Mathematical analysis of plasmonics resonances for nanoparticles and applications

Matias Ruiz

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PSL Research University

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Analyse Mathématique de Résonances Plasmoniques pour des Nanoparticules et Applications

Mathematical Analysis of Plasmonic Resonances for Nanoparticles and Applications

par Matias Ruiz

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Composition du Jury :

M. Habib Ammari
ETH Zurich
Directeur de thèse

M. Oscar Bruno
California Institute of Technology
Rapporteur

M. Gilles Lebeau
Université de Nice - Sophia Antipolis
Rapporteur

Mme. Liliana Borcea
University of Michigan
Examinatrice

M. Josselin Garnier
École Polytechnique
Examinateur

M. Bertrand Maury
Université Paris Sud
Examinateur

M. Gabriel Peyré
École Normale Supérieure
Président du jury

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Contents

Notations

- $D \Subset \mathbb{R}^d$ denotes that D is an open, bounded and simply connected subset of $\mathbb{R}^d$;
- ∂D denotes the boundary of the open set $D \in \mathbb{R}^d$;
- We say that ∂D is of type $\mathcal{C}^{1,\alpha}$, for $0 < \alpha < 1$, if ∂D is locally Lipschitz of order $0 < \alpha < 1$;
- ν denotes the outward normal to ∂D and $\frac{\partial}{\partial \nu}$ the outward normal derivative;
- $\varphi\big|_{\pm}(x) = \lim_{t \to 0^+} \varphi(x \pm t\nu)$, $x \in \partial D$;
- Id denotes the identity operator;
- $H^s(\partial D)$ denotes the usual Sobolev space of order s on ∂D;
- $(\cdot, \cdot)_{-\frac{1}{2}, \frac{1}{2}}$ denotes the duality pairing between $H^{-\frac{1}{2}}(\partial D)$ and $H^{\frac{1}{2}}(\partial D)$;
- For any functional space $F(\partial D)$ defined on ∂D, $F_0(\partial D)$ denotes its zero mean subspace;
- $\mathcal{L}(E, F)$ denotes the set of bounded linear applications from E to F and $\mathcal{L}(E) := \mathcal{L}(E, E)$;
- For $\alpha = (\alpha_1, \alpha_2) \in \mathbb{N}^2$, $\partial_\alpha := \partial_1^{\alpha_1} \partial_d^{\alpha_2}$ and $\alpha! := \alpha_1! \alpha_2!$;
- $\chi(S)$, denotes the characteristic function of the set S;
- $\Re z$ denotes the real part of z;
- $\Im z$ denotes the imaginary part of z;
- $|x|$ denotes the norm of $x \in \mathbb{R}^d$;
- We denote by the Sommerfeld radiation condition for a function u in dimension $d = 2, 3$, the following condition:

$$\left| \frac{\partial u}{\partial |x|} - ik_m u \right| \leq C |x|^{-(d+1)/2}$$

as $|x| \to +\infty$ for some constant C independent of x.

to my parents, Cesar and Olga

Introduction

Light has been a major field of scientific curiosity and study since the beginning of science. Despite the age of the field, research in photonics is more active than ever, as evidenced by 2015 being proclaimed by the United Nations General Assembly as the "International Year of Light and Light-based Technologies". In the last decades, the field of photonics has seen a revolution due to the study of the anomalous properties of metallic particles, no bigger than some tens of nanometers, and their interaction with light. At this scale, and for some specific range of frequencies, this nanoparticles have the unique capability of enhancing the brightness and directivity of light, confining strong electromagnetic fields into advantageous directions. This phenomenon, called "plasmonic resonances for nanoparticles" or "surface plasmons", open a door for a wide range of applications, from novel healthcare techniques to efficient solar panels. To harvest such opportunities, a deep mathematical understanding of the interactive effects between the particle size, shape and contrasts in the electromagnetic parameters is required.

Although very significant experimental and modeling advances have been achieved in the field of nanoplasmonic during recent decades, very few properties have been introduced and analyzed in the mathematical literature. There is a clear lack of deep understanding of the theory of plasmonic resonance. The goal of this work is to fill some of these gaps - understand the mathematical structure of inverse problems arising in nanophotonics and propose, from a better mathematical basis, pertinent applications of plasmonic nanoparticles that will best meet the challenges of emerging nanotechnologies.

Plasmonic nanoparticles

Plasmonic nanoparticles are particles, typically made of gold or silver, whose size range in the order of a few to a hundred nanometers. At this scale, they behave as metamaterials, meaning that their conductivity and/or premeabilitty has negative real part. When an external light wave is incident on the nanoparticle, the cloud of free electrons on the surface of the particule oscilates at some specific frequencies, entering in a resonance mode; see Figure 1. These resonances depend on the electromagnetic parameters of the nanoparticle, those of the surrounding material, and the particle shape and size. High scattering and absorption cross sections (see [43] for precise definitions of these quantities) and strong near-fields are unique effects of plasmonic resonant nanoparticles; see Figure 2.

Even though plasmonic nanoparticles have drawn the attention of scientists mainly in the 20th century, they have been first put into use thousands of years ago, when ancient civilizations made use of them for decoration and artistic purposes. Figure 3 shows the Lycurgus cup, a decorative Roman

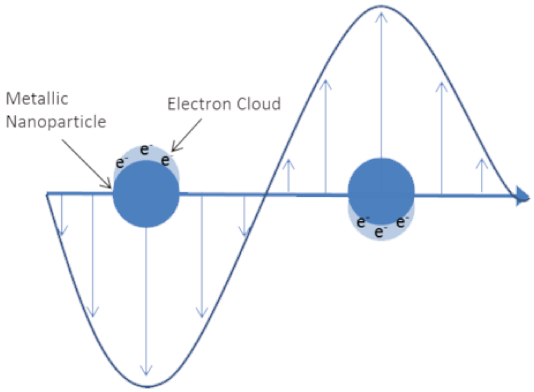

FIGURE 1: Electron cloud oscilating under the presence of an electric field. (nanohybrids.net)

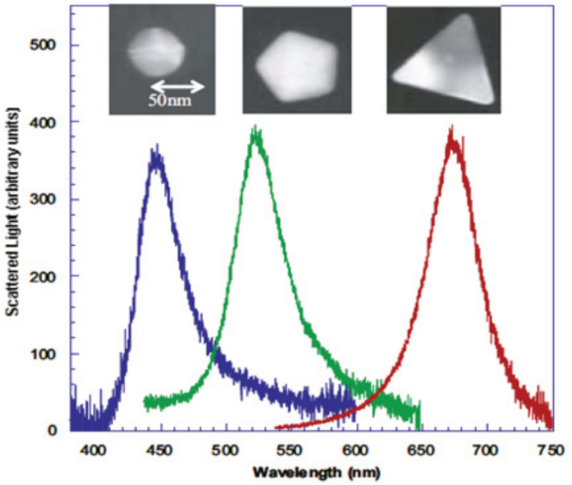

FIGURE 2: Scattering response as a function of the wavelength of the incoming electromagnetic wave, for different geometries of the plasmonic nanoparticle. (V.A.G. Rivera et al.)

treasure from about AD400; it is made of a glass containing gold-silver alloyed nanoparticles and reveals a brilliant red when light is shone through it. The reason for this? the nanoparticles in the glass resonate at the red-colored frequencies. For this and other examples one can observe the range of different colors (blue to red) mainly arose from different metal/metal oxides particles embedded in a dielectric (ceramic) matrix. In the late 8th century, the Iranian chemist Jaber-ibn-Hayyan, was one of the first researchers who studied the technical recipes dealing with the manufacture of colored glasses, making lustre-painted glass (stained glass) and coloring gemstones.

The physical understanding of this phenomenon started in the first decade of the 20th century with the work of Mie and Ritchie for small particles and flat interfaces, respectively. However, it was the work of Otto, Economou and Kretschmann that started the modern plasmonics field by providing a detailed theoretical description and experimental methods to excite surface plasmon polaritons on films of noble metals.

FIGURE 3: Lycurgus cup. Its galss contains gold-silver nanoparticles that resonates with red frequencies of incoming ight. (trustees of the British museum)

Driven by the search for new materials with interesting and unique optical properties, the field of plasmonic nanoparticles has grown immensely in the last decade [68]. Recent advances in nanofabrication techniques have made it possible to construct complex nanostructures such as arrays using plasmonic nanoparticles as components, allowing the design of new kinds of materials. Among this structures we find the so called "metasurfaces", consisting in a thin layer of periodically arranged nanoparticles mounted over a dielectric. This kind of composites are capable to control and transform optical waves in order to reduce scattering and make objects invisible or even trap electromagnetic waves in the goal of making efficient photovoltaic cells.

Another thriving interest for optical studies of plasmon resonant nanoparticles is due to their recently proposed use in molecular biology, where the strong field enhancement can be used as efficient contrast for biological and cell imaging applications [48].

Nanoparticles are also being used in thermotherapy as nanometric heat-generators that can be activated remotely by external electromagnetic fields. Nanotherapy relies on a simple mechanism. First nanoparticles become attached to tumor cells using selective biomolecular linkers. Then heat generated by optically-simulated plasmonic nanoparticles destroys the tumor cells [51].

Scientists have long dreamt of an optical microscope that can be used to see, noninvasively and in vivo, the details of living matter and other materials. When attempting to image nanoscale structures with visible light, a fundamental problem arises: diffraction effects limit the resolution to a dimension of roughly half the wavelength. Recently, the use of plasmonics nanoparticles has been proposed in a number of emerging techniques that achieve resolution below the conventional resolution limit into what is called super-resolution techniques.

Contributions

It is important to understand the collective behavior of plasmonic nanoparticles to derive the macroscopic optical properties of materials with a dilute set of plasmonic inclusions. In this regard, we have obtained effective properties

of a periodic arrangement of arbitrarily-shaped nanoparticles and derived a condition on the volume fraction of the nanoparticles that insures the validity of the Maxwell-Garnett theory for predicting the effective optical properties of systems embedded in a dielectric host material at the plasmonic resonances.

One of the most important parameters in the context of applications is the position of the resonances in terms of the wavelength or frequency. A longstanding problem is to tune this position by changing the particle size or the concentration of the nanoparticles in a solvent [49, 68]. It was experimentally observed, for instance, in [49, 89] that the scaling behavior of nanoparticles is critical. The question of how the resonant properties of plasmonic nanoparticles develops with increasing size or/and concentration is therefore fundamental.

According to the quasi-static approximation for small particles, the surface plasmon resonance peak occurs when the particle's polarizability is maximized. At this limit, since resonances are directly related to the Neumann-Poincaré integral operator, they are size-independent. However, as the particle size increases, a shift in the value of the resonances can be observed, for instance, in [49, 81, 89]. Using the Helmholtz equation to model light propagation we have precisely quantified the shift of the plasmonic resonance and the scattering absorption enhancement for a single nanoparticle.

At the quasi-static limit, we gave a proof that the averages over the orientation of scattering and extinction cross-sections of a randomly oriented nanoparticle are given in terms of the imaginary part of the polarization tensor. Moreover, we have derived bounds in dimension two (optimal bounds) and three for the absorption and scattering cross-sections.

Later on, we have generalized these results, providing the first mathematical study of the shift in plasmon resonance using the full Maxwell equations. Surprisingly, it turns out that in this case not only the spectrum of the Neumann-Poincaré operator plays a role in the resonance of the nanoparticles, but also its negative. We have explained how in the quasi-static limit, only the spectrum of the Neumann-Poincaré operator can be excited and that its negative can only be excited as in higher-order terms in the expansion of the electric field versus the size of the particle.

Due to their high absorption enhancement, monitoring the temperature generated by the nanoparticles in the plasmonic resonance could be crucial for thermoterapy success. We have established an asymptotic expansion for the temperature in the border of arbitrary shaped particles, which turns out to be related, again, to the eigenvalues of the Neumann-Poincaré operator.

If we consider the scattering by a layer of periodic plasmonic nanoparticles mounted on a perfectly conducting sheet, as the thickness of the layer, which is of the same order as the diameter of the individual nanoparticles, is negligible compared with the wavelength, it can be approximated by an impedance boundary condition. We have proved that at some resonant frequencies, the thin layer has anomalous reflection properties and can be viewed as a metasurface allowing the control and transformation of electromagnetic waves.

We have also proved that using plasmonic resonances one can classify the shape of a class of domains with real algebraic boundaries and on the other hand recover the separation distance between two components of multiple

connected domains. These results have important applications in nanophotonics. They can be used in order to identify the shape and separation distance between plasmonic nanoparticles having known material parameters from measured plasmonic resonances, for which the scattering cross-section is maximized.

The main objective of super-resolution is to create imaging approaches for objects significantly smaller than half the wavelenght, based on the use of resonant plasmonic nanoparticles. In a homogeneous space, particles smaller than half the wavelength cannot be resolved because the point spread function, which is the imaginary part of the Green function, has a width of roughly half the wavelenght. By following the methodology of [30], we have shown that super-resolution can be achieved when replacing the homogeneous media by a composite made of plasmonic nanoparticles.

Moreover, we have shown that we can make use of plasmonic nanoparticles to recover fine details of a subwavelength non plasmonic nanoparticles, providing a mathematical foundation for plasmonic biosensing. These results open a door for the ill-posed inverse problem of reconstructing small objects from far-field measurements.

The results obtained in this thesis have been published in [22–27].

Part I

Mathematical Analysis of Plasmonic Resonances

Chapter 1

The Quasi-Static Limit

Contents

1.1 Introduction

Consider the scattering problem of iluminating a nanoparticle immersed in a homogeneous medium. When the size of the nanoparticle is significantly smaller than the wavelength of the incomming light, Maxwell equations can be approximated by the equation (1.3) [87]. We say that we are working in the quasi-static limit. This regime have been extensively used by the physics community to model the scattering of light by small nanoparticles such as plasmonic nanoparticles. In the mathematics community, the first efforts to give rigourous results on the plasmonic resonance phenomena have been done in this framwork [52]. In the first part of this chapter we give a brief review of the mathematical analysis of the plasmonic resonances for nanoparticles in the quasi-static regime. This analysis rely strongly in the use of layer potential techniques for the Laplace equation.

Secondly, we investigate the overall optical properties of a collection of plasmonic nanoparticles. We treat a composite material in which plasmonic nanoparticles are embedded and isolated from each other. The Maxwell-Garnett theory provides a simple model for calculating the macroscopic optical properties of materials with a dilute inclusion of spherical nanoparticles [18]. Here, we extend the validity of the Maxwell-Garnett effective medium theory in order to describe the behavior of a system of arbitrary-shaped plasmonic resonant nanoparticles. We rigorously derive a condition on the volume fraction of the nanoparticles that insures its validity at the plasmonic resonances. To do so, we introduce the notion of plasmonic resonances for particles with anisotropic electromagnetic materials. This notion is introduced here for the first time.

In section 1.4 we analyze the anisotropic quasi-static problem in terms of layer potentials and define the plasmonic resonances for anisotropic nanoparticles. Formulas for a small anisotropic perturbation of resonances of the isotropic formulas are derived.

Section 1.5 is devoted to establish a Maxwell-Garnett type theory for approximating the plasmonic resonances of a periodic arrangement of arbitrary-shaped nanoparticles.

1.2 Preliminaries

In this section we recall important properties of the layer potentials for the Laplacian that will be of great use throughout this thesis.

1.2.1 Layer potentials for the Laplace equation

Consider a domain D with boundary ∂D of type $\mathcal{C}^{1,\alpha}$ for $0 < \alpha < 1$. Let ν denote the outward normal to ∂D. Define the single layer potential

$$\mathcal{S}_D[\varphi](x) = \int_{\partial D} G(x,y)\varphi(y)d\sigma(y), \qquad x \in \partial D, x \in \mathbb{R}^d \backslash \partial D,$$

where $G(x,y)$ is the green function for the Laplacian.
For $d = 2$

$$G(x,y) = \frac{1}{2\pi} \log|x-y|.$$

For $d = 3$

$$G(x, y) = -\frac{1}{4\pi|x - y|}.$$

We have the following lemma

Lemma 1.2.1. *1. For $\varphi \in H^{-\frac{1}{2}}(\partial D)$, $\mathcal{S}_D[\varphi] \in H^1(\mathbb{R}^d \backslash \partial D)$ is an Harmonic function on $\mathbb{R}^d \backslash \partial D$;*

2. $\mathcal{S}_D[\varphi](x)\big|_+ = \mathcal{S}_D[\varphi](x)\big|_-$;

3. For $d = 3$, $\mathcal{S}_D[\varphi](x) = O(\frac{1}{|x|^2})$ as $|x| \to \infty$ and $\mathcal{S}_D[\cdot] : H^{-\frac{1}{2}}(\partial D) \to H^{\frac{1}{2}}(\partial D)$ is invertible, negative definite and self-adjoint for the duality paring $H^{-\frac{1}{2}}(\partial D), H^{\frac{1}{2}}(\partial D)$;

4. Same for $d = 2$ under condition that $\int_{\partial D} \varphi d\sigma = 0$.

A more detailed analysis for the case $d = 2$ is given in Appendix A.

The Neumann-Poincaré operator (NP) $\mathcal{K}_D^*$ associated with D is defined as follows:

$$\mathcal{K}_D^*[\varphi](x) = \frac{1}{2\pi} \int_{\partial D} \frac{\langle x - y, \nu_x \rangle}{|x - y|^d} \varphi(y) d\sigma(y), \quad x \in \partial D.$$

It is related to the single layer potential $\mathcal{S}_D$ by the following jump relation:

$$\frac{\partial \mathcal{S}_D[\varphi]}{\partial \nu}\Big|_\pm = (\pm\frac{1}{2} I + \mathcal{K}_D^*)[\varphi] \quad \text{for } \varphi \in H^{-1/2}(\partial D). \tag{1.1}$$

It can be shown that the operator $\lambda I - \mathcal{K}_D^* : H^{-1/2}(\partial D) \to H^{-1/2}(\partial D)$ is invertible for any $|\lambda| > 1/2$. Furthermore, $\mathcal{K}_D^*$ is compact, its spectrum is discrete and contained in $]-1/2, 1/2]$ with 0 being an accumulation point; see for instance [18, 32] for more details.

In general, $\mathcal{K}_D^*$ is not symmetric for the pairing $(\cdot, \cdot)_{\frac{1}{2}, -\frac{1}{2}}$. Nevertheless, using Calderon's identity

$$\mathcal{K}_D \mathcal{S}_D = \mathcal{S}_D \mathcal{K}_D^*,$$

$\mathcal{K}_D^*$ can be symmetrized with the following inner product

$$(u, v)_{\mathcal{H}^*} := -(\mathcal{S}_D[v], u)_{\frac{1}{2}, -\frac{1}{2}}.$$

It can be shown that in $\mathbb{R}^3$, $(\cdot, \cdot)_{\mathcal{H}^*}$ defines a Hilbert space, equivalent to $H^{-1/2}(\partial D)$ [12, 32, 61, 65]. In $\mathbb{R}^2$ a similar analysis can be done to symmetrize $\mathcal{K}_D^*$. We refer the reader to Appendix A.

Let (λ_j, φ_j), $j = 0, 1, 2, \ldots$ be the eigenvalue and normalized eigenfunction pair of $\mathcal{K}_D^*$ in $\mathcal{H}^*(\partial D)$. From the spectral theorem, we know that $\lambda_j \to 0$ for $j \to \infty$ and φ_j form a base of $\mathcal{H}^*(\partial D)$. Therefore, the following representation formula holds: for any $\varphi \in H^{-1/2}(\partial D)$,

$$\mathcal{K}_D^*[\varphi] = \sum_{j=0}^{\infty} \lambda_j (\varphi, \varphi_j)_{\mathcal{H}^*} \otimes \varphi_j.$$

From the jump formula (1.1), we can see that $1/2$ is always an eigenvalue of $\mathcal{K}_D^*$. If the D is simply connected, there is only one eigenvalue taking the value $1/2$. We denote this eigenvalue by λ_0 and its corresponding eigenfunction φ_0.

In $\mathbb{R}^3$, let $\mathcal{H}(\partial D)$ be the space $H^{\frac{1}{2}}(\partial D)$ equipped with the following equivalent inner product

$$(u, v)_{\mathcal{H}} = ((-\mathcal{S}_D)^{-1}[u], v)_{-\frac{1}{2},\frac{1}{2}}. \tag{1.2}$$

Then, $\mathcal{S}_D$ is an isometry between $\mathcal{H}^*(\partial D)$ and $\mathcal{H}(\partial D)$.

A smilar result can be found in $\mathbb{R}^2$, see Appendix A.

1.3 Layer potential formulation for the scattering problem

We consider the scattering problem of a time-harmonic wave u^i incident on a plasmonic nanoparticle. The homogeneous medium is characterized by its electric permittivity ε_m that we assume to be real and strictly positive. The particle occupying a bounded and simply connected domain $D \Subset \mathbb{R}^d$ of class $\mathcal{C}^{1,\alpha}$ for some $0 < \alpha < 1$ is characterized by electric isotropic permittivity ε_c which may depend on the frequency of the incoming wave ω by the Drude model as

$$\varepsilon_c = \varepsilon_c(\omega) = \left(1 - \frac{\omega_p^2}{\omega(\omega + i\gamma)}\right)\varepsilon_0.$$

Here, ω_p is called the plasmon frequency, γ the damping parameter and ε_0 is the permittivity of the free space.

Assume that $\Re\varepsilon_c < 0$, $\Im\varepsilon_c > 0$, and define

$$\varepsilon_D = \varepsilon_m \chi(\mathbb{R}^d \backslash \overline{D}) + \varepsilon_c \chi(D).$$

where χ denotes the characteristic function. When the wavelength of the incoming wave is much larger than the particle's size, the following is a good approximation of the Maxwell equations.

$$\begin{cases} \nabla \cdot \varepsilon_D \nabla u = 0 \quad \text{in } \mathbb{R}^d \backslash \partial D, \\ u_+ - u_- = 0 \quad \text{on } \partial D, \\ \varepsilon_c \frac{\partial u}{\partial \nu}\big|_+ - \varepsilon_m \frac{\partial u}{\partial \nu}\big|_- = 0 \quad \text{on } \partial D, \\ u - u^i = O(\frac{1}{|x|^{d-1}}), \quad |x| \to \infty. \end{cases} \tag{1.3}$$

Here u corresponds to the electric potential. For some $\varphi \in H^{-\frac{1}{2}}(\partial D)$, the solution u can be written as

$$u(x) = u^i + \mathcal{S}_D[\varphi].$$

From Lemma 1.2.1 we can see that only the transmission conditions

$$\varepsilon_c \frac{\partial u}{\partial \nu}\big|_+ - \varepsilon_m \frac{\partial u}{\partial \nu}\big|_- = 0 \quad \text{on } \partial D,$$

need to be satisfied. This translates into

$$\varepsilon_c \frac{\partial \mathcal{S}_D[\varphi]}{\partial \nu}(x)\big|_+ - \varepsilon_m \frac{\partial \mathcal{S}_D[\varphi]}{\partial \nu}(x)\big|_- = (\varepsilon_m - \varepsilon_c)\frac{\partial u^i}{\partial \nu} \quad \text{on } \partial D.$$

From the jump formula for the single layer potential $\mathcal{S}_D$, i.e

$$\frac{\partial \mathcal{S}_D[\varphi]}{\partial \nu}(x)\big|_\pm = \big(\pm\frac{1}{2}Id + \mathcal{K}_D^*\big)[\varphi](x), \quad x \in \partial D.$$

we have

$$\big(\lambda - \mathcal{K}_D^*\big)[\varphi] = \frac{\partial u^i}{\partial \nu}, \tag{1.4}$$

with

$$\lambda = \frac{\varepsilon_m + \varepsilon_c}{2(\varepsilon_m - \varepsilon_c)}.$$

Finally

$$\begin{aligned} u &= u^i + \mathcal{S}_D(\lambda - \mathcal{K}_D^*)^{-1}[\frac{\partial u^i}{\partial \nu}] \\ &= u^i + \sum_{j=1}^{\infty} \frac{(\frac{\partial u^i}{\partial \nu}, \varphi_j)_{\mathcal{H}^*}}{\lambda - \lambda_j}\mathcal{S}_D[\varphi_j]. \end{aligned} \tag{1.5}$$

Recall that λ_j are eigenvalues $\mathcal{K}_D^*$ and they satisfy $|\lambda_j| < 1/2$. In the plasmonic case, $\Re\varepsilon_c(\omega)$ can take negative values. Then it holds that $|\Re\lambda(\omega)| < 1/2$ and $0 \leq \Im\varepsilon_c(\omega) \ll 1$. So, for a certain frequency ω_j, the value of $\lambda(\omega_j)$ can be very close to an eigenvalue λ_j of the NP operator. Then, in (1.5), the mode $\mathcal{S}_D[\varphi_j]$ will be amplified provided that $(\frac{\partial u^i}{\partial \nu}, \varphi_j)_{\mathcal{H}^*}$ is non-zero. As a result, the scattered field $u - u^i$ will show a resonant behavior. This phenomenon is called the plasmonic quasi-static resonance.

1.3.1 Contracted generalized polarization tensors

Decomposition (1.5) of u together with

$$u^i(x) = \sum_{\alpha \in \mathbb{N}^d} \frac{1}{\alpha!}\partial^\alpha u^i(0)x^\alpha$$

and

$$G(x,y) = \sum_{|\beta|=0}^{+\infty} \frac{(-1)^{|\beta|}}{\beta!}\partial_x^\beta \Gamma(x)y^\beta, \quad y \text{ in a compact set, } |x| \to +\infty,$$

where $G(x,y)$ is the fundamental solution to the Laplacian, yields the far-field behavior [4, p. 77]

$$(u - u^i)(x) = \sum_{|\alpha|,|\beta| \geq 1} \frac{1}{\alpha!\beta!}\partial^\alpha u^i(0)\left[\int_{\partial D} y^\beta (\lambda I - \mathcal{K}_D^*)^{-1}[\frac{\partial x^\alpha}{\partial \nu}](y)d\sigma(y)\right]\partial^\beta G(x,0) \tag{1.6}$$

as $|x| \to +\infty$. Introduce the *generalized polarization tensors* [4]:

$$M_{\alpha\beta}(\lambda, D) := \int_{\partial D} y^\beta (\lambda I - \mathcal{K}_D^*)^{-1}[\frac{\partial x^\alpha}{\partial \nu}](y)\, d\sigma(y), \qquad \alpha, \beta \in \mathbb{N}^d.$$

They will be of great use in chapter 8. We call $M := M_{\alpha\beta}$ for $|\alpha| = |\beta| = 1$ the first-order polarization tensor.

Suppose that $D = z + \delta B$, where B has size of order 1. Then, from (1.6) we have

Theorem 1.3.1. *In the far field*

$$u^s(x) = u^i(x) - \nabla_y G(x, 0) M(\lambda, D) \nabla u^i(0) + O\big(\frac{\delta^{d+1}}{\operatorname{dist}(\lambda, \sigma(\mathcal{K}_D^*))}\big).$$

For a positive integer m, let $P_m(x)$ be the complex-valued polynomial

$$P_m(x) = (x_1 + ix_2)^m := \sum_{|\alpha|=m} a_\alpha^m x^\alpha + i \sum_{|\beta|=m} b_\beta^m x^\beta. \tag{1.7}$$

Using polar coordinates $x = re^{i\theta}$, the above coefficients a_α^m and b_β^m can also be characterized by

$$\sum_{|\alpha|=m} a_\alpha^m x^\alpha = r^m \cos m\theta, \text{ and } \sum_{|\beta|=m} b_\beta^m x^\beta = r^m \sin m\theta. \tag{1.8}$$

We introduce the *contracted generalized polarization tensors* to be the following linear combinations of generalized polarization tensors using the coefficients in (1.7):

$$M_{mn}^{cc} = \sum_{|\alpha|=m} \sum_{|\beta|=n} a_\alpha^m a_\beta^n M_{\alpha\beta}, \quad M_{mn}^{cs} = \sum_{|\alpha|=m} \sum_{|\beta|=n} a_\alpha^m b_\beta^n M_{\alpha\beta},$$
$$M_{mn}^{sc} = \sum_{|\alpha|=m} \sum_{|\beta|=n} b_\alpha^m a_\beta^n M_{\alpha\beta}, \quad M_{mn}^{ss} = \sum_{|\alpha|=m} \sum_{|\beta|=n} b_\alpha^m b_\beta^n M_{\alpha\beta}.$$

It is clear that

$$M_{mn}^{cc} = \int_{\partial D} \Re(P_n)(\lambda I - \mathcal{K}_D^*)^{-1}[\frac{\partial \Re(P_m)}{\partial \nu}]\, d\sigma,$$
$$M_{mn}^{cs} = \int_{\partial D} \Im(P_n)(\lambda I - \mathcal{K}_D^*)^{-1}[\frac{\partial \Re(P_m)}{\partial \nu}]\, d\sigma,$$
$$M_{mn}^{sc} = \int_{\partial D} \Re(P_n)(\lambda I - \mathcal{K}_D^*)^{-1}[\frac{\partial \Im(P_m)}{\partial \nu}]\, d\sigma,$$
$$M_{mn}^{ss} = \int_{\partial D} \Im(P_n)(\lambda I - \mathcal{K}_D^*)^{-1}[\frac{\partial \Im(P_m)}{\partial \nu}]\, d\sigma.$$

We refer to [18] for further details

As recently shown [11, 18], the contracted generalized polarization tensors can efficiently be used for domain classification. They provide a natural tool for describing shapes. In imaging applications, they can be stably reconstructed from the data by solving a least-squares problem. They capture

high-frequency shape oscillations as well as topology. High-frequency oscillations of the shape of a domain are only contained in its high-order contracted generalized polarization tensors.

1.4 Plasmonic resonances for the anisotropic problem

In this section, we consider the scattering problem of a time-harmonic wave u^i, incident on a plasmonic anisotropic nanoparticle. The homogeneous medium is characterized by its electric permittivity ε_m, while the particle occupying a bounded and simply connected domain $\Omega \Subset \mathbb{R}^3$ of class $\mathcal{C}^{1,\alpha}$ for $0 < \alpha < 1$ is characterized by electric anisotropic permittivity A. We consider A to be a positive-definite symmetric matrix.

In the quasi-static regime, the problem can be modeled as follows:

$$\begin{aligned} \nabla \cdot \big(\varepsilon_m Id\chi(\mathbb{R}^3\backslash\bar{\Omega}) + A\chi(\Omega)\big)\nabla u = 0, \\ |u - u^i| = O(|x|^{-2}), \quad |x| \to +\infty, \end{aligned} \tag{1.9}$$

where χ denotes the characteristic function and u^i is a harmonic function in $\mathbb{R}^3$.
We are interested in finding the plasmonic resonances for problem (1.9).
First, introduce the fundamental solution to the operator $\nabla \cdot A\nabla$ in dimension three

$$G^A(x) = -\frac{1}{4\pi\sqrt{\det(A)}|A_* x|}$$

with $A_* = \sqrt{A^{-1}}$. From now on, we denote $G^A(x,y) := G^A(x-y)$.
The single-layer potential associated with A is

$$\begin{aligned} \mathcal{S}^A_\Omega[\varphi] : H^{-\frac{1}{2}}(\partial\Omega) &\longrightarrow H^{\frac{1}{2}}(\partial\Omega) \\ \varphi &\longmapsto \mathcal{S}^A_\Omega[\varphi](x) = \int_{\partial\Omega} G^A(x,y)\varphi(y)d\sigma(y), \quad x \in \mathbb{R}^3. \end{aligned}$$

We can represent the unique solution to (1.9) in the following form [18]:

$$u(x) = \begin{cases} u^i + \mathcal{S}_\Omega[\psi], & x \in \mathbb{R}^3\backslash\bar{\Omega}, \\ \mathcal{S}^A_\Omega[\phi], & x \in \Omega, \end{cases}$$

where $(\psi, \phi) \in \big(H^{-\frac{1}{2}}(\partial\Omega)\big)^2$ is the unique solution to the following system of integral equations on $\partial\Omega$:

$$\begin{cases} \mathcal{S}_\Omega[\psi] - \mathcal{S}^A_\Omega[\phi] & = -u^i, \\ \varepsilon_m \dfrac{\partial \mathcal{S}_\Omega[\psi]}{\partial\nu}\Big|_+ - \nu \cdot A\nabla\mathcal{S}^A_\Omega[\phi]\Big|_- & = -\varepsilon_m \dfrac{\partial u^i}{\partial\nu}. \end{cases} \tag{1.10}$$

Lemma 1.4.1. *The operator $\mathcal{S}^A_\Omega : H^{-\frac{1}{2}}(\partial\Omega) \to H^{\frac{1}{2}}(\partial\Omega)$ is invertible. Moreover, we have the jump formula*

$$\nu \cdot A\nabla\mathcal{S}^A_\Omega\Big|_\pm = \pm\frac{1}{2}Id + (\mathcal{K}^A_\Omega)^*$$

with

$$(\mathcal{K}_\Omega^A)^*[\varphi](x) = \int_{\partial\Omega} -\frac{\big(x-y,\nu(x)\big)}{4\pi\sqrt{\det(A)}|A_*(x-y)|^3}\varphi(y)d\sigma(y).$$

Proof. Let $\mathcal{T}_{A_*} \in \mathcal{L}(H^s(\partial\widetilde{\Omega}), H^s(\partial\Omega))$ be such that $\mathcal{T}_{A_*}[\varphi](x) = \varphi(A_*x)$ for $\varphi \in H^s(\partial\widetilde{\Omega})$ and $\widetilde{\Omega} = A_*\Omega$. Let $r_\nu \in \mathcal{L}(H^s(\partial\Omega), H^s(\partial\Omega))$ be such that $r_\nu[\varphi](x) = |A_*^{-1}\nu(x)|\varphi(x)$. It follows by the change of variables $\widetilde{y} = A_*y$ that $d\sigma(\widetilde{y}) = \det\sqrt{A_*}|A_*^{-1}\nu(y)|d\sigma(y)$. Thus,

$$\mathcal{S}_\Omega^A = \mathcal{T}_{A_*}\mathcal{S}_{\widetilde{\Omega}}\mathcal{T}_{A_*}^{-1}r_\nu^{-1},$$

and in particular $\mathcal{S}_\Omega^A$ is invertible and its inverse $(\mathcal{S}_\Omega^A)^{-1} = r_\nu\mathcal{T}_{A_*}\mathcal{S}_{\widetilde{\Omega}}^{-1}\mathcal{T}_{A_*}^{-1}$.

Note that, for $x \in \partial\Omega$,

$$\widetilde{\nu}(\widetilde{x}) = \frac{A_*^{-1}\nu(x)}{|A_*^{-1}\nu(x)|},$$

where $\widetilde{\nu}(\widetilde{x})$ is the outward normal to $\partial\widetilde{\Omega}$ at $\widetilde{x} = A_*x$. We have

$$\begin{aligned}
\nu\cdot A\nabla\mathcal{S}_\Omega^A\Big|_\pm &= \nu\cdot A\nabla_x\Big(\mathcal{T}_{A_*}\mathcal{S}_{\widetilde{\Omega}}\mathcal{T}_{A_*}^{-1}r_\nu^{-1}\Big)\Big|_\pm \\
&= \nu\cdot AA_*\Big(\mathcal{T}_{A_*}\nabla_{\widetilde{x}}\mathcal{S}_{\widetilde{\Omega}}\mathcal{T}_{A_*}^{-1}r_\nu^{-1}\Big)\Big|_\pm \\
&= |A_*^{-1}\nu|\widetilde{\nu}\cdot\Big(\mathcal{T}_{A_*}\nabla_{\widetilde{x}}\mathcal{S}_{\widetilde{\Omega}}\mathcal{T}_{A_*}^{-1}r_\nu^{-1}\Big)\Big|_\pm \\
&= \pm\frac{1}{2}Id + (r_\nu\mathcal{T}_{A_*})\mathcal{K}_{\widetilde{\Omega}}^*(r_\nu\mathcal{T}_{A_*})^{-1}. \qquad (1.11)
\end{aligned}$$

The result follows from a change of variables in the expression of the operator

$$(\mathcal{K}_\Omega^A)^* := (r_\nu\mathcal{T}_{A_*})\mathcal{K}_{\widetilde{\Omega}}^*(r_\nu\mathcal{T}_{A_*})^{-1}.$$

□

Lemma 1.4.2. *$\mathcal{S}_\Omega^A$ is negative definite for the duality pairing $(\cdot,\cdot)_{-\frac{1}{2},\frac{1}{2}}$ and we can define a new inner product*

$$(u,v)_{\mathcal{H}_A^*} = -(u,\mathcal{S}_\Omega^A[v])_{-\frac{1}{2},\frac{1}{2}}$$

with which $\mathcal{H}_A^*(\partial\Omega)$, the space induced by $(\cdot,\cdot)_{\mathcal{H}_A^*}$, is equivalent to $H^{-\frac{1}{2}}(\partial\Omega)$.

Proof. Let $\varphi \in H^{-\frac{1}{2}}(\partial\Omega)$. Using Lemma 1.4.1, we have

$$\varphi = \nu\cdot A\nabla\mathcal{S}_\Omega^A[\varphi]\Big|_+ - \nu\cdot A\nabla\mathcal{S}_\Omega^A[\varphi]\Big|_-.$$

Thus

$$\begin{aligned}
&\int_{\partial\Omega} \varphi(x)\mathcal{S}_\Omega^A[\varphi](x)d\sigma(x)\\
=\ & \int_{\partial\Omega} \nu\cdot A\nabla\mathcal{S}_\Omega^A[\varphi]\Big|_+(x)\mathcal{S}_\Omega^A[\varphi](x)d\sigma(x) - \int_{\partial\Omega} \nu\cdot A\nabla\mathcal{S}_\Omega^A[\varphi]\Big|_-(x)\mathcal{S}_\Omega^A[\varphi](x)d\sigma(x)\\
=\ & -\int_{\mathbb{R}^3\setminus\bar\Omega} \nabla\mathcal{S}_\Omega^A[\varphi](x)\cdot A\nabla\mathcal{S}_\Omega^A[\varphi](x)d\sigma(x) - \int_{\mathbb{R}^3\setminus\bar\Omega} \mathcal{S}_\Omega^A[\varphi](x)\nabla\cdot A\nabla\mathcal{S}_\Omega^A[\varphi](x)d\sigma(x)\\
& -\int_{\Omega} \nabla\mathcal{S}_\Omega^A[\varphi](x)\cdot A\nabla\mathcal{S}_\Omega^A[\varphi](x)d\sigma(x) + \int_{\Omega} \mathcal{S}_\Omega^A[\varphi](x)\nabla\cdot A\nabla\mathcal{S}_\Omega^A[\varphi](x)d\sigma(x)\\
=\ & -\int_{\mathbb{R}^3} \nabla\mathcal{S}_\Omega^A[\varphi](x)\cdot A\nabla\mathcal{S}_\Omega^A[\varphi](x)d\sigma(x) \le 0,
\end{aligned}$$

where the equality is achieved if and only if $\varphi = 0$. Here we have used an integration by parts, the fact that $\mathcal{S}_\Omega^A[\varphi](x) = O(|x|^{-1})$ as $|x| \to \infty$, $\nabla\cdot A\nabla\mathcal{S}_\Omega^A[\varphi](x) = 0$ for $x \in \mathbb{R}^3\setminus\partial\Omega$ and that A is positive-definite.
In the same manner, it is known that

$$\|\varphi\|^2_{\mathcal{H}^*} = -\int_{\partial\Omega} \varphi(x)\mathcal{S}_\Omega[\varphi](x)d\sigma(x) = \int_{\mathbb{R}^3} |\nabla\mathcal{S}_\Omega[\varphi](x)|^2 d\sigma(x).$$

Since A is positive-definite we have

$$c\|\varphi\|^2_{\mathcal{H}^*} \le -\int_{\partial\Omega} \varphi(x)\mathcal{S}_\Omega^A[\varphi](x)d\sigma(x) \le C\|\varphi\|^2_{\mathcal{H}^*}$$

for some constants $c, C > 0$.
Using the fact that $\mathcal{H}^*(\partial\Omega)$ is equivalent to $H^{-\frac{1}{2}}(\partial\Omega)$, we get the desired result. □

From (1.10) we have $\phi = (\mathcal{S}_\Omega^A)^{-1}(\mathcal{S}_\Omega[\psi] + u^i)$, whereas, by Lemma 1.4.1, the following equation holds for ψ:

$$\mathcal{Q}_A[\psi] = F \tag{1.12}$$

with

$$\mathcal{Q}_A = \frac{1}{2}\big(\varepsilon_m Id + (\mathcal{S}_\Omega^A)^{-1}\mathcal{S}_\Omega\big) + \big(\varepsilon_m\mathcal{K}_\Omega^* - (\mathcal{K}_\Omega^A)^*(\mathcal{S}_\Omega^A)^{-1}\mathcal{S}_\Omega\big), \tag{1.13}$$

and

$$F = -\varepsilon_m\frac{\partial u^i}{\partial\nu} + \nu\cdot A\nabla\mathcal{S}_\Omega^A[(\mathcal{S}_\Omega^A)^{-1}u^i]\Big|_-.$$

Propsition 1.4.1. *$\mathcal{Q}_A$ has a countable number of eigenvalues.*

Proof. It is clear that $(\mathcal{K}_\Omega^A)^* : H^{-\frac{1}{2}}(\partial\Omega) \to H^{-\frac{1}{2}}(\partial\Omega)$ is a compact operator. Hence, $\varepsilon_m\mathcal{K}_\Omega^* - (\mathcal{K}_\Omega^A)^*(\mathcal{S}_\Omega^A)^{-1}\mathcal{S}_\Omega$ is compact as well. Therefore, only the invertibility of $\frac{1}{2}\big(\varepsilon_m Id + (\mathcal{S}_\Omega^A)^{-1}\mathcal{S}_\Omega\big)$ needs to be proven.
Since $\mathcal{S}_\Omega^A$ is invertible, the invertibility of $\frac{1}{2}\big(\varepsilon_m Id + (\mathcal{S}_\Omega^A)^{-1}\mathcal{S}_\Omega\big)$ is equivalent to that of $\varepsilon_m\mathcal{S}_\Omega^A + \mathcal{S}_\Omega$.
Consider now, the bilinear form, for $(\varphi,\psi) \in (H^{-\frac{1}{2}}(\partial\Omega))^2$

$$B(\varphi,\psi) = -\varepsilon_m\int_{\partial\Omega} \varphi(x)\mathcal{S}_\Omega^A[\psi](x)d\sigma(x) - \int_{\partial\Omega} \varphi(x)\mathcal{S}_\Omega[\psi](x)d\sigma(x),$$

where $\varepsilon_m > 0$. From Lemma 1.4.2, we have

$$B(\psi,\psi) \geq c\|\psi\|^2_{H^{-\frac{1}{2}}(\partial\Omega)}$$

for some constant $c > 0$.
It follows then, from the Lax-Milgram theorem that $\varepsilon_m \mathcal{S}^A_\Omega + \mathcal{S}_\Omega$ is invertible in $H^{-\frac{1}{2}}(\partial\Omega)$, whence the result. □

Recall that the electromagnetic parameter of the problem, A, depends on the frequency, ω of the incident field. Therefore the operator $\mathcal{Q}_A$ is frequency dependent and we should write $\mathcal{Q}_A(\omega)$.
We say that ω is a plasmonic resonance if

$$|\mathrm{eig}_j(\mathcal{Q}_A(\omega))| \ll 1 \quad \text{and is locally minimal for some } j \in \mathbb{N},$$

where $\mathrm{eig}_j(\mathcal{Q}_A(\omega))$ stands for the j-th eigenvalue of $\mathcal{Q}_A(\omega)$.
Equivalently, we can say that ω is a plamonic resonance if

$$\omega = \arg\max_\omega \|\mathcal{Q}_A^{-1}(\omega)\|_{\mathcal{L}(\mathcal{H}^*(\partial\Omega))}. \tag{1.14}$$

From now on, we suppose that A is an anisotropic perturbation of an isotropic parameter, i.e., $A = \varepsilon_c(Id + P)$, with P being a symmetric matrix and $\|P\| \ll 1$.

Lemma 1.4.3. *Let $A = \varepsilon_c(Id + \delta R)$, with R being a symmetric matrix, $\|R\| = O(1)$ and $\delta \ll 1$. Let* Tr *denote the trace of a matrix. Then, as $\delta \to 0$, we have the following asymptotic expansions:*

$$\begin{aligned}
\mathcal{S}^A_\Omega &= \frac{1}{\varepsilon_c}\big(\mathcal{S}_\Omega + \delta\mathcal{S}_{\Omega,1} + o(\delta)\big),\\
(\mathcal{S}^A_\Omega)^{-1} &= \varepsilon_c\big(\mathcal{S}_\Omega^{-1} + \delta\mathcal{B}_{\Omega,1} + o(\delta)\big),\\
(\mathcal{K}^A_\Omega)^* &= \mathcal{K}^*_\Omega + \delta\mathcal{K}^*_{\Omega,1} + o(\delta)
\end{aligned}$$

with

$$\begin{aligned}
\mathcal{S}_{\Omega,1}[\varphi](x) &= -\frac{1}{2}\mathrm{Tr}(R)\mathcal{S}_\Omega[\varphi](x) - \frac{1}{2}\int_{\partial\Omega}\frac{\big(R(x-y),x-y\big)}{4\pi|x-y|^3}\varphi(y)d\sigma(y),\\
\mathcal{B}_{\Omega,1} &= -\mathcal{S}_\Omega^{-1}\mathcal{S}_{\Omega,1}\mathcal{S}_\Omega^{-1},\\
\mathcal{K}^*_{\Omega,1} &= -\frac{1}{2}\mathrm{Tr}(R)\mathcal{K}^*_\Omega[\varphi](x) - \frac{3}{2}\int_{\partial\Omega}\frac{\big(R(x-y),x-y\big)\big(x-y,\nu(x)\big)}{4\pi|x-y|^5}\varphi(y)d\sigma(y).
\end{aligned}$$

Proof. Recall that, for δ small enough,

$$\begin{aligned}
\sqrt{(I+\delta R)^{-1}} &= Id - \frac{\delta}{2}R + O(\delta^2),\\
\det(I+\delta R) = &= 1 + \delta\mathrm{Tr}(R) + o(\delta),\\
(1+\delta x + o(\delta))^s &= 1 + \delta s x + o(\delta), \quad s \in \mathbb{R}.
\end{aligned}$$

The results follow then from asymptotic expansions of $-\dfrac{1}{4\pi\sqrt{\det(A)}|A_* x|^\beta}$, $\beta = 1, 3$ and the identity

$$(\mathcal{S}^A_\Omega)^{-1} = \varepsilon_c(Id + \delta\mathcal{S}_\Omega^{-1}\mathcal{S}_{\Omega,1} + o(\delta))^{-1}\mathcal{S}_\Omega^{-1}.$$

□

Plugging the expressions above into the expression of $\mathcal{Q}_A$ we get the following result.

Lemma 1.4.4. *As $\delta \to 0$, the operator $\mathcal{Q}_A$ has the following asymptotic expansion*

$$\mathcal{Q}_A = \mathcal{Q}_{A,0} + \delta \mathcal{Q}_{A,1} + o(\delta),$$

where

$$\begin{aligned} \mathcal{Q}_{A,0} &= \frac{\varepsilon_m + \varepsilon_c}{2} Id + (\varepsilon_m - \varepsilon_c)\mathcal{K}_\Omega^*, \\ \mathcal{Q}_{A,1} &= \varepsilon_c\big((\frac{1}{2}Id - \mathcal{K}_\Omega^*)\mathcal{B}_{\Omega,1}\mathcal{S}_\Omega - \mathcal{K}_{\Omega,1}^*\big). \end{aligned}$$

We regard the operator $\mathcal{Q}_A$ as a perturbation of $\mathcal{Q}_{A,0}$. As in section 3.3, we use the standard perturbation theory to derive the perturbed eigenvalues and eigenvectors in $\mathcal{H}^*(\partial\Omega)$.

Let (λ_j, φ_j) be the eigenvalue and normalized eigenfunction pairs of $\mathcal{K}_\Omega^*$ in $\mathcal{H}^*(\partial\Omega)$ and τ_j the eigenvalues of $\mathcal{Q}_{A,0}$. We have

$$\tau_j = \frac{\varepsilon_m + \varepsilon_c}{2} + (\varepsilon_m - \varepsilon_c)\lambda_j.$$

For simplicity, we consider the case when λ_j is a simple eigenvalue of the operator $\mathcal{K}_\Omega^*$. Define

$$P_{j,l} = (\mathcal{Q}_{A,1}[\varphi_j], \varphi_l)_{\mathcal{H}^*}.$$

As $\delta \to 0$, the perturbed eigenvalue and eigenfunction have the following form:

$$\begin{aligned} \tau_j(\delta) &= \tau_j + \delta\tau_{j,1} + o(\delta), \\ \varphi_j(\delta) &= \varphi_j + \delta\varphi_{j,1} + o(\delta), \end{aligned}$$

where

$$\begin{aligned} \tau_{j,1} &= P_{jj}, \\ \varphi_{j,1} &= \sum_{l \neq j} \frac{P_{jl}}{(\varepsilon_m - \varepsilon_c)(\lambda_j - \lambda_l)} \varphi_l. \end{aligned}$$

1.5 A Maxwell-Garnett theory for plasmonic nanoparticles

In this subsection we derive effective properties of a system of plasmonic nanoparticles. To begin with, we consider a bounded and simply connected domain $\Omega \Subset \mathbb{R}^3$ of class $\mathcal{C}^{1,\alpha}$ for $0 < \alpha < 1$, filled with a composite material that consists of a matrix of constant electric permittivity ε_m and a set of periodically distributed plasmonic nanoparticles with (small) period η and electric permittivity ε_c.

Let $Y =]-1/2, 1/2[^3$ be the unit cell and denote $\delta = \eta^\beta$ for $\beta > 0$. We set

the (re-scaled) periodic function

$$\gamma = \varepsilon_m \chi(Y \backslash \bar{D}) + \varepsilon_c \chi(D),$$

where $D = \delta B$ with $B \Subset \mathbb{R}^3$ being of class $\mathcal{C}^{1,\alpha}$ and the volume of B, $|B|$, is assumed to be equal to 1. Thus, the electric permittivity of the composite is given by the periodic function

$$\gamma_\eta(x) = \gamma(x/\eta),$$

which has period η. Now, consider the problem

$$\nabla \cdot \gamma_\eta \nabla u_\eta = 0 \quad \text{in } \Omega, \tag{1.15}$$

with an appropriate boundary condition on $\partial\Omega$. Then, there exists a homogeneous, generally anisotropic, permittivity γ^*, such that the replacement, as $\eta \to 0$, of the original equation (1.15) by

$$\nabla \cdot \gamma^* \nabla u_0 = 0 \quad \text{in } \Omega$$

is a valid approximation in a certain sense. The coefficient γ^* is called an effective permittivity. It represents the overall macroscopic material property of the periodic composite made of plasmonic nanoparticles embedded in an isotropic matrix.

The (effective) matrix $\gamma^* = (\gamma^*_{pq})_{p,q=1,2,3}$ is defined by [18]

$$\gamma^*_{pq} = \int_Y \gamma(x) \nabla u_p(x) \cdot \nabla u_q(x) dx,$$

where u_p, for $p = 1, 2, 3$, is the unique solution to the cell problem

$$\begin{cases} \nabla \cdot \gamma \nabla u_p = 0 \quad \text{in } Y, \\ u_p - x_p \quad \text{periodic (in each direction) with period 1}, \\ \int_Y u_p(x) dx = 0. \end{cases} \tag{1.16}$$

Using Green's formula, we can rewrite γ^* in the following form:

$$\gamma^*_{pq} = \varepsilon_m \int_{\partial Y} u_q(x) \frac{\partial u_p}{\partial \nu}(x) d\sigma(x). \tag{1.17}$$

The matrix γ^* depends on η as a parameter and cannot be written explicitly.

The following lemmas are from [18].

Lemma 1.5.1. *For $p = 1, 2, 3$, problem (1.16) has a unique solution u_p of the form*

$$u_p(x) = x_p + C_p + \mathcal{S}_{D\sharp}(\lambda_\varepsilon Id - \mathcal{K}^*_{D\sharp})^{-1}[\nu_p](x) \quad \text{in } Y,$$

where C_p is a constant, ν_p is the p-component of the outward unit normal to ∂D, λ_ε is defined by (3.17), and

$$\begin{aligned}\mathcal{S}_{D\sharp}[\varphi](x) &= \int_{\partial D} G_\sharp(x,y)\varphi(y)d\sigma(y),\\ \mathcal{K}^*_{D\sharp}[\varphi](x) &= \int_{\partial D} \frac{\partial G_\sharp(x,y)}{\partial \nu(x)}\varphi(y)d\sigma(y)\end{aligned}$$

with $G_\sharp(x,y)$ being the periodic Green function defined by

$$G_\sharp(x,y) = -\sum_{n\in\mathbb{Z}^3\setminus\{0\}} \frac{e^{i2\pi n\cdot(x-y)}}{4\pi^2|n|^2}.$$

Lemma 1.5.2. *Let $\mathcal{S}_{D\sharp}$ and $\mathcal{K}^*_{D\sharp}$ be the operators defined as in Lemma 1.5.1. Then the following trace formula holds on ∂D*

$$(\pm\frac{1}{2}Id + \mathcal{K}^*_{D\sharp})[\varphi] = \frac{\partial \mathcal{S}_{D\sharp}[\varphi]}{\partial \nu}\Big|_\pm.$$

For the sake of simplicity, for $p = 1, 2, 3$, we set

$$\phi_p(y) = (\lambda_\varepsilon Id - \mathcal{K}^*_{D\sharp})^{-1}[\nu_p](y) \quad \text{for } y \text{ in } \partial D. \tag{1.18}$$

Thus, from Lemma 1.5.1, we get

$$\gamma^*_{pq} = \varepsilon_m \int_{\partial Y} \big(y_q + C_q + \mathcal{S}_{D\sharp}[\phi_q](y)\big)\frac{\partial\big(y_p + \mathcal{S}_{D\sharp}[\phi_p](y)\big)}{\partial \nu}d\sigma(y).$$

Because of the periodicity of $\mathcal{S}_{D\sharp}[\phi_p]$, we get

$$\gamma^*_{pq} = \varepsilon_m\Big(\delta_{pq} + \int_{\partial Y} y_q \frac{\partial \mathcal{S}_{D\sharp}[\phi_p]}{\partial \nu}(y)d\sigma(y)\Big). \tag{1.19}$$

In view of the periodicity of $\mathcal{S}_{D\sharp}[\phi_p]$, the divergence theorem applied on $Y\setminus\bar{D}$ and Lemma 1.5.2 yields (see [18])

$$\int_{\partial Y} y_q \frac{\partial \mathcal{S}_{D\sharp}[\phi_p]}{\partial \nu}(y) = \int_{\partial D} y_q\phi_p(y)d\sigma(y).$$

Let

$$\psi_p(y) = \phi_p(\delta y) \quad \text{for } y \in \partial B.$$

Then, by (1.19), we obtain

$$\gamma^* = \varepsilon_m(Id + fP), \tag{1.20}$$

where $f = |D| = \delta^3 (= \eta^{3\beta})$ is the volume fraction of D and $P = (P_{pq})_{p,q=1,2,3}$ is given by

$$P_{pq} = \int_{\partial B} y_q\psi_p(y)d\sigma(y). \tag{1.21}$$

To proceed with the computation of P we will need the following Lemma [18].

Lemma 1.5.3. *There exists a smooth function $R(x)$ in the unit cell Y such that*

$$G_\sharp(x,y) = -\frac{1}{4\pi|x-y|} + R(x-y).$$

Moreover, the Taylor expansion of $R(x)$ at 0 is given by

$$R(x) = R(0) - \frac{1}{6}(x_1^2 + x_2^2 + x_3^2) + O(|x|^4).$$

Now we can prove the main result of this section, which shows the validity of the Maxwell-Garnett theory uniformly with respect to the frequency under the assumptions that

$$f \ll \mathrm{dist}(\lambda_\varepsilon(\omega), \sigma(\mathcal{K}_B^*))^{3/5} \quad \text{and } (Id - \delta^3 R_{\lambda_\varepsilon(\omega)}^{-1} T_0)^{-1} = O(1), \tag{1.22}$$

where $R_{\lambda_\varepsilon(\omega)}^{-1}$ and T_0 are to be defined and $\mathrm{dist}(\lambda_\varepsilon(\omega), \sigma(\mathcal{K}_D^*))$ is the distance between $\lambda_\varepsilon(\omega)$ and the spectrum of $\mathcal{K}_B^*$.

Theorem 1.5.1. *Assume that (1.22) holds. Then we have*

$$\gamma^* = \varepsilon_m\Big(Id + fM(Id - \frac{f}{3}M)^{-1}\Big) + O\Big(\frac{f^{8/3}}{\mathrm{dist}(\lambda_\varepsilon(\omega), \sigma(\mathcal{K}_B^*))^2}\Big), \tag{1.23}$$

uniformly in ω. Here, $M = M(\lambda_\varepsilon(\omega), B)$ is the polarization tensor (3.36) associated with B and $\lambda_\varepsilon(\omega)$.

Proof. In view of Lemma 1.5.3 and (1.18), we can write, for $x \in \partial D$,

$$(\lambda_\varepsilon(\omega)Id - \mathcal{K}_D^*)[\phi_p](x) - \int_{\partial D} \frac{\partial R(x-y)}{\partial \nu(x)} \phi_p(y) d\sigma(y) = \nu_p(x),$$

which yields, for $x \in \partial B$,

$$(\lambda_\varepsilon(\omega)Id - \mathcal{K}_B^*)[\psi_p](x) - \delta^2 \int_{\partial B} \frac{\partial R(\delta(x-y))}{\partial \nu(x)} \psi_p(y) d\sigma(y) = \nu_p(x).$$

By virtue of Lemma 1.5.3, we get

$$\nabla R(\delta(x-y)) = -\frac{\delta}{3}(x-y) + O(\delta^3)$$

uniformly in $x, y \in \partial B$. Since $\int_{\partial B} \psi_p(y) d\sigma(y) = 0$, we now have

$$(R_{\lambda_\varepsilon(\omega)} - \delta^3 T_0 + \delta^5 T_1)[\psi_p](x) = \nu_p(x),$$

and so

$$(Id - \delta^3 R_{\lambda_\varepsilon(\omega)}^{-1} T_0 + \delta^5 R_{\lambda_\varepsilon(\omega)}^{-1} T_1)[\psi_p](x) = R_{\lambda_\varepsilon(\omega)}^{-1}[\nu_p](x), \tag{1.24}$$

where

$$\begin{aligned} R_{\lambda_\varepsilon(\omega)}[\psi_p](x) &= (\lambda_\varepsilon(\omega)Id - \mathcal{K}_B^*)[\psi_p](x), \\ T_0[\psi_p](x) &= \frac{\nu(x)}{3} \cdot \int_{\partial B} y \psi_p(y) d\sigma(y), \\ \|T_1\|_{\mathcal{L}(\mathcal{H}^*(\partial B))} &= O(1). \end{aligned}$$

Since $\mathcal{K}_B^*$ is a compact self-adjoint operator in $\mathcal{H}^*(\partial B)$ it follows that [50]

$$\|(\lambda_\varepsilon(\omega)Id - \mathcal{K}_B^*)^{-1}\|_{\mathcal{L}(\mathcal{H}^*(\partial B))} \leq \frac{c}{\mathrm{dist}(\lambda_\varepsilon(\omega), \sigma(\mathcal{K}_B^*))} \tag{1.25}$$

for a constant c.
It is clear that T_0 is a compact operator. From the fact that the imaginary part of $R_{\lambda_\varepsilon(\omega)}$ is nonzero, it follows that $Id - \delta^3 R^{-1}_{\lambda_\varepsilon(\omega)} T_0$ is invertible.
Under the assumption that

$$\begin{aligned}(Id - \delta^3 R^{-1}_{\lambda_\varepsilon(\omega)} T_0)^{-1} &= O(1),\\ \delta^5 &\ll \mathrm{dist}(\lambda_\varepsilon(\omega), \sigma(\mathcal{K}_B^*)),\end{aligned}$$

we get from (1.24) and (1.25)

$$\begin{aligned}\psi_p(x) &= (Id - \delta^3 R^{-1}_{\lambda_\varepsilon(\omega)} T_0 + \delta^5 R^{-1}_{\lambda_\varepsilon(\omega)} T_1)^{-1} R^{-1}_{\lambda_\varepsilon(\omega)}[\nu_p](x),\\ &= (Id - \delta^3 R^{-1}_{\lambda_\varepsilon(\omega)} T_0)^{-1} R^{-1}_{\lambda_\varepsilon(\omega)}[\nu_p](x) + O\Big(\frac{\delta^5}{\mathrm{dist}(\lambda_\varepsilon(\omega), \sigma(\mathcal{K}_B^*))}\Big).\end{aligned}$$

Therefore, we obtain the estimate for ψ_p

$$\psi_p = O\Big(\frac{1}{\mathrm{dist}(\lambda_\varepsilon(\omega), \sigma(\mathcal{K}_B^*))}\Big).$$

Now, we multiply (1.24) by y_q and integrate over ∂B. We can derive from the estimate of ψ_p that

$$P(Id - \frac{f}{3}M) = M + O\Big(\frac{\delta^5}{\mathrm{dist}(\lambda_\varepsilon(\omega), \sigma(\mathcal{K}_B^*))^2}\Big),$$

and therefore,

$$P = M(Id + \frac{f}{3}M)^{-1} + O\Big(\frac{\delta^5}{\mathrm{dist}(\lambda_\varepsilon(\omega), \sigma(\mathcal{K}_B^*))^2}\Big)$$

with P being defined by (1.21). Since $f = \delta^3$ and

$$M = O\Big(\frac{\delta^3}{\mathrm{dist}(\lambda_\varepsilon(\omega), \sigma(\mathcal{K}_B^*))}\Big),$$

it follows from (1.20) that the Maxwell-Garnett formula (1.23) holds (uniformly in the frequency ω) under the assumption (1.22) on the volume fraction f. □

Remark 1.5.1. *As a corollary of Theorem 1.5.1, we see that in the case when $fM = O(1)$, which is equivalent to the scale $f = O\Big(\mathrm{dist}(\lambda_\varepsilon(\omega), \sigma(\mathcal{K}_B^*))\Big)$, the matrix $fM(Id - \frac{f}{3}M)^{-1}$ may have a negative-definite symmetric real part. This implies that the effective medium is plasmonic as well as anisotropic.*

Remark 1.5.2. *It is worth emphasizing that Theorem 1.5.1 does not only prove the validity of the Maxwell-Garnett theory but it can also be used together with the results in section 1.4 in order to derive the plasmonic resonances of the effective medium made of a dilute system of arbitrary-shaped*

plasmonic nanoparticles, following (1.14)

$$\omega = \arg\max_{\omega} \|\mathcal{Q}_{\gamma^*}^{-1}(\omega)\|_{\mathcal{L}(\mathcal{H}^*(\partial\Omega))}.$$

1.6 Concluding remarks

In this chapter we have analyzed the plasmonic resonance phenomena assuming a quasi-static approximation, which is valid for particles considerably smaller than the wavelength of the incoming wave. We have presented a rigorous mathematical framework for its analysis, given beforehand the necessary mathematical tools, relying mainly in layer potential techniques. The plasmonic resonances depend strongly in the spectral properties of the Neumann-Poincaré operator $\mathcal{K}_D^*$ associated with D. We remark that this operator is scale invariant. This imply that the quasi-static model cannot explain changes in the resonances given by the scaling of nanoparticiles. This problem is analyzed in chapter 2 and 3 with the study of Helmholtz and Maxwell equations, respectively

We have also studied the anisotropic quasi-static problem in terms of layer potentials and defined the plasmonic resonances for anisotropic nanoparticles. Formulas for a small anisotropic perturbation of resonances of the isotropic formulas have been derived.

The Maxwell-Garnett theory provides a simple model for calculating the macroscopic optical properties of materials with a dilute inclusion of spherical nanoparticles [18]. In this chapter we have rigorously obtained effective properties of a periodic arrangement of arbitrary-shaped nanoparticles and derived a condition on the volume fraction of the nanoparticles that insures the validity of the Maxwell-Garnett theory for predicting the effective optical properties of systems of embedded in a dielectric host material at the plasmonic resonances.

Chapter 2

The Helmholtz Equation

Contents

2.1 Introduction

As seen in chapter 1, plasmon resonances in nanoparticles can be treated at the quasi-static limit as an eigenvalue problem for the Neumann-Poincaré integral operator. This leads to direct calculation of resonance values of permittivity and optimal design of nanoparticles that resonate at specified frequencies. At this limit, they are size-independent. However, as the particle size increases, they are determined from scattering and absorption blow up and become size-dependent. This was experimentally observed, for instance, in [49, 81, 89].

In this chapter, using the Helmholtz equation to model light propagation, we first prove that, as the particle size increases and crosses its critical value for dipolar approximation which is justified in [9], the plasmonic resonances become size-dependent. The resonance condition is determined from absorption and scattering blow up and depends on the shape, size and electromagnetic parameters of both the nanoparticle and the surrounding material. Then, we precisely quantify the scattering absorption enhancements in plasmonic nanoparticles. We derive new bounds on the enhancement factors given the volume and electromagnetic parameters of the nanoparticles. At the quasi-static limit, we prove that the averages over the orientation of scattering and extinction cross-sections of a randomly oriented nanoparticle are given in terms of the imaginary part of the polarization tensor. Moreover, we show that the polarization tensor blows up at plasmonic resonances and derive bounds for the absorption and scattering cross-sections. We also prove the blow-up of the first-order scattering coefficients at plasmonic resonances. The concept of scattering coefficients was introduced in [20] for scalar wave propagation problems and in [21] for the full Maxwell equations, rendering a powerful and efficient tool for the classification of the nanoparticle shapes. Using such a concept, we have explained in [6] the experimental results reported in [35].

The chapter is organized as follows. In section 2.3 we introduce a layer potential formulation for plasmonic resonances and derive asymptotic formulas for the plasmonic resonances and the near- and far-fields in terms of the size. In section 7.2 we consider the case of multiple plasmonic nanoparticles. Section 6.3 is devoted to the study of the scattering and absorption enhancements. The scattering coefficients are simply the Fourier coefficients of the scattering amplitude [20, 21]. In section 6.4 we investigate the behavior of the scattering coefficients at the plasmonic resonances.

2.2 Preliminaries

2.2.1 Layer potentials for the Helmholtz equation

Let G be the Green function for the Helmholtz operator $\Delta + k^2$ satisfying the Sommerfeld radiation condition.

The Sommerfeld radiation condition can be expressed in dimension $d = 2, 3$, as follows:

$$\left| \frac{\partial u}{\partial |x|} - ik_m u \right| \leq C|x|^{-(d+1)/2}$$

as $|x| \to +\infty$ for some constant C independent of x.

In $\mathbb{R}^3$, G is given by

$$G(x,y,k) = -\frac{e^{ik|x-y|}}{4\pi|x-y|}.$$

The single-layer potential and the Neumann-Poincaré integral operator for the Helmholtz equation are defined as follows

$$\begin{aligned}
\mathcal{S}_D^k[\varphi](x) &= \int_{\partial D} G(x,y,k)\varphi(y)d\sigma(y), \qquad x \in \mathbb{R}^3,\\
(\mathcal{K}_D^k)^*[\varphi](x) &= \int_{\partial D} \frac{\partial G(x,y,k)}{\partial \nu(x)}\varphi(y)d\sigma(y), \qquad x \in \partial D,
\end{aligned}$$

Let us recall some well known properties [12]:

(i) $\mathcal{S}_D^k : H^{-\frac{1}{2}}(\partial D) \to H^{\frac{1}{2}}(\partial D), H^1_{loc}(\mathbb{R}^2\backslash\partial D)$ is bounded;

(ii) $(\Delta + k^2)\mathcal{S}_D^k[\varphi](x) = 0$ for $x \in \mathbb{R}^2\backslash\partial D$, $\varphi \in H^{-\frac{1}{2}}(\partial D)$;

(iii) $(\mathcal{K}_D^k)^* : H^{-\frac{1}{2}}(\partial D) \to H^{-\frac{1}{2}}(\partial D)$ is compact;

(iv) $\mathcal{S}_D^k[\varphi]$, $\varphi \in H^{-\frac{1}{2}}(\partial D)$, satisfies the Sommerfeld radiation condition at infinity;

(v) $\dfrac{\partial \mathcal{S}_D^k[\varphi]}{\partial \nu}\Big|_{\pm} = (\pm\frac{1}{2}I + (\mathcal{K}_D^k)^*)[\varphi]$.

We have that, for any $\psi, \phi \in H^{-\frac{1}{2}}(\partial D)$,

$$u := \begin{cases} u^i + \mathcal{S}_D^{k_m}[\psi], & x \in \mathbb{R}^2\backslash\overline{D},\\ \mathcal{S}_D^{k_c}[\phi], & x \in D, \end{cases} \tag{2.1}$$

with $k_m = \omega\sqrt{\varepsilon_m\mu_m}$ and $k_c = \omega\sqrt{\varepsilon_c\mu_c}$, satisfies $\Delta u + k_m^2 u = 0$ in $\mathbb{R}^2\backslash\bar{D}$, $\Delta u + k_c^2 u = 0$ in D and $u - u^i$ satisfies the Sommerfeld radiation condition.

To satisfy the boundary transmission conditions, $\psi, \phi \in H^{-\frac{1}{2}}(\partial D)$ need to satisfy the following system of integral equations on ∂D

$$\begin{cases} \mathcal{S}_D^{k_m}[\psi] - \mathcal{S}_D^{k_c}[\phi] &= -u^i,\\ \frac{1}{\varepsilon_m}\big(\frac{1}{2}I + (\mathcal{K}_D^{k_m})^*\big)[\psi] + \frac{1}{\varepsilon_c}\big(\frac{1}{2}I - (\mathcal{K}_D^{k_c})^*\big)[\phi] &= -\dfrac{1}{\varepsilon_m}\dfrac{\partial u^i}{\partial \nu}. \end{cases} \tag{2.2}$$

The following result shows the existence of such a representation [19].

Theorem 2.2.1. *The operator*

$$\begin{aligned}
\mathcal{T} : \Big(H^{-\frac{1}{2}}(\partial D)\Big)^2 &\to H^{\frac{1}{2}}(\partial D) \times H^{-\frac{1}{2}}(\partial D)\\
(\psi,\phi) &\mapsto \left(\mathcal{S}_D^{k_m}[\psi] - \mathcal{S}_D^{k_c}[\phi],\ \frac{1}{\varepsilon_m}\big(\frac{1}{2}I + (\mathcal{K}_D^{k_m})^*\big)[\psi] + \frac{1}{\varepsilon_c}\big(\frac{1}{2}I - (\mathcal{K}_D^{k_c})^*\big)[\phi]\right)
\end{aligned}$$

is invertible.

2.3 Layer potential formulation for the scattering problem

2.3.1 Problem formulation and some basic results

We consider the scattering problem of a time-harmonic wave incident on a plasmonic nanoparticle. We use the Helmholtz equation instead of the full Maxwell equations. The homogeneous medium is characterized by its electric permittivity ε_m and its magnetic permeability μ_m, while the particle occupying a bounded and simply connected domain $D \Subset \mathbb{R}^3$ (the two-dimensional case can be treated similarly using results from Appendix B.3) of class $\mathcal{C}^{1,\alpha}$ for some $0 < \alpha < 1$ is characterized by electric permittivity ε_c and magnetic permeability μ_c, both of which may depend on the frequency. Assume that $\Re\mu_c < 0, \Im\mu_c > 0, \Im\varepsilon_c > 0$, and define

$$k_m = \omega\sqrt{\varepsilon_m\mu_m}, \quad k_c = \omega\sqrt{\varepsilon_c\mu_c},$$

and

$$\varepsilon_D = \varepsilon_m\chi(\mathbb{R}^3\backslash\bar{D}) + \varepsilon_c\chi(\bar{D}), \quad \mu_D = \varepsilon_m\chi(\mathbb{R}^3\backslash\bar{D}) + \varepsilon_c\chi(D),$$

where χ denotes the characteristic function. Let $u^i(x) = e^{ik_m d\cdot x}$ be the incident wave. Here, ω is the frequency and d is the unit incidence direction. Throughout this chapter, we assume that ε_m and μ_m are real and strictly positive and that $\Im k_c > 0$.

Using dimensionless quantities, we assume that the particle D has size of order one and also the following condition holds.

Condition 2.1. *We assume that the numbers $\varepsilon_m, \mu_m, \varepsilon_c, \mu_c$ are dimensionless and are of order one. In addition, $\Im\mu_c = o(1)$. We also assume that ω is dimensionless and is of order $o(1)$.*

It is worth emphasizing that in this section the variables ω refers to the ratio between the size of the particle and the incident wavelength. For real plasmonic nanoparticles made of noble metals such as silver and gold, their electric permittivity is only negative over a small range of frequencies in the optical regime. This is also the frequency range in which Condition 2.1 holds and also plasmonic resonance occurs. For the frequencies that are beyond that range, especially those near the origin, we shall assume that ε_c and μ_c are constant there. This assumption avoids complicated discussion on the dispersive property of electromagnetic parameters in that regime, and enables us to focus on the interesting frequency range when plasmonic resonance occurs. We also note that $\omega = o(1)$ implies that the plamsmonic nanoparticles have size much smaller than the incident wavelength. This is the case when plamsonic resonance occurs.

The scattering problem can be modeled by the following Helmholtz equation

$$\begin{cases} \nabla\cdot\dfrac{1}{\mu_D}\nabla u + \omega^2\varepsilon_D u = 0 \quad \text{in } \mathbb{R}^3\backslash\partial D, \\ u_+ - u_- = 0 \quad \text{on } \partial D, \\ \dfrac{1}{\mu_m}\dfrac{\partial u}{\partial\nu}\Big|_+ - \dfrac{1}{\mu_c}\dfrac{\partial u}{\partial\nu}\Big|_- = 0 \quad \text{on } \partial D, \\ u^s := u - u^i \text{ satisfies the Sommerfeld radiation condition.} \end{cases} \tag{2.3}$$

Here, $\partial/\partial\nu$ denotes the normal derivative and the Sommerfeld radiation condition.

The model problem (2.3) is referred to as the transverse magnetic case. Note that all the results of this chapter hold true in the transverse electric case where ε_D and μ_D are interchanged.

Let

$$\begin{aligned} F_1(x) &= -u^i(x) = -e^{ik_m d\cdot x}, \\ F_2(x) &= -\frac{1}{\mu_m}\frac{\partial u^i}{\partial \nu}(x) = -\frac{i}{\mu_m}k_m e^{ik_m d\cdot x} d\cdot\nu(x) \end{aligned}$$

with $\nu(x)$ being the outward normal at $x \in \partial D$.

By using the following single-layer potential and Neumann-Poincaré integral operator of section 2.2 we can represent the solution u in the following form

$$u(x) = \begin{cases} u^i + \mathcal{S}_D^{k_m}[\psi], & x \in \mathbb{R}^3\backslash\bar{D}, \\ \mathcal{S}_D^{k_c}[\phi], & x \in D, \end{cases} \tag{2.4}$$

where $\psi, \phi \in H^{-\frac{1}{2}}(\partial D)$ satisfy the following system of integral equations on ∂D [12]:

$$\begin{cases} \mathcal{S}_D^{k_m}[\psi] - \mathcal{S}_D^{k_c}[\phi] & = F_1, \\ \frac{1}{\mu_m}\big(\frac{1}{2}Id + (\mathcal{K}_D^{k_m})^*\big)[\psi] + \frac{1}{\mu_c}\big(\frac{1}{2}Id - (\mathcal{K}_D^{k_c})^*\big)[\phi] & = F_2, \end{cases} \tag{2.5}$$

where Id denotes the identity operator. In the sequel, we set $\mathcal{S}_D^0 = \mathcal{S}_D$.

We are interested in the scattering in the quasi-static regime, i.e., for $\omega \ll 1$. Note that for ω small enough, $\mathcal{S}_D^{k_c}$ is invertible [12]. We have $\phi = (\mathcal{S}_D^{k_c})^{-1}\big(\mathcal{S}_D^{k_m}[\psi] - F_1\big)$, whereas the following equation holds for ψ

$$\mathcal{A}_D(\omega)[\psi] = f, \tag{2.6}$$

where

$$\mathcal{A}_D(\omega) = \frac{1}{\mu_m}\big(\frac{1}{2}Id + (\mathcal{K}_D^{k_m})^*\big) + \frac{1}{\mu_c}\big(\frac{1}{2}Id - (\mathcal{K}_D^{k_c})^*\big)(\mathcal{S}_D^{k_c})^{-1}\mathcal{S}_D^{k_m}, \tag{2.7}$$

$$f = F_2 + \frac{1}{\mu_c}\big(\frac{1}{2}Id - (\mathcal{K}_D^{k_c})^*\big)(\mathcal{S}_D^{k_c})^{-1}[F_1]. \tag{2.8}$$

It is clear that

$$\mathcal{A}_D(0) = \mathcal{A}_{D,0} = \frac{1}{\mu_m}\big(\frac{1}{2}Id + \mathcal{K}_D^*\big) + \frac{1}{\mu_c}\big(\frac{1}{2}Id - \mathcal{K}_D^*\big) = \big(\frac{1}{2\mu_m} + \frac{1}{2\mu_c}\big)Id - \big(\frac{1}{\mu_c} - \frac{1}{\mu_m}\big)\mathcal{K}_D^*, \tag{2.9}$$

where the notation $\mathcal{K}_D^* = (\mathcal{K}_D^0)^*$ is used for simplicity.

We are interested in finding $\mathcal{A}_D(\omega)^{-1}$. We first recall some basic facts about the Neumann-Poincaré operator $\mathcal{K}_D^*$ stated in chaper 1. See also [12, 32, 61, 65].

Lemma 2.3.1. *(i) The following Calderón identity holds: $\mathcal{K}_D\mathcal{S}_D = \mathcal{S}_D\mathcal{K}_D^*$;*

(ii) The operator $\mathcal{K}_D^*$ is self-adjoint in the Hilbert space $H^{-\frac{1}{2}}(\partial D)$ equipped with the following inner product

$$(u, v)_{\mathcal{H}^*} = -(u, \mathcal{S}_D[v])_{-\frac{1}{2},\frac{1}{2}} \tag{2.10}$$

with $(\cdot,\cdot)_{-\frac{1}{2},\frac{1}{2}}$ *being the duality pairing between* $H^{-\frac{1}{2}}(\partial D)$ *and* $H^{\frac{1}{2}}(\partial D)$*, which is equivalent to the original one;*

(iii) Let $\mathcal{H}^*(\partial D)$ *be the space* $H^{-\frac{1}{2}}(\partial D)$ *with the new inner product. Let* (λ_j, φ_j)*,* $j = 0, 1, 2, \ldots$ *be the eigenvalue and normalized eigenfunction pair of* $\mathcal{K}_D^*$ *in* $\mathcal{H}^*(\partial D)$*, then* $\lambda_j \in (-\frac{1}{2}, \frac{1}{2}]$ *and* $\lambda_j \to 0$ *as* $j \to \infty$*;*

(iv) The following trace formula holds: for any $\psi \in \mathcal{H}^*(\partial D)$*,*

$$(-\frac{1}{2}Id + \mathcal{K}_D^*)[\psi] = \frac{\partial \mathcal{S}_D[\psi]}{\partial \nu}\Big|_-;$$

(v) The following representation formula holds: for any $\psi \in H^{-1/2}(\partial D)$*,*

$$\mathcal{K}_D^*[\psi] = \sum_{j=0}^{\infty} \lambda_j (\psi, \varphi_j)_{\mathcal{H}^*} \otimes \varphi_j.$$

It is clear that the following result holds.

Lemma 2.3.2. *Let* $\mathcal{H}(\partial D)$ *be the space* $H^{\frac{1}{2}}(\partial D)$ *equipped with the following equivalent inner product*

$$(u, v)_{\mathcal{H}} = ((-\mathcal{S}_D)^{-1}[u], v)_{-\frac{1}{2},\frac{1}{2}}. \tag{2.11}$$

Then, $\mathcal{S}_D$ *is an isometry between* $\mathcal{H}^*(\partial D)$ *and* $\mathcal{H}(\partial D)$*.*

We now present other useful observations and basic results. The following holds.

Lemma 2.3.3. *(i) We have* $(-\frac{1}{2}Id + \mathcal{K}_D^*)\mathcal{S}_D^{-1}[1] = 0$*.*

(ii) Let $\lambda_0 = \frac{1}{2}$*. Then the corresponding eigenspace has dimension one and is spanned by the function* $\varphi_0 = c\mathcal{S}_D^{-1}[1]$ *for some constant* c *such that* $||\varphi_0||_{\mathcal{H}^*} = 1$*.*

(iii) Moreover, $\mathcal{H}^*(\partial D) = \mathcal{H}_0^*(\partial D) \oplus \{\mu\varphi_0,\ \mu \in \mathbb{C}\}$*, where* $\mathcal{H}_0^*(\partial D)$ *is the zero mean subspace of* $\mathcal{H}^*(\partial D)$ *and* $\varphi_j \in \mathcal{H}_0^*(\partial D)$ *for* $j \geq 1$*, i.e.,* $(\varphi_j, 1)_{-\frac{1}{2},\frac{1}{2}} = 0$ *for* $j \geq 1$*. Here,* $\{\varphi_j\}_j$ *is the set of normalized eigenfunctions of* $\mathcal{K}_D^*$*.*

From (2.9), it is easy to see that

$$\mathcal{A}_{D,0}[\psi] = \sum_{j=0}^{\infty} \tau_j (\psi, \varphi_j)_{\mathcal{H}^*} \varphi_j, \tag{2.12}$$

where

$$\tau_j = \frac{1}{2\mu_m} + \frac{1}{2\mu_c} - \big(\frac{1}{\mu_c} - \frac{1}{\mu_m}\big)\lambda_j. \tag{2.13}$$

From (2.9), it is easy to see that

$$\mathcal{A}_{D,0}[\psi] = \sum_{j=0}^{\infty} \tau_j (\psi, \varphi_j)_{\mathcal{H}^*} \varphi_j, \tag{2.14}$$

where

$$\tau_j = \frac{1}{2\mu_m} + \frac{1}{2\mu_c} - \big(\frac{1}{\mu_c} - \frac{1}{\mu_m}\big)\lambda_j. \tag{2.15}$$

We now derive the asymptotic expansion of the operator $\mathcal{A}(\omega)$ as $\omega \to 0$. Using the asymptotic expansions in terms of k of the operators $\mathcal{S}_D^k$, $(\mathcal{S}_D^k)^{-1}$ and $(\mathcal{K}_D^k)^*$ proved in Appendix B.1, we can obtain the following result.

