

MODELLING SINGLE MOLECULE SINGLE PHOTON
SOURCES USING MASTER EQUATION

by

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To my family

ABSTRACT

Single photons are considered to be typical carriers of quantum information and hence are the main constituents of quantum communication protocols. The generation of single photons is a great interest for future quantum information applications. Ideally, the creation of single photon means providing exactly one photon while it is required for quantum optical systems such as quantum computing and quantum cryptography.

Here we propose a full quantum optical analysis of a certain single molecule as a single photon source based on the transition of the states inside a cavity. To calculate the dynamics of this open quantum system we use stochastic methods, especially density matrix formalism and the master equation. Characterization of the source will be examined by theoretical analysis of interferometric setups such as Hanbury Brown-Twiss interferometer, by calculating second order coherence functions using quantum trajectory methods. Quantum correlations in the system and their coherent control by external fields, such as microwaves, shall be considered for improvement of the single photon source qualities. From fundamental point of view, we shall also address the relation among identical particles, quantum entanglement and single photon generation and quantum measurements.

ÖZETÇE

Tek fotonlar kuantum bilgi iletimi konusunda önemli bir taşıyıcı olarak düşünölmekte ve iletişim protokolleri için temel ögelerden biri olarak nitelendirilmektedirler. Kuantum bilgi teknolojileri uygulamaları için tek fotonların üretimi ise ilgi çekmektedir. İdeal olarak, tek foton üretimi, kuantum kriptoloji ve kuantum hesaplama gibi sistemlerin bir geređi olarak her defasında kesin bir doğrulukta tek foton üretimi demektir. Bu yüksek lisans tezi kapsamında, bir kovuk içinde bulunan tek molekülün seviyeleri arasındaki geçişlerin kuantum optiksel analizi yapılarak tek foton kaynađı olarak kullanılabilceđi incelenmektedir. Bu şekildeki bir kuantum sisteminin dinamiđi ise, yoğunluk matrisi formalizmi ve ana denklemin yazılması gibi stokastik metotlar kullanılarak yapılmaktadır. Sonuç olarak nerilen tek foton kaynađı ise Hanbury-Brown-Twiss interferometresi gibi kaynakların teorik analizi ile birinci ve ikinci korelasyon fonksiyonlarının hesaplanmasıyla yapılacaktır. Sistemdeki kuantum korelasyonlar ve uygulanan mikrodalga benzeri dış alanlarla tek foton kaynađının kalitesinin artırılabilmesi tartışılacak ve ayrıca benzer parçacıklar, kuantum dolanıklık, kuantum ölçüm teorisi ve tek foton yaratımı arasında ilişkiler de incelenecektir.

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Chapter 1

INTRODUCTION

The development of efficient single photon sources is a main requirement for quantum information processing in the forms of quantum cryptography [1][2], quantum computing [3], quantum teleportation[4][5]. Quantum cryptography is based on the no cloning theorem [6], which states that a quantum state can not be copied exactly. Hence, any attempt of eavesdropping can be detected by the two communicating parties through the fact that a measurement changes the quantum state. The fundamental prerequisite in these protocols is therefore that information is coded in single qubits only. This makes single-photon sources an essential ingredient for quantum communication. Single photon sources have been produced by using two level rubidium atoms [7], pumping individual quantum dots [8][9], nitrogen vacancy color centers [10] and single molecules [11][12][13].

In principle, atom-cavity systems can generate single photons deterministically into a well defined mode which is easy to detect, making them much more efficient than other sources. The time evolution of the system is given by the master equation, which is of the type of a Fokker-Planck equation, is used to derive the equation of motion for the mean values of various atomic operators characterizing the physical properties of the system [14].

In order to identify rapidly single photon emission, a standard HBT setup is usually used to split the beam and detect single photons with two identical single photon detectors. Therefore, a HBT measurement proves that only single photons emerge from the cavity. This can be done by calculating the second order correlation function. To calculate the second-order correlation function for optical pumping, we

use numerical techniques based on a quantum trajectory approach[15][16][17] which presents the master equation approach in the interaction picture to study evolution of a certain single molecule. In addition, we report photon correlation measurements on a single molecule show antibunching behaviour as a requirement of the efficient single photon sources.

Besides single photon sources, sources of pairs of photons are also important, due to useful properties of indistinguishable photons and entanglement.[18][19] Many quantum information processing applications of a single-photon source require indistinguishable photons. To test for the indistinguishability of the produced photons a two-photon interference experiment has to be performed. According to Hong-Ou-Mandel effect, two photons that simultaneously enter different entrance ports of a 50:50 beam splitter will always leave through the same output port if they are indistinguishable.

This thesis presents the modelling of single photon sources using an atom-photon interface based on an atom-cavity system with the ability to generate single photons. In Chapter 1 the principle of the single photon source is introduced, significant experiments and properties of single photon sources are described. Chapter 2 reports using master equation and some stochastics methods under the key approximations. General discussion of the fluctuation-dissipation theorem and applications of these stochastic methods to the three level system in a cavity given in the Chapter 3. Using single molecule inside a cavity as a single photon source and signature of single photon sources like second order correlation function and Mandel Q parameter given in chapter 4.

Chapter 2

FUNDAMENTAL PROPERTIES OF SINGLE PHOTON SOURCES

Photons are the elementary constituents of light. They can be viewed in some sense as energy wave-packets and moving at the speed of light. A single photon is a quantum system in which quantum information can be encoded in various ways and transported over long distances safely. It is a non-classical state of light, in the sense that it cannot be described in terms of a classical electric and magnetic field.

The generation of single photons is a great interest for future quantum information applications. Ideally, the creation of single photon means providing exactly one photon while it is required for quantum optical systems such as quantum computing and quantum cryptography. An efficient single photon source has to fulfill some criteria such as indistinguishability and a high collection efficiency.

2.1 Signatures of Single Photon Sources

The quantum properties of light have been put to practical use in recent years in various forms of quantum information processing. The basic idea here is to use the laws of quantum mechanics to enhance the capabilities of transferring or manipulating data. We begin discussion by considering quantum cryptography which is a key distribution system that attempts to link the security of the system to the correctness of the uncertainty principle of quantum. Uncertainty is a major constituent in all quantum mechanics, and basically states the indeterminism of the universe around us. This indeterminism gives rise to certain mechanisms that can be put to great use in cryptography.

The best-known application of quantum cryptography is quantum key distribution

(QKD). QKD describes the process of using quantum communication to establish a shared key between two parties (usually called Alice and Bob) without a third party (Eve) learning anything about that key [21]. This is achieved by letting Alice encode the bits of the key as quantum data before sending them to Bob; if Eve tries to learn these bits, the messages will be disturbed and Alice and Bob will notice. Experimental quantum key distribution was demonstrated for the first time in 1989 and it was published only in 1992 [20]. Today, several groups have shown that quantum key distribution is possible, even outside the laboratory. In principle, any two-level quantum system could be used to implement Quantum Cryptography. In practice, optical quantum cryptography is based on the use of single photon Fock state. The reason is that their interaction with the environment can be controlled and moderated. The ideal single photon source is a device that when one pulls the trigger, and only then, emits one and only one photon. Although photon antibunching has been demonstrated already years ago, a practical and handy device is still needs to be improved. A single two-level quantum system that can obviously not emit two photons at a time. The manipulation of single trapped atoms or ions requires a much too involved technical effort. Single single molecules are easier to handle but only offer limited stability at room temperature. The fluorescence of single molecule exhibits strong photon antibunching.

It is the main purpose of the present chapter to investigate the signatures of ideal single photon sources by introducing the quantum coherence functions, indistinguishability and photon antibunching properties of single photons.

2.1.1 Photon Detection and Quantum Coherence Functions

In this section we discuss the photon detection and quantum coherence functions. Let us begin with the separation of field operator $E(r, t)$ into the two parts

$$E(r, t) = E^{(+)}(r, t) + E^{(-)}(r, t) \quad (2.1)$$

Here $E^+(r, t)$ and $E^-(r, t)$ are

$$E^{(+)} = \sum_k \epsilon_k \epsilon_k a_k e^{-i\nu_k t + i \vec{k} \cdot \vec{r}} \quad (2.2)$$

$$E^{(-)} = \{E^{(+)}\}^\dagger \quad (2.3)$$

Evidently, $E^+(r, t)$ is the positive frequency component part of the field, which contains the annihilation operators only and $E^-(r, t)$ is the negative frequency component part, which is associated with the creation operators only. The photoelectrons produced by photoionization are then observed [22]. In such absorbtive detectors, the measurements are destructive. Therefore only the annihilation operator $E^{(+)}(r, t)$ contributes to the process. Schematically an atom is placed in the radiational field at position r in its ground state. When we consider the atomic transition between the ground state $|g\rangle$ and an excited state $|e\rangle$ which associates with a transtion of a radiation field from initial state $|i\rangle$ to a final state $|f\rangle$. In this case, the photon counting rate of the detector can be written as

$$w_1(r, t) = |\langle f | E^{(+)}(r, t) | i \rangle|^2 \quad (2.4)$$

where $|i\rangle$ is the initial state and $|f\rangle$ is the final state. The final state of the field is never measured. Hence summing over the final states yields

$$w_1(r, t) = \sum_f |\langle f | E^{(+)}(r, t) | i \rangle|^2 \quad (2.5)$$

$$= \sum_f \langle i | E^{(-)}(r, t) | f \rangle \langle f | E^{(+)}(r, t) | i \rangle \quad (2.6)$$

$$= \langle i | E^{(-)}(r, t) E^{(+)}(r, t) | i \rangle \quad (2.7)$$

where we used the completeness relation

$$\sum_f |f\rangle \langle f| = 1. \quad (2.8)$$

We write the photon counting rate w_1 with averaging over all the possible realizations of the initial fields

$$w_1(r, t) = \sum_{P_i} \langle i | E^{(-)}(r, t) E^{(+)}(r, t) | i \rangle. \quad (2.9)$$

