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Thèse de doctorat de l'Université Paris VI
en co-tutelle avec
l'École Normale Supérieure de l'Est de la Chine

Spécialité : Optique Quantique

présentée par

Zhan ZHENG

*Pour obtenir le grade de
DOCTEUR DE L'UNIVERSITÉ PIERRE ET MARIE CURIE et de
DOCTEUR DE L'ÉCOLE NORMALE SUPÉRIEURE DE L'EST DE LA CHINE*

sur le sujet :

**Two-Photon–Two-Atom Excitation
With Quantum Multimode Light States**

Soutenue le 28 juillet 2014 devant le jury composé de :

M. Claude FABRE Co-Directeur de thèse
M. Longsheng MA Co-Directeur de thèse
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PhD thesis of the Université Paris VI

co-supervised with

East China Normal University

Speciality : Quantum Optics

presented by

Zhan ZHENG

for the degree of

*the UNIVERSITÉ PIERRE ET MARIE CURIE and of
the EAST CHINA NORMAL UNIVERSITY*

on the subject :

**Two-Photon–Two-Atom Excitation
With Quantum Multimode Light States**

defended on 28th July 2014 with the jury made up of :

M. Claude FABRE	Co-Director of thesis
M. Longsheng MA	Co-Director of thesis
Mme. Agnès MAÎTRE.	President
M. Antoine BROWAEYS	Reporter
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M. Guoxiang HUANG	Examiner
Mme. Alice SINATRA.	Invited

June 8, 2014

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In 2009, Prof. Claude FABRE gave a series of lectures on Quantum Optics in East China Normal University (ECNU), Shanghai, China. I still remember that all the attendants were motivated by his clear, lucid explanations with deep insights to quantum optics. We, the master or PhD students among all the attendants, had very strong wishes that we could work as his PhD student. After his lectures, before his flying back to France, I was too excited to say a word properly when he told me that I could come to Paris to be one of his PhD students in Laboratoire Brossel Kastler (LKB). I realized a new life was coming.

At that moment, I was a future doctoral student candidate in a cooperation program between le Group des Ecole Normale Suprieure and ECNU. One main aim of this program is to establish or strengthen, deepen the cooperation relationships, long-terms wise, between both sides. I asked Prof. Longsheng MA, who is also one of the leading physicists, whether he could be the supervisor of my thesis on ECNU side. It was just so kind of him that he agreed on it.

The new life in Paris, things seemed quite different, and most of them are quite fantastic. Several weeks later, I begun to suffer from the ignorance of the French education system. It was really not easy to do the registration work by myself alone, because I was a bit timid by nature, having an anxiety to make my French understood. I remember clearly the first week when I started my paperwork, Mr. Nicolas TREPS, who is also a leader of our group in LKB, came to me and said: "If you have any difficulty in the registration, just come to tell us, we can even take you to the right places and help you to do them."

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This subject is related to quantum spectroscopy. Professor MA is world-widely known for his excellent work in metrology and precision measurement, especially on the time/frequency standards. I hope this work can be a first preparation to establish the connection between two sides, and I wish both sides can find more and more interesting subjects, perhaps on quantum spectroscopy and metrology, to work productively together in future.

During my stay in Paris, I also worked on temporal multimode quantum memory based

on optical frequency comb. This is a very fascinating subject, and I should work more on it in future.

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Résumé

Deux photons - deux atomes (2P2A) excitation s'avère être un banc de test important de propriétés optiques quantiques. Depuis 1980, on sait que la probabilité de transition 2P2A peut être grandement améliorée en utilisant la source monochromatique cohérente avec la présence d'interactions atomiques qui induisent un pic de résonance de 2P2A. En 2004, Muthukrishnan et al. ont trouvés une grande amélioration de la transition ainsi que d'une résonance de 2P2A, quand une source de deux photons intriqués, provenant de la radiation d'un atome cascade à trois niveaux, interagit avec les deux atomes différents de l'interaction. Leur conclusion est que : dans certaines situations, l'intrication peut remplacer un vrai hamiltonien d'interaction, qui est une déclaration de grande portée et une propriété importante liée à l'intrication. Ce phénomène mérite plus d'attention et de favoriser la compréhension.

Dans cette thèse, nous étudions en détail l'efficacité des différents états multimodes à deux photons de lumière pour provoquer l'excitation simultanée de deux atomes de différents types sous condition de résonance 2P2A. Nous constatons que les états séparables, corrélés, produites soit par une cascade atomique ou conversion paramétrique, ou même des combinaisons appropriées des états cohérents, ont des efficacités comparables comme l'état de photons produit par une cascade atomique. Nous concluons que la véritable origine physique de l'amélioration est les anti-corrélations des fréquences, ni les corrélations temporelles, ni le temps de génération chronologique, ni l'intrication. Pour présenter, nous ne sommes pas sûrs si oui ou non l'amélioration est un effet quantique.

Nous avons également étudié : 1. la 2P2A probabilité de transition optimisée pour un spectre de photons donné; 2. l'influence venant de la durée de vie finie des atomes excités; 3. la 2P2A probabilité de transition en présence d'interaction atomique.

Mots-clefs : optique quantique, excitation 2P2A, amélioration de la transition, intrication, corrélation, nonclassicality

Abstract

Two-photon–two-atom (2P2A) excitation turns out to be an important test bench of quantum optical properties. Since 1980s, it is known that the 2P2A transition probability can be greatly enhanced by using monochromatic coherent source with the presence of atomic interactions which induce a 2P2A resonance peak. In 2004, Muthukrishnan et al. found a great enhancement of the transition as well as a 2P2A resonance, when an entangled two-photon source, coming from the radiation of a cascade three-level atom, interacts with two non-interacting atoms. Their conclusion is that: in some situations, entanglement can replace a real interaction Hamiltonian, which is a far reaching statement and an important property related to entanglement. This phenomenon deserves more attention and further comprehension.

In this thesis, We study in detail the efficiency of various multimode two-photon states of light to induce the simultaneous excitation of two atoms of different kinds under 2P2A resonance condition. We find that several separable, correlated states, produced either by an atomic cascade or parametric down-conversion, or even appropriate combinations of coherent states, have comparable efficiencies as the photon state produced by an atomic cascade. We conclude that the true physical origin of the enhancement is the frequency anti-correlations, neither the temporal correlations, time ordering, nor entanglement. To present, we are not sure whether or not the enhancement is a quantum effect. We also studied: 1. the optimized 2P2A transition probability for a given photon spectrum; 2. the influence coming from finite lifetimes of the excited detecting atoms; 3. the 2P2A transition probability in the presence of atomic interaction.

Keywords : quantum optics, two-photon–two-atom excitation, enhancement of transition probability, entanglement, correlation, nonclassicality

摘要

双光子双原子(2P2A)激发技术是检验量子光学性质很有效的一种手段。从上世纪80年代开始,人们知道:使用单色相干光源,原子间的相互作用会引起2P2A共振,从而使得激发效率有明显地提高。在2004年,Muthukrishnan等人发现:在没有原子间相互作用的情况下,使用产生于级联三能级原子辐射的双光子纠缠源去激发两个原子,同样出现了2P2A共振,显著地提高了激发效率。这意味着:在某些情况下,纠缠可以代替一个实在的相互作用哈密顿量。它揭示了与纠缠有关的一个很重要的性质,这一论断在物理学里的影响是深远的。人们理当更多地去注意和理解这一现象。

在本论文里,我们详细地研究了多种多模双光子态在2P2A共振条件下引起不同种类的两个原子同时受激的效率。我们发现一些关联的可分离态,源自原子级联或参量下转换过程甚至是一些相干态的组合,可以有相当于产生自级联原子的纠缠光子所引起的激发效率。我们得出的结论是:物理上导致激发效率增强的原因不是态在时域上的关联,不是产生光子的时序性,也不是态的纠缠性质,而是态中频率反关联。到目前,我们尚不确定这种增强是否源自量子效应。本论文还讨论了:1.对于给定光子谱的2P2A激发效率的优化问题;2.原子激发态有限的寿命对激发效率的影响;3.在有原子相互作用的情况下2P2A的跃迁概率问题。

关键词:

量子光学, 双原子双光子激发, 跃迁效率增强, 纠缠, 关联, 非经典性

Introduction

Relativity and quantum mechanics are the two foundations of modern physics. Both theories were firstly linked to the strange properties of light, such the light speed c being the velocity upper bound to transmit a signal in vacuum [EinsteinSR1905], discrete energy distribution [Planck1901] in blackbody radiation [Kirchhoff1859,1860] and particle property of light quanta [EinsteinPE1905] in photoelectric effect [EinsteinPE1905]. Together with the researches of spectroscopy on exploring the internal structures of atoms from absorption lines or radiation spectrum, they paved the road to the discovery of Bohr's quantum theory on atoms. Though the quantum properties of the field were highlighted before the ones of atoms, later researches were focussed more on matter corpuscles. On the way to the foundation of a complete quantum mechanics, the most important ideas, such like Heisenberg's matrices, Schrödinger's stationary wave-function explanation to de Broglie's hypothesis, Pauli's exclusion principle, were inspired by spectroscopy data. A full quantum theory on atom-light interaction, Quantum Electrodynamics (QED), was developed till [Dirac1927, Fermi1932]. With the advent of Dirac equation and the successful explanation of the Lamb shift and radiative corrections to the anomalous magnetic moment, it is believed that everything in light-matter interaction can be explained within the frame of QED .

In those early days, it was known that optical field should be treated as an ensemble of photons which have the same mathematical representation as quantum harmonic oscillators, but because of the lack of quantum sources, exhibiting the quantum nature of light was quite difficult. For example the light scattering in the name of Raman effect was observed by means of sunlight [Raman1928]. On the other hand, though phenomena such like blackbody radiation could be rigorously derived from QED theory [Dirac1927], there was no evident practical superiority of using QED theory instead of the semi-classical theory of light-matter interaction, and in particular of the phenomenological description developed by Einstein [Einstein1917] by introducing the simple and intuitive concepts of stimulated emission and spontaneous emission induced by classical light and acting on atoms having quantized energy levels. On the contrary, thanks to these two concepts, the former atomic molecule and optical physics (AMO) was brought forth to a height of development with the help of statistical methods, culminating in the invention and development of the maser and the laser.

Due to this lack of appropriate light sources, basic quantum conceptions, like the wave function collapse, could only be discussed via thought experiments. This situation was not improved until the invention of maser and laser. Owing to the coherence of fields from both, one became able to perform more and more precise control on atom and light

quantum states. Nowadays, one can control and manipulate photons and atoms at single particle level and the quantum fluctuations down to the vacuum fluctuation level. It has then been possible to test some thought experiments of the pioneers of Quantum Mechanics with a high precision.

In classical wave optics, coherence of light is of special importance, since the performance of imaging, resolution abilities of interferometers are determined by it. Logically, the coherence of a quantum light is also important. Experiments on optical correlations were made and some of them, like the experiments on photon bunching and antibunching initiated by Hanbury Brown and Twiss (HBT), showed the importance of photon statistics [Purcell1956]. To understand these results and also inspired by the success of QED theory, a quantum theory of optical coherence (in fact correlations) was developed [Glauber1963, Mandel1965].

The concept of coherence in quantum optics is built on all orders of correlation functions. The n -order correlation function G_n or g_n can provide rich information about the photon state and possible quantum interference phenomena. They also play an important role in nonlinear processes, such as n -photon absorption in a two-level atom: in these processes, the n -photon absorption rate is mainly determined by the normalized correlation function g_n [Teich1966]. It has been showed that the value of absorption rate corresponding to a thermal photon state exceeds the one corresponding to a coherent state by a factor $n!$ [Mandel1965, Lambropoulos1966] due to phenomenon of photon bunching. In addition, in a two-photon absorption [Göpert-Mayer1931] process induced by an entangled photon state, it has been shown that there is a linear dependence of absorption rate on photon flux density [Friberg1985, Javanainen1990] instead of a quadratic one. N -photon lithography performed with a NOON state $|NOON\rangle = (e^{iN\varphi}|N0\rangle + |0N\rangle)/\sqrt{2}$, for instance, has been theoretically shown to produce a remarkable improvement to the diffraction limit from classical light [Boto2000]. More generally, using a quantum light source may provide an appreciable improvement in absorption rates by matter, either in quantity or in its variation with the relevant parameters.

Quantum light sources are needed to produce highly non-classical correlation functions required to strongly modify multi-photon absorption rates. Various new light sources are available now, generating single photons, entangled two-photon states, squeezed states, Schrödinger's cat states, entangled coherent states, twin-beam states, NOON states ..., that exhibit genuine quantum properties such as photon anti-bunching, reduced quantum fluctuations, strong quantum correlations, negativity in quasi-probability distribution and violation of Bell inequality. The completeness of quantum mechanics, the upper bound of the wave function collapse duration, etc. have been experimentally checked. These sources are also widely used in quantum optical information science and quantum metrology.

On the other hand, coherent control of atomic processes by light shaping is a very important subject that has attracted a lot of interest from spectroscopists. For example, in ultrashort pulse spectroscopy, the use of so-called bright pulses and dark pulses [Meshulach1998] in two-photon absorption by a two-level atom with a given power spectrum has been shown to change significantly the transition probabilities.

Coherent quantum control is actually the extension of the same techniques to the quantum domain: one plays now not only with light mode shapes, but with quantum states which have more detailed characteristics than the power spectrum and phase distribution

of field. As a result, quantum assisted processes can benefit from the optimization of more features than in regular coherent control and be more efficient in the enhancement of processes. For example, the discovery of entanglement-induced two-photon transparency [Fei1997] phenomenon gave rise to entangled-photon virtual-state spectroscopy [Saleh1998] by entangled photon pairs.

Among all the light-matter systems, two-level atoms interacting with light are simple and of great interest, in particular because they implement systems of qubits, a basic tool of quantum computation. The global interaction between two different two-level atoms and light is in this respect important, especially in conditions where the photons are not resonant with each atom taken individually but when the sum of the energies of two photons matches the sum of the energies of the two atoms (a situation called 2-Photon 2-Atom absorption, or 2P2A absorption). Such a simultaneous two-photon absorption by two atoms of different species was observed in a mixture of Ba/Ti [White1981]. The detailed scheme of this process was considered in a theoretical article [RiosLeite1980] and then in [Andrews1983]. Their results show that a resonance can be found in the 2P2A configuration when there exists a physical interaction between the two atoms. This phenomena was verified by [Hettich2002, P-Penãfiel2012]. Because each atom is far off resonance with light, there is no overlap between the wings of the individual single photon transitions, and the height of the induced 2P2A peak depends on the strength of the inter-atomic interaction. Such a feature can be used to study cold collisions in cold atoms, or the quantum interference effects like cooperative emissions, etc. In addition to the case of a direct potential interaction between the atoms, like the dipole-dipole coupling, a cooperative 2P2A effect has also been predicted for pairs of atoms inside an optical cavity [Kim1998]. In this case the physical interaction is mediated by the radiation background surrounding the atoms.

In 2004, a theoretical paper was published [Muthukrishnan2004], addressing the case of 2P2A absorption in absence of interaction between the two atoms, but using some particular entangled two-photon state of light. It showed that a particular entangled state was able to efficiently induce the 2P2A transition, whereas factorized states could not induce it. The conclusion of the authors of [Muthukrishnan2004] is that *in some situations entanglement can replace a real physical interaction*, which is a far reaching statement and an important physical property attributed to entanglement. The same subject was also considered, but in the context of spin entanglement in Electron Paramagnetic Resonance by K. Salikhov [Salikhov2003]. Surprisingly, this question did not attract much attention during several years. It was only recently that the related problem of interaction with pairs of broadband spectrum photons [Richter2011, Dayan2007] has been discussed.

We have estimated that the physical problem of 2P2A absorption studied in this paper was a good test bench to examine in detail the role of entanglement, of correlations not related to entanglement, and of interactions in a simple situation. Our aim is to examine the two-photon two-atom excitation process in various configurations in order to find out the true physical origin to the remarkable enhancement of 2P2A transition probability announced in [Muthukrishnan2004].

More precisely, in this thesis, we will calculate the two-photon transition probability in two different two-level systems with various of quantum light source as well as the physical origins to these transitions. In chapter 1, we will recall briefly some fundamentals

of quantum mechanics and quantum optics as well as some measures of entanglement in a bipartite system. We start the discussion of two-photon transition in chapter 2 in the case of the interaction with a single effective two-level atom and highlight the dark pulse and entanglement-induced transparency phenomena. They are two important examples to the coherent control of atom-field interaction. In chapter 3, we will introduce a complete model to the interaction between light and an expression for two-photon transition. With these results, we discuss the physical origin of the enhancement of transition probability with two non-interacting two-level atoms in Chapter 4. In a following chapter, we also consider a case that both two atoms that have finite lifetime. In chapter 5, we focus our attention on the transition in the case of two interacting two-level atoms. Chapter 6 is designed to discuss the possible genuine quantum aspects of the light sources that we have used in the previous chapters.

Toolbox from Quantum Optics

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IN this chapter, we will give some introductions to basic knowledge of quantum optics. They are necessary for our later discussions. These concepts are quantized field, multi-modal photon states, the statistical properties of a photon state such like photon spacing, entanglement and correlations. In this chapter, we also give a brief introduction to two quantum two-photon state: 1. that emitted from an excited cascade three-level atom, and 2. that generated by nonlinear optical parameter down conversion process.

A Field quantization and correlation functions

A.1 Global field description

It is widely accepted that a light system should be described by a universal quantum field $\hat{\mathbf{A}}(\mathbf{r})$ where \mathbf{r} is a 3 dimensional spatial parameter and its corresponding quantum state ρ . The universal quantum field is usually expressed as a sum of a quantum analytical signal $\hat{\mathbf{A}}^{(+)}(\mathbf{r})$ and its Hermitian conjugate $\hat{\mathbf{A}}^{(+)\dagger}(\mathbf{r})$,

$$\hat{\mathbf{A}}(\mathbf{r}) = \hat{\mathbf{A}}^{(+)}(\mathbf{r}) + \hat{\mathbf{A}}^{(+)\dagger}(\mathbf{r}). \quad (1.1)$$

Quantum analytical signal $\hat{\mathbf{A}}^{(+)}(\mathbf{r})$ follows an integral form,

$$\hat{\mathbf{A}}^{(+)}(\mathbf{r}) = \sum_{\lambda=1}^2 \int d\mathbf{k} \sqrt{\frac{\hbar}{2(2\pi)^3 \epsilon_0 \omega_{\mathbf{k}}}} \mathbf{e}_{\mathbf{k},\lambda} \hat{a}_{\lambda}(\mathbf{k}) e^{i\mathbf{k}\cdot\mathbf{r}}. \quad (1.2)$$

The point $\mathbf{k} = \mathbf{0}$ is excluded, otherwise it will pose some problems. In this expression, $\hat{a}_{\lambda}(\mathbf{k}), \hat{a}_{\lambda}^{\dagger}(\mathbf{k})$ are the annihilation operator and the creation operator linked to wave-vector \mathbf{k} and polarization λ , they yield canonical Bosonic commutation relation:

$$[\hat{a}_{\lambda}(\mathbf{k}), \hat{a}_{\lambda'}^{\dagger}(\mathbf{k}')] = \delta_{\lambda\lambda'} \delta(\mathbf{k} - \mathbf{k}'). \quad (1.3)$$

The polarization can have only two independent components since the EM field is a transverse field. The Hamiltonian of such a field is given by

$$\hat{H} = \sum_{\lambda=1}^2 \int d\mathbf{k} \omega_{\mathbf{k}} \hat{a}_{\lambda}^{\dagger}(\mathbf{k}) \hat{a}_{\lambda}(\mathbf{k}). \quad (1.4)$$

The frequency $\omega_{\mathbf{k}}$ is a function of \mathbf{k} , when there is no transverse current, for example a free field in vacuum, the relation between them is simple,

$$\omega_{\mathbf{k}} = c\|\mathbf{k}\|, \quad (1.5)$$

where $\|\cdot\|$ is the length of a vector. For any quantum state of light $|\psi\rangle$, the mean value of the Hamiltonian is

$$\langle \psi | \hat{H} | \psi \rangle = \sum_{\lambda=1}^2 \int d\mathbf{k} \omega_{\mathbf{k}} \|\hat{a}_{\lambda}(\mathbf{k})|\psi\rangle\|^2 \geq 0. \quad (1.6)$$

The Hamiltonian is a positive operator. The state that gives rise to vanishing mean value of Hamiltonian is the vacuum state, denoted by $|0\rangle$, it yields $\hat{a}_{\lambda}(\mathbf{k})|0\rangle = 0$ for any \mathbf{k} . Owing to the canonical commutation relation Eq.(1.3), the mean value of an operator $\hat{a}_{\lambda}(\mathbf{k})\hat{a}_{\lambda}^{\dagger}(\mathbf{k})$ of any given polarization λ and wave-vector \mathbf{k} on vacuum state is $\langle 0 | \hat{a}_{\lambda}(\mathbf{k})\hat{a}_{\lambda}^{\dagger}(\mathbf{k}) | 0 \rangle = \infty$ which implies that $\hat{a}_{\lambda}^{\dagger}(\mathbf{k})|0\rangle$ is not normalizable, therefore it is not a vector in Hilbert space. A physical state should of course be normalized, and the conventional way to make the previous unnormalized state physical is to introduce a weight function, and define the state as

$$|I : u\rangle = \sum_{\lambda=1}^2 \int d\mathbf{k} u_{\lambda}(\mathbf{k}) \hat{a}_{\lambda}^{\dagger}(\mathbf{k}) | 0 \rangle, \quad \sum_{\lambda=1}^2 \int d\mathbf{k} |u_{\lambda}(\mathbf{k})|^2 = 1. \quad (1.7)$$

Now the state satisfying $\langle I : u | I : u \rangle = 1$ is indeed a physical one. In view of this, a physical light state should be assigned to some special function like $u_\lambda(\mathbf{k})$. Since $u_\lambda(\mathbf{k})$ can be an arbitrary usual (complex) function, it is convenient to find a basis $\{u_{\lambda,n}(\mathbf{k})\}_{n=0}^\infty$, whose orthonormal completeness relations follow

$$\int d^3\mathbf{k} u_{\lambda,m}^*(\mathbf{k}) u_{\lambda',n}(\mathbf{k}) = \delta_{\lambda\lambda'} \delta_{mn}, \quad (1.8)$$

$$\sum_n u_{\lambda,n}^*(\mathbf{k}) u_{\lambda,n}(\mathbf{k}') = \delta(\mathbf{k} - \mathbf{k}'), \quad (1.9)$$

to describe them all. Let's introduce some new annihilation operators as

$$\hat{b}_{\lambda,m} = \int d\mathbf{k} u_{\lambda,m}^*(\mathbf{k}) \hat{a}_\lambda(\mathbf{k}), \quad m = 0, 1, 2, \dots \quad (1.10)$$

Then the non-vanishing commutators are

$$[\hat{b}_{\lambda,m}, \hat{b}_{\lambda',n}^\dagger] = \iint d^3\mathbf{k} d^3\mathbf{k}' u_{\lambda,m}^*(\mathbf{k}) u_{\lambda',n}(\mathbf{k}') [\hat{a}_\lambda(\mathbf{k}), \hat{a}_{\lambda'}^\dagger(\mathbf{k}')] = \delta_{\lambda\lambda'} \delta_{mn}. \quad (1.11)$$

They are indeed equivalent to usual canonical commutation relations with respect to harmonic oscillators. Following a same procedure, a Fock state, defined as

$$|N : u_{\lambda,m}\rangle = \frac{\hat{b}_{\lambda,m}^{\dagger N}}{\sqrt{N!}} |0\rangle, \quad N \in \mathbb{N}, \quad (1.12)$$

describes a state with N photons in basis mode $u_{\lambda,m}$. In last equation, we have tacitly used a symbol like an integer or a complex number before ":" to signify the photon statistics of a state while the symbol after ":" stands for the corresponding mode in our semiotics. The total number operator is defined as a sum of number operators for all the modes in any basis

$$\hat{N} = \sum_{\lambda,m} \hat{b}_{\lambda,m}^\dagger \hat{b}_{\lambda,m}. \quad (1.13)$$

According to Eqs.(1.9,1.10), this total number operator is exactly equal to

$$\hat{N} = \sum_{\lambda=1}^2 \int d\mathbf{k} \hat{a}_\lambda^\dagger(\mathbf{k}) \hat{a}_\lambda(\mathbf{k}). \quad (1.14)$$

Now, we can conclude that the number operator is universal as it is independent of the choices of bases.

Operator $\hat{a}_\lambda(\mathbf{k})$ can also be expressed in terms of $\hat{b}_{\lambda,m}$ s, according to Eq.(1.10), such reads

$$\hat{a}_\lambda(\mathbf{k}) = \sum_m \hat{b}_{\lambda,m} u_{\lambda,m}(\mathbf{k}), \quad (1.15)$$

correspondingly, the quantum field can also written as

$$\hat{\mathbf{A}}^{(+)}(\mathbf{r}) = \sum_{\lambda=1}^2 \sum_m \hat{b}_{\lambda,m} \int d\mathbf{k} \sqrt{\frac{\hbar}{2(2\pi)^3 \epsilon_0 \omega_{\mathbf{k}}}} \mathbf{e}_{\mathbf{k},\lambda} u_{\lambda,m}(\mathbf{k}) e^{i\mathbf{k}\cdot\mathbf{r}}. \quad (1.16)$$

This quantum field permits of a description of ploy-chromatic many-body system.

Let's note that the concepts of modes and mode basis are not quantum at all, because both two are pure mathematical concepts related to linearity of Maxwell equations, a classical electromagnetic field also admits a basis. If the creation and annihilation operators are replaced by a pair of conjugate complex numbers, then the field amplitude is regarded as a linear combination of the all the mode-functions in the basis. By using different bases, one should have different sets of coefficients. However, if one chooses the field amplitude as the first unnormalized mode function, then it is of no use to introduce the other mode-functions in classical electromagnetic field theory. In other words, any non-fluctuating classical electromagnetic field is indeed a single-mode field. In a quantum world, it is not true in most cases because of the high-dimensionality and the complexity of Hilbert space (photon states) as well as the fundamental importance of canonical commutation relations (field operators). We will detail the related fundamental concepts in the last section of this chapter.

A.2 Local field description

For some practical reasons, sometimes it is more convenient to use a local field than to use the field introduced in previous section to discuss the quantum optical phenomena, since most quantum optical experiments are carried out in a finite volume of space. For example, a plane wave field which propagates along z direction can be treat as

$$\hat{\mathbf{A}}^{(+)}(\mathbf{r}) = \sum_{\lambda=1}^2 \sum_m \mathbf{e}_\lambda \hat{b}_{\lambda,m} \int d\omega c^{-1} \sqrt{\frac{\hbar}{2(2\pi)^3 \epsilon_0 \omega}} u_{\lambda,m}(\omega/c) e^{iz\omega/c}, \quad (1.17)$$

where $u_{\lambda,m}(\omega/c)$ is taken as a piecewise function,

$$u_{\lambda,m}(\omega/c) = (\omega/\omega_m)^{1/2} u_{\lambda,m}(\omega_m/c) e^{iz(\omega_m - \omega)/c}, \quad \omega \in (\omega_m, \omega_{m+1}];$$

otherwise $u_{\lambda,m}(\omega/c) = 0$. We denote the annihilation operator with this mode by $\hat{a}_{\lambda,m}$, then the field can be expressed as

$$\hat{\mathbf{A}}^{(+)}(z) = \sum_{\lambda=1}^2 \sum_m \mathbf{e}_\lambda \hat{a}_{\lambda,m} c^{-1} \sqrt{\frac{\hbar}{2(2\pi)^3 \epsilon_0 \omega_m}} e^{iz\omega_m/c}, \quad (1.18)$$

where the annihilation operator and creation operator satisfies the usual canonical commutation relation $[\hat{a}_{\lambda,m}, \hat{a}_{\lambda',n}^\dagger] = \delta_{mn} \delta_{\lambda\lambda'}$. The electric field Correspondingly, the electric field is then

$$\hat{\mathbf{E}}^{(+)}(z) = \sum_\lambda \mathbf{e}_\lambda \hat{E}_\lambda^{(+)}(z), \quad \hat{E}_\lambda^{(+)}(z) = \sum_m i \mathcal{E}_m \hat{a}_{\lambda,m} e^{i\omega_m z/c}, \quad (1.19)$$

where \mathcal{E}_m is the field unit of the monochromatic frequency ω_m . For any integer m , if $\omega_{m+1} - \omega_m = L/2\pi c$, then one regards L as a spatial periodicity of a field, and such corresponding quantization of field can be derived from the method of quantization in a box.

Here-to-after, we will hide the polarization index for simplicity.

A.3 Correlation functions

Correlation plays a fundamental role in the theory of optical coherence. A key ingredient of field correlation comes from the scalar part of field operator

$$\begin{aligned}\hat{E}^{(+)}(z, t) &= e^{i\sum_m \omega_m \hat{a}_m^\dagger \hat{a}_m t} \hat{E}^{(+)}(z) e^{-i\sum_n \omega_n \hat{a}_n^\dagger \hat{a}_n t} \\ &= \sum_m i\mathcal{E}_m \hat{a}_m e^{i\omega_m(z/c-t)},\end{aligned}\quad (1.20)$$

which can take a photon away from a given state. An n -th order field correlation function is then defined as

$$G^{(n)} = \langle \hat{E}^{(+)\dagger}(z_1, t_1) \cdots \hat{E}^{(+)\dagger}(z_n, t_n) \hat{E}^{(+)}(z_{n+1}, t_{n+1}) \cdots \hat{E}^{(+)}(z_{2n}, t_{2n}) \rangle, \quad (1.21)$$

where the angle-bracket ensemble average is evaluated with the use of the density operator of a light beam. This function describes a process destroying n photons at the positions and times $(z_{n+1}, t_{n+1}), \dots, (z_{2n}, t_{2n})$ and creating n photons at $(z_1, t_1), \dots, (z_n, t_n)$. The amplitude correlation is represented by the first order correlation function

$$G^{(1)}(z_1, t_1; z_2, t_2) = \langle \hat{E}^{(+)\dagger}(z_1, t_1) \hat{E}^{(+)}(z_2, t_2) \rangle. \quad (1.22)$$

To describe the optical coherence, it is convenient to introduce normalized form of the correlation functions. Correspondingly, it is defined as

$$g^{(n)}(\mathbf{z}, \mathbf{t}) = \frac{\langle \hat{E}^{(+)\dagger}(z_1, t_1) \cdots \hat{E}^{(+)\dagger}(z_n, t_n) \hat{E}^{(+)}(z_{n+1}, t_{n+1}) \cdots \hat{E}^{(+)}(z_{2n}, t_{2n}) \rangle}{\prod_{j=1}^{2n} [G^{(1)}(z_j, t_j; z_j, t_j)]^{1/2}}. \quad (1.23)$$

For example, an interesting second order correlation functions follows [LoudonQTL]

$$g^{(2)}(\mathbf{z}, \mathbf{t}) = \frac{\langle \hat{E}^{(+)\dagger}(z_1, t_1) \hat{E}^{(+)\dagger}(z_2, t_2) \hat{E}^{(+)}(z_2, t_2) \hat{E}^{(+)}(z_1, t_1) \rangle}{\langle \hat{E}^{(+)\dagger}(z_1, t_1) \hat{E}^{(+)}(z_1, t_1) \rangle \langle \hat{E}^{(+)\dagger}(z_2, t_2) \hat{E}^{(+)}(z_2, t_2) \rangle}, \quad (1.24)$$

In a bipartite system, it is also quite interesting to know the details of the correlations between parts with the help of the following cross temporal correlation function

$$g_{\times}^{(2)}(\mathbf{z}, \mathbf{t}) = \frac{\langle \hat{E}_{\alpha}^{(+)\dagger}(z_1, t_1) \hat{E}_{\beta}^{(+)\dagger}(z_2, t_2) \hat{E}_{\beta}^{(+)}(z_2, t_2) \hat{E}_{\alpha}^{(+)}(z_1, t_1) \rangle}{\langle \hat{E}_{\alpha}^{(+)\dagger}(z_1, t_1) \hat{E}_{\alpha}^{(+)}(z_1, t_1) \rangle \langle \hat{E}_{\beta}^{(+)\dagger}(z_2, t_2) \hat{E}_{\beta}^{(+)}(z_2, t_2) \rangle}, \quad (1.25)$$

or with the help of cross frequency correlation function

$$g_{\times}^{(2)}(\mathbf{z}, \boldsymbol{\omega}) = \frac{\langle \hat{E}_{\alpha}^{(+)\dagger}(z_1, \omega_1) \hat{E}_{\beta}^{(+)\dagger}(z_2, \omega_2) \hat{E}_{\beta}^{(+)}(z_2, \omega_2) \hat{E}_{\alpha}^{(+)}(z_1, \omega_1) \rangle}{\langle \hat{E}_{\alpha}^{(+)\dagger}(z_1, \omega_1) \hat{E}_{\alpha}^{(+)}(z_1, \omega_1) \rangle \langle \hat{E}_{\beta}^{(+)\dagger}(z_2, \omega_2) \hat{E}_{\beta}^{(+)}(z_2, \omega_2) \rangle}, \quad (1.26)$$

where $\hat{E}(\omega)$ is Fourier transformed from $\hat{E}(t)$. If for any frequencies ω_1, ω_2 , one has $g_{\times}^{(2)}(\mathbf{z}, \boldsymbol{\omega}) = 1$, then the bipartite light beam is factorable; otherwise, the light beam has some frequency correlations between parts. Frequency anti-correlation is a special correlation. In bipartite two-photon states whose total energy is $\hbar\omega_{2\text{photon}}$, such a correlation can be understood in the following way : for majority of photons in ω_1 -frequency mode in the first partite, it has a higher probability to find the other photon in $\omega_{2\text{photon}} - \omega_1$ frequency mode in the second partite.

B Some photon sources

In quantum optics, there are many interesting quantum light sources. The bipartite frequency-anticorrelated photon sources will be extremely used in this thesis.

A quantum state, mixed or pure, can always be represented by a density matrix ρ . For example, a general bipartite two-photon state can be described by

$$\rho = \sum_{jkmn} p(\omega_j, \omega_k, \omega_m, \omega_n) |1 : \omega_j, \alpha; 1 : \omega_k, \beta\rangle \langle 1 : \omega_m, \alpha; 1 : \omega_n, \beta|. \quad (1.27)$$

The symbol $|1 : \omega_j, \alpha; 1 : \omega_k, \beta\rangle$ is an abbreviation of a tensor product $|1 : \omega_j, \alpha\rangle \otimes |1 : \omega_k, \beta\rangle$ of two single photon states in frequency mode ω_j of α subsystem and in frequency mode ω_k of β subsystem respectively. This density matrix can show some quantum interferences or optical coherence between different frequency modes because of the off-diagonal elements. If all these off-diagonal elements are averaged to zero by noises, that is, the state becomes completely dephased, then the corresponding density matrix turns out to be a diagonal one. Such a state is named as a completely dephased state in this thesis. For a bipartite two-photon source, its completely dephased state reads

$$\rho = \sum_{mn} p(\omega_m, \omega_n, \omega_m, \omega_n) |1 : \omega_m, \alpha; 1 : \omega_n, \beta\rangle \langle 1 : \omega_m, \alpha; 1 : \omega_n, \beta|. \quad (1.28)$$

For sake of simplicity, sometimes $p(\omega_m, \omega_n, \omega_m, \omega_n)$ will be abbreviated as $p(\omega_m, \omega_n)$ or p_{mn} which should not cause any confusions.

A pure bi-partite two-photon state can also be described by a ket vector in Hilbert space, its the general form follows

$$|\mathbb{I} : \text{source}\rangle = \sum_{m,n} c(\omega_m, \omega_n) |1 : \omega_m, \alpha; 1 : \omega_n, \beta\rangle. \quad (1.29)$$

The normalized coefficients $c(\omega_m, \omega_n)$ will be abbreviated as c_{mn} if need be.

Let's note that the dephased state represents a stationary field whereas the pure state gives rise to a time-dependent field.

In this section, we will introduce two pure states, namely, the cascade two-photon state $|\mathbb{I} : \text{cas}\rangle$ which describes a photon pair generated from the decay of a three-level atom excited in its top level and cascading down to ground state through its intermediate state, as well as the two-photon state $|\mathbb{I} : \text{pdc}\rangle$ generated by spontaneous parametric down conversion process.

B.1 Cascade two-photon state

A three-level atom contains a top excited level $|\text{top}\rangle$ with its Bohr frequency $\omega_\alpha + \omega_\beta$ and width γ_α , an intermediate level $|\text{med}\rangle$ with its Bohr frequency ω_β and width γ_β , and also a ground state $|\text{g}\rangle$ whose Bohr frequency is set to be 0. See the configuration in Figure 1.1.

The atom is initially excited at $t = 0$ to the top level $|\text{top}\rangle$. It can stay on this state for a duration of γ_α^{-1} . A first photon is radiated on the transition $|\text{g}\rangle$ to $|\text{med}\rangle$, at time t . This process is described by

$$-\sum_k g_\alpha \frac{e^{i(\omega_k - \omega_\alpha)t - \gamma_\alpha t} - e^{-\gamma_\beta t}}{\omega_k - \omega_\alpha + i(\gamma_\alpha - \gamma_\beta)} |\text{med}\rangle \otimes |1 : \omega_k, \alpha\rangle. \quad (1.30)$$

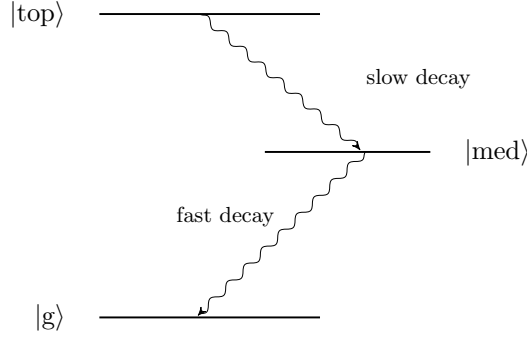


Figure 1.1: The generation scheme of a two-photon state from the excited cascade three-level atom. The atom was initially occupied in the state $|top\rangle$, hence via successional radiations, a photon pair is generated with frequency anti-correlation because of energy conservation

The both terms in numerator are decaying at rates of $\gamma_\alpha, \gamma_\beta$. The emitted single photon has a Lorentzian distribution in frequency and its width is equal to $|\gamma_\beta - \gamma_\alpha|$. If $\gamma_\alpha > \gamma_\beta$, then there will be some population accumulation, but if $\gamma_\alpha \ll \gamma_\beta$, then the state can not stay for a long time. As the excited state $|top\rangle$ has a lifetime, another photon is emitted soon, and at a given time t , the generated state is given by [ScullyQO]

$$\sum_{kq} \frac{g_\alpha g_\beta |1 : \omega_k, \alpha; 1 : \omega_q, \beta\rangle}{\omega_k - \omega_\alpha + i(\gamma_\alpha - \gamma_\beta)} \left[\frac{1 - e^{-\gamma_\beta t + i(\omega_q - \omega - \omega_\beta)t}}{\omega_q - \omega_\beta + i\gamma_\beta} - \frac{1 - e^{-\gamma_\alpha t + i(\omega_k + \omega_q - \omega_\alpha - \omega_\beta)t}}{\omega_k + \omega_q - \omega_\alpha - \omega_\beta + i\gamma_\alpha} \right] |g\rangle.$$

By projecting the atomic state on the ground state, one obtains a two-photon state $\rho = |\mathbb{II} : cas\rangle \langle \mathbb{II} : cas|$, where

$$|\mathbb{II} : cas\rangle = \sum_{kq} \frac{\mathcal{N} |1 : \omega_k, \alpha; 1 : \omega_q, \beta\rangle}{\omega_k - \omega_\alpha + i(\gamma_\alpha - \gamma_\beta)} \left[\frac{1 - e^{-\gamma_\beta t + i(\omega_q - \omega - \omega_\beta)t}}{\omega_q - \omega_\beta + i\gamma_\beta} - \frac{1 - e^{-\gamma_\alpha t + i(\omega_k + \omega_q - \omega_\alpha - \omega_\beta)t}}{\omega_k + \omega_q - \omega_\alpha - \omega_\beta + i\gamma_\alpha} \right], \quad (1.31)$$

with \mathcal{N} being a constant number for normalization. This state can not be factorized into two separable parts, therefore it is an entangled state. The first term in the square bracket stands for a usual single photon emission process while the second term represents frequency anti-correlated emission. This (anti-)correlation comes from the energy conservation, because the total energy $\hbar(\omega_k + \omega_q)$ of a photon pair should be close to $\hbar(\omega_\alpha + \omega_\beta)$. In other words, it is with higher probability to find a photon at frequency $\omega_\alpha + \omega_\beta - \omega_k$ when a photon at frequency ω_k is given in the pair. With a small width γ_α but a large width γ_β , one obtains significant frequency anti-correlations between the emitted photons. When the time t is large, $t \gg \gamma_\beta^{-1}, \gamma_\alpha^{-1}$, this state is equal to

$$|\mathbb{II} : cas\rangle = \sum_{kq} c_{kq} |1 : \omega_k, \alpha; 1 : \omega_q, \beta\rangle, \quad (1.32)$$

$$c_{kq} = \frac{\mathcal{N}}{(\omega_q - \omega_\beta + i\gamma_\beta)(\omega_k + \omega_q - \omega_\alpha - \omega_\beta + i\gamma_\alpha)}. \quad (1.33)$$

The central frequencies of photon pair in this state are $\omega_\alpha, \omega_\beta$ with corresponding widths $\gamma_\alpha + \gamma_\beta, \gamma_\beta$.

For full demonstration, see [ScullyQO].

If the atom excitation occurs no longer at a precise time $t = 0$, but at random times, the state is completely dephased, and all the off-diagonal elements of the density matrix are averaged to 0. Then the density matrix becomes

$$\Lambda_{cas} = \sum_{kq} \frac{\mathcal{N}^2 |1 : \omega_k, \alpha; 1 : \omega_q, \beta\rangle \langle 1 : \omega_k, \alpha; 1 : \omega_q, \beta|}{[(\omega_q - \omega_\beta)^2 + \gamma_\beta^2][(\omega_k + \omega_q - \omega_\alpha - \omega_\beta)^2 + \gamma_\alpha^2]}. \quad (1.34)$$

This is a diagonal density matrix which describes a separable state, without any entanglement. The quantum correlations of such a state will be discussed in chapter 6.

B.2 Spontaneous parametric down-converted two-photon states

Parametric down-conversion (PDC) is a coherent three-photon process in a nonlinear crystal. The crystal is illuminated by a pump laser with central frequency ω_p and an envelope of arbitrary shape $\mathcal{A}(z, t)$. The Fourier transform of shape function \mathcal{A} gives rise to its frequency spectrum $\mathcal{S}(\nu)$. Signal and idler beams with central frequencies at $\omega_\alpha, \omega_\beta$, degenerate or not, are generated by the nonlinear process of spontaneous down conversion. If the photon pairs are generated in same polarization, then the process belongs to type I ; if they are generated in orthogonal basis, then the process belongs to type II. The two beams can propagate either in different directions or collinearly [Pittman1996]. This process is described by an interaction Hamiltonian [Rubin1994, Keller1997, MandelOCQO]

$$\hat{H}_I = \int_V dV \epsilon_0 \chi \mathcal{A} e^{-i\omega_p t} \hat{E}_\alpha \hat{E}_\beta, \quad (1.35)$$

When \mathcal{A} is time-independent, this process is called optical parametric down-conversion, and each converted photon pair has perfect frequency anti-correlations. If the pump field consists of pulses, then the corresponding process is usually called spontaneous parametric down-conversion (SPDC).

For example, in the collinear case, in the perturbation theory, the first two terms of the state is

$$|\Psi\rangle = |0\rangle + \sum_{m,n} \mathcal{F}(k_\alpha(\omega_m), k_\beta(\omega_n)) a_\alpha^\dagger(\omega_m) a_\beta^\dagger(\omega_n) |0\rangle, \quad (1.36)$$

where $\mathcal{F}(k_\alpha(\omega_m), k_\beta(\omega_n))$ is the shape factor. Via second order nonlinear process, a photon at frequency ν splits into two photons at frequencies ω_m and ω_n on the condition that $\nu = \omega_m + \omega_n$. If it is a type II collinear SPDC process, the perfect phase matching conditions follow

$$\omega_\alpha + \omega_\beta = \omega_p, \quad (1.37a)$$

$$n_\alpha \omega_\alpha / c + n_\beta \omega_\beta / c = n_p \omega_p / c, \quad (1.37b)$$

where $n_\mu, \mu = \alpha, \beta, p$; are the refractive indices of the three fields. The wave-vector k_μ can be assessed at

$$k_\mu(\omega + \omega_\mu) = n_\mu \frac{\omega_\mu}{c} + \frac{\omega}{u_\mu(\omega_\mu)}, \quad \mu = \alpha, \beta; \quad (1.38)$$

where u_μ are the group velocities. Usually, the two-photon profile $\mathcal{F}(k_\alpha(\omega_m), k_\beta(\omega_n))$ can be expressed as a product of source part $\mathcal{S}(\omega_m + \omega_n)$ and material response part $\mathcal{M}(\omega_m, \omega_n)$,

$$\mathcal{F}(k_\alpha(\omega_m), k_\beta(\omega_n)) = \mathcal{S}(\omega_m + \omega_n) \mathcal{M}(\omega_m, \omega_n). \quad (1.39)$$

The material response functions $\mathcal{M}(\omega_m, \omega_n)$ can be very different from one crystal to another, as they are strongly dependent on the crystal parameters. Typically, the response functions of the interaction between crystal and light are related to the sine cardinal function because of the calculus of exponential functions over the domain of spatial and temporal variables. For simplicity, it is convenient to use Gaussian functions to replace the response functions. As an example, the two-photon state can be approximated by [Wang2006]

$$\begin{aligned} |\mathbb{II} : pdc\rangle \propto \sum_{m,n} \mathcal{S}(\omega_m + \omega_n) & \left[e^{-(\omega_m - \omega_\alpha)^2/2\Gamma_\alpha^2 - (\omega_n - \omega_\beta)^2/2\Gamma_\beta^2} \right. \\ & \left. + e^{i\theta} e^{-(\omega_n - \omega_\alpha)^2/2\Gamma_\alpha^2 - (\omega_m - \omega_\beta)^2/2\Gamma_\beta^2} \right] a_\alpha^\dagger(\omega_m) a_\beta^\dagger(\omega_n) |0\rangle, \end{aligned} \quad (1.40)$$

where $\Gamma_\alpha, \Gamma_\beta$ are two widths of peaks at $\omega = \omega_\alpha, \omega_\beta$; θ is a total phase difference between two beams.

When the two beams propagate collinearly, the common material response $\mathcal{M}(\omega_m, \omega_n)$ reads

$$\mathcal{M}(\omega_m, \omega_n) = \text{sinc}\{[k_\alpha(\omega_m) + k_\beta(\omega_n) - k_p(\omega_m + \omega_n)]l/2\}, \quad (1.41)$$

where l is the length of the $\chi^{(2)}$ crystal and $k_{\alpha,\beta,p}$ represent three wave-numbers of the corresponding fields. In most cases, the bands of the photons are quite small, it is convenient to make Taylor's expansions at the central frequencies to them:

$$k_\alpha(\omega_m) \simeq k_s(\omega_\alpha) + (\omega_m - \omega_\alpha)/u_\alpha, \quad u_\alpha = 1/(\partial_\omega k_\alpha(\omega))|_{\omega_\alpha}; \quad (1.42a)$$

$$k_\beta(\omega_m) \simeq k_\beta(\omega_\beta) + (\omega_m - \omega_\beta)/u_\beta, \quad u_\beta = 1/(\partial_\omega k_\beta(\omega))|_{\omega_\beta}; \quad (1.42b)$$

If the pump field has a very narrow band, one can treat it as a monochromatic source, then it is not necessary to make an expansion to the wave-number k_p ; otherwise, for example, the pump field consists of a train of short pulses, the band can be very large, therefore, one obtains

$$k_p(\omega) \simeq k_p(\omega_p) + (\omega - \omega_p)/u_p, \quad u_p = 1/(\partial k_p(\omega)\partial\omega)|_{\omega_p}. \quad (1.43)$$

So in the sinc function, one can define two effective coherent times:

$$T = (1/4u_\alpha + 1/4u_\beta - 1/2u_p)l, \quad T_c = (1/4u_\alpha - 1/4u_\beta)l. \quad (1.44)$$

In most cases, $T_c \ll T$. Subsequently, the response function $\mathcal{M}(\omega_m, \omega_n)$ which represents phase mismatch can be expressed as

$$\mathcal{M}(\omega_m, \omega_n) = \text{sinc}[(\omega_m + \omega_n - \omega_p)T - (\omega_m - \omega_n - \omega_{\alpha\beta})T_c], \quad (1.45)$$

where $\omega_{\alpha\beta} = \omega_\alpha - \omega_\beta$.

C "Quantumness" of light states

In 1935, Einstein, Podolsky and Rosen wrote a paper in questioning the completeness of quantum mechanics [Einstein1935]. The state introduced in their paper is named as EPR state, with which, intuitively speaking, a measurement on one part can instantly have an influence on the other regardless the spatial separation. This novel and mysterious property, in the name of (quantum) entanglement, inspired Schrödinger to the idea of cat state which was aiming at the classical-quantum boundary. Now it is commonly known that such property comes from inseparable quantum states, and an inseparable state is called an entangled state. The questions on separability as well as the quantumness are two main concerns to have a deeper knowledge of such states.

In a bipartite system, a separable state is defined as

$$\rho_{sep} = \sum_k p_k \rho_{\alpha,k} \otimes \rho_{\beta,k}, p_k \in \mathbb{R}^+, \quad (1.46)$$

where α, β are indices for two different parts. The density matrix of this separable state is a convex combination of some factorable density matrices pairs. To determine whether a state being entangled or not is a NP hard problem. There are some criteria for entangled state, for example, the Peres-Herodecki criterion and its variant-Simon's criterion, geometric measure [Simon2000]. We will detail it in chapter 6.

On the other hand, the data from local measurements on an entangled state usually have some correlations. The quantumness of correlations between parts is a key concept to the quantumness of entangled state. A popular measure of such quantum correlations is called quantum discord which is the mismatch of two mutual information, because of the statistical nature of quantum states. There are some other alternative measures, such like measurement induced disturbance, quantum deficits. In multimode quantum optics, the criteria of interests see [Trep2005, FabreQO].

C.1 Separability

Different separability criteria have been proposed. A simple situation for the bipartite separability problem is the pure state case, in which one can take advantage of the Schmidt decomposition.

C.1.1 Schmidt decomposition of a pure entangled state

Different separability criteria have been proposed. A simple situation for the bipartite separability problem is the pure state case, in which one can take advantage of the Schmidt decomposition. Any given pure state $|\Phi\rangle$ in Hilbert space $\mathcal{H}_\alpha \otimes \mathcal{H}_\beta$ with two sets of orthonormal bases $\{|\phi_{\alpha,m}\rangle\}_{m=1}^{\dim \mathcal{H}_\alpha}$ and $\{|\phi_{\beta,n}\rangle\}_{n=1}^{\dim \mathcal{H}_\beta}$ can be expressed as

$$|\Phi\rangle = \sum_{mn} c_{mn} |\phi_{\alpha,m}\rangle \otimes |\phi_{\beta,n}\rangle. \quad (1.47)$$

Here, $\dim \mathcal{H}_\alpha, \dim \mathcal{H}_\beta$ are the dimensions of subspaces $\mathcal{H}_\alpha, \mathcal{H}_\beta$, their value can be infinitely large. For a discussion of the entanglement, it is helpful to make use of the Schmidt decomposition. Mathematically, it is always possible to decompose the state in a simplest

way:

$$|\Phi\rangle = \sum_m \sqrt{\lambda_m} |f_m^\alpha\rangle \otimes |f_m^\beta\rangle, \quad \langle f_m^\mu | f_n^\mu \rangle = \delta_{mn}, \quad \mu = \alpha, \beta, \quad (1.48)$$

in which $\{\sqrt{\lambda_m}\}$ are called Schmidt coefficients and the minimum value of the dimensions, $\min\{\dim \mathcal{H}_\alpha, \dim \mathcal{H}_\beta\}$, is defined as the Schmidt rank or Schmidt number. The normalization condition imposes $\sum_m \lambda_m = 1$. Because of this constraint, one can know the main terms in the decomposition form from the Schmidt coefficients. A quantity, which is called Schmidt number sometimes or cooperativity parameter κ is introduced as

$$\kappa = \frac{1}{\sum_m \lambda_m^2}. \quad (1.49)$$

To avoid the confusions of the names, in this thesis, κ is only called cooperativity parameter. The cooperativity parameter can also be introduced as

$$\kappa = [\text{Tr}_b(\text{Tr}_a \rho)^2]^{-1}. \quad (1.50)$$

Schmidt rank and cooperativity parameter are two important complementary quantities for describing the complexity of entanglement. The Schmidt rank describes the abundance of modes that are in correlation while the cooperativity parameter reflects the homogeneity in the distribution of coefficients $\sqrt{\lambda_m}$.