Lemma 2.3.4. *As $\omega \to 0$, the operator $\mathcal{A}_D(\omega) : \mathcal{H}^*(\partial D) \to \mathcal{H}^*(\partial D)$ admits the asymptotic expansion*

$$\mathcal{A}_D(\omega) = \mathcal{A}_{D,0} + \omega^2 \mathcal{A}_{D,2} + O(\omega^3),$$

where

$$\mathcal{A}_{D,2} = (\varepsilon_m - \varepsilon_c)\mathcal{K}_{D,2} + \frac{\varepsilon_m\mu_m - \varepsilon_c\mu_c}{\mu_c}\big(\frac{1}{2}Id - \mathcal{K}_D^*\big)\mathcal{S}_D^{-1}\mathcal{S}_{D,2}. \tag{2.16}$$

Proof. Recall that

$$\mathcal{A}_D(\omega) = \frac{1}{\mu_m}\big(\frac{1}{2}Id + (\mathcal{K}_D^{k_m})^*\big) + \frac{1}{\mu_c}\big(\frac{1}{2}Id - (\mathcal{K}_D^{k_c})^*\big)(\mathcal{S}_D^{k_c})^{-1}\mathcal{S}_D^{k_m}. \tag{2.17}$$

By a straightforward calculation, it follows that

$$\begin{aligned}
(\mathcal{S}_D^{k_c})^{-1}\mathcal{S}_D^{k_m} &= Id + \omega\big(\sqrt{\varepsilon_c\mu_c}\mathcal{B}_{D,1}\mathcal{S}_D + \sqrt{\varepsilon_m\mu_m}\mathcal{S}_D^{-1}\mathcal{S}_{D,1}\big) + \\
&\quad \omega^2\big(\varepsilon_c\mu_c\mathcal{B}_{D,2}\mathcal{S}_D + \sqrt{\varepsilon_c\mu_c\varepsilon_m\mu_m}\mathcal{B}_{D,1}\mathcal{S}_{D,1} + \varepsilon_m\mu_m\mathcal{S}_D^{-1}\mathcal{S}_{D,2}\big) + O(\omega^3), \\
&= Id + \omega\big(\sqrt{\varepsilon_m\mu_m} - \sqrt{\varepsilon_c\mu_c}\big)\mathcal{S}_D^{-1}\mathcal{S}_{D,1} + \\
&\quad \omega^2\big((\varepsilon_m\mu_m - \varepsilon_c\mu_c)\mathcal{S}_D^{-1}\mathcal{S}_{D,2} + \sqrt{\varepsilon_c\mu_c}(\sqrt{\varepsilon_c\mu_c} - \sqrt{\varepsilon_m\mu_m})\mathcal{S}_D^{-1}\mathcal{S}_{D,1}\mathcal{S}_D^{-1}\mathcal{S}_{D,1}\big) \\
&\quad + O(\omega^3),
\end{aligned}$$

where $\mathcal{B}_{D,1}$ and $\mathcal{B}_{D,2}$ are defined by (B.5). Using the facts that

$$\big(\frac{1}{2}Id - \mathcal{K}_D^*\big)\mathcal{S}_D^{-1}\mathcal{S}_{D,1} = 0$$

and

$$\frac{1}{2}Id - (\mathcal{K}_D^k)^* = \big(\frac{1}{2}Id - \mathcal{K}_D^*\big) - k^2\mathcal{K}_{D,2} + O(k^3),$$

the lemma immediately follows. □

We regard $\mathcal{A}_D(\omega)$ as a perturbation to the operator $\mathcal{A}_{D,0}$ for small ω. Using standard perturbation theory [85], we can derive the perturbed eigenvalues and their associated eigenfunctions. For simplicity, we consider the case when λ_j is a simple eigenvalue of the operator $\mathcal{K}_D^*$.

We let

$$R_{jl} = \big(\mathcal{A}_{D,2}[\varphi_j], \varphi_l\big)_{\mathcal{H}^*}, \tag{2.18}$$

where $\mathcal{A}_{D,2}$ is defined by (2.16).

As ω goes to zero, the perturbed eigenvalue and eigenfunction have the following form:

$$\tau_j(\omega) = \tau_j + \omega^2\tau_{j,2} + O(\omega^3), \tag{2.19}$$

$$\varphi_j(\omega) = \varphi_j + \omega^2\varphi_{j,2} + O(\omega^3), \tag{2.20}$$

where

$$\tau_{j,2} = R_{jj}, \tag{2.21}$$

$$\varphi_{j,2} = \sum_{l\neq j} \frac{R_{jl}}{(\frac{1}{\mu_m}-\frac{1}{\mu_c})(\lambda_j-\lambda_l)}\varphi_l. \tag{2.22}$$

2.3.2 First-order correction to plasmonic resonances and field behavior at the plasmonic resonances

We first introduce different notions of plasmonic resonance as follows.

Definition 2.1. *(i) We say that ω is a plasmonic resonance if*

$$|\tau_j(\omega)| \ll 1 \quad \text{and is locally minimal for some } j.$$

(ii) We say that ω is a quasi-static plasmonic resonance if $|\tau_j| \ll 1$ and is locally minimized for some j. Here, τ_j is defined by (2.15).

(iii) We say that ω is a first-order corrected quasi-static plasmonic resonance if $|\tau_j + \omega^2\tau_{j,2}| \ll 1$ and is locally minimized for some j. Here, the correction term $\tau_{j,2}$ is defined by (2.21).

Note that quasi-static resonances are size independent and is therefore a zero-order approximation of the plasmonic resonance in terms of the particle size while the first-order corrected quasi-static plasmonic resonance depends on the size of the nanoparticle (or equivalently on ω in view of the non-dimensionalization adopted herein).

We are interested in solving the equation $\mathcal{A}_D(\omega)[\phi] = f$ when ω is close to the resonance frequencies, i.e., when $\tau_j(\omega)$ is very small for some j's. In this case, the major part of the solution would be the contributions of the excited resonance modes $\varphi_j(\omega)$. We introduce the following definition.

Definition 2.2. *We call $J \subset \mathbb{N}$ index set of resonance if τ_j's are close to zero when $j \in J$ and are bounded from below when $j \in J^c$. More precisely, we choose a threshold number $\eta_0 > 0$ independent of ω such that*

$$|\tau_j| \geq \eta_0 > 0 \quad \text{for } j \in J^c.$$

Remark 2.3.1. *Note that for $j = 0$, we have $\tau_0 = 1/\mu_m$, which is of size one by our assumption. As a result, throughout this chapter, we always exclude 0 from the index set of resonance J.*

From now on, we shall use J as our index set of resonances. We assume throughout that the following conditions hold.

Condition 2.2. *Each eigenvalue λ_j for $j \in J$ is a simple eigenvalue of the operator $\mathcal{K}_D^*$.*

Condition 2.3. *Let*

$$\lambda = \frac{\mu_m + \mu_c}{2(\mu_m - \mu_c)}. \tag{2.23}$$

We assume that $\lambda \neq 0$ or equivalently, $\mu_c \neq -\mu_m$.

Condition 2.3, which is crucial to our analysis, implies that the set J is finite. Otherwise, infinity resonance modes may be excited and the problem becomes unstable. We refer to [44, 47, 79] for detailed discussion on this case.

Remark 2.3.2. *Note that in the ideal case when $\Im\mu_c = 0$, we know that $\tau_j = 0$ if λ defined in (2.23) is equal to λ_j. This the usual definition in the quasi-static limiting case when the wavelength is infinite. In the case $\Im\mu_c \neq 0$ but $\Im\mu_c = o(1)$, one may neglect the imaginary part and still use the definition to find the resonance frequency. The draw back of this definition is that the resonance frequency is independent of the size of the particle. Now, with the asymptotic expansion (8.11), we may find ω, the resonance frequency, according to the criterion in Definition 3.3 (i) in a small neighborhood of the resonant frequency of the quasi-static limiting case. The difference of the two frequency yields the shift of resonance frequency with respect to size of the particle.*

We now define the projection $P_J(\omega)$ such that

$$P_J(\omega)[\varphi_j(\omega)] = \begin{cases} \varphi_j(\omega), & j \in J, \\ 0, & j \in J^c. \end{cases}$$

In fact, we have

$$P_J(\omega) = \sum_{j\in J} P_j(\omega) = \sum_{j\in J} \frac{1}{2\pi i} \int_{\gamma_j} (\xi - \mathcal{A}_D(\omega))^{-1} d\xi, \tag{2.24}$$

where γ_j is a Jordan curve in the complex plane enclosing only the eigenvalue $\tau_j(\omega)$ among all the eigenvalues.

To obtain an explicit representation of $P_J(\omega)$, we consider the adjoint operator $\mathcal{A}_D(\omega)^*$. By a similar perturbation argument, we can obtain its perturbed eigenvalue and eigenfunction, which have the following form

$$\widetilde{\tau}_j(\omega) = \overline{\tau_j(\omega)}, \tag{2.25}$$

$$\widetilde{\varphi_j}(\omega) = \varphi_j + \omega^2 \widetilde{\varphi}_{j,2} + o(\omega^2). \tag{2.26}$$

Using the eigenfunctions $\widetilde{\varphi}_j(\omega)$, we can show that

$$P_J(\omega)[x] = \sum_{j\in J} \big(x, \widetilde{\varphi}_j(\omega)\big)_{\mathcal{H}^*} \varphi_j(\omega). \tag{2.27}$$

Throughout this chapter, for two Banach spaces X and Y, by $\mathcal{L}(X, Y)$ we denote the set of bounded linear operators from X into Y.

We are now ready to solve the equation $\mathcal{A}_D(\omega)[\psi] = f$. First, it is clear that

$$\psi = \mathcal{A}_D(\omega)^{-1}[f] = \sum_{j\in J} \frac{\big(f, \widetilde{\varphi_j}(\omega)\big)_{\mathcal{H}^*}}{\tau_j(\omega)} + \mathcal{A}_D(\omega)^{-1}[P_{J^c}(\omega)[f]]. \tag{2.28}$$

The following lemma holds.

Lemma 2.3.5. *The norm $\|\mathcal{A}_D(\omega)^{-1} P_{J^c}(\omega)\|_{\mathcal{L}(\mathcal{H}^*(\partial D), \mathcal{H}^*(\partial D))}$ is uniformly bounded in ω for ω sufficiently small.*

Proof. Consider the operator

$$\mathcal{A}_D(\omega)|_{J^c} : P_{J^c}(\omega)\mathcal{H}^*(\partial D) \to P_{J^c}(\omega)\mathcal{H}^*(\partial D).$$

For ω small enough, we can show that $\mathrm{dist}(\sigma(\mathcal{A}_D(\omega)|_{J^c}), 0) \geq \frac{\eta_0}{2}$, where $\sigma(\mathcal{A}_D(\omega)|_{J^c})$ is the discrete spectrum of $\mathcal{A}_D(\omega)|_{J^c}$. Then, it follows that

$$\|\mathcal{A}_D(\omega)^{-1}(P_{J^c}(\omega)f)\| = \|\big(\mathcal{A}_D(\omega)|_{P_{J^c}}\big)^{-1}(P_{J^c}(\omega)f)\| \lesssim \frac{1}{\eta_0}\exp(\frac{C_1}{\eta_0^2})\|P_{J^c}(\omega)f\|,$$

where the notation $A \lesssim B$ means that $A \leq CB$ for some constant C.

On the other hand,

$$\begin{aligned} P_J(\omega)f &= \sum_{j\in J}\big(f, \widetilde{\varphi_j}(\omega)\big)_{\mathcal{H}^*}\varphi_j(\omega) = \sum_{j\in J}\big(f, \varphi_j + O(\omega)\big)_{\mathcal{H}^*}\big(\varphi_j + O(\omega)\big) \\ &= \sum_{j\in J}\big(f, \varphi_j\big)_{\mathcal{H}^*}\varphi_j(\omega) + O(\omega). \end{aligned}$$

Thus,

$$\|P_{J^c}(\omega)\| = \|(Id - P_J(\omega))\| \lesssim (1 + O(\omega)),$$

from which the desired result follows immediately. □

Second, we have the following asymptotic expansion of f given by (2.8) with respect to ω.

Lemma 2.3.6. *Let*

$$f_1 = -i\sqrt{\varepsilon_m\mu_m}e^{ik_m d\cdot z}\left(\frac{1}{\mu_m}[d\cdot\nu(x)] + \frac{1}{\mu_c}(\frac{1}{2}Id - \mathcal{K}_D^*)\mathcal{S}_D^{-1}[d\cdot(x-z)]\right)$$

and let z be the center of the domain D. In the space $\mathcal{H}^*(\partial D)$, as ω goes to zero, we have

$$f = \omega f_1 + O(\omega^2),$$

in the sense that, for ω small enough,

$$\|f - \omega f_1\|_{\mathcal{H}^*} \leq C\omega^2$$

for some constant C independent of ω.

Proof. A direct calculation yields

$$\begin{aligned} f &= F_2 + \frac{1}{\mu_c}(\frac{1}{2}Id - (\mathcal{K}_D^{k_c})^*)(\mathcal{S}_D^{k_c})^{-1}[F_1] \\ &= -\omega\frac{i}{\mu_m}\sqrt{\varepsilon_m\mu_m}e^{ik_m d\cdot z}[d\cdot\nu(x)] + O(\omega^2) + \\ &\quad \frac{1}{\mu_c}\Big((\frac{1}{2}Id - \mathcal{K}_D^*)\big((\mathcal{S}_D)^{-1} + \omega\mathcal{B}_{D,1}\big) + O(\omega^2)\Big)[-e^{ik_m d\cdot z}\big(1 + i\omega\sqrt{\varepsilon_m\mu_m}[d\cdot(x-z)]\big) \\ &\quad + O(\omega^2)] \\ &= -\frac{e^{ik_m d\cdot z}}{\mu_c}(\frac{1}{2}Id - \mathcal{K}_D^*)\mathcal{S}_D^{-1}[1] - \frac{\omega e^{ik_m d\cdot z}}{\mu_c}(\frac{1}{2}Id - \mathcal{K}_D^*)\mathcal{B}_{D,1}[1] - \\ &\quad \omega i\sqrt{\varepsilon_m\mu_m}e^{ik_m d\cdot z}\left(\frac{1}{\mu_m}[d\cdot\nu(x)] + \frac{1}{\mu_c}(\frac{1}{2}Id - \mathcal{K}_D^*)\mathcal{S}_D^{-1}[d\cdot(x-z)]\right) + O(\omega^2) \\ &= -\omega i\sqrt{\varepsilon_m\mu_m}e^{ik_m d\cdot z}\left(\frac{1}{\mu_m}[d\cdot\nu(x)] + \frac{1}{\mu_c}(\frac{1}{2}Id - \mathcal{K}_D^*)\mathcal{S}_D^{-1}[d\cdot(x-z)]\right) \\ &\quad + O(\omega^2), \end{aligned}$$

where we have made use of the facts that

$$(\frac{1}{2}Id - \mathcal{K}_D^*)\mathcal{S}_D^{-1}[1] = 0$$

and

$$\mathcal{B}_{D,1}[\chi(\partial D)] = c\mathcal{S}_D^{-1}[\chi(\partial D)]$$

for some constant c; see again Appendix B.1. □

Finally, we are ready to state our main result in this section.

Theorem 2.3.1. *Let D has size of order one. Under Conditions 2.1, 2.2, and 2.3 the scattered field $u^s = u - u^i$ due to a single plasmonic particle D has the following representation:*

$$u^s = \mathcal{S}_D^{k_m}[\psi],$$

where

$$\begin{aligned}\psi &= \sum_{j\in J} \frac{\omega\big(f_1, \widetilde{\varphi}_j(\omega)\big)_{\mathcal{H}^*}\varphi_j(\omega)}{\tau_j(\omega)} + O(\omega),\\ &= \sum_{j\in J} \frac{ik_m e^{ik_m d\cdot z}\big(d\cdot\nu(x), \varphi_j\big)_{\mathcal{H}^*}\varphi_j + O(\omega^2)}{\lambda - \lambda_j + O(\omega^2)} + O(\omega)\end{aligned}$$

with λ being given by (2.23).

Proof. We have

$$\begin{aligned}\psi &= \sum_{j\in J} \frac{\big(f, \widetilde{\varphi}_j(\omega)\big)_{\mathcal{H}^*}\varphi_j(\omega)}{\tau_j(\omega)} + \mathcal{A}_D(\omega)^{-1}(P_{J^c}(\omega)f),\\ &= \sum_{j\in J} \frac{\omega\big(f_1, \varphi_j\big)_{\mathcal{H}^*}\varphi_j + O(\omega^2)}{\frac{1}{2\mu_m} + \frac{1}{2\mu_c} - (\frac{1}{\mu_c} - \frac{1}{\mu_m})\lambda_j + O(\omega^2)} + O(\omega).\end{aligned}$$

We now compute $\big(f_1, \varphi_j\big)_{\mathcal{H}^*}$ with f_1 given in Lemma 2.3.6. We only need to show that

$$\Big((\frac{1}{2}Id - \mathcal{K}_D^*)\mathcal{S}_D^{-1}[d\cdot(x-z)]), \varphi_j\Big)_{\mathcal{H}^*} = (d\cdot\nu(x), \varphi_j)_{\mathcal{H}^*}. \tag{2.29}$$

Indeed, we have

$$\begin{aligned}
\Big((\frac{1}{2}Id-\mathcal{K}_D^*)\mathcal{S}_D^{-1}[d\cdot(x-z)],\varphi_j\Big)_{\mathcal{H}^*} &= -\Big(\mathcal{S}_D^{-1}[d\cdot(x-z)],(\frac{1}{2}Id-\mathcal{K}_D)\mathcal{S}_D[\varphi_j]\Big)_{-\frac{1}{2},\frac{1}{2}}\\
&= -\Big(\mathcal{S}_D^{-1}[d\cdot(x-z)],\mathcal{S}_D(\frac{1}{2}Id-\mathcal{K}_D^*)[\varphi_j]\Big)_{-\frac{1}{2},\frac{1}{2}}\\
&= -\Big(d\cdot(x-z),(\frac{1}{2}Id-\mathcal{K}_D^*)[\varphi_j]\Big)_{-\frac{1}{2},\frac{1}{2}}\\
&= -\Big(d\cdot(x-z),-\frac{\partial\mathcal{S}_D[\varphi_j]}{\partial\nu}\Big|_-\Big)_{-\frac{1}{2},\frac{1}{2}}\\
&= \int_{\partial D}\frac{\partial[d\cdot(x-z)]}{\partial\nu}\mathcal{S}_D[\varphi_j]d\sigma\\
&\quad -\int_D\Big(\Delta[d\cdot(x-z)]\mathcal{S}_D[\varphi_j]-\Delta\mathcal{S}_D[\varphi_j][d\cdot(x-z)]\Big)dx\\
&= -\Big(d\cdot\nu(x),\varphi_j\Big)_{\mathcal{H}^*},
\end{aligned}$$

where we have used the fact that $\mathcal{S}_D[\varphi_j]$ is harmonic in D. This proves the desired identity and the rest of the theorem follows immediately. □

Corollary 2.3.1. *Assume the same conditions as in Theorem 3.3.2. Under the additional condition that*

$$\min_{j\in J}|\tau_j(\omega)|\gg\omega^3, \tag{2.30}$$

we have

$$\psi=\sum_{j\in J}\frac{ik_me^{ik_md\cdot z}\big(d\cdot\nu(x),\varphi_j\big)_{\mathcal{H}^*}\varphi_j+O(\omega^2)}{\lambda-\lambda_j+\omega^2\big(\frac{1}{\mu_c}-\frac{1}{\mu_m}\big)^{-1}\tau_{j,2}}+O(\omega).$$

More generally, under the additional condition that

$$\min_{j\in J}\tau_j(\omega)\gg\omega^{m+1},$$

for some integer $m>2$, we have

$$\psi=\sum_{j\in J}\frac{ik_me^{ik_md\cdot z}\big(d\cdot\nu(x),\varphi_j\big)_{\mathcal{H}^*}\varphi_j+O(\omega^2)}{\lambda-\lambda_j+\omega^2\big(\frac{1}{\mu_c}-\frac{1}{\mu_m}\big)^{-1}\tau_{j,2}+\cdots+\omega^m\big(\frac{1}{\mu_c}-\frac{1}{\mu_m}\big)^{-1}\tau_{j,m}}+O(\omega).$$

Re-scaling back to original dimensional variables, we suppose that the magnetic permeability μ_c of the nanoparticle is changing with respect to the operating angular frequency ω while that of the surrounding medium, μ_m, is independent of ω. Then we can write

$$\mu_c(\omega)=\mu'(\omega)+i\mu''(\omega). \tag{2.31}$$

Because of causality, the real and imaginary parts of μ_c obey the following Kramer–Kronig relations:

$$\begin{aligned}\mu''(\omega) &= -\frac{1}{\pi}\text{p.v.}\int_{-\infty}^{+\infty}\frac{1}{\omega-s}\mu'(s)ds,\\ \mu'(\omega) &= \frac{1}{\pi}\text{p.v.}\int_{-\infty}^{+\infty}\frac{1}{\omega-s}\mu''(s)ds,\end{aligned} \tag{2.32}$$

where p.v. stands for the principle value.

The magnetic permeability $\mu_c(\omega)$ can be described by the Drude model; see, for instance, [86]. We have

$$\mu_c(\omega) = \mu_0(1 - F\frac{\omega^2}{\omega^2 - \omega_0^2 + i\tau^{-1}\omega}), \tag{2.33}$$

where $\tau > 0$ is the nanoparticle's bulk electron relaxation rate (τ^{-1} is the damping coefficient), F is a filling factor, and ω_0 is a localized plasmon resonant frequency. When

$$(1-F)(\omega^2-\omega_0^2)^2 - F\omega_0^2(\omega^2-\omega_0^2) + \tau^{-2}\omega^2 < 0,$$

the real part of $\mu_c(\omega)$ is negative.

We suppose that $D = z + \delta B$. The quasi-static plasmonic resonance is defined by ω such that

$$\Re\frac{\mu_m + \mu_c(\omega)}{2(\mu_m - \mu_c(\omega))} = \lambda_j$$

for some j, where λ_j is an eigenvalue of the Neumann-Poincaré operator $\mathcal{K}_D^*(= \mathcal{K}_B^*)$. It is clear that such definition is independent of the nanoparticle's size. In view of (8.11), the shifted plasmonic resonance is defined by

$$\text{argmin}\left|\frac{1}{2\mu_m} + \frac{1}{2\mu_c(\omega)} - \big(\frac{1}{\mu_c(\omega)} - \frac{1}{\mu_m}\big)\lambda_j + \omega^2\delta^2\tau_{j,2}\right|,$$

where $\tau_{j,2}$ is given by (2.21) with D replaced by B.

2.4 Scattering and absorption enhancements

In this section we analyze the scattering and absorption enhancements. We prove that, at the quasi-static limit, the averages over the orientation of scattering and extinction cross-sections of a randomly oriented nanoparticle are given by (2.43) and (2.44), where M given by (2.40) is the polarization tensor associated with the nanoparticle D and the magnetic contrast $\mu_c(\omega)/\mu_m$. In view of (2.48), the polarization tensor M blows up at the plasmonic resonances, which yields scattering and absorption enhancements. A bound on the extinction cross-section is derived in (2.50). As shown in (2.53) and (2.55), it can be sharpened for nanoparticles of elliptical or ellipsoidal shapes.

2.4.1 Far-field expansion

For simplicity, we assume throughout this section that D contains the origin. We first prove the following representation for the scattering amplitude.

Propsition 2.4.1. *Let $u^i = e^{ik_m d\cdot x}$ with d being a unit vector. Let $x \in \mathbb{R}^3$ be such that $|x| \gg 1/\omega$. Then, we have*

$$u^s(x) = \frac{e^{ik_m|x|}}{|x|} A_\infty\left(\frac{x}{|x|}, d\right) + O\left(\frac{1}{|x|^2}\right) \tag{2.34}$$

with

$$A_\infty\left(\frac{x}{|x|}, d\right) = -\frac{1}{4\pi}\int_{\partial D} e^{-ik_m \frac{x}{|x|}\cdot y}\psi(y)d\sigma(y) \tag{2.35}$$

being the scattering amplitude and ψ being defined by (2.5).

Proof. We recall that the scattered field u^s can be represented as follows:

$$\begin{aligned} u^s(x) &= \mathcal{S}_D^{k_m}[\psi](x) \\ &= -\frac{1}{4\pi}\int_{\partial D}\frac{e^{ik_m|x-y|}}{|x-y|}\psi(y)d\sigma(y). \end{aligned}$$

From

$$|x-y| = |x|\left(1 - \frac{x\cdot y}{|x|^2} + O(\frac{1}{|x|^2})\right),$$

it follows that

$$u^s(x) = -\frac{e^{ik_m|x|}}{4\pi|x|}\int_{\partial D} e^{-ik_m\frac{x}{|x|}\cdot y}\psi(y)\left(1 + \frac{(x\cdot y)}{|x|^2}\right)d\sigma(y) + o\left(\frac{1}{|x|^2}\right),$$

which yields the desired result. □

2.4.2 Energy flow

The following definitions are from [43]. We include them here for the sake of completeness. The analogous quantity of the Poynting vector in scalar wave theory is the *energy flux* vector; see [43]. We recall that for a real monochromatic field

$$U(x,t) = \Re\left[u(x)e^{-i\omega t}\right],$$

the averaged value of the energy flux vector, taken over an interval which is long compared to the period of the oscillations, is given by

$$F(x) = -iC\left[\overline{u}(x)\nabla u(x) - u(x)\nabla\overline{u}(x)\right],$$

where C is a positive constant depending on the polarization mode. In the transverse electric case, $C = \omega/\mu_m$ while in the transverse magnetic case $C = \omega/\varepsilon_m$. Assume that the particle is contained in the ball B_R of radius R and center the origin. We now consider the outward flow of energy through the sphere ∂B_R:

$$\mathcal{W} = \int_{\partial B_R} F(x)\cdot\nu(x)d\sigma(x),$$

where $\nu(x)$ is the outward normal at $x \in \partial B_R$.

As the total field can be written as $U = u^s + u^i$, the flow can be decomposed into three parts:

$$\mathcal{W} = \mathcal{W}^i + \mathcal{W}^s + \mathcal{W}',$$

where

$$\begin{aligned}
\mathcal{W}^i &= -iC \int_{\partial B_R} \left[\overline{u^i}(x)\nabla u^i(x) - u^i(x)\nabla\overline{u^i}(x)\right] \cdot \nu(x)\, d\sigma(x),\\
\mathcal{W}^s &= -iC \int_{\partial B_R} [\overline{u^s}(x)\nabla u^s(x) - u^s(x)\nabla\overline{u^s}(x)] \cdot \nu(x)\, d\sigma(x),\\
\mathcal{W}' &= -iC \int_{\partial B_R} \left[\overline{u^i}(x)\nabla u^s(x) - u^s(x)\nabla\overline{u^i}(x) - u^i(x)\nabla\overline{u^s}(x) + \overline{u^s}(x)\nabla u^i(x)\right] \cdot \nu(x)\, d\sigma(x).
\end{aligned}$$

It is straightforward to check that $\mathcal{W}$, $\mathcal{W}^i$, $\mathcal{W}^a$, and $\mathcal{W}'$ in the above definitions are independent of the radius R as long as the particle is contained in B_R. In the case where u^i is a plane wave, we can see that $\mathcal{W}^i = 0$:

$$\begin{aligned}
\mathcal{W}^i &= -iC \int_{\partial B_R} \left[\overline{u^i}(x)\nabla u^i(x) - u^i(x)\nabla\overline{u^i}(x)\right]\, d\sigma(x),\\
&= -iC \int_{\partial B_R} \left[e^{-ik_m d\cdot x} ik_m d e^{ik_m d\cdot x} + e^{ik_m d\cdot x} k_m d e^{-ik_m d\cdot x}\right] \cdot \nu(x)\, d\sigma(x),\\
&= 2Ck_m d \cdot \int_{\partial B_R} \nu(x)\, d\sigma(x),\\
&= 0.
\end{aligned}$$

In a non absorbing medium with non absorbing scatterer, $\mathcal{W}$ is equal to zero because the electromagnetic energy would be conserved by the scattering process. However, if the scatterer is an absorbing body, the conservation of energy gives the rate of absorption as

$$\mathcal{W}^a = -\mathcal{W}.$$

Therefore, we have

$$\mathcal{W}^a + \mathcal{W}^s = -\mathcal{W}'.$$

Here, $\mathcal{W}'$ is called the extinction rate. It is the rate at which the energy is removed by the scatterer from the illuminating plane wave, and it is the sum of the rate of absorption and the rate at which energy is scattered.

2.4.3 Extinction, absorption, and scattering cross-sections and the optical theorem

Denote by U^i the quantity $U^i(x) = \left|\overline{u^i}(x)\nabla u^i(x) - u^i(x)\nabla\overline{u^i}(x)\right|$. In the case of a plane wave illumination, $U^i(x)$ is independent of x and is given by $U^i = 2k_m$.

Definition 2.3. *The scattering cross-section Q^s, the absorption cross-section Q^a and the extinction cross-section are defined by*

$$Q^s = \frac{\mathcal{W}^s}{U^i}, \quad Q^a = \frac{\mathcal{W}^a}{U^i}, \quad Q^{ext} = \frac{-\mathcal{W}'}{U^i}.$$

Note that these quantities are independent of x for a plane wave illumination.

Theorem 2.4.1 (Optical theorem)**.** *If $u^i(x) = e^{ik_m d\cdot x}$, where d is a unit direction, then*

$$Q^{ext} = Q^s + Q^a = \frac{4\pi}{k_m}\Im\left[A_\infty(d,d)\right], \tag{2.36}$$

$$Q^s = \int_{\mathbb{S}^2} |A_\infty(\hat{x},d)|^2 d\sigma(\hat{x}) \tag{2.37}$$

with A_∞ being the scattering amplitude defined by (2.35).

Proof. The Sommerfeld radiation condition gives, for any $x \in \partial B_R$,

$$\nabla u^s(x)\cdot\nu(x) \sim ik_m u^s(x). \tag{2.38}$$

Hence, from (2.34) we get

$$u^s(x)\nabla\overline{u^s}(x)\cdot\nu(x) - \overline{u^s}(x)\nabla u^s(x)\cdot\nu(x) \sim -\frac{2ik_m}{|x|^2}\left|A_\infty\left(\frac{x}{|x|},d\right)\right|^2,$$

which yields (2.37). We now compute the extinction rate. We have

$$\nabla u^i(x)\cdot\nu(x) = ik_m d\cdot\nu(x)e^{ik_m d\cdot x}. \tag{2.39}$$

Therefore, using 2.38 and 2.39, it follows that

$$\begin{aligned}\overline{u^i}(x)\nabla u^s(x)\cdot\nu(x) - u^s(x)\nabla\overline{u^i}(x)\cdot\nu(x) &\sim \left(ik_m\frac{e^{ik_m(|x|-d\cdot x)}}{|x|}d\cdot\nu + ik_m\frac{e^{ik_m(|x|-d\cdot x)}}{|x|}\right)A_\infty\left(\frac{x}{|x|},d\right)\\ &= \frac{ik_m e^{ik_m|x|-d\cdot\nu(x)}}{|x|}\left(d\cdot\nu(x)+1\right)A_\infty\left(\frac{x}{|x|},d\right).\end{aligned}$$

For $x\in\partial B_R$, we can write

$$\overline{u^i}(x)\nabla u^s(x)\cdot\nu(x) - u^s(x)\nabla\overline{u^i}(x)\cdot\nu(x) \sim \frac{ik_m e^{-ik_m R\nu(x)\cdot(d-\nu(x))}}{R}\left(d\cdot\nu(x)+1\right)A_\infty\left(\frac{x}{|x|},d\right).$$

We now use Jones' lemma (see, for instance, [43, Chapter 13.3]) to write the following asymptotic expansion as $R\to\infty$

$$\frac{1}{R}\int_{\partial B_R}\mathcal{G}(\nu(x))e^{-ik_m d\cdot\nu(x)}d\sigma(x) \sim \frac{2\pi i}{k_m}\left(\mathcal{G}(d)e^{-ik_m R} - \mathcal{G}(-d)e^{ik_m R}\right),$$

to obtain

$$\int_{\partial B_R}\left[\overline{u^i}(x)\nabla u^s(x) - u^s(x)\nabla\overline{u^i}(x)\right]\cdot\nu(x) \sim -4\pi A_\infty(d,d) \quad \text{as } R\to\infty.$$

Therefore,

$$\mathcal{W}' = -i4\pi C\left[A_\infty(d) - \overline{A_\infty}(d)\right] = 8\pi C\Im\left[A_\infty(d)\right].$$

Since

$$\left|\overline{u^i}(x)\nabla u^i(x) - u^i(x)\nabla\overline{u^i}(x)\right| = 2k_m,$$

we get the result. □

2.4.4 The quasi-static limit

We start by recalling the small volume expansion for the far-field. Let λ be defined by (2.23) and let

$$M(\lambda, D) := \int_{\partial D} (\lambda Id - \mathcal{K}_D^*)^{-1}[\nu]x\, d\sigma(x) \tag{2.40}$$

be the polarization tensor. The following asymptotic expansion holds. It can be proved by exactly the same arguments as those in [9].

Propsition 2.4.2. *Assume that $D = \delta B + z$. As δ goes to zero the scattered field u^s can be written as follows:*

$$\begin{aligned} u^s(x) &= -k_m^2\left(\frac{\varepsilon_c}{\varepsilon_m} - 1\right)|D|G(x,z,k_m)u^i(z) - \nabla_z G(x,z,k_m)\cdot M(\lambda,D)\nabla u^i(z) \\ &\quad + O\left(\frac{\delta^4}{\mathrm{dist}(\lambda,\sigma(\mathcal{K}_D^*))}\right) \end{aligned} \tag{2.41}$$

for x away from D. Here, $\mathrm{dist}(\lambda,\sigma(\mathcal{K}_D^*))$ denotes $\min_j|\lambda-\lambda_j|$ with λ_j being the eigenvalues of $\mathcal{K}_D^*$.

We denote the first term in the right hand side of (2.41) by u_1^s and the second term by u_2^s. It is clear that u_1^s represent monopole radiation and u_2^s the dipole radiation. We explicitly compute the scattering amplitude A_∞ in (2.34). Take $u^i(x) = e^{ik_m d\cdot x}$ and assume again for simplicity that $z = 0$. Note that

$$u_2^s(x) = \frac{e^{ik_m|x|}}{4\pi|x|} ik_m\left(ik_m\frac{x}{|x|} - \frac{x}{|x|^2}\right)\cdot M(\lambda,D)d.$$

In the far-field region, i.e. for $|x| \gg \frac{1}{\omega}$,

$$u_2^s(x) = -k_m^2\frac{e^{ik_m|x|}}{4\pi|x|}\left(\frac{x}{|x|}\cdot M(\lambda,D)d\right) + O\left(\frac{1}{|x|^2}\right).$$

On the other hand,

$$u_1^s(x) = k_m^2\frac{e^{ik_m|x|}}{4\pi|x|}\left(\frac{\varepsilon_c}{\varepsilon_m} - 1\right)\cdot|D|.$$

Throughout the chapter, we are interested in the case when the frequency is near the plasmonic resonant frequency, then the polarization tensor $M(\lambda, D)$ blow up and hence the magnitude of the dipole part u_2^s is much

greater than that of the monopole part u_1^s. Therefore, the leading term in the scattered field (2.41) is given by the dipole part, i.e.

$$u^s(x) \approx -k_m^2 \frac{e^{ik_m|x|}}{4\pi|x|}\left(\frac{x}{|x|}\cdot M(\lambda,D)d\right). \tag{2.42}$$

In the next proposition we write the extinction and scattering cross-sections in terms of the polarization tensor.

Propsition 2.4.3. *Near plasmonic resonant frequency, the leading-order term (as δ goes to zero) of the average over the orientation of the extinction cross-section of a randomly oriented nanoparticle is given by*

$$Q_m^{ext} = \frac{4\pi k_m}{3}\Im\left[\mathrm{Tr}M(\lambda,D)\right], \tag{2.43}$$

where Tr *denotes the trace of a matrix. The leading-order term of the average over the orientation scattering cross-section of a randomly oriented nanoparticle is given by*

$$Q_m^s = \frac{k_m^4}{9\pi}\left|\mathrm{Tr}M(\lambda,D)\right|^2. \tag{2.44}$$

Proof. Remark from (2.42) that the scattering amplitude A_∞ in the case of a plane wave illumination is given by

$$A_\infty\left(\frac{x}{|x|},d\right) = -\frac{k_m^2}{4\pi}\frac{x}{|x|}\cdot M(\lambda,D)d. \tag{2.45}$$

Using Theorem 2.4.1, we can see that for a given orientation

$$Q^{ext} = -4\pi k_m \Im\left[d\cdot M(\lambda,D)d\right].$$

Therefore, if we integrate Q^{ext} over all illuminations we find that

$$Q_m^{ext} = -\,k_m\Im\left[\int_{\mathbb{S}^2} d\cdot M(\lambda,D)d\,d\sigma(d)\right].$$

Since $\Im M(\lambda,D)$ is symmetric, it can be written as $\Im M(\lambda,D) = P^tN(\lambda)P$ where P is unitary and N is diagonal and real. Then, by the change of variables $d = P^tx$ and using spherical coordinates, it follows that

$$Q_m^{ext} \;=\; -k_m\left[\int_{\mathbb{S}^2} x\cdot N(\lambda)x d\sigma(x)\right],$$

and therefore,

$$Q_m^{ext} \;=\; -\frac{4\pi k_m}{3}\left[\mathrm{Tr}N(\lambda)\right] = -\frac{4\pi k_m}{3}\Im\left[\mathrm{Tr}M(\lambda,D)\right]. \tag{2.46}$$

Now, we compute the averaged scattering cross-section. Let $\Re M(\lambda, D) = \widetilde{P}^t \widetilde{N}(\lambda) \widetilde{P}$ where $\widetilde{P}$ is unitary and $\widetilde{N}$ is diagonal and real. We have

$$\begin{aligned} Q_m^s &= \frac{k_m^4}{16\pi^2} \iint_{\mathbb{S}^2 \times \mathbb{S}^2} |x \cdot M(\lambda, D) d|^2 \, d\sigma(x) \, d\sigma(d), \\ &= \frac{k_m^4}{16\pi^2} \left[\iint_{\mathbb{S}^2 \times \mathbb{S}^2} \left| \widetilde{x} \cdot N(\lambda) \widetilde{d} \right|^2 d\sigma(\widetilde{x}) d\sigma(\widetilde{d}) + \iint_{\mathbb{S}^2 \times \mathbb{S}^2} \left| \widetilde{x} \cdot \widetilde{N}(\lambda) \widetilde{d} \right|^2 d\sigma(\widetilde{x}) \, d\sigma(\widetilde{d}) \right]. \end{aligned}$$

Then a straightforward computation in spherical coordinates gives

$$Q_m^s = \frac{k_m^4}{9\pi} \left| \mathrm{Tr} M(\lambda, D) \right|^2,$$

which completes the proof. □

From Theorem 2.4.1, we obtain that the averaged absorption cross-section is given by

$$Q_m^a = -\frac{4\pi k_m}{3} \Im \left[\mathrm{Tr} M(\lambda, D) \right] - \frac{k_m^4}{9\pi} \left| \mathrm{Tr} M(\lambda, D) \right|^2.$$

Therefore, under the condition (2.30), Q_m^a blows up at plasmonic resonances.

2.4.5 An upper bound for the averaged extinction cross-section

The goal of this section is to derive an upper bound for the modulus of the averaged extinction cross-section Q_m^{ext} of a randomly oriented nanoparticle. Recall that the entries $M_{l,m}(\lambda, D)$ of the polarization tensor $M(\lambda, D)$ are given by

$$M_{l,m}(\lambda, D) := \int_{\partial D} x_l (\lambda I - \mathcal{K}_D^*)^{-1} [\nu_m](x) \, d\sigma(x). \tag{2.47}$$

For a $\mathcal{C}^{1,\alpha}$ domain D in $\mathbb{R}^d$, $\mathcal{K}_D^*$ is compact and self-adjoint in $\mathcal{H}^*$. Thus, we can write

$$(\lambda Id - \mathcal{K}_D^*)^{-1} [\psi] = \sum_{j=0}^{\infty} \frac{(\psi, \varphi_j)_{\mathcal{H}^*} \otimes \varphi_j}{\lambda - \lambda_j},$$

with (λ_j, φ_j) being the eigenvalues and eigenvectors of $\mathcal{K}_D^*$ in $\mathcal{H}^*$ (see Lemma 2.3.1). Hence, the entries of the polarization tensor M can be decomposed as

$$M_{l,m}(\lambda, D) = \sum_{j=1}^{\infty} \frac{\alpha_{l,m}^{(j)}}{\lambda - \lambda_j}, \tag{2.48}$$

where $\alpha_{l,m}^{(j)} := (\nu_m, \varphi_j)_{\mathcal{H}^*} (\varphi_j, x_l)_{-\frac{1}{2}, \frac{1}{2}}$. Note that $(\nu_m, \chi(\partial D))_{-\frac{1}{2}, \frac{1}{2}} = 0$. So, considering the fact that $\lambda_0 = 1/2$, we have $(\nu_m, \varphi_0)_{\mathcal{H}^*} = 0$ and so, $\alpha_{l,m}^{(0)} = 0$.

The following lemmas are useful for us.

Lemma 2.4.1. *We have*

$$\alpha_{l,l}^{(j)} \geq 0, \quad j \geq 1.$$

Proof. For $d = 3$, we have

$$\begin{aligned}(\varphi_j, x_l)_{-\frac{1}{2},\frac{1}{2}} &= \Big((\frac{1}{2} - \lambda_j)^{-1}(\frac{1}{2}Id - \mathcal{K}_D^*)[\varphi_j], x_l\Big)_{-\frac{1}{2},\frac{1}{2}} \\ &= \frac{-1}{1/2 - \lambda_j}\Big(\frac{\partial \mathcal{S}_D[\varphi_j]}{\partial \nu}\Big|_-, x_l\Big)_{-\frac{1}{2},\frac{1}{2}} \\ &= \int_{\partial D} \frac{\partial x_l}{\partial \nu}\mathcal{S}_D[\varphi_j]d\sigma - \int_D \Big(\Delta x_l \mathcal{S}_D[\varphi_j] - x_l \Delta \mathcal{S}_D[\varphi_j]\Big)dx \\ &= \frac{(\nu_l, \varphi_j)_{\mathcal{H}^*}}{1/2 - \lambda_j},\end{aligned}$$

where we used the fact that $\mathcal{S}_D[\varphi_j]$ is harmonic in D. The same result holds for $d = 2$ if we change $\mathcal{S}_D$ by $\widetilde{\mathcal{S}}_D$ (see Appendix B.3). Since $|\lambda_j| < 1/2$ for $j \geq 1$, we obtain the result. □

Lemma 2.4.2. *Let*

$$M_{l,m}(\lambda, D) = \sum_{j=1}^{\infty} \frac{\alpha_{l,m}^{(j)}}{\lambda - \lambda_j}$$

be the (l, m)-entry of the polarization tensor M associated with a $\mathcal{C}^{1,\alpha}$ domain $D \Subset \mathbb{R}^d$. Then, the following properties hold:

(i)

$$\sum_{j=1}^{\infty} \alpha_{l,m}^{(j)} = \delta_{l,m}|D|;$$

(ii)

$$\sum_{j=1}^{\infty} \lambda_i \sum_{l=1}^{d} \alpha_{l,l}^{(j)} = \frac{(d-2)}{2}|D|;$$

(iii)

$$\sum_{j=1}^{\infty} \lambda_j^2 \sum_{l=1}^{d} \alpha_{l,l}^{(j)} = \frac{(d-4)}{4}|D| + \sum_{l=1}^{d} \int_D |\nabla \mathcal{S}_D[\nu_l]|^2 dx.$$

Proof. The proof can be found in Appendix C. □

Let $\lambda = \lambda' + i\lambda''$. We have

$$\big|\Im(\mathrm{Tr}(M(\lambda, D)))\big| = \sum_{j=1}^{\infty} \frac{|\lambda''| \sum_{l=1}^{d} \alpha_{l,l}^{(j)}}{(\lambda' - \lambda_j)^2 + \lambda''^2}. \tag{2.49}$$

For $d = 2$ the spectrum $\sigma(\mathcal{K}_D^*)\backslash\{1/2\}$ is symmetric. For $d = 3$ this is no longer true. Nevertheless, for our purposes, we can assume that $\sigma(\mathcal{K}_D^*)\backslash\{1/2\}$ is symmetric by defining $\alpha_{l,l}^{(j)} = 0$ if λ_j is not in the original spectrum.

Without loss of generality we assume for ease of notation that Conditions 2.2 and 2.3 hold. Then we define the bijection $\rho : \mathbb{N}^+ \to \mathbb{N}^+$ such that

$\lambda_{\rho(j)} = -\lambda_j$ and we can write

$$\begin{aligned}\left|\Im(\mathrm{Tr}(M(\lambda,D)))\right| &= \frac{1}{2}\left(\sum_{j=1}^{\infty}\frac{|\lambda''|\beta_j}{(\lambda'-\lambda_j)^2+\lambda''^2}+\sum_{j=1}^{\infty}\frac{|\lambda''|\beta^{(\rho(j))}}{(\lambda'+\lambda_j)^2+\lambda''^2}\right)\\ &= \frac{|\lambda''|}{2}\sum_{j=1}^{\infty}\frac{(\lambda'^2+\lambda''^2+\lambda_j^2)(\beta^{(j)}+\beta^{(\rho(j))})+2\lambda'\lambda_j(\beta^{(j)}-\beta^{(\rho(j))})}{\big((\lambda'-\lambda_j)^2+\lambda''^2\big)\big((\lambda'+\lambda_j)^2+\lambda''^2\big)},\end{aligned}$$

where $\beta_j = \sum_{l=1}^{d}\alpha_{l,l}^{(j)}$.

From Lemma 2.4.1 it follows that

$$\frac{(\lambda'^2+\lambda''^2+\lambda_j^2)(\beta^{(j)}+\beta^{(\rho(j))})+2\lambda'\lambda_j(\beta^{(j)}-\beta^{(\rho(j))})}{\big((\lambda'-\lambda_j)^2+\lambda''^2\big)\big((\lambda'+\lambda_j)^2+\lambda''^2\big)} \geq 0.$$

Moreover,

$$\begin{aligned}&\frac{(\lambda'^2+\lambda''^2+\lambda_j^2)(\beta^{(j)}+\beta^{(\rho(j))})+2\lambda'\lambda_j(\beta^{(j)}-\beta^{(\rho(j))})}{\big((\lambda'-\lambda_j)^2+\lambda''^2\big)\big((\lambda'+\lambda_j)^2+\lambda''^2\big)} \leq\\ &\frac{(\lambda'^2+\lambda''^2+\lambda_j^2)(\beta^{(j)}+\beta^{(\rho(j))})+2\lambda'\lambda_j(\beta^{(j)}-\beta^{(\rho(j))})}{\lambda''^2(4\lambda'^2+\lambda''^2)}+O(\frac{\lambda''^2}{4\lambda'^2+\lambda''^2}).\end{aligned}$$

Hence,

$$\begin{aligned}&\left|\Im(\mathrm{Tr}(M(\lambda,D)))\right| \leq\\ &\frac{|\lambda''|}{2}\sum_{j=1}^{\infty}\frac{(\lambda'^2+\lambda''^2+\lambda_j^2)(\beta^{(j)}+\beta^{(\rho(j))})+2\lambda'(\lambda_j\beta^{(j)}+\lambda_{\rho(j)}\beta^{(\rho(j))})}{\lambda''^2(4\lambda'^2+\lambda''^2)}+O(\frac{\lambda''^2}{4\lambda'^2+\lambda''^2}).\end{aligned}$$

Using Lemma 2.4.2 we obtain the following result.

Theorem 2.4.2. *Let $M(\lambda,D)$ be the polarization tensor associated with a $\mathcal{C}^{1,\alpha}$ domain $D \Subset \mathbb{R}^d$ with $\lambda = \lambda' + i\lambda''$ such that $|\lambda''| \ll 1$ and $|\lambda'| < 1/2$. Then,*

$$\begin{aligned}&\left|\Im(\mathrm{Tr}(M(\lambda,D)))\right| \leq \frac{d|\lambda''||D|}{\lambda''^2+4\lambda'^2}+\\ &\frac{1}{|\lambda''|(\lambda''^2+4\lambda'^2)}\left(d\lambda'^2|D|+\frac{(d-4)}{4}|D|+\sum_{l=1}^{d}\int_D|\nabla\mathcal{S}_D[\nu_l]|^2dx+2\lambda'\frac{(d-2)}{2}|D|\right)+\\ &O(\frac{\lambda''^2}{4\lambda'^2+\lambda''^2}).\end{aligned}$$

The bound in the above theorem depends not only on the volume of the particle but also on its geometry. Nevertheless, we remark that, since $|\lambda_j| < \frac{1}{2}$,

$$\sum_{j=1}^{\infty}\lambda_j^2\sum_{l=1}^{d}\alpha_{l,l}^{(j)} < \frac{d|D|}{4}.$$

Hence, we can find a geometry independent, but not optimal, bound.

Corollary 2.4.1. *We have*

$$\left|\Im(\mathrm{Tr}(M(\lambda,D)))\right| \leq \frac{1}{|\lambda''|(\lambda''^2+4\lambda'^2)}\left(d|D|\big(\lambda'^2+\frac{1}{4}\big)+2\lambda'\frac{(d-2)}{2}|D|\right)+\frac{d|\lambda''||D|}{\lambda''^2+4\lambda'^2}+O(\frac{\lambda''^2}{4\lambda'^2+\lambda''^2}). \tag{2.50}$$

Bound for ellipses

If D is an ellipse whose semi-axes are on the x_1- and x_2- axes and of length a and b, respectively, then its polarization tensor takes the form [12]

$$M(\lambda,D)=\begin{pmatrix} \dfrac{|D|}{\lambda-\frac{1}{2}\frac{a-b}{a+b}} & 0 \\ 0 & \dfrac{|D|}{\lambda+\frac{1}{2}\frac{a-b}{a+b}} \end{pmatrix}. \tag{2.51}$$

On the other hand, it is known that in $\mathcal{H}^*(\partial D)$ [65]

$$\sigma(\mathcal{K}_D^*)\backslash\{1/2\}=\left\{\pm\frac{1}{2}\left(\frac{a-b}{a+b}\right)^j,\quad j=1,2,\dots\right\}.$$

Then, from (2.48), we also have

$$M(\lambda,D)=\begin{pmatrix} \displaystyle\sum_{j=1}^{\infty}\frac{\alpha_{1,1}^{(j)}}{\lambda-\frac{1}{2}\left(\frac{a-b}{a+b}\right)^j} & \displaystyle\sum_{j=1}^{\infty}\frac{\alpha_{1,2}^{(j)}}{\lambda-\frac{1}{2}\left(\frac{a-b}{a+b}\right)^j} \\ \displaystyle\sum_{j=1}^{\infty}\frac{\alpha_{1,2}^{(j)}}{\lambda-\frac{1}{2}\left(\frac{a-b}{a+b}\right)^j} & \displaystyle\sum_{j=1}^{\infty}\frac{\alpha_{2,2}^{(j)}}{\lambda-\frac{1}{2}\left(\frac{a-b}{a+b}\right)^j} \end{pmatrix}.$$

Let $\lambda_1=\dfrac{1}{2}\dfrac{a-b}{a+b}$ and $\mathcal{V}(\lambda_j)=\{i\in\mathbb{N}$ such that $\mathcal{K}_D^*[\varphi_i]=\lambda_j\varphi_i\}$. It is clear now that

$$\sum_{i\in\mathcal{V}(\lambda_1)}\alpha_{1,1}^{(i)}=\sum_{i\in\mathcal{V}(-\lambda_1)}\alpha_{2,2}^{(i)}=|D|,\quad \sum_{i\in\mathcal{V}(\lambda_j)}\alpha_{1,1}^{(i)}=\sum_{i\in\mathcal{V}(-\lambda_j)}\alpha_{2,2}^{(i)}=0 \tag{2.52}$$

for $j\geq 2$ and

$$\sum_{i\in\mathcal{V}(\lambda_j)}\alpha_{1,2}^{(i)}=0$$

for $j\geq 1$.

In view of (2.52), we have

$$\frac{\beta^{(j)}}{(\lambda'-\lambda_j)^2+\lambda''^2}+\frac{\beta^{(\rho(j))}}{(\lambda'+\lambda_j)^2+\lambda''^2}\leq\frac{4\lambda'^2\beta^{(j)}+\lambda''^2(\beta^{(j)}+\beta^{(j)})}{\lambda''^2(4\lambda'^2+\lambda''^2)}+O(\frac{\lambda''^2}{4\lambda'^2+\lambda''^2}).$$

Hence,

$$|\Im(\mathrm{Tr}(M(\lambda,D)))|\leq\frac{|\lambda''|}{2}\sum_{j=1}^{\infty}\frac{4\lambda'^2\beta^{(j)}+\lambda''^2(\beta^{(j)}+\beta^{(j)})}{\lambda''^2(4\lambda'^2+\lambda''^2)}+O(\frac{\lambda''^2}{4\lambda'^2+\lambda''^2}).$$

Note that for for any ellipse $\widetilde{D}$ of semi-axes of length a and b, $\Im(\mathrm{Tr}(M(\lambda,\widetilde{D}))) = \Im(\mathrm{Tr}(M(\lambda,D)))$. Then using Lemma 2.4.2 we obtain the following result.

Corollary 2.4.2. *For any ellipse $\widetilde{D}$ of semi-axes of length a and b, we have*

$$|\Im(Tr(M(\lambda,\widetilde{D})))| \leq \frac{|\widetilde{D}|4\lambda'^2}{|\lambda''|(\lambda''^2+4\lambda'^2)} + \frac{2|\lambda''||\widetilde{D}|}{\lambda''^2+4\lambda'^2} + O(\frac{\lambda''^2}{4\lambda'^2+\lambda''^2}). \quad (2.53)$$

Figure 2.1 shows (2.53) and the average extinction of two ellipses of semi-axis a and b, where the ratio $a/b = 2$ and $a/b = 4$, respectively.

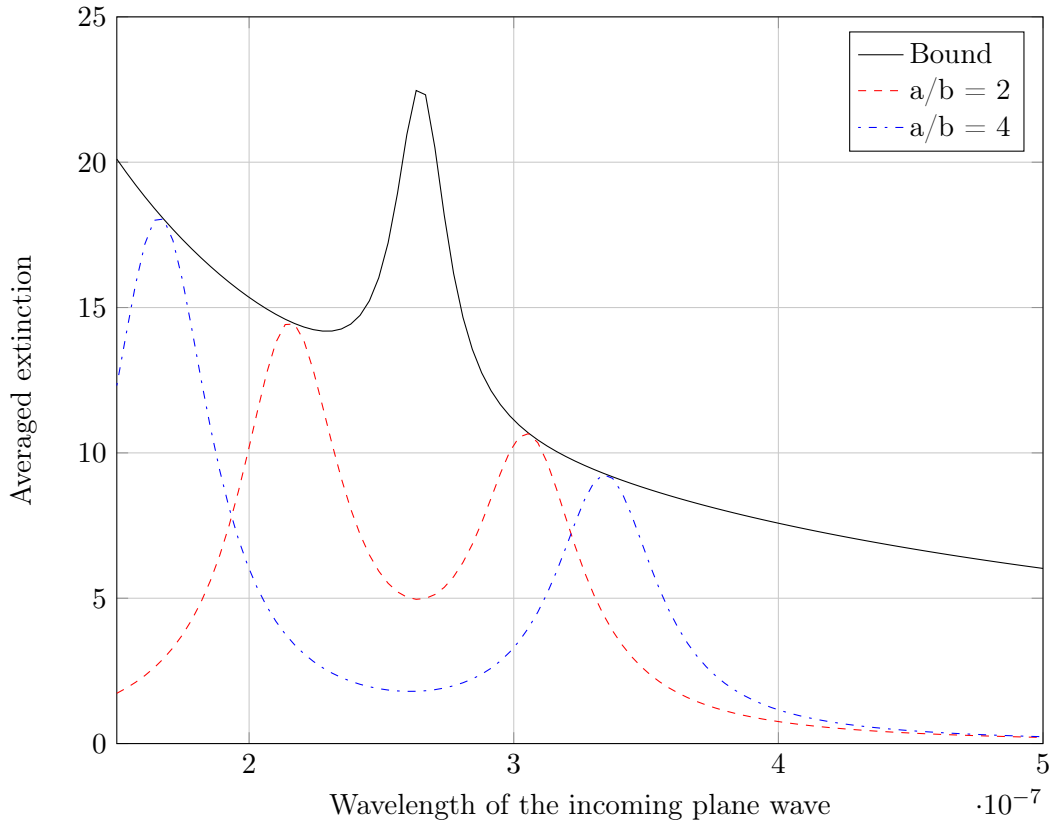

FIGURE 2.1: Optimal bound for ellipses.

We can see from (2.49), Lemma 2.4.1 and the first sum rule in Lemma 2.4.2 that for an arbitrary shape B, $|\Im(\mathrm{Tr}(M(\lambda,B)))|$ is a convex combination of $\frac{|\lambda''|}{(\lambda'-\lambda_j)^2+\lambda''^2}$ for $\lambda_j \in \sigma(\mathcal{K}_B^*)\backslash\{1/2\}$. Since ellipses put all the weight of this convex combination in $\pm\lambda_1 = \pm\frac{1}{2}\frac{a-b}{a+b}$, we have for any ellipse $\widetilde{D}$ and any shape B such that $|B| = |\widetilde{D}|$,

$$|\Im(\mathrm{Tr}(M(\lambda^*,B)))| \leq |\Im(\mathrm{Tr}(M(\lambda^*,\widetilde{D})))|$$

with $\lambda^* = \pm\frac{1}{2}\frac{a-b}{a+b} + i\lambda''$.

Thus, bound (2.53) applies for any arbitrary shape B in dimension two. This implies that, for a given material and a given desired resonance frequency ω^*, the optimal shape for the extinction resonance (in the quasi-static limit) is an ellipse of semi-axis a and b such that $\lambda'(\omega^*) = \pm\frac{1}{2}\frac{a-b}{a+b}$.

Bound for ellipsoids

Let D be an ellipsoid given by

$$\frac{x_1^2}{p_1^2}+\frac{x_2^2}{p_2^2}+\frac{x_3^2}{p_3^2}=1. \tag{2.54}$$

The following holds [12].

Lemma 2.4.3. *Let D be the ellipsoid defined by (2.54). Then, for $x \in D$,*

$$\mathcal{S}_D[\nu_l](x) = s_l x_l, \quad l=1,2,3,$$

where

$$s_l = -\frac{p_1p_2p_3}{2}\int_0^\infty \frac{1}{(p_l^2+s)\sqrt{(p_1^2+s)(p_2^2+s)(p_3^2+s)}}ds.$$

Then we have

$$\sum_{l=1}^{3}\int_D |\nabla \mathcal{S}_D[\nu_l]|^2 dx = (s_1^2+s_2^2+s_3^2)|D|.$$

For a rotated ellipsoid $\widetilde{D}=\mathcal{R}D$ with $\mathcal{R}$ being a rotation matrix, we have $M(\lambda,\widetilde{D}) = \mathcal{R}M(\lambda,D)\mathcal{R}^T$ and so $\mathrm{Tr}(M(\lambda,\widetilde{D})) = \mathrm{Tr}(M(\lambda,D))$. Therefore, for any ellipsoid $\widetilde{D}$ of semi-axes of length p_1,p_2 and p_3 the following result holds.

Corollary 2.4.3. *For any ellipsoid $\widetilde{D}$ of semi-axes of length p_1,p_2 and p_3, we have*

$$\Im(\mathrm{Tr}(M(\lambda,\widetilde{D}))) \leq \frac{|\widetilde{D}|\left(3\lambda'^2+\lambda'-\frac{1}{4}+(s_1^2+s_2^2+s_3^2)\right)}{|\lambda''|(\lambda''^2+4\lambda'^2)}+\frac{3|\lambda''||\widetilde{D}|}{\lambda''^2+4\lambda'^2}+O\left(\frac{\lambda''^2}{4\lambda'^2+\lambda''^2}\right), \tag{2.55}$$

where for $j=1,2,3$,

$$s_j = -\frac{p_1p_2p_3}{2}\int_0^\infty \frac{1}{(p_j^2+s)\sqrt{(p_1^2+s)(p_2^2+s)(p_3^2+s)}}ds.$$

2.5 Link with the scattering coefficients

Our aim in this section is to exhibit the mechanism underlying plasmonic resonances in terms of the scattering coefficients corresponding to the nanoparticle. The concept of scattering coefficients was first introduced in [20]. It plays a key role in constructing cloaking structures. It was extended in [21] to the full Maxwell equations. The scattering coefficients are simply the Fourier coefficients of the scattering amplitude A_∞. In Theorem 2.5.1 we provide an asymptotic expansion of the scattering amplitude in terms of the scattering coefficients of order ± 1. Our formula shows that, under physical conditions, the scattering coefficients of orders ± 1 are the only scattering coefficients inducing the scattering cross-section enhancement. For simplicity we only consider here the two-dimensional case.

2.5.1 The notion of scattering coefficients

From Graf's addition formula [12] and (2.4) the following asymptotic formula holds as $|x| \to \infty$

$$u^s(x) = (u - u^i)(x) = -\frac{i}{4} \sum_{n\in\mathbb{Z}} H_n^{(1)}(k_m|x|)e^{in\theta_x} \int_{\partial D} J_n(k_m|y|)e^{-in\theta_y}\psi(y)d\sigma(y),$$

where $x = (|x|, \theta_x)$ in polar coordinates, $H_n^{(1)}$ is the Hankel function of the first kind and order n, J_n is the Bessel function of order n and ψ is the solution to (2.6).
For $u^i(x) = e^{ik_m d\cdot x}$ we have

$$u^i(x) = \sum_{m\in\mathbb{Z}} a_m(u^i)J_m(k_m|x|)e^{im\theta_x},$$

where $a_m(u^i) = e^{im(\frac{\pi}{2}-\theta_d)}$. By the superposition principle, we get

$$\psi = \sum_{m\in\mathbb{Z}} a_m(u^i)\psi_m,$$

where ψ_m is solution to (2.6) replacing f by

$$f^{(m)} := F_2^{(m)} + \frac{1}{\mu_c}\big(\frac{1}{2}Id - (\mathcal{K}_D^{k_c})^*\big)(\mathcal{S}_D^{k_c})^{-1}[F_1^{(m)}]$$

with

$$\begin{aligned} F_1^{(m)}(x) &= -J_m(k_m|x|)e^{im\theta_x}, \\ F_2^{(m)}(x) &= -\frac{1}{\mu_m}\frac{\partial J_m(k_m|x|)e^{im\theta_x}}{\partial\nu}. \end{aligned}$$

We have

$$u^s(x) = (u - u^i)(x) = -\frac{i}{4} \sum_{n\in\mathbb{Z}} H_n^{(1)}(k_m|x|)e^{in\theta_x} \sum_{m\in\mathbb{Z}} W_{nm}e^{im(\frac{\pi}{2}-\theta_d)},$$

where

$$W_{nm} = \int_{\partial D} J_n(k_m|y|)e^{-in\theta_y}\psi_m(y)d\sigma(y). \tag{2.56}$$

The coefficients W_{nm} are called the scattering coefficients.

Lemma 2.5.1. *In the space $\mathcal{H}^*(\partial D)$, as ω goes to zero, we have*

$$\begin{aligned} f^{(0)} &= O(\omega^2), \\ f^{(\pm1)} &= \omega f_1^{(\pm1)} + O(\omega^2), \\ f^{(m)} &= O(\omega^m), \quad |m| > 1, \end{aligned}$$

where

$$f_1^{(\pm1)} = \mp\frac{\sqrt{\varepsilon_m\mu_m}}{2}\Big(\frac{1}{\mu_m}e^{i\pm\theta_\nu} + \frac{1}{\mu_c}(\frac{1}{2}Id - \mathcal{K}_D^*)\widetilde{\mathcal{S}}_D^{-1}[|x|e^{i\pm\theta_x}]\Big).$$

Proof. Recall that $J_0(x) = 1 + O(x^2)$. By virtue of the fact that

$$\big(\frac{1}{2}Id - (\mathcal{K}_D^{k_c})^*\big)(\mathcal{S}_D^{k_c})^{-1}[\chi(\partial D)] = O(\omega^2),$$

we arrive at the estimate for $f^{(0)}$ (see Appendix B.3). Moreover,

$$J_{\pm 1}(x) = \pm\frac{x}{2} + O(x^3)$$

together with the fact that

$$\big(\frac{1}{2}Id - (\mathcal{K}_D^{k_c})^*\big)(\mathcal{S}_D^{k_c})^{-1} = (\frac{1}{2}Id - \mathcal{K}_D^*)\widetilde{\mathcal{S}}_D^{-1} + O(\omega^2 \log \omega)$$

gives the expansion of $f^{(\pm 1)}$ in terms of ω (see Appendix B.3).

Finally, $J_m(x) = O(x^m)$ immediately yields the desired estimate for $f^{(m)}$. □

From Theorem 3.3.2, we can see that

$$\psi_m = \sum_{j\in J} \frac{\big(f^{(m)}, \widetilde{\varphi}_j(\omega)\big)_{\mathcal{H}^*}\varphi_j(\omega)}{\tau_j(\omega)} + \mathcal{A}_D(\omega)^{-1}(P_{J^c}(\omega)f). \tag{2.57}$$

Hence, from the definition of the scattering coefficients,

$$W_{nm} = \sum_{j\in J} \frac{\big(f^{(m)}, \widetilde{\varphi}_j(\omega)\big)_{\mathcal{H}^*}\Big(\varphi_j(\omega), J_n(k_m|x|)e^{-in\theta_x}\Big)_{-\frac{1}{2},\frac{1}{2}}}{\tau_j(\omega)} + \int_{\partial D} J_n(k_m|y|)e^{-in\theta_y}O(\omega)d\sigma(y). \tag{2.58}$$

Since

$$J_m(x) \sim \frac{1}{\sqrt{(2\pi|m|)}}\Big(\frac{ex}{2|m|}\Big)^{|m|}$$

as $m \to \infty$, we have

$$|f^{(m)}| \le \frac{C^{|m|}}{|m|^{|m|}}.$$

Using the Cauchy-Schwarz inequality and Lemma 2.5.1, we obtain the following result.

Propsition 2.5.1. *For $|n|, |m| > 0$, we have*

$$|W_{nm}| \le \frac{O(\omega^{|n|+|m|})}{\min_{j\in J}|\tau_j(\omega)|}\frac{C^{|n|+|m|}}{|n|^{|n|}|m|^{|m|}}$$

for a positive constant C independent of ω.

2.5.2 The leading-order term in the expansion of the scattering amplitude

In the following, we analyze the first-order scattering coefficients.

Lemma 2.5.2. *Assume that Conditions* 1 *and* 2 *hold. Then,*

$$\begin{aligned}\psi_0 &= \sum_{j\in J}\frac{O(\omega^2)}{\tau_j(\omega)}+O(\omega),\\ \psi_{\pm 1} &= \sum_{j\in J}\frac{\pm\omega\frac{\sqrt{\varepsilon_m\mu_m}}{2}\left(\frac{1}{\mu_m}-\frac{1}{\mu_c}\right)(e^{\pm i\theta_\nu},\varphi_j)_{\mathcal{H}^*}\varphi_j+O(\omega^3\log\omega)}{\tau_j(\omega)}+O(\omega).\end{aligned}$$

Proof. The expression of ψ_0 follows from (2.57) and Lemma 2.5.1. Changing $\mathcal{S}_D$ by $\widetilde{\mathcal{S}}_D$ in Theorem 3.3.2 gives $\left((\frac{1}{2}Id-\mathcal{K}_D^*)\widetilde{\mathcal{S}}_D^{-1}[|x|e^{i\theta_x}],\varphi_j\right)_{\mathcal{H}^*}=-(e^{i\theta_\nu},\varphi_j)_{\mathcal{H}^*}$. Using now Lemma 2.5.1 in (2.57) yields the expression of $\psi_{\pm1}$. □

Recall that in two dimensions,

$$\tau_j(\omega)=\frac{1}{2\mu_m}+\frac{1}{2\mu_c}-(\frac{1}{\mu_c}-\frac{1}{\mu_m})\lambda_j+O(\omega^2\log\omega),$$

where λ_j is an eigenvalue of $\mathcal{K}_D^*$ and $\lambda_0=1/2$. Recall also that for $0\in J$ we need $\tau_j\to 0$ and so $\mu_m\to\infty$, which is a limiting case that we can ignore. In practice, $P_J(\omega)[\varphi_0(\omega)]=0$. We also have $(\varphi_j,\chi(\partial D))_{-\frac{1}{2},\frac{1}{2}}=0$ for $j\neq 0$. It follows then from the above lemmas and the expression (2.58) of the scattering coefficients that

$$\begin{aligned}W_{00} &= \sum_{j\in J}\frac{O(\omega^4\log\omega)}{\tau_j(\omega)}+O(\omega),\\ W_{0\pm1} &= \sum_{j\in J}\frac{O(\omega^3\log\omega)}{\tau_j(\omega)}+O(\omega),\\ W_{\pm10} &= \sum_{j\in J}\frac{O(\omega^3)}{\tau_j(\omega)}+O(\omega^2).\end{aligned}$$

Note that $W_{\pm1\pm1}$ has a special structure. Indeed, from Lemma 2.5.2 and equation (2.58), we have

$$\begin{aligned}W_{\pm1\pm1} &= \sum_{j\in J}\frac{\pm\pm\omega\frac{\sqrt{\varepsilon_m\mu_m}}{2}\left(\frac{1}{\mu_m}-\frac{1}{\mu_c}\right)(\varphi_j,J_1(k_m|x|)e^{\mp i\theta_x})_{-\frac{1}{2},\frac{1}{2}}(e^{\pm i\theta_\nu},\varphi_j)_{\mathcal{H}^*}+O(\omega^4\log\omega)}{\tau_j(\omega)}\\ &\quad +O(\omega^2),\\ &= \sum_{j\in J}\frac{\pm\pm\omega^2\frac{\varepsilon_m\mu_m}{4}\left(\frac{1}{\mu_m}-\frac{1}{\mu_c}\right)(\varphi_j,|x|e^{\mp i\theta_x})_{-\frac{1}{2},\frac{1}{2}}(e^{\pm i\theta_\nu},\varphi_j)_{\mathcal{H}^*}+O(\omega^4\log\omega)}{\tau_j(\omega)}+O(\omega^2),\\ &= \frac{k_m^2}{4}\left(\sum_{j\in J}\frac{\pm\pm(\varphi_j,|x|e^{\mp i\theta_x})_{-\frac{1}{2},\frac{1}{2}}(e^{\pm i\theta_\nu},\varphi_j)_{\mathcal{H}^*}+O(\omega^2\log\omega)}{\lambda-\lambda_j+O(\omega^2\log\omega)}+O(1)\right),\end{aligned}$$

where λ is defined by (2.23). Now, assume that $\min_{j\in J}|\tau_j(\omega)| \gg \omega^2 \log\omega$. Then,

$$W_{\pm 1\pm 1} = \frac{k_m^2}{4}\left(\sum_{j\in J}\frac{\pm\pm\left(\varphi_j, |x|e^{\mp i\theta_x}\right)_{-\frac{1}{2},\frac{1}{2}}\left(e^{\pm i\theta_\nu},\varphi_j\right)_{\mathcal{H}^*}}{\lambda-\lambda_j} + O(1)\right). \quad (2.59)$$

Define the contracted polarization tensors by

$$N_{\pm,\pm}(\lambda, D) := \int_{\partial D} |x|e^{\pm i\theta_x}(\lambda I - \mathcal{K}_D^*)^{-1}[e^{\pm i\theta_\nu}](x)\, d\sigma(x).$$

It is clear that

$$\begin{aligned}
N_{+,+}(\lambda, D) &= M_{1,1}(\lambda, D) - M_{2,2}(\lambda, D) + i2M_{1,2}(\lambda, D),\\
N_{+,-}(\lambda, D) &= M_{1,1}(\lambda, D) + M_{2,2}(\lambda, D),\\
N_{-,+}(\lambda, D) &= M_{1,1}(\lambda, D) + M_{2,2}(\lambda, D),\\
N_{-,-}(\lambda, D) &= M_{1,1}(\lambda, D) - M_{2,2}(\lambda, D) - i2M_{1,2}(\lambda, D),
\end{aligned}$$

where $M_{l,m}(\lambda, D)$ is the (l, m)-entry of the polarization tensor given by (2.40).

Finally, considering the above we can state the following result.

Theorem 2.5.1. *Let A_∞ be the scattering amplitude in the far-field defined in (2.35) for the incoming plane wave $u^i(x) = e^{ik_m d\cdot x}$. Assume Conditions 1 and 2 and*

$$\min_{j\in J}|\tau_j(\omega)| \gg \omega^2\log\omega.$$

Then, A_∞ admits the following asymptotic expansion

$$A_\infty\left(\frac{x}{|x|}\right) = \frac{x}{|x|}^T W_1 d + O(\omega^2),$$

where

$$W_1 = \begin{pmatrix} W_{-11} + W_{1-1} - 2W_{11} & i\left(W_{1-1} - W_{-11}\right)\\ i\left(W_{1-1} - W_{-11}\right) & -W_{-11} - W_{1-1} - 2W_{11}\end{pmatrix}.$$

Here, W_{nm} are the scattering coefficients defined by (2.56).

Proof. From (2.45), we have

$$A_\infty\left(\frac{x}{|x|}\right) = -k_m^2\frac{x}{|x|}^T M(\lambda, D)d.$$

Since $\mathcal{K}_D^*$ is compact and self-adjoint in $\mathcal{H}^*$, we have

$$\begin{aligned}
N_{\pm,\pm}(\lambda, D) &= \sum_{j=1}^{\infty}\frac{\left(\varphi_j, |x|e^{\pm i\theta_x}\right)_{-\frac{1}{2},\frac{1}{2}}\left(e^{\pm i\theta_\nu},\varphi_j\right)_{\mathcal{H}^*}}{\lambda-\lambda_j}\\
&= \sum_{j\in J}\frac{\left(\varphi_j, |x|e^{\pm i\theta_x}\right)_{-\frac{1}{2},\frac{1}{2}}\left(e^{\pm i\theta_\nu},\varphi_j\right)_{\mathcal{H}^*}}{\lambda-\lambda_j} + O(1).
\end{aligned}$$

We have then from (2.59) that

$$\begin{aligned}
-\frac{k_m^2}{4} N_{+,+}(\lambda, D) &= W_{-11} + O(\omega^2), \\
-\frac{k_m^2}{4} N_{+,-}(\lambda, D) &= -W_{11} + O(\omega^2), \\
-\frac{k_m^2}{4} N_{-,+}(\lambda, D) &= -W_{11} + O(\omega^2), \\
-\frac{k_m^2}{4} N_{-,-}(\lambda, D) &= W_{1-1} + O(\omega^2).
\end{aligned}$$

In view of

$$\begin{aligned}
M_{11} &= \frac{1}{4}\left(N_{+,+} + N_{-,-} + 2N_{+,-}\right), \\
M_{22} &= \frac{1}{4}\left(-N_{+,+} - N_{-,-} + 2N_{+,-}\right), \\
M_{12} &= \frac{-i}{4}\left(N_{+,+} - N_{-,-}\right),
\end{aligned}$$

we get the result. □

2.6 Concluding remarks

In this chapter, based on perturbation arguments, we studied the scattering by plasmonic nanoparticles when the frequency of the incoming light is close to a resonant frequency.

We have derived the shift and broadening of the plasmon resonance with changes in size. The localization algorithms developed in [12, 41] can be extended to the problem of imaging plasmonic nanoparticles. We have precisely quantified the scattering and absorption cross-section enhancements and gave optimal bounds on the enhancement factors. We have also linked the plasmonic resonances to the scattering coefficients and showed that the leading-order term of the scattering amplitude can be expressed in terms of the $\pm$-one order of the scattering coefficients.

The generalization to the full Maxwell equations of the methods and results of the chapter are the subject of chapter 3.

Chapter 3

The Full Maxwell Equations

Contents

3.1 Introduction

The optical response of plasmon resonant nanoparticles is dominated by the appearance of plasmon resonances over a wide range of wavelengths [68]. For individual particles or very low concentrations in a solvent of non-interacting nanoparticles, separated from one another by distances larger than the wavelength, these resonances depend on the electromagnetic parameters of the nanoparticle, those of the surrounding material, and the particle shape and size. High scattering and absorption cross sections and strong near-fields are unique effects of plasmonic resonant nanoparticles. One of the most important parameters in the context of applications is the position of the resonances in terms of wavelength or frequency. A longstanding problem is to tune this position by changing the particle size or the concentration of the nanoparticles in a solvent [49, 68]. It was experimentally observed, for instance, in [49, 89] that the scaling behavior of nanoparticles is critical. The question of how the resonant properties of plasmonic nanoparticles develops with increasing size or/and concentration is therefore fundamental.

At the quasi-static limit, plasmon resonances in nanoparticles can be treated as an eigenvalue problem for the Neumann-Poincaré integral operator [9, 52, 72, 73]. Unfortunately, at this limit, they are size-independent.

This chapter provides the first mathematical study of the shift in plasmon resonance using the full Maxwell equations. It generalizes to the full Maxwell equations the results obtained in chapter 2 where the Helmholtz equation was used to model light propagation. Theorem 3.3.1 gives an asymptotic expansion of the plasmonic resonances in terms of the size of the nanoparticle. Theorem 3.3.2 provides the near field behavior of the electromagnetic fields near the plasmonic resonant frequencies. The far-field behavior is described in Theorem 3.4.1. Theorem 3.4.2 shows the blow up rate of the extinction cross section (the sum of the absorption and scattering cross sections) at the plasmonic resonance. Theorem 3.5.1 in section 3.5 considers the special case of spherical nanoparticles.

The chapter is organized as follows. In section 3.2 we first review commonly used function spaces. Then we introduce layer potentials associated with the Laplace operator and recall their mapping properties. Of particular interest is the Neumann-Poincaré operator $\mathcal{K}_D^*$ associated with the particle D defined in (3.4). We state some of its important properties in Lemma 3.2.1.

In section 3.3 we first derive a layer potential formulation for the scattering problem for the full Maxwell equations in (3.11). Then we obtain a first-order correction to plasmonic resonances in terms of the size of the nanoparticle in Theorem 3.3.1.

This enables us to analyze the shift and broadening of the plasmon resonance with changes in size and shape of the nanoparticles. The resonance condition is determined from absorption and scattering blow up and depends on the shape, size and electromagnetic parameters of both the nanoparticle and the surrounding material. Surprisingly, it turns out that in this case not only the spectrum of the Neumann-Poincaré operator plays a role in the resonance of the nanoparticles, but also its negative, i.e., $-\sigma(\mathcal{K}_D^*)$. We explain how in the quasi-static limit, only the spectrum of the Neumann-Poincaré operator can be excited. This is an important finding in our chapter. Note that it is not clear for what kind of geometries in $\mathbb{R}^3$ the spectrum of the

Neumann-Poincaré operator has symmetry, that is, if $\lambda \in \sigma(\mathcal{K}_D^*)$ so does $-\lambda$. For instance, such symmetry is not present in the case of a spherical nanoparticle while for a spherical shell the spectrum of the associated Neumann-Poincaré operator is symmetric around zero.

When the particle size increases and deviates from the dipole approximation, the resonances become size-dependent. Moreover, a part of the spectrum of negative of the Neumann-Poincaré operator can be excited as in higher-order terms in the expansion of the electric field versus the size of the particle.

In section 3.4, using the quasi-static limit for the electromagnetic fields, we derive a formula for the enhancement of the extinction cross-section.

Finally, in seccion 3.5 we provide calculations for the case of spherical nanoparticles and explicitly compute the shift in the spectrum of the Neumann-Poincaré operator and the extinction cross-section. In section 3.6 we consider the case of a spherical shell and apply degenerate perturbation theory since the eigenvalues associated with the corresponding Neumann-Poincaré operator are not simple. The explicit results obtained in sections 3.5 and 3.6 illustrate our main findings in sections 3.3 and 3.4.

3.2 Preliminaries

Here and throughout this chapter, we assume that D is bounded, simply connected, and of class $\mathcal{C}^{1,\alpha}$ for $0 < \alpha < 1$. We note by $\nabla\times$ the curl operator for a vector field in $\mathbb{R}^3$. We denote by $H^s(\partial D)$ the usual Sobolev space of order s on ∂D and

$$H_T^s(\partial D) = \left\{ \varphi \in \left(H^s(\partial D)\right)^3, \nu \cdot \varphi = 0 \right\}.$$

We also need the space $H^1_{\text{loc}}(\mathbb{R}^3)$ of functions locally in $H^1(\mathbb{R}^3)$.

We introduce the surface gradient, surface divergence and Laplace-Beltrami operator and denote them by $\nabla_{\partial D}$, $\nabla_{\partial D}\cdot$ and $\Delta_{\partial D}$, respectively. We define the vectorial and scalar surface curl by $\vec{\text{curl}}_{\partial D}\varphi = -\nu \times \nabla_{\partial D}\varphi$ for $\varphi \in H^{\frac{1}{2}}(\partial D)$ and $\text{curl}_{\partial D}\varphi = -\nu \cdot (\nabla_{\partial D} \times \varphi)$ for $\varphi \in H_T^{-\frac{1}{2}}(\partial D)$, respectively. We remind that

$$\begin{aligned} \nabla_{\partial D} \cdot \nabla_{\partial D} &= \Delta_{\partial D}, \\ \text{curl}_{\partial D}\vec{\text{curl}}_{\partial D} &= -\Delta_{\partial D}, \\ \nabla_{\partial D} \cdot \vec{\text{curl}}_{\partial D} &= 0, \\ \text{curl}_{\partial D}\nabla_{\partial D} &= 0. \end{aligned}$$

We introduce the following functional space:

$$H_T^{-\frac{1}{2}}(\text{div}, \partial D) = \left\{ \varphi \in H_T^{-\frac{1}{2}}(\partial D), \nabla_{\partial D} \cdot \varphi \in H^{-\frac{1}{2}}(\partial D) \right\}.$$

Let G be the Green function for the Helmholtz operator $\Delta + k^2$, that is,

$$(\Delta + k^2)G(x, y, k) = \delta_y,$$

where δ_y is the Dirac mass at y, subject to the Sommerfeld radiation condition in dimension three

$$\lim_{|x|\to+\infty} |x|\left(\frac{\partial G}{\partial |x|} - ikG\right) = 0,$$

uniformly in $x/|x|$.

The Green function G is given by

$$G(x,y,k) = -\frac{e^{ik|x-y|}}{4\pi|x-y|}, \quad x \neq y. \tag{3.1}$$

Define the following boundary integral operators and refer to [18,78] for their mapping properties:

$$\begin{aligned} \vec{\mathcal{S}}_D^k[\varphi] : H_T^{-\frac{1}{2}}(\partial D) &\longrightarrow H_T^{\frac{1}{2}}(\partial D) \text{ or } H^1_{\mathrm{loc}}(\mathbb{R}^3)^3 \\ \varphi &\longmapsto \vec{\mathcal{S}}_D^k[\varphi](x) = \int_{\partial D} G(x,y,k)\varphi(y)d\sigma(y), \quad x \in \partial D \text{ or } x \in \mathbb{R}^3; \end{aligned} \tag{3.2}$$

$$\begin{aligned} \mathcal{S}_D^k[\varphi] : H^{-\frac{1}{2}}(\partial D) &\longrightarrow H^{\frac{1}{2}}(\partial D) \text{ or } H^1_{\mathrm{loc}}(\mathbb{R}^3) \\ \varphi &\longmapsto \mathcal{S}_D^k[\varphi](x) = \int_{\partial D} G(x,y,k)\varphi(y)d\sigma(y), \quad x \in \partial D \text{ or } x \in \mathbb{R}^3; \end{aligned} \tag{3.3}$$

$$\begin{aligned} \mathcal{K}_D^*[\varphi] : H^{-\frac{1}{2}}(\partial D) &\longrightarrow H^{-\frac{1}{2}}(\partial D) \\ \varphi &\longmapsto \mathcal{K}_D^*[\varphi](x) = \int_{\partial D} \frac{\partial G(x,y,0)}{\partial \nu(x)}\varphi(y)d\sigma(y), \quad x \in \partial D; \end{aligned} \tag{3.4}$$

$$\begin{aligned} \mathcal{M}_D^k[\varphi] : H_T^{-\frac{1}{2}}(\mathrm{div}, \partial D) &\longrightarrow H_T^{-\frac{1}{2}}(\mathrm{div}, \partial D) \\ \varphi &\longmapsto \mathcal{M}_D^k[\varphi](x) = \int_{\partial D} \nu(x) \times \nabla_x \times G(x,y,k)\varphi(y)d\sigma(y), \quad x \in \partial D; \end{aligned} \tag{3.5}$$

$$\begin{aligned} \mathcal{L}_D^k[\varphi] : H_T^{-\frac{1}{2}}(\mathrm{div}, \partial D) &\longrightarrow H_T^{-\frac{1}{2}}(\mathrm{div}, \partial D) \\ \varphi &\longmapsto \mathcal{L}_D^k[\varphi](x) = \nu(x) \times \left(k^2 \vec{\mathcal{S}}_D^k[\varphi](x) + \nabla \mathcal{S}_D^k[\nabla_{\partial D} \cdot \varphi](x)\right), \quad x \in \partial D. \end{aligned} \tag{3.6}$$

Throughout this chapter, we denote $\vec{\mathcal{S}}_D^0, \mathcal{S}_D^0, \mathcal{M}_D^0$ by $\vec{\mathcal{S}}_D, \mathcal{S}_D, \mathcal{M}_D$, respectively. We also denote $\mathcal{K}_D$ by the $(\cdot,\cdot)_{-\frac{1}{2},\frac{1}{2}}$-adjoint of $\mathcal{K}_D^*$, where $(\cdot,\cdot)_{-\frac{1}{2},\frac{1}{2}}$ is the duality pairing between $H^{-\frac{1}{2}}(\partial D)$ and $H^{\frac{1}{2}}(\partial D)$.

