

Acousto-optic and photoacoustic imaging of scattering media using wavefront adaptive holography techniques in NdYO4

Baptiste Jayet

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Baptiste Jayet. Acousto-optic and photoacoustic imaging of scattering media using wavefront adaptive holography techniques in NdYO4. Other [cond-mat.other]. Université Pierre et Marie Curie - Paris VI, 2015. English. NNT: 2015PA066039. tel-01137779

HAL Id: tel-01137779 https://theses.hal.science/tel-01137779

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THÈSE DE DOCTORAT DE L'UNIVERSITÉ PIERRE ET MARIE CURIE

Spécialité

Optique physique expérimentale

(ED 564 - Physique en Île-de-France)

Préparée à l'Institut Langevin - Ondes et Images

Présentée par

Baptiste Jayet

Pour obtenir le grade de

DOCTEUR de l'UNIVERSITÉ PIERRE ET MARIE CURIE

Sujet de la thèse :

Acousto-optic and photoacoustic imaging of scattering media using wavefront adaptive holography techniques in $Nd:YVO_4$

Soutenue publiquement le 04/02/2015

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Remerciements

Bien que ce manuscrit n'ait qu'un unique auteur, sa réalisation a grandement bénéficié des soutiens, encouragements et aides que j'ai pu trouver tant au sein de l'Institut Langevin que dans mon entourage. Je tiens donc maintenant à remercier toutes ces personnes, ami(e)s, familles, collègues, ou simples connaissances, dont la présence m'a été d'une grande aide pour l'achèvement de ces travaux.

Tout d'abord je souhaite remercier mon directeur de thèse, François Ramaz, pour la confiance qu'il m'a accordée. Son soutien s'est manifesté dès le début par la recherche de financement pour le projet. Puis tout au long de la thèse à travers d'une part son enthousiasme pour mes résultats, souvent plus grand que le miens. Et d'autre part, par la grande autonomie qu'il m'a laissée ce qui m'as permis d'explorer, de proposer des idées (plus au moins farfelues), de les tester, bref... de chercher!

Quand on commence dans un domaine nouveau et qu'on arrive un peu perdu, avoir un interlocuteur d'expérience est un vrai bonus. Pour cela je tiens à remercier Jean-Pierre Huignard pour les longues discussions que l'on a eues sur la physique des mélanges d'ondes, la diffusion multiple, la modulation acousto-optique et bien d'autres sujets... Son expérience en holographie dynamique m'a été d'une grande aide tout au long de ces trois ans.

Si j'ai pu travailler sur ce projet, c'est aussi grâce aux financements qui lui ont été accordés. Je tiens donc à remercier la Direction Générale de l'Armement et l'Agence National de la Recherche pour nous avoir fourni les moyens financiers nécessaires à l'achat de matériel et pour que je puisse manger pendant ces trois ans.

Ma thèse s'est conclue par la rédaction de ce manuscrit et par la soutenance de mes travaux. Ma gratitude va donc à Anabela Da Silva pour avoir pris le temps de rapporter cette thèse ainsi que pour ses commentaires sur le manuscrit et ma présentation. *I would like add Michael Damzen to these acknowledgments. He also accepted to read and evaluate my work had very kind comments regarding my manuscript and my presentation.* Je remercie aussi les autres membres du jury, Arnaud Brignon et Claude Fabre pour l'intérêt qu'ils ont porté à ce travail. Enfin je souhaite aussi remercier Philippe Adam, notre correspondant avec la DGA, pour sa curiosité envers mon travail à travers les discussions que nous avons eues, notamment durant les longues heures de vol vers San Francisco pour *Photonic West*.

Je souhaite remercier l'ensemble des membres de l'Institut Langevin pour leur accueil chaleureux. J'ai vraiment pris plaisir à avoir de nombreuses discussions et débats quotidiens avec plein de gens différents sur des sujets pas nécessairement scientifiques.

Mes remerciements s'adressent aussi à l'ensemble du personnel technique et de gestion du laboratoire qui m'a aidé pour les nombreuses tâches administratives et les commandes de matériel.

Un merci particulier s'adresse à celles et ceux avec qui j'ai travaillé durant ces trois années. D'abord François et Jean-Pierre, cité plus haut, mais aussi les doctorants qui m'ont précédés, Max Lesaffre et Salma Farahi qui m'ont éclairés au départ sur l'acoustooptique. Puis les doctorants et post-doctorants avec qui j'ai partagé la salle de manip et les problèmes expérimentaux, Emilie Benoit, Jean-Baptiste Laudereau, Clément Dupuy et Kevin Contreras. A cela viennent s'ajouter Emmanuel Bossy, Olivier Simandoux, Florian Poisson et Jérôme Gateau pour leurs conseils sur la photoacoustique, et le prêt du laser nanoseconde pour mes expériences du dernier chapitre!

Comment ne pas remercier les joyeux lurons qui ont partager mon quotidien dans mes deux bureaux. D'abord à l'ESPCI : Marc, Karlaa et Emilie, puis après le déménagement, dans le bureau R31 à l'IPG : Emilie (et oui encore!), Nicolas V., Camille, Sylvain, Fabien, Amir, Anne (la génération initiale!) et JB, Olivier T. (continue à parler Handball avec les autres, même quand je ne serai plus la!!), Vincent, Marion (bon courage dans ce bureau masculin!), Clément D., Peng, et sans oublier ni le ficus, ni Yann "Présence" Desailly (les nouveaux occupants!). Merci à vous pour les multiples discussions, utiles et inutiles qu'on a pu avoir, les gâteaux partagés au moment du goûter, les moments en dehors du labo, et la bonne ambiance qui fait que même quand on a la flemme de travailler on vient quand même au labo, et du coup on travaille histoire de ne pas être venu pour rien! Pour ceux d'entre vous qui sont encore là, gardez cette ambiance, essentielle pour avoir une bonne santé mentale! Je vous souhaite bon courage pour la suite (et notamment pour la rédaction)!

Ils et elles n'ont pas partagé mon bureau, mais ça ne nous a pas empêché de passer plein de bon moment. Un merci particulier va donc à Ariadna, Daria et Francois B. pour votre bonne humeur, les pauses cafés, les soirées et les débats (surtout avec Daria!). Merci aussi à Olivier S. pour tes nombreuses questions de physique qui m'ont fait beaucoup re-réfléchir!

Merci également a tous les autres doctorants et post-doctorants du laboratoire, Pierre, Thomas, Matthieu, Nadège (je repars un peu triste qu'on ai pas fini OF), Florian, Lermu, Charlie, Clément P., Nikos, Thu-Mai,... et bien d'autres pour la bonne ambiance générale, les pauses midi avec QPUC, Haxball, et autre jeux sur internet, les verres du vendredi (et des autres soirs)! Je vous souhaite bon courage à tous pour la suite, et particulièrement à ceux et celles d'entre vous qui vont démarrer la rédaction de leur thèse! En dehors du laboratoire je souhaite remercier mes amis d'horizons divers, de Meudon, du Hand,... pour leur présence! Merci particulièrement à ceux qui se sont prêtés au jeu de la relecture de mon manuscrit (sans vraiment comprendre pour certains) : Olivier, Manu, Jonas, Benoit et Dr. BenJ.

Enfin mes derniers remerciements vont à ma famille pour leur soutien. Un merci particulier à mes parents qui m'ont toujours fait confiance dans mes études et m'ont laissé une liberté totale dans mes choix d'orientation en faisant en sorte que je puisse faire mes études dans de bonnes conditions! Je remercie aussi mes frères Cyril (merci pour la relecture d'un chapitre!) et Mikaël ainsi que ma belle-soeur Laetitia pour leur soutien! J'espère que les deux petits nouveaux membres de la famille, Aliénor et Constantin, que j'ai eu la joie d'accueillir pendant cette thèse, s'intéresseront de près ou de loin à la science.

Merci à tous!

Résumé

L'imagerie optique des tissus biologiques est un défi du fait de la diffusion de la lumière. Pour sonder les propriétés optiques à quelques cm de profondeur, on peut coupler l'information optique des ultrasons. De cette idée sont nées les imageries acousto-optique et photoacoustique. La première repose sur la modulation de la lumière par des ultrasons balistiques. La seconde se base sur la génération d'ultrasons lors de l'absorption de lumière par un objet.

Que ce soit pour l'une ou pour l'autre, l'enregistrement du signal nécessite la mesure de très faibles modulations de phase dans une figure de speckle. L'holographie dynamique est une bonne solution. En effet, les techniques interférométriques sont suffisamment sensibles pour mesurer de telles modulations et l'holographie permet de corriger la nature speckle de la lumière. Dans cette thèse nous démontrons la faisabilité de fabriquer un système d'holographie adaptative basé sur un milieu laser (Nd:YVO₄). Un des grands avantages de ce type de milieu est le temps de réponse. On montrera que le rafraîchissement d'un hologramme dans notre cristal peut se faire en moins de 100 µs, bien inférieur au temps de décorrélation du speckle (\approx ms) qui pourrait grandement perturber les techniques de détection plus lentes lors d'expériences *in vivo*. Trois montages sont présentés ici, le premier pour la détection acousto-optique par adaptation de front d'onde et enfin le troisième pour détection photoacoustique. Dans les trois cas on mesure un temps de réponse entre 15 µs et 50 µs, et on utilise le montage pour imager un échantillon.

Mots clés : Imagerie acousto-optique, Imagerie photoacoustique, Holographie dynamique, Conjugaison de phase, Lasers, Milieu diffusant.

Abstract

Strong scattering properties of biological media make their optical imaging in depth a challenge. A solution to probe the local optical properties is to couple the optical information with ultrasound. Two imaging techniques were born from this idea, acousto-optic imaging and photoacoustic imaging. The first technique is based on the local modulation of light by ballistic ultrasound. The latter relies on the emission of ultrasound following the absorption of light by an object.

Whether it is acousto-optic imaging or photoacoustic imaging, the recording of the the signal requires a detection system sensitive to weak phase modulation. In addition, the detection system must be compatible with a speckle pattern. Dynamic holography is a good solution. Indeed, as it is based on interferometry, it is very sensitive to small phase variations and holography can be used to correct the speckle nature of light. In this manuscript, we show the use of an holographic detection system based on a laser medium (Nd:YVO₄). One of the main advantage of this type of material is the very fast response time. It will be highlighted that the recording of a hologram inside our crystal can be done in less than 100 µs, much faster than the speckle decorrelation time (\approx ms), which is one of the major obstacle towards *in vivo* imaging. Three optical setups will be presented in this manuscript. The first one is a phase conjugation setup for acousto-optic detection. Finally, the third setup is an adaptive vibrometry setup for photoacoustic detection. In each setups the measured response time is between 15 µs and 50 µs.

Keywords: Acousto-optic imaging, Photoacoustic imaging, Dynamic holography, Phase conjugation, Lasers, Scattering medium.

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Introduction

In order to diagnose pathologies, physicians need accurate information about the body. Likewise, biologists also look for information within biological tissues to have a better understanding of the living. These information can take numerous forms such as qualitative descriptions (sensations, pain,...), values over time (cardiac frequency, blood pressure,...) and sometime images. To see what is inside a biological body, the most obvious path is to send light through the body and try to detect something on the other side. Very quickly, researchers and physicians noticed that light did not work well on biological tissues because of their very strong scattering properties.

Physicists looked for new methods of imaging biological tissues. It is during the 20th century that most of the biomedical imaging techniques used nowadays have been developed. Indeed, the discoveries of radioactivity, X-rays, piezoelectricity, and nuclear magnetic resonance have enabled the invention of new imaging modalities such as PET, X-ray imaging, ultrasound imaging and MRI. These very powerful methods have been rapidly adopted by physicians and biologists in order to get information from deep inside human and animal bodies. They all worked really well because they rely on waves (ultrasonic waves, RF electromagnetic waves,...) that propagate well inside biological tissues. Due to these good performances, the biomedical community lost interest into biomedical optics for a few decades. Nevertheless, recently the interest for optics in biology came back because of the limitations of certain imaging techniques. Indeed, as an example, the early detection of tumours remains a challenge, yet, it is crucial in cancer treatment. The difficulty resides in the small size of early tumours and in the fact that current techniques cannot always differentiate healthy tissues from cancer cells.

Optic, because it provides access to a lot of new information, can be a great support to current imaging techniques. As light interacts with matter constituents, optical imaging gives access to information such as chemical composition. In addition, the changes in composition of biological tissues can be linked to structural information. If added to current imaging techniques, the richness of information provided by optics can greatly enhance medical diagnosis and biological research. To overcome the problems of light scattering, physicists developed promising new techniques coupling light with ultrasound in order to retrieve the optical information deep inside tissues with the good resolution provided by ultrasound imaging. From these investigations, acousto-optic and photoacoustic imaging were born during the 1990s. These two imaging techniques are based on the eponymous effects.

Acousto-optic is the modulation of light with an acoustic field. By locally tagging photons within a biological tissue with an acoustic field - in the ultrasound domain it is possible to retrieve the local optical properties of the sample. The main challenge is the detection of the so called *tagged photons*, since there are very few of them and the tagging corresponds to a very small frequency shift compared to the light frequency. During the past twenty years, research was aimed at developing detection techniques of the tagged photons to retrieve acousto-optic signals with a good SNR. They were based either on narrow spectral filtering or on adaptive holography. The team at Institut Langevin, formerly Laboratoire d'Optique Physique, worked on acousto-optic imaging for the past fifteen years. They mainly developed detection methods based on adaptive holography using the photorefractive effect or using digital holography. This led to promising results on phantoms or ex vivo samples. The next step, performing real in vivo imaging, requires to overcome the issue of speckle decorrelation. This concern, shared with the other teams around the world working with coherent detections, can be a major obstacle since the light coming from a living sample is not stable in time. Due to inherent biological movements (blood flow, heart beats, breathing,...), it decorrelates in around 1 ms. Hence, holographic systems need to be able to record holograms faster than this decorrelation time. In common photorefractive crystals, the response time is usually between 1 ms and 100 ms, which may not be fast enough for *in vivo* imaging.

This was the main motivation for the work described in this manuscript. Our goal was to demonstrate the feasibility of an holographic detection system based on a gain medium (Nd:YVO₄) with a sub-millisecond response time. In addition, our optical setup was used for another biomedical imaging technique: photoacoustic imaging. We managed to perform the detection of photoacoustic signals without any contact on the sample.

This manuscript is organised into three parts. The first part describes, through two chapters, light propagation in biological tissues and how it can be used to perform imaging. The first chapter is dedicated to the optical properties of biological tissues and the propagation of light inside them. Chapter 2 describes the interactions of light with ultrasound and how they can be used to perform acousto-optic imaging and photoacoustic imaging. The second part of the manuscript is dedicated to the main tool used for the detection of acousto-optic and photoacoustic signal: holography in gain media. It is divided into two chapters. The first one explains the processes of amplification and wave mixing in a gain medium. And the second one details the particular cases of two-wave mixing and four-wave mixing through mathematical models. Finally, part III of this manuscript reports our experimental investigations. It is divided into three chapters. First, chapter 5 shows the detection of acousto-optic signals using phase conjugation by four-wave mixing in Nd:YVO₄. We demonstrate acousto-optic imaging with a response time around 15 µs. In chapter 6, another geometric configuration is investigated. It produced interesting results using wavefront adaption by two-wave mixing as a detection process. We performed

the acousto-optic imaging of a phantom with a response time of $45 \,\mu\text{s}$. The final chapter explores the use of two-wave mixing for the detection of vibration at the surface of a multiple scattering sample. We applied this detection to contactless photoacoustic imaging of wires hidden behind chicken breast with a response time of $25 \,\mu\text{s}$.

The conclusion of this manuscript will sum up the work achieved during the past three years. Then the perspectives opened by this work will be discussed in order to propose new ideas. The stakes being either the improvements of biomedical imaging (acousto-optic and photoacoustic) or the development of new applications of holography in gain media non necessarily related to biomedical imaging, *e.g.*, non-destructive testing.

Part I

Optical imaging of biological media

chapter 1

Optical properties of biological media

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B iological tissues are composed of complex structures (cells, membranes, fiber,...) that make the propagation of light very complicated. These structures strongly interact with light through two main phenomena: absorption and scattering. The first decreases the intensity of the light whereas the latter changes the trajectory of the photons travelling through the sample. This results in a large dispersion of light in every direction after a few millimetres of propagation in a biological tissue.

This chapter starts by presenting the different phenomena that govern the interactions of light with matter. We will see the different regimes of propagation and how they affect light along its propagation in a biological sample. Then, we will expose the interests of optical imaging inside biological media despite the complications due to the strong interaction of light with those media. Finally, we will discuss the different solutions that currently exist in order to retrieve the optical properties of biological tissues.

1.1 Propagation of light in multiple scattering media.

1.1.1 Interaction of light with matter.

The two main phenomena that occur when light propagates inside a multiple scattering medium are absorption and scattering. Depending on their relative magnitude, they can have a strong effect on how light exit a sample and on our ability to get optical information from inside the sample.

Absorption Absorption is the result of the interaction of light with the transition levels of the atoms (electronic transition) and/or molecules (electronic, vibrational,...) that compose the medium. If the energy of the photons matches a transition energy, it can be absorbed. The absorbed energy is then dissipated through various processes that can be non radiative, *e. g.* heat, or radiative, *e. g.* fluorescence or phosphorescence. Absorption is characterised by the absorption coefficient μ_a which links the decrease of the *flux* of a collimated light beam to the distance travelled by light in the medium through the following equation:

$$\Phi(z) = \Phi_0 \ e^{-\mu_a z} \tag{1.1}$$

where $\Phi(z)$ is the *flux* at depth z, Φ_0 is the incident *flux* and z is the distance travelled by light. The absorption coefficient μ_a is usually expressed in cm⁻¹. This relation is known as the Beer-Lambert law and is valid for non scattering medium. Absorption can also be described by the absorption mean free path l_a , which is the inverse of the absorption coefficient $l_a = 1/\mu_a$. **Scattering** Some media have a complicated microstructure, their refractive index is inhomogeneous at the microscopic scale or less. When light travels in such medium, it takes random path because it encounters scatterers, which randomly change the direction of propagation as illustrated in Fig. 1.1. As a result, photons exit the sample with a



Figure 1.1 – Trajectories of photons inside a scattering medium.

random direction and phase, this process is called scattering. It can be described by Mie theory which works for inhomogeneities, or scatterers, that have the size of the wavelength or larger. For scatterers much smaller than the wavelength, approximation can be made and Mie scattering is simplified into Rayleigh scattering.

In the same way absorption is described by the absorption coefficient μ_a , scattering is described by the scattering coefficient μ_s . It characterises the loss of photons travelling in a straight line through the sample, which we will call ballistic light. This coefficient is expressed in cm⁻¹, so the *flux* transmitted in a ballistic way trough a medium of thickness z can be written:

$$\Phi_{bal}(z) = \Phi_0 \ e^{-\mu_s z} \tag{1.2}$$

The scattering mean free path, which is the inverse of the scattering coefficient: $l_s = 1/\mu_s$, can be interpreted as the average distance travelled by a photon between two scattering events.

1.1.2 Multiple scattering and diffusion regime.

The scattering mean free path can be used to introduce the notion of multiple scattering. When propagating through a medium, photons can be scattered many times depending on the relative thickness of the medium compared to its scattering mean free path. Three regimes can be defined for a medium with a thickness L:

- $L \ll l_s$: Ballistic regime, photons are almost not scattered.
- $L \simeq l_s$: Simple scattering regime.
- $L \gg l_s$: Multiple scattering regime.

In the ballistic regime, light propagating in a ballistic way is almost not attenuated because of scattering, only by absorption (if the medium is absorbent). The multiple scattering regime is the opposite: the collimated beam is totally extinguished, the transport of the electromagnetic energy is done only through diffused light. Finally, the simple scattering regime is a combination of both: collimated light and diffused light coexist. **Extinction coefficient** In an absorbent and scattering medium, the loss of energy of a collimated beam is due to both absorption and scattering. It is then possible to define the extinction coefficient: $\mu_e = \mu_a + \mu_s$, which reflects the total attenuation of a collimated beam propagating through a medium with the relation: $\Phi_{col}(z) = \Phi_{col}(0) \exp(-\mu_e z)$.

Anisotropy In fact, when scatterers are larger than the wavelength, the probability distribution of θ , defined as the angle between the initial direction, and the direction after the scattering event (see Fig. 1.2), is no longer uniform. In this case the scattering is said to be anisotropic, as opposed to the isotropic scattering when the photons have the same probability to go in any direction. The anisotropy can be quantified using the anisotropy



Figure 1.2 – Isotropic scattering and anisotropic scattering.

coefficient defined as the expected value of the cosine of θ :

$$g = \langle \cos(\theta) \rangle \tag{1.3}$$

From this, it is possible to derive a new scattering coefficient μ'_s and a new mean free path l'_s , respectively called the reduced scattering coefficient and the transport mean free path. They can be expressed as functions of the scattering coefficient μ_s , the scattering mean free path l_s and the anistropy g:

$$\mu'_{s} = \mu_{s}(1-g) \tag{1.4}$$

$$l'_{s} = \frac{1}{\mu'_{s}} = \frac{l_{s}}{1-g} \tag{1.5}$$

The transport mean free path l'_s can be interpreted as the depth at which a photon, that underwent multiple scattering event, has lost the information about its initial direction and polarisation. The difference between l_s and l'_s is illustrated in Fig. 1.3.

Effective extinction coefficient If the thickness of the medium is larger than the transport mean free path, $L \gg l'_s$, it is possible to describe the total attenuation of light



Figure 1.3 – Scattering mean free path and transport mean free path.

by the effective extinction coefficient defined as:

$$\mu_{eff} = \sqrt{3\mu_a(\mu_a + \mu'_s)} \tag{1.6}$$

In this case, the flux of the light at a distance z from the entrance of the medium is given by:

$$\Phi(z) = \Phi_0 e^{-\mu_{eff} z} \tag{1.7}$$

1.1.3 Orders of magnitude in biological media.

Biological tissues are composed of a lot of different substances having various refractive indices. As an example, water has a refractive index of 1.33, lipids of 1.46 and proteins of 1.54. Depending on the exact composition of a tissue, its refractive index is usually between 1.33 and 1.5.

Optical therapeutic window Absorption in biological tissue is mainly due to water and haemoglobin. Their absorption spectra is given in Fig. 1.4, where we can see that haemoglobin strongly absorbs at wavelength shorter than 600 nm, and water is strongly absorbent at wavelength longer than 1300 nm. As a result, to exploit light for imaging inside biological media, one needs to stay within the so called *optical therapeutic window* which is usually the wavelength range [600 nm; 1300 nm].



Figure 1.4 – Absorption coefficient spectra of endogenous tissue chromophores. (taken from [1])

Scattering Light in the optical therapeutic window can propagate quite deeply, a few cm, without being absorbed in biological sample. Yet, imaging deep inside tissues remains a challenge due to their strong scattering properties. Despite a very strong scattering coefficient μ_s , between 50 cm⁻¹ and 200 cm⁻¹, light still propagates deeply in biological tissues because of the anisotropy of the scattering. Indeed, during a scattering event, photons are mainly forward scattered enabling a deep penetration of light inside tissues. The anisotropy coefficient is between 0.8 to 0.99, so the usual value of the reduced scattering coefficient used to describe tissues is $\mu'_s = 10 \text{ cm}^{-1}$.

Table 1.1 sums up the optical properties of biological sample in the optical therapeutic window. These values are issued from [2].

n	Refractive index	1.33 - 1.5
μ_a	Absorption coefficient	$10^{-2} \mathrm{cm}^{-1} - 10 \mathrm{cm}^{-1}$
μ_s	Scattering coefficient	$50{\rm cm^{-1}} - 200{\rm cm^{-1}}$
g	Anisotropy	0.9 - 0.99
μ'_s	Reduced scattering coefficient	$10 {\rm cm}^{-1}$
μ_{eff}	Effective extinction coefficient	$1.5{\rm cm^{-1}}-60{\rm cm^{-1}}$

Table 1.1 – Optical properties of biological tissues.

1.2 Optical imaging in biology and medicine - Interests and problems.

The variety of imaging techniques (MRI, ultrasound imaging, CT scan, X-ray imaging,...) that have been developed during the 20th century gives access to a lot of information on the studied subject, but they are mainly structural. They can be completed with methods such as Positron Emission Tomography (PET) to have metabolic information. In this section we first discuss how optical imaging can improve diagnosis by giving physicians new tools and new information to come up with accurate diagnosis and perform the right treatment. Indeed, as light interacts with the constituent of the body it can provide helpful information about composition or metabolism. The second part of this section is focused on the problems due to multiple scattering of light inside biological tissues and the difficulty of mapping their local optical properties.

1.2.1 Interest of optical imaging

Non-invasive Light in the visible and Near Infra-Red (NIR) part of the spectrum is attractive for biological imaging purposes. At these wavelengths, electromagnetic radiations are non-ionising and relatively harmless for biological tissues. Optical imaging is thus completely safe and provides information about tissues without causing any damage.

Enhancing detection Interaction of light with matter is mainly due to interaction of light with the atoms and molecules constituting the tissues. This means that the contrast given by an optical imaging technique can provide information about composition whereas techniques such as ultrasound imaging or X-Ray provide structural information. As an example, Pogue *et al.* showed in 2001 that diffuse optical tomography enabled to image haemoglobin concentration, which led them to image breast tumour (fibroadenoma and carcinoma) [3]. In 2014, Laudereau *et al.* used acousto-optic imaging in addition to ultrasound imaging to see tumour on a liver biopsy [4] as shown in Fig. 1.5. It represents the photography of an *ex vivo* liver sample with tumours (d), the ultrasound image(a) and the acousto-optic image (b) and (c). These two examples show that light has a great potential to add meaningful information to current imaging techniques.



Figure 1.5 – (a) Ultrasound image of a liver biopsy embedded in a scattering gel made of Agar and Intralipid. (b) Acousto-Optic image of the liver biopsy. (c) Acousto-Optic image with a subtracted envelop. (d) Picture of the biopsy before being embedded in the scattering gel. (taken from [4])

Metabolic information Figure 1.4, shows the absorption spectra of naturally present chromophores in biological tissues, especially haemoglobin and oxyhaemoglobin. There is a significant difference in the absorption coefficient of those two species for wavelengths between 750 nm and 800 nm. This means that it is theoretically possible to map the concentration of haemoglobin and oxyhaemoglobin by performing optical imaging at two different wavelengths. This is very interesting for medical purposes since the information of blood oxygenation is an essential parameters that physicians use everyday to monitor their patients' health.

Molecular imaging Some techniques, such as PET, use radioactive markers that are fixed on specific types of cells. Then, by detecting the products of the radioactive decays

it is possible to localise the marked cells. A similar imaging system can be realised by combining diffuse optical tomography and fluorescent markers. With this method, it is possible to map the local concentration of some fluorescent probes without using radioactivity [5]. This can lead to the detection and the study of tumours directly *in vivo* "without any stress or damage to the animal" as written by Koenig *et al.* [6].

1.2.2 Problems related to light scattering.

We have outlined that using light for *in vivo* imaging has very interesting applications. As light from the red and NIR part of the spectrum is not strongly absorbed, it can be used to perform optical imaging. Unfortunately, biological tissues have very strong scattering properties for light waves, which makes optical imaging difficult. Indeed, usual optical imaging techniques such as photography or microscopy rely on ballistic light. When light propagates in a scattering medium, standard imaging techniques are not possible anymore. The picture in Fig. 1.6, illustrates this phenomenon, the further the mountains in the background are, the less visible they are. It is due to the fact that, when the



Figure 1.6 – Effect of light scattering on background contrast.

picture was taken, the atmosphere was not very clear and scattered light. As a result, light from far away underwent more scattering events and was more attenuated resulting in a lower contrast. In addition, scattering made parasitic light going into the direction of the camera. In the case of this picture, the atmosphere is very lightly scattering, enabling taking pictures of object a few kilometres away, biological tissues are so scattering that after a few millimetres, it is impossible to perform standard optical imaging.

A quick calculation gives an idea of the attenuation of ballistic light in multiple scattering media such as biological tissues. Let us consider light at 1064 nm (this wavelength is chosen because it is the one used in all the experimental work described in this thesis). At this wavelength, biological tissues have the following characteristics: $\mu_a \approx 0.1 \,\mathrm{cm}^{-1}$, $\mu_s \approx 100 \,\mathrm{cm}^{-1}$ and $\mu'_s \approx 10 \,\mathrm{cm}^{-1}$ (see Fig. 1.4 and Tab. 1.1). The attenuation of ballistic light is given by the extinction coefficient μ_e , meaning that after propagating through L = 1 cm, the transmitted ballistic light is $e^{-\mu_e L} \simeq e^{-100} \approx 10^{-44}$, we might as well consider that there is no exploitable ballistic light. Besides, the attenuation of diffused light is much weaker and can be calculated using the effective extinction coefficient μ_{eff} . For the same thickness of L = 1 cm, the remaining diffused light is $e^{-\mu_e ff L} \simeq e^{-2} \approx 10^{-1}$. This shows that using ballistic light for biological tissue imaging is difficult, and the solution might be in finding techniques that exploits diffused light.

1.3 How to perform optical imaging of biological tissue.

Despite the difficulty of doing optical imaging in biological sample, a few solutions exists and are investigated. They can be separated into two categories: solutions that rely on ballistic or snake photons¹, and solutions that choose to take advantage of the multiple scattered light. This section describes both approach starting with solutions that use ballistic light. The techniques that rely on multiple scattered light to probe the local optical properties of biological medium are presented in the second part of this section.

1.3.1 Imaging using ballistic light.

Trying to exploit ballistic light is very attractive since it is equivalent to achieving imaging in the conventional sense. As ballistic photons do not undergo scattering event, their trajectories are governed by the laws of geometric optics, which means that they are easy to exploit for imaging with a very good resolution. The main problem is that their number decreases very rapidly inside biological tissues. As a result, after a few millimetres, they are very few compared to the multiple scattered ones that constitute parasitic light. In order to use ballistic photons, it is necessary to reject diffused light so that only the ballistic or snake photons reach the detector.

Snake photons Photons exiting a multiple scattering medium can be sorted in three types. First the ballistic photons, they are the ones that have not been scattered and that are labelled (a) on Fig. 1.7. In the case of biological medium, which have strong scattering properties, there are very few of them. The second type of photon are the *snake photons*, labelled (b) on Fig. 1.7. These photons underwent only several scattering events so their trajectory is very close to the trajectory of the ballistic photons, consequently, they can be used in imaging techniques based on ballistic light, at the cost of degrading the resolution. Finally, the third type of photons is the scattered ones (c), as they have been multiply scattered by the medium, their path inside the medium is random. They are the most numerous at the exit of a biological sample.

¹The snake photons are the photons that have been scattered only several times so their trajectory is very close to the ballistic photons trajectory.



Figure 1.7 – Types of photons exiting a multiple scattering medium. (a) Ballistic photons, (b) Snake photons and (c) Scattered photons.

It is possible to isolate ballistic light from diffused light using spatial or time gates [7]. Gating light makes optical imaging of multiple scattering medium possible, but with a very limited depth, usually a few millimetres, because of the very fast attenuation of ballistic light.

Spatial gating The definition of ballistic photons is that they have not been deviated by scattering. As they travel in straight lines, they can be isolated using a simple collimator. With a collimator in front of the detector aligned with the source, it is possible to isolate the photons that travelled in straight line between the source and the detector. This way only the photons with a propagation direction parallel to the initial direction will be detected. Another idea consists in using Fourier filtering with a lens in order to eliminate high frequency components. Then, with a point by point scanning it is possible to reconstruct the absorbing part of the sample. These techniques are still very limited because of, first, the small number of ballistic photons and second, the scattered photons that still manage to go through the collimator.

Temporal gating Another way of filtering the ballistic and snake photons is to use time gating. As mentioned earlier, the ballistic and snake photons travel in almost straight line. This means that in a short light pulse, they are the first to arrive at the detector because they took the shortest possible path. As a result, a femtosecond laser pulse will be enlarged in the time domain because of scattering and with a fast time gate, around a few picosecond, it is possible to isolate the ballistic and snake photons from the multiple scattered ones. A few methods exist in order to create short time gate such as the use of streak cameras or non-linear processes using a femtosecond laser pulse. The non linear processes can be Kerr effect [8], stimulated Raman scattering [9] or Optical Parametric Amplification [10].

Optical Coherence Tomography The last method of filtering ballistic light from multiple scattered light is by using coherence. When illuminating a sample with a low time-coherence light source, the only photons that remain coherent during their travel through the multiple scattering sample are the ballistic photons or the backscattered photons that have been scattered only once. This is the basic principle of the imaging technique called Optical Coherence Tomography (OCT) proposed by Fujimoto's team [11] in 1991. A classic scanning OCT setup is represented in Fig. 1.8.(a) along with images of human breast tissues obtained by Full-Field OCT (Fig. 1.8.(b)). Since then, OCT has been developed a lot and has numerous applications in various medical field such as ophthalmology, dermatology and even dentistry [12]. It is probably the most advanced technique based on the use of ballistic light in order to perform imaging on multiple scattering media. It is able to generate images with a resolution around 1 µm at depth up to 1 mm. More recent work tried to combine OCT with other imaging techniques in order to add new contrasts to OCT images such as elastography [13] or photothermal imaging [14].



Figure 1.8 – (a) Classical scanning OCT setup (From Wikipedia). (b) En face Full-Field OCT images of fixed human breast tissue at depth 40 µm on two different areas (From [15])

The use of ballistic photons is a great advantage because it leads to a very good spatial resolution, but it also has limitations. As mentioned earlier, the number of ballistic photons decreases very rapidly. The imaging depth is therefore very limited since after a few millimetres there is not enough light to perform imaging. In order to get optical information from deep inside tissues, it is necessary to find methods that exploit multiple scattered light.

1.3.2 Imaging techniques based on multiple scattered light.

Diffused light coming from a multiple scattering medium creates very complicated patterns that seem to carry no information about the medium. In fact, the interesting information (local absorption for example) is blurred into the diffused light pattern and is very hard to isolate. In this section, we will explore different solutions that have been implemented in order to retrieve interesting information from multiple scattered light. These methods can be separated into two categories:

• Methods that use detailed model and equation describing the propagation of light in complex media in order to reconstruct the optical field inside the sample (NIRS and DOT)
• Methods that couple optics to another type of wave that propagates ballistically (ultrasound) in order to carry the useful information (photoacoustic and acousto-optic).

NIRS NIRS is for Near-InfraRed Spectroscopy, this method enables to probe the optical properties of tissues at depth around 50 mm. It uses the fact that between a light source and a detector on the surface of a tissue, light propagates in a banana-shaped volume as shown in Fig. 1.9. As a result, light collected by the output fiber will carry information about the absorption along the "banana-shaped" path. This technique is not, strictly



Figure 1.9 – Principle of Near-InfraRed Spectroscopy – The path travelled by light in a multiple scattering media between a source and a receptor spaced by D has the shape of a banana that goes at a depth of D/2

speaking, an imaging technique since the collected information is not localised in space, it is the global absorption along a certain path. Nevertheless, it is possible to probe the absorption at different depth by changing the distance between the input fiber and the output fiber. For a distance D between the emitter and the receptor, the probed depth will be around D/2. Moreover, as the name suggests, it is possible to perform spectroscopy by scanning a range of wavelength, in order to calculate, for example, blood oxygenation. Since it has been proposed, by F. Jöbsis in 1977 [16], NIRS has been developed in a variety of modalities (NIRS-CW, NIRS-TD and NIRS-FD, for Continuous Wave, Time Domain and Frequency Domain) in order to improve depth and reduce measuring time.

Diffuse Optical Tomography The propagation of light in multiple scattering medium can be mathematically modelled using the Radiative Transfer Equation (RTE). The idea of DOT is to use the RTE to mathematically model the propagation of light inside a sample using the local optical properties (absorption and scattering coefficients) as parameters. Then a large number of optical fibres placed all around the sample are used as light sources and detectors in order to record the scattered light at the edge of the sample for different boundary conditions. Using the different boundary conditions, it is possible to solve the inverse problem in order to calculate the optical properties of the sample.

DOT has been implemented in commercial devices and can be used for brain imaging [17] or breast cancer imaging [18]. It gives 3D images of the optical properties of biological medium with a spatial resolution that depends on the algorithm and numerical parameters such as the mesh, the number of light sources and the number of receptors. The thinner the mesh is, the better the resolution, but the longer the numerical calculations. The resolution is typically 5 mm which is very limited for, *e. g.*, early detection of tumours which can be smaller than 1 mm. Nevertheless, as mentioned in 1.2.1, DOT can be used to track fluorescent probes with specific markers injected inside a patient. This method, called Fluorescence Diffuse Optical Tomography (FDOT) [19], is not, strictly speaking, an optical imaging method because the images generated do not give information about the optical properties of the sample. However coupled with standard DOT it is possible to add useful information to DOT images.

Another way of using diffused light to gather information from inside a multiple scattering medium is to couple light with another wave that propagates well in the medium such as ultrasound (in the case of biological sample). This can be done using the photoacoustic effect or the acousto-optic effect.

Photoacoustic imaging It is possible to generate acoustic waves with light. When a short light pulse hits an absorber, it briefly expands and generates an acoustic wave in the medium. This is called the photoacoustic effect. The position and size of the absorber can then be determined by detecting the generated acoustic wave. A paragraph of this manuscript, in chapter 2, is dedicated to the principles of photoacoustic imaging in order to introduce the experiments from chapter 7 on an optical detection of photoacoustic signals.

Acousto-optic imaging Light and ultrasound can interact through the acousto-optic effect which is the basic principle behind Acousto-Optic imaging, also known as Ultrasound modulated Optical Tomography. As photons travel through an acoustic field, they are phase modulated. Then, by detecting the phase modulated signal, it is possible to image the local optical properties of the medium. As acousto-optic imaging is the main subject of this thesis, a large part of chapter 2 describes its functioning. Chapters 5 and 6 are dedicated to our experimental investigations on the use of holography in gain media for the detection of the acousto-optic phase modulation.

1.4 Conclusion.

In this introduction chapter we presented the main optical properties of biological tissues. We saw that they were strongly scattering for optical waves, which implies that it is quite hard to perform optical imaging at high depth. Yet, information provided by optics could be very helpful for physicians to improve diagnosis by adding new informations when used in conjunction with other imaging techniques. Moreover, light waves are virtually harmless for biological tissue. Using them for imaging is therefore a very promising approach. We presented a few techniques that tried to exploit light that propagates in a ballistic way inside biological tissues, but we saw that these methods were very limited in terms of imaging depth.

Imaging deep inside biological tissues requires to use multiple-scattered light. A few

purely optical techniques exists such as diffuse optical tomography or NIRS but their resolution is very limited. Finally we introduced two techniques based on coupling light with ultrasound: Acousto-Optic imaging and Photoacoustic imaging. They will be detailed in the next chapter.

$\mathsf{CHAPTER}\ 2$

Ultrasound and light mixing - Acousto-optic and Photoacoustic imaging

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Due to multiple scattering of light inside biological tissues, it is no longer possible to have local information about the properties of the medium. One way of mapping the optical properties of the medium such as the absorption coefficient or the scattering coefficient is to couple light with another wave that propagates "well" (in a ballistic way). In biological tissues, ultrasonic waves are a good candidate since they propagate with almost no scattering and little attenuation at echographic frequencies (several MHz).

There are two ways of using light and ultrasound in order retrieve the local optical properties of a multiple scattering medium. The first idea is to use the acousto-optic effect, which is the modulation of light by an acoustic field. This technique is called *Acousto-Optic Imaging* (AOI) or *Ultrasound Optical Tomography* (UOT). The second idea is to use the generation of ultrasounds inside the medium through the photoacoustic effect. The original idea of this thesis was to develop a detection system for AOI based on holography in gain media. The first two sections of this chapter are dedicated to the principles and the challenges of acousto-optic imaging (our experimental investigations concerning AOI is the subject of chapters 5 and 6). As we noticed that our setup could be used to detect vibrations on a scattering surface, we also demonstrated the detection of photoacoustic signals. Therefore, the last section of this chapter outlines the principles of photoacoustic imaging in order to introduce the experiments described in chapter 7.

2.1 Acousto-Optic interaction within multiple scattering media.

Diffraction of light by an acoustic wave has been predicted in 1921 by Brillouin. A few years later, in 1932, P. Debye and R. Lucas independently brought the experimental demonstration of this phenomena [20, 21]. The pressure wave creates a refractive index grating that diffracts light waves propagating through the acoustic field. There are two effects of this diffraction on a beam travelling in an acoustic field (see Fig. 2.1). First the diffracted components (in a normal incidence) are deflected by an angle θ_m given by the following equation:

$$\Lambda \sin(\theta_m) = m\lambda \tag{2.1}$$

where Λ is the acoustic wavelength, λ is the optic wavelength and m is the diffraction order. In addition to the angular deflection, the diffracted components are frequency shifted, the light diffracted in order m has a frequency of $f_m = f_L + m f_{ac}$, where f_L is the input light frequency, and f_{ac} is the acoustic frequency. The frequency shift, which is due to a phase modulation, is the key of AOI of thick scattering medium, as it is the origin of the signal we will try to measure.



Figure 2.1 – Diffraction of light, in the Raman-Nath regime, by a refractive index grating generated by an acoustic wave.

2.1.1 Phase modulation of light by an acoustic wave in a multiple scattering medium.

In a scattering medium, the interaction between light and an acoustic field is different from the case without scattering. As the light is not propagating in a straight line anymore, but taking random path through the medium, talking about deflection of the beam has no longer any meaning.