It is also worth mentioning that some entangled states may not be genuinely entangled in bosonic system. The concept of entanglement sometimes relies on the choice of bases. For example, a single photon state $|\text{single}\rangle = (|1 : \omega_1\rangle \otimes |0 : \omega_2\rangle \otimes |0 : \omega_3\rangle \otimes |0 : \dots\rangle + |0 : \omega_1\rangle \otimes |1 : \omega_2\rangle \otimes |0 : \omega_3\rangle \otimes |0 : \dots\rangle) / \sqrt{2}$, according to the definition, we know the Schmidt rank of this state is 2, the cooperativity parameter reads $\kappa = 2 > 1$. One may conclude that such a state is an entangled state. However, if one introduces two new annihilation operators: $\hat{a}_+ = [\hat{a}(\omega_1) + \hat{a}(\omega_2)] / \sqrt{2}$, $\hat{a}_- = [\hat{a}(\omega_1) - \hat{a}(\omega_2)] / \sqrt{2}$, then in the new bases, the state $|\text{single}\rangle = |1 : +\rangle \otimes |0 : -\rangle \otimes |0 : \omega_3\rangle \otimes |0 : \dots\rangle$ is a factorable state, accordingly, the Schmidt rank is 1, which is equal to the cooperativity parameter. We conclude that both the magnitudes of Schmidt rank and of the cooperativity parameter are not unique in a bosonic system, they are strongly related to the choice of modes. So, in order to make κ be a good measure, one need define precisely the set of photon modes.

C.1.2 Simon's criterion and Mandel matrix

In quantum physics, a state is described by an abstract element in a Hilbert space. What one measures are its coefficients by projecting the state on all possible elements of an orthonormal basis rather than the abstract state itself which should not be observed by any type of apparatus. However, by performing measurements on the system, one can obtain enough data of its coefficients with a given representation of the state to reconstruct the abstract state. Then, one can apply some criteria to check whether the state is classical, non-classical or highly non-classical. In some simple systems, for example a two qubits system, this paradigm works quite well. In multimode quantum optics, in which each state usually contains many modes, to reconstruct an abstract state, in general, is very difficult. Even when one knows the exact expression of the state, it is still not so convenient to use the criteria such as the PPT criterion or its variant in quantum optics, the Simon's version of PPT criterion [Simon2000]. Some alternative criteria directly based on measurements

can be helpful to show some nonclassicality properties. Among them all, the other Simon criterion is one of the widely-used one. The basic idea is to introduce a covariance matrix of the field quadratures $\hat{p}_m = i\sqrt{\hbar/2}(\hat{a}_m^\dagger - \hat{a}_m)$, $\hat{q}_m = \sqrt{\hbar/2}(\hat{a}_m^\dagger + \hat{a}_m)$. The simplest form of the covariance matrix which is of 2×2 dimension, coming from the single mode state, reads

$$\begin{pmatrix} (\Delta\hat{q})^2 & \Delta(\hat{p}, \hat{q}) \\ \Delta(\hat{q}, \hat{p}) & (\Delta\hat{p})^2 \end{pmatrix} \quad (1.51)$$

A bipartite two-photon pure state can be regarded as a post-selection from the state which is obtained from an evolution operator that is determined by a quadratic Hamiltonian acting on the vacuum state. Before selection, it should be a multimode squeezing state. If the interaction strength is weak, then the two-photon part is the key component, which implies the squeezing properties of the state before selection can be shown in the covariance matrix of the two-photon state.

The variances of field quadratures of a coherent state is $\hbar/2$. Therefore a state is squeezed, when either of its field quadrature variances is smaller than this value (of course not both). It can be extended to multimode states, see details of the invariant squeezing criterion in [Arvind1995].

The quantum optical nonclassicality of multimode state is represented by the non-positive diagonal weight function in Glauber-Sudarshan representation [Ivan2011]. In general, the Mandel-type nonclassicality [Mandel1979], which is weaker than the general notation of nonclassicality, is a simpler to be formulated in light of the Mandel matrix. For example, with a bi-partite two-mode state ρ , there are two annihilation operators \hat{a}, \hat{b} and two corresponding creation operators. One can introduce two operator-value vectors as

$$\hat{C} = \begin{pmatrix} \hat{a}^\dagger \\ \hat{b}^\dagger \end{pmatrix} \otimes \begin{pmatrix} \hat{a} \\ \hat{b} \end{pmatrix} = \begin{pmatrix} \hat{a}^\dagger \hat{a} \\ \hat{a}^\dagger \hat{b} \\ \hat{b}^\dagger \hat{a} \\ \hat{b}^\dagger \hat{b} \end{pmatrix}; \quad (1.52)$$

$$\hat{C}^\dagger = (\hat{a}^\dagger \hat{a}, \hat{b}^\dagger \hat{a}, \hat{a}^\dagger \hat{b}, \hat{b}^\dagger \hat{b}), \quad (1.53)$$

then the Mandel matrix is defined as

$$M = \text{Tr} \left[\rho \begin{pmatrix} 1 & \hat{C}^\dagger \\ \hat{C} & \hat{C}^\dagger \hat{C} \end{pmatrix} \right], \quad (1.54)$$

where the two $:$ symbols impose all the creation and annihilation operators in between should be normally ordered. According to [Ivan2011], one has the following conclusions:

$$\rho \text{ is quantum optical classical} \Rightarrow M \geq 0; \quad (1.55a)$$

$$M \not\geq 0 \Leftrightarrow \rho \text{ is quantum optical nonclassical.} \quad (1.55b)$$

The Mandel nonclassicality can be regarded as the photon statistics effect. This method can be extended and then applied to multimode states, see [Ivan2011].

C.2 Quantum discord

In classical information theory, there are two equivalent definitions of the mutual information. In quantum theory, the mutual information of a bipartite state $\rho_{\alpha\beta}$ is defined

as

$$I(\alpha : \beta) = S_\alpha + S_\beta - S_{\alpha\beta}, \quad (1.56)$$

where $S_\alpha, S_\beta, S_{\alpha\beta}$ are von Neumann entropies of reduced density matrices ρ_α, ρ_β and the total density matrix $\rho_{\alpha\beta}$. The classical correlation is defined as the maximal gained information of one subsystem by performing a POVM measurements $E_m^\alpha = M_{\alpha,m}^\dagger M_{\alpha,m}$ on the other. Its mathematical formula reads

$$J(\beta|\alpha) = S_\beta - S_{\beta|\alpha}, \quad (1.57)$$

where the conditional entropy $S_{\beta|\alpha}$ is the minimum of an average on all the entropies of all possible POVMs

$$S_{\beta|\alpha} = \min_{\{E_m^\alpha\}} S_{\beta|\{E_m^\alpha\}}. \quad (1.58)$$

In quantum physics, the two definitions of mutual information give rise to different results. This mismatch,

$$\mathcal{D}_{\alpha\beta} = I(\alpha : \beta) - J(\beta|\alpha), \quad (1.59)$$

is called quantum discord. This quantity is always positive, since the quantum mutual information $I(\alpha : \beta)$ represents the total one while $J(\beta|\alpha)$ is an information based on measurements and therefore a reduction of information from an increase of entropy originated in measurements. In classical physics, this difference is definitely vanishing, therefore, a non-vanishing quantum discord indeed reveals some genuine quantumness of a state. It has also been shown that some separable states can have non-vanishing discord, that is, classical communication can give rise to quantum correlation. In this sense, we conclude that [1. quantum discord is a measure of quantum correlation rather than entanglement;](#) and [2. separability does not imply the absence of quantum effect.](#)

D Single/Multi-mode criterion

The concept of photon depends on the choice a mode-function, and a mode-function is an element in a linear space. As a result, for a given light system, the state can be regarded as a description of a collective behaviors which can be discussed in the contents of mode transformation.

D.1 Mode transformation

The concept of photon is strongly related to the mode function, this function can be expanded in a basis. A state with complicated superposition of several modes may be written in a simpler form by choosing proper basis. From one mode basis to another, it always permits a linear transformation. Let $\{a_m^\dagger\}$ and $\{b_m^\dagger\}$ be two complete sets of creation operators that are concomitant with the bases. So, in general, one has the transformation relations

$$\hat{b}_m^\dagger = \sum_{\ell} U_{m\ell} \hat{a}_{\ell}^\dagger. \quad (1.60)$$

However, the statistical nature of bosonic system is inherent, should not be changed by the representations and mode transformations. Such transformation is symplectic. The canonical commutation relations follow

$$[\hat{b}_m, \hat{b}_n^\dagger] = \sum_{\ell, \ell'} (U_{m\ell})^* U_{n\ell'} [\hat{a}_{\ell}, \hat{a}_{\ell'}^\dagger] = \sum_{\ell} U_{n\ell} (U^\dagger)_{\ell m} = \delta_{mn}. \quad (1.61)$$

All these relations are equivalent to

$$UU^\dagger = \mathbb{I}. \quad (1.62)$$

Therefore, any such transformation is unitary symplectic.

Because of this transformation relations, a multimode state in one basis may be a single mode state in another basis. The non-fluctuating classical field can be described by only one mode-function, while for some quantum light states and therefore the quantum field, there exist some photon state that cannot be fully described by only one mode-function, see examples in the intrinsic multimode states. That is, the multi-modality of a non-fluctuating light field is a property of non-classicality. To find minimum basis to describe an optical state is not only fundamentally important for the classification of the state therefore the complexity of the structure of a state, but also convenient for analysis and calculations. The following introduction to intrinsic single/multi- mode state will follow [Trep2005].

D.2 Intrinsic single-mode quantum light

In order to give a proper definition of a single mode state, let us firstly consider a state that has already written in a single mode u_m form

$$|\text{single mode}\rangle = \cdots |0\rangle \otimes |\psi : u_m\rangle \otimes |0\rangle \cdots. \quad (1.63)$$

We denote the annihilation operator that is in mode u_m by \hat{A}_m and other annihilation operators corresponding to other orthogonal vector in the basis by $\hat{A}_{\perp, n}$. So, we know

$$\hat{A}_m |\text{single mode}\rangle \neq 0; \quad \hat{A}_{\perp, n} |\text{single mode}\rangle = 0. \quad (1.64)$$

From the previous representation theory, we know that any annihilation operator can be expressed in terms of \hat{A}_m and other $\hat{A}_{\perp,n}$. So any operator acting on the single mode state $|\text{single mode}\rangle$ gives rise to a same final state. One concludes that [Treps2005]:

A quantum state of the field is single mode if and only if the action on it of all the annihilation operators of a given basis gives proportional vectors.

D.3 Multi-mode quantum light

A state of light is said multi-mode, if it is *not* single mode. So, not all the vectors generated from the action on a multi-mode state of all the annihilation operators are proportional to each other. We characterize such a state by the rank of these induced vectors, say degree n , (the rank of a single mode light is equal to 1) and call it an n -mode state. If, by coincidence, $\{u_1, u_2, \dots, u_n\}$ are n modes that are sufficient to describe the state, then these modes are called minimum basis.

Though the single/multi mode state is introduced in the case of pure state, these ideas can be generalized to mixed states.

D.4 Some examples

In this part, we will first consider two examples from [FabreQO]. Let's consider two annihilation operators \hat{a}_1, \hat{a}_2 of frequencies ω_1, ω_2 respectively. A first example is a state defined as

$$|\mathbb{I}\rangle = (|2 : \omega_1, 0 : \omega_2\rangle + |0 : \omega_1, 2 : \omega_2\rangle)/2 + |1 : \omega_1, 1 : \omega_2\rangle/\sqrt{2}. \quad (1.65)$$

It is a two-mode entangled state in common sense. However, one can verify directly that $\hat{a}_1|\mathbb{I}\rangle = \hat{a}_2|\mathbb{I}\rangle = (|1 : \omega_1, 0 : \omega_2\rangle + |0 : \omega_1, 1 : \omega_2\rangle)/\sqrt{2}$, thus the rank of the state is 1. We can construct two new annihilation operators: $\hat{A}_{\pm} = (\hat{a}_1 \pm \hat{a}_2)/\sqrt{2}$, so the state

$$\begin{aligned} |\mathbb{I}\rangle &= (\hat{a}_1^{\dagger 2} + 2\hat{a}_1^{\dagger}\hat{a}_2^{\dagger} + \hat{a}_2^{\dagger 2})|0\rangle/2\sqrt{2} = \frac{\hat{A}_+^{\dagger 2}}{\sqrt{2}}|0\rangle \\ &= |2 : +\rangle \otimes |0, -\rangle. \end{aligned} \quad (1.66)$$

The only non-vanishing induced vector is $\hat{A}_+|\mathbb{I}\rangle = \sqrt{2}|1 : +\rangle \otimes |0, -\rangle$, therefore state $|\mathbb{I}\rangle$ is an intrinsic single-mode state. From this example, we conclude that **1. a two-mode entangled state at the first sight can be an intrinsic single-mode state** and **2. entanglement is not a genuine property in some bosonic system.**

Consider another state that is a tensor product of two single-photon states at frequencies ω_1 and ω_2 respectively, $|11\rangle = |1 : \omega_1, 1 : \omega_2\rangle$. The action of both annihilation operators \hat{a}_1, \hat{a}_2 gives rise to

$$\hat{a}_1|11\rangle = |0 : \omega_1, 1 : \omega_2\rangle, \quad \hat{a}_2|11\rangle = |1 : \omega_1, 0 : \omega_2\rangle. \quad (1.67)$$

The above states are orthogonal to each other and the two states together span a 2D space. So the rank of induced vectors is 2, and state $|11\rangle$ is a 2-mode state.

Now let's consider a general bipartite pure two-photon state $|\mathbb{I}\rangle = \sum_{mn} c(\omega_m, \omega_n)|1 : \omega_m, \alpha\rangle \otimes |1 : \omega_n, \beta\rangle$ where the coefficients $c(\omega_m, \omega_n)$ are normalized as $\sum_{mn} |c(\omega_m, \omega_n)|^2 =$

1. By performing any annihilation operation $\hat{a}_{\alpha,k}$ or $\hat{a}_{\beta,q}(k, q \in \mathbb{Z})$ on a subsystem, we have

$$\hat{a}_{\alpha,k}|\mathbb{I}\rangle = |0 : \alpha\rangle \otimes \left(\sum_n c(\omega_k, \omega_n) |1 : \omega_n, \beta\rangle \right); \quad (1.68)$$

$$\hat{a}_{\beta,q}|\mathbb{I}\rangle = \left(\sum_m c(\omega_m, \omega_q) |1 : \omega_m, \alpha\rangle \right) \otimes |0 : \beta\rangle. \quad (1.69)$$

Both the two vectors, in general, are not equal to 0. The inner product of the two vectors in Eqs. (1.68-1.69) yields

$$(\langle \mathbb{I} | \hat{a}_{\alpha,k}^\dagger)(\hat{a}_{\beta,q} | \mathbb{I} \rangle) = 0. \quad (1.70)$$

That is, the two vectors are independent, and such a two-photon state has at least 2 modes. The orthogonality of the two vectors are valid for any integer k, q , therefore, the intrinsic mode number of the photon states should be equal to the minimum of the number of modes in both subsystems. Note that the matrix \mathbf{c} whose element at position (m, n) being $c(\omega_m, \omega_n)$, admits the following Schmidt decomposition

$$c(\omega_m, \omega_n) = \sum_{k=1}^{N_{sch}} \sqrt{\lambda_k} \phi_k(\omega_m) \psi_k^*(\omega_n), \quad (1.71)$$

where N_{sch} is the Schmidt rank. Then one finds possible bases are

$$\{|1 : \phi_k, \alpha\rangle = \sum_m \phi_k(\omega_m) |1 : \omega_m, \alpha\rangle\}_{k=1}^{N_{sch}}; \quad \{|1 : \psi_k^*, \beta\rangle = \sum_n \psi_k^*(\omega_n) |1 : \omega_n, \beta\rangle\}_{k=1}^{N_{sch}}, \quad (1.72)$$

and the intrinsic number of modes of a two-photon multimode state is twice large than the Schmidt number, that is the two-photon state $|\mathbb{I}\rangle$ is an intrinsic $(2N_{sch})$ -mode states.

For example, in cascade two-photon state or type II SPDC two-photon state discussed in the last chapter, we see in both cases the Schmidt numbers are infinitely large, so, the two two-photon states has infinitely many intrinsic mode.

However, it is not universally true that the intrinsic mode number is twice larger than the Schmidt number of the state. For example, consider a similar quantum superposed coherent state

$$|\text{QMC}\rangle = \sum_{mn} c(\omega_m, \omega_n) |z : \omega_m, \alpha\rangle \otimes |z : \omega_n, \beta\rangle, |z| \gg 1. \quad (1.73)$$

Though the orthogonality of the counterparts can still be valid, these vectors such as

$$\sum_m \phi_k(\omega_m) |z : \omega_m, \alpha\rangle, \quad (1.74a)$$

$$\sum_m \psi_k^*(\omega_m) |z : \omega_m, \beta\rangle, \quad (1.74b)$$

cannot represent single-mode vectors unless both functions $\phi_k(\omega_m)$ and $\psi_k^*(\omega_m)$ attain non-zero value only at single-frequencies. For example, a product coherent state $|z : \omega_1, \alpha\rangle \otimes |z : \omega_2, \beta\rangle$ is an intrinsic single-mode state, the rank of mode is less than $2N_{sch} = 2$.

When we consider a general $|\text{QMC}\rangle$ state such that the value of the function $\phi_k(\omega_m)$ or $\psi_k^*(\omega_m)$ is non-zero almost everywhere, then the ranks of modes of the vectors in (1.74) are really large. For example, consider a state

$$\left(\sum_{m=1}^M \phi_1(\omega_m) |z : \omega_m, \alpha\rangle \right) \otimes \left(\sum_{n=1}^N \psi_1^*(\omega_n) |z : \omega_n, \beta\rangle \right), \quad (1.75)$$

whose Schmidt rank is 1, but its rank of modes can be as large as $M \times N$, which is much larger than 2 in general.

We stress that the relation

$$\text{rank of modes} = 2N_{sch}, \quad (1.76)$$

is valid for bipartite pure two-photon states, and the cascade two-photon state as well as the type II SPDC two-photon state are intrinsic infinitely-many-mode states because $N_{sch} = \infty$ which will be shown in the last chapter of this thesis.

Two-photon quantum optics in one atom

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There are fundamental quantum statistical properties in a bipartite quantum system, such as entanglement or non-locality, but even in a system like an atom which is not bipartite, genuine quantum correlations play important role, such as the quantum correlation in a divisible system [Lapkiewicz2011]. In quantum optics, nowadays, scientists can produce various quantum light sources with complex correlation structures. A basic way to characterize these correlations is to use the correlation function $g^{(n)}$ which is also an important factor in n -photon transition process. With these quantum light sources, it is then also possible to take advantage of quantum coherent control to the atom-light interaction process to give rise to strongly constructive as well as destructive quantum interference in the probability of various processes such as the excitation of a target atom. These processes can then be helpful in spectroscopy.

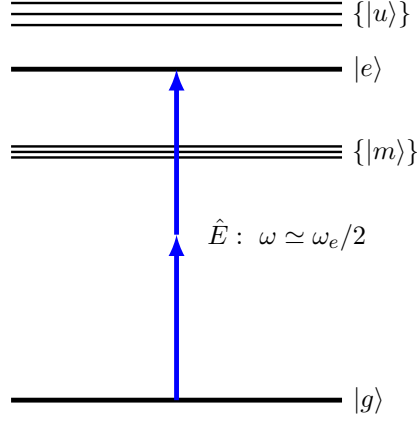


Figure 2.1: The interaction configuration. In this model, any single-photons are far away from resonance to all energy levels. Possible two-photon resonances occurs only between the ground state $|g\rangle$ and an excited state $|e\rangle$. In this figure, $\{|u\rangle\}, \{|m\rangle\}$ represent two collections of other levels

A Model

Let's consider the interaction between an one-dimensional quantum field and a single atom that is located at position $z = 0$ (see the configuration in Figure 2.1). The quantum field is defined in Eq.(1.19). Under long wavelength approximation, the Hamiltonian of field-atom dipole interaction [GAFIQO] reads

$$\hat{V} = -\hat{\mathbf{d}} \cdot (\hat{\mathbf{E}}^{(+)} + \hat{\mathbf{E}}^{(+)\dagger}), \quad (2.1)$$

where we have removed the z -dependence in field operators and $\hat{\mathbf{d}}$ is atomic dipole operator. Combined with total free Hamiltonian \hat{H}_{free} of atom and field, the evolution of the system is described by

$$\hat{U}(t) = \exp[-i(\hat{H}_{free} + \hat{V})t/\hbar]. \quad (2.2)$$

The atom is initially in the ground state $|g\rangle$ and photons at time $t = 0$ are described by density matrix ρ . The density matrix at a later time t is determined by $\hat{U}(t)(|g\rangle\langle g| \otimes \rho)\hat{U}^\dagger(t)$. The physics of on resonance interaction between a two-level atom and a monochromatic field has been thoroughly investigated [AllenORTLA], in particular in the context of Jaynes-Cummings model [Jaynes1963].

In this chapter, we consider two-photon excitation phenomena with a single atom in which noticeable two photon excitations only occur between ground state and an excited state $|e\rangle$. Single photon excitations are forbidden between this energy level pair because of selection rules. The probability to find the atom in state $|e\rangle$ is therefore equal to the one of two-photon excitation. It reads

$$P = \text{Tr}[\langle e|\hat{U}(t)(|g\rangle\langle g| \otimes \rho)\hat{U}^\dagger(t)|e\rangle] = \text{Tr}[\langle g|\hat{U}^\dagger(t)|e\rangle\langle e|\hat{U}(t)|g\rangle\rho]. \quad (2.3)$$

It most cases, it is really hard to find an analytical solution for the evolution operator $\hat{U}(t)$. The difficulties come from the fact that \hat{H}_{free}, \hat{V} are not commuting with each

other, even in an ideal two-level atom model. Conventionally, the evolution operator $\hat{U}(t)$ is usually expanded as Dyson series

$$\hat{U}(t) = e^{-i\hat{H}_{free}t/\hbar} \left[\mathbb{I} - i\hbar^{-1} \int_0^t d\tau \hat{V}(\tau) - \hbar^{-2} \int_0^t d\tau \int_0^{\tau_1} d\tau_1 \hat{V}(\tau) \hat{V}(\tau_1) + \dots \right], \quad (2.4)$$

All perturbative terms in Dyson series have a same form

$$\hat{V}(\tau) = e^{i\hat{H}_{free}\tau/\hbar} \hat{V} e^{-i\hat{H}_{free}\tau/\hbar} = -e^{i\hat{H}_{free}\tau/\hbar} \hat{\mathbf{d}} \cdot (\hat{\mathbf{E}}^{(+)} + \hat{\mathbf{E}}^{(+)\dagger}) e^{-i\hat{H}_{free}\tau/\hbar}. \quad (2.5)$$

According to Baker-Compel-Hausdorff formula, one has

$$e^{i\hat{H}_{free}\tau/\hbar} (\hat{\mathbf{E}}^{(+)} + \hat{\mathbf{E}}^{(+)\dagger}) e^{-i\hat{H}_{free}\tau/\hbar} = \mathbf{e}_0 \hat{E}^{(+)}(\tau) + h.c., \quad (2.6)$$

which gives rise to time-dependent quantum field,

$$\hat{E}^{(+)}(t) = i \sum_m \mathcal{E}_m \hat{a}_m e^{-i\omega_m t}. \quad (2.7)$$

Now let's consider a two-photon excitation process. The leading effect of such process is described by the second term $U^{(2)}$ in Dyson series,

$$\langle e|U^{(2)}|g\rangle = \langle e|e^{-i\hat{H}_{free}t/\hbar}|e\rangle \int_0^t d\tau \int_0^{\tau} d\tau_1 \mathcal{F}_{eg}(\tau, \tau_1) [\hat{E}^{(+)}(\tau) + h.c.] [\hat{E}^{(+)}(\tau_1) + h.c.], \quad (2.8)$$

where the factor of atomic part reads

$$\mathcal{F}_{eg}(\tau, \tau_1) = -\hbar^{-2} \sum_m e^{i\omega_e\tau} e^{i\omega_m(\tau_1-\tau)} \langle e|\hat{\mathbf{d}} \cdot \mathbf{e}_0|m\rangle \langle m|\hat{\mathbf{d}} \cdot \mathbf{e}_0|g\rangle. \quad (2.9)$$

If we take atomic decoherence into account, and the width of atomic state $|m\rangle$ is denoted by γ_m , then

$$\mathcal{F}_{eg}^\gamma(\tau, \tau_1) = -\hbar^{-2} \sum_m e^{\gamma_e(\tau-t)} e^{i\omega_e\tau} e^{(\gamma_m+i\omega_m)(\tau_1-\tau)} \langle e|\hat{\mathbf{d}} \cdot \mathbf{e}_0|m\rangle \langle m|\hat{\mathbf{d}} \cdot \mathbf{e}_0|g\rangle. \quad (2.10)$$

As the transition dipole elements $\langle e|\hat{\mathbf{d}} \cdot \mathbf{e}_0|m\rangle \langle m|\hat{\mathbf{d}} \cdot \mathbf{e}_0|g\rangle$ are largely independent of frequency, we then remove the atomic projector $|m\rangle \langle m|$ in it and rewrite the $\mathcal{F}_{eg}(\tau, \tau_1)$ as

$$\mathcal{F}_{eg}(\tau, \tau_1) = -\hbar^{-2} e^{i\omega_e\tau} \left(\sum_m e^{i\omega_m(\tau_1-\tau)} \right) \langle e|\hat{\mathbf{d}} \cdot \mathbf{e}_0 \hat{\mathbf{d}} \cdot \mathbf{e}_0|g\rangle. \quad (2.11)$$

In a real atomic system, usually it contains lots of (in fact infinitely many) energy levels, therefore, we have infinitely many terms of phases to make temporal interferences since the index m in the sum runs over all positive integer. when $\tau \neq \tau_1$, the sum gives a vanishing net value, because such interference-induced decoherence [Zurek1991, Dürr1998] originates in the fact that lots of phase functions $e^{i\omega_m(\tau_1-\tau)}$ are oscillating with small periods at high frequencies. When $\tau = \tau_1$, the sum diverges. In general, we approximate the sum as

$$\sum_m e^{i\omega_m(\tau_1-\tau)} \simeq \frac{L}{2\pi c} \int_{\mathbb{R}^+} d\omega_m e^{i\omega_m(\tau_1-\tau)} = \frac{L}{2c} \delta(\tau_1 - \tau). \quad (2.12)$$

The factor 2 in the denominator comes from the fact that frequency must be positive. Now we get

$$\langle e|U^{(2)}|g\rangle = -\frac{Le^{-i\hat{H}_{free}t/\hbar}}{4c\hbar^2} \langle e|\hat{\mathbf{d}} \cdot \mathbf{e}_0 \hat{\mathbf{d}} \cdot \mathbf{e}_0|g\rangle \int_0^t d\tau e^{i\omega_e\tau} [\hat{E}^{(+)}(\tau) + h.c.]^2. \quad (2.13)$$

where we have used the following identity

$$\int_{-\infty}^t d\tau \delta(t - \tau) = 1/2. \quad (2.14)$$

As a result, the transition probability is

$$P = P_{eg} \iint_0^t d\tau d\tau_1 e^{i\omega_e(\tau - \tau_1)} G^{(2)}(\tau_1, \tau), \quad P_{eg} \equiv \frac{L^2}{16c^2\hbar^4} |\langle e | \hat{\mathbf{d}} \cdot \mathbf{e}_0 \hat{\mathbf{d}} \cdot \mathbf{e}_0 | g \rangle|^2; \quad (2.15)$$

where the kernel reads

$$G^{(2)}(\tau_1, \tau) = \langle [\hat{E}^{(+)}(\tau_1) + h.c.] [\hat{E}^{(+)}(\tau_1) + h.c.] [\hat{E}^{(+)}(\tau) + h.c.] [\hat{E}^{(+)}(\tau) + h.c.] \rangle. \quad (2.16)$$

In the case of two photon absorption, the leading transition probability rate is described by $G_{Ladder}^{(2)}$ function

$$G_{Ladder}^{(2)}(t_1, t_2) = \langle \hat{E}^{(+)\dagger}(t_1) \hat{E}^{(+)\dagger}(t_1) \hat{E}^{(+)}(t_2) \hat{E}^{(+)}(t_2) \rangle, \quad (2.17)$$

One can take into account the atomic decay rate Γ of the excited state $|e\rangle$ in the following way [Mollow1968, Gea-Banacloche1989]

$$w_2 = 2|g|^2 \int_{\mathbb{R}} d\tau e^{2i\omega_e t - \Gamma|\tau|} G^{(2)}(-\tau, -\tau, \tau, \tau). \quad (2.18)$$

In Raman type two-photon transitions, the key kernel is

$$G_{Raman}^{(2)}(t_1, t_2) = \langle \hat{E}^{(+)\dagger}(t_1) \hat{E}^{(+)}(t_1) \hat{E}^{(+)\dagger}(t_2) \hat{E}^{(+)}(t_2) \rangle, \quad (2.19)$$

Though the two-photon transition processes with Raman type configuration (Quantum Raman spectroscopy) are quite different from the one with Ladder type configuration, we will not consider this interesting topic in this thesis which is not central in the present discussion. In the following of this chapter, we will give a brief introduction to two-photon absorption process in a two-level atom in the context of quantum coherent control.

B Two-photon absorption in ladder configuration

In this part, we will give a brief introduction to quantum two-photon absorption phenomena such like dark pulse, entanglement-induced transparency with continuous frequency modes.

If photons are initially in a mixed state, then the transition probability is a sum of the weighted contributions from all pure density matrices parts. Since any those contributions are positive, the minimum transition probability from mixed state should be not less than the one from pure states. So, a pure state which gives rise to the minimum two-photon excitation probability is what we are interested in here.

The correlation function $G^{(2)}$ of a generic pure quantum state $|\psi\rangle$ can be expanded as a sum of 16 terms. With an interaction of ladder configuration, only one term gives rise to two-photon absorption. It reads

$$G_L^{(2)}(\tau_1, \tau) = \langle \psi | \hat{E}^{(+)\dagger}(\tau_1) \hat{E}^{(+)\dagger}(\tau_1) \hat{E}^{(+)}(\tau) \hat{E}^{(+)}(\tau) | \psi \rangle, \quad (2.20)$$

Here we will consider two kinds of quantum states: 1. a class of most classical states, multimode coherent states, $|\alpha\rangle = [\otimes_m \hat{D}(\alpha f_m, a_m)]|0\rangle$ where $f_m \equiv f(\omega_m)$ is a normalized coefficient of a generic mode distribution $\sum_m |f_m|^2 = 1$; 2. two-photon states $|\mathbb{I}\rangle = \sum_{mn} c_{mn} |1 : \omega_m; 1 : \omega_n\rangle$ where coefficients $c_{mn} \equiv c(\omega_m, \omega_n)$ are also normalized $\sum_{mn} |c_{mn}|^2 = 1$. The multimode coherent state $|\alpha\rangle$ is an intrinsic single-mode state.

B.1 Dark pulse

By sending a general pulse into an atomic system, in general, the atoms will be excited more or less if the transition is allowed, then one can observe such transition phenomena via some widely used techniques. Interestingly, for a given pulse spectrum, the previously allowed transition can be sharply suppressed by adjusting the phase distributions. As a result, one may even find no transitions during a very long time owing to a very small transition probability. Such a corresponding pulse is called dark pulse [Meshulach1998].

Let's consider the minimum transition probability of multimode coherent state $|\alpha\rangle$ in this part. The correlation function $G_L^{(2)}$ can be factorized as

$$G_{L,|\alpha\rangle}^{(2)}(\tau_1, \tau) = \mathcal{E}^4 |\alpha|^4 \left(\sum_{jk} f_j^* f_k^* e^{i(\omega_j + \omega_k)\tau_1} \right) \left(\sum_{mn} f_m f_n e^{-i(\omega_m + \omega_n)\tau} \right), \quad (2.21)$$

which leads to

$$P(t) = P_{eg} \mathcal{E}^4 |\alpha|^4 \left| \sum_{mn} f_m f_n \int_0^t d\tau \exp[-i(\omega_m + \omega_n - \omega_e)\tau] \right|^2. \quad (2.22)$$

We notice the varying phase in the integral

$$\left| \sum_{mn} f_m f_n \int_0^t d\tau e^{-i(\omega_m + \omega_n - \omega_e)\tau} \right|^2 = \left| \sum_{mn} f_m f_n \frac{\sin[(\omega_m + \omega_n - \omega_e)t/2]}{(\omega_m + \omega_n - \omega_e)/2} \right|^2,$$

reveals the fact that when time t is large, the two-photon transition can only occur in a cooperative way $\omega_m + \omega_n = \omega_e$. The stationary two-photon transition can then occur by

destroying two photons at a pairwise anti-correlated frequencies. Replacing the double sums by double integrals and making Dirac delta function approximation, we have a following convolution function [Meshulach1998]:

$$P_\infty = \frac{P_{eg}\mathcal{E}^4|\alpha|^4L^4}{16\pi^2c^4} \left| \int_0^{\omega_e} d\omega_m f(\omega_m) f(\omega_e - \omega_m) \right|^2. \quad (2.23)$$

$f(\omega)$ is a complex number which can be rewritten in the polar form $f(\omega) = A_f(\omega)e^{i\phi_f(\omega)}$. If the amplitude is a slowly varying function in contrast to the phase function and if $\phi_f(\omega) \neq -\phi_f(\omega_e - \omega)$, then the integral is vanishing in general. This is because if a photon at a frequency is ready to excite the atom, then a photon at a close neighboring frequency is prone to de-excite back to ground state, which in general gives rise to no net excitation accumulation. Let's consider here a special class of distribution, for any frequency ω , $f(\omega)$ is symmetric about the line $\omega = \omega_e/2$, $f(\omega) = f(\omega_e - \omega)$, the convolution function is now simplified to

$$\int_0^{\omega_e} d\omega_m f(\omega_m) f(\omega_e - \omega_m) = 2 \int_0^{\omega_e/2} d\omega_m A_f^2(\omega_m) e^{2i\phi(\omega_m)}. \quad (2.24)$$

For a given power spectrum $A_f(\omega_m)$, the variation principle imposes that the extrema of functionals of $\phi_f(\omega_m)$ satisfies

$$e^{4i\phi(\omega_m)} = \frac{\int_0^{\omega_e/2} d\omega A_f^2(\omega) e^{2i\phi(\omega)}}{\int_0^{\omega_e/2} d\omega A_f^2(\omega) e^{-2i\phi(\omega)}}. \quad (2.25)$$

On the right hand side, the expression is frequency ω_m independent. This shows that the extrema of functionals is

$$P_\infty = \frac{P_{eg}\mathcal{E}^4|\alpha|^4L^4}{16\pi^2c^4} \left(\int_0^{\omega_e} d\omega_m A_f^2(\omega_m) \right)^2 \simeq \frac{P_{eg}\mathcal{E}^4|\alpha|^4L^2}{4c^2}. \quad (2.26)$$

However, it is the maximum, not a minimum. We hence conclude from the variation principle that there does not exist a minimum of this functionals.

To illustrate the dark pulse phenomena, let's consider a special case:

$$f(\omega) \propto e^{-(\omega - \omega_e/2)^2/2\sigma^2 + i(a/2) \sin[2b(\omega - \omega_e/2) + \varphi]}, \quad -\pi < \varphi \leq \pi. \quad (2.27)$$

which is a Gaussian distribution modulated by a sinusoidal phase $(a/2) \sin[2b(\omega - \omega_e/2) + \varphi]$. Such modulation brings sidebands to the original one as

$$f^2(\omega) \propto e^{-(\omega - \omega_e/2)^2/\sigma^2} \sum_{n=-\infty}^{\infty} J_n(a) e^{in[2b(\omega - \omega_e/2) + \varphi]}. \quad (2.28)$$

In a case of small a , $J_0(a) \simeq 1$ but the values of $J_{n \neq 0}(a)$ are small and hence the sidebands are not noticeable. When a becomes larger, all the $J_n(a)$ are small and therefore interferences between sidebands now are significant. Since every frequency in original mode has a set of sidebands, these frequency lines coincide with the ones from other bands, but their linked phases $e^{in\varphi}$ are different providing that $\varphi \neq 0$. These terms, which are out of phase, induce decoherence in superposition.

The photon source has a narrow band, whose value is much less than ω_e . We will extend the integral interval $0 < \omega \leq \omega_e$ to whole real axis at the cost of introducing some tiny errors. According to the following formulae,

$$\int_{-\infty}^{\infty} dx e^{-p^2 x^2 + qx} = \exp\left(\frac{q^2}{4p^2}\right) \frac{\sqrt{\pi}}{p}, \quad \text{Re } p^2 > 0. \quad (2.29)$$

the transition probability

$$P_{\infty} \propto \sigma^2 \left| \sum_{m=-\infty}^{\infty} J_m(a) e^{-\sigma^2 b^2 m^2} e^{im\varphi} \right|^2. \quad (2.30)$$

One can verify that for any φ , the transition probability is invariant after a π phase shift $\varphi + \pi$.

On the condition that there's no phase modulation, $a = 0$, the corresponding probability attains the maximum value. Accordingly, the sum in the modulus is equal to 1. For given b and φ , the value of transition probability decreases as the value of a increases. See the upper graph in Figure 2.2. If we take a as an n -th root x_n of Bessel function $J_0(x)$,

$$P_{\infty} \propto \sigma^2 \left[\left| \sum_{m=1}^{\infty} J_{2m}(x_n) e^{-4\sigma^2 b^2 m^2} \cos(2m\varphi) \right|^2 + \left| \sum_{m=1}^{\infty} J_{2m-1}(x_n) e^{-\sigma^2 b^2 (2m-1)^2} \sin(2m-1)\varphi \right|^2 \right]. \quad (2.31)$$

Intuitively, because the factor $\exp(-\sigma^2 b^2 m^2)$ decreases radically with increase of integer m , the crucial terms are those with small integer numbers, and therefore, the probability attains its minima around $\varphi = 0, \pi$ and attains its maxima around $\varphi = \pm\pi/2$, see the middle graph in Figure 2.2.

We are quite interested in the relation between transition probability and parameter b around $\varphi = 0$, since it shows a radical reduction of transition probability. The relation follows

$$P_{\infty} \propto \sigma^2 \left| \sum_{m=1}^{\infty} J_{2m}(x_n) e^{-4\sigma^2 b^2 m^2} \right|^2. \quad (2.32)$$

The transition probability decreases fast as the value of σb increases and with larger root x_n of $J_0(x) = 0$, the transition probability decreases more sharply. See the bottom graph in Figure 2.2. Roughly, the pulse duration can be assessed at σ^{-1} . Under the condition that the lifetime of the excited state is sufficiently large, even after elapsing a long time, the transmitted population accumulates. When there is a strong phase modulation ($b \gg \sigma^{-1}$), the reduction of transition probability at long time can be extremely suppressed in contrast to the case $b = 0$. Such a reduction is genuine. In this case, the two-photon transition should be very weak, and such light source, can hardly be absorbed by the energy level pair $|g\rangle - |e\rangle$, hence the name dark pulse. Since essentially no photon pairs in such state will be absorbed by this atom, it is indeed a kind of transparency induced by interference.

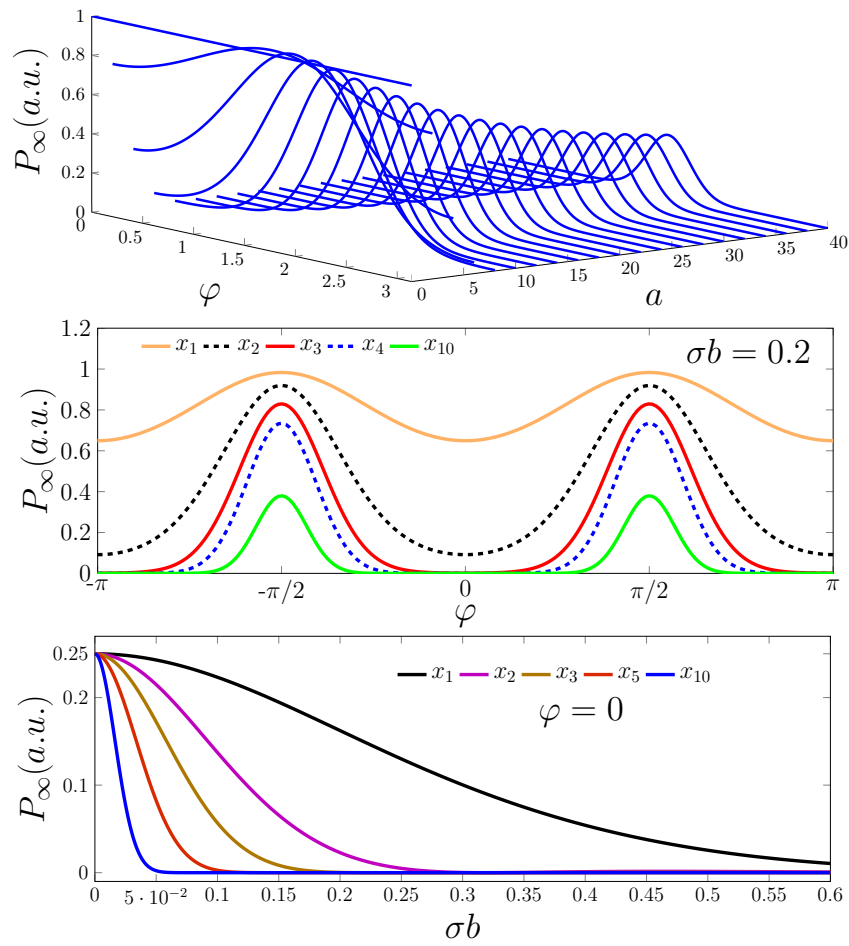


Figure 2.2: Graphs of transition probability P . The middle graph shows the phase φ dependence of P_∞ in which the curves attain their maxima around $\pm\pi/2$ and attain their minima around $0, \pi$. In this graph, we set $\sigma b = 0.2$. The bottom graph shows the relation between transition probability and parameter b at phase $\varphi = 0$. In both graphs, we find that transition probability decreases by choosing larger root of $J_0(x) = 0$ for parameter a , see the curves in the upper graph.

B.2 Two-photon transparency

Now let's consider a transition process with two-photon source $|\mathbb{II}\rangle = \sum_{mn} c(\omega_m, \omega_n) |1 : \omega_m, \alpha; 1 : \omega_n, \beta\rangle$. The correlation function $G_L^{(2)}$ can be factorized as

$$G_{L,|\mathbb{II}\rangle}^{(2)}(\tau_1, \tau) \propto \left(\sum_{jk} c_{jk}^* e^{i(\omega_j + \omega_k)\tau_1} \right) \left(\sum_{mn} c_{mn} e^{-i(\omega_m + \omega_n)\tau} \right), \quad (2.33)$$

which leads to

$$P(t) \propto \left| \iint_{\mathbb{R}^+ \times \mathbb{R}^+} d\omega_m d\omega_n c(\omega_m, \omega_n) \int_0^t d\tau \exp[-i(\omega_m + \omega_n - \omega_e)\tau] \right|^2. \quad (2.34)$$

For large time t , by applying the Dirac delta function approximation,

$$P(t) \propto \left| \int_0^{\omega_e} d\omega c(\omega, \omega_e - \omega) \right|^2. \quad (2.35)$$

If we take $c(\omega, \omega_e - \omega') = c'(\omega)c''(\omega')$, $c'(\omega) \propto c''(\omega) \propto f(\omega)$ where $f(\omega)$ is the distribution defined in multimode coherent state in B.1, that is, the state is factorable, we once again find the transparency phenomenon.

We therefore conclude that entanglement is not a necessary condition for transparency in two-photon absorption with two-photon state. In a paper [Fei1997], the authors found that when the central frequencies of a type II SPDC two-photon attain some special value, the no two-photon absorptions will be found, though it is expected to excite the atom and the curves should be quite flat in common sense. This counter-intuition result, should come from quantum origin and therefore the name: entanglement induced transparency. However, they did not show that any other states with no entanglement can not induce the transparency. However, we are more interested in possible cancellation for the excitation probability due to quantum entanglement. For simplicity, let's rewrite $c(\omega_m, \omega_n)$ as $C(\Omega = \omega_m + \omega_n, \nu = \omega_m - \omega_n)$. Then according to Eq.(2.35), the transition probability can be changed to

$$P(t) \propto \left| \int_{-\omega_e}^{\omega_e} d\omega C(\omega_e, \omega) \right|^2. \quad (2.36)$$

One can rewrite the coefficient $c(\omega_m, \omega_n)$ of the type II SPDC two-photon state as

$$C(\Omega, \nu) \propto e^{-(\Omega - \omega_p)^2/2\sigma^2} \text{sinc} \frac{(\Omega - \omega_p)T + \nu T_c - \phi_\Delta}{2\pi}. \quad (2.37)$$

In this case, the transition probability $P \propto |A|^2$,

$$A = e^{-\delta^2/2\sigma^2} \int_{-1}^1 \text{sinc} \frac{\omega_e T_c}{2\pi} \left(\omega - \frac{\phi_\Delta + T\delta}{\omega_e T_c} \right) d\omega, \quad \delta = \omega_p - \omega_e. \quad (2.38)$$

or equivalently in terms of sine integral function $\text{Si}(x) = \int_0^x \text{sinc} t dt$,

$$A = \frac{2\pi}{\omega_e T_c} e^{-\delta^2/2\sigma^2} \left[\text{Si} \left(\frac{\phi_\Delta + T\delta + \omega_e T_c}{2\pi} \right) - \text{Si} \left(\frac{\phi_\Delta + T\delta - \omega_e T_c}{2\pi} \right) \right]. \quad (2.39)$$

In a quantum optical system, the typical value of frequency $\omega_e/2\pi$ is 10^{14} Hz, the difference $|(1/u_1 - 1/u_2)|$ between the inverses of group velocities is about $10^{-9} \sim 10^{-10} \text{sm}^{-1}$,

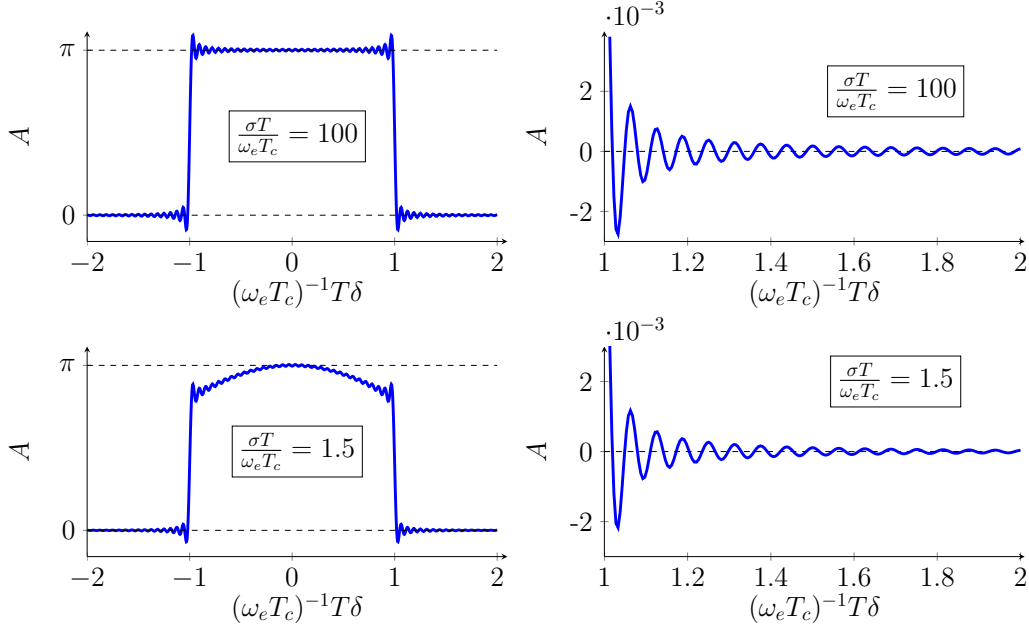


Figure 2.3: Figures of induced transparency in transition probability. As one can see in these figures, the zeros in the transition amplitude is induced by the large ratio T/T_c .

the common crystal length can be at the order of $1\mu\text{m} \sim 1\text{mm}$. Here we take $\omega_e/2\pi = 10^{14}$ Hz, $l = 2\text{mm}$, $|(1/u_1 - 1/u_2)| = 10^{-9}\text{sm}^{-1}$, $(1/u_1 + 1/u_2) = 10^{-6}\text{sm}^{-1}$ and $\phi_\Delta = 0$ therefore $\omega_e T_c/2\pi = 100$, $T = 1000T_c$. In Figure 2.3, we see that when $|\phi_\Delta + T\delta| \gg \omega_e T_c$, there exists points for vanishing amplitude A , therefore for vanishing transition probability. These point with vanishing transition probability are of great interest.

Recalling that $\omega_e T_c/2\pi (\sim 100)$ is a large parameter, and those points are out of the interval $-\omega_e T_c/2\pi \leq \omega \leq \omega_e T_c/2\pi$. Let's denote $x = (\phi_\Delta + T\delta)/2\pi$, $y = \omega_e T_c/2\pi$. Owing to the asymptotic expression of sine integral function for large argument x ,

$$\int_0^x dt \frac{\sin t}{t} \simeq \frac{\pi}{2} - \frac{\cos x}{x}, \quad (2.40)$$

the asymptotic expression of amplitude A follows

$$A_{asym} = \frac{8\pi^2}{\omega_e T_c} e^{-\delta^2/2\sigma^2} \left[\frac{(\phi_\Delta + T\delta) \sin \frac{\phi_\Delta + T\delta}{2\pi} \sin \frac{\omega_e T_c}{2\pi} + \omega_e T \cos \frac{\phi_\Delta + T\delta}{2\pi} \cos \frac{\omega_e T_c}{2\pi}}{(\phi_\Delta + T\delta)^2 - (\omega_e T_c)^2} \right]. \quad (2.41)$$

One can verify directly for $N_1 \in \mathbb{N}$, $Z \in \mathbb{Z}$ when

$$\omega_e T_c = (2N_1 + 1)\pi^2, \quad \phi_\Delta + T\delta = 2Z\pi^2; \text{ or} \quad (2.42a)$$

$$\omega_e T_c = 2N_1\pi^2, \quad \phi_\Delta + T\delta = (2Z + 1)\pi^2; \quad (2.42b)$$

the value of asymptotic expression of A is

$$A_{asym} = 0. \quad (2.43)$$

Roots from asymptotic expression are in good conformity with real ones, see the deviations of the n -th root from the right graph in Figure 2.4.

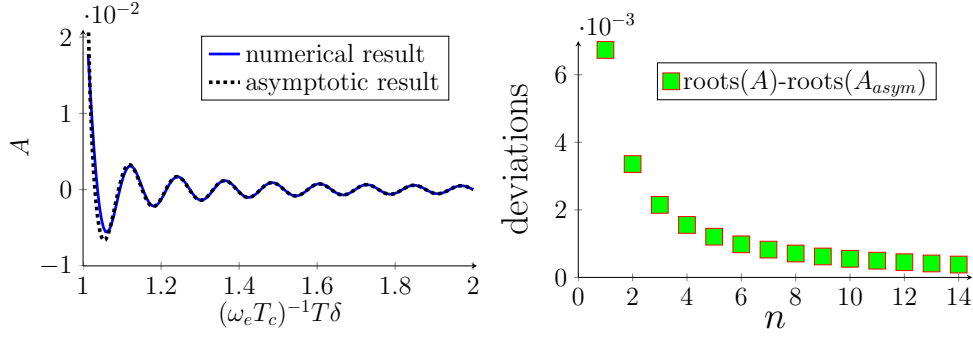


Figure 2.4: Comparison between the numerical result and the asymptotic result. The figure on the left hand side shows that the two results are well matched. The figure on the right hand side shows the deviations of the roots between the ones from numerical calculations and the ones from the asymptotic expression.

We have shown in the case of multimode coherent state source or factorable two-photon source, the two-photon absorption probability can be infinitely small, but never attains zero. In a real situation, one cannot tell the differences between a truly vanishing transition probability and a very small one. However, we observe noticeable consecutive hills and dales of the transition curve with growing two-photon transition detuning δ from the left graph in Figure 2.4 in contrast to the previous case. Such extra structure comes from 2 facts: 1. the factor T/T_c which stands for coherent quantum interferences ; 2. the detuning δ which represents frequency anti-correlations between photon pairs. This pure two-photon state with frequency anti-correlations between pairs are more likely entangled.

B.2.1 Entanglement induced transparency

Now consider a real atom and pick up a level pair that admits perfect two-photon resonance at frequency ω_e by shining a type II two-photon source and between which there exists other intermediate levels. From the second order perturbation theory, the transition cross section is proportional to

$$\sigma_e|_{j=s} \propto T_c^{-1} [\omega_s - \omega_p/2]^{-2} \delta(\omega_e - \omega_p) \sin^2[(\omega_s - \omega_p/2)T_c/2], \quad (2.44)$$

where ω_s is the Bohr frequency of s intermediate level. See more details in [Fei1997] where their notations $\varepsilon_s - \varepsilon_i, T_e$ were replaced by ω_e, T_c respectively. When $\omega_s - \omega_p/2 = 2m\pi/T_c$, then the transition cross section will be exactly zero. In general, every intermediate level should contribute to the transition cross section, and one should find some extra dips in the spectrum while in ordinary case, the spectrum is quite flat in these zones.

C Enhanced two-photon transition probability

By sending a pure photon source to an atom, the transition amplitude consists of different excitation contributions from the photons in different frequency modes. This superposition in transition amplitude can lead to transition enhancement or reduction by coherent control method. The reduction phenomena has been discussed in the previous sections in this Chapter. The enhancement of the transition is another aspect. It has been widely

studied during the past several decades, see [[LoudonQTL](#)]. In general, such phenomena are strongly related to the second order temporal correlation $g^{(2)}(0)$. With larger value of $g^{(2)}$, the transition probability is higher. It implies why a chaotic light source can give rise to higher transition probability than the coherent laser source. The interpretation is straightforward: because the two-photon transition takes place in a single atom, the absorbed photons should come at the same time.

With a multimode light state, especially with a continuous frequency-mode state, the field should show some temporal correlations but also some frequency anti-correlations because of the two-photon resonance condition. One concludes that an ideal condition to enhance the two-photon transition probability is to add both temporal correlations and frequency anti-correlations to the photon state. However, large value of $g^{(2)}(\tau = 0)$ (≥ 1 , that is, photon-bunching effect) can be explained by wave statistics from classical language. It is therefore regarded as a classical character if without any other details. Recently, some researches on the improvement of signal/noise ratio of a two-photon process on the quantum origins are studied, for example quantum illumination [[Lloyd2008](#)].