Introducing the density operator

$$\rho = \sum_i |i\rangle\langle i|, \quad (2.10)$$

the photocounting rate can be written as

$$w_1(r, t) = \sum_i \rho \langle i | E^{(-)}(r, t) E^{(+)}(r, t) | i \rangle \quad (2.11)$$

$$= \text{Tr}[\rho E^{(-)}(r, t) E^{(+)}(r, t)]. \quad (2.12)$$

Now we define first order correlation function

$$\begin{aligned} G^{(1)}(r_1, r_2; t_1, t_2) &= \text{Tr}[\rho E^-(r_1, t_1) E^+(r_2, t_2)] \\ &= \langle E^-(r_1, t_1) E^+(r_2, t_2) \rangle. \end{aligned}$$

In terms of the time difference $\tau = t_2 - t_1$, we can write the first order correlation function

$$G^{(1)}(r_1, r_2; t_1, t_2) \equiv G^{(1)}(r_1, r_2; \tau). \quad (2.13)$$

We now consider two photodetectors and calculate joint counting rate of one photoionization at point r_2 between t_2 and $t_2 + dt_2$ and another one at point r_1 between t_1 and $t_1 + dt_1$ with $t_1 \leq t_2$. In this case photocounting rate can be written as

$$w_2(r_1, t_1; r_2, t_2) = |\langle f | E^-(r_2, t_2) E^+(r_1, t_1) | i \rangle|^2. \quad (2.14)$$

Summing over the all final states and average over the all initial states as previous discussions, yields

$$\begin{aligned} G^{(2)}(r_1, r_2, r_3, r_4; t_1, t_2, t_3, t_4) &= \text{Tr}[\rho E^{-(r_1, t_1)} E^{-(r_2, t_2)} E^{+(r_3, t_3)} E^{+(r_4, t_4)}] \\ &= \langle E^-(r_1, t_1) E^-(r_2, t_2) E^+(r_3, t_3) E^+(r_4, t_4) \rangle. \end{aligned}$$

We can now define the normalized quantum mechanical first and second order correlation function at the position r

$$g^{(1)}(r; \tau) = \frac{\langle E^-(r, t) E^+(r, t + \tau) \rangle}{\sqrt{\langle E^-(r, t) E^+(r, t + \tau) \rangle \langle E^-(r, t) E^+(r, t + \tau) \rangle}}, \quad (2.15)$$

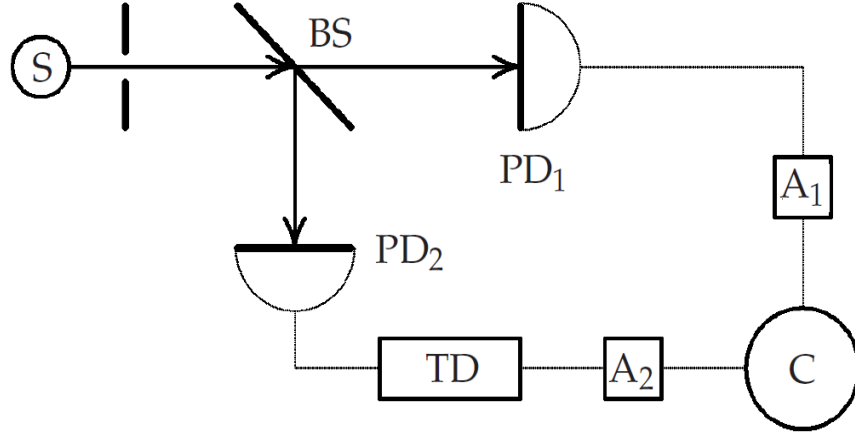


Figure 2.1: HBT experiment scheme where S is a light source; BS is beam splitter; PD1 and PD2, photodetectors; A1 and A2, amplifiers; TD, delay-time control; C, correlator.[23]

$$g^{(2)}(r; \tau) = \frac{\langle E^-(r, t)E^-(r, t + \tau)E^+(r, t)E^+(r, t + \tau) \rangle}{\sqrt{\langle E^-(r, t)E^+(r, t) \rangle \langle E^-(r, t + \tau)E^+(r, t + \tau) \rangle}}. \quad (2.16)$$

In the definition of $g^{(2)}(r; \tau)$, we have chosen not only normal ordering of the field operators but also certain time ordering due to fact that $t_2 \geq t_1$. Using the mode expansions of $E^{(+)}$ and $E^{(-)}$ as

$$E^{(+)} = \sum_k \epsilon_k \epsilon_k a_k e^{-i\nu_k t + i\vec{k} \cdot \vec{r}}, \quad (2.17)$$

$$E^{(-)} = \sum_k \epsilon_k \epsilon_k a_k^\dagger e^{i\nu_k t - i\vec{k} \cdot \vec{r}} \quad (2.18)$$

with cancelling most factors in these expressions, we finally write normalized first and second order correlation functions in the following way

$$g^{(1)}(\tau) = \frac{\langle a^\dagger(t)a(t + \tau) \rangle}{\langle a^\dagger a \rangle}, \quad (2.19)$$

$$g^{(2)}(\tau) = \frac{\langle a^\dagger(t)a^\dagger(t + \tau)a(t + \tau)a(t) \rangle}{\langle a^\dagger a \rangle^2}. \quad (2.20)$$

2.1.2 Hanbury Brown and Twiss Experiment

In the previous section we have studied how light beams can be detected by photodetectors and we saw that correlation functions may be seen as a clear signature of the quantum nature of the light. We will also discuss that antibunched light is only possible in photon interpretation and quantified according to second-order correlation function. To understand the non-classical nature of light we present the Hanbury Brown-Twiss (HBT) experiment which was first investigated by R. Hanbury Brown and R. Q. Twiss in 1950's [24]. The principle behind the HBT experiment is that the intensity fluctuations of a beam of light is related to its coherence. For the HBT experiment shown in Figure 2.1, if the distance between the photodetectors PD_1 and PD_2 and the 50 : 50 beam splitters are adjusted to be equal and the intensity of the incident field $I(z, t)$,

$$I_1(z, t) = I_2(z, t) = \frac{I(z, t)}{2} \quad (2.21)$$

their average values during the time T

$$\bar{I}_1 = \bar{I}_2 = \frac{\bar{I}}{2} \quad (2.22)$$

Then the output of the correlator C gives

$$C = \langle (I_1(z, t) - \bar{I}_1)(I_2(z, t) - \bar{I}_2) \rangle \quad (2.23)$$

$$= \frac{1}{4} [\langle I^2(z, t) - \bar{I}^2 \rangle] \geq 0. \quad (2.24)$$

Therefore, normalized output of C is

$$\frac{\langle I^2(z, t) - \bar{I}^2 \rangle}{\bar{I}^2} = g^{(2)} - 1 \geq 1. \quad (2.25)$$

In the classical description of the HBT experiment results are always positive and zero only when the instantaneous intensities are equal.

Historically HBT experiment caused great controversy. In 1963 R. Glauber wrote his paper [25] to explain HBT using the quantum theory of photon counting. His major contribution to discussion was considering quantum expressions of measuring quantities. We now turn to study the coherence properties of light in the quantum

theory. Our main goal here is to characterize the phenomena which are not described by classical statistical properties. Assuming the lengths between the experimental tools do not vary, photons impinge on the detectors and incoming light consists of a stream of photons output pulses recorded by the electronic counter. In this case photons will be detected by the two detectors with equal probability, so generation of a start pulse at the first detector and trigger the electronic counter means that there is zero probability of a generation of stop pulse from other detector of this photon. It means that if photon goes to the detector 1 then nothing happens and we have to wait for the next photon to have a chance of having a stop pulse. The process proceeds until a stop pulse eventually achieved. We therefore have a situation where we expect no events at delay time between two photons, $\tau = 0$, but some events for larger values of τ . This is clearly controversial with classical discussions above.

2.1.2.1 Photon Antibunching

As discussed above, R. Hanbury Brown and R. Q. Twiss investigated temporal correlations of the intensity fluctuations in a light beam emitted by a thermal source. According to their explanations photons in the light beam tend to arrive in bunches, rather than strictly at random; this phenomenon is called photon bunching. Nonetheless, their observations can also be explained classically as a wave behaviour of the electromagnetic field.

Due to Glauber's explanation of the quantum theory of optical coherences, in contrast to photon bunching, photons can also tend to arrive more evenly spread out than strictly at random. This phenomenon is called photon antibunching, and experimentally observed by Kimble *et al.* in 1977 [26]. Antibunching is understandable only in the quantum mechanical description of light. The fact that it has no classical explanation can be seen as a direct evidence for the existence of photons.