We recall now some useful results on the operator $\mathcal{K}_D^*$. See chapter 1 and [12,32,61,65].

Lemma 3.2.1. *(i) The following Calderón identity holds: $\mathcal{K}_D\mathcal{S}_D = \mathcal{S}_D\mathcal{K}_D^*$;*

(ii) The operator $\mathcal{K}_D^*$ is compact self-adjoint in the Hilbert space $H^{-\frac{1}{2}}(\partial D)$ equipped with the following inner product

$$(u,v)_{\mathcal{H}^*} = -(u, \mathcal{S}_D[v])_{-\frac{1}{2},\frac{1}{2}}, \tag{3.7}$$

with which $\mathcal{H}^*(\partial D)$, the space induced by $(\cdot,\cdot)_{\mathcal{H}^*}$, is equivalent to $H^{-\frac{1}{2}}(\partial D)$;

(iii) Let (λ_j, φ_j), $j = 0, 1, 2, \ldots$ be the eigenvalue and normalized eigenfunction pair of $\mathcal{K}_D^*$ in $\mathcal{H}^*(\partial D)$. Then, $\lambda_j \in (-\frac{1}{2}, \frac{1}{2}]$, $\lambda_j \neq 1/2$ for $j \geq 1$, $\lambda_j \to 0$ as $j \to \infty$ and $\varphi_j \in \mathcal{H}_0^*(\partial D)$ for $j \geq 1$, where $\mathcal{H}_0^*(\partial D)$ is the zero mean subspace of $\mathcal{H}^*(\partial D)$;

(iv) The following representation formula holds: for any $\psi \in H^{-1/2}(\partial D)$,

$$\mathcal{K}_D^*[\psi] = \sum_{j=0}^{\infty} \lambda_j (\psi, \varphi_j)_{\mathcal{H}^*} \otimes \varphi_j;$$

(v) The following trace formula holds: for any $\psi \in \mathcal{H}^*(\partial D)$,

$$(\pm\frac{1}{2} Id + \mathcal{K}_D^*)[\varphi] = \frac{\partial \mathcal{S}_D[\varphi]}{\partial \nu}\Big|_{\pm}.$$

(vi) Let $\mathcal{H}(\partial D)$ be the space $H^{\frac{1}{2}}(\partial D)$ equipped with the following equivalent inner product

$$(u, v)_{\mathcal{H}} = -(\mathcal{S}_D^{-1}[u], v)_{-\frac{1}{2}, \frac{1}{2}}. \tag{3.8}$$

Then, $\mathcal{S}_D$ is an isometry between $\mathcal{H}^*(\partial D)$ and $\mathcal{H}(\partial D)$.

In *(vi)* in Lemma 3.2.1, we refer to [18] for the invertibility of the single-layer potential $\mathcal{S}_D$ in three dimensions.

The following result holds.

Lemma 3.2.2. *The following Helmholtz decomposition holds [38]:*

$$H_T^{-\frac{1}{2}}(\mathrm{div}, \partial D) = \nabla_{\partial D} H^{\frac{3}{2}}(\partial D) \oplus \vec{\mathrm{curl}}_{\partial D} H^{\frac{1}{2}}(\partial D).$$

Remark 3.2.1. *The Laplace-Beltrami operator $\Delta_{\partial D} : H_0^{\frac{3}{2}}(\partial D) \to H_0^{-\frac{1}{2}}(\partial D)$ is invertible. Here $H_0^{\frac{3}{2}}(\partial D)$ and $H_0^{-\frac{1}{2}}(\partial D)$ are the zero mean subspaces of $H^{\frac{3}{2}}(\partial D)$ and $H^{-\frac{1}{2}}(\partial D)$ respectively.*

The following results on the operator $\mathcal{M}_D$ are of great importance. We refer to [78] for a proof of the following compactness property of $\mathcal{M}_D$.

Lemma 3.2.3. *The operator $\mathcal{M}_D : H_T^{-\frac{1}{2}}(\mathrm{div}, \partial D) \longrightarrow H_T^{-\frac{1}{2}}(\mathrm{div}, \partial D)$ is a compact operator.*

Lemma 3.2.4. *The following identities hold [9, 53]:*

$$\begin{aligned} \mathcal{M}_D[\vec{\mathrm{curl}}_{\partial D}\varphi] &= \vec{\mathrm{curl}}_{\partial D}\mathcal{K}_D[\varphi], \quad \forall \varphi \in H^{\frac{1}{2}}(\partial D), \\ \mathcal{M}_D[\nabla_{\partial D}\varphi] &= -\nabla_{\partial D}\Delta_{\partial D}^{-1}\mathcal{K}_D^*[\Delta_{\partial D}\varphi] + \vec{\mathrm{curl}}_{\partial D}\mathcal{R}_D[\varphi], \quad \forall \varphi \in H^{\frac{3}{2}}(\partial D), \end{aligned}$$

where

$$\mathcal{R}_D = -\Delta_{\partial D}^{-1}\mathrm{curl}_{\partial D}\mathcal{M}_D\nabla_{\partial D}. \tag{3.9}$$

3.3 Layer potential formulation for the scattering problem

We consider the scattering problem of a time-harmonic electromagnetic wave incident on a plasmonic nanoparticle. The homogeneous medium is characterized by electric permittivity ε_m and magnetic permeability μ_m, while the particle occupying a bounded and simply connected domain $D \Subset \mathbb{R}^3$ of class $\mathcal{C}^{1,\alpha}$ for $0 < \alpha < 1$ is characterized by electric permittivity ε_c and magnetic permeability μ_c, both of which depend on the frequency. Define

$$k_m = \omega\sqrt{\varepsilon_m\mu_m}, \quad k_c = \omega\sqrt{\varepsilon_c\mu_c},$$

and

$$\varepsilon_D = \varepsilon_m\chi(\mathbb{R}^3\backslash\bar{D}) + \varepsilon_c\chi(D), \quad \mu_D = \varepsilon_m\chi(\mathbb{R}^3\backslash\bar{D}) + \varepsilon_c\chi(D),$$

where χ denotes the characteristic function.

For a given incident plane wave (E^i, H^i), solution to the Maxwell equations in free space

$$\begin{aligned} \nabla \times E^i &= i\omega\mu_m H^i \quad \text{in } \mathbb{R}^3, \\ \nabla \times H^i &= -i\omega\varepsilon_m E^i \quad \text{in } \mathbb{R}^3, \end{aligned}$$

the scattering problem can be modeled by the following system of equations

$$\begin{aligned} \nabla \times E &= i\omega\mu_D H \quad \text{in } \mathbb{R}^3\backslash\partial D, \\ \nabla \times H &= -i\omega\varepsilon_D E \quad \text{in } \mathbb{R}^3\backslash\partial D, \\ \nu \times E\big|_+ - \nu \times E\big|_- &= \nu \times H\big|_+ - \nu \times H\big|_- = 0 \quad \text{on } \partial D, \end{aligned} \tag{3.10}$$

subject to the Silver-Müller radiation condition:

$$\lim_{|x|\to\infty} |x|\big(\sqrt{\mu_m}(H - H^i)(x) \times \frac{x}{|x|} - \sqrt{\varepsilon_m}(E - E^i)(x)\big) = 0$$

uniformly in $x/|x|$. Here and throughout the chapter, the subscripts $\pm$ indicate, as said before, the limits from outside and inside D, respectively.

Using the boundary integral operators (3.2) and (3.5), the solution to (3.10) can be represented as [90]

$$E(x) = \begin{cases} E^i(x) + \mu_m\nabla \times \vec{\mathcal{S}}_D^{k_m}[\psi](x) + \nabla \times \nabla \times \vec{\mathcal{S}}_D^{k_m}[\phi](x), & x \in \mathbb{R}^3\backslash\bar{D}, \\ \mu_c\nabla \times \vec{\mathcal{S}}_D^{k_c}[\psi](x) + \nabla \times \nabla \times \vec{\mathcal{S}}_D^{k_c}[\phi](x), & x \in D, \end{cases} \tag{3.11}$$

and

$$H(x) = -\frac{i}{\omega\mu_D}(\nabla \times E)(x) \quad x \in \mathbb{R}^3\backslash\partial D, \tag{3.12}$$

where the pair $(\psi, \phi) \in \left(H_T^{-\frac{1}{2}}(\mathrm{div}, \partial D)\right)^2$ is the unique solution to

$$\left(\begin{array}{cc} \frac{\mu_c + \mu_m}{2} Id + \mu_c \mathcal{M}_D^{k_c} - \mu_m \mathcal{M}_D^{k_m} & \mathcal{L}_D^{k_c} - \mathcal{L}_D^{k_m} \\ \mathcal{L}_D^{k_c} - \mathcal{L}_D^{k_m} & \left(\frac{k_c^2}{2\mu_c} + \frac{k_m^2}{2\mu_m} \right) Id + \frac{k_c^2}{\mu_c} \mathcal{M}_D^{k_c} - \frac{k_m^2}{\mu_m} \mathcal{M}_D^{k_m} \end{array} \right) \left(\begin{array}{c} \psi \\ \phi \end{array} \right) = \left(\begin{array}{c} \nu \times E^i \\ i\omega \nu \times H^i \end{array} \right) \Bigg|_{\partial D}. \tag{3.13}$$

Let $D = z + \delta B$ where B contains the origin and $|B| = O(1)$. For any $x \in \partial D$, let $\widetilde{x} = \frac{x-z}{\delta} \in \partial B$ and define for each function f defined on ∂D, a corresponding function defined on B as follows

$$\eta(f)(\widetilde{x}) = f(z + \delta \widetilde{x}). \tag{3.14}$$

Throughout this chapter, for two Banach spaces X and Y, by $\mathcal{L}(X, Y)$ we denote the set of bounded linear operators from X into Y. We will also denote by $\mathcal{L}(X)$ the set $\mathcal{L}(X, X)$.

Lemma 3.3.1. *For $\varphi \in H_T^{-\frac{1}{2}}(\mathrm{div}, \partial D)$, the following asymptotic expansion holds*

$$\mathcal{M}_D^k[\varphi](x) = \mathcal{M}_B[\eta(\varphi)](\tilde{x}) + \sum_{j=2}^{\infty} \delta^j \mathcal{M}_{B,j}^k[\eta(\varphi)](\tilde{x}),$$

where

$$\mathcal{M}_{B,j}^k[\eta(\varphi)](\widetilde{x}) = \int_{\partial B} \frac{-(ik)^j}{4\pi j!} \nu(\tilde{x}) \times \nabla_{\tilde{x}} \times |\tilde{x} - \tilde{y}|^{j-1} \eta(\varphi)(\tilde{y}) d\sigma(\tilde{y}).$$

Moreover, $\|\mathcal{M}_{B,j}^k\|_{\mathcal{L}\left(H_T^{-\frac{1}{2}}(\mathrm{div}, \partial B)\right)}$ is uniformly bounded with respect to j. In particular, the convergence holds in $\mathcal{L}\left(H_T^{-\frac{1}{2}}(\mathrm{div}, \partial B)\right)$ and $\mathcal{M}_D^k$ is analytic in δ.

Proof. We can see, after a change of variables, that

$$\mathcal{M}_D^k[\varphi](x) = \int_{\partial B} \nu(\tilde{x}) \times \nabla_{\tilde{x}} \times G(\tilde{x}, \tilde{y}, \delta k) \eta(\varphi)(\tilde{y}) d\sigma(\tilde{y}).$$

A Taylor expansion of $G(\tilde{x}, \tilde{y}, \delta k)$ yields

$$G(\tilde{x}, \tilde{y}, \delta k) = -\sum_{j=0}^{\infty} \frac{(i\delta k |\tilde{x} - \tilde{y}|)^j}{j! 4\pi |\tilde{x} - \tilde{y}|} = -\frac{1}{4\pi |\tilde{x} - \tilde{y}|} + \sum_{j=1}^{\infty} \delta^j \frac{(ik)^j}{4\pi j!} |\tilde{x} - \tilde{y}|^{j-1}.$$

Hence,

$$\mathcal{M}_D^k[\varphi](x) = \mathcal{M}_B[\eta(\varphi)](\tilde{x}) + \sum_{j=2}^{\infty} \delta^j \int_{\partial B} \frac{-(ik)^j}{4\pi j!} \nu(\tilde{x}) \times \nabla_{\tilde{x}} \times |\tilde{x} - \tilde{y}|^{j-1} \eta(\varphi)(\tilde{y}) d\sigma(\tilde{y}).$$

Note that from the regularity of $|\tilde{x} - \tilde{y}|^{j-1}$, $j \geq 2$, $\|\mathcal{M}_{B,j}^k[\eta(\varphi)]\|_{H_T^{-\frac{1}{2}}(\mathrm{div}, \partial B)}$ is uniformly bounded with respect to j, and therefore, $\|\mathcal{M}_{B,j}^k\|_{\mathcal{L}\left(H_T^{-\frac{1}{2}}(\mathrm{div}, \partial B)\right)}$

is uniformly bounded with respect to j as well. □

Lemma 3.3.2. *For $\varphi \in H_T^{-\frac{1}{2}}(\mathrm{div}, \partial D)$, the following asymptotic expansion holds*

$$(\mathcal{L}_D^{k_c} - \mathcal{L}_D^{k_m})[\varphi](x) = \sum_{j=1}^{\infty} \delta^j \omega \mathcal{L}_{B,j}[\eta(\varphi)](\tilde{x}),$$

where

$$\mathcal{L}_{B,j}[\eta(\varphi)](\tilde{x}) =$$
$$C_j \nu(\tilde{x}) \times \Big(\int_{\partial B} |\tilde{x} - \tilde{y}|^{j-2} \eta(\varphi)(\tilde{y}) d\sigma(\tilde{y}) - \int_{\partial B} \frac{|\tilde{x} - \tilde{y}|^{j-2}(\tilde{x} - \tilde{y})}{j+1} \nabla_{\partial B} \cdot \eta(\varphi)(\tilde{y}) d\sigma(\tilde{y}) \Big),$$

and

$$C_j = \frac{i^j (k_c^{j+1} - k_m^{j+1})}{\omega 4\pi (j-1)!}.$$

Moreover, $\|\mathcal{L}_{B,j}\|_{\mathcal{L}\left(H_T^{-\frac{1}{2}}(\mathrm{div}, \partial B)\right)}$ is uniformly bounded with respect to j. In particular, the convergence holds in $\mathcal{L}\left(H_T^{-\frac{1}{2}}(\mathrm{div}, \partial B)\right)$ and $\mathcal{L}_D^k$ is analytic in δ.

Proof. The proof is similar to that of Lemma 3.3.1. □

Using Lemma 3.3.1 and Lemma 3.3.2, we can write the system of equations (3.13) as follows:

$$\mathcal{W}_B(\delta) \begin{pmatrix} \eta(\psi) \\ \omega \eta(\phi) \end{pmatrix} = \begin{pmatrix} \dfrac{\eta(\nu \times E^i)}{\mu_m - \mu_c} \\ \dfrac{\eta(i\nu \times H^i)}{\varepsilon_m - \varepsilon_c} \end{pmatrix} \Bigg|_{\partial B}, \tag{3.15}$$

where

$$\mathcal{W}_B(\delta) = \begin{pmatrix} \lambda_\mu Id - \mathcal{M}_B + \delta^2 \dfrac{\mu_m \mathcal{M}_{B,2}^{k_m} - \mu_c \mathcal{M}_{B,2}^{k_c}}{\mu_m - \mu_c} & \dfrac{1}{\mu_m - \mu_c}(\delta \mathcal{L}_{B,1} + \delta^2 \mathcal{L}_{B,2}) \\ \dfrac{1}{\varepsilon_m - \varepsilon_c}(\delta \mathcal{L}_{B,1} + \delta^2 \mathcal{L}_{B,2}) & \lambda_\varepsilon Id - \mathcal{M}_B + \delta^2 \dfrac{\varepsilon_m \mathcal{M}_{B,2}^{k_m} - \varepsilon_c \mathcal{M}_{B,2}^{k_c}}{\varepsilon_m - \varepsilon_c} \end{pmatrix} + O(\delta^3), \tag{3.16}$$

and the material parameter contrasts λ_μ and λ_ε are given by

$$\lambda_\mu = \frac{\mu_c + \mu_m}{2(\mu_m - \mu_c)}, \quad \lambda_\varepsilon = \frac{\varepsilon_c + \varepsilon_m}{2(\varepsilon_m - \varepsilon_c)}. \tag{3.17}$$

It is clear that

$$\mathcal{W}_B(0) = \mathcal{W}_{B,0} = \begin{pmatrix} \lambda_\mu Id - \mathcal{M}_B & 0 \\ 0 & \lambda_\varepsilon Id - \mathcal{M}_B \end{pmatrix}.$$

Moreover,

$$\mathcal{W}_B(\delta) = \mathcal{W}_{B,0} + \delta \mathcal{W}_{B,1} + \delta^2 \mathcal{W}_{B,2} + O(\delta^3),$$

in the sense that

$$\|\mathcal{W}_B(\delta) - \mathcal{W}_{B,0} - \delta\mathcal{W}_{B,1} - \delta^2\mathcal{W}_{B,2}\| \leq C\delta^3$$

for a constant C independent of δ. Here $\|A\| = \sup_{i,j} \|A_{i,j}\|_{H_T^{-\frac{1}{2}}(\mathrm{div},\partial B)}$ for any operator-valued matrix A with entries $A_{i,j}$.
We are now interested in finding $\mathcal{W}_B^{-1}(\delta)$. For this purpose, we first consider solving the problem

$$(\lambda Id - \mathcal{M}_B)[\psi] = \varphi \tag{3.18}$$

for $(\psi, \varphi) \in \left(H_T^{-\frac{1}{2}}(\mathrm{div}, \partial B)\right)^2$ and $\lambda \notin \sigma(\mathcal{M}_B)$, where $\sigma(\mathcal{M}_B)$ is the spectrum of $\mathcal{M}_B$.
Using the Helmholtz decomposition of $H_T^{-\frac{1}{2}}(\mathrm{div}, \partial B)$ in Lemma 3.2.2, we can reduce (3.18) to an equivalent system of equations involving some well known operators.

Definition 3.1. *For $u \in H_T^{-\frac{1}{2}}(\mathrm{div}, \partial B)$, we denote by $u^{(1)}$ and $u^{(2)}$ any two functions in $H_0^{\frac{3}{2}}(\partial B)$ and $H^{\frac{1}{2}}(\partial B)$, respectively, such that*

$$u = \nabla_{\partial B} u^{(1)} + \vec{\mathrm{curl}}_{\partial B} u^{(2)}.$$

Note that $u^{(1)}$ is uniquely defined and $u^{(2)}$ is defined up to a constant function.

Lemma 3.3.3. *Assume $\lambda \neq \frac{1}{2}$, then problem (3.18) is equivalent to*

$$(\lambda Id - \widetilde{\mathcal{M}}_B) \begin{pmatrix} \psi^{(1)} \\ \psi^{(2)} \end{pmatrix} = \begin{pmatrix} \varphi^{(1)} \\ \varphi^{(2)} \end{pmatrix}, \tag{3.19}$$

where $(\varphi^{(1)}, \varphi^{(2)}) \in H_0^{\frac{3}{2}}(\partial B) \times H^{\frac{1}{2}}(\partial B)$ and

$$\widetilde{\mathcal{M}}_B = \begin{pmatrix} -\Delta_{\partial B}^{-1} \mathcal{K}_B^* \Delta_{\partial B} & 0 \\ \mathcal{R}_B & \mathcal{K}_B \end{pmatrix}.$$

Here, $\mathcal{R}_B$ is defined by (3.9) with D replaced with B.

Proof. Let $(\psi^{(1)}, \psi^{(2)}) \in H_0^{\frac{3}{2}}(\partial B) \times H^{\frac{1}{2}}(\partial B)$ be a solution (if there is any) to (3.19) where $(\varphi^{(1)}, \varphi^{(2)}) \in H_0^{\frac{3}{2}}(\partial B) \times H^{\frac{1}{2}}(\partial B)$ satisfies

$$\varphi = \nabla_{\partial B} \varphi^{(1)} + \vec{\mathrm{curl}}_{\partial B} \varphi^{(2)}.$$

We have

$$\left(\lambda Id + \Delta_{\partial B}^{-1} \mathcal{K}_B^* \Delta_{\partial B}\right)[\psi^{(1)}] = \varphi^{(1)}, \tag{3.20}$$

$$\lambda\psi^{(2)} - \mathcal{R}_B[\psi^{(1)}] - \mathcal{K}_B[\psi^{(2)}] = \varphi^{(2)}. \tag{3.21}$$

Taking $\nabla_{\partial B}$ in (3.20), $\vec{\mathrm{curl}}_{\partial B}$ in (3.21), adding up and using the identities of Lemma 3.2.4 yields

$$(\lambda Id - \mathcal{M}_B)\left[\nabla_{\partial B}\psi^{(1)} + \vec{\mathrm{curl}}_{\partial B}\psi^{(2)}\right] = \nabla_{\partial B}\varphi^{(1)} + \vec{\mathrm{curl}}_{\partial B}\varphi^{(2)}.$$

Therefore

$$\psi = \nabla_{\partial B}\psi^{(1)} + \vec{\text{curl}}_{\partial B}\psi^{(2)},$$

is a solution of (3.18).
Conversely, let ψ be the solution to (3.18). There exist $(\psi^{(1)}, \psi^{(2)}) \in H_0^{\frac{3}{2}}(\partial B) \times H^{\frac{1}{2}}(\partial B)$ and $(\varphi^{(1)}, \varphi^{(2)}) \in H_0^{\frac{3}{2}}(\partial B) \times H^{\frac{1}{2}}(\partial B)$ such that

$$\begin{aligned} \psi &= \nabla_{\partial B}\psi^{(1)} + \vec{\text{curl}}_{\partial B}\psi^{(2)}, \\ \varphi &= \nabla_{\partial B}\varphi^{(1)} + \vec{\text{curl}}_{\partial B}\varphi^{(2)}, \end{aligned}$$

and we have

$$(\lambda Id - \mathcal{M}_B)\left[\nabla_{\partial B}\psi^{(1)} + \vec{\text{curl}}_{\partial B}\psi^{(2)}\right] = \nabla_{\partial B}\varphi^{(1)} + \vec{\text{curl}}_{\partial B}\varphi^{(2)}. \tag{3.22}$$

Taking $\nabla_{\partial B}\cdot$ in the above equation and using the identities of Lemma 3.2.4 yields

$$\Delta_{\partial B}\big(\lambda Id + \Delta_{\partial B}^{-1}\mathcal{K}_B^*\Delta_{\partial B}\big)[\psi^{(1)}] = \Delta_{\partial B}\varphi^{(1)}.$$

Since $(\psi^{(1)}, \varphi^{(1)}) \in (H_0^{\frac{3}{2}}(\partial B))^2$ we get

$$\big(\lambda Id + \Delta_{\partial B}^{-1}\mathcal{K}_B^*\Delta_{\partial B}\big)[\psi^{(1)}] = \varphi^{(1)}.$$

Taking $\text{curl}_{\partial B}$ in (3.22) and using the identities of Lemma 3.2.4 yields

$$\Delta_{\partial B}(\lambda\psi^{(2)} - \mathcal{R}_B[\psi^{(1)}] - \mathcal{K}_B[\psi^{(2)}]) = \Delta_{\partial B}\varphi^{(2)}.$$

Therefore, there exists a constant c such that

$$\lambda\psi^{(2)} - \mathcal{R}_B[\psi^{(1)}] - \mathcal{K}_B[\psi^{(2)}] = \varphi^{(2)} + c\chi(\partial B).$$

Since $\mathcal{K}_B(\chi(\partial B)) = \dfrac{1}{2}\chi(\partial B)$ we have

$$\lambda\Big(\psi^{(2)} - \frac{c}{\lambda - 1/2}\Big) - \mathcal{R}_B[\psi^{(1)}] - \mathcal{K}_B\Big[\psi^{(2)} - \frac{c}{\lambda - 1/2}\Big] = \varphi^{(2)}.$$

Hence, $\Big(\psi^{(1)}, \psi^{(2)} - \dfrac{c}{\lambda - 1/2}\Big) \in H_0^{\frac{3}{2}}(\partial B) \times H^{\frac{1}{2}}(\partial B)$ is a solution to (3.19) □

Let us now analyze the spectral properties of $\widetilde{\mathcal{M}}_B$ in

$$H(\partial B) := H_0^{\frac{3}{2}}(\partial B) \times H^{\frac{1}{2}}(\partial B), \tag{3.23}$$

equipped with the inner product

$$(u, v)_{H(\partial B)} = (\Delta_{\partial B}u^{(1)}, \Delta_{\partial B}v^{(1)})_{\mathcal{H}^*} + (u^{(2)}, v^{(2)})_{\mathcal{H}},$$

which is equivalent to $H_0^{\frac{3}{2}}(\partial B) \times H^{\frac{1}{2}}(\partial B)$.
By abuse of notation we call $u^{(1)}$ and $u^{(2)}$ the first and second components of any $u \in H(\partial B)$.
We will assume for simplicity the following condition.

Condition 3.1. *The eigenvalues of $\mathcal{K}_B^*$ are simple.*

Recall that $\mathcal{K}_B^*$ and $\mathcal{K}_B$ are compact and self-adjoint in $\mathcal{H}^*(\partial B)$ and $\mathcal{H}(\partial B)$, respectively. Since $\mathcal{K}_B$ is the $(\cdot,\cdot)_{-\frac{1}{2},\frac{1}{2}}$ adjoint of $\mathcal{K}_B^*$, we have $\sigma(\mathcal{K}_B) = \sigma(\mathcal{K}_B^*)$, where $\sigma(\mathcal{K}_B)$ (resp. $\sigma(\mathcal{K}_B^*)$) is the (discrete) spectrum of $\mathcal{K}_B$ (resp. $\mathcal{K}_B^*$).
Define

$$\begin{aligned} \sigma_1 &= \sigma(-\mathcal{K}_B^*)\backslash\Big(\sigma(\mathcal{K}_B)\cup\{-\frac{1}{2}\}\Big), \\ \sigma_2 &= \sigma(\mathcal{K}_B)\backslash\sigma(-\mathcal{K}_B^*), \\ \sigma_3 &= \sigma(-\mathcal{K}_B^*)\cap\sigma(\mathcal{K}_B). \end{aligned} \tag{3.24}$$

Let $\lambda_{j,1}\in\sigma_1$, $j=1,2\ldots$ and let $\varphi_{j,1}$ be an associated normalized eigenfunction of $\mathcal{K}_B^*$ as defined in Lemma 3.2.1. Note that $\varphi_{j,1}\in H_0^{-\frac{1}{2}}(\partial B)$ for $j\geq 1$. Then,

$$\psi_{j,1} = \begin{pmatrix} \Delta_{\partial B}^{-1}\varphi_{j,1} \\ (\lambda_{j,1}Id-\mathcal{K}_B)^{-1}\mathcal{R}_B[\Delta_{\partial B}^{-1}\varphi_{j,1}] \end{pmatrix}$$

satisfies

$$\widetilde{\mathcal{M}}_B[\psi_{j,1}] = \lambda_{j,1}\psi_{j,1}.$$

Let $\lambda_{j,2}\in\sigma_2$ and let $\varphi_{j,2}$ be an associated normalized eigenfunction of $\mathcal{K}_B$. Then,

$$\psi_{j,2} = \begin{pmatrix} 0 \\ \varphi_{j,2} \end{pmatrix}$$

satisfies

$$\widetilde{\mathcal{M}}_B[\psi_{j,2}] = \lambda_{j,2}\psi_{j,2}.$$

Now, assume that Condition 3.1 holds. Let $\lambda_{j,3}\in\sigma_3$, let $\varphi_{j,3}^{(1)}$ be the associated normalized eigenfunction of $\mathcal{K}_B^*$ and let $\varphi_{j,3}^{(2)}$ be the associated normalized eigenfunction of $\mathcal{K}_B$. Then,

$$\psi_{j,3} = \begin{pmatrix} 0 \\ \varphi_{j,3}^{(2)} \end{pmatrix}$$

satisfies

$$\widetilde{\mathcal{M}}_B[\psi_{j,3}] = \lambda_{j,3}\psi_{j,3},$$

and $\lambda_{j,3}$ has a first-order generalized eigenfunction given by

$$\psi_{j,3,g} = \begin{pmatrix} c\Delta_{\partial B}^{-1}\varphi_{j,3}^{(1)} \\ (\lambda_{j,3}Id-\mathcal{K}_B)^{-1}\mathcal{P}_{\mathrm{span}\{\varphi_{j,3}^{(2)}\}^\perp}\mathcal{R}_B[c\Delta_{\partial B}^{-1}\varphi_{j,3}^{(1)}] \end{pmatrix} \tag{3.25}$$

for a constant c such that $\mathcal{P}_{\mathrm{span}\{\varphi_{j,3}^{(2)}\}}\mathcal{R}_B[c\Delta_{\partial B}^{-1}\varphi_{j,3}^{(1)}] = -\varphi_{j,3}^{(2)}$. Here, $\mathrm{span}\{\varphi_{j,3}^{(2)}\}$ is the vector space spanned by $\varphi_{j,3}^{(2)}$, $\mathrm{span}\{\varphi_{j,3}^{(2)}\}^\perp$ is the orthogonal space to

$\mathrm{span}\{\varphi^{(2)}_{j,3}\}$ in $\mathcal{H}(\partial B)$ (Lemma 3.2.1), and $\mathcal{P}_{\mathrm{span}\{\varphi^{(2)}_{j,3}\}}$ (resp. $\mathcal{P}_{\mathrm{span}\{\varphi^{(2)}_{j,3}\}^{\perp}}$ is the orthogonal (in $\mathcal{H}(\partial B)$) projection on $\mathrm{span}\{\varphi^{(2)}_{j,3}\}$ (resp. $\mathrm{span}\{\varphi^{(2)}_{j,3}\}^{\perp}$).

We remark that the function $\psi_{j,3,g}$ is determined by the following equation

$$\widetilde{\mathcal{M}}_B[\psi_{j,3,g}] = \lambda_{j,3}\psi_{j,3,g} + \psi_{j,3}.$$

Consequently, the following result holds.

Propsition 3.3.1. *The spectrum* $\sigma(\widetilde{\mathcal{M}}_B) = \sigma_1 \cup \sigma_2 \cup \sigma_3 = \sigma(-\mathcal{K}^*_B) \cup \sigma(\mathcal{K}^*_B)\backslash\{-\frac{1}{2}\}$ *in* $H(\partial B)$. *Moreover, under Condition 3.1,* $\widetilde{\mathcal{M}}_B$ *has eigenfunctions* $\psi_{j,i}$ *associated to the eigenvalues* $\lambda_{j,i} \in \sigma_i$ *for* $j = 1, 2, \dots$ *and* $i = 1, 2, 3$, *and generalized eigenfunctions of order one* $\psi_{j,3,g}$ *associated to* $\lambda_{j,3} \in \sigma_3$, *all of which form a non-orthogonal basis of* $H(\partial B)$ *(defined by (3.23)).*

Proof. It is clear that $\lambda - \widetilde{\mathcal{M}}_B$ is bijective if and only if $\lambda \notin \sigma(-\mathcal{K}^*_B) \cup \sigma(\mathcal{K}^*_B) \setminus \{-\frac{1}{2}\}$.
It is only left to show that $\psi_{j,1}, \psi_{j,2}, \psi_{j,3}, \psi_{j,3,g}$, $j = 1, 2, \dots$ form a non-orthogonal basis of $H(\partial B)$.
Indeed, let

$$\psi = \begin{pmatrix} \psi^{(1)} \\ \psi^{(2)} \end{pmatrix} \in H(\partial B).$$

Since $\psi^{(1)}_{j,1} \cup \psi^{(1)}_{j,3,g}$, $j = 1, 2, \dots$ form an orthogonal basis of $\mathcal{H}^*_0(\partial B)$, which is equivalent to $H_0^{-\frac{1}{2}}(\partial B)$, there exist $\alpha_\kappa, \kappa \in I_1 := \{(j,1) \cup (j,3,g) : j = 1, 2, \dots\}$ such that

$$\psi^{(1)} = \sum_{\kappa \in I_1} \alpha_\kappa \Delta^{-1}_{\partial B} \psi^{(1)}_\kappa,$$

and

$$\sum_{\kappa \in I_1} |\alpha_\kappa|^2 \leq \infty.$$

It is clear that $\|\psi^{(2)}_\kappa\|_{H^{\frac{1}{2}}(\partial B)}$ is uniformly bounded with respect to $\kappa \in I_1$. Then

$$h := \sum_{\kappa \in I_1} \alpha_\kappa \psi^{(2)}_\kappa \in H^{\frac{1}{2}}(\partial B).$$

Since $\psi^{(2)}_{j,2} \cup \psi^{(2)}_{j,3}$, $j = 1, 2, \dots$ form an orthogonal basis of $\mathcal{H}(\partial B)$, which is equivalent to $H^{\frac{1}{2}}(\partial B)$, there exist $\alpha_\kappa, \kappa \in I_2 := \{(j,2) \cup (j,3) : j = 1, 2, \dots\}$ such that

$$\psi^{(2)} - h = \sum_{\kappa \in I_2} \alpha_\kappa \psi^{(2)}_\kappa,$$

and

$$\sum_{\kappa\in I_2} |\alpha_\kappa|^2 \leq \infty.$$

Hence, there exist $\alpha_\kappa, \kappa \in I_1 \cup I_2$ such that

$$\psi = \sum_{\kappa\in I_1\cup I_2} \alpha_\kappa \psi_\kappa,$$

and

$$\sum_{\kappa\in I_1\cup I_2} |\alpha_\kappa|^2 \leq \infty.$$

□

To have the compactness of $\widetilde{\mathcal{M}}_B$, we need the following condition.

Condition 3.2. *σ_3 is finite.*

Indeed, if σ_3 is not finite we have $\widetilde{\mathcal{M}}_B(\{\psi_{j,3,g};\quad j \geq 1\}) = \{\lambda_{j,3}\psi_{j,g,3} + \psi_{j,3};\quad j \geq 1\}$ whose adherence is not compact. However, if σ_3 is finite, using Proposition 3.3.1 we can approximate $\widetilde{\mathcal{M}}_B$ by a sequence of finite-rank operators.
Throughout this chapter, we assume that Condition 3.2 holds, even though an analysis can still be done for the case where σ_3 is infinite; see section 3.6.

Definition 3.2. *Let $\mathcal{B}$ be the basis of $H(\partial B)$ formed by the eigenfunctions and generalized eigenfunctions of $\widetilde{\mathcal{M}}_B$ as stated in Lemma 3.3.1. For $\psi \in H(\partial B)$, we denote by $\alpha(\psi, \psi_\kappa)$ the projection of ψ into $\psi_\kappa \in \mathcal{B}$ such that*

$$\psi = \sum_\kappa \alpha(\psi, \psi_\kappa)\psi_\kappa.$$

The following lemma follows from the Fredholm alternative.

Lemma 3.3.4. *Let*

$$\psi = \begin{pmatrix} \psi^{(1)} \\ \psi^{(2)} \end{pmatrix} \in H(\partial B).$$

Then,

$$\alpha(\psi, \psi_\kappa) = \begin{cases} \dfrac{(\psi, \widetilde{\psi}_\kappa)_{H(\partial B)}}{(\psi_\kappa, \widetilde{\psi}_\kappa)_{H(\partial B)}}, & \kappa = (j,i),\ i = 1,2, \\ \dfrac{(\psi, \widetilde{\psi}_{\kappa'})_{H(\partial B)}}{(\psi_\kappa, \widetilde{\psi}_{\kappa'})_{H(\partial B)}}, & \kappa = (j,3,g), \kappa' = (j,3), \\ \dfrac{(\psi, \widetilde{\psi}_{\kappa_g})_{H(\partial B)} - \alpha(\psi, \psi_{\kappa_g})(\psi_{\kappa_g}, \widetilde{\psi}_{\kappa_g})_{H(\partial B)}}{(\psi_\kappa, \widetilde{\psi}_{\kappa_g})_{H(\partial B)}}, & \kappa = (j,3), \kappa_g = (j,3,g), \end{cases}$$

where $\widetilde{\psi}_\kappa \in \mathrm{Ker}(\bar{\lambda}_\kappa - \widetilde{\mathcal{M}}_B^*)$ for $\kappa = (j,i)$, $i = 1,2,3$; $\widetilde{\psi}_\kappa \in \mathrm{Ker}(\bar{\lambda}_\kappa - \widetilde{\mathcal{M}}_B^*)^2$ for $\kappa = (j,3,g)$ and $\widetilde{\mathcal{M}}_B^*$ is the $H(\partial B)$-adjoint of $\widetilde{\mathcal{M}}_B$.

The following remark is in order.

Remark 3.3.1. *Note that, since $\varphi_{j,1}$ and $\varphi^{(1)}_{j,3}$ form an orthogonal basis of $\mathcal{H}^*_0(\partial B)$, equivalent to $H_0^{-\frac{1}{2}}(\partial B)$, we also have*

$$\alpha(\psi,\psi_\kappa)=\begin{cases}(\Delta_{\partial B}\psi^{(1)},\varphi_{j,1})_{\mathcal{H}^*}, & \kappa=(j,1),\\ \frac{1}{c}(\Delta_{\partial B}\psi^{(1)},\varphi^{(1)}_{j,3})_{\mathcal{H}^*}, & \kappa=(j,3,g),\end{cases}$$

where c is defined in (3.25).

Remark 3.3.2. *For $i=1,2,3$, and $j=1,2,\ldots,$*

$$\begin{aligned}(\lambda Id-\widetilde{\mathcal{M}}_B)^{-1}[\psi_{j,i}] &= \frac{\psi_{j,i}}{\lambda-\lambda_{j,i}},\\ (\lambda Id-\widetilde{\mathcal{M}}_B)^{-1}[\psi_{j,3,g}] &= \frac{\psi_{j,3,g}}{\lambda-\lambda_{j,3}}+\frac{\psi_{j,3}}{(\lambda-\lambda_{j,3})^2}.\end{aligned}$$

Now we turn to the original equation (3.13). The following result holds.

Lemma 3.3.5. *The system of equations (3.13) is equivalent to*

$$W_B(\delta)\begin{pmatrix}\eta(\psi)^{(1)}\\ \eta(\psi)^{(2)}\\ \omega\eta(\phi)^{(1)}\\ \omega\eta(\phi)^{(2)}\end{pmatrix}=\left.\begin{pmatrix}\frac{\eta(\nu\times E^i)^{(1)}}{\mu_m-\mu_c}\\ \frac{\eta(\nu\times E^i)^{(2)}}{\mu_m-\mu_c}\\ \frac{\eta(i\nu\times H^i)^{(1)}}{\varepsilon_m-\varepsilon_c}\\ \frac{\eta(i\nu\times H^i)^{(2)}}{\varepsilon_m-\varepsilon_c}\end{pmatrix}\right|_{\partial B},\tag{3.26}$$

where

$$W_B(\delta) = W_{B,0}+\delta W_{B,1}+\delta^2 W_{B,2}+O(\delta^3)$$

with

$$\begin{aligned}W_{B,0} &= \begin{pmatrix}\lambda_\mu Id-\widetilde{\mathcal{M}}_B & O\\ O & \lambda_\varepsilon Id-\widetilde{\mathcal{M}}_B\end{pmatrix},\\ W_{B,1} &= \begin{pmatrix}O & \frac{1}{\mu_m-\mu_c}\widetilde{\mathcal{L}}_{B,1}\\ \frac{1}{\varepsilon_m-\varepsilon_c}\widetilde{\mathcal{L}}_{B,1} & O\end{pmatrix},\\ W_{B,2} &= \begin{pmatrix}\frac{1}{\mu_m-\mu_c}\widetilde{\mathcal{M}}^{\mu}_{B,2} & \frac{1}{\mu_m-\mu_c}\widetilde{\mathcal{L}}_{B,2}\\ \frac{1}{\varepsilon_m-\varepsilon_c}\widetilde{\mathcal{L}}_{B,2} & \frac{1}{\varepsilon_m-\varepsilon_c}\widetilde{\mathcal{M}}^{\varepsilon}_{B,2}\end{pmatrix},\end{aligned}$$

and

$$\widetilde{\mathcal{M}}_B=\begin{pmatrix}-\Delta^{-1}_{\partial B}\mathcal{K}^*_B\Delta_{\partial B} & 0\\ \mathcal{R}_B & \mathcal{K}_B\end{pmatrix},$$

$$\widetilde{\mathcal{M}}^{\mu}_{B,2}=\begin{pmatrix}\Delta^{-1}_{\partial B}\nabla_{\partial B}\cdot(\mu_m\mathcal{M}^{k_m}_{B,2}-\mu_c\mathcal{M}^{k_c}_{B,2})\nabla_{\partial B} & \Delta^{-1}_{\partial B}\nabla_{\partial B}\cdot(\mu_m\mathcal{M}^{k_m}_{B,2}-\mu_c\mathcal{M}^{k_c}_{B,2})\vec{\mathrm{curl}}_{\partial B}\\ -\Delta^{-1}_{\partial B}\mathrm{curl}_{\partial B}(\mu_m\mathcal{M}^{k_m}_{B,2}-\mu_c\mathcal{M}^{k_c}_{B,2})\nabla_{\partial B} & -\Delta^{-1}_{\partial B}\mathrm{curl}_{\partial B}(\mu_m\mathcal{M}^{k_m}_{B,2}-\mu_c\mathcal{M}^{k_c}_{B,2})\vec{\mathrm{curl}}_{\partial B}\end{pmatrix},$$

$$\widetilde{\mathcal{M}}^{\varepsilon}_{B,2} = \begin{pmatrix} \Delta^{-1}_{\partial B}\nabla_{\partial B}\cdot(\varepsilon_m\mathcal{M}^{k_m}_{B,2} - \varepsilon_c\mathcal{M}^{k_c}_{B,2})\nabla_{\partial B} & \Delta^{-1}_{\partial B}\nabla_{\partial B}\cdot(\varepsilon_m\mathcal{M}^{k_m}_{B,2} - \varepsilon_c\mathcal{M}^{k_c}_{B,2})\vec{\text{curl}}_{\partial B} \\ -\Delta^{-1}_{\partial B}\text{curl}_{\partial B}(\varepsilon_m\mathcal{M}^{k_m}_{B,2} - \varepsilon_c\mathcal{M}^{k_c}_{B,2})\nabla_{\partial B} & -\Delta^{-1}_{\partial B}\text{curl}_{\partial B}(\varepsilon_m\mathcal{M}^{k_m}_{B,2} - \varepsilon_c\mathcal{M}^{k_c}_{B,2})\vec{\text{curl}}_{\partial B} \end{pmatrix},$$

$$\widetilde{\mathcal{L}}_{B,s} = \begin{pmatrix} \Delta^{-1}_{\partial B}\nabla_{\partial B}\cdot\mathcal{L}_{B,s}\nabla_{\partial B} & \Delta^{-1}_{\partial B}\nabla_{\partial B}\cdot\mathcal{L}_{B,s}\vec{\text{curl}}_{\partial B} \\ -\Delta^{-1}_{\partial B}\text{curl}_{\partial B}\mathcal{L}_{B,s}\nabla_{\partial B} & -\Delta^{-1}_{\partial B}\text{curl}_{\partial B}\mathcal{L}_{B,s}\vec{\text{curl}}_{\partial B} \end{pmatrix},$$

for $s = 1, 2$.
Moreover, the eigenfunctions of $W_{B,0}$ *in* $H(\partial B)^2$ *are given by*

$$\begin{aligned} \Psi_{1,j,i} &= \begin{pmatrix} \psi_{j,i} \\ O \end{pmatrix}, \quad j = 0, 1, 2, \ldots; i = 1, 2, 3, \\ \Psi_{2,j,i} &= \begin{pmatrix} O \\ \psi_{j,i} \end{pmatrix}, \quad j = 0, 1, 2, \ldots; i = 1, 2, 3, \end{aligned}$$

associated to the eigenvalues $\lambda_\mu - \lambda_{j,i}$ *and* $\lambda_\varepsilon - \lambda_{j,i}$*, respectively, and generalized eigenfunctions of order one*

$$\begin{aligned} \Psi_{1,j,3,g} &= \begin{pmatrix} \psi_{j,3,g} \\ O \end{pmatrix}, \\ \Psi_{2,j,3,g} &= \begin{pmatrix} O \\ \psi_{j,3,g} \end{pmatrix}, \end{aligned}$$

associated to eigenvalues $\lambda_\mu - \lambda_{j,3}$ *and* $\lambda_\varepsilon - \lambda_{j,3}$*, respectively, all of which form a non-orthogonal basis of* $H(\partial B)^2$.

Proof. The proof follows directly from Lemmas 3.3.1 and 3.3.3. □

We regard the operator $W_B(\delta)$ as a perturbation of the operator $W_{B,0}$ for small δ. Using perturbation theory, we can derive the perturbed eigenvalues and their associated eigenfunctions in $H(\partial B)^2$.
We denote by $\Gamma = \{(k, j, i) : k = 1, 2; j = 1, 2, \ldots; i = 1, 2, 3\}$ the set of indices for the eigenfunctions of $W_{B,0}$ and by $\Gamma_g = \{(k, j, 3, g) : k = 1, 2; j = 1, 2, \ldots\}$ the set of indices for the generalized eigenfunctions. We denote by γ_g the generalized eigenfunction index corresponding to eigenfunction index γ and vice-versa. We also denote by

$$\tau_\gamma = \begin{cases} \lambda_\mu - \lambda_{j,i}, & k = 1, \\ \lambda_\varepsilon - \lambda_{j,i}, & k = 2. \end{cases} \tag{3.27}$$

Condition 3.3. $\lambda_\mu \neq \lambda_\varepsilon$.

In the following we will only consider $\gamma \in \Gamma$ with which there is no generalized eigenfunction index associated. In other words, we only consider $\gamma = (k, i, j) \in \Gamma$ such that $\lambda_{j,i} \in \sigma_1 \cup \sigma_2$ (see (3.24) for the definitions). We call this subset Γ_{sim}. Note that Conditions 3.1 and 3.3 imply that the eigenvalues of $W_{B,0}$ indexed by $\gamma \in \Gamma_{\text{sim}}$ are simple.

Theorem 3.3.1. *As* $\delta \to$*, the perturbed eigenvalues and eigenfunctions indexed by* $\gamma \in \Gamma_{\text{sim}}$ *have the following asymptotic expansions:*

$$\begin{aligned} \tau_\gamma(\delta) &= \tau_\gamma + \delta\tau_{\gamma,1} + \delta^2\tau_{\gamma,2} + O(\delta^3), \\ \Psi_\gamma(\delta) &= \Psi_\gamma + \delta\Psi_{\gamma,1} + O(\delta^2), \end{aligned} \tag{3.28}$$

where

$$\begin{aligned}
\tau_{\gamma,1} &= \frac{(W_{B,1}\Psi_\gamma, \widetilde{\Psi}_\gamma)_{H(\partial B)^2}}{(\Psi_\gamma, \widetilde{\Psi}_\gamma)_{H(\partial B)^2}} = 0, \\
\tau_{\gamma,2} &= \frac{(W_{B,2}\Psi_\gamma, \widetilde{\Psi}_\gamma)_{H(\partial B)^2} - (W_{B,1}\Psi_{\gamma,1}, \widetilde{\Psi}_\gamma)_{H(\partial B)^2}}{(\Psi_\gamma, \widetilde{\Psi}_\gamma)_{H(\partial B)^2}} \\
(\tau_\gamma - W_{B,0})\Psi_{\gamma,1} &= -W_{B,1}\Psi_\gamma.
\end{aligned} \tag{3.29}$$

Here, $\widetilde{\Psi}_{\gamma'} \in \mathrm{Ker}(\bar{\tau}_{\gamma'} - W^*_{B,0})$ *and* $W^*_{B,0}$ *is the* $H(\partial B)^2$ *adjoint of* $W_{B,0}$.

Using Lemma 3.3.4 and Remark 3.3.2 we can solve $\Psi_{\gamma,1}$. Indeed,

$$\Psi_{\gamma,1} = \sum_{\substack{\gamma'\in\Gamma \\ \gamma'\neq\gamma}} \frac{\alpha(-W_{B,1}\Psi_\gamma, \Psi_{\gamma'})\Psi_{\gamma'}}{\tau_\gamma - \tau_{\gamma'}} + \sum_{\substack{\gamma'_g\in\Gamma_g \\ \gamma'\neq\gamma}} \alpha(-W_{B,1}\Psi_\gamma, \Psi_{\gamma'_g}) \left(\frac{\Psi_{\gamma'_g}}{\tau_\gamma - \tau_{\gamma'}} + \frac{\Psi_{\gamma'}}{(\tau_\gamma - \tau_{\gamma'})^2} \right) + \alpha(-W_{B,1}\Psi_\gamma, \Psi_\gamma)\Psi_\gamma.$$

By abuse of notation,

$$\alpha(x, \Psi_\gamma) = \begin{cases} \alpha(x_1, \psi_\kappa) & \gamma = (1, j, i),\ \kappa = (j, i), \\ \alpha(x_2, \psi_\kappa) & \gamma = (2, j, i),\ \kappa = (j, i), \end{cases} \tag{3.30}$$

for

$$x = \begin{pmatrix} x_1 \\ x_2 \end{pmatrix} \in H(\partial B)^2$$

with α being introduced in Definition 3.2.

Consider now the degenerate case $\gamma \in \Gamma \backslash \Gamma_{\mathrm{sim}} =: \Gamma_{\mathrm{deg}} = \{\gamma = (k, i, j) \in \Gamma$ s.t $\lambda_{j,i} \in \sigma_3\}$. It is clear that, for $\gamma \in \Gamma_{\mathrm{deg}}$, the algebraic multiplicity of the eigenvalue τ_γ is 2 while the geometric multiplicity is 1.
In this case every eigenvalue τ_γ and associated eigenfunction Ψ_γ will split into two branches, as δ goes to zero, represented by a convergent Puiseux series as [28]:

$$\begin{aligned}
\tau_{\gamma,h}(\delta) &= \tau_\gamma + (-1)^h \delta^{1/2}\tau_{\gamma,1} + (-1)^{2h}\delta^{2/2}\tau_{\gamma,2} + O(\delta^{3/2}), \quad h = 0, 1, \\
\Psi_{\gamma,h}(\delta) &= \Psi_\gamma + (-1)^h \delta^{1/2}\Psi_{\gamma,1} + (-1)^{2h}\delta^{2/2}\Psi_{\gamma,2} + O(\delta^{3/2}), \quad h = 0, 1,
\end{aligned} \tag{3.31}$$

where $\tau_{\gamma,j}$ and $\Psi_{\gamma,j}$ can be recovered by recurrence formulas. We refer to [62] for more details.

3.3.1 First-order correction to plasmonic resonances and field behavior at the plasmonic resonances

Recall that the electric and magnetic parameters, ε_c and μ_c, depend on the frequency of the incident field, ω, following the Drude model [9]. Therefore, the eigenvalues of the operator $W_{B,0}$ and perturbation in the eigenvalues depend on the frequency as well, that is,

$$\begin{aligned}
\tau_\gamma(\delta, \omega) &= \tau_\gamma(\omega) + \delta^2\tau_{\gamma,2}(\omega) + O(\delta^3), \quad \gamma \in \Gamma_{\mathrm{sim}}, \\
\tau_{\gamma,h}(\delta, \omega) &= \tau_\gamma + \delta^{1/2}(-1)^h\tau_{\gamma,1}(\omega) + \delta^{2/2}(-1)^{2h}\tau_{\gamma,2}(\omega) + O(\delta^{3/2}), \quad \gamma \in \Gamma_{\mathrm{deg}}, \quad h = 0, 1.
\end{aligned}$$

In the sequel, we will omit frequency dependence to simplify the notation. However, we will keep in mind that all these quantities are frequency dependent.
We first recall different notions of plasmonic resonance, see chapter 2.

Definition 3.3. *(i) We say that ω is a plasmonic resonance if $|\tau_\gamma(\delta)| \ll 1$ and is locally minimized for some $\gamma \in \Gamma_{\text{sim}}$ or $|\tau_{\gamma,h}(\delta)| \ll 1$ and is locally minimized for some $\gamma \in \Gamma_{\text{deg}}$, $h = 0, 1$.*

(ii) We say that ω is a quasi-static plasmonic resonance if $|\tau_\gamma| \ll 1$ and is locally minimized for some $\gamma \in \Gamma$. Here, τ_γ is defined by (3.27).

(iii) We say that ω is a first-order corrected quasi-static plasmonic resonance if $|\tau_\gamma + \delta^2 \tau_{\gamma,2}| \ll 1$ and is locally minimized for some $\gamma \in \Gamma_{\text{sim}}$ or $|\tau_\gamma + \delta^{1/2}(-1)^h \tau_{\gamma,1}| \ll 1$ and is locally minimized for some $\gamma \in \Gamma_{\text{deg}}$, $h = 0, 1$. Here, the correction terms $\tau_{\gamma,2}$ and $\tau_{\gamma,1}$ are defined by (3.29) and (3.31).

Note that quasi-static resonance is size independent and is therefore a zero-order approximation of the plasmonic resonance in terms of the particle size while the first-order corrected quasi-static plasmonic resonance depends on the size of the nanoparticle.

We are interested in solving equation (3.26)

$$W_B(\delta)\Psi = f,$$

where

$$\Psi = \begin{pmatrix} \eta(\psi)^{(1)} \\ \eta(\psi)^{(2)} \\ \omega\eta(\phi)^{(1)} \\ \omega\eta(\phi)^{(2)} \end{pmatrix}, f = \left. \begin{pmatrix} \dfrac{\eta(\nu \times E^i)^{(1)}}{\mu_m - \mu_c} \\ \dfrac{\eta(\nu \times E^i)^{(2)}}{\mu_m - \mu_c} \\ \dfrac{\eta(i\nu \times H^i)^{(1)}}{\varepsilon_m - \varepsilon_c} \\ \dfrac{\eta(i\nu \times H^i)^{(2)}}{\varepsilon_m - \varepsilon_c} \end{pmatrix} \right|_{\partial B}$$

for ω close to the resonance frequencies, i.e., when $\tau_\gamma(\delta)$ is very small for some γ's $\in \Gamma_{\text{sim}}$ or $\tau_{\gamma,h}(\delta)$ is very small for some γ's $\in \Gamma_{\text{deg}}$, $h = 0, 1$. In this case, the major part of the solution would be the contributions of the excited resonance modes $\Psi_\gamma(\delta)$ and $\Psi_{\gamma,h}(\delta)$.

We introduce the following definition.

Definition 3.4. *We call $J \subset \Gamma$ index set of resonances if τ_γ's are close to zero when $\gamma \in \Gamma$ and are bounded from below when $\gamma \in \Gamma^c$. More precisely, we choose a threshold number $\eta_0 > 0$ independent of ω such that*

$$|\tau_\gamma| \geq \eta_0 > 0 \quad \text{for } \gamma \in J^c.$$

From now on, we shall use J as our index set of resonances. For simplicity, we assume throughout this chapter that the following condition holds.

Condition 3.4. *We assume that $\lambda_\mu \neq 0$, $\lambda_\varepsilon \neq 0$ or equivalently, $\mu_c \neq -\mu_m$, $\varepsilon_c \neq -\varepsilon_m$.*

It follows that the set J is finite.
Consider the space $\mathcal{E}_J = \text{span}\{\Psi_\gamma(\delta), \Psi_{\gamma,h}(\delta);\ \gamma \in J,\ h = 0, 1\}$. Note that, under Condition 3.4, $\mathcal{E}_J$ is finite dimensional. Similarly, we define $\mathcal{E}_{J^c}$ as the spanned by $\Psi_\gamma(\delta), \Psi_{\gamma,h}(\delta);\ \gamma \in J^c,\ h = 0, 1$ and eventually other vectors to complete the base. We have $H(\partial B)^2 = \mathcal{E}_J \oplus \mathcal{E}_{J^c}$.

We define $P_J(\delta)$ and $P_{J^c}(\delta)$ as the (non-orthogonal) projection into the finite-dimensional space $\mathcal{E}_J$ and infinite-dimensional space $\mathcal{E}_{J^c}$, respectively. It is clear that, for any $f \in H(\partial B)^2$

$$f = P_J(\delta)[f] + P_{J^c}(\delta)[f].$$

Moreover, we have an explicit representation for $P_J(\delta)$

$$P_J(\delta)[f] = \sum_{\gamma \in J \cap \Gamma_{\text{sim}}} \alpha_\delta(f, \Psi_\gamma(\delta))\Psi_\gamma(\delta) + \sum_{\substack{\gamma \in J \cap \Gamma_{\text{deg}} \\ h=0,1}} \alpha_\delta(f, \Psi_{\gamma,h}(\delta))\Psi_{\gamma,h}(\delta). \tag{3.32}$$

Here, as in Lemma 3.3.4,

$$\begin{aligned}
\alpha_\delta(f, \Psi_\gamma(\delta)) &= \frac{(f, \tilde{\Psi}_\gamma(\delta))_{H(\partial B)^2}}{(\Psi_\gamma(\delta), \tilde{\Psi}_\gamma(\delta))_{H(\partial B)^2}}, \quad \gamma \in J \cap \Gamma_{\text{sim}}, \\
\alpha_\delta(f, \Psi_{\gamma,h}(\delta)) &= \frac{(f, \tilde{\Psi}_{\gamma,h}(\delta))_{H(\partial B)^2}}{(\Psi_{\gamma,h}(\delta), \tilde{\Psi}_{\gamma,h}(\delta))_{H(\partial B)^2}}, \quad \gamma \in J \cap \Gamma_{\text{deg}},\ h = 0, 1,
\end{aligned}$$

where $\widetilde{\Psi}_\gamma \in \text{Ker}(\bar{\tau}_{\gamma,h}(\delta) - W_B^*(\delta))$, $\widetilde{\Psi}_{\gamma,h} \in \text{Ker}(\bar{\tau}_{\gamma,h}(\delta) - W_B^*(\delta))$ and $W_B^*(\delta)$ is the $H(\partial B)^2$-adjoint of $W_B(\delta)$.

We are now ready to solve the equation $W_B(\delta)\Psi = f$. In view of Remark 3.3.2,

$$\Psi = W_B^{-1}(\delta)[f] = \sum_{\gamma \in J \cap \Gamma_{\text{sim}}} \frac{\alpha_\delta(f, \Psi_\gamma(\delta))\Psi_\gamma(\delta)}{\tau_\gamma(\delta)} + \sum_{\substack{\gamma \in J \cap \Gamma_{\text{deg}} \\ h=0,1}} \frac{\alpha_\delta(f, \Psi_{\gamma,h}(\delta))\Psi_{\gamma,h}(\delta)}{\tau_{\gamma,h}(\delta)} + W_B^{-1}(\delta)P_{J^c}(\delta)[f]. \tag{3.33}$$

The following lemma holds. A similar result was proved for $\delta = 0$ in [6].

Lemma 3.3.6. *The norm $\|W_B^{-1}(\delta)P_{J^c}(\delta)\|_{\mathcal{L}(H(\partial B)^2, H(\partial B)^2)}$ is uniformly bounded in ω and δ.*

Proof. Consider the operator

$$W_B(\delta)|_{J^c} : P_{J^c}(\delta)H(\partial B)^2 \to P_{J^c}(\delta)H(\partial B)^2.$$

We can show that for every ω and δ, $\text{dist}(\sigma(W_B(\delta)|_{J^c}), 0) \geq \frac{\eta_0}{2}$, where $\sigma(W_B(\delta)|_{J^c})$ is the discrete spectrum of $W_B(\delta)|_{J^c}$. Here and throughout the chapter, dist denotes the distance. Then, it follows that

$$\|W_B^{-1}(\delta)P_{J^c}(\delta)[f]\| = \|W_B^{-1}(\delta)|_{J^c}P_{J^c}(\delta)[f]\| \lesssim \frac{1}{\eta_0}\exp(\frac{C_1}{\eta_0^2})\|P_{J^c}(\delta)[f]\| \lesssim \frac{1}{\eta_0}\exp(\frac{C_1}{\eta_0^2})\|f\|,$$

where the notation $A \lesssim B$ means that $A \leq CB$ for some constant C independent of A and B. □

Finally, we are ready to state our main result in this section.

Theorem 3.3.2. *Let η be defined by (3.14). Under Conditions 3.1, 3.2, 3.3 and 3.4, the scattered field $E^s = E - E^i$ due to a single plasmonic particle has the following representation:*

$$E^s = \mu_m \nabla \times \vec{\mathcal{S}}_D^{k_m}[\psi](x) + \nabla \times \nabla \times \vec{\mathcal{S}}_D^{k_m}[\phi](x) \qquad x \in \mathbb{R}^3 \backslash \bar{D},$$

where

$$\begin{aligned} \psi &= \eta^{-1}\big(\nabla_{\partial B}\widetilde{\psi}^{(1)} + \vec{\text{curl}}_{\partial B}\widetilde{\psi}^{(2)}\big), \\ \phi &= \frac{1}{\omega}\eta^{-1}\big(\nabla_{\partial B}\widetilde{\phi}^{(1)} + \vec{\text{curl}}_{\partial B}\widetilde{\phi}^{(2)}\big), \end{aligned}$$

$$\begin{aligned} \Psi &= \begin{pmatrix} \widetilde{\psi}^{(1)} \\ \widetilde{\psi}^{(2)} \\ \widetilde{\phi}^{(1)} \\ \widetilde{\phi}^{(2)} \end{pmatrix} \\ &= \sum_{\gamma \in J \cap \Gamma_{\text{sim}}} \frac{\alpha(f, \Psi_\gamma)\Psi_\gamma + O(\delta)}{\tau_\gamma(\delta)} + \sum_{\gamma \in J \cap \Gamma_{\text{deg}}} \frac{\zeta_1(f)\Psi_\gamma + \zeta_2(f)\Psi_{\gamma,1} + O(\delta^{1/2})}{\tau_{\gamma,0}(\delta)\tau_{\gamma,1}(\delta)} + O(1), \end{aligned}$$

and

$$\begin{aligned} \zeta_1(f) &= \frac{(f, \tilde{\Psi}_{\gamma,1})_{H(\partial B)^2}\tau_\gamma - (f, \tilde{\Psi}_\gamma)_{H(\partial B)^2}(\tau_{\gamma,1} + \tau_\gamma \frac{a_2}{a_1})}{a_1}, \\ \zeta_2(f) &= \frac{(f, \tilde{\Psi}_\gamma)_{H(\partial B)^2}}{a_1}, \\ a_1 &= (\Psi_\gamma, \tilde{\Psi}_{\gamma,1})_{H(\partial B)^2} + (\Psi_{\gamma,1}, \tilde{\Psi}_\gamma)_{H(\partial B)^2}, \\ a_2 &= (\Psi_\gamma, \tilde{\Psi}_{\gamma,2})_{H(\partial B)^2} + (\Psi_{\gamma,2}, \tilde{\Psi}_\gamma)_{H(\partial B)^2} + (\Psi_{\gamma,1}, \tilde{\Psi}_{\gamma,1})_{H(\partial B)^2}. \end{aligned}$$

Proof. Recall that

$$\Psi = \sum_{\gamma \in J \cap \Gamma_{\text{sim}}} \frac{\alpha_\delta(f, \Psi_\gamma(\delta))\Psi_\gamma(\delta)}{\tau_\gamma(\delta)} + \sum_{\substack{\gamma \in J \cap \Gamma_{\text{deg}} \\ h=0,1}} \frac{\alpha_\delta(f, \Psi_{\gamma,h}(\delta))\Psi_{\gamma,h}(\delta)}{\tau_{\gamma,h}(\delta)} + W_B^{-1}(\delta)P_{J^c}(\delta)[f].$$

By Lemma 3.3.6, we have $W_B^{-1}(\delta)P_{J^c}(\delta)[f] = O(1)$.
If $\gamma \in J \cap \Gamma_{\text{sim}}$, an asymptotic expansion on δ yields

$$\alpha_\delta(f, \Psi_\gamma(\delta))\Psi_\gamma(\delta) = \alpha(f, \Psi_\gamma)\Psi_\gamma + O(\delta).$$

If $\gamma \in J \cap \Gamma_{\text{deg}}$ then $(\Psi_\gamma, \tilde{\Psi}_\gamma)_{H(\partial B)^2} = 0$. Therefore, an asymptotic expansion on δ yields

$$\begin{aligned} \alpha_\delta(f, \Psi_{\gamma,h}(\delta))\Psi_{\gamma,h}(\delta) &= \frac{(-1)^h(f, \tilde{\Psi}_\gamma)_{H(\partial B)^2}\Psi_\gamma}{\delta^{-1/2}a_1} + \\ &\quad \frac{1}{a_1}\left(\big((f, \tilde{\Psi}_{\gamma,1})_{H(\partial B)^2} - (f, \tilde{\Psi}_\gamma)_{H(\partial B)^2}\frac{a_2}{a_1}\big)\Psi_\gamma + (f, \tilde{\Psi}_\gamma)_{H(\partial B)^2}\Psi_{\gamma,1}\right) \\ &\quad + O(\delta^{1/2}) \end{aligned}$$

with

$$\begin{aligned} a_1 &= (\Psi_\gamma, \tilde{\Psi}_{\gamma,1})_{H(\partial B)^2} + (\Psi_{\gamma,1}, \tilde{\Psi}_\gamma)_{H(\partial B)^2}, \\ a_2 &= (\Psi_\gamma, \tilde{\Psi}_{\gamma,2})_{H(\partial B)^2} + (\Psi_{\gamma,2}, \tilde{\Psi}_\gamma)_{H(\partial B)^2} + (\Psi_{\gamma,1}, \tilde{\Psi}_{\gamma,1})_{H(\partial B)^2}. \end{aligned}$$

Since $\tau_{\gamma,h}(\delta) = \tau_\gamma + \delta^{1/2}(-1)^h \tau_{\gamma,1} + O(\delta)$, the result follows by adding the terms

$$\frac{\alpha_\delta(f, \Psi_{\gamma,0}(\delta))\Psi_{\gamma,0}(\delta)}{\tau_{\gamma,0}(\delta)} \quad \text{and} \quad \frac{\alpha_\delta(f, \Psi_{\gamma,1}(\delta))\Psi_{\gamma,1}(\delta)}{\tau_{\gamma,1}(\delta)}.$$

The proof is then complete. □

Corollary 3.3.1. *Assume the same conditions as in Theorem 3.3.2. Under the additional condition that*

$$\min_{\gamma \in J \cap \Gamma_{\text{sim}}} |\tau_\gamma(\delta)| \gg \delta^3, \quad \min_{\gamma \in J \cap \Gamma_{\text{deg}}} |\tau_\gamma(\delta)| \gg \delta, \tag{3.34}$$

we have

$$\Psi = \sum_{\gamma \in J \cap \Gamma_{\text{sim}}} \frac{\alpha(f, \Psi_\gamma)\Psi_\gamma + O(\delta)}{\tau_\gamma + \delta^2 \tau_{\gamma,2}} + \sum_{\gamma \in J \cap \Gamma_{\text{deg}}} \frac{\zeta_1(f)\Psi_\gamma + \zeta_2(f)\Psi_{\gamma,1} + O(\delta^{1/2})}{\tau_\gamma^2 - \delta \tau_{\gamma,1}^2} + O(1).$$

Corollary 3.3.2. *Assume the same conditions as in Theorem 3.3.2. Under the additional condition that*

$$\min_{\gamma \in J \cap \Gamma_{\text{sim}}} |\tau_\gamma(\delta)| \gg \delta^2, \quad \min_{\gamma \in J \cap \Gamma_{\text{deg}}} |\tau_\gamma(\delta)| \gg \delta^{1/2}, \tag{3.35}$$

we have

$$\Psi = \sum_{\gamma \in J \cap \Gamma_{\text{sim}}} \frac{\alpha(f, \Psi_\gamma)\Psi_\gamma + O(\delta)}{\tau_\gamma} + \sum_{\gamma \in J \cap \Gamma_{\text{deg}}} \frac{\alpha(f, \Psi_\gamma)\Psi_\gamma}{\tau_\gamma} + \alpha(f, \Psi_{\gamma,g}) \left(\frac{\Psi_{\gamma,g}}{\tau_\gamma} + \frac{\Psi_\gamma}{\tau_\gamma^2} \right) + O(1).$$

Proof. We have

$$\begin{aligned} \lim_{\delta \to 0} W_B^{-1}(\delta) P_{\text{span}\{\Psi_{\gamma,0}(\delta), \Psi_{\gamma,1}(\delta)\}}[f] &= \lim_{\delta \to 0} \frac{\alpha_\delta(f, \Psi_{\gamma,0}(\delta))\Psi_{\gamma,0}(\delta)}{\tau_{\gamma,0}(\delta)} + \frac{\alpha_\delta(f, \Psi_{\gamma,1}(\delta))\Psi_{\gamma,1}(\delta)}{\tau_{\gamma,1}(\delta)} \\ &= W_{B,0}^{-1}(\delta) P_{\text{span}\{\Psi_\gamma, \Psi_{\gamma g}\}}[f] \\ &= \frac{\alpha(f, \Psi_\gamma)\Psi_\gamma}{\tau_\gamma} + \alpha(f, \Psi_{\gamma,g}) \left(\frac{\Psi_{\gamma,g}}{\tau_\gamma} + \frac{\Psi_\gamma}{\tau_\gamma^2} \right), \end{aligned}$$

where $\gamma \in J \cap \Gamma_{\text{deg}}$, $f \in H(\partial B)^2$ and $)P_{\text{span}E}$ is the projection into the linear space generated by the elements in the set E. □

Remark 3.3.3. *Note that for $\gamma \in J$,*

$$\tau_\gamma \approx \min \left\{ \text{dist}\big(\lambda_\mu, \sigma(\mathcal{K}_B^*) \cup -\sigma(\mathcal{K}_B^*)\big), \text{dist}\big(\lambda_\varepsilon, \sigma(\mathcal{K}_B^*) \cup -\sigma(\mathcal{K}_B^*)\big) \right\}.$$

It is clear, from Remark 3.3.3, that resonances can occur when exciting the spectrum of $\mathcal{K}_B^*$ or/and that of $-\mathcal{K}_B^*$. We substantiate in the following that only the spectrum of $\mathcal{K}_B^*$ can be excited to create the plasmonic resonances in the quasi-static regime.

Recall that

$$f = \left(\begin{array}{c} \dfrac{\eta(\nu \times E^i)^{(1)}}{\mu_m - \mu_c} \\ \dfrac{\eta(\nu \times E^i)^{(2)}}{\mu_m - \mu_c} \\ \dfrac{\eta(i\nu \times H^i)^{(1)}}{\varepsilon_m - \varepsilon_c} \\ \dfrac{\eta(i\nu \times H^i)^{(2)}}{\varepsilon_m - \varepsilon_c} \end{array} \right) \Bigg|_{\partial B},$$

and therefore,

$$f_1 := \frac{\eta(\nu \times E^i)^{(1)}}{\mu_m - \mu_c} = \frac{\Delta_{\partial B}^{-1} \nabla_{\partial B} \cdot \eta(\nu \times E^i)}{\mu_m - \mu_c}.$$

Now, suppose $\gamma = (1, j, 1) \in J$ (recall that J is the index set of resonances). Then $\tau_\gamma = \lambda_\mu - \lambda_{1,j}$, where $\lambda_{1,j} \in \sigma_1 = \sigma(-\mathcal{K}_B^*) \backslash \sigma(\mathcal{K}_B^*)$. From Remark 3.3.1,

$$\alpha(f, \Psi_\gamma) = (\Delta_{\partial B} f_1, \varphi_{j,1})_{\mathcal{H}^*} = \alpha(f, \Psi_\gamma) = \frac{1}{\mu_m - \mu_c} (\nabla_{\partial B} \cdot \eta(\nu \times E^i), \varphi_{j,1})_{\mathcal{H}^*},$$

where $\varphi_{j,1} \in \mathcal{H}_0^*(\partial B)$ is a normalized eigenfunction of $\mathcal{K}_B^*(\partial B)$.

A Taylor expansion of E^i gives, for $x \in \partial D$,

$$E^i(x) = \sum_{\beta \in \mathbb{N}^3}^{\infty} \frac{(x - z)^\beta \partial^\beta E^i(z)}{|\beta|!}.$$

Thus,

$$\eta(\nu \times E^i)(\tilde{x}) = \eta(\nu)(\tilde{x}) \times E^i(z) + O(\delta),$$

and

$$\begin{aligned} \nabla_{\partial B} \cdot \eta(\nu \times E^i)(\tilde{x}) &= -\eta(\nu)(\tilde{x}) \cdot \nabla \times E^i(z) + O(\delta) \\ &= O(\delta). \end{aligned}$$

Therefore, the zeroth-order term of the expansion of $\nabla_{\partial B} \cdot \eta(\nu \times E^i)$ in δ is zero. Hence,

$$\alpha(f, \Psi_\gamma) = 0.$$

In the same way, we have

$$\begin{aligned} \alpha(f, \Psi_\gamma) &= 0, \\ \alpha(f, \Psi_{\gamma_g}) &= 0 \end{aligned}$$

for $\gamma = (2, j, 1) \in J$ and γ_g such that $\gamma \in J$.

As a result we see that the spectrum of $-\mathcal{K}_B^*$ is not excited in the zeroth-order term. However, we note that $\sigma(-\mathcal{K}_B^*)$ can be excited in higher-order terms.

3.4 The extinction cross-section at the quasi-static limit

The aim of this section is to derive an expression of the extinction cross section and estimate its blow up at the plasmonic resonances. We first recall the quasi-static limit of the electric field at plasmonic resonances. The formula was first obtained in [9], but it can be derived by pursuing further computations in Corollary 3.3.2. In this formula, the polarization tensor is a key ingredient. It allows to express the quasi-static limit or zeroth-order approximation of the electromagnetic fields far away from the particle. The polarization tensor is given by [18]

$$M(\lambda, D) = \int_{\partial D} (\lambda Id - \mathcal{K}_D^*)^{-1}[\nu](x)\, x\, d\sigma(x), \tag{3.36}$$

where $\lambda \in \mathbb{C}\backslash(-1/2, 1/2)$. In view of Lemma 3.2.1, we have

$$M(\lambda, D) = \sum_{j=1}^{\infty} \frac{1}{\lambda - \lambda_j} (\nu, \varphi_j)_{\mathcal{H}^*} (\varphi_j, x)_{-\frac{1}{2},\frac{1}{2}}, \tag{3.37}$$

since $(\nu, \varphi_0)_{\mathcal{H}^*} = 0$;

The following result follows from [9].

Theorem 3.4.1. *Let* $d_\sigma = \min\left\{\operatorname{dist}\big(\lambda_\mu, \sigma(\mathcal{K}_D^*) \cup -\sigma(\mathcal{K}_D^*)\big), \operatorname{dist}\big(\lambda_\varepsilon, \sigma(\mathcal{K}_D^*) \cup -\sigma(\mathcal{K}_D^*)\big)\right\}$. *Then, for* $D = z + \delta B \Subset \mathbb{R}^3$ *of class* $\mathcal{C}^{1,\alpha}$ *for* $0 < \alpha < 1$, *the following uniform far-field expansion holds*

$$E^s = -\frac{i\omega\mu_m}{\varepsilon_m} \nabla \times G_d(x, z, k_m) M(\lambda_\mu, D) H^i(z) - \omega^2 \mu_m G_d(x, z, k_m) M(\lambda_\varepsilon, D) E^i(z) + O(\frac{\delta^4}{d_\sigma}),$$

where

$$G_d(x, z, k_m) = \varepsilon_m \big(G(x, z, k_m) Id + \frac{1}{k_m^2} D_x^2 G(x, z, k_m)\big)$$

is the Dyadic Green (matrix valued) function for the full Maxwell equations.

In order to express the extinction cross section we need to write the far-field behavior of the electric and magnetic fields. We first recall the representation for the scattering amplitude [78]. It is well-known that the solution (E, H) to the system (3.10) has the following far-field expansion as $|x| \to +\infty$:

$$E^s(x) = -\frac{e^{ik_m|x|}}{4\pi|x|} A_\infty(\hat{x}) + O\left(\frac{1}{|x|^2}\right),$$

and

$$H^s(x) = -\frac{e^{ik_m|x|}}{4\pi|x|} \hat{x} \times A_\infty(\hat{x}) + O\left(\frac{1}{|x|^2}\right),$$

where

$$A_\infty(\hat{x}) = -i\mu_m k_m \hat{x} \times \int_{\partial D} e^{-ik_m \hat{x}\cdot y} \psi(y) d\sigma(y) - k_m^2 \hat{x} \times \hat{x} \times \int_{\partial D} e^{-ik_m \hat{x}\cdot y} \phi(y) d\sigma(y),$$

and $\hat{x} = \frac{x}{|x|}$.

We also need the optical cross-section theorem for the scattering of electromagnetic waves [43], which can be stated as follows. Assume that the incident fields are plane waves given by

$$\begin{aligned} E^i(x) &= pe^{ik_m d\cdot x}, \\ H^i(x) &= d \times pe^{ik_m d\cdot x}, \end{aligned}$$

where $p \in \mathbb{R}^3$ and $d \in \mathbb{R}^3$ with $|d| = 1$ are such that $p \cdot d = 0$. Then, the extinction cross-section is given by

$$Q^{ext} = \frac{4\pi}{k_m} \Im \left[\frac{p \cdot A_\infty(d)}{|p|^2} \right],$$

where A_∞ is the scattering amplitude.

From Taylor expansions on the formula of Theorem 3.4.1, it follows that the following far-field asymptotic expansion holds:

$$\begin{aligned} &E^s = \\ &-\frac{e^{ik_m|x|}}{4\pi|x|} \Big(\omega\mu_m k_m e^{ik_m(d-\hat{x})\cdot z} \big(\hat{x} \times Id\big) M(\lambda_\mu, D)(d \times p) - k_m^2 e^{ik_m(d-\hat{x})\cdot z} \big(Id - \hat{x}\hat{x}^t\big) M(\lambda_\varepsilon, D)p \Big) \\ &+O(\frac{1}{|x|^2}) + O(\frac{\delta^4}{d_\sigma}), \end{aligned}$$

where $\hat{x} = x/|x|$. Therefore, up to an error term of order $O(\frac{\delta^4}{d_\sigma})$, we have

$$A_\infty(\hat{x}) = \omega\mu_m k_m e^{ik_m(d-\hat{x})\cdot z} \big(\hat{x}\times Id\big) M(\lambda_\mu, D)(d\times p) - k_m^2 e^{ik_m(d-\hat{x})\cdot z} \big(Id - \hat{x}\hat{x}^t\big) M(\lambda_\varepsilon, D)p. \tag{3.38}$$

Formula (3.38) allows us to compute the extinction cross-section Q^{ext} in terms of the polarization tensors associated with the particle D and the material parameter contrasts. Moreover, an estimate for the blow up of Q^{ext} at the plasmonic resonances follows immediately from (3.37).

Theorem 3.4.2. *We have*

$$Q^{ext} = \frac{4\pi}{k_m|p|^2} \Im \left[p \cdot \left[\omega\mu_m k_m \big(d \times Id\big) M(\lambda_\mu, D)(d \times p) - k_m^2 \big(Id - dd^t\big) M(\lambda_\varepsilon, D)p \right] \right],$$

where $M(\lambda_\mu, D)$ and $M(\lambda_\varepsilon, D)$ are the polarization tensors associated with D and $\lambda = \lambda_\mu$ and $\lambda = \lambda_\varepsilon$, respectively.

3.5 Explicit computations for a spherical nanoparticle

In this section we consider a spherical nanoparticle and explicitly compute the first order correction in terms of radius of its plasmonic resonances. We also derive and explicit formula for the extinction cross section.

3.5.1 Vector spherical harmonics

Let $\hat{x} = \frac{x}{|x|}$. For $m = -n, ..., n$ and $n = 1, 2, ...$, set Y_n^m to be the spherical harmonics defined on the unit sphere $S = \{x \in \mathbb{R}^3, |x| = 1\}$. For a wave

number $k > 0$, the function

$$v_{n,m}(k;x) = h_n^{(1)}(k|x|)Y_n^m(\hat{x})$$

satisfies the Helmholtz equation $\Delta v + k^2 v = 0$ in $\mathbb{R}^3 \setminus \{0\}$ together with the Sommerfeld radiation condition

$$\lim_{|x|\to\infty} |x|\Big(\frac{\partial v_{n,m}}{\partial |x|}(k;x) - ikv_{n,m}(k;x)\Big) = 0.$$

Similarly, let $\widetilde{v}_{n,m}(x)$ be defined by

$$\widetilde{v}_{n,m}(x) = j_n(k|x|)Y_n^m(\hat{x}),$$

where j_n is the spherical Bessel function of the first kind. Then the function $\widetilde{v}_{n,m}$ satisfies the Helmholtz equation in $\mathbb{R}^3$.

Next, define the vector spherical harmonics by

$$U_{n,m} = \frac{1}{\sqrt{n(n+1)}}\nabla_S Y_n^m(\hat{x}) \quad \text{and} \quad V_{n,m} = \hat{x} \times U_{n,m}$$

for $m = -n, ..., n$ and $n = 1, 2, ...$ Here, $\hat{x} \in S$ and ∇_S denote the surface gradient on the unit sphere S. The vector spherical harmonics form a complete orthogonal basis for $L_T^2(S)$.