Two-photon two-atom interaction: Model

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Interference between quantum states rooted in the superposition principle is a fundamental phenomenon in quantum physics. In comparison with the interference between classical waves, the quantum interference relies on a quantized field as well as the state itself rather than its mean field. When it comes to optical system, the richness of internal structure of photon states provides many kinds of non-classical properties such as squeezing, bunching and anti-bunching statistics, negativity, entanglement, contextual correlations. These non-classical properties give rise to numerous interference phenomena and subsequently some missions impossible in classical world such like teleportation.

In order to fully display these extraordinary and fantastic beauties in the quantum world, some kind of coupling between the light and matter is a necessity.

It is expected that with denser atomic ensemble, the field-matter coupling is stronger and therefore the interface is more efficient. However, when atoms get closer, their interaction will modify the properties of each individual atom, such as frequency shift and change of spontaneous emission rate [FicekQIC], etc.

There are two simple classes of systems that may exhibit nonclassical properties induced by various photon states, namely, three-level atom systems and two two-level atom systems. We have studied the first class in the previous chapter. In the present one, **we** will focus our attention on the latter class.

First we will precise the model for two two-level atoms interacting with a quantized field, then by using perturbation theory, we derive an expression for transition probability. This expression is a base for subsequent chapters. An exactly solvable model is also introduced and discussed in the remaining part.

A Introduction

The system consisting of two two-level atoms, either identical or different, is a good one to perform a joint measurement on a light state and assess its correlation properties. In this way, novel spectroscopic and interferometric properties of specific photon states can be put in evidence. It is in this sense that studies of light interacting with two two-level atoms are of importance and interest both theoretically and experimentally.

From the study of the two-atom excitation by a photon state, interesting properties of pairs of atoms can be inferred from experimental data. For example common processes for a pair of atoms to absorb a single photon are a key ingredient [P-Penãfiel2012] to the development of the understanding of cold collisions [Weiner1999] and the determination of scattering length values [Weiner1999] in laser cooling of atoms [CohenTannoudjiAAP].

The excitation of two atoms of different species by two-photon was first considered in a theoretical article [RiosLeite1980] and then in [Andrews1983]. They considered tunable monochromatic classical light of frequency ω and showed that, in presence of interatomic interaction, a new resonance appears when ω is equal to the half sum of the two Bohr frequencies of the excited states of the atoms. Such effect was first observed in a mixture of Ba/Ti [White1981], then in neutral molecules [Hettich2002] and in cold Na atoms [P-Penãfiel2012].

It is indeed true that the interaction with entangled photons can induce in some cases correlations and entanglement of otherwise uncorrelated atoms [Richter2011]. In the year 2004, an insightful theoretical article [Muthukrishnan2004] reported that two photon resonance can be induced by using some particular entangled state of light and in absence of interaction between the two atoms. The same subject was also considered, but in the context of spin entanglement in Electron Paramagnetic Resonance by K. Salikhov [Salikhov2003]. More recently, [Richter2011, Salazar2012, Dayan2007] the related problem of interaction with pairs of broadband spectrum photons has been discussed.

It has also been widely studied that atomic interference can dramatically change the decoherence of the two atoms [Ficek2002]. With these backgrounds, one can now develop a theory to investigate the optical response to all kinds of quantum light states theoretically. However, even without considering atomic interaction and atomic decoherence as well as only concerning with monochromatic field, it is in general difficult to find an analytical solution. Under such simple conditions, the evolution of system is usually described by SU(8) Lie group or the corresponding Lie algebra [Khaneja2001]. However, it is still too complicated to play with. In this thesis, we will give an analytical solution in a really special system that can be described by two-photon Lie algebra h_6 [Ballesteros1998, Ballesteros2001]. In more general cases, we will apply perturbation theory since it is a good approximation that meets most of our most needs.

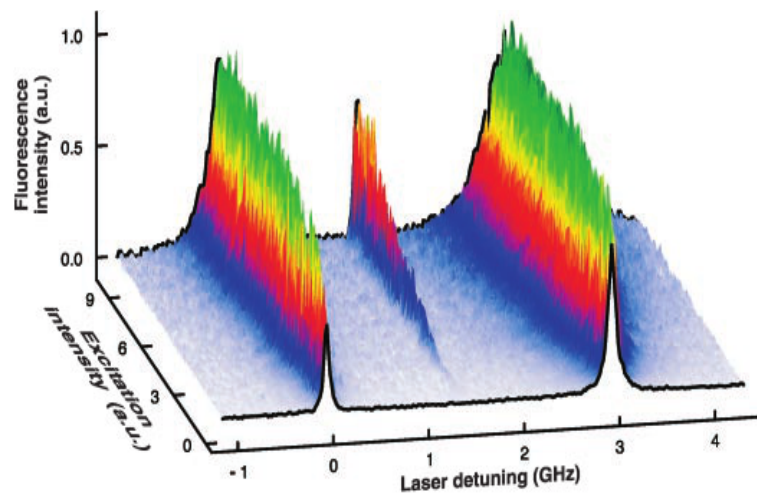


Figure 3.1: the transition spectra on the two-photon-two-atom detunings with a monochromatic field. On the left side, the spectrum only has two peaks at each single-photon-single-atom resonances in the absence of atomic interaction; On the right side, the spectrum shows an extra structure in the middle of two peaks that represent single-photon-single-atom resonance because of the presence of the atomic interaction. This new structure comes from a Fano type interference. The graph comes from [Hettich2002].

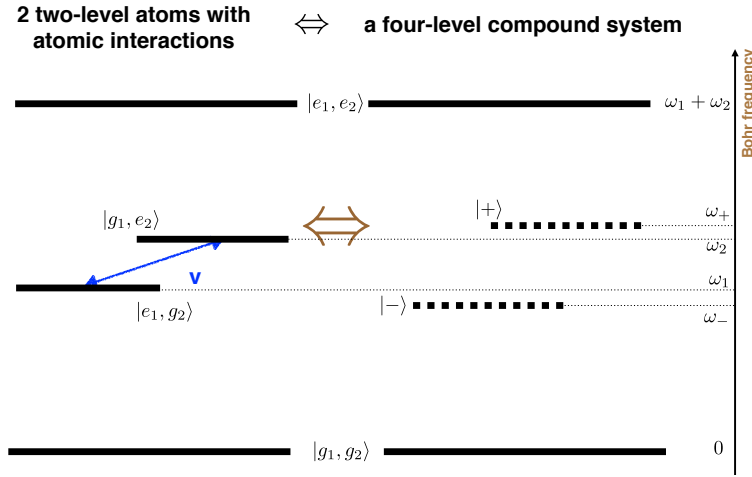


Figure 3.2: Figures of the atomic energy-level configurations. On the left hand side, the representation is based on the bare atomic energy-level basis, with ω_1, ω_2 being the Bohr frequencies of states $|e_1 g_2\rangle, |e_2 g_1\rangle$ respectively. The atomic interaction between two atoms is approximated by $\hbar v(|e_1 g_2\rangle\langle e_2 g_1| + |e_2 g_1\rangle\langle e_1 g_2|)$. On the right hand side, the energy-levels are represented by new eigenstates in a single system, because of the atomic interaction. Two new eigenstates, labeled by $|\pm\rangle$, have Bohr frequencies ω_{\pm} respectively. For sake of simplicity, we take $\omega_2 > \omega_1, \omega_+ > \omega_-$.

B Model

B.1 Two-atom system

The precise control of the spatial arrangement of two two-level systems is experimentally difficult. Historically speaking, two-atom experiment to show collective damping effect was first carried out with an atom before a reflecting surface in the late of 1960s by Drexhage [DrexhagePO]. Then techniques for ion trapping were developed. However, in order to perform a strong interaction between the two atoms, one needs to put the both objects at a distance that is smaller than the wavelength λ_0 ($\lambda_0 \simeq 1\mu\text{m}$) of radiation from the excited states [FicekQIC]. If the separation of the two atoms are not less than $10\lambda_0$, the interaction between atoms can be neglected. Because of the repulsive interactions between ions, it is really hard to keep both ions strongly interacting with each other while maintaining in a stable location. It is the development of laser cooling techniques that made it possible to push molecules to sufficiently close distance [Hettich2002, Weisenburger2013].

B.2 Atomic Hamiltonian

In this part, both atoms, which are different and labeled (1) and (2), are taken as two-level systems. If there is no interaction between them, the two atoms have ground and excited states $|g_i\rangle$ and $|e_i\rangle$ ($i = 1, 2$). For each excited state, we denote its atomic frequency by ω_i

and its decay constant by γ_i . The Hamiltonian of such a two-atom system is a sum of the two single-atom Hamiltonians,

$$H_{a,bare} = \hbar[\omega_1|e_1\rangle\langle e_1| \otimes \mathbb{I}_2 + \omega_2\mathbb{I}_1 \otimes |e_1\rangle\langle e_1|], \quad (3.1)$$

where we have taken the energy of the ground state to be zero. In the matrix representation, the atomic Hamiltonian can be written in a diagonal form,

$$H_{a,bare} = \hbar \begin{pmatrix} 0 & & & \\ & \omega_2 & & \\ & & \omega_1 & \\ & & & \omega_1 + \omega_2 \end{pmatrix}. \quad (3.2)$$

In this expression, the symbol of huge zero represents a 2×2 null matrix. The eigenstates of such a system are $|g\rangle = |g_1\rangle \otimes |g_2\rangle$, $|g_1\rangle \otimes |e_2\rangle$, $|e_1\rangle \otimes |g_2\rangle$, $|e\rangle = |e_1\rangle \otimes |e_2\rangle$, which correspond to atomic frequencies $0, \omega_2, \omega_1, \omega_1 + \omega_2$ respectively.

If there exists an interaction between the two atoms, for example, a dipole-dipole interaction $V_{aa}(\mathbf{r}_1, \mathbf{r}_2)$ that depending on their relative locations, then the two atoms are now a compound system.

Though the precise form of this Hamiltonian is complicated, in most cases, the dominant part is of the form

$$V = \hbar v |e_1 g_2\rangle\langle g_1 e_2| + h.c., \quad (3.3)$$

which describes an interaction between the two two-level atoms only when one atom is excited and the other in the ground state. For sake of simplicity, we neglect all the small terms and regard V as the interaction Hamiltonian. The total atomic Hamiltonian then reads $H_a = \hbar\omega_1|e_1\rangle\langle e_1| + \hbar\omega_2|e_2\rangle\langle e_2| + \hbar v(|e_1 g_2\rangle\langle g_1 e_2| + h.c.)$, or equivalently in the matrix representation,

$$H_a = \hbar \begin{pmatrix} 0 & & & \\ & \omega_2 & v & \\ & v & \omega_1 & \\ & & & \omega_1 + \omega_2 \end{pmatrix}. \quad (3.4)$$

Because the interaction happens only when one atom is in the ground state while the other is excited, the global ground state $|g\rangle$ and the co-excited state $|e\rangle$, which are out of the interaction, are eigenstates of the new total Hamiltonian. However, the other two eigenstates are different from the previous ones. The new eigenstates are $|\pm\rangle$ with atomic frequencies ω_{\pm} respectively, and the explicit solutions are

$$\omega_{\pm} = \frac{\omega_1 + \omega_2}{2} \pm \sqrt{\left(\frac{\omega_1 - \omega_2}{2}\right)^2 + |v|^2}; \quad (3.5)$$

$$|+\rangle = \frac{v|g_1 e_2\rangle + (\omega_+ - \omega_2)|e_1 g_2\rangle}{\sqrt{|v|^2 + (\omega_+ - \omega_2)^2}}, \quad (3.6)$$

$$|-\rangle = \frac{(\omega_+ - \omega_2)|g_1 e_2\rangle - v|e_1 g_2\rangle}{\sqrt{|v|^2 + (\omega_+ - \omega_2)^2}}. \quad (3.7)$$

One verifies that $|\pm\rangle$ are two orthonormal states. Note that the two new eigenstates are entangled states. The strength of the entanglement of the two states is determined by the ratio between the interaction parameter v and atomic frequency-difference $\omega_2 - \omega_1$.

The two factors appear naturally in the discriminant of the quadratic equation from the eigenvalue problem of the total Hamiltonian. According to the solutions (3.5,3.6,3.7), if v is much greater than ω_1, ω_2 , then both eigenstates are maximally entangled. Physically, the parameter v cannot be so large in general, however, it is still true that both states are more entangled with a larger v , since the cooperative Schmidt numbers that are the same in both states, are described by a monotonic increasing function of variable v , according to the explicit expression

$$\kappa = \frac{[v^2 + (\omega_+ - \omega_2)^2]^2}{v^4 + (\omega_+ - \omega_2)^4} = 2 - \frac{(\omega_1 - \omega_2)^2}{(\omega_1 - \omega_2)^2 + 2v^2}. \quad (3.8)$$

The range of κ is $1 \leq \kappa < 2$.

In the above expressions, one finds the sum of the two eigenfrequencies, is the same as the previous one,

$$\omega_+ + \omega_- = \omega_1 + \omega_2 \equiv \omega_e. \quad (3.9)$$

And therefore $\omega_+ - \omega_2 = -(\omega_- - \omega_1)$. That is, the two new frequencies are shifted with a same amount in opposite directions.

Now let's move to the new basis $\{|g\rangle, |-\rangle, |+\rangle, |e\rangle\}$. The completeness relation reads

$$|g\rangle\langle g| + |-\rangle\langle -| + |+\rangle\langle +| + |e\rangle\langle e| = \mathbb{I}_4, \quad (3.10)$$

and the total Hamiltonian is written in a diagonal form

$$H_a = \hbar(\omega_- |-\rangle\langle -| + \omega_+ |+\rangle\langle +| + \omega_e |e\rangle\langle e|). \quad (3.11)$$

Atomic transitions are described by the raising and lowering operators, denoted by $b_j^\dagger = |e_j\rangle\langle g_j|$ and b_j ($j = 1, 2$). b_j^\dagger , for example, makes the electron in the ground state to jump to the excited state $|e_j\rangle$. The excitation of one atom may partially be transferred to the other atom because of the coupling. b_j^\dagger has the following expression in the new basis

$$b_j^\dagger = \mathbb{I}_4 b_j^\dagger \mathbb{I}_4 = A_j |e\rangle\langle +| + B_j |e\rangle\langle -| + C_j |+\rangle\langle g| + D_j |-\rangle\langle g|, \quad j = 1, 2. \quad (3.12)$$

The components of the four vectors **A**, **B**, **C**, **D** with subscripts $j = 1, 2$ are

$$A_j = \alpha \delta_{j2} + \beta \delta_{j1}, \quad B_j = \alpha \delta_{j1} - \beta \delta_{j2}, \quad (3.13a)$$

$$C_j = \alpha \delta_{j1} + \beta \delta_{j2}, \quad D_j = \alpha \delta_{j2} - \beta \delta_{j1}, \quad (3.13b)$$

$$\alpha = \frac{\omega_+ - \omega_2}{\sqrt{(\omega_+ - \omega_2)^2 + v^2}}, \quad \beta = \frac{v}{\sqrt{(\omega_+ - \omega_2)^2 + v^2}}. \quad (3.13c)$$

where δ_{jk} is the Kronecker δ function. We can rewrite Eq.(3.12) as

$$(b_1^\dagger, b_2^\dagger)^T = \mathbf{A}|e\rangle\langle +| + \mathbf{B}|e\rangle\langle -| + \mathbf{C}|+\rangle\langle g| + \mathbf{D}|-\rangle\langle g|. \quad (3.14)$$

B.3 Free quantum electric field

A free quantum electric field \hat{E} propagating in the z direction can be decomposed as positive frequency part $\hat{E}^{(+)}$ and its conjugate part $(\hat{E}^{(+)})^\dagger$, where

$$\hat{E}^{(+)}(z, t) = i \sum_m \mathcal{E}_m a_m e^{-i\omega_m(t-z/c)} \quad (3.15)$$

\mathcal{E}_m is the unit of the single photon electric field at frequency ω_m , and the bosonic operator obeys the non-vanishing commutator $[a_m, a_n^\dagger] = \delta_{mn}$. The frequency difference between neighboring components is $2\pi c/L$ where L is the length of a quantized box along the z direction. The length of a quantized box can be chosen sufficiently long at will.

The total field Hamiltonian reads

$$H_f = \hbar \sum_m \omega_m a_m^\dagger a_m \quad (3.16)$$

The annihilation operators a_m are time independent.

B.4 Interaction between atoms and light

Here we consider the dipole interaction. At the rotating wave approximation, the interaction Hamiltonian is given by :

$$V = \hbar b_1^\dagger \sum_\ell f_1(\omega_\ell) a_\ell + \hbar b_2^\dagger \sum_\ell f_2(\omega_\ell) a_\ell + h.c., \quad (3.17)$$

where $f_k(\omega_\ell) = -i \langle e_k | \mathbf{p} \cdot \boldsymbol{\epsilon}_\ell | g_k \rangle \sqrt{\omega_\ell / 2\hbar\epsilon_0 S L} e^{i\omega_\ell(z_i/c-t)} = f_{i\ell} e^{i\omega_\ell(z_i/c-t)}$, $f_{i\ell}$ being a slowly varying function of the photon frequency. S is the transverse section of the beam which is focused on the atoms, z_i the position of atom i and L the length of the quantization box, the mode density in terms of frequencies ω_ℓ being $2\pi c/L$. For simplicity, we will set $z_1 = z_2 = 0$ and hence will not consider propagation effects and take $f_i(\omega_\ell)$ as a constant $f_i \equiv f_i(\omega_i)$.

Let $\mathbf{f} = (f_1, f_2)^T$, $F(t) = \sum_\ell e^{-i\omega_\ell t} a_\ell$. Recalling the expression (3.14), the interaction Hamiltonian in the coupling basis reads

$$\begin{aligned} V &= \hbar e^{iH_a t/\hbar} (b_1^\dagger f_1 F + b_2^\dagger f_2 F + h.c.) e^{-iH_a t/\hbar} \\ &= \hbar F e^{iH_a t/\hbar} (\mathbf{A}^T \mathbf{f} |e\rangle\langle +| + \mathbf{B}^T \mathbf{f} |e\rangle\langle -| + \mathbf{C}^T \mathbf{f} |+\rangle\langle g| + \mathbf{D}^T \mathbf{f} |-\rangle\langle g|) e^{-iH_a t/\hbar} + h.c. \\ &= \hbar (A_t |e\rangle\langle +| + B_t |e\rangle\langle -| + C_t |+\rangle\langle g| + D_t |-\rangle\langle g|) + h.c. \end{aligned} \quad (3.18)$$

where

$$A_t = \mathbf{A}^T \mathbf{f} F e^{i\omega_- t}, \quad B_t = \mathbf{B}^T \mathbf{f} F e^{i\omega_+ t}, \quad C_t = \mathbf{C}^T \mathbf{f} F e^{i\omega_+ t}, \quad D_t = \mathbf{D}^T \mathbf{f} F e^{i\omega_- t}. \quad (3.19)$$

B.5 Evolution and transition probability

The whole system at time t can be described by a density matrix $\tilde{\rho}$. Because of the finite lifetime of the excited detecting atoms, the evolution of system is non-unitary. For simplicity, we assume our system is Markovian, that is, the dynamics of the total system are governed by a master equation when the atomic interaction, atomic decoherence and multimode field are all incorporated. The governing master equation can be derived from the treatments of Louisell [LouisellSPR], Lehmberg [Lehmberg1970] and Agarwal [AgarwalQSTSE], etc. Here, we will use super-operator method to derive a general expression for 2P2A transition probability. In the following model, we also neglect the photon exchange during the interaction process where cooperative effect can occur in the nonlinear response.

In interaction picture, the master equation for density matrix can be formally written as:

$$i\partial_t\tilde{\rho} = L_t\tilde{\rho} - iR\tilde{\rho} \quad (3.20)$$

where $L_t\tilde{\rho} = [V/\hbar, \tilde{\rho}]$ is a commutator describing the field-atom interaction and $R\tilde{\rho}$ is decoherence operator. All the properties of the entire system are determined by the initial state $|g_1g_2\rangle\langle g_1g_2| \otimes \rho_0$ and a non-unitary evolution super operator $U(t)$, with which notation the density matrix of the whole system at any time t yields $\tilde{\rho} = U|g_1g_2\rangle\langle g_1g_2| \otimes \rho_0$. The two-atom excitation probability is related to the projection measurement $|e_1e_2\rangle\langle e_1e_2|$ on the atomic part according to Born's rule, consequently

$$P = \text{Tr}\langle e_1e_2|(U|g_1g_2\rangle\langle g_1g_2|\rho_0)|e_1e_2\rangle \quad (3.21)$$

B.6 Perturbative result

In principle, if one has the precise expression of the evolution operator $U(t)$, one has a maximum information about the entire system. However, even without decoherence terms, it is very difficult to find out the precise expression of $U(t)$. On the other hand, we are more interested in the two-atom resonances while keeping any of the both atoms sufficiently far away from single-atom single-photon resonances. In such two-atom excitation processes, the leading effects can be accurately described by the lowest order perturbative results.

As mentioned before, the commutator between the interaction Hamiltonian and density matrix $[\hbar^{-1}V, \tilde{\rho}]$ is labeled as a operator function $L_t\tilde{\rho}$ with subscript t to signify the time dependence. For example,

$$L_t|g\rangle\langle g| \otimes \rho_0 = (C_t|+\rangle\langle g| + D_t|-\rangle\langle g|)\rho_0 - h.c.$$

The explicit form of the super operator is

$$L_t\tilde{\rho} = [A_t|e\rangle\langle +| + B_t|e\rangle\langle -| + C_t|+\rangle\langle g| + D_t|-\rangle\langle g| + h.c., \tilde{\rho}]. \quad (3.22)$$

The operator

$$\tilde{L}_t \equiv A_t|e\rangle\langle +| + B_t|e\rangle\langle -| + C_t|+\rangle\langle g| + D_t|-\rangle\langle g| \quad (3.23)$$

is able to raise the atomic ket state vector and lower the bra state vector, therefore, the two terms $\tilde{\rho}\tilde{L}_t, \tilde{L}_t^\dagger\tilde{\rho}$ do not contribute to the two-atom excitation probability in the lowest order perturbation theory provided that the initial atomic system in the ground state. This leads us to simplify the super operator as

$$L_t\tilde{\rho} = \tilde{L}_t\tilde{\rho} - \tilde{\rho}\tilde{L}_t^\dagger. \quad (3.24)$$

Notice that now \tilde{L}_t is an operator, not a super operator.

During the interaction process, noises from the environment always disturb the whole system, and make the evolution of the atoms and field non-unitary. We will use the a Markovian super operator $R\tilde{\rho}$ to describe this phenomena. In the perturbation calculations, we take $R|g\rangle\langle g| = 0$ and $R|\mu\rangle\langle \nu| = (\gamma_\mu + \gamma_\nu)|\mu\rangle\langle \nu|$ for other density matrix element $|\mu\rangle\langle \nu|$.

Notice that the decay rates γ_\pm of $|+\rangle$ are different in general sense, they come from the re-partition of the $\gamma_{1,2}$ and also the interaction induced de-phasing rate γ_{12} which

is typically related to the $1/r^n (n > 2)$ interaction. In most situations, we neglect the difference between γ_+ and γ_- and then take them both as $\gamma = \gamma_e/2$. That is, we assume the two atoms are not so close such that the decay rate originates from the interaction between the two atoms are negligible since the value is proportional to the inverse cubic power of the distance and therefore is very sensitive. In a real experimental situation [Science 2002], this value can be 2 orders of magnitude smaller than the spontaneous emission rate of the bare atoms.

The transition probability given by the 4-th order perturbation theory

$$P = \text{Tr} \langle e | \left(\int d^4 t e^{R(\tau-t)} L_\tau e^{R(\tau_1-\tau)} L_{\tau_1} e^{R(\tau_2-\tau_1)} L_{\tau_2} e^{R(\tau_3-\tau_2)} L_{\tau_3} e^{-R\tau_3} |g\rangle \langle g| \otimes \rho_0 \right) |e\rangle,$$

includes $4! = 24$ terms. For simplicity, we introduce new notations $\tilde{A}_t = A_t e^{\gamma-t}$, $\tilde{B}_t = B_t e^{\gamma+t}$, $\tilde{C}_t = C_t e^{\gamma+t}$, $\tilde{D}_t = D_t e^{\gamma-t}$ and consequently, their hermitian conjugates read

$$\tilde{A}_t^\dagger = A_t^\dagger e^{\gamma-t}, \tilde{B}_t^\dagger = B_t^\dagger e^{\gamma+t}, \tilde{C}_t^\dagger = C_t^\dagger e^{\gamma+t}, \tilde{D}_t^\dagger = D_t^\dagger e^{\gamma-t}.$$

The transition probability can be written in a more compact form

$$\begin{aligned} P = & e^{-2\gamma_e t} \int_0^t d\tau \int_0^\tau d\tau_1 \int_0^{\tau_1} d\tau_2 \int_0^{\tau_2} d\tau_3 [(\tilde{A}_{\tau_2} \tilde{C}_{\tau_3} + \tilde{B}_{\tau_2} \tilde{D}_{\tau_3}) \rho_0 (\tilde{C}_{\tau_1}^\dagger \tilde{A}_\tau^\dagger + \tilde{D}_{\tau_1}^\dagger \tilde{B}_\tau^\dagger) \\ & + (\tilde{A}_\tau \tilde{C}_{\tau_1} + \tilde{B}_\tau \tilde{D}_{\tau_1}) \rho_0 (\tilde{C}_{\tau_3}^\dagger \tilde{A}_{\tau_2}^\dagger + \tilde{D}_{\tau_3}^\dagger \tilde{B}_{\tau_2}^\dagger) + (\tilde{A}_\tau \tilde{C}_{\tau_3} + \tilde{B}_\tau \tilde{D}_{\tau_3}) \rho_0 (\tilde{C}_{\tau_2}^\dagger \tilde{A}_{\tau_1}^\dagger + \tilde{D}_{\tau_2}^\dagger \tilde{B}_{\tau_1}^\dagger) \\ & + (\tilde{A}_{\tau_1} \tilde{C}_{\tau_3} + \tilde{B}_{\tau_1} \tilde{D}_{\tau_3}) \rho_0 (\tilde{C}_{\tau_2}^\dagger \tilde{A}_\tau^\dagger + \tilde{D}_{\tau_2}^\dagger \tilde{B}_\tau^\dagger) + (\tilde{A}_\tau \tilde{C}_{\tau_2} + \tilde{B}_\tau \tilde{D}_{\tau_2}) \rho_0 (\tilde{C}_{\tau_3}^\dagger \tilde{A}_{\tau_1}^\dagger + \tilde{D}_{\tau_3}^\dagger \tilde{B}_{\tau_1}^\dagger) \\ & + (\tilde{A}_{\tau_1} \tilde{C}_{\tau_2} + \tilde{B}_{\tau_1} \tilde{D}_{\tau_2}) \rho_0 (\tilde{C}_{\tau_3}^\dagger \tilde{A}_\tau^\dagger + \tilde{D}_{\tau_3}^\dagger \tilde{B}_\tau^\dagger)]. \end{aligned}$$

If we define $\mathbf{u}_t = (\tilde{A}_t, \tilde{B}_t)$, $\mathbf{u}_\tau = (\tilde{C}_\tau, \tilde{D}_\tau)$ and its hermitian transpose $\mathbf{u}_t^\dagger = (\tilde{A}_t^\dagger, \tilde{B}_t^\dagger)$ and a binary product \odot between \mathbf{u}_t and \mathbf{v}_τ as

$$\mathbf{u}_t \odot \mathbf{v}_\tau = \tilde{A}_t \tilde{C}_\tau + \tilde{B}_t \tilde{D}_\tau,$$

therefore,

$$\mathbf{v}_\tau^\dagger \odot \mathbf{u}_t^\dagger = \tilde{C}_\tau^\dagger \tilde{A}_t^\dagger + \tilde{D}_\tau^\dagger \tilde{B}_t^\dagger.$$

Now the transition probability can be expressed as

$$\begin{aligned} P(t) = & e^{-2\gamma_e t} \int_0^t d\tau \int_0^\tau d\tau_1 \int_0^{\tau_1} d\tau_2 \int_0^{\tau_2} d\tau_3 [\mathbf{u}_{\tau_2} \odot \mathbf{v}_{\tau_3} \rho_0 \mathbf{v}_{\tau_1}^\dagger \odot \mathbf{u}_\tau^\dagger \\ & + \mathbf{u}_\tau \odot \mathbf{v}_{\tau_1} \rho_0 \mathbf{v}_{\tau_3}^\dagger \odot \mathbf{u}_{\tau_2}^\dagger + \mathbf{u}_\tau \odot \mathbf{v}_{\tau_3} \rho_0 \mathbf{v}_{\tau_2}^\dagger \odot \mathbf{u}_{\tau_1}^\dagger + \mathbf{u}_{\tau_1} \odot \mathbf{v}_{\tau_2} \rho_0 \mathbf{v}_{\tau_3}^\dagger \odot \mathbf{u}_\tau^\dagger \\ & + \mathbf{u}_{\tau_1} \odot \mathbf{v}_{\tau_3} \rho_0 \mathbf{v}_{\tau_2}^\dagger \odot \mathbf{u}_\tau^\dagger + \mathbf{u}_\tau \odot \mathbf{v}_{\tau_2} \rho_0 \mathbf{v}_{\tau_3}^\dagger \odot \mathbf{u}_{\tau_1}^\dagger] u(t-\tau) u(\tau-\tau_1) u(\tau_1-\tau_2) u(\tau_2-\tau_3), \end{aligned} \quad (3.25)$$

where $u(\theta)$ is the Heaviside function whose value is equal to 1 when $\theta \geq 0$ and otherwise is zero. The product of the four Heaviside functions provides a time ordering to all the terms in square bracket. A possible way to simplify the expression is to change all the temporal arguments for different terms in the square bracket in a same order, say, $\mathbf{u}_{\tau_3} \odot \mathbf{v}_{\tau_2} \rho_0 \mathbf{v}_{\tau_1}^\dagger \odot \mathbf{u}_\tau^\dagger$. By doing so, it gives rise to the following combination of Heaviside functions:

$$\begin{aligned} U_{\{\tau\}} = & u(t-\tau) u(\tau-\tau_3) u(\tau_3-\tau_1) u(\tau_1-\tau_2) + u(t-\tau) u(\tau-\tau_1) u(\tau_1-\tau_3) u(\tau_3-\tau_2) \\ & + u(t-\tau_3) u(\tau_3-\tau) u(\tau-\tau_1) u(\tau_1-\tau_2) + u(t-\tau) u(\tau-\tau_3) u(\tau_3-\tau_2) u(\tau_2-\tau_1) \\ & + u(t-\tau_3) u(\tau_3-\tau_2) u(\tau_2-\tau) u(\tau-\tau_1) + u(t-\tau_3) u(\tau_3-\tau) u(\tau-\tau_2) u(\tau_2-\tau_1). \end{aligned} \quad (3.26)$$

Observe that the time ordering of $t \geq \tau, \tau \geq \tau_1, t \geq \tau_3, \tau_3 \geq \tau_2$ can be found in all the terms, therefore $u(t-\tau)u(\tau-\tau_1)u(t-\tau_3)u(\tau_3-\tau_2)$ is a common term and can be written outside the square bracket, subsequently,

$$\begin{aligned} U_{\{\tau\}} &= u(t-\tau)u(\tau-\tau_1)u(t-\tau_3)u(\tau_3-\tau_2) \\ &\quad [1 + u(\tau_1-\tau_3)(1 - u(\tau-\tau_3)u(\tau_1-\tau_2)) + u(\tau_2-\tau)(1 - u(\tau_3-\tau)u(\tau_2-\tau_1))] \\ &= u(t-\tau)u(\tau-\tau_1)u(t-\tau_3)u(\tau_3-\tau_2)[1 + u(\tau_1-\tau_3)(1-1) + u(\tau_2-\tau)(1-1)] \\ &= u(t-\tau)u(\tau-\tau_1)u(t-\tau_3)u(\tau_3-\tau_2), \end{aligned} \quad (3.27)$$

which separates the temporal arguments into two sets and brings light to a simple expression of the 4-th order perturbation transition probability

$$P = \text{Tr}(\mathcal{K}\rho_0\mathcal{K}^\dagger), \quad \mathcal{K} = e^{-\gamma e t} \int_0^t d\tau \int_0^\tau d\tau_1 [A_\tau C_{\tau_1} e^{\gamma-\tau} e^{\gamma+\tau_1} + B_\tau D_{\tau_1} e^{\gamma+\tau} e^{\gamma-\tau_1}]. \quad (3.28)$$

We notice that a double integral in the expression of \mathcal{K} , which differs from the product form of two integrals in the case with absence of the atomic interaction. This is because the two atoms are different and photons can choose whichever atoms to excite if the atoms are without interaction in contrast to the case with interaction, in which the two-atom system should be excited firstly to their mutual state $|+\rangle$ or $|-\rangle$ which can be "felt" by both atoms because they interchange electronic energies.

Combined with Eqs.(3.13,3.19),

$$\mathcal{K} = - \sum_{mn} a_m a_n \mathcal{K}_{mn}, \quad (3.29)$$

$$\begin{aligned} \mathcal{K}_{mn} &= \frac{\mathbf{A}^T \mathbf{f} \mathbf{C}^T \mathbf{f}}{\omega_+ \omega_m - \omega_+ + i\gamma_+} \left(\frac{e^{i(\omega_e - \omega_m - \omega_n)t} - e^{-\gamma e t}}{\omega_m + \omega_n - \omega_e + i\gamma_e} - \frac{e^{-\gamma+t+i(\omega_- - \omega_n)t} - e^{-\gamma e t}}{\omega_n - \omega_- + i\gamma_-} \right) \\ &\quad + \frac{\mathbf{B}^T \mathbf{f} \mathbf{D}^T \mathbf{f}}{\omega_m - \omega_- + i\gamma_-} \left(\frac{e^{i(\omega_e - \omega_m - \omega_n)t} - e^{-\gamma e t}}{\omega_m + \omega_n - \omega_e + i\gamma_e} - \frac{e^{-\gamma-t+i(\omega_+ - \omega_n)t} - e^{-\gamma e t}}{\omega_n - \omega_+ + i\gamma_+} \right). \end{aligned} \quad (3.30)$$

In the two brackets, one can find 2 terms that are oscillating at the two-photon detuning frequency $\omega_m + \omega_n - \omega_e$ and the other 6 terms are exponentially decaying. Therefore, when $t \gg \gamma_+^{-1}, \gamma_-^{-1}$, these terms are swept away, and \mathcal{K}_{mn} becomes

$$\mathcal{K}_{mn}^\infty = \left[\frac{\mathbf{A}^T \mathbf{f} \mathbf{C}^T \mathbf{f}}{\omega_m - \omega_+ + i\gamma_+} + \frac{\mathbf{B}^T \mathbf{f} \mathbf{D}^T \mathbf{f}}{\omega_m - \omega_- + i\gamma_-} \right] \frac{e^{i(\omega_e - \omega_m - \omega_n)t}}{\omega_m + \omega_n - \omega_e + i\gamma_e}. \quad (3.31)$$

B.6.1 Coupling strength

Since $v^2 = (\omega_+ - \omega_-)^2/4 - (\omega_1 - \omega_2)^2/4 = (\omega_+ - \omega_2)(\omega_+ - \omega_1)$, we have $(\omega_+ - \omega_2)^2 + v^2 = (\omega_+ - \omega_2)(\omega_+ - \omega_-)$. From Eqs.(3.13), one has

$$\mathbf{A}^T \mathbf{f} \mathbf{C}^T \mathbf{f} = f_1 f_2 + v(f_1^2 + f_2^2)/(\omega_+ - \omega_-), \quad (3.32a)$$

$$\mathbf{B}^T \mathbf{f} \mathbf{D}^T \mathbf{f} = f_1 f_2 - v(f_1^2 + f_2^2)/(\omega_+ - \omega_-). \quad (3.32b)$$

We notice a sign difference between the two lines. This difference, induced by the v parameter, is crucially important in some cases. From the two expressions, we also notice that when the interaction parameter v is small, $v/(\omega_- - \omega_+) \approx v/|\omega_1 - \omega_2|$.

C Case of infinite lifetimes of the excited detecting atoms

When both excited states of the detecting atoms have infinite lifetime, the decoherence effect is then not considered in the process, therefore, the evolution is reversible. In this case, the transition probability is determined by

$$\mathcal{K} = - \sum_{mn} a_m a_n \mathcal{K}_{mn}, \quad (3.33)$$

$$\begin{aligned} \mathcal{K}_{mn} = & \frac{\mathbf{A}^T \mathbf{f} \mathbf{C}^T \mathbf{f}}{\omega_m - \omega_+} \left(\frac{e^{i(\omega_e - \omega_m - \omega_n)t} - 1}{\omega_m + \omega_n - \omega_e} - \frac{e^{i(\omega_- - \omega_n)t} - 1}{\omega_n - \omega_-} \right) \\ & + \frac{\mathbf{B}^T \mathbf{f} \mathbf{D}^T \mathbf{f}}{\omega_m - \omega_-} \left(\frac{e^{i(\omega_e - \omega_m - \omega_n)t} - 1}{\omega_m + \omega_n - \omega_e} - \frac{e^{i(\omega_+ - \omega_n)t} - 1}{\omega_n - \omega_+} \right). \end{aligned} \quad (3.34)$$

The last terms in brackets are far off resonance and quite small, and therefore sometimes can be neglected. The photons of frequencies ω_m and ω_n , both of which are far away from the atomic frequencies but the sum might be close to ω_e , can lead to a noticeable transition. This transition at 2P2A resonance $\omega_m + \omega_n = \omega_e = \omega_1 + \omega_2$ is described by

$$\mathcal{K}_{mn} \simeq \left(\frac{\mathbf{A}^T \mathbf{f} \mathbf{C}^T \mathbf{f}}{\omega_m - \omega_+} + \frac{\mathbf{B}^T \mathbf{f} \mathbf{D}^T \mathbf{f}}{\omega_m - \omega_-} \right) \frac{e^{i(\omega_e - \omega_m - \omega_n)t} - 1}{\omega_m + \omega_n - \omega_e}. \quad (3.35)$$

If we use the explicit form the inner products, we then find the exact expression \mathcal{K}_{mn}

$$\begin{aligned} \mathcal{K}_{mn} = & f^2 \frac{e^{i(\omega_+ - \omega_m)t} - 1}{\omega_m - \omega_+} \frac{e^{i(\omega_- - \omega_n)t} - 1}{\omega_n - \omega_-} \\ & + \frac{2v f^2}{(\omega_m - \omega_+)(\omega_m - \omega_-)} \frac{e^{i(\omega_e - \omega_m - \omega_n)t} - 1}{\omega_m + \omega_n - \omega_e} \\ & + \frac{2v f^2}{\omega_+ - \omega_-} \frac{e^{i(\omega_+ - \omega_m)t} - 1}{(\omega_m - \omega_+)} \frac{e^{i(\omega_- - \omega_n)t} - 1}{(\omega_n - \omega_-)}. \end{aligned} \quad (3.36)$$

Consider finally the limit case of a two-atom excitation without atomic interaction. Then ω_{\pm} degrade into ω_1, ω_2 and therefore the response function is a product of two individual single-photon–single-atom response functions

$$\mathcal{K}_{mn} = f_1 f_2 \frac{e^{i(\omega_1 - \omega_m)t} - 1}{\omega_m - \omega_1} \frac{e^{i(\omega_2 - \omega_n)t} - 1}{\omega_n - \omega_2}. \quad (3.37)$$

The response kernel \mathcal{K}_{mn} has been written in a product form of two terms, each of which represents a response of single-photon–single-atom process. At a single-photon–single-atom resonance, one has

$$\lim_{\omega \rightarrow \omega_a} h(\omega - \omega_a, t) = \lim_{\omega \rightarrow \omega_a} \frac{e^{i(\omega_a - \omega)t} - 1}{\omega - \omega_a} = -it, \quad \omega_a = \omega_1, \omega_2; \quad (3.38)$$

that is, a linearly t -dependent factor. When time t is sufficiently large, the single-photon–single-atom response $h(\omega - \omega_a, t)$ is a fast oscillating function except at the point $\omega = \omega_a$, so the transition probability should be mainly determined by the numeric properties of the coefficients of a multimode photon state at $\omega = \omega_a$ from the asymptotic analysis point of view.

D An analytical solution in the case of two degenerate atoms with infinite lifetime

As we have mentioned, it is not simple to find an exact solution to the evolution of a system in which a quantized field interacts with two two-level atoms, even in a simple situation where the atoms are initially in the ground state and that no noise enters during the interaction process and no interaction occurs between the two atoms which have different atomic frequencies. By using a perturbation theory, at its leading order, one finds the probability in two-photon absorption is, generally speaking, expressed as a product of a factor dependency on photon statistics and a dependency of spectrum which has t^2 dependence for stationary photon states [Mollow1968]. This is also true in two-atom cases.

In both cases, the expression shows that each photon in the field interacts with atoms individually. Consequently, some special photon states of a give frequency mode are able to make dramatically enhancement or reductions of the transition probability. If we disregard the common response function and only focus on our attention on the photon statistics, in fact it is a double slit type experiment in quantum optics.

By manipulation of the photon state, one can have dark pulses [Meshulach1998] and bright pulses. An interesting and important question is *how bright the transition signal can be*. To get a full answer to this question, we need to go further than the leading term in the perturbation theory.

On the other hand, as mentioned above, the transition probability of a completely dephased state (as defined in Eq. (1.28) represented by a diagonal density matrix) is accumulated from time to time since the photon flux is fixed and the atomic decays are negligible. For these states, the transition probabilities are expected to be $(ct/L)^2$ dependent. Time can be sufficiently large even though $t \leq L/c$ in a quantized box and hence does not violate the upper bound 1 of the probability, but the transition probability still somehow depends on the way of choosing the length of quantization box, is not physical. This problem in this expression has perhaps its origin from the cut-off of the high order perturbation terms. Therefore, an analytical solution to the model is of great desire even it is an approximate one.

We will consider now an exactly solvable model in which a monochromatic field interacts with two interacting two-level atoms that are frequency degenerate and initially in the ground state. After doing so, we will modify the model by going to a quasi-monochromatic field.

D.1 Monochromatic field interacting with two-atom system

D.1.1 Introduction

In this part, we only focus our attention on the interaction with the narrow band photon source. For sake of simplicity, we will take the light field as the monochromatic one. Then the Hamiltonian of the free field and of field atoms interactions are:

$$\begin{aligned} H_f^0 &= \hbar\omega a^\dagger a, \quad [a, a^\dagger] = 1; \\ V &= \hbar(f_1|e_1\rangle\langle g_1| + f_2|e_2\rangle\langle g_2|)a + h.c.. \end{aligned} \quad (3.39)$$

The coupling strength are given previously in Eq. (3.17).

Since the interaction strength v between the two degenerate atoms is not vanishing, there should not be resonant peaks at frequency ω_a for excitations when a monochromatic classical field is applied. This is because the interaction has shifted the eigen-frequencies of singly excited state. In this sense, to observe the co-excitation phenomena, it is not necessary to use the non-degenerate atoms in the presence of the interaction between atoms. In a real situation, especially when the interaction between atoms is quite weak, there should be a strong peak which represents the double single-photon–single-atom resonance, and therefore might hide the effects originated from the parameter v . However, when v is small, it is also weak to observe such a resonance in non-identical two atoms system, and typically, v is about 1 GHz, and the laser width is about 1 MHz, therefore, two identical atoms should also deserve a place to show such a striking effect.

Because of the symmetry of the two degenerate two-level atoms, a theoretical group way to describe the dynamics of the system will be helpful. Here we take $K_0 = \sum_{i=1}^2 (|e_i\rangle\langle e_i| - |g_i\rangle\langle g_i|)/2$, $K_- = \sum_{i=1}^2 |g_i\rangle\langle e_i|/\sqrt{2}$, and $K_+ = K_-^\dagger$, which thus yields the commutation relation

$$[K_0, K_\pm] = \pm K_\pm, \quad [K_+, K_-] = K_0; \quad (3.40)$$

which are the typical relations in $\mathfrak{su}(2)$ Lie algebra. From these relations, one also obtains $[K_0, K_+K_-] = 0$, $[a^\dagger a + K_0, a^\dagger K_- + aK_+] = 0$. The total Hamiltonian is now can be summed by two commuting parts: $H_0 = \hbar\omega(a^\dagger a + K_0 + 1)$ and $H_I = H - H_0$, more explicitly,

$$H_I = \hbar[(\omega_a - \omega - v)(1 + K_0) + 2vK_+K_- + \sqrt{2}f(a^\dagger K_- + aK_+)]. \quad (3.41)$$

The evolution operator $U(t) = e^{-iHt/\hbar}$ can be disentangled as $U(t) = e^{-iH_0t/\hbar}e^{-iH_I t/\hbar}$. Both the two parts have compact forms of the generators, therefore, it would be convenient to use the four eigenstates of the operator K_0 as bases:

$$|-\rangle = |g_1g_2\rangle, |0\rangle = K_+|-\rangle, |+\rangle = K_+^2|-\rangle; |0'\rangle = (|g_1e_2\rangle - |e_1g_2\rangle)/\sqrt{2}, \quad (3.42)$$

correspondingly, $K_0|\pm\rangle = \pm|\pm\rangle$, $K_0|0\rangle = K_0|0'\rangle = 0$.

Notice that, whichever operator K_0, K_\pm acting on state $|0'\rangle$, the result is zero and the other states form a complete set under the three operations as well as identity operation. That is, the dynamical behavior of atomic part can be described by $U(1) \oplus SO(3)$. We have also the completeness relationship

$$|-\rangle\langle -| + |+\rangle\langle +| + |0\rangle\langle 0| = \mathbb{I}_3, \quad \mathbb{I}_3 + |0'\rangle\langle 0'| = \mathbb{I}_4.$$

When the evolution operator acts on $|0'\rangle\langle 0'|$, it gives rise to a trivial result:

$$e^{-iHt/\hbar}|0'\rangle\langle 0'| = e^{-i\omega t a^\dagger a}|0'\rangle\langle 0'|. \quad (3.43)$$

The non-trivial evolution of the states now is restricted in the 3-dimensional Hilbert space spanned by the bases $\{|\pm\rangle, |0\rangle\}$. In the new matrix representation, we have $K_- = |0\rangle\langle +| + |-\rangle\langle 0|$, $K_0 = |+\rangle\langle +| - |-\rangle\langle -|$ and $K_+K_- = \mathbb{I}_3 - |-\rangle\langle -|$. The interaction Hamiltonian now becomes

$$H_I = \hbar \begin{pmatrix} 0 & \sqrt{2}fa^\dagger & 0 \\ \sqrt{2}fa & 2v+w & \sqrt{2}fa^\dagger \\ 0 & \sqrt{2}fa & 2(v+w) \end{pmatrix}, \quad (3.44)$$

where

$$w = \omega_a - \omega - v. \quad (3.45)$$

The evolution of the entire system is governed by its initial state $|g_1g_2\rangle\langle g_1g_2| \otimes \rho_0$ and a unitary evolution operator $U(t) = e^{-iH_0t/\hbar}e^{-iH_I t/\hbar}$. The density matrix of the whole system at time t is $\tilde{\rho} = U|g_1g_2\rangle\langle g_1g_2| \otimes \rho_0 U^\dagger$. According to Born's rule, the two-atom excitation probability is related to the projection measurement $|e_1e_2\rangle\langle e_1e_2|$ on the atomic part, yielding $P = \text{Tr}[|e_1e_2\rangle\langle e_1e_2|\tilde{\rho}(t)]$, or equivalently

$$\begin{aligned} P &= \text{Tr}_F(\rho_0\langle g_1g_2|U^\dagger|e_1e_2\rangle\langle e_1e_2|U|g_1g_2\rangle) \\ &= \text{Tr}_F(\rho_0\langle g_1g_2|e^{iH_I t/\hbar}|e_1e_2\rangle\langle e_1e_2|e^{-iH_I t/\hbar}|g_1g_2\rangle). \end{aligned} \quad (3.46)$$

The Tr_F stands for the trace operation over the field variable. Without causing any confusion, we will drop the subscript hereafter.

Now let's focus our attention on how to solve the evolution operator $U_I(t) \equiv e^{-iH_I t/\hbar}$. The evolution operator is exactly solvable by following the procedures [PuriMMQO] in the three-level atom model. However, The expression is very complicated. What we need is the exact expression of $U_I|-\rangle$, obeying

$$i\hbar\partial_t U_I|-\rangle = H_I U_I|-\rangle. \quad (3.47)$$

In this matrix representation, one can always write $\hat{U}_I(t) = \sum_{jk} \hat{\psi}_{jk}|j\rangle\langle k|$, then $\hat{U}_I|-\rangle\langle -| = \sum_j \hat{\psi}_{j-}|j\rangle\langle -|$, the equations of interest are

$$i\partial_t \hat{\psi}_{--} = \sqrt{2}fa^\dagger\psi_{0-}; \quad (3.48a)$$

$$(i\partial_t - 2v - 2w)\hat{\psi}_{+-} = \sqrt{2}fa\hat{\psi}_{0-}; \quad (3.48b)$$

$$(i\partial_t - w - 2v)\hat{\psi}_{0-} = \sqrt{2}fa\hat{\psi}_{--} + \sqrt{2}fa^\dagger\psi_{+-}. \quad (3.48c)$$

To solve this equation, it is convenient to take $\hat{\phi}(t) = (a^\dagger\psi_{+-}, \hat{\psi}_{0-}, a\hat{\psi}_{--})^T$ as unknown operator-valued functions,

$$i\partial_t \hat{\phi} = \begin{pmatrix} 2(v+w) & \sqrt{2}fa^\dagger a & 0 \\ \sqrt{2}f & 2v+w & \sqrt{2}f \\ 0 & \sqrt{2}faa^\dagger & 0 \end{pmatrix} \hat{\phi}, \quad \hat{\phi}(0) = (0, 0, a)^T. \quad (3.49)$$

All the elements in the time independent coefficient matrix are commuting with each other, so that one can treat the operator-valued elements as common numbers. Let \hat{p}_\pm, \hat{p}_0 are three operator-valued eigenvalues of the coefficient matrix, by following a routine eigenvalue decomposition procedure of a square matrix, formally, the solution to the above equation reads

$$\hat{\phi}(t) = e^{-i\hat{p}_- t} \hat{\phi}_- + e^{-i\hat{p}_0 t} \hat{\phi}_0 + e^{-i\hat{p}_+ t} \hat{\phi}_+, \quad \hat{\phi}_- + \hat{\phi}_0 + \hat{\phi}_+ = \hat{\phi}(t=0). \quad (3.50)$$

The eigenvalue decomposition of the coefficient indeed shows the peculiar structure of the transition behaviors. The simple solution is determined in the characteristic cubic equation,

$$\hat{p}^3 - (4v+3w)\hat{p}^2 - 2[f^2(2aa^\dagger - 1) - (v+w)(2v+w)]\hat{p} + 4(v+w)f^2aa^\dagger = 0, \quad (3.51)$$

whose coefficients commute and which can be solved as an algebraic one and gives solutions \hat{p}_\pm, \hat{p}_0 . The exact solution of $\hat{\psi}_{+-} = 2f^2 a \hat{\Psi}_{aa^\dagger} a$ is expressed as

$$\hat{\Psi}_{aa^\dagger} = \frac{e^{-i\hat{p}_- t}}{(\hat{p}_- - \hat{p}_0)(\hat{p}_- - \hat{p}_+)} + \frac{e^{-i\hat{p}_0 t}}{(\hat{p}_0 - \hat{p}_-)(\hat{p}_0 - \hat{p}_+)} + \frac{e^{-i\hat{p}_+ t}}{(\hat{p}_+ - \hat{p}_0)(\hat{p}_+ - \hat{p}_-)}. \quad (3.52)$$

The transition probability now reads $P = \text{Tr}_F(\rho_0 \hat{\psi}_{+-}^\dagger \hat{\psi}_{+-})$. Function $\hat{\Psi}_{aa^\dagger}$ depends on aa^\dagger and therefore $\hat{\Psi}_{aa^\dagger} a = a \hat{\Psi}_{a^\dagger a}$. Thus the transition probability now becomes

$$P = \text{Tr}_F(\rho_0 \hat{\psi}_{+-}^\dagger \hat{\psi}_{+-}) = 4f^4 \text{Tr}_F(\rho_0 a^{\dagger 2} a \hat{\Psi}_{aa^\dagger}^\dagger \hat{\Psi}_{aa^\dagger} a) = 4f^4 \text{Tr}_F(\rho_0 a^{\dagger 2} a^2 \hat{\Psi}_{a^\dagger a}^\dagger \hat{\Psi}_{a^\dagger a}). \quad (3.53)$$

As we can see from the above expression, the response function $\hat{\Psi}_{a^\dagger a}^\dagger \hat{\Psi}_{a^\dagger a}$ also depends on the photon number operator $a^\dagger a$, and in general $\langle \hat{\Psi}_{a^\dagger a}^\dagger \hat{\Psi}_{a^\dagger a} \rangle \neq \hat{\Psi}_{\langle a^\dagger a \rangle}^\dagger \hat{\Psi}_{\langle a^\dagger a \rangle}$ unless the photon state is a number state $|n\rangle$. In this sense, the photon number statistics are important to the excitation probability, not only the intensity.