2.1.3 Hong-Ou-Mandel Experiment

Hong-Ou-Mandel effect is a quantum interference effect that occurs when two indistinguishable photons impinge onto a symmetric beam splitter [27]. Indistinguishable photons arrive simultaneously at a 50:50 beamsplitter's two input parts. We expect that both photons exit together from beamsplitter's output parts. With perfect overlap and a perfect 50:50 beamsplitter, the probability that a single photon exits from each of the two output parts are exactly zero. As an example with operator transformations, let us consider the incident state $a^\dagger b^\dagger |0, 0\rangle_{ab} = |1, 1\rangle_{ab}$ where a , a^\dagger , b , b^\dagger are the annihilation and creation operators of two optical modes and $|1\rangle$ is a single photon state. When two modes are mixed in a 50:50 beam splitter, using the unitary transformation, they turn into new modes c and d

$$\begin{pmatrix} a \\ b \end{pmatrix} \rightarrow \frac{1}{\sqrt{2}} \begin{pmatrix} 1 & 1 \\ 1 & -1 \end{pmatrix} \begin{pmatrix} c \\ d \end{pmatrix}. \quad (2.26)$$

Then

$$a^\dagger \rightarrow \frac{c^\dagger + d^\dagger}{\sqrt{2}} \quad b^\dagger \rightarrow \frac{c^\dagger - d^\dagger}{\sqrt{2}}.$$

When two photons enter a beamsplitter, the state of the two modes becomes

$$\begin{aligned} |1, 1\rangle_{ab} &= a^\dagger b^\dagger |0, 0\rangle_{ab} \rightarrow \frac{1}{2} (c^\dagger + d^\dagger)(c^\dagger - d^\dagger) |0, 0\rangle_{cd} \\ &= \frac{1}{2} (c^{\dagger 2} - d^{\dagger 2}) |0, 0\rangle_{cd} \\ &= \frac{|2, 0\rangle_{cd} - |0, 2\rangle_{cd}}{\sqrt{2}} \end{aligned} \quad (2.27)$$

where we labeled the output modes c and d in accordance with the original paper of Hong, Ou and Mandel.

The Hong-Ou-Mandel effect can be used to test the degree of indistinguishability of the two incoming photons which is very important to model single photon source. For example, the Hong-Ou-Mandel interferometer was used, in 2002, to demonstrate the purity of a solid state single photon source by feeding two successive photons from a single photon source into a 50:50 beam splitter [28].

Chapter 3

OPEN QUANTUM SYSTEMS

An open quantum system, S , is a quantum mechanical system that interacts with an external quantum system. Since we assume here the environment is infinitely large, we refer it as the reservoir R . The open quantum system, S , thus represents a subsystem of the combined total system $S \otimes R$. The state of the subsystem S , changes as a consequence of its internal dynamics and interaction with its surroundings.

Due to dissipative nature of the open quantum systems, we generally do not have complete knowledge of the system state. In many cases it is useful to formulate the dynamics of an open system by means of an appropriate equation of motion for its density matrix, a quantum master equation. The advantage of the master equation is that it allows the derivation simpler, solvable equations using only a few assumptions and approximations for both the interaction between S and R , and the properties of R which is in the content of the following section.

3.1 Born and Markov Approximations

Here we discuss the derivation of the Born and Markov approximations as commonly used in optics. We derive where these approximations come from and where they are used in quantum optics. Particularly, deriving a quantum optical master equation makes good understanding of these approximations. Born and Markov approximations involve assuming that the reservoir correlations decay rapidly compared to the system damping time and that the reservoir does not change significantly with time due to the effect of the system. Besides, its mathematical benefits in derivations, these approximations are not valid in many regimes.

3.1.1 Born Approximation

In this section, we propose to show that, how one can demonstrate the Born approximation in quantum mechanics with introducing the integral scattering equation for stationary states. [29, 30] We can write the eigenvalue equation of H ,

$$H\phi(\mathbf{r}) = E\phi(\mathbf{r}) \quad (3.1)$$

where

$$H = \frac{p^2}{2m} + \frac{\hbar^2}{2m}U(\mathbf{r}) \quad (3.2)$$

and

$$E = \frac{\hbar^2 k^2}{2m} \quad (3.3)$$

can be put into the form

$$(\nabla^2 + k^2)\phi(\mathbf{r}) = U(\mathbf{r})\phi(\mathbf{r}). \quad (3.4)$$

The solution to this equation may be written in the form

$$\Phi(r) = \Phi_0(r) + \int G(r - r')U(r')\Phi(r')d^3r' \quad (3.5)$$

where $\Phi_0(r)$ is the solution of the homogeneous equation and $G(r)$ is the “Green’s function“ which satisfies the relation.

$$(\nabla^2 + k^2)G(r) = \delta(r) \quad (3.6)$$

For a stationary scattering state we might choose $\Phi_0(r) = e^{(ikr)}$ and $G(r) = G^\dagger(r)$, and we get

$$\Phi_k(r) = e^{ikr} + \int G^\dagger(r - r')U(r')\Phi_k(r')d^3r' \quad (3.7)$$

A simple change of notation, $r \rightarrow r'$ and $r' \rightarrow r''$ and applying same procedure again and a permits us to write

$$\begin{aligned} \Phi_k(r) &= e^{ikr} + \int G^\dagger(r - r')U(r')\Phi_k(r')d^3r' \\ &+ \int d^3r' \int G^\dagger(r - r')U(r')\Phi_k(r')G^\dagger(r' - r'')U(r'')\Phi_k(r'')d^3r'' \end{aligned}$$

where the first two terms are known; the unknown function $\Phi_k(r)$ has been pushed back into the fourth term.

Thus we can construct, step by step, what is called the Born expansion. If the potential is weak, each successive term is smaller than the preceding one. If we push the expansion far enough, we neglect the last terms in the right hand side and thus we called "Born Approximation" is the first term in the Born expansion.

3.1.2 Markov Process and Markov Approximation

We can describe stochastic processes using the system which evolve probabilistically in time or systems in which a certain time-dependent random variable $X(t)$ exist [31]. We can measure values $x_1, x_2, x_3 \dots$ etc of $X(t)$ at times $t_1, t_2, t_3 \dots$ and we can also define conditional probability densities as

$$P_{n,k}(x_{k+n}, t_{k+n}; \dots; x_{k+1}, t_{k+1} | x_k, t_k; \dots, x_1, t_1) \quad (3.8)$$

$$= P_{n,1}(x_{k+n}, t_{k+n}; \dots; x_{k+1}, t_{k+1} | x_k, t_k) \quad (3.9)$$

for $(t_1 \leq t_2 \leq t_3 \leq \dots)$

Here we can see that only the most recent past governs the time evolution of the process. We can express this by saying that the future of $x(t)$ is influenced by the present, but not by its earlier history. such Process is known as a "Markov Process."

The concept of a Markov Process is a very powerful and surprisingly general tool in the study of classical irreversible phenomena. Classically, the essence of the Markov idea is that we need only know the probability distribution at time t in order to predict for all future times. At the deepest level in classical physics this is exactly true: Hamilton's equations of motion are first order in time, so that the knowledge of the canonical quadratures at one time determines them for all futures. But even classically, Markov property must be regarded as an approximation. [32]

In classical or quantum mechanical sense, damping of a macroscopic system comes from the transfer of energy to microscopic degrees of freedom, which, in the macroscopic description, give rise to noise terms, in the macroscopic equations of motion.

The next section develops the theory of quantum stochastic differential equations using Born and Markov approximations giving theoretical discussion of the quantum optical master equation.

3.2 The Master Equation Approach

In this section we derive the master equation of an open quantum system using some approximations. As we introduced in the previous sections, Born-Markov approximation involves assuming that reservoir correlations decay rapidly compared with the system damping time. In that case, reservoir does not change significantly with time due to the effect of the system.

We consider the system which is described by the Hamiltonians for the system, heat bath, and the interaction respectively.

$$H = H_S + H_R + H_{int} \quad (3.10)$$

The total density operator for the system and reservoir, $\rho_{tot}(t)$, in the Schrödinger picture, satisfies the Liouville Equation

$$\dot{\rho}_{tot} = -\frac{i}{\hbar}[H_S + H_R + H_{int}, \rho_{tot}] \quad (3.11)$$

We now eliminate the field operators and get the system operators, by using the reduced density operator

$$\hat{\rho}(t) = Tr_R \{ \rho_{tot} \} \quad (3.12)$$

and it is most convenient to transform into the interaction picture, then setting $\rho_{int}(t)$, density operator

$$\rho_{int}(t) = exp\left[\frac{i}{\hbar}(H_{sys} + H_R)t\right]\rho_{tot}(t)exp\left[-\frac{i}{\hbar}(H_{sys} + H_R)t\right] \quad (3.13)$$

obeys the interaction picture equation of motion

$$\dot{\rho}_I(t) = -\frac{i}{\hbar}[H_{int}, \rho_I(t)] \quad (3.14)$$

where we also transformed the Hamiltonian as

$$H_{int}(t) = \exp\left[\frac{i}{\hbar}(H_{sys} + H_R)t\right]H_{int}\exp\left[-\frac{i}{\hbar}(H_{sys} + H_R)t\right] \quad (3.15)$$

As an initial condition, we assume that the system and the bath are initially independent, so that the total density operator factorizes into a direct product

$$\rho_{tot}(0) = \hat{\rho}(0) \otimes \rho_R \quad (3.16)$$

And we can also say that reservoir is so large that its statistical properties are unaffected by the weak coupling to the system.