Nevertheless, light is still phase modulated by the acoustic wave. Two mechanisms are responsible for this modulation of the optical wave. First, the modulation of the refractive index, due to the pressure changes, modulates the phase of the optical beam. In



Figure 2.2 – Principles of acousto-optic imaging of multiple scattering media. The scattered photons propagating through the sample have the optical path changing due to an acoustic field. Taken from [22]

addition, the acoustic wave makes scatterers vibrate, which results in a modulation of the optical paths, and thus contributes to a modulation of the global phase of the light. Ultrasound and light mixing in a multiple scattering medium is illustrated in Fig. 2.2 (taken from [22]). It shows the effects of the changes of the optical pathlength due to the vibration of scatterers and the changes of the refractive index. The study and modelling of the acousto-optic interaction in multiple scattering media has only started in the 1990s. The first theories took into account only one of these phenomena, usually the vibration of scatterers. The first model was proposed by Leutz and Maret in 1995 [23], were they considered that the scatterers in a liquid vibrates both because of the Brownian motion and the acoustic field. This model was completed in 1997 by Kempe *et al.* where they considered the non uniformity of the ultrasonic field [24]. In 2001, L. Wang analytically modelled the phase modulation of the light taking into account both the vibrations of the scatterers and the modulation of the refractive index [25].

In the following, we will consider that photons propagate in straight lines between scatterers at position \mathbf{r}_j , as shown in the diagram from Fig. 1.1. The length of the j^{th} mean free path between scatterers \mathbf{r}_{j-1} and \mathbf{r}_j will be called l_j and s_j will be the coordinates along this path.

Phase modulation by modulation of the refractive index An acoustic wave is a periodic modulation of the pressure inside its propagation medium. As a result, the local density of the material changes causing local variations of the refractive index of the medium. Let us consider an acoustic wave described by the following pressure field:

$$p_{ac}(\mathbf{r},t) = A_{ac}\sin(\mathbf{k}_{ac}\cdot\mathbf{r} - \omega_{ac}t) \tag{2.2}$$

where A_{ac} is the amplitude of the acoustic field, ω_{ac} its angular frequency and \mathbf{k}_{ac} the wave vector. It creates a local change of the refractive index Δn which causes a phase shift during the j^{th} mean free path given by:

$$\Phi_{nj}(t) = \int_0^{l_j} k_0 \Delta n(\mathbf{r}_j, s_j, \theta_j, t) ds_j$$
(2.3)

where θ_j is the angle between the acoustic wave vector \mathbf{k}_{ac} and the j^{th} mean free path. The modulation of the refractive index due to the acoustic field can be written [25]:

$$\Delta n = n_0 \eta k_{ac} A_{ac} \sin(\mathbf{k}_{ac} \cdot \mathbf{r}_{j-1} - \omega_{ac} t + k_{ac} s_j \cos \theta_j)$$
(2.4)

where n_0 is the refractive index of the medium at rest, η is linked to the adiabatic piezooptical coefficient of the material $\partial n/\partial p$ ($\approx 1.4 \times 10^{-10} \,\mathrm{Pa^{-1}}$ in water [26]), the density of the medium ρ and the acoustic velocity ν_{ac} by: $\eta = (\partial n/\partial p)\rho \nu_{ac}^2$. The phase shift by modulation of the refractive index is accumulated along each scattering mean free path, consequently the total phase accumulated along the travel through the acoustic field will depend on the value of $k_{ac}l_s$.

Phase modulation by vibration of scatterers The other phenomenon that modulates the phase of the light propagating through an acoustic field in a multiple scattering medium is the vibration of the scatterers. Indeed, the acoustic wave makes the scatterers vibrate around their rest position; so, when a photon is scattered, its frequency is shifted by Doppler effect. If we consider an isotropic scattering, and a scattering mean free path l_s larger than the optical wavelength, the phase shift introduced during the j^{th} scattering event is given by [25];

$$\Phi_{dj}(t) = -n_0 k_0 (\mathbf{k}_{j+1} - \mathbf{k}_j) \cdot \mathbf{A}_{US} \sin(\mathbf{k}_{US} \cdot \mathbf{r}_j - \omega_{US} t)$$
(2.5)

where \mathbf{k}_j is the unitary wave vector of the light along the j^{th} scattering mean free path, and \mathbf{A}_{US} is the amplitude vector of the acoustic wave.

Coherent acousto-optic modulation When the sample is illuminated by a coherent light, the illumination pattern at the exit of the sample is a *speckle* pattern because of the multiple interferences due to the numerous paths inside the medium. The amplitude of the modulation of the light at $n\omega_{ac}$ can be retrieved with the autocorrelation function of the electric field and the Wiener-Khinchin theorem. It states that the intensity is the Fourier transform of the autocorrelation function $G(\tau)$ of the electric field. The intensity of the component modulated at $n\omega_{ac}$ can thus be written:

$$I_n = \frac{1}{T_{ac}} \int_0^{T_{ac}} G(\tau) \cos(n\omega_{ac}\tau) d\tau$$
(2.6)

The autocorrelation function of the electric field is the summation on every possible path of the autocorrelation of the electric field E_s along a given path s. The summation is weighted by the probability p(s) of the path:

$$G(\tau) = \int_0^\infty p(s)G_s(t,\tau)ds$$
(2.7)

where $G_s(t,\tau) = \langle E_s(t)E_s^*(t+\tau)\rangle$ is the autocorrelation of the electric field on the path s. It is a function of the phase shifts given by Eqs. 2.3 and 2.5. Supposing that they are independent of each other, and if the phase variations are much less than unity, $G_s(t,\tau)$ can be written [25]:

$$G_s(t,\tau) = \exp\left\{-\frac{1}{2}\left\langle \left(\sum_{j=1}^{s/ls+1} \Delta\phi_{nj}(t,\tau) + \sum_{j=1}^{s/ls} \Delta\phi_{dj}(t,\tau)\right)^2 \right\rangle\right\}$$
(2.8)

where $\Delta \phi_{nj}(t,\tau) = \Phi_{nj}(t+\tau) - \Phi_{nj}(t)$ is the phase variation induced by the modulated index of refraction along the j^{th} free path, and $\Delta \phi_{dj}(t,\tau) = \Phi_{dj}(t+\tau) - \Phi_{dj}(t)$ is the phase variation induced by the modulated displacement of the j^{th} scatterer following the j^{th} mean free path. Detailed calculation of G can be found in the previously cited article [25]. Finally, the autocorrelation function can be expressed:

$$G(\tau) = \int_0^\infty p(s) \exp\left\{-\frac{2s}{l_s}(\delta_n + \delta_d)(n_0 k_0 A_{ac})^2 [1 - \cos(\omega_{ac}\tau)]\right\} ds$$
(2.9)

where δ_d and δ_n are constants that respectively characterise the influence of the displacement of the scatterers and the influence of the modulation of the index of refraction. Using this equation, it is possible to assess which of the two contributions has the most influence. In his model, Wang says that the ratio between δ_n and δ_d reaches a critical point when $k_{ac}l_s = 0.56$. When $k_{ac}l_s$ becomes larger than this critical value, the contribution of the phase modulation from the index of refraction increases rapidly and can be significantly larger than the contribution from the displacement. Indeed, when the scattering mean free path becomes longer than the acoustic wavelength, the optical path between two scattering events increases, so the accumulated phase increases, whereas the number of scattering events does not change (so the phase shift due to the vibration of scatterers stay constant).

2.1.2 Properties of the exiting light.

Speckle modulation Equation 2.9 shows that G has a dependency in $[1 - \cos(\omega_{ac}\tau)]$. It indicates the modulation of the speckle light at the acoustic frequency. Thus, photons exiting the multiple scattering sample can be separated into two categories:

- The photons that have been modulated by the acoustic field. Their frequency is the original frequency of the light shifted by the frequency of the acoustic wave: $\omega = \omega_L \pm \omega_{ac}$. These photons will be called the *tagged photons* in the rest of this manuscript.
- The photons that have not been modulated by the acoustic field. Their frequency did not change, it is the original frequency ω_L . They will be called the *untagged photons*.

One can consider that the speckle pattern exiting the sample is the superposition of speckle patterns from both components, *tagged* and *untagged* (as illustrated in Fig. 2.3.(a)). Hence, the spectrum of the light presents sidebands shifted by the frequency of the acoustic field (see Fig. 2.3.(b)). In terms of wavelength, if one considers near-infrared light



Figure 2.3 – Tagged and untagged photons exiting the sample. (a) shows the decorrelated speckle patterns from the tagged and the untagged components of the light. (b) shows the optical spectrum of the exiting light. It is comprised of a main peak at the original frequency ω_L and sidebands at $\omega_L \pm \omega_{ac}$.

 $(\lambda_L = 1 \,\mu\text{m})$ going through an acoustic field at $f_{ac} = 5 \,\text{MHz}$, the frequency shift corresponds to a wavelength shift of $\Delta \lambda \approx 10^{-5} \,\text{nm}$. This gives a glimpse of one of the major

difficulty of the detection of the acousto-optic signal: isolate the *tagged* component from *untagged* component. Section 2.2.3 will explain several ways of detecting the acousto-optic signal. It will especially be focused on the ones that are the main subject of our work: the holographic detection techniques.

Optical contrast The autocorrelation function G, given by Eq. 2.9, depends on the probability density p(s) of a path s. This highlights the influence of the local intensity of the light inside the medium on G. Indeed, if there is an absorbing inclusion inside the acoustic field, light will have a very small probability of taking the paths that goes through the inclusion. Thus, the contributions of these paths on G will be small resulting in a decrease of the acousto-optic signal. An illustration is given in Fig. 2.4 in which three possible paths for the light are represented (numbered from 1 to 3). In the presence of the absorbing inclusion, the path number 1 will not contribute anymore causing a decrease of the autocorrelation. An absorption contrast can thus be easily measured



Figure 2.4 – Origin of the optical contrast in acousto-optic imaging. In presence of an absorber in the acoustic field, there are much less light paths that enable photon tagging, as a result the signal is smaller.

by detecting the acousto-optic signal. The possible methods for localising it will be detailed in section 2.2. Theoretically, tagging light with ultrasound is also sensitive to scattering contrast. It has been reported by Kothapalli *et al.* that inclusions with different scattering coefficients, give different acousto-optic signals [27]. They observe a decrease of the modulation for objects with a higher scattering coefficient and an increase of the modulation for object with a lower scattering coefficient. In their model, the acousto-optic signal consists of three contributions: the modulated refractive index contribution, the modulated displacement of scatterers contribution and the anticorrelation between the two mechanisms (this name comes from a negative value of the correlation). They state that when the scattering coefficient is high, the absolute value of this anticorrelation increases and becomes dominant compared to the two other terms which results in a decrease of the amplitude of the modulation and *vice versa*.

2.2 Optical imaging with sound - Acousto-Optic Imaging.

Now that we have described the interaction between light and an acoustic field inside a multiple scattering medium, we will show how to use this interaction to locally probe the optical properties of a sample. We will also describe the techniques that exist to detect the acoustic modulation of the light.

2.2.1 Mapping the local optical properties.

To map the optical properties of a thick scattering medium using the acousto-optic effect, one needs to create a limited volume where light and the acoustic wave can interact.

Sample scan If the volume of interaction is small, then the *tagged photons* that reach the detection system will give information about this particular volume of the sample. This volume of interaction is a *tagged photons* source. The next step it to move this *tagged photons* source and record the signal for each position to create a map of the optical properties of the sample. This is illustrated by Fig. 2.5, the sample is represented at different time T_i , with the position of the volume being scanned. The left side shows



Figure 2.5 – Image reconstruction by scanning the acoustic focal spot in the sample. The acoustic focal spot is tagging photons in a specific volume. By moving the volume it is possible to retrieve the optical properties inside scattering media such a biological tissues.

a sample with the diffuse light pattern and a black inclusion. The position of the *tagged* photons source at time T_i is represented by a green square, and the positions for time

 $t < T_i$ are represented by light green squares. The right side shows the image obtained at T_i . The resolution depends on the size of the *tagged photons* source, which is linked to the properties of the acoustic wave.

2.2.2 Resolution of the imaging system.

Transverse (US) resolution The size of the *tagged photons* source can be reduced by sending a focused acoustic wave inside the sample. An acoustic wave propagating along the y axis (see Fig. 2.5) can be focused in the transverse directions, x and z. The size of the acoustic focal spot in those directions, and therefore the resolution, is limited by diffraction. For a circular transducer with a diameter D and a focal length F, the size of the acoustic focal spot, and therefore the resolution is:

$$dx = dz \approx \frac{1.22v_{ac}F}{Df_{ac}} \tag{2.10}$$

where v_{ac} is the celerity of the acoustic wave, and f_{ac} is its frequency. The size of the acoustic focal spot in the propagation direction is around:

$$l_y \approx 7 \frac{v_{ac}}{f_{ac}} \left(\frac{F}{D}\right)^2 \tag{2.11}$$

In biological media, ultrasonic frequencies are commonly used (e. g. ultrasound imaging). Their frequency is usually between 1 MHz and 20 MHz with a celerity around $1.5 \text{ mm } \text{µs}^{-1}$. If one uses a transducer with a numerical aperture (D/F) equals to 0.5, the transverse resolution given by Eq. 2.10 is dx = dz = 0.7 mm, which is very interesting for medical imaging purposes. The axial size of the focal spot will be $l_u \approx 8.5 \text{ mm}$.

Achieving a good axial resolution, along the y direction, is harder. Indeed, if one uses continuous ultrasound, photons will be tagged all along the acoustic column under the transducer resulting in no resolution along the y axis. Moreover, with continuous ultrasound, safety limits (in terms of energy sent into tissues) imposed by medical standards are often exceeded. Various strategies exists in order to have a good axial resolution.

Axial (US) resolution with US bursts Instead of working with continuous acoustic waves, one can use short bursts. Limiting the temporal extension of the burst, limits also its spatial extension. The resolution will be given by the spatial extension of the burst, as illustrated in Fig. 2.6. As an example, if a burst of ultrasound at $f_{ac} = 5$ MHz lasts 2 cycles and propagates in a biological tissues, its spatial extension will be: $dy = v_{ac}N_c/f_{ac} = 0.6$ mm. The burst regime works well. It gives a good resolution and enables to stay below the safety limits. Nevertheless, it suffer from a few drawbacks. First, because the acoustic burst is short, there are very few tagged photons. As a result the signal is weak. Secondly, using short bursts requires a detection system that is either able to follow the bursts or to record the signal during a short enough time to freeze the burst.



Figure 2.6 – Axial resolution of acousto-optic imaging in burst regime. The resolution is given by the spatial extension of the burst.

Axial (US) resolution with a *chirp* This method has been proposed by L. Wang et G. Ku [28,29]. It consist on sending a frequency-swept acoustic wave to modulate the laser light. Then the light is heterodyned with a frequency-swept reference beam, and analysed in the Fourier domain to obtain a one dimensional image. The resolution is given by the span of the frequency sweep Δf by: $dy \approx v_{ac}/\Delta f$ (where v_{ac} is the speed of the acoustic wave). The transducer used to create the acoustic wave needs to have a flat frequency response over a large bandwidth in order to achieve a good resolution. A frequency sweep over $\Delta f = 1$ MHz, which is quite large, would yield a resolution of 1.5 mm. This technique requires the light to be heterodyned, meaning that it will not work with the incoherent detection method that will be presented in 2.2.3.

Axial (US) resolution by random phase jumps The last method is the technique of random phase jumps. It has been developed by Lesaffre *et al.* at Institut Langevin. It consists in modulating the ultrasonic wave with a random phase pattern. It is a series of jumps between 0 and π that occurs at random time. This phase pattern is also applied to a



Figure 2.7 – Axial resolution of acousto-optic imaging using random phase jumps. The resolution is given by the width of the autocorrelation function of the random phase jumps. Image taken from [30]

reference light with a delay τ . This reference is used to heterodyne the diffused light from the sample. The random phase jumps reduce the optical coherence length of the diffused

light. As a result, by changing τ , it is possible to choose the depth one wants to probe. The principle of this method is illustrated in Fig. 2.7, the resolution is given by the width of the autocorrelation function of the random pattern. It is directly linked to the delay t_j between two jumps. This method is called *Acousto-Optical Coherence Tomography* and works only with coherent detection systems such as photorefractive holography [30] or digital holography [31].

2.2.3 Detection of the acousto-optic signal.

In section 2.1.2, we saw that the spectrum of the light exiting an insonified scattering medium had several components. There is the component oscillating at the light frequency ω_L , and two sidebands shifted by the frequency of the acoustic wave: $\omega = \omega_L \pm \omega_{ac}$.

Remark : In fact there are also components at higher orders ($\omega = \omega_L \pm n\omega_{ac}$), but we will not take them into account because they are generally negligible.

Tagged or untagged photons The detection of the acousto-optic signal requires the separation of the untagged component at ω_L from the tagged components at $\omega = \omega_L \pm \omega_{ac}$. In the previous paragraph, we assumed, for the sake of clarity, that the acousto-optic signal was carried by the tagged photons. Actually, the information provided by the acousto-optic interaction can be retrieved on both components. Indeed, as illustrated by figure 2.8, when the acoustic transducer is off, there is a certain number, N_p , of photons detected. Then, when the transducer is turned on, the acoustic field is creating tagged photons, as a result, the number of tagged photons increases. Because the total number of photons, N_p , has to be conserved, the number of untagged photons decreases ($N_p = N_t + N_u$, where N_t is the number of tagged photons and N_u is the number of untagged photons). When the



Figure 2.8 – Evolution of the number of *tagged* and *untagged* photons as the piezoelectric transducer is moved above the sample.

acoustic field is on an absorbing inclusion, there are no *tagged photons* created anymore so their number decreases. Mechanically, the number of *untagged* photons goes back up. In conclusion, the information provided by the acousto-optic interaction can be retrieved by detecting either component. The main challenge is to be able to separate one component from the other with a good sensitivity. As mentioned in 2.1.2, the acoustooptic frequency shift leads to a wavelength shift around $\Delta \lambda = 10^{-5}$ nm. In addition, the number of *tagged photons* is very small compared to the number of *untagged photons* (usually 1/1000 in short burst regime), which means that a very sensitive detection is required. The acousto-optic detection techniques can be sorted into two categories:

- The **incoherent** methods that are aimed at creating very narrow spectral filters in order to detect either one of the components by a simple measurement of *flux*.
- The **coherent** methods that select the *tagged* or *untagged* component using interferometric setups.

Incoherent methods They are based on spectral filtering. If one manages to create a spectral filter narrow enough to separate two wavelengths as close as 10^{-5} nm, then the use of a single photodiode with a large area is sufficient to detect the *tagged* (or *untagged*) photons. Moreover, as photodiodes are very fast, it is possible to work with ultrasonic bursts in order to have an axial resolution. Several solutions exist to create spectral filters that are narrow enough for acousto-optic imaging.

One solution is to use a very high finesse confocal Fabry-Perot interferometer that acts as very narrow bandpass filter. This solution has been proposed by S. Sakadzic and L. V. Wang. In 2004, they managed to image several millimeters deep into biological tissues with 15 MHz ultrasounds and a confocal Fabry-Perot tuned to one of the sideband of the ultrasound-modulated light [32] (see Fig. 2.9.(a)). Rousseau *et al.* improved this technique a lot using double-pass configuration. In 2009 they managed to achieve AOI,



Figure 2.9 – (a) 1D acousto-optic scan obtained by Sakadzic *et al.*. The sample is a 4 mm piece of chicken with an inclusion made of black latex (extracted from [32]). (b) 2D image of jellified India ink in the center of a 60 mm thick piece of chicken breast (extracted from [33]).

much deeper, up to several centimeters with a millimetric resolution [33]. They created a bandpass filter with a full width at half maximum of 2.4 MHz in order to filter sidebands

at 5 MHz (see Fig. 2.9.(b)). The main issue of the Fabry-Perot based detection is the very limited optical etendue¹, which can be a problem when working with diffused light.

Very narrow bandpass filter can also be created using spectral holeburning. In rare-earth doped crystals, it is possible to create transparency bands in the absorption spectrum at a given frequency by saturating absorption. This method has been implemented by Li *et al.* [34] and Xu *et al.* [35]. Another strategy is to use dispersion effects to delay one spectral component, so they are detected at different time on the detector [36]. These spectral holeburning methods requires to cool the crystal at very low temperature (a few K), which requires heavy equipments.

Coherent methods Those methods are based on interferometry to detect either component. To perform interferometry with light coming out a multiple scattering medium, it is necessary to use a coherent source such as a laser. As a result, the intensity at the exit of the sample is a speckle pattern because of the multiple paths light followed through the medium. In order to compensate this loss of spatial coherence, one has to use techniques such as adaptive holography or phase conjugation. When performed analogically, these processes use non linear effect in crystal to record aberrated wavefronts as holograms. The work described in this thesis will expose how one can use a gain medium (Nd:YVO₄) to perform holography for acousto-optic detection. The use of phase conjugation by fourwave mixing will be described in chapter 5, and chapter 6 will be dedicated to adaptive holography by two-wave mixing.

Speckle decorrelation Coherent detection methods retrieve the acousto-optic signal by correcting the aberrations introduced by the sample. As they use coherent light, the aberrations take the form of a speckle pattern. In biological media, those aberrations and thus the speckle pattern changes overtime and decorrelates in about 1 ms [37]. It is, therefore, essential to have a detection system that is not sensitive to these problems of speckle decorrelation. This means that whether it is the wavefront adaption or the phase conjugation, it has to respond faster than this decorrelation time.

This speckle decorrelation problem is critical to the field of acousto-optic imaging. As we will mention in chapters 5 and 6, the current setups that are using coherent detection are based on photorefractive crystals which response time are at best a few ms. None of the current setup has yet successfully manage to perform a real *in vivo* acousto-optic image through several centimetres of biological tissues. This problem is one of the main motivation of this thesis work since we will demonstrate that the holography in Nd:YVO₄ has a response time of a few tenth of microseconds.

2.3 Principles of Photoacoustic imaging.

Absorbers hidden inside a biological sample can be detected by making them emit an acoustic wave. Then by detecting this acoustic wave, it is possible to pinpoint the position

¹The optical etendue is defined by the product of the surface of the aperture by the collection solid angle.

of the absorbers using image reconstruction algorithms. Sound generation with light is called *photoacoustic* effect and is the phenomenon exploited in the eponymous technique. As the detection of photoacoustic signals by speckle vibrometry was not the main subject of this thesis work, this section only outlines how the photoacoustic effect works and how it is used to perform biological imaging.

2.3.1 Generate sound with light.

A little bit of history The photoacoustic effect has been observed in 1880 by Alexander Graham Bell [38]. At the beginning, this effect was not really exploited because the conversion between light and sound was very weak, and the acoustic detectors were not efficient enough. Two developments during the 20th century rekindled the interest for the photoacoustic effect. First the development of very sensitive sensors based on piezoelectricity, and secondly, the invention of laser in 1960. Laser effect provided new high intensity light source, that could also emit very short pulses. Two essential characteristics for photoacoustic generation. The idea of using photoacoustic effect for optical imaging deep inside biological tissues came during the 1990s. It was mainly motivated by the poor resolution of medical imaging techniques based on optics.

From light to heat We suppose a medium in which acoustic and thermal properties are homogeneous, but with inhomogeneous optical properties. Embedded inside the scattering medium are absorbing inclusions. When light propagates through this medium and encounters an inclusion, it is absorbed. The absorbed electromagnetic energy is converted into heat and acts as a heat source:

$$P_V(\mathbf{r}, t) = \mu_a(\mathbf{r})\Phi(\mathbf{r}, t) \tag{2.12}$$

where P_V is the volumic density of heat absorbed (in W cm⁻³), μ_a is the absorption coefficient (in cm⁻¹) and Φ is the optical flux (in W cm⁻²).

From heat to sound This heat source P_V will increase the temperature of the inclusion, which will result in a dilatation of the inclusion and the generation of an acoustic wave. The temperature increase, and the propagation of the resulting pressure wave are described by a system of two equations:

$$\rho_0 c_p \frac{\partial T}{\partial t}(\mathbf{r}, t) - \kappa \Delta T(\mathbf{r}, t) = P_V(\mathbf{r}, t)$$
(2.13)

$$\Delta p(\mathbf{r},t) - \frac{1}{v_{ac}^2} \frac{\partial^2 p}{\partial t^2}(\mathbf{r},t) = -\rho_0 \frac{\partial}{\partial t} \left(\beta(T) \frac{\partial T}{\partial t}(\mathbf{r},t)\right)$$
(2.14)

where $T(\mathbf{r}, t)$ and $p(\mathbf{r}, t)$ are respectively the temperature field and the pressure field, v_{ac} is the speed of the acoustic wave, ρ_0 is the volumetric mass density, β is the thermal expansion coefficient, c_p is the isobaric thermal capacity and finally κ is the thermal conductivity.

Equation 2.14 is a wave equation with a source term. This source is given by equation 2.13, which is a diffusion equation of the temperature inside the medium. Its source is the electromagnetic energy absorbed by the inclusion $P_V(\mathbf{r}, t)$. These equations show the cascade of events that leads to the generation of a pressure wave from the absorption of light. This manuscript will not detail any further the processes behind photoacoustic generation. More details about the theory of photoacoustic can be found in [39].

In biological medium From the acoustic wave point of view, biological tissues can be considered as homogeneous with a constant speed of sound around $v_{ac} = 1.5 \text{ mm µs}^{-1}$. If one considers that the absorbing inclusions are spheric with a typical size being a few millimetres, the frequency of the acoustic wave emitted by the photoacoustic effect is in the ultrasound range. Typically around a few megahertz, which propagate very well in biological tissues.

2.3.2 Ultrasound detection and image reconstruction.

Detection As the photoacoustic effect generates an acoustic wave, it seems natural to use an acoustic detector such as a piezoelectric transducer to record the signal. Moreover, piezoelectric transducers are very sensitive and available with various bandwidth which makes them a very good solution. In order to use this type of detection, one has to make sure that there are no impedance mismatch between the sample and the transducer. Otherwise the acoustic wave will be reflected at the interface. This problem can be solved with the use of a coupling agent such as gel for ultrasound imaging or by putting the sample into water. Since the beginning of photoacoustic for biomedical imaging, most of the experiments were done using piezoelectric transducers.

Image reconstruction The most direct solution to generate a photoacoustic image is to use a focused mono-element piezoelectric transducer and scan it above the sample. When a photoacoustic emitter, or an optical absorber, is in the focal spot of the transducer, a signal will be detected (see Fig. 2.10). The method enables to map point by point the distribution of absorber inside the sample.



Figure 2.10 – Image reconstruction methods for photoacoustic imaging using a focused transducer. In (a) the inclusion is not in the focal spot of the transducer so no signal is recorded. In (b) the inclusion is in the focal spot of the transducer so it detects a photoacoustic signal.

Another solution consists in using an array of plane transducers. Then, by analysing the temporal signals recorded on each channel of the array, it is possible to reconstruct the position of the absorber such as illustrated in Fig. 2.11, taken from [1]. The temporal



Figure 2.11 – Image reconstruction methods for photoacoustic imaging using an array of transducer. The temporal signal on each transducer is recorded, then by back propagation, each photoacoustic emitter can be reconstructed. Image taken from [1]

signals recorded by the array of transducers are series of value $(S_i)_k$ where k represent the k^{th} transducer and i is the i^{th} sample over time. For each value of $(S_i)_k$, the algorithm computes every point of the sample that have emitted a wave that propagated from the point and arrive at the k^{th} transducer at time t_i . Then the amplitude of the photoacoustic signal at each point of the sample is computed by doing the summation of the signals that arrive at the right time on the right transducer.

2.3.3 All optical detection methods.

Acoustic waves can also be detected using displacement sensitive optical systems. Interferometry is a powerful tool since interferometers can be sensitive to very small displacement such as those created by an acoustic wave. One of the main advantage of going for an optical approach is the bandwidth. Indeed, in piezoelectric based detection, the bandwidth is usually limited by the piezoelectric element. Optical system on the other hand have very wide bandwidth, from several hundredth of kilohertz to several hundredth of megahertz, the upper limitation will usually come from the electronic used to acquire the signal.

Fabry-Perot In 1999, Beard *et al.* proposed an optical based concept for the wideband detection of ultrasound [40]. They used a thin film of PET (polyethylene terephthalate) as a Fabry-Perot interferometer. This film, in contact with the sample, is illuminated by a tunable laser diode to probe its characteristics such as its thickness. When an ultrasonic wave arrives on the interferometer, its thickness changes, which is detected by the probe laser (see Fig. 2.12, taken from [40]). Ten years later, this technology has progressed and



Figure 2.12 – Schematics of the sensing Fabry-Perot interferometer for photoacoustic detection. Figure taken from [40].

is now able to generate three-dimensional photoacoustic images of biological tissues with spatial resolution smaller than $100 \,\mu m$ [41, 42].

Rousseau *et al.* adopted another strategy also based on a Fabry-Perot. In 2011, they reported the use of a confocal Fabry-Perot interferometer to demodulate light coming from the surface of a biological tissue in order to detect ultrasound generated by photoacoustic [43]. With this method they were able to detect metallic wires inside chicken breast completely remotely.

Laser vibrometry Another solution to optically probe the vibrations caused by photoacoustically generated ultrasounds, is to use an adaptive interferometer. This idea proposed by Horchreiner *et al.* [44], is very interesting. The use of an interferometer enables the generation of photoacoustic images completely remotely. As the vibrations are probed by a laser spot, the measurement is done without any contact between the detection system and the sample, which can be interesting in situations where it is not convenient to be in contact with the sample.

To understand the need for an adaptive interferometer, let us imagine a basic optical setup such as a Michelson's interferometer. It can be used to measure very precisely phase



Figure 2.13 – (a) Michelson interferometer for the detection of the vibration of a mirror. (b) Problem of spatial coherence in the detection of the vibration of a rough sample with a Michelson interferometer.

differences between the two arms of the interferometer. The changes in intensity of the fringes are directly linked to the phase difference between the signal and the reference arm. As show in Fig. 2.13.(a), the phase difference can be the result of a small displacement of the surface reflecting the signal beam while the mirror reflecting the reference is fixed. Supposing that M1 and M2 are perfectly reflecting, the intensity of the light measured on the photodiode is given by the following equation:

$$I_d \propto |E_R + E_S|^2 \\ \propto I_R + I_S + 2\sqrt{I_R I_S} \cos\left(\frac{2\pi\delta}{\lambda}\right)$$
(2.15)

Troubles arrive when the probed surface is not reflective but scattering. In this case, the light coming back from the sample is no longer a plane wave, but a speckle pattern as illustrated in Fig. 2.13.(b). The loss of spatial coherence blurs the interference pattern on the photodiode. The electric field backscattered by the sample can be written:

$$E_S(x,y) = A_S(x,y)e^{i\left(\frac{2\pi(l+\delta)}{\lambda} + \phi_S(x,y)\right)}$$
(2.16)

where $A_S(x, y)$ and $\phi_S(x, y)$ are the amplitude and phase of the signal wave. They are random functions reflecting the speckle nature of the backscattered light. The total intensity measured by the detector will then be:

$$I_d \propto \iint_{S_{Det}} |E_R + E_S(x, y)|^2 dx dy$$

$$\propto I_R + I_S + 2 \iint_{S_{Det}} A_R A_S(x, y) \cos\left(\frac{2\pi\delta}{\lambda} + \phi(x, y)\right) dx dy$$
(2.17)

Because $\phi(x, y)$ is random, the integration of the intensity over the surface of the detector will not improve the SNR. Despite that the signal increases like $\sqrt{N_{speckle}}$, so does the noise. As a result, the integration over a large number of speckle grains will yield the same SNR as the measurement on one speckle grain [24]. One solution to compensate



Figure 2.14 – Addition of a wavefront adaption element inside an interferometer in order to measure the vibration of rough surfaces.

for this problem is to add, in the interferometer, a system that shapes the reference wavefront to fit the signal wavefront as illustrated by Fig. 2.14. This method is exactly the same as the wavefront adaption technique used for acousto-optic imaging, which is detailed in chapter 6 (section 6.1.1). The system based on a Nd:YVO₄ gain medium for acousto-otpic imaging can thus be used for the detection of vibrations on rough surface and photoacoustic signals. The experiments we carried out to achieve these goals are presented in chapter 7.

2.4 Conclusion.

In this chapter we explained the principles of the two imaging techniques we will focus on in this manuscript. First, section 2.1 explained the specificities of the acousto-optic interaction within a multiple scattering medium. We saw that the phase modulation of the light has two origins: the modulation of the refractive index and the vibration of the scatterers. Then, in 2.2 we saw how to use this phase modulation to retrieve local optical properties inside the multiple scattering medium. We also described the challenges of detecting the acousto-optic signals and the methods that currently exists.

The last part of this chapter, part 2.3, dealt with the other optical imaging technique of multiple scattering media coupling light and ultrasound: Photoacoustic imaging. We outlined the principles behind photoacoustic imaging, namely, the generation of ultrasound due to the thermal expansion following the absorption of light. The classic detection methods, based on ultrasonic transducers were exposed, as long as more exotic techniques using optical setups. Two of them were presented, the thin Fabry-Perot film as long as a non-contact detection based on laser vibrometry. This latter method is the one we tried to develop in this thesis using a gain medium-based holographic setup.

Part II

Holography in $Nd:YVO_4$

chapter $\mathbf{3}$

Wave-mixing in laser media

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In this chapter, and the next one, we put aside the problems of imaging inside multiple scattering media. The focus will be on the main tool that was used for the detection of acousto-optic and photoacoustic signals: holography in gain media. Optical amplifiers are based on *stimulated emission* which has been theoretically discovered in 1917 by A. Einstein. The main and most well-known application of optical amplifiers is the LASER, which has been invented in 1960 by T. Maiman¹ [45]. Yet, optical amplifiers can be used for other applications such as wave mixing in order to perform wavefront adaption or phase conjugation. Indeed, since they can be saturated, it is possible to perform operation such as two-wave mixing or four-wave mixing through non-linearities.

To understand how wave mixing works inside an optical amplifier, we first explain how light is amplified through stimulated emission. Then, the second section is dedicated to gain saturation, which is the key property to record holograms in a gain medium. Finally, the last section deals with the use of gain saturation to actually record an hologram through a spatial modulation of gain and of refractive index.

3.1 Amplification of light.

Before going into wave-mixing in gain media, let us explain the processes underlying the amplification of light in matter. For the sake of clarity, the word "atom" will be used, in this chapter, to designate the optically active elements in the gain medium. They can either be atoms (*e. g.* in He-Ne lasers), molecules (*e. g.* in CO_2 lasers) or ions (*e. g.* in Nd:YVO₄ lasers where Nd³⁺ ions are responsible for the amplification).

3.1.1 Spontaneous and stimulated emission of light.

When an atom receives energy, its electrons jumps from one energy level to another. The atom is said to be in an excited state. In this state, it can release its energy by different processes that can be either non-radiative (no emission of light) or radiative (with emission of a photon). The emission of light during a deexcitation can happen through two different processes: spontaneous emission or stimulated emission.

Spontaneous emission In an excited state, an atom can randomly emit a photon to go back to its ground state. The direction of propagation, polarisation and phase of the emitted photon are totally random. Only its frequency is set and is given by the energy difference between the two energy levels: $\nu = \Delta E/h$ where ΔE is the energy difference and h is Planck's constant. This process is called *spontaneous emission* and is represented

¹Actually, a few years before, C. Townes created the first MASER, which works with the same principles as the LASER but in the microwave range of the electromagnetic spectrum.

in Fig. 3.1.(a). It is characterised by the spontaneous emission lifetime τ . The inverse of τ represents the probability of a deexcitation event per unit of time. Depending on the value of τ , this luminescent process is either called fluorescence (when the lifetime is short, less than a second) or phosphorescence (when the lifetime is long, more than a few seconds).



Figure 3.1 – Radiative deexcitation processes. (a) Spontaneous emission of a photon by an excited atom. (b) Stimulated emission of a photon by an excited atom in an electromagnetic field.

Stimulated emission It is possible to make an excited atom deexcite in a certain mode of the electromagnetic field. When an excited atom interacts with an electromagnetic field which frequency equals $\Delta E/h$, the atom has a high probability of emitting photons in the same mode as the electromagnetic field. The characteristics of the emitted photons (direction, polarisation and phase) are identical to the ones of the surrounding photons. The atom transfers its energy to the electromagnetic field, resulting in its amplification. This process is called *stimulated emission* and is sketched in Fig. 3.1.(b). It is the reciprocal process of absorption. Thus, the probability per unit of time for an atom to deexcite through stimulated emission is the same as the one for an atom in the ground state to absorb photons (considering that the levels have no degeneracy).

3.1.2 Amplification of light by stimulated emission of light.

Stimulated emission can be used to amplify a light beam propagating through a material of which atoms are in an excited state. At first, let us suppose that there is an exterior energy source that is transferring atoms from their ground state to an excited state. This exterior source will be called the pump. It can be either a flash lamp, electrical discharges or a laser. We will suppose that the atoms of the material can be modelled by a two energy levels system separated by an energy ΔE as represented in 3.2. The population densities of the ground state and the excited state will be called respectively N_0 and N_1 . Finally we will suppose that a light beam is entering the medium with an initial intensity I_0 and a frequency $\omega_L = \Delta E/\hbar$. When the beam propagates inside the medium, it can loose and gain energy through various processes. The variation of its intensity can be written²:

$$dI = dI_{abs} + dI_{stim} + dI_{loss} \tag{3.1}$$

 $^{^{2}}$ The problem is simplified by considering that the beam is not diverging so that there is no intensity variations due to modifications of its geometry



Figure 3.2 – Amplification of light along its travel through a pumped gain medium. On the left is represented the increasing intensity by stimulated emission, with loss of a few photons (scattering,...). The gain medium is sketched as a two level system with a population inversion between the ground state (0) and the excited state (1). The population inversion is maintained thanks to the pumping.

where dI_{abs} is the amount of light absorbed by the medium, dI_{stim} is the amount of light given by the medium as stimulated emission and dI_{loss} covers all other types of loss (absorption followed by non radiative deexcitation, scattering,...). Each of these terms can be made explicit:

- $dI_{abs} = -\sigma_a I N_0 dz$ where σ_a is the absorption cross section (in cm²) at the frequency of light. dI_{abs} is proportional to the intensity of light and population density of the ground level (the more atoms are in the ground level, the more photons can be absorbed). It is negative because it represent an absorption process.
- $dI_{stim} = \sigma_e I N_1 dz$, where σ_e is the emission cross section at the frequency of the light. Since stimulated emission is the reciprocal process of absorption, we have $\sigma_e = \sigma_a$. dI_{stim} is proportional to the population density of excited state. Indeed, the more atoms are in the excited state, the more photons can be generated through stimulated emission.
- $dI_{loss} = -\alpha_L I dz$ where α_L represents all types of loss per unit length.

As a result, the variations of the intensity can be written:

$$\frac{dI}{dz} = -\sigma_e I N_0 + \sigma_e I N_1 - \alpha_L I$$

= $\sigma_e I (N_1 - N_0) - \alpha_L I$ (3.2)

Equation 3.2 reveals a parameter essential to light amplification. Indeed, with this equation we can see that in order to amplify light using stimulated emission, $N_1 - N_0 > 0$ is a necessary condition. The parameter $N_1 - N_0$ is called the *population inversion* and will now be referred to as $\Delta N = N_1 - N_0$. The linear gain can be defined by $\alpha = \sigma_e \Delta N$, it is proportional to the population inversion and is a function of the frequency of the light. When amplified, light may also be phase shifted. The gain, α , and the phase shift, ϕ are linked to the complex susceptibility of the active atoms in the medium by [46]:

$$\alpha(\nu) = k\chi''(\nu) \quad \text{and} \quad \phi(\nu) = \frac{k}{2}\chi'(\nu) \tag{3.3}$$

where χ' and χ'' are respectively the real part and imaginary part of the susceptibility $(\chi = \chi' + i\chi'')$, they are linked by the Kramers-Kronig relations. The intensity of the light propagating through a pumped medium is the solution of Eq. 3.2, and if we suppose that σ_e , ΔN and α_L are independent of z we have:

$$I(z) = I_0 e^{(\alpha - \alpha_L)z} \tag{3.4}$$

Amplified spontaneous emission The process of amplification of light happens to every photon inside the medium. In particular, spontaneously emitted photons can also be amplified due to the population inversion. This phenomenon is called *Amplified Spontaneous Emission* and is often referred to as ASE. It is usually a source of noise in optical amplifiers and limits the performances of the system.

3.1.3 Multi-level systems.

In fact, it is not possible to create a population inversion in a two-level system. In order to have a positive population inversion, the system needs to have a transition more energetic than the amplification transition. This can be modelled either by three-level of four-level systems.

Three-level systems In a three-level system, there is a ground state (called 0) and two excited states (called 1 and 2). In the system represented in Fig. 3.3, the amplification transition is $1 \rightarrow 0$ and the pumping transition is $0 \rightarrow 2$. The pump brings atoms from the ground state to state 2. Then the atoms quickly deexcite (in a non radiative way) from state 2 to state 1, where they can deexcite by stimulated emission to amplify light. In order to have a strong population inversion between the ground state and state 1, it is necessary for the fluorescence lifetime between levels 2 and 1, τ_{21} , to be very short compared to all other fluorescence lifetimes (τ_{20} between state 2 and ground and τ between state 1 and ground). This way, as soon as atoms arrive in state 2, they relax in state 1 keeping state 2 always empty ($N_2 \approx 0$) and maintaining a population inversion between the two levels of the amplification transition.