Because of completeness relationship $\sum_n |n\rangle \langle n| = \mathbb{I}$ in the Fock space, we have

$$a^{\dagger 2} a^2 \hat{\Psi}_{a^\dagger a}^\dagger \hat{\Psi}_{a^\dagger a} = \sum_n (n^2 - n) |\Psi_n|^2 |n\rangle \langle n|, \quad (3.54)$$

and thus the transition probability is the sum of contributions from all the diagonal elements of the state,

$$P = 4f^4 \text{Tr}(\rho_0 \hat{\Psi}_{a^\dagger a}^\dagger a^{\dagger 2} a^2 \hat{\Psi}_{a^\dagger a}) = 4f^4 \sum_{n \geq 2} (n^2 - n) |\Psi_n|^2 \langle n | \rho_0 | n \rangle. \quad (3.55)$$

We see that the coherence or the interference between different Fock state do not play a role in the probability.

D.1.2 Evolution at the perfect resonance

The phenomena of resonances in Physics are of great interest since they are the witnesses of the strongest correlations or couplings between subsystems. In most quantum optical systems, even for some ultrafast pulses, phase is the fastest varying quantity, the perfect resonance condition in fact manifests an offset to bring the whole system to a more stationary situation with respect to the fastest varying factors. Now let's have a closer look at the behaviors under perfect 2P2A condition $2\omega = \omega_+ + \omega_-$ which is equivalent to $v + w = 0$.

The solution of the cubic characteristic equation (3.51) is

$$p_0 = 0, \hat{p}_\pm = v/2 \pm \sqrt{v^2/4 + 2f^2(2aa^\dagger - 1)}. \quad (3.56)$$

Correspondingly,

$$\hat{\Psi}_{aa^\dagger}^{rsn} = \frac{1}{\hat{p}_+ - \hat{p}_-} \left[\frac{1 - e^{-i\hat{p}_- t}}{\hat{p}_-} - \frac{1 - e^{-i\hat{p}_+ t}}{\hat{p}_+} \right]. \quad (3.57)$$

When the photon number is large $n \gg (v/f)^2$, the dominant term in the solution for p in (3.56) comes from the photon number, approximate values to \hat{p}_\pm are $\pm 2f\sqrt{n}$, as a result

$$2f^2 a^2 \hat{\Psi}_{a^\dagger a}^{rsn} |n\rangle \simeq -\sin^2(f\sqrt{nt}) |n-2\rangle. \quad (3.58)$$

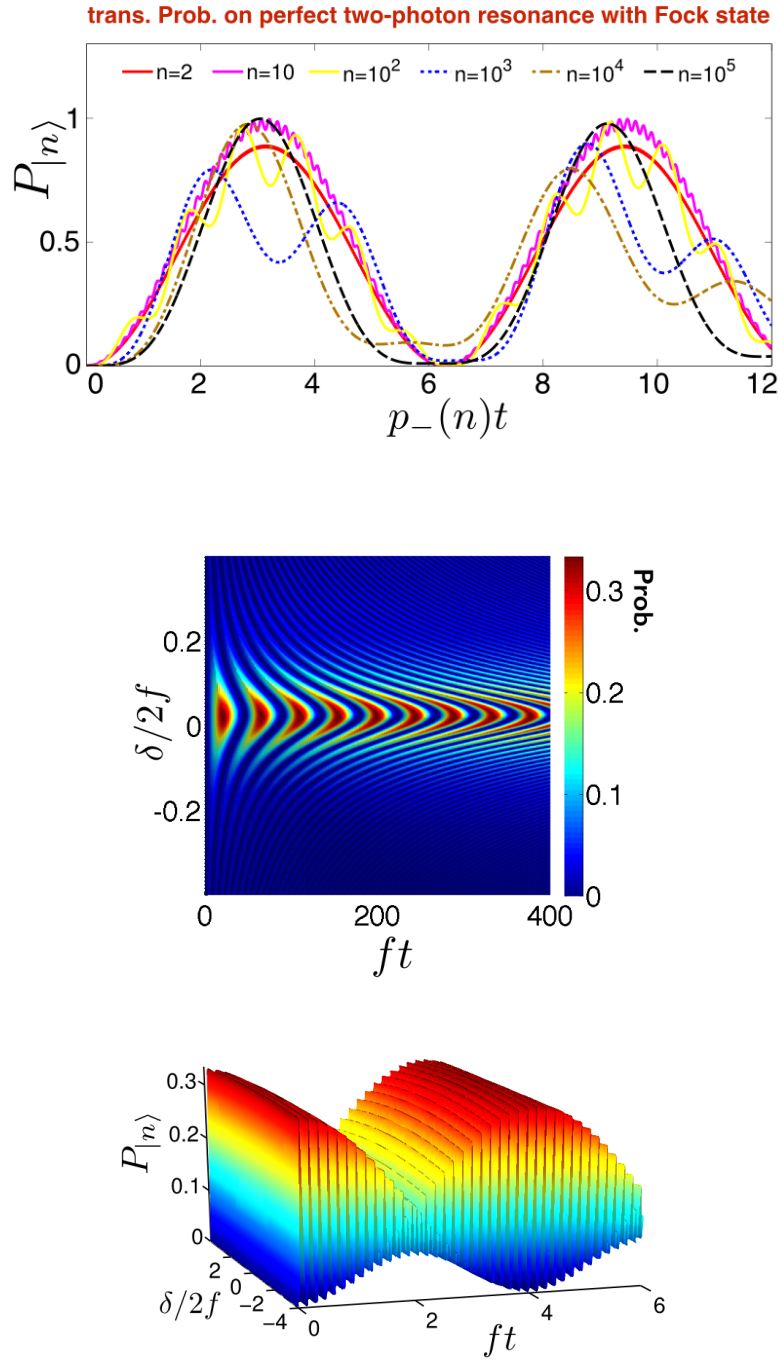


Figure 3.3: Figures of the response function $|\Psi_n|^2$ (hence $P_{|n\rangle}$) for different monochromatic Fock states. From the top graph, one finds some wrinkles which are induced by interferences between photons in a Fock state. These wrinkles can dramatically change the time dependence of the transition probability. With the increase of the photon number, the quantum collapse phenomenon become more noticeable, for example, the peaks become much narrower, see the case in which $n = 10^5$. The bottom two graphs are the 3d visualization for the transition probability with respect to the arguments of time ft and 2P2A detuning $\delta/2f$. One sees clearly the quantum collapses and revivals in this two graphs.

Note that $f\sqrt{n} \propto \sqrt{n/L}$ is related to the square root of the photon linear density in the 1D quantized box. We stress that this linear density should be fixed even when the length along z -direction becomes infinitely large.

The transition probability with a Fock state $|n\rangle$ yields

$$P = (n-1)n \sin^4(f\sqrt{n}t). \quad (3.59)$$

D.1.3 Comparison with the perturbation theory

The final result from perturbation theory in a case of monochromatic field and two frequency degenerate atoms at perfect resonance is

$$\mathcal{K} = 2f^2 \left(\frac{1 - e^{-ivt} - ivt}{v^2} \right) a^2. \quad (3.60)$$

When the response operator Eq.(3.57) is applied to a Fock state $|n\rangle$,

$$-2f^2 a^2 \hat{\Psi}_{a^\dagger a}^{rsn} |n\rangle = a^2 \frac{2f^2}{p_+ - p_-} \left[\frac{1 - e^{-ip_+ t}}{p_+} - \frac{1 - e^{-ip_- t}}{p_-} \right] |n\rangle, \quad (3.61)$$

where $p_{\pm} = v/2 \pm \sqrt{v^2/4 + 4f^2(n-1/2)}$. It is only when

$$n \ll \frac{v^2}{16f^2} + \frac{1}{2}, \quad (3.62)$$

therefore, $p_- \simeq 0, p_+ \simeq v$ and

$$|p_- T| \simeq 2Tf^2(2n-1)/v \ll 1 \Leftrightarrow T \ll v/2f^2(2n-1), \quad (3.63)$$

that $-2f^2 a^2 \hat{\Psi}_{a^\dagger a}^{rsn} |n\rangle$ for any time $t \in [0, T]$ degrades itself into $\mathcal{K}|n\rangle$.

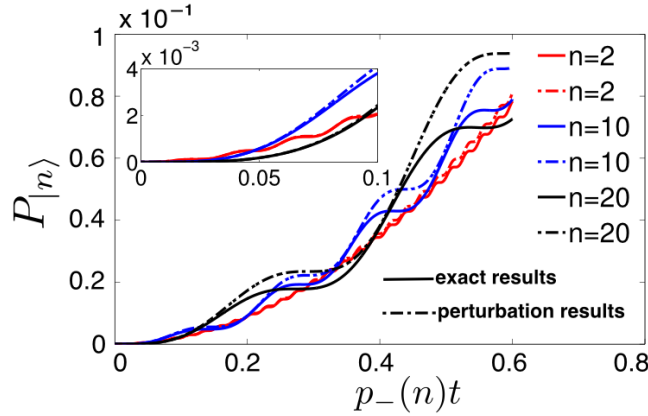


Figure 3.4: The transition probability for different Fock states. Here we take $v/f = 40$. One finds for the Fock state with small photon number, the mismatch between the exact result and the approximate result is small. For large-number Fock state, the mismatch becomes noticeable even at a short time. This figure shows the perturbation result is valid for short times. For longer times, one needs consider higher order corrections.

With a monochromatic field, in the most general case, noticing that the varying phase functions are in terms of $\omega - \omega_{\pm}$ in perturbation theory while those factors are in terms of $p_{0,\pm}(n) = \langle n | \hat{p}_{0,\pm} (a^\dagger a) | n \rangle$ which are n -dependent in the exact solution, $p_{0,\pm}(n)$ are in fact the three roots of $P_3(p, f^2) = 0$,

$$P_3(p, f^2) = p^3 - (4v + 3w)p^2 - 2[f^2(2n - 1) - (v + w)(2v + w)]p + 4(v + w)f^2n. \quad (3.64)$$

If there are only few photons in the total system, the field intensity is then weak and $p_{0,\pm}(n)$ are approximately $p_{0,\pm}(n) \simeq \tilde{p}_{0,\pm} + \Delta p_{0,\pm}$, where $\tilde{p}_{0,\pm} = p_{0,\pm}|_{f=0}$

$$\tilde{p}_0 = 0, \quad \tilde{p}_- = w + 2v; \quad \tilde{p}_+ = 2(v + w); \quad (3.65)$$

are three roots of $P_3(\tilde{p}, 0) = 0$ and hence

$$P_3(p_j, f^2) \simeq \frac{\partial P_3(\tilde{p}_j, 0)}{\partial p} \Delta p_j + \frac{\partial P_3(\tilde{p}_j, 0)}{\partial f^2} f^2 + \dots \quad (3.66)$$

if all the derivatives exist. Such expansion holds on the condition that higher order corrections of p_j are much less than the lower order ones. The above analyses do not apply when one of \tilde{p}_{\pm} is by accident equal to 0, which represents a resonance of single-photon or two-photon where often exists singularity.

Function $\langle n - 2 | \hat{\psi}_{+-} | n \rangle$ degrades into $\langle n - 2 | \mathcal{K} | n \rangle$ during the whole period $[0, T]$ provided that $|T \Delta p_j| \ll 1$, or more explicitly,

$$|T \Delta p_j| = 2f^2 T \left| \frac{2n(v + w) - (2n - 1)\tilde{p}_j}{3\tilde{p}_j^2 - 2\tilde{p}_j(4v + 3w) + 2(v + w)(2v + w)} \right| \ll 1. \quad (3.67)$$

Therefore the upper bound T for the validation of the perturbation theory with respect to time should satisfy

$$T \ll \min \left\{ \frac{v}{2(n - 1)f^2}, \frac{v}{2(2n - 1)f^2}, \frac{v}{2nf^2} \right\} = \frac{v}{2(2n - 1)f^2}. \quad (3.68)$$

when it is close to two-photon resonance.

D.2 Quasi mono-chromatic pure field

In this section, we will try to extend our conclusion to the case of a quasi-monochromatic field case which contains M frequency modes. Such a model is suitable for description of the interaction between two frequency-degenerate atoms and frequency comb field (or a pulse train), providing that the bandwidth of the whole comb is much less than its central frequency.

The total Hamiltonian now changes to two commuting parts: $H_0 = \hbar\omega \sum (a_m^\dagger a_m + K_0 + 1)$ and $H_I = H - H_0$, more explicitly,

$$H_I = \hbar[(\omega_a - \omega - v)(1 + K_0) + 2vK_+K_- + \sqrt{2M}f(a^\dagger K_- + aK_+)], \quad (3.69)$$

where $a = M^{-1/2} \sum_m a_m$. Because of the Bosonic commutation relations, one finds the non vanishing commutation relation $[a, a^\dagger] = 1$. Because of the normalized factor $M^{-1/2}$, the previous coupling constant f scales as $\sqrt{M}f$. The evolution operator $U(t) = e^{-iHt/\hbar}$

can also be disentangled as $U(t) = e^{-iH_0t/\hbar}e^{-iH_I t/\hbar}$. Following the same procedures, we once again have

$$P = 4M^2 f^4 \text{Tr}_F(\rho_0 a^{\dagger 2} a^2 \hat{\Psi}_{a^\dagger a}^\dagger \hat{\Psi}_{a^\dagger a}). \quad (3.70)$$

but with $\hat{p}_{\pm,0}$ now being the solutions of a new cubic equation

$$p^3 - (4v + 3w)p^2 - 2[Mf^2(2aa^\dagger - 1) - (v + w)(2v + w)]p + 4M(v + w)f^2aa^\dagger = 0. \quad (3.71)$$

Though from direct calculation, one finds $[\sum_m a_m^\dagger a_m, a^\dagger a] = 0$, a Fock state $|2_k\rangle$ of k -th frequency mode is an eigenstate of $\sum_m a_m^\dagger a_m$ but not an eigenstate of $a^\dagger a$. This kind of facts reveals the importance of the diffusion during the interaction process.

For M discrete frequency modes, one can choose the discrete Fourier series as basis: $\mathbf{F}_n = M^{-1/2}(\dots, e^{2i\pi nm/M}, \dots)^T$. The completeness relation for the mode distribution reads

$$\mathbb{I} = \sum_n \mathbf{F}_n \mathbf{F}_n^H. \quad (3.72)$$

Let $\hat{\mathbf{a}}$ be a column vector with $(\hat{\mathbf{a}})_m = a_m$ and $A_m = \mathbf{F}_m^H \hat{\mathbf{a}}$ or the inverse transformation $a_n = M^{-1/2} \sum_m e^{-2i\pi nm/M} A_m$, then $a = M^{-1/2} \sum_m a_m = \mathbf{F}_0^H \hat{\mathbf{a}} = A_0$. One can also have the non vanishing commutation relation:

$$[A_m, A_n^\dagger] = [\mathbf{F}_m^H \hat{\mathbf{a}}, (\mathbf{F}_n^H \hat{\mathbf{a}})^\dagger] = M^{-1} \sum_{jk} e^{2i\pi(nj - mk)/M} [a_k, a_j^\dagger] = \delta_{mn}. \quad (3.73)$$

These relations combined with Eq.(3.72) imply the invariability of the total photon number operator under mode transformations:

$$\hat{N}_{tot} = \sum_m a_m^\dagger a_m = \sum_m A_m^\dagger A_m. \quad (3.74)$$

We will use the notation $|\cdot, \mathbf{F}_n\rangle$ to represent whatever state of the \mathbf{F}_n mode. Notice that, for any states $|\psi, \mathbf{F}_0^\perp\rangle$ which do not contain any photons in the \mathbf{F}_0 mode, one has $a^\dagger a |\psi, \mathbf{F}_0^\perp\rangle = A_0^\dagger A_0 |\psi, \mathbf{F}_0^\perp\rangle = 0$. Subsequently, the transition probability is simplified to

$$P = 4M^2 f^4 \text{Tr}(\rho_{0, \mathbf{F}_0} a^{\dagger 2} a^2 \hat{\Psi}_{a^\dagger a}^\dagger \hat{\Psi}_{a^\dagger a}). \quad (3.75)$$

The reduced density matrix ρ_{0, \mathbf{F}_0} is traced over all the modes which are perpendicular to the mode \mathbf{F}_0 . At the first sight, for any states with given photon spectra $(\dots, \text{Tr}(a_m^\dagger a_m \rho_0), \dots)$, because of the partial trace operation, it seems that the entanglement or even correlations between modes are irrelevant to the enhancement of the co-excitation. However, this conjecture may not true, at least, is not obviously true. The formula for transition probability can be re-expressed as

$$P = 4M^2 f^4 \sum_{n \geq 2} (n^2 - n) |\Psi_n|^2 \langle n, \mathbf{F}_0 | \rho_{0, \mathbf{F}_0} | n, \mathbf{F}_0 \rangle, \quad (3.76)$$

from which we know that if there is no possibility to have more than one photon in the \mathbf{F}_0 mode even though there may exist a very large number of photons in the total state, the phenomenon of the co-excitation of the both atoms will not happen. Note that, the scale factor $n^2 - n$ is different for different Fock states $|n \geq 2, \mathbf{F}_0\rangle$, not for those Fock states $|n, \omega_m\rangle$ of a specific frequency mode ω_m . The value of the transition probability is highly related to the mean photon number of the mode \mathbf{F}_0 and the all order correlation functions

$g_{\mathbf{F}0}^{(n)}$. This is a good hint of the elements to prepare a photon state that manipulates the excitation of the atoms: a proper mode and its corresponding photon statistics.

One may notice a factor $M^2 f^4$ that is before the sum operation in Eq.(3.76). This M^2 term will be removed by the explicit expressions of $|\Psi_n|^2$.

E Conclusion

In this chapter, we have presented in detail the model of two-photon–two-atom interaction. By using perturbation theory, we have derived a general expression for the leading transition probability depending on the quantum state which is used for the excitation. In a special exactly solvable model, we obtained an exact solution to the evolution and found the breakdown conditions to the perturbation theory with respect to long time and large photon number therefore strong coupling between field and atoms. We then considered the quasi-monochromatic or pulsed field case, in the same section and found that what really matters is the photon statistics in a special mode in which the photons of different frequency components are in phase, rather than the total photon number.

Two-photon–two-atom transition without atomic interaction

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Two-photon–two-atom excitation is a most fundamental process in quantum optics. When there is no interaction between the two atoms, each atom can be excited independently. However, the total transition probability is not a simple product of two transition probabilities with respect to both atoms even the photon source is monochromatic. This is because a photon can be firstly absorbed either by one atom or by the other one, therefore, a which-way excitation is naturally embedded in the 2P2A transition. Besides, the quantum interferences from photon pairs in the source should also

be embedded. Though a two-atom system without an atomic interaction is a simplest situation for 2P2A transition, it is able to show some complicated quantum interferences from a photon source.

In this chapter, we focus our attention on a simplest situation: a two-atom system without any atomic interactions and the mean frequencies of the photons and the proper frequencies corresponding to the atomic Bohr energy being far apart from each other. Under this condition, the which-way interferences are not important, so the 2P2A excitation is determined by the properties of photons. At a first step, we will consider a model that without atomic decoherence which means if an atom is excited, it can stay in that state forever and the physical origins of an enhancement of transition probability. **This work is published [Zheng2013]**. Then we will try to find which kind of photon source are optimized to the maxima transition probability with a given photon spectra. In the third part, we will discuss the best time for observation of the transition probability in the case of small decay rates of the excited atoms.

A Do entangled photons induce 2P2A transitions more efficiently than other states of light ?

Quantum entanglement and its inherent non local properties are among the most fascinating and challenging features of the quantum world. In addition, entanglement plays a central role in quantum information [Plenio2007, Gerardo2007, Reid2009, Horodecki2009, Pan2012]. Since its first description in the 1930's [Schrödinger1935], and in spite of the decisive contribution of J. Bell [Bell1964] and the subsequent experimental studies [Aspect1982], entanglement appears as a rather mysterious and puzzling property even for bipartite systems. In particular distinguishing between effects related to genuine entanglement and those related to the quantum correlations measured on a single quantum observable is a difficult task [Treps2005], as can be seen for example by the great number of papers about quantum discord [Ollivier2001, Modi2012]. Some time ago, a paper was published [Muthukrishnan2004] which showed that some entangled states are able to induce transitions in quantum systems that factorized states cannot excite. The physical problem studied in that paper is therefore a good test bench to examine in detail in a simple situation the role of entanglement and of correlations not related to entanglement. This is the purpose of the present subject.

The problem under consideration is the probability of two-photon two-atom (2P2A) excitation, in the situation where the two atoms are of different species and have different transition frequencies and the light to which the atoms are submitted is in general non-resonant for each one, but resonant for the system of two atoms. Two photon absorption by single atoms or molecules have been studied since 1931 [Göpert-Mayer1931] and remains a current subject of theoretical and experimental research [Mollow1968, Bjorkholm1974, Fei1997, Lloyd2008, Kastella2011]. When the atoms have more than one intermediate state, many important features, including cross section cancellation and enhancement, are observed [Bjorkholm1974]. These features have recently been shown to be applicable in characterizing the two-photon quantum states [Kastella2011].

It has also been known for a long time that the two photons resonant excitation of two different atoms is indeed possible when the two atoms are interacting [RiosLeite1980]. A nearly monochromatic light beam will have a resonant two photon absorption peak when tuned across the average frequency of the two atoms. Different experiments have since then confirmed this theoretical prediction [White1981, Hettich2002, P-Penãfiel2012]. In addition to a direct potential interaction between the atoms, like the dipole-dipole, cooperative 2P2A has also been predicted for pairs of atoms inside an optical cavity [Kim1998]. In this case the physical interaction is mediated by the radiation background surrounding the atoms. Ref [Muthukrishnan2004] addresses the case of two photon absorption in absence of interaction between the two atoms, with the excitation made using some particular entangled state of light. The conclusion of the authors of [Muthukrishnan2004] is that *in some situations entanglement can replace a real physical interaction*, which is a far reaching statement and an important physical property related to entanglement. Surprisingly, this question did not attract much attention for several years. The same subject was also considered, but in the context of spin entanglement in electron paramagnetic resonance by Salikhov [Salikhov2003]. More recently [Richter2011, Dayan2007], the related problem of interaction with pairs of broadband spectrum photons has been discussed.

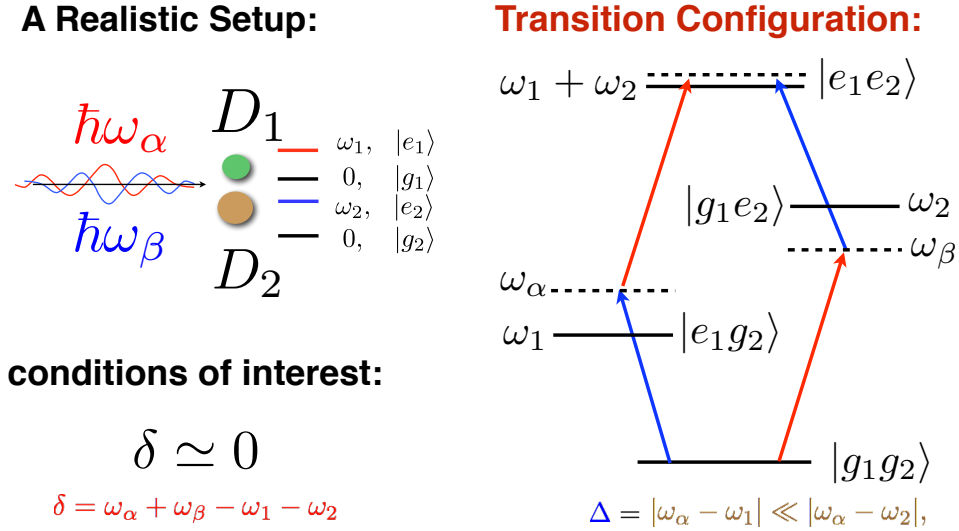


Figure 4.1: A sketch of the two-atom–two-photon interaction model. The two atoms here are without atomic interactions between each other, and the Bohr frequencies of excited states are ω_1, ω_2 respectively. Both the two central frequencies of the two incident beams ω_α and ω_β are far from resonances with single-atoms but their sum is almost equal to the sum of the two Bohr frequencies ($\delta = \omega_\alpha + \omega_\beta - \omega_1 - \omega_2 \simeq 0$). For sake of simplicity, we will take $\Delta = |\omega_1 - \omega_\alpha| (\simeq |\omega_2 - \omega_\beta|)$ as the minimum single-photon–single-atom detuning.

In this section, we consider in more detail the problem of two-atom excitation in order to determine the exact role of entanglement in the process. More precisely, we determine the probability of 2P2A excitation by different multi-modal states of light. From these results we draw conclusions on the respective role of entanglement and of correlations not related to entanglement in such a process. Sec. A.1 gives the general framework in which the problem is treated and the expression of relevant transition probability from a second order perturbation theory. Results for various different two photon states, introduced in Sec. A.2, are given in sections from A.3 to A.6. Finally, in Sec. A we discuss different hypotheses for the physical origin of the enhancement of the 2P2A process.

A.1 Model

We consider here the model used in Chap. 3 that two different two-level atoms labeled (1) and (2), having ground and excited states $|g_i\rangle$ and $|e_i\rangle$ ($i = 1, 2$), corresponding Bohr frequencies ω_i , and spontaneous emission rates γ_i , interacting with a quantized field. We assume that the mean atomic excitation time is much shorter than the lifetimes of the two excited atoms so that we can consider that the two excited states have infinite lifetimes ($\gamma_{1,2} \simeq 0$) and keep for ever their excitation. For the sake of simplicity we will assume that the light source is far from the atoms, so that the only non-empty modes are plane-wave modes having a single propagation direction Oz and a single polarization. Thus one can use annihilation operators depending only on the frequency $a(\omega_\ell) = a_\ell$. For simplicity, we will set the locations of atoms $z_1 = z_2 = 0$ and hence will not consider propagation effects. In this case, we may approximate the coupling strength defined in Chapter 3 as

$$f_i(\omega_\ell) \simeq f_i(\omega_i).$$

The evolution of the whole system is described by a unitary operator U , and the input light ρ_0 can be either a pure two-photon state $|\Psi_\mu\rangle = \sum_{kq} c_{kq}^\mu |1_k, 1_q\rangle$ or a mixed state in its spectral decomposition form $\rho_0 = \sum_\mu p_\mu |\Psi_\mu\rangle\langle\Psi_\mu|$. The probability of 2P2A excitation is given in this case by

$$P(t) = \sum_\mu p_\mu \langle\Psi_\mu, g_1, g_2|U^\dagger(t)|0, e_1, e_2\rangle\langle 0, e_1, e_2|U(t)|\Psi_\mu, g_1, g_2\rangle. \quad (4.1)$$

Due to the weak coupling between the light field and the two atoms, the leading term in the evolution is determined by

$$P(t) \simeq \sum_{jkmn} \mathcal{K}_{jk}^* \mathcal{K}_{mn} \text{Tr}(a_j^\dagger a_k^\dagger a_m a_n \rho_0). \quad (4.2)$$

according to the results in Chapter 3. The explicit expression of the response function without atomic decoherence, which has been derived in Eq. (3.37), reads

$$\mathcal{K}_{mn} = f_1(\omega_m) f_2(\omega_n) \frac{1 - e^{i(\omega_1 - \omega_m)t}}{\omega_m - \omega_1} \frac{1 - e^{i(\omega_2 - \omega_n)t}}{\omega_n - \omega_2}. \quad (4.3)$$

Note that $\omega_m = \omega_1$ and $\omega_n = \omega_2$ are not singular points of the function \mathcal{K}_{mn} . In fact, when the two variables take their magnitudes that are equal to the atomic frequencies respectively, the response function \mathcal{K}_{mn} gives rise to a simple result

$$\mathcal{K}_{mn}|_{\omega_m=\omega_1, \omega_n=\omega_2} = -f_1(\omega_1) f_2(\omega_2) t^2, \quad (4.4)$$

which shows the t^2 dependence. Such a dependence is common by using perturbation method in dealing with short-time behaviors of an interacting system. In the case of a continuous frequency distribution of photons in a pure two-photon state $|\Psi_\mu\rangle$, it is also convenient to introduce the transition probability amplitude:

$$A_\mu = \frac{L^2}{4\pi^2 c^2} \iint d\omega_m d\omega_n \mathcal{K}_{mn} (c_{mn}^\mu + c_{nm}^\mu). \quad (4.5)$$

From this equation, we see there are two terms in the bracket. The sum which implies a quantum interference between two paths to excite the atoms, is invariant by exchanging their subscripts of the two terms.

Note that the coefficient \mathcal{K}_{mn} is the product of two factors which represent the response of each atom to the field. When time t goes to infinity these two factors behave roughly like two independent Dirac delta functions centered on the atom resonances (we will detail this argument in Sec. A.6). The 2P2A excitation probability is indeed induced by the individual wings of the incident light spectrum which are resonant with the atoms. Consequently, if one photon is absorbed by one atom, there is no reason why the second photon should be absorbed by the second atom in a way correlated to the absorption of the first photon. In other words, the 2P2A excitation phenomenon has no reason a priori to have a resonant behavior when the 2P2A resonance condition $\omega_1 + \omega_2 = \omega_m + \omega_n$ is fulfilled.

However, as the process is non-linear and involves two atoms, it can be enhanced by taking advantage of correlation effects between the atoms or between the photons :

- A first possibility consists in introducing an interaction between the two atoms. Let $\omega_u \simeq \omega_1 + \omega_2$ be the maximal Bohr frequency of the two-atom system. If one photon with frequency ω_k is absorbed, then the two atoms will be more likely to absorb another photon with frequency $\omega_u - \omega_k$ in a resonant two-photon process [RiosLeite1980, Andrews1983]. This was experimentally demonstrated in [White1981, Hettich2002, P-Penãfiel2012] using nearly degenerate photon pairs.
- A second possibility is to use correlated photons to interact with the two atoms. Let us consider a source that emits correlated photons. If a photon is absorbed by one atom, then the remaining atom will interact with its correlated photon with a higher probability, leading to enhanced 2P2A resonance (we will detail this argument and the kind of correlation needed in Sec. A).

We will now discuss the ideas by having a closer look at different possible light states likely to induce such a 2P2A transition.

A.2 Entangled, correlated-separable and factorized two-photon states

Before we go further, let us discuss the different kinds of two-photon states that we will consider in the following. Starting from entangled pure quantum state $|\Psi\rangle$, having a density matrix $\rho_0 = |\Psi\rangle\langle\Psi|$ of matrix elements $\rho_{0;kk'qq'} = \langle 1_k, 1_q | \rho_0 | 1_{k'}, 1_{q'} \rangle$, one can construct others that have the same mean energy and the same single photon spectrum, and hence that would give the same transition probabilities for a single photon resonance. We choose two special cases that will allow a quantitative evaluation of the role of correlations :

- The first one is defined as

$$\rho_1 = \sum_{k,q} \rho_1(\omega_k, \omega_q) |1_k, 1_q\rangle\langle 1_k, 1_q|, \quad \rho_1(\omega_k, \omega_q) = \rho_{0;kkqq}. \quad (4.6)$$

It is the diagonal part of ρ_0 . It has lost any temporal field coherence and is time independent. It is actually a *correlated-separable state* [Duan2000], which results from the "disentanglement" of the previous one. It gives rise, however, to correlations between its two parties.

- The second one is defined as

$$\rho_2 = \left(\sum_{k,q'} \rho_1(\omega_k, \omega_{q'}) |1_k\rangle\langle 1_k| \right) \otimes \left(\sum_{k',q} \rho_1(\omega_{k'}, \omega_q) |1_q\rangle\langle 1_q| \right). \quad (4.7)$$

This is a fully factorized state, which does not give rise to any correlation whatsoever. The diagonal element at $|1_k, 1_q\rangle\langle 1_k, 1_q|$ is $\rho_2(\omega_k, \omega_q) = \sum_{m,n} \rho_1(\omega_k, \omega_m) \rho_1(\omega_n, \omega_q)$.

These states will induce 2P2A excitation with respective probabilities $P_1(t)$ and $P_2(t)$.

The two diagonal density matrices (ρ_1, ρ_2) describe two cw fields while the entangled pure state ρ_0 describes a pulse. As a result, at a time t , the flow of energy having interacted with atoms in each state is different. However, as shown in Appendix B, this quantity is quite the same in each state when $t = L/c$. For comparison, we will take $t = L/c$ through the whole section.

We are of course interested in cases where an increase in the excitation probability is expected. When this happens with $P(L/c) \gg P_2(L/c)$ and $P(L/c) \gg P_1(L/c)$, then entanglement is indeed the key to efficient 2P2A transition, whereas if $P(L/c) \simeq P_1(L/c) \gg P_2(L/c)$, correlations, of quantum or classical origin, are more important than entanglement in the present problem.

A.3 2P2A induced by different two-photon quantum field states

We will now examine the efficiency of various multimode light states for the simultaneous excitation of the two atoms.

A.3.1 Two quasi-monochromatic uncorrelated photons

Let us begin by the simplest case : two uncorrelated photon wavepackets of mean frequencies ω_α (much closer to ω_1) and ω_β , and respective spectral widths γ_α and γ_β much bigger than the detecting atom spectral widths γ_1 and γ_2 , emitted by two uncorrelated atoms excited at the same time in the past and arriving at the detecting atoms position at $t = 0$, described therefore by the two-photon state $|\psi^{11}\rangle$ with

$$|\psi^{11}\rangle = \sum_k \frac{g_\alpha(\omega_k)}{\omega_k - \omega_\alpha + i\gamma_\alpha} |1_k\rangle \otimes \sum_q \frac{g_\beta(\omega_q)}{\omega_q - \omega_\beta + i\gamma_\beta} |1_q\rangle. \quad (4.8)$$

It is the tensor product of two single-photon wave packets [GAFIQO] of duration γ_α^{-1} and γ_β^{-1} . In the calculation of the probability amplitude in Eq.(4.5), we will replace the sum over modes in Eq.(4.8) by the double integral $(L^2/4\pi^2c^2) \iint_0^\infty d\omega_k d\omega_q$, extend each integration domain to the whole real axis and use the residue theorem [ScullyQO]. When $\gamma_\alpha t \gg 1$ and $\gamma_\beta t \gg 1$ while keeping $\gamma_1 t \ll 1$ and $\gamma_2 t \ll 1$, the transition probability amplitude is

$$A^{11} \simeq \frac{L^2 f_1(\omega_1) f_2(\omega_2) g_\alpha(\omega_1) g_\beta(\omega_2)}{c^2 (\omega_1 - \omega_\alpha + i\gamma_\alpha) (\omega_2 - \omega_\beta + i\gamma_\beta)}. \quad (4.9)$$

The coefficients $g_\mu(\omega_\ell)$ can be taken as constant and the normalization of the two-photon state imposes

$$g_\alpha g_\beta = \frac{2c\sqrt{\gamma_\alpha \gamma_\beta}}{L}, \quad (4.10)$$

so that the transition probability P^{11} is

$$P^{11} = \frac{P_0 \gamma_\alpha \gamma_\beta}{[(\omega_1 - \omega_\alpha)^2 + \gamma_\alpha^2][(\omega_2 - \omega_\beta)^2 + \gamma_\beta^2]}, \quad (4.11)$$

where $P_0 = d_1^2 d_2^2 \omega_1 \omega_2 / \hbar^2 \epsilon_0^2 c^2 S^2 = 36\pi^2 \gamma_1 \gamma_2 c^4 / \omega_1^2 \omega_2^2 S^2$, in which we have used the expression for the spontaneous emission rate $\Gamma_i = 2\gamma_i$, $\Gamma_i = d_i^2 \omega_i^3 / (3\pi\epsilon_0 \hbar c^3)$ [GAFIQO, ScullyQO].

A.3.2 Double resonance

According to (4.11), the best transition probability will be achieved when the photons are separately resonant with the two atoms $\omega_\alpha = \omega_1$ and $\omega_\beta = \omega_2$, namely, the double-resonance (DR) condition, under which the transition probability is then equal to

$$P_{DR}^{11} = \frac{P_0}{\gamma_\alpha \gamma_\beta}, \quad (4.12)$$

which can be written in a more general way

$$P_{DR}^{11} = \frac{P_0}{S_{fr}}, \quad (4.13)$$

where S_{fr} is the effective area of frequency distribution $|c_{kq}|^2$ in the (ω_k, ω_q) plane (see Fig. 6.2). This result turns out to be general and implies that all pure states having the same effective areas S_{fr} , entangled or not, will produce the same doubly resonant transition probability. Thus we regard (4.12) as a universal result under the double resonance condition, and its value will serve as a reference for all subsequent transition probabilities.

A.3.3 Two-photon two-atom resonance

When none of the two photons are resonant with the two atoms, but where the sum of their two energies almost matches the sum of the two atomic energies $\omega_\alpha + \omega_\beta \simeq \omega_1 + \omega_2$, the transition probability (4.11) has in this case no resonant variation as a function of the 2P2A detuning

$$\delta = \omega_\alpha + \omega_\beta - \omega_1 - \omega_2. \quad (4.14)$$

When $\delta = 0$ the transition probability is :

$$P_{2P2A}^{11} = \frac{P_0 \gamma_\alpha \gamma_\beta}{\Delta^4} = P_{DR}^{11} \frac{\gamma_\alpha^2 \gamma_\beta^2}{\Delta^4}, \quad (4.15)$$

where Δ is the smallest frequency mismatch between the emitting atom frequencies, and the detecting atom frequencies are supposed to be much larger than the atomic widths. Without loss of generality, we have taken $\Delta = |\omega_\alpha - \omega_1| = |\omega_2 - \omega_\beta|$.

We then conclude that the special case of 2P2A excitation probability by uncorrelated photons is also non zero for any couple of frequencies $\omega_\alpha, \omega_\beta$; thus such a two-photon transition turns out not to be disallowed but simply induced by the wings of the two single photon frequency resonances. It is therefore very weak, as witnessed by the Δ^{-4} variation of probability.

A.3.4 Two photons produced by an atomic cascade

Let us now envision the case considered in [Muthukrishnan2004] of a two-photon light state produced by a three-level atom excited at a given time in the upper state that cascades down to the ground state on two successive transitions of Bohr frequencies successively equal to ω_α and ω_β . The corresponding spontaneous emission rates are γ_α and γ_β . We assume that the emitted light is wholly directed in the Oz direction of atoms (1) and (2) (by means of a parabolic mirror for example) and can only arrive after $t = 0$. It is described by a two-photon wave-packet with a coefficient c_{kq}^{cas} equal, at a time t long compared to the lifetimes of the two transitions, to [Muthukrishnan2004, ScullyQO]:

$$c_{kq}^{cas} = \frac{g_\alpha(\omega_k)g_\beta(\omega_q)}{[\omega_k + \omega_q - \omega_\alpha - \omega_\beta + i\gamma_\alpha][\omega_q - \omega_\beta + i\gamma_\beta]}. \quad (4.16)$$

Here this entangled non stationary state is produced by a cascade, so that the photon of frequency ω_q always arrives just after the photon of frequency ω_k . In addition, the probability to have photons of frequency sum $\omega_k + \omega_q$ close to $\omega_\alpha + \omega_\beta$ is high. We

have therefore an entangled state which is not only correlated in time but also anti-correlated in frequency. It is the time-energy analog of the position-momentum entangled state introduced by EPR, or of the field quadrature entangled state [ScullyQO, Reid2009, Khan2006].

Using the residue theorem to integrate over the frequencies ω_k and ω_q , the transition probability amplitude reads exactly as follows

$$A^{cas} = \frac{L^2}{c^2} \frac{g_\alpha(\omega_1)g_\beta(\omega_2)f_1(\omega_1)f_2(\omega_2)}{\omega_{\beta 2} - \delta - i(\gamma_\beta - \gamma_\alpha)} \left[\frac{1 - e^{-(\gamma_\beta + i\omega_{\beta 2})t}}{\omega_{\beta 2} - i\gamma_\beta} - \frac{1 - e^{-(\gamma_\alpha + i\delta)t}}{\delta - i\gamma_\alpha} \right] + (1 \leftrightarrow 2). \quad (4.17)$$

When $\gamma_{1,2}^{-1} \gg t \gg \gamma_{\alpha,\beta}^{-1}$, the four decaying terms in Eq. (4.17) are negligible, leading to a compact expression

$$A^{cas} = -\frac{L^2}{c^2} \frac{f_1(\omega_1)f_2(\omega_2)}{\delta - i\gamma_\alpha} \left[\frac{g_\alpha(\omega_1)g_\beta(\omega_2)}{\omega_{\beta 2} - i\gamma_\beta} + \frac{g_\alpha(\omega_2)g_\beta(\omega_1)}{\omega_{\beta 1} - i\gamma_\beta} \right], \quad (4.18)$$

where $\omega_{\mu\nu} = \omega_\mu - \omega_\nu$ is the frequency difference between frequency ω_μ and frequency ω_ν ; $\mu = \alpha, \beta, k, q$; $\nu = 1, 2$.

A.3.5 Double resonance

Starting from (4.18) and keeping only the largest term, one obtains in this case for the probability amplitude,

$$A_{DR}^{cas} \simeq \frac{L^2 f_1(\omega_1) f_2(\omega_2) g_\alpha(\omega_1) g_\beta(\omega_2)}{c^2 \gamma_\alpha \gamma_\beta}. \quad (4.19)$$

Using the same assumption as in the previous calculation, one finds for the probability

$$P_{DR}^{cas} = \frac{P_0}{\gamma_\alpha \gamma_\beta} = P_{DR}^{11}. \quad (4.20)$$

It is time independent because we are considering times much longer than the two-photon pulse of duration $\gamma_\alpha^{-1} + \gamma_\beta^{-1}$. As it is equal to the probability obtained with uncorrelated photons, we conclude that entanglement does not help in the fully resonant case, but does not harm either.

A.3.6 Two-photon two-atom resonance

Let us now turn to the 2P2A resonance case. One obtains in this case for the probability :

$$P_{2P2A}^{cas} \simeq \frac{L^2}{4c^2} \frac{P_0}{\delta^2 + \gamma_\alpha^2} \left[\frac{g_\alpha(\omega_1)g_\beta(\omega_2)}{\omega_2 - \omega_\beta} + \frac{g_\alpha(\omega_2)g_\beta(\omega_1)}{\omega_1 - \omega_\beta} \right]^2. \quad (4.21)$$

This expression, already obtained in [ScullyQO], shows that for this state the probability has indeed a resonant character around the 2P2A resonance $\delta = 0$. The transition probability P_{2P2A}^{cas} at the exact 2P2A resonance is then

$$P_{2P2A}^{cas} \simeq \frac{P_0}{\gamma_\alpha \gamma_\beta} \frac{\gamma_\beta^2}{\Delta^2} = P_{DR}^{11} \frac{\gamma_\beta^2}{\Delta^2}. \quad (4.22)$$

One therefore finds that the transition probability is in the present case smaller than P_{DR}^{11} by a factor $(\gamma_\beta/\Delta)^2$ at exact 2P2A resonance, as expected because one is now less

resonant than in the double-resonance case. One finds more importantly that P_{2P2A}^{casc} is larger than P_{2P2A}^{11} , i.e. than in the two-uncorrelated-photon case, by a factor $(\Delta/\gamma_\alpha)^2$, which can be very large. This enhancement of the 2P2A transition probability is the main result of [Muthukrishnan2004]. Entanglement may indeed significantly enhance the two-photon–two-atom process. To the best of our knowledge no experiment has been undertaken to show such a striking effect.

It must be emphasized that the present considerations do not imply that the atom cascade entangled state is the only one likely to produce such a significant increase in the transition probability. This is the reason why we will now consider other light quantum states which may also be of interest in the present problem.

A.4 Correlated and factorized states analogous to the atomic cascade

Let us now consider the two states that have the same energy and the same spectrum that we have introduced in Sec. A.2, namely, the correlated-separable state,

$$\rho_1 = \left(\frac{2c}{L}\right)^2 \sum_{kq} \frac{\gamma_\beta}{(\omega_{q\beta}^2 + \gamma_\beta^2)} \frac{\gamma_\alpha}{[(\omega_{q\beta} + \omega_{k\alpha})^2 + \gamma_\alpha^2]} |1_k, 1_q\rangle \langle 1_k, 1_q|, \quad (4.23)$$

and the factorized state,

$$\rho_2 = \left(\frac{2c}{L}\right)^2 \left(\sum_k \frac{\gamma_\alpha + \gamma_\beta}{\omega_{k\alpha}^2 + (\gamma_\alpha + \gamma_\beta)^2} |1_k\rangle \langle 1_k| \right) \otimes \left(\sum_q \frac{\gamma_\beta}{\omega_{q\beta}^2 + \gamma_\beta^2} |1_q\rangle \langle 1_q| \right). \quad (4.24)$$

The first one corresponds to an atomic cascade for which the starting time is random, thereby averaging to zero all the off-diagonal time-dependent terms in the density matrix; the second one characterizes a mixed state with two uncorrelated photons having the same spectrum as the initial cascade state. They give rise to the following transition probabilities

$$P_1 \simeq P_0 \frac{\gamma_\alpha \gamma_\beta}{\delta^2 + \gamma_\alpha^2} \left(\frac{1}{(\omega_1 - \omega_\beta)^2} + \frac{1}{(\omega_2 - \omega_\beta)^2} \right) \frac{t^2}{(L/c)^2}, \quad (4.25)$$

$$P_2 \simeq P_0 \gamma_\beta (\gamma_\alpha + \gamma_\beta) \left(\frac{1}{(\omega_1 - \omega_\beta)^4} + \frac{1}{(\omega_2 - \omega_\beta)^4} \right) \frac{t^2}{(L/c)^2}. \quad (4.26)$$

At exact 2P2A resonance, we have $P_1 \simeq P_{DR}^{11} \gamma_\beta^2 c^2 t^2 / (\Delta^2 L^2)$ and $P_2 \simeq P_{DR}^{11} \gamma_\alpha \gamma_\beta^2 (\gamma_\alpha + \gamma_\beta) c^2 t^2 / (\Delta^4 L^2)$. At any time t , one finds $P_1 \gg P_2$, since the spectral widths are much smaller than single-photon detunings. This fact shows that correlations indeed play an important role in the efficiency of the excitation.

Note that P_1 and P_2 depend on time, as can be expected in a situation where the detecting atoms, which have an infinite lifetime, are submitted to a stationary quantum state, and therefore to cw light. In order to compare P_1 and P_2 to P_{2P2A}^{cas} [Eq. (4.21)], which is induced by a pulse of light, we need to fix an interaction time t . It is shown in appendix B that the two atoms are submitted to the same energy flow at time $t = L/c$. One then obtains at this time and at exact resonance

$$P_1 \simeq P_{DR}^{11} \frac{\gamma_\beta^2}{\Delta^2} \simeq P_{2P2A}^{cas}. \quad (4.27)$$

We thus find the result that *a correlated-separable state like ρ_1 can induce the 2P2A transition as efficiently as the entangled cascade state*. This statement constitutes the main result of the present section.

Let us stress that ρ_1 , though not entangled, indeed has genuine quantum properties, being a mixture of single-photon states which are highly nonclassical. It exhibits strong correlations that we will study in more detail in Sec. [A.6](#).

A.5 Two-photon state produced by parametric down conversion

The two-photon state $|\Psi_{pdc}\rangle$ produced by non-degenerate spontaneous parametric down-conversion (SPDC), which has been under wide and in-depth investigation for many years is another important photon source in quantum optics. Because of its $\chi^{(2)}$ nonlinearity, a non-linear crystal submitted to a pulsed pump field of central frequency $\omega_\alpha + \omega_\beta$ and narrow bandwidth σ_α emits a signal field (central frequency ω_α) and an idler field (central frequency ω_β). Let σ_β be the frequency width of the phase matching curve. For the sake of computational simplicity we will use a Gaussian approximation for both the laser lineshape and the phase matching curve. The crystal generates in such a case an entangled state which is described by a wavepacket with a coefficient c_{kq}^{pdc1} [[Wang2006](#)] given by

$$c_{kq}^{pdc} = \mathcal{N} e^{-\frac{(\omega_{k\alpha} + \omega_{q\beta})^2}{2\sigma_\alpha^2} + i(\omega_{k\alpha} + \omega_{q\beta})t_0} \left(e^{-\frac{\omega_{k\alpha}^2 + \omega_{q\beta}^2}{2\sigma_\beta^2}} + i e^{-\frac{\omega_{k\beta}^2 + \omega_{q\alpha}^2}{2\sigma_\beta^2}} \right), \quad (4.29)$$

where \mathcal{N} is the normalized coefficient, satisfying

$$\left(\frac{L}{2\pi c} \right)^2 \mathcal{N}^2 \frac{2\pi\sigma_\alpha\sigma_\beta^2}{\sqrt{\sigma_\alpha^2 + 2\sigma_\beta^2}} = 1.$$

In expression [\(4.29\)](#), we have assumed that the pump laser pulse had a Gaussian temporal shape centered at time $t_0 \gg \sigma_\alpha^{-1} + \sigma_\beta^{-1}$ to provide most of the photons a chance to interact with the two detecting atoms. The factor i in the second term originates from a relative phase (depending on the birefringence) which is set to be $\pi/2$ for the sake of simplicity in our case.

Here we will also extend the double integral to the whole plane and find, when t is sufficient large [[GradshteynTISP](#)], the transition probability

$$P^{pdc} = \pi P_0 \frac{\sqrt{\sigma_\alpha^2 + 2\sigma_\beta^2}}{\sigma_\alpha\sigma_\beta^2} e^{-\frac{\delta^2}{\sigma_\alpha^2}} \left(e^{-\frac{\omega_{1\alpha}^2 + \omega_{2\beta}^2}{2\sigma_\beta^2}} + e^{-\frac{\omega_{2\alpha}^2 + \omega_{1\beta}^2}{2\sigma_\beta^2}} \right)^2. \quad (4.30)$$

¹In fact, when the coefficient is given as

$$c_{kq}^{pdc} \propto e^{-\frac{(\omega_{k\alpha} + \omega_{q\beta})^2}{2\sigma_\alpha^2} + i(\omega_{k\alpha} + \omega_{q\beta})t_0} e^{-\frac{\omega_{q\beta}^2}{2\sigma_\beta^2}}, \quad (4.28)$$

one will obtain similar results by following the same procedures. Because the expression of the coefficients of type II two-photon state in this chapter are complicated, it is not easy to find the analytical expressions to write the state in Schmidt decomposition form. In Chapter [6](#), we will consider the SPDC state given in this footnote rather than the one discussed in this section.

The two mixed two-photon states ($\rho_1^{pdc}, \rho_2^{pdc}$), defined in Sec. A.2 and pertaining to the pure SPDC two-photon state (4.29), are

$$\rho_1^{pdc} = \mathcal{N}^2 \sum_{kq} e^{-\frac{(\omega_{k\alpha} + \omega_{q\beta})^2}{\sigma_\alpha^2}} \left(e^{-\frac{\omega_{k\alpha}^2 + \omega_{q\beta}^2}{\sigma_\beta^2}} + e^{-\frac{\omega_{k\beta}^2 + \omega_{q\alpha}^2}{\sigma_\beta^2}} \right) |1_k, 1_q\rangle \langle 1_k, 1_q|, \quad (4.31)$$

$$\rho_2^{pdc} = \frac{\zeta \pi c^2}{L^2 \sigma_\beta^2} \left[\sum_k \left(e^{-\zeta \frac{\omega_{k\alpha}^2}{\sigma_\beta^2}} + e^{-\zeta \frac{\omega_{k\beta}^2}{\sigma_\beta^2}} \right) |1_k\rangle \langle 1_k| \right] \otimes \left[\sum_q \left(e^{-\zeta \frac{\omega_{q\alpha}^2}{\sigma_\beta^2}} + e^{-\zeta \frac{\omega_{q\beta}^2}{\sigma_\beta^2}} \right) |1_q\rangle \langle 1_q| \right], \quad (4.32)$$

where $\zeta = 1 + \sigma_\beta^2/(\sigma_\alpha^2 + \sigma_\beta^2)$. The first one corresponds to an SPDC process in which all the off-diagonal time-dependent terms in the density matrix are averaged to zero by random processes, while the second one characterizes a mixed state with two uncorrelated photons having the same spectrum than the initial SPDC state. When t is sufficient large, their corresponding transition probabilities read

$$P_1^{pdc} = \pi P_0 \frac{\sqrt{\sigma_\alpha^2 + 2\sigma_\beta^2}}{\sigma_\alpha \sigma_\beta^2} e^{-\frac{\delta^2}{\sigma_\alpha^2}} \left(e^{-\frac{\omega_{1\alpha}^2 + \omega_{2\beta}^2}{\sigma_\beta^2}} + e^{-\frac{\omega_{1\beta}^2 + \omega_{2\alpha}^2}{\sigma_\beta^2}} \right) \left(\frac{t}{L/c} \right)^2, \quad (4.33)$$

$$P_2^{pdc} = \pi \frac{P_0}{2} \frac{\zeta}{\sigma_\beta^2} \left(e^{-\zeta \frac{\omega_{1\alpha}^2}{\sigma_\beta^2}} + e^{-\zeta \frac{\omega_{1\beta}^2}{\sigma_\beta^2}} \right) \left(e^{-\zeta \frac{\omega_{2\alpha}^2}{\sigma_\beta^2}} + e^{-\zeta \frac{\omega_{2\beta}^2}{\sigma_\beta^2}} \right) \left(\frac{t}{L/c} \right)^2. \quad (4.34)$$

We will once again take $t = L/c$ to be able to compare in a fair way the pulsed and cw excitations through the rest of the following discussions.

A.5.1 Double resonance

Taking $\omega_\alpha = \omega_1, \omega_\beta = \omega_2$ and keeping the largest term, one finds the probability

$$P_{DR}^{pdc} = P_{1,DR}^{pdc} \simeq \pi P_0 \frac{\sqrt{\sigma_\alpha^2 + 2\sigma_\beta^2}}{\sigma_\alpha \sigma_\beta^2}, \quad (4.35)$$

which once again implies that entanglement is not active in enhancing the transition probability in the double resonance case.