We now return the equation of motion and integrate from 0 to t

$$\int_0^t dt \dot{\rho}_I(t) = - \int_0^t dt \frac{i}{\hbar} [H_{int}, \rho_I(t)] \quad (3.17)$$

subject to initial conditions we write

$$\rho_I(t) = \rho_I(0) - \frac{i}{\hbar} \int_0^t dt' [H_{int}(t'), \rho_I(0)] \quad (3.18)$$

and iterating as taking derivative with respect to t and replacing $\frac{d}{dt} \rightarrow [H_{Int}(t), \cdot]$ yields

$$\begin{aligned} \dot{\rho}_I(t) &= \frac{i}{\hbar} [H_{int}(t), \rho_I(0)] \\ &- \frac{1}{\hbar^2} \int_0^t dt' Tr_B \{ [H_{int}(t), [H_{int}(t'), \rho_I(t')]] \} \end{aligned} \quad (3.19)$$

Next step is defining reduced density operator in the interaction picture

$$\hat{\rho}_I(t) = \exp\left[-\frac{i}{\hbar} H_{sys}t\right]\rho(t)\exp\left[\frac{i}{\hbar} H_{sys}t\right] \quad (3.20)$$

where we can obtain

$$\rho(t) \equiv Tr_R \{ \hat{\rho}_I \} \quad (3.21)$$

using this expression yields

$$\dot{\rho}_I(t) = -\frac{1}{\hbar^2} \int_0^t dt' Tr_R \{ [H_{Int}(t), [H_{Int}(t'), \rho_I(t')]] \} \quad (3.22)$$

where we assumed with considering initial condition

$$\text{Tr}_R \{H_{int}(t)\rho_I\} = 0 \quad (3.23)$$

and

$$\rho_I(0) = \rho_{tot}(0) = \hat{\rho}(0) \otimes \hat{\rho}_B \quad (3.24)$$

Born Approximation, we now consider the weak coupling case via assuming that H_{int} is very much less than either H_R or H_{sys} , and the reservoir density operator is not significantly affected by the interaction. In this case, we may write

$$\rho_I(t') \approx \rho(t') \otimes \rho_R \quad (3.25)$$

and it is straightforward to see that, the bath density operator is not significantly changed by the interaction. However we have allowed that the system density operator to change significantly, and we also know that system is much smaller than the reservoir, hence the fractional effect of the interaction is much bigger on the system. Using the weak coupling assumption, we can reduce the (23) to an equation

$$\dot{\rho}(t) = -\frac{1}{\hbar^2} \int_0^t dt' \text{Tr}_R \{[H_{int}(t), [H_{int}(t'), \rho_I(t') \otimes \rho_R]]\}. \quad (3.26)$$

Markov Approximation, since the interaction is assumed weak, the rate of change of the interaction picture system density operator will be quite slow compared to that of the bath operators, which will var on a time scale determined by H_R . In fact, bath correlation functions are generally determined by the choice of ρ_R which are much shorter than the typical time constant expected from $\rho(t)$. In this case, we can say that $\rho(t')$ changes insignificantly over the time and setting $\rho(t') \rightarrow \rho(t)$ and due to fact that $t \gg$ thermal correlation time, we can let the lower limit of the time integral go to $-\infty$. Finally we get the master equation:

$$\dot{\rho}_I(t) = -\frac{1}{\hbar^2} \int_0^t dt' \text{Tr}_R \{[H_{int}(t), [H_{int}(t - \tau), \rho_I(t) \otimes \rho_R]]\} \quad (3.27)$$

The approximation, $\rho(t') \rightarrow \rho(t)$ is called the Markov approximation and this means that the knowledge of $\rho(t)$ at one point in time $t = t_0$ is sufficient to determine $\rho(t)$ for

all $t > t_0$. The Markov property is a highly desirable property from the mathematical point of view, because whole structure of measurement theory can be built around it in a compact way.

3.2.1 The Master Equation for Two-Level Systems

In this subsection, we introduce the master equation for two level system. Master equation approach allows us to take the effect of the applied field (the Rabi oscillations) and the effect of spontaneous emission. Master equations are the main quantum optical tool to predict the time evolution of electromagnetically driven systems undergoing spontaneous emission. They are especially suited to calculate expectation values, whose calculation would otherwise require an averaging over all the possible trajectories of the system. However, before we can write down the master equations for two level system, we have to introduce the density matrix operator ρ . The main motivation for the introduction of this operator is, that it allows us to calculate the probability to find the atom at a time t in the excited state or the probability density for a photon emission at a time t without having to know anything else. As an simple example for master equation aprocah we now discuss simple two level atom in a free space. Such a system has two stationary energy levels, denoted by $E_a = \hbar\omega_a$ (upper level) and $E_b = \hbar\omega_b$ (lower level). the wave functions of these two levels are denoted by the state vectors $|a\rangle$ (upper state) and $|b\rangle$ (lower state). These state vectors are normalized as

$$\begin{aligned}\langle a | a \rangle &= \langle b | b \rangle = 1 \\ \langle a | b \rangle &= \langle b | a \rangle = 0\end{aligned}\tag{3.28}$$

and the completeness of the basis requires

$$\langle a | a \rangle + \langle b | b \rangle\tag{3.29}$$

In general, any state vector, wave function, of the two level system can be written in the form,

$$|\psi\rangle = C_a |a\rangle + C_b |b\rangle\tag{3.30}$$

Here C_a and C_b are the probability amplitudes of finding the system in states, $|a\rangle$ and $|b\rangle$, respectively. The total Hamiltonian is,

$$H = H_0 + H_{int} \quad (3.31)$$

where H_0 is the free part and given by

$$H_0 = \hbar\omega_a \langle a | a \rangle + \hbar\omega_b \langle b | b \rangle \quad (3.32)$$

In this case, we consider a particular case where two level system is free, $H_{int} = 0$.

In this case the Schrödinger equation is,

$$\begin{aligned} i\hbar \frac{d}{dt} |\psi\rangle &= H_0 |\psi\rangle \\ &= (\hbar\omega_a \langle a | a \rangle + \hbar\omega_b \langle b | b \rangle) |\psi\rangle \end{aligned} \quad (3.33)$$

which has a solution like

$$\begin{aligned} |\psi\rangle &= C_a(t) |a\rangle + C_b(t) |b\rangle \\ &= e^{-i\omega_a t} C_a(0) |a\rangle + e^{-i\omega_b t} C_b(0) |b\rangle \end{aligned} \quad (3.34)$$

then we introduce the density operator

$$\rho = |\psi\rangle \langle \psi| \quad (3.35)$$

and we derive the evolution equation for the density operator ρ

$$\begin{aligned} \frac{d}{dt} \rho &= \frac{d}{dt} |\psi\rangle \langle \psi| \\ &= \left(\frac{d}{dt} |\psi\rangle \right) \langle \psi| + |\psi\rangle \left(\frac{d}{dt} \langle \psi| \right) \\ &= \frac{1}{i\hbar} H |\psi\rangle \langle \psi| - \frac{1}{i\hbar} |\psi\rangle \langle \psi| H \\ &= \frac{1}{i\hbar} [H, \rho] \end{aligned} \quad (3.36)$$

which is a master equation for a two level atom in a free space where $H_{int} = 0$. In case of two-level atom in a cavity we use the general form of the master equation to

evaluate the time evolution of the density matrices. Interaction Hamiltonian is now given

$$\begin{aligned} H_{int} &= \hbar g(\hat{a}\hat{\sigma}_+ + \hat{a}^\dagger\hat{\sigma}_-) \\ &= \hbar g \begin{pmatrix} 0 & \hat{a} \\ \hat{a}^\dagger & 0 \end{pmatrix} \end{aligned} \quad (3.37)$$

and the initial state

$$\begin{aligned} \hat{\rho}_{af}(t) &= \hat{\rho}_f \otimes \begin{pmatrix} \rho_{aa} & 0 \\ 0 & \rho_{bb} \end{pmatrix} \\ &= \begin{pmatrix} \rho_{aa}\hat{\rho}_f & 0 \\ 0 & \rho_{bb}\hat{\rho}_f \end{pmatrix} \end{aligned}$$

Iterative solution of the master equation can be written as

$$\begin{aligned} \hat{\rho}_f(t + \tau) &= \hat{\rho}_f(t) + \frac{\tau}{i\hbar} Tr_{atom}[H_{int}, \hat{\rho}_{af}(t)] \\ &+ \frac{\tau^2}{2(i\hbar)^2} Tr_{atom}[H_{int}, [H_{int}, \hat{\rho}_{af}(t)]] \end{aligned} \quad (3.38)$$

and according to the Born-Markov approximation second term does not change during a time interval τ and using the matrix representation, third term can be calculated as

$$\begin{aligned} Tr_{atom}[H_{int}, [H_{int}, \hat{\rho}_{af}(t)]] &= (\hbar g)^2 (aa^\dagger \rho_{aa} \hat{\rho}_f - a \rho_{bb} \hat{\rho}_f a^\dagger + a^\dagger a \rho_{bb} \hat{\rho}_f - a^\dagger \rho_{bb} \hat{\rho}_f a) \\ &- (\hbar g)^2 (a \rho_{bb} \hat{\rho}_f a^\dagger - \rho_{aa} \hat{\rho}_f a a^\dagger + a^\dagger \rho_{aa} \hat{\rho}_f a - \rho_{bb} \hat{\rho}_f a^\dagger a) \end{aligned}$$

and using the expression we find that

$$\begin{aligned} \hat{\rho}_f(t + \tau) &= \hat{\rho}_f(t) + \frac{1}{2}(g\tau)^2 [(aa^\dagger \rho_f - 2a^\dagger \rho_f a + \rho_f a a^\dagger) \rho_{aa} \\ &+ (a^\dagger a \rho_f - 2a \rho_f a^\dagger + \rho_f a^\dagger a) \rho_{bb}] \end{aligned} \quad (3.39)$$

If r atoms injected per second, the change in $\hat{\rho}_f(t)$ should be multiplied by $r\tau$. Then

$$\begin{aligned} \frac{d}{dt} \rho &= \frac{r\tau[\hat{\rho}_f(t + \tau) - \hat{\rho}_f(t)]}{\tau} \\ &= -\frac{1}{2} R_a [aa^\dagger \rho_f - 2a^\dagger \rho_f a + \rho_f a a^\dagger] - \frac{1}{2} R_b [a^\dagger a \rho_f - 2a \rho_f a^\dagger + \rho_f a^\dagger a] \end{aligned} \quad (3.40)$$

where $R_a = r\rho_{aa}(g\tau)^2$ is the photon emission rate per second and $R_b = r\rho_{bb}(g\tau)^2$ is the photon absorption rate per second and the equation called the master equation for the two level atom in a cavity. There are many examples, where a more detailed analysis of the time evolution of a system, using the master equations. In general, the description of any quantum optical system with more than two levels or involving a more complex, time dependent Hamiltonian requires a solution of the corresponding master equations.