Figure 3.3 – (Left) Energy levels of a three-level system with the fluorescence lifetimes. (Right) Population density of each level. Since τ_{21} is faster than τ and τ_{20} , level 2 is almost always empty and level 1 is more populated than the ground state.

Four-level systems In a four-level system, the amplification transition $(2 \rightarrow 1)$ is surrounded by two other levels. The ground state (noted 0) below and a higher excited state (noted 3) as represented in Fig. 3.4. With the pumping, atoms are brought from ground state to state 3. Then, as in a three-level system, they decay (in a non radiative way) to state 2 where they can amplify light by stimulated emission. Finally, they decay again, still in a non radiative way, from state 1 to the ground state. Like in a three-level system, there are conditions on the fluorescence lifetimes for a four-level system to work. Indeed, the fluorescence lifetimes of transitions $3 \rightarrow 2$ and $1 \rightarrow 0$ have to be very short compared to all other lifetimes. This way, the upper state is always empty, and pumped atoms go almost directly from ground level to level 2. The main advantages of four-level systems compared to three-level systems is the possibility to maintain the lower level of the amplification transition almost always empty. This is because this level is not the ground state, as a result it empties itself very fast in the ground state. Indeed, as soon as atoms arrive in level 1, they deexcite to go into the ground state. This means that the population inversion $\Delta N = N_2 - N_1$ is approximately equal to N_2 because $N_1 \approx 0$. Four-level systems can thus achieve very high population inversion leading to strong gain (since the gain, α , is proportional to ΔN). One drawback is that they usually generate more heat than three-level system because of the two non radiative decays.



Figure 3.4 – (Left) Energy levels of a four-level system with the fluorescence lifetimes. (Right) Population density of each level. Since τ_{32} is faster than τ , τ_{30} and τ_{31} , level 3 is almost always empty. In addition, the fast decay time τ_{10} guarantee a large population inversion between levels 2 and 1.

3.2 Gain saturation phenomenon.

When the intensity of the light inside the gain medium increases, the amplification decreases. This phenomenon is called *gain saturation*. A simple way of explaining this, is to consider that when the light is very intense, deexcitation by stimulated emission happens too fast. As a result the medium is not able to maintain a large population inversion anymore (there is a finite number of atoms in the medium), and the gain decreases. This section presents a simple model of the gain saturation in the steady state regime based on the rate equations inside a four-level system.

3.2.1 Rate equations.

The rate equations consist in modelling the population density of each level to have an expression of the population inversion. The notations are the one given in Fig. 3.4. First we will consider that level 1 is almost empty, so that $\Delta N = N_2 - N_1 \approx N_2$. As a result:

$$\frac{\partial \Delta N}{\partial t} \simeq \frac{\partial N_2}{\partial t} = \frac{N_3}{\tau_{32}} - \frac{N_2}{\tau_{20}} - \frac{N_2}{\tau} - \frac{\sigma_e I}{h\nu} N_2 + \frac{\sigma_e I}{h\nu} N_1 \qquad (3.5)$$

We can assume that the fluorescence lifetime of transition $2 \to 1, \tau$, is very short compared to the lifetime of transition $2 \to 0, \tau_{20}$ $(1/\tau \gg 1/\tau_{20})$. The previous equation can thus be simplified as:

$$\frac{\partial \Delta N}{\partial t} = \frac{N_3}{\tau_{32}} - \frac{\Delta N}{\tau} - \frac{\sigma_e I}{h\nu} \Delta N \tag{3.6}$$

The rate equation for level 3 is:

$$\frac{\partial N_3}{\partial t} = \frac{\stackrel{\text{Pumping}}{R_P N_0}}{R_P N_0} - \frac{\stackrel{\text{Stimulated}}{R_P N_3}}{R_P N_3} - \frac{\stackrel{\text{Deexcitation}}{N_3}}{\tau_{32}} - \frac{\stackrel{\text{Spontaneous}}{N_3}}{\tau_{31}} - \frac{\stackrel{\text{Nometric}}{N_3}}{\tau_{30}} \qquad (3.7)$$

$$= R_P (N_0 - N_3) - N_3 \left(\frac{1}{\tau_{32}} + \frac{1}{\tau_{31}} + \frac{1}{\tau_{30}}\right)$$

where R_P is the effective pumping rate. The lifetime of transition $3 \rightarrow 2$, τ_{32} , is very short compared to τ_{31} and τ_{30} $(1/\tau_{32} \gg 1/\tau_{31} + 1/\tau_{30})$. As a result, $N_3 \ll N_0$ so if we suppose that $\partial N_3/\partial t \approx 0$, the previous equation yields:

$$\frac{N_3}{\tau_{32}} = R_P N_0 \tag{3.8}$$

Which, when inserted into 3.6, gives:

$$\frac{\partial \Delta N}{\partial t} = R_P N_0 - \frac{\Delta N}{\tau} - \frac{\sigma_e I}{h\nu} \Delta N \tag{3.9}$$

3.2.2 Steady state regime.

To be in the steady state regime, we need to consider that the electromagnetic wave is continuous or that the pulse duration, t_p , is longer than the fluorescence lifetime τ . In addition, the pumping is supposed to be continuous or to last a long enough time for the gain to be constant over time. In this regime the population inversion can simply be written using Eq. 3.9 with the left hand term equal to 0:

$$\Delta N = \frac{\tau R_P N_{tot}}{1 + \frac{I\sigma_e \tau}{h\nu}} \tag{3.10}$$

Here we have added one last hypothesis stating that $N_0 \approx N_{tot}$ (very often true in practice).

From this equation, we can see that the population inversion, and thus the gain, depends on the intensity of the light inside the medium. The gain starts to saturate when $(I\sigma_e\tau)/(h\nu) \lesssim 1$. The saturation intensity of the amplifier is then defined by:

$$I_{sat} = \frac{h\nu}{\sigma_e \tau} \tag{3.11}$$

We can also define the *small-signal gain*, α_0 , which is the unsaturated gain:

$$\alpha_0 = \sigma_e R_P \tau N_{tot} \tag{3.12}$$

Using these notations the linear gain becomes:

$$\alpha = \sigma_e \Delta N = \frac{\alpha_0}{1 + \frac{I}{I_{sat}}} \tag{3.13}$$

The saturation intensity is the value of the intensity inside the medium for which the linear gain is equal to half of its small-signal value.

3.3 Hologram recording in gain medium.

Gain saturation is the key phenomenon for holography in gain media. We will now see how one can use it in order to record a hologram within the medium by spatially modulating the gain.

3.3.1 Propagation of light inside a gain medium.

Before introducing the spatial modulation of gain, let us look at how light propagates inside a pumped gain medium. We suppose that it is a dielectric medium with an external source of energy bringing atoms in the higher state of the laser transition. From Maxwell's equation, it is possible to derive the propagation equation of an electric field \mathbf{E} inside this medium:

$$\Delta \mathbf{E} - \frac{1}{c^2} \frac{\partial^2 \mathbf{E}}{\partial t^2} = \frac{1}{\epsilon_0 c^2} \frac{\partial^2 \mathbf{P}}{\partial t^2}$$
(3.14)

where c is the speed of light in vacuum, ϵ_0 is the vacuum permittivity and **P** is the polarisation vector of the medium. A solid gain medium is usually composed of a crystalline matrix with optically active atoms embedded inside. The polarisation of the medium can be decomposed into two contributions. One from the crystalline matrix and one from the active atoms: $\mathbf{P} = \mathbf{P}_{host} + \mathbf{P}_{at}$ with \mathbf{P}_{host} being the linear, non resonant polarisation of the crystalline matrix and \mathbf{P}_{at} is the polarisation due to the active atoms. They can be written, using Siegman's notation [46]:

$$\mathbf{P}_{host} = \chi_{host} \epsilon_0 \mathbf{E} \tag{3.15}$$

$$\mathbf{P}_{at} = \chi \epsilon_{host} \mathbf{E} \tag{3.16}$$

where χ_{host} is the crystalline matrix susceptibility. It is linked to its refractive index by: $n^2 = 1 + \chi_{host} = \epsilon_{host}/\epsilon_0$. ϵ_{host} is the matrix permittivity and χ is the susceptibility of the active atoms (the same as the one introduced in Eqs. 3.3). Using Eqs. 3.15 and 3.16, the propagation equation 3.14 can be changed into:

$$\Delta \mathbf{E} - \frac{n^2}{c^2} (1+\chi) \frac{\partial^2 \mathbf{E}}{\partial t^2} = 0$$
(3.17)

3.3.2 Spatial modulation of gain.

The spatial modulation of the gain is achieved through the modulation of the susceptibility. First, we will suppose that there is only gain and no phase shift. However in certain conditions, there is an induced refractive index grating in addition to the gain grating. According to Eq. 3.3, χ is linked to the gain α :

$$\chi = \frac{i}{k} \alpha$$

$$= \frac{i}{k} \frac{\alpha_0}{1 + \frac{I}{I_{sat}}}$$
(3.18)

Now, in order to record an hologram inside a gain medium, it is necessary to have two interfering beams. The total electric field inside the medium is the interference between a signal beam E_S and a reference beam E_R :

$$E(x, y, z, t) = |A_S(x, y)|e^{i(\omega t - \mathbf{k}_S \cdot \mathbf{r} + \phi_S)} + |A_R(x, y)|e^{i(\omega t - \mathbf{k}_R \cdot \mathbf{r} + \phi_R)}$$
(3.19)

The intensity in the crystal is thus:

$$\frac{I(x, y, z, t) \propto |E(x, y, z, t)|^2}{\propto |A_S(x, y)|^2 + |A_R(x, y)|^2 + 2|A_S||A_R|\cos(\Delta \mathbf{K} \cdot \mathbf{r} - \Delta \Phi)}$$
(3.20)

where $\Delta \mathbf{K} = \mathbf{k}_R - \mathbf{k}_S$ and $\Delta \Phi$ is the phase difference between the signal and the reference. This expression of the intensity of the light can be inserted into the susceptibility given by Eq. 3.18:

$$\chi = \frac{i}{k} \frac{\alpha_0}{1 + \frac{|A_S(x,y)|^2 + |A_R(x,y)|^2}{I_{sat}} + 2\frac{|A_S||A_R|}{I_{sat}}\cos(\Delta \mathbf{K} \cdot \mathbf{r} - \Delta \Phi)}$$
(3.21)

The mean intensity of the interference pattern I_M , and the amplitude of the intensity of the interference fringes I_F can be defined as:

$$I_M = \frac{|A_S(x,y)|^2 + |A_R(x,y)|^2}{I_{sat}}$$
(3.22)

$$I_F = 2 \frac{|A_S||A_R|}{I_{sat}}$$
(3.23)

The expression of χ is thus simplified as:

1

$$\chi = \frac{i}{k} \frac{\alpha_0}{1 + I_M + I_F \cos(\Delta \mathbf{K} \cdot \mathbf{r} - \Delta \Phi)}$$
(3.24)

The expression of the susceptibility given by Eq. 3.24 shows that, in the presence of an interference pattern, light propagating in the medium "sees" a spatially modulated gain pattern. In the case of weak saturation $(I_R, I_S \ll I_{sat})$, we have $I_M \ll 1$, thus Eq. 3.24 can be simplified using Taylor series as follows:

$$\chi = \frac{i\alpha_0}{k} \left[1 - I_F \cos(\Delta \mathbf{K} \cdot \mathbf{r} - \Delta \Phi) \right]$$

$$\Rightarrow \alpha = \alpha_0 \left[1 - I_F \cos(\Delta \mathbf{K} \cdot \mathbf{r} - \Delta \Phi) \right]$$
(3.25a)

In this case the hologram is sinusoidal and the gain grating is true to the interference pattern. The modulation of the gain by a sinusoidal intensity pattern is represented in Fig. 3.5. When there is no light propagating through the pumped medium, the population



Figure 3.5 – (Left) In the absence of incoming light in the medium the population inversion is the same in the whole volume. (Right) When two beams create an interference pattern inside the gain media, the bright fringes saturate the population inversion creating a small gain region. Whereas the dark fringes do not saturate the population inversion keeping the gain high.

inversion is spatially constant. Whereas, with an interference pattern, the intensity of the light spatially varies. Bright fringes tend to deplete the population inversion resulting in a lower gain. And conversely, dark fringes do not affect the population inversion so the gain remains high. This effect is local, meaning that the gain grating is aligned with the interference pattern. Actually, as a bright fringe corresponds to a low gain and *vice versa*, one can say that the gain grating has a π shift relative to the interference pattern³. The

³Some materials, such as certain kind of photorefractive crystals, have a non-local response. The refractive index grating has a $\pi/2$ shift relative the interference pattern, which result in $\pi/2$ phase shift in the holographic process.

gain grating can then be used in order to perform wave mixing. The next chapter details two types of wave mixing:

- Two-wave mixing (TWM): It results from the diffraction of the reference beam on the hologram it has imprinted through the interference with the signal. This process generates a beam that has the same wavefront as the signal beam.
- Four-wave mixing (FWM): It results from the diffraction of a second reference beam on the hologram imprinted through the interference of the first reference with the signal. The second reference has to be counter propagating compared to the first reference. The result of this diffraction is the generation of the phase conjugate of the signal beam.

These two processes will be detailed in chapter 4 through mathematical models that describe the interactions of two or four beams inside a gain medium.

3.3.3 Spatial modulation of the index of refraction.

In the previous section, we supposed the light was not phase shifted due to the amplification. However, in reality, the gain grating also induces a refractive index grating. Here we simply give an overview of the mechanisms responsible for the refractive index modulation. In solid state laser such as Nd:YAG or Nd:YVO₄ (which is used in our experiments), the modulation of the index of refraction is due to the variations of the polarisability of atoms when they are in different states [47,48]. There are two contributions to this phenomenon: a non-resonant contribution due to the UV absorption bands and a resonant contribution that appears when there is a frequency mismatch between the light and the laser transition. These refractive index changes are usually neglected, however recent works studied them in order to better understand the physics behind these phenomena [49, 50].

In our experiments described in part III, we used a Nd:YVO₄ gain medium, while our laser source is a Nd:YAG. The emission wavelength of Nd:YAG is 1064.2 nm whereas the emission wavelength of Nd:YVO₄ is 1064.3 nm. As mentioned earlier this wavelength detuning adds a resonant contribution to the imaginary part of the susceptibility χ as illustrated by the variation of the gain and the refractive index in Fig. 3.6 extracted from [50]. The detuning is quantified by the normalised frequency difference:

$$\delta = \frac{\nu_0 - \nu_C}{\Delta \nu} = \frac{\frac{1}{\lambda_0} - \frac{1}{\lambda_C}}{\frac{\Delta \lambda}{\lambda_C^2}}$$

Where ν_0 , ν_C and $\Delta\nu$ are respectively the maximum gain frequency of the amplifier, the working frequency and the frequency width of the gain curve. λ_0 , λ_C and $\Delta\lambda$ are the wavelength equivalents. In the same reference, there are a few formulae to quantify the variation of the refractive index in Nd:YVO₄ with an illumination coming from a Nd:YAG laser. First these variations are linked to the Henry factor by:

$$\Delta n = \frac{\beta \alpha}{2k_0} \tag{3.26}$$


Figure 3.6 – Theoretical variations of $\alpha_0 L$ and ΔnL near the laser resonance as a function of the frequency detuning. It was calculated for $\alpha_0 L(\delta = 0) = 1.5$ and $\beta_{NR} = 3$ at the resonance wavelength (taken from [50]).

where α is the small signal gain, $k_0 = 2\pi/\lambda_0$ the illumination wave vector amplitude, and β is the Henry factor. It is the ratio between the refractive index contribution to the susceptibility, χ' , and gain contribution to the susceptibility, χ'' : $\beta = \chi'/\chi''$. The Henry factor can be expressed as the sum of the resonant contribution and the non-resonant contribution: $\beta = \beta_R + \beta_{NR}$ which respective expressions are:

$$\beta_R \approx 2 \frac{\frac{1}{\lambda_0} - \frac{1}{\lambda_C}}{\frac{\Delta\lambda}{\lambda_C^2}} \quad \text{and} \quad \beta_{NR} = \frac{8\pi^2 F_l^2 \Delta\alpha}{n_0 \lambda_0 \sigma_e} (1 + \delta^2) \quad \text{with} \quad F_l = \frac{n_0^2 + 2}{3}$$
(3.27)

where $\Delta \alpha$ is the polarisability variation of Nd:YVO₄, σ_e is the emission cross section at λ_C and F_l is the Lorentz factor. The values of all these parameters can be found in [51,52] and are given in Tab. 3.1. They yield for β_R and β_{NR} respectively 0.22 and 0.082, and for

n_0	2	
$\Delta \alpha$	0.095	\AA^3
λ_0	1064.2	nm
σ_e	18×10^{-19}	cm^2
λ_C	1064.3	nm
$\Delta\lambda$	0.9	nm
α_0	7	$\rm cm^{-1}$ (measured in chapter 5)

Table 3.1 – Optical characteristics of Nd: YVO_4 .

the refractive index variation we have $\Delta n = 3.6 \times 10^{-5}$. This calculated value of Δn is, indeed, not that small. Changes of the order of 10^{-5} in the refractive index is not small (it is the same order of magnitude as in photorefractive crystals [53]). The small phase variations introduced by these refractive index changes become non negligible in our case since the recorded grating is thick.

3.4 Conclusion.

In this chapter, we have described the phenomena that take place inside a pumped gain medium illuminated by light with a frequency equal or close to the frequency of the transition of amplification. This led us to explain the functioning of an optical amplifier and in particular the gain saturation. We then saw how this saturation could be exploited in order to dynamically write holograms inside the medium as a spatial modulation of the gain. Even if most of the time it will be neglected, we saw that the gain grating was also accompanied by a refractive index grating. As explained later, in chapter 6, this spatial modulation of the refractive index enables a linear detection in addition to a quadratic one. This linear detection, due to the refractive index grating, is crucial to the detection of photoacoustic signals, reported in chapter 7.

The next chapter explains the two kinds of wave mixing interactions used in our experimental work: two-wave mixing and four-wave mixing. They are used to perform respectively wavefront adaption and phase conjugation.

CHAPTER 4

Two and Four wave mixing in $\ensuremath{\mathsf{Nd}}\xspace{\mathsf{YVO}}_4$

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In the previous chapter we explained how light was amplified in a gain medium. In addition, we saw that, it was possible to saturate the gain, which could be exploited to record hologram within the medium in the form of a gain grating.

Two interesting holographic processes are treated in this second chapter of part II: twowave mixing (TWM) in section 4.1 and four-wave mixing (FWM) in section 4.2. The first one is used to perform wavefront adaption whereas FWM is a way to generate phase conjugate beams. In the whole chapter we suppose that our gain medium is a crystal of Nd:YVO₄, pumped by a high-power laser diode at 808 nm. In Nd:YVO₄, the optically active objects are the Nd³⁺ ions embedded inside the matrix host. They can be modelled by a four-level system. We will suppose that the crystal is doped at 1% with Nd³⁺, which gives a fluorescence lifetime of the laser transition: $\tau \approx 90 \,\mu s$ and a saturation intensity around $I_{sat} \approx 1 \,\mathrm{kW \, cm^{-2}}$.

4.1 Two wave mixing in $Nd:YVO_4$.

In this section we develop a model of the interactions between two coherent beams inside a pumped gain medium. We will see that the propagation of the signal beam can be described using a system of four equations (given by Eq. 4.7), that can be solved either numerically or analytically using the Laplace transform. These calculations have been reported in [54].

4.1.1 Description of the geometric configurations.

Let us suppose a uniform pumping and constant overtime. It creates a population inversion ΔN generating a gain $\alpha(\mathbf{r}, t)$. The crystal is also illuminated by a signal beam $E_S(\mathbf{r}, t)$ and a reference $E_R(\mathbf{r}, t)$ (see Fig. 4.1). The angle between the beams is supposed



Figure 4.1 – Geometric configuration and notations for the TWM model.

to be small $(2\theta \ll 1)$ so we can neglect each beam component along the z-axis. Finally, we will consider that there is no refractive index grating recorded inside the crystal.

4.1.2 Evolution of the gain.

The temporal evolution of the intensity gain, α , is governed by a differential equation [55]:

$$\frac{\partial \alpha(\mathbf{r},t)}{\partial t} = \frac{\alpha_0}{\tau} - S(\mathbf{r},t) \frac{\alpha(\mathbf{r},t)}{\tau}$$
(4.1)

where α_0 is the small signal intensity gain, τ is the spontaneous emission lifetime, and $S(\mathbf{r},t) = 1 + \frac{I(\mathbf{r},t)}{I_{sat}}$ is a saturation parameter (I_{sat} is the saturation intensity defined by Eq. 3.11). As explained in 3.3.2, it is possible to spatially modulate the gain through this saturation parameter. The two fields interfering inside the medium create the following intensity pattern:

$$I(\mathbf{r},t) \propto I_R + I_S + E_R E_S^* e^{i\mathbf{K}\cdot\mathbf{r}} + E_R^* E_S e^{-i\mathbf{K}\cdot\mathbf{r}}$$
(4.2)

where I_R and I_S are the intensity of the reference and signal beams and $\mathbf{K} = \mathbf{k}_R - \mathbf{k}_S$ is the interference pattern wave vector. Equation 4.1 can be solved by expanding $\alpha(\mathbf{r}, t)$ as a Fourier series in spatial harmonics. Thus, by keeping only the first two terms we have:

$$\alpha(\mathbf{r},t) = \alpha^{(0)} + \frac{1}{2}\alpha^{(1)}e^{i\mathbf{K}\cdot\mathbf{r}} + \frac{1}{2}\alpha^{(1)*}e^{-i\mathbf{K}\cdot\mathbf{r}}$$
(4.3)

where $\alpha^{(0)}$ is called the uniform gain (it is the gain without the spatial intensity modulation) and $\alpha^{(1)}$ is the grating contribution to the gain. Under these conditions, two coupled equations for $\alpha^{(0)}$ and $\alpha^{(1)}$ can be derived from Eq. 4.1 by identifying the terms for $\mathbf{K} = 0$ and $\mathbf{K}.\mathbf{r}$ (we have neglected the higher order terms):

$$\frac{\partial \alpha^{(0)}}{\partial t} + \frac{1}{\tau} \left(1 + \frac{I_S + I_R}{I_{sat}} \right) \alpha^{(0)} + \frac{1}{\tau} \left(\frac{E_S E_R^*}{2I_{sat}} \right) \alpha^{(1)} + \frac{1}{\tau} \left(\frac{E_S^* E_R}{2I_{sat}} \right) \alpha^{(1)*} = \frac{\alpha_0}{\tau}$$
(4.4a)

$$\frac{\partial \alpha^{(1)}}{\partial t} + \frac{1}{\tau} \left(1 + \frac{I_S + I_R}{I_{sat}} \right) \alpha^{(1)} + \frac{2}{\tau} \left(\frac{E_S E_R^*}{I_{sat}} \right) \alpha^{(0)} = 0$$
(4.4b)

In our case, the amplitude of the signal field E_S is much smaller than the reference field E_R . The intensity of the reference field, is supposed to be either small or of the order of magnitude of I_{sat} :

- $I_S \ll I_R$
- $I_R \ll I_{sat}$ or $I_R \lesssim I_{sat}$
- $|E_S E_R| \ll I_{sat}$

Under these assumptions, Eqs. 4.4 can be simplified into:

$$\frac{\partial \alpha^{(0)}}{\partial t} + \frac{1}{\tau} \left(1 + \frac{I_R}{I_{sat}} \right) \alpha^{(0)} = \frac{\alpha_0}{\tau}$$
(4.5a)

$$\frac{\partial \alpha^{(1)}}{\partial t} + \frac{1}{\tau} \left(1 + \frac{I_R}{I_{sat}} \right) \alpha^{(1)} = -\frac{2}{\tau} \left(\frac{E_S E_R^*}{I_{sat}} \right) \alpha^{(0)}$$
(4.5b)

This final system gives the temporal evolution of the gain under the illumination by two optical fields E_R and E_S that can be time dependent. In order to complete the model, we have to know the evolution of the optical fields, which is given by a coupled wave model.

4.1.3 Coupled wave propagation in a gain medium.

The modelling of the propagation in a medium of two waves interacting with each other *via* a non linearity has been done by P. Yeh. From Maxwell's equations it is possible to derive a system of coupled equation for the amplitude of each field [53]. If we consider a propagation along the z-axis we have, under the slow varying envelope approximation:

$$\frac{\partial E_S}{\partial z} = \frac{\alpha^{(0)}}{2} E_S + \frac{\alpha^{(1)}}{4} E_R \tag{4.6a}$$

$$\frac{\partial E_R}{\partial z} = \frac{\alpha^{(0)}}{2} E_R + \frac{\alpha^{(1)*}}{4} E_S \tag{4.6b}$$

The four equations given by 4.5 and 4.6 can be simplified by taking into account some experimental conditions:

- The pumping is in a steady state regime so the uniform gain $\alpha^{(0)}$ is established: $\frac{\partial \alpha^{(0)}}{\partial t} = 0$
- The reference beam is a continuous wave: $\frac{\partial I_R}{\partial t} = 0$

In the end, the propagation of the signal and reference beams inside the gain medium is governed by these four equations:

$$\alpha^{(0)}(z) = \frac{\alpha_0}{1 + \frac{I_R(z)}{I_{sat}}}$$
(4.7a)

$$\frac{\partial I_R(z)}{\partial z} = \alpha^{(0)}(z)I_R(z) \tag{4.7b}$$

$$\frac{\partial \alpha^{(1)}(z,t)}{\partial t} + \frac{1}{\tau} \left(1 + \frac{I_R(z)}{I_{sat}} \right) \alpha^{(1)}(z,t) = -\frac{2}{\tau} \left(\frac{E_S(z,t)E_R^*(z)}{I_{sat}} \right) \alpha^{(0)}(z)$$
(4.7c)

$$\frac{\partial E_S(z,t)}{\partial z} = \frac{\alpha^{(0)}(z)}{2} E_S(z,t) + \frac{\alpha^{(1)}(z,t)}{4} E_R(z)$$
(4.7d)

First, Eqs. 4.7a and 4.7b can be considered independently of Eqs. 4.7c and 4.7d in order to calculate $\alpha^{(0)}(z)$ and $I_R(z)$. Then the other equations can be numerically solved using a step by step spatio-temporal Euler's method.

4.1.4 Analytical resolution.

Another way of finding a solution to Eqs. 4.7 is to use the Laplace transform. This method leads to an integral solution of the electric field of the signal beam. Let us take

the Laplace transform of Eqs. 4.7c and 4.7d:

$$\left(\widetilde{s\alpha^{(1)}}(z,s) - \alpha^{(1)}(z,0)\right) + \frac{1}{\tau} \left(1 + \frac{I_R}{I_{sat}}\right) \widetilde{\alpha^{(1)}}(z,s) = -\frac{2}{\tau} \left(\frac{\widetilde{E_S}(z,s)E_R^*(z)}{I_{sat}}\right) \alpha^{(0)}(z) \quad (4.8a)$$

$$\frac{\partial \widetilde{E_S}(z,s)}{\partial z} = \frac{\alpha^{(0)}(z)}{2} \widetilde{E_S}(z,s) + \frac{\alpha^{(1)}(z,s)}{4} E_R(z)$$
(4.8b)

At t = 0 the signal beam is turned ON so there is no interference pattern, which means that: $\alpha^{(1)}(z, 0) = 0$. Equation 4.8a becomes:

$$\widetilde{\alpha^{(1)}}(z,s) = -\frac{2}{\tau} \frac{E_R^*(z)}{I_{sat}} \alpha^{(0)}(z) \frac{\widetilde{E_S}(z,s)}{s + \frac{1}{\tau} \left(1 + \frac{I_R(z)}{I_{sat}}\right)}$$
(4.9)

We can now insert 4.9 into 4.8b. With a few manipulations this yields:

$$\frac{\partial \widetilde{E}_{S}(z,s)}{\widetilde{E}_{S}(z,s)} = \frac{\alpha^{(0)}(z)}{2} dz - \frac{\alpha^{(0)}(z)}{2\tau} \frac{I_{R}(z)}{I_{sat}} \frac{1}{s + \frac{1}{\tau} \left(1 + \frac{I_{R}(z)}{I_{sat}}\right)} dz
= \frac{\alpha^{(0)}(z)}{2} dz - \frac{1}{2} \frac{\frac{\partial}{\partial z} \left[s + \frac{1}{\tau} \left(1 + \frac{I_{R}(z)}{I_{sat}}\right)\right]}{s + \frac{1}{\tau} \left(1 + \frac{I_{R}(z)}{I_{sat}}\right)} dz$$
(4.10)

This equation can now be integrated between 0 and z:

$$\ln\left(\widetilde{E_S}(z,s)\right) = \frac{1}{2} \int_0^z \alpha^{(0)}(z') dz' - \frac{1}{2} \ln\left[s + \frac{1}{\tau} \left(1 + \frac{I_R(z)}{I_{sat}}\right)\right] + K_1(s)$$
$$\widetilde{E_S}(z,s) = K_2(s) \exp\left[\frac{1}{2} \int_0^z \alpha^{(0)}(z') dz'\right] \left[s + \frac{1}{\tau} \left(1 + \frac{I_R(z)}{I_{sat}}\right)\right]^{-\frac{1}{2}}$$
(4.11)

The boundary condition for z = 0 is:

$$\widetilde{E_S}(0,s) = K_2(s) \left[s + \frac{1}{\tau} \left(1 + \frac{I_R(0)}{I_{sat}} \right) \right]^{-\frac{1}{2}}$$

We can now replace $K_2(s)$ in the expression of $\widetilde{E}_S(z,s)$ given by Eq. 4.11:

$$\widetilde{E_S}(z,s) = \widetilde{E_S}(0,s) \exp\left[\frac{1}{2} \int_0^z \alpha^{(0)}(z') dz'\right] \left[\frac{s + \frac{1}{\tau} \left(1 + \frac{I_R(0)}{I_{sat}}\right)}{s + \frac{1}{\tau} \left(1 + \frac{I_R(z)}{I_{sat}}\right)}\right]^{\frac{1}{2}}$$

$$= \widetilde{E_S}(0,s) \exp\left[\frac{1}{2} \int_0^z \alpha^{(0)}(z') dz'\right] \left[1 - \frac{\frac{I_R(z) - I_R(0)}{\tau I_{sat}}}{s + \frac{1}{\tau} \left(1 + \frac{I_R(z)}{I_{sat}}\right)}\right]^{\frac{1}{2}}$$
(4.12)

In order to clarify the expression, we can define P and Q as:

$$P = \frac{I_R(z) - I_R(0)}{\tau I_{sat}} \quad \text{and} \quad Q = s + \frac{1}{\tau} \left(1 + \frac{I_R(z)}{I_{sat}} \right)$$

As we have P/Q < 1, Eq. 4.12 can be expanded as a power series:

$$\widetilde{E_S}(z,s) = \widetilde{E_S}(0,s) \exp\left[\frac{1}{2} \int_0^z \alpha^{(0)}(z') dz'\right] \sum_{n=0}^\infty (-1)^n \frac{\Gamma(1/2+1)}{\Gamma(n+1)\Gamma(1/2-n+1)} \left(\frac{P}{Q}\right)^n$$
(4.13)

with Γ being the usual Gamma function. The last step towards the temporal evolution of E_S is to take the inverse Laplace transform of Eq. 4.13. Only Q has a dependency in the variable s. Thus, we only need to find the inverse Laplace transform of $1/Q^n$. Well, we know that:

$$TL^{-1}\left(\frac{1}{Q^n}\right) = TL^{-1}\left(\frac{1}{\left(s + \frac{1}{\tau}\left[1 + \frac{I_R(z)}{I_{sat}}\right]\right)^n}\right)$$
$$= \begin{cases} \delta(t) \quad \text{(Dirac function)} & \text{for } n = 0\\ \exp\left[-\frac{t}{\tau}\left(1 + \frac{I_R(z)}{I_{sat}}\right)\right]\frac{t^{n-1}}{\Gamma(n)} & \text{for } n \ge 1 \end{cases}$$

Consequently, the inverse Laplace transform of Eq. 4.13 is:

$$E_{S}(z,t) = E_{S}(0,t) \exp\left[\frac{1}{2} \int_{0}^{z} \alpha^{(0)}(z')dz'\right] * \left\{\delta(t) + \exp\left[-\frac{t}{\tau} \left(1 + \frac{I_{R}(z)}{I_{sat}}\right)\right] \sum_{n=1}^{\infty} (-1)^{n} P^{n} \frac{\Gamma(1/2+1)}{\Gamma(n+1)\Gamma(1/2-n+1)} \frac{t^{n-1}}{\Gamma(n)}\right\} (4.14)$$

Where f * g is an "incomplete" convolution operator defined by:

$$(f * g)(t) = \int_0^t f(t-\tau)g(\tau)d\tau$$

This equation can be rearranged into:

$$E_S(z,t) = \exp\left(\frac{1}{2}\int_0^z \alpha^{(0)}(z')dz'\right) \times \left[E_S(0,t) + E_S(0,t) * C(z,t)\right]$$
(4.15)

Where C(z,t) is defined by:

$$C(z,t) = \frac{1}{\tau} \exp\left[-\frac{t}{\tau} \left(1 + \frac{I_R(z)}{I_{sat}}\right)\right] \times \sum_{n=1}^{\infty} \left[(-1)^n \left(\frac{I_R(z) - I_R(0)}{I_{sat}}\right)^n \frac{\Gamma(1/2+1)}{\Gamma(n+1)\Gamma(1/2-n+1)} \frac{\left(\frac{t}{\tau}\right)^{n-1}}{\Gamma(n)} \right] \quad (4.16)$$

The formula given by Eq. 4.15 is convenient to understand the effects of the crystal on the signal beam. Indeed, the intensity at the exit of the crystal can be written:

$$E_{S}(L,t) = \exp\left(\frac{1}{2}\int_{0}^{L} \alpha^{(0)}(z')dz'\right) \times \left[E_{S}(0,t) + E_{S}(0,t) * C(L,t) \right]$$
Amplification inside the crystal

The first term corresponds to the amplification of the light throughout its propagation inside the crystal. The second term is the sum of two contributions: the transmitted signal through the crystal, and a version of the signal filtered by the crystal with a transfer function given by Eq. 4.16.

At the first order, the transfer function of the crystal is the one of a lowpass filter with a response time $\tau_R = \tau \left(1 + \frac{I_R}{I_{sat}}\right)^{-1}$. This value of the response time was expected considering Eqs. 4.5. Hence, the uniform gain, $\alpha^{(0)}$ and the modulated gain, $\alpha^{(1)}$ have a temporal behaviour governed by τ_R . The holographic process in a gain medium, such as Nd:YVO₄, is therefore very fast since its time constant is shorter than the fluorescence lifetime of the laser transition (around 90 µs in our case). In conclusion, gain media are capable of fast wavefront adaption if the fluorescence lifetime is short enough, which is usually the case. Finally, it can be noted that, if we consider only the first term of the summation in C(z, t), the formula is consistent with the formalism from [55] in the case of an established modulation.

4.1.5 Numerical solution of the coupled wave system.

The last equation (Eq. 4.15) can be used to numerically calculate the signal coming out of the crystal after the TWM process. It has been used to compute the temporal evolution of the gain factor for a signal beam that is turned ON at t = 0. The gain factor is defined by:

$$G = \frac{I_S(L,t)}{I_S(0,t)}$$

We chose to calculate this step response because it gives a good idea of the time needed



Figure 4.2 – Theoretical value of the gain versus time in a TWM configuration for several value of the ratio I_{R0}/I_{sat} .

for the system to reach a steady state regime. Also, it is easy to realise experimentally so

we will be able to compare the results with the experiment¹. The computed gain versus time (we normalised the output intensity by the input intensity) is represented in Fig. 4.2.

With this simulation, we can see the influence of the saturation parameter on the signal beam. First, the more saturated the gain is, the lower the initial gain. In addition, the wave mixing and the hologram writing can be seen in the temporal behaviour. The gain on the signal beam decreases overtime to reach a stationary value. This gain loss is larger when the gain is strongly saturated. We can also comment on the dynamic of the holography. The hologram writing process is fast in Nd:YVO₄, since it needs only about 100 µs to reach a stationary value. As mentioned in 2.2.3, this fast holographic response time is the most attractive features of holography in gain media. Indeed, as we aim to use this technique for the detection of modulation in speckle that decorrelates in about 1 ms, it is necessary to have a faster response time.

4.1.6 With a refractive index modulation.

One of the main weakness of the previous model, is that it does not consider the refractive index contribution to the wave mixing. Indeed, as explained in 3.3.3, in addition to the gain grating, there is a refractive index grating recorded within the crystal. In this section we will modify a little the equations to take into account this additional contribution.

The gain α is described by a real number. One way of considering the refractive index modulation is to add an imaginary part to α . The gain is now a complex number that we we will write:

$$\alpha(\mathbf{r},t) = \alpha_g(\mathbf{r},t) + i\alpha_n(\mathbf{r},t) \tag{4.17}$$

where $\alpha_g(\mathbf{r}, t) = k\chi''$ describes the gain contribution and $\alpha_n(\mathbf{r}, t)$ the refractive index contribution. As $\alpha_g(\mathbf{r}, t)$ is the gain contribution, all the work done previously on the evolution of the gain (section 4.1.2) is still valid if we switch α by α_g . Especially, the system of Eqs. 4.5 remains the same:

$$\frac{\partial \alpha_g^{(0)}}{\partial t} + \frac{1}{\tau} \left(1 + \frac{I_R}{I_{sat}} \right) \alpha_g^{(0)} = \frac{\alpha_{g0}}{\tau}$$
(4.18a)

$$\frac{\partial \alpha_g^{(1)}}{\partial t} + \frac{1}{\tau} \left(1 + \frac{I_R}{I_{sat}} \right) \alpha_g^{(1)} = -\frac{2}{\tau} \left(\frac{E_S E_R^*}{I_{sat}} \right) \alpha_g^{(0)}$$
(4.18b)

The same way α_g is link to the susceptibility by $\alpha_g = k\chi''$, we can link α_n to the susceptibility by $\alpha_n = k\chi'$. Yet, χ' is proportional to χ'' through the Henry factor (defined in section 3.3.3), as a result:

$$\chi' = \beta \chi'' \tag{4.19a}$$

$$\Rightarrow \alpha_n = k\chi' = k\beta\chi'' = \beta\alpha_g \tag{4.19b}$$

Which means that the complex gain can be expressed simply: $\alpha(\mathbf{r}, t) = (1 + i\beta)\alpha_g(\mathbf{r}, t)$. The coupled wave equations 4.6 stay the same, but with the complex value of α . If we

¹This is done in chapter 6, section 6.2.2

neglect the second term of the propagation equation of the reference field (the signal field is much small than the reference field), we have:

$$\frac{\partial E_S}{\partial z} = \frac{\alpha_g^{(0)}(1+i\beta)}{2} E_S + \frac{\alpha_g^{(1)}(1+i\beta)}{4} E_R \tag{4.20a}$$

$$\frac{\partial E_R}{\partial z} = \frac{\alpha_g^{(0)}(1+i\beta)}{2} E_R \tag{4.20b}$$

Ultimately, the propagation of the electric fields is governed by a system close to 4.7:

$$\alpha_g^{(0)}(z) = \frac{\alpha_{g0}}{1 + \frac{I_R(z)}{I_{sat}}}$$
(4.21a)

$$\frac{\partial E_R(z)}{\partial z} = (1+i\beta) \frac{\alpha_g^{(0)}(z)}{2} E_R(z)$$
(4.21b)

$$\frac{\partial \alpha_g^{(1)}(z,t)}{\partial t} + \frac{1}{\tau} \left(1 + \frac{I_R(z)}{I_{sat}} \right) \alpha_g^{(1)}(z,t) = -\frac{2}{\tau} \left(\frac{E_S(z,t)E_R^*(z,t)}{I_{sat}} \right) \alpha_g^{(0)}(z)$$
(4.21c)

$$\frac{\partial E_S(z,t)}{\partial z} = \frac{\alpha_g^{(0)}(z)(1+i\beta)}{2} E_S(z,t) + \frac{\alpha_g^{(1)}(z,t)(1+i\beta)}{4} E_R(z)$$
(4.21d)

This system can be solved using the same strategy as in the previous section. Therefore we will detail less the calculations than in section 4.1.4. We will just write the main steps.