One also finds $P_{DR}^{pdc} \simeq P_{DR}^{11}$ when $\sigma_\alpha = \gamma_\alpha, \sigma_\beta = \gamma_\beta$. In the following we will take this correspondences of spectral widths for comparisons. Henceforth, P_{DR}^{pdc} or P_{DR}^{11} will be regarded as a reference in the discussions related to SPDC two-photon states.

A.5.2 Two-photon two-atom resonance

The transition probability P^{pdc} has indeed a resonant character around $\delta = 0$ according to (4.30). At the exact 2P2A resonance, its value is assessed at

$$P_{2P2A}^{pdc} \simeq P_{DR}^{11} e^{-2\Delta^2/\sigma_\beta^2}, \quad (4.36)$$

which is much smaller than for the atom cascade state because the factor Δ^2/σ_β^2 enters now as an exponent in a Gaussian function and the detuning Δ is much greater than the spectral widths.

For the factorized, uncorrelated state ρ_2^{pdc} , the transition probability in this case reads

$$P_{2,2P2A}^{pdc} \simeq P_{DR}^{11} (1 + 2\sigma_\beta^2/\sigma_\alpha^2)^{-1/2} e^{-2\zeta\Delta^2/\sigma_\beta^2} \lesssim e^{-2\Delta^2/(\sigma_\alpha^2+\sigma_\beta^2)} P_{2P2A}^{pdc}. \quad (4.37)$$

Thus P_{2P2A}^{pdc} is much greater than the probability given by the factorized state because of the scale factor $e^{2\Delta^2/(\sigma_\alpha^2+\sigma_\beta^2)}$. So we obtain in the parametric down-conversion configuration the same conclusion as the one drawn in [Muthukrishnan2004] for the atomic cascade : the entangled state $|\Psi_{pdc}\rangle$ is much more efficient for inducing a 2P2A resonance than the factorized, uncorrelated state.

For the correlated-separable state ρ_1^{pdc} , the transition probability reads

$$P_{1,2P2A}^{pdc} \simeq P_{DR}^{11} e^{-2\Delta^2/\sigma_\beta^2} \simeq P_{2P2A}^{pdc}, \quad (4.38)$$

Thus, one has $P_{1,2P2A}^{pdc} \gg P_{2,2P2A}^{pdc}$. The same conclusion is found as in the cascade case : the correlated-separable state is as efficient as the entangled state to boost the 2P2A resonance. The fact that $P_{1,2P2A}^{pdc}$ is much larger than $P_{2,2P2A}^{pdc}$, and $P_{1,2P2A}^{pdc} \simeq P_{2P2A}^{pdc}$, once again shows that correlations, which are not necessarily related to entanglement, indeed play a crucial role in the efficiency of the excitation.

A.6 Enhancement of 2P2A resonance for more general classes of light states

We have so far studied interesting but specific states of light and showed an enhancement effect for some of them, entangled or correlated-separable. It would be interesting to consider now more general classes of light states.

A.6.1 Light pulses starting at a given time

Let us go back to the initial equations (4.3) and (4.2). They contain functions such as $[1 - \exp(i\omega_{1m}t)]/\omega_{1m}$. When $t \rightarrow \infty$, as explained in the Appendix A, even though this function does not act as a Dirac function when it is applied to integrations with any function, it indeed tends to $2i\pi\delta(\omega_1 - \omega_m)$ if applied to functions of ω_m that have a Fourier transform which is strictly zero for $t < 0$. Such will be the case here.

The initial two-photon light state $|\Psi\rangle$ is the pure state,

$$|\Psi\rangle = \sum_{kq} c_{kq} |1_k, 1_q\rangle, \quad (4.39)$$

that describes a "switched-on" light which is not vacuum only after time $t = 0$. One can then use the delta function approximation. The probability that the two atoms are found in the excited state at long times compared to the pulse duration is now

$$P \simeq \frac{P_0}{4} \frac{L^2}{c^2} |c(\omega_1, \omega_2) + c(\omega_2, \omega_1)|^2. \quad (4.40)$$

Mathematically, if $|c(\omega_1, \omega_2)| \sim |c(\omega_2, \omega_1)|$, this interference, which has been studied in the literature [Fei1997], may lead to strong variations according to the relative phase. According to the Cauchy-Schwartz inequality, one has

$$0 \leq P \leq 2 \left(\frac{P_0}{4} \frac{L^2}{c^2} (|c(\omega_1, \omega_2)|^2 + |c(\omega_2, \omega_1)|^2) \right). \quad (4.41)$$

However, physically speaking, only one component between $c(\omega_1, \omega_2)$ and $c(\omega_2, \omega_1)$ dominates in the expression (4.40). This is because we have assumed that the quantities $\omega_1, \omega_2, \omega_\alpha, \omega_\beta$ are sufficiently separated from each other but with a small 2P2A detuning $\delta \simeq 0$; as a result, ω_1 should be closer to one of the central frequencies of the fields than to the other ones. Under this condition, one has

$$P \simeq \frac{P_0}{4} \frac{L^2}{c^2} (|c(\omega_1, \omega_2)|^2 + |c(\omega_2, \omega_1)|^2). \quad (4.42)$$

The correlated and factorized states ρ_1, ρ_2 analogous to the initial state $|\Psi\rangle\langle\Psi|$, give rise to the following 2P2A transition probabilities :

$$P_1 = \frac{P_0}{4} t^2 (|c(\omega_1, \omega_2)|^2 + |c(\omega_2, \omega_1)|^2), \quad (4.43)$$

$$P_2 = \frac{P_0}{4} t^2 \sum_{mn} (|c(\omega_1, \omega_n)c(\omega_m, \omega_2)|^2 + |c(\omega_2, \omega_n)c(\omega_m, \omega_1)|^2). \quad (4.44)$$

The enhancement of the 2P2A transition probability is characterized by the quotient G_p between P and P_1 at $t = L/c$,

$$G_p = \frac{P}{P_1} \Big|_{t=L/c} = \frac{|c(\omega_1, \omega_2) + c(\omega_2, \omega_1)|^2}{|c(\omega_1, \omega_2)|^2 + |c(\omega_2, \omega_1)|^2}. \quad (4.45)$$

Thus, one finds $0 \leq G_p \leq 2$. The maximum value of 2 is achieved when $c(\omega_1, \omega_2) = c(\omega_2, \omega_1)$.

One has $G_p \simeq 1$ under the physical conditions we stated before. That is, the entangled and the correlated-separable state yield almost equal transition probabilities. This implies that the conclusion that we had drawn in the special previous cases is valid for a large class of two-photon states : correlated states are as efficient as entangled states in 2P2A co-excitation when they have delivered the same amount of energy to the two atoms.

Another important discriminability index is the ratio between the two transition rates P_1 and P_2 :

$$G_{12} = \frac{|c(\omega_1, \omega_2)|^2 + |c(\omega_2, \omega_1)|^2}{\sum_{mn} (|c(\omega_1, \omega_n)c(\omega_m, \omega_2)|^2 + |c(\omega_2, \omega_n)c(\omega_m, \omega_1)|^2)}. \quad (4.46)$$

The value of the enhancement factor G_{12} can be used as a witness for the correlation needed in such a problem.

Note in addition that, while P is sensitive to possible destructive interference effects between $c(\omega_1, \omega_2)$ and $c(\omega_2, \omega_1)$, P_1 is not. Therefore, the enhancement effect as indicated by G , and due to correlations not related to entanglement, turns out to be more "robust" than the one related to it.

A.6.2 Coherent states

So far we have only considered two-photon states of different shapes, which are all strongly non-classical objects, as they are produced by spontaneous emission or parametric fluorescence which are specifically quantum processes with no classical equivalent. But one can also envision superpositions of two-mode coherent states of the form

$$|\Psi_{coh}\rangle = \sum_{kq} c_{kq} |z : \omega_k, \alpha\rangle \otimes |z : \omega_q, \beta\rangle, \quad (4.47)$$

or its corresponding dephased state (which is classically mixed coherent state),

$$\rho_{CMC} = \sum_{kq} |c_{kq}|^2 |z : \omega_k, \alpha\rangle\langle z : \omega_k, \alpha| \otimes |z : \omega_q, \beta\rangle\langle z : \omega_q, \beta|, \quad (4.48)$$

where $|z : \omega, \mu\rangle$ is the coherent state $|z\rangle$ in the mode of frequency ω of μ subsystem, z being the same complex number for all modes.

By using the approximation $\langle \alpha|0\rangle \approx 0$ valid for $|\alpha| \gg 1$, one finds that the probability of 2P2A excitation is given by:

$$P_{coh}(t) = |z|^4 P(t), \quad (4.49)$$

where $P(t)$ is the probability (4.2) obtained for two-photon states. Apart from the energy scaling factor $|z|^4$, the conclusions of the previous paragraphs hold in the present case. These states are interesting for experimental demonstration, for example, the classically mixed coherent states can be produced by classical means. See further details in Appendix C and Appendix D.

A.7 Conclusion

In the previous sections of this chapter, we have shown that the transition probability by using specific entangled photon sources are approximately equal to the ones by using their corresponding completely dephased states which do not have any entanglement. We can now answer the question raised in the Introduction of this chapter about the role of entanglement in the two-photon excitation process considered in this chapter. We have shown that what is necessary for the enhancement of the transition probability is not quantum entanglement. However, the real physical origins are not clear at this stage. Candidates are correlations, temporal ones or frequency ones, or some other non-classical properties that related to the states that have been discussed. These are main topics in Chap. 6, in which we will find possible physical origin is not temporal correlation, but rather frequency anticorrelation, which can be due to the presence of entanglement in the state, but also to correlations that are not related to entanglement. This result brings new light to the problem of assessing the exact role of quantum correlations and quantum entanglement in physical processes.

As for any nonlinear process, such as two-photon absorption in a single atom [LoudonQTL], 2P2A transition probability can be modified by changing the quantum state of light, and therefore the enhancement effect that we have studied in this chapter is due to the partial optimization of the quantum state. The absorption of two photons by the two atoms occurs when each photon is resonant with the atomic transition of the atom that absorbs it. This has a greater probability to happen in frequency-anticorrelated photon states than in uncorrelated states.

We have not treated the important question of the full optimization of the quantum state with respect to the 2P2A probability maximization, given some constant spectral energy distributions. It will be addressed in the next section.

B The optimization of the transition probability with a given photon spectrum

In previous part of this chapter, we came to the conclusion that a state with entanglement does not induce a two-photon–two-atom transition more efficiently than its corresponding correlated-separable state. In fact, the two states have the same transition probability when both the photons are absorbed at the time L/c in asymptotical sense. For a general pure two-photon state $|\mathbb{II}\rangle = \sum_{mn} c(\omega_m, \omega_n) |1 : \omega_m, \alpha; 1 : \omega_n, \beta\rangle$, its photon spectrum follows

$$S(\omega) = \sum_m [|c(\omega_m, \omega)|^2 + |(\omega, \omega_m)|^2], \quad (4.50)$$

which is only determined by the diagonal elements of the corresponding density matrix. This fact suggests that the photon spectrum of a pure two-photon state is the same as the one of the corresponding diagonal density matrix. Furthermore, any convex linear combination of the two density matrices sharing such spectrum should also yield the same spectrum. In addition to these states, there should exist more photon states, pure or mixed, having the same photon spectrum.

The following question then arises: among all these states, which state is likely to attain the maximum (at least, in an asymptotical sense) transition probability, for a given photon spectrum and energy content ?

The photon spectrum is obtained by performing photon number measurement of specific mode on a given state. Here, we only consider the frequency modes since we are interested in the frequency-correlations. For a given two-photon state ρ , its spectrum is given by:

$$S(\omega) = \text{Tr}[\hat{a}^\dagger(\omega)\hat{a}(\omega)\rho]. \quad (4.51)$$

For any two-photon state, the total number of photons should be equal to 2 which is exactly the total photon number of the state,

$$\sum_\omega S(\omega) = 2. \quad (4.52)$$

On the contrary, for any given shape of the spectrum, it is always possible to find some density matrices according to the functional definition of a density matrix [PetzQITQS]. Thanks to it, we can discuss this optimization problem directly without the knowledge of a possible state in advance.

Assuming that we are in the same physical situation as in Sec. A, we know that the quantum interference from different excitation paths is not important. In other words, if the states, pure or mixed, have a same diagonal distribution, their corresponding transition probability are asymptotically equivalent. In view of this, we can restrict our discussion to diagonal density matrices.

B.1 Optimization of a diagonal density matrix

The density matrix of a generic bipartite two-photon state can be written as

$$\Lambda = \sum_{kq} p_{kq} |1 : \omega_k, 1 : \omega_q\rangle \langle 1 : \omega_k, 1 : \omega_q|, \quad \sum_{kq} p_{kq} = 1, \quad \forall k, q; p_{kq} \geq 0. \quad (4.53)$$

For a given spectrum $S(\omega)$, we denote all the diagonal density matrices that give rise to the spectrum, by

$$\Lambda = \sum_{kq} |c_{kq}|^2 |1 : \omega_k, 1 : \omega_q\rangle \langle 1 : \omega_k, 1 : \omega_q|, \quad \sum_{kq} |c_{kq}|^2 = 1. \quad (4.54)$$

Accordingly, the coefficients yield

$$S(\omega) = \sum_m (|c(\omega, \omega_m)|^2 + |c(\omega_m, \omega)|^2). \quad (4.55)$$

A first solution to this equation will be

$$|c(\omega_m, \omega_n)|^2 = \delta_{mn} S(\omega_m) / 2. \quad (4.56)$$

In principle, if the coefficients satisfy

$$|c(\omega_m, \omega_n)|^2 = \delta_{mn} S(\omega_m) / 2 + f(\omega_m - \omega_n), \quad (4.57)$$

where $f(x)$ is a bounded odd function,

$$f(\omega_m - \omega_n) = -f(\omega_n - \omega_m); \quad S(\omega_m) \geq 2|f(\omega_m - \omega_n)|, \quad (4.58)$$

then one can verify that they are also a solution of the spectrum function $S(\omega)$. From this second example, we also conclude that there should be infinitely many solutions of Eq. (4.55).

Now let's revisit the transition probability with a two photon source that has an arrival time or a two-photon state whose majority of photon pairs arrive at the two detecting atoms after time $t = 0$. Asymptotically, the transition probability follows

$$P = P_0(L/2c)^2 \lambda_P, \quad \lambda_P = |c(\omega_1, \omega_2)|^2 + |c(\omega_2, \omega_1)|^2, \quad (4.59)$$

at the time $t = L/c$ which is sufficiently large. This transition probability is determined by point-wise property of the diagonal elements ($|c(\omega_m, \omega_n)|^2; m, n \in \mathbb{N}$) of a state at frequencies $(\omega_m, \omega_n) = (\omega_1, \omega_2)$ and (ω_2, ω_1) .

Because of the positivity of the absolute value, according to Eq. (4.55), we come to the following two inequalities:

$$S(\omega_1) = \sum_m (|c(\omega_1, \omega_m)|^2 + |c(\omega_m, \omega_1)|^2) \geq |c(\omega_1, \omega_2)|^2 + |c(\omega_2, \omega_1)|^2 = \lambda_P, \quad (4.60a)$$

$$S(\omega_2) = \sum_m (|c(\omega_2, \omega_m)|^2 + |c(\omega_m, \omega_2)|^2) \geq |c(\omega_1, \omega_2)|^2 + |c(\omega_2, \omega_1)|^2 = \lambda_P. \quad (4.60b)$$

The two inequalities set an upper bound to the parameter λ_P ,

$$\lambda_P \leq B_\lambda, \quad B_\lambda \equiv \frac{S(\omega_1) + S(\omega_2)}{2} - \frac{|S(\omega_1) - S(\omega_2)|}{2}. \quad (4.61)$$

For sake of simplicity, let's assume $S(\omega_1) \geq S(\omega_2)$. The parameter λ_P saturates when

$$|c(\omega_1, \omega_2)|^2 \neq 0, |c(\omega_2, \omega_1)|^2 \neq 0; \quad (4.62a)$$

$$|c(\omega_2, \omega_m)|^2 = |c(\omega_m, \omega_2)|^2 = 0, \omega_m \neq \omega_2. \quad (4.62b)$$

Then all the density matrices whose diagonal elements satisfy the Eqs. (4.55,4.62) can attain the maximum transition probability at the value of

$$P = P_0(L/2c)^2 S(\omega_2). \quad (4.63)$$

One possible state to give rise such a transition probability is a diagonal mixed-state which reads

$$\begin{aligned} \Lambda = & \frac{S(\omega_2)}{2} [|1 : \omega_1, 1 : \omega_2\rangle\langle 1 : \omega_1, 1 : \omega_2| + |1 : \omega_2, 1 : \omega_1\rangle\langle 1 : \omega_2, 1 : \omega_1|] \\ & + \frac{S(\omega_1) - S(\omega_2)}{2} |1 : \omega_1, 1 : \omega_1\rangle\langle 1 : \omega_1, 1 : \omega_1| \\ & + \sum_{\omega_m \neq \omega_1, \omega_2} \frac{S(\omega_m)}{2} |1 : \omega_m, 1 : \omega_m\rangle\langle 1 : \omega_m, 1 : \omega_m|. \end{aligned} \quad (4.64)$$

B.2 An example

Now let's apply our results to a concrete example: the same spectrum as the one of the cascade two-photon state (1.32). It follows

$$S_{cas}(\omega) = \frac{2c}{L} \left[\frac{\gamma_\alpha + \gamma_\beta}{(\omega - \omega_\alpha)^2 + (\gamma_\alpha + \gamma_\beta)^2} + \frac{\gamma_\beta}{(\omega - \omega_\beta)^2 + \gamma_\beta^2} \right]. \quad (4.65)$$

This spectrum is a combination of two Lorentzian functions with central frequencies $\omega_\alpha, \omega_\beta$, and widths $\gamma_\alpha + \gamma_\beta, \gamma_\beta$ respectively. So at frequencies $\omega = \omega_1, \omega_2$, we have

$$S_{cas}(\omega_1) \simeq \frac{2c}{L} \left[\frac{\gamma_\alpha + \gamma_\beta}{(\omega_1 - \omega_\alpha)^2} + \frac{\gamma_\beta}{(\omega_1 - \omega_\beta)^2} \right], \quad (4.66)$$

$$S_{cas}(\omega_2) \simeq \frac{2c}{L} \left[\frac{\gamma_\alpha + \gamma_\beta}{(\omega_1 - \omega_\beta)^2} + \frac{\gamma_\beta}{(\omega_1 - \omega_\alpha)^2} \right]. \quad (4.67)$$

The difference between the two terms is

$$S_{cas}(\omega_2) - S_{cas}(\omega_1) \simeq \gamma_\alpha \frac{2c}{L} \left[\frac{1}{(\omega_1 - \omega_\beta)^2} - \frac{1}{(\omega_1 - \omega_\alpha)^2} \right] < 0. \quad (4.68)$$

When γ_α is much smaller than γ_β , the difference can be regarded as 0. The transition probability saturates at the value of

$$P_{opt} = P_0 \frac{L}{2c} \left[\frac{\gamma_\alpha + \gamma_\beta}{(\omega_1 - \omega_\beta)^2} + \frac{\gamma_\beta}{(\omega_1 - \omega_\alpha)^2} \right]. \quad (4.69)$$

One possible density matrix is

$$\begin{aligned} \Lambda = & \frac{S(\omega_2)}{2} |1 : \omega_1, 1 : \omega_2\rangle\langle 1 : \omega_1, 1 : \omega_2| + \frac{S(\omega_1)}{2} |1 : \omega_2, 1 : \omega_1\rangle\langle 1 : \omega_2, 1 : \omega_1| \\ & + \sum_{\omega_m \neq \omega_1, \omega_2} \frac{S(\omega_m)}{2} |1 : \omega_m, 1 : \omega_m\rangle\langle 1 : \omega_m, 1 : \omega_m|. \end{aligned} \quad (4.70)$$

At perfect 2P2A resonance, the single photon detuning between atomic frequency ω_1 and optical central frequency ω_α is the smallest one, and $\Delta = |\omega_1 - \omega_\alpha| \ll |\omega_1 - \omega_\beta|$, therefore the optimized transition probability can be approximate as

$$P_{opt} \simeq \frac{\gamma_\alpha L}{2c} P_{DR}^{11} \frac{\gamma_\beta^2}{\Delta^2} = \frac{\gamma_\alpha L}{2c} P_{2P2A}^{cas}. \quad (4.71)$$

This problematic increase should be perhaps corrected by taking into consideration the higher order perturbation results. The exact physical value of the optimal transition probability is therefore a problem which remains to be solved.

C A remark on the length of quantization box

In Sec. A, we have derived formulae of the 2P2A transition probability with a generic class of two-photon states. From these formulae, we know the dominant elements to give a net 2P2A transition probability are those around $|c(\omega_1, \omega_2)|^2 |1 : \omega_1, 1 : \omega_2\rangle \langle 1 : \omega_1, 1 : \omega_2|$, $|c(\omega_2, \omega_1)|^2 |1 : \omega_2, 1 : \omega_1\rangle \langle 1 : \omega_2, 1 : \omega_1|$. What's more, the two elements are the components which are on perfect resonance with the two atoms. The perfect resonance gives rise to a contribution of probability that has a t^4 dependence, and at time L/c , it will cause some problems in some situation, such as the optimized states we considered in Sec. B. Therefore, we should take into account the higher order terms in the perturbation series for the two components that are on perfect resonance.

For sake of simplicity, we only consider the element $|c(\omega_m, \omega_n)|^2 |1 : \omega_m, 1 : \omega_n\rangle \langle 1 : \omega_m, 1 : \omega_n|$. It is a two-monochromatic-mode state. The transition probability is dominated by a product of transition probabilities that represent the perfect resonances of both atoms. In this sense, we can only consider the transition probability of a two-level atom interacting with a monochromatic field.

C.1 Single-atom–single-photon transition probability

Here we only consider the transition probability of atom 1 by shining a monochromatic light of single-photon state $|1 : \omega_m\rangle$. According to [PuriMMQO], the evolution operator on light part is

$$\langle e_1 | \hat{U}_I(t) | g_1 \rangle = -2if_1 \frac{\sin[t\sqrt{(\omega_m - \omega_1)^2 + 4f_1^2 \hat{a}_m \hat{a}_m^\dagger}/2}]}{\sqrt{(\omega_m - \omega_1)^2 + 4f_1^2 \hat{a}_m \hat{a}_m^\dagger}} \hat{a}_m. \quad (4.72)$$

The transition probability amplitude is

$$A = \langle 0 | \langle e_1 | \hat{U}_I(t) | g_1 \rangle | 1 : \omega_m \rangle = -2if_1 \frac{\sin[t\sqrt{(\omega_m - \omega_1)^2/4 + f_1^2}]}{\sqrt{(\omega_m - \omega_1)^2 + 4f_1^2}}, \quad (4.73)$$

which is sinusoidal function with its period $4\pi/\sqrt{4f_1^2 + (\omega_m - \omega_1)^2}$. This result shows the quantum Rabi oscillation in which f_1 is named as single photon Rabi frequency[GAFIQO]. Accordingly, the transition probability is

$$P_1(\omega_m) = \frac{4f_1^2}{(\omega_m - \omega_1)^2 + 4f_1^2} \sin^2[t\sqrt{(\omega_m - \omega_1)^2/4 + f_1^2}]. \quad (4.74)$$

The single-photon–single-atom transition probability of atom 2 and photon state $|1 : \omega_n\rangle$ has the same expression by changing ω_m and subscript 1 to ω_n and subscript 2. Therefore the corresponding 2P2A transition probability is

$$P \simeq \sum_{mn} |c(\omega_m, \omega_n)|^2 [P_1(\omega_m)P_2(\omega_n) + P_1(\omega_n)P_2(\omega_m)]. \quad (4.75)$$

where we have neglected the cross terms which are not important in our model introduced in Sec. A. Only when $2\pi c/L \gg 2f_1$, one can use the following approximation at a long time t^2 ,

$$P_j(\omega_m) \simeq f_j^2 t^2 \text{sinc}^2[t(\omega_m - \omega_j)/2] \simeq 2\pi f_j^2 t \delta(\omega_m - \omega_j). \quad (4.76)$$

²In fact, time t is much smaller than f_1^{-1} . The approximation can be derived in more rigorous way by using the techniques from asymptote analysis.

By some direct calculations, once again, we derived the same expression in Eq.(4.43) which was previously obtained from second order perturbation theory. We then conclude: the expression of the 2P2A transition probability in Eq. (4.43) is correct only when $L \ll \pi c f_1^{-1}$.

If f_1 is expressed in terms of the spontaneous emission rate γ which is introduced in Sec. A, then the inequality can also be written as

$$L \ll \frac{\pi^2}{3} c \gamma^{-1} \simeq \pi c \gamma^{-1}. \quad (4.77)$$

Since γ^{-1} is regarded as the typical lifetime of both excited detecting atoms and the value of γ is small, the detecting atoms can stay in the excited state for very long time. The inequality claims that during the whole time interval, $0 \leq t \leq L/c$, if atom 1 is excited, its decay is extremely small and can be neglected. This condition is a necessity to our assumption on infinite life of excited detecting atoms in Sec. A.

Now the optimized transition probability of the spectra given by Eq. (4.66) and Eq. (4.67) satisfying

$$P_{opt} \ll \frac{\pi}{2} \frac{\gamma_\alpha}{\gamma} P_{2P2A}^{cas}. \quad (4.78)$$

If γ_α are several thousands times larger than γ , one takes c/L as hundreds times larger than γ , then the optimized transition probability can be several tens times larger than the previous one P_{2P2A}^{cas} .

At first sight, one may say that the magnitude of the term on the right side of inequality (4.78), can also exceed the value of 1 by decreasing γ . However, P_0 , therefore P_{2P2A}^{cas} , is proportional to γ^2 , to decrease the decay constant γ is to weaken the interaction strengths between atoms and light, hence is to give rise to a smaller transition probability.

D 2P2A transition with long-lived detecting atoms

In the previous sections, the two-photon–two-atom transition with different kinds of photons are discussed when both detecting atoms are of infinite lifetimes. We find that after sending a same amount energy of photon state of frequency-entangled kind or its corresponding separable-correlated kind, the transition probabilities for comparison at time L/c are the same.

For any real detecting atoms, their lifetime, though, can be sufficiently long, are still finite. It imposes an inequality to the length of quantization box along z direction for our detecting atoms in last section. However, in principle, the length of a quantization box can be sufficiently large at will, and an astronomical size of the box indeed breaks down our assumption on the infinite lifetime of the excited detecting atoms. In this case, one should consider the influences from atomic decays.

Owing to the atomic decays, a portion of the population in the excited state may transfer back to the ground state. Such de-excitations are more evident for the pulsed light sources in contrast to the cw sources. So when is the best time to observe the excitations for different light sources if the atoms are excited is a question we should answer.

For sake of simplicity, in this section, we will take the decay constants of both excited detecting atoms as a same γ . The magnitude of γ is small in comparison with the width of frequency bands of photon sources. We will also restrict ourselves in discussing the transition probability with photon sources which are related to the cascade two-photon state, rather than taking consideration of some generic states.

As introduced in Chap. 3 Sec. B, the response from the two detecting atoms is given by

$$\mathcal{K}_{mn} = f_1 f_2 \frac{[e^{i(\omega_1 - \omega_n)t} - e^{-\gamma t}][e^{i(\omega_2 - \omega_m)t} - e^{-\gamma t}]}{(\omega_n - \omega_1 + i\gamma)(\omega_m - \omega_2 + i\gamma)}, \quad (4.79)$$

when two photons at frequencies ω_m, ω_n are injected. This is a product of two single-photon–single-atom responses, from which we know that atom 1 (2) destroys a photon at frequency ω_m (ω_n). As we can see from Eq. (4.79), the kernel \mathcal{K}_{mn} varies slowly around $\omega_m = \omega_1$ and $\omega_n = \omega_2$. So if the photon source has a slowly varying distribution, then the transition probability is determined by its components of the photon distribution around $\omega_m = \omega_1$ and $\omega_n = \omega_2$, in the view of asymptotic analyses.

Consider the cascade two-photon source introduced in Chap. 1, its coefficients c_{mn} are of the following form,

$$c_{mn}^{cas} = \frac{2c}{L} \frac{\sqrt{\gamma_\alpha \gamma_\beta}}{[\omega_m + \omega_n - \omega_\alpha - \omega_\beta + i\gamma_\alpha][\omega_n - \omega_\beta + i\gamma_\beta]}. \quad (4.80)$$

It takes a time as long as γ_α^{-1} to generate two photons from a cascade three-level source atom, and the population in the co-excited state $|e\rangle = |e_1 e_2\rangle$ decays at a rate 2γ . If $\gamma_\alpha < 2\gamma$, then there won't be any noticeable population accumulation in the state $|e\rangle$ after both photons arrived at the detecting atoms. So we assume $\gamma_\alpha \geq 2\gamma$.

By applying the residue integral theorem, we obtain

$$P_{pure}^{cas} \simeq 4 \frac{L^2}{c^2} \gamma_\alpha \gamma_\beta \left(\frac{f_1 f_2}{\omega_\alpha - \omega_2} + \frac{f_1 f_2}{\omega_\alpha - \omega_1} \right)^2 \frac{e^{-2\gamma_\alpha t} + e^{-4\gamma t} - 2e^{-(\gamma_\alpha + 2\gamma)t} \cos(\delta t)}{\delta^2 + (2\gamma - \gamma_\alpha)^2}. \quad (4.81)$$

It shows a resonance of 2P2A transition at $\delta = 0$, and the width $\gamma_\alpha - 2\gamma$ of peak is modified by atomic decays. Though there exists frequency anti-correlations in the light source promising an enhancement of the 2P2A transition, because of the atomic decays, the population in the excited state vanishes gradually.

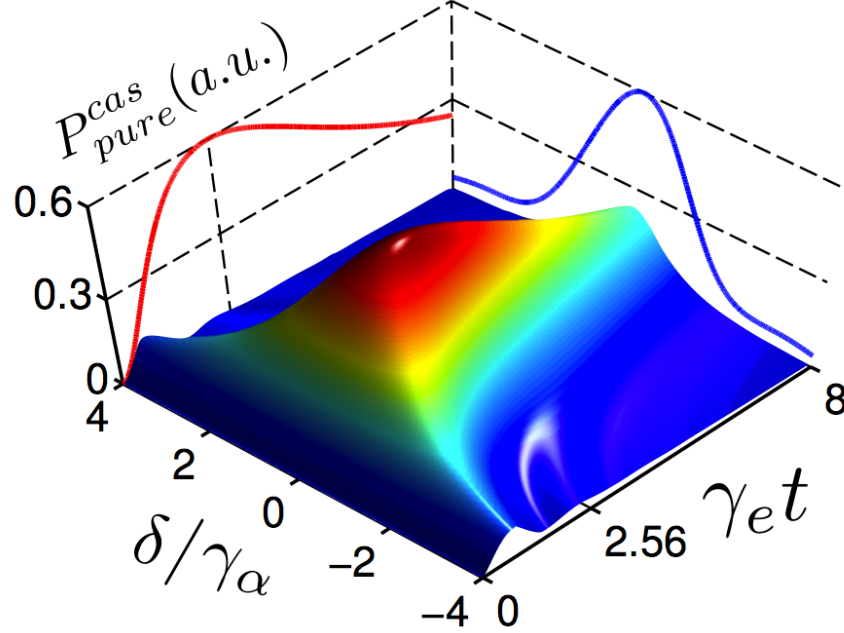


Figure 4.2: The plotting of the transition probability of two-photon–two-atom detuning δ and time t . Quantities here are reexpressed by taking the coherence length γ_α of the two-photon state as unit. In this plotting, we set $2\gamma/\gamma_\alpha = 0.1$.

When the 2P2A detuning $\delta = 0$, the transition probability is proportional to following function

$$p(t) \equiv \left(\frac{e^{-\gamma_\alpha t} - e^{-2\gamma t}}{2\gamma - \gamma_\alpha} \right)^2. \quad (4.82)$$

It attains its maximum of

$$p_{max} = \left(\frac{2\gamma}{\gamma_\alpha} \right)^{\frac{4\gamma}{\gamma_\alpha - 2\gamma}} \gamma_\alpha^{-2}, \quad \text{at } t = \frac{\ln \gamma_\alpha - \ln 2\gamma}{\gamma_\alpha - 2\gamma}. \quad (4.83)$$

The expression of p_{max} can be re-expressed as

$$p_{max} = \gamma_\alpha^{-2} \left[(1+x)^{\frac{1}{x}} \right]^{-2}, \quad x = \frac{\gamma_\alpha - 2\gamma}{2\gamma}. \quad (4.84)$$

The maximum p_{max} is a monotonic decreasing function of the parameter γ . This is because with smaller γ , the detecting atoms have longer time to accumulate the excitation population, so larger transition probability. It is of course reasonable if the detecting

atoms have no decays, then all the population transferred to the excited state can stay there forever.

If $\gamma_\alpha \gg \gamma$, then the maximum of

$$p_{max} = \left(\frac{2\gamma}{\gamma_\alpha} \right)^{\frac{4\gamma}{\gamma_\alpha - 2\gamma}} \gamma_\alpha^{-2} \rightarrow \gamma_\alpha^{-2}, \quad (4.85)$$

can be attained at a very long time

$$t \simeq \frac{\ln(\gamma_\alpha/2\gamma)}{\gamma_\alpha}. \quad (4.86)$$

At this time, the transition probability yields

$$P_{pure}^{cas} \simeq 4 \frac{L^2}{c^2} \gamma_\alpha^{-1} \gamma_\beta \left(\frac{f_1 f_2}{\omega_\alpha - \omega_2} + \frac{f_1 f_2}{\omega_\alpha - \omega_1} \right)^2. \quad (4.87)$$

This is a result we have known from Sec. A. When $\gamma_\alpha = 2\gamma$, the function $p(t)$ degrades to $(te^{-\gamma_\alpha t})^2$ which attains a maximum of $(e\gamma_\alpha)^{-2}$ where $e \simeq 2.71828$ at $t = 1/\gamma_\alpha$. This result is incorporated in the previous one since

$$\lim_{2\gamma \rightarrow \gamma_\alpha} \frac{\ln \gamma_\alpha - \ln 2\gamma}{\gamma_\alpha - 2\gamma} = \frac{1}{\gamma_\alpha}, \quad (4.88)$$

$$\lim_{2\gamma \rightarrow \gamma_\alpha} \left(\frac{2\gamma}{\gamma_\alpha} \right)^{\frac{2\gamma}{\gamma_\alpha - 2\gamma}} = \left[\lim_{2\gamma \rightarrow \gamma_\alpha} \left(1 + \frac{\gamma_\alpha - 2\gamma}{2\gamma} \right)^{\frac{2\gamma}{\gamma_\alpha - 2\gamma}} \right]^{-1} = e^{-1}. \quad (4.89)$$

This is the worst case for 2P2A transition for any $\gamma \leq \gamma_\alpha/2$. The results are in good agreement with our intuition because after passing a time γ_α^{-1} , the two photons are absorbed by the detecting atoms, then we see the pure decays leading to atomic de-excitations.

For the pure two-photon state, the best time for observation of 2P2A transition is at

$$t = \frac{\ln \gamma_\alpha - \ln 2\gamma}{\gamma_\alpha - 2\gamma}. \quad (4.90)$$

Since at time $t = L/c$, the photons are completely absorbed by the atoms, we can set

$$L = c \frac{\ln \gamma_\alpha - \ln 2\gamma}{\gamma_\alpha - 2\gamma}. \quad (4.91)$$

However, for the correlated-separable photon state, all the transition probability are accumulated separately from each local state, and the energy supply at any time is the same. It suggests: if the total atomic decay rate is less than the energy flux, then the transition probability should be monotonic increasing with time.

By applying the residue theorem to the mixed cascade two-photon source,

$$|c_{mn}^{cas}|^2 = \frac{4c^2}{L^2} \frac{\gamma_\alpha \gamma_\beta}{[(\omega_m + \omega_n - \omega_\alpha - \omega_\beta)^2 + \gamma_\alpha^2][(\omega_n - \omega_\beta)^2 + \gamma_\beta^2]},$$

we then have the transition probability

$$\begin{aligned} P \simeq & \frac{\gamma_\beta}{\gamma^2} \left(\frac{f_1 f_2}{\omega_\alpha - \omega_2} + \frac{f_1 f_2}{\omega_\alpha - \omega_1} \right)^2 \left[\frac{\gamma_\alpha + 2\gamma}{\delta^2 + (\gamma_\alpha + 2\gamma)^2} - 2 \frac{\gamma_\alpha e^{-2\gamma t}}{\delta^2 + \gamma_\alpha^2} + \frac{\gamma_\alpha - 2\gamma}{\delta^2 + (\gamma_\alpha - 2\gamma)^2} e^{-4\gamma t} \right. \\ & + 2\gamma e^{-(\gamma_\alpha + 2\gamma)t} \frac{[\delta^2 + \gamma^2 - (\gamma_\alpha - \gamma)^2] \cos \delta t + 2(\gamma_\alpha - \gamma)\delta \sin \delta t}{[\delta^2 + (\gamma_\alpha - 2\gamma)^2][\delta^2 + \gamma_\alpha^2]} \\ & \left. - 2\gamma e^{-(\gamma_\alpha + 2\gamma)t} \frac{[\delta^2 + \gamma^2 - (\gamma_\alpha + \gamma)^2] \cos \delta t + 2(\gamma_\alpha + \gamma)\delta \sin \delta t}{[\delta^2 + (\gamma_\alpha + 2\gamma)^2][\delta^2 + \gamma_\alpha^2]} \right]. \quad (4.92) \end{aligned}$$

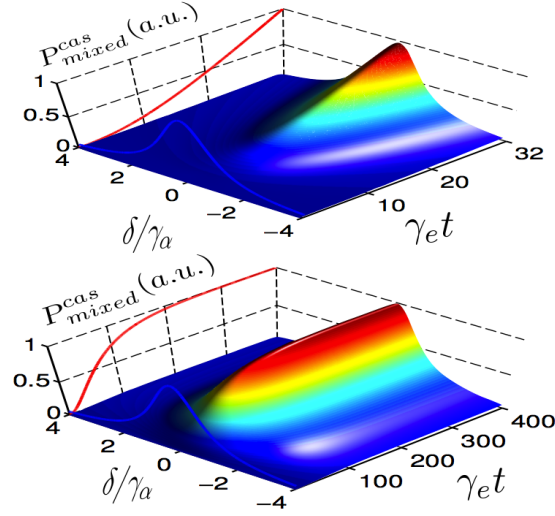


Figure 4.3: The plotting of the transition probability of two-photon–two-atom detuning δ and time t . Quantities here are reexpressed by taking the coherence length γ_α of the two-photon state as unit. In this plotting, we set $\gamma/\gamma_\alpha = 0.01$.

The time and the 2P2A dependence of the transition probability, see Figure 4.3.

There are some decaying terms which are originated in the imperfect correlations of the photon source (non vanishing value of the parameter γ_α) in the Eq. (4.92). These terms vanishes in a duration of γ_α^{-1} . We are more interested in the case $\gamma_\alpha \gg \gamma$. Under this condition, at a time much longer than γ_α^{-1} , the remaining long-lived terms dominate in the transition probability

$$P \simeq \frac{\gamma_\beta}{\gamma^2} \left(\frac{f_1 f_2}{\omega_\alpha - \omega_2} + \frac{f_1 f_2}{\omega_\alpha - \omega_1} \right)^2 \left[\frac{\gamma_\alpha + 2\gamma}{\delta^2 + (\gamma_\alpha + 2\gamma)^2} - 2 \frac{\gamma_\alpha e^{-2\gamma t}}{\delta^2 + \gamma_\alpha^2} + \frac{\gamma_\alpha - 2\gamma}{\delta^2 + (\gamma_\alpha - 2\gamma)^2} e^{-4\gamma t} \right]. \quad (4.93)$$

Though there are three Lorentzian peaks around $\delta = 0$, in view of $\gamma \ll \gamma_\alpha$, the modification of width in a graphic representation is not noticeable. By taking $\gamma_\alpha \pm 2\gamma$ as γ_α , the long-lived transition probability can be simplified as

$$P \simeq \frac{\gamma_\alpha \gamma_\beta}{\delta^2 + \gamma_\alpha^2} \left(\frac{f_1 f_2}{\omega_\alpha - \omega_2} + \frac{f_1 f_2}{\omega_\alpha - \omega_1} \right)^2 \left(\frac{1 - e^{-2\gamma t}}{\gamma} \right)^2. \quad (4.94)$$

This is a monotonic increasing function of time t and the transition probability attains its maximum at time $t = L/c$,

$$P_{max} \propto \left(\frac{1 - e^{-2\gamma L/c}}{\gamma} \right)^2. \quad (4.95)$$

And it is a monotonic decreasing function of γ . When $\gamma L/c$ is much greater than 1, we have

$$P_{max} \propto \frac{1}{\gamma^2}. \quad (4.96)$$

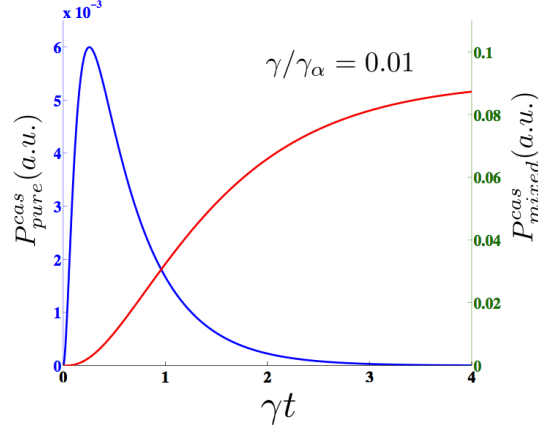


Figure 4.4: The relationship between the 2P2A transition probability and time t . Quantities here are reexpressed by taking the coherence length γ_α of the two-photon state as unit. The decay rate of the excited state of the detecting atoms are much smaller than the frequency coherence length γ_α (here we set $\gamma/\gamma_\alpha = 0.01$). From this figure, one find the transition probability for a pulsed light state can attains its peak value at a small time while the transition probability induced by a completely dephased state increase with time developments and gradually saturates at a constant value.

When $\delta = 0$ and $\gamma \ll \gamma_\alpha$, at time $t = L/c$ where L is optimized as $L = c(\ln \gamma_\alpha - \ln 2\gamma)/(\gamma_\alpha - 2\gamma)$, the ratio between the two transition probability At two-photon–two-atom resonance, the ratio between the two probability is

$$\frac{P}{P_{pure}^{cas}} \simeq 1. \quad (4.97)$$

This result shows, when the atomic decays are smaller than the correlation length γ_α , by choosing the proper L , we still have the same conclusion as in Sec. A. Also, this length L satisfies

$$L = c \frac{\ln \gamma_\alpha - \ln 2\gamma}{\gamma_\alpha - 2\gamma} \simeq c \frac{\ln(\gamma_\alpha/2\gamma)}{\gamma_\alpha} \ll \frac{c}{2\gamma} < \pi c \gamma^{-1}, \quad (4.98)$$

which do not contradict the restriction in Sec. C.

We then conclude that: in consideration of small atomic decays, the best length of quantization box along z direction is $L = c(\ln \gamma_\alpha - \ln 2\gamma)/(\gamma_\alpha - 2\gamma)$, and consequently, the best time for observation is at $t = (\ln \gamma_\alpha - \ln 2\gamma)/(\gamma_\alpha - 2\gamma)$.

E Conclusions

In this chapter, we have discussed the 2P2A transition with various two-photon states interacting two atoms that have no atomic interaction between each other and have infinite lifetimes. We further show the transition probability can be optimized from the original

states given in Sec. A. We also discussed the validation condition on the assumption of the infinite lifetime of excited detecting atoms, and found a restriction on the length of quantization box along the field propagation direction. Finally, we consider a more real condition that the detecting atoms have long-lived lifetimes rather than infinite ones and we found a optimized length of quantization box as well as a best time for observation of the 2P2A transition probability.

Two-photon–two-atom transition with an atomic interaction

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IN the last chapter, we have shown that the 2P2A transition probability can be greatly enhanced by frequency anti-correlated two-photon pairs. This is because the photon pairs share a correlation between themselves, and the atoms have a higher priority to destroy the more correlated photons. Intuitively, it is quite similar to the case when the atoms are interacting: in this case they share in some way the frequency information on the firstly absorbed photon and "know" which frequency of another photon to be taken.

In this chapter, we will focus our attention on the 2P2A transition process with an atomic interaction. In its first part, we will discuss the 2P2A transition by taking two photons from a monochromatic field, because it is an alternative situation as the one in previous chapter. In the second part, we will consider the case that the absorbed two photons came from a bipartite frequency anti-correlated two-photon source, since now one has both atomic interaction and frequency anti-correlations of photon source in the transition process and a competition or cooperation between the two physical origins can be discussed.

A Two-photon absorption with monochromatic sources

Consider a system with two two-level atoms that are coupled by an interaction as introduced in 3. By shining a monochromatic light of frequency ω on them, the two atoms can be excited by destroying two photons from the light. The leading effect of such a transition is described by a 2nd order optical process, a general expression for the transition probability of which has been derived in Chap. 3.

Let's denote the frequency of field by ω and the corresponding annihilation operator by \hat{a} , then the transition probability reads

$$P(t) \simeq \mathcal{R}(\omega, t) \text{Tr}(a^{\dagger 2} a^2 \rho_0), \quad \mathcal{R}(\omega, t) = |\mathcal{A}(\omega, t) + \mathcal{B}(\omega, t)|^2. \quad (5.1)$$

The function $\mathcal{R}(\omega, t)$ is independent of the photon number, hence the name transition profile.

In view of the monochromaticity, the above expression can be rewritten as

$$P(t) = \mathcal{R}(\omega, t) \langle \hat{N} \rangle^2 g^{(2)}(0). \quad (5.2)$$

For those light sources with given mean energy $\langle \hat{N} \rangle$, the one with stronger photon bunching will cause a higher transition probability. Super bunching and anti-bunching of photon spacing will give rise to two completely different excitations.

The explicit expressions of \mathcal{A} and \mathcal{B} , that come from the Eq.(3.30) in Chap. 3, follow

$$\mathcal{A}(\omega, t) = f_1 f_2 \frac{e^{-\gamma_+ t} - e^{i(\omega_+ - \omega)t}}{\omega - \omega_+ + i\gamma_+} \frac{e^{-\gamma_- t} - e^{i(\omega_- - \omega)t}}{\omega - \omega_- + i\gamma_-}, \quad (5.3)$$

$$\begin{aligned} \mathcal{B}(\omega, t) &= 2v f_1 f_2 \frac{1 + i(\gamma_- - \gamma_+)/(\omega_+ - \omega_-)}{(\omega - \omega_+ + i\gamma_+)(\omega - \omega_- + i\gamma_-)} \\ &\times \left[\frac{e^{i(\omega_+ + \omega_- - 2\omega)t} - e^{-2\gamma t}}{2\omega - \omega_+ - \omega_- + 2i\gamma} + \frac{e^{i(\omega_+ - \omega)t - \gamma_- t} - e^{i(\omega_- - \omega)t - \gamma_+ t}}{\omega_+ - \omega_- + i(\gamma_- - \gamma_+)} \right], \end{aligned} \quad (5.4)$$

where $2\gamma = \gamma_+ + \gamma_-$. From both expressions, one knows: 1. \mathcal{A} represents a process that the states $|\pm\rangle$ absorb photons independently, leading to a product of two independent responses; 2. \mathcal{B} stands for a cooperative absorption process, since without interactions ($v = 0$), it does not appear. Moreover, the first term in square bracket implies a two photon resonance at $\omega = (\omega_+ + \omega_-)/2$. We denote this value by ω_0 .

From Eqs.(5.2-5.4), we know that the statistics of photons can only change the transition strength, and the shape of a transition profile $\mathcal{R}(\omega, t)$ is only determined by frequency relations at a very-long time. In other words, such transition profile is independent of whichever photon sources, therefore is unique.

A.1 Transition profile \mathcal{R}

We have seen in chapter 3 that the interaction between two atoms gives rise to a new set of stationary states, of the form

$$|+\rangle = a(v)|e_1 g_2\rangle + b(v)|g_1 e_2\rangle, \quad (5.5a)$$

$$|-\rangle = a(v)|g_1 e_2\rangle - b(v)|e_1 g_2\rangle, \quad (5.5b)$$

$$a^2(v) + b^2(v) = 1, \quad (5.5c)$$

where $|e_1g_2\rangle$ and $|g_1e_2\rangle$ are the excited states of the two two-level atoms while there is no atomic interaction. The coefficients $a(v), b(v)$ are dependent on the interaction strength v , therefore the newly defined states in Eqs.(5.5) are mixed from $|e_1g_2\rangle$ and $|g_1e_2\rangle$ by v . When this parameter is equal to 0, the state mixing disappears and states $|\pm\rangle$ turn out to be $|e_1g_2\rangle$ and $|g_1e_2\rangle$. It is also worth noting, the atomic interaction also gives rise to cooperative emissions with a factor γ_{12} . According to [Hettich2002], the decay rates of $|\pm\rangle$ yield

$$\gamma_+ = a^2\gamma_1 + b^2\gamma_2 + 2ab\gamma_{12}, \quad (5.6a)$$

$$\gamma_- = a^2\gamma_2 + b^2\gamma_1 - 2ab\gamma_{12}, \quad (5.6b)$$

where γ_1, γ_2 are the dephasing rates of $|e_1g_2\rangle$ and $|g_1e_2\rangle$ respectively in absence of interaction. The contribution of cooperation term $2ab\gamma_{12}$ indeed brings about the super-radiation and sub-radiation phenomena to state $|+\rangle$ and $|-\rangle$ respectively. It is also worth mentioning that the value of γ is fixed because $\gamma = (\gamma_+ + \gamma_-)/2 = (\gamma_1 + \gamma_2)/2$. We then conclude that the super-radiation and sub-radiation phenomena are stucked together when both two-level atoms are excited.

A.1.1 A general response shape at long time

At a sufficiently large time t which is much greater than both γ_{\pm}^{-1} , the time dependence in transition profile becomes very weak, thus $\mathcal{R}(t) \simeq \mathcal{R}_{\infty}$,

$$\mathcal{R}_{\infty} = \frac{f_1^2 f_2^2}{[(\omega - \omega_+)^2 + \gamma_+^2][(\omega - \omega_-)^2 + \gamma_-^2]} \left| 1 + v \frac{1 - i(\gamma_+ - \gamma_-)/(\omega_+ - \omega_-)}{\omega - \omega_0 + i\gamma} \right|^2, \quad (5.7)$$

where we have used the fact that $\omega_+ + \omega_- = \omega_1 + \omega_2$ and the sum is defined as $2\omega_0$. In between the sign of absolute value, $\gamma_+ - \gamma_- = 4ab\gamma_{12}$ is a factor that represents the cooperation emission while $|\omega_+ - \omega_-|$ is the energy difference which is much greater than $\gamma_+ - \gamma_-$. As a result, the transition profile can be further simplified as

$$\mathcal{R}_{\infty} = \frac{f_1^2 f_2^2}{[(\omega - \omega_+)^2 + \gamma_+^2][(\omega - \omega_-)^2 + \gamma_-^2]} \left| 1 + \frac{v}{\omega - \omega_0 + i\gamma} \right|^2. \quad (5.8)$$

In this expression, one finds the new structure induced by two-atom cooperative absorption is strongly dependent on the parameter v . If v is much larger than γ , which is half of the dephasing rate of the state $|e_1e_2\rangle$, then a notable peak at $\omega = \omega_0$ should be observed.

With an increase of v , it is possible to make the super-radiation and sub-radiation phenomena more noticeable, because the width of the two peaks are strongly dependent on the interaction parameter v , see the relationship of the width difference between two peaks around $\omega = \omega_{\pm}$ on v in the bottom right graph in Figure 5.1. We also notice that the width $\gamma = (\gamma_1 + \gamma_2)/2$ of the peak in the middle is fixed whichever the value of the parameter v . What's more, the peak in the center is asymmetric because of the constant term 1 in the absolute sign. It is an important factor, since the transition profile will be greatly modified by increasing the interaction strength v . As we know, the energies $\hbar\omega_{\pm}$ are shifted from $\hbar\omega_1, \hbar\omega_2$ by an equal amount in opposite directions, so when $\gamma_1 = \gamma_2$, there is no reason to show for monochromatic two-photons whose frequency is less than $\omega_0 = (\omega_1 + \omega_2)/2$ should have a higher average transition probability than the one of a

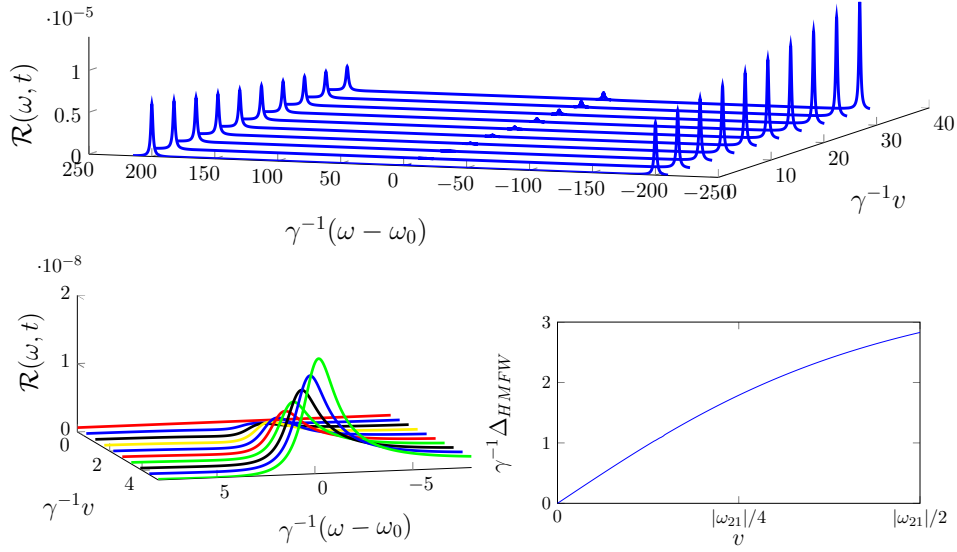


Figure 5.1: Graphs giving the properties of transition profiles at a sufficiently long time. In all the plots, $\omega_0 = (\omega_+ + \omega_-)/2$ is a fixed quantity and $\mathcal{R}(t)$ is in a unit of $f_1^2 f_2^2$. When there is no atomic interaction between atoms, there are two peaks at $\omega = \omega_1$ and $\omega = \omega_2$ which are ω_{\pm} at $v = 0$ respectively. When there is an interaction between atoms, a peak in the window center of the transition profile $\mathcal{R}(t)$ appears, see the lines in the upper graph. Though the peak value is small, with a increase of interaction strength v , asymptotically, it grows as a parabolic function. Because of the constant term 1 in the sign of absolute value, such a transition profile is asymmetric, see the lines in the bottom left graph. Owing to the cooperative absorption, the (HMF) width of the peaks around $\omega = \omega_-$ is small in contrast with the width around $\omega = \omega_+$. The difference between the two width is positive and monotonic increasing function of v , see the bottom right graph. In it, we abbreviate $\omega_2 - \omega_1$ as ω_{21} .

pair whose frequency is on the other side, and vice versa. Then we regard the integral of response function \mathcal{R}_{∞} over the two parts are equal. However, since the peak in the center is asymmetric about $\omega = \omega_0$ and a sharply slope appears on the side of $\omega = \omega_-$, see the bottom left graph in Figure 5.1, it is quite natural that the peak value around $\omega = \omega_-$ should be larger than the one around $\omega = \omega_+$ (see the upper graph in Figure 5.1). Such a phenomenon will be more remarkable with a larger v .