3.3 Quantum Regression Theorem and Fluctuation-Dissipation Theorem

We discuss here the evolution of the quantum regression theorem and the fluctuation-dissipation theorem with considering the both classical and quantum mechanical point of view and provide examples to show the power of these theorems. Then we discuss the two-level system density matrices using the both quantum regression theorem and the fluctuation-dissipation theorem and relate with the quantum optics with discussing the second-order correlation function and the concept of the photon anti-bunching.

3.3.1 Quantum Regression Theorem

Linear systems are always particular interest in physics due to their simplicity. It is often the case that the equations of motion for the means of certain operators are linear. It was shown by Lax [33] that it is often possible to express multi-time correlation functions of certain quantum mechanical operators in terms of single time expectations. The result is known as the regression theorem [34].

We consider the quantum system, S, which is coupled with the reservoir, R, and suppose that the system has a complete orthonormal set of eigenstates $|\Phi_i\rangle \dots i = 1, 2, 3 \dots$ as a basis. Whether these eigenstates are discrete or continuous, we will here treat as discrete for the simplicity. The density operator picture spans the Hilbert

space of S and R and reduced density operators $\hat{\rho}_S$ and $\hat{\rho}_R$ can be derived as

$$\hat{\rho}_S(t) = Tr_R \hat{\rho}(t) \quad (3.41)$$

$$\hat{\rho}_R(t) = Tr_S \hat{\rho}(t) \quad (3.42)$$

In particular, $\hat{\rho}_S(t)$ can be given in terms of the basis $|\Phi_i\rangle$ in the form

$$\hat{\rho}_S(t) = \sum_{ij} \rho_{ij}(t) |\Phi_i\rangle \langle \Phi_j| \quad (3.43)$$

where

$$\begin{aligned} \rho_{ij}(t) &= Tr[\hat{\rho}(t) |\Phi_j\rangle \langle \Phi_i|] \\ &= \langle \Phi_i | \hat{\rho}_S(t) | \Phi_j \rangle \end{aligned} \quad (3.44)$$

Time evolution of the density operator $\hat{\rho}(t)$ is governed by the unitary time evolution operator $\hat{U}(t, t_0)$, such that

$$\hat{\rho}(t) = \hat{U}(t, t_0) \hat{\rho}(t_0) \hat{U}^\dagger(t, t_0) \quad (t \geq t_0) \quad (3.45)$$

and as an assumption we take the system and reservoir are uncoupled at the initial time t_0

$$\hat{\rho}(t_0) = \hat{\rho}_R(t_0) \times \hat{\rho}_S(t_0) \quad (3.46)$$

We can now determine the $\rho_{ij}(t)$ such as

$$\begin{aligned} \rho_{ij}(t) &= Tr[\hat{U}(t, t_0) \hat{\rho}(t_0) \hat{U}^\dagger(t, t_0) |\Phi_i\rangle \langle \Phi_j|] \\ &= Tr[\hat{U}(t, t_0) \hat{\rho}_R(t_0) \hat{\rho}_S(t_0) \hat{U}^\dagger(t, t_0) |\Phi_i\rangle \langle \Phi_j|] \\ &= Tr[\hat{U}^\dagger(t, t_0) |\Phi_i\rangle \langle \Phi_j| \hat{U}(t, t_0) \hat{\rho}_R(t_0) \hat{\rho}_S(t_0)] \end{aligned} \quad (3.47)$$

Now, making representation of the operator $\hat{U}^\dagger(t, t_0) |\Phi_i\rangle \langle \Phi_j| \hat{U}(t, t_0)$ in the basis $|\Phi_p\rangle$ yields

$$\hat{U}^\dagger(t, t_0) |\Phi_i\rangle \langle \Phi_j| \hat{U}(t, t_0) = \sum_{pq} \hat{C}_{ijpq}^{(R)}(t, t_0) |\Phi_p\rangle \langle \Phi_q| \quad (3.48)$$

where the coefficients $\hat{C}_{ijpq}^{(R)}(t, t_0)$ are c-numbers with respect to the Hilbert space, S, but operators in the Hilbert space, R. By substituting this expression in the Eq. (3.47) gives

$$\rho_{ji}(t) = Tr \sum_{pq} \hat{C}_{ijpq}^{(R)}(t, t_0) |\Phi_p\rangle \langle \Phi_q| \hat{\rho}_R(t_0) \hat{\rho}_S(t_0) \quad (3.49)$$

and according to the Eq. (3.43)

$$\begin{aligned} \rho_{ji}(t) &= Tr \sum_{pq,lm} \hat{C}_{ijpq}^{(R)}(t, t_0) \hat{\rho}_R(t_0) |\Phi_p\rangle \langle \Phi_q| \rho_{lm}(t_0) |\Phi_l\rangle \langle \Phi_m| \quad (3.50) \\ &= Tr \sum_{pq,lm} \hat{C}_{ijpq}^{(R)}(t, t_0) \hat{\rho}_R(t_0) |\Phi_p\rangle \langle \Phi_q| \rho_{lm}(t_0) |\Phi_l\rangle \langle \Phi_m| \rho_{lm}(t_0) \\ &= Tr \sum_{pq} \hat{C}_{ijpq}^{(R)}(t, t_0) \hat{\rho}_R(t_0) \sum_{lm} \langle \Phi_m| \rho_{lm}(t_0) |\Phi_p\rangle \langle \Phi_q| |\Phi_l\rangle \\ &= Tr \sum_{pq} \hat{C}_{ijpq}^{(R)}(t, t_0) \rho_{lm}(t_0) |\Phi_p\rangle \langle \Phi_q| \\ &= Tr \sum_{pq} \hat{C}_{ijpq}^{(R)}(t, t_0) \rho_{qp}(t_0) \\ &= Tr \sum_{pq} G_{ijpq}(t, t_0) \rho_{qp}(t_0) \end{aligned}$$

where we have written

$$G_{ijpq}(t, t_0) = Tr_R \hat{C}_{ijpq}^{(R)}(t, t_0) \hat{\rho}_R(t_0) \quad (3.51)$$

as the Green function for the time evolution of the system. As an example of the regression theorem, we are going to use Green function $G(t, t_0)$ to calculate the single time expectation value of any dynamical variable \hat{M} belonging to system S. Thus we have

$$\begin{aligned} \langle \hat{M}(t) \rangle &= Tr[\hat{M}\rho(t)] \quad (3.52) \\ &= Tr_S[\hat{M}\hat{\rho}_S(t)] \\ &= Tr_S\left[\sum_i |\Phi_i\rangle \langle \Phi_i| \hat{M} \sum_j |\Phi_j\rangle \langle \Phi_j| \hat{\rho}_S(t)\right] \\ &= \sum_{ij} \langle \Phi_i | \Phi_i \rangle \langle \Phi_i | \hat{M} | \Phi_j \rangle \langle \Phi_j | \hat{\rho}_S(t) | \Phi_i \rangle \\ &= \sum_{ij} \hat{M}_{ij} \hat{\rho}_{ji}(t) \end{aligned}$$

where $M_{ij} = \langle \Phi_i | M | \Phi_j \rangle$ is the matrix representation of the \hat{M} . Let us use Eq. (3.43) for $\hat{\rho}_{ji}(t)$

$$\langle \hat{M}(t) \rangle = \sum_{ij} \sum_{pq} \hat{M}_{ij} G_{ijpq}(t, t_0) \hat{\rho}_{qp}(t_0) \quad (3.53)$$

The Green function $G_{ijpq}(t, t_0)$ allows the expectation value of \hat{M} at any time t to be determined.

To consider another example of the regression theorem, let \hat{L} , \hat{M} , \hat{N} be the dynamical variables belonging to the system S and suppose we wish to evaluate the multi time expectation values of $\langle \hat{L}(t_0) \hat{M}(t) \hat{N}(t_0) \rangle$ with $t \geq t_0$ then since

$$\hat{M}(t) = U^\dagger(t, t_0) \hat{M}(t_0) \hat{U}(t, t_0) \quad (3.54)$$

we write that the equation and using the unit projectors yields

$$\begin{aligned} \langle \hat{L}(t_0) \hat{M}(t) \hat{N}(t_0) \rangle &= Tr[\hat{L}(t_0) \hat{M}(t) \hat{N}(t_0) \hat{\rho}(t_0)] \\ &= Tr\left[\sum_{ijklmn} L(t_0) |\Phi_k\rangle \langle \Phi_k| U^\dagger(t, t_0) |\Phi_i\rangle \langle \Phi_i| M(t_0) \right. \\ &\quad \times \left. |\Phi_j\rangle \langle \Phi_j| U(t, t_0) |\Phi_l\rangle \langle \Phi_l| N(t_0) |\Phi_m\rangle \langle \Phi_m| \hat{\rho}_R(t_0) \hat{\rho}_S(t_0) |\Phi_n\rangle \langle \Phi_n| \right] \\ &= Tr_R \sum_{ijklmn} L_{nk} \langle \Phi_k | U^\dagger(t, t_0) | \Phi_i \rangle M_{ij} \langle \Phi_j | U(t, t_0) | \Phi_l \rangle N_{lm} \hat{\rho}_R(t_0) \hat{\rho}_{mn}(t_0) \end{aligned} \quad (3.55)$$

Finally, we use the expansion (3.48) for $U^\dagger(t, t_0) |\Phi_i\rangle \langle \Phi_j| U(t, t_0)$ and taking the trace over R as in Eq. (3.51) leads the equation

$$\begin{aligned} \langle \hat{L}(t_0) \hat{M}(t) \hat{N}(t_0) \rangle &= \sum_{ijklmnpq} L_{nk} \langle \Phi_k | \Phi_p \rangle \\ &\quad \times G_{ijpq}(t, t_0) \langle \Phi_q | \Phi_l \rangle M_{ij} N_{lm} \rho_{mn}(t_0) \\ &= \sum_{ijklmnpq} G_{ijpq}(t, t_0) L_{nk} M_{ij} \\ &\quad \times N_{lm} \hat{\rho}_{mn}(t_0) \end{aligned} \quad (3.56)$$

which is called the Quantum Regression Theorem, implies that the fluctuations regress in time like the macroscopic averages. In the interaction is efficiently turned on at time t_0 , or the coupled system is measured at that time, so that the states $\hat{\rho}_S(t_0)$ and $\hat{\rho}_R(t_0)$ are known separately. In this case the quantum regression theorem can be applied to evaluate the correlation functions.