The Laplace transform of Eq. 4.21c and 4.21d leads to modified version of Eqs. 4.8a and 4.10:

$$\widetilde{\alpha_g^{(1)}}(z,s) = -\frac{2}{\tau} \frac{E_R^*(z)}{I_{sat}} \alpha_g^{(0)}(z) \frac{\widetilde{E_S}(z,s)}{s + \frac{1}{\tau} \left(1 + \frac{I_R(z)}{I_{sat}}\right)}$$
(4.22a)

$$\frac{\partial \widetilde{E}_S(z,s)}{\widetilde{E}_S(z,s)} = \frac{(1+i\beta)\alpha_g^{(0)}(z)}{2}dz - \frac{1+i\beta}{2}\frac{\frac{\partial}{\partial z}\left[s + \frac{1}{\tau}\left(1 + \frac{I_R(z)}{I_{sat}}\right)\right]}{s + \frac{1}{\tau}\left(1 + \frac{I_R(z)}{I_{sat}}\right)}dz$$
(4.22b)

After the same manipulations as in the previous section, the Laplace transform of E_S , with a refractive index modulation, is given by:

$$\widetilde{E_S}(z,s) = \widetilde{E_S}(0,s) \exp\left[\frac{1}{2} \int_0^z \alpha^{(0)}(z') dz'\right] \left[1 - \frac{\frac{I_R(z) - I_R(0)}{\tau I_{sat}}}{s + \frac{1}{\tau} \left(1 + \frac{I_R(z)}{I_{sat}}\right)}\right]^{\frac{1+i\beta}{2}}$$
(4.23)

The only difference with the case without the modulation of the refractive index (given by Eq. 4.12) is the exponent which is now a complex number. The temporal evolution of the signal field is given by the inverse Laplace transform of Eq. 4.23 using the Gamma function generalised to complex arguments:

$$E_S(z,t) = \exp\left(\frac{1+i\beta}{2}\int_0^z \alpha_g^{(0)}(z')dz'\right) \times [E_S(0,t) + E_S(0,t) * C_n(z,t)]$$
(4.24)

with $C_n(z,t)$ is the transfer function of the crystal taking into account the refractive index

modulation. It is defined by:

$$C_n(z,t) = \frac{1}{\tau} \exp\left[-\frac{t}{\tau} \left(1 + \frac{I_R(z)}{I_{sat}}\right)\right] \times \sum_{n=1}^{\infty} \left[(-1)^n \left(\frac{I_R(z) - I_R(0)}{I_{sat}}\right)^n \frac{\Gamma(\frac{1+i\beta}{2} + 1)}{\Gamma(n+1)\Gamma(\frac{1+i\beta}{2} - n + 1)} \frac{\left(\frac{t}{\tau}\right)^{n-1}}{\Gamma(n)} \right] \quad (4.25)$$

The physical phenomena behind this mathematical expression can be interpreted using a few simplifications. If we go back to Eq. 4.23 and simplify it using a Taylor expansion at the first order:

$$\widetilde{E_S}(z,s) = \widetilde{E_S}(0,s) \exp\left[\frac{1}{2} \int_0^z \alpha^{(0)}(z') dz'\right] \left[1 - \frac{1+i\beta}{2} \frac{\frac{I_R(z) - I_R(0)}{\tau I_{sat}}}{s + \frac{1}{\tau} \left(1 + \frac{I_R(z)}{I_{sat}}\right)}\right]$$
(4.26)

We can take the inverse Laplace transform and write:

$$E_{S}(z,t) = \underbrace{e^{\frac{1}{2}\int_{0}^{z} \alpha^{(0)}(z')dz'}}_{\text{Amplification inside the crystal}} \left(\underbrace{E_{S}(0,t)}_{\text{Transmitted signal}} + \underbrace{E_{S}(0,t) * C'_{n}(z,t)}_{\text{Oiffracted signal on the gain grating}} + \underbrace{i\beta E_{S}(0,t) * C'_{n}(z,t)}_{\text{Oiffracted signal on the refractive index grating}} \right)$$
(4.27)

The signal exiting the crystal is now the sum of three terms each of them amplified by a factor $\exp\left(\frac{1}{2}\int_0^z \alpha^{(0)}(z')dz'\right)$:

- The transmitted signal: $E_S(0,t)$
- The diffracted signal on the gain grating: $E_S(0,t) * C'_n(z,t)$. The simplified crystal transfer function $C'_n(z,t)$ is given by: $C'_n(z,t) = -\frac{I_R(z) I_R(0)}{2\tau I_{sat}} e^{-\frac{t}{\tau} \left(1 + \frac{I_R(z)}{I_{sat}}\right)}$
- The diffracted signal on the refractive index grating: $i\beta E_S(0,t) * C'_n(z,t)$. The refractive index component has the same transfer function has the gain, the only difference is the pre-factor $i\beta$.

Now, in addition to the transmitted signal, there are two contributions generated by the diffraction of the reference on each grating. Both these diffracted beams are filtered version of the signal beam by a transfer function of the crystal. They have the same temporal form, only the amplitude changes. At the first order, the dynamic of the holographic process is given by the response time $\tau_R = \tau \left(1 + \frac{I_R}{I_{sat}}\right)^{-1}$. One can notice that the diffracted component on the refractive index grating has a $\pi/2$ phase shift (multiplication by *i*). This features plays a fundamental role in the detection of photoacoustic signal as we will detail in chapter 6.

Continuous modulation regime. A first verification of this model can be made in a continuous modulation regime. Let us take a phase modulated wave at a frequency f_{mod} and use Eq. 4.24 to compute the output light intensity on the detector. The signal wave

at the entrance of the crystal can be written:

$$E_S(0,t) = \exp\left(i\phi\sin(2\pi f_{mod}t)\right)$$

The output intensity is modulated, and its Fourier transform presents peaks at nf_{mod} . The amplitude of the peaks at f_{mod} and $2f_{mod}$ versus the amplitude of the modulation ϕ is represented in Fig. 4.3. To check the validity of the computed signal, the curves have



Figure 4.3 – Geometric configuration and notations for the FWM model.

been fitted by theoretical models that involve Bessel functions of the first kind J_n . Indeed, if we consider a purely phase modulated wave, and a perfect filtering by the crystal, the interference on the detector of the transmitted wave with the diffraction on the gain grating gives a signal at $2f_{mod}$ with an amplitude proportional to $J_0(\phi)J_2(\phi)$. And the interference on the detector of the transmitted wave with the diffraction on the refractive index grating yields a signal at f_{mod} with an amplitude proportional to $J_0(\phi)J_1(\phi)$. We can see on Fig. 4.3 that it fits well. The model seems to work well, the confrontation with the experiment in chapter 7 will confirm it.

The presence of peaks at f_{mod} – generally at nf_{mod} with n odd – is due to the refractive index contribution. As we can see on Fig. 4.3, for small phase variation this contribution is much higher than the gain contribution. This is because it is a linear contribution, whereas the gain contribution is quadratic. Therefore the system is very sensitive, even for small phase variations.

4.2 Four-wave mixing in $Nd:YVO_4$.

In this second section we describe a model of the interactions between three waves inside a gain medium: one signal beam, and two counter-propagating reference beams. The result of this interaction is a generation of a fourth wave, which is the phase conjugate of the signal beam. Hence the name *four-wave mixing*. The first models of FWM in saturable media have been proposed by Abrams and Lind [56] in 1978 where they studied the interaction of optical waves in saturable absorbers. In 1982, Reintjes and Palumbo did a model of phase conjugation by four wave mixing in saturable amplifiers [57]. As we did not bring any additional theory to FWM in gain media during this PhD work, the goal of this section is simply to understand how the waves interact with each other during phase conjugation by FWM. Hence, we will present FWM through a simple model in steady state regime that can be found in the last chapter² of [58]. This model does not take into account the modulation of the refractive index, we simply consider the grain grating.

4.2.1 Description of the geometric configuration.

Like in 4.1, we consider a continuously pumped crystal. The pumping creates a gain $\alpha(\mathbf{r}, t)$ governed by Eq. 4.1. The crystal is illuminated by two waves in z = 0 at $\lambda_L = 1064$ nm: a signal beam E_3 and a reference beam E_1 ; and in z = L by a second reference beam E_2 also at λ_L . The two references are counter-propagative. The interaction between those three waves generates a fourth wave called E_4 . This configuration is sketched in Fig. 4.4. The angle 2θ is supposed to be small enough so that we can neglect the z-axis component of each beam.



Figure 4.4 – Geometric configuration and notations for the FWM model.

4.2.2 Coupled wave model in steady-sate regime.

Intensity pattern In the most general case, the intensity pattern inside the crystal is the result of the summation of four two-waves interference pattern $(E_1 + E_2, E_1 + E_3, E_2 + E_3 \text{ and } E_3 + E_4)$. To simplify the problem, we will consider that there is only one

²Written by M. J. Damzen, page 367

grating written inside the crystal ³. The four electric fields will be noted:

$$\mathbf{E}_{1} = E_{1}(\mathbf{r}, t)e^{i(\omega t - \mathbf{k}_{R} \cdot \mathbf{r})}\mathbf{x}$$

$$\mathbf{E}_{2} = E_{2}(\mathbf{r}, t)e^{i(\omega t + \mathbf{k}_{R} \cdot \mathbf{r})}\mathbf{y}$$

$$\mathbf{E}_{3} = E_{3}(\mathbf{r}, t)e^{i(\omega t - \mathbf{k}_{S} \cdot \mathbf{r})}\mathbf{x}$$

$$\mathbf{E}_{4} = E_{4}(\mathbf{r}, t)e^{i(\omega t + \mathbf{k}_{S} \cdot \mathbf{r})}\mathbf{y}$$

The resulting intensity pattern, normalised by the saturation intensity, is given by [58]:

$$\frac{I(\mathbf{r},t)}{I_{sat}} = I_M + |I_F| \cos(\Delta \mathbf{K} \cdot \mathbf{r} - \phi_F)$$
(4.28)

with:

$$I_M = \frac{1}{I_{sat}} \sum_{j=1}^4 |E_j(\mathbf{r}, t)|^2$$
(4.29a)

$$I_F = |I_F|e^{i\phi_F} = \frac{2}{I_{sat}}(E_1E_3^* + E_2E_4^*)$$
(4.29b)

$$\Delta \mathbf{K} = \mathbf{k}_R - \mathbf{k}_S \tag{4.29c}$$

 I_M is the amplitude of the averaged value of the light intensity pattern and I_F is the amplitude of the modulated component of the fringes intensity pattern.

Gain modulation in steady-state regime Equation 4.28 can be inserted into Eq. 4.1 to have the modulation of the gain. First, let us suppose that we are in a steady state regime, the equation giving the evolution of the gain becomes:

$$\alpha(\mathbf{r}) = \frac{\alpha_0}{1 + \frac{I(\mathbf{r})}{I_{sat}}} \tag{4.30}$$

By substituting the interference pattern given by Eq. 4.28 in the previous equation, we obtain:

$$\alpha = \frac{\alpha_0}{1 + I_M + |I_F| \cos(\Delta \mathbf{K} \cdot \mathbf{r} - \phi_F)} = \frac{\Gamma_0}{1 + M_F \cos(\Delta \mathbf{K} \cdot \mathbf{r} - \phi_F)}$$
(4.31)

where Γ_0 is the averaged saturated gain, and M_F defines the gain modulation depth. They are given by:

$$\Gamma_0 = \frac{\alpha_0}{1 + I_M}$$
 and $M_F = \frac{|I_F|}{1 + I_M}$

Like in the TWM case, the gain is periodically modulated, so it can be decomposed as a Fourier series in spatial harmonics as follows:

$$\alpha(\mathbf{r},t) = \frac{1}{2} \sum_{n=0}^{\infty} \left(\alpha^{(n)} e^{-in(\Delta \mathbf{K} \cdot \mathbf{r} - \phi_F)} + \alpha^{(n)*} e^{in(\Delta \mathbf{K} \cdot \mathbf{r} - \phi_F)} \right)$$
(4.32)

³This is can be experimentally realised by having the second reference E_2 orthogonally polarised to E_1 and E_3

With the Fourier coefficient given by:

$$\alpha^{(0)} = \frac{\Gamma_0}{\sqrt{1 - M_F}} \tag{4.33a}$$

$$\alpha^{(n)} = (-1)^n \frac{2\Gamma_0}{\sqrt{1 - M_F}} \left(\frac{1 - \sqrt{1 - M_F}}{M_F}\right)^n$$
(4.33b)

Coupled equations The coupled equations describing the propagation of the optical fields inside the gain medium can be obtained by substituting the Fourier decomposition of α into the propagation equations of each field. Then, the selection of the phase-matched components yields:

$$\frac{dE_1}{dz} = \gamma E_1 + \kappa_F E_3 \tag{4.34a}$$

$$-\frac{dE_2}{dz} = \gamma E_2 + \kappa_F^* E_4 \tag{4.34b}$$

$$\frac{dE_3}{dz} = \gamma E_3 + \kappa_F^* E_1 \tag{4.34c}$$

$$-\frac{dE_4}{dz} = \gamma E_4 + \kappa_F E_2 \tag{4.34d}$$

where γ and κ_F are the coupling coefficient given by:

$$\gamma = \alpha^{(0)} = \frac{\Gamma_0}{\sqrt{1 - M_F}}$$
 and $\kappa_F = \frac{1}{2}\alpha^{(1)}e^{i\phi_F} = -\frac{\Gamma_0}{M_F}\left(\frac{1}{\sqrt{1 - M_F}} - 1\right)e^{i\phi_F}$

Several observations can be made on the system of coupled equations given by 4.34. In each equation, the first term conveys the amplification of the field in the medium. Therefore, γ is responsible for the gain of the medium and its capacity to amplify light from each optical beam. The second term of each equation represents the coupling between the waves because of the gain grating. Hence, κ_F quantifies the ability of the medium to transfer energy from one field to another. Since it is negative, the coupling between two waves in a gain medium is destructive, the diffraction of a beam on the self-induced grating is in antiphase with the second beam that wrote the hologram. Physically, this negative value is due to the π phase difference between the intensity pattern and the gain grating. Indeed, as explained in 3.3.2 (see Fig. 3.5), bright fringes of the interference pattern correspond to low gain and *vice versa*, as a result the gain grating is in antiphase with the interference pattern. This destructive nature can be seen in the TWM case (Fig. 4.2), where the signal decreases overtime as the hologram is recorded inside the medium.

Finally, it is worth noting that if $E_2 = 0$ and $E_4 = 0$, *i.e.*, if there are only two beams interfering inside the medium, the system of equations 4.34, becomes the system describing the TWM process.

Co-polarised beams In our experiments (reported in chapter 5), we will be in the case where all the beams have the same polarisation. This case is much more complicated

since the interference pattern can be decomposed into four contributions represented in Fig. 4.5. Two of these gratings (Figs. 4.5.(a) and 4.5.(b)) participate to the FWM. They are the transmission grating between A_1 and A_3 (read by A_2), and the reflection



Figure 4.5 – Main gratings in a co-polarised FWM experiment. (a) and (b) are the gratings participating in the FWM process. (c) and (d) are parasitic gratings between the references and the signal beams.

grating between A_2 and A_3 (read by A_1). In addition there are parasitic gratings (Figs. 4.5.(c)) and (4.5.(d)) formed by the interferences between the two references, A_1 and A_2 or between the two signal beams, A_3 and A_4 .

4.3 Conclusion

In this chapter we explained the cases of two-wave mixing and four wave-mixing in gain media. The first part was dedicated to a model we developed in order to calculate the output of an holographic system based on TWM in a gain medium. We managed, under certain conditions, to give an analytic formula that describes the amplitude of the electric field after its propagation through the gain medium. More complicated cases will require numerical methods such as Euler or Runge-Kutta to solve the system of differential equations. We used the analytic formula to show the establishment of a static hologram in the medium. The theoretical curves presented in Fig. 4.2 will be compared in chapter 6 to experimental data, which will highlight the limitations of our model. Finally, in 4.1.6, we did a first try at improving the model by taking into account the refractive index modulation. The new set of equations was confronted to a theoretical model in the simple case of a continuous phase modulated field with a small amplitude of the modulation. The agreement between the computed points and the model is good, the next step will be to confront it with experimental data, which will be done in chapter 7.

The second part of this chapter was focused on FWM. As we did not developed any additional theory for this mechanism, this part only described FWM through a model taken from the last chapter of the book *Phase Conjugate Laser Optics* [58] written by M. J. Damzen. We used the model in a simple configuration (steady-state and probe beam orthogonally polarised to the signal and reference) to understand how the waves interact

with each other. This enabled to show the specificities of wave mixing in gain medium, particularly the effects of the π phase shift between the illumination pattern inside the medium and the grain grating.

Part III

Detection of Acousto-optic and Photoacoustic signals by holography in $Nd:YVO_4$

Chapter 5

Acousto-optic detection by four-wave mixing in $\ensuremath{\mathsf{Nd}}\xspace{\mathsf{YVO}}_4$

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In this chapter we describe how the use of four-wave mixing with acousto-optic can lead to various interesting applications. Indeed, the wavefront control enabled by phase conjugation coupled with the ultrasound tagging of light allows optical imaging or focusing of light inside multiple-scattering media.

The first part of this chapter presents the distinctive features of phase conjugate beams and how they can be used to perform imaging or light focusing inside biological samples. The second part reports our experimental realisation of an acousto-optic imaging setup of a biological sample with a detection based on phase conjugation.

5.1 Acousto-optic and optical phase conjugation.

5.1.1 Properties of phase conjugate beams.

The process of degenerate four-wave mixing has the property to generate the phase conjugate of an initial incoming beam. This type of beam has two main features: they travel back along the same path as the original wave and the phase pattern is reversed.

Back propagation When a wave is reflected on a classical mirror, the direction of the reflected wave is given by Snell-Descartes' laws. The angle of reflection equals the angle of incidence, but on the other side of the normal to the mirror. Mathematically, this means that only the component of the incident wave vector normal to the mirror plane is inverted. If the incident wave vector is $\mathbf{k}_i = k_x \mathbf{x} + k_y \mathbf{y} + k_z \mathbf{z}$ and the mirror is in the XY plane, the wave vector of the reflected beam will be $\mathbf{k}_i = k_x \mathbf{x} + k_y \mathbf{y} - k_z \mathbf{z}$. In the



Figure 5.1 – Difference between a classical mirror and a *phase conjugate mirror*. (a) shows the reflection on a classic mirror and (b) the reflection on a *phase conjugate mirror*.

case of the reflection on a phase conjugate mirror, the reflected beam goes back in the

same direction as the incident beam whatever its initial direction. This means that phase conjugation generates a wave with a wave vector \mathbf{k}_r with all the components of the initial wave vector \mathbf{k}_i inverted: $\mathbf{k}_r = -\mathbf{k}_i$. The difference between these two types of mirror is illustrated in Fig. 5.1.

Reversed phase The second interesting property of a phase conjugate beam is the inversion of the phase pattern of the original wave. This is very useful as it can correct the aberrations created on a wavefront by a medium. If the phase conjugate beam travels through the aberrating medium a second time, which happens because of the back propagation, every phase aberration will be corrected after this second pass through the medium. This is illustrated in Fig. 5.2. Let us imagine that we have a plane wave sent on an aberrator that is a piece of glass ($n_g = 1.5$) surrounded by water ($n_W = 1.33$). After



Figure 5.2 – Aberration correction using phase conjugation. Effect of a round-trip in an aberrator with a reflection on a classical mirror (a) and a phase conjugate mirror (b).

this sample, the center of the wavefront has travelled through a more refractive medium thus it has a phase delay. The reflection on a *phase conjugate mirror* turns the phase delay into a phase lead as shown in Fig. 5.2.(b). This phase lead is then compensated when the wave travels back through the sample therefore correcting the aberrations introduced in the first place.

5.1.2 State of the art of the use of phase conjugation for imaging.

Analog phase conjugation The first experiments using phase conjugation for imaging date from the mid 1960s. They were performed by Kogelnik [59] in 1965 and Leith and Upatnieks [60] in 1966. They both used a similar method, which consists in recording on a spectroscopic plate the hologram of an object seen through an aberrating medium. Then, they illuminated the hologram with a counter propagating reference wave leading to the generation of phase conjugate beam that goes back through the aberrator to recreate the object in the right plane. The main difference between their experiments is the

nature of the aberrating medium. Kogelnik used an inhomogeneous glass plate, whereas Leith and Upatnieks used a ground glass. Both reported the importance of carefully aligning the hologram so that the aberrator is "brought into the exact coincidence with its reconstructed image".

During the 1970s, the community of non-linear media demonstrated many ways to generate new kinds of beams with interesting properties. These experiments relied on wave mixing with different categories of non-linear effect: photorefractive effect, Brillouin scattering or gain saturation in lasers. It is only in the late 1970s that it was understood that the wave mixing experiments were equivalent to dynamic hologram recording [61] inside the non-linear medium through the spatial modulation of its properties (refractive index, absorption, gain...).

Since then, much work has been done using phase conjugation for imaging and beam propagation, through phase disturbing media [62]. During the 1990s, Brignon *et al.* used gain media such as Nd:YAG or Nd:YVO₄ to phase conjugate images [63]. Then the next experiment of imaging through scattering media using phase conjugation was only performed in 2008 by C. Yang's team [64]. In this experiment, they used a LiNbO₃ photorefractive crystal to perform the optical phase conjugation of a wave coming from a point source, which travelled through a 0.7 mm piece of chicken breast. They showed that the phase conjugate light refocused back on the initial point source. Finally Wang's team used phase conjugation in photorefractive crystals (BSO) on ultrasonically tagged photons to focus inside a multiple scattering sample with a technique called TRUE focusing [65], which will be explained in the next section.

Digital phase conjugation The 2000s have seen the rapid development of spatial light modulators (SLM) and CCD cameras. Used together, they constitute a powerful tool for active wavefront control and digital phase conjugation. The flexibility given by digital technologies has enabled interesting applications related to imaging or focusing light through complex media. Using SLMs and CCD cameras, Vellekoop and Mosk used wavefront control to focus through strongly scattering media [66]. Following this idea, Popoff *et al.* studied the control of light propagation in disordered media, and transmitted an image using the theory of transmission matrix [67, 68]. Complex media are not necessary scattering media, Papadopoulos *et al.* managed to focus through a multimode optical fiber [69] using digital phase conjugation.

Some people used digital phase conjugation coupled with other technique to focus inside multiple scattering media. For example, Wang *et al.* used the acousto-optic effect to focus on fluorescent dyes embedded in a multiple scattering phantom [70]. Another example of the coupling of digital phase conjugation with ultrasound, is the experiment from Chaigne *et al.* which used the photoacoustic effect as a feedback loop to enhance focusing inside a scattering phantom [71, 72].

Acoustic and electromagnetism During the late 1980s, and the 1990s, the ideas developed by the community of optics inspired the community of ultrasound. The concept of phase conjugation for monochromatic waves has been extended by Fink *et al.* to large bandwidth pulses used in ultrasound and led to the first experiments on focus-

ing broadband acoustic signals through multiple scattering samples [73, 74] using timereversal. Then in 2004, Lerosey *et al.* reported the first experimental demonstration of time-reversal focusing with electromagnetic waves in the GHz domain [75].

5.1.3 Optical phase conjugation for acousto-optic imaging.

Optical phase conjugation of untagged photons As mentioned in chapter 2, acoustooptic imaging requires the detection of the tagged or the untagged photons. In order to detect the acousto-optic signal, and retrieve the local optical properties of the sample, our idea is to use phase conjugation to correct the aberrations introduced on the untagged photons wavefront. After a "round trip", the original plane wave is recovered and can be separated from the initial wave using a beam-splitter, and detected on a single photodiode. If the original wave is continuous, the intensity measured on the photodiode is constant as shown in Fig. 5.3.(a).

Then, when an ultrasonic burst travels through the scattered light inside the sample, the acousto-optic effect will "tag" some of the photons, and their frequency will be shifted $(\omega_T = \omega_L \pm \omega_{US})$. The crystal records only a time averaged hologram because its holographic response time is not fast enough to follow the ultrasonic burst. The effects of the tagging will therefore not be recorded in the hologram. As a result, the intensity of the phase conjugate beam before its second pass through the scattering medium will be constant whether the ultrasounds are ON or OFF (see Fig. 5.3.(a)). However, when the



Figure 5.3 – Amplitude of the phase conjugate signal from a multiple scattering medium in the absence of ultrasound (a) and with ultrasound (b).

phase conjugate beam re-propagates through the sample to re-generate the plane wave, it travels back through the acoustic field and some of the photons from the phase conjugate beam undergo a frequency shift by acousto-optic effect. These shifted photons, will not be retrieved in the reconstructed plane wave because the will not be coherent with the untagged photons anymore. Indeed, as demonstrated by Gross *et al.* in 2005, the various acoustic frequency components contained in an ultrasonically tagged speckle pattern are uncorrelated [76]. Consequently, when the plane wave is reconstructed, the photons that are tagged will loose their correlation with the untagged ones and will not travel back to reconstruct the plane wave as illustrated in Fig. 5.3.(b). The number of these tagged photons - and therefore the decrease of the phase conjugate beam magnitude - is directly linked to the local light intensity where the ultrasonic burst is. It is thus possible to extract the optical properties locally inside the sample with a resolution given by the ultrasound as explained in chapter 2.

Optical phase conjugation of tagged photons - TRUE focusing Another way of using optical phase conjugation in concert with acousto-optic is to set up the *phase conjugate mirror* so it performs the phase conjugation of the tagged photons instead of the untagged photons as described previously. The idea, proposed by L. V. Wang's team, is to use the acoustic focal spot as a light source, a sort of internal "guide star" inside the scattering sample. As a result, the phase conjugate of the tagged photons, will focus back on this virtual source inside the scattering medium. With this method, so called TRUE focusing, for Time-Reversed Ultrasonically Encoded focusing, they managed to focus light back inside a 10 mm-thick scattering phantom with a transport mean free path $l^* = 4 \text{ mm}$ [65]. They performed four-wave mixing in a photorefractive crystal of BSO to create their *phase conjugate mirror*. One year later, they managed to improve their setup in order to perform the TRUE focusing on a real biological tissue [77].

5.2 Experimental realisation of an acousto-optic imaging setup by phase conjugation in $Nd:YVO_4$.

This section, describes the preliminary experiments on phase conjugation in a $Nd:YVO_4$ crystal and the first results obtained using this type of crystal to perform acousto-optic imaging on a biological sample (chicken breast).

5.2.1 Preliminary experiments.

The first step towards using phase conjugation in Nd:YVO₄ for acousto-optic imaging is to build a *phase conjugate mirror* with this crystal. The setup uses a cubic crystal, whereas the crystal used for the two-wave mixing experiments in chapter 6 is a parallelepipoid in order to test another geometric configuration. In this section, we will describe the preliminary experiments we performed in order to test and verify our *phase conjugate mirror* using a Nd:YVO₄ crystal. The experimental conditions, such as the pumping power or the intensities of the different beams (reference and signal), will not be indicated because the purpose of these experiments is purely qualitative. An in-depth description of the complete experimental setup and conditions used for acousto-optic imaging will be given in section 5.2.2.

Characteristics of the crystal Our setup uses a $5 \times 5 \times 5 \text{ mm}^3$ crystal of Nd:YVO₄ doped at 1%-at with Nd³⁺ ions which gives it the optical characteristics presented in table 5.1. Because it is a-cut, the light at 1064 nm has a polarisation parallel to the c-axis to have a high gain. The crystal will be used in a double-pass configuration [78] because it enables a good overlap between the pump, the two references and the signal beams. Therefore, as shown in Fig. 5.4, the crystal has a high-reflectivity (HR) coating

5.2. Experimental realisation of an acousto-optic imaging setup by phase conjugation in $Nd:YVO_4$.

Characteristic (units)	Value
Emission wavelength $\lambda_0(nm)$	1064.3
Emission cross section $\sigma_e(\mathrm{cm}^2)$	18.10^{-19}
Emission polarisation	Along c-axis
Fluorescence lifetime τ (µs)	90
Absorption wavelength λ_{abs} (nm)	808
Absorption coefficient at λ_{abs} (cm ⁻¹)	30 (// c)
	10 (// a)
Refractive index	$n_o = 1.96$
	$n_e = 2.17$
Saturation intensity $(kW cm^{-2})$	1

Table 5.1 – Characteristics of the crystal: Nd:YVO₄ doped at 1% with Nd³⁺ ions.

for 1064 nm and an anti-reflection coating (AR) for 808 nm on its back side (in blue) and an AR coating for 1064 nm and its front side (in red). The HR coating on the back side of the crystal is used to generate the second reference beam by reflecting the input reference beam. The four other sides of the crystal are not polished, they are in contact with a water-cooled copper mount through an indium seal in order to dissipate heat generated by the pumping.



Figure 5.4 – Coating on each side of the crystal. The red side is the front side where the signal and reference beam enter and the blue side is the back side where the pump at 808 nm enters.

Realisation of an optical amplifier Before doing any phase conjugation experiments, we realized an optical amplifier with the crystal in order to optimise the alignment of the reference beam within the pumped volume. The light travels through the crystal and is amplified a first time, it is then reflected on the HR coating on the back side of the crystal to go back on its track and is amplified a second time on the way. Finally after exiting the crystal, it is separated from the input light by a non-polarising beam-splitter to be sent on a photodiode in order to measure the amplification factor. Figure 5.5 shows a simplified drawing of the setup, this double-pass configuration gives very high amplification factor defined as the ratio between the output intensity I_{OUT} to the input intensity I_{IN} : $G = I_{OUT}/I_{IN}$. Figure 5.6 represents G versus the input intensity I_{IN} . The pump works in a quasi-continuous-wave regime (with a pumping power of



Figure 5.5 – Setup for an double-pass optical amplifier.

100 W and a beam diameter on the crystal around 500 µm). I_{IN} can be measured directly on the photodiode by taking the signal when the pump is OFF, this avoids any correction due to the beam-splitter. The output intensity I_{OUT} is corrected by subtracting the amplified spontaneous emission (ASE) intensity measured when the laser is turned OFF. The saturation effect is clearly visible and the saturation intensity can be calculated by fitting the experimental data by a theoretical model [79] that yields the intensity inside the crystal in this double-pass configuration. This led to a saturation intensity of



Figure 5.6 – Amplification factor vs. the input intensity.

 $1.25 \,\mathrm{kW} \,\mathrm{cm}^{-2}$ and an amplification factor up to G = 1000 which corresponds to a small signal gain per unit length of $\alpha_0 = 7 \,\mathrm{cm}^{-1}$. These data show the great ability of Nd:YVO₄ to amplify light. It is interesting considering that the acousto-optic signal is very weak because of the speckle nature of light and the small efficiency of ultrasound tagging.

Such a double-pass amplifier, can be used as a *phase conjugate mirror* by sending a new wave S inside to perform the four-wave mixing that will generate the phase conjugate S^* . The references being the input light (R) and its reflection on the back side of the crystal (R^*) .

Generation of a phase conjugate beam in free space Our first phase conjugation experiment consisted in generating the phase conjugate of an image in free space, without any aberrator (see Fig. 5.7). The initial laser at 1064 nm is split into two beams by a polarising beam-splitter:

- The reference 1, which is reflected on the back side of the crystal and generates reference 2.
- The signal, which is enlarged and pass through an object before being focused inside the crystal.

Three half-wave plates are used to change the power ratio between the reference and the signal (HW1), and correct the polarisation of the reference (HW2) and the signal (HW3). Along the path of the signal beam is added a beam-splitter plate after the object in order to separate the counter-propagating phase conjugate from the signal beam. The collimated phase conjugate is finally send on a CCD camera. The object we put on the



Figure 5.7 – Setup for free space phase conjugation of an object.

path of the signal beam was a USAF target and we tried to generate the phase conjugate of the various structures of the target. Figure 5.8.(a) shows a picture of the USAF target and Fig. 5.8.(b) shows the images recorded by the CCD camera. The features of the target we imaged are annotated from A to D. We can see that the letters of the name "Edmund"



Figure 5.8 – Free space phase conjugation of a collimated wave travelling through an object. (a) Pictures of the USAF target used with features identified by letters A, B, C and D. (b) Pictures from the CCD camera of the identified features.

or the logo are quite true to the original. The same goes with the horizontal lines from the target. As the final goal is to use the *phase conjugation mirror* as a detection methods for acousto-optic imaging rather than as an imaging system, we did not try to measure the resolution of this system.

Phase conjugation through a diffuser The second step towards using phase conjugation for the detection of the acousto-optic signal is to use our setup to perform the phase conjugation of an aberrated wavefront. To do that, we focused the signal beam a few centimetres before a ground glass plate into a point source A (represented by a green star on Fig. 5.9). The light scattered by the plate is collected inside the crystal and, as



Figure 5.9 – Setup for the reconstruction of a point source through a diffuser by phase conjugation

in the previous paragraph, the phase conjugated light is separated from the initial beam by a beam-splitter. The CCD camera is equipped with a lens focused on the plane where A is. A diaphragm as been placed around point A in order to correctly focus the camera. Each arm (reference and signal) of our setup is equipped with acousto-optic modulators (not represented on Fig. 5.9) in order to add a frequency shift between the signal and the references. Figure 5.10.(a) shows a picture of the ground glass used as a diffuser. Figure 5.10.(b) shows the images recorded by the CCD camera with and without a frequency shift – of $0.2 \,\mathrm{MHz}$ – between the references and the signal beams. The large grey area



Figure 5.10 – Reconstruction of a point source through a diffuser by phase conjugation. (a) Picture of the diffuser. (b) Images acquired by the CCD with a lens focused on the plane of the point source with and without a 200 kHz frequency detuning.

5.2. Experimental realisation of an acousto-optic imaging setup by phase conjugation in $Nd: YVO_4$.

in the middle of the totally black image is the opening of the diaphragm we added to identify plane A. The bright spot in the center of the opening on the picture without detuning is the phase conjugated light. It disappears when the reference is detuned from the signal. Indeed, with a 0.2 MHz frequency shift between the references and the signal, the interference pattern oscillates at 200 kHz. The hologram recording dynamic inside the crystal is slower. Consequently, the hologram is not recorded by the crystal and the process of four-wave mixing can not take place anymore.

In order to get closer to our goal of imaging biological samples, the same experiment can be performed by replacing the ground glass by a 5 mm thick slice of chicken breast. The resulting images on the CCD camera are visible on Fig. 5.11. As for the experiment with the ground glass, a picture without detuning and a pictures with a 200 kHz detuning has been recorded. As the signal was weaker, the diaphragm has a very small aperture to be able to identify precisely the phase conjugate point. The brightness of the images has also been increased to be able to see clearly the point and the diaphragm limits. The





results are quite the same as the phase conjugation on a ground glass, the signal is just much lower due to a smaller light transmission through the chicken breast. In the end, the fact that we managed to detect a phase conjugate signal trough a relatively thick piece of biological tissue is encouraging.

These preliminary experiments were tests to get familiar with phase conjugation by fourwave mixing. Now that we have built a *phase conjugation mirror*, we will characterise some of its performances and use it in conjunction with ultrasounds to perform acoustooptic imaging of a scattering sample.

5.2.2 Optical setup and experimental conditions.

This section describes the optical setup and the experimental conditions we used in order to obtain the results presented in section 5.3.

Optical setup Figure 5.12 represents a schematic of the optical setup. The initial laser source is a continuous wave (CW) Nd:YAG laser injected in an Ytterbium doped fiber amplifier. The output of the amplifier is a 4.5 W collimated beam at 1064 nm. It passes through an optical isolator to avoid back reflection inside the amplifier. After the

isolator, the beam is split into two beams (reference and signal) by a polarising beamsplitter (PBS1). A half-wave plate (HW1) can be used to change the intensity ratio between the reference and the signal. Then each arm goes through a group {half-wave plate + polarising beam-splitter}, {HW2 + PBS3} for the signal path and {HW3 + PBS2} for the reference path; every beam intensity can thus be tuned independently. Then, each beam passes through an acousto-optic modulator (AOM1 for the reference path and AOM2 for the signal path) supplied with a 80 MHz signal and set up to have the maximum power diffracted in the +1 order. The reference beam is focused on the back side of the crystal by a f' = 30 mm lens (L2) so it is reflected on the HR coating on the back side to generate the second reference beam. The signal beam goes through a non-polarising beam-splitter (NPBS), which is used to separate the phase conjugate beam from the signal beam. It is then sent on the sample, the diffused light from the sample is collected by a lens (L1) and focused in the crystal. L1 has a focal length $f' = 5 \,\mathrm{mm}$, a 2" diameter, and works in a 2f configuration. The back propagating phase conjugate beam (dashed line on Fig. 5.12) is sent back on the sample to regenerate the original plane wave and separated from the initial signal wave by a non-polarising beam-splitter (NPBS) and collected on a Si-photodiode (Thorlabs PDA36A-EC).



Figure 5.12 – Optical setup for acousto-optic imaging by phase conjugation. HWi are Half-Wave plates, PBSi are Polarising Beam-Splitters, AOMi are Acousto-Optic Modulators, NPBS is a Non-Polarising Beam-Splitter, PZT is a Piezoelectric Transducer, PD is a PhotoDiode.

The crystal is held in a water-cooled copper mount, with an indium seal in order to have a good thermal contact between the crystal and the mount. It is pumped on the back side by a 808 nm laser diode (LIMO100-F200-DL808).

Pumping conditions and acquisition In most experiments, the pump is set to send 100 W long pulses. The duration of each pulse is 1 ms with a delay between each pulse of 59 ms. This quasi-continuous-wave regime (QCW) is used to prevent any thermal damage to the crystal despite the high pumping power. When a different regime is used, it will be indicated. The output of the pumping diode is a 200 µm multimode fiber. It is collimated and then focused inside the crystal by a lens with a focal length of f' = 18 mm.

The signal from the photodiode is digitized using an ADLINK PCI9646D data acquisition (DAQ) board inside the computer that can be driven directly *via* MATLAB. This DAQ device can acquire up to 40 MS/s encoded in 16 bit between two ranges: ± 1 V or ± 200 mV.

5.2.3 Setup characterisation.

Before performing the acousto-optic experiment, we measured the performances of the setup in terms of phase conjugation reflectivity, defined as the power of the phase conjugate beam P_{S^*} over the power of the initial signal beam P_S : $R = P_{S^*}/P_S$, and response time τ_R .

Phase conjugation reflectivity To measure the phase conjugation reflectivity of our setup, we used it on a plane wave. The setup from Fig. 5.12 was used without a sample. The power of the phase conjugate beam was calculated using the characteristics of the photodiode (sensitivity, gain) and taking into account the different elements that attenuate the signal (NPBS, filters,...). As for the input signal beam power, it was measured using a power meter.



Figure 5.13 – Reflectivity of the *phase conjugate mirror* vs. Intensity of the signal for 3 value of the intensity of the input reference beam.

The measurement of the reflectivity versus the power of the input signal beam was done for three different intensities of the reference beam. As we can see from Fig. 5.13, the setup has a reflectivity between 10% and 30% depending on the conditions. Another interesting observation is that the reflectivity seems to be independent of the power of the input signal beam, which was expected considering that the signal beam is not intense enough to saturate the amplifier (as a reminder, $I_{SAT} \approx 1 \,\mathrm{kW \, cm^{-2}}$).

Response time The second characterisation of our setup is its response time. It was measured by recording the evolution of the magnitude of the phase conjugated beam while the signal was detuned in frequency from the reference. This was done using the two AOMs in the setup described in Fig. 5.12, AOM1, on the reference path, stayed at 80 MHz and AOM2, on the signal path, swept the frequency range between 79.8 MHz and 80.2 MHz. We worked with plane waves, without any sample in the setup.

The baseline of each signal is corrected by subtracting the signal measured with a $\Delta f = -0.2$ MHz frequency detuning because it is large enough to cancel the phase conjugate signal. The magnitude of the phase conjugate is then calculated by integrating the signal during the whole pump pulse. Finally this experiment has been repeated for several intensities of the reference beam. The experimental data, with a Lorentzian fit, are shown in Fig. 5.14. The Lorentzian model has been chosen because we supposed that the crystal was acting as a first-order low-pass filter. This fit has been used to experimentally calculate the value of the response time τ_R of our setup.



Figure 5.14 – Response time of our phase conjugation setup for different intensity of the reference beam.

We can see that under the strong pumping conditions, $P_{pump} = 100$ W, and with saturation effects, the response time of our setup can be much shorter than the fluorescence lifetime of Nd:YVO₄ (90 µs). Indeed with an input reference intensity of 650 W cm⁻² we

have a reponse time as fast as $12 \,\mu\text{s}$. This really fast response time is much smaller than the speckle decorrelation time in living tissues (around $1 \,\text{ms} [37]$), which makes our setup much less sensitive to these problems in the scope of *in vivo* applications.

5.3 Acousto-optic imaging on a biological sample using phase conjugation

Now the acoustic component of the experiment is added to our setup in order to perform acousto-optic imaging on a biological sample.

5.3.1 Sample and ultrasonic conditions

Sample Our sample is a piece of chicken breast placed in a sample holder. It consists of two slabs of plexiglas spaced using chocks, and held together with tape. The space between the plexiglas slabs, and thus the thickness of the sample, is controlled using stacks of 1 mm-thick microscope slides as chocks. The top of the sample holder is opened to allow the ultrasound to go in the sample. Finally, the sample holder is placed inside a water tank to make the acoustic coupling.

Ultrasound In the water tank, above the sample holder, is placed an ultrasonic transducer. It is an Olympus A395S transducer which characteristics are given in table 5.2. The signal used for the imaging process consisted of 5 cycles of a sinusoid at f_{US} , giving

Characteristic (units)	Value
Central frequency f_{US} (MHz)	2.25
$-6 \mathrm{dB}$ Bandwidth (%)	40.7
Diameter (mm inches)	38.1 1.5
Focal length (mm)	76

Table 5.2 – Characteristics of the Olympus A395S transducer used on the phase conjugation setup.

an axial US resolution of dy = 3 mm and a transverse US resolution of dx = dz = 1.6 mmlimited by the transducer aperture. In order to be able to scan our sample, the horizontal position of the transducer is controlled by two high-precision motorized translation stages. The vertical position of the transducer is set so its focal point is at the same height as the incoming laser and can be adjusted *via* a manual mechanical stage.