Using the vectorial spherical harmonics, we can separate the solutions of Maxwell's equations into multipole solutions; see [78, Section 5.3]. Define the exterior transverse electric multipoles, i.e., $E \cdot x = 0$, as

$$\begin{cases} E_{n,m}^{TE}(x) = -\sqrt{n(n+1)}h_n^{(1)}(k|x|)V_{n,m}(\hat{x}), \\ H_{n,m}^{TE}(x) = -\dfrac{i}{\omega\mu}\nabla\times\Big(-\sqrt{n(n+1)}h_n^{(1)}(k|x|)V_{n,m}(\hat{x})\Big), \end{cases} \tag{3.39}$$

and the exterior transverse magnetic multipoles, i.e., $H \cdot x = 0$, as

$$\begin{cases} E_{n,m}^{TM}(x) = \dfrac{i}{\omega\epsilon}\nabla\times\Big(-\sqrt{n(n+1)}h_n^{(1)}(k|x|)V_{n,m}(\hat{x})\Big), \\ H_{n,m}^{TM}(x) = -\sqrt{n(n+1)}h_n^{(1)}(k|x|)V_{n,m}(\hat{x}). \end{cases} \tag{3.40}$$

The exterior electric and magnetic multipoles satisfy the Sommerfeld radiation condition. In the same manner, one defines the interior multipoles $(\widetilde{E}_{n,m}^{TE}, \widetilde{H}_{n,m}^{TE})$ and $(\widetilde{E}_{n,m}^{TM}, \widetilde{H}_{n,m}^{TM})$ with $h_n^{(1)}$ replaced by j_n, i.e.,

$$\begin{cases} \widetilde{E}_{n,m}^{TE}(x) = -\sqrt{n(n+1)}j_n(k|x|)V_{n,m}(\hat{x}), \\ \widetilde{H}_{n,m}^{TE}(x) = -\dfrac{i}{\omega\mu}\nabla \times \widetilde{E}_{n,m}^{TE}(x), \end{cases} \tag{3.41}$$

and

$$\begin{cases} \widetilde{H}_{n,m}^{TM}(x) = -\sqrt{n(n+1)}j_n(k|x|)V_{n,m}(\hat{x}), \\ \widetilde{E}_{n,m}^{TM}(x) = \dfrac{i}{\omega\epsilon}\nabla \times \widetilde{H}_{n,m}^{TM}(x). \end{cases} \tag{3.42}$$

Note that one has

$$\nabla \times E^{TE}_{n,m}(k;x) = \frac{\sqrt{n(n+1)}}{|x|}\mathcal{H}_n(k|x|)U_{n,m}(\hat{x}) + \frac{n(n+1)}{|x|}h^{(1)}_n(k|x|)Y^m_n(\hat{x})\hat{x} \tag{3.43}$$

and

$$\nabla \times \widetilde{E}^{TE}_{n,m}(k;x) = \frac{\sqrt{n(n+1)}}{|x|}\mathcal{J}_n(k|x|)U_{n,m}(\hat{x}) + \frac{n(n+1)}{|x|}j_n(k|x|)Y^m_n(\hat{x})\hat{x}, \tag{3.44}$$

where

$$\mathcal{J}_n(t) = j_n(t) + tj'_n(t), \qquad \mathcal{H}_n(t) = h^{(1)}_n(t) + t(h^{(1)}_n)'(t).$$

For $|x| > |y|$, the following addition formula holds:

$$\begin{aligned} G(x,y,k)I = &-\sum_{n=1}^{\infty}\frac{ik}{n(n+1)}\frac{\epsilon}{\mu}\sum_{m=-n}^{n}E^{TM}_{n,m}(x)\overline{\widetilde{E}^{TM}_{n,m}(y)}^T \\ &-\sum_{n=1}^{\infty}\frac{ik}{n(n+1)}\sum_{m=-n}^{n}E^{TE}_{n,m}(x)\overline{\widetilde{E}^{TE}_{n,m}(y)}^T \\ &-\frac{i}{k}\sum_{n=1}^{\infty}\sum_{m=-n}^{n}\nabla v_{n,m}(x)\overline{\nabla\widetilde{v}_{n,m}(y)}^T. \end{aligned} \tag{3.45}$$

Alternatively, for $|x| < |y|$, we have

$$\begin{aligned} G(x,y,k)I = &-\sum_{n=1}^{\infty}\frac{ik}{n(n+1)}\frac{\epsilon}{\mu}\sum_{m=-n}^{n}\overline{\widetilde{E}^{TM}_{n,m}(x)}E^{TM}_{n,m}(y)^T \\ &-\sum_{n=1}^{\infty}\frac{ik}{n(n+1)}\sum_{m=-n}^{n}\overline{\widetilde{E}^{TE}_{n,m}(x)}E^{TE}_{n,m}(y)^T \\ &-\frac{i}{k}\sum_{n=1}^{\infty}\sum_{m=-n}^{n}\overline{\nabla\widetilde{v}_{n,m}(x)}\nabla v_{n,m}(y)^T. \end{aligned} \tag{3.46}$$

3.5.2 Explicit representations of boundary integral operators

Let D be a sphere of radius $r > 0$. We have the following results.

Lemma 3.5.1. *Let $\partial D = \{|x| = r\}$. Then, for $r' > r$, we have*

$$\nu \times \nabla \times \vec{S}^k_D[U_{n,m}]\big|^+_{|x|=r'} = (-ikr)h^{(1)}_n(kr')\mathcal{J}_n(kr)U_{n,m}, \tag{3.47}$$

$$\nu \times \nabla \times \vec{S}^k_D[V_{n,m}]\big|^+_{|x|=r'} = ik\frac{r^2}{r'}j_n(kr)\mathcal{H}_n(kr')V_{n,m}, \tag{3.48}$$

$$\nu \times \nabla \times \nabla \times \vec{S}^k_D[U_{n,m}]\big|^+_{|x|=r'} = -ik\frac{r}{r'}\mathcal{J}_n(kr)\mathcal{H}_n(kr')V_{n,m}, \tag{3.49}$$

$$\nu \times \nabla \times \nabla \times \vec{S}^k_D[V_{n,m}]\big|^+_{|x|=r'} = ik(kr)^2 j_n(kr)h^{(1)}_n(kr')U_{n,m}. \tag{3.50}$$

For $r' < r$,

$$\nu \times \nabla \times \vec{S}_D^k[U_{n,m}]\big|_{|x|=r'}^{+} = (-ikr) j_n(kr') \mathcal{H}_n(kr) U_{n,m}, \tag{3.51}$$

$$\nu \times \nabla \times \vec{S}_D^k[V_{n,m}]\big|_{|x|=r'}^{+} = ik \frac{r^2}{r'} \mathcal{J}_n(kr') h_n^{(1)}(kr) V_{n,m}, \tag{3.52}$$

$$\nu \times \nabla \times \nabla \times \vec{S}_D^k[U_{n,m}]\big|_{|x|=r'}^{+} = -ik \frac{r}{r'} \mathcal{J}_n(kr') \mathcal{H}_n(kr) V_{n,m}, \tag{3.53}$$

$$\nu \times \nabla \times \nabla \times \vec{S}_D^k[V_{n,m}]\big|_{|x|=r'}^{+} = ik(kr)^2 j_n(kr') h_n^{(1)}(kr) U_{n,m}. \tag{3.54}$$

Proof. We only consider (3.47). The other formulas can be proved in a similar way.

From (3.43), (3.44), and the definitions of $E_{n,m}^{TE}, E_{n,m}^{TM}, \widetilde{E}_{n,m}^{TE}$ and $\widetilde{E}_{n,m}^{TM}$, we have

$$\begin{aligned}
&\nabla_x \times G(x,y,k) U_{n,m}(\hat{y}) \\
&= -\sum_{n=1}^{\infty} \frac{ik}{n(n+1)} \frac{\epsilon}{\mu} \sum_{m=-n}^{n} \nabla \times E_{n,m}^{TM}(x) \overline{\widetilde{E}_{n,m}^{TM}(y)} \cdot U_{p,q}(\hat{y}) \\
&\quad + \sum_{n=1}^{\infty} \frac{ik}{n(n+1)} \sum_{m=-n}^{n} \nabla \times E_{n,m}^{TE}(x) \overline{\widetilde{E}_{n,m}^{TE}(y)} \cdot U_{p,q}(\hat{y}) \\
&= -\sum_{n=1}^{\infty} \frac{ik}{\sqrt{n(n+1)}} \frac{\epsilon}{\mu} \sum_{m=-n}^{n} \nabla \times E_{n,m}^{TM}(x) \frac{-i}{\omega\varepsilon} \frac{1}{r} \mathcal{J}_n(kr) U_{n,m}(\hat{y}) \cdot U_{p,q}(\hat{y}) \\
&\quad + \sum_{n=1}^{\infty} \frac{ik}{\sqrt{n(n+1)}} \sum_{m=-n}^{n} \nabla \times E_{n,m}^{TE}(x)(-1) j_n(kr) V_{n,m}(\hat{y}) \cdot U_{p,q}(\hat{y})
\end{aligned}$$

for $|y| = r$ and $|x| > |y|$. Therefore, we get on $|x| = r$

$$\begin{aligned}
\nabla \times \vec{S}_D^k[U_{n,m}]\big|_{+} &= \nabla_x \times \int_{|y|=r} G(x,y,k) U_{n,m}(\hat{y}) \\
&= \frac{kr}{\sqrt{n(n+1)}} \frac{1}{\omega\mu} \mathcal{J}_n(kr) (\nabla \times E_{n,m}^{TM}(x))|_{|x|=r}.
\end{aligned} \tag{3.55}$$

Since

$$\nabla \times E_{p,q}^{TM} = \frac{i}{\omega\varepsilon} \nabla \times \nabla \times E_{p,q}^{TE} = \frac{i}{\omega\varepsilon} k^2 E_{p,q}^{TE},$$

we obtain

$$\begin{aligned}
\hat{x} \times \nabla \times \vec{S}_D^k[U_{n,m}]\big|_{+} &= \frac{ikr}{\sqrt{n(n+1)}} \mathcal{J}_n(kr) (\hat{x} \times E_{n,m}^{TE}(x))|_{|x|=r} \\
&= (-ikr) h_n^{(1)}(kr) \mathcal{J}_n(kr) U_{n,m} \quad \text{on } |x| = r,
\end{aligned}$$

which completes the proof. □

Note that

$$\nu \times \nabla \times \vec{\mathcal{S}}_D^k[\phi]\big|_{\pm} = (\mp \frac{1}{2} I + \mathcal{M}_D^k)[\phi] \quad \text{on } \partial D,$$

and recall the following identity, which was proved in [90],

$$\nu \times \nabla \times \nabla \times \vec{\mathcal{S}}_D^k[\phi] = \mathcal{L}_D^k[\phi] \quad \text{on } \partial D.$$

For $m = -n, \ldots, n$ and $n = 1, 2, 3, \ldots$, let $H_{n,m}(\partial D)$ be the subspace of $H(\partial D)$ defined by

$$H_{n,m}(\partial D) = \mathrm{span}\{U_{n,m}, V_{n,m}\}.$$

Let us represent the operators $\mathcal{M}_D^k$ and $\mathcal{L}_D^k$ explicitly on the subspace $H_{m,n}(\partial D)$. Using $U_{n,m}, V_{n,m}$ as basis vectors, we obtain the following matrix representations for $\mathcal{M}_D^k$ and $\mathcal{L}_D^k$ on the subspace $H_{n,m}(\partial D)$:

$$\mathcal{M}_D^k = \begin{pmatrix} \frac{1}{2} - ikrh_n^{(1)}(kr)\mathcal{J}_n(kr) & 0 \\ 0 & \frac{1}{2} + ikrj_n(kr)\mathcal{H}_n(kr) \end{pmatrix}, \tag{3.56}$$

and

$$\mathcal{L}_D^k = \begin{pmatrix} 0 & ik(kr)^2 j_n(kr)h_n^{(1)}(kr) \\ -ik\mathcal{J}_n(kr)\mathcal{H}_n(kr) & 0 \end{pmatrix}. \tag{3.57}$$

3.5.3 Asymptotic behavior of the spectrum of $\mathcal{W}_B(r)$

Now we consider the asymptotic expansions of the operator $\mathcal{W}_B(r)$ and its spectrum when $r \ll 1$.

It is well-known that, as $t \to 0$,

$$\begin{aligned} j_n(t) &= \frac{t^n}{(2n+1)!!}\Big(1 - \frac{1}{2(2n+3)}t^2 + O(t^4)\Big), \\ h_n^{(1)}(t) &= -i((2n-1)!!)t^{-n-1}\Big(1 + \frac{1}{2(2n-1)}t^2 + O(t^4)\Big). \end{aligned} \tag{3.58}$$

By making use of these asymptotics of the spherical Bessel functions, we obtain that

$$\begin{aligned} i\mathcal{J}_n(t)h_n^{(1)}(\tilde{t}) &= \frac{n+1}{2n+1}\Big(\frac{t}{\tilde{t}}\Big)^n\frac{1}{\tilde{t}} + \frac{n+1}{2(2n-1)(2n+1)}\Big(\frac{t}{\tilde{t}}\Big)^n\tilde{t} - \frac{n+3}{2(2n+1)(2n+3)}\Big(\frac{t}{\tilde{t}}\Big)^{n+1}t + O(t^3), \\ ij_n(t)\mathcal{H}_n(\tilde{t}) &= \frac{-n}{2n+1}\Big(\frac{t}{\tilde{t}}\Big)^n\frac{1}{\tilde{t}} + \frac{-n+2}{2(2n-1)(2n+1)}\Big(\frac{t}{\tilde{t}}\Big)^n\tilde{t} + \frac{n}{2(2n+1)(2n+3)}\Big(\frac{t}{\tilde{t}}\Big)^{n+1}t + O(t^3), \\ ij_n(t)h_n^{(1)}(\tilde{t}) &= \frac{1}{2n+1}\Big(\frac{t}{\tilde{t}}\Big)^n\frac{1}{\tilde{t}} + \frac{1}{2(2n-1)(2n+1)}\Big(\frac{t}{\tilde{t}}\Big)^n\tilde{t} - \frac{1}{2(2n+1)(2n+3)}\Big(\frac{t}{\tilde{t}}\Big)^{n+1}t + O(t^3), \\ i\mathcal{J}_n(t)\mathcal{H}_n(\tilde{t}) &= \frac{(-n)(n+1)}{2n+1}\Big(\frac{t}{\tilde{t}}\Big)^n\frac{1}{\tilde{t}} + \frac{(n+1)(-n+2)}{2(2n-1)(2n+1)}\Big(\frac{t}{\tilde{t}}\Big)^n\tilde{t} + \frac{n(n+3)}{2(2n+1)(2n+3)}\Big(\frac{t}{\tilde{t}}\Big)^{n+1}t \\ &\quad + O(t^3), \end{aligned} \tag{3.59}$$

for small $t, \tilde{t} \ll 1$ with $t \approx \tilde{t}$.

So, we have

$$\mathcal{M}_D^k = \begin{pmatrix} \frac{(-1)}{2(2n+1)} + (kr)^2 r_n & 0 \\ 0 & \frac{1}{2(2n+1)} + (kr)^2 s_n \end{pmatrix} + O(r^4), \tag{3.60}$$

and

$$\mathcal{L}_D^k = \begin{pmatrix} 0 & k^2 r p_n \\ \dfrac{n(n+1)}{2n+1}\dfrac{1}{r} + k^2 r q_n & 0 \end{pmatrix} + O(r^3), \tag{3.61}$$

where

$$\begin{aligned} p_n &= \frac{1}{2n+1}, \\ q_n &= \frac{(n+1)(n-2)}{2(2n-1)(2n+1)} - \frac{n(n+3)}{2(2n+1)(2n+3)}, \\ r_n &= -\frac{n+1}{2(2n-1)(2n+1)} + \frac{(n+3)}{2(2n+1)(2n+3)}, \\ s_n &= -\frac{n-2}{2(2n-1)(2n+1)} + \frac{n}{2(2n+1)(2n+3)}. \end{aligned} \tag{3.62}$$

Therefore, we can obtain

$$\mathcal{W}_B(r) = \mathcal{W}_{B,0} + r\mathcal{W}_{B,1} + r^2\mathcal{W}_{B,2} + O(r^3),$$

where

$$\mathcal{W}_{B,0} = \begin{pmatrix} \lambda_\mu - \dfrac{(-1)}{2(2n+1)} & 0 & 0 & 0 \\ 0 & \lambda_\mu - \dfrac{1}{2(2n+1)} & 0 & 0 \\ 0 & 0 & \lambda_\varepsilon - \dfrac{(-1)}{2(2n+1)} & 0 \\ 0 & 0 & 0 & \lambda_\varepsilon - \dfrac{1}{2(2n+1)} \end{pmatrix}, \tag{3.63}$$

$$\mathcal{W}_{B,1} = \begin{pmatrix} 0 & 0 & 0 & \omega C_\mu p_n \\ 0 & 0 & \omega C_\mu q_n & 0 \\ 0 & \omega C_\varepsilon p_n & 0 & 0 \\ \omega C_\varepsilon q_n & 0 & 0 & 0 \end{pmatrix}, \tag{3.64}$$

$$\mathcal{W}_{B,2} = \begin{pmatrix} \omega^2 D_\mu r_n & 0 & 0 & 0 \\ 0 & \omega^2 D_\mu s_n & 0 & 0 \\ 0 & 0 & \omega^2 D_\varepsilon r_n & 0 \\ 0 & 0 & 0 & \omega^2 D_\varepsilon s_n \end{pmatrix}, \tag{3.65}$$

and

$$C_\mu = \frac{\mu_c\varepsilon_c - \mu_m\varepsilon_m}{\mu_m - \mu_c}, \quad C_\varepsilon = \frac{\mu_c\varepsilon_c - \mu_m\varepsilon_m}{\varepsilon_m - \varepsilon_c}, \tag{3.66}$$

$$D_\mu = \frac{\varepsilon_c\mu_c^2 - \varepsilon_m\mu_m^2}{\mu_m - \mu_c}, \quad D_\varepsilon = \frac{\varepsilon_c^2\mu_c - \varepsilon_m^2\mu_m}{\varepsilon_m - \varepsilon_c}. \tag{3.67}$$

By applying the standard perturbation theory, the asymptotics of eigenvalues of $\mathcal{W}_B(r)$ are obtained as follows: up to an error term of order $O(r^3)$,

$$\lambda_\mu - \frac{(-1)}{2(2n+1)} + (r\omega)^2\left[C_\varepsilon C_\mu \frac{p_n q_n}{\lambda_\mu - \lambda_\varepsilon + p_n} + D_\mu r_n\right] + O(r^3),$$
$$\lambda_\mu - \frac{1}{2(2n+1)} + (r\omega)^2\left[C_\varepsilon C_\mu \frac{p_n q_n}{\lambda_\mu - \lambda_\varepsilon - p_n} + D_\mu s_n\right] + O(r^3),$$
$$\lambda_\varepsilon - \frac{(-1)}{2(2n+1)} + (r\omega)^2\left[C_\varepsilon C_\mu \frac{p_n q_n}{\lambda_\varepsilon - \lambda_\mu + p_n} + D_\varepsilon r_n\right] + O(r^3),$$
$$\lambda_\varepsilon - \frac{1}{2(2n+1)} + (r\omega)^2\left[C_\varepsilon C_\mu \frac{p_n q_n}{\lambda_\varepsilon - \lambda_\mu - p_n} + D_\varepsilon s_n\right] + O(r^3),$$

and the asymptotics of the associated eigenfunction are given by

$$[1,0,0,0]^T + r\omega \frac{C_\varepsilon q_n}{\lambda_\mu - \lambda_\varepsilon + p_n}[0,0,0,1]^T + O(r^2),$$
$$[0,1,0,0]^T + r\omega \frac{C_\varepsilon}{2n+1}\frac{1}{\lambda_\mu - \lambda_\varepsilon - p_n}[0,0,1,0]^T + O(r^2),$$
$$[0,0,1,0]^T + r\omega \frac{C_\mu q_n}{\lambda_\varepsilon - \lambda_\mu + p_n}[0,1,0,0]^T + O(r^2),$$
$$[0,0,0,1]^T + r\omega \frac{C_\mu}{2n+1}\frac{1}{\lambda_\varepsilon - \lambda_\mu - p_n}[1,0,0,0]^T + O(r^2).$$

3.5.4 Extinction cross-section

In this subsection, we compute the extinction cross-section Q^{ext}. We need the following lemma.

Lemma 3.5.2. *Let D be a sphere with radius $r > 0$ and suppose that E^i is given by*

$$E^i(x) = \sum_{n=1}^{\infty}\sum_{l=-n}^{n} \alpha_{nl}^{TE}\widetilde{E}_{n,l}^{TE}(x;k_m) + \alpha_{nl}^{TM}\widetilde{E}_{n,l}^{TM}(x;k_m),$$

for some coefficients $\alpha_{nl}^{TE}, \alpha_{nl}^{TM}$. Then the scattered wave can be represented as follows: for $|x| > r$,

$$E^s(x) = \sum_{n=1}^{\infty}\sum_{l=-n}^{n} \alpha_{nl}^{TE}S_n^{TE}E_{n,l}^{TE}(x;k_m) + \alpha_{nl}^{TM}S_n^{TM}E_{n,l}^{TM}(x;k_m),$$

where S_n^{TE} and S_n^{TM} are given by

$$S_n^{TE} = \frac{\mu_c j_n(k_c r)\mathcal{J}_n(k_m r) - \mu_m j_n(k_m r)\mathcal{J}_n(k_c r)}{\mu_m \mathcal{J}_n(k_c r)h_n^{(1)}(k_m r) - \mu_c j_n(k_c r)\mathcal{H}(k_m r)},$$
$$S_n^{TM} = \frac{\varepsilon_c j_n(k_c r)\mathcal{J}_n(k_m r) - \varepsilon_m j_n(k_m r)\mathcal{J}_n(k_c r)}{\varepsilon_m \mathcal{J}_n(k_c r)h_n^{(1)}(k_m r) - \varepsilon_c j_n(k_c r)\mathcal{H}(k_m r)}.$$

Proof. Let $E^i = \widetilde{E}^{TE}_{n,l}(x;k_m)$. We look for a solution of the following form:

$$E = \begin{cases} a\,\widetilde{E}^{TE}_{n,l}(x;k_c), & |x| < r \\ \widetilde{E}^{TE}_{n,l}(x;k_m) + b\,E^{TE}_{n,l}(x;k_m), & |x| > r. \end{cases}$$

Then, from the boundary condition on ∂D, we easily see that

$$\begin{pmatrix} j_n(k_m r) \\ \frac{1}{\mu_m}\mathcal{J}_n(k_m r) \end{pmatrix} = \begin{pmatrix} j_n(k_c r) & -h^{(1)}_n(k_m r) \\ \frac{1}{\mu_c}\mathcal{J}_n(k_c r) & -\frac{1}{\mu_m}\mathcal{H}_n(k_m r) \end{pmatrix} \begin{pmatrix} a \\ b \end{pmatrix}. \tag{3.68}$$

Therefore, the coefficient a and b can be obtained as follows:

$$\begin{aligned} \begin{pmatrix} 1/a \\ b/a \end{pmatrix} &= \begin{pmatrix} j_n(k_m r) & h^{(1)}_n(k_m r) \\ \frac{1}{\mu_m}\mathcal{J}_n(k_m r) & \frac{1}{\mu_m}\mathcal{H}_n(k_m r) \end{pmatrix}^{-1} \begin{pmatrix} j_n(k_c r) & h^{(1)}_n(k_c r) \\ \frac{1}{\mu_c}\mathcal{J}_n(k_c r) & \frac{1}{\mu_c}\mathcal{H}_n(k_c r) \end{pmatrix} \begin{pmatrix} 1 \\ 0 \end{pmatrix}, \\ &= \frac{\mu_m k_m r}{i} \begin{pmatrix} \frac{1}{\mu_m}\mathcal{H}_n(k_m r) & -h^{(1)}_n(k_m r) \\ -\frac{1}{\mu_m}\mathcal{J}_n(k_m r) & j_n(k_m r) \end{pmatrix} \begin{pmatrix} j_n(k_c r) \\ \frac{1}{\mu_c}\mathcal{J}_n(k_c r) \end{pmatrix}, \\ &= -ik_m r \begin{pmatrix} \mathcal{H}_n(k_m r) j_n(k_c r) - \dfrac{\mu_m}{\mu_c} h^{(1)}_n(k_m r)\mathcal{J}_n(k_c r) \\ -\mathcal{J}_n(k_m r) j_n(k_c r) + \dfrac{\mu_m}{\mu_c} j_n(k_m r)\mathcal{J}_n(k_c r) \end{pmatrix}, \end{aligned} \tag{3.69}$$

where we have used the following Wronskian identity for the spherical Bessel function:

$$j_n(t)\mathcal{H}_n(t) - h^{(1)}_n(t)\mathcal{J}_n(t) = t\Big(j_n(t)(h^{(1)}_n)'(t) - j_n'(t)h^{(1)}_n(t)\Big) = \frac{i}{t}.$$

Therefore, we immediately see that

$$b = \frac{\mu_c j_n(k_c r)\mathcal{J}_n(k_m r) - \mu_m j_n(k_m r)\mathcal{J}_n(k_c r)}{\mu_m \mathcal{J}_n(k_c r) h^{(1)}_n(k_m r) - \mu_c j_n(k_c r)\mathcal{H}(k_m r)}.$$

Now suppose that $E^i = \widetilde{E}^{TM}_{n,l}(x;k_m)$. We look for a solution in the following form:

$$E = \begin{cases} c\,\widetilde{E}^{TM}_{n,l}(x;k_c), & |x| < r, \\ \widetilde{E}^{TM}_{n,l}(x;k_m) + d\,E^{TM}_{n,l}(x;k_m), & |x| > r. \end{cases}$$

Then, from the boundary conditions on $|x| = r$, we obtain

$$\begin{pmatrix} \frac{1}{\varepsilon_c}\mathcal{J}_n(k_c r) & \frac{1}{\varepsilon_c}\mathcal{H}_n(k_c r) \\ j_n(k_c r) & h^{(1)}_n(k_c r) \end{pmatrix} \begin{pmatrix} c \\ 0 \end{pmatrix} = \begin{pmatrix} \frac{1}{\varepsilon_m}\mathcal{J}_n(k_m r) & \frac{1}{\varepsilon_m}\mathcal{H}_n(k_m r) \\ j_n(k_m r) & h^{(1)}_n(k_m r) \end{pmatrix} \begin{pmatrix} 1 \\ d \end{pmatrix}. \tag{3.70}$$

By solving (3.70), we get

$$d = \frac{\varepsilon_c j_n(k_c r)\mathcal{J}_n(k_m r) - \varepsilon_m j_n(k_m r)\mathcal{J}_n(k_c r)}{\varepsilon_m \mathcal{J}_n(k_c r) h^{(1)}_n(k_m r) - \varepsilon_c j_n(k_c r)\mathcal{H}(k_m r)}.$$

By the principle of superposition, the conclusion immediately follows.

□

We also need the following lemma concerning the scattering amplitude A_∞.

Lemma 3.5.3. *Suppose that the scattered electric field E^s is given by*

$$E^s(x) = \sum_{n=1}^{\infty} \sum_{l=-n}^{n} \beta_{nl}^{TE} E_{n,l}^{TE}(x; k_m) + \beta_{nl}^{TM} E_{n,l}^{TM}(x; k_m)$$

for $\mathbb{R}^3 \setminus \overline{D}$. Then the scattering amplitude A_∞ can be represented as follows:

$$A_\infty(\hat{x}) = \sum_{n=1}^{\infty} \sum_{l=-n}^{n} \frac{4\pi(-i)^n}{ik_m} \sqrt{n(n+1)} \left(\beta_{nl}^{TE} V_{n,l}(\hat{x}) + \sqrt{\frac{\mu_m}{\varepsilon_m}} \beta_{nl}^{TM} U_{n,l} \right).$$

Proof. It is well-known that

$$h_n^{(1)}(t) \sim \frac{1}{t} e^{it} e^{-i\frac{n+1}{2}\pi} \quad \text{as } t \to \infty,$$

and

$$(h_n^{(1)})'(t) \sim \frac{1}{t} e^{it} e^{-i\frac{n}{2}\pi} \quad \text{as } t \to \infty.$$

Then one can easily see that as $|x| \to \infty$,

$$E_{n,m}^{TE}(x; k_m) \sim -\frac{e^{ik_m|x|}}{k_m|x|} e^{-i\frac{n+1}{2}\pi} \sqrt{n(n+1)} V_{n,l}(\hat{x})$$

and

$$E_{n,m}^{TM}(x; k_m) \sim -\frac{e^{ik_m|x|}}{k_m|x|} \sqrt{\frac{\mu_m}{\varepsilon_m}} e^{-i\frac{n+1}{2}\pi} \sqrt{n(n+1)} U_{n,l}(\hat{x}).$$

By applying these asymptotics to the series expansion of E^s, the conclusion follows.

□

A plane wave can be represented as a series expansion. The following lemma is proved in [66].

Lemma 3.5.4. *Let E^i be a plane wave, that is, $E^i(x) = p\, e^{ik_m d\cdot x}$ with $d \in S$ and $p \cdot d = 0$. Then we have the following series representation for a plane wave as follows:*

$$E^i(x) = \sum_{n=1}^{\infty} \sum_{l=-n}^{n} \alpha_{nl}^{pw,TE} \widetilde{E}_{n,l}^{TE}(x; k_m) + \alpha_{nl}^{pw,TM} \widetilde{E}_{n,l}^{TM}(x; k_m),$$

where

$$\begin{cases} \alpha_{nl}^{pw,TE} = \dfrac{(-1)4\pi i^n}{\sqrt{n(n+1)}} i\big(V_{n,l}(d) \cdot p\big), \\ \alpha_{nl}^{pw,TM} = \dfrac{(-1)4\pi i^n}{\sqrt{n(n+1)}} \sqrt{\dfrac{\varepsilon_m}{\mu_m}} \big(U_{n,l}(d) \cdot p\big). \end{cases}$$

Now we are ready to compute the extinction cross-section Q^{ext}.

Theorem 3.5.1. *Assume that $E^i(x) = p\, e^{ik_m d\cdot x}$ with $d \in S$ and $p \cdot d = 0$. Let D be a sphere with radius r. Then the extinction cross-section is given*

by

$$Q^{ext} = \sum_{n=1}^{\infty}\sum_{l=-n}^{n} \frac{(4\pi)^3}{k_m^2|p|^2}\Im\left((-1)S_n^{TE}(V_{n,l}(d)\cdot p)^2 + iS_n^{TM}(U_{n,l}(d)\cdot p)^2\right).$$

Moreover, for small $r > 0$, we have

$$Q^{ext} = \sum_{l=-1}^{1} \frac{(-1)(4\pi k_m r)^3}{k_m^2|p|^2}\Im\left(i\frac{2}{3}\frac{\mu_c-\mu_m}{2\mu_m+\mu_c}(V_{1,l}(d)\cdot p)^2 + \frac{2}{3}\frac{\varepsilon_c-\varepsilon_m}{2\varepsilon_m+\varepsilon_c}(U_{1,l}(d)\cdot p)^2\right) + O((k_m r)^4).$$

Proof. Let us first compute the scattering amplitude A_∞ when E^i is a plane wave. From

$$\begin{aligned} A_\infty(\hat{x}) &= \sum_{n=1}^{\infty}\sum_{l=-n}^{n} \frac{4\pi(-i)^n}{ik_m}\sqrt{n(n+1)} \\ &\quad\times\left(\alpha_{nl}^{pw,TE}S_n^{TE}V_{n,l}(\hat{x}) + \sqrt{\frac{\mu_m}{\varepsilon_m}}\alpha_{nl}^{pw,TM}S_n^{TM}U_{n,l}\right) \\ &= \sum_{n=1}^{\infty}\sum_{l=-n}^{n} \frac{(4\pi)^2}{k_m}\left((-1)S_n^{TE}(V_{n,l}(d)\cdot p)V_{n,l} + iS_n^{TM}(U_{n,l}(d)\cdot p)U_{n,l}\right). \end{aligned}$$

Therefore, we have

$$\begin{aligned} Q^{ext} &= \frac{4\pi}{k_m}\Im\left[\frac{p\cdot A_\infty(d)}{|p|^2}\right] \\ &= \sum_{n=1}^{\infty}\sum_{l=-n}^{n} \frac{(4\pi)^3}{k_m^2|p|^2}\Im\left((-1)S_n^{TE}(V_{n,l}(d)\cdot p)^2 + iS_n^{TM}(U_{n,l}(d)\cdot p)^2\right). \end{aligned}$$

Now we assume that $r \ll 1$. By applying (3.58), one can easily see that

$$\begin{aligned} S_1^{TE} &= i\frac{2}{3}\frac{(\mu_c-\mu_m)(k_m r)^3}{2\mu_m+\mu_c} + O(r^4), \\ S_1^{TM} &= i\frac{2}{3}\frac{(\varepsilon_c-\varepsilon_m)(k_m r)^3}{2\varepsilon_m+\varepsilon_c} + O(r^4), \\ S_n^{TE}, S_n^{TM} &= O(r^4), \quad \text{for } n \geq 2. \end{aligned}$$

Therefore, we obtain, up to an error term of order $O(r^4)$,

$$Q^{ext} = \sum_{l=-1}^{1} \frac{(-1)(4\pi)^3}{k_m^2|p|^2}\Im\left(i\frac{2}{3}\frac{(\mu_c-\mu_m)(k_m r)^3}{2\mu_m+\mu_c}(V_{1,l}(d)\cdot p)^2 + \frac{2}{3}\frac{(\varepsilon_c-\varepsilon_m)(k_m r)^3}{2\varepsilon_m+\varepsilon_c}(U_{1,l}(d)\cdot p)^2\right).$$

The proof is complete. □

3.6 Explicit computations for a spherical shell

In this section we consider a spherical shell. Since, in this case, the eigenvalues associated with the corresponding Neumann-Poincaré operator are not

simple, we apply degenerate perturbation theory in order to compute the first order effect of the size.

3.6.1 Explicit representation of boundary integral operators

Let D_s and D_c be a spherical shell with radius r_s and r_c with $r_s > r_c > 0$. Let

$$(\varepsilon, \mu) = \begin{cases} (\varepsilon_m, \mu_m) & \text{in } D_c, \\ (\varepsilon_s, \mu_s) & \text{in } D_s \setminus \bar{D}_c, \\ (\varepsilon_m, \mu_m) & \text{in } \mathbb{R}^3 \setminus \bar{D}_s. \end{cases}$$

Let

$$\rho = \frac{r_c}{r_s}.$$

The solution to the transmission problem can be represented as follows

$$E(x) = \begin{cases} \mu_c \nabla \times \vec{\mathcal{S}}^{k_c}_{D_s}[\psi_s](x) + \nabla \times \nabla \times \vec{\mathcal{S}}^{k_c}_{D_s}[\phi_s](x) \\ \qquad +\mu_c \nabla \times \vec{\mathcal{S}}^{k_c}_{D_c}[\psi_c](x) + \nabla \times \nabla \times \vec{\mathcal{S}}^{k_c}_{D_c}[\phi_c](x) & x \in D_c, \\ \mu_s \nabla \times \vec{\mathcal{S}}^{k_s}_{D_s}[\psi_s](x) + \nabla \times \nabla \times \vec{\mathcal{S}}^{k_s}_{D_s}[\phi_s](x) \\ \qquad +\mu_s \nabla \times \vec{\mathcal{S}}^{k_s}_{D_c}[\psi_c](x) + \nabla \times \nabla \times \vec{\mathcal{S}}^{k_s}_{D_c}[\phi_c](x) & x \in D_s \setminus \bar{D}_c, \\ E^i + \mu_m \nabla \times \vec{\mathcal{S}}^{k_m}_{D_s}[\psi_s](x) + \nabla \times \nabla \times \vec{\mathcal{S}}^{k_m}_{D_s}[\phi_s](x) \\ \qquad +\mu_m \nabla \times \vec{\mathcal{S}}^{k_m}_{D_c}[\psi_c](x) + \nabla \times \nabla \times \vec{\mathcal{S}}^{k_m}_{D_c}[\phi_c](x) & x \in \mathbb{R}^3 \setminus \bar{D}_s, \end{cases} \tag{3.71}$$

and

$$H(x) = -\frac{i}{\omega \mu_D}(\nabla \times E)(x) \quad x \in \mathbb{R}^3 \setminus \partial D, \tag{3.72}$$

where the pair $(\psi_s, \phi_s, \psi_c, \phi_c) \in \left(H_T^{-\frac{1}{2}}(\mathrm{div}, \partial D_s)\right)^2 \times \left(H_T^{-\frac{1}{2}}(\mathrm{div}, \partial D_c)\right)^2$ is the unique solution to

$$W^{sh} \begin{pmatrix} \psi_s \\ \phi_s \\ \psi_c \\ \phi_c \end{pmatrix} := \begin{pmatrix} W^{sh}_{11} & W^{sh}_{12} \\ W^{sh}_{21} & W^{sh}_{22} \end{pmatrix} \begin{pmatrix} \psi_s \\ \phi_s \\ \psi_c \\ \phi_c \end{pmatrix} = \begin{pmatrix} \nu \times E^i \\ i\omega \nu \times H^i \\ 0 \\ 0 \end{pmatrix}$$

with

$$W^{sh}_{11} = \begin{pmatrix} \frac{\mu_s + \mu_m}{2} Id + \mu_s \mathcal{M}^{k_s}_{D_s} - \mu_m \mathcal{M}^{k_m}_{D_s} & \mathcal{L}^{k_s}_{D_s} - \mathcal{L}^{k_m}_{D_s} \\ \mathcal{L}^{k_s}_{D_s} - \mathcal{L}^{k_m}_{D_s} & \left(\frac{k_s^2}{2\mu_s} + \frac{k_m^2}{2\mu_m}\right) Id + \frac{k_s^2}{\mu_s}\mathcal{M}^{k_s}_{D_s} - \frac{k_m^2}{\mu_m}\mathcal{M}^{k_m}_{D_s} \end{pmatrix}, \tag{3.73}$$

$$W^{sh}_{12} = \begin{pmatrix} \mu_s \nu \times \nabla \times \vec{\mathcal{S}}^{k_s}_{D_c} - \mu_m \nu \times \nabla \times \vec{\mathcal{S}}^{k_m}_{D_c} & \nu \times \nabla \times \nabla \times \vec{\mathcal{S}}^{k_s}_{D_c} - \nu \times \nabla \times \nabla \times \vec{\mathcal{S}}^{k_m}_{D_c} \\ \nu \times \nabla \times \nabla \times \vec{\mathcal{S}}^{k_s}_{D_c} - \nu \times \nabla \times \nabla \times \vec{\mathcal{S}}^{k_m}_{D_c} & \frac{k_s^2}{\mu_s}\nu \times \nabla \times \vec{\mathcal{S}}^{k_s}_{D_c} - \frac{k_m^2}{\mu_m}\nu \times \nabla \times \vec{\mathcal{S}}^{k_m}_{D_c} \end{pmatrix}, \tag{3.74}$$

$$W_{21}^{sh}=\begin{pmatrix} -\mu_c\nu\times\nabla\times\vec{\mathcal{S}}_{D_s}^{k_c}+\mu_s\nu\times\nabla\times\vec{\mathcal{S}}_{D_s}^{k_s} & -\nu\times\nabla\times\nabla\times\vec{\mathcal{S}}_{D_s}^{k_c}+\nu\times\nabla\times\nabla\times\vec{\mathcal{S}}_{D_s}^{k_s} \\ -\nu\times\nabla\times\nabla\times\vec{\mathcal{S}}_{D_s}^{k_c}+\nu\times\nabla\times\nabla\times\vec{\mathcal{S}}_{D_s}^{k_s} & -\frac{k_c^2}{\mu_c}\nu\times\nabla\times\vec{\mathcal{S}}_{D_s}^{k_c}+\frac{k_s^2}{\mu_s}\nu\times\nabla\times\vec{\mathcal{S}}_{D_s}^{k_s} \end{pmatrix}, \tag{3.75}$$

$$W_{22}^{sh}=\begin{pmatrix} -\frac{\mu_c+\mu_s}{2}Id-\mu_c\mathcal{M}_{D_c}^{k_c}+\mu_s\mathcal{M}_{D_c}^{k_s} & -\mathcal{L}_{D_c}^{k_c}+\mathcal{L}_{D_c}^{k_s} \\ -\mathcal{L}_{D_c}^{k_c}+\mathcal{L}_{D_c}^{k_s} & -\left(\frac{k_c^2}{2\mu_c}+\frac{k_s^2}{2\mu_s}\right)Id-\frac{k_c^2}{\mu_c}\mathcal{M}_{D_c}^{k_c}+\frac{k_s^2}{\mu_s}\mathcal{M}_{D_c}^{k_s} \end{pmatrix}. \tag{3.76}$$

Note that W_{11}^{sh} and W_{22}^{sh} are similar to the operator in left-hand side of (3.13). In the previous section for the sphere case, we have already obtained the matrix representation of this operator and its asymptotic expansion.

By Lemma 3.5.1, we can represent $\nu\times\nabla\times\vec{\mathcal{S}}_D^k|_{|x|=r'}$ and $\nu\times\nabla\times\nabla\times\vec{\mathcal{S}}_D^k|_{|x|=r'}$ in a matrix form as follows(using $U_{n,m},V_{n,m}$ as basis):
(i) For $r'>r$,

$$\nu\times\nabla\times\vec{\mathcal{S}}_D^k|_{|x|=r'}=\begin{pmatrix} (-ikr)\mathcal{J}_n(kr)h_n^{(1)}(kr') & 0 \\ 0 & ik\frac{r^2}{r'}j_n(kr)\mathcal{H}_n(kr') \end{pmatrix}, \tag{3.77}$$

$$\nu\times\nabla\times\nabla\times\vec{\mathcal{S}}_D^k|_{|x|=r'}=\begin{pmatrix} 0 & ik(kr)^2j_n(kr)h_n^{(1)}(kr') \\ -ik\frac{r}{r'}\mathcal{J}_n(kr)\mathcal{H}_n(kr') & 0 \end{pmatrix}; \tag{3.78}$$

(ii) For $r'<r$,

$$\nu\times\nabla\times\vec{\mathcal{S}}_D^k|_{|x|=r'}=\begin{pmatrix} (-ikr)j_n(kr')\mathcal{H}_n(kr) & 0 \\ 0 & ik\frac{r^2}{r'}\mathcal{J}_n(kr')\langle_n^{(1)}(kr) \end{pmatrix}, \tag{3.79}$$

$$\nu\times\nabla\times\nabla\times\vec{\mathcal{S}}_D^k|_{|x|=r'}=\begin{pmatrix} 0 & ik(kr)^2j_n(kr')h_n^{(1)}(kr) \\ -ik\frac{r}{r'}\mathcal{J}_n(kr')\mathcal{H}_n(kr) & 0 \end{pmatrix}. \tag{3.80}$$

Using the above formulas, the matrix representation of the operators W_{12}^{sh} and W_{21}^{sh} can be easily obtained.

We now consider scaling of W^{sh}. First, we need some definitions. Let $D_s=z+r_sB_s$ where B_s contains the origin and $|B_s|=O(1)$. Let B_c be defined in a similar way. For any $x\in\partial D_s$ (or ∂D_c), let $\widetilde{x}=\frac{x-z}{r_s}\in\partial B_s$ (or ∂B_c with r_s replaced by r_c) and define for each function f defined on ∂D_s (or ∂D_c), a corresponding function defined on B as follows

$$\eta_s(f)(\widetilde{x})=f(z+r_s\widetilde{x}),\quad \eta_c(f)(\widetilde{x})=f(z+r_c\widetilde{x}). \tag{3.81}$$

Then, in a similar way to the sphere case, let us write

$$W_B^{sh}(r_s)\begin{pmatrix}\eta_s(\psi_s)\\ \omega\eta_s(\phi_s)\\ \eta_c(\psi_c)\\ \omega\eta_c(\phi_c)\end{pmatrix}=\begin{pmatrix}\frac{\eta(\nu\times E^i)}{\mu_m-\mu_s}\\ \frac{\eta(i\nu\times H^i)}{\varepsilon_m-\varepsilon_s}\\ 0\\ 0\end{pmatrix}.$$

Using $(U_{n,m},V_{n,m},U_{n,m},V_{n,m})\times(U_{n,m},V_{n,m},U_{n,m},V_{n,m})$ as basis, we can represent $W_B^{sh}(r_s)$ in a 8×8 matrix form in a subspace $H_{n,m}(\partial B_s)\times H_{n,m}(\partial B_c)$. Then, by using (3.59), their asymptotic expansion can also be obtained.

Here, the resulting asymptotics of the matrix W_B^{sh} are given as follows. Write

$$\mathcal{W}_B^{sh}(r_s)=\mathcal{W}_{B,0}^{sh}+r_s\mathcal{W}_{B,1}^{sh}+r_s^2\mathcal{W}_{B,2}^{sh}+O(r_s^3), \tag{3.82}$$

where

$$\mathcal{W}_{B,0}^{sh}=\begin{pmatrix}\Lambda_{\mu,\varepsilon} & \\ & \Lambda_{\mu,\varepsilon}\end{pmatrix}+\begin{pmatrix}P_{0,n} & Q_{0,n}\\ R_{0,n} & -P_{0,n}\end{pmatrix}, \tag{3.83}$$

$$\mathcal{W}_{B,1}^{sh}=\begin{pmatrix}P_{1,n} & Q_{1,n}\\ R_{1,n} & -P_{1,n}\end{pmatrix},\quad \mathcal{W}_{B,2}^{sh}=\begin{pmatrix}P_{2,n} & Q_{2,n}\\ R_{2,n} & -P_{2,n}\end{pmatrix}.$$

Here, the matrix $P_{j,n},Q_{j,n}$ and $R_{j,n}$ are given by

$$\Lambda_{\mu,\varepsilon}=\begin{pmatrix}\lambda_\mu & & & \\ & \lambda_\mu & & \\ & & \lambda_\varepsilon & \\ & & & \lambda_\varepsilon\end{pmatrix},\quad P_{0,n}=\begin{pmatrix}p_n & & & \\ & -p_n & & \\ & & p_n & \\ & & & -p_n\end{pmatrix},$$

$$Q_{0,n}=\rho^2\begin{pmatrix}g_n & & & \\ & f_n & & \\ & & g_n & \\ & & & f_n\end{pmatrix},\quad R_{0,n}=\begin{pmatrix}f_n & & & \\ & g_n & & \\ & & f_n & \\ & & & g_n\end{pmatrix},$$

$$P_{1,n}=\omega\begin{pmatrix} & & C_\mu p_n & \\ & & & C_\mu q_n\\ C_\varepsilon q_n & & & \\ & C_\varepsilon p_n & & \end{pmatrix},\quad P_{2,n}=\omega^2\begin{pmatrix}D_\mu r_n & & & \\ & D_\mu s_n & & \\ & & D_\varepsilon r_n & \\ & & & D_\varepsilon s_n\end{pmatrix},$$

$$Q_{1,n}=\omega\rho\begin{pmatrix} & & C_\mu\tilde{p}_n & \\ & & & C_\mu\tilde{q}_n\\ C_\varepsilon\tilde{q}_n & & & \\ & C_\varepsilon\tilde{p}_n & & \end{pmatrix},\quad Q_{2,n}=\omega^2\rho\begin{pmatrix}D_\mu\tilde{r}_n & & & \\ & D_\mu\tilde{s}_n & & \\ & & D_\varepsilon\tilde{r}_n & \\ & & & D_\varepsilon\tilde{s}_n\end{pmatrix},$$

$$R_{1,n}=-\omega\rho^{-1}\begin{pmatrix} & & C_\mu\tilde{p}_n & \\ & & & C_\mu\tilde{q}_n\\ C_\varepsilon\tilde{q}_n & & & \\ & C_\varepsilon\tilde{p}_n & & \end{pmatrix},$$

$$R_{2,n} = \omega^2 \rho^{-1} \begin{pmatrix} D_\mu \tilde{s}_n & & & \\ & -D_\mu \tilde{r}_n & & \\ & & D_\varepsilon \tilde{s}_n & \\ & & & -D_\varepsilon \tilde{r}_n \end{pmatrix}.$$

Here, p_n, q_n, r_n, s_n are defined as (3.62) and $\tilde{p}_n, \tilde{q}_n, \tilde{r}_n, \tilde{s}_n, D_\mu$ and D_ε are defined as follows:

$$f_n = \rho^n \frac{n}{2n+1}, \quad g_n = \rho^{n-1} \frac{n+1}{2n+1}, \tag{3.84}$$

$$\tilde{p}_n = \frac{1}{2n+1} \rho^{n+1}, \tag{3.85}$$

$$\tilde{q}_n = \frac{(n+1)(n-2)}{2(2n-1)(2n+1)} \rho^n - \frac{n(n+3)}{2(2n+1)(2n+3)} \rho^{n+2}, \tag{3.86}$$

$$\tilde{r}_n = -\frac{n+1}{2(2n-1)(2n+1)} \rho^n + \frac{(n+3)}{2(2n+1)(2n+3)} \rho^{n+2}, \tag{3.87}$$

$$\tilde{s}_n = -\frac{n-2}{2(2n-1)(2n+1)} \rho^{n+1} + \frac{n}{2(2n+1)(2n+3)} \rho^{n+3}, \tag{3.88}$$

and

$$D_\mu = \frac{\varepsilon_s \mu_s^2 - \varepsilon_m \mu_m^2}{\mu_m - \mu_s}, \quad D_\varepsilon = \frac{\varepsilon_s^2 \mu_s - \varepsilon_m^2 \mu_m}{\varepsilon_m - \varepsilon_s}. \tag{3.89}$$

3.6.2 Asymptotic behavior of the spectrum of $\mathcal{W}_B^{sh}(r_s)$

Let us define

$$\lambda_n^{sh} = \frac{1}{2(2n+1)} \sqrt{1 + 4n(n+1)\rho^{2n+1}}.$$

Note that $\pm\lambda_n^{sh}$ are eigenvalues of the Neumann-Poincaré operator on the shell.

It turns out that the eigenvalues of $W_{B,0}^{sh}$ are as follows

$$\lambda_\mu + \lambda_n^{sh}, \quad \lambda_\mu - \lambda_n^{sh}, \quad \lambda_\varepsilon + \lambda_n^{sh}, \quad \lambda_\varepsilon - \lambda_n^{sh},$$

for $n = 0, 1, 2, ...$, and their multiplicities is 2. Their associated eigenfunctions are as follows:

$$\begin{aligned}
\lambda_\mu + \lambda_n^{sh} \quad &\longrightarrow \quad E_1^0 := (\lambda_n^{sh} + p_n)\mathbf{e}_1 + f_n \mathbf{e}_5, \quad E_2^0 := (\lambda_n^{sh} - p_n)\mathbf{e}_2 + g_n \mathbf{e}_6, \\
\lambda_\mu - \lambda_n^{sh} \quad &\longrightarrow \quad E_3^0 := (-\lambda_n^{sh} + p_n)\mathbf{e}_1 + f_n \mathbf{e}_5, \quad E_4^0 := (-\lambda_n^{sh} - p_n)\mathbf{e}_2 + g_n \mathbf{e}_6, \\
\lambda_\varepsilon + \lambda_n^{sh} \quad &\longrightarrow \quad E_5^0 := (\lambda_n^{sh} + p_n)\mathbf{e}_3 + f_n \mathbf{e}_7, \quad E_6^0 := (\lambda_n^{sh} - p_n)\mathbf{e}_4 + g_n \mathbf{e}_8, \\
\lambda_\varepsilon - \lambda_n^{sh} \quad &\longrightarrow \quad E_7^0 := (-\lambda_n^{sh} + p_n)\mathbf{e}_3 + f_n \mathbf{e}_7, \quad E_8^0 := (-\lambda_n^{sh} - p_n)\mathbf{e}_4 + g_n \mathbf{e}_8,
\end{aligned}$$

where $\{\mathbf{e}_i\}_{i=1}^8$ is standard unit basis in $\mathbb{R}^8$.

To derive asymptotic expansions of the eigenvalues, we apply degenerate eigenvalue perturbation theory (since the multiplicity of each of these

eigenvalues is 2). To state the result, we need some definitions. Let

$$T_{16,n} = C_\varepsilon \frac{(\lambda_n^{sh} - p_n)a_{1,n} - b_{1,n}}{|E_1^0||E_6^0|}, \quad T_{18,n} = C_\varepsilon \frac{(-\lambda_n^{sh} - p_n)a_{1,n} - b_{1,n}}{|E_1^0||E_8^0|},$$
$$T_{25,n} = C_\varepsilon \frac{(\lambda_n^{sh} + p_n)a_{2,n} - b_{2,n}}{|E_2^0||E_5^0|}, \quad T_{27,n} = C_\varepsilon \frac{(-\lambda_n^{sh} + p_n)a_{2,n} - b_{2,n}}{|E_2^0||E_7^0|},$$
$$T_{36,n} = C_\varepsilon \frac{(\lambda_n^{sh} - p_n)a_{3,n} - b_{3,n}}{|E_3^0||E_6^0|}, \quad T_{38,n} = C_\varepsilon \frac{(-\lambda_n^{sh} - p_n)a_{3,n} - b_{3,n}}{|E_3^0||E_8^0|},$$
$$T_{45,n} = C_\varepsilon \frac{(\lambda_n^{sh} + p_n)a_{4,n} - b_{4,n}}{|E_4^0||E_5^0|}, \quad T_{47,n} = C_\varepsilon \frac{(-\lambda_n^{sh} + p_n)a_{4,n} - b_{4,n}}{|E_4^0||E_7^0|},$$
$$T_{52,n} = \frac{C_\mu}{C_\varepsilon} T_{16,n}, \quad T_{54,n} = \frac{C_\mu}{C_\varepsilon} T_{18,n}, \quad T_{61,n} = \frac{C_\mu}{C_\varepsilon} T_{25,n}, \quad T_{63,n} = \frac{C_\mu}{C_\varepsilon} T_{27,n},$$
$$T_{72,n} = \frac{C_\mu}{C_\varepsilon} T_{36,n}, \quad T_{74,n} = \frac{C_\mu}{C_\varepsilon} T_{38,n}, \quad T_{81,n} = \frac{C_\mu}{C_\varepsilon} T_{45,n}, \quad T_{83,n} = \frac{C_\mu}{C_\varepsilon} T_{47,n},$$

where

$$a_{1,n} = (\lambda_n^{sh} + p_n)q_n + \rho f_n \tilde{q}_n,$$
$$a_{2,n} = (\lambda_n^{sh} - p_n)p_n + \rho g_n \tilde{p}_n,$$
$$a_{3,n} = (-\lambda_n^{sh} + p_n)q_n + \rho f_n \tilde{q}_n,$$
$$a_{4,n} = (-\lambda_n^{sh} - p_n)p_n + \rho g_n \tilde{p}_n,$$

and

$$b_{1,n} = f_n g_n q_n + \rho^{-1}(\lambda_n^{sh} + p_n) g_n \tilde{q}_n,$$
$$b_{2,n} = f_n g_n p_n + \rho^{-1}(\lambda_n^{sh} - p_n) f_n \tilde{p}_n,$$
$$b_{3,n} = f_n g_n q_n + \rho^{-1}(-\lambda_n^{sh} + p_n) g_n \tilde{q}_n,$$
$$b_{4,n} = f_n g_n p_n + \rho^{-1}(-\lambda_n^{sh} - p_n) f_n \tilde{p}_n.$$

We also define

$$K_{1,n} = D_\mu \frac{(\lambda_n^{sh} + p_n)((\lambda_n^{sh} + p_n)r_n + \rho f_n \tilde{r}_n) + f_n((\lambda_n^{sh} + p_n)\rho^{-1}\tilde{s}_n - f_n r_n)}{|E_1^0|^2},$$
$$K_{2,n} = D_\mu \frac{g_n((-\lambda_n^{sh} + p_n)\rho^{-1}\tilde{r}_n - g_n s_n) + (\lambda_n^{sh} - p_n)((\lambda_n^{sh} - p_n)s_n + \rho g_n \tilde{s}_n)}{|E_2^0|^2},$$
$$K_{3,n} = D_\mu \frac{(-\lambda_n^{sh} + p_n)((-\lambda_n^{sh} + p_n)r_n + \rho f_n \tilde{r}_n) + f_n((-\lambda_n^{sh} + p_n)\rho^{-1}\tilde{s}_n - f_n r_n)}{|E_3^0|^2},$$
$$K_{4,n} = D_\mu \frac{g_n((\lambda_n^{sh} + p_n)\rho^{-1}\tilde{r}_n - g_n s_n) + (-\lambda_n^{sh} - p_n)((-\lambda_n^{sh} - p_n)s_n + \rho g_n \tilde{s}_n)}{|E_4^0|^2},$$
$$K_{5,n} = \frac{D_\varepsilon}{D_\mu} K_{1,n}, \quad K_{6,n} = \frac{D_\varepsilon}{D_\mu} K_{2,n}, \quad K_{7,n} = \frac{D_\varepsilon}{D_\mu} K_{3,n}, \quad K_{8,n} = \frac{D_\varepsilon}{D_\mu} K_{4,n}.$$

Now we are ready to state the result. The followings are asymptotics of eigenvalues of $W_B^{sh}(r_s)$

$$\lambda_\mu + \lambda_\varepsilon + (r_s\omega)^2 \left(\frac{T_{16,n}T_{61,n}}{\lambda_\mu - \lambda_\varepsilon} + \frac{T_{18,n}T_{81,n}}{\lambda_\mu - \lambda_\varepsilon + 2\lambda_n^{sh}} + K_{1,n} \right) + O(r_s^3),$$
$$\lambda_\mu + \lambda_\varepsilon + (r_s\omega)^2 \left(\frac{T_{16,n}T_{61,n}}{\lambda_\mu - \lambda_\varepsilon} + \frac{T_{18,n}T_{81,n}}{\lambda_\mu - \lambda_\varepsilon + 2\lambda_n^{sh}} + K_{2,n} \right) + O(r_s^3),$$
$$\lambda_\mu - \lambda_\varepsilon + (r_s\omega)^2 \left(\frac{T_{36,n}T_{63,n}}{\lambda_\mu - \lambda_\varepsilon - 2\lambda_n^{sh}} + \frac{T_{38,n}T_{83,n}}{\lambda_\mu - \lambda_\varepsilon} + K_{3,n} \right) + O(r_s^3),$$
$$\lambda_\mu - \lambda_\varepsilon + (r_s\omega)^2 \left(\frac{T_{36,n}T_{63,n}}{\lambda_\mu - \lambda_\varepsilon - 2\lambda_n^{sh}} + \frac{T_{38,n}T_{83,n}}{\lambda_\mu - \lambda_\varepsilon} + K_{4,n} \right) + O(r_s^3),$$
$$\lambda_\varepsilon + \lambda_\mu + (r_s\omega)^2 \left(\frac{T_{52,n}T_{25,n}}{\lambda_\varepsilon - \lambda_\mu} + \frac{T_{54,n}T_{45,n}}{\lambda_\varepsilon - \lambda_\mu + 2\lambda_n^{sh}} + K_{5,n} \right) + O(r_s^3),$$
$$\lambda_\varepsilon + \lambda_\mu + (r_s\omega)^2 \left(\frac{T_{52,n}T_{25,n}}{\lambda_\varepsilon - \lambda_\mu} + \frac{T_{54,n}T_{45,n}}{\lambda_\varepsilon - \lambda_\mu + 2\lambda_n^{sh}} + K_{6,n} \right) + O(r_s^3),$$
$$\lambda_\varepsilon - \lambda_\mu + (r_s\omega)^2 \left(\frac{T_{72,n}T_{27,n}}{\lambda_\varepsilon - \lambda_\mu - 2\lambda_n^{sh}} + \frac{T_{74,n}T_{47,n}}{\lambda_\varepsilon - \lambda_\mu} + K_{7,n} \right) + O(r_s^3),$$
$$\lambda_\varepsilon - \lambda_\mu + (r_s\omega)^2 \left(\frac{T_{72,n}T_{27,n}}{\lambda_\varepsilon - \lambda_\mu - 2\lambda_n^{sh}} + \frac{T_{74,n}T_{47,n}}{\lambda_\varepsilon - \lambda_\mu} + K_{8,n} \right) + O(r_s^3).$$

We also have the following asymptotic expansions of the eigenfunctions:

$$E_1^0 + r_s\omega \left(\frac{T_{16,n}}{\lambda_\mu - \lambda_\varepsilon} E_6^0 + \frac{T_{18,n}}{\lambda_\mu - \lambda_\varepsilon + 2\lambda_n^{sh}} E_8^0 \right) + O(r_s^2),$$
$$E_2^0 + r_s\omega \left(\frac{T_{25,n}}{\lambda_\mu - \lambda_\varepsilon} E_5^0 + \frac{T_{27,n}}{\lambda_\mu - \lambda_\varepsilon + 2\lambda_n^{sh}} E_7^0 \right) + O(r_s^2),$$
$$E_3^0 + r_s\omega \left(\frac{T_{36,n}}{\lambda_\mu - \lambda_\varepsilon - 2\lambda_n^{sh}} E_6^0 + \frac{T_{38,n}}{\lambda_\mu - \lambda_\varepsilon} E_8^0 \right) + O(r_s^2),$$
$$E_4^0 + r_s\omega \left(\frac{T_{45,n}}{\lambda_\mu - \lambda_\varepsilon - 2\lambda_n^{sh}} E_5^0 + \frac{T_{47,n}}{\lambda_\mu - \lambda_\varepsilon} E_7^0 \right) + O(r_s^2),$$
$$E_5^0 + r_s\omega \left(\frac{T_{52,n}}{\lambda_\mu - \lambda_\varepsilon} E_2^0 + \frac{T_{54,n}}{\lambda_\mu - \lambda_\varepsilon + 2\lambda_n^{sh}} E_4^0 \right) + O(r_s^2),$$
$$E_6^0 + r_s\omega \left(\frac{T_{61,n}}{\lambda_\mu - \lambda_\varepsilon} E_1^0 + \frac{T_{63,n}}{\lambda_\mu - \lambda_\varepsilon + 2\lambda_n^{sh}} E_3^0 \right) + O(r_s^2),$$
$$E_7^0 + r_s\omega \left(\frac{T_{72,n}}{\lambda_\mu - \lambda_\varepsilon - 2\lambda_n^{sh}} E_2^0 + \frac{T_{74,n}}{\lambda_\mu - \lambda_\varepsilon} E_4^0 \right) + O(r_s^2),$$
$$E_8^0 + r_s\omega \left(\frac{T_{81,n}}{\lambda_\mu - \lambda_\varepsilon - 2\lambda_n^{sh}} E_1^0 + \frac{T_{83,n}}{\lambda_\mu - \lambda_\varepsilon} E_3^0 \right) + O(r_s^2).$$

Interestingly, the first-order term (of order δ) is still zero in the asymptotic expansions of the eigenvalues. This is due to the fact that degenerate eigenfunctions does not interact with each other.

3.7 Concluding remarks

In this chapter, we have given the first rigorous detailed description of the scaling behavior of plasmonic resonances for the full Maxwell equations, improving our understanding of light scattering by plasmonic nanoparticles.

The particle dimension and interparticle distances are considered to be infinitely small compared with the wavelength of the interacting light.

We have also shown formulas indicating the blow up rate of the extinction cross section at the plasmonic resonance and give explicit formulas for the case of spherical and spherical shell nanoparticles.

Part II

Applications

Chapter 4

Heat Generation with Plasmonic Nanoparticles

Contents

4.1 Introduction

Our aim in this chapter is to provide a mathematical and numerical framework for analyzing photothermal effects using plasmonic nanoparticles. At or near the plasmonic resonant frequencies, strong enhancement of scattering and absorption occurs, see chapter 2, 3 and [86]. This translates into an efficient heat generation in the presence of electromagnetic radiation. Moreover, plasmonic nanoparticles biocompatibility makes them suitable for use in nanotherapy [36].

Nanotherapy relies on a simple mechanism. First nanoparticles become attached to tumor cells using selective biomolecular linkers. Then heat generated by optically-simulated plasmonic nanoparticles destroys the tumor cells [51]. In this nanomedical application, the temperature increase is the most important parameter [71, 83]. It depends in a highly nontrivial way on the shape, the number, and the arrangement of the nanoparticles. Moreover, it is challenging to measure it at the surface of the nanoparticles [51].

In this chapter, we derive an asymptotic formula for the temperature at the surface of plasmonic nanoparticles of arbitrary shapes. Our formula holds for clusters of simply connected nanoparticles. It allows to estimate the collective response of plasmonic nanoparticles. In particular, it shows that the total amount of heat generated by two interacting nanoparticles is significantly different from the heat created by two single nanoparticles. The more interacting nanoparticles, the stronger the temperature increase. Our results in this chapter formally explain the experimental observations reported in [51].

The chapter is organized as follows. In section 4.2 we describe the mathematical setting for the physical phenomena we are modeling. To this end, we use the Helmholtz equation to model the propagation of light which we couple to the heat equation. Later on, we present our main results in this chapter which consist on original asymptotic formulas for the inner field and the temperature on the boundaries of the nanoparticles. In section 4.3 we prove Theorems 4.2.1 and 4.2.2. These results clarify the strong dependency of the heat generation on the geometry of the particles as it depends on the eigenvalues of the associated Neumann-Poincaré operator. In section 4.4 we present numerical examples of the temperature at the boundary of single and multiple particles.

4.2 Setting of the problem and the main results

In this chapter, we use the Helmholtz equation for modeling the propagation of light. This can be thought of as a special case of Maxwell's equations, when the incident wave u^i is a transverse electric or transverse magnetic (TE or TM) polarized wave. This approximation, also called paraxial approximation [54], is a good model for a laser beam which are used, in particular, in full-field optical coherence tomography. We will therefore model the propagation of a laser beam in a host domain (tissue), hosting a nanoparticle.

Let the nanoparticle occupy a bounded domain $D \Subset \mathbb{R}^2$ of class $\mathcal{C}^{1,\alpha}$ for some $0 < \alpha < 1$. Furthermore, let $D = z + \delta B$, where B is centered at the origin and $|B| = O(1)$.

We denote by $\varepsilon_c(x)$ and $\mu_c(x)$, $x \in D$, the electric permittivity and magnetic permeability of the particle, respectively, both of which may depend

on the frequency ω of the incident wave. Assume that $\varepsilon_c(x) = \varepsilon_0\varepsilon_c'$, $\mu_c(x) = \mu_0\mu_c'$ and that $\Re\varepsilon_c' < 0, \Im\varepsilon_c' > 0, \Re\mu_c' < 0, \Im\mu_c' > 0$. Here and throughout, ε_0 and μ_0 are the permittivity and permeability of vacuum.

Similarly, we denote by $\varepsilon_m(x) = \varepsilon_0\varepsilon_m'$ and $\mu_m(x) = \mu_0\mu_m'$, $x \in \mathbb{R}^2\backslash\overline{D}$ the permittivity and permeability of the host medium, both of which do not depend on the frequency ω of the incident wave. Assume that ε_m and μ_m are real and strictly positive.

The index of refraction of the medium (with the nanoparticle) is given by

$$n(x) = \sqrt{\varepsilon_c'\mu_c'}\chi(D)(x) + \sqrt{\varepsilon_m'\mu_m'}\chi(\mathbb{R}^2\backslash\overline{D})(x),$$

where χ denotes the indicator function.

The scattering problem for a TE incident wave u^i is modeled as follows:

$$\begin{cases} \nabla\cdot\dfrac{c^2}{n^2}\nabla u + \omega^2 u = 0 \quad \text{in } \mathbb{R}^2\backslash\partial D, \\ u_+ - u_- = 0 \quad \text{on } \partial D, \\ \dfrac{1}{\varepsilon_m}\dfrac{\partial u}{\partial\nu}\Big|_+ - \dfrac{1}{\varepsilon_c}\dfrac{\partial u}{\partial\nu}\Big|_- = 0 \quad \text{on } \partial D, \\ u^s := u - u^i \text{ satisfies the Sommerfeld radiation condition at infinity,} \end{cases} \tag{4.1}$$

where $\frac{\partial}{\partial\nu}$ denotes the outward normal derivative and $c = \frac{1}{\sqrt{\varepsilon_0\mu_0}}$ is the speed of light in vacuum. We use the notation $\frac{\partial}{\partial\nu}\Big|_\pm$ indicating

$$\frac{\partial u}{\partial\nu}\Big|_\pm(x) = \lim_{t\to 0^+}\nabla u(x \pm t\nu(x))\cdot\nu(x),$$

with ν being the outward unit normal vector to ∂D.

The interaction of the electromagnetic waves with the medium produces a heat flow of energy which translates into a change of temperature governed by the heat equation [37]

$$\begin{cases} \rho C\dfrac{\partial\tau}{\partial t} - \nabla\cdot\gamma\nabla\tau = \dfrac{\omega}{2\pi}\Im(\varepsilon)|u|^2 \quad \text{in } (\mathbb{R}^2\backslash\partial D)\times(0,T), \\ \tau_+ - \tau_- = 0 \quad \text{on } \partial D, \\ \gamma_m\dfrac{\partial\tau}{\partial\nu}\Big|_+ - \gamma_c\dfrac{\partial\tau}{\partial\nu}\Big|_- = 0 \quad \text{on } \partial D, \\ \tau(x,0) = 0, \end{cases} \tag{4.2}$$

where $\rho = \rho_c\chi(D) + \rho_m\chi(\mathbb{R}^2\backslash\overline{D})$ is the mass density, $C = C_c\chi(D) + C_m\chi(\mathbb{R}^2\backslash\overline{D})$ is the thermal capacity, $\gamma = \gamma_c\chi(D) + \gamma_m\chi(\mathbb{R}^2\backslash\overline{D})$ is the thermal conductivity, $T \in \mathbb{R}$ is the final time of measurements and $\varepsilon = \varepsilon_c\chi(D) + \varepsilon_m\chi(\mathbb{R}^2\backslash\overline{D})$.

We further assume that $\rho_c, \rho_m, C_c, C_m, \gamma_c, \gamma_m$ are real positive constants.

Note that $\Im(\varepsilon) = 0$ in $\mathbb{R}^2\backslash\overline{D}$ and so, outside D, the heat equation is homogeneous.

The coupling of equations (4.1) and (4.2) describes the physics of our problem.

We remark that, in general, the index of refraction varies with temperature; hence, a solution to the above equations would imply a dependency on time for the electric field u, which contradicts the time-harmonic assumption leading to model (4.1). Nevertheless, the time-scale on the dynamics of the index of refraction is much larger than the time-scale on the dynamics of the interaction of the electromagnetic wave with the medium. Therefore, we will not integrate a time-varying component into the index of refraction.

Let $G(\cdot, k)$ be the Green function for the Helmholtz operator $\Delta + k^2$ satisfying the Sommerfeld radiation condition. In dimension two, G is given by

$$G(x,k) = -\frac{i}{4}H_0^{(1)}(k|x|),$$

where $H_0^{(1)}$ is the Hankel function of first kind and order 0. We denote $G(x,y,k) := G(x-y,k)$.

Recall the definition of the single-layer potential and Neumann-Poincaré integral operator for the Helmholtz equation

$$\mathcal{S}_D^k[\varphi](x) = \int_{\partial D} G(x,y,k)\varphi(y)d\sigma(y), \qquad x \in \partial D \text{ or } x \in \mathbb{R}^2,$$

and

$$(\mathcal{K}_D^k)^*[\varphi](x) = \int_{\partial D} \frac{\partial G(x,y,k)}{\partial \nu(x)}\varphi(y)d\sigma(y), \qquad x \in \partial D.$$

Our main results in this chapter are the following.

Theorem 4.2.1. *For an incident wave $u^i \in \mathcal{C}^2(\mathbb{R}^2)$, the solution u to (4.1), inside a plasmonic particle occupying a domain $D = z + \delta B$, has the following asymptotic expansion as $\delta \to 0$ in $L^2(D)$,*

$$u = u^i(z) + \left(\delta(x-z) + \mathcal{S}_D\left(\lambda_\varepsilon Id - \mathcal{K}_D^*\right)^{-1}[\nu]\right)\cdot\nabla u^i(z) + O\left(\frac{\delta^3}{\operatorname{dist}(\lambda_\varepsilon, \sigma(\mathcal{K}_D^*))}\right),$$

where ν is the outward normal to D, $\sigma(\mathcal{K}_D^*)$ denotes the spectrum of $\mathcal{K}_D^*$ in $H^{-\frac{1}{2}}(\partial D)$ and

$$\lambda_\varepsilon := \frac{\varepsilon_c + \varepsilon_m}{2(\varepsilon_c - \varepsilon_m)}.$$

Theorem 4.2.2. *Let u be the solution to (4.1). The solution τ to (4.2) on the boundary ∂D of a plasmonic particle occupying the domain $D = z + \delta B$ has the following asymptotic expansion as $\delta \to 0$, uniformly in $(x,t) \in \partial D \times (0,T)$,*

$$\tau(x,t) = F_D(x,t,b_c) - \mathcal{V}_D^{b_c}(\lambda_\gamma Id - \mathcal{K}_D^*)^{-1}\left[\frac{\partial F_D(\cdot,\cdot,b_c)}{\partial \nu}\right](x,t) + O\left(\frac{\delta^4 \log \delta}{\operatorname{dist}(\lambda_\varepsilon, \sigma(\mathcal{K}_D^*))^2}\right),$$

where ν is the outward normal to D and

$$\begin{aligned}
\lambda_\gamma &:= \frac{\gamma_c+\gamma_m}{2(\gamma_c-\gamma_m)},\\
b_c &:= \frac{\rho_c C_c}{\gamma_c},\\
F_D(x,t,b_c) &:= \frac{\omega}{2\pi\gamma_c}\Im(\varepsilon_c)\int_0^t\int_D \frac{e^{-\frac{|x-y|^2}{4b_c(t-t')}}}{4\pi b_c(t-t')}|u|^2(y)dydt',\\
\mathcal{V}_D^{b_c}[f](x,t) &:= \int_0^t\int_{\partial D}\frac{e^{-\frac{|x-y|^2}{4b_c(t-t')}}}{4\pi b_c(t-t')}f(y,t')dydt'.
\end{aligned}$$

Remark 4.2.1. *We remark that Theorem 4.2.1 and Theorem 4.2.2 are independent. A generalization of Theorem 4.2.2 to $\mathbb{R}^3$ is straightforward and the same type of small volume approximation can be found using the techniques presented in this chapter. In fact, in $\mathbb{R}^3$, the operators involved in the first term of the temperature small volume expansion are*

$$\begin{aligned}
F_D(x,t,b_c) &:= \frac{\omega}{2\pi\gamma_c}\Im(\varepsilon_c)\int_0^t\int_D \frac{e^{-\frac{|x-y|^2}{4b_c(t-t')}}}{\left(4\pi b_c(t-t')\right)^{\frac{3}{2}}}|E|^2(y)dydt',\\
\mathcal{V}_D^{b_c}[f](x,t) &:= \int_0^t\int_{\partial D}\frac{e^{-\frac{|x-y|^2}{4b_c(t-t')}}}{\left(4\pi b_c(t-t')\right)^{\frac{3}{2}}}f(y,t')dydt'.
\end{aligned}$$

Here E is the vectorial electric field as a result of Maxwell equations. A small volume expansion for E inside the nanoparticle for the plasmonic case can be found using the same techniques of chapter 3.

4.3 Heat generation

In this section we consider the coupling of equations (4.1) and (4.2), that is,

$$\begin{cases}
\nabla\cdot\dfrac{c^2}{n^2}\nabla u+\omega^2 u=0 \quad \text{in } \mathbb{R}^2\backslash\partial D,\\
u_+-u_-=0 \quad \text{on } \partial D,\\
\dfrac{1}{\varepsilon_m}\dfrac{\partial u}{\partial\nu}\Big|_+-\dfrac{1}{\varepsilon_c}\dfrac{\partial u}{\partial\nu}\Big|_-=0 \quad \text{on } \partial D,\\
u^s:=u-u^i \text{ satisfies the Sommerfeld radiation condition at infinity},\\
\dfrac{\rho_c C_c}{\gamma_c}\dfrac{\partial\tau}{\partial t}-\Delta\tau=\dfrac{\omega}{2\pi\gamma_c}\Im(\varepsilon_c)|u|^2 \quad \text{in } D\times(0,T),\\
\dfrac{\rho_m C_m}{\gamma_m}\dfrac{\partial\tau}{\partial t}-\Delta\tau=0 \quad \text{in } (\mathbb{R}^2\backslash\overline{D})\times(0,T),\\
\tau_+-\tau_-=0 \quad \text{on } \partial D,\\
\gamma_m\dfrac{\partial\tau}{\partial\nu}\Big|_+-\gamma_c\dfrac{\partial\tau}{\partial\nu}\Big|_-=0 \quad \text{on } \partial D,\\
\tau(x,0)=0.
\end{cases} \tag{4.3}$$

Under the assumption that the index of refraction n does not depend on the temperature, we can solve equation (4.1) separately from equation (4.2).

Our goal is to establish a small volume expansion for the resulting temperature at the surface of the nanoparticle as a function of time. To do so, we first need to compute the electric field inside the nanoparticle as a result of a plasmonic resonance. The results of the following sections rely heavily on the use of layer potentials for the Helmholtz equation. We refer to chapter 2 for a summary.

4.3.1 Small volume expansion of the inner field

We proceed in this section to prove Theorem 4.2.1.

Rescaling

Since we are working with nanoparticles, we want to rescale equation (2.2) to study the solution for a small volume approximation by using representation (2.1).

Recall that $D = z + \delta B$. For any $x \in \partial D$, $\widetilde{x} := \frac{x-z}{\delta} \in \partial B$ and for each function f defined on ∂D, we introduce a corresponding function defined on ∂B as follows

$$\eta(f)(\widetilde{x}) = f(z + \delta\widetilde{x}). \tag{4.4}$$

It follows that

$$\begin{aligned} \mathcal{S}_D^k[\varphi](x) &= \delta\mathcal{S}_B^{\delta k}[\eta(\varphi)](\widetilde{x}), \\ (\mathcal{K}_D^k)^*[\varphi](x) &= (\mathcal{K}_B^{\delta k})^*[\eta(\varphi)](\widetilde{x}), \end{aligned} \tag{4.5}$$

so system (2.2) becomes

$$\begin{cases} \mathcal{S}_B^{\delta k_m}[\eta(\psi)] - \mathcal{S}_B^{\delta k_c}[\eta(\phi)] &= -\dfrac{\eta(u^i)}{\delta}, \\ \frac{1}{\varepsilon_m}\big(\frac{1}{2}Id + (\mathcal{K}_B^{\delta k_m})^*\big)[\eta(\psi)] + \frac{1}{\varepsilon_c}\big(\frac{1}{2}Id - (\mathcal{K}_B^{\delta k_c})^*\big)[\eta(\phi)] &= -\dfrac{1}{\varepsilon_m}\eta(\dfrac{\partial u^i}{\partial \nu}). \end{cases} \tag{4.6}$$

Note that the system is defined on ∂B.

For δ small enough $\mathcal{S}_B^{\delta k_m}$ is invertible (see Appendix B.3). Therefore,

$$\eta(\psi) = (\mathcal{S}_B^{\delta k_m})^{-1}\mathcal{S}_B^{\delta k_c}[\eta(\phi)] - (\mathcal{S}_B^{\delta k_m})^{-1}[\frac{\eta(u^i)}{\delta}].$$

Hence, we have the following equation for $\eta(\phi)$:

$$\mathcal{A}_B^I(\delta)[\eta(\phi)] = f^I,$$

where

$$\begin{aligned} \mathcal{A}_B^I(\delta) &= \tfrac{1}{\varepsilon_m}\big(\tfrac{1}{2}I + (\mathcal{K}_B^{\delta k_m})^*\big)(\mathcal{S}_B^{\delta k_m})^{-1}\mathcal{S}_B^{\delta k_c} + \tfrac{1}{\varepsilon_c}\big(\tfrac{1}{2}Id - (\mathcal{K}_B^{\delta k_c})^*\big), \\ f^I &= -\frac{1}{\varepsilon_m}\eta(\frac{\partial u^i}{\partial \nu}) + \tfrac{1}{\varepsilon_m}\big(\tfrac{1}{2}Id + (\mathcal{K}_B^{\delta k_m})^*\big)(\mathcal{S}_B^{\delta k_m})^{-1}[\frac{\eta(u^i)}{\delta}]. \end{aligned} \tag{4.7}$$

Proof of Theorem 4.2.1

To express the solution to (4.1) in D, asymptotically on the size of the nanoparticle δ, we make use of the representation (2.1). We derive an asymptotic expansion for $\eta(\phi)$ on δ to later compute $\delta\mathcal{S}_B^{\delta k_c}[\eta(\phi)]$ and scale back to D. We divide the proof into three steps.

Step 1. We first compute an asymptotic for $\mathcal{A}_B^I(\delta)$ and f^I.

Let $\mathcal{H}^*(\partial B)$ be defined by (A.3) with D replaced by B. In $\mathcal{L}(\mathcal{H}^*(\partial B))$, we have the following asymptotic expansion as $\delta \to 0$ (see Appendix B.3)

$$\begin{aligned}(\mathcal{S}_B^{\delta k_m})^{-1}\mathcal{S}_B^{\delta k_c} &= \mathcal{P}_{\mathcal{H}_0^*} + \mathcal{U}_{\delta k_m}(\widetilde{\mathcal{S}}_B + \Upsilon_{\delta k_c}) + O(\delta^2 \log \delta),\\ \frac{1}{2}Id \pm (\mathcal{K}_B^{\delta k})^* &= (\frac{1}{2}Id \pm \mathcal{K}_B^*) + O(\delta^2 \log \delta).\end{aligned}$$

Let φ_0 be an eigenfunction of $\mathcal{K}_B^*$ associated to the eigenvalue $1/2$ (see Appendix A) and let $\mathcal{U}_{\delta k_m}$ be defined by (B.12) with k replaced with δk_m. Then it follows that

$$(\frac{1}{2}Id + \mathcal{K}_B^*)\mathcal{U}_{\delta k_m} = \mathcal{U}_{\delta k_m}.$$

Therefore, in $\mathcal{L}(\mathcal{H}^*(\partial B))$,

$$\mathcal{A}_B^I(\delta) = \left((\frac{1}{2\varepsilon_m} + \frac{1}{2\varepsilon_c})Id + (\frac{1}{\varepsilon_m} - \frac{1}{\varepsilon_c})\mathcal{K}_B^*\right)\mathcal{P}_{\mathcal{H}_0^*} + \frac{1}{\varepsilon_m}\mathcal{U}_{\delta k_m}(\widetilde{\mathcal{S}}_B + \Upsilon_{\delta k_c}) + O(\delta^2 \log \delta),$$

and from the definition of $\mathcal{U}_{\delta k_m}$ we get

$$\mathcal{A}_B^I(\delta) = \left((\frac{1}{2\varepsilon_m} + \frac{1}{2\varepsilon_c})Id + (\frac{1}{\varepsilon_m} - \frac{1}{\varepsilon_c})\mathcal{K}_B^*\right)\mathcal{P}_{\mathcal{H}_0^*} + \frac{1}{\varepsilon_m}\frac{\mathcal{S}_B[\varphi_0] + \tau_{\delta k_c}}{\mathcal{S}_B[\varphi_0] + \tau_{\delta k_m}}(\cdot, \varphi_0)_{\mathcal{H}^*}\varphi_0 + O(\delta^2 \log \delta). \tag{4.8}$$

In the same manner, in the space $\mathcal{H}^*(\partial B)$,

$$f^I = \frac{1}{\varepsilon_m}\left(-\eta(\frac{\partial u^i}{\partial \nu}) + (\frac{1}{2}Id + \mathcal{K}_B^*)\mathcal{P}_{\mathcal{H}_0^*}\widetilde{\mathcal{S}}_B^{-1}[\frac{\eta(u^i)}{\delta}] + \mathcal{U}_{\delta k_m}[\frac{\eta(u^i)}{\delta}] + O(\delta^2 \log \delta)\right).$$

We can further develop f^I. Indeed, for every $\tilde{x} \in \partial B$, a Taylor expansion yields

$$\begin{aligned}\eta(\frac{\partial u^i}{\partial \nu})(\tilde{x}) &= \nu(\tilde{x}) \cdot \nabla u^i(\delta\tilde{x} + z) = \nu(\tilde{x}) \cdot \nabla u^i(z) + O(\delta),\\ \frac{\eta(u^i)}{\delta}(\tilde{x}) &= \frac{u^i(\delta\tilde{x} + z)}{\delta} = \frac{u^i(z)}{\delta} + \tilde{x} \cdot \nabla u^i(z) + O(\delta).\end{aligned}$$

The regularity of u^i ensures that the previous formulas hold in $\mathcal{H}^*(\partial B)$.