3.3.2 The Fluctuation-Dissipation Theorem

It has been known by the work of Einstein on Brownian motion and molecular diffusion[35], and the work of Nyquist on thermal noise in resistors[36], that there exist a connectioning some physical systems between thermal fluctuations and the energy dissipation produced by an external disturbance. This connection was generalized by Collen and Welton[37] and has become known as fluctuaton-dissipation theorem. Another explanation of the theorem can be found in the Kubo's paper [38].

As an example of classical linear dissipative system, let us consider a physical system which has the equation of motion in the form

$$L \frac{dI}{dt} = -RI + \nu(t) \quad (3.57)$$

here L is the inductance, R is the resistor and the electromagnetic force (e.m.f.) $\nu(t)$ induced by the thermal motion of charges in the resistor.

By definition, $\langle \nu(t) \rangle = 0$ and the Eq. (3.57) can be integrated directly and we obtain

$$I(t) = I(0)e^{-\frac{Rt}{L}} + \frac{1}{L}e^{-\frac{Rt}{L}} \int_0^t e^{\frac{Rt'}{L}} \nu(t') dt' \quad (3.58)$$

If we calculate the mean of the each term, and make use of the fact that $\langle \nu(t) \rangle = 0$ we immediately find that

$$\langle I(t) \rangle = \langle I(0) \rangle e^{-\frac{Rt}{L}} \quad (3.59)$$

The average current therefore tends to be zero and its intial value is not important here. however, the mean squared current does not tend to zero, as we now show. Using the fact that, the current $I(0)$ at time $t = 0$ is not correlated with the fluctuating with the e.m.f. $\nu(t)$ at a later time t, so that

$$\langle I(0)\nu(t) \rangle = \langle I(0) \rangle \langle \nu(t) \rangle = 0 \quad (3.60)$$

if $t > 0$, we then have

$$\begin{aligned} \langle I^2(t) \rangle &= \langle I^2(0) \rangle e^{-\frac{2Rt}{L}} \\ &+ \frac{1}{L^2} e^{-\frac{2Rt}{L}} \int_0^t \int_0^t e^{\frac{R(t'+t'')}{L}} \langle \nu(t') \nu(t'') \rangle dt' dt'' \end{aligned} \quad (3.61)$$

Now $\langle \nu(t')\nu(t'') \rangle$ is the correlation function of the Johnson noise, which may be assumed to be stationary at a given temperature, so we may write

$$\langle \nu(t')\nu(t'') \rangle = \Gamma(t'' - t'). \quad (3.62)$$

Then, the substitution of new variables as

$$\begin{aligned} \frac{1}{2}(t' + t'') &= t_1, \\ t'' - t' &= t_2 \end{aligned}$$

we obtain $t' = t_1 - \frac{t_2}{2}$, $t'' = t_1 + \frac{t_2}{2}$ and the limits of the new variables are

$$\begin{aligned} 0 \leq t_1 \leq t_2 &\longrightarrow -2t_1 \leq t_2 \leq 2t_1 \\ \frac{1}{2}t \leq t_1 \leq t &\longrightarrow -2(t - t_1) \leq t_2 \leq 2(t - t_1). \end{aligned}$$

Finally we obtain the following formula

$$\begin{aligned} \langle I^2(t) \rangle &= \langle I^2(0) \rangle e^{-\frac{2Rt}{L}} + \frac{1}{L^2} e^{-\frac{2Rt}{L}} \left[\int_0^{\frac{t}{2}} dt_1 \int_{-2t_1}^{2t_1} dt_2 e^{\frac{2Rt_1}{L}} \Gamma(t_2) \right. \\ &\quad \left. + \int_{\frac{t}{2}}^t dt_1 \int_{-2(t-t_1)}^{2(t-t_1)} dt_2 e^{\frac{2Rt_1}{L}} \Gamma(t_2) \right]. \end{aligned} \quad (3.63)$$

Now the correlation function $\Gamma(t_2)$ always has a limited range governed by the random noise $\nu(t)$ and for the steady-state value of the $\langle I^2(t) \rangle$, we assume t to be very large. In this case, we approximated that

$$\int_{-\infty}^{\infty} \Gamma(t_2) dt_2 = \Phi(0) \quad (3.64)$$

where

$$\Phi(\omega) = \int_{-\infty}^{\infty} \Gamma(t_2) e^{i\omega t_2} dt_2 \quad (3.65)$$

is the spectral density of the Johnson noise. Then the equation simplifies to

$$\begin{aligned} \langle I^2(t) \rangle &= \langle I^2(0) \rangle e^{-\frac{2Rt}{L}} \\ &\quad + \frac{1}{L^2} e^{-\frac{2Rt}{L}} \int_0^t e^{\frac{2Rt_1}{L}} \Phi(0) dt_1 \\ &= \langle I^2(0) \rangle e^{-\frac{2Rt}{L}} + \frac{1}{2RL} (1 - e^{-\frac{2Rt}{L}}) \Phi(0). \end{aligned} \quad (3.66)$$

This reduces to

$$\langle I^2 \rangle = \frac{1}{2RL} \Phi(0) \quad (3.67)$$

for $t \rightarrow \infty$. Now $\frac{1}{2}\langle I^2 \rangle$ is the average energy stored in the inductance through the magnetic fields, and from the equipartition law it is equal to $\frac{1}{2}k_B T$ where k_B is the Boltzman constant. Then the last equation can be written in the form

$$\Phi(0) = 2k_B T R \quad (3.68)$$

which makes the connection between the magnitude of the voltage fluctuations $\Phi(0)$ and with the dissipation in the system represented by the resistance R .

Another example of a quantum system that obeys the fluctuation dissipation theorem is quantum noise in the quantum Langevin processes. Let the quantum system, S , in the form of a harmonic oscillator at frequency ω_0 , couple to a large number of reservoir, R , oscillators of various frequencies ω . The reservoir is so large that its state is not very much affected by the coupling to the system. We take the energy of coupled system and reservoir to be

$$\begin{aligned} \hat{H} &= \hbar\omega_0(\hat{a}^\dagger\hat{a} + \frac{1}{2}) + \sum_{\omega} \hbar\omega[\hat{A}^\dagger(\omega)\hat{A}(\omega) + \frac{1}{2}] \\ &+ \sum_{\omega} \hbar[g(\omega)\hat{a}^\dagger\hat{A}(\omega) + g^*(\omega)\hat{A}^\dagger(\omega)\hat{a}] \end{aligned} \quad (3.69)$$

in which \hat{a} , $\hat{A}(\omega)$ and their conjugates are creation and annihilation operators for the system and reservoir modes, respectively, and $g(\omega)$ is the freefrequency dependent coupling constant. The time evolution of $\hat{a}(t)$ and $\hat{A}(\omega, t)$ is governed by the Heisenberg equation of motion.

$$\begin{aligned} \dot{\hat{a}} &= -\frac{i}{\hbar}[\hat{a}(t), \hat{H}] \\ &= -i\omega_0\hat{a}(t) - i \sum_{\omega} g(\omega)\hat{A}(\omega, t) \end{aligned} \quad (3.70)$$

and

$$\begin{aligned} \dot{\hat{A}}(\omega, t) &= -\frac{i}{\hbar}[\hat{A}(\omega, t), \hat{H}] \\ &= -i\omega\hat{A}(\omega, t) - ig^*(\omega)\hat{a}(t) \end{aligned} \quad (3.71)$$

integrating last equation with respect to time

$$\dot{\hat{A}}(\omega, t) = \dot{\hat{A}}(\omega, 0)e^{-i\omega t} - ig^*(\omega)e^{-i\omega t} \int_0^t \hat{a}(t')e^{i\omega t'} dt' \quad (3.72)$$

and inserting into the Eq. (3.70) yields

$$\begin{aligned} \dot{\hat{a}}(t) = -i\omega_0 \hat{a}(t) & - i \sum_{\omega} g(\omega) \hat{A}(\omega, 0) e^{-i\omega t} \\ & - \int_0^t \sum_{\omega} |g(\omega)|^2 e^{-i\omega(t'-t)} \hat{a}(t') dt' \end{aligned} \quad (3.73)$$