5.3.2 Acousto-optic signal detection by optical phase conjugation

Before imaging a sample, we tried to detect an acousto-optic signal from our sample. We started by sending long US bursts consisting of 50 cycles to 100 cycles while the sample was illuminated by the signal beam. Figure 5.15 shows the normalized temporal signal
directly acquired on the photodiode with ultrasound (blue) and without ultrasound (red). This signal has been averaged 100 times in order to have a good SNR through the 5 mm



Figure 5.15 – Magnitude of the phase conjugate signal over time without ultrasound (red) and when a 70 cycles long ultrasonic burst travel in the sample (blue)

thick chicken breast sample. The ultrasonic burst lasted 70 cycles making the acoustooptic signal clearly visible but giving no axial US resolution. This first experiment shows that it is possible to detect the acousto-optic signal from a multiple scattering medium using phase conjugation. After that, we tried to increase the thickness of our sample. We managed to have an acousto-optic signal by phase conjugation through up to 8 mm of chicken breast as shown in Fig. 5.16 where part (a) shows the magnitude of the phase conjugate signal with (blue) and without (red) ultrasounds, and part (b) shows the difference between the two curves.



Figure 5.16 – (a) Magnitude of the phase conjugate signal over time without ultrasound (red) and when a 70 cycles long ultrasonic burst travel in the sample (8 mm thick slice of chicken breast) (blue). (b) Difference between the red and blue curves from (a).

The next step toward imaging, is to reduce the number of cycles to have a millimetric resolution and scan the sample along one axis.

5.3.3 Acousto-optic image

With this full setup, we performed a scan of a 5 mm thick piece of chicken breast without any inclusion inside. The purpose of this experiment was to image the diffuse light pattern inside the chicken breast tissues and get a 2D image. It was obtained by moving the transducer on the x-axis by step of 0.2 mm. The data were acquired on the photodiode at 40 MS/s by the DAQ device in the computer. Before being sampled, the signal was amplified by a FEMTO DHPVA and filtered by a low-pass filter with a 1 MHz cutoff frequency. Each line has been averaged a 100 times. The pump worked in the conditions described in section 5.2.2 except for the delay between each pulse, which was set to 19 ms (instead of 59 ms) to decrease the total acquisition time. The result of this scan is shown on figure 5.17, where the y-axis is converted into distance by the speed of ultrasound in biological tissues [80].



Figure 5.17 – Acousto-optic image of a 5 mm thick piece of chicken breast by phase conjugation.
(a) Whole normalized image smoothed by a 2D convolution.
(b) Time signal along the white dashed line on image (a).

Part (a), on the left, shows an image of each line concatenated, the image has been smoothed using a 2D convolution. Part (b), on the right, shows the time signal along the dashed line on part (a). The blue curve is the signal with ultrasound, whereas the red curve is the signal without the US. The diffused light pattern inside the sample is circular with a diameter of about 3 mm. This measurement is consistent with the theory of multiple scattered light, which states that at high depth $(z \gg l^*)$ the size of the diffused light spot is approximately equal to the depth. In our case, the sample is 5 mm thick and the imaging has been done approximatively in the middle meaning that the depth is between 2 mm and 3 mm.

5.4 Conclusions and perspectives.

Conclusion The topic of this chapter was the use of four-wave mixing in Nd:YVO₄ to perform optical phase conjugation and use it as a way to detect the acousto-optic signals for the purpose of imaging inside a multiple-scattering sample. First we demonstrated the possibility of doing optical phase conjugation in Nd:YVO₄ with speckle beams. Then, after a few characterisation of our *phase conjugate mirror*, we used it in conjunction with ultrasound in order to detect the travel of an ultrasonic burst inside a 5 mm-thick piece of chicken breast. This enabled us to map the local light intensity inside the chicken with a millimetric resolution. These first results are very encouraging and shows that phase conjugation by four-wave mixing in gain media such as Nd:YVO₄ is an attractive method for the detection of acousto-optic signals from multiple-scattering media.

We also showed that this method of holographic detection has a very fast response time. This fast response of the holographic processes in the gain media, between $10 \,\mu s$ to $100 \,\mu s$, is very interesting for *in vivo* experiments considering that in living samples, the speckle decorrelates in a millisecond time scale.

Perspectives Many improvements can be made on the setup in order to enhance the detection. One of the main problem is the square shaped signal carrying the acousto-optic signal that can be seen on Fig. 5.15. Indeed, this signal is the addition of the light from the amplified spontaneous emission (ASE) in the crystal and the phase conjugate (PC) level. As the level of the ASE plus the PC is high compared the variations of the PC because of the ultrasound, only a small portion of the DAQ device dynamic is used for the interesting signal - the variations of the PC. A solution for this problem is to use an adapted high pass filter between the photodiode and the DAQ card in order to bring the ASE plus PC level around 0V. This way the variations of the PC can be amplified much more. This has been implemented on the experiment described in chapter 6 and works quite well.

Another issue is the total acquisition time for one image. This is mainly due to two factors, the low SNR and the QCW regime. A lot of averaging is required to get a visible signal. The QCW regime imposes a delay of 20 ms between two consecutive acquisitions. As a result the imaging process is slow: around 1 min to have the image of an area of $4.2 \times 8 \text{ mm}^2$. One solution is to send several ultrasonic bursts within 1 pump pulse in order to average faster leading to a higher SNR and/or a faster acquisition rate. This has also been implemented for the experiments of chapter 6, where we sent 17 ultrasonic bursts during one pump pulse. Transferring those two improvements onto the phase conjugation setup would be quite interesting and can really enhance the signal.

Geometry Another way to improve the signal to noise ratio would be to study different geometrical configurations. One idea we had was to make every beam perpendicular to each other to prevent any parasitic light from the reference or the pump to go in the direction of the signal. The experiments did not work well for phase conjugation. However, they yielded interesting results for a two-wave mixing detection scheme. This lead us to implement the two-wave mixing detection presented in the next chapter.

Chapter 6

Acousto-optic detection by two-wave mixing in $\ensuremath{\mathsf{Nd}}\xspace{\mathsf{YVO}}_4$

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This second experimental chapter is dedicated to the experiments we performed in order to make a detection of the acousto-optic signal by two-wave mixing (TWM) in Nd:YVO₄. After the experiments on phase conjugation by four-wave mixing, we wanted to try a different geometry. The idea was, first to decrease the noise by making each wave (reference, signal and pump) perpendicular to each other, and second, to have a large area for the collection of the speckle light from the scattering sample. We looked into the solution presented in Fig. 6.1, where the pump is focused within the crystal by a cylindrical lens to have an active holographic volume with a large aperture, the two reference beams enter the crystal by the small sides and the large side is used to collect the speckle light. We rapidly realized, that adding an angle to each beam in order to



Figure 6.1 – Full orthogonal geometric configuration idea.

have a total internal reflection on the pumped side will give a much higher signal due to a better overlap of the beams. Unfortunately, despite this improvement, we still could not get a satisfying wave-mixing. Our hypothesis is that, first, the overlap between the signal beam and the other beams is not optimised and, second, the path travelled by the signal in the pumped volume is too short to lead to a high enough gain. This led us to give up the idea of using the large side of the crystal to collect the speckle light. As a result, we changed the arrangement of the beams around the crystal so that the reference and the signal enters by the same side at different angles. This new configuration will be presented in section 6.2.1.

The firsts tests we carried out were TWM experiments. As they seem to work well, it motivated us to try to use this detection for ultrasound optical tomography.

6.1 Two-wave mixing for metrology with speckle beams

As mentioned in 2.2.3, TWM in non-linear media can be used to detect the acoustooptic signal in a speckle pattern coming from a multiple scattering sample. This section describes how TWM in a generic non-linear medium can be used for such an application, then we particularly develop the case of gain media through the equations developed in section 4.1.

6.1.1 Principles of acousto-optic detection by two-wave mixing.

In the previous chapter, the problem of the low spatial coherence of the untagged photons speckle pattern was corrected using phase conjugation. Another way of correcting this low spatial coherence is to perform a wavefront adaption through TWM in a non linear media.

Wavefront adaption Each grain of the speckle pattern coming out of the sample carries the acousto-optic information. It is therefore possible to retrieve the information about the ultrasonic modulation by observing only one speckle grain. The problem is that the signal is very weak due to the small optical etendue of one speckle grain. Integrating over a lot of speckle grains on a single detector is not a solution because of the spatial incoherence of the speckle pattern. Indeed, the random phase of each speckle grain does not improve the SNR while integrating on a single detector [24]. A solution is to generate a phase matched wavefront that will interfere coherently with the wavefront of interest to phase each speckle grain, thus improving the SNR by a factor $\sqrt{N_{sp}}$ where N_{sp} is the number of speckle grains. For a pump volume that has a cross section around 300 µm, it corresponds to about 10⁴ speckle grains.

This wavefront adaption can be performed by two-wave mixing in a non-linear media such as a photorefractive crystal [81], or, in the case of this thesis work, a gain medium. Whatever the non-linear medium is, the principles of this technique can be summed up by the drawing in Fig. 6.2. The signal beam S, which carries the acousto-optic information,



Figure 6.2 – Two-wave mixing adaptive holography in a non-linear medium.

interferes inside the non-linear medium with a reference beam R. The resulting interference pattern is recorded inside the crystal as an hologram. Such recording is usually due to a non-linear behaviour of the crystal in which one optical property (refractive index, gain, absorption,...) of the crystal is modulated by the interference pattern. The reference beam is then diffracted on the hologram, generating a new wave with a wavefront identical to the signal beam's, which propagates in the same direction. The total electric field at the exit of our holographic medium is the coherent sum of those two waves. **Bragg condition** The interferences within the crystal create a volumetric hologram with a grating vector $\Delta \mathbf{K} = \mathbf{k}_S - \mathbf{k}_R$. Both the reference and the signal beams will diffract on this grating in only one direction with a wave vector that satisfies the Bragg's law:

$$\mathbf{k}_d = \mathbf{k}_i \pm \Delta \mathbf{K} \tag{6.1}$$

where \mathbf{k}_d and \mathbf{k}_i are respectively the diffracted beam wave vector and the incident beam wave vector. This leads to the diffraction of the reference in the direction of the signal and the diffraction of the signal in the direction of the reference as represented in Fig. 6.2.

$$\mathbf{k}_d^R = \mathbf{k}_R + \Delta \mathbf{K} = \mathbf{k}_S \tag{6.2}$$

$$\mathbf{k}_d^S = \mathbf{k}_S - \Delta \mathbf{K} = \mathbf{k}_R \tag{6.3}$$

Case of Nd: YVO_4 In the case of a gain medium such as Nd: YVO_4 , the output electric field is given by equation 4.24 obtained with the coupled wave approach developed in section 4.1.

$$E_S(x, y, L, t) = \exp\left(\frac{1}{2}\int_0^z \alpha^{(0)}(z')dz'\right) \times \left[E_S(x, y, 0, t) + E_S(x, y, 0, t) * C(L, t)\right]$$

It is convenient to understand the effect of the holographic process in the gain medium on the signal beam. We can see that the electric field is composed of two terms:

- The original signal $E_S(x, y, 0, t)$, which is the transmitted signal through the crystal and amplified.
- The diffracted signal $E_D(x, y, L, t) = E_S(x, y, 0, t) * C(L, t)$, which is the diffraction of the reference on the gain grating, also amplified.

The diffracted signal, $E_D(x, y, L, t)$, is the input signal, $E_S(x, y, 0, t)$, filtered by the frequency response of the crystal given by the function C(z, t) which expression in given by equation 4.25. Its spatial variations are identical to those of the transmitted signal beams: this is the wavefront adaption giving its name to this techniques so called adaptive gain holography.

6.1.2 State of the art

Interferometry is a powerful tool to make very accurate measurements. Unfortunately interferometric setups are very sensitive to low spatial coherence of light. The addition of a holographic medium in an interferometer to perform wavefront adaption is a solution to retrieve a high accuracy despite the low spatial coherence, even if the light is a speckle beam. Most of the time the holographic medium is a photorefractive crystal, but there are a few examples of setup that works with a gain medium.

Non-destructive testing In order to measure the displacement or the vibration of a reflective surface, interferometry is largely used because of its extremely high precision. Laser vibrometry is commonly used in non-contact and non-destructive testing in order

to probe the mechanical properties of materials. Problems arises when the tested surface is highly scattering. Indeed, if the reflected light has no specular component, the displacement information of the sample will be blurred in the speckle pattern. One way of compensating this issue is to integrate inside the interferometer a wavefront adaption device such as a photorefractive crystal. The wavefront adaption technique enables to retrieve a high accuracy in the displacement measurements even on rough surfaces that backscatter light. This method was first proposed by Huignard and Marrakchi [82] and is now employed in commercial interferometers which works at different wavelength using crystals of BSO (532 nm), GaAs (1064 nm) or CdTe (1.5 μ m) to perform the two-wave mixing¹.

Acousto-optic imaging Biomedical imaging can also benefit from these powerful wavefront adaption techniques. A few teams around the world have used photorefractive crystals in order to perform the detection of acousto-optic signals in order to create a new biomedical imaging modality. In 2004, both the teams from Murray *et al.* [83] and Ramaz *et al.* [84] proposed to use photorefractive holography to detect the acousto-optic signals from multiple scattering samples. The former used a crystal of BSO working at 532 nm, whereas the latter used gallium arsenide (GaAs) crystals working at 1064 nm. Several other teams followed, such as Wang *et al.* and Monchalin *et al.*, and used these materials to perform acousto-optic imaging of thick scattering samples [85, 86].

Since the first experiment on GaAs at Institut Langevin by Max Lesaffre [30], the quest for the right holographic detection method has not stopped. One of the issues of GaAs, as long as BSO, is their working wavelength which are respectively 1064 nm and 532 nm. As green light is very rapidly absorbed in biological tissue (mainly by blood), the acousto-optic detection using BSO is limited to very shallow depth. The wavelength 1064 nm is better but is still on the edge of the optical therapeutic window, where the absorption is still quite high ($\mu_a \approx 0.1 \text{ cm}^{-1}$). Ideally, the best working wavelength for *in vivo* application should be around 800 nm ($\mu_a \approx 0.01 \text{ cm}^{-1}$). This led to the PhD work of Salma Farahi followed by Emilie Benoit à La Guillaume whom both worked on a setup using a crystal of tellurium-doped tin thiohypodiphosphate (Te:SPS). This crystal has very interesting properties for acousto-optic imaging: a high photorefractive gain, a quite fast response time (several millisecond) and a very large sensitivity spectrum in the red and near-IR (650 nm - 1500 nm). It gave interesting results on thick phantoms [81] and even enabled the first *ex vivo* images of liver biopsy [4].

Despite all its advantages, Te:SPS may still encounter the problem of speckle decorrelation presented in 2.2.3 when the first *in vivo* experiments will be attempted. This leads to one of the goal of the thesis, exploring the possibility offered by gain media, such as Nd:YVO₄, which has a very fast holographic response time compared to the photorefractive crystals.

Gain media Despite the fact that wave mixing in gain media is a phenomenon that has been known for quite some time, it has not, as far as we know, been used by a lot

¹Two examples of companies manufacturing such systems: Bossa Nova Tech (http://www.bossanovatech.com/) and IOS. Inc (http://www.intopsys.com).

of people neither in industry, nor in academic research. The first experiments of wave mixing in a laser medium were carried out by Tomita [87], and were phase conjugation experiments. Two-wave mixing in gain media only appeared in 1993 with the experiment performed by Brignon *et al.* in Nd:YAG [88]. In 2005 Damzen *et al.* proposed to use a Nd:YVO₄ crystal to perform metrology with speckle beams [55] and showed that is was possible to detect a phase modulation through a rotating diffuser plate and suggests the possibility of using these mechanisms to measure vibrations of rough surfaces.

6.1.3 Two-wave mixing acousto-optic detection in Nd:YVO₄

The model from 4.1.6, and especially Eq. 4.24, is convenient to numerically compute the signal after the crystal and see the effect of a phase modulation which origin can be, e.g. an ultrasonic burst.

Detectable frequencies At first, let us consider the simple case of a phase modulated input wave:

$$E_S(x, y, 0, t) = A_S(x, y)e^{i\phi(t)}$$
(6.4)

Considering the simplified equation 4.27, the intensity of the light at the exit of the crystal can be written:

$$I_{S}(x, y, L, t) \propto |E_{S}(x, y, L, t)|^{2} \propto G|A_{S}(x, y)e^{i\phi(t)} + A_{S}(x, y)e^{i\phi(t)} * C(L, t) + i\beta A_{S}(x, y)e^{i\phi(t)} * C(L, t)|^{2} \propto GI_{S}(x, y)|e^{i\phi(t)} + e^{i\phi(t)} * C(L, t) + i\beta e^{i\phi(t)} * C(L, t)|^{2}$$
(6.5)

where G is the amplification factor of the crystal. Two cases can then be considered:

- 1. The frequency of modulation is smaller than the inverse of the response time: $f_{mod} \ll \frac{1}{\tau_R}$
- 2. The frequency of modulation is larger than the inverse of the response time: $f_{mod} \gg \frac{1}{\tau_R}$

In case (1), the crystal is able to adapt to the phase changes, it means that the diffracted components will not be filtered by the crystal and will carry the phase information. Therefore they can be written: $E_D(t) = m_1 e^{i\phi(t)}$, where m_1 is a complex constant. Equation 6.5 becomes:

$$I_S(x, y, L, t) \propto GI_S(x, y) |e^{i\phi(t)} + m_1 e^{i\phi(t)}|^2 \propto GI_S(x, y) |1 + m_1|^2$$
(6.6)

The output intensity does not depend on the phase modulation $\phi(t)$, which makes the system unable to detect it.

In case (2), the crystal cannot adapt to the phase changes due to the modulation. The diffracted components is filtered through the convolution product with the function C(z,t). We have $e^{i\phi(t)} * C(L,t) = \left\langle e^{i\phi(t)} \right\rangle_{\tau_B}$, it represents approximately the mean value

of $e^{i\phi(t)}$. When inserted into Eq. 6.5, this leads to:

$$I_S(x, y, L, t) \propto GI_S(x, y) |e^{i\phi(t)} + \left\langle e^{i\phi(t)} \right\rangle_{\tau_R} + i\beta \left\langle e^{i\phi(t)} \right\rangle_{\tau_R} |^2$$
(6.7)

This time, the output intensity has a dependency in $\phi(t)$, meaning that the system is sensitive to phase modulations of the signal.

To sum up, adaptive holographic systems are able to detect phase modulated signal provided that the frequency of modulation is high enough so that it cannot be followed by the crystal.

Small signal detection Moreover, in the case of a small amplitude phase modulation, the intensity can be simplified:

$$I_{S}(x, y, L, t) \propto GI_{S}(x, y)|1 + i\phi(t) + (1 + i\beta) \left\langle e^{i\phi(t)} \right\rangle_{\tau_{R}}|^{2}$$

$$\propto GI_{S}(x, y) \left[\left(1 + \left\langle e^{i\phi(t)} \right\rangle_{\tau_{R}} \right)^{2} + \beta^{2} \left\langle e^{i\phi(t)} \right\rangle_{\tau_{R}}^{2} + 2\beta \left\langle e^{i\phi(t)} \right\rangle_{\tau_{R}} \phi + \phi^{2} \right]$$
(6.8)

This last equation shows that the variations of the intensity have two components. One is proportional to $\phi(t)$ and the other is proportional to $\phi(t)^2$. Therefore, this setup has, a linear and a quadratic sensitivity of phase modulated signals. At small amplitude of modulation, the linear component is easier to detect because it is much stronger than the quadratic one.

Computed AO signal As an example, let us consider that the US phase modulation is a short signal, a few cycles of a sinusoid, with a Gaussian envelope that can be written as follows:

$$\phi(t) = \phi_0 e^{-\left(\frac{t - t_c}{0.5 * N_c/f_{US}}\right)^2} rect_{N_c/f_{US}}(t - t_c)sin(\omega_{US}t)$$
(6.9)

With ϕ_0 being the amplitude of the phase modulation, N_c the number of cycles of the burst, f_{US} and ω_{US} respectively the frequency and the angular frequency of the modulation, t_c the time of the center of the US burst ($t_c = t_0 + N_c/2f_{US}$, where t_0 is the beginning of the burst). Finally $rect_{N_c/f_{US}}(t)$ is the rectangular function, its value is 0 everywhere except on the interval $\left[-\frac{N_c}{2f_{US}}, \frac{N_c}{2f_{US}}\right]$ where it is equal to 1.

For the simulation, we suppose that we have a plane wave so $A_S(x, y, 0) = A_S$ with a small amplitude: $I_S = |A_S|^2 \ll I_R, I_{SAT}$. In order to simulate the ultrasonic burst, we chose the following values for the parameters:

- Number of cycles: $N_c = 5$.
- Phase modulation frequency: $f_{US} = 5 \text{ MHz}$.
- Phase modulation amplitude: $\phi_0 = \pi/10$.
- $t_0 = 100 \, \mu s.$

The shape of this phase modulation is represented in Fig. 6.3.(a). The modulated field can be used as an input signal in the computing script. The computed signal exiting the crystal is represented in Fig 6.3.(b).



Figure 6.3 – (a) Input phase modulation of the signal beam. (b) Computed response of the crystal to a short phase modulation.

It shows both the linear and the quadratic contribution. The linear contribution at f_{US} can be seen in the main oscillations of the signal, whereas the quadratic contribution at $2f_{US}$ causes the asymmetry regarding the x-axis.

6.2 Experimental realisation of an acousto-imaging detection by two-wave mixing in Nd:YVO₄

This section describe our experimental investigations towards the realisation of an acoustooptic imaging system by two-wave mixing in Nd:YVO₄. Before presenting the new setup and a few characterisations, we will detail the new geometric configuration introduced at the beginning of this chapter and the characteristics of the crystal.

6.2.1 Description of the new configuration

The two-wave mixing experiments were performed using a configuration implemented by J.E. Bernard *et al.* for high gain amplifiers [89] and used by M.J. Damzen for metrology using speckle beams [55].

Geometric configuration In this configuration, the reference and signal beams enter the crystal on one of the lateral sides with an incidence angle such as it is totally reflected on the top side and exit through the opposite lateral side. The light from the pump enters from the top side so that there is a good overlap between the interference pattern and the pumped volume as shown in Fig. 6.4. The pump is the same diode as the one used



Figure 6.4 – Geometric configuration of the signal, reference and pump beams on a Nd:YVO₄ cyrstal for two-wave mixing.

in chapter 5 but now it is focused inside the crystal using a cylindrical lens in order to have a large pumped volume. The cylindrical lens has a focal length of 18 mm so the pumping volume is about $0.3 \times 0.3 \times 6 \text{ mm}^3$. The reference and signal beams have angles of incidence of respectively $i_{0R} = 10^\circ$ and $i_{0S} = 20^\circ$, which results in incidence angle on the top side inside the crystal of 85° for the reference and 80° for the signal.

Remark : The refractive index of the vanadate is around 2, so the critical angle is approximately 30° .

Characteristic of the crystal This setup uses a $5 \times 5 \times 10 \text{ mm}^3$ crystal of Nd:YVO₄ with the same doping as the one used in chapter 5 (1%-at with Nd³⁺ ions). Therefore its optical characteristics are the same and can be found in Tab. 5.1. In addition, the crystal has AR coatings for 1064 nm on its lateral sides and an AR coating for 808 nm on the top side, the three other sides are not polished and are in contact of an aluminium mount through an indium seal in order to dissipate the heat generated by the pumping. A picture of the crystal in its mount is shown in Fig. 6.5



Figure 6.5 – Photo of the crystal in its aluminium mount cooled by a Peltier module.

Full optical setup This crystal is placed in an optical setup that is similar to the one used for the phase conjugation experiments. This new setup is represented in Fig. 6.6. The first part of the setup is the same as the one used for phase conjugation. Combinations



Figure 6.6 – Optical setup for acousto-optic imaging by TWM. HWi are Half-Wave plates, PBSi are Polarising Beam-Splitters, AOMi are Acousto-Optic Modulators, PZT is a Piezoelectric Transducer, PD is a PhotoDiode.

of polarising beam-splitters and half-wave plates are again used to set the flux and the polarisation of each beam. The main change is in the crystal configuration. As described earlier, in this new setup, the beams enter the crystal by one of the lateral side, undergo a total reflection on the pumped side and finally exit the crystal through the opposite lateral side. The light from the sample is collected by an optical system (OS1) to image the exit plan of the sample inside the crystal. After the crystal, light is collected by an another optical system (OS2) that images the holographic volume on the detector (Thorlabs DET10A photodiode). As in 5.2.2, the signal from the photodiode is amplified by a large bandwidth amplifier (FEMTO DHPVA) and sampled at up to 40 MHz by the ADLINK PCI9646D DAQ device.

Pumping conditions In this setup the pump works at a power of 80 W and sends 2.5 ms long pulses at a repetition rate of 40 Hz. We chose to change the repetition rate and the length of the pulses in order to increase the acquisition rate (this will be detailed in 6.3.1). The increase of the duty cycle is compensated by a lower peak power of each pulses (80 W instead of 100 W) and a larger focusing area (the light is focused only in one dimension by a cylindrical lens).

6.2.2 A few characterisation of the setup

Gain One of the first characterisation of the setup that can be done is the measurement of the light amplification with the crystal in this configuration. The gain of our setup was measured on the signal path without any scattering sample. The signal beam was focused inside the crystal with a lens of focal length f' = 15 cm, the amplified light was then collected on the photodiode. The gain was obtained by measuring the intensity of the signal with and without a pump pulse. The gain factor as a function of the input intensity is plotted in Fig. 6.7. The experimental data (red dots) have been fitted by a model that



Figure 6.7 – Gain versus the intensity of the signal beam with a pumping power of $80 \, \mathrm{W}$.

calculates the propagation of light in an amplifier taking into account saturation effect (blue solid line). This model led to an intensity of saturation of $I_{sat} = 1.3 \,\mathrm{kW} \,\mathrm{cm}^{-2}$ and a small signal gain per unit of length of $\alpha_0 = 5.6 \,\mathrm{cm}^{-1}$. This smaller value of the gain per unit of length compared to the value found in chapter 5 can be explained by the difference in the pumping conditions. Here, the pump beam is focused using a cylindrical lens instead of a standard one, resulting in a less intense pump in the crystal. Nevertheless the amplification is still high since the amplification factor reaches 150 without saturation.

Step response In section 4.1.5, we used a model to calculate the temporal evolution of the signal beam when it is turned "ON" and see the effect of several parameters, especially the ratio I_{R0}/I_{SAT} . Looking at the step response is convenient because it is easy to realise experimentally, indeed, the AOMs on the setup (see Fig. 6.6) can be used as fast shutters to create the step signal. The measured step response can then be compared to the theoretical one. The theoretical model requires 3 input parameters to calculate the signal: the unsaturated small signal gain by unit length α_0 , the ratio between the incoming reference and the saturation intensities I_{R0}/I_{SAT} , and the response time τ_R . The first two parameters are estimated using the experimental data, whereas the response time τ_R is entered manually and adjusted so the computed data best fit the experimental ones. The estimation of α_0 and I_{R0}/I_{SAT} is done as follows:

• α_0 is estimated by looking at the signal amplification in the absence of a reference beam: $\alpha_0 = \frac{1}{L} ln(G)$, where G is the ratio between the signal beam measured with

and without the pump.

• I_{R0}/I_{SAT} is estimated by measuring the power of the reference beam and calculating the waist of the reference inside the crystal.

Figure 6.8 shows the experimental data (dots) and the raw calculated gain (dashed line) and a corrected calculated gain (solid line) over time for several values of the ratio I_{R0}/I_{SAT}^2 . In order to have a good agreement between the calculated and the experimental gain, several corrections had to be made. First, the estimated small signal gain α_0 has been replaced by an effective gain equal to $0.72\alpha_0$ (determined empirically, see remark below). Then, an offset remained between the theoretical data and the experimental data. This offset has been corrected on the theoretical data in order to best fit the experiment.



Figure 6.8 – Experimental (dots), calculated (dashed lines) and corrected (solid lines) gain of a small signal beam in the presence of a reference beam for several values the the ratio between the reference intensity and the saturation intensity.

²This figure is Fig. 4.2 where we have added the experimental data and a correction

Remark : The corrections made on the computed data can be justified by the difference between the experiment and the model. Indeed, our model supposes three strong conditions: first, a perfect overlap between the reference beam, the signal beam and the pump, second, a spatially uniform gain in the pumped volume and third uniform in intensity plane waves. As described in section 6.2.1, the signal and reference beams enter the crystal with respective angles of 20° and 10° . Moreover the gain is not spatially uniform because the pump is strongly absorbed by the crystal, and finally neither the signal, nor the reference are plane waves with a uniform intensity, they are focused gaussian waves. Nevertheless, after the introduction of the corrections, the model shows a good agreement with the experimental data.

In addition of confirming the model developed in chapter 2, this experiment/model comparison gives us an idea of the response time of our setup. Indeed, as we had to enter manually the value of τ_R , we had to compute the theoretical data for several values. A good agreement was found for values of τ_R between 40 µs and 50 µs, which will be confirmed in the next paragraph.

Response Time As we did in chapter 5, we will characterise this configuration in term of response time. This can be done experimentally using a scattering phantom and ultrasound with a method proposed by Lesafre et al. in 2007 in [90]. It allows to directly measure in situ the response time of a self-adaptive-wavefront holography setup by sending US pulses at a high frequency rate on a scattering sample. We used the method explained in part 4, we sent on our sample an amplitude modulated ultrasonic excitation and recorded the temporal signal at the exit of the holographic system for several frequency shifts between the signal and the reference beams. Then, the amplitude of the signal at the modulation frequency is retrieved by detecting the peak on the Fourier spectrum. The method requires the modulation frequency to be large compared to the inverse of the response time τ_R , as we know that it is between 40 µs and 50 µs (which gives $1/\pi\tau_R$ between 6 kHz and 8 kHz), we chose to use a modulation frequency of $f_{mod} = 200$ kHz. The US are generated by a 25.4 mm diameter Olympus focused single element transducer (model A307S) with a focal length of 51.5 mm and a center frequency $f_{US} = 4.8 \text{ MHz}$. Figure 6.9 shows the amplitude of the peak at f_{mod} versus the frequency shift (Δf) between the signal and the reference beams. In order to estimate τ_R , the experimental points have been fitted by a Lorentzian model: $A(\Delta f) = \frac{A_0}{1+(2\pi\Delta f\tau_R)^2}$. This fit gives a value of $\tau_R = 44 \,\mu\text{s} \pm 6 \,\mu\text{s}$ with a coefficient of determination $r^2 = 0.91$. This result confirms the previous observations with the model that led to a response between $40 \,\mu\text{s}$ and $50 \,\mu\text{s}$.

6.3 Acousto-optic imaging on a scattering phantom using TWM

This last section details the experiments that led to the imaging of 3 black inclusions inside a scattering phantom. First, we explain the main characteristics of the sample



Figure 6.9 – Amplitude of the component of the acousto-optic signal at $f_{mod} = 200 \text{ kHz}$ vs. the frequency shift between the signal and the reference beam (dots) and lorentzian fit (solid line).

used and the specifities of the US excitation and the data acquisition. Then, we show the first result of a 1D profile and a 2D acousto-optic image obtained by TWM in Nd:YVO₄.

6.3.1 Samples, ultrasound and acquisition conditions

Samples The samples used for the two-wave mixing experiments were phantoms made of agar containing Zinc Oxyde (ZnO) powder to increase their scattering coefficient. The thickness of the samples was 1.5 cm with a reduced scattering coefficient around $\mu'_s = 3 \text{ cm}^{-1}$. Depending on the experiments we performed, the gel contained black inclusions of various shapes and sizes (lines, circles). They were made using agar and India ink. Then, the samples were placed inside a water tank under the same ultrasonic transducer as the one used in 6.2.2 for the experimental response time measurements. The transducer is aligned on the x and y axis so that its focal spot coincide with the incoming laser. In order to be able to map the sample, the water tank is placed on two PI-Micos Pollux Drive translation stages (not represented in Fig. 6.6), that moves in x and y directions.

Ultrasound and data acquisition As previously mentioned, the ultrasonic transducer used for these experiments is an Olympus A307S. Its characteristics are given in Tab. 6.3. The signal sent to the transducer was generated by a standard arbitrary function generator

Characteristic (units)	Value
Central frequency f_{US} (MHz)	4.8
$-6 \mathrm{dB}$ Bandwidth (%)	42.3
Diameter $(mm \mid inches)$	$25.4 \mid 1$
Focal length (mm)	51.5

Table 6.3 – Characteristics of the Olympus A307S transducer used in the TWM setup.

and amplified by a 25 W RF amplifier. It consisted of short sinusoidal bursts (a few cycles)

at $f_{US} = 4.8$ MHz. The transverse US resolution is given by the size of the transducer focal spot which is around 770 µm at 4.8 MHz in water, the axial resolution depends on the number of cycles used which will be indicated for each experiment.

As explained in section 5.4, several improvements on the ultrasonic excitation and the data acquisition have been implemented. Indeed, in order to exploit the whole dynamic of the data acquisition (DAQ) device for the useful signal, we first used a Dual Low-Pass Programmable Butterworth Filter (KROHN-HITE 3945) after the photodiode amplifier. It was set to work as a high-pass filter (2 kHz cutoff frequency) and a low-pass filter (2 MHz cutoff frequency). The signal was then amplified a second time with another FEMTO DHPVA voltage amplifier before being sampled by the computer. With a 2.5 ms pump pulse, it is possible to send several ultrasonic bursts during one pump pulse, meaning that the averaging process can be made a lot faster. As the signal is high-pass filtered, the artefact generated make the first 500 µs unusable to send ultrasound. That leaves a 2 ms interval during which we can sent the US burst and acquire the data. We used a pulse generator, triggered by the pump, to trigger the ultrasonic excitation and the DAQ device every 100 µs making the averaging of the signal 19 times faster than if we had sent only one US burst per pump pulse! A time diagram can be found in Fig. 6.10. The 30 µs delay between the US burst and the DAQ trigger corresponds to a little less than the time required for the US to travel from the transducer to the focal spot.



Figure 6.10 – Time diagram of the pump, ultrasound and DAQ triggers for high-speed averaging.

6.3.2 One dimension imaging of a phantom

The first step was to perform a one dimensional profile using long US bursts and by scanning the sample along the x axis. The long US bursts gives a higher signal, but no resolution in the US propagation direction (y axis). The acousto-optic signal acquired versus time in a scattering phantom using long US bursts (10 cycles) is represented in



Figure 6.11 – Acousto-optic signal vs. time in a two-wave mixing configuration.

figure 6.11. This signal does not look like the computed signal presented in Fig. 6.3 because it has been electronically filtered to remove the oscillation. As the filtering is not perfect, we can see that some oscillation at the ultrasound frequency remains. The first "imaging" experiment consisted in acquiring these signals on a gel with 2 black inclusions embedded inside. The inclusions were 2 long lines (1 cm) with respective thickness of 1 mm and 2 mm and spaced by 10 mm. A drawing of these inclusions is given in Fig. 6.12.(a). As the inclusions are only one dimensional, we used long US bursts (10 cycles), and the sample was scanned along a 2 cm interval with a 0.1 mm pitch. The amplitude of the acousto-optic signal, in arbitrary unit, is represented on Fig. 6.12.(b). The measurement of the thicknesses, full width at half maximum (FWHM), of the inclusions, as long as the space between them, gives 1.5 mm and 2.1 mm and a gap of 11.7 mm. These values are in agreement with the expected results. The inclusions are a bit larger, especially the small one, probably due to the limited resolution, 0.6 mm, and the uncertainty on their real size. Indeed, firstly, the instrument used to create the hole for the inclusion is not perfect and secondly, it happens that the ink diffuse inside the gel making it larger.

6.3.3 From 1D to 2D.

To go from a one dimensional scan to a two dimensional image, the step is not very high. First we can shorten the ultrasonic burst in order to have a better resolution along the y direction, and secondly we can scan the sample also in the y direction in order to map a large field.

2D scanning method In the previous chapter, the image inside the chicken breast was performed by scanning the ultrasonic transducer above the sample. Most of the time, the acousto-optic images are obtained using this method. In our experiments, we decided to scan the sample, because it has 2 major advantages:

- The field of view is not limited to the diffused light pattern inside the sample because the sample moves also relative to the incoming laser.
- The background light is more homogeneous. Indeed, usually, in the acousto-optic 2D images, the background light around the inclusions has a Gaussian envelop because



Figure 6.12 – (a)Drawing of the inclusions embedded in the gel used for a 1 dimensional scan. (b)Result of the acquisition of the acousto-optic signal along 1 dimension. The sample scanned was a 1.5 cm thick phantom of Agar + ZnO ($\mu'_s = 3 \text{ cm}^{-1}$).

the US transducer moves relative to the laser. When the sample is moved, the transducer and the laser are always aligned, making the background homogeneous.

One of the drawbacks of scanning the sample, is that the acquired lines cannot just be concatenated, the overlap has to be taken into account, especially if the translation pitch is smaller than the US focal spot. Despite the additional calculations required to reconstruct an image, a good point, is that a pixel that is at the intersection of two or more acquisition is more averaged. As an example, Fig. 6.13 shows what happens in that case. If each red ellipse represents the imaged area (the shape is given by the ultrasonic focal spot) for one position of the sample, the value of the acousto-optic signal in the pixel indicated in blue is acquired for 2 vertical positions of the sample. As a result, if each of the positions is averaged N_{avg} time, the blue pixel will be averaged N_{avg} for each vertical position thus $2N_{avg}$ in this case.

Sample The sample manufactured for this experiment contains 3 circular shaped inclusions with respective diameters of 1 mm, 2 mm and 3 mm and spaced by 6 mm (distance between the centers). A picture of the inclusions embedded inside the gel is represented on Fig. 6.14, the picture has been taken before covering the sample with another layer of scattering gel. The total thickness of the phantom is 1.5 cm with a reduced scattering coefficient of $l^* = 3.5 \text{ cm}^{-1}$.



Figure 6.13 – Effect of the overlapping of the US focal spot when the sample moves along the y axis.



Figure 6.14 – Photo of the inclusions inside the phantom before being covered with a layer of scattering gel.

Imaging To perfom the image presented in Fig. 6.15 [54], we moved the sample both on the x axis and y axis. The travel pitches on each axis were respectively 0.1 mm for the x axis and 0.75 mm for the y axis. Each position of the sample was the result of the averaging of 3000 signals giving a total acquisition time around 3 h. On Fig. 6.15.(a) is represented the picture of the gel superimposed with the acousto-optic image, whereas 6.15.(b) represent the acousto-optic image with a profile along the black dashed line. The profile can be used to measure the size of the inclusions and the spacing between them. The three inclusions inside the gel are clearly visible on the acousto-optic image. Their respective FWHMs measured on the profile are in a good agreement with the reality since they are 0.9 mm, 1.9 mm and 2.7 mm and the distance between them is 6 mm.

6.4 Conclusions and perspectives

Conclusions In this chapter we presented our experimental investigations on the use of two-wave mixing in Nd:YVO₄ for acousto-optic imaging of multiple scattering media. First we introduced the principles behind the detection of phase modulated signal using two-wave mixing in gain media. We relied on the model developed in chapter 4 to indicate the specificities of such detection. Then, after describing our setup and the geometrical configuration of the beams around the crystal, we characterised its step response to have an estimation of the response time, which was confirmed by a second method of measure-



Figure 6.15 – (a)Photo of the inclusions superimposed with the acousto-optic imaged obtained by TWM. (b)Acousto-optic image obtained by TWM and profile along the back dashed vertical line.

ment. Finally, the setup was used to detect absorbing inclusions inside multiple scattering phantoms. We did a 1D profile to detect two line-shaped inclusions of millimetric size, and a full image of three circular inclusions, and showed that it was possible to detect inclusions as small as 1 mm in a 1.5 cm thick gel of agar with a scattering coefficient of $\mu'_s = 3 \text{ cm}^{-1}$.

These results show that it is possible to use two-wave mixing in gain media to detect acousto-optic signals from multiple scattering sample. Moreover, the short response time offered by vanadate crystals, around 45 µs, is very interesting in the scope of *in vivo* application such as biological imaging.

Perspectives Using two-wave mixing in a gain medium to detect acousto-optic signal has a lot of interests, especially the response time of the crystal. However, it is possible that the geometric configuration used in this chapter is not the most efficient one. Compared to the previous configuration (the one used in chapter 5 for phase conjugation), the gain is much less, and a good overlapping of the beams seems harder to achieve experimentally. A solution could be to return to the former configuration and remove the HR coating on the back of the crystal to allow the signal beam to exit the crystal by the backside. A dichroic mirror could be placed right at the exit of the crystal to separate the signal beam at 1064 nm from the pumping at 808 nm. Such a configuration has been used in the experiment on photoacoustic detection presented in the next chapter, unfortunately, we did not have the time to try it for acousto-optic detection.

chapter 7

Photoacoustic imaging by speckle vibrometry

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This final chapter describes the last project of this thesis. It consisted in using an adaptive gain interferometer, based on a Nd:YVO₄ crystal, to perform the detection of photoacoustic signals. As explained in chapter 2, photoacoustic imaging is one of the two imaging techniques that use light and ultrasound in order to detect absorbers inside a multiple scattering medium. It relies on the ultrasonic wave emitted by an optically absorbing inclusion when hit by a short laser pulse. The position and shape of the absorber can be retrieved by detecting the emitted ultrasonic field. Several methods for optical detection of photoacoustic signals on a multiple scattering sample exist. For example Beard *et al.* use a small Fabry-Perot interferometer to probe the local vibration of the sample [40]. Another solution consists in using an adaptive interferometer to correct the low spatial coherence of the speckle. This method has been investigated by Hochreiner *et al.* [44]. They worked with a probe beam at 532 nm and a photoacoustic excitation Q-Switch laser at 1064 nm. They used a BSO photorefractive crystal to perform the TWM necessary for the wavefront adaption. The work described in this chapter has been inspired by these experiments.