The fact that $\tilde{x} \cdot \nabla u^i(z)$ is harmonic in B and Lemma A.0.4 imply that

$$-\nu \cdot \nabla u^i(z) = (\frac{1}{2}Id - \mathcal{K}_B^*)\mathcal{P}_{\mathcal{H}_0^*}\widetilde{\mathcal{S}}_B^{-1}[\tilde{x} \cdot \nabla u^i(z)]$$

in $\mathcal{H}^*(\partial B)$.

Thus, in $\mathcal{H}^*(\partial B)$,

$$f^I = \frac{1}{\varepsilon_m}\left(\mathcal{P}_{\mathcal{H}_0^*}\widetilde{\mathcal{S}}_B^{-1}[\tilde{x} \cdot \nabla u^i(z)] + \mathcal{U}_{\delta k_m}[\frac{u^i(z)}{\delta} + \tilde{x}\nabla u^i(z)] + O(\delta)\right).$$

From the definition of $\mathcal{U}_{\delta k_m}$ we get

$$f^I = \frac{1}{\varepsilon_m}\left(\mathcal{P}_{\mathcal{H}_0^*}\widetilde{\mathcal{S}}_B^{-1}[\tilde{x}\cdot\nabla u^i(z)] + \frac{u^i(z)\varphi_0}{\delta(\mathcal{S}_B[\varphi_0]+\tau_{\delta k_m})} - \frac{(\widetilde{\mathcal{S}}_B^{-1}[\tilde{x}\cdot\nabla u^i(z)],\varphi_0)_{\mathcal{H}^*}\varphi_0}{\mathcal{S}_B[\varphi_0]+\tau_{\delta k_m}} + O(\delta)\right). \tag{4.9}$$

Step 2. We compute $(\mathcal{A}_B^I(\delta))^{-1}f^I$.

We begin by computing an asymptotic expansion of $(\mathcal{A}_B^I(\delta))^{-1}$.

The operator $\mathcal{A}_0^I := \left(\left(\frac{1}{2\varepsilon_m}+\frac{1}{2\varepsilon_c}\right)Id + \left(\frac{1}{\varepsilon_m}-\frac{1}{\varepsilon_c}\right)\mathcal{K}_B^*\right)$ maps $\mathcal{H}_0^*$ into $\mathcal{H}_0^*$. Hence, the operator defined by (which appears in the expansion of $\mathcal{A}_B^I(\delta)$)

$$\mathcal{A}_{B,0}^I := \mathcal{A}_0^I\mathcal{P}_{\mathcal{H}_0^*} + \frac{1}{\varepsilon_m}\frac{\mathcal{S}_B[\varphi_0]+\tau_{\delta k_c}}{\mathcal{S}_B[\varphi_0]+\tau_{\delta k_m}}(\cdot,\varphi_0)_{\mathcal{H}^*}\varphi_0,$$

is invertible of inverse

$$(\mathcal{A}_{B,0}^I)^{-1} = (\mathcal{A}_0^I)^{-1}\mathcal{P}_{\mathcal{H}_0^*} + \varepsilon_m\frac{\mathcal{S}_B[\varphi_0]+\tau_{\delta k_m}}{\mathcal{S}_B[\varphi_0]+\tau_{\delta k_c}}(\cdot,\varphi_0)_{\mathcal{H}^*}\varphi_0.$$

Therefore, we can write

$$(\mathcal{A}_B^I)^{-1}(\delta) = \left(Id + (\mathcal{A}_{B,0}^I)^{-1}O(\delta^2\log\delta)\right)^{-1}(\mathcal{A}_{B,0}^I)^{-1}.$$

Since $\mathcal{K}_B^*$ is a compact self-adjoint operator in $\mathcal{H}^*(\partial B)$ it follows that

$$\|(\mathcal{A}_0^I)^{-1}\|_{\mathcal{L}(\mathcal{H}^*(\partial B))} \leq \frac{c}{\operatorname{dist}(0,\sigma(\mathcal{A}_0^I))}, \tag{4.10}$$

for a constant c. Therefore, for δ small enough, we obtain

$$\begin{aligned}
&(\mathcal{A}_B^I(\delta))^{-1}f^I = \left(Id + (\mathcal{A}_{B,0}^I)^{-1}O(\delta^2\log\delta)\right)^{-1}(\mathcal{A}_{B,0}^I)^{-1}f^I\\
=\ &\left(Id + (\mathcal{A}_{B,0}^I)^{-1}O(\delta^2\log\delta)\right)^{-1}\left(\frac{u^i(z)\varphi_0}{\delta(\mathcal{S}_B[\varphi_0]+\tau_{\delta k_c})} - \frac{(\widetilde{\mathcal{S}}_B^{-1}[\tilde{x}\cdot\nabla u^i(z)],\varphi_0)_{\mathcal{H}^*}\varphi_0}{\mathcal{S}_B[\varphi_0]+\tau_{\delta k_c}} + \right.\\
&\left.(\mathcal{A}_0^I)^{-1}\frac{1}{\varepsilon_m}\mathcal{P}_{\mathcal{H}_0^*}\widetilde{\mathcal{S}}_B^{-1}[\tilde{x}\cdot\nabla u^i(z)] + O\left(\frac{\delta}{\operatorname{dist}(0,\sigma(\mathcal{A}^I{}_0))}\right)\right)\\
=\ &\frac{u^i(z)\varphi_0}{\delta(\mathcal{S}_B[\varphi_0]+\tau_{\delta k_c})} - \frac{(\widetilde{\mathcal{S}}_B^{-1}[\tilde{x}\cdot\nabla u^i(z)],\varphi_0)_{\mathcal{H}^*}\varphi_0}{\mathcal{S}_B[\varphi_0]+\tau_{\delta k_c}} + (\mathcal{A}_0^I)^{-1}\frac{1}{\varepsilon_m}\mathcal{P}_{\mathcal{H}_0^*}\widetilde{\mathcal{S}}_B^{-1}[\tilde{x}\cdot\nabla u^i(z)] +\\
&O\left(\frac{\delta}{\operatorname{dist}(0,\sigma(\mathcal{A}^I{}_0))}\right).
\end{aligned}$$

Using the representation formula of $\mathcal{K}_B^*$ described in Lemma A.0.2 we can further develop the third term in the above expression to obtain

$$\begin{aligned}(\mathcal{A}_0^I)^{-1}\mathcal{P}_{\mathcal{H}_0^*}\widetilde{\mathcal{S}}_B^{-1}[\tilde{x}\cdot\nabla u^i(z)] &= \sum_{j=1}^{\infty}\frac{(\widetilde{\mathcal{S}}_B^{-1}[\tilde{x}\cdot\nabla u^i(z)],\varphi_j)_{\mathcal{H}^*}\varphi_j}{\left(\frac{1}{2}+\frac{\varepsilon_m}{2\varepsilon_c}\right)-\left(\frac{\varepsilon_m}{\varepsilon_c}-1\right)\lambda_j}\\ &= \sum_{j=1}^{\infty}\left(\frac{(\widetilde{\mathcal{S}}_B^{-1}[\tilde{x}\cdot\nabla u^i(z)],\varphi_j)_{\mathcal{H}^*}\varphi_j}{\left(\frac{1}{2}+\frac{\varepsilon_m}{2\varepsilon_c}\right)-\left(\frac{\varepsilon_m}{\varepsilon_c}-1\right)\lambda_j}-(\widetilde{\mathcal{S}}_B^{-1}[\tilde{x}\cdot\nabla u^i(z)],\varphi_j)_{\mathcal{H}^*}\varphi_j\right)\\ &\quad+\mathcal{P}_{\mathcal{H}_0^*}\widetilde{\mathcal{S}}_B^{-1}[\tilde{x}\cdot\nabla u^i(z)]\\ &= \mathcal{P}_{\mathcal{H}_0^*}\widetilde{\mathcal{S}}_B^{-1}[\tilde{x}\cdot\nabla u^i(z)]+\sum_{j=1}^{\infty}(\lambda_j-\frac{1}{2})\frac{(\widetilde{\mathcal{S}}_B^{-1}[\tilde{x}\cdot\nabla u^i(z)],\varphi_j)_{\mathcal{H}^*}\varphi_j}{\lambda-\lambda_j}.\end{aligned}$$

Using the same arguments as those in the proof of Lemma A.0.4, we have

$$(\lambda_j-\frac{1}{2})(\widetilde{\mathcal{S}}_B^{-1}[\tilde{x}\cdot\nabla u^i(z)],\varphi_j)_{\mathcal{H}^*}=\frac{(\nu\cdot\nabla u^i(z),\varphi_j)_{\mathcal{H}^*}}{\lambda_j-\frac{1}{2}},$$

and consequently,

$$(\mathcal{A}_0^I)^{-1}\frac{1}{\varepsilon_m}\mathcal{P}_{\mathcal{H}_0^*}\widetilde{\mathcal{S}}_B^{-1}[\tilde{x}\cdot\nabla u^i(z)]=\mathcal{P}_{\mathcal{H}_0^*}\widetilde{\mathcal{S}}_B^{-1}[\tilde{x}\cdot\nabla u^i(z)]+(\lambda_\varepsilon Id-\mathcal{K}_B^*)^{-1}[\nu]\cdot\nabla u^i(z).$$

Therefore,

$$\begin{aligned}(\mathcal{A}_B^I(\delta))^{-1}f^I = \ &\frac{u^i(z)\varphi_0}{\delta(\mathcal{S}_B[\varphi_0]+\tau_{\delta k_c})}-\frac{(\widetilde{\mathcal{S}}_B^{-1}[\tilde{x}\cdot\nabla u^i(z)],\varphi_0)_{\mathcal{H}^*}\varphi_0}{\mathcal{S}_B[\varphi_0]+\tau_{\delta k_c}}+\mathcal{P}_{\mathcal{H}_0^*}\widetilde{\mathcal{S}}_B^{-1}[\tilde{x}\cdot\nabla u^i(z)]+\\ &(\lambda_\varepsilon Id-\mathcal{K}_B^*)^{-1}[\nu]\cdot\nabla u^i(z)+O\left(\frac{\delta}{\operatorname{dist}(0,\sigma(\mathcal{A}^I{}_0))}\right).\end{aligned}$$

Step 3. Finally, we compute $\eta(u)=\delta\mathcal{S}_B^{\delta k_c}(\mathcal{A}_B^I(\delta))^{-1}f^I$.

From Appendix B.3, the following holds when $\mathcal{S}_B^{\delta k_c}$ is viewed as an operator from the space $\mathcal{H}^*(\partial B)$ to $\mathcal{H}(\partial B)$:

$$\mathcal{S}_B^{\delta k_c}=\widetilde{\mathcal{S}}_B+\Upsilon_{\delta k_c}+O(\delta^2\log\delta).$$

In particular, we have

$$\mathcal{S}_B^{\delta k_c}[\varphi_0]=\mathcal{S}_B[\varphi_0]+\tau_{\delta k_c}+O(\delta^2\log\delta).$$

It can be verified that the same expansion holds when viewed as an operator from $\mathcal{H}^*(\partial B)$ into $L^2(B)$.

Note that the following identity holds

$$\begin{aligned}&-\frac{(\widetilde{\mathcal{S}}_B^{-1}[\tilde{x}\cdot\nabla u^i(z)],\varphi_0)_{\mathcal{H}^*}\varphi_0}{\mathcal{S}_B[\varphi_0]+\tau_{\delta k_c}}+\mathcal{P}_{\mathcal{H}_0^*}\widetilde{\mathcal{S}}_B^{-1}[\tilde{x}\cdot\nabla u^i(z)]=\\ &-\frac{\Upsilon_{\delta k_c}[\widetilde{\mathcal{S}}_B^{-1}[\tilde{x}\cdot\nabla u^i(z)]]\varphi_0}{\mathcal{S}_B[\varphi_0]+\tau_{\delta k_c}}+\widetilde{\mathcal{S}}_B^{-1}[\tilde{x}\cdot\nabla u^i(z)].\end{aligned}$$

Straightforward calculations and the fact that $\mathcal{S}_B$ is harmonic in B yields

$$\delta\mathcal{S}_B^{\delta k_c}(\mathcal{A}_B^I(\delta))^{-1}f^I = u^i(z) + \delta\big(\tilde{x} + \mathcal{S}_B\big(\lambda_\varepsilon Id - \mathcal{K}_B^*\big)^{-1}[\nu]\big)\cdot\nabla u^i(z) + O\left(\frac{\delta^2}{\operatorname{dist}(\lambda_\varepsilon, \sigma(\mathcal{K}_B^*))}\right)$$

in $L^2(B)$. Using Lemma A.0.3 to scale back the estimate to D leads to the desired result.

4.3.2 Small volume expansion of the temperature

We proceed in this section to prove Theorem 4.2.2. To do so, we make use of the Laplace transform method [46, 58, 69].

Consider equation (4.3) and define the Laplace transform of a function $g(t)$ by

$$L(g)(s) = \int_0^\infty e^{-st}g(t)dt.$$

Taking the Laplace transform of the equations on τ in (4.3) we formally obtain the following system:

$$\begin{cases} s\dfrac{\rho_c C_c}{\gamma_c}\hat{\tau}(\cdot,s) - \Delta\hat{\tau}(\cdot,s) = L(g_u)(\cdot,s) \quad \text{in } D, \\ s\dfrac{\rho_m C_m}{\gamma_m}\hat{\tau}(\cdot,s) - \Delta\hat{\tau}(\cdot,s) = 0 \quad \text{in } \mathbb{R}^2\backslash\overline{D}, \\ \hat{\tau}_+(\cdot,s) - \hat{\tau}_-(\cdot,s) = 0 \quad \text{on } \partial D, \\ \gamma_m\dfrac{\partial\hat{\tau}}{\partial\nu}\Big|_+ - \gamma_c\dfrac{\partial\hat{\tau}}{\partial\nu}\Big|_- = 0 \quad \text{on } \partial D, \\ \hat{\tau}(\cdot,s) \text{ satisfies the Sommerfeld radiation condition at infinity,} \end{cases} \tag{4.11}$$

where $\hat{\tau}(\cdot,s)$ and $L(g_u)(\cdot,s)$ are the Laplace transforms of τ and $g_u := \frac{\omega}{2\pi\gamma_c}\Im(\varepsilon_c)|u|^2$, respectively, and $s \in \mathbb{C}\backslash(-\infty, 0]$.

A rigorous justification for the derivation of system (4.11) and the validity of the inverse transform of the solution can be found in [58].

Using layer potential techniques we have that, for any $\hat{p}, \hat{q} \in H^{-\frac{1}{2}}(\partial D)$, $\hat{\tau}$ defined by

$$\hat{\tau} := \begin{cases} -\mathcal{S}_D^{\beta_{\gamma_m}}[\hat{p}], & x \in \mathbb{R}^2\backslash\overline{D}, \\ -\hat{F}_D(\cdot, y, \beta_{\gamma_c}) - \mathcal{S}_D^{\beta_{\gamma_c}}[\hat{q}], & x \in D, \end{cases} \tag{4.12}$$

satisfies the differential equations in (4.11) together with the Sommerfeld radiation condition. Here $\beta_{\gamma_m} := i\sqrt{s\frac{\rho_m C_m}{\gamma_m}}$, $\beta_{\gamma_c} := i\sqrt{s\frac{\rho_c C_c}{\gamma_c}}$ and

$$\hat{F}_D(\cdot, \beta_{\gamma_c}) := \int_D G(\cdot, y, \beta_{\gamma_c})L(g_u)(y)dy.$$

To satisfy the boundary transmission conditions, $\hat{p}$ and $\hat{q} \in H^{-\frac{1}{2}}(\partial D)$ should satisfy the following system of integral equations on ∂D:

$$\begin{cases} -\mathcal{S}_D^{\beta_{\gamma_m}}[\hat{p}] + \mathcal{S}_D^{\beta_{\gamma_c}}[\hat{q}] &= -\hat{F}_D(\cdot, \beta_{\gamma_c}), \\ -\gamma_m\big(\frac{1}{2}Id + (\mathcal{K}_D^{\beta_{\gamma_m}})^*\big)[\hat{p}] + \gamma_c\big(-\frac{1}{2}Id + (\mathcal{K}_D^{\beta_{\gamma_c}})^*\big)[\hat{q}] &= -\gamma_c \dfrac{\partial \hat{F}_D(\cdot, \beta_{\gamma_c})}{\partial \nu}. \end{cases} \tag{4.13}$$

Re-scaling of the equations

Recall that $D = z + \delta B$, for any $x \in \partial D$, $\widetilde{x} := \frac{x-z}{\delta} \in \partial B$, for each function f defined on ∂D, η is such that $\eta(f)(\widetilde{x}) = f(z + \delta\widetilde{x})$ and

$$\begin{aligned} \mathcal{S}_D^k[\varphi](x) &= \delta \mathcal{S}_B^{\delta k}[\eta(\varphi)](\widetilde{x}), \\ (\mathcal{K}_D^k)^*[\varphi](x) &= (\mathcal{K}_B^{\delta k})^*[\eta(\varphi)](\widetilde{x}). \end{aligned}$$

We can also verify that

$$\begin{aligned} \hat{F}_D(x, \beta_{\gamma_c}) &= \delta^2 \hat{F}_B(\hat{x}, \delta\beta_{\gamma_c}), \\ \frac{\partial \hat{F}_D}{\partial \nu}(x, \beta_{\gamma_c}) &= \delta \frac{\partial \hat{F}_B}{\partial \nu}(\hat{x}.\delta\beta_{\gamma_c}). \end{aligned}$$

Note that in the above identity, in the left-hand side we differentiate with respect to x while in the right-hand side we differentiate with respect to $\tilde{x}$. To simplify the notation, we will use $\hat{F}_B$ to refer to $\hat{F}_B(\cdot, \delta\beta_{\gamma_c})$.

We rescale system (4.13) to arrive at

$$\begin{cases} -\mathcal{S}_B^{\delta\beta_{\gamma_m}}[\eta(\hat{p})] + \mathcal{S}_B^{\delta\beta_{\gamma_c}}[\eta(\hat{q})] &= -\delta\hat{F}_B, \\ -\gamma_m\big(\frac{1}{2}Id + (\mathcal{K}_B^{\delta\beta_{\gamma_m}})^*\big)[\eta(\hat{p})] + \gamma_c\big(-\frac{1}{2}Id + (\mathcal{K}_B^{\delta\beta_{\gamma_c}})^*\big)[\eta(\hat{q})] &= -\gamma_c\delta \dfrac{\partial \hat{F}_B}{\partial \nu}. \end{cases}$$

For δ small enough, $\mathcal{S}_B^{\delta\beta_{\gamma_c}}$ is invertible (see Appendix B.3). Therefore, it follows that

$$\eta(\hat{p}) = (\mathcal{S}_B^{\delta\beta_{\gamma_m}})^{-1}\mathcal{S}_B^{\delta\beta_{\gamma_c}}[\eta(\hat{q})] + (\mathcal{S}_B^{\delta\beta_{\gamma_m}})^{-1}\left[\delta\hat{F}_B\right].$$

Hence, we have the following equation for $\eta(\hat{q})$:

$$\mathcal{A}_B^h(\delta)[\eta(\hat{q})] = f^h,$$

where

$$\begin{aligned} \mathcal{A}_B^h(\delta) &= -\gamma_m\big(\tfrac{1}{2}Id + (\mathcal{K}_B^{\delta\beta_{\gamma_m}})^*\big)(\mathcal{S}_B^{\delta\beta_{\gamma_m}})^{-1}\mathcal{S}_B^{\delta\beta_{\gamma_c}} + \gamma_c\big(-\tfrac{1}{2}Id + (\mathcal{K}_B^{\delta\beta_{\gamma_c}})^*\big), \\ f^h &= -\gamma_c\delta\frac{\partial \hat{F}_B}{\partial \nu} + \gamma_m\big(\tfrac{1}{2}Id + (\mathcal{K}_B^{\delta\beta_{\gamma_m}})^*\big)(\mathcal{S}_B^{\delta\beta_{\gamma_m}})^{-1}\left[\delta\hat{F}_B\right]. \end{aligned} \tag{4.14}$$

Proof of Theorem 4.2.2

To express the solution of (4.2) on $\partial D \times (0, T)$, asymptotically on the size of the nanoparticle δ, we make use of the representation (4.12). We will compute

an asymptotic expansion for $\eta(\hat{q})$ on δ to later compute $\delta\mathcal{S}_B^{\delta\beta_{\gamma_c}}[\eta(\hat{q})]$ on ∂B, scale back to D and take Laplace inverse.

Using the asymptotic expansions of Appendix B.3 the following asymptotic for $\mathcal{A}_B^h(\delta)$ holds in $\mathcal{L}(\mathcal{H}^*(\partial B))$

$$\mathcal{A}_B^h(\delta) = \mathcal{A}_0^h + O(\delta^2 \log \delta),$$

where

$$\mathcal{A}_0^h = -\left(\frac{1}{2}(\gamma_c+\gamma_m)Id - (\gamma_c-\gamma_m)\mathcal{K}_B^*\right).$$

In the same manner, in $\mathcal{H}^*(\partial B)$,

$$\begin{aligned} f^h &= -\gamma_c\delta\frac{\partial \hat{F}_B}{\partial\nu} + \gamma_m\left(\frac{1}{2}Id + \mathcal{K}_B^*\right)\widetilde{\mathcal{S}}_B^{-1}[\delta\hat{F}_B] + O\left(\frac{\delta^5\log\delta}{\mathrm{dist}(\lambda_\varepsilon,\sigma(\mathcal{K}_D^*))^2}\right) \\ &= -\gamma_c\delta\frac{\partial \hat{F}_B}{\partial\nu} - \gamma_m\left(\frac{1}{2}Id - \mathcal{K}_B^*\right)\widetilde{\mathcal{S}}_B^{-1}[\delta\hat{F}_B] + \gamma_m\widetilde{\mathcal{S}}_B^{-1}[\delta\hat{F}_B] + O\left(\frac{\delta^5\log\delta}{\mathrm{dist}(\lambda_\varepsilon,\sigma(\mathcal{K}_D^*))^2}\right). \end{aligned}$$

Here the remainder comes from the fact that $\hat{F}_B = O\left(\frac{\delta^2}{\mathrm{dist}(\lambda_\varepsilon,\sigma(\mathcal{K}_D^*))^2}\right)$.

Note that $\Delta\hat{F}_B = \eta(L(g_u)) - \delta^2\beta_{\gamma_c}^2\hat{F}_B$ in B and $\Delta\hat{F}_B = 0$ in $\mathbb{R}^2\backslash\bar{D}$. We can further verify that $\hat{F}_B$ satisfies the assumption required in Lemma A.0.4. Thus we have

$$\left(\frac{1}{2}Id - \mathcal{K}_B^*\right)\widetilde{\mathcal{S}}_B^{-1}[\delta\hat{F}_B] = -\delta\frac{\partial\hat{F}_B}{\partial\nu} + C_u\varphi_0 + \gamma_m\widetilde{\mathcal{S}}_B^{-1}[\delta\hat{F}_B] + O\left(\frac{\delta^5}{\mathrm{dist}(\lambda_\varepsilon,\sigma(\mathcal{K}_D^*))^2}\right),$$

where C_u is a constant such that $C_u = O\left(\frac{\delta^3}{\mathrm{dist}(\lambda_\varepsilon,\sigma(\mathcal{K}_D^*))^2}\right)$.

After replacing the above in the expression of f^h we find that

$$\begin{aligned} \eta(\hat{q}) &= (\mathcal{A}_B^h(\delta))^{-1}f^h \\ &= (\lambda_\gamma Id - \mathcal{K}_B^*)^{-1}[\delta\frac{\partial\hat{F}_B}{\partial\nu}] + \frac{C_u\gamma_m}{(\gamma_c-\gamma_m)(\lambda_\gamma-\frac{1}{2})}\varphi_0 + O\left(\frac{\delta^5\log\delta}{\mathrm{dist}(\lambda_\varepsilon,\sigma(\mathcal{K}_D^*))^2}\right), \end{aligned} \tag{4.15}$$

where

$$\lambda_\gamma = \frac{\gamma_c+\gamma_m}{2(\gamma_c-\gamma_m)}.$$

Finally, in $\mathcal{H}^*(\partial B)$,

$$\eta(\hat{\tau}) = -\delta^2\hat{F}_B - \delta\mathcal{S}_B^{\delta\beta_{\gamma_c}}(\lambda_\gamma Id - \mathcal{K}_B^*)^{-1}[\frac{\partial\delta\hat{F}_B}{\partial\nu}] - \frac{C_u\gamma_m}{(\gamma_c-\gamma_m)(\lambda_\gamma-\frac{1}{2})}\delta\mathcal{S}_B^{\delta\beta_{\gamma_c}}[\varphi_0] + O\left(\frac{\delta^6\log\delta}{\mathrm{dist}(\lambda_\varepsilon,\sigma(\mathcal{K}_D^*))^2}\right). \tag{4.16}$$

It can be shown, from the regularity of the remainders, that the previous identity also holds in $L^2(\partial B)$.

Using Holder's inequality we can prove that

$$\|\mathcal{S}_B^{\delta\beta_{\gamma_c}}[\varphi]\|_{L^\infty(\partial B)} \leq C\|\varphi\|_{L^2(\partial B)},$$

for some constant C. Hence, we find that identity (4.16) also holds true uniformly on ∂B and $C_u\delta\mathcal{S}_B^{\delta\beta_{\gamma_c}f}[\varphi_0](\tilde{x}) = O\left(\frac{\delta^4\log\delta}{\mathrm{dist}(\lambda_\varepsilon,\sigma(\mathcal{K}_D^*))^2}\right)$, uniformly in

∂B. Scaling back to D gives

$$\hat{\tau}(x,s) = -\hat{F}_D(x,\beta_{\gamma_c}) - \mathcal{S}_D^{\beta_{\gamma_c}}(\lambda_\gamma Id - \mathcal{K}_D^*)^{-1}[\frac{\partial \hat{F}_D(\cdot,\beta_{\gamma_c})}{\partial \nu}] + O\left(\frac{\delta^4 \log \delta}{\text{dist}(\lambda_\varepsilon, \sigma(\mathcal{K}_D^*))^2}\right) \quad (4.17)$$

Before we take the inverse Laplace transform to (4.17) we note that (see [69])

$$L\big(K(x,\cdot,b_c)\big) = -G(x,\beta_{\gamma_c}),$$

where $b_c := \frac{\rho_c C_c}{\gamma_c}$ and $K(x,\cdot,b_c)$ is the fundamental solution of the heat equation. In dimension two, K is given by

$$K(x,t,\gamma) = \frac{e^{-\frac{|x|^2}{4b_c t}}}{4\pi b_c t}.$$

We denote $K(x,y,t,t',b_c) := K(x-y,t-t',b_c)$. By the properties of the Laplace transform, we have

$$-\hat{F}_D(x,\beta_{\gamma_c}) = -\int_D G(x,y,\beta_{\gamma_c}) L(g_u)(y)dy = L\left(\int_0^\cdot \int_D K(x,y,\cdot,t',b_c) g_u(y) dy dt'\right).$$

We define F_D as follows

$$F_D(x,t,b_c) := \int_0^t \int_D K(x,y,t,t',b_c) g_u(y) dy dt'. \quad (4.18)$$

Similarly, we have that for a function f

$$-\int_{\partial D} G(x,y,\beta_{\gamma_c}) L(f)(y)dy = L\left(\int_0^\cdot \int_{\partial D} K(x,y,\cdot,t',b_c) f(y,t') dy dt'\right).$$

We define $\mathcal{V}_D^{b_c}$ as follows

$$\mathcal{V}_D^{b_c}[f](x,t) := \int_0^t \int_{\partial D} K(x,y,t,t',b_c) f(y,t') dy dt'. \quad (4.19)$$

Finally, using Fubini's theorem and taking Laplace inverse we find that

$$\tau(x,t) = F_D(x,t,b_c) - \mathcal{V}_D^{b_c}(\lambda_\gamma Id - \mathcal{K}_D^*)^{-1}[\frac{\partial F_D(\cdot,\cdot,b_c)}{\partial \nu}](x,t) + O\left(\frac{\delta^4 \log \delta}{\text{dist}(\lambda_\varepsilon, \sigma(\mathcal{K}_D^*))^2}\right),$$

uniformly in $(x,t) \in \partial D \times (0,T)$.

4.3.3 Temperature elevation at the plasmonic resonance

Suppose that the incident wave is $u^i(x) = e^{ik_m d\cdot x}$, where d is a unit vector. For a nanoparticle occupying a domain $D = z + \delta B$, the inner field u solution to (4.1) is given by Theorem 4.2.1, which states that, in $L^2(D)$,

$$u \approx e^{ik_m d\cdot z}\big(1 + ik_m \mathcal{S}_D\big(\lambda_\varepsilon Id - \mathcal{K}_D^*\big)^{-1}[\nu]\cdot d\big),$$

and hence

$$|u|^2 \approx 1+2k_m\Re\left(i\mathcal{S}_D\big(\lambda_\varepsilon Id-\mathcal{K}_D^*\big)^{-1}[\nu]\cdot d\right)+\left|k_m\mathcal{S}_D\big(\lambda_\varepsilon Id-\mathcal{K}_D^*\big)^{-1}[\nu])\cdot d\right|^2. \tag{4.20}$$

Using Lemma A.0.2, we can write

$$\mathcal{S}_D\big(\lambda_\varepsilon Id-\mathcal{K}_D^*\big)^{-1}[\nu]\cdot d=\sum_{j=1}^{\infty}\frac{(\nu\cdot d,\varphi_j)_{\mathcal{H}^*}\mathcal{S}_D[\varphi_j]}{\lambda_\varepsilon-\lambda_j},$$

and therefore, for a given plasmonic frequency ω, we have

$$\mathcal{S}_D\big(\lambda_\varepsilon Id-\mathcal{K}_D^*\big)^{-1}[\nu]\cdot d\approx\frac{(\nu\cdot d,\varphi_{j^*})_{\mathcal{H}^*}\mathcal{S}_D[\varphi_{j^*}]}{\lambda_\varepsilon(\omega)-\lambda_{j^*}}.$$

Here j^* is such that $\lambda_{j^*}=\Re(\lambda_\varepsilon(\omega))$ and the eignevalue λ_{j^*} is assumed to be simple. If this was not the case, $(\nu\cdot d,\varphi_{j^*})_{\mathcal{H}^*}\mathcal{S}_D[\varphi_{j^*}]$ should be replaced by the corresponding sum over an orthonormal basis of eigenfunctions for the eigenspace associated to λ_{j^*}.

Replacing in (4.20) we find

$$|u|^2\approx 1+2k_m\frac{(\nu\cdot d,\varphi_{j^*})_{\mathcal{H}^*}\mathcal{S}_D[\varphi_{j^*}]}{|\lambda_\varepsilon(\omega)-\lambda_{j^*}|}+k_m^2\frac{(\nu\cdot d,\varphi_{j^*})^2_{\mathcal{H}^*}\mathcal{S}_D[\varphi_{j^*}]^2}{|\lambda_\varepsilon(\omega)-\lambda_{j^*}|^2}.$$

Thus, at a plasmonic resonance ω,

$$\begin{aligned}
F_D[g_u] &\approx \left(F_D[1]+2k_m\frac{(\nu\cdot d,\varphi_{j^*})_{\mathcal{H}^*}}{|\lambda_\varepsilon(\omega)-\lambda_{j^*}|}F_D[\mathcal{S}_D[\varphi_{j^*}]]+k_m^2\frac{(\nu\cdot d,\varphi_{j^*})^2_{\mathcal{H}^*}}{|\lambda_\varepsilon(\omega)-\lambda_{j^*}|^2}F_D[\mathcal{S}_D[\varphi_{j^*}]^2]\right),\\
\frac{\partial F_D}{\partial\nu} &\approx \left(2k_m\frac{(\nu\cdot d,\varphi_{j^*})_{\mathcal{H}^*}}{|\lambda_\varepsilon(\omega)-\lambda_{j^*}|}\frac{\partial F_D[\mathcal{S}_D[\varphi_{j^*}]]}{\partial\nu}+k_m^2\frac{(\nu\cdot d,\varphi_{j^*})^2_{\mathcal{H}^*}}{|\lambda_\varepsilon(\omega)-\lambda_{j^*}|^2}\frac{\partial F_D[\mathcal{S}_D[\varphi_{j^*}]^2]}{\partial\nu}\right).
\end{aligned}$$

Then, the temperature on the boundary of a nanoparticle at the plasmonic resonance can be estimated by plugging the above approximations of F_D and $\dfrac{\partial F_D(x,t,b_c)}{\partial\nu}$ into

$$\tau(x,t)=F_D(x,t,b_c)-\mathcal{V}_D^{b_c}(\lambda_\gamma Id-\mathcal{K}_D^*)^{-1}[\frac{\partial F_D(\cdot,\cdot,b_c)}{\partial\nu}](x,t)+O\left(\frac{\delta^4\log\delta}{\operatorname{dist}(\lambda_\varepsilon,\sigma(\mathcal{K}_D^*))^2}\right).$$

4.3.4 Temperature elevation for two close-to-touching particles

Lemma A.0.4 implies that

$$\frac{\partial F_D(x,t,b_c)}{\partial\nu}=-(\frac{1}{2}Id-\mathcal{K}_D^*)\widetilde{\mathcal{S}}_D^{-1}[F_D](x,t)+O\left(\frac{\delta^4\log\delta}{\operatorname{dist}(\lambda_\varepsilon,\sigma(\mathcal{K}_D^*))^2}\right).$$

Therefore, we can write the temperature on the boundary of the nanoparticle as

$$\tau(x,t)=F_D(x,t,b_c)+\mathcal{V}_D^{b_c}(\lambda_\gamma Id-\mathcal{K}_D^*)^{-1}\mathcal{P}_{\mathcal{H}^*\setminus E_{\frac{1}{2}}}[\frac{\partial F_D(\cdot,\cdot,b_c)}{\partial\nu}](x,t)+O\left(\frac{\delta^4\log\delta}{\operatorname{dist}(\lambda_\varepsilon,\sigma(\mathcal{K}_D^*))^2}\right), \tag{4.21}$$

where $\mathcal{P}_{\mathcal{H}^*\backslash E_{\frac{1}{2}}}$ is the projection into $\mathcal{H}^*\backslash E_{\frac{1}{2}}$: the complement in $\mathcal{H}^*(\partial\mathcal{D})$ of the eigenspace associated to the eigenvalue $\frac{1}{2}$ of $\mathcal{K}_D^*$. This implies that, even if λ_γ is close to $\frac{1}{2}$, the quantity $(\lambda_\gamma Id - \mathcal{K}_D^*)^{-1}\mathcal{P}_{\mathcal{H}^*\backslash E_{\frac{1}{2}}}[\frac{\partial F_D(\cdot,\cdot,b_c)}{\partial\nu}](x,t)$ will remain of order $O\left(\frac{\delta^2}{\text{dist}(\lambda_\varepsilon,\sigma(\mathcal{K}_D^*))^2}\right)$, provided that the second largest eigenvalue of $\mathcal{K}_D^*$ is not close to $\frac{1}{2}$.

Even if this is in general the case for smooth boundaries ∂D, it turns out that for nanoparticles with two connected close-to-touching subparts with contact of order m, a family of eigenvalues of $\mathcal{K}_D^*$ in $\mathcal{H}^*\backslash E_{\frac{1}{2}}$ approaches $\frac{1}{2}$ as (see [42])

$$\lambda_n^\zeta \sim \frac{1}{2} - c_n\zeta^{1-\frac{1}{m}} + o(\zeta^{1-\frac{1}{m}}),$$

where ζ is the distance between connected subparts and c_n is an increasing sequence of positive numbers.

Now, $\lambda_\gamma \approx \frac{1}{2}$ is the kind of situations encountered for metallic nanoparticles immersed in water or some biological tissue. As an example, the thermal conductivity of gold is $\gamma_c = 318\frac{W}{mK}$ and that of pure water is $\gamma_m = 0.6\frac{W}{mK}$. This gives $\lambda_\gamma \approx 0.5019$.

In view of this, the second term in (4.21) may increase considerably for some type of close-to-touching particles.

We stress, nevertheless, that this is not the general case. For a more refined analysis, asymptotics of the eigenfunctions of $\mathcal{K}_D^*$ should be also studied.

4.4 Numerical results

The numerical experiments for this work can be divided into two parts. The first one is the Helmholtz equation solution approximation, which is obtained by using Theorem 4.2.1. The second part is the Heat equation solution computation, which is obtained using Theorem 4.2.2.

The major tasks surrounding the numerical implementation of these formulas are integrating against a singular kernel. The numerical computations of the operators $F_D[\cdot]$ and $\partial_\nu F_D[\cdot]$ can be achieved by meshing the domain D and integrating semi-analytically inside the triangles that are close to the singularities. We used the following formula to avoid numerical differentiation:

$$\frac{\partial F_D(x,t,b_c)}{\partial\nu} = \frac{1}{2\pi b_c}\int_D \exp\left(\frac{-|x-y|^2}{4b_c t}\right)\frac{\langle y-x,\nu_x\rangle}{|x-y|^2}g_u(y)dy, \quad x\in\partial D. \tag{4.22}$$

For all the presented simulations, we considered an incident plane wave given by

$$u^i(x) = e^{ik_m d\cdot x},$$

where $d = (1,1)/\sqrt{2} \in \mathbb{R}^2$ is the illumination direction and $k_m = 2\pi/750\cdot 10^9$ is the frequency (in the red range). The considered nanoparticles are ellipses with semi-axes $30nm$ and $20nm$, respectively.

It is worth noticing that the illumination direction d is relevant solely in the asymptotic formula in Theorem 4.2.1. Its role is to define the coefficients

of a linear combination of both components of $\mathcal{S}_D(\lambda_\epsilon Id - \mathcal{K}_D^*)^{-1}[v] \in \mathbb{R}^2$. We will see from the numerical simulations that this is fundamental if we wish to maximize the produced electromagnetic field, and therefore the generated heat inside the nanoparticles.

With respect to the asymptotic formula established in Theorem 4.2.1, besides the nanoparticle's shape D, the sole parameter that is left is λ_ϵ. For all the following simulations we will consider this as a free parameter that we will use to excite the eigenvalues of the Neumann-Poincaré operator and hence to generate resonances. The physical justification that allows us to do this is based on the Drude model [9]. Whenever we mention that we approach a particular eigenvalue λ_j of $\mathcal{K}_D^*$, we will adopt $\lambda_\epsilon = \lambda_j + 0.001i$.

With respect to the heat equation coefficients, we use realistic values of gold for nanoparticles, and water for tissues.

4.4.1 Single-particle simulation

We consider one elliptical nanoparticle $D \Subset \mathbb{R}^2$ centered at the origin, with its semi-major axis aligned with the x-axis.

Single-particle Helmholtz resonance

Resonance is achieved by approaching the eigenvalues of the Neumann-Poincaré operator $\mathcal{K}_D^*$ with λ_ϵ, and afterwards applying it to each of the components of the normal ν to ∂D. It turns out that for some eigenfunctions of $\mathcal{K}_D^*$, the normal of the shape is almost orthogonal, in $\mathcal{H}^*(\partial D)$, to them. Therefore, we cannot observe resonance for their associated eigenvalues. In Figure 4.1 we can see values of the inner product between the eigenfunctions of $\mathcal{K}_D^*$ and the components ν_x and ν_y of ν. Figure 4.1 suggests us which are the available resonant modes with the respective strength of each coordinate. In

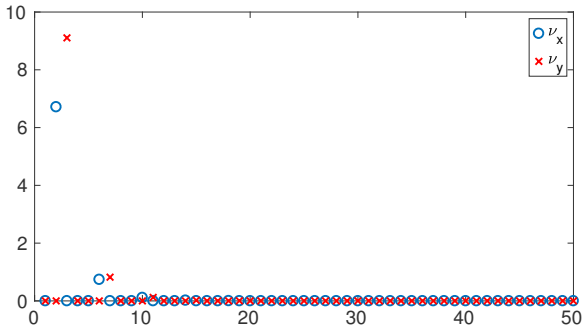

FIGURE 4.1: Inner product in $\mathcal{H}^*(\partial D)$ between the eigenfunctions of $\mathcal{K}_D^*$ and the components ν_x and ν_y of the normal ν to ∂D.

Figure 4.2 we present the absolute value of the inner field for the first three resonant modes, corresponding to the second, third and sixth eigenvalue of $\mathcal{K}_D^*$, respectively. In Figure 4.3 we decompose the inner field into the zeroth-order and the first-order terms respectively given by $u^i(z) + \delta(x-z)\nabla u^i(z)$ and $\mathcal{S}_D\big(\lambda_\varepsilon Id - \mathcal{K}_D^*\big)^{-1}[\nu] \cdot \nabla u^i(z)$. Figure 4.4 shows the components of the vector $\mathcal{S}_D(\lambda_\epsilon Id - \mathcal{K}_D^*)^{-1}[v]$.

From Figure 4.3, we can see that when we excite the nanoparticle at its resonant mode, the largest contribution to the electromagnetic field comes

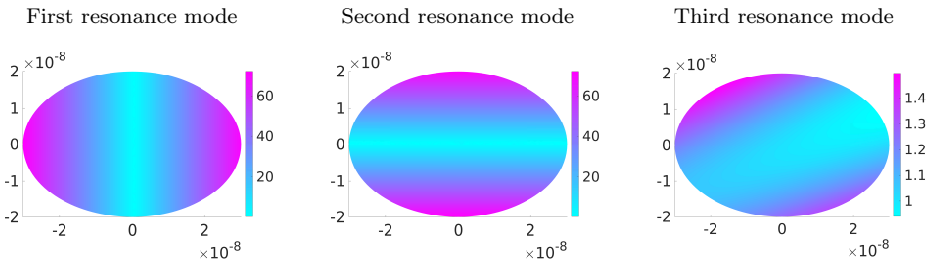

FIGURE 4.2: Absolute value of the electromagnetic field inside the nanoparticle at the first resonant modes, being those when λ_ϵ approaches the second, third and sixth eigenvalue of $\mathcal{K}_D^*$.

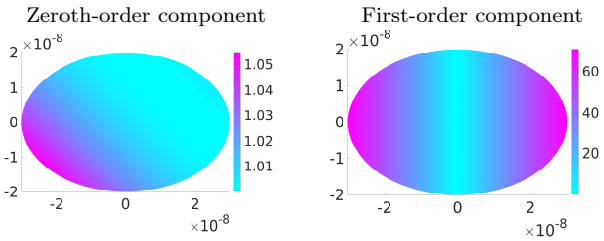

FIGURE 4.3: First resonant mode of the nanoparticle decomposed in its first- and second-order term in the formula given by Theorem 4.2.1. Both images are absolute values of the respective component.

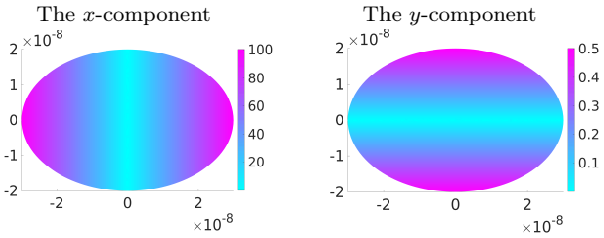

FIGURE 4.4: Absolute value of the vectorial components of the first-order term for the first resonant mode.

from the first-order term of the small volume expansion formula established in Theorem 4.2.1.

Observing the vectorial components of the first-order term in Figure 4.4 tells us how important is the illumination direction as the x-component is significantly stronger than the y-component. If we wish to maximize the electromagnetic field and therefore the generated heat, the recommended illumination direction would be around $d = (1, 0)^t$ (with t being the transpose), as it was initially suggested by Figure 4.1.

Single-particle surface heat generation

Considering the electromagnetic field inside the nanoparticle given by the first resonant mode presented in Figure 4.2, following the formula given by Theorem 4.2.2, we compute the generated heat on the surface of the nanoparticle. In Figure 4.5 we plot the generated heat in three dimensions and present a two dimensional plot obtained by parameterizing the boundary. In Figure 4.6 we decompose the heat in its first- and second-order terms given by

formula 4.2.2, being $F_D(x,t,b_c)$ and $-\mathcal{V}_D^{b_c}(\lambda_\gamma Id - \mathcal{K}_D^*)^{-1}[\frac{\partial F_D(\cdot,\cdot,b_c)}{\partial \nu}](x,t)$ respectively. In Figure 4.7, we integrate the total heat on the boundary and plot it as a function of time, for each component.

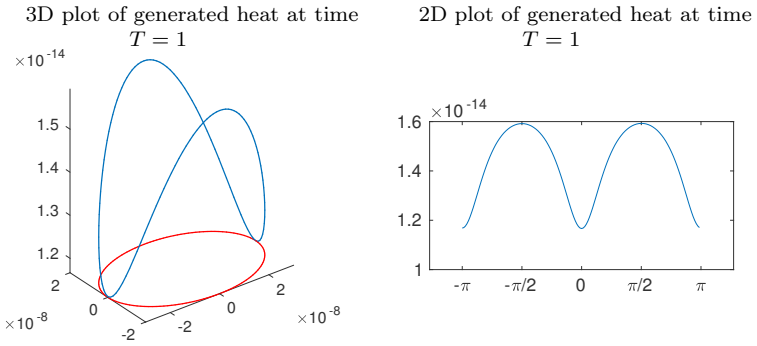

FIGURE 4.5: At the left-hand side, we can see a three-dimensional plot of the nanoparticle heat, the red shape is a reference value to show where the nanoparticle is located. At the right-hand side we can see a two-dimensional plot of the generated heat, where the boundary was parametrized following $p(\theta) = (a\cos(\theta), b\sin(\theta))$, $\theta \in [-\pi, \pi]$, with a and b being the semi-major and semi-minor axes, respectively.

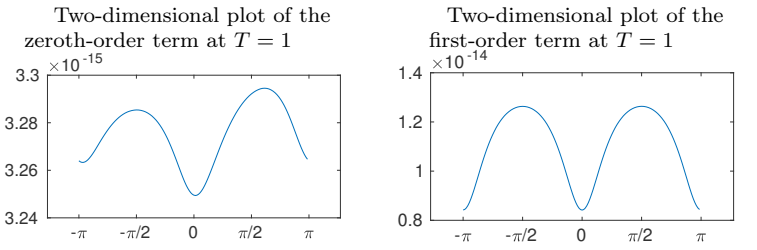

FIGURE 4.6: Two-dimensional plots of the zeroth- and first-order components of the heat on the boundary when time is equal to one. As time goes on, each point of the graph increases, but the general shape is preserved.

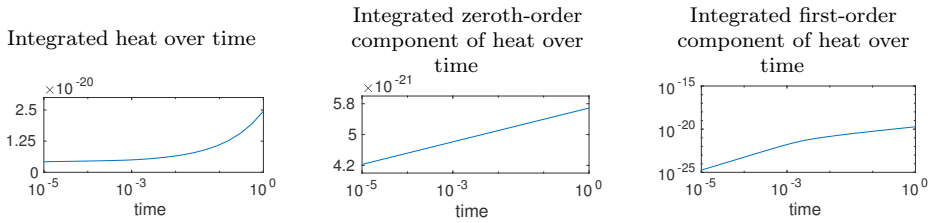

FIGURE 4.7: Time-logarithmic plots showing the total heat on the boundary for each component of the heat. The values were obtained for each fixed time, by integrating over the boundary the computed heat. From left- to right-hand side: The total heat, the zeroth-order and its first-order, according to formula given by Theorem 4.2.2. Notice that the first-order term is plotted in a log-log scale.

We can observe that the heat is not symmetric, this can be noticed from the total inner field for the first resonance mode in Figure 4.2. The reason behind this non symmetry is because we are illuminating with direction $d =$

$(1,1)^t/\sqrt{2}$ over an ellipse. From Figure 4.7 we can notice that the first-order term converges, while the zeroth-order term increases logarithmically, as it is expected from the known solution of the heat equation for constant source in two dimensions that the heat increases logarithmically.

4.4.2 Two particles simulation

We consider two elliptical nanoparticles D_1, D_2, $D = D_1 \cup D_2$, with the same shape and orientation as the nanoparticle considered in the above example. The particle D_1 is centered at the origin and D_2 is centered at $(0, 4.1 \cdot 10^{-9})$, resulting in a separation distance of $0.1nm$ between the two particles.

Two particles Helmholtz resonance

Following the same analysis as the one for one particle, in Figure 4.8 we present the inner product between the eigenfunctions of $\mathcal{K}_D^*$ with each component of the normal of D. We can observe that there are more available resonant modes. In particular we can see that when λ_ϵ approaches the 36th or 37th eigenvalues, we achieve strong resonant modes.

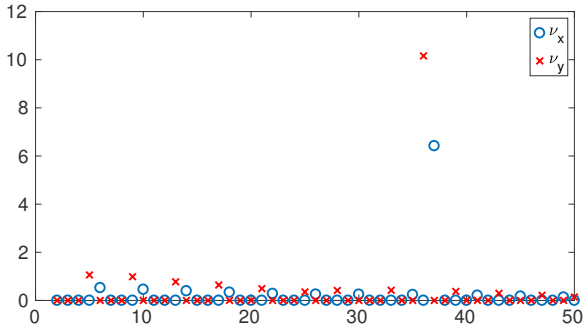

FIGURE 4.8: inner product in $\mathcal{H}^*\partial D$ between the eigenvalues of $\mathcal{K}_D^*$ and each component of the normal of ∂D, ν_x and ν_y.

In Figure 4.2 we present the absolute value of the inner field for the resonant modes corresponding to the 6th, 37th and 38th eigenvalues of $\mathcal{K}_D^*$. Similarly to the case with one particle, the dominant term in the electromagnetic field for each case is the first-order term. In Figure 4.10 we decompose the first-order term in its x-component and y-component.

As suggested by Figure 4.8, for the resonant mode associated to the 38th eigenfunction of $\mathcal{K}_D^*$, the stronger component is the one on the y direction, meaning that if we wish to maximize the electromagnetic field, and therefore the generated heat, it is suggested to consider the illumination vector $d = (0,1)^t$.

Two particles surface heat generation

Similarly to the analysis carried out for one particle, we now compute the generated heat for these two particles while undergoing resonance for the resonant mode associated to the 38th eigenvalue of $\mathcal{K}_D^*$. In Figure 4.11 we plot generated heat in the boundary of the two nanoparticles. In Figure 4.12 we decompose the generated heat in its zeroth and first-order component, for each of the two nanoparticles.

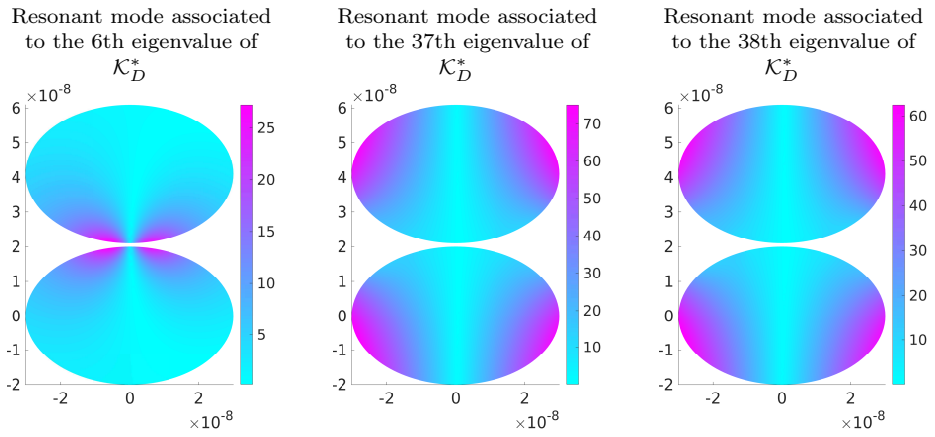

FIGURE 4.9: Absolute value of the electromagnetic field inside the nanoparticle at the resonant modes associated to the 6th, 37th and 38th resonant modes, obtained when λ_ϵ approaches the respective eigenvalues of $\mathcal{K}_D^*$.

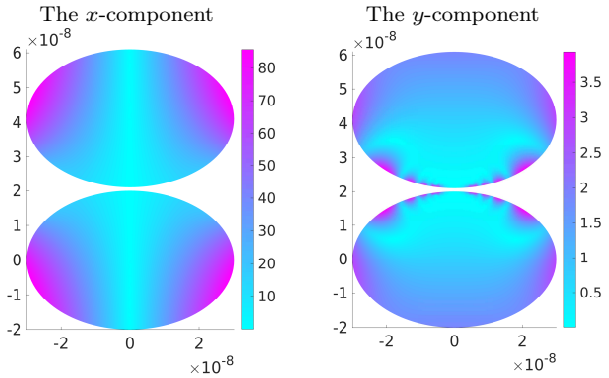

FIGURE 4.10: Absolute value of the vectorial components of the first-order term for the 38th resonant mode.

Similarly to the single nanoparticle case, there is no symmetry on the heat values on the boundary, which is due to the illumination. We have not provided the plots of the heat integrated along the boundary, as the conclusions are the same as the ones in the single nanoparticle case: The total heat on the boundary increases logarithmically, initially on time the dominant term is the fist-order one, but as time increases the zeroth-order term becomes the predominant one.

4.5 Concluding remarks

In this chapter we have derived an asymptotic formula for the temperature elevation due to plasmonic nanoparticles. We have considered thermal coupling within close-to-touching nanoparticles, where the temperature field deviates significantly from the one generated by single nanoparticles. Combined with the methods developed in [13, 14], our results can be used for the optical and thermal detection and localization of plasmonic nanoparticles. As reported in [77], the detection and localization of nanoparticles in highly scattering media such as biological tissue remains a challenge. They can also be used for monitoring temperature elevation due to plasmonic nanoparticles

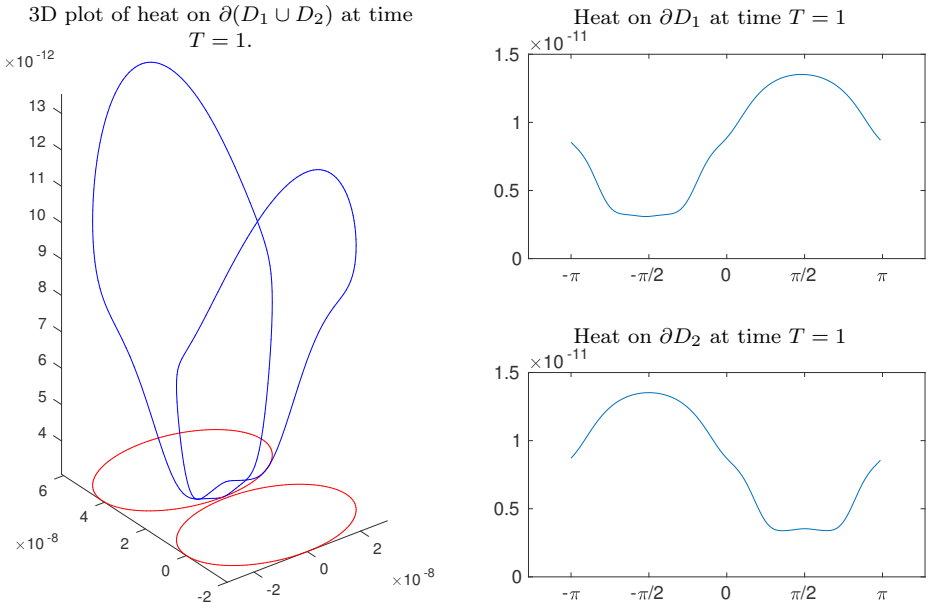

FIGURE 4.11: Generated heat on the boundary of the nanoparticles for time equal to 1. On the left we can see a three dimensional view of the heat, the red shapes are referential to show the location of the nanoparticles. On the right-hand side we can see the two dimensional heat plots corresponding to each nanoparticle. To obtain these plots we parameterized the boundary of each nanoparticle with $p(\theta) = (a\cos(\theta), b\sin(\theta)) + z$, $\theta \in [-\pi, \pi]$, where $z \in \mathbb{R}^2$ corresponds to the center of each nanoparticle. On the top we can see nanoparticle D_2 and on the bottom nanoparticle D_1.

based on the photoacoustic signal recently analyzed in [91]. Thermoacoustic signals generated by nanoparticle heating can be computed numerically based on the successive resolution of the thermal diffusion problem considered in this chapter and a thermoelastic problem, taking into account the size and shape of the nanoparticle, thermoelastic and elastic properties of both the particle and its environment, and the temperature-dependence of the thermal expansion coefficient of the environment. For sufficiently high illumination fluences, this temperature dependence yields a nonlinear relationship between the photoacoustic amplitude and the fluence [82].

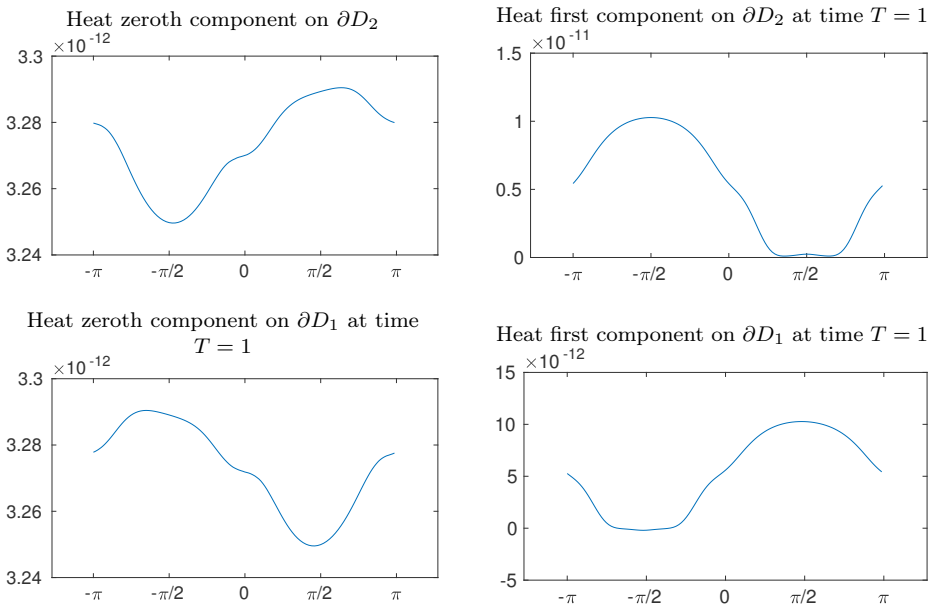

FIGURE 4.12: Two-dimensional plots of the zeroth and first component of the heat at time 1, for each nanoparticle. On the left column we have the zeroth component of the heat, on the right-hand side column we have the first component of the heat. On top we show the values for nanoparticle D_2, on the bottom we show the values for nanoparticle D_1.

Chapter 5

Plasmonic Metasurfaces

Contents

5.1 Introduction

In this chapter we consider the scattering by a layer of periodic plasmonic nanoparticles mounted on a perfectly conducting sheet. We design the layer in order to control and transform waves. Since the thickness of the layer, which is of the same order of the diameter of the individual nanoparticles, is negligible compared to the wavelength, it can be approximated by an impedance boundary condition. Our main result is to prove that at some resonant frequencies, which are fully characterized in terms of the periodicity, the shape and the material parameters of the nanoparticles, the thin layer has anomalous reflection properties and can be viewed as a metasurface. Since the period of the array is much smaller than the wavelength, the resonant frequencies of the array of nanoparticles differ significantly from those of single nanoparticles. As shown in this chapter, they are associated with eigenvalues of a periodic Neumann-Poincaré type operator. In contrast with quasi-static plasmonic resonances of single nanoparticles, they depend on the particle size. For simplicity, only one-dimensional arrays embedded in $\mathbb{R}^2$ are considered in this chapter. The extension to the two-dimensional case is straightforward and the dependence of the plasmonic resonances on the parameters of the lattice is easy to derive.

The array of plasmonic nanoparticles can be used to efficiently reduce the scattering of the perfectly conducting sheet. We present numerical results to illustrate our main findings in this chapter, which open a door for a mathematical and numerical framework for realizing full control of waves using metasurfaces [2, 76, 92]. Our approach applies to any example of periodic distributions of resonators having resonances in the quasi-static regime. It provides a framework for explaining the observed extraordinary or meta properties of such structures and for optimizing these properties. The results presented in this chapter hold for arbitrary-shaped nanoparticles. Simulations with disks, ellipses, and rings are shown. In this connection, we refer to the recent works [59, 70, 80, 93]. It is also worth highlighting that at optical frequencies, a perfectly conducting approximation breaks down and needs to be replaced by a proper material response. In this chapter, the perfectly conducting boundary condition is used only for simplicity of the presentation. Similar effective boundary conditions can be obtained by using exactly the same approach presented here for penetrable half-space.

The chapter is organized as follows. We first formulate the problem of approximating the effect of a thin layer with impedance boundary conditions and give useful results on the one-dimensional periodic Green function. Then we derive the effective impedance boundary conditions and give the shape derivative of the impedance parameter. In doing so, we analyze the spectral properties of the one-dimensional periodic Neumann-Poincaré operator defined by (5.10) and obtain an explicit formula for the equivalent boundary condition in terms of its eigenvalues and eigenvectors. Finally, we illustrate with a few numerical experiments the anomalous change in the equivalent impedance boundary condition due to the plasmonic resonances of the periodic array of nanoparticles. For simplicity, we only consider the scalar wave equation and use a two-dimensional setup. The results of this chapter can be readily generalized to higher dimensions as well as to the full Maxwell equations.

5.2 Setting of the problem

We use the Helmholtz equation to model the propagation of light. This approximation can be viewed as a special case of Maxwell's equations, when the incident wave u^i is transverse magnetic (TM) or transverse electric (TE) polarized.

Consider a particle occupying a bounded domain $D \Subset \mathbb{R}^2$ of class $\mathcal{C}^{1,\alpha}$ for some $0 < \alpha < 1$ and with size of order $\delta \ll 1$. The particle is characterized by electric permittivity ε_c and magnetic permeability μ_c, both of which may depend on the frequency of the incident wave. Assume that $\Im m\, \varepsilon_c > 0, \Re e\, \mu_c < 0, \Im m\, \mu_c > 0$ and define

$$k_m = \omega\sqrt{\varepsilon_m \mu_m}, \quad k_c = \omega\sqrt{\varepsilon_c \mu_c},$$

where ε_m and μ_m are the permittivity and permeability of free space respectively and ω is the frequency. Throughout this chapter, we assume that ε_m and μ_m are real and positive and k_m is of order 1.

We consider the configuration shown in Figure 5.1, where a particle D is repeated periodically in the x_1-axis with period δ, and is at a distance of order δ from the boundary $x_2 = 0$ of the half-space $\mathbb{R}^2_+ := \{(x_1, x_2) \in \mathbb{R}^2,\ x_2 > 0\}$. We denote by $\mathcal{D}$ this collection of periodically arranged particles and $\Omega := \mathbb{R}^2_+ \setminus \overline{\mathcal{D}}$.

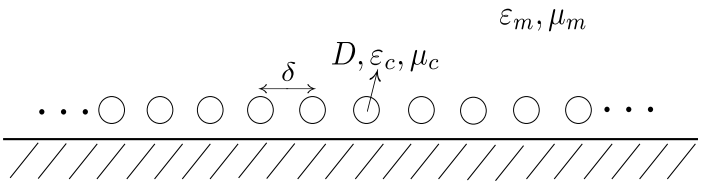

FIGURE 5.1: Thin layer of nanoparticles in the half space.

Let $u^i(x) = e^{ik_m d\cdot x}$ be the incident wave. Here, d is the unit incidence direction. The scattering problem is modeled as follows

$$\begin{cases} \nabla \cdot \dfrac{1}{\mu_{\mathcal{D}}} \nabla u + \omega^2 \varepsilon_{\mathcal{D}} u = 0 \quad \text{in } \mathbb{R}^2_+ \setminus \partial\mathcal{D}, \\ u_+ - u_- = 0 \quad \text{on } \partial\mathcal{D}, \\ \dfrac{1}{\mu_m} \dfrac{\partial u}{\partial \nu}\Big|_+ - \dfrac{1}{\mu_c} \dfrac{\partial u}{\partial \nu}\Big|_- = 0 \quad \text{on } \partial\mathcal{D}, \\ u - u^i \text{ satisfies an outgoing radiation condition at infinity}, \\ u = 0 \quad \text{on } \partial\mathbb{R}^2_+ = \{(x_1, 0),\ x_1 \in \mathbb{R}\}, \end{cases} \tag{5.1}$$

where

$$\varepsilon_{\mathcal{D}} = \varepsilon_m \chi(\Omega) + \varepsilon_c \chi(\mathcal{D}), \quad \mu_{\mathcal{D}} = \varepsilon_m \chi(\Omega) + \varepsilon_c \chi(\mathcal{D}),$$

and $\partial/\partial\nu$ denotes the outward normal derivative on $\partial\mathcal{D}$.

Following [1], under the assumption that the wavelength of the incident wave is much larger than the size of the nanoparticle, a certain homogenization occurs, and we can construct $z \in \mathbb{C}$ such that the solution to

$$\begin{cases} \Delta u_{\text{app}} + k_m^2 u_{\text{app}} = 0 \quad \text{in } \mathbb{R}^2_+, \\ u_{\text{app}} + \delta z \frac{\partial u_{\text{app}}}{\partial x_2} = 0 \quad \text{on } \partial\mathbb{R}^2_+, \\ u_{\text{app}} - u^i \text{ satisfies outgoing radiation condition at infinity,} \end{cases} \tag{5.2}$$

gives the leading order approximation for u. We will refer to $u_{\text{app}} + \delta z \frac{\partial u_{\text{app}}}{\partial x_2} = 0$ as the equivalent impedance boundary condition for problem (5.1). A proof of existence and uniqueness of a solution to (5.2) follows immediately from [45].

5.3 One-dimensional periodic Green function

Consider the function $G_\sharp : \mathbb{R}^2 \to \mathbb{C}$ satisfying

$$\Delta G_\sharp(x) = \sum_{n\in\mathbb{Z}} \delta(x + (n, 0)). \tag{5.3}$$

We call $G_\sharp$ the 1-d periodic Green function for $\mathbb{R}^2$.

Lemma 5.3.1. *Let $x = (x_1, x_2)$, then*

$$G_\sharp(x) = \frac{1}{4\pi} \log\big(\sinh^2(\pi x_2) + \sin^2(\pi x_1)\big),$$

satisfies (5.3).

Proof. We have

$$\begin{aligned} \Delta G_\sharp(x) &= \sum_{n\in\mathbb{Z}} \delta(x + (n, 0)) \\ &= \sum_{n\in\mathbb{Z}} \delta(x_2)\delta(x_1 + n) \\ &= \sum_{n\in\mathbb{Z}} \delta(x_2) e^{i2\pi n x_1}, \end{aligned} \tag{5.4}$$

where we have used the Poisson summation formula $\sum_{n\in\mathbb{Z}} \delta(x_1 + n) = \sum_{n\in\mathbb{Z}} e^{i2\pi n x_1}$.

On the other hand, since $G_\sharp$ is periodic in x_1 of period 1, we have

$$G_\sharp(x) = \sum_{n\in\mathbb{Z}} \beta_n(x_2) e^{i2\pi n x_1},$$

therefore

$$\Delta G_\sharp(x) = \sum_{n\in\mathbb{Z}} (\beta_n''(x_2) + (i2\pi n)^2 \beta_n) e^{i2\pi n x_1}. \tag{5.5}$$

Comparing (5.4) and (5.5) yields

$$\beta_n''(x_2) + (i2\pi n)^2 \beta_n = \delta(x_2).$$

A solution to the previous equation can be found by using standard techniques for ordinary differential equations. We have

$$\begin{aligned}\beta_0 &= \frac{1}{2}|x_2| + c,\\ \beta_n &= \frac{-1}{4\pi|n|}e^{-2\pi|n||x_2|}, \quad n \neq 0,\end{aligned}$$

where c is a constant. Subsequently,

$$\begin{aligned}G_\sharp(x) &= \frac{1}{2}|x_2| + c - \sum_{n\in\mathbb{Z}\setminus\{0\}} \frac{1}{4\pi|n|}e^{-2\pi|n||x_2|}e^{i2\pi n x_1}\\ &= \frac{1}{2}|x_2| + c - \sum_{n\in\mathbb{N}\setminus\{0\}} \frac{1}{2\pi n}e^{-2\pi n|x_2|}\cos(2\pi n x_1)\\ &= \frac{1}{4\pi}\log\big(\sinh^2(\pi x_2) + \sin^2(\pi x_1)\big),\end{aligned}$$

where we have used the summation identity (see, for instance, [57, pp. 813-814])

$$\begin{aligned}\sum_{n\in\mathbb{N}\setminus\{0\}} \frac{1}{2\pi n}e^{-2\pi n|x_2|}\cos(i2\pi n x_1) = \frac{1}{2}|x_2| - \frac{\log(2)}{2\pi}\\ -\frac{1}{4\pi}\log\big(\sinh^2(\pi x_2) + \sin^2(\pi x_1)\big),\end{aligned}$$

and defined $c = -\dfrac{\log(2)}{2\pi}$. □

Let us also denote by $G_\sharp(x,y) := G_\sharp(x-y)$. In the following we define the 1-d periodic single layer potential and 1-d periodic Neumann-Poincaré operator, respectively, for a bounded domain $B \Subset \big(-\frac{1}{2},\frac{1}{2}\big)\times\mathbb{R}$ which we assume to be of class $\mathcal{C}^{1,\alpha}$ for some $0<\alpha<1$. Let

$$\begin{aligned}\mathcal{S}_{B\sharp} : H^{-\frac{1}{2}}(\partial B) &\longrightarrow H^1_{\mathrm{loc}}(\mathbb{R}^2), H^{\frac{1}{2}}(\partial B)\\ \varphi &\longmapsto \mathcal{S}_{B,\sharp}[\varphi](x) = \int_{\partial B} G_\sharp(x,y)\varphi(y)d\sigma(y)\end{aligned}$$

for $x\in\mathbb{R}^2, x\in\partial B$ and let

$$\begin{aligned}\mathcal{K}^*_{B\sharp} : H^{-\frac{1}{2}}(\partial B) &\longrightarrow H^{-\frac{1}{2}}(\partial B)\\ \varphi &\longmapsto \mathcal{K}^*_{B,\sharp}[\varphi](x) = \int_{\partial B} \frac{\partial G_\sharp(x,y)}{\partial\nu(x)}\varphi(y)d\sigma(y)\end{aligned}$$

for $x\in\partial B$. As in [65], the periodic Neumann-Poincaré operator can be symmetrized. The following lemma holds.

Lemma 5.3.2. *(i) For any $\varphi\in H^{-\frac{1}{2}}(\partial B)$, $\mathcal{S}_{B\sharp}[\varphi]$ is harmonic in B and in $\big(-\frac{1}{2},\frac{1}{2}\big)\times\mathbb{R}\setminus\overline{B}$;*

(ii) The following trace formula holds: for any $\varphi \in H^{-\frac{1}{2}}(\partial B)$,

$$(-\frac{1}{2}Id + \mathcal{K}^*_{B\sharp})[\varphi] = \frac{\partial \mathcal{S}_{B\sharp}[\varphi]}{\partial \nu}\Big|_-;$$

(iii) The following Calderón identity holds: $\mathcal{K}_{B\sharp}\mathcal{S}_{B\sharp} = \mathcal{S}_{B\sharp}\mathcal{K}^*_{B\sharp}$, *where* $\mathcal{K}_{B\sharp}$ *is the* L^2*-adjoint of* $\mathcal{K}^*_{B\sharp}$;

(iv) The operator $\mathcal{K}^*_{B\sharp} : H_0^{-\frac{1}{2}}(\partial B) \to H_0^{-\frac{1}{2}}(\partial B)$ *is compact self-adjoint equipped with the following inner product*

$$(u, v)_{\mathcal{H}_0^*} = -(u, \mathcal{S}_{B\sharp}[v])_{-\frac{1}{2},\frac{1}{2}} \tag{5.6}$$

with $(\cdot,\cdot)_{-\frac{1}{2},\frac{1}{2}}$ *being the duality pairing between* $H_0^{-\frac{1}{2}}(\partial B)$ *and* $H_0^{\frac{1}{2}}(\partial B)$, *which makes* $\mathcal{H}_0^*$ *equivalent to* $H_0^{-\frac{1}{2}}(\partial B)$. *Here, by* E_0 *we denote the zero-mean subspace of* E.

(v) Let (λ_j, φ_j), $j = 1, 2, \ldots$ *be the eigenvalue and normalized eigenfunction pair of* $\mathcal{K}^*_{B\sharp}$ *in* $\mathcal{H}_0^*(\partial B)$, *then* $\lambda_j \in (-\frac{1}{2}, \frac{1}{2})$ *and* $\lambda_j \to 0$ *as* $j \to \infty$.

Proof. First, note that a Taylor expansion of $\sinh^2(\pi x_2) + \sin^2(\pi x_1)$ yields

$$G_\sharp(x) = \frac{\log|x|}{2\pi} + R(x),$$

where R is a smooth function such that

$$R(x) = \frac{1}{4\pi}\log(1 + O(x_2^2 - x_1^2)).$$

We can decompose the operators $\mathcal{S}_{B\sharp}$ and $\mathcal{K}^*_{B\sharp}$ on $\mathcal{H}_0^*(\partial B)$ accordingly. We have

$$\mathcal{S}_{B\sharp} = \mathcal{S}_B + \mathcal{G}_B, \quad \mathcal{K}^*_{B\sharp} = \mathcal{K}^*_B + \mathcal{F}_B,$$

where $\mathcal{S}_B$ and $\mathcal{K}^*_B$ are the single layer potential and Neumann-Poincaré operator (see [18]), respectively, and $\mathcal{G}_B, \mathcal{F}_B$ are smoothing operators. Using this fact, the proof of the Lemma follows the same arguments as those given in [12, 18]. □

5.4 Boundary layer corrector and effective impedance

In order to compute z, we introduce the following asymptotic expansion [1, 3]:

$$u = u^{(0)} + u^{(0)}_{BL} + \delta(u^{(1)} + u^{(1)}_{BL}) + \ldots \tag{5.7}$$

where the leading-order term $u^{(0)}$ is solution to

$$\begin{cases} \Delta u^{(0)} + k_m^2 u^{(0)} = 0 \quad \text{in } \mathbb{R}^2_+, \\ u^{(0)} = 0 \quad \text{on } \partial\mathbb{R}^2_+, \\ u^{(0)} - u^i \text{ satisfies an outgoing radiation condition at infinity.} \end{cases}$$

The boundary-layer correctors $u_{BL}^{(0)}$ and $u_{BL}^{(1)}$ have to be exponentially decaying in the x_2-direction. Note that according to [1, 3], $u_{BL}^{(0)}$ is introduced in order to correct (up to the first-order in δ) the transmission condition on the boundary of the nanoparticles, which is not satisfied by the leading-order term $u^{(0)}$ in the asymptotic expansion of u, while $u_{BL}^{(1)}$ is a higher-order correction term and does not contribute to the first-order equivalent boundary condition in (5.2).

We next construct the corrector $u_{BL}^{(0)}$. We first introduce a function α and a complex constant α_∞ such that they satisfy the rescaled problem:

$$\begin{cases} \Delta\alpha = 0 \quad \text{in } \left(\mathbb{R}^2_+ \backslash \overline{\mathcal{B}}\right) \cup \mathcal{B}, \\ \alpha|_+ - \alpha|_- = 0 \quad \text{on } \partial\mathcal{B}, \\ \dfrac{1}{\mu_m}\dfrac{\partial\alpha}{\partial\nu}\Big|_+ - \dfrac{1}{\mu_c}\dfrac{\partial\alpha}{\partial\nu}\Big|_- = \left(\dfrac{1}{\mu_c} - \dfrac{1}{\mu_m}\right)\nu_2 \quad \text{on } \partial\mathcal{B}, \\ \alpha = 0 \quad \text{on } \partial\mathbb{R}^2_+, \\ \alpha - \alpha_\infty \text{ is exponentially decaying as } x_2 \to +\infty. \end{cases} \tag{5.8}$$

Here, $\nu = (\nu_1, \nu_2)$ and $B = D/\delta$ is repeated periodically in the x_1-axis with period 1 and $\mathcal{B}$ is the collection of these periodically arranged particles.

Then $u_{BL}^{(0)}$ is defined by

$$u_{BL}^{(0)}(x) := \delta \frac{\partial u^{(0)}}{\partial x_2}(x_1, 0)\left(\alpha(\frac{x}{\delta}) - \alpha_\infty\right).$$

The corrector $u^{(1)}$ can be found to be the solution to

$$\begin{cases} \Delta u^{(1)} + k_m^2 u^{(1)} = 0 \quad \text{in } \mathbb{R}^2_+, \\ u^{(1)} = \alpha_\infty \frac{\partial u^{(0)}}{\partial x_2} \quad \text{on } \partial\mathbb{R}^2_+, \\ u^{(1)} \text{ satisfies an outgoing radiation condition at infinity.} \end{cases}$$

By writing

$$u_{\text{app}} = u^{(0)} + \delta u^{(1)}, \tag{5.9}$$

we arrive at (5.2) with $z = -\alpha_\infty$, up to a second order term in δ. We summarize the above results in the following theorem.

Theorem 5.4.1. *The solution u_{app} to (5.2) with $z = -\alpha_\infty$ approximates pointwisely (for $x_2 > 0$) the exact solution u to (5.1) as $\delta \to 0$, up to a second order term in δ.*

In order to compute α_∞, we derive an integral representation for the solution α to (5.8). We make use of the periodic Green function $G_\sharp$ defined by (5.3). Let

$$G_\sharp^+(x, y) = G_\sharp\big((x_1 - y_1, x_2 - y_2)\big) - G_\sharp\big((x_1 - y_1, -x_2 - y_2)\big),$$

which is the periodic Green's function in the upper half space with Dirichlet boundary conditions, and define

$$\begin{aligned} \mathcal{S}^+_{B\sharp} : H^{-\frac{1}{2}}(\partial B) &\longrightarrow H^1_{\mathrm{loc}}(\mathbb{R}^2), H^{\frac{1}{2}}(\partial B) \\ \varphi &\longmapsto \mathcal{S}^+_{B,\sharp}[\varphi](x) = \int_{\partial B} G^+_\sharp(x,y)\varphi(y)d\sigma(y) \end{aligned}$$

for $x \in \mathbb{R}^2_+, x \in \partial B$ and

$$\begin{aligned} &(\mathcal{K}^*_{B\sharp})^+ : H^{-\frac{1}{2}}(\partial B) \longrightarrow H^{-\frac{1}{2}}(\partial B) \\ &\varphi \longmapsto (\mathcal{K}^*_{B,\sharp})^+[\varphi](x) = \int_{\partial B} \frac{\partial G^+_\sharp(x,y)}{\partial \nu(x)}\varphi(y)d\sigma(y) \end{aligned} \tag{5.10}$$

for $x \in \partial B$.

It can be easily proved that all the results of Lemma 5.3.2 hold true for $\mathcal{S}^+_{B\sharp}$ and $(\mathcal{K}^*_{B\sharp})^+$. Moreover, for any $\varphi \in H^{-\frac{1}{2}}(\partial B)$, we have

$$\mathcal{S}^+_{B,\sharp}[\varphi](x) = 0 \quad \text{for } x \in \partial\mathbb{R}^2_+.$$

Now, we can readily see that α can be represented as $\alpha = \mathcal{S}^+_{B,\sharp}[\varphi]$, where $\varphi \in H^{-\frac{1}{2}}(\partial B)$ satisfies

$$\frac{1}{\mu_m}\frac{\partial \mathcal{S}^+_{B,\sharp}[\varphi]}{\partial \nu}\bigg|_+ - \frac{1}{\mu_c}\frac{\partial \mathcal{S}^+_{B,\sharp}[\varphi]}{\partial \nu}\bigg|_- = \Big(\frac{1}{\mu_c} - \frac{1}{\mu_m}\Big)\nu_2 \quad \text{on } \partial B.$$

Using the jump formula from Lemma 5.3.2, we arrive at

$$\big(\lambda_\mu Id - (\mathcal{K}^*_{B\sharp})^+\big)[\varphi] = \nu_2,$$

where

$$\lambda_\mu = \frac{\mu_c + \mu_m}{2(\mu_c - \mu_m)}.$$

Therefore, using item (v) in Lemma 5.3.2 on the characterization of the spectrum of $\mathcal{K}^*_{B\sharp}$ and the fact that the spectra of $(\mathcal{K}^*_{B\sharp})^+$ and $\mathcal{K}^*_{B\sharp}$ are the same, we obtain that

$$\alpha = \mathcal{S}^+_{B,\sharp}\big(\lambda_\mu Id - (\mathcal{K}^*_{B\sharp})^+\big)^{-1}[\nu_2].$$

Lemma 5.4.1. *Let $x = (x_1, x_2)$. Then, for $x_2 \to +\infty$, the following asymptotic expansion holds:*

$$\alpha = \alpha_\infty + O(e^{-x_2}),$$

with

$$\alpha_\infty = -\int_{\partial B} y_2\big(\lambda_\mu Id - (\mathcal{K}^*_{B\sharp})^+\big)^{-1}[\nu_2](y)d\sigma(y).$$

Proof. The result follows from an asymptotic analysis of $G_\sharp^+(x,y)$. Indeed, suppose that $x_2 \to +\infty$, we have

$$\begin{aligned}
&G_\sharp^+(x,y) = \tfrac{1}{4\pi}\log\big(\sinh^2(\pi(x_2-y_2)) + \sin^2(\pi(x_1-y_1))\big)\\
&-\frac{1}{4\pi}\log\big(\sinh^2(\pi(x_2+y_2)) + \sin^2(\pi(x_1-y_1))\big)\\
&= \frac{1}{4\pi}\log\big(\sinh^2(\pi(x_2-y_2))\big)\\
&-\frac{1}{4\pi}\log\big(\sinh^2(\pi(x_2+y_2))\big)\\
&+O\Big(\log\Big(1+\frac{1}{\sinh^2(x_2)}\Big)\Big)\\
&= \frac{1}{2\pi}\Big(\log\Big(\frac{e^{\pi(x_2-y_2)}-e^{-\pi(x_2+y_2)}}{2}\Big)\\
&-\log\Big(\frac{e^{\pi(x_2+y_2)}-e^{-\pi(x_2-y_2)}}{2}\Big)\Big) + O\Big(\log\Big(1+e^{-x_2^2}\Big)\Big)\\
&= -y_2 + O(e^{-x_2}),
\end{aligned}$$

which yields the desired result. □

Finally, it is important to note that α_∞ depends on the geometry and size of the particle B.

Since $(\mathcal{K}^*_{B\sharp})^+ : \mathcal{H}_0^* \to \mathcal{H}_0^*$ is a compact self-adjoint operator, where $\mathcal{H}_0^*$ is defined as in Lemma 5.3.2, we can write

$$\begin{aligned}
\alpha_\infty &= -\int_{\partial B} y_2\big(\lambda_\mu Id - (\mathcal{K}^*_{B\sharp})^+\big)^{-1}[\nu_2](y)d\sigma(y),\\
&= -\int_{\partial B} y_2\sum_{j=1}^{\infty}\frac{(\varphi_j,\nu_2)_{\mathcal{H}_0^*}\varphi_j(y)}{\lambda_\mu-\lambda_j}d\sigma(y),\\
&= \sum_{j=1}^{\infty}\frac{(\varphi_j,\nu_2)_{\mathcal{H}_0^*}(\varphi_j,y_2)_{-\frac{1}{2},\frac{1}{2}}}{\lambda_\mu-\lambda_j},
\end{aligned}$$

where $\lambda_1,\lambda_2,\dots$ are the eigenvalues of $(\mathcal{K}^*_{B\sharp})^+$ and $\varphi_1,\varphi_2,\dots$ is a corresponding orthornormal basis of eigenfunctions.