If the reservoir oscillators are densely distributed, we can approximate the sum on ω by an integral. Then we may write the last term as

$$= \int_0^t dt' \int_0^{\infty} d\omega \eta(\omega) |g(\omega)|^2 e^{-i\omega(t'-t)} \hat{a}(t') \quad (3.74)$$

where $\eta(\omega)\delta(\omega)$ is the number of oscillators within the width of $\delta(\omega)$. Substituting $\omega = \omega_0 + \omega'$ and $t - t' = \tau$ makes the last term

$$\int_0^t d\tau \int_{-\infty}^{\infty} d\omega' \eta(\omega_0 + \omega') |g(\omega_0 + \omega')|^2 e^{-i\omega'\tau} a(t - \tau) e^{-i\omega_0\tau} \quad (3.75)$$

and the combination $a(t - \tau)e^{-i\omega_0\tau}$ is a slowly varying function of τ . By the Wiener-Khintchine theorem the ω' -integral gives

$$\frac{1}{2\pi} \int_{-\omega_0}^{\infty} \eta(\omega_0 + \omega') |g(\omega_0 + \omega')|^2 e^{-i\omega'\tau} d\omega' = \Gamma(\tau) \quad (3.76)$$

which plays the role of a memory function for the reservoir. the memory or correlation time is determined by the spectral width of $\eta(\omega_0 + \omega') |g(\omega_0 + \omega')|^2$ and this memory is very short if the frequency spread is great. In this case we may approximate that

$$\begin{aligned} \Gamma(\tau) &= \frac{1}{2\pi} \int_{-\infty}^{\infty} \eta(\omega_0 + \omega') |g(\omega_0 + \omega')|^2 e^{-i\omega'\tau} d\omega' \\ &\approx \eta(\omega_0) |g(\omega_0)|^2 \frac{1}{2\pi} \int_{-\infty}^{\infty} e^{-i\omega'\tau} d\omega' \\ &\approx \eta(\omega_0) |g(\omega_0)|^2 \delta(\tau) \end{aligned} \quad (3.77)$$

so $\Gamma(\tau)$ reduces to a $\delta(\tau)$ function in the sense of Markov approximation. With the help of Eq. (3.76) we write the Eq. (3.75) as

$$\int_0^t \sum_{\omega} |g(\omega)|^2 e^{i\omega t' - t} \hat{a}(t') dt' = 2\pi \int_0^t \Gamma(t) \hat{a}(t - \tau) e^{-i\omega_0\tau} d\tau \quad (3.78)$$

in which $\Gamma(\tau)$ is a strongly peaked ($\delta(\tau)$ – like function) whereas $\hat{a}(t - \tau)e^{-i\omega_0\tau}$ varies slowly with τ so we may approximate that

$$\begin{aligned} \int_0^t \sum_{\omega} |g(\omega)|^2 e^{i\omega t' - t} \hat{a}(t') dt' &= 2\pi \hat{a}(t) \int_0^t \Gamma(\tau) d\tau \\ &= \pi \hat{a}(t) \eta(\omega_0) |g(\omega_0)|^2 \end{aligned} \quad (3.79)$$

and setting

$$\pi \eta(\omega_0) |g(\omega_0)|^2 = \kappa \quad (3.80)$$

which has the dimension of frequency or rate, Eq. (3.75) becomes

$$\dot{\hat{a}}(t) = (-i\omega_0 - \kappa)\hat{a}(t) - \hat{F}(t) \quad (3.81)$$

where

$$\hat{F}(t) = i \sum_{\omega} g(\omega) \hat{A}(\omega, 0) e^{i\omega t} \quad (3.82)$$

This equation of motion for $\hat{a}(t)$ has precisely the form of Langevin equation with damping κ and $\hat{F}(t)$ describes a form of quantum noise. In this case, the spectral density function of the quantum noise determines the damping of the system, which is another example of the fluctuation-dissipation theorem.

3.3.3 Two-Level System With Little Dephasing

In this section, a general discussion of using quantum regression theorem and the fluctuation-dissipation theorem is given by introducing the two-level system. We describe the two-level system as in Figure 3.1 that has spontaneous emission, with the Hamiltonian

$$\begin{aligned} \hat{H} = \hbar\omega_{eg}\hat{\sigma}_{ee} &+ \hbar\omega_L\hat{a}^\dagger\hat{a} + \sum_k \hbar\omega_k\hat{a}^\dagger\hat{a} + \sum_k \hbar g_k(\hat{a}_k^\dagger\hat{\sigma}_{ge} + \hat{\sigma}_{eg}\hat{a}_k) \\ &+ \hbar g(\hat{a}^\dagger\hat{\sigma}_{ge} + \hat{\sigma}_{eg}\hat{a}) \end{aligned} \quad (3.83)$$

where \hat{a} and \hat{a}^\dagger are creation and annihilation operators while $\hat{\sigma}_{ge} = |g\rangle\langle e|$ and $\hat{\sigma}_{eg} = |e\rangle\langle g|$ are atomic projector operators. g_k and g correspond to the coupling between

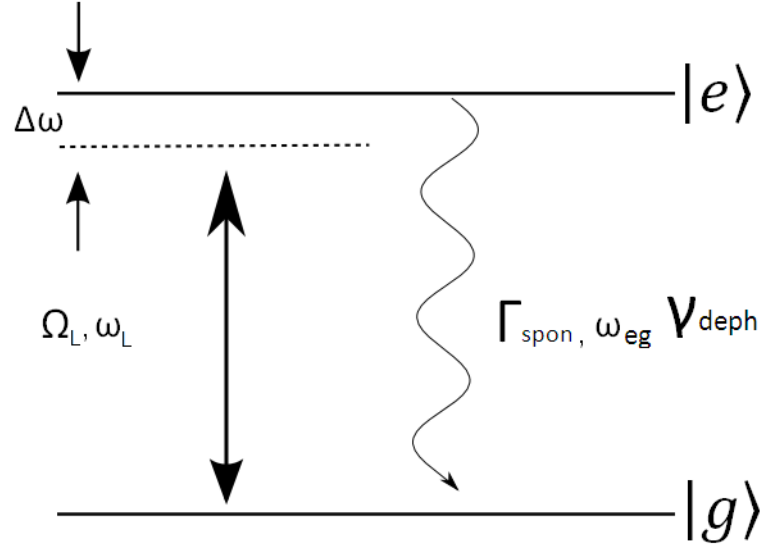


Figure 3.1: Two level system with little dephasing

the two-level emitter and the vacuum mode k , and the laser field, respectively. To discuss the properties of the two-level atom, we use the Liouville equation, that we can introduce couplings with a reservoir like the decay of the atomic levels due to spontaneous emission. We write the Liouville equation as

$$\dot{\hat{\rho}} = -i\frac{i}{\hbar}[\hat{H}, \hat{\rho}] - \frac{1}{2}\{\hat{\Gamma}, \hat{\rho}\} \quad (3.84)$$

where $\langle i|\hat{\Gamma}|j\rangle = \gamma_i\delta_{ij}$ back to the two-level atom, we write Hamiltonian in this way,

$$\hat{H} = H_0 + H'$$

where

$$\hat{H}_0 \rightarrow \begin{pmatrix} E_g & 0 \\ 0 & E_e \end{pmatrix}$$

and

$$\hat{H}' \rightarrow -\vec{d}_{eg}\vec{E}_0 \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix}$$

Using this Hamiltonian for the Liouville equation, we get the time evolution ex-

pression for the density matrix elements in the following set of equations

$$\begin{aligned}\dot{\rho}_{gg} &= -\gamma_g \rho_{gg} + -\frac{i}{2} \Omega_L (\rho_{ge} - \rho_{eg}) \\ \dot{\rho}_{ee} &= -\gamma_e \rho_{ee} + -\frac{i}{2} \Omega_L (\rho_{ge} - \rho_{eg}) \\ \dot{\rho}_{ge} &= (i\omega_0 - \gamma_g) \rho_{gg} + -\frac{i}{2} \Omega_L (\rho_{ee} - \rho_{gg})\end{aligned}\quad (3.85)$$

called Optical Bloch equations of the system. Then to consider the fluctuations terms as a result of the dissipation induced by the spontaneous emission we use the Hamiltonian given Eq. (3.83) and Liouville equation

$$\dot{\hat{\sigma}} = -\frac{i}{\hbar} [\hat{\sigma}, \hat{H}]$$

we found the time evolution of the density matrices of the given two-level system as

$$\begin{aligned}\hat{\sigma}_{ee} &= \hat{\sigma}_{ee} \left(\sum_k \hbar g_k \hat{a}_k^\dagger + \hbar a^\dagger \right) - \left(\sum_k \hbar g_k \hat{a}_k + \hbar g a \right) \hat{\sigma}_{ge} \\ \hat{\sigma}_{gg} &= \hat{\sigma}_{ge} \left(\sum_k \hbar g_k \hat{a}_k + \hbar g a \right) - \hat{\sigma}_{gg} \left(\sum_k \hbar g_k \hat{a}_k^\dagger + \hbar a^\dagger \right) \\ \hat{\sigma}_{eg} &= \left(\sum_k \hbar g_k \hat{a}_k + \hbar g a \right) (\hat{\sigma}_{ee} - \hat{\sigma}_g)\end{aligned}\quad (3.86)$$

then $f(t)$, $g(t)$ and Γ_{spon} are calculated as a same procedure given in the previous section

$$\begin{aligned}f(t) &= -i \sum_k g_k \hat{a}_k(0) (\hat{\sigma}_{gg}(t) - \hat{\sigma}_{gg}(t) - \hat{\sigma}_{gg}(t)) e^{-i(\omega_k - \omega_l)t} \\ g(t) &= i \sum_k g_k \hat{a}_k^\dagger(0) \hat{\sigma}_{ge}(t) e^{i(\omega_k - \omega_l)t} - i \sum_k g_k \hat{\sigma}_{eg}(t) \hat{a}_k(0) e^{-i(\omega_k - \omega_l)t} \\ \frac{\Gamma_{spon}}{2} &= \sum_k g_k^2 \int_{-\infty}^t dt' e^{-i(\omega_k - \omega_l)(t-t')}\end{aligned}\quad (3.87)$$

then we write the time evolution of the system operators as

$$\begin{aligned}\frac{d}{dt} \begin{bmatrix} \hat{\sigma}_{eg}(t) \\ \hat{\sigma}_{ee}(t) - \hat{\sigma}_{gg}(t) \\ \hat{\sigma}_{ge}(t) \end{bmatrix} &= \begin{bmatrix} i\Delta\omega - \gamma_{tot} & -i\frac{\Omega_l}{2} & 0 \\ -i\Omega_l & -\Gamma_{spon} & i\Omega_L \\ 0 & i\frac{\Omega_l}{2} & -i\Delta\omega - \gamma_{tot} \end{bmatrix} \\ &\times \begin{bmatrix} \hat{\sigma}_{eg}(t) \\ \hat{\sigma}_{ee}(t) - \hat{\sigma}_{gg}(t) \\ \hat{\sigma}_{ge}(t) \end{bmatrix} + \begin{bmatrix} f^\dagger(t) \\ 2g(t) - \Gamma_{spon} \\ f(t) \end{bmatrix}\end{aligned}\quad (3.88)$$

where $\Delta\omega = \omega_{eg} - \omega_L$, $\gamma_{tot} = \frac{\Gamma_{spon}}{2} + \gamma_{deph}$ and $\hbar\Omega_L = 2\hbar g = 2\langle e\vec{r}\vec{D}\rangle$ denote laser detuning, coherence dephasing rate, and the Rabi frequency respectively. Eq. (3.88) reduces to the Bloch equation when the average over the dissipative modes taken, using $\langle f(t)\rangle = \langle g(t)\rangle = 0$ we get the Eq. (3.85).