According to Eq.(5.8), the response function \mathcal{R}_{∞} attains two local maxima around $\omega = \omega_{\pm}$. In principle, $\omega = \omega_{\pm}$ are not the element where the function attains extrema, however, from the expression we know that the maxima are quite far separated and $\omega = \omega_{\pm}, \omega = \omega_0$ are indeed good approximations to the solutions of local maxima where the Half-Maximum-Full-Widths (HMF) of corresponding peaks are assessed at $2\gamma_{\pm}, 2\gamma$. The ratio between the values of \mathcal{R}_{∞} at ω_{\pm} is

$$\frac{\mathcal{R}_{\infty}(\omega = \omega_+)}{\mathcal{R}_{\infty}(\omega = \omega_-)} \simeq \frac{\gamma_-^2 (\omega_+ - \omega_0 + v)^2 + \gamma^2}{\gamma_+^2 (\omega_+ - \omega_0 - v)^2 + \gamma^2}. \quad (5.9)$$

When $\gamma_1 = \gamma_2 = \gamma$, this ratio can be re-expressed as

$$\frac{\mathcal{R}_{\infty}(\omega = \omega_+)}{\mathcal{R}_{\infty}(\omega = \omega_-)} \simeq \omega_{21}^4 \frac{1 + 4\gamma^2 / [(\omega_{21}^2 + 4v^2)^{1/2} + 2v]^2}{\omega_{21}^4 + 4\gamma^2 [(\omega_{21}^2 + 4v^2)^{1/2} + 2v]^2}, \quad (5.10)$$

from which one finds the ratio is a monotonic decreasing function of v ($v \geq 0$). When

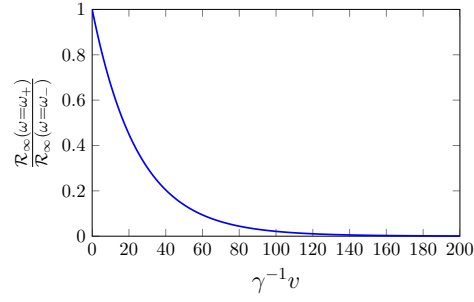


Figure 5.2: Graphs of the relation between the ratio of two local maxima and the interaction strength v . In this plotting, ω_{21} is set as 400γ

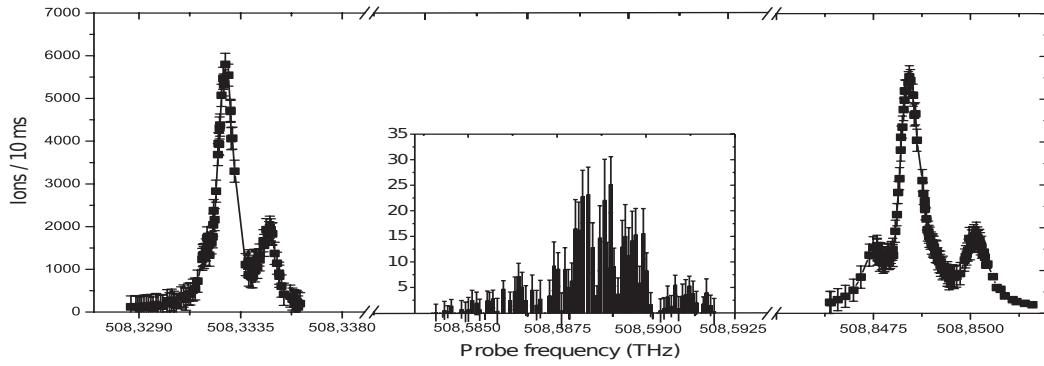


Figure 5.3: Graph of experimental data from [P-Penãfiel2012] in cold Na atom system. In their experiments, they observed an evident peak in the window center. The shoulders in the peaks on both sides are coming from the interference between atomic states.

$v = 0$, this ratio is exactly equal to 1, and the two peaks have an equal height. So, when there is an atomic interaction, the peak around $\omega = \omega_-$ is always higher than the one around $\omega = \omega_+$, see Figure 5.2.

In [Hettich2002], it was shown in experiment that the width of the peak on one side was always larger than the one on the other side. In [P-Penãfiel2012], the asymmetric peak in center was clearly shown in their data from experiments on two sodium atoms. See Figure 5.3. In both experiments, because the atomic interaction strength is weak, there is no evident height difference between the two peaks around $\omega = \omega_{\pm}$.

A.1.2 Strong decoherence-suppression case

With the growth of v , the decay rate γ_- becomes small, and is possible even to be vanishing when $\gamma_1 = \gamma_2$. In this case, when $t \gg \gamma_+^{-1}$ and $t \lesssim \gamma_-^{-1}$, the response function $\mathcal{R}(t)$ turns out to be

$$\begin{aligned} \mathcal{R}(t) = & \frac{f_1^2 f_2^2}{[(\omega - \omega_+)^2 + \gamma_+^2][(\omega - \omega_-)^2 + \gamma_-^2]} \\ & \times \left| 1 - \frac{v}{\omega - \omega_0 + i\gamma} - \frac{\omega_+ - \omega_- + 2v}{\omega_+ - \omega_-} e^{-\gamma_- t - i(\omega_- - \omega)t} \right|^2. \end{aligned} \quad (5.11)$$

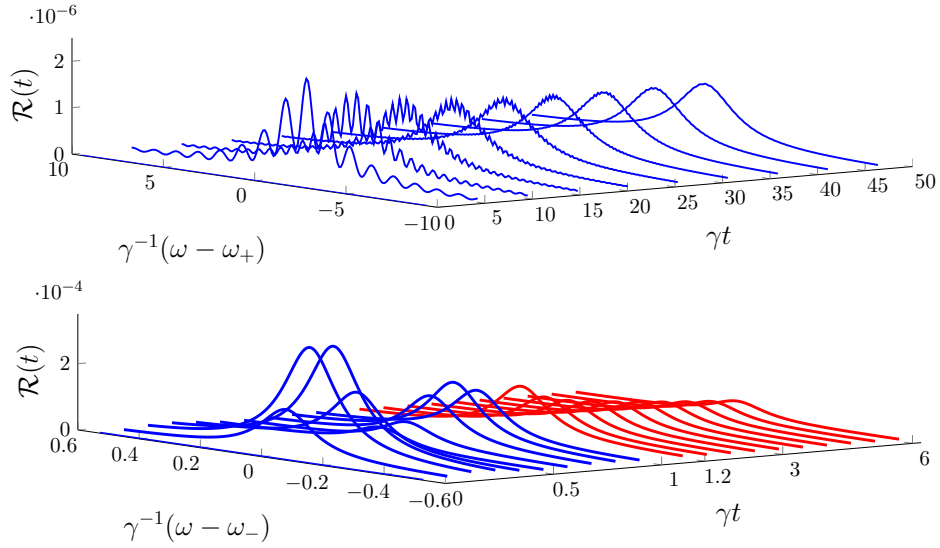


Figure 5.4: Graphs of transition profile. In both graphs, we take $v = 2|\omega_2 - \omega_1|$. The width of transition profiles in upper graph is $\gamma_+ = 1.9\gamma$ and the one in bottom graph is $\gamma_- = 0.1\gamma$. With the increase of time, the shapes of lines $\mathcal{R}(t)$ in upper graph become fixed while those lines in bottom graph become very attenuate. However, its value is still much larger.

As we can see from this expression that peak around $\omega = \omega_0$ is almost unchanged because $v \sim \omega_+ - \omega_- \gg \gamma$, the dominant term in the sign of absolute value is the one of Lorentzian type. We conclude that the shape of the induced peak around center is the same as \mathcal{R}_∞ even when γ_- becomes quite small where v is comparative to $\omega_+ - \omega_-$.

The possible changes in the transition profile will be the global profile and the most noticeable ones are the profile around $\omega = \omega_\pm$. See Figure 5.4.

A.2 Various photon states

The perturbation theory gives rise to Eq.(5.2), which implies that the transition probability is described by a universal response part and $g^{(2)}$ function of light field at a vanishing delay time, and this factor is indeed introduced as

$$g^{(2)} = \langle \hat{a}^{\dagger 2} \hat{a}^2 \rangle / \langle \hat{a}^\dagger \hat{a} \rangle^2. \quad (5.12)$$

When $g^{(2)} = 1$, the photon spacing is equidistant, hence the statistics yield Poissonian distribution. If $g^{(2)} > 1$, the photon statistics is super-Poissonian as it is bunched photon spacing while if $g^{(2)} < 1$, the photon statistics is sub-Poissonian as its photon spacing is anti-bunched. The photon bunching effect is well known from the wave statistics, but the anti-bunching effect is a genuine quantumness that is strongly dependent on the not commuting relation between operator \hat{a}, \hat{a}^\dagger . For example, for any given pure number state $|N\rangle$, the $g^{(2)}$ function reads $g^{(2)} = 1 - N^{-1}$ which claims that any Fock state yield photon anti-bunching. So such a state $|N\rangle$ is highly quantum. We can construct a diagonal state as following

$$\rho = (1 - \epsilon)|N\rangle\langle N| + \epsilon \sum_{m \neq N} p_m |m\rangle\langle m|, \quad \sum_{m \neq N} p_m = 1, \quad (5.13)$$

whose $g^{(2)}$ function follows

$$g^{(2)} = \frac{(1 - \epsilon)N^2 + \epsilon \sum_{m \neq N} p_m m^2}{[(1 - \epsilon)N + \epsilon \sum_{m \neq N} m p_m]^2} - \frac{1}{(1 - \epsilon)N + \epsilon \sum_{m \neq N} m p_m}. \quad (5.14)$$

When the parameter ϵ is sufficiently small, $g^{(2)} \simeq (N - 1)/N$. We conclude that the diagonal density matrix is highly quantum. This is quite surprising since intuitively speaking, a diagonal density matrix should be most classical one, because it does not possess any quantum interferences.

For any given density matrix ρ , we have

$$\langle \hat{N}^n \rangle = \text{Tr}[\rho \hat{N}^n] = \sum_m \langle m | \rho | m \rangle m^n. \quad (5.15)$$

This equation suggests that the photon statistics only relies on the diagonal elements as the non-diagonal elements have nothing to do with it. Therefore, for any pure state $|\Psi\rangle = \sum_m c_m |m\rangle$, its photon statistics is exactly equal to a diagonal density matrix,

$$\rho = \sum_m |c_m|^2 |m\rangle \langle m|. \quad (5.16)$$

This property leads to a conclusion that the corresponding diagonal density matrix can attains a same transition probability as the pure state and as any other states that imply a same photon spectra.

According to Eq.(5.2), if the photon state is a monochromatic coherent state $|\alpha, \omega_0\rangle$ which has a Poissonian photon statistic, then the transition probability is $P_{|\alpha\rangle}(t) = |\alpha|^2 \mathcal{R}(\omega, t)$, and if it is a Fock state $|n, \omega_0\rangle$ whose photon spacing is anti-bunched, then the probability is exactly equal to

$$P_{|n\rangle}(t) = n(n - 1) \mathcal{R}(\omega, t). \quad (5.17)$$

Let's consider a thermal state $\rho = (1 - \lambda) \sum_{n \geq 0} \lambda^n |n\rangle \langle n|$, $0 \neq \lambda < 1$ as the photon source. Such a parameter is sometimes named as Boltzmann factor, because the parameter λ is related to the temperature of the atoms in thermal motions, and it is larger with higher temperature in thermal equilibrium statistics. The corresponding value of correlation function is $g^{(2)} = 2$ hence the photon spacing is bunched. Its corresponding transition probability is

$$P_{thermal}(t) = 2 \frac{\lambda^2}{(1 - \lambda)^2} \mathcal{R}(t). \quad (5.18)$$

Of course, there are other states with high photon bunching, for example, in a semiconductor, photon extra-bunching ($g^{(2)} = 3$) was found in a twin beams [Boitier2011].

A.2.1 A two-photon state whose coefficients are related to Riemann $\zeta(s)$ function

Let's consider here a special mixed state, defined as

$$\Lambda = \left(1 - \bar{n} \frac{\zeta(s+2)}{\zeta(s+1)}\right) |0\rangle \langle 0| + \sum_{n \geq 1} \frac{\bar{n}}{\zeta(s+1)} n^{-(2+s)} |n\rangle \langle n|, \quad (5.19)$$

where $\zeta(x) = \sum_m 1/m^x$ is the Riemann function¹ and \bar{n} is mean photon number which is quite small because of the positivity of the coefficient $1 - \bar{n}\zeta(s+2)/\zeta(s+1)$, or $\bar{n} < \zeta(s+1)/\zeta(s+2)$. One can verify that this density matrix is normalized to 1. By direct calculations, this value

$$g^{(2)}(s, \bar{n}) = \frac{1}{\bar{n}} \left(\frac{\zeta(s)}{\zeta(s+1)} - 1 \right), \quad (5.20)$$

from which we see clearly that when the mean photon number \bar{n} is fixed, and $s \rightarrow 1$, the value of $g^{(2)}$ can be very large. This is because the photon spectrum is really flat and the part of many-photon mixture is indeed the key part in which the photon number fluctuations are really large. The transition probability is then

$$P_\Lambda(t) = \bar{n} \left(\frac{\zeta(s)}{\zeta(s+1)} - 1 \right) \mathcal{R}(t). \quad (5.21)$$

As we know, the key ingredient to enhance the transition probability is the photon number fluctuations from large photon number components, even though the mean photon number of this state is smaller than $\zeta(s+1)/\zeta(s+2)$, it is possible to enlarge the mean photon number by consider similar density matrices, such as

$$\Lambda' = [\zeta(s+2) - \sum_{n=1}^{N-1} n^{-(2+s)}]^{-1} \sum_{n \geq N} n^{-(2+s)} |n\rangle \langle n|. \quad (5.22)$$

These states keep the flatness of photon spectrum and the essential photon fluctuations. One will find a $\zeta(s)$ term in the expression of $g^{(2)}$, which is the dominant term when s goes to 0. That is, one recovers a large transition probability in this new state.

Because all these density matrices are diagonal, they are much easier to produce than their corresponding pure states.

¹This is an important function in both Mathematics and Physics. For example, $\zeta(3/2) \simeq 2.612$ is employed in calculating the critical temperature of a Bose-Einstein condensate in a box, and $\zeta(4) = \pi^4/90$ is used to derive the Stefan-Boltzmann law in the study of blackbody radiations. Other common numbers are $\zeta(2) = \pi^2/6$, $\zeta(3) \simeq 1.202$. The Riemann function diverges at $x = 1$, $\zeta(1) = \infty$.

B Two-photon absorption with a bipartite frequency anti-correlated source

In Chapter 3, we have derived a general expression of transition probability according to Eqs. (3.29,3.31), at any time t . Here we consider the influence of atomic interaction in the process of two-photon–two-atom transition.

Let's consider the cascade two-photon state defined in Eq. (1.32), the transition probability follows $P(t) = |\sum_{mn} \mathcal{K}_{mn}(c_{mn} + c_{nm})|^2$, where c_{mn} is the coefficient of the photon pair in which one photon of frequency ω_m is in the first part and a second photon of frequency ω_n is in the other part in the two-photon state. We assume that in the cascade two-photon state, $\gamma_\alpha \ll \gamma_\beta$, so the decaying terms related to $e^{-\gamma_\beta t}$ vanish at a short time $t \sim \gamma_\beta^{-1}$, then the remaining terms are those long-lived ones.

$$P(t) \simeq 4\gamma_\alpha\gamma_\beta \frac{L^2}{c^2} \frac{|e^{i\delta t - \gamma_\alpha t} - e^{-2\gamma t}|^2}{\delta^2 + (2\gamma - \gamma_\alpha)^2} \times \left| \frac{\mathbf{A}^T \mathbf{f} \mathbf{C}^T \mathbf{f}}{\omega_- - \omega_\beta + i(\gamma_\beta + \gamma_+ - \gamma_\alpha)} + \frac{\mathbf{B}^T \mathbf{f} \mathbf{D}^T \mathbf{f}}{\omega_+ - \omega_\beta + i(\gamma_\beta + \gamma_- - \gamma_\alpha)} \right|^2. \quad (5.23)$$

where δ is the two-photon–two-atom detuning $\omega_\alpha + \omega_\beta - \omega_1 - \omega_2$. From Eq.(5.23), we observe a 2P2A resonance at $\delta = 0$ whose width is equal to $2|\gamma_\alpha - 2\gamma|$. The 2P2A transition profile at $\delta = 0$ is the same as the one in Eq.(4.81) in Chapter 4. This is universally true in our model, because the atomic interaction yields the equalities $\omega_+ + \omega_- = \omega_1 + \omega_2$, $\gamma_+ + \gamma_- = \gamma_1 + \gamma_2$. Intuitively, we set $\tilde{\omega}_\pm = \omega_\pm - i\gamma_\pm$, we conclude that for any photon sources, the profile of induced two-photon–two-atom peak is independent of the interaction strength v and this parameter can only enhance or reduce the transition probability.

For simplicity, let's take $f_1 = f_2 = f$ and $|\gamma_\alpha - \gamma_\pm| \gg \gamma_\beta$, then we neglect $\gamma_\alpha - \gamma_\pm$ in the denominator and get

$$P(t) \simeq 4f^4 \gamma_\alpha \gamma_\beta \frac{L^2}{c^2} \frac{|e^{i\delta t - \gamma_\alpha t} - e^{-2\gamma t}|^2}{\delta^2 + (2\gamma - \gamma_\alpha)^2} \frac{(\omega_\alpha - \omega_\beta - \delta + 2v)^2 + 4\gamma_\beta^2}{[\gamma_\beta^2 + \omega_\alpha \omega_\beta + v^2 - \omega_1 \omega_2]^2 + \gamma_\beta^2 (\omega_\alpha - \omega_\beta)^2}. \quad (5.24)$$

If we take $x = \omega_\alpha - (\omega_1 + \omega_2)/2$, then the shape

$$\frac{(\omega_\alpha - \omega_\beta - \delta + 2v)^2 + 4\gamma_\beta^2}{[\gamma_\beta^2 + \omega_\alpha \omega_\beta + v^2 - \omega_1 \omega_2]^2 + \gamma_\beta^2 (\omega_\alpha - \omega_\beta)^2}$$

can be expressed as

$$F(x, v) = 4 \frac{(x - v)^2 + \gamma_\beta^2}{(\gamma_\beta^2 + \omega_{21}^2/4 - x^2 + v^2)^2 + 4\gamma_\beta^2 x^2}. \quad (5.25)$$

If $x^2 - \gamma_\beta^2 - \omega_{21}^2/4 > 0$, then for fixed x , the transition probability can be greatly enhanced at $v^2 = x^2 - \gamma_\beta^2 - \omega_{21}^2/4$. It happens only when $\max\{\omega_\alpha, \omega_\beta\} > \max\{\omega_1, \omega_2\}$. The emergence of enhancement is originated in the fact that ω_+ eventually attains its value which is equal to ω_α or ω_β by increasing v . That is, the mean photons are resonant with the new eigenstates of $|\pm\rangle$.

In a degenerate situation, $\omega_\alpha = \omega_\beta$, then we find that $\delta = 2x$ and in this case, we have

$$F(\delta/2, v) = 16 \frac{(\delta - 2v)^2 + 4\gamma_\beta^2}{(4\gamma_\beta^2 + \omega_{21}^2 - \delta^2 + 4v^2)^2 + 16\gamma_\beta^2 \delta^2}. \quad (5.26)$$

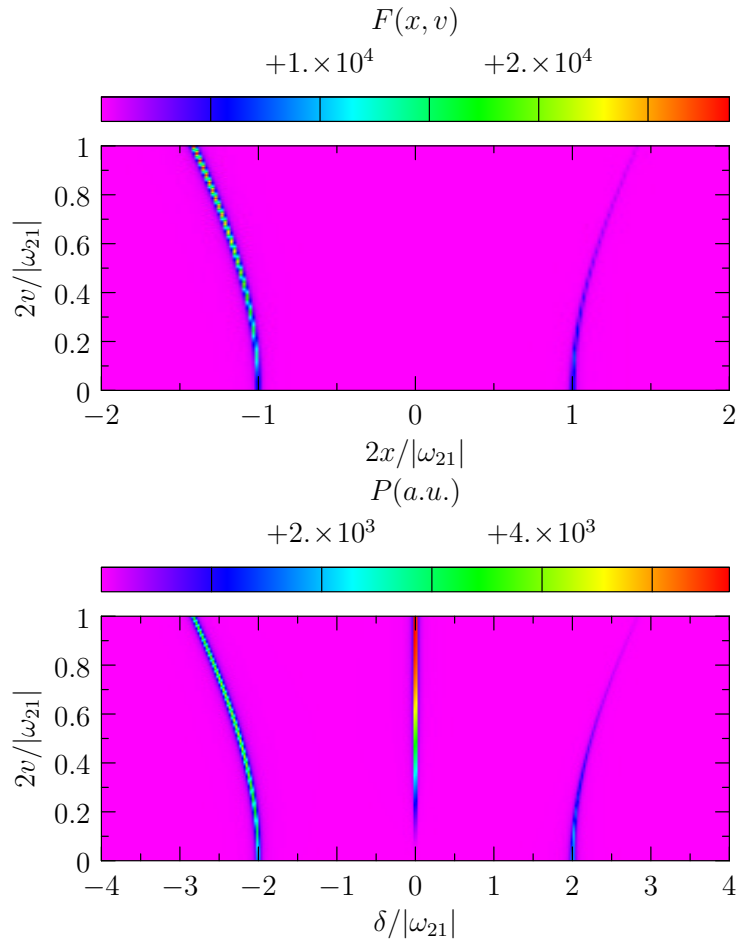


Figure 5.5: The 3D graph of transition probability function of x and interaction parameter v . In both graphs, we set $|\omega_{21}| = 200\gamma_\beta, \gamma_\beta = 100\gamma_\alpha$. In the upper graph, one sees the 3D distribution on variables $x = \omega_\alpha - (\omega_1 + \omega_2)/2$ and atomic interaction parameter v . In this graph, we find two ranges at $\omega = \omega_\pm(v)$. In the bottom one, the graph is obtained at a large time $t \gg \gamma_\alpha^{-1}$. We observe clearly in this graph that there are three local maxima of transition probabilities at ω_\pm, ω_0 . The discontinuities in both graphs come from a poor numerical precision

At a long time $t \gg \gamma_\alpha^{-1}$, one observes three peaks where two of them are at $\omega = \omega_\pm$ respectively and a third peak in the center. When $\omega_\alpha = (\omega_1 + \omega_2)/2$, the transition probability reads

$$P(t) = 16f^4 \gamma_\alpha \gamma_\beta \frac{L^2}{c^2} \frac{|e^{-\gamma_\alpha t} - e^{-2\gamma t}|^2}{(2\gamma - \gamma_\alpha)^2} \frac{v^2 + \gamma_\beta^2}{[\gamma_\beta^2 + \omega_{21}^2/4 + v^2]^2}. \quad (5.27)$$

The ratio between the transition probability at $v \neq 0$ and the one at $v = 0$ is

$$\frac{P|_{v \neq 0}}{P|_{v=0}} = \frac{[\omega_{21}^2 + 4\gamma_\beta^2]^2}{[\omega_{21}^2 + 4\gamma_\beta^2 + 4v^2]^2} \frac{\gamma_\beta^2 + v^2}{\gamma_\beta^2}. \quad (5.28)$$

In general, the interaction strength v is not so strong, it should not exceed the magnitude of $|\omega_{21}|/2$, roughly speaking, this ratio is assessed at

$$\frac{P|_{v \neq 0}}{P|_{v=0}} \simeq 1 + v^2/\gamma_\beta^2. \quad (5.29)$$

The interaction parameter can be much larger than the decay rate γ_β , therefore the transition probability can be greatly enhanced.

Recalling the case of the 2P2A transition by taking two photons from a monochromatic field, the peak induced in the center is notably asymmetrical. However in this case, one observes a symmetrical peak in the center.

The absence of asymmetry of the peak in the center is because in the latter case, there are infinitely many possible photon pairs in the source and the interference of photon pairs killed this asymmetry. We then conclude that the 2P2A transition shape is mainly determined by the properties of frequency anti-correlated photon pairs while in the transition strength part, the interaction parameter is a crucial factor.

C Two-photon absorption with a parametrically generated pulse of light

C.1 Type II SPDC two-photon source

A common way to generate two-photon states is via parametric down-conversion process in which three fields (pump (p), signal (s), and idler (i)) are involved because of $\chi^{(2)}$ nonlinearity. The frequency modes of newly generated two-photon state of the signal field and idler field can be continuous or discrete. It is well known that [?] by shining a train of periodic pulses on a parametric crystal, the generated photon state whose field is indeed a frequency comb with quantum properties whereas a single pump pulse generates a single down-converted pulse having a continuous spectrum.

Here we will focus our attention on a specific and concrete example: **an** SPDC two-photon collinear state generated from 1-dimensional photonic crystal, whose length can be very long, and pumped by a train of T_r -periodic pulses. Such two-photon state can be expressed as [Corona2007]

$$|\mathbb{II}\rangle = \sum_{\ell mnjk} f_{\ell,m,n}(\omega_j, \omega_k) |1_s : K_s^{(m)}(\omega_j); 1_i : K_i^{(n)}(\omega_k)\rangle, \quad (5.30)$$

where $K_\mu^{(m)}(\omega) = m2\pi/\Lambda + \tilde{K}_\mu^{(m)}(\omega)$, $0 \leq \tilde{K}_\mu^{(m)}(\omega) < 2\pi/\Lambda$, $\mu = s, i$; $m = 0, 1, 2, \dots$ are the Bloch wave-numbers and Λ is the length of the first Brillouin zone of the 1-dimensional crystal. The dispersion relations follow

$$K_\mu^{(m)}(\omega) = m\frac{\pi}{\Lambda} \pm \sqrt{(\beta_\mu^{(m)})^2 - |\kappa_\mu^{(m)}|^2}, \quad \beta_\mu^{(m)} = \bar{n}_\mu\omega/c - m\pi/\Lambda. \quad (5.31)$$

as well as

$$\kappa_\mu^{(m)} = i[1 - \cos(m\pi)]\alpha\bar{n}_\mu\omega/(4mc\pi), \quad (5.32)$$

where \bar{n}_μ is the mean refractive index. For simplicity, we assume the value of all three mean refractive indices are the same, hence denoted by \bar{n} . In general, there should be gaps between different bands if $\kappa_\mu^{(m)} \neq 0$, for frequencies whose corresponding momenta are close to the band gaps, the dispersion curves will dramatically changed in contrast to the optical branches and therefore the group velocities can be greatly reduced. The band width $\pi c/\bar{n}\Lambda$ is typically at the order of 10^{15} Hz if the length $\Lambda \simeq 300$ nm. We assume that the central frequency of the signal (idler) field ω is very close to a gap

$$\bar{\omega} = \omega \pmod{\pi c/\bar{n}\Lambda}, \quad \bar{\omega} \ll \pi c/\bar{n}\Lambda; \quad (5.33)$$

and the spectral width of all three fields are much smaller than the band width. The group velocities u_s, u_i of both signal field and idler field respectively become quite small. Under these conditions, due to the phase matching conditions, a common joint spectral amplitude $f_{\ell,m,n}$ is usually given as

$$f_{\ell,m,n}(\omega_j, \omega_k) = C_0\delta_{m,n}\delta_{\ell,2m}\delta_{m,m_0}\mathcal{E}_p\mathcal{E}_s\mathcal{E}_i e^{-(\omega_j+\omega_k-\omega_p)^2\omega_r^2/2\sigma^2} \text{sinc}(\phi_{m_0})e^{i\phi_{m_0}}. \quad (5.34)$$

The waist σ of the pump field which is assumed to be a Gaussian beam, is large and therefore the photon pairs have a broad distribution band whose central frequency is at

2ω . The contributions of the mismatched phase ϕ_{m_0} are coming from the the differences among those momenta of the three modes

$$\phi_{m_0} = L[K_s^{(m_0)}(\omega_j + \omega_\alpha) + K_i^{(m_0)}(\omega_k + \omega_\beta) - K_p^{(2m_0)}(\omega_j + \omega_k - \omega_p)]/2, \quad (5.35)$$

where L is the length of the photonic crystal. Let's make the Tylor's expansion to the three Bloch wave numbers,

$$K_\mu(\omega_\ell + \omega_\mu) = K_\mu(\omega_\mu) + \omega_\ell/u_\mu + \dots; \mu = s, i, p, \quad (5.36)$$

under perfect phase matching condition $K_s^{(m_0)}(\omega_\alpha) + K_i^{(m_0)}(\omega_\beta) = K_p^{(2m_0)}(\omega_\alpha + \omega_\beta)$, the phase becomes

$$\phi_{m_0}(\omega_j, \omega_k) = (\omega_j + \omega_k - \omega_p)T + (\omega_j - \omega_k - \omega_{\alpha\beta})T_c, \quad (5.37a)$$

$$T = L(u_s^{-1} + u_i^{-1} - 2u_p^{-1})L/4, \quad T_c = L(u_s^{-1} - u_i^{-1})L/4. \quad (5.37b)$$

It is worth mentioning that u_p can be quite large. Now these states can be expressed as

$$|\mathbb{I}\rangle = \sum_{mn} c_{m,n} |1_s : \omega_m, \alpha; 1_i : \omega_n, \beta\rangle, \quad (5.38)$$

where

$$c_{m,n} = C_0 e^{-(\omega_m + \omega_n - \omega_p)^2 / 2\sigma^2} \text{sinc}[\phi_{m_0}(\omega_m, \omega_n)] e^{i\phi_{m_0}(\omega_m, \omega_n)}. \quad (5.39)$$

If we make a selection such that the final state can only have two photons, then C_0 should be normalized, reads

$$C_0^2 = \left(\sum_{mn} e^{-\frac{(\omega_m + \omega_n - \omega_p)^2}{\sigma^2}} \text{sinc}^2[(\omega_m + \omega_n - \omega_p)T + (\omega_m - \omega_n - \omega_{\alpha\beta})T_c] \right)^{-1} \simeq 2 \frac{T_c \omega_r^2}{\sigma \pi^{3/2}}. \quad (5.40)$$

We have replaced the double sum by double integral as well as have extended the integral domain to the whole real plane to get the approximate result. When T_c is quite small, that is, the group velocities between two newly-generated beams in the crystal are close, two beams are well matched, the coefficients are small.

C.2 Transition behaviors at a long time $t \gg \gamma^{-1}$

The energy difference between two atoms is quite large, its magnitude is usually at several GHz, while the atomic decay rate is much smaller ($\gamma \sim 10$ MHz). As we have mentioned, when the frequencies of photons are close to the band gap in the crystal in which they were generated, their group velocities have great reductions, accordingly, the coherence time T and T_c ($T \gg T_c$) can be very large. If the coherence time is quite large, it satisfies $T \gg \gamma^{-1}, \sigma^{-1}$, then the transition probability amplitude follows

$$\begin{aligned} \mathcal{A} \simeq & -2\pi i C_0 e^{-2\gamma T_c t/T} \frac{f^2}{\omega_r^2 T} \\ & \left[\left(1 + \frac{2v}{\omega_+ - \omega_-} \right) \left(1 - \frac{\delta + 2i\gamma}{\omega_+ - \omega_-} \right) \frac{e^{-\frac{2T_c^2}{\sigma^2 T^2} (\omega_+ - \omega_\beta)^2 - 2i(\omega_+ - \omega_\beta)(t-t_0)T_c/T}}{\delta + 2(\omega_+ - \omega_\beta)T_c/T + 2i\gamma} \right. \\ & \left. + \left(1 - \frac{2v}{\omega_+ - \omega_-} \right) \left(1 + \frac{\delta + 2i\gamma}{\omega_+ - \omega_-} \right) \frac{e^{-\frac{2T_c^2}{\sigma^2 T^2} (\omega_- - \omega_\beta)^2 - 2i(\omega_- - \omega_\beta)(t-t_0)T_c/T}}{\delta + 2(\omega_- - \omega_\beta)T_c/T + 2i\gamma} \right]. \quad (5.41) \end{aligned}$$

This transition probability amplitude has a relatively fast oscillating term, hence the transition probability. The transition probability over a period $\pi T/(\omega_+ - \omega_-)T_c$ during which the decay is not noticeable, is a stable quantity. This slowly varying transition probability follows

$$\begin{aligned} \bar{P} \simeq \frac{8\sqrt{\pi}f^4T_c}{\sigma\omega_r^2T^2} e^{-4\gamma T_c t/T} & \left[\frac{(\omega_+ - \omega_- + 2v)^2 (\omega_+ - \omega_- - \delta)^2 e^{-\frac{4T_c^2}{\sigma^2 T^2} (\omega_+ - \omega_\beta)^2}}{(\omega_+ - \omega_-)^4 [\delta - 2(\omega_\beta - \omega_+)T_c/T]^2 + 4\gamma^2} \right. \\ & \left. + \frac{(\omega_+ - \omega_- - 2v)^2 (\omega_+ - \omega_- + \delta)^2 e^{-\frac{4T_c^2}{\sigma^2 T^2} (\omega_- - \omega_\beta)^2}}{(\omega_+ - \omega_-)^4 [\delta - 2(\omega_\beta - \omega_-)T_c/T]^2 + 4\gamma^2} \right]. \end{aligned} \quad (5.42)$$

One can find 2P2A resonance peaks around $\delta = 2(\omega_\beta - \omega_+)T_c/T$ and $\delta = 2(\omega_\beta - \omega_-)T_c/T$. The distant between the two peaks can be assessed at $\Delta_{peaks} = 2(\omega_+ - \omega_-)T_c/T$. If the two peaks are fairly separated, the widths of peaks are about 2γ . See Fig. 5.6.

Because of the smallness of T_c/T , the large single photon detunings become sensitive to the 2P2A resonance spectrum. We stress that this phenomenon mainly originates in the phase interferences rather than a pure frequency anti-correlations. This can be seen from the absence of the peak-splitting in the bottom graph of Fig. 5.6 or from the Eq. (5.42) when $T_c \rightarrow 0$.

C.3 Transition probability of some diagonal matrices that have the same spectra

Consider the completely dephased state corresponding to the state discussed in last subsections,

$$\Lambda = \sum_{m,n} |c_{m,n}|^2 |1 : \omega_m, s; 1 : \omega_n, i\rangle \langle 1 : \omega_m, s; 1 : \omega_n, i|. \quad (5.43)$$

Consequently, when $T_c \ll T$, the transition probability follows

$$P \simeq 4f^4 \frac{T_c \pi^{1/2}}{\sigma \gamma T} \left[1 + \frac{4v^2}{(\omega_+ - \omega_-)^2} \right] \left(\frac{e^{-4\omega_{\beta-}^2 T_c^2 / \sigma^2 T^2}}{[\delta - 2\omega_{\beta-} T_c/T]^2 + 4\gamma^2} + \frac{e^{-4\omega_{\beta+}^2 T_c^2 / \sigma^2 T^2}}{[\delta - 2\omega_{\beta+} T_c/T]^2 + 4\gamma^2} \right). \quad (5.44)$$

When $|\omega_{\beta-}|T_c/T \sim \gamma$, one can find the two peaks around the center. If the two peaks are found, then the magnitude of T_c/T is assessed at $\gamma/|\omega_{\beta-} - \omega_-|$, therefore the transition probability

$$P \simeq f^4 \frac{\pi^{1/2}}{\sigma \Delta \gamma^2}, \quad \Delta = \{|\omega_{\beta-}|, |\omega_{\beta+}|\}. \quad (5.45)$$

In fact this transition probability is much larger than the one of the factorized completely dephased state

$$\Lambda_{(} = \sum_{mn} \left(\sum_j |c_{m,j}|^2 \right) \times \left(\sum_k |c_{k,n}|^2 \right) |1 : \omega_m, s; 1 : \omega_n, i\rangle \langle 1 : \omega_m, s; 1 : \omega_n, i|, \quad (5.46)$$

that has the same spectra.

From this expression (5.44), we see that the two two-photon–two-atom resonance peaks around the center can also induced by using a correlated state without any entanglement.

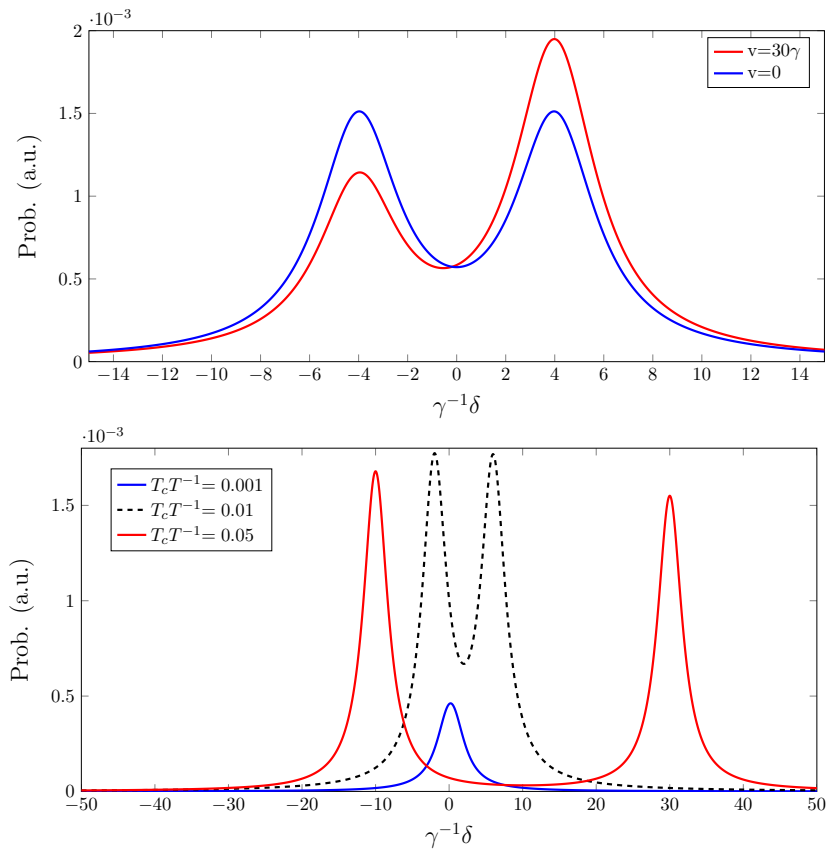


Figure 5.6: 2P2A resonance with the type II SPDC two-photon states. Here we set $\omega_+ - \omega_0 = 200\gamma$, $\omega_- - \omega_0 = -200\gamma$, $\sigma = 100\gamma$, $\omega_\beta - \omega_0 = 100\gamma$. One finds the ratio T_c/T between the group velocities of the new beams, therefore T_c , and the mismatch between three beams, thus T , is quite important. When this ratio is small, one finds a weak single peak in the middle; when the ratio becomes larger, the single peak splits into two peaks. The distance between two peaks is monotonely increasing with T_c/T .

The atomic interaction parameter v enhances the total transition probability but does not modify the shape of the transition spectrum.

One then concludes that the entanglement is not a necessity for inducing such two peaks. It is a special spectrum distribution of photon states.

D Conclusion

In this chapter, we have discussed the two-photon transition in two-atom system that has an atomic interaction. In the first case, the two photons are from a same monochromatic field. When any photon in the pair is far off resonance with both atoms, but the two photons are resonant with both atoms, a peak at frequency $\omega = \omega_0 = (\omega_1 + \omega_2)/2$ is induced by atomic interaction. The width of this peak is independent of the interaction parameter v . However, this parameter gives rise to an absorption suppression at frequency ω_+ and an absorption enhancement at frequency ω_- . Another important feature in this 2P2A transition is that the probability is independent of the pureness of a state, the real physical factor is $g^{(2)}$ which represents the photon spacing. A large transition probability is attained by using the states with large photon fluctuations.

In a case of 2P2A transition with a bipartite continuous two-photon state whose frequencies are anti-correlated, we find the transition profile is determined by the photon source and the shape of peak is symmetrical in contrast to the previous monochromatic field case. At the perfect 2P2A resonance condition, $\delta = 0$, we find a remarkable enhancement of the transition probability by the emergence of interaction parameter v in both cases.

Is the enhancement of 2P2A transition a quantum effect ?

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IN previous chapters, we have shown that transition probabilities of various processes crucially depend on the quantum state which is used. In this chapter we address the issue of the physical origin of such a behavior. In particular, is the enhancement effect of 2P2A transition due to some kind of quantum effect or not ? Its answer is obviously linked to the precise meaning attributed to the words "quantum effect", and more precisely to the definition of a "quantum correlation". This is what we will consider in the first section of this chapter. We then identify the type of conclusion which is used to enhance 2P2A probability and discuss its properties using the time/frequency Wigner representation of the state.

A What kind of correlation is required to enhance the 2P2A transition probability ?

We have found in Chap. 4 that the 2P2A transition probability depends crucially on the specific state of light used for the excitation, even when all the considered states have the same energy spectrum. The question we address now is the physical origin of an enhanced transition probability. We have seen that entangled and unentangled states may give comparable results, so a first answer to the question is obviously that entanglement is not at the origin of the effect but rather is some kind of correlation effect which is shared by entangled and unentangled states.

Candidates likely to play a role in the present problem are time correlation and frequency correlation. We will now examine them successively

A.1 Temporal correlation effect

It is well characterized by the cross-second-order correlation function $g_{\times}^{(2)}(t, \tau)$

$$g_{\times}^{(2)}(t, \tau) = \frac{\text{Tr}[\rho_0 \hat{E}_{\alpha}^{(-)}(\tau) \hat{E}_{\beta}^{(-)}(t) \hat{E}_{\beta}^{(+)}(t) \hat{E}_{\alpha}^{(+)}(\tau)]}{\text{Tr}[\rho_0 \hat{E}_{\alpha}^{(-)}(t) \hat{E}_{\alpha}^{(+)}(t)] \text{Tr}[\rho_0 \hat{E}_{\beta}^{(-)}(t) \hat{E}_{\beta}^{(+)}(t)]}. \quad (6.1)$$

Assuming that the amplitude of the single-photon electric field is a smooth function of ω_k , one gets the following for the pure state $|\Psi\rangle = \sum_{kq} c_{kq} |1 : \omega_k, \alpha; 1 : \omega_q, \beta\rangle$:

$$G_{\times}^{(2)}(t, \tau) = \left| \sum_{kq} c_{kq} e^{-i\omega_k \tau - i\omega_q t} \right|^2. \quad (6.2)$$

It is the modulus square of two-time Fourier transform of the two-photon state.

1. In the case of the cascade state (4.16),

$$g_{\times}^{(2)}(t, \tau) = \left(\frac{\gamma_{\beta}}{\gamma_{\alpha}} - 1 \right) \frac{\gamma_{\alpha} \gamma_{\beta}}{\pi^2} \theta(\tau) \theta(t - \tau) e^{2\gamma_{\alpha} t - 2\gamma_{\beta}(t - \tau)}, \quad (6.3)$$

$\theta(t)$ being the step function. We notice here a time asymmetry between t and τ , expected in the case of a cascade in which the ω_{α} photon is always emitted before the ω_{β} photon.

2. For the SPDC state (4.29),

$$g_{\times}^{(2)}(t, \tau) = \frac{2}{\mathcal{N}^2} [1 + \sin \omega_{\alpha\beta}(t - \tau)] \exp \left[-\frac{\sigma_{\beta}^2(t - \tau)^2}{2} - \frac{2\sigma_{\alpha}^2 \sigma_{\beta}^2}{\sigma_{\alpha}^2 + 2\sigma_{\beta}^2} \left(t_0 - \frac{t + \tau}{2} \right)^2 \right]. \quad (6.4)$$

As can be seen in Figure (6.1), $g_{\times}^{(2)}(t, \tau)$ is in both cases significant only very close to the diagonal, which implies that both states exhibit strong temporal correlations, as expected. The width of the diagonal, which gives the characteristic time of this correlation, is equal to $\gamma_{\beta}^{-1}(\sigma_{\beta}^{-1})$ in both the cascade and SPDC cases.

It is easy to see that for the correlated-separable states (4.23) and (4.31), there is no time dependence for $g_{\times}^{(2)}(t, \tau)$ ($g_{\times}^{(2)}(t, \tau)|_{sep.cor} = 1$), and hence no temporal correlation, as

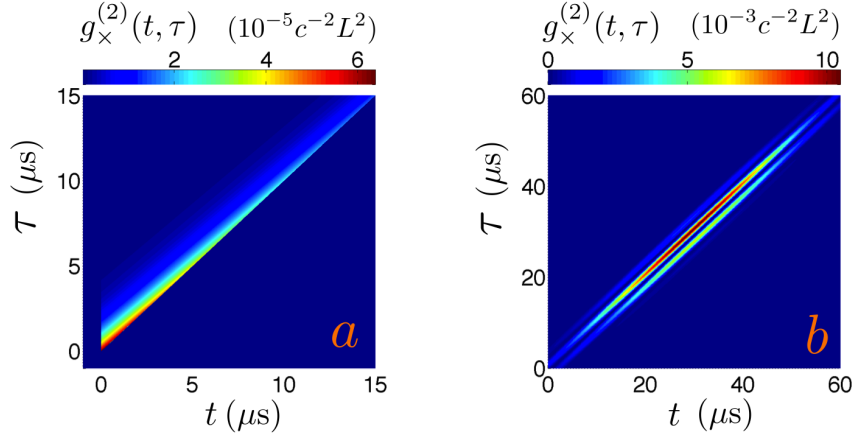


Figure 6.1: Plots of the cross temporal correlation function $g_x^{(2)}(t, \tau)$. The left one is for the atom cascade two-photon state, with $\gamma_\alpha = 0.05\text{MHz}$ and $\gamma_\beta = 0.5\text{MHz}$; the right plot is for the SPDC two-photon state, in which the pulse takes place around $t_0 = 30\mu\text{s}$, and with $\sigma_\alpha = 0.05\text{MHz}$, $\sigma_\beta = 0.5\text{MHz}$, $\omega_{\beta\alpha} = 2\text{MHz}$. Note the (t, τ) asymmetry in the first figure, and fringes in the second one are due to interferences from two temporal processes. In both plots, one finds significant temporal correlations along the diagonal line. In a real condition, the value of $\omega_{\beta\alpha}$ should be much greater, leading to a poorer graphic representation for interference patterns.

expected from a c.w. time averaged state in which the photons arrive at any time. It is also the case for the coherent states (4.47). As these states give 2P2A transition probabilities comparable to the entangled state, we must conclude that the temporal correlation is not the physical origin of the enhancement effect, nor is the time ordering of the photons present in the cascade state. The physical reason is that, as we have neglected their spontaneous emission, the two detecting atoms have an infinite memory time, and hence they can be excited separately at any time.

A.2 Frequency correlation effect

It is well characterized by the cross second order frequency correlation function $g_x^{(2)}(\omega, \omega')$,

$$g_x^{(2)}(\omega, \omega') = \frac{\text{Tr}[\rho_0 \hat{E}_\alpha^{(-)}(\omega') \hat{E}_\beta^{(-)}(\omega) \hat{E}_\beta^{(+)}(\omega) \hat{E}_\alpha^{(+)}(\omega')]}{\text{Tr}[\rho_0 \hat{E}_\alpha^{(-)}(\omega') \hat{E}_\alpha^{(+)}(\omega')] \text{Tr}[\rho_0 \hat{E}_\beta^{(-)}(\omega) \hat{E}_\beta^{(+)}(\omega)]}, \quad (6.5)$$

equal in the pure state case to $|c(\omega, \omega')|^2$ and to $p(\omega, \omega')$ in the diagonal mixed-state case.

This quantity is plotted in Figure 6.2 for the cascade and SPDC states, either entangled, correlated-separable, or factorized. One observes that the frequency correlation functions take significant values only on the anti-diagonal for the left-side plots, which implies that the corresponding states exhibit strong frequency anticorrelations. This is not the case for the right side plots. The width of the anti-diagonal, which gives the characteristic width of the frequency anticorrelation, is equal to $\gamma_\alpha(\sigma_\alpha)$ in both the cascade and SPDC entangled and correlated-separable cases.

The important point to notice is that such a frequency anticorrelation exists for all the states which exhibit 2P2A resonance enhancement, and is not present for the states

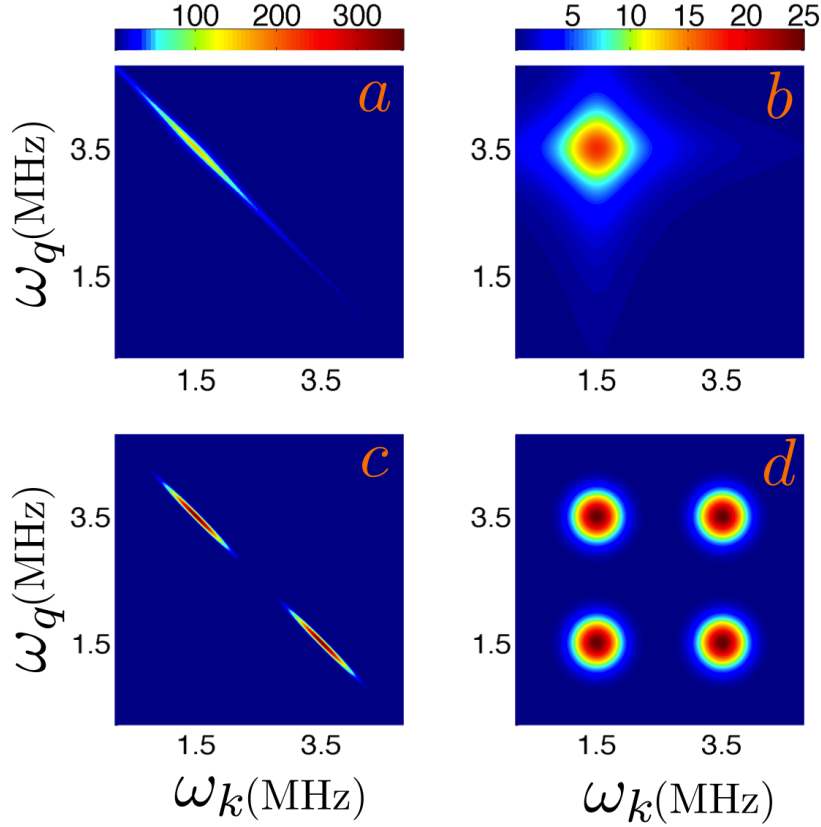


Figure 6.2: Plots of the cross frequency correlation function $g_x^{(2)}(\omega_k, \omega_q)$: (a) entangled, correlated-separable, and coherent cascade states; (b) factorized cascade state; (c) entangled, correlated-separable, and coherent SPDC states; and (d) factorized SPDC state. In all plots $\gamma_\alpha = \sigma_\alpha = 0.05$ MHz, $\gamma_\beta = \sigma_\beta = 0.5$ MHz, $\omega_\alpha = 1.5$ MHz, and $\omega_\beta = 3.5$ MHz. The color codes, in the unit of c^2/L^2 , on the top left (right) are shared by (a) and (c) (b and d). The left-side plots exhibit strong frequency anticorrelations along the line $\omega_k + \omega_q = \omega_\alpha + \omega_\beta$, while in the right-side plots, one finds no such a correlation. The SPDC two-photon source is nondegenerate and each photon has two distribution peaks; thus one sees two bright spots in the left side bottom plot and four bright spots in the factorized case in the right-side bottom plot. In a real condition, the distances of the peaks in the bottom plots are much greater, and the sizes of spots are much smaller.

which do not give rise to this effect. We are therefore led to the conclusion that *the property needed to enhance the 2P2A excitation is precisely the presence of strong frequency anticorrelations in the quantum state*. We will characterize more precisely the quantum aspects of frequency correlations in the next sections.

This conclusion, that we have demonstrated for the two specific examples considered in chapter 4, is far more general, as can be seen in the expression of the probability written for any switched-on two-photon state.