On the other hand, by integrating by parts we get

$$(\varphi_j,y_2)_{-\frac{1}{2},\frac{1}{2}} = \frac{1}{\frac{1}{2}-\lambda_j}(\varphi_j,\nu_2)_{\mathcal{H}_0^*}.$$

This together with the fact that $\Im m\,\lambda_\mu < 0$ (by the Drude model [9]), yield the following lemma.

Lemma 5.4.2. *We have* $\Im m\,\alpha_\infty > 0$.

Finally, we give a formula for the shape derivative [16] of α_∞. This formula can be used to optimize $|\alpha_\infty|$, for a given frequency ω, in terms of the shape B of the nanoparticle. Let B_η be an η-perturbation of B; i.e., let

$h \in \mathcal{C}^1(\partial B)$ and ∂B_η be given by

$$\partial B_\eta = \Big\{ x + \eta h(x)\nu(x), x \in \partial B \Big\}.$$

Following [17] (see also [12]), we can prove that

$$\begin{aligned} \alpha_\infty(B_\eta) &= \alpha_\infty(B) + \eta(\frac{\mu_m}{\mu_c} - 1) \\ &\quad \times \int_{\partial B} h \left[\frac{\partial v}{\partial \nu}|_- \frac{\partial w}{\partial \nu}|_- + \frac{\mu_c}{\mu_m} \frac{\partial v}{\partial \tau}|_- \frac{\partial w}{\partial \tau}|_- \right] d\sigma, \end{aligned}$$

where $\partial/\partial\tau$ is the tangential derivative on ∂B, v and w periodic with respect to x_1 of period 1 and satisfy

$$\begin{cases} \Delta v = 0 \quad \text{in } \left(\mathbb{R}^2_+ \backslash \overline{\mathcal{B}}\right) \cup \mathcal{B}, \\ v|_+ - v|_- = 0 \quad \text{on } \partial\mathcal{B}, \\ \left.\dfrac{\partial v}{\partial \nu}\right|_+ - \dfrac{\mu_m}{\mu_c} \left.\dfrac{\partial v}{\partial \nu}\right|_- = 0 \quad \text{on } \partial\mathcal{B}, \\ v - x_2 \to 0 \quad \text{as } x_2 \to +\infty, \end{cases}$$

and

$$\begin{cases} \Delta w = 0 \quad \text{in } \left(\mathbb{R}^2_+ \backslash \overline{\mathcal{B}}\right) \cup \mathcal{B}, \\ \frac{\mu_m}{\mu_c} w|_+ - w|_- = 0 \quad \text{on } \partial\mathcal{B}, \\ \left.\dfrac{\partial w}{\partial \nu}\right|_+ - \left.\dfrac{\partial w}{\partial \nu}\right|_- = 0 \quad \text{on } \partial\mathcal{B}, \\ w - x_2 \to 0 \quad \text{as } x_2 \to +\infty, \end{cases}$$

respectively. Therefore, the following lemma holds.

Lemma 5.4.3. *The shape derivative $d_S\alpha_\infty(B)$ of α_∞ is given by*

$$d_S\alpha_\infty(B) = (\frac{\mu_m}{\mu_c} - 1) \left[\frac{\partial v}{\partial \nu}|_- \frac{\partial w}{\partial \nu}|_- + \frac{\mu_c}{\mu_m} \frac{\partial v}{\partial \tau}|_- \frac{\partial w}{\partial \tau}|_- \right].$$

If we aim to maximize the functional $J := \frac{1}{2}|\alpha_\infty|^2$ over B, then it can be easily seen that J is Fréchet differentiable and its Fréchet derivative is given by $\Re e\, d_S\alpha_\infty(B)\overline{\alpha_\infty(B)}$. As in [11], in order to include cases where topology changes and multiple components are allowed, a level-set version of the optimization procedure described below can be developed.

5.5 Numerical illustrations

5.5.1 Setup and methods

We use the Drude model [9] to model the electromagnetic properties of the materials of our problem. We use water for the half space and gold for the metallic nanoparticles. We recall that, from the Drude model, the properties of the materials depend on the frequency of the incoming wave, or equivalently, on the wavelength. To compute $|\alpha_\infty|$ and the integral (geometry

dependent) operators involved on its expression, we make a simple uniform discretization with 200 points of the corresponding geometric figures and use a standard quadrature midpoint rule.

Figure 5.2 shows $|\alpha_\infty|$ as a function of the wavelength for disks of different sizes, all centered at $(0, 0.5)$.

Figure 5.3 shows $|\alpha_\infty|$ as a function of the wavelength for two disks of the same fixed radius equal to 0.2 but centered at two different distances from $x_2 = 0$.

In Figures 5.4 and 5.5 we plot $|\alpha_\infty|$ as a function of the wavelength for a disk and a group of three well-separated disks. We can see that a disk can be excited roughly at one single frequency whereas three disks can be excited at different frequencies but with lower values of $|\alpha_\infty|$.

The previous results consist only of nanodisks. Here we give a few other examples to confirm how general are the conclusions obtained. Figure 5.6 shows the blow up of $|\alpha_\infty|$ for an ellipse. In Figure 5.7 we consider a triangle with rounded corners. In Figure 5.8, values of $|\alpha_\infty|$ are computed for a circular ring.

5.5.2 Results and discussion

An important conclusion is that the spectrum of the periodic Neumann-Poincaré operator defined by (5.10) varies with the position and size of the particles. Our results hold for arbitrary-shaped nanoparticles. The resonances of the effective impedance α_∞ depend not only on the geometry of the particle B but also on its size and position. One can see (Figures 5.2 and 5.3) a change in the magnitude and a shift of the resonances. The plasmonic resonances shift to smaller wavelengths and the magnitude of the peak value increases with increasing volume. We remark that this is not particular to the examples considered here. In fact, this is the case for any particle. These two phenomena are due to the strong interaction between the particles and the ground that appears as their sizes increase while the period of the arrangement is fixed.

Note also that in our analysis we did not assume the particles to be simply connected. In fact, the theory is still valid for particles which have two or more components. This allows for more possibilities when choosing a particular geometry for the optimization of the effective impedance. For instance, one may want to design a geometry such that a single frequency is excited with a very pronounced peak or, on the other hand, to excite not only a specific frequency but rather a group of them.

5.6 Concluding remarks

In this chapter we have considered the scattering by an array of plasmonic nanoparticles mounted on a perfectly conducting plate and showed both analytically and numerically the significant change in the boundary condition induced by the nanoparticles at their periodic plasmonic frequencies. We have also proposed an optimization approach to maximize this change in terms of the shape of the nanoparticles. Our results in this chapter can be generalized in many directions. Different boundary conditions on the plate as well as curved plates can be considered. Our approach can be easily extended

to two-dimensional arrays embedded in $\mathbb{R}^3$ and the lattice effect can be included. Full Maxwell's equations to model the light propagation can be used. The observed extraordinary or meta properties of periodic distributions of subwavelength resonators can be explained by the approach proposed in this chapter.

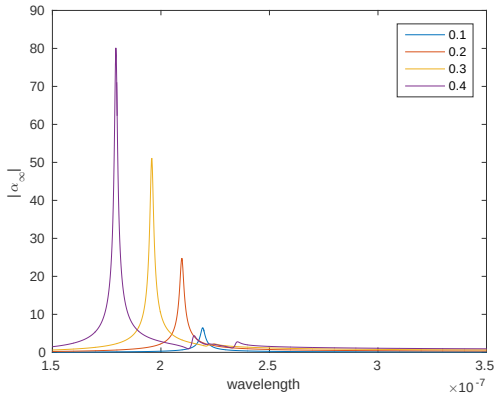

FIGURE 5.2: $|\alpha_\infty|$ as a function of the wavelength for disks of different radii, ranging from 0.1 to 0.4.

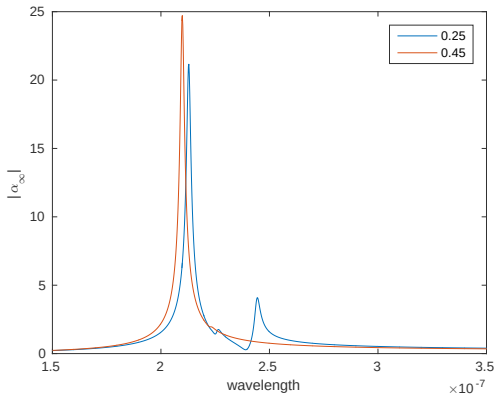

FIGURE 5.3: $|\alpha_\infty|$ as a function of the wavelength for a disk centered respectively at distance 0.25 and 0.45 from $x_2 = 0$.

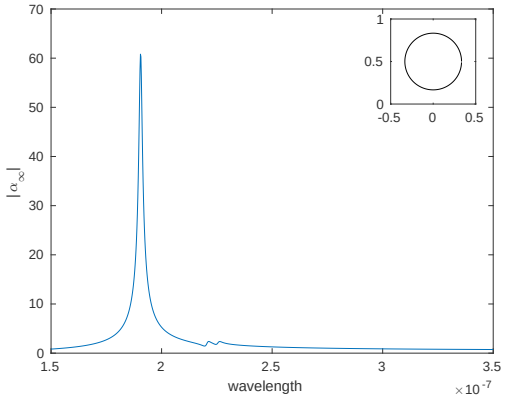

FIGURE 5.4: Well localized resonance for a disk.

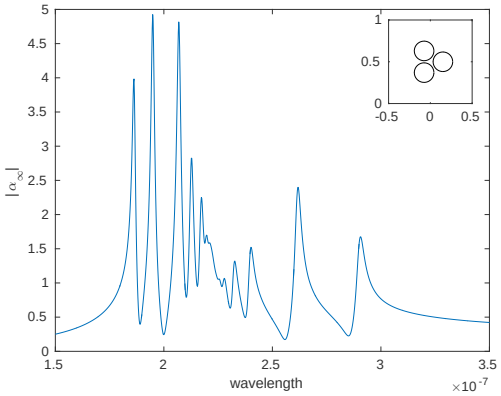

FIGURE 5.5: Delocalized resonances for three well-separated disks.

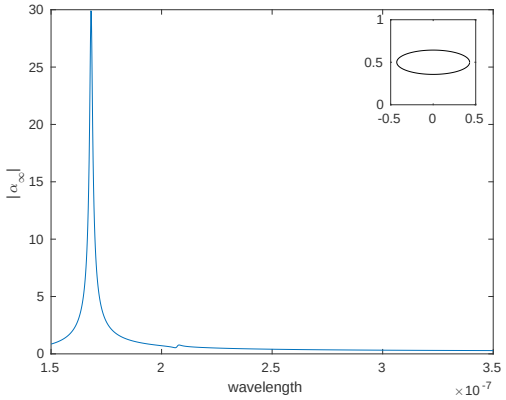

FIGURE 5.6: Well localized resonance for an ellipse.

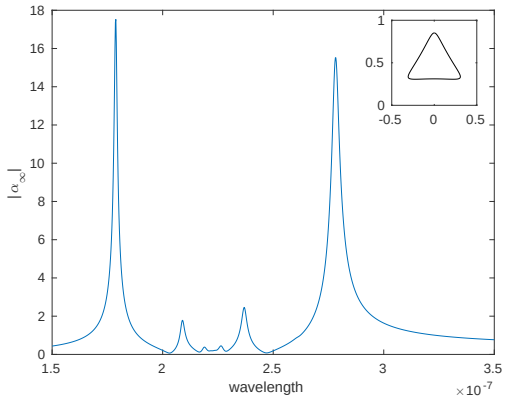

FIGURE 5.7: Delocalized resonances for a triangle with rounded corners.

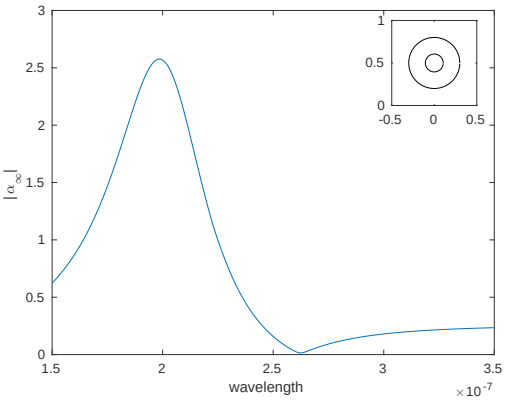

FIGURE 5.8: Wide resonance for a ring.

Chapter 6

Shape Recovery of Algebraic Domains

Contents

6.1 Introduction

In this chapter, we prove that based on plasmonic resonances we can on one hand classify the shape of a class of domains with real algebraic boundaries and on the other hand recover the separation distance between two components of multiple connected domains. These results have important applications in nanophotonics. They can be used in order to identify the shape and separation distance between plasmonic nanoparticles having known material parameters from measured plasmonic resonances, for which the scattering cross-section is maximized.

A real algebraic curve is the zero level set of a bivariate polynomial. Domains enclosed by real algebraic curves (henceforth simply called *algebraic domains*) are dense, in Hausdorff metric among all planar domains. On a simpler note, every smooth curve can be approximated by a sequence of algebraic curves. This observation turns algebraic curves into an efficient tool for describing shapes [64, 88, 94]. Note that an algebraic domain which is the sub level set of a polynomial of degree n can uniquely be determined from its set of two-dimensional moments of order less than or equal to $3n$ [55, 67]. In this chapter we consider a class of algebraic curves determined via conformal mappings by two parameters m and δ, with m being the order of the polynomial parametrizing the curve and δ being a shape parameter, see (6.1) and (6.2). One can think of algebraic domains as non-generic, but dense, among all planar domains, as much as polynomials are non-generic, but dense among all continuous functions on a compact set. In either case, the identifications/reconstructions have to be complemented by a fine analysis of the rate of convergence.

The main results of the present chapter are:

(i) Algebraic domains described by (6.2) have only two plasmonic resonances asymptotically (in δ). Based on these two plasmonic resonances, one can classify them;

(ii) Two nearly touching disks have an infinite number of plasmonic resonances and the separating distance can be determined from the measurement of the first plasmonic resonance.

The chapter is organized as follows. In section 6.2 we give explicit calculations of the Neumann-Poincaré operator associated with an algebraic domain. Moreover, we analyze its asymptotic behavior as δ approaches zero. We compute the first- and second-order contracted polarization tensors, and show how to use them to determine the two parameters describing the algebraic boundaries. In section 6.3 we consider two nearly touching disks. We use the bipolar coordinates to compute the spectrum of the associated Neumann-Poincaré operator. We show that all the eigenvalues of the associated Neumann-Poincaré operator contribute to the set of plasmonic resonances. From the first-order polarization tensor, we show that we can recover the separating distance between the disks. In section 6.4 we illustrate our main findings in this chapter with several numerical examples.

6.2 Plasmonic resonance for algebraic domains

6.2.1 Algebraic domains of class $\mathcal{Q}$

Let Ω be the unit disk in $\mathbb{C}$. For $m \in \mathbb{N}$ and $a \in \mathbb{R}$, define $\Phi_{m,a} : \mathbb{C} \setminus \overline{\Omega} \to \mathbb{C}$ by

$$\Phi_{m,a}(\zeta) = \zeta + \frac{a}{\zeta^m}.$$

Assume that $\Phi_{m,a}$ is injective on $\mathbb{C} \setminus \overline{\Omega}$. We introduce the class $\mathcal{Q}$ as the collection of all bounded domains $D \subset \mathbb{C}$ bounded by the curves

$$\partial D = \{\Phi_{m,a}(\zeta) : \quad |\zeta| = r_0\} \quad \text{for some } r_0 > 1, \quad m \in \mathbb{N} \text{ and } a \in \mathbb{R}.$$

Note that $\Phi_{m,a}$ is a conformal mapping from $\{|\zeta| > r_0\}$ onto $\mathbb{C} \setminus \overline{D}$. In what follows, we shall suppress the subscript m, a from $\Phi_{m,a}$ for the ease of notation.

Conformal images of the unit disc by rational functions are also called *quadrature domains*. We refer to [56, 84] for details and ramifications of the theory of quadrature domains. In particular, up to the inversion $z \mapsto 1/z$, the complements of the domains in class $\mathcal{Q}$ are quadrature domains. We write for convenience $\zeta = e^{\rho+i\theta}$. Let ρ_0 be such that $r_0 = e^{\rho_0}$. Let J be the Jacobian defined by

$$J = \left|\partial_\xi\big(\Phi(e^\xi)\big)|_{\xi=\rho+i\theta}\right|.$$

In the (ρ, θ) plane, the normal derivative $\partial/\partial\nu$ on ∂D is represented as

$$\frac{\partial}{\partial\nu} = \frac{1}{J}\frac{\partial}{\partial\rho}.$$

Moreover, the boundary ∂D is parametrized by

$$\theta \mapsto \Phi(e^{\rho_0+i\theta}) = e^{\rho_0+i\theta} + ae^{-m\rho_0-im\theta}.$$

If we fix the constant a and change ρ_0, then the size and the shape of ∂D will change accordingly. In order to leave the shape unchanged, we need to represent the constant a in a different way. We write

$$a = e^{(m+1)\rho_0}\delta. \tag{6.1}$$

Then the boundary ∂D can be represented as

$$\theta \mapsto \Phi(e^{\rho_0+i\theta}) = e^{\rho_0}(e^{i\theta} + \delta e^{-im\theta}). \tag{6.2}$$

Now, if we fix the constant δ and change ρ_0, then it is clear that only the size changes and the shape stays unaffected. The parameter e^{ρ_0} can be considered as a generalized radius of D because it determines the size. In conclusion, the shape of D is determined by the two parameters m and δ, while the size by the parameter ρ_0.

6.2.2 Explicit computation of the Neumann-Poincaré operator

In this section, we compute the Neumann-Poincaré operator on ∂D explicitly. We need to compute $\mathcal{K}_D^*[J^{-1}\cos n\theta]$ and $\mathcal{K}_D^*[J^{-1}\sin n\theta]$ explicitly. Our

strategy is as follows. Let $u = \mathcal{S}_D[J^{-1}\cos n\theta]$ and $v = \mathcal{S}_D[J^{-1}\sin n\theta]$. If u, v can be obtained explicitly, then $\mathcal{K}_D^*[J^{-1}\cos n\theta]$ and $\mathcal{K}_D^*[J^{-1}\sin n\theta]$ are immediately derived by using the following identity:

$$\mathcal{K}_D^*[\varphi] = \frac{1}{2}\left(\frac{\partial \mathcal{S}[\varphi]}{\partial \nu}\Big|_+ + \frac{\partial \mathcal{S}[\varphi]}{\partial \nu}\Big|_-\right), \tag{6.3}$$

which follows from (1.1). For simplicity, we consider only u. By using the continuity of the single layer potential and the jump relation (1.1), we can see that the function u is the solution to the following problem:

$$\begin{cases} \Delta u = 0 & \text{in } \mathbb{C}\setminus\partial D, \\ u|_- = u|_+ & \text{on } \partial D, \\ \dfrac{\partial u}{\partial \nu}\Big|_+ - \dfrac{\partial u}{\partial \nu}\Big|_- = J^{-1}\cos n\theta & \text{on } \partial D, \\ u = O(|z|^{-1}) & \text{as } |z|\to\infty. \end{cases} \tag{6.4}$$

Let $\widetilde{u}(\rho,\theta) = (u\circ\Phi)(e^{\rho+i\theta})$. Since $\Phi(\zeta)$ is conformal on $|\zeta| > e^{\rho_0}$, the above problem can be rewritten as follows:

$$\begin{cases} \Delta u = 0 & \text{for } \rho < \rho_0, \\ \Delta \widetilde{u} = 0 & \text{for } \rho > \rho_0, \\ \widetilde{u}|_- = \widetilde{u}|_+ & \text{on } \rho = \rho_0, \\ \dfrac{\partial \widetilde{u}}{\partial \rho}\Big|_+ - \dfrac{\partial \widetilde{u}}{\partial \rho}\Big|_- = \cos n\theta & \text{on } \rho = \rho_0, \\ \widetilde{u} = O(e^{-\rho}) & \text{as } \rho\to\infty. \end{cases} \tag{6.5}$$

Note that in (6.5), the first equation for $u|_D$ is not represented in terms of $\widetilde{u}$. This is due to the singularity of $\Phi(\zeta)$ near $\zeta = 0$. Hence, we need to consider $u|_D$ more carefully. If $a = 1$ and $m = 1$, then D becomes an ellipse and (ρ,θ) are called the elliptic coordinates. In this case, equation (6.5) for $\widetilde{u}$ can be easily solved by imposing some appropriate conditions on $\rho = \rho_0$ and $\rho = 0$. However, for general shaped domains, this is not easy.

Fortunately, we can overcome this difficulty by the fact that the shape of the domain D is defined by a rational function $\Phi(\zeta) = \zeta + a/\zeta^m$. Our strategy is to seek a solution to (6.5) such that

$$u(z) = \Re\{\text{a polynomial of degree } n \text{ in } z\} \quad \text{for } z\in D.$$

We can show that, for $1 \le n \le m$, $u|_D$ in equation (6.5) can be explicitly solved by using the following ansatz:

$$\begin{aligned} u|_D(z) \propto \Re\{z^n\} &= \Re\left\{\left(\zeta + \frac{a}{\zeta^m}\right)^n\right\} \\ &= \Re\sum_{k=0}^{n}\binom{n}{k}\zeta^{n-k}\left(\frac{a}{\zeta^m}\right)^k \qquad (\zeta = e^{\rho+i\theta}) \\ &= e^{n\rho}\cos n\theta + \sum_{k=1}^{n} a^k\binom{n}{k} e^{-t_k^{mn}\rho}\cos t_k^{mn}\theta, \end{aligned} \tag{6.6}$$

where the constant t_k^{mn} is defined by

$$t_k^{mn} = (m+1)k - n, \quad 0 \le k \le n.$$

As will be seen later, for the purpose of computing the polarization tensor, we consider only the case where $1 \le n \le m$. (If $n > m$, $u|_D(z)$ turns out to be more complicated polynomial than z^n but is still a polynomial of degree n.)

Let us assume $1 \le n \le m$. In view of (6.6), we define

$$w(\rho,\theta) := \begin{cases} e^{n\rho}\cos n\theta + \displaystyle\sum_{k=1}^{n} a^k \binom{n}{k} e^{-t_k^{mn}\rho}\cos t_k^{mn}\theta, & \rho < \rho_0, \\ e^{-n(\rho-2\rho_0)}\cos n\theta + \displaystyle\sum_{k=1}^{n} a^k \binom{n}{k} e^{-t_k^{mn}\rho}\cos t_k^{mn}\theta, & \rho > \rho_0. \end{cases}$$

Note that w is harmonic in $\{\rho < \rho_0\}$ and $\{\rho > \rho_0\}$ and $w = O(e^{-\rho})$ as $\rho \to \infty$. Moreover,

$$\begin{cases} w|_+ = w|_- & \text{on } \rho = \rho_0, \\ \dfrac{\partial w}{\partial \rho}\Big|_+ - \dfrac{\partial w}{\partial \rho}\Big|_- = (-2)ne^{n\rho_0}\cos n\theta & \text{on } \rho = \rho_0. \end{cases} \tag{6.7}$$

Therefore, the function w is equal to $\widetilde{u}$ up to a multiplicative constant. More precisely, we have

$$\widetilde{u}(\rho,\theta) = -\frac{1}{2n}e^{-n\rho_0}w(\rho,\theta). \tag{6.8}$$

Now we are ready to compute $\mathcal{K}_D^*[J^{-1}\cos n\theta]$. We can check that

$$\frac{1}{2}\left(\frac{\partial w}{\partial \rho}\Big|_{\rho=\rho_0}^{+} + \frac{\partial w}{\partial \rho}\Big|_{\rho=\rho_0}^{-}\right) = \sum_{k=1}^{n} -t_k^{mn}a^k \binom{n}{k} e^{-t_k^{mn}\rho_0}\cos t_k^{mn}\theta. \tag{6.9}$$

Then it follows from (6.3) and (6.8) that

$$\mathcal{K}_D^*[J^{-1}\cos n\theta] = \frac{1}{J}\sum_{k=1}^{n}\delta^k\frac{t_k^{mn}}{2n}\binom{n}{k}\cos t_k^{mn}\theta \tag{6.10}$$

for $1 \le n \le m$. In exactly the same manner, we can show that

$$\mathcal{K}_D^*[J^{-1}\sin n\theta] = -\frac{1}{J}\sum_{k=1}^{n}\delta^k\frac{t_k^{mn}}{2n}\binom{n}{k}\sin t_k^{mn}\theta. \tag{6.11}$$

It is worth mentioning that we can also compute the single layer potentials for $J^{-1}\cos n\theta$ and $J^{-1}\sin n\theta$:

$$\mathcal{S}_D[J^{-1}\cos n\theta] = -\frac{1}{2n}\cos n\theta - \frac{1}{2n}\sum_{k=1}^{n}\delta^k\binom{n}{k}\cos t_k^{mn}\theta, \tag{6.12}$$

and

$$\mathcal{S}_D[J^{-1}\sin n\theta] = -\frac{1}{2n}\sin n\theta + \frac{1}{2n}\sum_{k=1}^{n}\delta^k\binom{n}{k}\sin t_k^{mn}\theta. \tag{6.13}$$

6.2.3 Asymptotic behavior of the Neumann-Poincaré operator $\mathcal{K}_D^*$

If δ is small enough, then the shape of ∂D is close to a circle. Next we investigate the asymptotic behavior of the Neumann-Poincaré operator and its spectrum for small δ. From (6.10), we infer

$$\begin{aligned}\mathcal{K}_D^*[J^{-1}\cos n\theta] &= \delta\frac{(m+1-n)}{2}J^{-1}\cos(m+1-n)\theta + O(\delta^2),\\ \mathcal{K}_D^*[J^{-1}\sin n\theta] &= -\delta\frac{(m+1-n)}{2}J^{-1}\sin(m+1-n)\theta + O(\delta^2)\end{aligned} \tag{6.14}$$

for small δ and $1 \le n \le m$. One can verify the decay

$$\mathcal{K}_D^*[J^{-1}\cos n\theta],\ \mathcal{K}_D^*[J^{-1}\sin n\theta] = O(\delta^2) \tag{6.15}$$

for small δ and $n \ge m+1$.

Let us denote by

$$v_n^c = J^{-1}\cos n\theta, \quad v_n^s = J^{-1}\sin n\theta,$$

and let V_c and V_s be the subspaces defined by

$$V_c = \operatorname{span}\{v_1^c, v_2^c, \ldots, v_n^c, \ldots, v_m^c, \ldots\} \quad \text{and } V_s = \operatorname{span}\{v_1^s, v_2^s, \ldots, v_n^s, \ldots, v_m^s, \ldots\}.$$

In view of (6.14) and (6.15), we can easily see that the Neunamm-Poincaré operator $\mathcal{K}_D^*$ can be approximated by a finite rank operator for small δ. To state this fact, we define a finite rank operator $\mathcal{F}_m^c$ by

$$\begin{aligned}\mathcal{F}_m^c[v_n^c] &= \begin{cases}(m+1-n)v_{m+1-n}^c, & 1 \le n \le m,\\ 0, & n \ge m+1.\end{cases}\\ \mathcal{F}_m^c[v_n^s] &= 0, \quad n \ge 1.\end{aligned}$$

Similarly, we define $\mathcal{F}_m^s$ by

$$\begin{aligned}\mathcal{F}_m^s[v_n^s] &= \begin{cases}(m+1-n)v_{m+1-n}^s, & 1 \le n \le m,\\ 0, & n \ge m+1.\end{cases}\\ \mathcal{F}_m^s[v_n^c] &= 0, \quad n \ge 1.\end{aligned}$$

Then, on the subspace V_c, we have

$$\mathcal{K}_D^* = \frac{\delta}{2}\mathcal{F}_m^c + O(\delta^2).$$

Similarly, on the subspace V_s, we have

$$\mathcal{K}_D^* = -\frac{\delta}{2}\mathcal{F}_m^s + O(\delta^2). \tag{6.16}$$

Here, $O(\delta^2)$ is with respect to the operator norm.

Since the operators $\mathcal{F}_m^c$ and $\mathcal{F}_m^s$ are of finite rank, they have matrix representations. Using $\{v_n^c\}_{n=1}^m$ as basis, $\mathcal{F}_m^c$ can be represented as the following matrix $M_{D,m}$:

$$M_{D,m} := \begin{bmatrix} 0 & 0 & \dots & & 1 \\ 0 & \dots & & 2 & \\ \dots & & \dots & & \dots \\ & m-1 & & \dots & 0 \\ m & & \dots & 0 & 0 \end{bmatrix} \tag{6.17}$$

Clearly, $\mathcal{F}_m^s$ has the same matrix representation $M_{D,m}$ using $\{v_n^s\}_{n=1}^m$ as basis.

Let us now consider the eigenvalues and the associated eigenvectors of the matrix $M_{D,m}$. The following lemma can be easily proven.

Lemma 6.2.1. *(i) If m is odd, that is, $m = 2k-1$ for some $k \in \mathbb{N}$, then the matrix $M_{D,m}$ has the following eigenvalues:*

$$k, \pm\sqrt{1 \cdot m}, \pm\sqrt{2 \cdot (m-1)}, \dots, \pm\sqrt{(k-1) \cdot (k+1)},$$

and the associated eigenvectors are given by

$$\mathbf{e}_k, \quad \mathbf{e}_1 \pm \sqrt{m}\, \mathbf{e}_m, \quad \mathbf{e}_2 \pm \sqrt{m-1}\, \mathbf{e}_{m-1}, \quad \dots, \quad \sqrt{k-1}\, \mathbf{e}_{k-1} \pm \sqrt{k+1}\, \mathbf{e}_{k+1},$$

where $\mathbf{e}_i$ is the unit vector in the i-th direction.

(ii) If m is even, that is, $m = 2k$ for some $k \in \mathbb{N}$, then the matrix $M_{D,m}$ has the following eigenvalues:

$$\pm\sqrt{1 \cdot m}, \pm\sqrt{2 \cdot (m-1)}, \dots, \pm\sqrt{k \cdot (k+1)},$$

and the associated eigenvectors are given by

$$\mathbf{e}_1 \pm \sqrt{m}\, \mathbf{e}_m, \quad \sqrt{2}\, \mathbf{e}_2 \pm \sqrt{m-1}\, \mathbf{e}_{m-1}, \quad \dots, \quad \sqrt{k}\, \mathbf{e}_k \pm \sqrt{k+1}\, \mathbf{e}_{k+1}.$$

Using (6.16), Lemma 6.2.1 and the perturbation theory [62], we get the following asymptotic result for $\mathcal{K}_D^*$ on V_c.

Theorem 6.2.1. *For small δ, we have the following asymptotic expansions of eigenvalues and eigenfunctions of $\mathcal{K}_D^*$ on V_c:*

(i) If m is odd, that is, $m = 2k-1$ for some $k \in \mathbb{N}$:

Eigenvalues: up to order δ

$$\frac{\delta}{2} \quad \times \quad \left\{ k, \pm\sqrt{1 \cdot m}, \pm\sqrt{2 \cdot (m-1)}, \dots, \pm\sqrt{(k-1) \cdot (k+1)} \right\}.$$

Eigenfunctions: up to order δ^0

$$v_k^c, \quad v_1^c \pm \sqrt{m}\, v_m^c, \quad \sqrt{2}\, v_2^c \pm \sqrt{m-1}\, v_{m-1}^c, \quad \dots, \quad \sqrt{k-1}\, v_{k-1}^c \pm \sqrt{k+1}\, v_{k+1}^c.$$

(ii) If m is even, that is, $m = 2k$ for some $k \in \mathbb{N}$:

Eigenvalues: up to order δ

$$\frac{\delta}{2} \quad \times \quad \left\{ \pm\sqrt{1 \cdot m}, \pm\sqrt{2 \cdot (m-1)}, \dots, \pm\sqrt{k \cdot (k+1)} \right\}.$$

Eigenfunctions: up to order δ^0

$$v_1^c \pm \sqrt{m}\, v_m^c, \quad \sqrt{2}\, v_2^c \pm \sqrt{m-1}\, v_{m-1}^c, \quad \ldots, \quad \sqrt{k}\, v_k^c \pm \sqrt{k+1}\, v_{k+1}^c.$$

Similarly, we have the following result for $\mathcal{K}_D^*$ on the subspace V_s.

Theorem 6.2.2. *We have the following asymptotic expansion of eigenvalues and eigenfunctions of the Neumann-Poincaré operator* $\mathcal{K}_D^*$ *on the subspace* V_s *for small* δ*:*

(i) If m *is odd, that is,* $m = 2k-1$ *for some* $k \in \mathbb{N}$*:*

Eigenvalues: up to order δ

$$-\frac{\delta}{2} \quad \times \quad \left\{ k, \pm\sqrt{1\cdot m}, \pm\sqrt{2\cdot(m-1)}, \ldots, \pm\sqrt{(k-1)\cdot(k+1)} \right\}.$$

Eigenfunctions: up to order δ^0

$$v_k^s, \quad v_1^s \pm \sqrt{m} v_m^s, \quad \sqrt{2} v_2^s \pm \sqrt{m-1} v_{m-1}^s, \quad \ldots, \quad \sqrt{k-1}\, v_{k-1}^s \pm \sqrt{k+1}\, v_{k+1}^s.$$

(ii) If m *is even, that is,* $m = 2k$ *for some* $k \in \mathbb{N}$*:*

Eigenvalues: up to order δ

$$-\frac{\delta}{2} \quad \times \quad \left\{ \pm\sqrt{1\cdot m}, \pm\sqrt{2\cdot(m-1)}, \ldots, \pm\sqrt{k\cdot(k+1)} \right\}.$$

Eigenfunctions: up to order δ^0

$$v_1^s \pm \sqrt{m}\, v_m^s, \quad \sqrt{2}\, v_2^s \pm \sqrt{m-1}\, v_{m-1}^s, \quad \ldots, \quad \sqrt{k}\, v_k^s \pm \sqrt{k+1}\, v_{k+1}^s.$$

Corollary 6.2.1. *Suppose that* m *is odd, that is,* $m = 2k-1$ *for some* $k \in \mathbb{N}$*. In other words,* D *is a star-shaped domain with* $2k$ *petals. Then, up to order* δ*, the Neumann-Poincaré operator* $\mathcal{K}_D^*$ *has the following* $2k$ *eigenvalues:*

$$\frac{\delta}{2} \quad \times \quad \left\{ \pm\sqrt{1\cdot m}, \pm\sqrt{2\cdot(m-1)}, \ldots, \pm\sqrt{(k-1)\cdot(k+1)}, \pm\sqrt{k\cdot k} \right\}.$$

6.2.4 Generalized polarization tensors and their spectral representations

First-order polarization tensor

Let us compute the first-order polarization tensor associated with D and λ. Recall the definition of λ,

$$\lambda := \frac{\varepsilon_m + \varepsilon_c}{2(\varepsilon_m - \varepsilon_c)},$$

for a domain D with permittivity ε_c and backgroud with permittivity ε_m.

See chapter 1 for a brief introduction on generalized polarization tensors.

For simplicity, we consider only the case when m is odd, that is, $m = 2k-1$ for some $k \in \mathbb{N}$. The case where m is even can be treated analogously. Numerical results are presented in section 6.4 for both cases.

Since m is odd, the shape of D has even symmetry with respect to both x_1-axis and x_2-axis. Thanks to this symmetry, $M(\lambda, D)$ has the following simple form [18]:

$$M(\lambda, D) = m_{11} \begin{bmatrix} 1 & 0 \\ 0 & 1 \end{bmatrix},$$

where m_{11} is given by

$$m_{11} = \left(x_1, (\lambda I - \mathcal{K}_D^*)^{-1}[\nu_1]\right)_{\frac{1}{2},-\frac{1}{2}}.$$

Let λ_j and φ_j, $j \in \mathbb{N}$, be the eigenvalues and the (normalized) eigenfunctions of $\mathcal{K}_D^*$, respectively. Then, from the spectral decomposition of $\mathcal{K}_D^*$, we have (see chapter 1)

$$\begin{aligned} m_{11} &= \sum_j \frac{1}{\lambda - \lambda_j} \frac{(x_1, \varphi_j)_{\frac{1}{2},-\frac{1}{2}} (-\mathcal{S}_D[\nu_1], \varphi_j)_{\frac{1}{2},-\frac{1}{2}}}{(-\mathcal{S}_D[\varphi_j], \varphi_j)_{\frac{1}{2},-\frac{1}{2}}} \\ &= \sum_j \frac{(\frac{1}{2} - \lambda_j)}{\lambda - \lambda_j} \frac{|(x_1, \varphi_j)_{\frac{1}{2},-\frac{1}{2}}|^2}{(-\mathcal{S}_D[\varphi_j], \varphi_j)_{\frac{1}{2},-\frac{1}{2}}}. \end{aligned}$$

By Theorems 6.2.1 and 6.2.2, one can see that only the following two eigenvalues and two eigenfunctions contribute to m_{11} up to order δ:

$$\begin{aligned} &\text{eigenvalues } \lambda_\pm := \pm\frac{1}{2}\delta\sqrt{m}, \\ &\text{eigenfunctions } \varphi_\pm := v_1^c \pm \sqrt{m}\, v_m^c. \end{aligned}$$

In fact, for other eigenfunctions, we have $(x_1, \varphi_j)_{\frac{1}{2},-\frac{1}{2}} = O(\delta)$. In what follows we calculate $(x_1, \varphi_\pm)_{\frac{1}{2},-\frac{1}{2}}$ and $(x_1, \mathcal{S}_D[\varphi_\pm])_{\frac{1}{2},-\frac{1}{2}}$.

First, since $d\sigma = J d\theta$ and

$$x_1|_{\partial D} = \Re\{\Phi(e^{\rho_0 + i\theta})\} = e^{\rho_0}\cos\theta + a e^{-m\rho_0}\cos m\theta,$$

we have

$$\begin{aligned} (x_1, \varphi_\pm)_{\frac{1}{2},-\frac{1}{2}} &= \int_0^{2\pi} (e^{\rho_0}\cos\theta + a e^{-m\rho_0}\cos m\theta)(\cos\theta \pm \sqrt{m}\cos m\theta)\, d\theta, \\ &= \pi e^{\rho_0}(1 + 2\lambda_\pm). \end{aligned}$$

Now, we compute $(\mathcal{S}_D[\varphi_\pm], \varphi_\pm)_{\frac{1}{2},-\frac{1}{2}}$. Note that, from (6.12), we have

$$\mathcal{S}_D[v_n^c] = -\frac{1}{2n}\cos n\theta - \frac{1}{2}\delta\cos(m + 1 - n)\theta + O(\delta^2).$$

Consequently,

$$\begin{aligned} (-\mathcal{S}_D[\varphi_\pm], \varphi_\pm)_{\frac{1}{2},-\frac{1}{2}} &= (-\mathcal{S}_D[v_1^c \pm \sqrt{m}\, v_m^c], v_1^c \pm \sqrt{m}\, v_m^c)_{\frac{1}{2},-\frac{1}{2}} \\ &= \int_0^{2\pi} (\cos\theta \pm \sqrt{m}\,\cos m\theta) \\ &\qquad \times (\frac{1}{2}\cos\theta + \frac{\delta}{2}\cos m\theta \pm \frac{1}{2\sqrt{m}}\cos m\theta \pm \sqrt{m}\,\frac{\delta}{2}\cos\theta)\, d\theta + O(\delta^2) \\ &= \pi(1 + 2\lambda_\pm) + O(\delta^2). \end{aligned}$$

Finally, we are ready to obtain an approximation formula for m_{11}.

Theorem 6.2.3. *We have*

$$m_{11} = \frac{\pi}{2} e^{2\rho_0} \Big(\frac{1}{\lambda - \lambda_+} + \frac{1}{\lambda - \lambda_-} \Big) + O(\delta^2), \tag{6.18}$$

as $\delta \to 0$.

Second-order contracted generalized polarization tensors

Let $M^{cc}_{mn}, M^{ss}_{mn}, M^{sc}_{mn}$, and M^{cs}_{mn} be the contracted generalized polarization tensors. One can easily see that $M^{sc}_{22} = M^{cs}_{22} = 0$ and $M_{12} = M_{21} = 0$. We only need to consider M^{cc}_{22} and M^{ss}_{22}. It turns out that only the following two eigenvalues and two eigenfunctions contribute to M^{cc}_{22}(up to the order δ):

$$\begin{aligned} &\text{eigenvalues } \lambda'_\pm := \pm\frac{1}{2}\delta\sqrt{2\cdot(m-1)}, \\ &\text{eigenfunctions } \varphi'_\pm := \sqrt{2}\, v^c_2 \pm \sqrt{m-1}\, v^c_{m-1}. \end{aligned}$$

Let $H := \Re\big\{(x_1 + ix_2)^2\big\}$. Then we have

$$H|_{\partial D} = e^{2\rho_0}(\cos 2\theta + 2\delta \cos m\theta + \delta^2 \cos 2m\theta).$$

Therefore,

$$\begin{aligned} (H, \varphi'_\pm)_{\frac{1}{2},-\frac{1}{2}} &= \int_0^{2\pi} e^{2\rho_0}(\cos 2\theta + 2\delta\cos m\theta + \delta^2 \cos 2m\theta)(\sqrt{2}\,\cos 2\theta \pm \sqrt{m-1}\cos(m-1)\theta)\, d\theta \\ &= \sqrt{2}\pi e^{2\rho_0}. \end{aligned}$$

Now we compute $(-\mathcal{S}_D[\varphi'_\pm], \varphi'_\pm)_{\frac{1}{2},-\frac{1}{2}}$. Since

$$\mathcal{S}_D[v^c_n] = -\frac{1}{2n}\cos n\theta - \frac{1}{2}\delta\cos(m+1-n)\theta + O(\delta^2),$$

we obtain

$$\begin{aligned} (-\mathcal{S}_D[\varphi'_\pm], \varphi'_\pm)_{\frac{1}{2},-\frac{1}{2}} &= (-\mathcal{S}_D[\sqrt{2}\, v^c_2 \pm \sqrt{m-1}\, v^c_{m-1}], \sqrt{2}\, v^c_2 \pm \sqrt{m-1}\, v^c_{m-1})_{\frac{1}{2},-\frac{1}{2}} \\ &= \int_0^{2\pi} (\sqrt{2}\,\cos 2\theta \pm \sqrt{m-1}\,\cos(m-1)\theta) \\ &\quad \times (\frac{1}{2\sqrt{2}}\cos 2\theta + \frac{\delta}{\sqrt{2}}\cos(m-1)\theta \\ &\quad \pm \frac{1}{2\sqrt{m-1}}\cos(m-1)\theta \pm \sqrt{m-1}\,\frac{\delta}{2}\cos 2\theta)\, d\theta + O(\delta^2) \\ &= \pi(1 + 2\lambda'_\pm) + O(\delta^2). \end{aligned}$$

Finally, we find

$$\begin{aligned} M^{cc}_{22} &= \sum_j \frac{(\frac{1}{2} - \lambda_j)}{(\lambda - \lambda_j)} \frac{|(H, \varphi_j)_{\frac{1}{2},-\frac{1}{2}}|^2}{(-\mathcal{S}_D[\varphi_j], \varphi_j)_{\frac{1}{2},-\frac{1}{2}}}, \\ &= \pi e^{4\rho_0} \Big(\frac{(\frac{1}{2} - \lambda'_+)}{(\frac{1}{2} + \lambda'_+)(\lambda - \lambda'_+)} + \frac{(\frac{1}{2} + \lambda'_-)}{(\frac{1}{2} - \lambda'_-)(\lambda - \lambda'_-)} \Big) + O(\delta^2). \end{aligned} \tag{6.19}$$

Similarly, one can show that M_{22}^{ss} has similar asymptotic expansion.

6.2.5 Classification of algebraic domains in the class $\mathcal{Q}$

The identification of the parameters ρ_0 and m is now straightforward using the results of the previous subsection. Suppose that we can obtain the values of $\lambda_\pm, \lambda'_\pm$ approximately from m_{11} and M_{22}^{cc}. Then, by formula (6.18), we can easily find the parameter ρ_0, which determines the size of D. In order to reconstruct the parameters m and δ we turn to the definitions of λ_+ and λ'_+:

$$\lambda_+ = \frac{1}{2}\delta\sqrt{m}, \quad \lambda'_+ = \frac{1}{2}\delta\sqrt{2\cdot(m-1)}.$$

It is worth emphasizing that the eigenvalues λ_+ and λ'_+ are first-order approximations of the exact eigenvalues of Neumann-Poincaré operator $\mathcal{K}_D^*$ for small δ.

Solving the above equations for m and δ yields the exact formulas:

$$m = \frac{\lambda_+^2}{\lambda_+^2 - (\lambda'_+)^2/2}, \quad \delta = 2\sqrt{\lambda_+^2 - (\lambda'_+)^2/2}.$$

6.3 Plasmonic resonances for two separated disks

In this section, we consider the spectrum of the Neumann-Poincaré operator when two conductors are located closely to each other in $\mathbb{R}^2$. As an application of the spectral decomposition of the Neumann-Poincaré operator, we derive the $(1,1)$-entry, m_{11}, of the first-order polarization tensor associated with the two disks. See chapter 1 for a brief introduction on polarization tensors.

6.3.1 The bipolar coordinates and the boundary integral operators

Let B_1 and B_2 be two disks with conductivity σ embedded in the background with conductivity 1. The conductivity is such that $0 < \sigma \neq 1 < \infty$. Let $\sigma_{B_1\cup B_2}$ denote the conductivity distribution, *i.e.*,

$$\sigma_{B_1\cup B_2} = \sigma\chi(B_1) + \sigma\chi(B_2) + \chi(\mathbb{R}^2 \setminus (B_1 \cup B_2)), \tag{6.20}$$

where χ is the characteristic function. Let ϵ be the distance between two disks, that is,

$$\epsilon := \operatorname{dist}(B_1, B_2).$$

We set Cartesian coordinates (x_1, x_2) such that x_1-axis is parallel to the line joining the centers of the two disks.

(Definition) Each point $\mathbf{x} = (x_1, x_2)$ in the Cartesian coordinate system corresponds to $(\xi, \theta) \in \mathbb{R}\times(-\pi, \pi]$ in the bipolar coordinate system through the equations

$$x_1 = \alpha\frac{\sinh\xi}{\cosh\xi - \cos\theta} \quad \text{and} \quad x_2 = \alpha\frac{\sin\theta}{\cosh\xi - \cos\theta} \tag{6.21}$$

with a positive number α. In fact, the bipolar coordinates can be defined using a conformal mapping. Define a conformal map Ψ by

$$z = x_1 + ix_2 = \Psi(\zeta) = \alpha\frac{\zeta+1}{\zeta-1}.$$

If we write $\zeta = e^{\xi - i\theta}$, then we can recover (6.21).

(The coordinate curve) From the definition, we can derive that the coordinate curves $\{\xi = c\}$ and $\{\theta = c\}$ are, respectively, the zero-level set of the following two functions:

$$f_\xi(x,y) = \left(x - \alpha\frac{\cosh c}{\sinh c}\right)^2 + y^2 - \left(\frac{\alpha}{\sinh c}\right)^2 \tag{6.22}$$

and

$$f_\theta(x,y) = x^2 + \left(y - \alpha\frac{\cos c}{\sin c}\right)^2 - \left(\frac{\alpha}{\sin c}\right)^2.$$

(Basis vectors) Orthonormal basis vectors $\{\hat{\mathbf{e}}_\xi, \hat{\mathbf{e}}_\theta\}$ are defined as follows:

$$\hat{\mathbf{e}}_\xi := \frac{\partial \mathbf{x}/\partial\xi}{|\partial \mathbf{x}/\partial\xi|} \quad \text{and} \quad \hat{\mathbf{e}}_\theta := \frac{\partial \mathbf{x}/\partial\theta}{|\partial \mathbf{x}/\partial\theta|}.$$

(Normal- and tangential derivatives and line element) In the bipolar coordinates, the scaling factor h is

$$h(\xi,\theta) := \frac{\cosh\xi - \cos\theta}{\alpha}.$$

The gradient of any scalar function g is

$$\nabla g = h(\xi,\theta)\left(\frac{\partial g}{\partial\xi}\hat{\mathbf{e}}_\xi + \frac{\partial g}{\partial\theta}\hat{\mathbf{e}}_\theta\right). \tag{6.23}$$

Moreover, the normal and tangential derivatives of a function u in bipolar coordinates are

$$\begin{cases} \dfrac{\partial u}{\partial\nu}\Big|_{\xi=c} = \nabla u \cdot v_{\xi=c} = -\mathrm{sgn}(c)h(c,\theta)\dfrac{\partial u}{\partial\xi}\Big|_{\xi=c}, \\ \dfrac{\partial u}{\partial T}\Big|_{\xi=c} = -\mathrm{sgn}(c)h(c,\theta)\dfrac{\partial u}{\partial\theta}\Big|_{\xi=c}, \end{cases} \tag{6.24}$$

and the line element $d\sigma$ on the boundary $\{\xi = \xi_0\}$ is

$$d\sigma = \frac{1}{h(\xi_0,\theta)}d\theta.$$

(Separation of variables) The bipolar coordinate system admits separation of variables for any harmonic function f as follows:

$$f(\xi,\theta) = a_0 + b_0\xi + c_0\theta + \sum_{n=1}^{\infty}\left[(a_n e^{n\xi} + b_n e^{-n\xi})\cos n\theta + (c_n e^{n\xi} + d_n e^{-n\xi})\sin n\theta\right], \tag{6.25}$$

where a_n, b_n, c_n and d_n are constants.

For $\xi > 0$, we have

$$\frac{\sinh\xi - i\sin\theta}{\cosh\xi - \cos\theta} = \frac{e^{\zeta} + e^{-\zeta}}{e^{\zeta} - e^{-\zeta}} = 1 + 2\sum_{n=1}^{\infty} e^{-n\xi}(\cos n\theta - i\sin n\theta), \tag{6.26}$$

with $\zeta = (\xi + i\theta)/2$.

Using (6.21), we have the following harmonic expansions for the two linear functions x_1 and x_2:

$$x_1 = \mathrm{sgn}(\xi)\alpha\left[1 + 2\sum_{n=1}^{\infty} e^{-n|\xi|}\cos n\theta\right], \tag{6.27}$$

and

$$x_2 = 2\alpha\sum_{n=1}^{\infty} e^{-n|\xi|}\sin n\theta.$$

Let $\mathbb{K}^*$ be the Neumann-Poincaré operator given by

$$\mathbb{K}^* := \begin{bmatrix} \mathcal{K}^*_{B_1} & \dfrac{\partial}{\partial\nu^{(1)}}\mathcal{S}_{B_2} \\ \dfrac{\partial}{\partial\nu^{(2)}}\mathcal{S}_{B_1} & \mathcal{K}^*_{B_2} \end{bmatrix},$$

and define the operator $\mathbb{S}$ by

$$\mathbb{S} = \begin{bmatrix} \mathcal{S}_{B_1} & \mathcal{S}_{B_2} \\ \mathcal{S}_{B_1} & \mathcal{S}_{B_2} \end{bmatrix}.$$

Here, $\nu^{(i)}$ is the outward normal on ∂B_i, $i = 1, 2$.

Then, from [7], $\mathbb{K}^*$ is self-adjoint with the inner product

$$(\varphi,\psi)_{\mathcal{H}^*} := -(\mathbb{S}[\psi],\varphi)_{H^{1/2}(\partial B_1)\times H^{1/2}(\partial B_2),H^{-1/2}(\partial B_1)\times H^{-1/2}(\partial B_2)},$$

for $\varphi, \psi \in H_0^{-1/2}(\partial B_1) \times H_0^{-1/2}(\partial B_2)$.

6.3.2 Neumann Poincaré-operator for two separated disks and its spectral decomposition

First we introduce some notations. Set

$$\alpha = \sqrt{\epsilon(r + \frac{\epsilon}{4})} \quad \text{and} \quad \xi_0 = \sinh^{-1}\left(\frac{\alpha}{r}\right), \quad \text{for } j = 1, 2, \tag{6.28}$$

where r is the radius of the two disks and ϵ their separation distance. Note that

$$\partial B_j = \{\xi = (-1)^j\xi_0\}, \quad \text{for } j = 1, 2. \tag{6.29}$$

Let us denote the Neumann-Poincaré operator for two disks separated by a distance ϵ by $\mathbb{K}_\epsilon^*$. To find out the spectral decomposition of the Neumann-Poincaré operator $\mathbb{K}_\epsilon^*$, we use the following lemma [7].

Lemma 6.3.1. *Assume that there exists u a nontrivial solution to the following equation:*

$$\begin{cases} \Delta u = 0 & \text{in } B_1 \cup B_2 \cup \mathbb{R}^2 \setminus \overline{(B_1 \cup B_2)}, \\ u|_+ = u|_- & \text{on } \partial B_j, j = 1, 2, \\ \dfrac{\partial u}{\partial \nu}\Big|_+ = \sigma_0 \dfrac{\partial u}{\partial \nu}\Big|_- & \text{on } \partial B_j, j = 1, 2, \\ u(\mathbf{x}) \to 0 & \text{as } |\mathbf{x}| \to \infty, \end{cases} \tag{6.30}$$

where $\sigma_0 = -\dfrac{1+2\lambda_0}{1-2\lambda_0} < 0$. If we set

$$\psi_j := \frac{\partial u}{\partial \nu}\Big|_{\partial B_j}^{+} - \frac{\partial u}{\partial \nu}\Big|_{\partial B_j}^{-}, \quad \text{for } j = 1, 2,$$

then $\psi = \begin{bmatrix} \psi_1 \\ \psi_2 \end{bmatrix}$ is an eigenvector of $\mathbb{K}_\epsilon^*$ corresponding to the eigenvalue λ_0.

One can see that the following function u_n is a solution to (6.30):

$$u_n^{\pm}(\xi, \theta) = (\text{const.}) + \begin{cases} \mp \dfrac{1}{2|n|}(e^{|n|\xi_0} \mp e^{-|n|\xi_0}) e^{|n|\xi + in\theta}, & \text{for } \xi < -\xi_0, \\ \dfrac{1}{2|n|} e^{-|n|\xi_0}(e^{|n|\xi} \mp e^{-|n|\xi}) e^{in\theta}, & \text{for } -\xi_0 < \xi < \xi_0, \\ \dfrac{1}{2|n|}(e^{|n|\xi_0} \mp e^{-|n|\xi_0}) e^{-|n|\xi + in\theta}, & \text{for } \xi > \xi_0. \end{cases} \tag{6.31}$$

From (6.31) and Lemma 6.3.1, we obtain eigenvalues and eigenvectors to $\mathbb{K}_\epsilon^*$

$$\lambda_{\epsilon,n}^{\pm} = \pm \frac{1}{2} e^{-2|n|\xi_0} \quad \text{and} \quad \Phi_{\epsilon,n}^{\pm}(\theta) = e^{in\theta} \begin{bmatrix} h(-\xi_0, \theta) \\ \mp h(\xi_0, \theta) \end{bmatrix}.$$

Note that the above eigenvectors are not normalized.

We compute $(-\mathbb{S}[\Phi_{\epsilon,n}^{\pm}], \Phi_{\epsilon,n}^{\pm})_{\frac{1}{2},-\frac{1}{2}}$. From (6.31), one can see that

$$\mathbb{S}[\Phi_{\epsilon,n}^{\pm}] = (\text{const.}) + \begin{bmatrix} \mp \frac{1}{2|n|}(1 \mp e^{-2|n|\xi_0}) e^{in\theta} \\ \frac{1}{2|n|}(1 \mp e^{-2|n|\xi_0}) e^{in\theta} \end{bmatrix}.$$

It follows that

$$(-\mathbb{S}[\Phi_{\epsilon,n}^{\pm}], \Phi_{\epsilon,n}^{\pm})_{\frac{1}{2},-\frac{1}{2}} = \frac{2\pi}{|n|}(1 \mp e^{-2|n|\xi_0}).$$

Therefore, we arrive at the following result.

Theorem 6.3.1. *We have the following spectral decomposition of $\mathbb{K}_\epsilon^*$:*

$$\mathbb{K}_\epsilon^* = \sum_{n \neq 0} \frac{1}{2} e^{-2|n|\xi_0} \Psi_{\epsilon,n}^{+} \otimes \Psi_{\epsilon,n}^{+} + \sum_{n \neq 0} \left(-\frac{1}{2} e^{-2|n|\xi_0} \right) \Psi_{\epsilon,n}^{-} \otimes \Psi_{\epsilon,n}^{-},$$

where $\Psi^{\pm}_{\epsilon,n}$ *are the normalized eigenvectors defined by*

$$\Psi^{\pm}_{\epsilon,n}(\theta) := \frac{\sqrt{|n|}e^{in\theta}}{\sqrt{2\pi(1 \mp e^{-2|n|\xi_0})}} \begin{bmatrix} h(-\xi_0,\theta) \\ \mp h(\xi_0,\theta) \end{bmatrix}. \tag{6.32}$$

Note that

$$(\mathcal{S}_{B_1}[\Psi^{\pm}_{\epsilon,n,1}] + \mathcal{S}_{B_2}[\Psi^{\pm}_{\epsilon,n,2}])(\xi,\theta) = (\text{const.}) + \frac{\sqrt{|n|}}{\sqrt{2\pi(1 \mp e^{-2|n|\xi_0})}} \tag{6.33}$$

$$\times \begin{cases} \mp\dfrac{1}{2|n|}(e^{|n|\xi_0} \mp e^{-|n|\xi_0})e^{|n|\xi+in\theta}, & \text{for } \xi < -\xi_0, \\ \dfrac{1}{2|n|}e^{-|n|\xi_0}(e^{|n|\xi} \mp e^{-|n|\xi})e^{in\theta}, & \text{for } -\xi_0 < \xi < \xi_0, \\ \dfrac{1}{2|n|}(e^{|n|\xi_0} \mp e^{-|n|\xi_0})e^{-|n|\xi+in\theta}, & \text{for } \xi > \xi_0. \end{cases} \tag{6.34}$$

6.3.3 The Polarization tensor

Let us compute the $(1,1)$-entry m^{ϵ}_{11} of the first-order polarization tensor for two separated disks. Note that

$$m^{\epsilon}_{11} = \left(\varphi, (\lambda\mathbb{I} - \mathbb{K}^*_{\epsilon})^{-1}[\psi]\right)_{\frac{1}{2},-\frac{1}{2}},$$

where

$$\phi = \begin{bmatrix} x_1|_{\partial B_1} \\ x_1|_{\partial B_2} \end{bmatrix}, \quad \psi = \begin{bmatrix} \nu_1|_{\partial B_1} \\ \nu_1|_{\partial B_2} \end{bmatrix}.$$

The spectral decomposition of $\mathbb{K}^*_{\epsilon}$ implies

$$\begin{aligned} m^{\epsilon}_{11} &= \sum_{n\neq 0} \frac{\langle \phi, \Psi^{+}_{\epsilon,n}\rangle_{\frac{1}{2},-\frac{1}{2}} (\Psi^{+}_{\epsilon,n}, \psi)_{\mathcal{H}^*}}{\lambda - \lambda^{+}_{\epsilon,n}} + \sum_{n\neq 0} \frac{\langle \phi, \Psi^{-}_{\epsilon,n}\rangle_{\frac{1}{2},-\frac{1}{2}} (\Psi^{-}_{\epsilon,n}, \psi)_{\mathcal{H}^*}}{\lambda - \lambda^{-}_{\epsilon,n}} \\ &= \sum_{n\neq 0} \frac{\left(\frac{1}{2} - \lambda^{+}_{\epsilon,n}\right) |\langle \phi, \Psi^{+}_{\epsilon,n}\rangle_{\frac{1}{2},-\frac{1}{2}}|^2}{\lambda - \lambda^{+}_{\epsilon,n}} + \sum_{n\neq 0} \frac{\left(\frac{1}{2} - \lambda^{-}_{\epsilon,n}\right) |\langle \phi, \Psi^{-}_{\epsilon,n}\rangle_{\frac{1}{2},-\frac{1}{2}}|^2}{\lambda - \lambda^{-}_{\epsilon,n}}. \end{aligned}$$

From (6.27), we derive the expansion

$$x_1 = \operatorname{sgn}(\xi)\alpha \sum_{m=-\infty}^{\infty} e^{-|m||\xi|+im\theta}. \tag{6.35}$$

Therefore,

$$\begin{aligned} \langle \phi, \Psi^{+}_{\epsilon,n}\rangle_{\frac{1}{2},-\frac{1}{2}} &= 2\int_0^{2\pi} \left[-\alpha \sum_{m=-\infty}^{\infty} e^{-|m|\xi_0+im\theta}\right] \frac{\sqrt{|n|}h(-\xi_0,\theta)e^{-in\theta}}{\sqrt{2\pi(1-e^{-2|n|\xi_0})}} \frac{1}{h(-\xi_0,\theta)} d\theta \\ &= -2\sqrt{2\pi}\alpha \frac{\sqrt{|n|}e^{-|n|\xi_0}}{\sqrt{1-e^{-2|n|\xi_0}}}, \end{aligned}$$

and

$$\langle \phi, \Psi^{-}_{\epsilon,n}\rangle_{\frac{1}{2},-\frac{1}{2}} = 0.$$

As a consequence, we arrive at the following result.

Propsition 6.3.1. *We have*

$$m_{11}^{\epsilon} = \sum_{n\neq 0} \frac{4\pi\alpha^2 |n| e^{-2|n|\xi_0}}{\lambda - \lambda_{\epsilon,n}^{+}} = 8\pi\alpha^2 \sum_{n=1}^{\infty} \frac{n e^{-2n\xi_0}}{\lambda - \frac{1}{2}e^{-2n\xi_0}},$$

where α is given by (6.28).

6.3.4 Reconstruction of the separation distance

Suppose that the first eigenvalue

$$\lambda_{\epsilon,1}^{+} = \frac{1}{2}e^{-2\xi_0}$$

is measured. Then we immediately find the value of e^{ξ_0}. From (6.28), we have

$$r\cosh\xi_0 = \frac{\epsilon}{2} + r.$$

By solving the above quadratic equation, we can determine the distance ϵ between the two disks.

6.4 Numerical illustrations

In this section we illustrate our main findings in this chapter with several numerical examples.

We use the material parameters of gold nanoparticles and suppose that we can measure their first- and second-order polarization tensors for a range of wavelengths in the visible regime.

Figure 6.1 shows the variations of the real and imaginary parts of λ, defined by (2.23), as function of the wavelength using Drude's model for $\sigma = \sigma(\omega)$, which is depending on the operating frequency ω [9].

As shown in Figure 6.1, the imaginary part of λ is very small. Therefore, when the real part of λ hits an eigenvalue that contributes to the first-order polarization tensor (and therefore to the plasmonic resonances), we should see a peak in the graph of $|m_{11}|$ and $|M_{22}^{cc}|$ with respect to the wavelength. This allow us, in the case of class $\mathcal{Q}$ of algebraic domains, to recover λ_+ and λ'_+ and, in the case of two separated disks, to recover $\lambda_{\epsilon,1}^{+}$.

Figures 6.2, 6.3, and 6.4 present examples of algebraic domains and their reconstructions, where a circle of radius one has been transformed for different values of m and δ.

Figures 6.5 and 6.6 present examples of algebraic domains and their reconstructions, where a circle of radius one has been transformed for $m = 4$ and $m = 6$ and $\delta = 0.02$.

Figures 6.7, 6.8, and 6.9 show examples of two circles of radius one separated by a distance ϵ, and their reconstructions.

6.5 Concluding remarks

In this chapter we have proved for a class of algebraic domains that the associated plasmonic resonances can be used to classify them. It would be

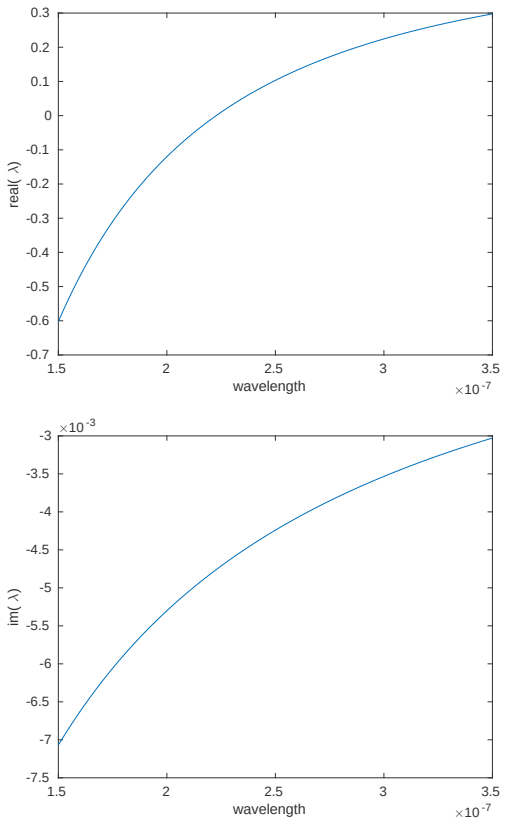

FIGURE 6.1: Real and imaginary parts of λ as function of the wavelength.

very interesting to prove a similar result for all quadrature domains or all algebraic domains. We have also reconstructed the separation distance between two nanoparticles of circular shape from measurements of their first collective plasmonic resonances. Another challenging problem would be to generalize this result to more components and arbitrary shaped particles.

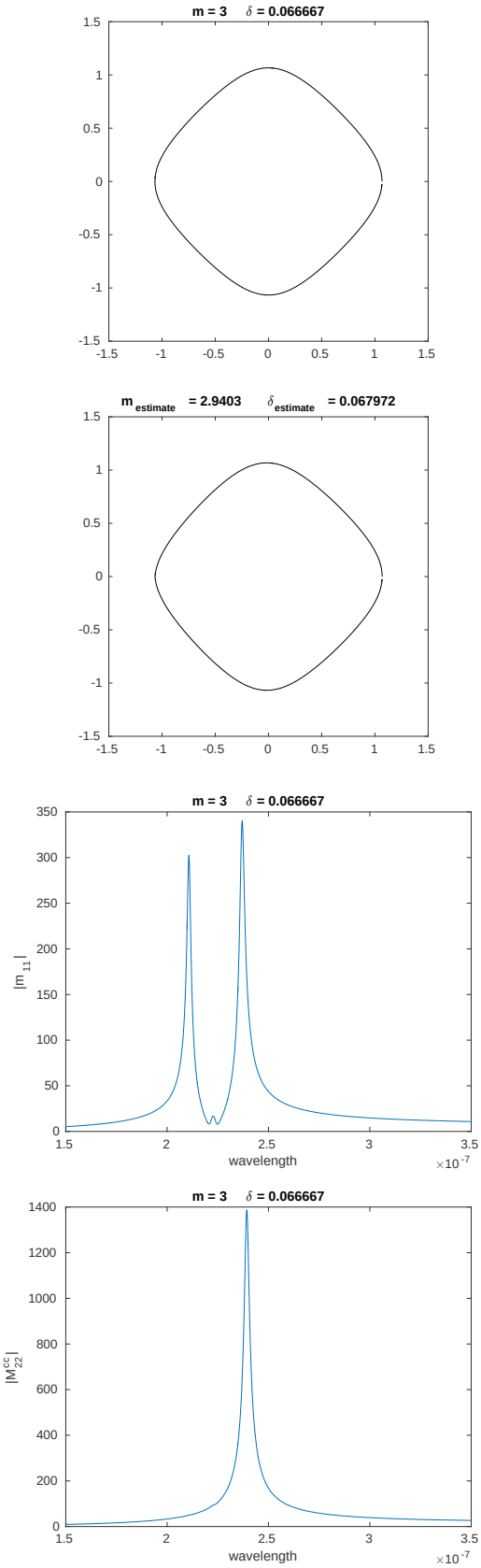

FIGURE 6.2: From top to bottom and left to right: initial shape, reconstructed shape, $|m_{11}|$ and $|M_{22}^{cc}|$ with respect to the wavelength for $m = 3$ and $\delta = 0.066667$.

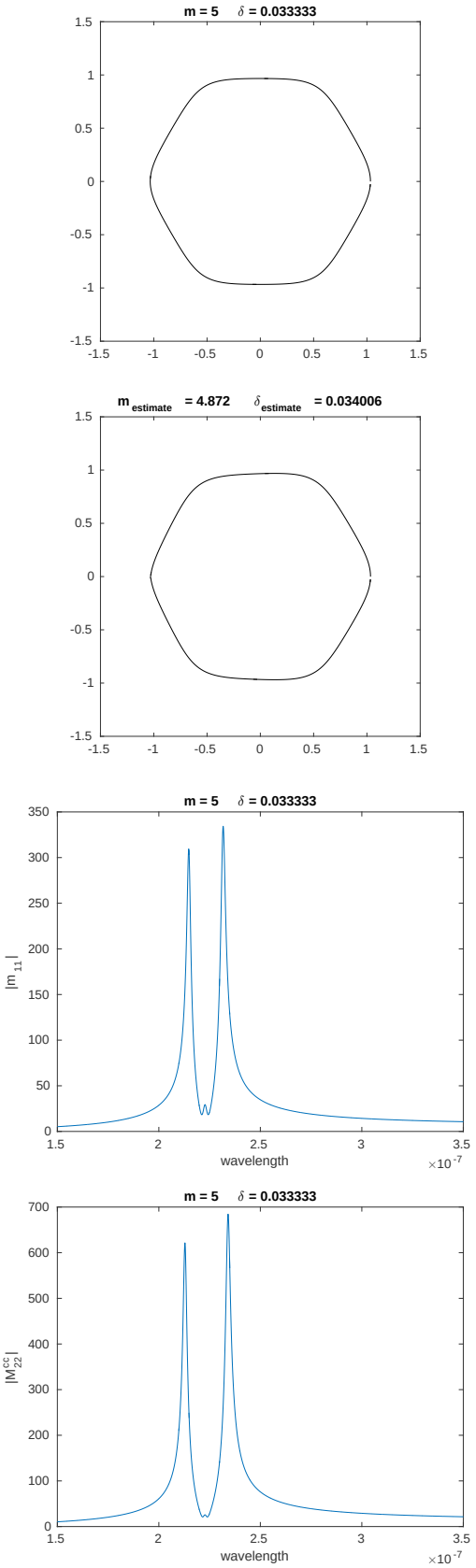

FIGURE 6.3: From top to bottom and left to right: initial shape, reconstructed shape, $|m_{11}|$ and $|M_{22}^{cc}|$ with respect to the wavelength for $m = 5$ and $\delta = 0.03333$.

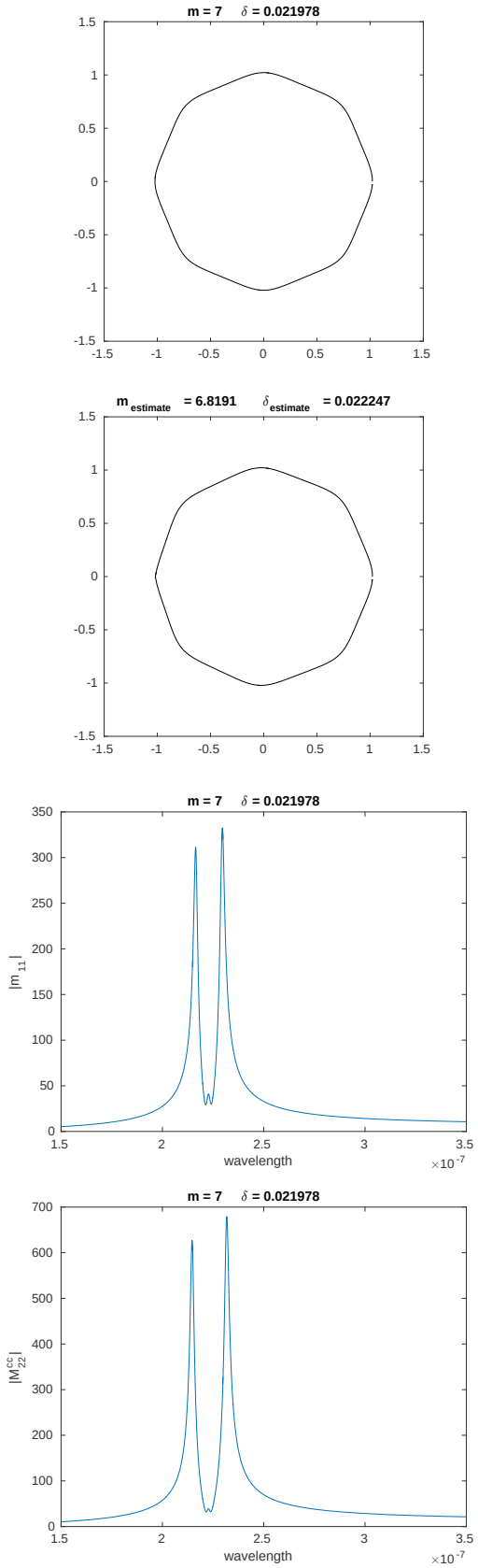

FIGURE 6.4: From top to bottom and left to right: initial shape, reconstructed shape, $|m_{11}|$ and $|M_{22}^{cc}|$ with respect to the wavelength for $m = 7$, $\delta = 0.021978$.

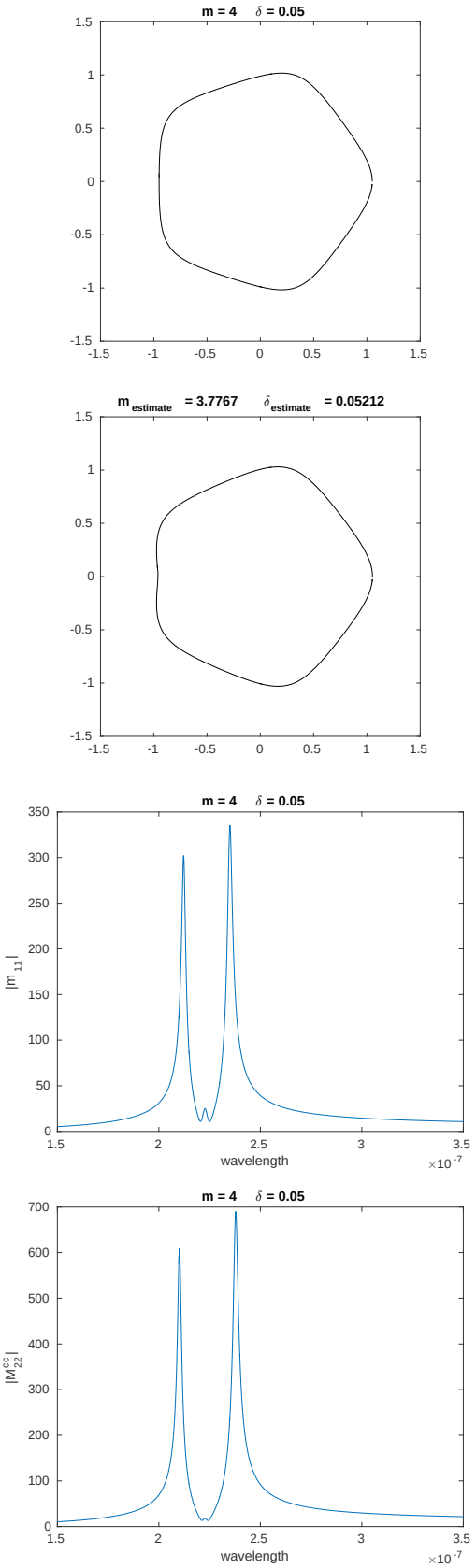

FIGURE 6.5: From top to bottom and left to right: initial shape, reconstructed shape, $|m_{11}|$ and $|M_{22}^{cc}|$ with respect to the wavelength for $m = 4$, $\delta = 0.05$.

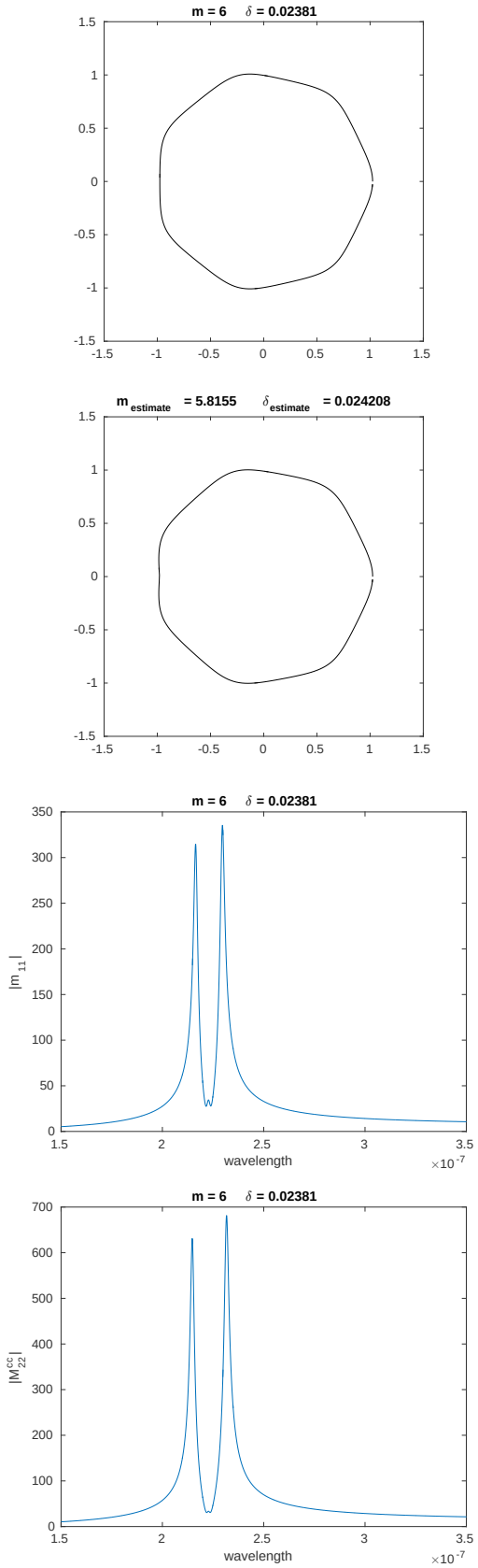

FIGURE 6.6: From top to bottom and left to right: initial shape, reconstructed shape, $|m_{11}|$ and $|M_{22}^{cc}|$ with respect to the wavelength for $m = 6$, $\delta = 0.02381$.

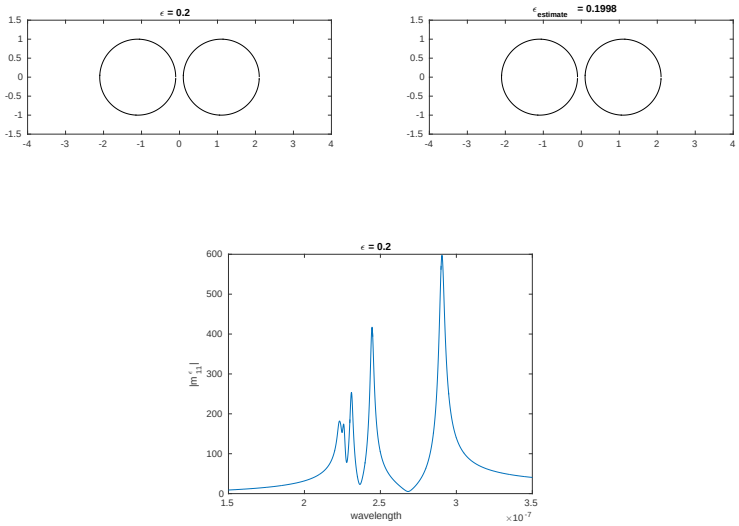

FIGURE 6.7: From top left to bottom: initial shape, reconstructed shape and $|m_{11}|$ with respect to the wavelength for $\epsilon = 0.2$.

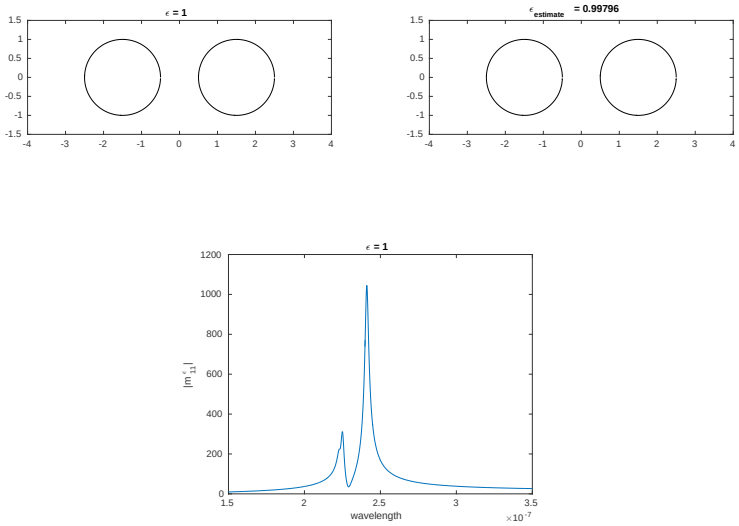

FIGURE 6.8: From top left to bottom: initial shape, reconstructed shape and $|m_{11}|$ with respect to the wavelength for $\epsilon = 1$.

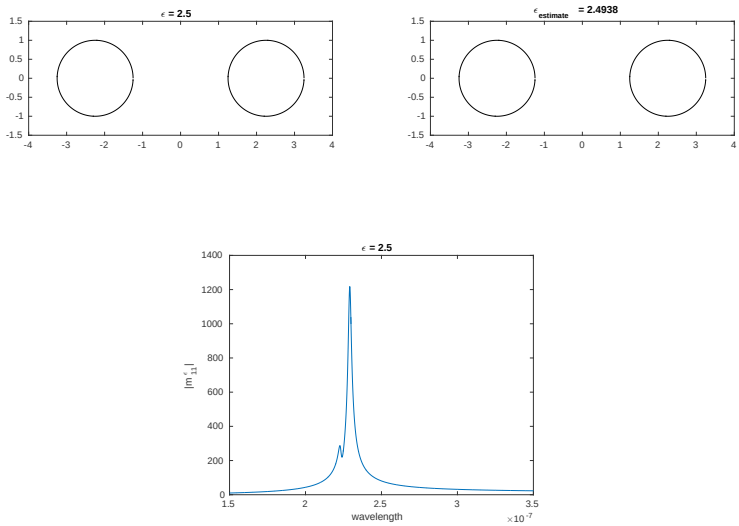

FIGURE 6.9: From top left to bottom right: initial shape, reconstructed shape and $|m_{11}|$ with respect to the wavelength for $\epsilon = 2.5$.

Chapter 7

Super-Resolution with Plasmonic Nanoparticles

Contents

7.1 Introduction

Super-resolution is a set of techniques meant to cross the barrier of diffraction limits by reducing the focal spot size. This resolution limit applies only to light that has propagated for a distance substantially larger than its wavelength [39,40]. It is known that the resolution limit (or the diffraction limit) in a general inhomogeneous space is determined by the imaginary part of the Green function in the associated space [4]. An idea to break the resolution limit is to insert subwavelength resonators in the homogeneous space. This way, we can introduce propagating subwavelength resonance modes which, when excited at the right frequency, encode subwavelength informations. This yield a Green's function whose imaginary part exhibits subwavelength peaks and therefore break the resolution limit (or diffraction limit) in the homogeneous space. The principle has been mathematically demonstrated in [30].

Super-focusing is the counterpart of super-resolution. It is a concept for waves to be confined to a length scale significantly smaller than the diffraction limit of the focused waves. As for the resolution problem, the focusing capacity is also determined by the imaginary part of the Green function in the associated space. The super-focusing phenomenon is being intensively investigated in the field of nanophotonics as a possible technique to focus electromagnetic radiation in a region of order of a few nanometers beyond the diffraction limit of light and thereby causing an extraordinary enhancement of the electromagnetic fields.

Here, using the fact that plasmonic particles are ideal subwavelength resonators, we consider the possibility of super-resolution (super-focusing) by using a system of identical plasmonic particles.

First, a precise analisys of field behavior of multiple plasmonic particles is in order.