After exposing our motivations for an optical detection of photoacoustic signals, we present the optical setup we used to perform this kind of detection. Our setup was tested with preliminary experiments showing that it was able to detect the vibrations of a piece of Silicon wafer and of a piece of paper. The second part of this chapter is dedicated to the detection of ultrasound inside a scattering gel made of Agar and Intralipid. We first show that it is possible to detect ultrasounds generated by a transducer inside the gel, and in a second time, we detect the ultrasound generated by photoacoustic in order to localise an absorber.

7.1 Motivation for optical detection of photoacoustic signals.

As explained in 2.3.2, photoacoustic signals are usually detected using either a single element piezoelectric transducer or an array of piezoelectric transducers. Piezoelectric detection of acoustic wave is very efficient and has given numerous interesting results over the past few years. It can be used to perform photoacoustic tomography and microscopy [1,91,92]. But also imaging of the vascular network in tumours [93] and brain [94], imaging of nanoparticles [95] and temperature mapping [96].

For some applications, it can be interesting to use a full optical detection of ultrasound. For instance, it may be impossible to apply the coupling medium on the sample; either because it is not directly accessible or because it would be damaged by the coupling medium. In these cases, optical remote sensing is an attractive solution because it can measure the vibrations of a surface through the air or even through vacuum (although biological imaging in vacuum is probably very rare!). Moreover, another advantage of optical detection is the very large bandwidth compared to a standard ultrasonic transducer. This is interesting because the ultrasonic frequency emitted by an optical absorber depends on its size. As a result, having a broadband probe enables to access various size scales with one probe. Usually, the maximum detectable frequency is limited by the photodetector rise time, which can be very fast (several nanosecond for photodiodes), leading to bandwidth of several hundreds of megahertz.

7.2 Adaptive gain laser vibrometry on rough surfaces using $Nd:YVO_4$.

In this section we will present our adaptive gain interferometer as well as two preliminary experiments in which we measured the vibrations of a reflective surface (a silicon wafer) and a scattering surface (a piece of standard white paper for printer).

7.2.1 Adaptive gain interferometer

In order to perform the two-wave mixing for the gain adaption we used a geometric configuration similar to the one used in chapter 5 for phase conjugation.

Geometric configuration We came back to a colinear configuration where the reference and the signal beams propagate along almost the same axis as the pump. As shown in Fig. 7.1, a small angle (5° to 10°) is introduced between the pump and the signal and between the pump and the reference in order to be able to separate all the beams. Most notably, this geometric configuration allowed us to separate the noise generated by ASE and the lasing effect between the crystal sides.



Figure 7.1 – Beam configurations in the crystal for the photoacoustic experiments. AR means "Anti-Reflection coating"

The crystal has no HR coating for 1064 nm on its backside as shown in Fig. 7.1. Instead, the signal beam is transmitted through the crystal and separated from the pump with a dichroic mirror. The other characteristics of the crystal are the same as the one of the crystal used in chapter 5. It is a $5 \times 5 \times 5 \text{ mm}^3$ YVO₄ crystal doped at 1%-at with Nd³⁺ ions so its optical characteristics are the same as the one given in Tab. 5.1. It is placed

inside a water-cooled copper mount to absorb the heat generated by the pumping, which can be seen in Fig. 7.2



Figure 7.2 – Photo of the crystal in its copper mount with the $808 \,\mathrm{nm}$ pump.

Optical setup The first elements of this optical setup are the same as what was presented in the two previous chapters (see Fig. 7.3). We use combinations of polarising



Figure 7.3 – Optical setup for detection of PA signals by TWM. HWi are Half-Wave plate, PBSi are Polarising Beam-Splitter, AOMi are Acousto-Optic Modulators, PZT is a Piezoelectric Transducer, PD is a PhotoDiode.

beam-splitters (PBS) and half-wave plates (HWi) to change the intensity and polarisation of each beam and AOMs to add a frequency shift between the beams or to switch "ON" or "OFF" each beam. The reference beam is sent inside the crystal and focused using a f' = 40 cm lens (L4). The signal beam is focused on the sample using a f' = 6 cm lens (L1). The back scattered light from the sample is collected by L1 and separated from the incoming light using a polarising beam-splitter (PBS4) and a quarter-wave plate (QW). The collimated backscattered light is then focused in the crystal by a $f' = 20 \,\mathrm{cm}$ lens (L2). Finally, light is separated from the pump by a dichroic mirror (DM) and sent on a photodiode via a f' = 5 cm lens (L3). The processing and recording of the signals is done the same way as in chapter 6. The Thorlabs DET10A photodiode is first amplified using a Femto HCA current amplifier, the signal is then filtered using a KHRON-HITE 3945 Dual Low-Pass Programmable Butterworth Filter, and finally the signal is amplified one last time in a FEMTO DHPVA voltage amplifier. The DAQ device also remains the same (ADLINK PCI9646D). To perform the photoacoustic experiment in section 7.3.2, the sample will be illuminated by a frequency-doubled Nd:YAG laser that sends 4 ns pulses at 532 nm (more information about this laser is given in Tab. 7.1 in section 7.3.2).

Pumping conditions Because the final goal of this setup is to perform photoacoustic imaging, we have to adapt the pumping regime to the photoacoustic excitation regime. The laser used for the photoacoustic experiment is limited to a repetition rate of 10 Hz, so the QCW regime used for the pumping is set to send 1 pulse every 100 ms, each pulse lasting 1 ms. The peak power of each pulse is 80 W, except specified otherwise.

Single-pass gain One of the first characteristic of this setup that can be measured is the gain relative to the incoming signal intensity. This can be done by placing a mirror at the position of the sample on Fig. 7.3 and turning OFF the reference. The amplified signal beam is recorded on the photodiode. The gain is obtained by recording and dividing



Figure 7.4 – Single-pass gain versus the intensity of the signal beam with a pumping power of $80 \,\mathrm{W}$

the signal with and without the pump pulse for several values of the power of the input light. Figure 7.4, shows the measured gain (red dots) as long as a fitted curve (blue solid line). It has been obtained using a simple model of the propagation of a single coherent beam in an amplifier taking into account saturation effects. The fit leads to a saturation intensity of $I_{sat} = 1.3 \text{ kW cm}^{-2}$ and a small signal gain per unit length of $\alpha_0 = 15 \text{ cm}^{-1}$. Again, as in chapter 5, we can see that this configuration, with the counter-propagating pump, is very efficient in terms of single-pass gain. The value of α_0 is higher than in chapter 5, we believe it is due to the focusing of the amplified light inside the crystal that creates a better overlap with the gain volume.

7.2.2 Preliminary experiment: Detecting vibration of a Silicon wafer.

The first experiment we carried out was the detection of the vibration of a Silicon wafer. Instead of the sample on Fig. 7.3, we put a small piece of reflective silicon glued onto a piezoelectric actuator in order to make it vibrate at 40 kHz. This experiment had two goals: first check that our vibrometer was working, and second, highlight the refractive index grating contribution in the two-wave mixing process.

Refractive index grating and linear detection As was mentioned in 4.1.6, the gain modulation comes with a refractive index modulation. Thus, the reference beam diffract on two gratings, and the diffraction on a refractive index grating causes a $\pi/2$ phase shift as shown by Eq. 4.27.

This $\pi/2$ phase shift is the origin of the linear sensitivity of our system, as we showed in section 6.1.3. The detection of photoacoustic signal requires to measure the displacement of the surface due to the ultrasound. Such information can be measured only if our system has a linear sensitivity on the phase since the phase shift is proportional to the displacement. Two experiments were performed to validate this idea of vibration measurement using a gain media with a detuned laser line: the detection of the vibrations of a silicon wafer and a blank sheet of white paper.

Vibrating mirror Our vibrating mirror is a small piece of silicon wafer glued onto a piezoelectric actuator. The actuator is given for a displacement of 2.2 µm for 100 V and a resonance frequency of 300 kHz without any load. A picture of this mirror is represented in Fig. 7.5.(a). The actuator is excited using a sinusoidal voltage at $f_{mirr} = 40$ kHz with a peak to peak amplitude varying from 0 V to 10 V. The temporal signals for several amplitudes of vibration are recorded, then the amplitude of the peaks at f_{mirr} and $2f_{mirr}$ for each value of the amplitude of vibrations are retrieved using Fourier transform. The experimental points have been fitted using Bessel functions in Fig. 7.5.(b) and with our theoretical model (from 4.1.6) in Fig. 7.5.(c).

The model using Bessel functions returned a value of k (proportionality coefficient between the phase shift ϕ and the voltage on the piezoelectric actuator V_{mirr}) of 0.1 rad V⁻¹. This value is linked to the piezoelectric coefficient r of the actuator by $k = 4\pi r/\lambda$. Thus we find $r = 8 \text{ nm V}^{-1}$. This has to be compared to the coefficient given in the technical notes: 22 nm V^{-1} when the actuator is not loaded. The difference can be explained by several factors. First the additional mass due to the load on the actuator changes its resonance frequency and the amplitude of vibrations. Moreover the fact that the silicon wafer



Figure 7.5 – (a) Photo of the vibrating silicon wafer. (b) Amplitude of the signals at f_{mirr} (blue) and $2f_{mirr}$ (red) versus the voltage applied on the piezoelectric actuator with a fit (dashed lines) by Bessel functions. (c) Amplitude of the signals at f_{mirr} (blue) and $2f_{mirr}$ (red) with a fit by the model presented in 4.1.6 (dashed lines).

is glued onto the actuator may bring additional non linearities in the displacement versus voltage characteristics. Finally, the angle of incidence on the wafer was not perfectly controlled which can bring an error on the evaluation of r.

The theoretical model requires to enter several parameters: the small signal gain α_0 , the ratio I_R/I_{sat} and the Henry coefficient β . The small signal gain and the ratio I_R/I_{sat} have been estimated using the experimental conditions at $\alpha_0 = 8 \text{ cm}^{-1}$ and $I_R/I_{sat} = 0.62$, whereas β has been estimated the same way as in section 3.3.3. A script on Matlab was used to compute the signal at f_{mirr} and $2f_{mirr}$ versus the amplitude of the phase modulation. The computed data were then fitted on the experimental data by expanding the abscissae in order to find the right relation between ϕ and V_{mirr} . With this method, the best agreement was found for $V_{mirr} = 10\phi$ which leads to the same value of the piezoelectric coefficient r as with the fit using Bessel function.

Vibration of a white sheet of paper The previous experiment was repeated using a scattering white sheet of paper instead of the reflecting silicon wafer. This new sample is represented in Fig. 7.6.(a). The same measurements as with the mirror were carried out, and the new set of experimental data were fitted by the same two models (see Fig. 7.6.(b) and Fig. 7.6.(c)). The results of both fits are similar to the mirror case. The value found for k is 0.1 rad V⁻¹ with the Bessel functions, and we had to multiply ϕ by ten in order to make both curves (for f_{mirr} and $2f_{mirr}$) from the model overlap the experimental points.



Figure 7.6 – (a)Photo of the vibrating paper. (b) Amplitude of the signals at f_{mirr} (blue) and $2f_{mirr}$ (red) versus the voltage applied on the piezoelectric actuator with a fit (dashed lines) by Bessel functions. (c) Amplitude of the signals at f_{mirr} (blue) and $2f_{mirr}$ (red) with a fit by the model presented in 4.1.6 (dashed lines).

These are the same results as in the previous experiment which lead to a piezoelectric coefficient of $r = 8 \text{ nm V}^{-1}$.

These two experiments confirm that our model is able to predict well the response of our setup to a phase modulated signal. In addition, it highlights the presence of a refractive index grating and shows that at small amplitude phase modulation, the linear component is much stronger than the quadratic component.

7.2.3 Non-destructive testing perspectives.

These two preliminary experiments show that this adaptive gain interferometer using a crystal of Nd:YVO₄ is quite interesting to probe the displacement of rough surfaces. Indeed, our system has a linear response through the refractive index grating. This linear response is necessary for the retrieval of the sample local displacement and thus reconstructing the waves propagating on the surface of a sample. Hence, setups of this kind can be used to remotely probe the displacement of various surfaces.

7.3 Ultrasound detection by adaptive gain interferometry.

The main goal of this project is to detect photoacoustically generated ultrasound on a biological sample in order to perform non-contact photoacoustic imaging. This section will describe the experiments performed with our adaptive gain interferometer that led to the detection of photoacoustic signals.

7.3.1 Detection of ultrasound inside a phantom.

Before trying to detect short US pulse generated by photoacoustic interaction, we tried to detect ultrasound sent inside a sample by an ultrasonic transducer.

Principles The idea of this experiment is to send ultrasound on one side of the sample and detect the vibration they cause on the opposite side. A representation of the principle of this experiment is given in Fig. 7.7. The sample used is a thick phantom of Agar with a thin multiple scattering layer used to perform the detection of the vibrations caused by the ultrasound. Our samples were between 8 cm and 10 cm thick and we used the



Figure 7.7 – Drawing of the phantom prepared for the detection of transducer-generated ultrasound by TWM.

A307S transducer already used in chapter 6. Its main characteristics are given in Tab. 6.3 in the previous chapter. The thickness of the samples was chosen to be larger than the focal length of the transducer (51.5 mm). In this configuration the acoustic signal reaching the surface of the sample consists of low-intensity diverging waves, which mimics a photoacoustic signal.

Ultrasound detection The detection of ultrasound in this configuration has been tried using either long US bursts or short US bursts. In order to simulate a biological sample, we used cooked chicken breasts to make the scattering slice of our sample. The thickness of a slice of cooked chicken breasts is between 1 mm and 2 mm so we stacked several of them in order to have a scattering part with a thickness around 5 mm (see Fig. 7.8). As the pump works in a QCW regime, in order not to heat our sample too much, the signal and reference beams are turned ON only during the pump pulse using the AOMs on each arm.



Figure 7.8 – Picture of the phantom prepared for the detection of ultrasound inside the gel generated by a transducer.

The transducer was working at a frequency of $f_{US} = 4.8$ MHz and sending N_c cycles in the sample. The signal for different values of N_c is represented in Fig. 7.9. An offset has been added to each signal for N_c equal to 10, 25, 50 and 100 for representation purposes. This experiment shows that the setup is able to detect ultrasound coming from deep inside the sample by probing the surface of the sample. With this method we are able to detect long US bursts (100 cycles) as well as short bursts (2 cycles). These signals were obtained by averaging over 100 acquisitions



Figure 7.9 – Time signals obtained with our setup while sending US burst through the sample. The length of the signals are 2, 10, 25, 50 and 100 cycles for the signal in blue, red, green, purple and black.

Response time Using this experiment, it is possible to measure the holographic response time of our setup using a method inspired by the technique (from [90]) used in the previous chapter. We sent on our sample an amplitude modulated ultrasonic excitation. The modulation signal is a square wave at a frequency $f_{mod} = 500 \text{ kHz}$ with a 0.5 duty cycle. Then, as in chapter 6, the time signal is recorded and the amplitude of the peak at $3f_{mod}$ is plotted as a function of a frequency shift Δf between the signal and the reference. The experimental data, fitted by a Lorentzian model is represented in Fig. 7.10, the fit is used to ascertain the response time of our setup in this configuration. This method yields a response time of $\tau_R = 25 \text{ µs} \pm 5 \text{ µs}$ (with a determination coefficient of $r^2 = 0.9$),



Figure 7.10 – Amplitude of the component of the acousto-optic signal at $3f_{mod} = 1.5$ MHz vs. the frequency shift between the signal and the reference beam (dots) and lorentzian fit (solid line).

which is consistent with the measurements made in the two previous chapters. This configuration has a stronger pumping than the orthogonal configuration of chapter 6 so the fluorescence lifetime is shorter. However, because there is only one pass of the reference through the crystal, the response time is a bit longer than the two-pass configuration of chapter 5. Once again, this highlights the capacity of Nd:YVO₄-based holographic setup to perform fast wavefront adaption, which is interesting for measurements on rapidly changing samples.

7.3.2 Detection of photoacoustic signals.

This final part presents the results we obtained using this adaptive holographic setup for photoacoustic imaging. These experiments were done using the same kind of sample as described earlier, illuminated by a Q-Switch laser at 532 nm.

Sample The samples are constituted of two parts. A thin (a few millimetres) highly scattering part that is used to perform the optical detection of ultrasounds, and a thick (a few centimetres) part of gel made with agar and water. In addition, a thin absorber



Figure 7.11 – Drawing of the sample prepared for the photoacoustic imaging experiment.

is hidden behind the scattering layer as illustrated in Fig. 7.11. The absorber used is either a $170 \,\mu\text{m}$ in diameter reddish fishing line, or a black sewing wire with a diameter around $500 \,\mu\text{m}$. As a first step, the sample is illuminated by the side, through the lightly scattering part of the sample in order to have a strong photoacoustic signal.

Excitation laser The excitation laser is a pulsed laser manufactured by Quantel. It works with a doubled Nd:YAG crystal to operate at 532 nm. Its main features are shown in Tab. 7.1.

Manufacturer	Quantel
Model	Brilliant
Oscillator material	Nd:YAG
Working wavelength	$532 \mathrm{nm}$ (doubled $1064 \mathrm{nm}$)
Pulse operation	Q-Switch
Pulse length	$4\mathrm{ns}$
Repetition rate	$10\mathrm{Hz}$
Max energy per pulse	$150\mathrm{mJ}$

Table 7.1 – Characteristics of the Quantel laser.

Photoacoustic signal With this configuration we managed to record photoacoustic signal from both the thin fishing line and the black sewing wire. In order to have a good SNR, the signal has to be averaged over 200 laser pulses. The averaged time signal for both the wires are plotted in Fig. 7.12.(a). Figure 7.12.(b) represents the Fourier spectrum of each photoacoustic signals. As mentioned in the description of the setup, the



Figure 7.12 – (a)Time signals obtained for a fishing line and a black sewing wire. (b)Fourier spectrum of the signal obtained for a fishing line and a black sewing wire.

signal is filtered by a High-Pass filter and a Low-Pass filter. Their cut off frequency are set up differently, depending on which target we use. Indeed, the larger the target, the lower the emitted ultrasonic frequency. For the sewing wire, which has a diameter around 500 µm, the filters are set to $f_{HP} = 200$ kHz and $f_{LP} = 3$ MHz, whereas for the fishing line they are set to $f_{HP} = 600$ kHz and $f_{LP} = 6$ MHz. The acquisition is delayed by 2 µs to avoid an artefact corresponding to parasitic light from the Q-Switch laser, which, despite all our effort to block it, manages to reach the photodiode. The 3.3 µs delay between the excitation laser pulse and the signal corresponds to the time of flight of the US between the emitter (the wire) and the probe laser spot on the surface of the sample.

1D Scanning Now that we are able to detect a photoacoustic signal from an absorber hidden behind a multiple scattering layer, it is possible to image this absorber. Indeed, as mentioned in 2.3.2, if we record the signal at several positions of the probe spot on the surface of the sample, it is then possible to numerically reconstruct the ultrasonic source. As our sample contains a 1 dimensional wire, we chose to scan our sample along only one dimension, perpendicular to the wire with a travel pitch of 100 µm over a total range of 8 mm. The time signals acquired for each positions are represented as an image in Fig. 7.13. On part (a) are the time signals for the fishing line, and on part (b) are the time signals for the sewing wire. These acquisitions were made by averaging the signal 200 times



Figure 7.13 – Concatenation of the time signals obtained for different position of the probe spot for the fishing line (a) and the sewing wire (b)

and with a mean power of 600 mW for the 532 nm laser. The time t = 0 corresponds to the Q-Switch laser pulse, the photoacoustic signal arrives a few microsecond later depending on the position of the probe on the sample. The time of flight of the US wave for each position draws a rounded shape on the images, which correspond to the wavefront emitted by the absorber due to the optical excitation. On the signals from the sewing wire, we can notice that there are extra echoes for $t > 5.5 \,\mu s$ and X between $-4 \,\mu m$ and $0 \,\mu m$. They are due to a secondary path of the wave from the absorber to the probed spot. Indeed as represented in Fig. 7.14, the wire is in a corner of the sample. In addition to the direct path from the wire to the probed spot (in blue), a second path (in purple), where
the ultrasonic pulse is reflected on the interface between the gel and the air can create additional signals after the direct signal. The closer to the edge the probed spot is, the closer in time this additional signal will be. Also, when the sample is scanned along the



Figure 7.14 – If the absorber is too close from the edge of the gel, there are two paths the link the absorber to the probed spot. This result in an artefact visible on Fig. 7.13.(b)

x-axis, the amount of backscattered light by the sample inside the crystal changes. This can have several effects that hinder the imaging process such as:

- 1. The amplitude of the photoacoustic signal may vary independently of the amplitude of the acoustic wave.
- 2. Backscattered light becomes dimmer than noise.
- 3. Backscattered light becomes too intense resulting in a saturation of one of the electronic component.

In the first case, the time of flight information can be recovered in order to localise the optical absorber, in the two other cases, no information can be recovered from the acquired data. Such missed lines can be seen on the images of Fig. 7.13. Also, in order to really see the wavefront shape, the previous images have been corrected by normalizing each line between -1 and +1. With this process, the information of the amplitude of the wave is lost, anyway, they could not be properly used because of (1).

Despite this issue, it is still possible to try to reconstruct the absorber using a beamforming algorithm, developed for photoacoustic imaging with an array of transducer, on the normalised data. The results of the reconstruction algorithm is represented in Fig. 7.15, part (a) being the reconstruction of the fishing line and part (b) the reconstruction of the sewing wire. The wires appear as dark spots on a gray background. We can notice that whether it is the fishing line or the sewing wire, the size along the Z-axis fits the reality, whereas the size along the X-axis is stretched. This can be explained by two phenomena: on the one hand, the finite collection angle of the ultrasonic wave and on the other hand, the loss of information due to the normalisation of each signals. The sample is scanned along an 8 mm range, with the wires being around 5 mm deep, if we suppose that the ultrasound propagates at 1.5 mm µs^{-1} , and that their frequency is between 1 MHz and 5 MHz, the resolution in the X direction is between 180 µm and 930 µm, which explains the spread in this direction. Also, as mentioned earlier, the normalisation of each signals



Figure 7.15 – Reconstructed images by the beamforming algorithm. (a) is the fishing line and (b) is the sewing wire.

should be more attenuated, the fact that we normalised the signals individually tricks the algorithm into reconstructing a larger source.

7.4 Conclusions and perspectives

Conclusions This final chapter reported our preliminary experiments aimed at using adaptive holography in Nd:YVO₄ for the detection of vibrations on the surface of samples. We showed that we could benefit from the additional refractive index modulation in order to have a linear detection of small phase modulation due to vibration of the sample. First, we used our setup to demonstrate the possibility of detecting high frequency vibrations of both a reflective surface (a silicon wafer), and a scattering surface such as a piece of white paper. In a second time, the setup was used to detect ultrasound from inside a phantom of biological tissue. We tested two means of generating the ultrasound. First, we simply sent US using a transducer and looked at the resulting vibration on the opposite side. Secondly we detected US generated by the photoacoustic effect in order to detect absorbers hidden behind a multiple scattering layer made of chicken breast. We showed that it was possible to detect thin wires with sub-millimetric diameters using this detection method, which can be interesting for *in vivo* applications.

These experiments are a first step to show the interests of using gain media to perform fast wavefront adaptive holography for the detection of vibration or ultrasound. Our detection system has several advantages. Firstly, because of its optical nature, there is no contact between the probe and the sample, which is interesting for applications that requires not to touch the samples. Secondly, we showed in this chapter and the two previous ones, that the response time of holography in gain media is quite fast (several tens of microsecond). This is very interesting for application in difficult conditions where the tested sample is vibrating or moving such as *in vivo* experiments or industrial testing in harsh environment. Thirdly, the system has a very broad bandwidth which is mainly limited by the electronic used for the signal processing. If we consider only the optical part of the system, the bandwidth is limited by the crystal for the lower frequency (so it is around several 10 kHz) and by the photodiode for the upper frequency (which means that it can go up to several 100 MHz).

Perspectives In spite of the promising results described in this chapter, there is still room of improvements on the setup. As an example, we could imagine having a system that adapts the power of the probe beam in order to fix the issue of the changes in the amount of backscattered light in the crystal. This way the information on the amplitude of the photoacoustic-generated ultrasound could be used for the reconstruction resulting in the truer image. Another possibility could be to record the signal before being filtered in order to have the DC level and normalise the PA signal. The noise level due to parasitic light from the ASE and lasing effect is quite strong. Using optical fibres in the system could be a way to reduce the noise by better guiding each beam. It could also make the system easier to align and more robust to misalignment. Another improvement could be in the image reconstruction. Indeed, the algorithm used was originally developed for measurement made with an array of transducers. Writing a new algorithm taking into account the specificities of the optical detection could yield better images.

Conclusions and perspectives

Conclusions

Optical imaging deep inside biological tissues is a relatively recent field. In the first part of this manuscript we have highlighted the difficulties of accessing optical information deep inside biological bodies. Because light is strongly scattered, direct optical imaging using ballistic light - is nearly impossible after a few millimetres. We saw that this issue could be solved by coupling light with another wave that propagates well inside living tissues: ultrasound. This led to the description, in the second chapter of this manuscript, of two imaging techniques: acousto-optic imaging and photoacoustic imaging. In order to detect acousto-optic signals and photoacoustic signals, we outlined the need for a detection system with the following characteristics:

- Sensitive to very small phase variations.
- Able to detect signal in a light field that has lost its spatial coherence: a speckle pattern.
- Can adapt fast enough to compensate the natural movements of the sample. Essential in the case of *in vivo* application.

As a result, the second part of this manuscript was dedicated to the description of wave mixing in laser gain media. Holographic techniques enable to perform wavefront adaption in order to compensate for the low spatial coherence of the light. In addition they are very sensitive to small phase variations. As a result, we described with mathematical models the processes of two-wave and four-wave mixing in a gain medium. These models highlighted the fast holographic response time of gain media with a relation linking the response time to the fluorescence lifetime: $\tau_R = \tau \left(1 + \frac{I}{I_{sat}}\right)^{-1}$. As a lot of gain media have a sub-millisecond fluorescence lifetime, this type of material is well suited for *in vivo* applications. In addition we explained, through the modelling of two-wave mixing, the presence of two types of grating inside the medium. Indeed, the gain grating induces an additional refractive index grating. The main consequence of this secondary grating, is the possibility to have a linear response which is much more sensitive in case of very small phase variations.

Finally, part III of this manuscript described two methods of detecting acousto-optic signals, and one method of detecting photoacoustic signals. First, we used phase conjugation by four-wave mixing in Nd:YVO₄ for the detection of acousto-optic signals. This enabled us to image the diffused light pattern inside a 5 mm thick slice of chicken breast. The response time of our setup in this configuration was around 15 µs, which is very fast compared to the speckle decorrelation time. Second, we investigated another detection technique of acousto-optic signals: wavefront adaption using two-wave mixing. With this method we managed to detect three black inclusions in a 1.5 cm thick scattering phantom with a reduced scattering coefficient around $\mu_s^* = 3.5 \text{ cm}^{-1}$. In this second configuration, the response time was a little bit longer, 45 µs, because of a less intense reference beam and a less intense pumping. Finally, we used the linear response of our setup to perform the detection of the vibration at the surface of scattering sample. It was applied to the detection of ultrasound generated by photoacoustic effect, which enabled us to remotely image thin wires hidden behind several millimetres of chicken breast. In this last configuration, our setup had a response time around 25 µs.

As a conclusion, gain medium such as Nd:YVO₄ are very interesting to perform holography on light coming from biological samples. Indeed, in each setup we developed, we saw that the holographic response time was always of the order of magnitude of a few tenth of microsecond. This very fast response time guarantees the detection setup to be more robust to speckle decorrelation that can arises when performing *in vivo* optical imaging. Moreover, the possibility of inducing a refractive index grating opens a lot a interesting applications. This refractive index grating has two main advantages: it enables a linear detection and is much more sensitive in the case of the detection of small phase modulation.

Perspectives

Our investigations about dynamic holography in $Nd:YVO_4$ for application such as acoustooptic imaging or photoacoustic imaging showed promising preliminary results. We highlighted interesting features of wave mixing in $Nd:YVO_4$:

- High gain due to the natural amplification of light inside the gain medium.
- Possibility of perfoming wavefront adaption by two-wave mixing as well as phase conjugation by four-wave mixing.
- Sub-millisecond holographic response time.
- Linear and quadratic response to a temporal phase modulation because of the refractive index grating and of the gain grating.

Therefore, pursuing the understanding of these processes can lead to various applications in domain such as biomedical imaging and non-destructive testing. The various setups described in this thesis were preliminary experiments. Lots of modifications can be made in order to improve the performances, the robustness and ease of use. For instance, using optical fiber could protect the setup from misalignment and make it easier to handle. In order to reduce the number of light source, we could imagine inserting the gain medium inside a cavity to make a laser, and use the crystal as both the light source and the wavemixing medium.

On acousto-optic imaging

Through two optical setups, we demonstrated that an acousto-optic detection system based on holography in Nd:YVO₄ was possible. We showed that this medium was an attractive candidate for interferometry with speckle beams for *in vivo* applications because of the relatively fast response time (< 100 µs). Despite being in the optical therapeutic windows, the working wavelength, 1064 nm, is not ideal for biological application. Indeed, above 1 µm light starts to be strongly absorbed by water and does not penetrate very deep inside inside biological tissues. Therefore, extending these experiments to gain media working around 800 nm could greatly benefit biological applications.

Among the existing gain media, some of them have the right combination of characteristics (fluoresence lifetime, saturation intensity,...) for *in vivo* application. In particular, experimenting with Ti:Sapphire, Alexandrite or Cr:LiSAF could bring very interesting results. Moreover, some of these materials are tunable which could give access to multispectral imaging which is impossible with Nd:YVO₄.

On photoacoustic imaging and non-destructive testing

Our third setup was aimed at detecting local displacements of a rough surface. It was possible thanks to the wavelength difference between the light source (Nd:YAG) and the amplifier (Nd:YVO₄) which induces a refractive index grating. The remote probing of vibrations at the surface of a multiple scattering sample opens the possibility of performing non contact photoacoustic imaging. In certain environment, such as a sterile operating room, this remote sensing is interesting since it does not requires to use sterile protection between the probe and the sample. Contrary to acousto-optic imaging, the wavelength 1064 nm is well adapted. Indeed, as we want the signal to come only from the surface of the sample, it is necessary that the light does not penetrate too deep inside the sample. Moreover it must not be absorbed too much, therefore 1064 nm seems a good compromise. The very large bandwidth offered by optical systems is also of great interest for photoacoustic imaging since it gives access to several different size scales of absorbers.

Another attractive application of this work is non destructive testing. Our experiments were performed on biological samples, but they can be extended to other materials. Measurement of the vibrations of any rough surface is possible which is interesting for a lot of industrial applications where materials are tested using surface acoustic waves. The remote testing possibility, plus the high speed of the dynamic holographic process makes our setup robust if materials need to be tested in difficult environments with a lot of parasitic vibrations.

Appendix A

Scientific production

Published peer-reviewed articles (Full text at the end of this list)

B. Jayet, J.-P. Huignard, and F. Ramaz. Optical phase conjugation in Nd:YVO₄ for acousto-optic detection in scattering media. *Opt. Lett.*, 38(8):1256–1258, April 2013.

B. Jayet, J.-P. Huignard, and F. Ramaz. Fast wavefront adaptive holography in Nd:YVO₄ for ultrasound optical tomography imaging. *Opt. Express*, 22(17):20622–20633, August 2014.

Oral presentations

B. Jayet, J.-P. Huignard, and F. Ramaz.

Optical phase conjugation in gain media: Detection of an acousto-optic signal for optical imaging of biological tissue.

OSA IONS12 - University of Naples Federico II, Naples, Italy (July 2012)

B. Jayet, J.-P. Huignard, and F. Ramaz.

Optical phase conjugation applied to acousto-optic imaging of thick scattering media with Nd:YVO₄ gain medium.

SPIE Photonic West 2013 - [8581-58] - The Moscone Center, San Francisco, USA (February 2013)

B. Jayet, J.-P. Huignard, and F. Ramaz.

Optical phase conjugation applied to acoust o-optic imaging of thick scattering media with $\rm Nd: YVO_4$ gain medium. 8^{ème} Journées d'Imagerie Optique Non Conventionelle - ESPCI ParisTech, Paris, France (March 2013)

B. Jayet, J.-P. Huignard, and F. Ramaz.

Optical phase conjugation in gain medium (Nd: YVO_4). Detection of acousto-optic signals in thick scattering media.

IOP PR'13 - The Winchester Hotel, Winchester, UK (September 2013)

B. Jayet, J.-P. Huignard, and F. Ramaz.

Wave-mixing in gain media (Nd:YVO₄) for acousto-optic imaging. 3AF OPTRO 2014 - OECD Conference Center, Paris, France (January 2014)

B. Jayet, J.-P. Huignard, and F. Ramaz.
Wave-mixing in gain media (Nd:YVO₄) for acousto-optical imaging.
SPIE Photonic West 2014 - [8943-44] - The Moscone Center, San Francisco, USA (February 2014)

B. Jayet, J.-P. Huignard, and F. Ramaz.

Holographie dans les milieux à gain pour l'imagerie acousto-optique des milieux diffusants epais.

OptDiag 2014 - Insitut de Physique du Globe, Paris, France (May 2014)

Poster presentations

B. Jayet, J.-P. Huignard, and F. Ramaz.

Détection d'un signal acousto-optique par conjugaison de phase dans un milieu à gain : Application à l'imagerie des milieux diffusant épais.

7^{ème} Journées d'Imagerie Optique Non Conventionelle - ESPCI ParisTech, Paris, France (March 2012)

E. Benoit, J.-L. Genisson, S. Farahi, A. A. Grabar, B. Jayet, M. Tanter, and F. Ramaz. Joint acousto-optic and ultrasound images of small animals. *SPIE Photonic West 2013 - [8581-95]* - The Moscone Center, San Francisco, USA (February 2013)

B. Jayet, J.-P. Huignard, and F. Ramaz.

4-wave mixing in laser media for acousto-optic detection in scattering media. Cargèse Summer School: Controlling the propagation of waves in complex media. - Institut Scientifique de Cargèse, Cargèse, France (June 2013)

Optical phase conjugation in Nd:YVO₄ for acousto-optic detection in scattering media

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Received February 14, 2013; accepted March 9, 2013; posted March 15, 2013 (Doc. ID 184637); published April 4, 2013

Acousto-optic imaging is a technique that maps the optical properties of a thick scattering sample with millimetric resolution. The detection of the acousto-optic signal represents a challenge, because it is very weak among a strong parasitic signal. Various methods based on holography in photorefractive crystals or digital holography have been studied. Here dynamic holography is obtained with the gain medium Nd:YVO₄. We study the experimental feasibility of a detection system based on holography in a gain medium and show acousto-optic results obtained in a 5 mm slice of chicken breast. © 2013 Optical Society of America

OCIS codes: 090.2880, 110.0113, 110.7170, 170.3660, 170.3880, 190.5040.

Tagging photons using ultrasound is a method to map the optical properties of thick scattering media, such as biological tissues [1]. To detect the ultrasonically tagged photons coming out of a scattering sample, dynamic holographic techniques are frequently used. A hologram of the tagged (or untagged) light is written inside a medium through the interference of a speckle pattern with a reference beam. Then the hologram can be either read by the reference (two-wave mixing) to generate a matched wavefront and detect the signal on a single photodiode, or read by a counterpropagating reference beam (four-wave mixing or phase conjugation) to generate a wave that will counterpropagate through the scattering sample and regenerate the original wave.

Usually, these nonlinear interactions are performed using photorefractive crystals, such as SPS [2], BSO [3], or GaAs [4] or, in the case of phase conjugation (PC), spatial light modulators (SLMs) [5] can also be used. Those methods have proven to be efficient and give good results on static samples. However, they have almost never been tested in vivo where the sample can be moving due to breathing, heart beats, or blood flow. In those cases, the speckle decorrelates overtime (1 ms [6]) and the recording of the hologram may not be fast enough. Indeed, most of the photorefractive crystals have a response time between 1 and 100 ms; thus, if the movements of the sample change the speckle with a typical time of around 1 ms, the hologram will become blurred, making the detection inefficient. That is why *in vivo* experiments may require the use of faster materials than those currently used.

In this Letter, we present the experimental realization of an acousto-optic detection based on PC in a Nd:YVO₄ laser crystal. The idea is to generate the PC of the scattered photons coming from the medium and send them back to regenerate the original wave, which is detected on a photodiode. When the ultrasonic burst travels trough the scattered light, it tags the photons. As these photons undergo a frequency shift ($\omega_L \pm \omega_{\rm US}$), they will no longer participate in the PC, resulting in a decrease of the magnitude of the phase conjugate beam. Moreover, the use of focused ultrasonic bursts gives millimetric resolution within the volume of the sample and thus enables the technique to be used as an imaging system. Holographic detection is achieved in gain media through the phenomenon of gain saturation [7]. Neodymiumdoped vanadate is a good answer to the problems of speckle decorrelation raised earlier, because it has a fast response time. In the case of a hologram written within a gain medium, the response time is typically limited by the spontaneous emission lifetime at 1064 nm, i.e., 100 μ s in Nd:YVO₄, thus 10–100 times faster than the usual photorefractive crystal response time. This highspeed dynamic holography should protect the detection system from speckle decorrelation that may occur when performing in vivo experiments. Fast holography using Nd:YVO₄ has already been implemented in an adaptive gain interferometer based on two-wave mixing for metrology with speckle beams [8], but this is the first time, to our knowledge, that it is used to perform PC on speckle beams for acousto-optical imaging.

Neodymium-doped vanadate is an anisotropic gain medium with a very high emission cross section at 1064 nm along its c axis. As near-infrared light is still in the optical therapeutic window, the use of Nd:YVO₄ should be valuable for in vivo experiments. Amplification or PC processes requires a population inversion of the Nd³⁺ ions in the crystal. It is achieved via optical pumping at the absorption peak around 808 nm with a high-power laser diode, as shown in Fig. 1. The hologram necessary for PC is written through the spatial modulation of the gain by an inhomogeneous illumination pattern coming from the interference between the signal and a reference beam. This gain hologram acts as a diffraction grating on a third beam that counterpropagates with the reference and, thus, generates the phase conjugate of the signal.



Fig. 1. (Color online) Gain modulation in the crystal.

0146-9592/13/081256-03\$15.00/0



The crystal used in our experiments is a $5 \text{ mm} \times 5 \text{ mm} \times 5 \text{ mm} c$ -cut cube (Crylight Photonics Inc.) with coatings on two opposite sides. The front side, where the reference and the signal beams enter the crystal, is antireflection coated for 1064 nm, whereas the back side, where the pump beam enters, has an antireflection coating for 809 nm and a high-reflectivity one at 1064 nm. This coating enables having a double-pass amplifier and generating the counterpropagating reference beam necessary to perform PC.

At first, the crystal is used as a double-pass amplifier. The laser at 1064 nm is a CW 1 W single longitudinal mode Nd:YAG (CrystaLaser Inc.), coupled to a 5 W Yb-doped fiber amplifier (Keopsys Inc.). It propagates within the crystal at normal incidence so that the reflection is superimposed with the incoming light. The crystal is pumped at 805 nm by a LIMO100-F200-DL808 laser diode for 1 ms with a 100 W pulse repeating at 16 Hz to allow the crystal to cool down. The pulse is focused by a 18 mm focal length lens a few millimeters outside the front side in order to have a large pumped volume. In these conditions, we obtain an amplification factor up to 1000, leading to a gain of 7 cm⁻¹, as shown in Fig. 2. The experimental data are fitted with a theoretical model from [7], leading to an intensity of saturation around $1.25 \text{ kW}/\text{cm}^2$, consistent with the theory.

The next step is to use this amplifier as a phase conjugate mirror. In our configuration, the reflection of the reference beam on the backside of the crystal is the second reference. We have then to add a signal beam coherent with the references to use our amplifier as a phase-conjugate mirror (see Fig. 3). The phase-conjugate beam is isolated from the original signal by a 50/50 nonpolarizing beam splitter (BS) cube and sent to a Thorlabs PDA36A-EC photodiode. The signal from the photodiode is recorded with an ADLINK-PCI9646D acquisition board in the computer driven by MATLAB. The board is triggered by the laser pump so the acquisition starts when the pump fires a pulse. Both reference and signal are modulated at 80 MHz using AAOptoelectronics acoustooptic modulators (AOMs). These modulators are used as fast shutters and also to record the noise by shifting the frequency of one beam to cancel the PC beam and measure parasitic light.

To characterize our system, we have looked at the reflectivity of the PC mirror, defined by $R = P_{S^*}/P_S$ where



Fig. 2. Amplification factor obtained in a two-pass configuration as a function of the power of the input beam. Experimental data are fitted according to [7].



Fig. 3. (Color online) PC setup. PBS, polarizing beam splitter; BS, 50/50 beam splitter; HWP*i*, *i*th half-wave plate; AOM*i*, *i*th acousto-optic modulator; PD, photodiode detection of the phase conjugate signal; UST, ultrasonic transducer with center frequency f = 2.25 MHz; M1, high-reflectivity coating at 1064 nm.

 P_{S^*} is the power of the phase conjugate beam and P_S is the power of the input signal beam. We initially worked without a sample, and used light coming directly from the Nd:YAG laser. The PC reflectivity is plotted as a function of the signal input power P_S in Fig. <u>4</u> with different reference power P_R . We can see that our system has a reflectivity that varies between 10% and 30%, depending on the conditions. It seems that the greater P_R , the greater the reflectivity.