Equations (37), (38), and (39) indeed show that the probability of 2P2A excitation is proportional to the component of the density matrix of the two-photon state corresponding to the existence of one photon with frequency ω_1 and one photon with frequency ω_2 . This gives a simple interpretation of the effect: there is 2P2A excitation only when each photon of the two-photon state is resonant with the atomic transition of the atom it excites. This is expected, since we are considering that the atomic excited states have a very long lifetime, and therefore very narrow linewidths. Since the spectrum of each photon of the source has a much larger bandwidth, the probability of excitation is small. If the photons are not correlated in frequency, the probability of double excitation is proportional to the product of the probabilities that each photon has the corresponding transition frequency, and this yields a very small transition probability. But when the photons are anti-correlated in frequency such that the sum of their frequencies is equal to the sum of the transition frequencies of the atoms, when one photon is resonant with one atomic transition, the correlated photon will be automatically resonant with the other transition, and the probability of 2P2A transition will in general be much higher than in the non-correlated case.

We can say that the 2P2A transition occurs with a higher probability when the sum of the photon frequencies is found inside a small interval around the sum of the atomic transition frequencies, so that the enhancement is associated with the inverse of the variance of the $|c(\omega_k, \omega_q)|^2$ distribution in the direction of the diagonal.

The question which arises now is whether this strong frequency anti-correlation responsible for the enhancement can be found only in non-classical states. This should be the sign of a pure quantum effect. This is what we examine in the following.

B Characterization of bipartite correlations

We have considered in this thesis various bipartite light sources, microscopic or macroscopic, pure or mixed. What still remains to be clarified are the characterization of the correlations that exist in such states.

Correlations in quantum context are fascinating and sometimes mysterious. For example, in classical physics, correlations can only exist in composite system while in quantum systems, correlations such like contextual correlations can even exist in an indivisible system [Lapkiewicz2011]. Interestingly, on one hand, there are some quantum entangled states that generate outcomes perfectly in accord with local-realism [Werner1989]. In other words, the correlations in such states are regarded as classical. On the other hand, the correlations in a separable mixed state can have a strong non-classical character, which can be assessed for example by a non-vanishing value of the quantum discord [Ollivier2001], which may contradict our intuition. It is important to distinguish which kinds of correlations are genuinely quantum. This question is still far from being totally solved, in spite of lots of efforts that have been devoted to address it.

Nowadays, there are some kinds of widely used measures [Modi2012] of quantum correlations, such as gemellity, quantum discord, quantum deficit, measurement induced disturbance (MID). The relations between various criteria in multimode quantum state are discussed in [Treps2005]. In this section, we will apply these concepts to our photon states.

B.1 Separability and purity

A bipartite diagonal density matrix

$$\Lambda_{\alpha\beta} = \sum_{kq} |c_{kq}|^2 |\phi_\alpha : \omega_k, \phi_\beta : \omega_q\rangle \langle \phi_\alpha : \omega_k, \phi_\beta : \omega_q|, \quad (6.6)$$

where ϕ stands for the photon state, for example $\phi = \alpha$ stands for the coherent state while $\phi = 1$, represents the single photon Fock state, can be regrouped as

$$\Lambda_{\alpha\beta} = \sum_k \left(\sum_n |c_{kn}|^2 \right) |\phi_\alpha : \omega_k\rangle \langle \phi_\alpha : \omega_k| \otimes \rho_{\beta k}, \quad \rho_{\beta k} = \sum_q \frac{|c_{kq}|^2}{\sum_m |c_{km}|^2} |\phi : \omega_q\rangle \langle \phi : \omega_q|. \quad (6.7)$$

Therefore, the density matrix $\Lambda_{\alpha\beta}$ is separable, do not have any entanglement.

It is always possible to write a pure bipartite state

$$|\psi, \alpha\beta\rangle = \sum_{kq} c_{kq} |\phi_\alpha : \omega_k, \phi_\beta : \omega_q\rangle \quad (6.8)$$

into the Schmidt decomposition form,

$$|\psi, \alpha\beta\rangle = \sum_m \sqrt{\lambda_m} |f_m^\alpha; f_m^\beta\rangle; \quad \langle f_m^\alpha | f_n^\alpha \rangle = \delta_{mn}, \quad \langle f_m^\beta | f_n^\beta \rangle = \delta_{mn}. \quad (6.9)$$

Here if symbol ψ represents two-photon state, then we will use \mathbb{I} explicitly; if ψ represents a coherent state, we replace ψ by symbol z .

According to the formula (1.50), the cooperativity parameter of bipartite two photon state $|\mathbb{I}, \alpha\beta\rangle$ can then be calculated with the following formula,

$$\kappa = \frac{1}{\sum_{mn} |\sum_k c_{km} c_{kn}^*|^2}. \quad (6.10)$$

This formula also applies (approximately) when dealing with a corresponding state $|z, \alpha\beta\rangle$ on the condition of large $|z|$. Therefore, when we are dealing with quantities that are related to this number, we will never try to distinguish the two states.

Take c_{kq} as the coefficient of a cascade two-photon state [see Eq. (1.32)] for example, the cooperativity parameter is found to be:

$$\kappa_{cascade} = 1 + \gamma_\beta/\gamma_\alpha. \quad (6.11)$$

It has a lower bound 1 when γ_α is infinitely large than γ_β . In this case, the population on the top state of cascade three-level source atom will de-excited to the mediate state within a very short duration, and then from the mediate level to the ground state. As a result, the total energy fluctuations of the two-photon state will be quite large and it gives rise to poorer frequency anti-correlations. When γ_α is extremely large, then the de-excitation from the top state finishes instantaneously after it is excited, one should hardly to observe such an emission. What one observes is that a photon is emitted from the de-excitation. The behavior of such a two-photon state is just as a combination of behaviors of uncorrelated photons. On the contrary, if γ_α is very small in contrast to γ_β , then a second photon will be emitted soon after the photon created from the transition from the top level of the intermediate level, and the total energy fluctuations of the two-photon state can be very small, that is, the two photons are in good frequency anti-correlations. In an extreme case where $\gamma_\alpha \rightarrow 0$, then the photon pairs are perfectly frequency-anticorrelated, and the expression of c_{kq} will degrade into $c(\omega_k, \omega_e - \omega_k)$, and the state is indeed in the form of Schmidt decomposition. Surely, there are incredibly many terms (the Schmidt rank then is infinity) in the expression. Due to the flatness of the Schmidt coefficients distribution, the corresponding cooperativity parameter can be very large.

It is also worth mentioning, a larger ratio of $\gamma_\beta/\gamma_\alpha$ gives rise to both better frequency correlations and stronger entanglement of the total state. It is still not clear that whether the enhancement of correlations is mainly determined by the quantum entanglement. However, if one dephases the state completely, then one has a correlated-separable state which **does** not have any entanglement. The distribution coefficients have indeed frequency anti-correlations. By increasing the ratio, once again, one find the state have better correlations. Then we conclude: **the cooperativity parameter is a good measure of frequency anti-correlations and entanglement; the frequency anti-correlations are, at least, not mainly determined by entanglement.**

At this stage, it is still not clear whether the correlations are classical or quantum.

We will come to a similar situation by considering a coefficient as the one in parametric down conversion two-photon state $|\psi_{pdc}\rangle = \sum_{kq} c_{kq}|1 : \omega_k, \alpha; 1 : \omega_q, \beta\rangle$,

$$c_{kq} = 2\pi cL^{-1}(\pi\sigma\Gamma)^{-1/2}e^{-(\omega_k+\omega_q-\omega_p^0)^2/2\sigma^2-(\omega_q-\omega_\beta)^2/2\Gamma^2+i(\omega_k+\omega_q-\omega_p^0)t_0}, \quad (6.12)$$

where σ, Γ are widths of pump field whose central frequency is ω_p^0 and of idle field with central frequency ω_β ; t_0 is the delay time. The cooperativity parameter in this case turns out to be

$$\kappa_{pdc} = \sqrt{1 + \Gamma^2/\sigma^2}. \quad (6.13)$$

B.1.1 Schmidt coefficients

It is quite difficult to find an analytical expression of the Schmidt decomposition of a pure bipartite state. However, in some situations, there exists such exact expressions, or

approximate analytical expressions.

Let's first consider the coefficients (6.12) of a type II SPDC two-photon state. One can deform it as

$$c_{kq}e^{-i(\omega_k+\omega_q-\omega_p^0)t_0} = \frac{2\pi cL^{-1}}{(\pi\sigma\Gamma)^{1/2}} \exp\left(-\frac{1-u}{1+u}\frac{(x+y)^2}{4} - \frac{1+u}{1-u}\frac{(x-y)^2}{4}\right), \quad (6.14)$$

which admits the following expansion

$$c_{kq}e^{-i(\omega_k+\omega_q-\omega_p^0)t_0} = \frac{2\pi c}{L} \sqrt{\frac{1-u^2}{\sigma\Gamma}} \sum_{n=0}^{\infty} u^n \psi_n(x)\psi_n(y), \quad (6.15)$$

$$\psi_n(x) = (2^n n! \sqrt{\pi})^{-1/2} e^{-x^2/2} H_n(x). \quad (6.16)$$

with the parameters

$$u = -\sqrt{1 - \frac{2}{\kappa_{pdc} + 1}}; \quad x = \frac{\omega_k + \omega_\beta - \omega_p^0}{\sigma\sqrt{\kappa_{pdc}}}; \quad y = \sqrt{\kappa_{pdc}} \frac{\omega_q - \omega_\beta}{\Gamma}, \quad (6.17)$$

and $H_n(x)$ is the Hermite polynomial of degree n . As expected, the cooperativity parameter κ_{pdc} which is an important measure of correlation, now enters the decomposition. In this new orthonormal basis

$$|\psi_n : \alpha\rangle = i^n \sqrt{\frac{2\pi c}{L\sigma\sqrt{\kappa_{pdc}}}} \sum_k \psi_n\left(\frac{\omega_k + \omega_\beta - \omega_p^0}{\sigma\sqrt{\kappa_{pdc}}}\right) e^{i\omega_k t_0} |1 : \omega_k, \alpha\rangle, \quad (6.18)$$

$$|\psi_n : \beta\rangle = i^n \sqrt{\frac{2\pi c}{\sqrt{\kappa_{pdc}}L\Gamma}} \sum_q \psi_n\left(\sqrt{\kappa_{pdc}} \frac{\omega_q - \omega_\beta}{\Gamma}\right) e^{i\omega_q t_0} |1 : \omega_q, \beta\rangle, \quad (6.19)$$

the cooperativity parameter rescales the distributions to make their frequency widths quite comparable. It is in agreement with the distribution function of $|c_{kq}|^2$. Thanks to these results, the state now has a simple decomposition form:

$$|\psi_{pdc}\rangle = \sqrt{1-u^2} \sum_{n=0}^{\infty} |u|^n |\psi_n : \alpha\rangle |\psi_n : \beta\rangle. \quad (6.20)$$

By tracing the total density matrix over one part, one has the reduced density matrices:

$$\rho_\alpha = (1-u^2) \sum_n u^{2n} |\psi_n : \alpha\rangle \langle \psi_n : \alpha|, \quad (6.21a)$$

$$\rho_\beta = (1-u^2) \sum_n u^{2n} |\psi_n : \beta\rangle \langle \psi_n : \beta|. \quad (6.21b)$$

Both of them are in the spectral decomposition form.

Now let's consider the Schmidt decomposition of the cascade two-photon state whose coefficients c_{kq} are defined in Eq. (1.32). One of its reduced density matrices is given by

$$\rho_{reduced}^{cas} = \sum_k \left(\sum_m c_{mk} |1 : \omega_m\rangle \right) \left(\sum_n c_{nk} |1 : \omega_n\rangle \right)^\dagger. \quad (6.22)$$

For simplicity, let us introduce normalized state $|k\rangle$ as

$$|k\rangle \propto \sum_m c_{mk} |1 : \omega_m\rangle. \quad (6.23)$$

We have the following inner product

$$\langle j|k\rangle \propto \sum_m c_{mk}c_{mj}^* = \frac{2c\gamma_\beta/L}{(\omega_k - \omega_\beta + i\gamma_\beta)(\omega_j - \omega_\beta - i\gamma_\beta)} \frac{2i\gamma_\alpha}{\omega_k - \omega_j + 2i\gamma_\alpha}. \quad (6.24)$$

When $\gamma_\alpha \ll \gamma_\beta$, if $|\omega_j - \omega_k| \gg 2\gamma_\alpha$, then $\langle j|k\rangle \simeq 0$; if $|\omega_j - \omega_k| \ll 2\gamma_\alpha$, then we have $\langle j|k\rangle \propto 2c\gamma_\beta/[(\omega_k - \omega_\beta)^2 + \gamma_\beta^2]L$. We then approximate $|j\rangle$ as a piecewise state $|N_j\rangle$,

$$|N_j\rangle = \sqrt{\frac{\pi}{\gamma_\beta} \frac{4N_j^2\gamma_\alpha^2 + \gamma_\beta^2}{4\gamma_\alpha^2}} \int_{N_j \cdot 2\gamma_\alpha}^{(N_j+1) \cdot 2\gamma_\alpha} \sum_m c(\omega_m, \omega) d\omega |1 : \omega_m\rangle. \quad (6.25)$$

where N_j is the integer part of $\omega_j/2\gamma_\alpha$. The orthonormal relations read

$$\langle N_j|N_k\rangle \simeq \delta_{jk}. \quad (6.26)$$

Then the reduced density matrix approximates at

$$\rho_{reduced}^{cas} = \sum_{N_k} \frac{\gamma_\beta}{\pi} \frac{2\gamma_\alpha}{(2N_k\gamma_\alpha - \omega_\beta)^2 + \gamma_\beta^2} |N_k\rangle \langle N_k|. \quad (6.27)$$

The Schmidt coefficients defined in Eq. (1.48) are approximately equal to

$$\sqrt{\lambda_k} \simeq \sqrt{\frac{\gamma_\beta}{\pi} \frac{2\gamma_\alpha}{(2N_k\gamma_\alpha - \omega_\beta)^2 + \gamma_\beta^2}}. \quad (6.28)$$

By using this result, one finds the cooperativity parameter

$$\tilde{\kappa}_{cas} = \pi\gamma_\beta/\gamma_\alpha, \quad (6.29)$$

which is 3 times larger than the exact value (6.11). Therefore, the approximation we have made is rather rough.

Anyway, we will still use the results in (6.28) for qualitative analyses, due to the difficulty to obtain an analytical expression of the Schmidt decomposition of the cascade two-photon state. We should bear in mind that the related results are not accurate. An alternative is using Gaussian distributions to replace those Lorentzian ones. Then the final expressions are the same as those of the type II SPDC two-photon state. This alternative is not so good, owing to the the different convergence rates for frequencies far away from the peak centers.

B.2 Simon's criterion on multimode states

In this part, two type of completely dephased states:

$$\Lambda_{\text{II}} = \sum_{m,n} |c(\omega_m, \omega_n)|^2 |1 : \omega_m, \alpha; 1 : \omega_n, \beta\rangle \langle 1 : \omega_m, \alpha; 1 : \omega_n, \beta|; \quad (6.30a)$$

$$\Lambda_c = \sum_{m,n} |c(\omega_m, \omega_n)|^2 |z : \omega_m, \alpha; z : \omega_n, \beta\rangle \langle z : \omega_m, \alpha; z : \omega_n, \beta|; \quad (6.30b)$$

where $z \in \mathbb{C}$, $|z| \gg 1$, are of special interest. The two states are just convex combination of single-frequency-mode product state, the nonclassical properties of the two states can only exist in those elements $\Lambda_{(mn)} = |1 : \omega_m, \alpha\rangle \langle 1 : \omega_m, \alpha| \otimes |1 : \omega_n, \beta\rangle \langle 1 : \omega_n, \beta|$ and $\Lambda_{c,(mn)} = |z : \omega_m, \alpha\rangle \langle z : \omega_m, \alpha| \otimes |z : \omega_n, \beta\rangle \langle z : \omega_n, \beta|$.

The Mandel matrix, which is introduced in the first chapter, of the state $\Lambda_{(mn)}$ is

$$M_{\mathbb{I},(mn)} = \begin{pmatrix} 1 & 1 & 0 & 0 & 1 \\ 1 & 0 & 0 & 0 & 1 \\ 0 & 0 & 1 & 0 & 0 \\ 0 & 0 & 0 & 1 & 0 \\ 1 & 1 & 0 & 0 & 0 \end{pmatrix}. \quad (6.31)$$

The eigenvalues of the Mandel matrices of these elements are

$$\{1, 1, 1 + \sqrt{2}, 1 - \sqrt{2}, -1\}. \quad (6.32)$$

Because the latter 2 eigenvalues are negative, one concludes any local element $\Lambda_{(mn)}$ is a quantum optical nonclassical state. This conclusion is well known since any single photon state cannot be produced by classical means. Therefore the dephased state is quantum optical non-classical state.

However, consider a coherent state,

$$\Lambda_{c,(kq)} = |z : \omega_k, \alpha\rangle \langle z : \omega_k, \alpha| \otimes |z : \omega_q, \beta\rangle \langle z : \omega_q, \beta|. \quad (6.33)$$

The Mandel matrix reads

$$M_{c,(mn)} = \begin{pmatrix} 1 & |z|^2 & |z|^2 & |z|^2 & |z|^2 \\ |z|^2 & |z|^4 & |z|^4 & |z|^4 & |z|^4 \\ |z|^2 & |z|^4 & |z|^4 & |z|^4 & |z|^4 \\ |z|^2 & |z|^4 & |z|^4 & |z|^4 & |z|^4 \\ |z|^2 & |z|^4 & |z|^4 & |z|^4 & |z|^4 \end{pmatrix}. \quad (6.34)$$

Follow the same procedures, the eigenvalues of the Mandel matrix of state $|z : \omega_k, \alpha\rangle \langle z : \omega_k, \alpha| \otimes |z : \omega_q, \beta\rangle \langle z : \omega_q, \beta|$ are

$$\{0, 0, 0, 0, 4|z|^4 + 1\}. \quad (6.35)$$

All the five eigenvalues are positive. So, the classically mixed coherent state Λ_c is a quantum optical classical state.

Follow the procedures in [Ivan2011], the Mandel matrix of a generic bipartite two-photon state defined there can be rearranged as

$$M = \begin{pmatrix} 1 & \langle : (\hat{\mathbf{a}} \otimes \hat{\mathbf{a}}_+)^T : \rangle & \langle : (\hat{\mathbf{b}} \otimes \hat{\mathbf{b}}_+)^T : \rangle \\ \langle : \hat{\mathbf{a}}_+ \otimes \hat{\mathbf{a}} : \rangle & 0 & \langle : \hat{\mathbf{a}}_+ \otimes \hat{\mathbf{a}} (\hat{\mathbf{b}} \otimes \hat{\mathbf{b}}_+)^T : \rangle \\ \langle : \hat{\mathbf{b}}_+ \otimes \hat{\mathbf{b}} : \rangle & \langle : \hat{\mathbf{b}}_+ \otimes \hat{\mathbf{b}} (\hat{\mathbf{a}} \otimes \hat{\mathbf{a}}_+)^T : \rangle & 0 \end{pmatrix} \oplus \begin{pmatrix} \langle : \hat{\mathbf{a}}_+ \hat{\mathbf{a}}^T \otimes \hat{\mathbf{b}} \hat{\mathbf{b}}_+^T : \rangle & 0 \\ 0 & \langle : \hat{\mathbf{b}}_+ \hat{\mathbf{b}}^T \otimes \hat{\mathbf{a}} \hat{\mathbf{a}}_+^T : \rangle \end{pmatrix}, \quad (6.36)$$

in which the notations $\hat{\mathbf{c}} = (\dots, \hat{c}_k, \dots)^T$, $\hat{\mathbf{c}}_+ = (\dots, \hat{c}_k^\dagger, \dots)^T$, $c = a, b$. It is a block matrix, and the majority of the blocks are zeros. The eigenvalues can be solved from the following equations

$$|\langle : \hat{\mathbf{b}}_+ \otimes \hat{\mathbf{a}} (\hat{\mathbf{b}} \otimes \hat{\mathbf{a}}_+)^T : \rangle - \lambda| = 0; \quad (6.37)$$

$$|\langle : \hat{\mathbf{a}}_+ \otimes \hat{\mathbf{b}} (\hat{\mathbf{a}} \otimes \hat{\mathbf{b}}_+)^T : \rangle - \lambda| = 0; \quad (6.38)$$

$$\begin{vmatrix} 1 - \lambda & \langle : (\hat{\mathbf{a}} \otimes \hat{\mathbf{a}}_+)^T : \rangle & \langle : (\hat{\mathbf{b}} \otimes \hat{\mathbf{b}}_+)^T : \rangle \\ \langle : \hat{\mathbf{a}}_+ \otimes \hat{\mathbf{a}} : \rangle & -\lambda & \langle : \hat{\mathbf{a}}_+ \otimes \hat{\mathbf{a}} (\hat{\mathbf{b}} \otimes \hat{\mathbf{b}}_+)^T : \rangle \\ \langle : \hat{\mathbf{b}}_+ \otimes \hat{\mathbf{b}} : \rangle & \langle : \hat{\mathbf{b}}_+ \otimes \hat{\mathbf{b}} (\hat{\mathbf{a}} \otimes \hat{\mathbf{a}}_+)^T : \rangle & -\lambda \end{vmatrix} = 0. \quad (6.39)$$

Now let's consider a pure bipartite two-photon state

$$|\mathbb{II}\rangle = \sum_{mn} c(\omega_m, \omega_n) |1 : \omega_m, \alpha; 1 : \omega_n, \beta\rangle, \quad (6.40)$$

which can also be rewritten in the Schmidt decomposition form

$$|\mathbb{II}\rangle = \sum_m \sqrt{p_m} \hat{a}_{\phi_m}^\dagger \hat{b}_{\psi_m}^\dagger |0, \alpha; 0, \beta\rangle, \quad (6.41)$$

Now the annihilation and creation operators $\hat{\mathbf{a}}, \hat{\mathbf{b}}, \hat{\mathbf{a}}_+, \hat{\mathbf{b}}_+$ are corresponding to the mode functions that are in the Schmidt basis, then the matrix

$$\mathcal{M} = \langle : \hat{\mathbf{a}}_+ \hat{\mathbf{a}}^T \otimes \hat{\mathbf{b}} \hat{\mathbf{b}}_+^T : \rangle = \begin{pmatrix} A_{11} & \cdots & A_{1N} \\ \vdots & \ddots & \vdots \\ A_{N1} & \cdots & A_{NN} \end{pmatrix}, \quad (6.42)$$

whose element satisfies $(A_{ij})_{mn} = \sqrt{p_m p_n} \delta_{in} \delta_{jm}$ and the $\sqrt{p_m}, \sqrt{p_n}$ are Schmidt coefficients. The determinant of the matrix \mathcal{M} reads

$$\det \mathcal{M} = (-1)^{N(N-1)/2} \left(\prod_j p_j \right)^N, \quad (6.43)$$

which can be negative for some integer number N . By direct calculations, the eigenvalues are

$$\{p_1, p_2, \dots, p_N; \pm\sqrt{p_1 p_2}, \dots, \pm\sqrt{p_1 p_N}; \dots; \pm\sqrt{p_{N-1} p_N}\} \quad (6.44)$$

That is, one explicitly finds some negative eigenvalues which shows that the state have sub-Poissonian photon distribution. So such two-photon states are classical. In the first part of this section, we have found that the Schmidt coefficients are increasing functions of the cooperativity parameter, hence, if the state is more entangled, one has larger cooperativity parameter, and larger Schmidt coefficients, then smaller negative eigenvalue of the Mandel matrix. Consequently, the state itself is certainly much more nonclassical.

This conclusion also applies when one considers a counterpart

$$|z : \alpha\beta\rangle = \sum_{mn} c(\omega_m, \omega_n) |z : \omega_m, \alpha; z : \omega_n, \beta\rangle, \quad (6.45)$$

which is superposed by coherent states, providing the field amplitude z is large, $|z| \gg 1$.

B.3 Quantum discord

Quantum discord is a well-known measure for quantum correlations. It has some variants, such as the measurements induced disturbance (MID). The quantity of quantum discord comes from the different value of two definitions of mutual information which are equivalent in classical statistics.

B.3.1 Quantum discord of a bipartite diagonal density matrix

Let's first consider a generic diagonal density matrix $\Lambda_{\alpha\beta}$ of a two-photon state, whose elements come from a normalized bivariate distribution,

$$\Lambda_{\alpha\beta} = \sum_{kq} p_{kq} |1 : \omega_k, \alpha; 1 : \omega_q, \beta\rangle \langle 1 : \omega_k, \alpha; 1 : \omega_q, \beta|. \quad (6.46)$$

The two reduced density matrices of subsystems are

$$\Lambda_\alpha = \sum_k \eta_k |1 : \omega_k, \alpha\rangle \langle 1 : \omega_k, \alpha|; \quad \Lambda_\beta = \sum_q \zeta_q |1 : \omega_q, \beta\rangle \langle 1 : \omega_q, \beta|, \quad (6.47)$$

with $\eta_k = \sum_q p_{kq}$, $\zeta_q = \sum_k p_{kq}$. Since all the three density matrices are diagonal, von Neumann entropies can be calculated by their distribution coefficients as

$$S_{\alpha\beta} = - \sum_{kq} p_{kq} \log p_{kq}; \quad S_\alpha = - \sum_k \eta_k \log \eta_k; \quad S_\beta = - \sum_q \zeta_q \log \zeta_q. \quad (6.48)$$

where we have hidden the base "2" in the logarithm function. The total mutual information $I(\alpha : \beta)$ is known because of Eq. (1.56).

Now let's consider a special POVM on the subsystem α :

$$E_m^\mu = |1 : \omega_m, \mu\rangle \langle 1 : \omega_m, \mu|, \quad m \in \mathbb{N}; \quad \mu = \alpha, \beta. \quad (6.49)$$

According to Born's rule, the probability to have a non-vanishing outcome of measurement E_m^α is then given by

$$x_m = \text{Tr}(|1 : \omega_m, \alpha\rangle \langle 1 : \omega_m, \alpha| \Lambda_{\alpha\beta}) = \sum_q p_{mq} = \eta_m, \quad (6.50)$$

and after the measurement, the final state now becomes

$$\Lambda_{\beta|m} = \text{Tr}_\alpha[E_m^\alpha \Lambda_{\alpha\beta}] / x_m = \sum_q \frac{p_{mq}}{\sum_n p_{mn}} |1 : \omega_q, \beta\rangle \langle 1 : \omega_q, \beta|. \quad (6.51)$$

The von Neumann entropy of such a conditional state is indeed a Shannon entropy,

$$S_{\beta|m} = - \sum_q \frac{p_{mq}}{x_m} \log \frac{p_{mq}}{x_m} = \log x_m - \sum_q \frac{p_{mq}}{x_m} \log p_{mq}, \quad (6.52)$$

and therefore the conditional entropy which is an average over all the conditional states $\Lambda_{\beta|m}$, yields

$$S_{\beta|\{E_m^\alpha\}} = \sum_m x_m S_{\beta|m} = S_{\alpha\beta} - S_\alpha. \quad (6.53)$$

The above equation suggests that the classical information in this state is not less than the total information since

$$J_{\beta|\alpha} \geq S_{\beta} - S_{\beta|\{E_m^{\alpha}\}} = S_{\alpha} + S_{\beta} - S_{\alpha\beta} = I(\alpha : \beta). \quad (6.54)$$

That is, the quantum discord defined as $\mathcal{D}_{\alpha\beta} = I_{\alpha\beta} - J_{\beta|\alpha}$ is no greater than 0. On the other hand, it is universally true that quantum discord is non-negative, $\mathcal{D}_{\alpha\beta} \geq 0$ [Modi2012]. Then the only possibility is

$$\mathcal{D}_{\alpha\beta} = 0, \quad I_{\alpha\beta} = J_{\beta|\alpha}. \quad (6.55)$$

That is, the quantum discord is zero and the amount of classical correlations are equal to the total correlations in such diagonal state.

Bearing in mind that the relation between total correlations, quantum correlations \mathcal{Q} and classical correlations is sub-additive [Groisman2005] in general,

$$\mathcal{Q} + J_{\beta|\alpha} \geq I(\alpha : \beta) \Leftrightarrow \mathcal{Q} \geq \mathcal{D}_{\alpha\beta}. \quad (6.56)$$

That is, even if a state has vanishing discord, it can still have quantum correlations.

We also noticed that the diagonal density matrix can be expressed as

$$\Lambda = \sum_{kq} p_{kq} E_k^{\alpha} \otimes E_q^{\beta}, \quad (6.57)$$

so it is a classical state [Luo2008, Li2008](only with respect to the statistics on the bivariate distribution), and the mutual information $J_{\beta|\alpha}$ is regarded as a measure of classical correlations.

B.3.2 Pure states

The von Neumann entropy of a pure state is 0, therefore the total mutual information is

$$I(\alpha : \beta) = S_{\beta} + S_{\alpha} = 2S_{\beta}, \quad (6.58)$$

The classical mutual information $J_{\beta|\alpha}$,

$$J_{\beta|\alpha} = S_{\beta}, \quad (6.59)$$

is achieved when a POVM with its element $E_m^{\alpha} = |f_m^{\alpha}\rangle\langle f_m^{\alpha}|$ is performed. The quantum discord is just the entropy of a subsystem

$$\mathcal{D}_{\alpha\beta} = J_{\beta|\alpha} = - \sum_n \lambda_n \log \lambda_n. \quad (6.60)$$

where λ_n are the square of the Schmidt coefficients in (1.48). We conclude that the measure of quantum correlations is equal to the measure of classical correlation in a generic pure state. So, if there are more bipartite quantum correlations in a pure state, there should be more classical correlations and vice versa. Together with the vanishing discord of diagonal density matrices, when the conclusions are applied to our results in Chap. 4, from the point of view of information scientists, we have:

The enhancement of 2P2A transition probability does not rely on the quantum correlations of a state.

Anyway, we are still interested in the classical correlations in the pure state in which its measure is exactly equal to the quantum discord.

In the case of type II SPDC two-photon state,

$$\begin{aligned} \mathcal{D}_{\alpha\beta}^{|pdc\rangle} &= - \sum_n [(1-u^2)u^{2n}] \log[(1-u^2)u^{2n}] \\ &= - \frac{u^2}{1-u^2} \log u^2 - \log(1-u^2). \end{aligned} \quad (6.61)$$

According to the definition of u in Eq. (6.17), the measure of classical correlations reads

$$\mathcal{D}_{\alpha\beta}^{|pdc\rangle} = \frac{1}{2} [(\kappa_{pdc} + 1) \log(\kappa_{pdc} + 1) - (\kappa_{pdc} - 1) \log(\kappa_{pdc} - 1)] - 1. \quad (6.62)$$

When the cooperativity parameter becomes large, the discord has a simpler expression

$$\mathcal{D}_{\alpha\beta}^{|pdc\rangle} \simeq \log(e\kappa_{pdc}/2). \quad (6.63)$$

It is a monotonic increasing function of the cooperativity parameter κ_{pdc} . See Fig 6.3 the two curves.

In any pure state, this discord is also a measure of the classical frequency anti-correlations because it reveals the classical correlations and κ_{pdc} represents the frequency anti-correlations. So the enhancement of 2P2A transition probability is determined by the classical frequency anti-correlations, from the viewpoint of the quantum information scientists.

B.3.3 Conclusions

In the whole section, we have discussed some quantumness of the states that may be relevant to the physical origins of the enhancement of 2P2A transition probabilities in Chap. 4. We found that the quantumness of a state that is related to entanglement, separability, negativity, geometric quantum discord as well as quantum correlations, is irrelevant to the enhancement of transition probability. However, whether the physical origins are related to the quantumness that breaks down the PPT criterion or Simon's criterion is still not clear. That is, though it seems that the enhancement of 2P2A transition probability does not have a quantum origin, it is still not completely verified. Therefore, some measurement-based criteria will be discussed in the frame frequency-time Wigner distribution in the following section.

From the discussions on quantum discord (quantum correlations), we found that in any pure state, the amount of classical correlations is equal to the one of quantum correlations, while in its completely dephased state, all the correlations are classical, no quantum correlations can be found. The common correlations in both pure or mixed states is of course of interest to be explored.

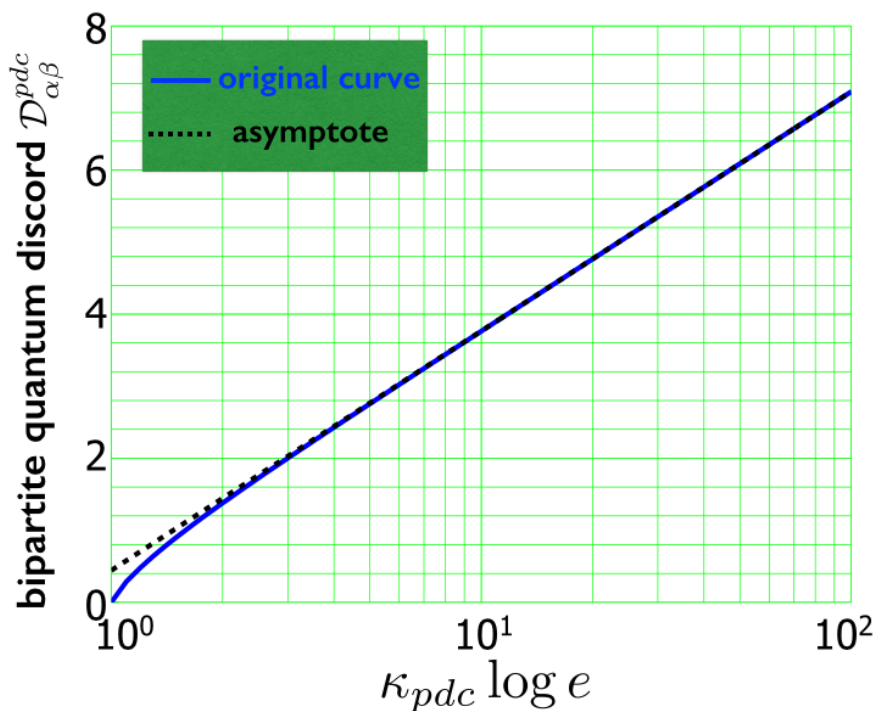


Figure 6.3: The relation between quantum discord (or the amount of classical correlations) and cooperativity parameter in the parametric down converted two-photon state. The quantities of $\kappa_{pdc} \log e$ in x -axis is given in logarithm form. One can see from the curve that with the increasing of cooperativity parameter κ_{pdc} , $D_{\alpha\beta}^{pdc}$ eventually turns out to be its asymptote, a logarithm function

C Characterization using the time-frequency Wigner-Ville distribution of a quantum system

An alternative way to describe a quantum state of a system is to use its quasi-probability distribution, so-called Wigner function, in the corresponding phase space. Such a way of description, which was originally introduced in quantum mechanical context for statistical equilibrium by Wigner in 1932 [Wigner1932] and which is really intuitive as it provides a quite similar way to calculate the statistical quantities of the operators that act on the state, has then been developed as a new and popular formulation of quantum mechanics which is equivalent to other formulations [Styer2002] such like matrix mechanics, wave mechanics and path-integral functional theory.

The quadrature arguments are induced by the field observables which are pairwise conjugate operators. Therefore, the conventional Wigner function is defined by the generalized canonical coordinates. Though the Wigner function was firstly introduced in quantum physics, its first triumph was in signal analysis since Ville's work [Ville1948] in 1948 on time-dependent signal field $E(t)$. Consequently, such a function with respect to the time and frequency arguments is sometimes named as Wigner-Ville distribution

of classical signals. When dealing with a running signal which contains both frequency and time information, Ville proposed to use a characteristic two variable kernel function, then followed by Moyal [Moyal1949] with non-commutative property between time and frequency. In ultrafast optics, such distribution was introduced for classical pulses with another name *chronocyclic* Wigner function [Paye1992] to highlight the different arguments from the conventional ones.

C.1 Quantum Wigner-Ville distribution

The quantum Wigner-Ville distribution of a quantum state of the electromagnetic field is introduced as

$$\mathcal{V}(t, \omega) = (2\pi)^{-1} \int ds e^{i\omega s} \langle \hat{E}^{(+)\dagger}(t - s/2) \hat{E}^{(+)}(t + s/2) \rangle, \quad (6.64)$$

where $\langle \cdot \rangle$ represents the quantum mechanical average. This function describes the time and frequency dependence of the distribution of a photon detected at a same point.

When we consider a bipartite state, sometimes the corresponding field can be separated because of different polarizations or central frequencies by polarized beam splitters or dichroic mirrors. We denote the two separated fields $\hat{E}_\alpha, \hat{E}_\beta$ with subscripts α, β . Then we can introduce 2×2 dimensional Wigner-Ville distribution

$$\mathcal{V}_2(\mathbf{t}, \boldsymbol{\omega}) = (2\pi)^{-2} \int d^2\mathbf{s} e^{i\boldsymbol{\omega}^T \mathbf{s}} \langle \hat{\mathcal{O}}(\mathbf{t}, \mathbf{s}) \rangle, \quad (6.65)$$

with 2 dimensional vectors $\mathbf{t} = (t_a, t_b)^T, \mathbf{s} = (s_a, s_b)^T, \boldsymbol{\omega} = (\omega_a, \omega_b)^T$ as well as the kernel

$$\hat{\mathcal{O}}(\mathbf{t}, \mathbf{s}) \equiv \hat{E}_\alpha^{(+)\dagger}(t_a - s_a/2) \hat{E}_\alpha^{(+)}(t_a + s_a/2) \hat{E}_\beta^{(+)\dagger}(t_b - s_b/2) \hat{E}_\beta^{(+)}(t_b + s_b/2), \quad (6.66)$$

that describes the temporal behaviors of photons detected at two different points. Here we have hidden the dependence of the 2 locations in a same z -plane where we detect the photons.

A Wigner-Ville distribution with higher dimensions can be introduced in a similar way.

C.1.1 A variant: chronocyclic Wigner distribution \mathcal{W}

In Heisenberg picture, the field operator $\hat{E}_\mu(t), \mu = \alpha, \beta$, can be written in following form,

$$\hat{E}_\mu^{(+)}(t) \propto i \int d\omega_\mu \mathcal{E}_{\omega_\mu} \hat{a}_\mu(\omega_\mu) e^{-i\omega_\mu t}, \quad (6.67)$$

with nonvanishing commutation relations $[\hat{a}_\mu(\omega), \hat{a}_\nu^\dagger(\omega')] = c\delta_{\mu\nu}\delta(\omega - \omega')$ where c is the velocity of light in vacuum. The monochromatic field units \mathcal{E}_{ω_μ} is a slowly varying quantity with respect to frequency and we treat it as a constant number \mathcal{E}_μ . The expectation value in Eq.(6.66) is

$$\langle \hat{\mathcal{O}}(\mathbf{t}, \mathbf{s}) \rangle = \langle \otimes_\mu [\iint d\omega_\mu d\omega'_\mu \mathcal{E}_\mu^2 \hat{a}_\mu^\dagger(\omega'_\mu) \hat{a}_\mu(\omega_\mu) e^{-i(\omega_\mu + \omega'_\mu)s_\mu/2} e^{i(\omega'_\mu - \omega_\mu)\tau_\mu}] \rangle. \quad (6.68)$$

According to the definition of the Dirac δ function, one has

$$\mathcal{V}_2(\boldsymbol{\tau}, \boldsymbol{\omega}) = 2^2 \int d^2\boldsymbol{\omega}' \langle \Pi_\mu \mathcal{E}_\mu^2 [\hat{a}_\mu^\dagger(2\omega_\mu - \omega'_\mu) \hat{a}_\mu(\omega'_\mu)] \rangle e^{2i(\boldsymbol{\omega} - \boldsymbol{\omega}')^T \boldsymbol{\tau}}. \quad (6.69)$$

Now let's consider a special photon state that has only one photon in each subsystem, its density matrix reads

$$\rho = \int d^2\omega' d^2\omega'' p(\omega', \omega'') |1 : \omega'\rangle \langle 1 : \omega''|, \quad (6.70a)$$

$$|1 : \omega\rangle = |1 : \omega_a, \alpha\rangle \otimes |1 : \omega_b, \beta\rangle. \quad (6.70b)$$

Only when all the annihilation operator acting on the correct frequency modes, the average in Eq. (6.69) has non-vanishing contribution, and after such operation, the output state turn out to be vacuum: $[\prod_\mu \hat{a}_\mu(\omega'_\mu)] |1 : \omega'\rangle = |0\rangle$.

$$\begin{aligned} \mathcal{V}_2(\boldsymbol{\tau}, \boldsymbol{\omega}) &= 2^2 (\Pi_\nu \mathcal{E}_\nu^2) \int d^2\omega' p(\omega', 2\boldsymbol{\omega} - \omega') \\ &\quad e^{2i(\boldsymbol{\omega} - \omega')^T \boldsymbol{\tau}} \langle 1 : 2\boldsymbol{\omega} - \omega' | \prod_\mu \hat{a}_\mu^\dagger(2\omega_\mu - \omega'_\mu) \hat{a}_\mu(\omega'_\mu) |1 : \omega'\rangle \\ &= 2^2 (\Pi_\nu \mathcal{E}_\nu^2) \int d^2\omega' p(\omega', 2\boldsymbol{\omega} - \omega') e^{2i(\boldsymbol{\omega} - \omega')^T \boldsymbol{\tau}}. \end{aligned} \quad (6.71)$$

The two frequency vectors in the inner product on the last line of the above equation can be rewritten as $2\boldsymbol{\omega} - \omega' = \boldsymbol{\omega} + (\boldsymbol{\omega} - \omega')$ and $\omega' = \boldsymbol{\omega} - (\boldsymbol{\omega} - \omega')$, by changing the integral variables $\boldsymbol{\nu} = 2(\boldsymbol{\omega} - \omega')$, one has

$$\mathcal{V}_2(\boldsymbol{\tau}, \boldsymbol{\omega}) = \mathcal{E}_\alpha^2 \mathcal{E}_\beta^2 \int d^2\nu p(\boldsymbol{\omega} - \boldsymbol{\nu}/2, \boldsymbol{\omega} + \boldsymbol{\nu}/2) e^{i\boldsymbol{\nu}^T \boldsymbol{\tau}}, \quad (6.72)$$

$$= \mathcal{E}_\alpha^2 \mathcal{E}_\beta^2 \int d^2\nu \langle 1 : \boldsymbol{\omega} - \boldsymbol{\nu}/2 | \rho | 1 : \boldsymbol{\omega} + \boldsymbol{\nu}/2 \rangle e^{i\boldsymbol{\nu}^T \boldsymbol{\tau}}. \quad (6.73)$$

The normalized Wigner-Ville distribution is

$$\mathcal{W}(\boldsymbol{\tau}, \boldsymbol{\omega}) = (2\pi)^{-2} \int \langle 1 : \boldsymbol{\omega} - \boldsymbol{\nu}/2 | \rho | 1 : \boldsymbol{\omega} + \boldsymbol{\nu}/2 \rangle e^{i\boldsymbol{\nu}^T \boldsymbol{\tau}} d^2\nu. \quad (6.74)$$

It is usually called: chronocyclic Wigner distribution. This expression has the same form as the definition of the conventional Wigner function, but it comes from the quantum Wigner-Ville distribution. For any other states, to introduce a time-frequency Wigner function by analogy with the conventional one can be problematic mainly because: 1. there is no definition of time operator; 2. there exists states, for example the completely dephased states (the statistical mixture), that are time-independent and therefore the Fourier integral transform does not apply.

It is worth mentioning that the quantum Wigner-Ville distribution of a state, that is an tensor product of two monochromatic coherent states which are Gaussian states in the phase-space that describes the quadratures, has some negative-value-zones [Cohen1989]. That is, the negative value of the quantum Wigner-Ville function is not an intrinsic character of non-classicality. Owing to this fact, I will only consider the variances of frequencies and times.

C.2 Bipartite Wigner distributions in continuous variable regime

Now let's consider bipartite two-photon states which have infinitely many frequency modes. Two types of states of our interest here are: 1. pure states $|\psi_{\alpha\beta}\rangle = \sum_{kq} c_{kq} |1 : \omega_k; 1 : \omega_q\rangle$,

and 2. a second type of the mixed type $\Lambda = \sum_{kq} |c_{kq}|^2 |1; \omega_k; 1 : \omega_q\rangle \langle 1 : \omega_k; 1 : \omega_q|$. We will treat them as bivariate continuous frequency functions.

We explicitly denote $\boldsymbol{\omega}, \boldsymbol{\tau}$ in Eq. (6.74) as $(\omega_a, \omega_b)^T, (\tau_a, \tau_b)^T$ where subscripts a, b are related to the parts α and β respectively.

The key elements of Wigner-Ville distribution or chronocyclic Wigner distribution are the following expectation:

$$\langle 1 : \omega_m; 1 : \omega_n | \hat{a}_{a,p}^\dagger \hat{a}_{a,q} \hat{a}_{b,r}^\dagger \hat{a}_{b,s} | 1 : \omega_j; 1 : \omega_k \rangle = \delta_{mp} \delta_{qj} \delta_{nr} \delta_{sk}, \quad (6.75)$$

according to formula (6.66). It imposes four constraints of the states and leads to a generic expression of chronocyclic Wigner distribution,

$$\mathcal{W}(\boldsymbol{\tau}, \boldsymbol{\omega}) \propto \iint_{\mathbb{R}} d\omega_m d\omega_n \langle 1 : \omega_a - \omega_m; 1 : \omega_b - \omega_n | \rho | 1 : \omega_a + \omega_m; 1 : \omega_b + \omega_n \rangle e^{2i\omega_m \tau_a + 2i\omega_n \tau_b}. \quad (6.76)$$

We will first take a closer look at the shapes of diagonal mixed states and pure states.

- diagonal density matrices.

If ρ is a diagonal matrix Λ ,

$$\Lambda = \iint_{\mathbb{R}} d\omega_m d\omega_n |c(\omega_m, \omega_n)|^2 |1 : \omega_m; 1 : \omega_n\rangle \langle 1 : \omega_m; 1 : \omega_n|,$$

then Eq. (6.75) imposes that $\omega_a - \omega_m = \omega_a + \omega_m$ and $\omega_b - \omega_n = \omega_b + \omega_n$. The two equalities imply that $\omega_m = \omega_n = 0$ and reveal the fact that there is no quantum interference between any frequency modes. Therefore the time-dependence introduced via the exponential function $e^{2i\omega_m \tau_a + 2i\omega_n \tau_b}$ disappears, resulting in the following frequency-dependent distribution

$$\mathcal{W}^\Lambda(\boldsymbol{\omega}) \propto |c(\omega_a, \omega_b)|^2. \quad (6.77)$$

So, for any diagonal density matrix, the chronocyclic Wigner distribution is positive and time-independent, and the temporal standard deviations are regarded as infinitely large. So, any time-frequency deviation-product of a poly-chromatic state should be infinitely large.

- pure states.

The density matrix ρ of a pure two-photon state is given by $|\mathbb{II}\rangle \langle \mathbb{II}|$, where

$$|\mathbb{II}\rangle = \iint_{\mathbb{R}} d\omega_m d\omega_n c(\omega_m, \omega_n) |1 : \omega_m; 1 : \omega_n\rangle.$$

Accordingly, the chronocyclic Wigner distribution follows

$$\mathcal{W}^{|\mathbb{II}\rangle}(\boldsymbol{\tau}, \boldsymbol{\omega}) \propto \iint_{\mathbb{R}} d\omega_m d\omega_n c(\omega_a - \omega_m, \omega_b - \omega_n) c^*(\omega_a + \omega_m, \omega_b + \omega_n) e^{2i\omega_m \tau_a + 2i\omega_n \tau_b}. \quad (6.78)$$

Such a distribution admits both time and frequency dependence.

Consider two concrete examples: 1. the chronocyclic Wigner distribution of cascade two-photon state, and 2. the chronocyclic Wigner distribution of a type II two-photon state.

For cascade two-photon state, its coefficients $c(\omega_m, \omega_n)$ are defined in Eq. (4.16), according to the formula (6.78), the chronocyclic Wigner distribution yields

$$\begin{aligned} \mathcal{W}^{|cas\rangle}(\boldsymbol{\tau}, \boldsymbol{\omega}) &= \frac{4}{\pi^2} \gamma_\alpha \gamma_\beta u(\tau_a) u(\tau_b - \tau_a) e^{-2\gamma_\alpha \tau_a - 2\gamma_\beta (\tau_b - \tau_a)} \\ &\times \frac{\sin 2(\omega_a + \omega_b - \omega_\alpha - \omega_\beta) \tau_a}{\omega_a + \omega_b - \omega_\alpha - \omega_\beta} \frac{\sin 2(\omega_b - \omega_\beta) (\tau_b - \tau_a)}{\omega_b - \omega_\beta}. \end{aligned} \quad (6.79)$$

where $u(t)$ is the Heaviside function which attains the value of 1 when $t \geq 0$, and the value of 0 else-wise.

Let us stress that, owing to the sinusoidal functions, these Wigner distributions take negative values, which is an attribute of a non-classical state, as expected from a two-photon pure state.

The marginal distributions on frequencies and times are

$$\mathcal{W}_{freq}^{|cas\rangle}(\omega_a, \omega_b) = \frac{\gamma_\alpha \gamma_\beta / \pi^2}{[(\omega_a + \omega_b - \omega_\alpha - \omega_\beta)^2 + \gamma_\alpha^2][(\omega_b - \omega_\beta)^2 + \gamma_\beta^2]}, \quad (6.80)$$

$$\mathcal{W}_{time}^{|cas\rangle}(\tau_a, \tau_b) = 4\gamma_\alpha \gamma_\beta u(\tau_a) u(\tau_b - \tau_a) \exp[-2\gamma_\alpha \tau_a - 2\gamma_\beta (\tau_b - \tau_a)]. \quad (6.81)$$

Recalling the chronocyclic Wigner distribution of the diagonal density matrices, we find that the marginal distribution on frequency is exactly equal to the chronocyclic Wigner distribution of the corresponding diagonal density matrix. The two marginal distributions display the frequency anticorrelations and temporal correlations respectively. See Figure 6.4.

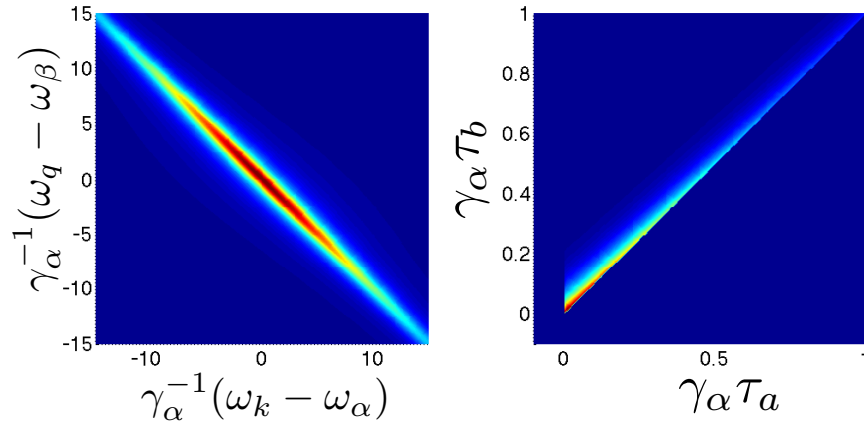


Figure 6.4: The marginal distribution distribution of the Wigner function of the cascade two-photon state. The left (right) graphic represents the reduced frequency (time) Wigner function $\mathcal{W}_{freq}^{|cas\rangle}(\omega_a, \omega_b)$ ($\mathcal{W}_{time}^{|cas\rangle}(\tau_a, \tau_b)$). In both graphics, we set $\Gamma = 10\sigma$.

Now let's consider the type II SPDC two-photon state with coefficients

$$c_{kq} \propto e^{-\frac{(\omega_k + \omega_q - \omega_p)^2}{2\sigma^2} + i(\omega_k + \omega_q - \omega_p)t_0 - \frac{(\omega_q - \omega_\beta)^2}{2\Gamma^2}}, \quad (6.82)$$

where ω_p is the total energy of the two-photon state and σ, Γ are two frequency widths of the pump field and newly generated field. Its chronocyclic Wigner distribution which can

be calculated via Gaussian integral, reads

$$\mathcal{W}^{|pdc\rangle} = \pi^{-2} e^{-\frac{(\omega_a + \omega_b - \omega_p)^2}{\sigma^2} - \Gamma^2 (\tau_b - \tau_a)^2} e^{-\frac{(\omega_b - \omega_\beta)^2}{\Gamma^2} - \sigma^2 (\tau_a - t_0)^2}. \quad (6.83)$$

This distribution is a product of four Gaussian distributions, and its value is positive everywhere. The two marginal distributions on frequencies and times are

$$\mathcal{W}_{freq}^{|pdc\rangle}(\omega_a, \omega_b) = \pi^{-1} e^{-\frac{(\omega_a + \omega_b - \omega_p)^2}{\sigma^2}} e^{-\frac{(\omega_b - \omega_\beta)^2}{\Gamma^2}}; \quad (6.84)$$

$$\mathcal{W}_{time}^{|pdc\rangle}(\tau_a, \tau_b) = \pi^{-1} e^{-\Gamma^2 (\tau_b - \tau_a)^2} e^{-\sigma^2 (\tau_a - t_0)^2}; \quad (6.85)$$

$$\mathcal{W}_{tf}^{|pdc\rangle}(\omega_a, \tau_a) = \frac{\sigma}{\pi \sqrt{\sigma^2 + \Gamma^2}} e^{-\frac{(\omega_a - \omega_\alpha)^2}{\sigma^2 + \Gamma^2}} e^{-\sigma^2 (\tau_a - t_0)^2}. \quad (6.86)$$

See the marginal distributions of the type II SPDC two-photon state in Figure 6.5.