7.2 Multiple plasmonic nanoparticles

7.2.1 Layer potential formulation in the multi-particle case

We consider the scattering of an incident time harmonic wave u^i by multiple weakly coupled plasmonic nanoparticles in three dimensions. Our motivation is to demonstrate the principle of super-resolution in resonant media; see Section 7.3. The analysis done in this section follows the same lines as those in chapter 2. The scattering from multiple weakly coupled, non-resonant small particles can be analyzed in the same way. However, no super-resolution can be achieved in this case.

For ease of exposition, we consider the case of L particles with an identical shape. We assume that Condition 2.1 holds. Moreover, in contrast to Section 2.3 where the size of the particle is assumed to be of order one, we assume the following condition in this section.

Condition 7.1. *All the identical particles have size of order δ which is a small parameter and the distances between neighboring ones are of order one.*

We write $D_l = z_l + \delta B$, $l = 1, 2, \ldots, L$, where B has size one and is centered at the origin. Moreover, we denote $D_0 = \delta B$ as our reference

nanoparticle. Denote by

$$D = \bigcup_{l=1}^{L} D_l, \quad \varepsilon_D = \varepsilon_m \chi(\mathbb{R}^3 \backslash \bar{D}) + \varepsilon_c \chi(\bar{D}), \quad \mu_D = \mu_m \chi(\mathbb{R}^3 \backslash \bar{D}) + \mu_c \chi(D).$$

The scattering problem can be modeled by the following Helmholtz equation:

$$\begin{cases} \nabla \cdot \dfrac{1}{\mu_D} \nabla u + \omega^2 \varepsilon_D u = 0 \quad \text{in } \mathbb{R}^3 \backslash \partial D, \\ u_+ - u_- = 0 \quad \text{on } \partial D, \\ \dfrac{1}{\mu_m} \dfrac{\partial u}{\partial \nu}\Big|_+ - \dfrac{1}{\mu_c} \dfrac{\partial u}{\partial \nu}\Big|_- = 0 \quad \text{on } \partial D, \\ u^s := u - u^i \text{ satisfies the Sommerfeld radiation condition.} \end{cases} \tag{7.1}$$

Let

$$\begin{aligned} u^i(x) &= e^{ik_m d \cdot x}, \\ F_{l,1}(x) &= -u^i(x)\big|_{\partial D_l} = -e^{ik_m d \cdot x}\big|_{\partial D_l}, \\ F_{l,2}(x) &= -\frac{\partial u^i}{\partial \nu}(x)\Big|_{\partial D_l} = -ik_m e^{ik_m d \cdot x} d \cdot \nu(x)\big|_{\partial D_l}, \end{aligned}$$

and define the operator $\mathcal{K}^k_{D_p,D_l}$ by

$$\mathcal{K}^k_{D_p,D_l}[\psi](x) = \int_{\partial D_p} \frac{\partial G(x,y,k)}{\partial \nu(x)} \psi(y) d\sigma(y), \quad x \in \partial D_l.$$

Analogously, we define

$$\mathcal{S}^k_{D_p,D_l}[\psi](x) = \int_{\partial D_p} G(x,y,k) \psi(y) d\sigma(y), \quad x \in \partial D_l.$$

The solution u of (7.1) can be represented as follows:

$$u(x) = \begin{cases} u^i + \displaystyle\sum_{l=1}^{L} \mathcal{S}^{k_m}_{D_l}[\psi_l], & x \in \mathbb{R}^3 \backslash \bar{D}, \\ \displaystyle\sum_{l=1}^{L} \mathcal{S}^{k_c}_{D_l}[\phi_l], & x \in D, \end{cases}$$

where $\phi_l, \psi_l \in H^{-\frac{1}{2}}(\partial D_l)$ satisfy the following system of integral equations

$$\begin{cases} \mathcal{S}^{k_m}_{D_l}[\psi_l] - \mathcal{S}^{k_c}_{D_l}[\phi_l] + \displaystyle\sum_{p \neq l} \mathcal{S}^{k_m}_{D_p,D_l}[\psi_p] = F_{l,1}, \\ \dfrac{1}{\mu_m}\Big(\dfrac{1}{2} Id + (\mathcal{K}^{k_m}_{D_l})^*\Big)[\psi_l] + \dfrac{1}{\mu_c}\Big(\dfrac{1}{2} Id - (\mathcal{K}^{k_c}_{D_l})^*\Big)[\phi_l] \\ \qquad + \dfrac{1}{\mu_m} \displaystyle\sum_{p \neq l} \mathcal{K}^{k_m}_{D_p,D_l}[\psi_p] = F_{l,2}, \end{cases}$$

and

$$\begin{cases} F_{l,1} = -u^i \quad \text{on } \partial D_l, \\ F_{l,2} = -\dfrac{1}{\mu_m}\dfrac{\partial u^i}{\partial \nu} \quad \text{on } \partial D_l. \end{cases}$$

7.2.2 Feld behavior at plasmonic resonances in the multi-particle case

We consider the scattering in the quasi-static regime, i.e., when the incident wavelength is much greater than one. With proper dimensionless analysis, we can assume that $\omega \ll 1$. As a consequence, $\mathcal{S}_D^{k_c}$ is invertible. Note that

$$\phi_l = (\mathcal{S}_{D_l}^{k_c})^{-1}\big(\mathcal{S}_{D_l}^{k_m}[\psi_l] + \sum_{p\neq l} \mathcal{S}_{D_p,D_l}^{k_m}[\psi_p] - F_{l,1}\big).$$

We obtain the following equation for ψ_l's,

$$\mathcal{A}_D(w)[\psi] = f,$$

where

$$\mathcal{A}_D(w) = \begin{pmatrix} \mathcal{A}_{D_1}(\omega) & & & \\ & \mathcal{A}_{D_2}(\omega) & & \\ & & \ddots & \\ & & & \mathcal{A}_{D_L}(\omega) \end{pmatrix} + \begin{pmatrix} 0 & \mathcal{A}_{1,2}(\omega) & \cdots & \mathcal{A}_{1,L}(\omega) \\ \mathcal{A}_{2,1}(\omega) & 0 & \cdots & \mathcal{A}_{2,L}(\omega) \\ \vdots & \cdots & 0 & \vdots \\ \mathcal{A}_{L,1}(\omega) & \cdots & \mathcal{A}_{L,L-1}(\omega) & 0 \end{pmatrix},$$

$$\psi = \begin{pmatrix} \psi_1 \\ \psi_2 \\ \vdots \\ \psi_L \end{pmatrix}, \quad f = \begin{pmatrix} f_1 \\ f_2 \\ \vdots \\ f_L \end{pmatrix},$$

and

$$\begin{aligned} \mathcal{A}_{D_l}(\omega) &= \frac{1}{\mu_m}\big(\frac{1}{2}Id + (\mathcal{K}_{D_l}^{k_m})^*\big) + \frac{1}{\mu_c}\big(\frac{1}{2}Id - (\mathcal{K}_{D_l}^{k_c})^*\big)(\mathcal{S}_{D_l}^{k_c})^{-1}\mathcal{S}_{D_l}^{k_m}, \\ \mathcal{A}_{l,p}(\omega) &= \frac{1}{\mu_c}\big(\frac{1}{2}Id - (\mathcal{K}_{D_l}^{k_c})^*\big)(\mathcal{S}_{D_l}^{k_c})^{-1}\mathcal{S}_{D_p,D_l}^{k_m} + \frac{1}{\mu_m}\mathcal{K}_{D_p,D_l}^{k_m}, \\ f_l &= F_{l,2} + \frac{1}{\mu_c}\big(\frac{1}{2}Id - (\mathcal{K}_{D_l}^{k_c})^*\big)(\mathcal{S}_{D_l}^{k_c})^{-1}[F_{l,1}]. \end{aligned}$$

The following asymptotic expansions hold (see chapter 1 for the definition of $\mathcal{H}^*(\partial D)$ and chapter 2 and Appendix B for the definition of the operators).

Lemma 7.2.1. *(i) Regarded as operators from $\mathcal{H}^*(\partial D_p)$ into $\mathcal{H}^*(\partial D_l)$, we have*

$$\mathcal{A}_{D_j}(\omega) = \mathcal{A}_{D_j,0} + O(\delta^2\omega^2),$$

(ii) Regarded as operators from $\mathcal{H}^*(\partial D_l)$ into $\mathcal{H}^*(\partial D_j)$, we have

$$\mathcal{A}_{l,p}(\omega) = \frac{1}{\mu_c}\big(\frac{1}{2}Id - \mathcal{K}_{D_l}^*\big)\mathcal{S}_{D_l}^{-1}\big(\mathcal{S}_{p,l,0,1} + \mathcal{S}_{p,l,0,2}\big) + \frac{1}{\mu_m}\mathcal{K}_{p,l,0,0} + O(\delta^2\omega^2) + O(\delta^4).$$

Moreover,

$$\begin{aligned}(\frac{1}{2}Id-\mathcal{K}_{D_l}^*)\circ\mathcal{S}_{D_l}^{-1}\circ\mathcal{S}_{p,l,0,1} &= O(\delta^2),\\ (\frac{1}{2}Id-\mathcal{K}_{D_l}^*)\circ\mathcal{S}_{D_l}^{-1}\circ\mathcal{S}_{p,l,0,2} &= O(\delta^3),\\ \mathcal{K}_{p,l,0,0} &= O(\delta^2).\end{aligned}$$

Proof. The proof of (i) follows from Lemmas 2.3.4 and B.2.3. We now prove (ii). Recall that

$$\begin{aligned}\frac{1}{2}Id-(\mathcal{K}_{D_l}^{k_c})^* &= \frac{1}{2}Id-\mathcal{K}_{D_l}^*+O(\delta^2\omega^2),\\ (\mathcal{S}_{D_l}^{k_c})^{-1} &= \mathcal{S}_{D_l}^{-1}-k_c\mathcal{S}_{D_l}^{-1}\mathcal{S}_{D_l,1}\mathcal{S}_{D_l}^{-1}+O(\delta^2\omega^2),\\ \mathcal{S}_{D_p,D_l}^{k_m} &= \mathcal{S}_{p,l,0,0}+\mathcal{S}_{p,l,0,1}+\mathcal{S}_{p,l,0,2}+k_m\mathcal{S}_{p,l,1}+k_m^2\mathcal{S}_{p,l,2,0}+O(\delta^4)+O(\omega^2\delta^2)\\ \mathcal{K}_{D_p,D_l}^{k_m} &= \mathcal{K}_{p,l,0,0}+O(\omega^2\delta^2).\end{aligned}$$

Using the identity

$$(\frac{1}{2}Id-\mathcal{K}_{D_l}^*)\mathcal{S}_{D_l}^{-1}[\chi(D_l)]=0,$$

we can derive that

$$\begin{aligned}\mathcal{A}_{l,p}(\omega) &= \frac{1}{\mu_c}(\frac{1}{2}Id-\mathcal{K}_{D_l}^*)(\mathcal{S}_{D_l}^{k_c})^{-1}\mathcal{S}_{D_p,D_l}^{k_m}+\frac{1}{\mu_m}\mathcal{K}_{p,l,0,0}+O(\delta^2\omega^2)\\ &= \frac{1}{\mu_c}(\frac{1}{2}Id-\mathcal{K}_{D_l}^*)\mathcal{S}_{D_l}^{-1}\mathcal{S}_{D_p,D_l}^{k_m}+\frac{1}{\mu_m}\mathcal{K}_{p,l,0,0}+O(\delta^2\omega^2)\\ &= \frac{1}{\mu_c}(\frac{1}{2}Id-\mathcal{K}_{D_l}^*)\mathcal{S}_{D_l}^{-1}\big(\mathcal{S}_{p,l,0,0}+\mathcal{S}_{p,l,0,1}+\mathcal{S}_{p,l,0,2}+k_m\mathcal{S}_{p,l,1}+k_m^2\mathcal{S}_{p,l,2,0}+O(\delta^4)\big)\\ &+\frac{1}{\mu_m}\mathcal{K}_{p,l,0,0}+O(\delta^2\omega^2)\\ &= \frac{1}{\mu_c}(\frac{1}{2}Id-\mathcal{K}_{D_l}^*)\mathcal{S}_{D_l}^{-1}\big(\mathcal{S}_{p,l,0,1}+\mathcal{S}_{p,l,0,2}\big)+\frac{1}{\mu_m}\mathcal{K}_{p,l,0,0}+O(\delta^2\omega^2)+O(\delta^4).\end{aligned}$$

The rest of the lemma follows from Lemmas B.2.3 and B.2.6. □

Denote by $\mathcal{H}^*(\partial D)=\mathcal{H}^*(\partial D_1)\times\ldots\times\mathcal{H}^*(\partial D_L)$, which is equipped with the inner product

$$(\psi,\phi)_{\mathcal{H}^*}=\sum_{l=1}^{L}(\psi_l,\phi_l)_{\mathcal{H}^*(\partial D_l)}.$$

With the help of Lemma 7.2.1, the following result is obvious.

Lemma 7.2.2. *Regarded as an operator from $\mathcal{H}^*(\partial D)$ into $\mathcal{H}^*(\partial D)$, we have*

$$\mathcal{A}_{\mathcal{D}}(\omega) = \mathcal{A}_{D,0}+\mathcal{A}_{D,1}+O(\omega^2\delta^2)+O(\delta^4),$$

where

$$\mathcal{A}_{D,0}=\begin{pmatrix}\mathcal{A}_{D_1,0} & & & \\ & \mathcal{A}_{D_2,0} & & \\ & & \cdots & \\ & & & \mathcal{A}_{D_L,0}\end{pmatrix},\ \mathcal{A}_{D,1}=\begin{pmatrix}0 & \mathcal{A}_{D,1,12} & \mathcal{A}_{D,1,13} & \cdots\\ \mathcal{A}_{D,1,21} & 0 & \mathcal{A}_{D,1,23} & \cdots\\ & & \cdots & \\ \mathcal{A}_{D,1,L1} & \cdots & \mathcal{A}_{D,1,LL-1} & 0\end{pmatrix}$$

with

$$\begin{aligned}\mathcal{A}_{D_l,0} &= \big(\frac{1}{2\mu_m}+\frac{1}{2\mu_c}\big)Id-\big(\frac{1}{\mu_c}-\frac{1}{\mu_m}\big)\mathcal{K}^*_{D_l},\\ \mathcal{A}_{D,1,pq} &= \frac{1}{\mu_c}\big(\frac{1}{2}Id-\mathcal{K}^*_{D_p}\big)\mathcal{S}^{-1}_{D_p}\big(\mathcal{S}_{q,p,0,1}+\mathcal{S}_{q,p,0,2}\big)+\frac{1}{\mu_m}\mathcal{K}_{q,p,0,0}.\end{aligned}$$

It is evident that

$$\mathcal{A}_{D,0}[\psi]=\sum_{j=0}^{\infty}\sum_{l=1}^{L}\tau_j(\psi,\varphi_{j,l})_{\mathcal{H}^*}\varphi_{j,l}, \tag{7.2}$$

where

$$\tau_j = \frac{1}{2\mu_m}+\frac{1}{2\mu_c}-\big(\frac{1}{\mu_c}-\frac{1}{\mu_m}\big)\lambda_j, \tag{7.3}$$

$$\varphi_{j,l} = \varphi_j e_l \tag{7.4}$$

with e_l being the standard basis of $\mathbb{R}^L$.

We take $\mathcal{A}_D(\omega)$ as a perturbation to the operator $\mathcal{A}_{D,0}$ for small ω and small δ. Using a standard perturbation argument, we can derive the perturbed eigenvalues and eigenfunctions. For simplicity, we assume that the following conditions hold.

Condition 7.2. *Each eigenvalue λ_j, $j\in J$, of the operator $\mathcal{K}^*_{D_0}$ is simple. Moreover, we have $\omega^2\ll\delta$.*

In what follows, we only use the first order perturbation theory and derive the leading order term, i.e., the perturbation due to the term $\mathcal{A}_{D,1}$. For each l, we define an $L\times L$ matrix R_l by letting

$$\begin{aligned}R_{l,pq} &= \big(\mathcal{A}_{D,1}[\varphi_{l,p}],\varphi_{l,q}\big)_{\mathcal{H}^*},\\ &= \Big(\mathcal{A}_{D,1}[\varphi_l e_p],\varphi_l e_q\Big)_{\mathcal{H}^*},\\ &= \big(\mathcal{A}_{D,1,pq}[\varphi_l],\varphi_l\big)_{\mathcal{H}^*}.\end{aligned}$$

Lemma 7.2.3. *The matrix $R_l=(R_{l,pq})_{p,q=1,\dots,L}$ has the following explicit expression:*

$$\begin{aligned}R_{l,pp} &= 0,\\ R_{l,pq} &= \frac{3}{4\pi\mu_c}(\lambda_j-\frac{1}{2})\sum_{|\alpha|=|\beta|=1}\int_{\partial D_0}\int_{\partial D_0}\frac{(z_p-z_q)^{\alpha+\beta}}{|z_p-z_q|^5}x^\alpha y^\beta\varphi_l(x)\varphi_l(y)d\sigma(x)d\sigma(y)\\ &\quad+\big(\frac{1}{4\pi\mu_c}-\frac{1}{4\pi\mu_m}\big)(\lambda_j-\frac{1}{2})\int_{\partial D_0}\int_{\partial D_0}\frac{x\cdot y}{|z_p-z_q|^3}\varphi_l(x)\varphi_l(y)d\sigma(x)d\sigma(y)\\ &= O(\delta^3),\quad p\neq q.\end{aligned}$$

Proof. It is clear that $R_{l,pp}=0$. For $p\neq q$, we have

$$R_{l,pq}=R^I_{l,pq}+R^{II}_{l,pq}+R^{III}_{l,pq},$$

where

$$\begin{aligned} R^I_{l,pq} &= \frac{1}{\mu_c}\Big((\frac{1}{2}Id-\mathcal{K}^*_{D_p})\mathcal{S}^{-1}_{D_p}\mathcal{S}_{q,p,0,1}[\varphi_l],\varphi_l\Big)_{\mathcal{H}^*(\partial D_l)},\\ R^{II}_{l,pq} &= \frac{1}{\mu_c}\Big((\frac{1}{2}Id-\mathcal{K}^*_{D_p})\mathcal{S}^{-1}_{D_p}\mathcal{S}_{q,p,0,2}[\varphi_l],\varphi_l\Big)_{\mathcal{H}^*(\partial D_l)},\\ R^{III}_{l,pq} &= \frac{1}{\mu_m}\big(\mathcal{K}_{q,p,0,0}[\varphi_l],\varphi_l\big)_{\mathcal{H}^*(\partial D_l)}. \end{aligned}$$

We first consider $R^I_{l,pq}$. By the following identity

$$(\frac{1}{2}Id-\mathcal{K}^*_{D_p})\mathcal{S}_{D_l}[\varphi_l]=\mathcal{S}_{D_l}(\frac{1}{2}Id-\mathcal{K}_{D_p})[\varphi_l]=(\lambda_j-\frac{1}{2})\varphi_l,$$

we obtain

$$\begin{aligned} R^I_{l,pq} &= -\frac{1}{\mu_c}\Big((\frac{1}{2}Id-\mathcal{K}^*_{D_p})\mathcal{S}^{-1}_{D_p}\mathcal{S}_{q,p,0,1}[\varphi_l],\mathcal{S}_{D_l}[\varphi_l]\Big)_{L^2(\partial D_l)},\\ &= \frac{1}{\mu_c}(\lambda_j-\frac{1}{2})\big(\mathcal{S}_{q,p,0,1}[\varphi_l],\mathcal{S}_{D_l}[\varphi_l]\big)_{L^2(\partial D_l)}. \end{aligned}$$

Using the explicit representation of $\mathcal{S}_{q,p,0,1}$ and the fact that $(\chi(\partial D_j),\phi_l)_{L^2(\partial D_j)}=0$ for $j\neq 0$, we further conclude that

$$R^I_{l,pq}=0.$$

Similarly, we have

$$\begin{aligned} R^{II}_{l,pq} &= \frac{1}{\mu_c}(\lambda_j-\frac{1}{2})\big(\mathcal{S}_{q,p,0,2}[\varphi_l],\mathcal{S}_{D_l}[\varphi_l]\big)_{L^2(\partial D_l)},\\ &= \frac{1}{\mu_c}(\lambda_j-\frac{1}{2})\cdot\\ &\quad \sum_{|\alpha|=|\beta|=1}\int_{\partial D_0}\int_{\partial D_0}\Big(\frac{3(z_p-z_q)^{\alpha+\beta}}{4\pi|z_p-z_q|^5}x^\alpha y^\beta+\frac{\delta_{\alpha\beta}x^\alpha y^\beta}{4\pi|z_p-z_q|^3}\Big)\varphi_l(x)\varphi_l(y)d\sigma(x)d\sigma(y),\\ &= \frac{3}{4\pi\mu_c}(\lambda_j-\frac{1}{2})\sum_{|\alpha|=|\beta|=1}\int_{\partial D_0}\int_{\partial D_0}\frac{(z_p-z_q)^{\alpha+\beta}}{|z_p-z_q|^5}x^\alpha y^\beta\varphi_l(x)\varphi_l(y)d\sigma(x)d\sigma(y)\\ &\quad +\frac{1}{4\pi\mu_c}(\lambda_j-\frac{1}{2})\sum_{|\alpha|=1}\int_{\partial D_0}\int_{\partial D_0}\frac{1}{|z_p-z_q|^3}x^\alpha y^\alpha\varphi_l(x)\varphi_l(y)d\sigma(x)d\sigma(y). \end{aligned}$$

Finally, note that

$$\mathcal{K}_{q,p,0,0}[\varphi_l]=\frac{1}{4\pi|z_p-z_q|^3}a\cdot\nu(x)=\frac{1}{4\pi|z_p-z_q|^3}\sum_{m=1}^{3}a_m\nu_m(x),$$

where $a_m=\big((y-z_q)_m,\varphi_l\big)_{L^2(\partial D_q)}$, and $a=(a_1,a_2,a_3)^T$.

By identity (2.29), we have

$$\begin{aligned} R^{III}_{l,pq} &= -\frac{1}{\mu_m}\big(\mathcal{K}_{q,p,0,0}[\varphi_l],\varphi_l\big)_{\mathcal{H}^*(\partial D_l)} \\ &= -\frac{1}{4\pi|z_p-z_q|^3\mu_m}\big(a\cdot\nu(x),\varphi_l\big)_{\mathcal{H}^*(\partial D_l)} \\ &= -\frac{1}{4\pi|z_p-z_q|^3\mu_m}\left((\frac{1}{2}Id-\mathcal{K}^*_{D_p})\mathcal{S}^{-1}_{D_p}(a\cdot(x-z_p)),\varphi_l\right)_{\mathcal{H}^*(\partial D_l)} \\ &= -\frac{1}{4\pi|z_p-z_q|^3\mu_m}(\lambda_j-\frac{1}{2})\big(a\cdot(x-z_p),\varphi_l\big)_{L^2(\partial D_p)} \\ &= -\frac{1}{4\pi|z_p-z_q|^3\mu_m}(\lambda_j-\frac{1}{2})\int_{\partial D_0}\int_{\partial D_0} x\cdot y\varphi_l(x)\varphi_l(y)d\sigma(x)d\sigma(y). \end{aligned}$$

This completes the proof of the lemma.

□

We now have an explicit formula for the matrix R_l. It is clear that R_l is symmetric, but not self-adjoint. For ease of presentation, we assume the following condition.

Condition 7.3. *R_l has L-distinct eigenvalues.*

We remark that Condition 7.3 is not essential for our analysis. Without this condition, the perturbation argument is still applicable, but the results may be quite complicated. We refer to [62] for a complete description of the perturbation theory.

Let $\tau_{j,l}$ and $X_{j,l}=(X_{j,l,1},\cdots,X_{j,l,L})^T$, $l=1,2,\ldots,L$, be the eigenvalues and normalized eigenvectors of the matrix R_j. Here, T denotes the transpose. We remark that each $X_{j,l}$ may be complex valued and may not be orthogonal to other eigenvectors.

Under perturbation, each τ_j is splitted into the following L eigenvalues of $\mathcal{A}(\omega)$,

$$\tau_{j,l}(\omega)=\tau_j+\tau_{j,l}+O(\delta^4)+O(\omega^2\delta^2). \tag{7.5}$$

The associated perturbed eigenfunctions have the following form

$$\varphi_{j,l}(\omega)=\sum_{p=1}^{L}X_{j,l,p}e_p\varphi_j+O(\delta^4)+O(\omega^2\delta^2). \tag{7.6}$$

We are interested in solving the equation $\mathcal{A}_D(\omega)[\psi]=f$ when ω is close to the resonance frequencies, i.e., when $\tau_{j,l}(\omega)$ are very small for some j's. In this case, the major part of the solution would be based on the excited resonance modes $\varphi_{j,l}(\omega)$. For this purpose, we introduce the index set of resonance J as we did in chapter 2 for a single particle case.

We define

$$P_J(\omega)\varphi_{j,m}(\omega)=\begin{cases}\varphi_{j,m}(\omega), & j\in J,\\ 0, & j\in J^c.\end{cases}$$

In fact,

$$P_J(\omega)=\sum_{j\in J}P_j(\omega)=\sum_{j\in J}\frac{1}{2\pi i}\int_{\gamma_j}(\xi-\mathcal{A}_D(\omega))^{-1}d\xi, \tag{7.7}$$

where γ_j is a Jordan curve in the complex plane enclosing only the eigenvalues $\tau_{j,l}(\omega)$ for $l=1,2,\ldots,L$ among all the eigenvalues.

To obtain an explicit representation of $P_J(\omega)$, we consider the adjoint operator $\mathcal{A}_D(\omega)^*$. By a similar perturbation argument, we can obtain its perturbed eigenvalue and eigenfunctions. Note that the adjoint matrix $\bar{R}_j^T = \bar{R}_j$ has eigenvalues $\overline{\tau_{j,l}}$ and corresponding eigenfunctions $\overline{X_{j,l}}$. Then the eigenvalues and eigenfunctions of $\mathcal{A}_D(\omega)^*$ have the following form

$$\begin{aligned}\widetilde{\tau}_{j,l}(\omega) &= \tau_j + \overline{\tau_{j,l}} + O(\delta^4) + O(\omega^2\delta^2),\\ \widetilde{\varphi}_{j,l}(\omega) &= \widetilde{\varphi}_{j,l} + O(\delta^4) + O(\omega^2\delta^2),\end{aligned}$$

where

$$\widetilde{\varphi}_{j,l} = \sum_{p=1}^{L} \widetilde{X}_{j,l,p} e_p \varphi_j$$

with $\widetilde{X}_{j,l,p}$ being a multiple of $\overline{X_{j,l,p}}$.

We normalize $\widetilde{\varphi}_{j,l}$ in a way such that the following holds

$$(\varphi_{j,p}, \widetilde{\varphi}_{j,q})_{\mathcal{H}^*(\partial D)} = \delta_{pq},$$

which is also equivalent to the following condition

$$\overline{X_{j,p}}^T \widetilde{X}_{j,q} = \delta_{pq}.$$

Then, we can show that the following result holds.

Lemma 7.2.4. *In the space $\mathcal{H}^*(\partial D)$, as ω goes to zero, we have*

$$f = \omega f_0 + O(\omega^2 \delta^{\frac{3}{2}}),$$

where $f_0 = (f_{0,1}, \ldots, f_{0,L})^T$ with

$$f_{0,l} = -i\sqrt{\varepsilon_m \mu_m} e^{ik_m d\cdot z_l} \left(\frac{1}{\mu_m} d \cdot \nu(x) + \frac{1}{\mu_c} (\frac{1}{2} Id - \mathcal{K}_{D_l}^*) \mathcal{S}_{D_l}^{-1} [d \cdot (x - z)] \right) = O(\delta^{\frac{3}{2}}).$$

Proof. We first show that

$$\|u\|_{\mathcal{H}^*(\partial D_0)} = \delta^{\frac{3}{2}+m} \|u\|_{\mathcal{H}^*(\partial B)}, \quad \|u\|_{\mathcal{H}(\partial D_0)} = \delta^{\frac{1}{2}+m} \|u\|_{\mathcal{H}(\partial B)}$$

for any homogeneous function u such that $u(\delta x) = \delta^m u(x)$. Indeed, we have $\eta(u)(x) = \delta^m u(x)$. Since $\|\eta(u)\|_{\mathcal{H}^*(\partial B)} = \delta^{-\frac{3}{2}} \|u\|_{\mathcal{H}^*(\partial D_0)}$ (see Appendix B.2), we obtain

$$\|u\|_{\mathcal{H}^*(\partial D_0)} = \delta^{\frac{3}{2}} \|\eta(u)\|_{\mathcal{H}^*(\partial B)} = \delta^{\frac{3}{2}+m} \|u\|_{\mathcal{H}^*(\partial B)},$$

which proves our first claim. The second claim follows in a similar way. Using this result, by a similar argument as in the proof of Lemma 2.3.6 we arrive at the desired asymptotic result. □

Denote by $Z = (Z_1, \ldots, Z_L)$, where $Z_j = ik_m e^{ik_m d\cdot z_j}$. We are ready to present our main result in this section.

Theorem 7.2.1. *Under Conditions 2.1, 2.2, 2.3, 7.1, and 7.3, the scattered field by L plasmonic particles has the following representation*

$$u^s = \mathcal{S}_D^{k_m}[\psi],$$

where

$$\begin{aligned}\psi &= \sum_{j\in J}\sum_{l=1}^{L}\frac{\left(f,\widetilde{\varphi}_{j,l}(\omega)\right)_{\mathcal{H}^*}\varphi_{j,l}(\omega)}{\tau_{j,l}(\omega)} + \mathcal{A}_D(\omega)^{-1}(P_{J^c}(\omega)f)\\ &= \sum_{j\in J}\sum_{l=1}^{L}\frac{(d\cdot\nu(x),\varphi_j)_{\mathcal{H}^*(\partial D_0)}Z\overline{\widetilde{X}_{j,l}}\,\varphi_{j,l}+O(\omega^2\delta^{\frac{3}{2}})}{\lambda-\lambda_j+\left(\frac{1}{\mu_c}-\frac{1}{\mu_m}\right)^{-1}\tau_{j,l}+O(\delta^4)+O(\delta^2\omega^2)}+O(\omega\delta^{\frac{3}{2}}).\end{aligned}$$

Proof. The proof is similar to that of Theorem 3.3.2. □

As a consequence, the following result holds.

Corollary 7.2.1. *With the same notation as in Theorem 7.2.1 and under the additional condition that*

$$\min_{j\in J}|\tau_{j,l}(\omega)| \gg \omega^q\delta^p,$$

for some integer p and q, and

$$\tau_{j,l}(\omega) = \tau_{j,l,p,q} + o(\omega^q\delta^p),$$

we have

$$\psi = \sum_{j\in J}\sum_{l=1}^{L}\frac{(d\cdot\nu(x),\varphi_j)_{\mathcal{H}^*(\partial D_0)}Z\overline{\widetilde{X}_{j,l}}\,\varphi_{j,l}+O(\omega^2\delta^{\frac{3}{2}})}{\tau_{j,l,p,q}}+O(\omega\delta^{\frac{3}{2}}).$$

7.3 Super-resolution (super-focusing) by using plasmonic particles

In [30,31], a rigorous mathematical theory is developed to explain the super-resolution phenomenon in microstructures with high contrast material around the source point. Such microstructures act like arrays of subwavelength sensors. A key ingredient is the calculation of the resonances and the Green function in the microstructure. By following the same methodology, we show in this section that one can achieve super-resolution using plasmonic nanoparticles as well.

7.3.1 Asymptotic expansion of the scattered field

In order to illustrate the super-resolution phenomenon, we set

$$u^i(x) = G(x,x_0,k_m) = -\frac{e^{ik_m|x-x_0|}}{4\pi|x-x_0|}.$$

Lemma 7.3.1. *In the space $\mathcal{H}^*(\partial D)$, as ω goes to zero, we have*

$$f = f_0 + O(\omega\delta^{\frac{3}{2}}) + O(\delta^{\frac{5}{2}}),$$

where $f_0 = (f_{0,1},\dots,f_{0,L})^T$ with

$$f_{0,l} = -\frac{1}{4\pi|z_l-x_0|^3}\left(\frac{1}{\mu_m}(z_l-x_0)\cdot\nu(x)+\frac{1}{\mu_c}(\frac{1}{2}Id-\mathcal{K}_{D_l}^*)\mathcal{S}_{D_l}^{-1}[(z_l-x_0)\cdot(x-z_l)]\right)=O(\delta^{\frac{3}{2}}).$$

Proof. The proof is similar to that of Lemma 2.3.6. Recall that

$$f_l = F_{l,2} + \frac{1}{\mu_c}\big(\frac{1}{2}Id - (\mathcal{K}_{D_l}^{k_c})^*\big)(\mathcal{S}_{D_l}^{k_c})^{-1}[F_{l,1}].$$

We can show that

$$F_{l,2} = -\frac{1}{\mu_m}\frac{\partial u^i}{\partial \nu} = -\frac{1}{4\pi\mu_m|z_l - x_0|^3}(z_l - x_0)\cdot\nu(x) + O(\delta^{\frac{5}{2}}) + O(\omega\delta^{\frac{3}{2}}) \quad \text{in } \mathcal{H}^*(\partial D_l).$$

Besides,

$$u^i(x)|_{\partial D_l} = -\frac{e^{ik_m|z_l - x_0|}}{4\pi|z_l - x_0|}\chi(\partial D_l) + \frac{1}{4\pi|z_l - x_0|^3}(z_l - x_0)\cdot(x - z_l) + O(\delta^{\frac{5}{2}}) + O(\omega\delta^{\frac{3}{2}}) \quad \text{in } \mathcal{H}(\partial D_l).$$

Using the identity $(\frac{1}{2}Id - \mathcal{K}_{D_l}^*)\mathcal{S}_{D_l}^{-1}[\chi(\partial D_l)] = 0$, we obtain that

$$\frac{1}{\mu_c}\big(\frac{1}{2}Id - (\mathcal{K}_{D_l}^{k_c})^*\big)(\mathcal{S}_{D_l}^{k_c})^{-1}[F_{l,1}] = -\frac{1}{4\pi|z_l - x_0|^3\mu_c}(\frac{1}{2}Id - \mathcal{K}_{D_l}^*)\mathcal{S}_{D_l}^{-1}[(z_l - x_0)\cdot(x - z_l)].$$

This completes the proof of the lemma. □

We now derive an asymptotic expansion of the scattered field in an intermediate regime which is neither too close to the plasmonic particles nor too far away. More precisely, let C be a fixed sufficient large positive number, we consider the following domain

$$\mathrm{D}_{\delta,k,C} = \big\{x \in \mathbb{R}^3;\ \min_{1\le l\le L}|x - z_l| \ge C\delta,\ \max_{1\le l\le L}|x - z_l| \le \frac{1}{Ck}\big\}.$$

Lemma 7.3.2. *Let $\psi_l \in \mathcal{H}^*(\partial D_l)$ and let $v(x) = \mathcal{S}_{D_l}^k[\psi_l](x)$. Then we have for $x \in \mathrm{D}_{\delta,k,C}$,*

$$\begin{aligned} v(x) &= G(x, z_l, k)\big(\frac{1}{|x - z_l|} - ik\big)\frac{x - z_l}{|x - z_l|}\cdot\int_{\partial D_0} y\psi_l(y)d\sigma(y) + O(\delta^{\frac{5}{2}})\|\psi_l\|_{\mathcal{H}^*(\partial D_l)} \\ &\quad + G(x, z_l, k)\int_{\partial D_0}\psi_l(y)d\sigma(y). \end{aligned}$$

Moreover, the following estimates hold

$$\begin{aligned} v(x) &= O(\delta^{\frac{3}{2}}) \quad \text{if } \int_{\partial D_0}\psi_l(y)d\sigma(y) = 0, \\ v(x) &= O(\delta^{\frac{1}{2}}) \quad \text{if } \int_{\partial D_0}\psi_l(y)d\sigma(y) \neq 0. \end{aligned}$$

Proof. We only consider the case when $l = 0$. The other case follows similarly or by coordinate translation. We have

$$v(x) = \mathcal{S}_{D_0}^k[\psi](x) = \int_{\partial D_0} G(x, y, k)\psi(y)d\sigma(y) = -\int_{\partial D_0}\frac{e^{ik|x-y|}}{4\pi|x - y|}\psi(y)d\sigma(y).$$

Since

$$G(x, y, k) = G(x, 0, k) + \sum_{|\alpha=1|}\frac{\partial G(x, 0, k)}{\partial y^\alpha}y^\alpha + \sum_{m\ge 2}\sum_{|\alpha=m|}\frac{\partial^m G(x, 0, k)}{\partial y^\alpha}y^\alpha,$$

and

$$\frac{\partial G(x,0,k)}{\partial y^\alpha} = -\frac{e^{ik|x|}}{4\pi|x|}\big(\frac{1}{|x|} - ik\big)\frac{x}{|x|} = G(x,0,k)\big(\frac{1}{|x|} - ik\big)\frac{x^\alpha}{|x|},$$

we obtain the required identity for the case $l = 0$. The estimate follows from the fact that

$$\|y^\alpha\|_{\mathcal{H}(\partial D_0)} = O(\delta^{\frac{2|\alpha|+1}{2}}).$$

This completes the proof of the lemma. □

Denote by

$$\begin{aligned} S_{j,l}(x,k) &= G(x,z_l,k)\frac{x-z_l}{|x-z_l|^2}\cdot\int_{\partial D_0} y\varphi_j(y)d\sigma(y), \\ S_l(x,k) &= G(x,z_l,k)\int_{\partial D_0}\varphi_0(y)d\sigma(y), \\ H_{j,l}(x_0) &= -\frac{1}{4\pi|z_l-x_0|^3}\big((z_l-x_0)\cdot\nu(x),\varphi_j\big)_{\mathcal{H}^*(\partial D_0)}. \end{aligned}$$

It is clear that the following size estimates hold

$$S_{j,l}(x,k) = O(\delta^{\frac{3}{2}}), \quad S_l(x,k) = O(\delta^{\frac{1}{2}}), \quad H_{j,l}(x_0) = O(\delta^{\frac{3}{2}}) \quad \text{for} j \neq 0, \quad H_{O,l}(x_0) = 0.$$

Theorem 7.3.1. *Under Conditions 2.1, 2.2, 2.3, 7.1, and 7.3, the Green function $\Gamma(x,x_0,k_m)$ in the presence of L plasmonic particles has the following representation in the quasi-static regime: for $x \in \mathrm{D}_{\delta,k_m,C}$,*

$$\begin{aligned} \Gamma(x,x_0,k_m) &= G(x,x_0,k_m) \\ &\quad + \sum_{j\in J}\sum_{l=1}^{L}\frac{H_{j,p}(x_0)\widetilde{X}_{j,l,p}X_{j,l,q}S_{j,q}(x,k_m) + O(\delta^4) + O(\omega\delta^3)}{\lambda-\lambda_j+\big(\frac{1}{\mu_c}-\frac{1}{\mu_m}\big)^{-1}\tau_{j,l} + O(\delta^4) + O(\delta^2\omega^2)} + O(\delta^3). \end{aligned}$$

Proof. With $u^i(x) = G(x,x_0,k_m)$, we have

$$\psi = \sum_{j\in J}\sum_{1\le l\le L} a_{j,l}\varphi_{j,l} + \sum_{1\le l\le L} a_{0,l}\varphi_{0,l} + O(\delta^{\frac{3}{2}}),$$

where

$$\begin{aligned} a_{j,l} &= (f,\widetilde{\varphi}_{j,l})_{\mathcal{H}^*(\partial D)} = (f_0,\widetilde{\varphi}_{j,l})_{\mathcal{H}^*(\partial D)} + O(\omega\delta^{\frac{3}{2}}) + O(\delta^{\frac{5}{2}}), \\ &= (\frac{1}{\mu_c}-\frac{1}{\mu_m})\widetilde{X}_{j,l,p}H_{j,p}(x_0) + O(\omega\delta^{\frac{3}{2}}) + O(\delta^{\frac{5}{2}}), \\ a_{0,l} &= (f,\widetilde{\varphi}_{0,l})_{\mathcal{H}^*(\partial D)} = O(\delta^{\frac{5}{2}}). \end{aligned}$$

By Lemma 7.3.2,

$$\begin{aligned} \mathcal{S}_D^{k_m}[\varphi_{j,l}](x) &= \sum_{1\le p\le L}\mathcal{S}_D^{k_m}[X_{j,l,p}\varphi_j e_p](x) = \sum_{1\le p\le L}X_{j,l,p}\mathcal{S}_{D_p}^{k_m}[\varphi_j](x) \\ &= \sum_{1\le p\le L}X_{j,l,p}S_{j,p}(x,k_m) + O(\delta^{\frac{5}{2}}) + O(\omega\delta^{\frac{3}{2}}). \end{aligned}$$

On the other hand, for $j = 0$, we have

$$\begin{aligned} \mathcal{S}_D^{k_m}[\varphi_{0,l}](x) &= O(\delta^{\frac{1}{2}}), \\ \tau_{0,l}(\omega) &= \tau_0 + O(\delta^4) + O(\delta^2\omega^2) = O(1). \end{aligned}$$

Therefore, we can deduce that

$$\begin{aligned} u^s &= \mathcal{S}_D^{k_m}[\psi](x) = \sum_{j\in J}\sum_{1\le l\le L} a_{j,l}\mathcal{S}_D^{k_m}[\varphi_{j,l}] + \sum_{1\le l\le L} a_{0,l}\mathcal{S}_D^{k_m}[\varphi_{0,l}] + O(\delta^3), \\ &= \sum_{j\in J}\sum_{l=1}^{L} \frac{1}{\tau_{j,l}(\omega)}\Big((\frac{1}{\mu_c} - \frac{1}{\mu_m})H_{j,p}(x_0)\widetilde{X}_{j,l,p}X_{j,l,q}S_{j,q}(x,k_m) + O(\omega\delta^3) + O(\delta^4)\Big) \\ &\quad + O(\delta^3), \\ &= \sum_{j\in J}\sum_{l=1}^{L} \frac{H_{j,p}(x_0)\widetilde{X}_{j,l,p}X_{j,l,q}S_{j,q}(x,k_m) + O(\omega\delta^3) + O(\delta^4)}{\lambda - \lambda_j + \big(\frac{1}{\mu_c} - \frac{1}{\mu_m}\big)^{-1}\tau_{j,l} + O(\delta^4) + O(\delta^2\omega^2)} + O(\delta^3). \end{aligned}$$

□

7.3.2 Asymptotic expansion of the imaginary part of the Green function

As a consequence of Theorem 7.3.1, we obtain the following result on the imaginary part of the Green function.

Theorem 7.3.2. *Assume the same conditions as in Theorem 7.3.1. Under the additional assumption that*

$$\begin{aligned} \lambda - \lambda_j + \big(\frac{1}{\mu_c} - \frac{1}{\mu_m}\big)^{-1}\tau_{j,l} &\gg O(\delta^4) + O(\delta^2\omega^2), \\ \Re\left(\lambda - \lambda_j + \big(\frac{1}{\mu_c} - \frac{1}{\mu_m}\big)^{-1}\tau_{j,l}\right) &\lesssim \Im\left(\lambda - \lambda_j + \big(\frac{1}{\mu_c} - \frac{1}{\mu_m}\big)^{-1}\tau_{j,l}\right) \end{aligned}$$

for each l and $j \in J$, we have

$$\begin{aligned} \Im\Gamma(x,x_0,k_m) &= \Im G(x,x_0,k_m) + O(\delta^3) + \\ &\quad \sum_{j\in J}\sum_{l=1}^{L} \Re\left(H_{j,p}(x_0)\widetilde{X}_{j,l,p}X_{j,l,q}S_{j,q}(x,0) + O(\omega\delta^3) + O(\delta^4)\right) \\ &\quad \times \Im\left(\frac{1}{\lambda - \lambda_j + \big(\frac{1}{\mu_c} - \frac{1}{\mu_m}\big)^{-1}\tau_{j,l}}\right), \end{aligned}$$

where $x, x_0 \in \mathrm{D}_{\delta,k_m,C}$.

Note that $\Re\left(H_{j,p}(x_0)\widetilde{X}_{j,l,p}X_{j,l,q}S_{j,q}(x,0)\right) = O(\delta^3)$. Under the conditions in Theorem 7.3.2, if we have additionally that

$$\Im\left(\frac{1}{\lambda - \lambda_j + \big(\frac{1}{\mu_c} - \frac{1}{\mu_m}\big)^{-1}\tau_{j,l}}\right) = O(\frac{1}{\delta^3})$$

for some plasmonic frequency ω, then the term in the expansion of $\Im\Gamma(x,x_0,k_m)$ which is due to resonance has size one and exhibits subwavelength peak with

width of order one. This breaks the diffraction limit $1/k_m$ in the free space. We also note that the term $\Im G(x, x_0, k_m)$ has size $O(\omega)$. Thus, we can conclude that super-resolution (super-focusing) can indeed be achieved by using a system of plasmonic particles.

7.4 Concluding remarks

In this chapter, by analyzing the imaginary part of the Green function of a medium populated by plasmonic resonators, we have shown that one can achieve super-resolution and super-focusing using plasmonic nanoparticles. We have assumed a weak interaction between nanoparticles. Results on strong interaction between plasmonic nanoparticles could be acheived using ideas of chapter 1 and [31]. Indeed, by considering a periodic arrangement of nanoparticles we could construct a high contrast media, thus allowing super-resolution.

Chapter 8

Sensing Beyond the Resolution Limit

Contents

8.1 Introduction

The inverse problem of reconstructing fine details of small objects by using far-field measurements is severally ill-posed. There are two main reasons for this. The first is the diffraction limit. When illuminated by an incident wave with wavelength Ω, the scattered field excited from the object which carries information on the scale smaller than Ω are confined near the object itself and only those with information on the scale greater than Ω can propagate into the far-field and be measured. As a result, from the far-field measurement one can only retrieve information about the object on the scale less than Ω. Especially in the case when the object is small (size smaller than Ω), one can only obtain very few information. The second is the low signal to noise ratio. We know that small objects scatter "weakly". This results very weak measurement in the far-field. In the presence of measurement noise, one has low signal to noise ratio and hence poor reconstruction. In this chapter, we propose a new methodology to overcome the ill-posedness of this inverse problem. Our method is motivated by plasmonic bio-sensing. The key is to use a plasmonic particle to interact with the object to propagate its near field information into far-field in term of shifts of plasmonic resonance frequencies.

The plasmon resonance frequency is one of the most important characterization of a plasmonic particle. It depends not only on the electromagnetic properties of the particle and its size and shape, but also the electromagnetic properties of the environment. It is the last property which enables the sensing application of plasmonic particles. Motivated by [34], we establish in this chapter a rigorous quantitative analysis for the sensing application. We show that plasmonic resonance can be used to reconstruct fine details of small objects. We also remark that plasmonic resonance can also be used to identify the shape of the plasmonic particle itself, see chapter 6.

The methodology we propose is closely related to super-resolution in imaging. Super-resolution is about the separation of point sources. In super-resolution technology near field microscopy, the basis idea is to obtain the near field of sources which contains high resolution information. This is made possible by propagating the near field information into the far field through certain near field interaction mechanism, see chapter 7. In this chapter, we are interested in reconstructing the fine details of small objects in comparison to their positions and separability which are the focus of super-resolution. The idea is similar. The near field information of the object is obtained from the near field interaction of the object and the plasmonic nanoparticle.

In this chapter we consider a system composed of a known plasmonic particle and the unknown object whose geometry and electromagnetic properties are the quantities of interest. Under the illumination of incident waves with frequencies in certain range, we observe the color of the system or measure the frequencies where the peaks in the scattering field occur. These are the resonant frequencies or spectroscopic data of the system. By varying the relative position of the two particles, we obtain different resonant frequencies due to the varying interactions between the two particles. We assume that the unknown particle is small compared to the plasmonic particle. In the intermediate regime when the distance of the two particles is comparable to the size of the plasmonic particle, we show that the presence of the small unknown particle can be viewed as a small perturbation to the homogenous environment of the plasmonic particle. As a result, it induces a

small shift to the plasmonic resonance frequencies of the plasmonic particle, which can be read from the observed spectroscopic data. By using rigorous asymptotic analysis, we obtain analytical formula for the shift which shows that the shift is determined by the generalized polarization tensors [18] of the unknown object. Therefore, from the far-field measurement of the shift of resonant frequencies, we can reconstruct the fine information of the object by using its generalized polarization tensors.

In this chapter, for the sake of simplicity, we consider the quasi-static approximation for the interaction between the electromagnetic field and the system of the two particles. Thus, we shall use the conductivity equation instead of the Helmholtz equation and the Maxwell equations. In addition, we only consider the intermediate interaction regime, the strong interaction regime when the object is close to the plasmonic particle is also very interesting and will be reported in future works.

This chapter is organized in the following way. In Section 8.2, we consider the forward scattering problem of the incident field interacting with a system composed of a normal particle and a plasmonic particle. We derive the asymptotic of the scattered field in the case of intermediate regime. In Section 8.3, we consider the inverse problem of reconstructing the geometry of the normal particle. This is done by first constructing the generalized polarization tensors of the particles through the resonance shift it induced to the plasmonic particle. In Section 8.4, we provide numerical examples to justify our theoretical results.

8.2 The forward problem

We consider a system composed of a small ordinary particle and a plasmonic particle embedded in a homogeneous medium; see Figure 8.1. The ordinary particle and the plasmonic particle occupy a bounded and simply connected domain $D_1 \subset \mathbb{R}^2$ and $D_2 \subset \mathbb{R}^2$ of class $\mathcal{C}^{1,\alpha}$ for some $0 < \alpha < 1$, respectively. We denote the permittivity of the ordinary particle D_1 (or the plasmonic particle D_2) by ε_1 (or ε_2), respectively. The permittivity of the background medium is denoted by ε_m. In other words, the permittivity distribution ε is given by

$$\varepsilon := \varepsilon_1 \chi(D_1) + \varepsilon_2 \chi(D_2) + \varepsilon_m \chi(\mathbb{R}^2 \backslash (\overline{D_1 \cup D_2})).$$

The permittivity ε_2 of the plasmonic particle depends on the operating frequency and is modeled by the Drude model as

$$\varepsilon_2 = \varepsilon_2(\omega) = 1 - \frac{\omega_p^2}{\omega(\omega + i\gamma)}.$$

We assume the following condition on the size of the particles D_1 and D_2.

Condition 8.1. *The plasmonic particle D_2 has size of order one and is centered at a position that we denote by z; the ordinary particle D_1 has size of order $\delta \ll 1$ and is centered at the origin. Specifically, we write $D_1 = \delta B$, where the domain B has size of order one.*

The total electric potential u satisfies the following equation:

$$\begin{cases} \nabla \cdot (\varepsilon \nabla u) = 0 & \text{in } \mathbb{R}^2 \backslash (\partial D_1 \cup \partial D_2), \\ u|_+ = u|_- & \text{on } \partial D_1 \cup \partial D_2, \\ \varepsilon_m \dfrac{\partial u}{\partial \nu}\Big|_+ = \varepsilon_1 \dfrac{\partial u}{\partial \nu}\Big|_- & \text{on } \partial D_1, \\ \varepsilon_m \dfrac{\partial u}{\partial \nu}\Big|_+ = \varepsilon_2 \dfrac{\partial u}{\partial \nu}\Big|_- & \text{on } \partial D_2, \\ (u - u^i)(x) = O(|x|^{-1}), & \text{as } |x| \to \infty, \end{cases} \tag{8.1}$$

where $u^i(x) = d \cdot x$ is the incident potential with a constant vector $d \in \mathbb{R}^2$.

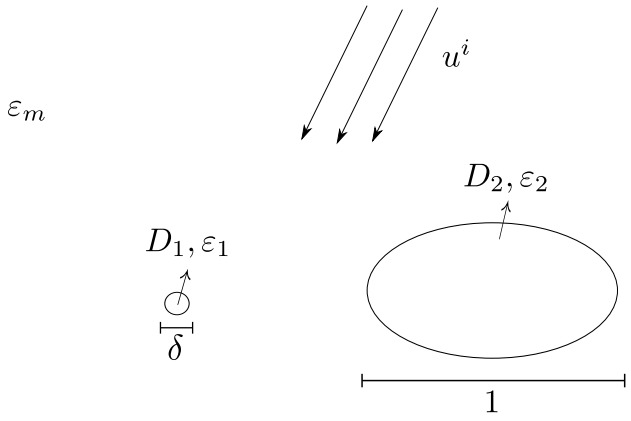

FIGURE 8.1: Scattering of an incident wave u^i by a system of a plasmonic (D_2) - non plasmonic (D_1) particles.

8.2.1 The Green function in the presence of a small particle

Let $G_{D_1}(\cdot, y)$ be the Green function at the source point y of a medium consisting of the particle D_1, which is embedded in the free space. For every $y \notin \overline{D_1}$, $G_{D_1}(\cdot, y)$ satisfies the following equation:

$$\begin{cases} \nabla \cdot \big(\varepsilon_1 \chi(D_1) + \varepsilon_m \chi(\mathbb{R}^2 \backslash \overline{D_1})\big) \nabla u = \delta_y & \text{in } \mathbb{R}^2 \backslash \partial D_1, \\ u|_+ = u|_- & \text{on } \partial D_1, \\ \varepsilon_m \dfrac{\partial u}{\partial \nu}\Big|_+ = \varepsilon_1 \dfrac{\partial u}{\partial \nu}\Big|_- & \text{on } \partial D_1, \\ u(x) = O(|x|^{-1}), & \text{as } |x| \to \infty. \end{cases} \tag{8.2}$$

We look for a solution of the form:

$$G_{D_1}(x, y) := \; G(x, y) + \mathcal{S}_{D_1}[\psi](x), \qquad x \in \mathbb{R}^2 \backslash \overline{D_1} \; . \tag{8.3}$$

Note that G_{D_1} satisfies the second and fourth conditions in (8.2). From the third condition in (8.2) and the jump formula (1.1) for the single layer

potential, the density ψ must satisfy the following equation on ∂D_1:

$$\varepsilon_m\big(\frac{1}{2}Id+\mathcal{K}_{D_1}^*\big)[\psi]+\varepsilon_1\big(\frac{1}{2}Id-\mathcal{K}_{D_1}^*\big)[\psi]=(\varepsilon_1-\varepsilon_m)\frac{\partial}{\partial\nu}G(\cdot,y). \tag{8.4}$$

So we obtain

$$\begin{aligned}\psi &= \big(\lambda_{D_1}Id-\mathcal{K}_{D_1}^*\big)^{-1}\Big[\frac{\partial}{\partial\nu}G(\cdot,y)\Big],\\ \lambda_{D_1} &= \frac{\varepsilon_1+\varepsilon_m}{2(\varepsilon_1-\varepsilon_m)}.\end{aligned}$$

Therefore, from (8.3) and the uniqueness of a solution to (8.2), we have the following representation for the Green's function G_{D_1}:

$$G_{D_1}(x,y)=G(x,y)+\mathcal{S}_{D_1}\left(\lambda_{D_1}Id-\mathcal{K}_{D_1}^*\right)^{-1}\Big[\frac{\partial}{\partial\nu}G(\cdot,y)\Big](x)\quad\text{for } x,y\in\mathbb{R}^2\backslash\overline{D_1}. \tag{8.5}$$

8.2.2 Representation of the total potential

Here we derive a layer potential representation of the total potential u, which is the solution to (8.1).

Let u_{D_1} be the total field resulting from the incident field u^i and the ordinary particle D_1 (without the plasmonic particle D_2). Note that u_{D_1} is given by

$$u_{D_1}(x)=u^i(x)+\mathcal{S}_{D_1}\left(\lambda_{D_1}Id-\mathcal{K}_{D_1}^*\right)^{-1}[\frac{\partial u^i}{\partial\nu_1}](x),\qquad\text{for } x\in\mathbb{R}^2\backslash\overline{D_1}.$$

To consider the total potential u, we also need to represent the field generated by the plasmonic particle D_2. For this, we introduce a new layer potential $\mathcal{S}_{D_2,D_1}$ as follows:

$$\mathcal{S}_{D_2,D_1}[\varphi](x)=\int_{\partial D_2}G_{D_1}(x,y)\varphi(y)d\sigma(y).$$

The total potential u can be represented in the following form:

$$u(x)=u_{D_1}(x)+\mathcal{S}_{D_2,D_1}[\psi](x),\quad x\in\mathbb{R}^2\backslash\overline{D_2}. \tag{8.6}$$

We need to find a boundary integral equation for the density ψ. It follows from (8.5) that, for any φ,

$$\mathcal{S}_{D_2,D_1}[\varphi](x)=\mathcal{S}_{D_2}[\varphi](x)+\mathcal{S}^1_{D_2,D_1}[\varphi](x),$$

where $\mathcal{S}^1_{D_2,D_1}$ is given by

$$\mathcal{S}^1_{D_2,D_1}[\varphi](x):=\int_{\partial D_2}\mathcal{S}_{D_1}\left(\lambda_{D_1}Id-\mathcal{K}_{D_1}^*\right)^{-1}[\frac{\partial}{\partial\nu_1}G(\cdot,y)](x)\varphi(y)d\sigma(y).$$

The expression of $\mathcal{S}^1_{D_2,D_1}[\varphi]$ can be further developed using the following spectral expansion of the free-space Green function $G(x,y)$ [32]:

$$G(x,y) = -\sum_{j=1}^{\infty} \mathcal{S}_{D_2}[\varphi_j](x)\mathcal{S}_{D_2}[\varphi_j](y) + \mathcal{S}_{D_2}[\varphi_0](x), \quad \text{for } x \in \mathbb{R}^2\backslash\overline{D_2} \text{ and } y \in \overline{D_2},$$

where $\varphi_j, j = 1, 2, ...$ are eigenfunctions of $\mathcal{K}^*_{D_2}$ on $\mathcal{H}^*(\partial D_2)$ and φ_0 is an eigenfunction associated to the eigenvalue $1/2$. Then, for any $\varphi \in \mathcal{H}^*(\partial D_2)$, we get

$$\begin{aligned}\int_{\partial D_2} G(x,y)\varphi(y)d\sigma(y) &= \sum_{j=1}^{\infty} \mathcal{S}_{D_2}[\varphi_j](x)(\varphi,\varphi_j)_{\mathcal{H}^*(\partial D_2)} + \mathcal{S}_{D_2}[\varphi_0](x)\int_{\partial D_2}\varphi(y)d\sigma(y)\\ &= \sum_{j=1}^{\infty} \mathcal{S}_{D_2}[\varphi_j](x)(\varphi,\varphi_j)_{\mathcal{H}^*(\partial D_2)}.\end{aligned}$$

Therefore, for any $\varphi \in \mathcal{H}^*(\partial D_2)$, we have,

$$\begin{aligned}\mathcal{S}^1_{D_2,D_1}[\varphi](x) &= \int_{\partial D_2} \mathcal{S}_{D_1}\left(\lambda_{D_1} Id - \mathcal{K}^*_{D_1}\right)^{-1}\left[\frac{\partial}{\partial\nu_1}G(\cdot,y)\right](x)\varphi(y)d\sigma(y)\\ &= \mathcal{S}_{D_1}\left(\lambda_{D_1} Id - \mathcal{K}^*_{D_1}\right)^{-1}\frac{\partial}{\partial\nu_1}\mathcal{S}_{D_2}\Big[\sum_{j=0}^{\infty}(\varphi,\varphi_j)_{\mathcal{H}^*}\varphi_j\Big](x)\\ &= \mathcal{S}_{D_1}\left(\lambda_{D_1} Id - \mathcal{K}^*_{D_1}\right)^{-1}\frac{\partial\mathcal{S}_{D_2}[\varphi]}{\partial\nu_1}(x),\end{aligned}$$

where we have used the notation $\frac{\partial}{\partial\nu_i}$ to indicate the outward normal derivative on ∂D_i.

Combining the boundary conditions in (8.1), the representation formula (8.6) and the jump formula (1.1) yields the following equation for ψ

$$\left(\mathcal{A}_{D_2,0} + \mathcal{A}_{D_2,1}\right)[\psi] = \frac{\partial u_{D_1}}{\partial\nu_2},$$

where

$$\begin{aligned}\mathcal{A}_{D_2,0} &= \lambda_{D_2} Id - \mathcal{K}^*_{D_2},\\ \lambda_{D_2} &= \frac{\varepsilon_2 + \varepsilon_m}{2(\varepsilon_2 - \varepsilon_m)}, \qquad (8.7)\\ \mathcal{A}_{D_2,1} &= \frac{\partial\mathcal{S}^1_{D_2,D_1}}{\partial\nu_2} = \frac{\partial}{\partial\nu_2}\mathcal{S}_{D_1}\left(\lambda_{D_1} Id - \mathcal{K}^*_{D_1}\right)^{-1}\frac{\partial\mathcal{S}_{D_2}}{\partial\nu_1}. \qquad (8.8)\end{aligned}$$

8.2.3 Intermediate regime and asymptotic expansion of the scattered field

Here we introduce the concept of intermediate regime and derive the asymptotic expansion of the scattered field $u - u^i$ for small δ.

Definition 8.1 (Intermediate regime). *We say that D_2 is in the intermediate regime with respect to the origin if there exist positive constants C_1 and C_2 such that $C_1 < C_2$ and*

$$C_1 \leq \text{dist}(0, D_2) \leq C_2.$$

Definition 8.1 says that the plasmonic particle D_2 is located not too close to D_1 nor far from D_1. Throughout this chapter, we assume the plasmonic particle D_2 is in the intermediate regime. We have the following result.

Propsition 8.2.1. *If D_2 is in the intermediate regime, then $\|\mathcal{A}_{D_2,1}\|_{\mathcal{H}*} = O(\delta^2)$ as $\delta \to 0$.*

Proof. Fix $\varphi \in \mathcal{H}^*(\partial D_2)$ and let

$$\widetilde{\varphi} := (\lambda_{D_1} Id - \mathcal{K}^*_{D_1})^{-1}\Big[\frac{\partial \mathcal{S}_{D_2}[\varphi]}{\partial \nu_1}\Big].$$

Since $\mathcal{S}_{D_2}[\varphi]$ is harmonic in D_1, the Green's identity gives $\int_{\partial D_1} \frac{\partial}{\partial \nu_1}\mathcal{S}_{D_2}[\varphi] = 0$. Then it can be proved that $\int_{\partial D_1} \widetilde{\varphi} = 0$. So we get

$$\begin{aligned}\mathcal{S}_{D_1}[\widetilde{\varphi}](x) &= \int_{\partial D_1} (\log|x-y| - \log|x|)\widetilde{\varphi}(y)d\sigma(y) + \log|x| \int_{\partial D_1} \widetilde{\varphi}(y)d\sigma(y) \\ &= \int_{\partial D_1} (\log|x-y| - \log|x|)\widetilde{\varphi}(y)d\sigma(y).\end{aligned}$$

Therefore, since $|y-x| \geq C'$ and $|y| \leq C\delta$ for $(y,x) \in (\partial D_1, \partial D_2)$, we obtain

$$\|\mathcal{A}_{D_2,1}[\varphi]\|_{\mathcal{H}^*(\partial D_2)} = \big\|\frac{\partial}{\partial \nu_2}\mathcal{S}_{D_1}[\widetilde{\varphi}]\big\|_{\mathcal{H}^*(\partial D_2)} \leq C\delta \|\widetilde{\varphi}\|_{\mathcal{H}^*(\partial D_1)}.$$

Now it suffices to prove that

$$\|\widetilde{\varphi}\|_{\mathcal{H}^*(\partial D_1)} \leq C\delta. \tag{8.9}$$

Recall that $D_1 = \delta B$. Let $f_\delta(y) = f(\delta y)$. Then the function f_δ belongs to $\mathcal{H}^*(\partial B)$ for $f \in \mathcal{H}^*(\partial D_1)$. Since it is known that $\mathcal{K}^*_\Omega$ is scale-invariant for any Ω, we have $\mathcal{K}^*_{D_1}[f] = \mathcal{K}^*_B[f_\delta]$. Therefore,

$$\widetilde{\varphi} = \big(\lambda_{D_1} Id - \mathcal{K}^*_{D_1}\big)^{-1}[f]d(\delta\sigma(y)) = (\lambda_{D_1} Id - \mathcal{K}^*_B)^{-1}[f_\delta]d(\delta\sigma(y)).$$

Again, since $|y-x| \geq C'$ for $(y,x) \in (\partial D_1, \partial D_2)$ and $|\partial D_1| = O(\delta)$, we arrive at

$$\begin{aligned}\|\widetilde{\varphi}\|_{\mathcal{H}^*(\partial D_1)} &= \|\,(\lambda_{D_1} Id - \mathcal{K}^*_B)^{-1}\Big[\Big(\frac{\partial \mathcal{S}_{D_2}[\varphi]}{\partial \nu_1}\Big)_\delta\Big]\|_{\mathcal{H}^*(\partial B)} \\ &\leq C\|\frac{\partial}{\partial \nu_1}\mathcal{S}_{D_2}[\varphi]\|_{\mathcal{H}^*(\partial D_1)} \leq C\delta.\end{aligned}$$

The proof is completed. □

From Proposition 8.2.1, we can view $\mathcal{A}_{D_2,1}$ as a perturbation of $\mathcal{A}_{D_2,0}$. Using standard perturbation theory [85], we can derive the perturbed eigenvalues and associated eigenfunctions.

Let λ_j and φ_j be the eigenvalues and eigenfunctions of $\mathcal{K}^*_{D_2}$ on $\mathcal{H}^*(\partial D_2)$. For simplicity, we consider the case when λ_j is a simple eigenvalue of the operator $\mathcal{K}^*_{D_2}$. Let us define

$$R_{jl} = \big(\mathcal{A}_{D_2,1}[\varphi_l], \varphi_j\big)_{\mathcal{H}^*(\partial D_2)}, \tag{8.10}$$

where $\mathcal{A}_{D_2,1}$ is given by (8.8). Note that $R_{jl} = O(\delta^2)$.

The perturbed eigenvalues have the following form:

$$\tau_j(\delta) = \lambda_{D_2} - \lambda_j + \mathcal{P}_j,$$

where $\mathcal{P}_j$ are given by

$$\mathcal{P}_j = R_{jj} + \sum_{l\neq j} \frac{R_{jl}R_{lj}}{\lambda_j - \lambda_l} + \sum_{(l_1,l_2)\neq j} \frac{R_{jl_2}R_{l_2l_1}R_{l_1j}}{(\lambda_j - \lambda_{l_1})(\lambda_j - \lambda_{l_2})} + \sum_{(l_1,l_2,l_3)\neq j} \frac{R_{jl_3}R_{l_3l_2}R_{l_2l_1}R_{l_1j}}{(\lambda_j - \lambda_{l_1})(\lambda_j - \lambda_{l_2})(\lambda_j - \lambda_{l_3})} + \cdots . \tag{8.11}$$

Also, the perturbed eigenfunctions have the following form:

$$\varphi_j(\delta) = \varphi_j + O(\delta^2). \tag{8.12}$$

Here the remainder term is with respect to the norm $\|\cdot\|_{\mathcal{H}^*(\partial D_2)}$.

Remark 8.2.1. *Note that $\mathcal{P}_j$ depends not only on the geometry and material properties of D_1, but also on D_2's properties, in particular its position z.*

Theorem 8.2.1. *If D_2 is in the intermediate regime, the scattered field $u^s_{D_2} = u - u_{D_1}$ by the plasmonic particle D_2 has the following representation:*

$$u^s_{D_2} = \mathcal{S}_{D_2,D_1}[\psi],$$

where ψ satisfies

$$\psi = \sum_{j=1}^{\infty} \frac{\left(\nabla u^i(z)\cdot\nu, \varphi_j\right)_{\mathcal{H}^*(\partial D_2)} \varphi_j + O(\delta^2)}{\lambda_{D_2} - \lambda_j + \mathcal{P}_j}$$

with λ_{D_2} being given by (8.7).

As a corollary, we have the following asymptotic expansion of the scattered field $u - u^i$.

Theorem 8.2.2. *We have the following far field expantion:*

$$(u-u^i)(x) = \nabla u^i(z)\cdot M(\lambda_{D_1},\lambda_{D_2},D_1,D_2)\nabla G(x,z) + O(\delta^2) + O\left(\frac{\delta^3}{\operatorname{dist}(\lambda_{D_2},\sigma(\mathcal{K}^*_{D_2}))}\right),$$

as $|x|\to\infty$. Here, $M(\lambda_{D_1},\lambda_{D_2},D_1,D_2)$ is the polarization tensor satisfying

$$M(\lambda_{D_1},\lambda_{D_2},D_1,D_2)_{l,m} = \sum_{j=1}^{\infty} \frac{(\nu_l,\varphi_j)_{\mathcal{H}^*(\partial D_2)}(\varphi_j,x_m)_{-\frac{1}{2},\frac{1}{2}} + O(\delta^2)}{\lambda_{D_2} - \lambda_j + \mathcal{P}_j}, \tag{8.13}$$

for $l,m = 1,2$.

We remark that the scattered field in the above expression depends on the frequency (since λ_{D_2} does so) and exhibit local peaks at certain frequencies when one of the denominators is close to zero and is minimized while the associated nominator is not zero. These frequencies are called the resonant frequencies of the system. It is clear that these resonant frequencies also depend on the geometry and the electric permittivity of D_1 through the

perturbative terms $\mathcal{P}_j$'s. We shall use this fact in the next section to solve the associated inverse problem of reconstructing D_1 by using those frequencies.

8.2.4 Representation of the shift $\mathcal{P}_j$ using CGPTs

Here we show that the term $\mathcal{P}_j$ in the plasmonic resonances can be expressed in terms of the contracted generalized polarization tensors (CGPTs), see chapter 1. The CGPTs carry information on the geometry and material properties of D_1. See [18] for a detailed reference. We shall reconstruct the ordinary particle D_1 from the measurement of the shift $\mathcal{P}_j$.

Propsition 8.2.2. *If D_2 is in the intermediate regime, then the perturbative terms R_{jl} can be represented using CGPTs $M_{m,n}(\lambda_{D_1}, D_1)$ associated with D_1 as follows:*

$$R_{jl} = \left(\frac{1}{2} - \lambda_j\right) \sum_{m=1}^{M} \sum_{n=1}^{N} a_m^j M_{m,n}(\lambda_{D_1}, D_1)(a_n^l)^t + O(\delta^{M+N+1}), \tag{8.14}$$

where the superscript t denotes the transpose and $a_m^j = (a_{m,c}^j, a_{m,s}^j)$ with

$$\begin{aligned} a_{m,c}^j &= -\frac{1}{2\pi m} \int_{\partial D_2} \frac{\cos(m\theta_y)}{r_y^m} \varphi_j(y) d\sigma(y), \\ a_{m,s}^j &= -\frac{1}{2\pi m} \int_{\partial D_2} \frac{\sin(m\theta_y)}{r_y^m} \varphi_j(y) d\sigma(y). \end{aligned}$$

Here, (r_y, θ_y) denote the polar coordinates of y and $\{\varphi_j\}_j$ is an orthonormal basis of eigenfunctions of $\mathcal{K}_{D_2}^*$ on $\mathcal{H}^*$.

Proof. To simplify the notation, let us denote

$$F_l = \mathcal{S}_{D_1} \left(\lambda_{D_1} Id - \mathcal{K}_{D_1}^*\right)^{-1} \frac{\partial \mathcal{S}_{D_2}[\varphi_l]}{\partial \nu_1}.$$

Then, from the Green's identity and the jump formula (1.1), we obtain

$$\begin{aligned} R_{jl} &= \left(F_l, \varphi_j\right)_{\mathcal{H}^*} = -\left(\frac{\partial F_l}{\partial \nu_2}, \mathcal{S}_{D_2}[\varphi_j]\right)_{\frac{1}{2},-\frac{1}{2}} \\ &= -\left(F_l, \frac{\partial \mathcal{S}_{D_2}[\varphi_j]}{\partial \nu_2}\bigg|_{-}\right)_{\frac{1}{2},-\frac{1}{2}} = -\left(F_l, (-\frac{1}{2} + \mathcal{K}_{D_2}^*)[\varphi_j]\right)_{\frac{1}{2},-\frac{1}{2}}. \end{aligned}$$

Since φ_j is an eigenfunction of $\mathcal{K}_{D_2}^*$ with an eigenvalue λ_j, we have

$$R_{jl} = \left(\frac{1}{2} - \lambda_j\right)\left(F_l, \varphi_j\right)_{\frac{1}{2},-\frac{1}{2}}.$$

Let (r_x, θ_x) be the polar coordinates of x. It is known from [12] that, for $|x| < |y|$,

$$G(x,y) = \sum_{n=0}^{\infty} \frac{(-1)}{2\pi n} \frac{\cos(n\theta_y)}{r_y^n} r_x^n \cos(n\theta_x) + \frac{(-1)}{2\pi n} \frac{\sin(n\theta_y)}{r_y^n} r_x^n \sin(n\theta_x). \tag{8.15}$$

By interchanging x and y and the fact that $G(x,y) = G(y,x)$, we have, for $|x| > |y|$,

$$G(x,y) = \sum_{n=0}^{\infty} \frac{(-1)}{2\pi n} \frac{\cos(n\theta_x)}{r_x^n} r_y^n \cos(n\theta_y) + \frac{(-1)}{2\pi n} \frac{\sin(n\theta_x)}{r_x^n} r_y^n \sin(n\theta_y). \quad (8.16)$$

If $x \in \partial D_1$ and $y \in \partial D_2$, then $|x| < |y|$. So, applying (8.15) gives

$$\begin{aligned} \frac{\partial \mathcal{S}_{D_2}[\varphi_l]}{\partial \nu_1}(x) &= \frac{\partial}{\partial \nu_1} \int_{\partial D_2} G(x,y)\varphi_l d\sigma(y) \\ &= \sum_{n=1}^{\infty} \frac{\partial r_x^n \cos(n\theta_x)}{\partial \nu_1} a_{n,c}^l + \frac{\partial r_x^n \sin(n\theta_x)}{\partial \nu_1} a_{n,s}^l. \end{aligned}$$

On the contrary, if $y \in \partial D_1$ and $x \in \partial D_2$, then $|x| > |y|$. We have from (8.16) that, for any f,

$$\begin{aligned} \mathcal{S}_{D_1}[f](x) &= \int_{\partial D_1} G(x,y)[f](y) d\sigma(y) \\ &= \sum_{m=0}^{\infty} -\frac{1}{2\pi m} \frac{\cos(m\theta_x)}{r_x^m} \int_{\partial D_1} r_y^m \cos(m\theta_y)[f](y) d\sigma(y) \\ &\quad + \sum_{m=0}^{\infty} -\frac{1}{2\pi m} \frac{\sin(m\theta_x)}{r_x^m} \int_{\partial D_1} r_y^m \sin(m\theta_y)[f](y) d\sigma(y). \end{aligned}$$

Therefore, from the definition of $M_{m,n}$, we get

$$\begin{aligned} R_{jl} &= \left(\frac{1}{2} - \lambda_j\right) \left(\mathcal{S}_{D_1} \left(\lambda_{D_1} Id - \mathcal{K}_{D_1}^*\right)^{-1} \frac{\partial \mathcal{S}_{D_2}[\varphi_l]}{\partial \nu_1}, \varphi_j\right)_{\frac{1}{2}, -\frac{1}{2}} \\ &= \left(\frac{1}{2} - \lambda_j\right) \sum_{m=0,n=1}^{\infty} (a_{m,c}^j, a_{m,s}^j) M_{m,n}(\lambda_{D_1}, D_1)(a_{n,c}^l, a_{n,s}^l)^t. \end{aligned}$$

For any $\lambda \in \mathbb{C}$ and $D = \delta B$, it is easy to check that $M_{m,n}(\lambda, D) = \delta^{m+n} M_{m,n}(\lambda, B)$. Since D_2 is in the intermediate regime, $a_{n,c}^l$ and $a_{n,s}^l$ satisfy

$$|a_{m,c}^j|, |a_{m,s}^j| \leq \frac{1}{m} C^{-m}, \quad |a_{n,c}^l|, |a_{n,s}^l| \leq \frac{1}{n} C^{-n},$$

for some constant $C > 1$ independent of δ. Moreover, it can be shown that (see [15])

$$\sum_{n=1}^{\infty} (a_{0,c}^j, a_{0,s}^j) M_{0,n}(\lambda_{D_1}, D_1)(a_{n,c}^l, a_{n,s}^l)^t = 0.$$

Then the conclusion immediately follows. □

Corollary 8.2.1. *We have*

$$\begin{aligned} &\mathcal{P}_j(z) - \sum_{l \neq j} \frac{R_{jl}(z) R_{lj}(z)}{\lambda_j - \lambda_l} - \sum_{(l_1,l_2) \neq j} \frac{R_{jl_2} R_{l_2 l_1} R_{l_1 j}}{(\lambda_j - \lambda_{l_1})(\lambda_j - \lambda_{l_2})} \cdots \\ &= \left(\frac{1}{2} - \lambda_j\right) \sum_{m=1}^{M} \sum_{n=1}^{N} a_m^j M_{m,n}(\lambda_{D_1}, D_1)(a_n^l)^t + O(\delta^{M+N+1}). \end{aligned}$$

In the LHS, the summation should be truncated so that all the terms which contain $R_{jl_k}\cdots R_{l_kj} = O(\delta^{2(k+1)})$ *with* $2(k+1) \leq M+N+1$ *are ignored.*

8.3 The inverse problem

In this section, we consider the inverse problem associated with the forward system (8.1). We assume that the plasmonic particle D_2 is known, *i.e.*, we know its electric permittivity $\varepsilon_2 = \varepsilon_2(\omega)$, its shape D_2 and position z. The ordinary particle D_1 is unknown. For simplicity, we assume that its permittivity ε_1 is known. For each of many different positions z of the plasmonic particle D_2, we measure the resonant frequency and use these resonant frequencies to reconstruct the shape of the ordinary particle D_1.

As illustrated by Theorem 8.2.2, the resonance in the scattered field occurs when $\lambda_{D_2}(\omega) - \lambda_j + \mathcal{P}_j$ is minimized and $(\nu_l, \varphi_j)_{\mathcal{H}^*}(\varphi_j, x_m)_{-\frac{1}{2},\frac{1}{2}} \neq 0$. So by varying the frequency ω, we can measure the value of $\lambda_j - \mathcal{P}_j$. Moreover, in the absence of the ordinary particle, the resonance occurs when $\lambda_{D_2}(\omega) - \lambda_j$ is minimized and $(\nu_l, \varphi_j)_{\mathcal{H}^*}(\varphi_j, x_m)_{-\frac{1}{2},\frac{1}{2}} \neq 0$. Since we assume that the plasmonic particle D_2 is known, we can get the value of λ_j a priori. Therefore, by comparing $\lambda_j - \mathcal{P}_j$ and λ_j, we can measure the shift $\mathcal{P}_j$ of the eigenvalue.

Finding $\mathcal{P}_j$ for many different positions of D_2 will yield a linear system of equations that will allow the recovery of the CGPTs associated with D_1. From the recovered CGPTs, we will reconstruct the ordinary particle D_1. Here, we only consider the shape reconstruction problem. Nevertheless, by using the CGPTs associated with D_1, it is possible to reconstruct the permittivity ε_1 of D_1 in the case it is not a priori given [12].

From now on, we denote $M_{m,n} = M_{m,n}(\lambda_{D_1}, D_1)$.

8.3.1 CGPTs recovery algorithm

We propose a recurrent algorithm to recover the GPTs of order less or equal to k up to an order δ^{2k-1}, using measurements of P_j at different positions of D_2. For simplicity, we only consider the shift of a single eigenvalue λ_j with a fixed j. To gain robustness and efficiency, the shift in other resonant frequencies could also be considered.

We now explain our method for reconstructing GPTs $M_{m,n}, m+n \leq K$ for a given $K \in \mathbb{N}$ from the measurements of the shift $\mathcal{P}_j$.

Suppose we measure precisely $\mathcal{P}_j$ for three different positions z_1, z_2, z_3 of the plasmonic particle D_2. First we reconstruct $M_{1,1}$ approximately. Since $M_{1,1}^t = M_{1,1}$, the matrix $M_{1,1}$ is symmetric. We look for a symmetric matrix $M_{1,1}^{(2)}$ satisfying

$$\begin{aligned}
\mathcal{P}_j(z_1) &= \left(\frac{1}{2} - \lambda_j\right) a_1^j(z_1) M_{1,1}^{(2)} (a_1^j)^t(z_1) \\
\mathcal{P}_j(z_2) &= \left(\frac{1}{2} - \lambda_j\right) a_1^j(z_2) M_{1,1}^{(2)} (a_1^j)^t(z_2) \\
\mathcal{P}_j(z_3) &= \left(\frac{1}{2} - \lambda_j\right) a_1^j(z_3) M_{1,1}^{(2)} (a_1^j)^t(z_3).
\end{aligned}$$

The above equations can be seen as a linear system of equations for three independent components $(M_{1,1}^{(2)})_{11}, (M_{1,1}^{(2)})_{12}$ and $(M_{1,1}^{(2)})_{22}$. We emphasize

that $a_m^j(z_i)$ can be a priori given because the particle D_2 is known. Since, from Corollary 8.2.1 and the fact that $R_{jl} = O(\delta^2)$, we have

$$\mathcal{P}_j(z_k) = \left(\frac{1}{2} - \lambda_j\right) a_1^j(z_k) M_{1,1} (a_1^j)^t(z_k) + O(\delta^3), \quad k = 1, 2, 3,$$

we see that $M_{1,1}$ is well approximated by $M_{1,1}^{(2)}$. Specifically, we have $M_{1,1} - M_{1,1}^{(2)} = O(\delta^3)$.

Next we reconstruct and update the higher order GPTs $M_{n,m}$ in a recursive way. Towards this, we need more measurement data of the shift $\mathcal{P}_j$. Let $k \geq 3$. Due to the symmetry of harmonic combinations of the non contracted GPTs (see [18]), we have $M_{m,n} = M_{n,m}^t$. One can see that, by using this symmetry property, the set of GPTs $M_{m,n}$ satisfying $m + n \leq k$ contains e_k independent variables where e_k is given by

$$e_k = \begin{cases} k(k-1) + k/2, & \text{if } k \text{ is even,} \\ k(k-1) + (k-1)/2, & \text{if } k \text{ is odd.} \end{cases}$$

Therefore, we need e_k measurement data for $\mathcal{P}_j$ to reconstruct the GPTs $M_{m,n}$ for $m + n \leq k$.

Suppose we have $e_k - 2$ more measurement data $\mathcal{P}_j$ at different positions $z_4, z_5, ..., z_{e_k}$. Let $\{M_{m,n}^{(k)}\}_{m+n\leq k}$ be the set of matrices satisfying $[M_{n,m}^{(k)}]^t = M_{m,n}^{(k)}$ and the following linear system:

$$\begin{aligned}
\widetilde{\mathcal{P}}_j^{(k-1)}(z_1) &= \left(\frac{1}{2} - \lambda_j\right) \sum_{m+n\leq k} a_m^j(z_1) M_{m,n}^{(k)} (a_n^j)^t(z_1) \\
\widetilde{\mathcal{P}}_j^{(k-1)}(z_2) &= \left(\frac{1}{2} - \lambda_j\right) \sum_{m+n\leq k} a_m^j(z_2) M_{m,n}^{(k)} (a_n^j)^t(z_2) \\
\vdots \quad &= \qquad\qquad \vdots \\
\widetilde{\mathcal{P}}_j^{(k-1)}(z_{e_k}) &= \left(\frac{1}{2} - \lambda_j\right) \sum_{m+n\leq k} a_m^j(z_{e_k}) M_{m,n}^{(k)} (a_n^j)^t(z_{e_k}),
\end{aligned} \tag{8.17}$$

where

$$\widetilde{\mathcal{P}}_j^{(k-1)}(z_i) := \mathcal{P}_j(z_i) - \sum_{l\neq j} \frac{R_{jl}^{(k-1)}(z_i) R_{lj}^{(k-1)}(z_i)}{\lambda_j - \lambda_l} - \ldots, \quad i = 1, 2, ..., e_k, \tag{8.18}$$

and

$$R_{jl}^{(k-1)}(z) := \left(\frac{1}{2} - \lambda_j\right) \sum_{m+n\leq k-1} a_m^j(z) M_{m,n}^{(k-1)} (a_n^l)^t(z).$$

Note that $M_{m,n}^{(k)}$ are defined recursively. In (8.18), the summation should be truncated as in Corollary 8.2.1.