Now that our PC mirror works with a plane wave, the final step is to use it with a speckle from a multiscattering medium as the signal and see if it is possible to detect an acousto-optic signal. The pumped volume is around 0.4 mm^3 , which contains around 2.5×10^4 speckle grain according to wavelength and the NA of the system. Our sample is a few-millimeter-thick slice of chicken breast held between two slabs of plexiglass. The thickness of the sample is controlled using chocks made with several 1 mm thick microscope slides. To keep enough power to go through several millimeters of scattering sample, the reference power is around $P_R = 150$ mW. This value has been chosen after a few experiments using scattering samples with a thickness between 2 and 5 mm. The chicken is illuminated with a power of 550 mW and the light coming out is collected by a lens (f = 5 cm, NA = 1) in a 2f configuration. To send ultrasound into the chicken breast, the sample holder is placed under an ultrasonic transducer immersed in a water tank. The transducer sends one short burst consisting of several cycles of a sinusoid at $f_{\rm US} = 2.25$ MHz generated by a



Fig. 4. Reflectivity of the phase conjugate mirror as a function of the signal power for different reference power.



Fig. 5. (Color online) (a) Temporal signal without ultrasound and with ultrasound. (b) 2D scan of the chicken breast.

Tektronix signal generator and a 25 W RF amplifier. The generator is triggered by the signal used to trigger the acquisition board, i.e., the pulse from the pump.

Figure 5(a) shows part of the temporal phaseconjugate signal as the pulse from the pump is launched. In this experiment, the sample was a 5 mm thick piece of chicken breast. The acquisition board samples the signal at 40 MS/s and enables averaging the signal over 100 times in order to have a clear signal. The solid and the dotted lines, respectively, represent the signal with and without ultrasound. The burst lasts five cycles—giving a resolution of around 3 mm—and is fired 200 µs after the pump pulse. Despite fluctuations, the hole caused by the passage of the ultrasonic burst within the scattered light inside the sample is clear.

To have a better visualization of the acousto-optic signal, both signals (with and without ultrasound) have been corrected by subtracting a baseline calculated by fitting the curve with a linear model. Moreover, the signal has been inverted so the hole appears as a peak. An attempt at doing a 2D image of the light scattering pattern within the sample has been made by scanning transversally the ultrasonic transducer with a high-precision motor, and the resulting image is shown in Fig. 5(b). In this image, we can see the light spot at a depth of $\overline{2.5}$ mm in a 5 mm thick piece of chicken breast, corresponding to $5l^*$ (the transport mean free path in biological tissue is around 1 mm). In those conditions, the spot of the scattered light at the imaging depth is 2-3 mm wide, which is close to what appears on the scan. One of the next steps for the experiment is to expand the spot both by using a thicker sample and also by using a divergent or an expended beam to light the sample.

In conclusion, various detection methods exist to retrieve the acousto-optical signal coming from a multiscattering sample. Currently, the most commonly used are holography in photorefractive crystals or digital holography using a camera and an SLM. These techniques have shown good results but have never been tested in vivo, where their response time could be a problem for imaging. An alternative to this problem could be the use of a gain medium, such as $Nd:YVO_4$. Its ability to amplify light gives this method the necessary sensitivity to detect the acousto-optic signal and its fast response time practically avoids sensitivity to speckle decorrelation. The experiments described in this Letter show promising first results with vanadate. The next steps are to study the influence of the pumping (especially the length of the pulse and the time between two pulses), reduce the noise coming from the spontaneous emission of the crystal, by thinking of new pumping configuration, for example, and investigate the possibility of two-wave mixing.

The authors gratefully acknowledge the Délégation générale de l'Armement, the Agence Nationale de la Recherche under the project ICLM-ANR-2011-BS04-017-01, and, finally, the Laboratory of Excellence with the French Program "Investments for the futur" (LABEX WIFI) under the reference ANR-10-IDEX-0001-02 PSL* for financial support.

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Fast wavefront adaptive holography in Nd:YVO₄ for ultrasound optical tomography imaging

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Several approaches exist to perform acousto-optic imaging of Abstract: multiple-scattering media such as biological samples. Up to now, most of the coherent detection methods use holographic setup based on photorefractive crystals such as BSO or SPS. One of the issue of these techniques is the moderate response time compared to the speckle decorrelation time in biological sample. We introduce a new approach for the holographic detection based on two-wave mixing in a Nd:YVO₄ gain medium enabling us to perform a fast wavefront adaption (50 μs) of the speckle field from a multiple-scattering sample.

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OCIS codes: (090.2880) Holographic interferometry; (110.0113) Imaging through turbid media; (110.7170) Ultrasound; (170.3660) Light propagation in tissues; (170.3880) Medical and biological imaging; (190.7070) Two-wave mixing.

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1. Introduction

Tagging photons with ultrasound is an attractive method to image the optical properties inside multiple-scattering media such as biological tissues [1]. Indeed, conventional optical imaging techniques cannot be used on biological tissues because they rely on ballistic light, which is attenuated very fast in such media due to a very high scattering coefficient. A way to harvest information from the diffused light is to couple it with ultrasound (US) through the acousto-optic effect. As the US at some MHz are ballistic inside most biological tissues, it is possible to focus them in a small volume thus creating a small virtual source of ultrasonically tagged photons. These so called *tagged photons* are slightly shifted in frequency, due to the US phase modulation. Then, with an appropriate detection, it is possible to probe the local optical properties of the sample by scanning the focus spot of the US within the sample. As an increase of the tagged photons results in a decrease of the untagged photons and vice versa, the information from the acousto-optic signal can be retrieved on either component. The resolution of this acousto-optic imaging technique, given by the size of the virtual tagged photons source, is connected to the size of the acoustic focal spot (typically 1 mm at echographic frequencies), whereas the contrast of the image is given by the intensity of light at the US focal point which means that it is of optical nature.

If a single detector is used at the exit of a multiple-scattering sample, the signal to noise ratio (SNR) for the detection of the modulation created by the US is weak due to first, the spatial random phase of each speckle grain [2] and second, the small fraction of photons tagged by the acoustic field. Several methods exist in order to improve this SNR, they can be either incoherent or coherent. The incoherent methods are based on creating a very sharp spectral filter to separate the tagged from the untagged photons and do only a *flux* detection. They are usually based on Fabry-Perot cavities [3] or spectral hole burning [4, 5]. The coherent methods, on the other hand, are based on the idea of phasing each speckle grain of one component (tagged or untagged) of the speckle field in order to increase the SNR. This can be performed by creating a new wavefront that is matched with, for example, the wavefront of the untagged photons. All the speckle grains of both wavefronts will interfere coherently on the single detector resulting in a higher SNR. The wavefront adaption can be realized with dynamic two-wave mixing (TWM) holography in non linear photosensitive media.

Usually photorefractive crystals are used to perform the TWM. Photorefractive dynamic holography has enabled the detection of absorbing inclusions embedded inside scattering phantoms [6] and has also been used to perform phase conjugation of the ultrasonically tagged photons in order to focus back behind a scattering sample [7]. One of the issue that can come up with *in vivo* experiments is that the speckle has a decorrelation time constant of about 1 ms [8, 9] in biological tissues, while the required time to write the hologram inside a photorefractive crystal is usually between a few ms to 100 ms [6, 7]. To overcome this problem, we propose in this article a new approach of TWM based on a gain media (Nd:YVO₄). The response time

in vanadate crystal is governed by the spontaenous emission lifetime which is typically 90 μs . Nevertheless, this lifetime, and also the response time can be reduced under certain conditions such as strong pumping or saturation effects [10].

Wave mixing in gain media has already been theoretically and experimentally investigated [11, 12]. It has also been used to perform phase conjugate laser cavities [13], vibrometry of rough surfaces [14] and detection of acousto-optic signals from a multiple-scattering sample by four-wave mixing (FWM, phase conjugation) [15]. Here we report our investigations on the use of gain media for the detection of the acousto-optic signal by TWM.

At first, we will introduce a model of the interactions of two coherent plane waves interfering inside a diode pumped Nd:YVO₄ crystal. This model will then be compared to experimental measurements. We will also investigate the experimental response time of the holography in Nd:YVO₄ in our setup. Finally, the first experimental results of imaging a scattering phantom with 3 black inclusions will be presented before discussing the outcomes and the perspectives of the use of gain media for this imaging process.

2. Model for TWM in Nd:YVO₄

Gain media are traditionally used either as amplifiers or in laser cavities. Light amplification in these media is the result of a diode pumping and stimulated emission at a higher wavelength. In Nd:YVO₄, Nd³⁺ ions are responsible for the amplification. A simplified diagram of their energy levels (Fig. 1(a)) shows that they absorb light around 808 nm and re-emit photons at 1064 nm by stimulated emission. If the pumping is strong enough, it is possible to create a population inversion between the two levels of the laser transition (level 1 and 2 in Fig. 1(a)). The intensity gain per unit of length of the crystal, $\alpha(\mathbf{r}, t)$, defined by the following relation



Fig. 1. (a) 4 levels energy model of Nd^{3+} ions with time constants. (b)Gain modulation with a transverse diode pumping when two beams A_R and A_S are interfering inside the crystal.

 $I_{OUT} = I_{IN}e^{\alpha L}$ (where L is the length of the gain region), is proportional to the population inversion $\Delta N = N_2 - N_1$ created by the pumping. As indicated in Fig. 1(a), the dynamic of these processes is mainly governed by the lifetime τ of the transition $2 \rightarrow 1$, which is slow compared to the other transitions. In the presence of an incident optical field, the dynamic of

 $\alpha(\mathbf{r},t)$ is given by [14] :

$$\frac{\partial \alpha(\mathbf{r},t)}{\partial t} = \frac{\alpha_0}{\tau} - S(\mathbf{r},t) \frac{\alpha(\mathbf{r},t)}{\tau}$$
(1)

where α_0 is the small signal intensity gain, τ the spontaneous emission lifetime of the laser transition and $S(\mathbf{r},t) = 1 + \frac{I(\mathbf{r},t)}{I_S}$ is the saturation parameter of the amplifier where I_S is the saturation intensity of the gain medium. As the saturation parameter depends on the local light intensity (in W/cm²), it is possible to spatially modulate the gain using an interference pattern and thus to record the spatial phase information of a complex signal beam as shown in Fig. 1(b). Let us consider the interference of two coherent beams (A_R and A_S) inside the crystal. The fringes intensity pattern can be written: $I(\mathbf{r},t) = I_R + I_S + A_R A_S^* e^{j\mathbf{K}\cdot\mathbf{r}} + c.c$ where I_R and I_S are the intensities of the optical fields A_R and A_S and $\mathbf{K} = \mathbf{k}_r - \mathbf{k}_s$ is the interference wave vector. In order to solve Eq. (1), we expanded $\alpha(\mathbf{r},t)$ as a Fourier series in spatial harmonics and kept only the first two terms: $\alpha(\mathbf{r},t) = \alpha^{(0)} + \frac{1}{2}\alpha^{(1)}e^{j\mathbf{K}\cdot\mathbf{r}} + c.c$, where $\alpha^{(0)}$ is the uniform gain (the gain without the spatial intensity modulation) and $\alpha^{(1)}$ is the grating contribution to the gain. Under these conditions, two coupled equations for $\alpha^{(0)}$ and $\alpha^{(1)}$ can be derived from Eq. (1) by only identifying the terms for $\mathbf{K} = 0$ and $\mathbf{K}\cdot\mathbf{r}$ (with terms in 2**K**.**r** neglected):

$$\frac{\partial \alpha^{(0)}}{\partial t} + \frac{1}{\tau} \left(1 + \frac{I_S + I_R}{I_{sat}} \right) \alpha^{(0)} + \frac{1}{\tau} \left(\frac{A_S A_R^*}{2I_{sat}} \right) \alpha^{(1)} + \frac{1}{\tau} \left(\frac{A_S^* A_R}{2I_{sat}} \right) \alpha^{(1)*} = \frac{\alpha_0}{\tau}$$
(2)

$$\frac{\partial \alpha^{(1)}}{\partial t} + \frac{1}{\tau} \left(1 + \frac{I_S + I_R}{I_{sat}} \right) \alpha^{(1)} + \frac{2}{\tau} \left(\frac{A_S A_R^*}{I_{sat}} \right) \alpha^{(0)} = 0$$
(3)

If we assume that the input signal field A_S is small compared to the reference field A_R and that the reference field is either small or of the order of magnitude of the saturation intensity ($I_S \ll I_R$ and $A_S A_R < I_{sat}$), Eqs. (2) and (3) can be simplified:

$$\frac{\partial \alpha^{(0)}}{\partial t} + \frac{1}{\tau} \left(1 + \frac{I_R}{I_{sat}} \right) \alpha^{(0)} = \frac{\alpha_0}{\tau}$$
(4)

$$\frac{\partial \alpha^{(1)}}{\partial t} + \frac{1}{\tau} \left(1 + \frac{I_R}{I_{sat}} \right) \alpha^{(1)} = -\frac{2}{\tau} \left(\frac{A_S A_R^*}{I_{sat}} \right) \alpha^{(0)}$$
(5)

In order to complete the model, we have to consider the evolution of the amplitude of the two optical fields within the crystal. The system of coupled equations for A_S and A_R in the crystal can be derived from Maxwell's equations [16]. If we consider a light propagation along the z-axis we have (with the slow varying envelope approximation):

$$\frac{\partial A_S}{\partial z} = \frac{\alpha^{(0)}}{2} A_S + \frac{\alpha^{(1)}}{4} A_R \tag{6}$$

$$\frac{\partial A_R}{\partial z} = \frac{\alpha^{(0)}}{2} A_R + \frac{\alpha^{(1)*}}{4} A_S \tag{7}$$

Equations (4)-(7) can be simplified by taking into account our experimental conditions which consider that:

- The intensity of the signal beam (I_S) is weak compared to the intensity of the reference (I_R) and to the saturation intensity (I_{sat}) .
- The diode pumping is "ON" and the uniform gain $\alpha^{(0)}$ is established $\left(\frac{d\alpha^{(0)}}{dt} = 0\right)$.
- The reference beam is "ON" and is also established $\left(\frac{dI_R}{dt} = 0\right)$.

Under these assumptions, Eqs. (4)-(7) can be simplified into the following system of coupled equations that can be numerically solved using, for example, a finite difference methods (Euler):

$$\alpha^{(0)}(z) = \frac{\alpha_0}{1 + \frac{I_R(z)}{I_{sat}}} \tag{8}$$

$$\frac{\partial I_R(z)}{\partial z} = \alpha^{(0)}(z)I_R(z) \tag{9}$$

$$\frac{\partial \alpha^{(1)}(z,t)}{\partial t} + \frac{1}{\tau} \left(1 + \frac{I_R(z)}{I_{sat}} \right) \alpha^{(1)}(z,t) = -\frac{2}{\tau} \left(\frac{A_S(z,t)A_R^*(z,t)}{I_{sat}} \right) \alpha^{(0)}(z)$$
(10)

$$\frac{\partial A_S(z,t)}{\partial z} = \frac{\alpha^{(0)}(z)}{2} A_S(z,t) + \frac{\alpha^{(1)}(z,t)}{4} A_R(z)$$
(11)

First, Eqs. (8) and (9) can be considered independently of Eqs. (10) and (11) in order to calculate $I_R(z)$ and $\alpha^{(0)}(z)$. Then, Eqs. (10) and (11) can be solved step by step using a spatio-temporal Euler's method. Another method of solving Eqs. (10) and (11) is by using the Laplace transform of each equations. This leads to an analytical formula for the amplitude of the signal field at the output of the crystal given by Eq. (12). The details of the calculation are shown in the appendix.

$$A_{S}(z,t) = \exp\left(\frac{1}{2}\int_{0}^{z} \alpha^{(0)}(z')dz'\right) \times [A_{S}(0,t) + A_{S}(0,t) * C(z,t)]$$
(12)

where f * g is an "incomplete" convolution operator of function f and g defined as $(f * g)(t) = \int_0^t f(t - \tau)g(\tau)d\tau$ and C(z,t) is the transfer function of the crystal on the input signal beam. It is given by:

$$C(z,t) = \frac{1}{\tau} \exp\left[-\frac{t}{\tau} \left(1 + \frac{I_R(z)}{I_{sat}}\right)\right] \times \sum_{n=1}^{\infty} \left[(-1)^n \left(\frac{I_R(z) - I_R(0)}{I_{sat}}\right)^n \frac{\Gamma(1/2+1)}{\Gamma(n+1)\Gamma(1/2-n+1)} \frac{\left(\frac{t}{\tau}\right)^{n-1}}{\Gamma(n)}\right]$$
(13)

where Γ is the usual Gamma function. Equation (12) is interesting to understand the effects of the crystal amplifier on the signal. The first term, $\exp\left(\frac{1}{2}\int_0^z \alpha^{(0)}(z')dz'\right)$, exhibits the input signal amplification in the crystal and the second term is the time dependent component of the output signal, it is composed of:

- The original signal $A_S(0,t)$.
- The diffracted signal $A_S(0,t) * C(z,t)$ by the gain grating.

This last term represents the input signal filtered by the crystal with a transfer function C(z,t), which expression is given in Eq. (13). This transfer function is a first order transfer function with corrective terms given in the summation. The response time of this filter is, at the first order, $\tau_R = \tau \left(1 + \frac{J_R}{I_{sat}}\right)^{-1}$ as expected from Eqs. (4) and (5). This means that uniform gain $(\alpha^{(0)})$ and modulated gain $(\alpha^{(1)})$ almost exhibit the same temporal behavior, shorter than the fluorescence lifetime if the reference beam is intense enough to saturate the gain. This makes the wavefront adaption in vanadate crystals really fast. If only the first term of the summation in C(z,t) is considered, the formula is consistent with the formalism given by Damzen *et al.* [14] in the case of an established modulation.

3. Experimental conditions

This section describes our experimental conditions. The crystal is a 5 x 5 x 10 mm³ YVO₄ crystal doped at 1%-at with Nd³⁺ ions responsible for the amplification, and corresponding to a saturation intensity around 1 kW/cm² (manufactured by Crylight Photonics). One of the large side is anti-reflection (AR) coated at 808 nm for the pump and the two small sides are AR coated at 1064 nm for the signal and reference beams to enter the crystal in the geometrical configuration presented in Fig. 2(a). It is pumped by a 808 nm laser diode (LIMO100-F200-DL808), the output of this diode is a multimode fiber collimated, then focused inside the crystal by a 18 mm focal length cylindrical lens (CL on Fig. 1(a)). The pump works in a quasi continuous wave (QCW) regime, it consists of 2.5 ms long pulses at a repetition rate of 40 Hz. The peak power of each pulse is 80 W, the QCW regime is used to achieve a high gain and prevent thermal damages from the pumping. The crystal is cooled in an aluminium structure with a peltier module. The signal and reference beams are generated by the same laser source, a



Fig. 2. (a)Geometric configuration of the signal and reference beams inside the crystal. (b) Temporal diagram of the pump, signal and reference beams.

1W-CW single longitudinal mode Nd:YAG laser (CrystaLaser inc.), coupled to a 5W Yb-doped fiber amplifier (Keopsys inc.). Each arm of the setup includes an acousto-optic modulator (AA Optoelectronic) that is used either as a fast shutter (for the experiment in part 4) or to generate a frequency shift between the signal and the reference (for the experiment in part 5). The beams enter the crystal with a small incidence angle (10° for the reference and 20° for the signal) to obtain a total internal reflection [17] on the pumped surface and ensure a good overlap of the interference pattern and the gain volume, which has a typical size of $0.3 \times 0.3 \times 5 \text{ mm}^3$. At the output of the crystal, the signal beam is collected on a fast photodiode (Thorlabs DET10A) amplified by a large bandwidth amplifier (FEMTO DHPVA) and sampled at 40MHz using an ADLINK-PCI9646D DAQ board in the computer driven by MatLab(r).

4. Model versus experiment

In order to verify the model, we performed measurements of the temporal evolution of the gain on a signal beam when it is switched ON in the presence of a continuous reference beam. A 1.5 ms long signal pulse is switched ON with a delay of $\delta t = 500\mu s$ after the pump pulse to have an established population inversion (Fig. 2(b)). In order to observe the temporal behavior of the gain I_{Sout}/I_{Sin} and compare it with the model, we measured the signal both with (I_{Sout}) and without (I_{Sin}) a pump pulse. The signal with pump I_{Sout} is corrected of the amplified spontaneous emission (ASE), which is the main source of noise, by recording the signal on the photodiode between t = 0 and $t = \delta t$. Solving Eqs. (8)-(11) requires the values of the small signal gain α_0 , the ratio between the incoming reference intensity and saturation intensity

 I_{R0}/I_{sat} and the response time τ_R . The first two are estimated using the experimental measurements whereas τ_R is adjusted to best fit the experimental data. Figure 3 shows the experimental data (dots) and the calculated gain (solid line) for several value of I_{R0}/I_{sat} . In order to have a good agreement between the experimental and the calculated data, we had to introduce two corrections. First the small signal gain α_0 , which was estimated by measuring the gain without the reference beam, had to be replaced by an effective gain equal to 0.75 α_0 . After this gain correction, there still was an offset difference between the calculated data and the experimental data which has been corrected to fit the temporal behavior.



Fig. 3. Experimental (dots) and calculated (solid lines) gain of a small signal beam for several value of the ratio between the intensity of the reference and saturation intensity

As shown in Fig. 3, after these corrections, the model predicts justly the temporal evolution of the gain hologram. Several observations can be made on this figure. Firstly, the effects of the saturation can be observed, the initial gain on the signal beam decreases as the intensity of the reference beam increases. Secondly, the effect of the wave mixing and the writing of the hologram can be seen in the temporal behavior. Over time, the gain on the signal beam decreases to reach a stationary value when the static hologram is recorded inside the medium. The calculation has been made with a value of $\tau_R = 50 \ \mu s$ which gives a good estimation of the temporal dynamic of the holographic recording. The magnitude of τ_R will be confirmed in section 5 where we experimentally measure the response time of our setup by using a method proposed by Lesaffre *et al.* [18].

The corrections on the calculated data to fit the experimental signals can be justified by the different conditions between the model and the experiment. The model supposes a perfect overlap between the signal, the reference and the pump beams and also a uniform pumping inside the crystal; whereas in reality, the signal and reference beams are not parallel, and also the pump is not uniform as it is absorbed by the crystal as described in section 3. Nevertheless, after introduction of correcting factors, this model shows a good agreement with the experimental data.

5. Dynamic of the TWM in Nd:YVO₄

In this part we experimentally investigate the response time by using a method proposed in [18]. It allows to directly measure *in situ* the response time of a self-adaptive wavefront holography

setup using the scattering sample and US. We used the method explained in part 4, we sent on our sample an amplitude modulated ultrasonic excitation and recorded the temporal signal at the exit of the holographic system for several frequency shifts between the signal and the reference beams. Then the amplitude of the signal at the modulation frequency is retrieved by detecting the peak on the Fourier spectrum. The method requires the modulation frequency to be large compared to the inverse of the response time τ_R , as we know that τ_R is about 50 μs ($\tau_R^{-1} = 20 \text{ kHz}$) we chose to do the experiment with a modulation frequency $f_{mod} = 200\text{ kHz}$. The US are generated by a 25.4 mm diameter Olympus focused single element transducer (model A307S) with a focal length of 51.5 mm and a center frequency $f_{US} = 4.8MHz$. Figure 4 shows the amplitude of the peak at $f_{mod} = 200\text{ kHz}$ versus the frequency shift (Δf) between the signal and the reference beams.



Fig. 4. Amplitude of the component of the acousto-optic signal at $f_{mod} = 200kHz$ vs. the frequency shift between the signal and the reference beam (dots) and lorentzian fit (solid line).

In order to estimate τ_R , the experimental points have been fitted by a Lorentzian model: $A(\Delta f) = \frac{A_0}{1+(2\pi\Delta f\tau_R)^2}$. The fit gives $\tau_R = 44 \ \mu s \pm 6 \ \mu s$ with a coefficient of determination $r^2 = 0.91$. This result confirms the previous observations with the model that lead to a response time of 50 μs .

6. Acousto-optic imaging using holography in Nd:YVO₄

This holographic detection is used to perform acousto-optic imaging of a multiple-scattering sample. In order to test this detection method, we put a sample into a water tank on the signal beam. An optical system (OS1, see Fig. 5(a)) is used to collect the scattered light and focus it inside the crystal within the pumped volume. After the crystal, light is collected by OS2 and focused on the photodiode (Thorlabs DET10A). A simplified drawing of the setup is shown in Fig. 5(a). In this setup, we chose to detect the untagged photons [6] thus the reference beam is not shifted from the US frequency. The pumping conditions are the same as the one described in section 3. The sample used is a 1.5 cm thick scattering phantom made of agar and zinc oxide particle for scattering ($\mu_s^* = 3 \text{ cm}^{-1}$). In this gel we put three circular black inclusions made with Indian ink and agar with a diameter of 1 mm, 2 mm and 3 mm. A picture of the inclusions before being covered with a layer of scattering gel is shown in Fig. 5(b). The sample is placed inside a water tank (not shown in Fig. 5(a)) and placed under the same ultrasonic transducer as the one used in part 5. It sends short bursts (4 cycles) at $f_{US} = 4.8 \text{ MHz}$ giving a resolution of 650 μm laterally and 1.2 mm axially. The transducer is aligned on the x axis in order to be al-



Fig. 5. (a)Simplified setup for the acousto-optic imaging of a sample. OSi are optical system to collect light, HWi are half-wave plate to change the ratio between the intensities of the reference and signal beams and correct the polarization of each beams, AOM is an acousto-optic modulator and US is an ultrasonic transducer. (b)Drawing of the sample with the inclusions (left) and picture (right) of the sample before being covered with another layer of scattering gel.

ways above the laser and on the y axis so that its focal spot is at the same height as the incoming laser on the sample. To get a large image, the sample is moved along the x and y axes by two PI-Micos Pollux Drive translation stages (not shown in Fig. 5(a)) while the incoming laser and the US stay aligned. The displacement step along the x and y directions are respectively 0.1 mm and 0.75 mm. The photodiode signal is first amplified by a large bandwidth amplifier (FEMTO



Fig. 6. Acousto-optic image of the sample obtained by two-wave mixing detection in Nd:YVO₄ at 1064 nm (left) and profile along the black dashed line (right)

DHPVA), then filtered by a KROHN-HITE 3945 Dual Low-Pass Programmable Butterworth Filter. It is adjusted to operate as a high-pass filter (2kHz cutoff frequency) and a low-pass filter (2MHz cutoff frequency). The signal is then amplified a second time by another FEMTO DHPVA before being sampled at 40MHz by an ADLINK-PCI9646D acquisition board in the computer driven by MatLab^(T) and averaged 3000 times for each position. The whole acquisition took a total time of three hours for this proof of concept. The reconstructed image as well as a profile along the y axis is shown on Fig. 6. The reconstruction algorithm simply concatenates all the signals measured for each position and takes into account the overlapping of the different signals because the displacement step is smaller than the diffuse light pattern inside the sample. The three inclusions inside the gel are clearly visible on the acousto-optic image. The full width at half maximum (FWHM) of the inclusions measured on the profile are respectively 0.9 mm, 1.9 mm and 2.7 mm. These values are in agreement with their real size.

7. Conclusion

As outlined in the article, adaptive wavefront holography is a fast process in gain media such as Nd:YVO₄. The response time is a main advantage for ultrasound optical tomography imaging of biological tissues due to the problems of speckle decorrelation that arises when performing in vivo experiments. We have shown through a model and measurements that our setup, with a neodymium doped vanadate crystal, has a response time around 50 μ s. This value was confirmed by a method for in situ monitoring of the response time in a self-adaptive wavefront holography experiment. We then used our setup to perform the detection of an acousto-optic signal from a multiple-scattering phantom in order to map the local optical properties inside a sample. We were able to detect inclusions as small as one mm using US at 4.8 MHz. The acquisition time is still too long because of the need for a lot of averaging due to a low SNR. For future experiments, improving the SNR is a necessity to make data acquisition faster (less averaging) on samples with scattering properties closer to biological tissue ($\mu_s^* = 10 \ cm^{-1}$). These experiments show that gain media such as Nd:YVO₄ are attractive candidates for interferometry with speckle beams with application such as in vivo UOT, where the speckle decorrelates rapidly, or for metrology of moving or vibrating rough surfaces. Even if we focused on the use of two-wave mixing for acousto-optic imaging, other application based on four-wave mixing (phase conjugation) can also be considered as we demonstrated last year (see [15]). For example, a technique such as TRUE focusing could benefit from the speed of gain media to overcome the speckle decorrelation problems inherent to in vivo experiments. The working wavelength, 1064 nm, is at the edge of the optical therapeutic windows so it does not penetrate very deep inside biological tissues in comparison to light at 800 nm. Therefore, extending these experiments to gain media operating in the 700 nm - 900 nm part of the spectrum, such as Ti:Sapphire, Alexandrite or Cr:LiSAF, would be attractive for further in vivo biomedical applications.

Appendix : Analytical solution for the signal exiting the gain medium

This appendix details the calculation to go from the system of Eqs. (10) and (11) to the analytical solution given in Eq. (12). As mentioned in section 2, if we take the Laplace Transform of Eqs. (10) and (11), we obtain:

$$\left(\widetilde{s\alpha^{(1)}}(z,s) - \alpha^{(1)}(z,0)\right) + \frac{1}{\tau} \left(1 + \frac{I_R}{I_{sat}}\right) \widetilde{\alpha^{(1)}}(z,s) = -\frac{2}{\tau} \left(\frac{\widetilde{A_S}(z,s)A_R^*(z)}{I_{sat}}\right) \alpha^{(0)}(z) \quad (14)$$

$$\frac{\partial \widetilde{A}_1(z,s)}{\partial z} = \frac{\alpha^{(0)}(z)}{2} \widetilde{A}_S(z,s) + \frac{\alpha^{(\overline{1})}(z,s)}{4} A_R(z)$$
(15)

Considering that at t = 0 there is no interference pattern, we can write $\alpha^{(1)}(z, 0) = 0$ so Eq. (14) can be simplified:

$$\widetilde{\alpha^{(1)}}(z,s) = -\frac{2}{\tau} \frac{A_R^*(z)}{I_{sat}} \alpha^{(0)}(z) \frac{\widetilde{A_S}(z,s)}{s + \frac{1}{\tau} \left(1 + \frac{I_R(z)}{I_{sat}}\right)}$$
(16)

Then inserting Eq. (16) into Eq. (15) yields:

$$\frac{\partial \widetilde{A}_{S}(z,s)}{\widetilde{A}_{S}(z,s)} = \frac{\alpha^{(0)}(z)}{2} dz - \frac{\alpha^{(0)}(z)}{2\tau} \frac{I_{R}(z)}{I_{sat}} \frac{1}{s + \frac{1}{\tau} \left(1 + \frac{I_{R}(z)}{I_{sat}}\right)} dz \tag{17}$$

Which can be rewritten:

$$\frac{\partial \widetilde{A}_{S}(z,s)}{\widetilde{A}_{S}(z,s)} = \frac{\alpha^{(0)}(z)}{2} dz - \frac{1}{2} \frac{\frac{\partial}{\partial z} \left[s + \frac{1}{\tau} \left(1 + \frac{I_{R}(z)}{I_{sat}}\right)\right]}{s + \frac{1}{\tau} \left(1 + \frac{I_{R}(z)}{I_{sat}}\right)} dz$$
(18)

Equation (18) can then be integrated between 0 and z:

$$\ln\left(\widetilde{A}_{S}(z,s)\right) = \frac{1}{2} \int_{0}^{z} \alpha^{(0)}(z') dz' - \frac{1}{2} \ln\left[s + \frac{1}{\tau}\left(1 + \frac{I_{R}(z)}{I_{sat}}\right)\right] + K_{1}(s)$$
(19)

$$\widetilde{A_1}(z,s) = K_2(s) \exp\left[\frac{1}{2} \int_0^z \alpha^{(0)}(z') dz'\right] \left[s + \frac{1}{\tau} \left(1 + \frac{I_R(z)}{I_{sat}}\right)\right]^{-\frac{1}{2}}$$
(20)

The boundary conditions for z = 0 gives: $\widetilde{A_1}(0,s) = K_2(s) \left[s + \frac{1}{\tau} \left(1 + \frac{I_R(0)}{I_{sat}}\right)\right]^{-\frac{1}{2}}$ which when inserted into (20) yields:

$$\widetilde{A}_{S}(z,s) = \widetilde{A}_{S}(0,s) \exp\left[\frac{1}{2} \int_{0}^{z} \alpha^{(0)}(z') dz'\right] \left[\frac{s + \frac{1}{\tau} \left(1 + \frac{I_{R}(0)}{I_{sat}}\right)}{s + \frac{1}{\tau} \left(1 + \frac{I_{R}(z)}{I_{sat}}\right)}\right]^{\frac{1}{2}}$$

$$= \widetilde{A}_{S}(0,s) \exp\left[\frac{1}{2} \int_{0}^{z} \alpha^{(0)}(z') dz'\right] \left[1 - \frac{\frac{I_{R}(z) - I_{R}(0)}{\tau I_{sat}}}{s + \frac{1}{\tau} \left(1 + \frac{I_{R}(z)}{I_{sat}}\right)}\right]^{\frac{1}{2}}$$

$$= \widetilde{A}_{S}(0,s) \exp\left[\frac{1}{2} \int_{0}^{z} \alpha^{(0)}(z') dz'\right] \left[1 - \frac{P}{Q}\right]^{\frac{1}{2}}$$
(21)

With:

$$P = \frac{I_R(z) - I_R(0)}{\tau I_{sat}}$$
$$Q = s + \frac{1}{\tau} \left(1 + \frac{I_R(z)}{I_{sat}} \right)$$
$$I_R(z) = I_R(0) e^{\int_0^z \alpha^{(0)}(z') dz'} \text{ (see Eq. (9))}$$

As we have R = P/Q < 1, Eq. (21) can be expanded as a power series:

$$\widetilde{A}_{S}(z,s) = \widetilde{A}_{S}(0,s) \exp\left[\frac{1}{2} \int_{0}^{z} \alpha^{(0)}(z') dz'\right] \sum_{n=0}^{\infty} (-1)^{n} \frac{\Gamma(1/2+1)}{\Gamma(n+1)\Gamma(1/2-n+1)} \left(\frac{P}{Q}\right)^{n}$$
(22)

The temporal evolution of the signal field is given by the inverse Laplace transform of Eq. (22). In this formula, the only term that has a dependency in the variable *s* is *Q*. This means that we have to find the inverse Laplace transform of $1/Q^n$. For $n \ge 1$:

$$TL^{-1}\left(\frac{1}{Q^n}\right) = TL^{-1}\left(\frac{1}{\left(s + \frac{1}{\tau}\left[1 + \frac{I_R(z)}{I_{sat}}\right]\right)^n}\right) = \exp\left[-\frac{t}{\tau}\left(1 + \frac{I_R(z)}{I_{sat}}\right)\right]\frac{t^{n-1}}{\Gamma(n)}$$

And for n = 0:

$$TL^{-1}\left(\frac{1}{Q^0}\right) = TL^{-1}(1) = \delta(t)$$
 where δ is the Dirac function

We can now take the inverse Laplace transform of Eq. (22) and have the temporal evolution of the signal beam:

$$A_{S}(z,t) = A_{S}(0,t) \exp\left[\frac{1}{2} \int_{0}^{z} \alpha^{(0)}(z') dz'\right] * \left\{\delta(t) + \exp\left[-\frac{t}{\tau} \left(1 + \frac{I_{R}(z)}{I_{sat}}\right)\right] \sum (-1)^{n} P^{n} \frac{\Gamma(1/2+1)}{\Gamma(n+1)\Gamma(1/2-n+1)} \frac{t^{n-1}}{\Gamma(n)}\right\}$$
(23)

Where f * g is the "incomplete" convolution operator define in section 2. This equation can be rearranged:

$$A_{S}(z,t) = \exp\left(\frac{1}{2}\int_{0}^{z} \alpha^{(0)}(z')dz'\right) \times \left[A_{S}(0,t) + A_{S}(0,t) * C(z,t)\right]$$
(24)

Where C(z,t) is defined by:

$$C(z,t) = \frac{1}{\tau} \exp\left[-\frac{t}{\tau} \left(1 + \frac{I_R(z)}{I_{sat}}\right)\right] \times \sum_{n=1}^{\infty} \left[(-1)^n \left(\frac{I_R(z) - I_R(0)}{I_{sat}}\right)^n \frac{\Gamma(1/2+1)}{\Gamma(n+1)\Gamma(1/2-n+1)} \frac{\left(\frac{t}{\tau}\right)^{n-1}}{\Gamma(n)}\right]$$

Acknowledgment

The authors gratefully acknowledge the *Direction générale de l'Armement*, the *Agence Nationale de la Recherche* under the project ICLM-ANR-2011-BS04-017-01, and finally the *LABEX WIFI (Laboratory of Excellence with the French Program "Investments for the future")* under the references ANR-10-LABX-24 and ANR-10-IDEX-0001-02 PSL* for financial support.

APPENDIX B

Version courte (FR)

L es techniques d'imagerie médicale sont couramment utilisées par les médecins pour arriver à des diagnostics de plus en plus précis. Que ce soit l'échographie, la radiographie, l'IRM, etc, toutes apportent une information différente. C'est cette multiplicité des informations fournies par l'imagerie médicale qui permet des diagnostics précis d'une large gamme de pathologies. Malgré cela, il est toujours impossible pour les médecins et les biologistes d'avoir accès à l'information optique en profondeur dans les tissus biologiques.

L'optique biomédicale peut avoir un grand intérêt. En effet, comme les interactions lumière/matière se font à l'échelle atomique et moléculaire, la lumière peut donc fournir des informations importantes comme la composition chimique. Le principal problème des milieux biologiques, qui rend impossible toute imagerie en profondeur, est la forte diffusion de la lumière. Néanmoins, en couplant la lumière avec les ultrasons, qui se propage bien aux fréquences de l'ordre du mégahertz, il est possible de cartographier en profondeur les propriétés optiques des tissus biologiques. C'est le principe de deux nouvelles modalités d'imagerie : l'imagerie acousto-optique et l'imagerie photoacoustique.

Cette thèse présente l'étude de l'utilisation d'un milieu à gain, le Nd:YVO₄, comme milieu holographique pour la détection de signaux acousto-optique et photoacoustique. Ce résumé est divisé en quatre sections. La première section est consacrée à l'intérêt de l'optique dans les milieux biologiques ainsi que les difficultés posées par la forte diffusion de la lumière. La deuxième section décrit les processus gouvernant le mélange d'ondes et l'holographie dans un milieu à gain comme Nd:YVO₄. Enfin les sections 3 et 4 parlent respectivement de l'imagerie acousto-optique et de l'imagerie photoacoustique. Dans chacune de ces sections, on présentera brièvement les montages d'imagerie ainsi que les principaux résultats obtenues. Enfin la thèse se termine sur une discussion des principaux résultats et des perspectives de ces travaux.

B.1 L'optique dans les milieux biologiques.

Dans cette section on va décrire comment la lumière se propage dans les milieux biologiques ainsi que les grandeurs qui caractérisent cette propagation. On parlera ensuite des deux méthodes d'imagerie optique de ces milieux qui seront utilisées dans cette thèse : l'imagerie acousto-optique et l'imagerie photoacoustique.

B.1.1 Propagation de la lumière dans les milieux biologiques.

Lorsqu'elle traverse la matière, la lumière interagit avec ses constituants de deux manières. Elle est soit absorbée, soit diffusée.

L'absorption correspond à la disparition des photons, l'énergie ainsi absorbée est rendue soit sous forme non radiative (ex : chaleur), soit sous forme radiative (ex : luminescence). La capacité d'un milieu à absorber la lumière est caractérisée par le coefficient d'absorption μ_a qui relie le flux d'un faisceau collimaté au flux incident et à la distance parcourue. La loi de Beer-Lambert (en l'absence de diffusion) donne :

$$\Phi(z) = \Phi_0 e^{-\mu_a z} \tag{B.1}$$

Dans les milieux biologiques, l'absorption est très variable dans le spectre visible. Afin de travailler en profondeur il est nécessaire d'utiliser des longueurs d'ondes dans la *fenêtre thérapeutique optique* qui correspond au rouge/proche infrarouge (entre 600 nm et 1300 nm) où l'absorption est autour de 1 cm^{-1} [1]. Les longueurs d'ondes plus courtes sont fortement absorbées par l'hémoglobine alors que les longueurs d'ondes plus grandes sont absorbées par l'eau.