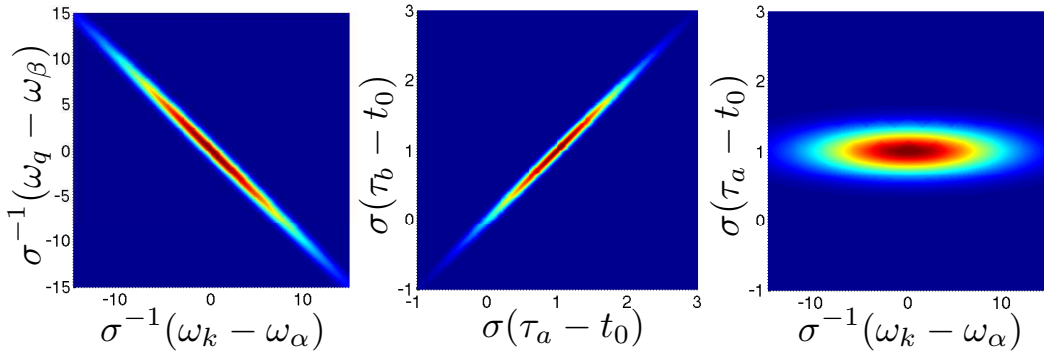


Figure 6.5: The marginal distributions of the Wigner function of the SPDC two photon state. The graphic on the left side, in the middle, and on the right side represent the reduced Wigner function $\mathcal{W}_{freq}^{|pdc\rangle}(\omega_k, \omega_q)$, $\mathcal{W}_{time}^{|pdc\rangle}(\tau_a, \tau_b)$, $\mathcal{W}_{tf}^{|pdc\rangle}(\omega_k, \tau_b)$. In all the three graphics, we set $\Gamma = 10\sigma$ and $\sigma t_0 = 1$.

Once again, we find the marginal distribution $\mathcal{W}_{freq}^{|pdc\rangle}(\omega_a, \omega_b)$ is the same as the chronocyclic Wigner distribution of the corresponding diagonal density matrix.

This conclusion turns out to be correct in any case, because if one integrates the temporal variables in Eq. (6.78), one obtains two Dirac δ functions. They impose the expression of the marginal distribution as the chronocyclic Wigner distribution of the respective diagonal density matrix.

From the marginal distribution of frequencies both in Figure 6.4 and Figure 6.5, one sees obvious band-like frequency distributions. Intuitively, correlations is sometimes roughly represented by a ratio between the maximum diameter and the minimum one in an (effective) area. The larger the ratio is, the stronger the correlations are. In the two figures, the minimum diameters are found along the direction of line $\omega_a - \omega_b = 0$ and their magnitudes are at the order of γ_α or σ respectively; the maximum ones are found along the direction of line $\omega_a + \omega_b = \omega_p$ and the magnitude are at several γ_β or Γ respectively. Since $\Gamma_\beta/\sigma \gg 1, \gamma_\beta/\gamma_\alpha \gg 1$, the frequency (anti-) correlations shown in both figures are quite strong.

C.2.1 Apparent "violation" of the Heisenberg inequality

The famous EPR paper [Einstein1935] showed that there exist states with strong correlations which "violate" the Heisenberg inequality. They can be characterized by conditional variances the product of which can be smaller than the limit ascertained by Heisenberg [Reid2009]. Let us apply such an analysis to the correlated states considered here.

Consider a type II two-photon state $|\text{II}\rangle = \sum_{kq} c_{kq} |1 : \omega_k, \alpha; 1 : \omega_q, \beta\rangle$ where the coefficients c_{kq} reads

$$c_{kq} \propto e^{-\frac{(\omega_k + \omega_q - \omega_p)^2}{2\sigma^2} + i(\omega_k + \omega_q - \omega_p)t_0 - \frac{(\omega_q - \omega_\beta)^2}{2\Gamma^2}}. \quad (6.87)$$

where ω_p is the central frequency of the pump field whose bandwidth is σ , Γ is the bandwidth caused by phase mismatch, t_0 is the typical arrival time. The corresponding chronocyclic Wigner function follows

$$\mathcal{W}^{|pdc\rangle} = \pi^{-2} e^{-\frac{(\omega_a + \omega_b - \omega_p)^2}{\sigma^2} - \Gamma^2(\tau_b - \tau_a)^2} e^{-\frac{(\omega_b - \omega_\beta)^2}{\Gamma^2} - \sigma^2(\tau_a - t_0)^2}. \quad (6.88)$$

Then one can calculate the conditioned mean value and momentum of a variable $A(\omega_a, \tau_a)$ by

$$\overline{A}_{\text{cond}} = \iint d\tau_a d\omega_a A(\omega_a, \tau_a) \mathcal{W}^{|pdc\rangle}(\omega_a, \tau_a, \omega_b, \tau_b); \quad (6.89a)$$

$$\overline{A^2}_{\text{cond}} = \iint d\tau_a d\omega_a A^2(\omega_a, \tau_a) \mathcal{W}^{|pdc\rangle}(\omega_a, \tau_a, \omega_b, \tau_b); \quad (6.89b)$$

and the variance of $(\Delta A)_{\text{cond}} = \sqrt{\overline{A^2}_{\text{cond}} - \overline{A}_{\text{cond}}^2}$ according to the definition. The unconditioned value quantities are given by the integration on the other part,

$$\overline{A}_{\text{uncond}} = \iint d^2\boldsymbol{\tau} d^2\boldsymbol{\omega} A(\omega_a, \tau_a) \mathcal{W}^{|pdc\rangle}(\omega_a, \tau_a, \omega_b, \tau_b); \quad (6.90a)$$

$$\overline{A^2}_{\text{uncond}} = \iint d^2\boldsymbol{\tau} d^2\boldsymbol{\omega} A^2(\omega_a, \tau_a) \mathcal{W}^{|pdc\rangle}(\omega_a, \tau_a, \omega_b, \tau_b); \quad (6.90b)$$

hence the unconditioned variance $(\Delta A)_{\text{uncond}} = \sqrt{\overline{A^2}_{\text{uncond}} - \overline{A}_{\text{uncond}}^2}$.

By direct calculations, the unconditioned product of the variances reads $(\Delta\omega_a \Delta\tau_a)_{\text{uncond}} = \kappa_{pdc}/2 \geq 1/2$, which does not show any violation of Heisenberg inequality, as expected. But the conditioned quantities follow

$$(\overline{\omega_a})_{\text{cond}} = (\omega_p - \omega_b) f(\omega_b, \tau_b); \quad (\overline{\tau_a})_{\text{cond}} = \frac{\Gamma^2 \tau_b + \sigma^2 t_0}{\Gamma^2 + \sigma^2} f(\omega_b, \tau_b); \quad (6.91)$$

$$(\overline{\omega_a^2})_{\text{cond}} = [(\omega_p - \omega_b)^2 + \sigma^2/2] f(\omega_b, \tau_b); \quad (6.92)$$

$$(\overline{\tau_a^2})_{\text{cond}} = \left[\left(\frac{\Gamma^2 \tau_b + \sigma^2 t_0}{\Gamma^2 + \sigma^2} \right)^2 + \frac{1}{2} \frac{1}{\Gamma^2 + \sigma^2} \right] f(\omega_b, \tau_b); \quad (6.93)$$

$$f(\omega_b, \tau_b) = \frac{1}{\pi} \frac{\sigma}{\sqrt{\Gamma^2 + \sigma^2}} e^{-\sigma^2 \Gamma^2 \frac{(\tau_b - t_0)^2}{\Gamma^2 + \sigma^2}} e^{-\frac{(\omega_b - \omega_\beta)^2}{\Gamma^2}}. \quad (6.94)$$

In fact, $f(\omega_b, \tau_b)$ is a marginal Wigner function that represents the time-frequency distribution of the β subsystem. As a result, we have the following two variances

$$(\Delta\omega_a)_{\text{cond}} = \sqrt{[(\omega_p - \omega_b)^2 + \sigma^2/2] f(\omega_b, \tau_b) - (\omega_p - \omega_b)^2 f^2(\omega_b, \tau_b)}; \quad (6.95)$$

$$(\Delta\tau_a)_{\text{cond}} = \sqrt{\left[\left(\frac{\Gamma^2 \tau_b + \sigma^2 t_0}{\Gamma^2 + \sigma^2} \right)^2 + \frac{1}{2} \frac{1}{\Gamma^2 + \sigma^2} \right] f(\omega_b, \tau_b) - \left(\frac{\Gamma^2 \tau_b + \sigma^2 t_0}{\Gamma^2 + \sigma^2} \right)^2 f^2(\omega_b, \tau_b)}. \quad (6.96)$$

Therefore, when $|\omega_p - \omega_b| \gg \Gamma, \sigma\Gamma|\tau_b - t_0| \gg \sqrt{\Gamma^2 + \sigma^2}$, the value of the product

$$(\Delta\omega_a\Delta\tau_a)_{\text{cond}} \simeq f(\omega_b, \tau_b) \sqrt{\left[\left(\frac{\Gamma^2\tau_b + \sigma^2 t_0}{\Gamma^2 + \sigma^2} \right)^2 + \frac{1}{2} \frac{1}{\Gamma^2 + \sigma^2} \right] [(\omega_p - \omega_b)^2 + \frac{\sigma^2}{2}]} \quad (6.97)$$

is much less than 1. When $\sigma/\Gamma \ll 1$, once again, the product reveals the EPR entanglement of the state according to the Duan-Mancini criterion [Duan2000, Mancini2002].

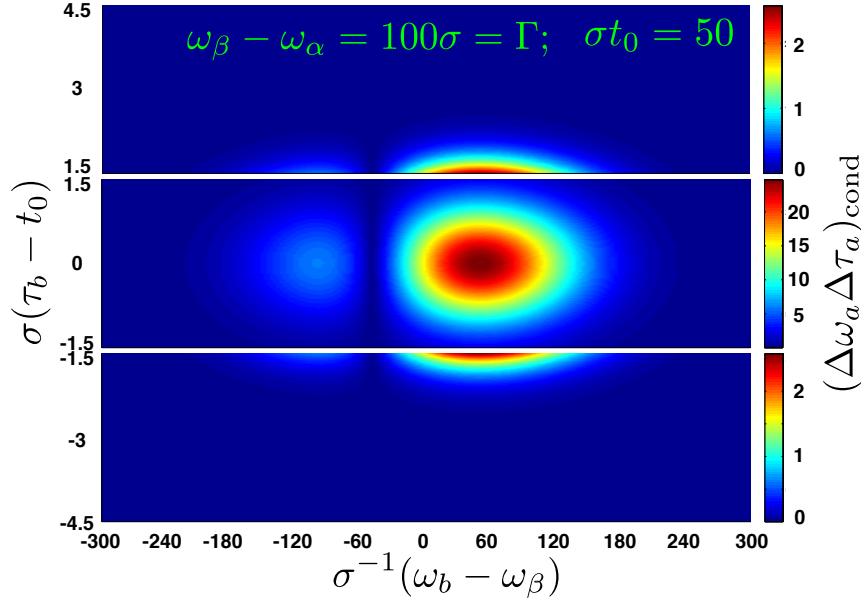


Figure 6.6: The distribution of conditioned value of the product of deviations. The parameters are set as $\omega_\beta - \omega_\alpha = 100\sigma$, $\Gamma = 10\sigma$ and $\sigma t_0 = 50$. The value in most parts of the top and the bottom graphs is much less than 1, which reveals the entanglement of the state.

Therefore, in such a type II SPDC two-photon state, those photons, whose frequencies are far away from their central frequencies, in the pulse are EPR correlated.

Conclusion

Two-photon–two-atom excitation turns out to be an important test bench of quantum optical properties. In this thesis, this subject has been discussed in two cases: 2P2A transition probability with and without atomic interaction. In the absence of atomic interaction, the 2P2A transition probability can be dramatically enhanced by using cascade two-photon state. The cascade two-photon state is a typical intrinsic multimode entangled state. It is a pulsed source which is characterized by frequency anti-correlations. To find out the physical origin of transition enhancement is one of key ingredients of the work in this thesis. The possible candidates such as entanglement, time ordering, temporal correlation have been excluded. We have found that actually the physical origin of enhancement is the frequency anti-correlation. However, the negative eigenvalues of the Mandel matrices of the two-photon states, pure multimode states and their corresponding completely dephased mixed states, exhibit the sub-Poissonian character of photon statistics, which are nonclassical. The classically mixed coherent state, which is regarded as a most classical state (according to all aspects like quantum optical nonclassicality, separability, quantum discord. etc), has a $|z|^4$ scale factor when $|z| \gg 1$, in comparison with the transition probability that is caused by the two-photon dephased state of the same form. Unlike the completely dephased state that can be obtained by averaging from noises, the classically mixed coherent state cannot. Therefore, it is very likely that the anti-correlations necessary for the enhancement of the transition probability are classical.

The 2P2A transition probability can also be greatly enhanced by atomic interaction even when a monochromatic coherent field is applied on the condition that the atomic interaction strength is much larger than atomic linewidths of the excited state of the two atoms. We have found when the monochromatic photon state is highly bunched, the transition probability can also be enhanced. This is very evident for those photon states that have very flat distribution, like a sequence that takes its terms from Riemann $\zeta(x)$ function. By using the SPDC two-photon state, the transition spectrum exhibits a new structure of 2P2A resonance.

The subjects presented in the second chapter are taken from the existing literatures. The contents in later chapters, from chapter 3 to chapter 6, are original to my best of knowledge. The findings on the transition enhancement by various two-photon states in chapter 4 have been published on Physical Review A [Zheng2013]. Another publication on the case with interaction is in preparation.

Appendix

A When can $(\exp(-i\omega t) - 1)/(2i\pi\omega)$ be a good approximation of the delta function?

Let us note $s_t(\omega)$ the function $(\exp(-i\omega t) - 1)/(2i\pi\omega)$. One can also write it as $s_t(\omega) = -\sin \omega t/(2\pi\omega) + i(1 - \cos \omega t)/(2\pi\omega)$. Whereas the real part of $s_t(\omega)$ is a sinc function which tends indeed to a delta function when $t \rightarrow \infty$, the imaginary part, being not a peaked function whose area is constant, is not an approximation of the delta function. So, in general, $s_t(\omega)$ does not tend to the delta distribution when it acts on the general set of integrable functions; however, it can be so on a smaller set of functions. This set includes, for example, all the odd functions in ω , a subset which is not relevant for the present paper. We show in this appendix that $s_t(\omega)$ behaves also as a δ function when it acts on functions which have a Fourier transform which is strictly zero before $t = 0$.

Let us consider a function $F(t)$ that is zero for $t < 0$ and admits a well-behaved Fourier transform $f(\omega)$. Then

$$f(\omega) = \frac{1}{2\pi} \int_{-\infty}^{\infty} dt F(t) e^{i\omega t} = \frac{1}{2\pi} \int_0^{\infty} dt F(t) e^{i\omega t}, \quad (\text{A-1})$$

$$F(t) = \int_{-\infty}^{\infty} d\omega f(\omega) e^{-i\omega t}, \quad (\text{A-2})$$

where $f(\omega)$ is absolutely integrable, which excludes functions such as $1/(\omega + i\gamma)$ from the present discussion. Let us now calculate the integral:

$$I = \int_{-\infty}^{\infty} d\omega \frac{\exp(-i\omega t) - 1}{\omega} f(\omega) = i \int_0^t d\tau \int_{-\infty}^{\infty} d\omega f(\omega) \exp(-i\omega\tau) = i \int_0^t d\tau \theta(\tau) F(\tau) \quad (\text{A-3})$$

Then $I \rightarrow i \int_0^{\infty} d\tau F(\tau) = 2\pi i f(0)$ when $t \rightarrow \infty$. This proves that $s_t(\omega)$ acts as a delta function for the set of functions that have a Fourier transform strictly null for $t < 0$.

B Why do we take $t = L/c$ in the comparison of transition probabilities ?

In order to compare the probabilities of transitions induced by pulsed and cw light in a fair way, we must be careful to take the same amount of energy flow $\mathcal{F}(t)$ on the detecting atoms in both cases. This quantity is nothing else than the integral over time and transverse

section S of the Poynting vector. It is equal to, at a given time t and for a state ρ ,

$$\mathcal{F}(t) = 2\varepsilon_0 c S \int_0^t \text{Tr}[\rho \hat{E}^{+\dagger}(\tau) \hat{E}^+(\tau)] d\tau \simeq \hbar\omega \frac{c}{L} \int_0^t \text{Tr}[\rho \hat{b}^\dagger(\tau) \hat{b}(\tau)] d\tau, \quad (\text{B-1})$$

where $\hat{b}(\tau) = \sum_m \hat{a}_m \exp(-i\omega_m \tau)$ and ω is the mean frequency of the state under consideration.

For any diagonal density matrix Λ , since $\text{Tr}[\Lambda \hat{b}^\dagger(\tau) \hat{b}(\tau)] = 2$ is time independent, one finds a linear relationship between the energy flow and time t ,

$$\mathcal{F}_\Lambda(t) = 2\hbar\omega \frac{ct}{L}. \quad (\text{B-2})$$

For any entangled pure state $|\Psi\rangle = \sum_{kq} c_{kq} |1_k, 1_q\rangle$,

$$\text{Tr}[|\Psi\rangle\langle\Psi| \hat{b}^\dagger(\tau) \hat{b}(\tau)] = \sum_k \left| \sum_q c_{kq} e^{-i\omega_q \tau} \right|^2 + \sum_q \left| \sum_k c_{kq} e^{-i\omega_k \tau} \right|^2. \quad (\text{B-3})$$

The energy flow at time t is

$$\mathcal{F}_\Psi(t) = \int_0^t d\tau \text{Tr}[|\Psi\rangle\langle\Psi| \hat{b}^\dagger(\tau) \hat{b}(\tau)] \approx \int_{-\infty}^t d\tau \text{Tr}[|\Psi\rangle\langle\Psi| \hat{b}^\dagger(\tau) \hat{b}(\tau)], \quad (\text{B-4})$$

when most photons arrive at the detecting atoms after $t = 0$. One assumes that at sufficiently large time t (much greater than the temporal coherence length of the field), the photons in state $|\Psi\rangle$ have fully interacted with the detecting atoms; therefore one extends t to $+\infty$ without introducing notable error. By using the Parseval identity, one has

$$\begin{aligned} \mathcal{F}_\Psi(t) &\approx \hbar\omega \frac{c}{L} \int_{-\infty}^{\infty} d\tau \text{Tr}[|\Psi\rangle\langle\Psi| \hat{b}^\dagger(\tau) \hat{b}(\tau)] \\ &= \hbar\omega \frac{c}{L} \int_{-\infty}^{\infty} d\tau \left[\sum_k \left| \sum_q c_{k,q} e^{-i\omega_q \tau} \right|^2 + \sum_q \left| \sum_k c_{k,q} e^{-i\omega_k \tau} \right|^2 \right] \\ &= \hbar\omega \left[\sum_{kq} |c_{kq}|^2 + \sum_{kq} |c_{kq}|^2 \right] = 2\hbar\omega, \end{aligned} \quad (\text{B-5})$$

as expected. By comparison with Eq. (B-2), one finds that at time $t = L/c$, the energies supplied by the cw field and by the pulse are equal. Under this situation, one can make legitimate comparisons between the corresponding transition probabilities.

C Superposed coherent states

C.1 States that give rise to large field amplitudes

So far we have only considered two-photon states of different shapes, which are all strongly non-classical objects, as they are produced by spontaneous emission or parametric fluorescence which are specifically quantum processes with no classical equivalent. But one can also envision superpositions of two-mode coherent states of the form

$$|QMC\rangle = \sum_{kq} c_{kq} |z : \omega_k\rangle \otimes |z : \omega_q\rangle, \quad (\text{C-1})$$

or mixed states of the form

$$\rho_{CMC} = \sum_{kq} |c_{kq}|^2 |z : \omega_k\rangle\langle z : \omega_k| \otimes |z : \omega_q\rangle\langle z : \omega_q|, \quad (\text{C-2})$$

where $|z : \omega_k\rangle$ is the coherent state $|z\rangle$ in the mode of frequency ω_k , z being the same complex number for all modes. Because both the two macroscopic states whose components are of a product form with two coherent states from different subsystems are either quantum superposed or classically combined, we give names *QMC* and *CMC*.

By using the approximation $\langle z|0\rangle \simeq 0$ valid for $|z| \gg 1$, one has the following relations for transition probability:

$$P_{QMC} = |z|^4 P \simeq P_{CMC} \quad (\text{C-3})$$

where P is the probability (4.2) obtained for two-photon states at time $t = L/c$. Apart from the energy scaling factor $|z|^4$, the states seem much more classical than the previously studied ones in Chap. 4, as the states of ρ_{CMC} can be produced by classical means.

If we restrict ourselves to the case of large $|z|$, then we interpret *QMC* as "quantum mixed coherent states"; *CMC* as "classical mixed coherent states".

Even with large $|z|$, it is not clear at this stage whether the pure state $|QMC\rangle$ can be produced in classical means or not. Before we go further, let first check its multi-modality.

For frequency modes of any two frequencies ω_k, ω_q , the product coherent state $|z : \omega_k; z : \omega_q\rangle$ is indeed an intrinsic single mode state [Treps2005, FabreQO]. Now let's consider a superposition of two states with intrinsic single-modes ψ_1, ψ_2 ,

$$|0 : \psi_1; z : \psi_2\rangle + |z : \psi_1; 0 : \psi_2\rangle. \quad (\text{C-4})$$

We find the rank of induced vectors from the action of the annihilation operators is 2, according to the definition in Chap. 1, this is an intrinsic 2-mode state. Similarly, one can show the state $|QMC\rangle$ is an intrinsic infinitely-many-mode state for a generic expression c_{kq} .

C.2 Positiveness of the Wigner distribution of the state $|QMC\rangle$

If any $|QMC\rangle$ can be produced in classical means, then the quantum state related to the unnormalized state $|z : \omega_1; z : \omega_q\rangle + |z : \omega_2; z : \omega_q\rangle$ can also be produced in classical means. The coherent state in frequency mode of ω_q can be displaced to the origin of phase space by a displacement operator, then we have the following unnormalized state

$$|coco\rangle = (|z : \omega_1, 0 : \omega_2\rangle + |0 : \omega_1, z : \omega_2\rangle) \otimes |0 : \omega_q\rangle. \quad (\text{C-5})$$

For simplicity, we will hide the vacuum state in the frequency mode of ω_q .

Let (x_j, k_j) be the two field quadratures related to the coherent state in the frequency mode of $\omega_j, j = 1, 2$. In phase space, the Wigner function is a tetra-variate function. If the state $|cooc\rangle$ can be produced in classical means, then the Wigner distribution should be positive. So are the marginal distributions. Otherwise, the state $|cooc\rangle$ is really quantum, cannot be produced in any classical means.

A single frequency mode coherent state is a Gaussian state which is completely determined by its first and second momenta. Its projection on the x quadrature [HarocheETQ]

$$\langle x|z\rangle = \pi^{-1/4} e^{-x^2/2 + \sqrt{2}zx - (z^2 + |z|^2)/2}, \quad (\text{C-6})$$

is a Gaussian function. Subsequently, the kernel of our tetra-variate Wigner function is

$$\begin{aligned} & \langle x_1 + \xi_1/2 : \omega_1, x_2 + \xi_2/2 : \omega_2 | cooc \rangle \langle cooc | x_1 - \xi_1/2 : \omega_1, x_2 - \xi_2/2 : \omega_2 \rangle \\ &= \pi^{-1} e^{-|z|^2 - (z^2 + z^{*2})/2 - (x_1^2 + x_2^2) - (\xi_1^2 + \xi_2^2)/4} \\ & \times [e^{\sqrt{2}z(x_1 + \xi_1/2)} + e^{\sqrt{2}z(x_2 + \xi_2/2)}] [e^{\sqrt{2}z^*(x_1 - \xi_1/2)} + e^{\sqrt{2}z^*(x_2 - \xi_2/2)}]. \end{aligned} \quad (\text{C-7})$$

Making Fourier transform to it, we have

$$\begin{aligned} W(x_1, k_1; x_2, k_2) &\propto e^{-(|z_1|^2 + |z_2|^2)} e^{-|z|^2} \left[e^{\sqrt{2}(zz_2^* + z^*z_1)} + \right. \\ & \left. e^{\sqrt{2}(zz_1^* + z^*z_2)} + e^{-|z|^2} (e^{\sqrt{2}(zz_1^* + z^*z_1)} + e^{\sqrt{2}(zz_2^* + z^*z_2)}) \right], \end{aligned} \quad (\text{C-8})$$

where $z_j = x_j + ik_j$. One finds the two complex exponential terms in the square bracket. The sum of the two terms give rise to sinusoidal function, which may lead to a negative value of the tetra-variate Wigner function. For simplicity, let's take z as a real number and then consider the marginal distribution $W_{mrg}(x_1, k_1) = \iint dx_2 dk_2 W(x_1, k_1; x_2, k_2)$ of variables x_1, k_1 . It reads,

$$W_{mrg}(x_1, k_1) \propto e^{-(x_1^2 + k_1^2 + z^2)} \left[1 + e^{-z^2} e^{2\sqrt{2}zx_1} + 2e^{\sqrt{2}zx_1} \cos(\sqrt{2}zk_1) \right]. \quad (\text{C-9})$$

At the point $(x_1 = 0, k_1 = \pi/\sqrt{2}z)$, the value of Wigner function

$$W_{mrg}(0, \pi/\sqrt{2}z) \propto e^{-z^2 - \pi^2/2z^2} [e^{-z^2} - 1] \quad (\text{C-10})$$

is negative since $1 - e^{-z^2} > 0$ for any nonvanishing z . We then conclude that *such state, therefore a generic QMC states, cannot be produced in classical means.*

The negative value of $W_{mrg}(0, \pi/\sqrt{2}z)$, see Figure 7.1. One finds when the value of z becomes very large, the negative value becomes very small, owing to the term in front of the square bracket in Eq. (C-10). In fact, it is also true for the tetra-variate Wigner distribution that all the negative value will be very small with a large z . It is possible to construct a similar shape of the Wigner function whose value is positive everywhere. But we are still not sure whether such an alternative can be produced in classical means or not, because the fact that a positive-everywhere Wigner distribution does not mean that the corresponding state should be classical.

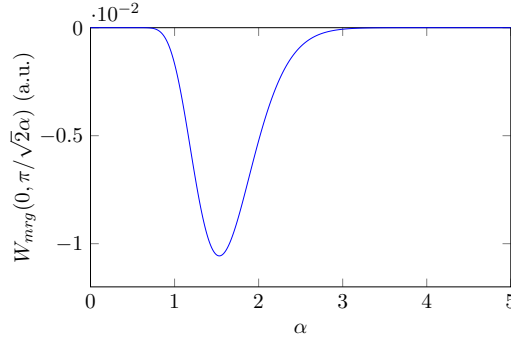


Figure 7.1: Plot of the value of the marginal Wigner function $W_{mrg}(0, \pi/\sqrt{2}z)$ with different value of field amplitude z . The function attains its minimum ($\simeq -1.1 \times 10^{-2}$) around $z = 1.6$. When z becomes very large or very small, the negativeness is not noticeable.

D Possible physical realization of QMC states

To produce a CMC state as defined above is not so difficult. In principle, one can use classical random number generator and two local systems to generate the coherent states[Glauber1963] with classical communications[Werner1989]. But to produce a QMC state, a pure macroscopic state, is not so easy.

D.1 Micro-macro coupling via two-atom interaction

Consider two individual three-level atoms (a) and (b) which are initially occupied on the found states interacting with a bipartite EM field, see fig. 7.4. A bipartite two-photon state $|\psi_{\alpha\beta}\rangle$ is prepared in advance in other system and the central frequencies of subparts can be adjustable. In atom a (b), photon in its own subsystem $a(b)$ optically connects to the ground state and upper state with a large detuning Δ which is greater than the width of photon energy spectrum, photons in a superposition of coherent state

$$|mac : a(b)\rangle = \text{dim}^{-1/2} \sum_k |\alpha(\beta) : \omega_k\rangle \quad (\text{D-1})$$

in which we signify symbol $\alpha(\beta)$ as a complex number, connect to the metastable state $|m_{a(b)}\rangle$ and the upper state with a same detuning Δ . The Bohr frequency $\omega_{m;a(b)}$ usually is different from the ground state, in other words, the central frequencies of the micro- and macro-scopic states are different. For a desired central frequency of the macroscopic state, one can always adjust the central frequency of the microscopic state in advance. For sake of simplicity, we will assume the central frequencies of both are identical, so as to the Bohr frequencies of metastable state and ground state.

Furthermore, we also assume that the atomic frequency difference between the excited states are large, and photon in subsystem $a(b)$ only sensitive to atom a (b). In this sense, we split the quantized field at origin into several effective fields for practical reason,

$$\hat{E}_\mu = i\mathcal{E}_\mu \sum_m \hat{a}_{j,m} e^{-i\omega_m t} - h.c., \quad \mu = a, b; \quad (\text{D-2a})$$

$$\hat{E}_u = i\mathcal{E}_u \sum_m \hat{b}_{j,m} e^{-i\omega_m t} - h.c., \quad u = a, b. \quad (\text{D-2b})$$

The extra subscript j signify which atom a frequency component couples. A photon

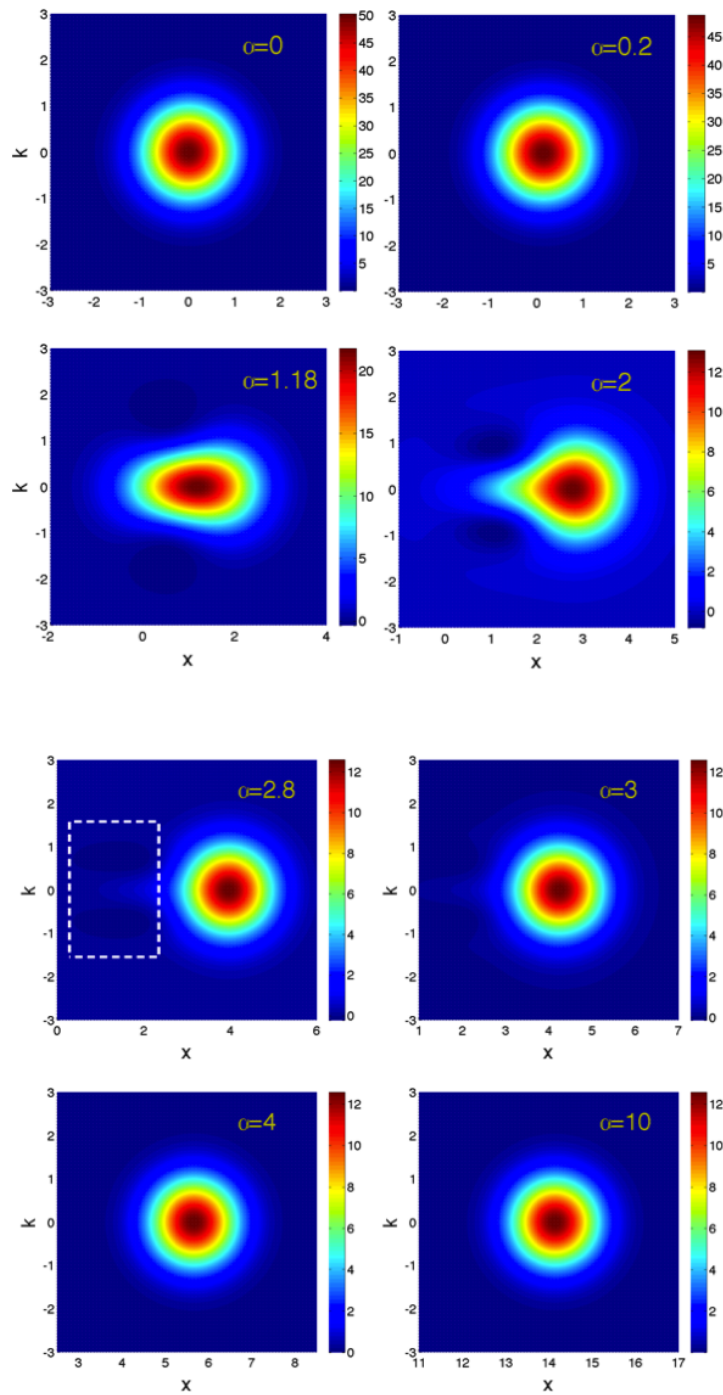


Figure 7.2: Figures of the Wigner function of the state $|0, z\rangle + |z, 0\rangle$. From the two figures, one finds zones whose value is negative. The negative value of the Wigner function is a witness of non-classicality that reveals the fact that such state cannot be produced in any classical means.

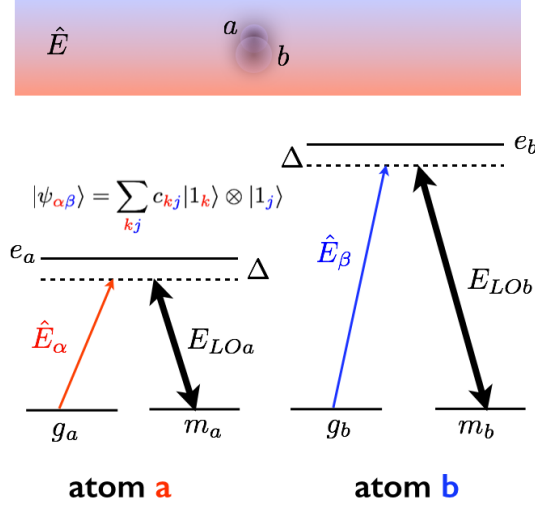


Figure 7.3: A schematic sketch for transferring the correlations in the microscopic state to macroscopic one by performing an atomic measurement. Each subsystem of a bipartite microscopic state is coupling to a three level atom in which the other energy level pair allowed for dipole transition is coupled by a macroscopic state that is superposed by equal wighted coherent states. Stationary coupling between two fields connected to the ground state and metastable state in an atom impose that a frequency mode in one field can only interacts with its corresponding frequency mode in another. If the populations which initially occupied on the ground states are measured on both metastable states, then the macroscopic state is prepared

in subsystem a does not have a direct impact on the interaction between a photon in subsystem b and the other atom, and hence we consider the whole process as two individual but similar subprocesses. Take the process related to atom (i) for detail consideration. At the rotating wave approximation the hamiltonian of the system is then given by :

$$H = \hbar\omega|e_i\rangle\langle e_i| + \sum_{\ell} \hbar\omega(\hat{a}_{i,\ell}^{\dagger}\hat{a}_{i,\ell} + \hat{b}_{i,\ell}^{\dagger}\hat{b}_{i,\ell}) + V_i; \quad (\text{D-3a})$$

$$V = \hbar|e_i\rangle\langle g_i| \sum_{\ell} f_{e_i g_i} \hat{a}_{i,\ell} + \hbar|e_i\rangle\langle m_i| \sum_{\ell} f_{e_i m_i} \hat{b}_{i,\ell} + h.c., \quad (\text{D-3b})$$

$$[\hat{a}_{i,\ell}, \hat{a}_{j,\ell'}^{\dagger}] = [\hat{b}_{i,\ell}, \hat{b}_{j,\ell'}^{\dagger}] = \delta_{ij}\delta_{\ell,\ell'}, \quad [\hat{a}_{i,\ell}, \hat{b}_{j,\ell'}^{\dagger}] = [\hat{a}_{i,\ell}, \hat{b}_{j,\ell'}] = 0; \quad (\text{D-3c})$$

where $f_{e_i g_i}$ and $f_{e_i m_i}$ are two vacuum Rabi frequencies. For simplicity, we will set the location of the two atoms are at origin and hence will not consider propagation effects in the phase factor. The vacuum Rabi frequencies are largely frequency independent, we then take them as constant numbers.

In each subsystems, whichever the microscopic state or the macro-superposed state, one can extract one photon from one frequency mode but never two photons from two different frequency modes. This model can be exactly described by the method of disentangling an operator (time ordered) exponential function[PuriMMQO] with the absence

of decoherence. In the case here, it is logical to neglect these decoherence effects in the light of large single photon detuning Δ .

Via the optical connections to a mutual upper state, the photons in both states establish a stable coherence. Since only one photon in the subsystem of the microscopic state, if one observes a population transferring to the metastable state $|m_i\rangle$, then the photon in microscopic state should have been destroyed and its correlation with the other photon in another party should be transferred to the respective macroscopic state. Therefore the desired state, according to Born's rule, is the one after performing a projection measurement $|m_a m_b\rangle\langle m_a m_b|$ on atomic part of the total density matrix,

$$\rho = \frac{\text{Tr}_{atoms}[\tilde{\rho}(t)|m_a m_b\rangle\langle m_a m_b|]}{\text{Tr}[\tilde{\rho}(t)|m_a m_b\rangle\langle m_a m_b|]} = \frac{\langle m_a m_b|\tilde{\rho}(t)|m_a m_b\rangle}{\text{Tr}[\tilde{\rho}(t)|m_a m_b\rangle\langle m_a m_b|]} \quad (\text{D-4})$$

where the total density matrix $\tilde{\rho}$ at time t is developed from the initial state $|g_a g_b\rangle\langle g_a g_b| \otimes \rho_{\alpha\beta}$ by a given set-up characterized by a function \hat{V} , therefore, can be expressed as $\tilde{\rho}(t) = \hat{V}(|g_a g_b\rangle\langle g_a g_b| \otimes \rho_{\alpha\beta})$.

To simplify the discussion, we will use lowest order perturbation theory in which we needn't consider the correlations induced by the interaction with photons between atoms a and b. The leading term which describes a population transferring from ground state $|g_j\rangle$ to the metastable state $|m_j\rangle$ certainly stands for a process $\mathcal{V}_j^{(2)}$ that destroy one photon in the microscopic state and create another one in the macroscopic state. So, the second order perturbation theory meets the need and formally

$$\langle m_a m_b|\tilde{\rho}(t)|m_a m_b\rangle \simeq \langle m_a m_b|\mathcal{V}_a^{(2)}\mathcal{V}_b^{(2)}|g_a g_b\rangle\rho_0\langle g_a g_b|\mathcal{V}_a^{(2)\dagger}\mathcal{V}_b^{(2)\dagger}|m_a m_b\rangle, \quad (\text{D-5})$$

By direct calculations, one finds

$$\langle m_j|\mathcal{V}_j^{(2)}|g_j\rangle = -f_{m_j e_j} f_{e_j g_j} \sum_{nl} \hat{b}_{j,n}^\dagger \hat{a}_{j,l} \frac{e^{-\gamma_j t - iH_{j,f}^0 t/\hbar}}{i\omega_{e_j l} + \gamma_j} \left[\frac{1 - e^{-i\omega_{nl} t}}{i\omega_{nl}} + \frac{e^{-i\omega_{nl} t} - e^{-(i\omega_{e_j l} + \gamma_j)t}}{i\omega_{e_j l} - \gamma_j} \right]$$

where $\hat{b}_{j,n}^\dagger$ is the creation operator of n -th frequency mode and $H_{j,f}^0$ is a free field Hamiltonian. The last term in the square bracket is at a magnitude of Δ^{-1} and therefore can be neglected. We also noticed that $\omega_{e_i l} \pm i\gamma_i \simeq \Delta$. As a result,

$$\langle m_j|\mathcal{V}_j^{(2)}|g_j\rangle \simeq -f_{m_j e_j} f_{e_j g_j} \sum_{nl} \frac{\hat{b}_{j,n}^\dagger \hat{a}_{j,l}}{i\Delta} \frac{1 - e^{-i\omega_{nl} t}}{i\omega_{nl}} e^{-\gamma_j t - iH_{j,f}^0 t/\hbar} \quad (\text{D-6})$$

By using an approximation to a limit representation of the Dirac δ function, $[1 - e^{-i\omega_{nl} t}]/i\omega_{nl} \simeq 2\pi\delta(\omega_{nl})$, we have

$$\begin{aligned} \langle m_j|\mathcal{V}_j^{(2)}|g_j\rangle &\simeq f_{m_j e_j} f_{e_j g_j} \frac{iL}{c} \frac{e^{-\gamma_j t}}{\Delta} \int d\omega_l \sum_n \hat{b}_{j,n}^\dagger \hat{a}_{j,l} \delta(\omega_{nl}) e^{-iH_{j,f}^0 t/\hbar} \\ &= f_{m_j e_j} f_{e_j g_j} \frac{iL}{c} \frac{e^{-\gamma_j t}}{\Delta} \sum_n \hat{b}_{j,n}^\dagger \hat{a}_{j,n} e^{-iH_{j,f}^0 t/\hbar} \\ &= e^{-iH_{j,f}^0 t/\hbar} f_{m_j e_j} f_{e_j g_j} \frac{iL}{c} \frac{e^{-\gamma_j t}}{\Delta} \sum_n \hat{b}_{j,n}^\dagger \hat{a}_{j,n} \end{aligned} \quad (\text{D-7})$$

To obtain the last equality we have used the assumption that the central frequencies of both fields are the same. However, it is safe to use the last equality even in a real situation

when there is a difference between both field. Recalling the formula in Eq.(D-5), any global (pure) phase function in the expression of $\langle m_j | \mathcal{V}_j^{(2)} | g_j \rangle$ does imply a product to its complex conjugate.

The final photon state, according to Eq.(D-4), is indeed a pure state $\rho = |\Psi\rangle\langle\Psi|$, where

$$\begin{aligned} |\Psi\rangle &= e^{-iH_f^0 t/\hbar} \frac{\sum_{mn} \hat{b}_{a,m}^\dagger \hat{b}_{b,n}^\dagger \hat{a}_{a,m} \hat{a}_{b,n} |\psi_{ab}\rangle \otimes |mac : a\rangle \otimes |mac ; b\rangle}{\|\sum_{mn} \hat{b}_{a,m}^\dagger \hat{b}_{b,n}^\dagger \hat{a}_{a,m} \hat{a}_{b,n} |\psi_{ab}\rangle \otimes |mac : a\rangle \otimes |mac ; b\rangle\|} \\ &= e^{-iH_f^0 t/\hbar} \frac{\sum_{mn} c_{mn} \hat{b}_{a,m}^\dagger \hat{b}_{b,n}^\dagger |mac : a\rangle \otimes |mac : b\rangle}{\|\sum_{mn} c_{mn} \hat{b}_{a,m}^\dagger \hat{b}_{b,n}^\dagger |mac : a\rangle \otimes |mac : b\rangle\|} \end{aligned} \quad (D-8)$$

In a macroscopic coherent state $|z_\mu : \omega_n\rangle$ whose complex amplitude is large, approximately, we can take

$$\hat{b}_{\mu,m}^\dagger |z_\mu : \omega_n\rangle \simeq \delta_{mn} z_\mu^* |z_\mu : \omega_n\rangle. \quad (D-9)$$

Then the remarkable terms are only those with same subscript indices,

$$\sum_{mn} c_{mn} \hat{b}_{a,m}^\dagger \hat{b}_{b,n}^\dagger |mac : a\rangle \otimes |mac ; b\rangle \simeq (\alpha\beta)^* \sum_{mn} c_{mn} |\alpha : \omega_m\rangle \otimes |\beta : \omega_n\rangle \quad (D-10a)$$

$$\|\sum_{mn} c_{mn} \hat{b}_{a,m}^\dagger \hat{b}_{b,n}^\dagger |mac : a\rangle \otimes |mac ; b\rangle\| \simeq |\alpha\beta| \quad (D-10b)$$

Therefore, $|\Psi\rangle$ is indeed the macroscopic state $|QMC\rangle$ by setting $\alpha = \beta$,

$$|\Psi\rangle = e^{-iH_f^0 t/\hbar} e^{-2i \arg \alpha} |QMC\rangle. \quad (D-11)$$

D.2 Micro-macro coupling via optical beam splitter

As we have seen in the previous treatment, the key ingredient to produce the QMC state is to create an one-one coupling between frequency modes. Such an interaction indeed can be found in a beam splitter. In general, a beam splitter is a physical realization of a unitary

Figure 7.4: A schematic sketch for transferring correlations in a microscopic state to a macroscopic one by performing a measurement on the output light. Each subsystem of the bipartite microscopic state together with a state $|mac : a(b)\rangle$ that is superposed by equal wighted coherent states, is sent to couple each other in a beam splitter a(b). The one-one correspondence of frequency coupling between a single photon state and a coherent state gives rise to a displacement operation only on the microscopic part. It is thus expected that the output light should include a new coherent state.

operation acting on the state who passing through it, such operation is characterized by a unitary evolution operator

$$\hat{\mathcal{B}}_s = \exp[i \sum_j \phi_j (\hat{a}_j \hat{b}_j^\dagger + \hat{a}_j^\dagger \hat{b}_j)], \quad (D-12)$$

where \hat{a}_j, \hat{b}_j are two annihilation operators of different modes but corresponding to a same frequency ω_j , ϕ_j is set real and stands for the "interaction" strength. This interaction is also not a real one in the standard sense, since any photons do not have interaction between each other, it is an effective one by averaging the microscopic details. The fact

of a vanishing commutator between the effective action $\hbar\phi_j(\hat{a}_j\hat{b}_j^\dagger + \hat{a}_j^\dagger\hat{b}_j)$ and the true Hamiltonian of free fields $\hbar\sum_j\omega_j(\hat{a}_j^\dagger\hat{a}_j + \hat{b}_j^\dagger\hat{b}_j)$ suggests such transform is energy preserved in quantum sense.

Let's first consider a most simple case: a beam splitter only working on two modes of a same frequency ω_j ,

$$\hat{\mathcal{B}}_{s,j} = \exp[i\phi_j(\hat{a}_j\hat{b}_j^\dagger + \hat{a}_j^\dagger\hat{b}_j)]. \quad (\text{D-13})$$

This is an exponential function of elements in $\mathfrak{su}(2)$ algebra because of the commutative relations:

$$[K_+^j, K_-^j] = 2K_0^j, \quad [K_0^j, K_\pm^j] = \pm K_\pm^j, \quad (\text{D-14})$$

where $K_+^j = \hat{a}_j^\dagger\hat{b}_j$, $K_-^j = \hat{a}_j\hat{b}_j^\dagger$, $K_0^j = (\hat{a}_j^\dagger\hat{a}_j - \hat{b}_j^\dagger\hat{b}_j)/2$. Such an exponential function can be disentangled as [PuriMMQO]

$$\hat{\mathcal{B}}_{s,j} = \exp(iK_+^j \tan \phi_j) \exp(-2K_0^j \ln \cos \phi_j) \exp(iK_-^j \tan \phi_j) \quad (\text{D-15})$$

Such disentanglement has a clear physical picture: the equipment works in a way by firstly taking away photons in one part and add them to another then assessing the photon number differences and lastly putting some photons back according to the "policies" from the previous judgements.

Note that the beam splitter operator $\hat{\mathcal{B}}_{s,j}$ is not a displacement operator $\hat{\mathcal{D}}(i\alpha\phi_j, \hat{a}_j) = \exp[i\alpha\phi_j\hat{a}_j^\dagger - h.c.]$ in general, except by taking $(\hat{b}_j, \hat{b}_j^\dagger) = (\alpha_j, \alpha_j^*)$ as well as weak coupling strength ϕ_j . This is universally true since a beam splitter operator is energy-preserving while a displacement operator should add energy to or take energy away from the original state in most cases.

Consider an input state $|1 : \omega_k\rangle \otimes |\alpha : \omega_q\rangle$ where α is a large complex number therefore one can apply the approximation (D-9) for creation operator \hat{b}_j^\dagger . The output state is then

$$\hat{\mathcal{B}}_{s,j}|1 : \omega_k\rangle \otimes |\alpha : \omega_q\rangle \simeq \delta_{jk}\delta_{jq}(\cos \phi_j)^{|\alpha|^2} e^{i(\alpha \tan \phi_j)\hat{a}_j^\dagger} [|1 : \omega_j\rangle + i\alpha^*|0\rangle] \otimes |\alpha : \omega_j\rangle. \quad (\text{D-16})$$

The microscopic state $(\cos \phi_j)^{|\alpha|^2} e^{i(\alpha \tan \phi_j)\hat{a}_j^\dagger} [|1 : \omega_j\rangle + i\alpha^*|0\rangle] = \hat{\mathcal{D}}(i\alpha\phi_j, \hat{a}_j)|1 : \omega_j\rangle$ is a single photon state of frequency ω_j displaced by $i\alpha\phi_j$ in phase space, then we rewrite the output as

$$\hat{\mathcal{B}}_{s,j}|1 : \omega_k\rangle \otimes |\alpha : \omega_q\rangle \simeq \delta_{jk}\delta_{jq}[\hat{\mathcal{D}}(i\alpha\phi_j, \hat{a}_j)|1 : \omega_j\rangle] \otimes |\alpha : \omega_j\rangle. \quad (\text{D-17})$$

The effective actions of different frequencies are irrelevant, as one can see from the result of any the commutation operation

$$\forall i, j; [\phi_i(\hat{a}_i\hat{b}_i^\dagger + \hat{a}_i^\dagger\hat{b}_i), \phi_j(\hat{a}_j\hat{b}_j^\dagger + \hat{a}_j^\dagger\hat{b}_j)] = 0,$$

as a result, the beam splitter operator $\hat{\mathcal{B}}_s$ can be regarded as a product combination of those single frequency beam splitter operators which works individually,

$$\hat{\mathcal{B}}_s = \otimes_j \hat{\mathcal{B}}_{s,j}. \quad (\text{D-18})$$

Then the whole output state can be expressed as

$$|\Psi\rangle = \hat{\mathcal{B}}_s^a \hat{\mathcal{B}}_s^b |\psi_{\alpha\beta}\rangle \otimes |mac : a\rangle \otimes |mac : b\rangle \quad (\text{D-19})$$

$$\begin{aligned} &= \sum_{mn} c_{mn} \otimes_{jk} \hat{\mathcal{B}}_{s,j}^a \hat{\mathcal{B}}_{s,k}^b |1 : \omega_m^a; 1 : \omega_n^b\rangle \otimes |mac : a\rangle \otimes |mac : b\rangle \\ &\simeq \sum_{mn} c_{mn} \hat{\mathcal{B}}_{s,m}^a \hat{\mathcal{B}}_{s,n}^b |1 : \omega_m^a; 1 : \omega_n^b\rangle \otimes |mac : a\rangle \otimes |mac : b\rangle \end{aligned} \quad (\text{D-20})$$

The superscript $a(b)$ signify a process related to beam splitter a(b). Let's take all the ϕ_j s to be identical, denoted by ϕ as well as take $|\alpha| \gg 1$, then the total output state reads

$$|\Psi\rangle = \dim^{-1} \sum_{mn} c_{mn} \hat{\mathcal{D}}^a(i\alpha\phi, \hat{a}_m) \hat{\mathcal{D}}^b(i\alpha\phi, \hat{a}_n) |1 : \omega_m^a; 1 : \omega_n^b\rangle \otimes |\alpha : \omega_m^a\rangle \otimes |\alpha : \omega_n^b\rangle. \quad (\text{D-21})$$

Now the macroscopic coherent states are well-coupled at the cost of the presence of some newly induced microscopic displaced Fock states $\hat{\mathcal{D}}^a(i\alpha\phi, \hat{a}_m) \hat{\mathcal{D}}^b(i\alpha\phi, \hat{a}_n) |1 : \omega_m^a; 1 : \omega_n^b\rangle$. Unfortunately, it is really difficult (or impossible) to partially displace all these states back to Fock state while keep other joint macroscopic coherent states unchanged.

Recalling the Eq.(D-16), because of the ratio between the vacuum state and single photon state in its square bracket, the large portion of the superposition is vacuum state when with large $|\alpha|$, therefore, the displaced single photon state $\hat{\mathcal{D}}(i\alpha\phi_j, \hat{a}_j) |1 : \omega_j\rangle$ behaves like a coherent state

$$\hat{\mathcal{D}}(i\alpha\phi_j, \hat{a}_j) |1 : \omega_j\rangle \simeq i\alpha^* \delta_{jk} \delta_{jq} (\cos \phi_j)^{|\alpha|^2} e^{(|\alpha| \tan \phi_j)^2 / 2} |i\alpha \tan \phi_j : \omega_j\rangle, \quad (\text{D-22})$$

and then the total output state approximates

$$|\Psi\rangle \simeq \mathcal{N}_0 \sum_{mn} c_{mn} |i\alpha \tan \phi : \omega_m^a; i\alpha \tan \phi : \omega_n^b\rangle \otimes |\alpha : \omega_m^a\rangle \otimes |\alpha : \omega_n^b\rangle \quad (\text{D-23})$$

where $\mathcal{N}_0 = -\dim^{-1} \alpha^{*2} (\cos \phi)^{2|\alpha|^2} e^{|\alpha|^2 \tan^2 \phi}$ is a constant number.

In order to get the desired output state, now we should make these microscopic coherent states disappear in the expression. We notice that the weakness of coupling strength ϕ can give rise to a universal single photon detection to all the frequency modes in each part,

$$\hat{M}_j = |I_j\rangle \langle I_j|, \quad |I_j\rangle = \dim^{-1/2} \sum_m \hat{a}_m^{\dagger} |0\rangle. \quad (\text{D-24})$$

$|I_j\rangle$ is an intrinsic single mode state according to the criteria in [Treps2005]. After performing such measurement on both microscopic parts, the final state reads

$$\rho_f = \frac{\text{Tr}_{micro}[\hat{M}_1 \hat{M}_2 |\Psi\rangle \langle \Psi|]}{\text{Tr}[\hat{M}_1 \hat{M}_2 |\Psi\rangle \langle \Psi|]} \quad (\text{D-25})$$

or alternatively,

$$|\Psi\rangle_f = \sum_{mn} c_{mn} |\alpha : \omega_m^a\rangle \otimes |\alpha : \omega_n^b\rangle = |QMC\rangle. \quad (\text{D-26})$$

D.3 Some remarks

We have demonstrated two ways to produce the QMC state. To make both proposals work, one has to prepare two macroscopic superposition states $|mac : a, b\rangle$. If either is replaced by mixed states or multimode coherent state, then they fail to give the desired result. The two states are necessities, but, such superposition states are very difficult to produce experimentally and we should find a way to produce them in future.

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