Then $M_{m,n}^{(k)}$ becomes a good approximation of the CGPT $M_{m,n}$ for $m + n \leq k$. Moreover, the accuracy improves as the iteration goes on. Indeed, we can see that

$$M_{m,n} - M_{m,n}^{(k)} = O(\delta^{2k-1}), \quad m + n \leq k. \tag{8.19}$$

In fact, (8.19) can be verified by induction. We already know that this is true when $k = 2$. Let us assume $M_{m,n} - M_{m,n}^{(k-1)} = O(\delta^{2k-3})$, $m+n \leq k-1$. Then, from Proposition 8.2.2, we have

$$R_{jl}(z) - R_{jl}^{(k-1)}(z) \quad = \quad O(\delta^{2k-3}).$$

Hence, from Corollary 8.2.1 and the fact that $R_{jl} = O(\delta^2)$, we obtain

$$\widetilde{\mathcal{P}}_j^{(k-1)}(z_i) - \left(\mathcal{P}_j(z_i) - \sum_{l \neq j} \frac{R_{jl}(z_i) R_{lj}(z_i)}{\lambda_j - \lambda_l} - \cdots \right) = O(\delta^{2k-1}).$$

Therefore, in view of Corollary 8.2.1 and the linear system (8.17), we obtain (8.19). In conclusion, $M_{m,n}^{(k)}$ is indeed precise up to an order δ^{2k-1}.

Remark 8.3.1. *In practice, $\mathcal{P}_j$ might be subject to noise and could not be measured precisely. In this case only the low order CGPTs could be recovered.*

8.3.2 Shape recovery from CGPTs

To recover the shape of D_1 from its CGPTs, we search to minimize the following shape functional ([12])

$$\mathcal{J}_c^{(l)}[B] := \frac{1}{2} \sum_{n+m \leq k} \left| N_{mn}^{(1)}(\lambda_{D_1}, B) - N_{mn}^{(1)}(\lambda_{D_1}, D_1) \right|^2 , \tag{8.20}$$

where

$$N_{m,n}^{(1)}(\lambda, D) = (M_{m,n}^{cc} - M_{m,n}^{ss}) + i(M_{m,n}^{cs} - M_{m,n}^{sc}).$$

To minimize $\mathcal{J}^{(l)}[B]$ we need to compute the shape derivative, $d_S \mathcal{J}_c^{(l)}$, of $\mathcal{J}_c^{(l)}$.

For ϵ small, let B_ϵ be an ϵ-deformation of B, *i.e.*, there is a scalar function $h \in \mathcal{C}^1(\partial B)$, such that

$$\partial B_\epsilon := \{x + \epsilon h(x)\nu(x) \ : x \in \partial B\}.$$

Then, according to [11, 12, 17], the perturbation of a harmonic sum of GPTs due to the shape deformation is given as follows:

$$N_{m,n}^{(1)}(\lambda_{D_1}, B_\epsilon) - N_{m,n}^{(1)}(\lambda_{D_1}, D_1)$$

$$= \epsilon(k_{\lambda_{D_1}} - 1) \int_{\partial B} h(x) \left[\frac{\partial u}{\partial \nu}\Big|_- \frac{\partial v}{\partial \nu}\Big|_- + \frac{1}{k_{\lambda_{D_1}}} \frac{\partial u}{\partial T}\Big|_- \frac{\partial v}{\partial T}\Big|_- \right](x)\, d\sigma(x) + O(\epsilon^2),$$

where

$$k_{\lambda_{D_1}} = (2\lambda_{D_1} + 1)/(2\lambda_{D_1} - 1), \tag{8.21}$$

and u and v are respectively the solutions to the problems:

$$\begin{cases} \Delta u = 0 & \text{in } B \cup (\mathbb{R}^2 \backslash \overline{B}) \,, \\ u|_+ - u|_- = 0 & \text{on } \partial B \,, \\ \dfrac{\partial u}{\partial \nu}\Big|_+ - k_{\lambda_{D_1}} \dfrac{\partial u}{\partial \nu}\Big|_- = 0 & \text{on } \partial B \,, \\ (u - (x_1 + ix_2)^m)(x) = O(|x|^{-1}) & \text{as } |x| \to \infty \,, \end{cases} \tag{8.22}$$

and

$$\begin{cases} \Delta v = 0 & \text{in } B \cup (\mathbb{R}^2 \backslash \overline{B}) \,, \\ k_{\lambda_{D_1}} v|_+ - v|_- = 0 & \text{on } \partial B \,, \\ \dfrac{\partial v}{\partial \nu}\Big|_+ - \dfrac{\partial v}{\partial \nu}\Big|_- = 0 & \text{on } \partial B \,, \\ (v - (x_1 + ix_2)^n)(x) = O(|x|^{-1}) & \text{as } |x| \to \infty \,. \end{cases} \tag{8.23}$$

Here, $\partial/\partial T$ is the tangential derivative.

Let

$$w_{m,n}(x) = (k_{\lambda_{D_1}} - 1) \left[\frac{\partial u}{\partial \nu}\Big|_- \frac{\partial v}{\partial \nu}\Big|_- + \frac{1}{k_{\lambda_{D_1}}} \frac{\partial u}{\partial T}\Big|_- \frac{\partial v}{\partial T}\Big|_- \right](x), \quad x \in \partial B \,.$$

The shape derivative of $\mathcal{J}_c^{(l)}$ at B in the direction of h is given by

$$\langle d_S \mathcal{J}_c^{(l)}[B], h \rangle = \sum_{m+n \le k} \delta_N \langle w_{m,n}, h \rangle_{L^2(\partial B)} \,,$$

where

$$\delta_N = N_{m,n}^{(1)}(\lambda_{D_1}, B) - N_{m,n}^{(1)}(\lambda_{D_1}, D_1) \,.$$

Next, using a gradient descent algorithm we can minimize, at least locally, the functional $\mathcal{J}_c^{(l)}$.

8.4 Numerical Illustrations

In this section, we support our theoretical results by numerical examples. In the sequel, we assume that D_2 is an ellipse with semi-axes $a = 1$ and $b = 2$, as shown in Figure 8.2. In this case the resonances in the far-field can only occur at $\lambda_1 = \frac{1}{2}\frac{a-b}{a+b} = -\frac{1}{6}$ and $\lambda_2 = -\frac{1}{2}\frac{a-b}{a+b} = \frac{1}{6}$. Thus, for a fixed position of D_2, we can measure two shifts of the plasmonic resonance: $\mathcal{P}_1$ and $\mathcal{P}_2$.

We consider the case of D_1 being a triangular-shaped and a rectangular-shaped particle with known contrast $\lambda_{D_1} = 1$, as shown in Figure 8.3.

Figure 8.4 shows the shift in the plasmonic resonance around λ_1, for random positions of D_2 around a triangular-shaped particle D_1. From these measurements, $\mathcal{P}_1$ can be precisely estimated from the resonance peaks and the equation $\mathcal{P}_j = \lambda_j - \lambda_r$, where λ_r is the value at which we achieve the maximum of the resonant peak.

It is worth mentioning that, for the sake of simplicity and clarity, we plot the graph not by varying the frequency but the parameter λ directly. We assume $\mathrm{Re}(\lambda_{D_2})$ ranges from $-1/2$ to $1/2$ and $\mathrm{Im}(\lambda_{D_2}) = 10^{-4}$. In a more

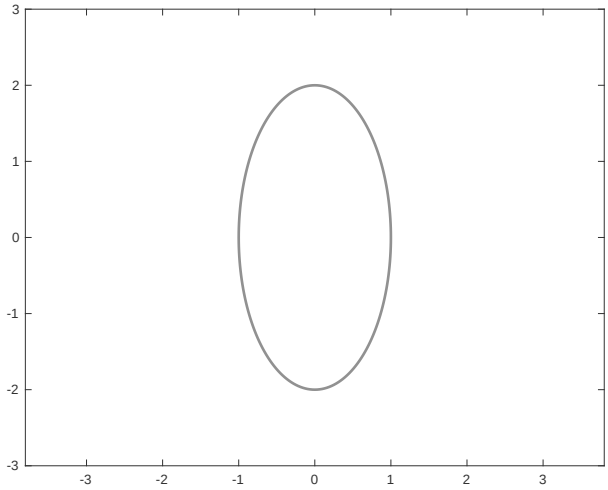

FIGURE 8.2: Plasmonic particle D_2.

realistic setting, corrections in the peaks of resonances should be included, by considering the Drude model for λ_{D_2}. But they are essentially equivalent.

To recover geometrical properties of D_1 from measurements of $\mathcal{P}_1$, we recover the CGPTs using the algorithm described in 8.3.1 and then minimize functional (8.20) to reconstruct an approximation of D_1.

To recover the first CGPTs of order 5 or less we make 22 measurements around D_1 as shown in Figure 8.5, and measure the shift from $\lambda_1 = -\frac{1}{6}$.

In the following we show a comparison between the recovered CGPTs of order less or equal to 4 and their theoretical value, for each iteration.

Triangle-shaped D_1:

Theoretical values:

$$M_{11} = \begin{pmatrix} 0.2426 & 0 \\ 0 & 0.2426 \end{pmatrix} \quad M_{12} = \begin{pmatrix} 0 & -0.0215 \\ -0.0215 & 0 \end{pmatrix}$$
$$M_{22} = \begin{pmatrix} 0.043 & 0 \\ 0 & 0.043 \end{pmatrix} \quad M_{13} = \begin{pmatrix} 0 & 0 \\ 0 & 0 \end{pmatrix}$$

Recovered:

$$M_{11}^{(2)} = \begin{pmatrix} 0.2444 & -0.0007 \\ -0.0007 & 0.2408 \end{pmatrix} \quad M_{11}^{(3)} = \begin{pmatrix} 0.2438 & 0 \\ 0 & 0.2414 \end{pmatrix}$$
$$M_{11}^{(4)} = \begin{pmatrix} 0.2429 & -0.0001 \\ -0.0001 & 0.2430 \end{pmatrix} \quad M_{11}^{(5)} = \begin{pmatrix} 0.2426 & 0 \\ 0 & 0.2426 \end{pmatrix}$$

$$M_{12}^{(3)} = \begin{pmatrix} 0.0008 & -0.2414 \\ -0.0212 & -0.0087 \end{pmatrix} \quad M_{12}^{(4)} = \begin{pmatrix} 0 & -0.2413 \\ -0.0213 & 0 \end{pmatrix}$$
$$M_{12}^{(5)} = \begin{pmatrix} 0 & -0.2415 \\ -0.0215 & 0 \end{pmatrix}$$

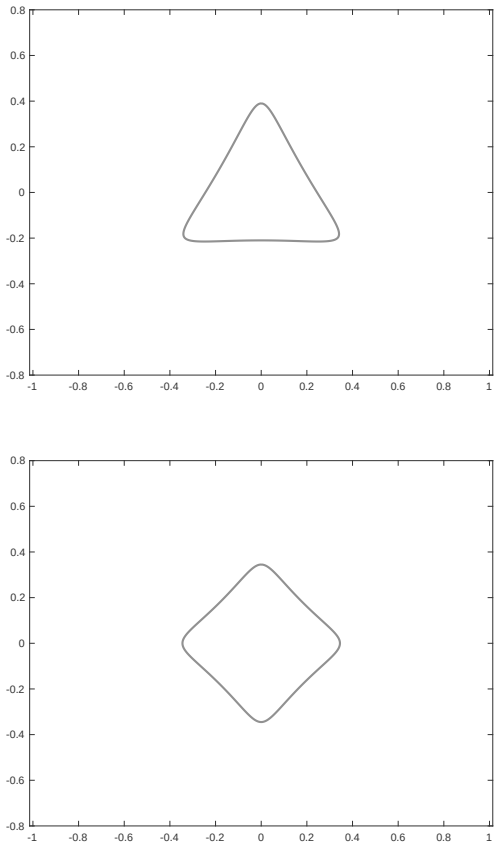

FIGURE 8.3: Non plasmonic particles D_1. Triangular-shaped (left) and rectangular-shaped (right).

$$M_{22}^{(4)} = \begin{pmatrix} 0.0180 & 0.2204 \\ 0.2204 & 0.0389 \end{pmatrix} \quad M_{22}^{(5)} = \begin{pmatrix} 0.0368 & 0.0010 \\ 0.0010 & 0.0497 \end{pmatrix}$$
$$M_{13}^{(4)} = \begin{pmatrix} 0.0093 & -0.1126 \\ -0.1123 & -0.0019 \end{pmatrix} \quad M_{13}^{(5)} = \begin{pmatrix} 0.0032 & -0.0005 \\ -0.0005 & -0.0032 \end{pmatrix}$$

Rectangular-shaped D_1:

Theoretical values:

$$M_{11} = \begin{pmatrix} 0.2682 & 0.0000 \\ 0 & 0.2682 \end{pmatrix} \quad M_{12} = \begin{pmatrix} 0 & 0 \\ 0 & 0 \end{pmatrix}$$
$$M_{22} = \begin{pmatrix} 0.0544 & 0 \\ 0 & 0.0402 \end{pmatrix} \quad M_{13} = \begin{pmatrix} 0.0054 & 0 \\ 0 & -0.0054 \end{pmatrix}$$

Recovered:

$$M_{11}^{(2)} = \begin{pmatrix} 0.2703 & 0.0001 \\ 0.0001 & 0.2661 \end{pmatrix} \quad M_{11}^{(3)} = \begin{pmatrix} 0.2696 & 0 \\ 0 & 0.2662 \end{pmatrix}$$
$$M_{11}^{(4)} = \begin{pmatrix} 0.2682 & 0 \\ 0 & 0.2681 \end{pmatrix} \quad M_{11}^{(5)} = \begin{pmatrix} 0.2682 & 0 \\ 0 & 0.2681 \end{pmatrix}$$

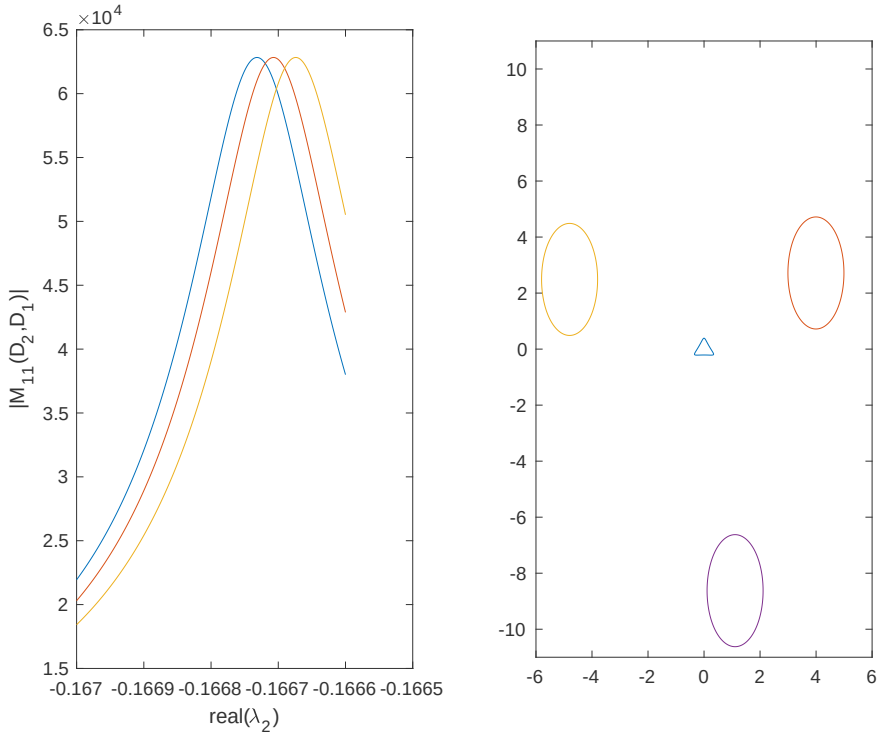

FIGURE 8.4: (right) Modulus of the entry (1,1) of the first order polarization tensor given in Theorem 8.2.2, for different positions of D_2 around a triangular-shaped particle D_2 (left).

$$M_{12}^{(3)} = \begin{pmatrix} 0.0038 & -0.0001 \\ 0 & -0.0112 \end{pmatrix} \quad M_{12}^{(4)} = \begin{pmatrix} 0 & 0 \\ 0 & 0 \end{pmatrix}$$
$$M_{12}^{(5)} = \begin{pmatrix} 0 & 0 \\ 0 & 0 \end{pmatrix}$$

$$M_{22}^{(4)} = \begin{pmatrix} 0.0530 & -0.0007 \\ -0.0007 & 0.0425 \end{pmatrix} \quad M_{22}^{(5)} = \begin{pmatrix} 0.0537 & 0.0006 \\ 0.0006 & 0.0416 \end{pmatrix}$$
$$M_{13}^{(4)} = \begin{pmatrix} 0.0064 & 0.0003 \\ 0.0004 & -0.0063 \end{pmatrix} \quad M_{13}^{(5)} = \begin{pmatrix} 0.0060 & -0.0003 \\ -0.0003 & -0.0059 \end{pmatrix}$$

The results of minimizing the functional (8.20) with a gradient descent approach and using the recovered CGPTs of order less or equal to 5 are shown in Figures 8.6 and 8.7. We take as initial point the equivalent ellipse to D_1, given by the first order polarization recovered with Algorithm 8.3.1, i.e $M_{11}^{(5)}$.

8.5 Concluding remarks

In this chapter, using the quasi-static model, we have shown that the fine details of a small object can be reconstructed from the shift of resonant frequencies it induces to a plasmonic particle in the intermediate regime. This provides a solution for the ill-posed inverse problem of reconstructing small

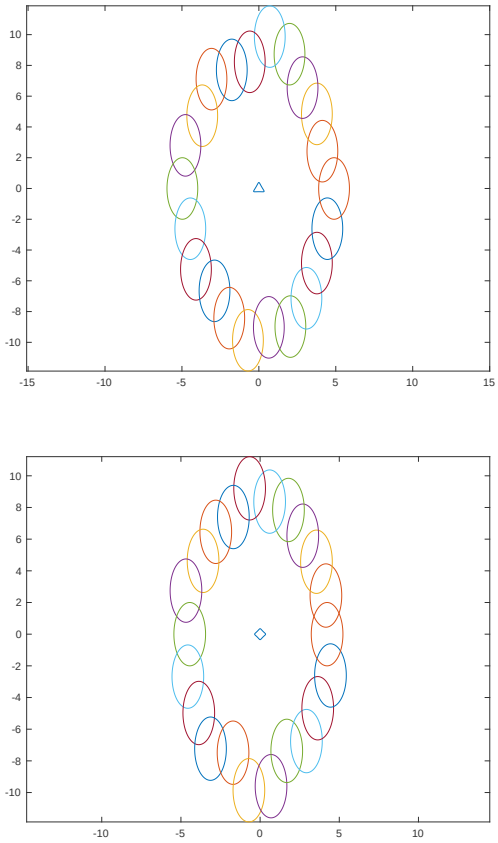

FIGURE 8.5: Positions of D_2 for which we measure $\mathcal{P}_1$. (left) Triangular-shaped particle D_1, (right) rectangular-shaped particle D_1.

objects from far-field measurements and also laid a mathematical foundation for plasmonic bio-sensing. The idea can be extended in several directions: (i) to investigate the strong interaction regime when the small object is close to the plasmonic particle; (ii) to study the case when the size of object is comparable to the size of plasmonic particle; (iii) to analyze the case with multiple small objects and multiple plasmonic particles; (iv) to consider the more practical model of Maxwell equations, and (v) to investigate other types of subwavelength resonances such as Minnaert resonance [10, 75] in bubbly fluids. These new developments will be reported in forthcoming works.

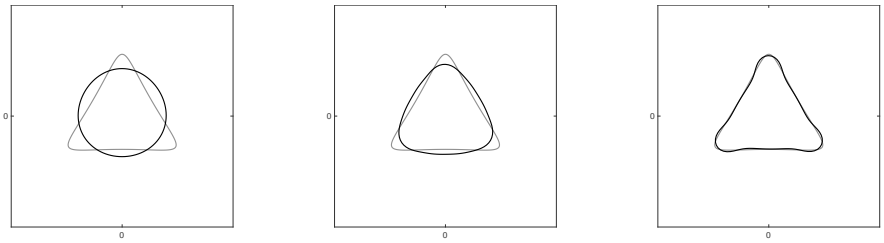

FIGURE 8.6: Shape recovery of a triangular-shaped particle D_1. From left to right, we show both, the original shape and the recovered one after 0 iterations, after 8 iterations and after 30 iterations.

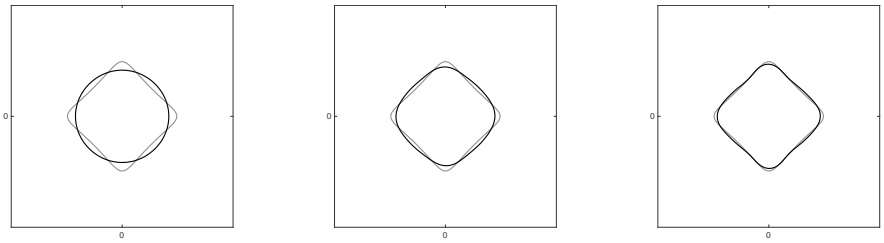

FIGURE 8.7: Shape recovery of a rectangular-shaped particle D_1. From left to right, we show both, the original shape and the recovered one after 0 iterations, after 30 iterations and after 100 iterations.

Appendix A

Layer Potentials for the Laplacian in two Dimensions

In $\mathbb{R}^2$ the single-layer potential $\mathcal{S}_D : H^{-1/2}(\partial D) \to H^{1/2}(\partial D)$ is not, in general, invertible. Hence, $-(u, \mathcal{S}_D[v])_{-\frac{1}{2},\frac{1}{2}}$ does not define an inner product and the symmetrization technique described in [1] is no longer valid.

Here and throughout, $(\cdot,\cdot)_{-\frac{1}{2},\frac{1}{2}}$ denotes the duality pairing between $H^{-1/2}(\partial D)$ and $H^{1/2}(\partial D)$.

To overcome this difficulty, we will introduce a substitute of $\mathcal{S}_D$, in the same way as in [32].

We first need the following lemma.

Lemma A.0.1. *Let* $\mathcal{C} = \{\varphi \in H^{-1/2}(\partial D);\ \exists\, \alpha \in \mathbb{C},\ \mathcal{S}_D[\varphi] = \alpha\}$. *We have* $\dim(\mathcal{C}) = 1$.

Proof. It is known that

$$\begin{aligned} \mathcal{A}_D : H^{-1/2}(\partial D) \times \mathbb{C} &\to H^{1/2}(\partial D) \times \mathbb{C} \\ (\varphi, a) &\to \Big(\mathcal{S}_D[\varphi] + a, \int_{\partial D} \varphi d\sigma\Big), \end{aligned}$$

is invertible [18, Theorem 2.26].

We can see that $\mathcal{C} = \Pi_1 \mathcal{A}_D^{-1}(0, \mathbb{C})$, where $\Pi_1[(\varphi, a)] = \varphi$. The invertibility of $\mathcal{A}_D$ implies that $\mathrm{Ker}(\Pi_1 \mathcal{A}_D^{-1}(0,\cdot)) = \{0\}$. Thus, by the range theorem we have

$$1 = \dim(\mathrm{Im}(\Pi_1 \mathcal{A}_D^{-1}(0,\cdot))) + \dim(\mathrm{Ker}(\Pi_1 \mathcal{A}_D^{-1}(0,\cdot))) = \dim(\mathrm{Im}(\Pi_1 \mathcal{A}_D^{-1}(0,\cdot))) = \dim(\mathcal{C}).$$

□

Definition A.1. *We call* φ_0 *the unique element of* $\mathcal{C}$ *such that* $\int_{\partial D} \varphi_0 d\sigma = 1$.

Note that for every $\varphi \in H^{-1/2}(\partial D)$ we have the decomposition

$$\varphi = \varphi - \Big(\int_{\partial D} \varphi d\sigma\Big)\varphi_0 + \Big(\int_{\partial D} \varphi d\sigma\Big)\varphi_0 := \psi + \Big(\int_{\partial D} \varphi d\sigma\Big)\varphi_0,$$

where we can see that $(\psi, 1)_{-\frac{1}{2},\frac{1}{2}} = 0$. This kind of decomposition, $\varphi = \psi + \alpha\varphi_0$, with $(\psi, 1)_{-\frac{1}{2},\frac{1}{2}} = 0$ is unique.

Note that we can decompose $H^{-1/2}$ as a direct sum of elements with zero-mean and multiples of φ_0, $H^{-1/2}(\partial D) = H_0^{-1/2}(\partial D) \oplus \{\mu\varphi_0, \mu \in \mathbb{C}\}$. This allows us to define the following operator.

Definition A.2. *Let $\widetilde{\mathcal{S}}_D$ be the linear operator that satisfies*

$$\begin{aligned} \widetilde{\mathcal{S}}_D : H^{-1/2}(\partial D) &\to H^{1/2}(\partial D) \\ \varphi &\to \begin{cases} \mathcal{S}_D[\varphi] & \text{if } (\varphi,1)_{-\frac{1}{2},\frac{1}{2}} = 0, \\ -1 & \text{if } \varphi_0 = \varphi. \end{cases} \end{aligned}$$

Remark A.0.1. *When $\mathcal{S}_D$ is invertible, $\widetilde{\mathcal{S}}_D$ is similar enough to keep the invertibility. When $\mathcal{S}_D$ is not invertible, then $\mathcal{C} = ker(\mathcal{S}_D)$ and the operator $\widetilde{\mathcal{S}}_D$ becomes an invertible alternative to $\mathcal{S}_D$ that images the kernel $\mathcal{C}$ to the space $\{\mu\chi(\partial D), \mu \in \mathbb{C}\}$.*

Remark A.0.2. *$\widetilde{\mathcal{S}}_D : H^{-1/2}(\partial D) \to H^1(D)$ follows the same definition.*

Theorem A.0.1. *$\widetilde{\mathcal{S}}_D$ is invertible, self-adjoint and negative for $(\cdot,\cdot)_{-\frac{1}{2},\frac{1}{2}}$ and satisfies the following Calderón identity: $\widetilde{\mathcal{S}}_D\mathcal{K}_D^* = \mathcal{K}_D\widetilde{\mathcal{S}}_D$.*

Proof. The invertibility is a direct consequence of Lemma A.0.1.

Indeed, since $\mathcal{S}_D$ is Fredholm of zero index, so is $\widetilde{\mathcal{S}}_D$. Therefore, we only need the injectivity. Suppose that, $\exists\, \varphi \neq 0$ such that $\widetilde{\mathcal{S}}_D[\varphi] = 0$. This mean that, $\exists\, \alpha \neq 0 \in \mathbb{C}$ such that $\varphi = \alpha\varphi_0$. Therefore, $\widetilde{\mathcal{S}}_D[\varphi] = \alpha\widetilde{\mathcal{S}}_D[\varphi_0] = -\alpha = 0$, which is a contradiction. Hence $\varphi = 0$.

The self-adjointness comes directly form that of $\mathcal{S}_D$. Noticing that φ_0 is an eigenfunction of eigenvalue $1/2$ of $\mathcal{K}_D^*$ we get the Calderón identity from a similar one satisfied by $\mathcal{S}_D$: $\mathcal{S}_D\mathcal{K}_D^* = \mathcal{K}_D\mathcal{S}_D$; see [12, Lemma 2.12].

It is known that $\int_{\partial D} \psi\mathcal{S}_D[\psi]d\sigma < 0$ if $(\psi,1)_{-\frac{1}{2},\frac{1}{2}} = 0$ and $\psi \neq 0$, see [12, Lemma 2.10]. Therefore, writing $\varphi = \psi + \Big(\int_{\partial D}\varphi d\sigma\Big)\varphi_0$, with $\psi = \varphi - \Big(\int_{\partial D}\varphi d\sigma\Big)\varphi_0$, and noticing that $\int_{\partial D}\varphi_0\widetilde{\mathcal{S}}_D[\psi]d\sigma = \int_{\partial D}\widetilde{\mathcal{S}}_D[\varphi_0]\psi d\sigma = -\int_{\partial D}\psi d\sigma = 0$, we have

$$\begin{aligned} \int_{\partial D}\varphi\widetilde{\mathcal{S}}_D[\varphi]d\sigma &= \int_{\partial D}\psi\widetilde{\mathcal{S}}_D[\psi]d\sigma + \Big(\int_{\partial D}\varphi d\sigma\Big)^2\widetilde{\mathcal{S}}_D[\varphi_0] \\ &= \int_{\partial D}\psi\mathcal{S}_D[\psi]d\sigma - \Big(\int_{\partial D}\varphi d\sigma\Big)^2 < 0, \end{aligned}$$

if $\varphi \neq 0$.

□

Definition A.3. *We define the space $\mathcal{H}^*(\partial D)$ as the Hilbert space resulting from endowing $H^{-1/2}(\partial D)$ with the inner product*

$$(u,v)_{\mathcal{H}^*} := -(u, \widetilde{\mathcal{S}}_D[v])_{-\frac{1}{2},\frac{1}{2}}. \tag{A.1}$$

Similarly, we let $\mathcal{H}$ to be the Hilbert space resulting from endowing $H^{1/2}$ with the inner product

$$(u,v)_{\mathcal{H}} = -(\widetilde{\mathcal{S}}_D^{-1}[u], v)_{-\frac{1}{2},\frac{1}{2}}. \tag{A.2}$$

If D is $\mathcal{C}^{1,\alpha}$, we have the following result.

Lemma A.0.2. *Let D be a $\mathcal{C}^{1,\alpha}$ bounded domain of $\mathbb{R}^2$ and let $\widetilde{\mathcal{S}}_D$ be the operator introduced in Definition A.2. Then*

(i) The operator $\mathcal{K}_D^*$ is compact self-adjoint in the Hilbert space $\mathcal{H}^*(\partial D)$ and $\mathcal{H}^*(\partial D)$ is equivalent to $H^{-\frac{1}{2}}(\partial D)$; Similarly, the Hilbert space $\mathcal{H}(\partial D)$ is equivalent to $H^{\frac{1}{2}}(\partial D)$.

(ii) Let (λ_j, φ_j), $j = 0, 1, 2, \ldots$, be the eigenvalue and normalized eigenfunction pair of $\mathcal{K}_D^*$ with $\lambda_0 = \frac{1}{2}$. Then, $\lambda_j \in (-\frac{1}{2}, \frac{1}{2}]$ and $\lambda_j \to 0$ as $j \to \infty$;

(iii) The following representation formula holds: for any $\varphi \in H^{-1/2}(\partial D)$,

$$\mathcal{K}_D^*[\varphi] = \sum_{j=0}^{\infty} \lambda_j (\varphi, \varphi_j)_{\mathcal{H}^*} \otimes \varphi_j.$$

The following lemmas are needed in the proof of Theorem 4.2.1 and Theorem 4.2.2.

Lemma A.0.3. *Let $D = z + \delta B$ and η be the function such that, for every $\varphi \in \mathcal{H}^*(\partial D)$, $\eta(\varphi)(\tilde{x}) = \varphi(z + \delta\tilde{x})$, for almost all $\tilde{x} \in \partial B$. Then*

$$\|\varphi\|_{\mathcal{H}^*(\partial D)} = \delta \|\eta(\varphi)\|_{\mathcal{H}^*(\partial B)}.$$

Similarly, if for every $\varphi \in L^2(D)$, $\eta(\varphi)(\tilde{x}) = \varphi(z+\delta\tilde{x})$, for almost all $\tilde{x} \in B$, then

$$\|\varphi\|_{L^2(D)} = \delta \|\eta(\varphi)\|_{L^2(B)}.$$

Proof. We only prove the scaling in $\mathcal{H}^*(\partial D)$. From the proof of Theorem A.0.1, we have

$$\|\varphi\|^2_{\mathcal{H}^*(\partial D)} = -\int_{\partial D} \psi \mathcal{S}_D[\psi] d\sigma + \Big(\int_{\partial D} \varphi d\sigma \Big)^2,$$

where $\psi = \varphi - \Big(\int_{\partial D} \varphi d\sigma \Big) \varphi_0$. Note that $(\psi, 1)_{-\frac{1}{2}, \frac{1}{2}} = 0$ and so, $(\eta(\psi), 1)_{-\frac{1}{2}, \frac{1}{2}} = 0$ as well.

By a rescaling argument we find that

$$\begin{aligned}
\|\varphi\|^2_{\mathcal{H}^*(\partial D)} &= -\delta^2 \int_{\partial B} \int_{\partial B} \frac{1}{2\pi} \log|\delta(\tilde{x} - \tilde{y})| \eta(\psi)(\tilde{x}) \eta(\psi)(\tilde{y}) d\sigma(\tilde{x}) d\sigma(\tilde{y}) + \delta^2 \Big(\int_{\partial B} \eta(\varphi) d\sigma \Big)^2 \\
&= -\frac{1}{2\pi} \delta^2 \log(\delta) \Big(\int_{\partial B} \eta(\psi) d\sigma \Big)^2 + \delta^2 \left(- \int_{\partial B} \eta(\psi) \mathcal{S}_D[\eta(\psi)] d\sigma + \Big(\int_{\partial B} \eta(\varphi) d\sigma \Big)^2 \right) \\
&= \delta^2 \|\eta(\varphi)\|^2_{\mathcal{H}^*(\partial B)}.
\end{aligned}$$

□

Lemma A.0.4. *Let $g \in H^1(D)$ be such that $\Delta g = f$ with $f \in L^2(D)$. Then, in $\mathcal{H}^*(\partial D)$,*

$$(\frac{1}{2} I - \mathcal{K}_D^*) \widetilde{\mathcal{S}}_D^{-1}[g] = -\frac{\partial g}{\partial \nu} + \mathcal{T}_f.$$

For some $\mathcal{T}_f \in \mathcal{H}^*(\partial D)$ and $\|\mathcal{T}_f\|_{\mathcal{H}^*} \leq C \|f\|_{L^2(D)}$ for a constant C.

Moreover, if $g \in H^1_{loc}(\mathbb{R}^2)$, $\Delta g = 0$ in $\mathbb{R}^2 \backslash \bar{D}$, $\lim_{|x| \to \infty} g(x) = 0$, then

$$\mathcal{T}_f = c_f \varphi_0 + \widetilde{\mathcal{S}}_D^{-1}[g],$$

with

$$c_f = \int_D f(x)dx - \int_{\partial D} \widetilde{\mathcal{S}}_D^{-1}[g](y)d\sigma(y),$$

where φ_0 is given in Definition A.1. Here, by an abuse of notation, we still denote by g the trace of g on ∂D.

Proof. Let $\varphi \in \mathcal{H}^*(\partial D)$. Then

$$\begin{aligned}
\Big((\frac{1}{2}I - \mathcal{K}_D^*)\widetilde{\mathcal{S}}_D^{-1}[g], \varphi\Big)_{\mathcal{H}^*} &= -\Big(\widetilde{\mathcal{S}}_D^{-1}[g], (\frac{1}{2}I - \mathcal{K}_D)\widetilde{\mathcal{S}}_D[\varphi]\Big)_{-\frac{1}{2},\frac{1}{2}} \\
&= -\Big(\widetilde{\mathcal{S}}_D^{-1}[g], \widetilde{\mathcal{S}}_D(\frac{1}{2}I - \mathcal{K}_D^*)[\varphi]\Big)_{-\frac{1}{2},\frac{1}{2}} \\
&= -\Big(g, (\frac{1}{2}I - \mathcal{K}_D^*)[\varphi]\Big)_{-\frac{1}{2},\frac{1}{2}} \\
&= -\Big(g, -\frac{\partial \widetilde{\mathcal{S}}_D[\varphi]}{\partial \nu}\Big|_-\Big)_{-\frac{1}{2},\frac{1}{2}} \\
&= \int_{\partial D} \frac{\partial g}{\partial \nu}\widetilde{\mathcal{S}}_D[\varphi]d\sigma - \int_D \Big(f\widetilde{\mathcal{S}}_D[\varphi] - \Delta\widetilde{\mathcal{S}}_D[\varphi](g)\Big)dx \\
&= -\Big(\frac{\partial g}{\partial \nu}, \varphi\Big)_{\mathcal{H}^*} - \int_D f\widetilde{\mathcal{S}}_D[\varphi]dx.
\end{aligned}$$

We have used the fact that $\widetilde{\mathcal{S}}_D$ is harmonic in D.

Consider the linear application $\mathcal{T}_f[\varphi] := -\int_D f\widetilde{\mathcal{S}}_D[\varphi]dx$. We have

$$|\mathcal{T}_f[\varphi]| \leq C\|f\|_{L^2(D)}\|\widetilde{\mathcal{S}}_D[\varphi]\|_{L^2(D)} \leq C_f\|\widetilde{\mathcal{S}}_D[\varphi]\|_{H^1(D)} \leq C_f\|\widetilde{\mathcal{S}}_D[\varphi]\|_{H^{\frac{1}{2}}(\partial D)} \leq C_f\|\varphi\|_{H^{-\frac{1}{2}}(\partial D)}.$$

Here we have used Holder's inequality, a standard Sobolev embedding, the trace theorem and the fact that $\widetilde{\mathcal{S}}_D : H^{-\frac{1}{2}}(\partial D) \to H^{\frac{1}{2}}(\partial D)$ is continuous. By the Riez representation theorem, there exists $v \in \mathcal{H}^*(\partial D)$ such that $\mathcal{T}_f[\varphi] = (v, \varphi)_{\mathcal{H}^*}$, $\forall \varphi \in \mathcal{H}^*(\partial D)$.

By abuse of notation we still denote $\mathcal{T}_f := v$ to make explicit the dependency on f. It follows that

$$\begin{aligned}
\|\mathcal{T}_f\|_{\mathcal{H}^*}^2 = -\int_D f\widetilde{\mathcal{S}}_D[\mathcal{T}_f]dx &\leq C\|f\|_{L^2(D)}\|\widetilde{\mathcal{S}}_D[\mathcal{T}_f]\|_{L^2(D)} \\
&\leq C\|f\|_{L^2(D)}\|\widetilde{\mathcal{S}}_D[\mathcal{T}_f]\|_{H^1(D)} \\
&\leq C\|f\|_{L^2(D)}\|\widetilde{\mathcal{S}}_D[\mathcal{T}_f]\|_{H^{\frac{1}{2}}(\partial D)} \\
&\leq C\|f\|_{L^2(D)}\|\mathcal{T}_f\|_{\mathcal{H}^*}.
\end{aligned}$$

We now show that in $\mathcal{H}_0^*(\partial D)$, $\mathcal{T}_f = \widetilde{\mathcal{S}}_D^{-1}[g]$.

Indeed, let $\varphi \in \mathcal{H}_0^*(\partial D)$, then

$$\begin{aligned}
\Big(\widetilde{\mathcal{S}}_D^{-1}[g],\varphi\Big)_{\mathcal{H}^*} &= -\Big(\widetilde{\mathcal{S}}_D^{-1}[g],\widetilde{\mathcal{S}}_D[\varphi]\Big)_{-\frac{1}{2},\frac{1}{2}} \\
&= -\Big(g,\varphi\Big)_{-\frac{1}{2},\frac{1}{2}} \\
&= -\Big(g,\frac{\partial\widetilde{\mathcal{S}}_D[\varphi]}{\partial\nu}\Big|_+ - \frac{\partial\widetilde{\mathcal{S}}_D[\varphi]}{\partial\nu}\Big|_-\Big)_{-\frac{1}{2},\frac{1}{2}} \\
&= \int_{\partial D}\frac{\partial g}{\partial\nu}\widetilde{\mathcal{S}}_D[\varphi]d\sigma - \int_{\partial D}\frac{\partial g}{\partial\nu}\widetilde{\mathcal{S}}_D[\varphi]d\sigma + \int_{\partial B_\infty}\frac{\partial g}{\partial\nu}\widetilde{\mathcal{S}}_D[\varphi]d\sigma - \int_{\partial B_\infty} g\frac{\partial\widetilde{\mathcal{S}}_D[\varphi]}{\partial\nu}d\sigma \\
&\quad - \int_{\mathbb{R}^2}\Big(f\widetilde{\mathcal{S}}_D[\varphi] - \Delta\widetilde{\mathcal{S}}_D[\varphi](g)\Big)dx \\
&= -\int_D f\widetilde{\mathcal{S}}_D[\varphi]dx.
\end{aligned}$$

Here we have used the assumption on g, the fact that $\widetilde{\mathcal{S}}_D[\varphi]$ is harmonic in D and $\mathbb{R}^2\backslash\bar{D}$ and that for $\varphi \in \mathcal{H}_0^*(\partial D)$ we have $\widetilde{\mathcal{S}}_D[\varphi](x) = O(\frac{1}{|x|})$ and $\frac{\partial\widetilde{\mathcal{S}}_D[\varphi]}{\partial\nu}(x) = O(\frac{1}{|x|})$ for $|x| \to \infty$.

Therefore,

$$\mathcal{T}_f = (\mathcal{T}_f - \widetilde{\mathcal{S}}_D^{-1}[g],\varphi_0)_{\mathcal{H}^*}\varphi_0 + \widetilde{\mathcal{S}}_D^{-1}[g].$$

Finally, re-scaling the definition of φ_0 given in Definition A.1 we obtain that

$$(\mathcal{T}_f - \widetilde{\mathcal{S}}_D^{-1}[g],\varphi_0)_{\mathcal{H}^*} = \int_D f(x)dx - \int_{\partial D}\widetilde{\mathcal{S}}_D^{-1}[g](y)d\sigma(y).$$

□

Appendix B

Asymptotic Expansions

In this section, we derive asymptotic expansions for the Helmholtz integral operators with respect to k, of some boundary integral operators defined on the boundary of a bounded and simply connected smooth domain D.

B.1 Asymptotic expansions in $\mathbb{R}^3$

We consider a domain $D \Subset \mathbb{R}^3$ whose size is of order one.

Recall the definition of the single layer potential

$$\mathcal{S}_D^k[\psi](x) = \int_{\partial D} G(x,y,k)\psi(y)d\sigma(y), \quad x \in \partial D,$$

where

$$G(x,y,k) = -\frac{e^{ik|x-y|}}{4\pi|x-y|}$$

is the Green function of Helmholtz equation in $\mathbb{R}^3$, subject to the Sommerfeld radiation condition. Note that

$$G(x,y,k) = -\sum_{j=0}^{\infty} \frac{(ik|x-y|)^j}{j!4\pi|x-y|} = -\frac{1}{4\pi|x-y|} - \frac{ik}{4\pi}\sum_{j=1}^{\infty}\frac{(ik|x-y|)^{j-1}}{j!}.$$

We get

$$\mathcal{S}_D^k = \mathcal{S}_D + \sum_{j=1}^{\infty} k^j \mathcal{S}_{D,j}, \tag{B.1}$$

where

$$\mathcal{S}_{D,j}[\psi](x) = -\frac{i}{4\pi}\int_{\partial D}\frac{(i|x-y|)^{j-1}}{j!}\psi(y)d\sigma(y).$$

In particular, we have

$$\mathcal{S}_{D,1}[\psi](x) = -\frac{i}{4\pi}\int_{\partial D}\psi(y)d\sigma(y), \tag{B.2}$$

$$\mathcal{S}_{D,2}[\psi](x) = -\frac{1}{4\pi}\int_{\partial D}|x-y|\psi(y)d\sigma(y). \tag{B.3}$$

Lemma B.1.1. *$\|\mathcal{S}_{D,j}\|_{\mathcal{L}((\mathcal{H}^*(\partial D),\mathcal{H}(\partial D))}$ is uniformly bounded with respect to j. Moreover, the series in (B.1) is convergent in $\mathcal{L}(\mathcal{H}^*(\partial D),\mathcal{H}(\partial D))$.*

Proof. It is clear that

$$\|\mathcal{S}_{D,j}\|_{\mathcal{L}(L^2(\partial D),H^1(\partial D))} \leq C,$$

where C is independent of j. On the other hand, a similar estimate also holds for the operator $\mathcal{S}^*_{D,j}$. It follows that

$$\|\mathcal{S}_{D,j}\|_{\mathcal{L}(H^{-1}(\partial D),L^2(\partial D))} \leq C.$$

Thus, we can conclude that $\|\mathcal{S}_{D,j}\|_{\mathcal{L}(H^{-\frac{1}{2}}(\partial D),H^{\frac{1}{2}}(\partial D))}$ is uniformly bounded by using interpolation theory. By the equivalence of norms in the $H^{-\frac{1}{2}}(\partial D)$ and $H^{\frac{1}{2}}(\partial D)$, the lemma follows immediately. □

Note that $\mathcal{S}_D$ is invertible in dimension three, so is $\mathcal{S}^k_D$ for small k. By formally writing

$$(\mathcal{S}^k_D)^{-1} = \mathcal{S}_D^{-1} + k\mathcal{B}_{D,1} + k^2\mathcal{B}_{D,2} + \dots, \tag{B.4}$$

and using the identity $(\mathcal{S}^k_D)^{-1}\mathcal{S}^k_D = Id$, we can derive that

$$\mathcal{B}_{D,1} = -\mathcal{S}_D^{-1}\mathcal{S}_{D,1}\mathcal{S}_D^{-1}, \quad \mathcal{B}_{D,2} = -\mathcal{S}_D^{-1}\mathcal{S}_{D,2}\mathcal{S}_D^{-1} + \mathcal{S}_D^{-1}\mathcal{S}_{D,1}\mathcal{S}_D^{-1}\mathcal{S}_{D,1}\mathcal{S}_D^{-1}. \tag{B.5}$$

We can also derive other lower-order terms $\mathcal{B}_{D,j}$.

Lemma B.1.2. *The series in (B.4) converges in $\mathcal{L}(\mathcal{H}(\partial D),\mathcal{H}^*(\partial D))$ for sufficiently small k.*

Proof. The proof can be deduced from the identity

$$(\mathcal{S}^k_D)^{-1} = (Id + \mathcal{S}_D^{-1}\sum_{j=1}^{\infty} k^j\mathcal{S}_{D,j})^{-1}\mathcal{S}_D^{-1}.$$

□

We now consider the expansion for the boundary integral operator $(\mathcal{K}^k_D)^*$. We have

$$(\mathcal{K}^k_D)^* = \mathcal{K}^*_D + k\mathcal{K}_{D,1} + k^2\mathcal{K}_{D,2} + \dots, \tag{B.6}$$

where

$$\mathcal{K}_{D,j}[\psi](x) = -\frac{i}{4\pi}\int_{\partial D}\frac{\partial(i|x-y|)^{j-1}}{j!\partial\nu(x)}\psi(y)d\sigma(y) = -\frac{i^j(j-1)}{4\pi j!}\int_{\partial D}|x-y|^{j-3}(x-y)\cdot\nu(x)\psi(y)d\sigma(y).$$

In particular, we have

$$\mathcal{K}_{D,1} = 0, \quad \mathcal{K}_{D,2}[\psi](x) = \frac{1}{4\pi}\int_{\partial D}\frac{(x-y)\cdot\nu(x)}{|x-y|}\psi(y)d\sigma(y). \tag{B.7}$$

Lemma B.1.3. *The norm $\|\mathcal{K}_{D,j}\|_{\mathcal{L}(\mathcal{H}^*(\partial D),\mathcal{H}^*(\partial D))}$ is uniformly bounded for $j \geq 1$. Moreover, the series in (B.6) is convergent in $\mathcal{L}(\mathcal{H}^*(\partial D),\mathcal{H}^*(\partial D))$.*

B.2 Asymptotic expansion in $\mathbb{R}^3$: multiple particles

In this section, we consider the multiple particle case in dimension three. We assume that the particles have size of order δ which is a small number and the distance between them is of order one. We write $D_j = z_j + \delta\widetilde{D}$, $j = 1, 2, \dots, M$, where $\widetilde{D}$ has size one and is centered at the origin. Our goal is to derive estimates for various boundary integral operators that are

defined on small particles in terms of their size. For this purpose, we denote by $D_0 = \delta\widetilde{D}$. For each function f defined on ∂D_0, we define a corresponding function on $\widetilde{D}$ by

$$\eta(f)(\widetilde{x}) = f(\delta\widetilde{x}).$$

In this section, we denote by $\chi(\partial D_j)$ the constant function equal one over the border of D_j.

We first state some useful results.

Lemma B.2.1. *The following scaling properties hold:*

(i) $\|\eta(f)\|_{L^2(\partial\widetilde{D})} = \delta^{-1}\|f\|_{L^2(\partial D_0)}$;

(ii) $\|\eta(f)\|_{\mathcal{H}(\partial\widetilde{D})} = \delta^{-\frac{1}{2}}\|f\|_{\mathcal{H}(\partial D_0)}$;

(iii) $\|\eta(f)\|_{\mathcal{H}^*(\partial\widetilde{D})} = \delta^{-\frac{3}{2}}\|f\|_{\mathcal{H}^*(\partial D_0)}$.

Proof. The proof of (i) is straightforward and we only need to prove (ii) and (iii). To prove (iii), we have

$$\begin{aligned}\|f\|^2_{\mathcal{H}^*(\partial D_0)} &= \int_{\partial D_0}\int_{\partial D_0}\frac{f(x)f(y)}{4\pi|x-y|}d\sigma(x)d\sigma(y)\\ &= \delta^3\int_{\partial\widetilde{D}}\int_{\partial\widetilde{D}}\frac{\eta(f)(\widetilde{x})\eta(f)(\widetilde{y})}{4\pi|\widetilde{x}-\widetilde{y}|}d\sigma(\widetilde{x})d\sigma(\widetilde{x})\\ &= \delta^3\|\eta(f)\|^2_{\mathcal{H}^*(\partial\widetilde{D})},\end{aligned}$$

whence (iii) follows. To prove (ii), recall that

$$\|f\|_{\mathcal{H}(\partial D_0)} = \|\mathcal{S}^{-1}_{D_0}f\|_{\mathcal{H}^*(\partial D_0)}.$$

Let $u = \mathcal{S}^{-1}_{D_0}[f]$. Then $f = \mathcal{S}_{D_0}[u]$. We can show that

$$\eta(f) = \delta\mathcal{S}_{\widetilde{D}}(\eta(u)).$$

As a result, we have

$$\|\eta(f)\|_{\mathcal{H}(\partial\widetilde{D})} = \delta\|\mathcal{S}_{\widetilde{D}}(\eta(u))\|_{\mathcal{H}(\partial\widetilde{D})} = \delta\|\eta(u)\|_{\mathcal{H}^*(\partial\widetilde{D})} = \delta^{-\frac{1}{2}}\|u\|_{\mathcal{H}^*(\partial D_0)} = \delta^{-\frac{1}{2}}\|f\|_{\mathcal{H}(\partial D_0)},$$

which proves (ii). □

Lemma B.2.2. *Let X and Y be bounded and simply connected smooth domains in $\mathbb{R}^3$. Assume $0 \in X, Y$ and $X = \delta\widetilde{X}$, $Y = \delta\widetilde{Y}$. Let $\mathcal{R}$ and $\widetilde{\mathcal{R}}$ be two boundary integral operators from $\mathcal{D}'(\partial Y)$ to $\mathcal{D}'(\partial X)$ and $\mathcal{D}'(\partial\widetilde{Y})$ to $\mathcal{D}'(\partial\widetilde{X})$, respectively. Here, $\mathcal{D}'$ denotes the Schwartz space. Assume that both operators have the same Schwartz kernel R with the following homogeneous scaling property*

$$R(\delta x, \delta y) = \delta^m R(x, y).$$

Then,

$$\begin{aligned}\|\mathcal{R}\|_{\mathcal{L}(\mathcal{H}^*(\partial Y),\mathcal{H}^*(\partial X))} &= \delta^{2+m}\|\widetilde{\mathcal{R}}\|_{\mathcal{L}(\mathcal{H}^*(\partial\widetilde{Y}),\mathcal{H}^*(\partial\widetilde{X}))},\\ \|\mathcal{R}\|_{\mathcal{L}(\mathcal{H}^*(\partial Y),\mathcal{H}(\partial X))} &= \delta^{1+m}\|\widetilde{\mathcal{R}}\|_{\mathcal{L}(\mathcal{H}^*(\partial\widetilde{Y}),\mathcal{H}(\partial\widetilde{X}))}.\end{aligned}$$

Proof. The result follows from Lemma B.2.1 and the following identity

$$\mathcal{R} = \delta^{2+m}\eta^{-1} \circ \widetilde{\mathcal{R}} \circ \eta.$$

□

We first consider the operators $\mathcal{S}_{D_j}^k$ and $(\mathcal{K}_{D_j}^k)^*$. The following asymptotic expansions hold.

Lemma B.2.3. *(i) Regarded as operators from $\mathcal{H}^*(\partial D_j)$ into $\mathcal{H}(\partial D_j)$, we have*

$$\mathcal{S}_{D_j}^k = \mathcal{S}_{D_j} + k\mathcal{S}_{D_j,1} + k^2\mathcal{S}_{D_j,2} + O(k^3\delta^3),$$

where $\mathcal{S}_{D_j} = O(1)$ and $\mathcal{S}_{D_j,m} = O(\delta^m)$;

(ii) Regarded as operators from $\mathcal{H}(\partial D_j)$ into $\mathcal{H}^*(\partial D_j)$, we have

$$(\mathcal{S}_{D_j}^k)^{-1} = \mathcal{S}_{D_j}^{-1} + k\mathcal{B}_{D_j,1} + k^2\mathcal{B}_{D_j,2} + O(k^3\delta^3),$$

where $\mathcal{S}_{D_j}^{-1} = O(1)$ and $\mathcal{B}_{D_j,m} = O(\delta^m)$;

(iii) Regarded as operators from $\mathcal{H}^*(\partial D_j)$ into $\mathcal{H}^*(\partial D_j)$, we have

$$(\mathcal{K}_{D_j}^k)^* = \mathcal{K}_{D_j}^* + k^2 O(\delta^2),$$

where $\mathcal{K}_{D_j}^* = O(1)$.

Proof. The proof immediately follows from Lemmas B.2.2, B.1.1, and B.1.3. □

We now consider the operator $\mathcal{S}_{D_j,D_l}^k$. By definition,

$$\mathcal{S}_{D_j,D_l}^k[\psi](x) = \int_{\partial D_j} G(x,y,k)\psi(y)d\sigma(y), \quad x \in \partial D_l.$$

Using the expansion

$$G(x,y,k) = \sum_{m=0}^{\infty} k^m Q_m(x,y),$$

where

$$Q_m(x,y) = -\frac{i^m|x-y|^{m-1}}{4\pi},$$

we can derive that

$$\mathcal{S}_{D_j,D_l}^k = \sum_{m\geq 0} k^m \mathcal{S}_{j,l,m},$$

where

$$\mathcal{S}_{j,l,m}[\psi](x) = \int_{\partial D_j} Q_m(x,y)\psi(y)d\sigma(y).$$

We can further write

$$\mathcal{S}_{j,l,m} = \sum_{n\geq 0} \mathcal{S}_{j,l,m,n},$$

where $\mathcal{S}_{j,l,m,n}$ is defined by

$$\mathcal{S}_{j,l,m,n}[\psi](x) = \int_{\partial D_j} \sum_{|\alpha|+|\beta|=n} \frac{1}{\alpha!\beta!} \frac{\partial^{|\alpha|+|\beta|}}{\partial x^\alpha \partial y^\beta} Q_m(z_l, z_j)(x-z_l)^\alpha (y-z_j)^\beta \psi(y)\, d\sigma(y).$$

In particular, we have

$$\begin{aligned}
\mathcal{S}_{j,l,0,0}[\psi](x) &= -\frac{1}{4\pi|z_j - z_l|}(\psi, \chi(\partial D_j))_{H^{-1/2}(\partial D_j), H^{1/2}(\partial D_j)} \chi(D_l), \\
\mathcal{S}_{j,l,0,1}[\psi](x) &= \sum_{|\alpha|=1} \frac{(z_l - z_j)^\alpha}{4\pi|z_l - z_j|^3} \Big((x - z_l)^\alpha (\psi, \chi(\partial D_l))_{H^{-1/2}(\partial D_j), H^{1/2}(\partial D_j)} + \\
&\qquad \big((y - z_j)^\alpha, \psi\big)\chi(D_l)\Big), \\
\mathcal{S}_{j,l,0,2}[\psi](x) &= \sum_{|\alpha|+|\beta|=2} \frac{1}{\alpha!\beta!} \frac{\partial^2 Q_0(z_l, z_j)}{\partial x^\alpha \partial y^\beta} (x - z_l)^\alpha (y - z_j)^\beta \psi(y) d\sigma(y), \\
\mathcal{S}_{j,l,1}[\psi](x) &= -\frac{i}{4\pi}(\psi, \chi(\partial D_j))_{H^{-1/2}(\partial D_j), H^{1/2}(\partial D_j)} \chi(D_l), \\
\mathcal{S}_{j,l,2,0}[\psi](x) &= \frac{1}{4\pi}|z_l - z_j|(\psi, \chi(\partial D_j))_{H^{-1/2}(\partial D_j), H^{1/2}(\partial D_j)} \chi(D_l).
\end{aligned}$$

The following estimate holds.

Lemma B.2.4. *We have* $\|\mathcal{S}_{j,l,m,n}\|_{\mathcal{L}(\mathcal{H}^*(\partial D), \mathcal{H}(\partial D))} \lesssim O(\delta^{n+1})$.

Proof. After a translation of coordinates, the stated estimate immediately follows from Lemma B.2.2. □

Similarly, for the operator $\mathcal{K}^{k_m}_{D_j, D_l}$ defined in the following way

$$\mathcal{K}^k_{D_j, D_l}[\psi](x) = \int_{\partial D_j} \frac{\partial G(x, y, k)}{\partial \nu(x)} \psi(y) d\sigma(y), \quad x \in \partial D_l,$$

we have

$$\mathcal{K}^k_{D_j, D_l} = \sum_{m \geq 0} k^m \sum_{n \geq 0} \mathcal{K}_{j,l,m,n},$$

where

$$\mathcal{K}_{j,l,m,n}[\psi](x) = \int_{\partial D_j} \sum_{|\alpha|+|\beta|=n} \frac{1}{\alpha!\beta!} \frac{\partial^n K_m(z_l, z_j)}{\partial x^\beta \partial y^\alpha} (x - z_l)^\beta (y - z_j)^\alpha (x - y) \cdot \nu(x) \psi(y) d\sigma(y)$$

with

$$K_m(x, y) = -\frac{i^m (m-1)|x - y|^{m-3}}{4\pi m!}.$$

In particular, we have

$$\begin{aligned}
\mathcal{K}_{j,l,0,0}[\psi](x) &= \frac{1}{4\pi|z_l - z_j|^3} \Big[(x - z_l) \cdot \nu(x) \big(\psi, \chi(\partial D_j)\big)_{H^{-1/2}(\partial D_j), H^{1/2}(\partial D_j)} \\
&\quad - \big(\psi, (y - z_j) \cdot \nu(x)\big)_{H^{-1/2}(\partial D_j), H^{1/2}(\partial D_j)} \\
&\quad + (z_l - z_j) \cdot \nu(x) \big(\psi, \chi(\partial D_j)\big)_{H^{-1/2}(\partial D_j), H^{1/2}(\partial D_j)} \Big], \qquad \text{(B.8)} \\
\mathcal{K}_{j,l,1,m}[\psi] &= 0 \quad \text{for all } m. \qquad \text{(B.9)}
\end{aligned}$$

Lemma B.2.5. *We have* $\|\mathcal{K}_{j,l,m,n}\|_{\mathcal{L}(\mathcal{H}^*(\partial D_j),\mathcal{H}^*(\partial D_l))} \lesssim O(\delta^{n+2})$.

Proof. Note that

$$\begin{aligned}\mathcal{K}_{j,l,m,n}[\psi](x) &= \int_{\partial D_j} \sum_{|\alpha|+|\beta|=n} \frac{1}{\alpha!\beta!} \frac{\partial^n K_m(z_l,z_j)}{\partial x^\beta \partial y^\alpha} (x-z_l)^\beta (y-z_j)^\alpha (x-z_l)\cdot\nu(x)\psi(y)d\sigma(y),\\ &\quad - \int_{\partial D_j} \sum_{|\alpha|+|\beta|=n} \frac{1}{\alpha!\beta!} \frac{\partial^n K_m(z_l,z_j)}{\partial x^\beta \partial y^\alpha} (x-z_l)^\beta (y-z_j)^\alpha (y-z_j)\cdot\nu(x)\psi(y)d\sigma(y),\\ &\quad + \int_{\partial D_j} \sum_{|\alpha|+|\beta|=n} \frac{1}{\alpha!\beta!} \frac{\partial^n K_m(z_l,z_j)}{\partial x^\beta \partial y^\alpha} (x-z_l)^\beta (y-z_j)^\alpha (z_l-z_j)\cdot\nu(x)\psi(y)d\sigma(y).\end{aligned}$$

After a translation of coordinates, we can apply Lemma B.2.2 to each one of the three terms above to conclude that $\mathcal{K}_{j,l,m,n} = O(\delta^{n+3}) + O(\delta^{n+2})$. This completes the proof of the lemma. □

To summarize, we have proven the following results.

Lemma B.2.6. *(i) Regarded as an operator from* $\mathcal{H}^*(\partial D_j)$ *into* $\mathcal{H}(\partial D_l)$ *we have,*

$$\mathcal{S}^k_{D_j,D_l} = \mathcal{S}_{j,l,0,0} + \mathcal{S}_{j,l,0,1} + \mathcal{S}_{j,l,0,2} + k\mathcal{S}_{j,l,1} + k^2\mathcal{S}_{j,l,2,0} + O(\delta^4) + O(k^2\delta^2).$$

Moreover,

$$\mathcal{S}_{j,l,m,n} = O(\delta^{n+1}).$$

(ii) Regarded as an operator from $\mathcal{H}^*(\partial D_j)$ *into* $\mathcal{H}^*(\partial D_l)$*, we have*

$$\mathcal{K}^k_{D_j,D_l} = \mathcal{K}_{j,l,0,0} + O(k^2\delta^2).$$

Moreover,

$$\mathcal{K}_{j,l,0,0} = O(\delta^2).$$

B.3 Asymptotic expansions in $\mathbb{R}^2$

Let us now consider the single-layer potential for the Helmholtz equation in $\mathbb{R}^2$ given by

$$\mathcal{S}^k_D[\varphi](x) = \int_{\partial D} G(x,y,k)\varphi(y)d\sigma(y), \quad x \in \partial D,$$

where $G(x,y,k) = -\frac{i}{4}H_0^{(1)}(k|x-y|)$ and $H_0^{(1)}$ is the Hankel function of first kind and order 0. We have, for $k \ll 1$,

$$-\frac{i}{4}H_0^{(1)}(k|x-y|) = \frac{1}{2\pi}\log|x-y| + \tau_k + \sum_{j=1}^{\infty}(b_j \log k|x-y| + c_j)(k|x-y|)^{2j},$$

where

$$\tau_k = \frac{1}{2\pi}(\log k + \gamma_e - \log 2) - \frac{i}{4}, \quad b_j = \frac{(-1)^j}{2\pi}\frac{1}{2^{2j}(j!)^2}, \quad c_j = -bj\left(\gamma_e - \log 2 - \frac{i\pi}{2} - \sum_{n=1}^{j}\frac{1}{n}\right),$$

and γ_e is the Euler constant. Thus, we get

$$\mathcal{S}_D^k = \hat{\mathcal{S}}_D^k + \sum_{j=1}^{\infty} \left(k^{2j} \log k\right) \mathcal{S}_{D,j}^{(1)} + \sum_{j=1}^{\infty} k^{2j} \mathcal{S}_{D,j}^{(2)}, \tag{B.10}$$

where

$$\begin{aligned}
\hat{\mathcal{S}}_D^k[\varphi](x) &= \mathcal{S}_D[\varphi](x) + \tau_k \int_{\partial D} \varphi d\sigma, \\
\mathcal{S}_{D,j}^{(1)}[\varphi](x) &= \int_{\partial D} b_j |x-y|^{2j} \varphi(y) d\sigma(y), \\
\mathcal{S}_{D,j}^{(2)}[\varphi](x) &= \int_{\partial D} |x-y|^{2j} (b_j \log|x-y| + c_j) \varphi(y) d\sigma(y).
\end{aligned}$$

Lemma B.3.1. *The norms* $\|\mathcal{S}_{D,j}^{(1)}\|_{\mathcal{L}(\mathcal{H}^*(\partial D),\mathcal{H}(\partial D))}$ *and* $\|\mathcal{S}_{D,j}^{(2)}\|_{\mathcal{L}(\mathcal{H}^*(\partial D),\mathcal{H}(\partial D))}$ *are uniformly bounded with respect to* j*. Moreover, the series in* (B.10) *is convergent in* $\mathcal{L}(\mathcal{H}^*(\partial D), \mathcal{H}(\partial D))$ *for* $k < 1$*.*

Observe that

$$\left(\mathcal{S}_D - \widetilde{\mathcal{S}}_D\right)[\varphi] = \left(\mathcal{S}_D - \widetilde{\mathcal{S}}_D\right)[\mathcal{P}_{\mathcal{H}_0^*}[\varphi] + (\varphi, \varphi_0)_{\mathcal{H}^*} \varphi_0] = (\varphi, \varphi_0)_{\mathcal{H}^*} \left(\mathcal{S}_D[\varphi_0] + 1\right).$$

Then it follows that

$$\hat{\mathcal{S}}_D^k[\varphi] = \widetilde{\mathcal{S}}_D[\varphi] + (\varphi, \varphi_0)_{\mathcal{H}^*} \left(\mathcal{S}_D[\varphi_0] + 1\right) + \tau_k \int_{\partial D} \mathcal{P}_{\mathcal{H}_0^*}[\varphi] + (\varphi, \varphi_0)_{\mathcal{H}^*} \varphi_0 d\sigma = \widetilde{\mathcal{S}}_D[\varphi] + \Upsilon_k[\varphi],$$

where

$$\Upsilon_k[\varphi] = (\varphi, \varphi_0)_{\mathcal{H}^*} \left(\mathcal{S}_D[\varphi_0] + 1 + \tau_k\right). \tag{B.11}$$

Therefore, we arrive at the following result.

Lemma B.3.2. *For* k *small enough,* $\hat{\mathcal{S}}_D^k : \mathcal{H}^*(\partial D) \to \mathcal{H}(\partial D)$ *is invertible.*

Proof. Υ_k is clearly a compact operator. Since $\widetilde{\mathcal{S}}_D$ is invertible, the invertibility of $\hat{\mathcal{S}}_D^k$ is equivalent to that of $\hat{\mathcal{S}}_D^k \widetilde{\mathcal{S}}_D^{-1} = I + \Upsilon_k \widetilde{\mathcal{S}}_D^{-1}$. By the Fredholm alternative, we only need to prove the injectivity of $I + \Upsilon_k \widetilde{\mathcal{S}}_D^{-1}$.
Since $\forall\, v \in H^{1/2}(\partial D)$, $\Upsilon_k \widetilde{\mathcal{S}}_D^{-1}[v] \in \mathbb{C}$, for $\left(I + \Upsilon_k \widetilde{\mathcal{S}}_D^{-1}\right)[v] = 0$, we need to show that $v = \widetilde{\mathcal{S}}_D[\alpha\varphi_0] = -\alpha \in \mathbb{C}$.
We have

$$\left(I + \Upsilon_k \widetilde{\mathcal{S}}_D^{-1}\right) \widetilde{\mathcal{S}}_D[\alpha\varphi_0] = \alpha(\mathcal{S}_D[\varphi_0] + \tau_k) = 0 \quad \text{iff} \quad \mathcal{S}_D[\varphi_0] = -\tau_k \text{ or } \alpha = 0.$$

Since we can always find a small enough k such that $\mathcal{S}_D[\varphi_0] \neq -\tau_k$, we need $\alpha = 0$. This yields the stated result. □

Lemma B.3.3. *For* k *small enough, the operator* $\mathcal{S}_D^k : \mathcal{H}^*(\partial D) \to \mathcal{H}(\partial D)$ *is invertible.*

Proof. The operator $\mathcal{S}_D^k - \hat{\mathcal{S}}_D^k : \mathcal{H}^*(\partial D) \to \mathcal{H}(\partial D)$ is a compact operator. Because $\hat{\mathcal{S}}_D^k$ is invertible for k small enough, by the Fredholm alternative only the injectivity of $\mathcal{S}_D^k$ is necessary. From the uniqueness of a solution to the Helmholtz equation we get the result. □

Lemma B.3.4. *The following asymptotic expansion holds for k small enough:*

$$(\mathcal{S}_D^k)^{-1} = \mathcal{P}_{\mathcal{H}_0^*}\widetilde{\mathcal{S}}_D^{-1} + \mathcal{U}_k - k^2\log k\mathcal{P}_{\mathcal{H}_0^*}\widetilde{\mathcal{S}}_D^{-1}\mathcal{S}_{D,1}^{(1)}\mathcal{P}_{\mathcal{H}_0^*}\widetilde{\mathcal{S}}_D^{-1} + O(k^2)$$

with

$$\mathcal{U}_k = -\frac{(\widetilde{\mathcal{S}}_D^{-1}[\cdot],\varphi_0)_{\mathcal{H}^*}}{\mathcal{S}_D[\varphi_0]+\tau_k}\varphi_0. \tag{B.12}$$

Note that $\mathcal{U}_k = O(1/\log k)$.

Proof. We can write (B.10) as

$$\mathcal{S}_D^k = \hat{\mathcal{S}}_D^k + \mathcal{G}_k,$$

where $\mathcal{G}_k = k^2\log k\mathcal{S}_{D,1}^{(1)} + O(k^2)$. From Lemma B.3.2 and Lemma B.3.3 we get the identity

$$(\mathcal{S}_D^k)^{-1} = \left(I + (\hat{\mathcal{S}}_D^k)^{-1}\mathcal{G}_k\right)^{-1}(\hat{\mathcal{S}}_D^k)^{-1}.$$

Hence, we have

$$(\hat{\mathcal{S}}_D^k)^{-1} = \underbrace{\left(\widetilde{\mathcal{S}}_D^{-1}\hat{\mathcal{S}}_D^k\right)^{-1}}_{\Lambda_k^{-1}}\widetilde{\mathcal{S}}_D^{-1}.$$

Here,

$$\begin{aligned}\Lambda_k &= I - (\cdot,\varphi_0)_{\mathcal{H}^*}(\mathcal{S}_D[\varphi_0]+1+\tau_k)\varphi_0\\ &= \mathcal{P}_{\mathcal{H}_0^*} - (\cdot,\varphi_0)_{\mathcal{H}^*}(\mathcal{S}_D[\varphi_0]+\tau_k)\varphi_0.\end{aligned}$$

Then,

$$\Lambda_k^{-1} = \mathcal{P}_{\mathcal{H}_0^*} - (\cdot,\varphi_0)_{\mathcal{H}^*}\frac{1}{\mathcal{S}_D[\varphi_0]+\tau_k}\varphi_0,$$

and therefore,

$$(\hat{\mathcal{S}}_D^k)^{-1} = \mathcal{P}_{\mathcal{H}_0^*}\widetilde{\mathcal{S}}_D^{-1} - \frac{(\widetilde{\mathcal{S}}_D^{-1}[\cdot],\varphi_0)_{\mathcal{H}^*}}{\mathcal{S}_D[\varphi_0]+\tau_k}\varphi_0.$$

It is clear that $\|(\hat{\mathcal{S}}_D^k)^{-1}\|_{\mathcal{L}(\mathcal{H}(\partial D),\mathcal{H}^*(\partial D))}$ is bounded for k small. Since $||\mathcal{G}_k||_{\mathcal{L}(\mathcal{H}(\partial D),\mathcal{H}^*(\partial D))}$ goes to zero as k goes to zero, for k small enough, we can write

$$(\mathcal{S}_D^k)^{-1} = (\hat{\mathcal{S}}_D^k)^{-1} - (\hat{\mathcal{S}}_D^k)^{-1}\mathcal{G}_k(\hat{\mathcal{S}}_D^k)^{-1} + O\left(k^4(\log k)^2\right),$$

which yields the desired result. □

We now consider the expansion for the boundary integral operator $(\mathcal{K}_D^k)^*$. We have

$$(\mathcal{K}_D^k)^* = \mathcal{K}_D^* + \sum_{j=1}^{\infty}\left(k^{2j}\log k\right)\mathcal{K}_{D,j}^{(1)} + \sum_{j=1}^{\infty}k^{2j}\mathcal{K}_{D,j}^{(2)}, \tag{B.13}$$

where

$$\begin{aligned}\mathcal{K}_{D,j}^{(1)}[\varphi](x) &= \int_{\partial D} b_j \frac{\partial |x-y|^{2j}}{\partial \nu(x)} \varphi(y) d\sigma(y),\\ \mathcal{K}_{D,j}^{(2)}[\varphi](x) &= \int_{\partial D} \frac{\partial \left(|x-y|^{2j}(b_j \log|x-y| + c_j)\right)}{\partial \nu(x)} \varphi(y) d\sigma(y).\end{aligned}$$

Lemma B.3.5. *The norms* $\|\mathcal{K}_{D,j}^{(1)}\|_{\mathcal{L}(\mathcal{H}^*(\partial D),\mathcal{H}^*(\partial D))}$ *and* $\|\mathcal{K}_{D,j}^{(2)}\|_{\mathcal{L}(\mathcal{H}^*(\partial D),\mathcal{H}^*(\partial D))}$ *are uniformly bounded for* $j \geq 1$. *Moreover, the series in (B.13) is convergent in* $\mathcal{L}(\mathcal{H}^*(\partial D), \mathcal{H}^*(\partial D))$.

Appendix C

Sum Rules for the Polarization Tensor

Let f be a holomorphic function defined in an open set $U \subset \mathbb{C}$ containing the spectrum of $\mathcal{K}_D^*$. Then, we can write $f(z) = \sum_{j=0}^{\infty} a_j z^j$ for every $z \in U$.

Definition C.1. *Let*

$$f(\mathcal{K}_D^*) := \sum_{j=0}^{\infty} a_j (\mathcal{K}_D^*)^j,$$

where $(\mathcal{K}_D^*)^j := \underbrace{\mathcal{K}_D^* \circ \mathcal{K}_D^* \circ .. \circ \mathcal{K}_D^*}_{j \text{ times}}$.

Lemma C.0.1. *We have*

$$f(\mathcal{K}_D^*) = \sum_{j=1}^{\infty} f(\lambda_j)(\cdot, \varphi_j)_{\mathcal{H}^*} \varphi_j.$$

Proof. We have

$$\begin{aligned} f(\mathcal{K}_D^*) &= \sum_{i=0}^{\infty} a_i (\mathcal{K}_D^*)^i = \sum_{i=0}^{\infty} a_i \sum_{j=1}^{\infty} \lambda_j^i (\cdot, \varphi_j)_{\mathcal{H}^*} \varphi_j \\ &= \sum_{j=1}^{\infty} \left(\sum_{i=0}^{\infty} a_i \lambda_j^i \right) (\cdot, \varphi_j)_{\mathcal{H}^*} \varphi_j \\ &= \sum_{j=1}^{\infty} f(\lambda_j)(\cdot, \varphi_j)_{\mathcal{H}^*} \varphi_j. \end{aligned}$$

□

From Lemma C.0.1, we can deduce that

$$\int_{\partial D} x_l f(\mathcal{K}_D^*)[\nu_m](x)\, d\sigma(x) = \sum_{j=1}^{\infty} f(\lambda_j) \alpha_{l,m}^{(j)}. \tag{C.1}$$

Equation (C.1) yields the summation rules for the entries of the polarization tensor.

In order to prove that $\sum_{j=1}^{\infty} \alpha_{l,m}^{(j)} = \delta_{l,m}|D|$, we take $f(\lambda) = 1$ in (C.1) to get

$$\sum_{j=1}^{\infty} \alpha_{l,m}^{(j)} = \int_{\partial D} x_l \nu_m(x)\, d\sigma(x) = \delta_{l,m}|D|.$$

Next, we prove that

$$\sum_{j=1}^{\infty} \lambda_j \sum_{l=1}^{d} \alpha_{l,l}^{(j)} = \frac{(d-2)}{2}|D|.$$

Taking $f(\lambda) = \lambda$ in (C.1), we obtain

$$\begin{aligned}
\sum_{j=1}^{\infty} \lambda_j \sum_{l=1}^{d} \alpha_{l,l}^{(j)} &= \sum_{l=1}^{d} \int_{\partial D} x_l \mathcal{K}_D^*[\nu_l](x)\, d\sigma(x), \\
\int_{\partial D} x_l \mathcal{K}_D^*[\nu_l](x)\, d\sigma(x) &= \int_{\partial D} x_l \left(\frac{1}{2}\nu_l(x) + \frac{\partial \mathcal{S}_D[\nu_l]}{\partial \nu}\Big|_-(x) \right) d\sigma(x), \\
&= \frac{|D|}{2} + \int_{\partial D} x_l \frac{\partial \mathcal{S}_D[\nu_l]}{\partial \nu}\Big|_-(x) d\sigma(x). \qquad \text{(C.2)}
\end{aligned}$$

Integrating by parts we arrive at

$$\int_{\partial D} x_l \frac{\partial \mathcal{S}_D[\nu_l]}{\partial \nu}\Big|_-(x) d\sigma(x) = \int_D e_l(x) \cdot \nabla \mathcal{S}_D[\nu_l](x) dx + \int_D x_l \Delta \mathcal{S}_D[\nu_l](x) dx.$$

Since the single-layer potential is harmonic on D,

$$\int_{\partial D} x_l \frac{\partial \mathcal{S}_D[\nu_l]}{\partial \nu}\Big|_-(x) d\sigma(x) = \int_D e_l(x) \cdot \left(\int_{\partial D} \nabla_x \Gamma(x, x') \nu_l(x') d\sigma(x') \right) dx.$$

Summing on i and using $\nabla_x \Gamma(x, x') = -\nabla_{x'} \Gamma(x, x')$, we get

$$\begin{aligned}
\sum_{l=1}^{d} \int_{\partial D} x_l \frac{\partial \mathcal{S}_D[\nu_l]}{\partial \nu}\Big|_-(x) d\sigma(x) &= -\int_D \left(\int_{\partial D} \nu(x') \cdot \nabla_{x'} \Gamma(x, x') d\sigma(x') \right) dx, \\
&= -\int_D \mathcal{D}_D[1](x) dx, \\
&= -|D|, \qquad \text{(C.3)}
\end{aligned}$$

where $\mathcal{D}_D$ is the double-layer potential. Hence, summing equation (C.2) for $i = 1, \ldots, d$, we get the result.

Finally, we show that

$$\sum_{j=1}^{\infty} \lambda_j^2 \sum_{l=1}^{d} \alpha_{l,l}^{(j)} = \frac{d-4}{4}|D| + \sum_{l=1}^{d} \int_D |\nabla \mathcal{S}_D[\nu_l]|^2 dx.$$

Taking $f(\lambda) = \lambda^2$ in (C.1) yields

$$\begin{aligned}
\sum_{j=1}^{\infty} \lambda_j^2 \sum_{l=1}^{d} \alpha_{l,l}^{(j)} &= \sum_{l=1}^{d} \int_{\partial D} x_l (\mathcal{K}_D^*)^2[\nu_l](x)\, d\sigma(x) \\
&= \sum_{l=1}^{d} \int_{\partial D} \mathcal{K}_D[y_l](x) \mathcal{K}_D^*[\nu_l](x)\, d\sigma(x) \\
&= \sum_{l=1}^{d} \int_{\partial D} \mathcal{K}_D[y_l] \frac{\nu_l}{2} d\sigma + \sum_{l=1}^{d} \int_{\partial D} \mathcal{K}_D[y_l] \frac{\partial \mathcal{S}_D[\nu_l]}{\partial \nu}|_- d\sigma \\
&= \frac{(d-2)}{4}|D| - \underbrace{\sum_{l=1}^{d} \int_{\partial D} \frac{y_l}{2} \frac{\partial \mathcal{S}_D[\nu_l]}{\partial \nu}\Big|_- d\sigma}_{I_1} + \underbrace{\sum_{l=1}^{d} \int_{\partial D} \mathcal{D}_D[y_l]\Big|_- \frac{\partial \mathcal{S}_D[\nu_l]}{\partial \nu}\Big|_- d\sigma}_{I_2}.
\end{aligned}$$

From (C.3) it follows that

$$I_1 = -\frac{|D|}{2}.$$

Since x_l is harmonic, we have $x_l = \mathcal{D}_D[y_l](x)|_- - \mathcal{S}_D[\nu_l](x)$ on ∂D, and thus,

$$\begin{aligned}
I_2 &= \sum_{l=1}^{d} \int_{\partial D} (x_l + \mathcal{S}_D[\nu_l](x)) \frac{\partial \mathcal{S}_D[\nu_l]}{\partial \nu}\Big|_- (x) d\sigma(x), \\
&= -|D| + \sum_{l=1}^{d} \int_{\partial D} \mathcal{S}_D[\nu_l] \frac{\partial \mathcal{S}_D[\nu_l]}{\partial \nu}\Big|_- d\sigma, \\
&= -|D| + \sum_{l=1}^{d} \int_{D} |\nabla \mathcal{S}_D[\nu_l]|^2 dx.
\end{aligned}$$

Replacing I_1 and I_2 by their expressions gives the desired result.

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Résumé

Cette thèse porte sur l'étude mathématique des interactions entre la lumière et certains types de nanoparticules. A l'échelle du nanomètre, des particules métalliques comme l'or ou l'argent subissent un phénomène de résonance lorsque leurs électrons libres interagissent avec un champ électromagnétique. Cette interaction produit une augmentation du champs électrique proche et lointain, leur permettant d'améliorer la luminosité et la directivité de la lumière, confinant des champs électromagnétiques dans des directions avantageuses. Ce phénomène, appelé "résonances plasmoniques pour des nanoparticules" ouvre une porte sur une large gamme d'applications, des nouvelles techniques d'imagerie médicale à des panneaux solaires efficaces. En utilisant des techniques issues des potentiels de couches et de la théorie de la perturbation, nous proposons une étude de la dispersion d'ondes électromagnétiques par une et plusieurs nanoparticules plasmoniques, dans le cadre quasi-statique, Helmholtz et Maxwell. Nous étudions ensuite certaines applications tel que la génération de chaleur, les métasurfaces et l'imagerie super-résolue.

Mots Clés

operateur de Neumann-Poincaré, potentiels de couche, nanoparticules plasmoniques, resonance de plasmon, analyse asymptotique

Abstract

This thesis deals with the mathematical study of the interactions between light and certain types of nanoparticles. At the nanometer scale, metal particles such as gold or silver undergo a resonance phenomenon when their free electrons interact with an electromagnetic field. This interaction results in an enhancement of the near and far electric field, enabling them to improve the brightness and the directivity of the light, confining electromagnetic fields in advantageous directions. This phenomenon, called "plasmonic resonances for nanoparticles", opens a door to a wide range of applications, from new medical imaging techniques to efficient solar panels. Using layer potentials techniques and perturbation theory, we propose a study of the scattering of electromagnetic waves by one and several plasmonic nanoparticles in the quasi-static, Helmholtz and Maxwell framework. We then study some applications such as heat generation, metasurfaces and super-resolution.

Keywords

Neumann Poincaré operator, layer potentials, plasmonic nanoparticles, plasmonic resonances, asymptotic analysis