La diffusion, quant à elle, correspond à une perturbation de la trajectoire des photons qui n'est plus rectiligne. Elle est caractérisée par deux grandeurs :

- Le coefficient de diffusion μ_s , qui correspond au nombre d'évènements de diffusion par unité de longueur. Dans les milieux biologiques, on a : $\mu_s \approx 50 \,\mathrm{cm}^{-1} - 200 \,\mathrm{cm}^{-1}$ [2]
- Le coefficient d'anisotropie g, qui caractérise la direction majoritaire dans laquelle se fait la diffusion. Il est défini comme $g = \langle \cos \theta \rangle$ où θ est l'angle de diffusion par rapport à la direction initial, c'est une variable aléatoire. Dans les milieux biologiques la diffusion est très anisotrope et on a $g \approx 0.8 - 0.99$ [2]

La diminution du flux **balistique** d'un faisceau collimaté en présence de diffusion (et sans absorption) est donnée par :

$$\Phi_{bal} = \Phi_0 e^{-\mu_s z} \tag{B.2}$$

Néanmoins, comme dans les milieux biologiques la diffusion est très anisotrope et se fait majoritairement vers l'avant, il est plus intéressant d'utiliser le coefficient de diffusion réduit qui caractérise le nombre de photons qui "perdent la mémoire" de leur direction initial et de leur polarisation par unité de longueur :

$$\mu'_s = \mu_s (1 - g) \tag{B.3}$$

Il est d'environ $\mu'_s \approx 10 \text{ cm}^{-1}$ dans les milieux biologiques. Il peut être plus commode de parler en libre parcours moyen de diffusion, défini comme l'inverse du coefficient de diffusion : $l_s = 1/\mu_s$. C'est une distance qui représente la distance moyenne que parcours un photon entre chaque évènements de diffusion. De même on peut définir le libre parcours moyen de transport comme l'inverse du coefficient de diffusion réduit : $l'_s = 1/\mu'_s$. Le libre parcours moyen de transport représente la distance moyenne à partir de laquelle un photon ayant subi des diffusions multiples à perdu la mémoire de sa direction initiale. Une représentation de ces différentes grandeurs, et le trajet des photons dans un milieu multiplement diffusant est donné dans la figure B.1.



Figure B.1 – Trajet des photons dans un milieu multiplement diffusant. Différence entre libre parcours moyen de diffusion l_s et libre parcours moyen de transport l'_s .

B.1.2 Imagerie optique des milieux biologiques - Intérêts et problèmes.

L'imagerie optique des milieux biologiques peut fournir de nouvelles informations inaccessibles auxquelles les médecins et le biologistes n'ont pas actuellement accès. Les nouvelles informations fournis par l'optique peut améliorer le diagnostic. Par exemple, Laudereau *et al.* ont montrés qu'il était possible de détecter des tumeurs à la surface du foie en utilisant l'imagerie acousto-optique. Un autre exemple est l'expérience de Pogue *et al.*. En utilisant la technique de Tomographie Optique Diffuse, ils ont imagé la concentration en hémoglobine ce qui a permis de détecter des tumeurs dans le sein [3].

Un autre intérêt de l'utilisation de l'optique est le fait de pouvoir faire la différence entre l'hémoglobine oxygénée et l'hémoglobine désoxygénée en utilisant plusieurs longueurs d'ondes. On peut ainsi théoriquement cartographier la saturation en oxygène du sang, qui est un paramètre important pour les médecins.

Comme expliqué au paragraphe précédent, les milieux biologiques sont très diffusants. Le libre parcours moyen de transport est autour de $l'_s = 1$ mm. Cela signifie qu'au delà de quelques millimètres, il n'y a pratiquement plus de lumière balistique, nécessaire pour faire de l'imagerie au sens classique. Il faut donc trouver d'autres stratégies afin de récupérer l'information optique en profondeur dans les tissus biologiques. De nombreuses méthodes existent. Dans cette thèse on va particulièrement développer les méthodes qui exploitent les effets acousto-optique et photoacoustique.

B.1.3 Imageries acousto-optique et photoacoustique.

Imagerie acousto-optique.

Modulation acousto-optique L'imagerie acousto-optique (AO) est basé sur la modulation de la phase de la lumière par une onde acoustique. Dans les milieux diffusants, comme les tissus biologiques, cette modulation à deux origines (cf Fig. B.2.(a))

- La modulation de l'indice de réfraction : Comme l'onde acoustique est une onde de pression, la densité locale du milieu dans lequel elle se propage est aussi modulée. Il en résulte une modulation de l'indice de réfraction du milieu, les zones de forte pression ont une densité plus grande et donc un indice de réfraction plus fort et inversement.
- La vibration des diffuseurs : Lors de la propagation d'une onde acoustique, les constituants de la matière vibrent autour d'une position d'équilibre. Par conséquent les photons qui traversent le champ acoustique sont diffusés par des diffuseurs vibrants à la fréquence acoustique. Le chemin optique des photons est donc modulé ce qui provoque une modulation de phase de la lumière.

La modulation de phase par l'onde acoustique se traduit sur le spectre de la lumière (à ω_L) traversant un milieu diffusant en présence d'une onde acoustique (à ω_{US}) par des bandes latérales à $\omega = \omega_L + n\omega_{US}$ ($n \in \mathbb{Z}$) comme le montre la figure B.2.(b) (On négligera les bandes pour $|n| \geq 2$). Les photons des bandes latérales à $\omega_L \pm \omega_{US}$ sont appelés *photons marqués* et inversement les photons à ω_L sont les *photons non marqués*. Dans les tissus biologiques, les fréquences acoustiques qui se propagent bien, et permettent une bonne résolution d'imagerie (autour du millimètre), sont les fréquences ultra-sonore, de l'ordre du mégahertz (couramment utilisées en échographie). A ces fréquences, le décalage en fréquence sur la lumière créé par les ultrasons correspond à un décalage en longueur d'onde de l'ordre de 10^{-5} nm (pour de la lumière à $\lambda = 1$ µm).



Figure B.2 – (a) Modulation acousto-optique dans un milieu multiplement diffusant (b) Spectre de sortie de la lumière.

Détection du signal acousto-optique La première étape pour faire de l'imagerie acousto-optique est d'avoir un système de détection des photons marqués avec un bon rapport signal à bruit. En réalité, comme la quantité de photons est conservée, l'information

acousto-optique peut aussi être récupérée en détectant les photons non marqués. La détection de si faible décalage en fréquence peut se faire en utilisant l'interférométrie. Ces techniques nécessitent une lumière cohérente et sont efficaces tant que l'on conserve une cohérence spatiale. Or dans des milieux très diffusants comme les milieux biologiques, la cohérence spatiale est très vite perdue, le lumière en sortie est un speckle. Le rapport signal à bruit devient limité à celui d'un grain de speckle [24]. Il est possible de l'améliorer grandement en utilisant des méthodes d'adaptation de front d'onde par holographie dynamique comme illustré dans la figure B.3.(a). Dans le cadre de cette thèse, le montage holographique est basé sur un cristal à gain de Nd:YVO₄ dont les principes de bases sont expliqués au paragraphe B.2.



Figure B.3 – (a) Détection par adaptation de front d'onde des photons non marqués. (b) Détection par conjugaison de phase des photons non marqués.

Un autre moyen de détecter le signal acousto-optique est de "corriger" les aberrations dues à la diffusion multiple. Ceci peut être fait en utilisant le processus de conjugaison de phase qui consiste à créer le front d'onde conjugué en phase du front d'onde issu du milieu à imager. Ensuite, lors de la repropagation dans le milieu biologique, les retards locaux de phase sont entièrement compensés et l'onde initiale est recréée. Le marquage de la lumière par l'onde acoustique perturbe la reformation de l'onde initiale ce qui peut être utilisé pour détecter le signal acousto-optique (cf Fig. B.3.(b)). La conjugaison de phase peut aussi être utilisée pour focaliser dans le milieu biologique [65, 77].

Imagerie La quantité de photons marqués est proportionnelle à l'intensité locale de la lumière dans le milieu. Celle-ci est plus faible dans une zone absorbante, il est donc possible de détecter des absorbeurs en localisant les ultrasons dans un petit volume. Comme la propagation ultra-sonore se fait de manière balistique, il est facile de focaliser l'onde acoustique, ce qui garantit une bonne résolution dans les axes perpendiculaires (axes X et Z) à sa direction de propagation. Afin d'avoir une bonne résolution sur l'axe Y, une solution consiste à limiter l'extension spatiale de l'onde acoustique en travaillant avec des salves. En effet, une salve courte dans le temps a une extension spatiale faible donnée par : $dy = N_c v_{US}/f_{US}$ où N_c est le nombre de cycles de la salve, v_{US} est la vitesse de propagation de l'onde acoustique dans le milieu et f_{US} sa fréquence. Si la salve conserve une faible extension spatiale selon X et Z suffisamment longtemps, il suffit de suivre sa propagation dans la zone de lumière pour acquérir le signal acousto-optique sur une ligne entière de l'image. Ensuite il faut balayer le transducteur US dans une direction pour avoir une image dans un plan comme le montre la figure B.4.



Figure B.4 – Imagerie acousto-optique.

Les travaux décrits dans cette thèse utilisent des salves pour avoir une bonne résolution selon Y. Néanmoins, il existe d'autres méthodes qui ne seront pas détaillées ici, basées sur une modulation de la phase ou de la fréquence des US. Plus d'informations peuvent être trouvées dans les références suivantes [28–31]. Le paragraphe B.3 décrit un peu plus le principe des deux méthodes de détection (adaptation de front d'onde et conjugaison de phase).

Imagerie photoacoustique.

De la lumière aux ultrasons L'imagerie photoacoustique (PA) est basée sur la génération d'une onde acoustique par la lumière [38]. Lorsqu'une impulsion laser très courte (quelques nanosecondes) se propageant dans une milieu diffusant rencontre une inclusion absorbante, celle-ci chauffe et se dilate brusquement provoquant l'émission d'une onde de pression : une onde acoustique. Plus de détails sont données ici [39].

Détection et imagerie L'imagerie photoacoustique repose sur ce phénomène. En effet, en détectant l'onde acoustique émise, il est possible de reconstruire la position et la forme des absorbeurs dans une milieu diffusant. La fréquence de l'onde émise est directement reliée à la taille et la forme de l'inclusion. Pour des inclusions ayant des tailles entre 100 µm et 1 mm, les fréquences acoustiques émises sont de quelques mégahertz.

La méthode classique de détection de l'onde photoacoustique est avec un transducteur piézoélectrique. En déplaçant le transducteur ou en utilisant une barrette, il est possible d'enregistrer le front d'onde acoustique dans un plan et ainsi reconstruire numériquement les émetteurs. Le principe de l'imagerie photoacoustique est résumé sur la figure B.5. Dans cette thèse nous proposerons une méthode de détection de l'onde acoustique 100% optique. Un des avantages d'utiliser l'optique est d'avoir une détection sans contact contrairement à l'utilisation de transducteur qui nécessite d'assurer un couplage entre le milieu biologique et la sonde. Le fonctionnement de notre détection optique est décrit au paragraphe B.4.



Figure B.5 – Imagerie photoacoustique.

B.2 Mélange d'onde et holographie dans Nd:YVO₄.

Dans ce paragraphe on va présenter l'outil qui va servir de base à la détection de signaux acousto-optiques et photoacoustiques : le mélange d'ondes dans un milieu à gain. Deux types de mélange d'ondes sont présentés, le mélange à deux ondes qui permet de faire de l'adaptation de front d'onde et le mélange à quatre ondes qui permet de faire de la conjugaison de phase.

B.2.1 Saturation du gain dans un laser et holographie.

Saturation du gain Les phénomènes de mélange d'ondes sont possible à travers la saturation du gain laser. Dans un milieu à gain pompé, la lumière est amplifiée par émission stimulée, l'intensité de la lumière dans le milieu à gain s'écrit :

$$I(z) = I_0 e^{\alpha z} \text{ avec } \alpha = \frac{\alpha_0}{1 + \frac{I}{I_{sat}}}$$
(B.4)

Où α_0 est le gain petit signal et I_{sat} est l'intensité de saturation. Avec cette équation on voit que tant que l'intensité lumineuse est faible devant l'intensité de saturation, le gain est constant et a la valeur du gain petit signal α_0 . Par contre pour des intensités lumineuses non négligeables devant I_{sat} , le gain chute. Il est possible d'utiliser ce phénomène de saturation de gain pour moduler spatialement le gain dans un milieu laser et ainsi enregistrer dynamiquement un hologramme.

Holographie Un hologramme est la figure d'interférence d'un faisceau signal E_S , dont on veut avoir les informations sur la phase, avec un faisceau référence E_R , dont la phase est connue (généralement une onde plane). Elle peut s'écrire :

$$I(x, y, z, t) \propto |A_S(x, y)e^{i\left(\omega t - \overrightarrow{k_S} \cdot \overrightarrow{\tau} + \phi_S\right)} + A_R(x, y)e^{i\left(\omega t - \overrightarrow{k_R} \cdot \overrightarrow{\tau} + \phi_R\right)}|^2$$

$$\propto |A_S(x, y)|^2 + |A_S(x, y)|^2 + 2|A_R||A_S|\cos\left(\Delta \overrightarrow{K} \cdot \overrightarrow{\tau} - \Delta \Phi\right)$$
(B.5)

Avec $\Delta \overrightarrow{K} = \overrightarrow{k_S} - \overrightarrow{k_R}$ et $\Delta \Phi = \phi_S - \phi_R$. Afin d'enregistrer cette figure d'interférence dans un hologramme de gain, il faut envoyer dans un milieu à gain pompé le faisceau signal et la référence afin de saturer le gain. Généralement, le faisceau référence est celui qu'on peut contrôler, il faut donc choisir son intensité afin que l'intensité moyenne de la figure d'interférence, $|A_S(x,y)|^2 + |A_S(x,y)|^2$, soit proche de I_{sat} . Dans le milieu que l'on va utiliser, Nd:YVO₄ avec un dopage à 1% en ions Nd³⁺, l'intensité de saturation est autour de $I_{sat} = 1 \text{ kW cm}^{-2}$.

En réalité, la modulation spatiale du gain laser dans un cristal de Nd:YVO₄ s'accompagne de la modulation spatiale de son indice de réfraction. Ce deuxième hologramme, qui est un hologramme d'indice, possède deux contributions : une contribution non résonante due aux bandes d'absorption UV et une contribution résonante due à la variation d'indice d'origine électronique (la polarisabilité des ions Nd³⁺ change lorsque les ions sont dans un état excité) [49,50]. Cette deuxième contribution est plus importante lorsque la longueur d'onde la lumière se propageant dans le milieu n'est pas la même que la longueur d'onde du maximum de gain du milieu. C'est notre cas puisque notre milieu à gain est Nd:YVO₄ ($\lambda_C = 1064.3$ nm) et notre laser est un Nd:YAG ($\lambda_0 = 1064.2$ nm). Dans nos expériences nous aurons donc deux réseaux inscrits dans le milieux : un réseau de gain et un réseau d'indice de réfraction. Le réseau d'indice de réfraction va surtout être important dans le cadre de la détection photoacoustique, puisque, comme on le verra au paragraphe B.4, il permet une détection linéaire qui est essentielle pour la reconstruction du signal photoacoustique.

B.2.2 Mélanges d'ondes dans Nd:YVO₄

Mélange à deux ondes Le mélange à deux ondes se produit lorsqu'on éclaire le milieu avec seulement le faisceau signal et un faisceau de référence (contrairement au mélange à quatre ondes qui nécessite deux références). La diffraction de la référence sur les hologrammes inscrits par saturation du gain génère un faisceau ayant les mêmes caractéristiques (direction de propagation et front d'onde) que le faisceau signal. Un schéma de principe de l'adaptation de front d'onde par mélange à deux ondes est représenté sur la figure B.6.(a). Sous certaines conditions, le champ électrique en sortie du cristal peut s'écrire :

$$E_{S}(z,t) = \underbrace{e^{\frac{1}{2} \int_{0}^{z} \alpha^{(0)}(z')dz'}}_{\text{Amplification par}} \left(\underbrace{E_{S}(0,t) + E_{S}(0,t) * C'_{n}(z,t)}_{\text{Signal diffracté sur}} + \underbrace{E_{S}(0,t) * C'_{n}(z,t)}_{\text{Signal diffracté sur}} + \underbrace{E_{S}(0,t) * C'_{n}(z,t)}_{\text{le réseau de gain}} \right)$$
(B.6)

Cette formule est commode pour comprendre l'effet du cristal (le détail des calculs ainsi que les conditions d'applications de cette formule se trouvent dans le chapitre 4 de cette thèse). On voit que le champ signal à la sortie du cristal est la somme de trois termes, chacun amplifié par un facteur $\exp\left(\frac{1}{2}\int_0^z \alpha^{(0)}(z')dz'\right)$:

- Le signal transmis : $E_S(0,t)$
- Le signal diffracté sur le réseau de gain et filtré par la "fonction de transfert" du cristal : $E_S(0,t) * C'_n(z,t)$
- Le signal diffracté sur le réseau d'indice et filtré par la "fonction de transfert" du cristal : $i\beta E_S(0,t) * C'_n(z,t)$. Cette contribution est déphasée de $\pi/2$ par rapport au champ diffracté sur le réseau de gain.

Une expression simplifiée de la fonction de transfert est :

$$C'_{n}(z,t) = -\frac{I_{R}(z) - I_{R}(0)}{2\tau I_{sat}} e^{-\frac{t}{\tau} \left(1 + \frac{I_{R}(z)}{I_{sat}}\right)}$$
(B.7)

Cette formule fait apparaître un paramètre important de l'holographie dans Nd:YVO₄, à savoir son temps de réponse $\tau_R = \frac{\tau}{1+\frac{I_R}{I_{sat}}}$ où τ est le temps de vie de fluorescence du niveau excité. Ce temps de réponse est très rapide puisqu'il est inférieur à τ qui est de l'ordre de 100 µs (pour notre cristal).

Pour comprendre l'intérêt de cette dynamique, il faut d'abord examiner le fonctionnement de la détection par mélange à deux ondes. Supposons que le champ électrique signal incident a simplement une modulation de phase $\phi(t)$: $E_S(0, t) = e^{i\phi(t)}$. L'intensité à la sortie du cristal (z = L) peut alors s'écrire:

$$I_S(L,t) \propto G |e^{i\phi(t)} + e^{i\phi(t)} * C'_n(L,t) + i\beta e^{i\phi(t)} * C'_n(L,t)|^2$$
(B.8)

où G est le facteur d'amplification. Comme le temps de réponse du cristal est de l'ordre de quelques dizaines de microsecondes, les composantes diffractées sur les réseaux d'indice et de gain ne portent pas l'information de la modulation du phase si celle-ci a une fréquence suffisamment élevée ($f_{mod} \gg 1/\tau_R$). Les termes $e^{i\phi(t)} * C'_n(L,t)$ s'écrivent alors sous la forme d'une constante du temps que l'on notera $\langle e^{i\phi(t)} \rangle_{\tau_R}$. L'intensité à la sortie du cristal s'écrit alors :

$$I_S(L,t) \propto G |e^{i\phi(t)} + \left\langle e^{i\phi(t)} \right\rangle_{\tau_R} + i\beta \left\langle e^{i\phi(t)} \right\rangle_{\tau_R} |^2$$
(B.9)

 $I_S(L,t)$ possède bien une dépendance temporelle qui aurait disparu avec l'opération $|\bullet|^2$ si les informations de la modulation de phase étaient aussi présentes dans les composantes diffractées. Le cristal de Nd:YVO₄ est donc capable d'enregistrer et restituer les modulations temporelles de phase lente ($f_{mod} \ll 1/\tau_R$), or il a été mesuré par Gross *et al.* que le speckle issu d'un milieu biologique se décorrèle en environ 1 ms à cause des mouvements naturels (respiration, battements cardiaques,...) [37]. Notre temps de réponse inférieur à 100 µs permet donc de s'adapter à ces variations lentes et ainsi de les éliminer. Ce matériau est donc intéressant pour les applications biologiques.



Figure B.6 – (a) Principe du mélange à deux ondes. (b) Principe du mélange à quatre ondes.

Mélange à quatre ondes Le mélange à quatre ondes fait intervenir deux faisceaux références. Comme le montre la figure B.6.(b), cette fois l'hologramme, enregistré par l'interférence entre le faisceau signal et le faisceau référence 1, est relu par une deuxième

référence contra-propageante avec la référence 1. Pour fabriquer un faisceau conjugué en phase du signal, il faut que la référence 2 soit elle même le conjugué en phase de la référence 1. Or comme celle-ci est une onde plane, la référence 2 peut être générée soit par réflexion sur un miroir plan, soit par simple séparation avec un beamsplitter. La diffraction de la deuxième référence sur l'hologramme inscrit dans le cristal génère un quatrième faisceau qui est le conjugué en phase du faisceau signal.

Une modélisation des interactions entre les quatre ondes dans un milieu à gain peut être trouvée au dernier chapitre de *Phase Conjugate Laser Optics* [58]. Le modèle présenté dans ce livre est une modèle d'ondes couplées, c'est à dire que l'évolution des champs est régie par un système de quatre équations différentielles couplées faisant intervenir les champs dans chacune d'elles à travers des constantes de couplages. Une étude basée sur ce modèle du régime stationnaire peut être trouvé dans le chapitre 4 de cette thèse.

B.3 Détection AO par mélange à deux et quatre ondes.

Cette partie est consacrée à l'utilisation de l'holographie dans $Nd:YVO_4$ pour la première technique d'imagerie décrite au paragraphe précédent, l'imagerie acousto-optique. Dans un premier temps on va décrire la détection par conjugaison de phase, puis on verra comment le montage mis en place nous a amené à faire du mélange à deux ondes.

B.3.1 Détection AO par conjugaison de phase.

L'idée originale de ce travail de thèse était d'utiliser la conjugaison de phase dans $Nd:YVO_4$ pour faire de la détection de signaux acousto-optique avec un temps de réponse inférieur à la milliseconde.

Montage expérimentale Après quelques expériences préliminaires que nous ne décrirons pas ici, le montage schématisé sur la figure B.7 a été mis en place afin de faire une image acousto-optique d'un échantillon multiplement diffusant.

La partie supérieure de ce montage, constituée par trois cubes séparateur de polarisation (PBS1 à PBS3) et cinq lames demi ondes (HW1 à HW5), est utilisée pour séparer le faisceau source en deux bras (signal et référence) puis régler indépendamment l'intensité de chaque bras et changer le rapport d'intensité entre les bras. Chaque bras est équipé de modulateurs acousto-optique qui sont utilisés afin de couper rapidement les faisceaux ou alors pour mettre un décalage en fréquence suffisant afin de ne plus avoir de mélange d'onde pour enregistrer le signal d'offset. Le faisceau référence est légèrement focalisé dans le cristal sur le traitement haute réflectivité de manière à générer la deuxième référence pour faire la conjugaison de phase. Le faisceau signal est envoyé sur l'échantillon qui est placé dans une cuve d'eau afin d'assurer le couplage acoustique avec le transducteur (qui fonctionne à $f_{US} = 2.25$ MHz). La lumière diffuse issue de l'échantillon est récoltée et focalisée dans le cristal par une lentille de focale f' = 5 cm en configuration 2f - 2f. Le faisceau conjugué en phase repart dans l'échantillon pour reformer l'onde plane initiale



Figure B.7 – Schéma du montage de détection acousto-optique par conjugaison de phase. HWi sont des lames demi-onde, PBSi sont des cube séparateur de polarisation, AOMi sont des modulateurs acousto-optique, NPBS est un cubes séparateur non polarisant, PD est une photodiode, PZT est un transducteur piézoélectrique, et HR et AR sont des traitements Haute Réflectivité et Anti Reflets.

qui est séparée du faisceau signal par un cube séparateur d'intensité (NPBS) pour être détectée sur une photodiode (PD, modèle Thorlabs PDA36A-EC). Avant d'être numérisé, le signal issue de la photodiode est amplifié par une amplificateur de tension (FEMTO DHPVA) et filtré passe bas (fréquence de coupure de 1 MHz). Le signal filtré est ensuite enregistré directement sur un ordinateur par une carte d'acquisition fonctionnant à 40 MS/s (ADLINK PCI-9646D).

Le cristal, qui a le rôle de miroir à conjugaison de phase, est un cristal de Nd:YVO₄ dopé à 1% en ions Nd³⁺. Il a la forme d'un cube de 5 mm de coté. Deux faces opposées sont polies, la face avant est traitée anti-reflet pour 1064 nm afin de laisser la lumière entrer, et la face arrière est traitée haute réflectivité pour 1064 nm afin de générer la deuxième référence. En supplément, la face arrière est traité anti-reflet à 808 nm pour le faisceau de pompe. Celui-ci est issu d'une diode de haute puissance et arrive sur le cristal à travers une fibre multimode de 200 µm de cœur. Pour l'expérience d'imagerie la pompe fonctionne en régime quasi-continu, elle envoie une suite d'impulsions de durée 1 ms séparée par une pause de 19 ms. La puissance crête de chaque impulsion est autour de 100 W. Le cristal est placé dans une monture en cuivre refroidie par eau afin de dissiper la chaleur du pompage.
Temps de réponse Une estimation du temps de réponse du montage a été faite en ondes planes (sans échantillon) en regardant l'intensité du conjugué de phase en fonction du décalage en fréquence entre les deux modulateurs acousto-optique. Cette expérience préliminaire a donnée un temps de réponse de l'ordre de 15 µs. Cette rapidité, bien en dessous du temps de vie de fluorescence, est due au pompage très intense et au faisceau référence intense puisqu'il est lui même fortement amplifié par le cristal lors des deux passages (aller et retour) dans la zone de gain.

Image acousto-optique Afin de tester notre montage nous avons essayé de détecter un signal acousto-optique issu d'un milieu biologique. Pour cela, nous avons pris un morceau de blanc de poulet cru de 5 mm d'épaisseur qui a été placé dans une cuve sous un transducteur ultrasonore monté sur un moteur afin de pouvoir balayer l'échantillon à plusieurs positions. Pour chaque impulsion de la pompe, le transducteur envoie une impulsion ultrasonore de quatres cycles d'une sinusoïde à $f_{US} = 2.25$ MHz. L'enregistrement du trajet de l'impulsion dans le milieu diffusant permet d'obtenir le signal acousto-optique sur une ligne avec une résolution autour de 2 mm. Afin d'obtenir une image acousto-optique, le transducteur est déplacé par pas de 200 µm sur un intervalle total de 4 mm. L'image de la figure B.8.(a) est obtenue en moyennant chaque ligne à partir de 100 acquisitions. La partie (b) représente la trace temporelle du passage des ultrasons dans la tâche de lumière le long de la ligne pointillée blanche. La résolution transverse (selon X) est donnée par les caractéristiques du transducteur. Ici elle est autour de 1.5 mm.



Figure B.8 – Image acousto-optique d'un morceau de blanc de poulet de 5 mm d'épaisseur. (a) Image entière normalisée et lissée par convolution 2D. (b) Signal temporel le long de la ligne pointillée blanche sur (a).

On peut voir sur cette image que le diamètre de la tâche de lumière dans le milieu est autour de 3 mm. Cette mesure est cohérente avec la théorie de la diffusion multiple qui indique qu'en profondeur $(z \gg l^*)$, la taille de la tâche de lumière diffuse est de l'ordre de grandeur de la profondeur. Ici on est à peu près au milieu d'un échantillon de 5 mm d'épaisseur, on s'attend donc à une tâche de quelques millimètres de diamètre.

Conclusion Dans ce paragraphe nous avons démontré qu'il était possible d'utiliser la conjugaison de phase par mélange à quatre ondes dans $Nd:YVO_4$ pour détecter un signal

acousto-optique issue d'un milieu biologique et ainsi faire une image acousto-optique. Le montage réalisé possède un temps de réponse très court, autour de 15 µs, ce qui nous garantit une robustesse face à la décorrélation du speckle dans un milieu biologique. La lumière parasite, notamment celle issue de l'émission spontanée amplifiée, représente une source importante de bruit. Afin de la diminuer une autre configuration géométrique où tous les faisceaux sont perpendiculaires a été pensé. Cette nouvelle configuration a présentée des résultats intéressants en mélange à deux ondes ce qui nous a amené à faire un montage de détection acousto-optique par mélange à deux ondes.

B.3.2 Détection AO par adaptation de front d'onde.

Les expériences préliminaires, avec une géométrie où tous les faisceaux sont perpendiculaires, n'ont pas données de résultats satisfaisants, le gain était très faible dû à une faible longueur d'interaction entre le signal et la zone de gain. Nous avons donc décidé de se mettre dans une configuration où le signal et la référence sont pratiquement colinéaires avec un pompage perpendiculaire comme illustré sur le figure B.9.

Schéma du montage Le montage réalisé pour ces expériences est très similaire au montage précédent. La partie du montage qui change est celle concernant le cristal. Comme indiqué sur le zoom à droite du schéma complet du montage (cf. figure B.9), le cristal est un parallélépipède de $5 \times 5 \times 10 \text{ mm}^3$. Il est pompé par la même diode de pompe qui est cette fois focalisée par une lentille cylindrique pour avoir une grande longueur d'interaction entre la zone de gain et les faisceaux signal et référence. Ceux-ci



Figure B.9 – Schéma du montage de détection acousto-optique par mélange à deux ondes. HWi sont des lames demi-onde, PBSi sont des cube séparateur de polarisation, AOMi sont des modulateurs acousto-optique, PD est une photodiode, PZT est un transducteur piézoélectrique, et HR et AR sont des traitements Haute Réflectivité et Anti Reflets.

arrivent dans le cristal par une des petite faces et avec un angle (environ 10° pour la

référence et environ 20° pour le signal) pour avoir une réflexion totale interne sur le face pompée. Ils ressortent par la petite face opposée et sont détectés par une photodiode.

Pour ce montage les conditions de pompage ont été modifiées. Le cristal est maintenant pompé par des impulsions de 2.5 ms d'une durée de 80 W avec une cadence de répétition de 40 Hz. La détection a aussi été modifiée, elle est basée sur une photodiode plus rapide (Thorlabs DET10A). Le signal issu de la photodiode est amplifié une première fois, puis filtré passe-haut ($f_C = 2 \text{ kHz}$) et passe-bas ($f_C = 2 \text{ MHz}$) et enfin amplifié une dernière fois avant d'être enregistré sur l'ordinateur. La longue impulsion de la pompe permet d'envoyer plusieurs salves d'ultrasons dans une impulsion de pompe ce qui permet de moyenner plus rapidement.

Temps de réponse Le temps de réponse du montage dans cette configuration a été déterminé en utilisant une méthode inspirée de la technique de mesure décrite dans [90]. Cette méthode consiste à utiliser le milieu diffusant sur lequel on envoie la lumière en continu et des ultrasons modulés en intensité par une fonction créneau de fréquence f_{mod} . Ensuite, on observe l'intensité du signal à f_{mod} en fonction du décalage en fréquence entre signal et référence. La courbe obtenue peut être ajustée par une fonction Lorentzienne (fonction de transfert d'un filtre passe bas du premier ordre) et donne un temps de réponse de $\tau_R = 45 \,\mu$ s. Ce temps de réponse est un peu plus long que la mesure dans l'autre configuration. C'est probablement dû au pompage moins intense (focalisation dans une seule dimension) et à l'intensité plus faible de la référence.

Imagerie L'échantillon est un gel d'agar de 1.5 cm d'épaisseur avec des particules de ZnO pour assurer la diffusion. Le coefficient de diffusion réduit est estimé à $\mu'_s = 3 \text{ cm}^{-1}$. Le gel comporte trois inclusions noires (encre de chine) circulaires de diamètre 1 mm, 2 mm et 3 mm. Cet échantillon est placé dans une cuve d'eau sous un transducteur ultrasonore qui envoie quatre cycles d'une sinusoïde à 4.8 MHz donnant une résolution axiale autour de 1.2 mm. Étant donnée les caractéristiques géométriques du transducteur, la résolution transverse est autour de 800 µm. Cette fois-ci, c'est la cuve qui est placée sur des moteurs pour déplacer l'échantillon en X et en Y tout en gardant la lumière alignée avec les ultrasons de manière à avoir un fond d'imagerie uniforme.

L'image présentée sur la figure B.10 a été obtenue en faisant l'acquisition de 3000 signaux acousto-optique pour chaque position de l'échantillon. Celui-ci a été déplacé par pas de 0.1 mm le long de l'axe X et par pas de 0.75 mm le long de l'axe Y. Du fait du grand nombre de moyennes et de positions d'acquisition, le temps total d'acquisition est relativement long (environ trois heures). On observe bien les trois inclusions sur l'image acousto-optique avec des tailles (largeur à mi-hauteur) et un espacement cohérent avec la réalité.

Conclusion Après la conjugaison de phase, nous venons de démontrer que l'adaptation de front d'onde par mélange à deux ondes était aussi une solution envisageable pour la détection de signaux acousto-optique issu de milieu multiplement diffusant. De plus, le montage utilisé a un temps de réponse bien inférieur à la milliseconde. Néanmoins, cette configuration n'est pas très pratique et assez compliquée à mettre en place. Il est possible



Figure B.10 – Image acousto-optique d'un gel d'agar de 1.5 cm d'épaisseur. (a) Image entière normalisée superposée avec une photo des inclusions. (b) Image entière normalisé et coupe transverse le long de la ligne noire.

qu'une configuration géométrique similaire à celle employée pour la conjugaison de phase donne de meilleurs résultats. Pour cela il faut utiliser un cristal sans traitement HR sur la face arrière afin de récupérer la lumière à 1064 nm derière le cristal avec un miroir dichroïque pour la séparer de la pompe par exemple.

B.4 Détection PA par vibrometrie de speckle.

Dans ce dernier paragraphe on va mettre en place un montage de mélange à deux ondes pour la détection optique de signaux photoacoustiques.

Principe de la détection Les ultrasons générés par photoacoustique se propagent jusqu'à la surface de l'échantillon. Le principe de la détection optique est de sonder les vibrations engendrées par ces ultrasons à l'aide d'un laser. En effet, comme les ultrasons font vibrer l'interface entre l'échantillon et le milieu environnant, le faisceau laser se retrouve modulé en phase, ce qui peut être détecté par interférométrie. Du fait de la diffusion multiple dans les milieux biologiques, il est nécessaire de faire de l'adaptation de front d'onde afin d'améliorer le rapport signal à bruit.

Une fois l'onde acoustique mesurée en plusieurs points de la surface de l'échantillon, on peut reconstruire la forme et la position des émetteurs à l'aide d'algorithme de calcul numérique. On obtient alors une image photoacoustique des absorbeurs dans le milieu diffusant. Afin d'obtenir la forme de l'onde acoustique, il est essentiel que notre système de détection soit linéaire, c'est à dire qu'il soit sensible à la modulation de phase engendrée par les ultrasons et non à son carré. La détection linéaire est possible grâce à la présence de la modulation locale d'indice de réfraction du milieu. En effet, la diffraction sur un réseau de gain ne donne qu'une composante quadratique.

Schéma du montage La détection optique de signaux photoacoustiques a été réalisée à l'aide du montage schématisé sur la figure B.11. Le début du montage est identique

aux montages précédents. Le cristal est sensiblement le même que pour la conjugaison de phase aux traitements près. Ici il n'est pas équipé de traitement HR afin de pouvoir récolter la lumière qui a traversée la zone de gain. Pour la séparer de la lumière de la pompe à 808 nm, on utilise un miroir dichroïque. Le bras signal est aussi très différent



Figure B.11 – Schéma du montage de détection photoacoustique par mélange à deux ondes. HWi sont des lames demi-onde, PBSi sont des cubes séparateur de polarisation, AOMi sont des modulateurs acousto-optique, PD est une photodiode, DM est un miroir dichroïque.

puisque ce montage est basé sur la lumière rétro-diffusé et plus sur la lumière transmise par l'échantillon comme les montages précédents. Le faisceau de sonde est envoyé sur l'échantillon à travers un cube séparateur de polarisation et une lame quart d'onde, ainsi la lumière rétro-diffusé repasse dans la lame quart d'onde et est séparée du faisceau sonde par le cube séparateur. Elle est ensuite focalisée dans la zone de gain par une lentille avant d'être recollectée sur une photodiode.

Temps de réponse Comme pour les montages précédents la première étape a été la mesure du temps de réponse de l'holographie dans cette configuration. Pour cela, la méthode utilisée dans le paragraphe précédent (issue de [90]) a été adaptée. La mesure donne un temps de réponse autour de 25 µs. Ce temps est compris entre les temps de réponse précédemment mesurés. En effet, le pompage est plus intense que dans le cas du montage de mélange à deux ondes (focalisation dans deux dimensions au lieu d'une) ce qui conduit à un temps de réponse plus court. D'un autre coté, il n'y a plus qu'une référence dans le cristal qui est moins saturé que dans le cas du montage de conjugaison

de phase. Par conséquent le temps de réponse est un peu plus long.

Imagerie Afin de faire un premier test d'imagerie photoacoustique dans des conditions "confortables", nous avons fabriqué un gel d'agar cubique non diffusant de plusieurs centimètres d'épaisseur. Un des cotés du gel est constitué d'un morceau de quelques millimètres de blanc de poulet cuit derrière lequel est caché un fil absorbant. L'autre morceau du gel est de l'agar pur très légèrement diffusant. La cible est éclairée par un laser nanoseconde à travers la partie non diffusante du gel afin d'avoir un signal photoacoustique intense. La détection se fait en observant la lumière du laser de sonde rétrodiffusée sur le poulet. Pour faire une image, on balaye l'échantillon le long d'un axe perpendiculaire au faisceau sonde. Le déplacement se fait par pas de 100 µm sur une longueur totale de 8 mm. Les figures B.12.(a) et B.12.(b) représentent la concaténation des signaux temporels enregistrés pour chaque position (chaque signal est la moyenne de 200 acquisitions) pour deux cibles différentes. On y voit bien la forme du front d'onde qui atteint la surface de l'échantillon.



Figure B.12 – Image photoacoustique par détection optique. Signaux temporel pour : (a) un fil de pêche de diamètre 170 µm et (b) un fil de couture de diamètre 500 µm. Image reconstruite pour : (c) le fil de pêche et (d) le fil de couture.

A partir de ces fronts d'ondes, on peut reconstruire la cible par propagation inverse. Le résultat du calcul par un algorithme de beamforming, développé initialement pour l'imagerie photoacoustique avec un transducteur, est donné dans les figures B.12.(c) et B.12.(d). On y voit bien les cibles, elles sont un peu élargies dans la dimension X. Cela est dû à la faible ouverture de collection de l'onde acoustique et à la renormalisation appliquée à chaque signal temporel pour compenser les variations d'intensité de lumière rétrodiffusée.

Conclusion Dans cette dernière partie nous avons démontré la faisabilité d'une détection de signaux photoacoustiques basés sur de l'holographie dans un milieu à gain $(Nd:YVO_4)$. L'étape suivante est de confirmer ces expériences sur un échantillon entièrement diffusant. Bien que plus compliqué que la détection classique par un transducteur piézoélectrique, cette méthode de détection entièrement optique à quelques avantages. Tout d'abord elle est totalement sans contact. Elle ne nécessite pas de mettre un milieu de couplage sur l'échantillon, ce qui peut être utile dans certains cas où il ne faut pas "polluer" l'échantillon. Un autre avantage est la bande passante qui est extrêmement large. En effet, la bande passante de notre système est limité vers le bas par le temps de réponse du cristal. L'adaptation de front d'onde empêche de détecter les variations lentes (de fréquence inférieure à quelques dizaine de kilohertz), et la bande passante de la photodiode limite la bande passante du système vers le haut. Or les photodiodes peuvent être des détecteurs extrêmement rapides avec des bandes passantes de plusieurs centaines de mégahertz. Ce dernier point est intéressant étant donné que la fréquence d'un signal photoacoustique dépend de la taille de l'émetteur. Notre système a donc accès à plusieurs échelles de taille.

B.5 Conclusion et perspectives

Conclusion Au cours de cette thèse nous avons montré la faisabilité d'un système holographique basé sur un milieux à gain (Nd:YVO₄) pour la détection de signaux acoustooptique ou photoacoustique. On a démontré que la rapidité des processus de mélange d'ondes dans notre cristal, permettait de faire de l'holographie dynamique avec un temps de réponse sub-milliseconde. Ce temps de réponse court est particulièrement intéressant pour des applications *in vivo* étant donné que les mouvements naturels dans un milieu biologique décorrèle le speckle en quelques millisecondes. Les milieux à gain sont donc intéressants pour ce type d'application.

Perspectives Les résultats des expériences préliminaires sur l'holographie dans $Nd:YVO_4$ pour l'imagerie acousto-optique et l'imagerie photoacoustique ouvrent des perspectives intéressantes. En effet, les différents montages présentés dans cette thèse ont mis en évidence la rapidité de l'holographie dans notre milieu à gain. Ce type de milieu permet une holographie dynamique avec un temps de réponse plutôt court, largement inférieur au temps de décorrélation du speckle dans un milieu biologique.

Pour faire de l'imagerie acousto-optique, en profondeur, il est nécessaire que la lumière pénètre dans les tissus. Or la longueur d'onde utilisée ici, 1064 nm, n'est pas la plus adaptée. Il serait intéressant d'utiliser des milieux qui ont des caractéristiques similaires (en termes de gain et de temps de vie de fluorescence) mais qui fonctionnent à des longueurs plus basse, autour de 800 nm. Par exemple on peut penser à Ti:Sapphire, Alexandrite ou Cr:LiSAF. De plus, il serait intéressant de transférer le travail effectué sur la détection du mélange à deux ondes (nouveaux filtres, et envoi de plusieurs salves US pendant une impulsion de pompe) au montage de mélange à quatre ondes.

A travers cette thèse nous avons aussi mis en évidence la détection linéaire grâce au réseau d'indice présent dans le milieu à gain. Cette détection linéaire a été exploitée pour faire de l'imagerie photoacoustique sur un échantillon biologique. Mais les applications d'une telle technique ne sont pas limitées à l'imagerie biologique. On pourrait imaginer utiliser ce type de détection pour sonder les vibrations de plaques pour leur caractérisation (il existe actuellement des appareil commerciaux à base de milieux photoréfractifs). La rapidité de notre montage permettrait de faire de genre de tests en conditions difficiles par exemple.

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