Then the unnormalized second order correlation function $G^2(\tau) = \langle \hat{\sigma}_{eg}(t')\hat{\sigma}_{ee}(t' + \tau)\hat{\sigma}_{ge}(t') \rangle$ can be obtained from Eq. (3.88) by multiplying with $\hat{\sigma}_{eg}(t')$ from left and $\hat{\sigma}_{ge}(t')$ from right, and taking the average over all dissipative modes. Quantum regression theorem reveals $\langle \hat{\sigma}_{eg}(t')f(t)\hat{\sigma}_{ge}(t') \rangle = \langle \hat{\sigma}_{eg}(t')g(t)\hat{\sigma}_{ge}(t') \rangle = 0$. Introducing the variable $\tau = t - t'$, the equations becomes

$$\begin{aligned} \frac{d}{dt} \begin{bmatrix} \hat{G}_{eg}(\tau) \\ \hat{G}_{ee}(\tau) - \hat{G}_{gg}(\tau) \\ \hat{G}_{ge}(\tau) \end{bmatrix} &= \begin{bmatrix} i\Delta\omega - \gamma_{tot} & -i\frac{\Omega_L}{2} & 0 \\ -i\Omega_L & -\Gamma_{spon} & i\Omega_L \\ 0 & i\frac{\Omega_L}{2} & -i\Delta\omega - \gamma_{tot} \end{bmatrix} \\ &\times \begin{bmatrix} \hat{G}_{eg}(\tau) \\ \hat{G}_{ee}(\tau) - \hat{G}_{gg}(\tau) \\ \hat{G}_{ge}(\tau) \end{bmatrix} + \begin{bmatrix} 0 \\ -\Gamma_{spon}\langle \hat{\sigma}_{ee}(t') \rangle \\ 0 \end{bmatrix} \end{aligned} \quad (3.89)$$

in the above equations

$$\begin{aligned} \hat{G}_{eg}(\tau) &= \langle \hat{\sigma}_{eg}(t')\hat{\sigma}_{eg}(t)\hat{\sigma}_{ge}(t') \rangle \\ \hat{G}_{ee}(\tau) - \hat{G}_{gg}(\tau) &= \langle \hat{\sigma}_{eg}(t')(\hat{\sigma}_{ee}(t) - \hat{\sigma}_{gg}(t))\hat{\sigma}_{ge}(t') \rangle \\ \hat{G}_{ge}(\tau) &= \langle \hat{\sigma}_{eg}(t')\hat{\sigma}_{ge}(t)\hat{\sigma}_{ge}(t') \rangle \end{aligned} \quad (3.90)$$

Assuming that the correlation functions are calculated when system reached its steady state, we can find the initial values of the equations at $t = t'$

$$\begin{aligned} \hat{G}_{eg}(\tau = 0) &= \langle \hat{\sigma}_{ee}(t') \rangle \\ \hat{G}_{ee}(\tau) - \hat{G}_{gg}(\tau) &= 0 \\ \hat{G}_{ge}(\tau) &= 0 \end{aligned} \quad (3.91)$$

and using the relation that

$$\begin{aligned} \langle \hat{\sigma}_{ee}(t) \rangle + \langle \hat{\sigma}_{gg}(t) \rangle &= 1 \\ \Rightarrow \hat{G}_{ee}(\tau) + \hat{G}_{gg}(\tau) &= \langle \hat{\sigma}_{ee}(t') \rangle \end{aligned} \quad (3.92)$$

and the solution of the $\hat{G}_{ee}(\tau) - \hat{G}_{gg}(\tau)$ second order correlation function can be calculated as [17]

$$G^2(\tau) = \hat{G}_{ee}(\tau) = \langle \hat{\sigma}_{eg}(t') \hat{\sigma}_{ee}(t' + \tau) \hat{\sigma}_{ge}(t') \rangle \quad (3.93)$$

We now discuss the physical meaning of the correlation functions. The first-order correlation function is required for calculating the spectrum of the field. The second-order correlation function gives information about the photon statistics and describes photon bunching or antibunching. For the two-level system in which the dephasing rates much larger than the Rabi frequency, we write the Optical Bloch equations as,

$$\frac{d}{dt} \begin{bmatrix} \langle \hat{\sigma}_{ee}(t) \rangle \\ \langle \hat{\sigma}_{gg}(t) \rangle \end{bmatrix} = \begin{bmatrix} -\Gamma_{spon} - \frac{\Omega_L^2}{\gamma_{tot}} & \frac{\Omega_L^2}{\gamma_{tot}} \\ \Gamma_{spon} + \frac{\Omega_L^2}{\gamma_{tot}} & -\frac{\Omega_L^2}{\gamma_{tot}} \end{bmatrix} \begin{bmatrix} \langle \hat{\sigma}_{ee}(t) \rangle \\ \langle \hat{\sigma}_{gg}(t) \rangle \end{bmatrix} \quad (3.94)$$

Finally we get the $G^2(\tau)$ as

$$\frac{d}{dt} G^2(\tau) = -(\Gamma_{spon} + \frac{\Omega_L^2}{\gamma_{tot}}) G^2(\tau) + (\frac{\Omega_L^2}{2\gamma_{tot}}) \langle \hat{\sigma}_{ee}(t) \rangle \quad (3.95)$$

substituting $\langle \hat{\sigma}_{ee}(t) \rangle = \langle \hat{\sigma}_{ee}(t) \rangle_{ss} = \Omega_L^2 / 2\gamma_{tot} \Gamma_{spon} + \frac{\Omega_L^2}{\gamma_{tot}}$ solving for the initial condition $G^2(\tau = 0) = 0$ the normalized second order correlation function is found as

$$g^2(\tau) = 1 - e^{-(\Gamma_{spon} + \frac{\Omega_L^2}{\gamma_{tot}})\tau} \quad (3.96)$$

and means that a relative detection probability of a photon at time τ given that another photon detection has occurred at time 0. In this case, at $\tau = 0$ the detection of one photon sets the atom in its ground state, and a second photon cannot be detected until the atom has been reexcited so we just predicted that the probability for detecting two photons is just the probability for detecting the first photon, multiplied by the probability for detecting second photon at time $t = \tau$, given that the atom was in its ground state at $t = 0$.

3.4 General Lindblad form of the Master Equation

Dissipative processes imply irreversibility, which means a preferred direction in time. Therefore, it is generally thought that a rigorous introduction of dissipation into a

quantum system can be done using quantum dynamical semigroups and most general form of these semigroups given by Lindblad. In quantum optics, master equation in the Lindblad form is the most general type of Markovian master equation describing non-unitary evolution of the density matrix ρ that is trace preserving and completely positive for any initial condition. According to the theory of Lindblad[45], the most general quantum Markovian master equation for the density operator ρ has the following form:

$$\dot{\rho} = -\frac{i}{\hbar}[H, \rho] + \sum_{n,m} h_{n,m}(-\rho L_m^\dagger L_n - L_m^\dagger L_n \rho + 2L_n \rho L_m^\dagger) \quad (3.97)$$

where H is a Hamiltonian of the system and L_m are the arbitrary operators on the Hilbert space and $h_{n,m}$ are constants which determine the dynamics of the system. Here $h_{n,m}$ must be positive to guarantee that the master equation is trace preserving and positive. The terms $-L_m^\dagger L_n \rho$ and $-\rho L_m^\dagger L_n$ in the Lindblad form of the master equation describe the loss of population from the current states, $L_n \rho L_m^\dagger$ term describe the gain of population. In this section we described Lindblad type of master equation which will have essential role of quantum trajectory approach that we will discuss later. As a preparation of next discussions it is useful here to derive the master equation for the reduced density operator

$$\dot{\rho} = L\rho(t) \quad (3.98)$$

where L is generally called Liouvillian or superoperator.

Chapter 4

ANALYTICAL INVESTIGATIONS OF THE MASTER EQUATION

After analyzing two-level systems we now move on to three level systems for semi-classical and fully quantum mechanical point of views. Two level systems well known in physics for spontaneous and stimulated emissions, Rabi oscillations, superradiance etc. Three level systems become interesting for their quantum optical effects such as electromagnetically induced transparency, coherent population trapping etc. In this section we will discuss the analytical investigation of the master equation under the special case of three level atoms.

We also investigate here the generation and evolution of continuous-variable entanglement for an incoherently pumped single-atom cavity electrodynamics system. Continuous-variable entanglement is important for quantum information theory due to the simplicity and high efficiency in the generation, manipulation and detection of optical states. It is shown that the intensity of the driving field can influence effectively the entanglement between two cavity modes.

4.1 Microwave-driven Λ -type Three Level Atom

We first consider a typical problem for a two level system with a small state space to illustrate how equations of motions derived. Suppose that a three-level atom in a lambda configuration is driven by Microwave as shown in Figure 4.1. In order to include the effects of spontaneous emission, a master equation for the density matrix required.

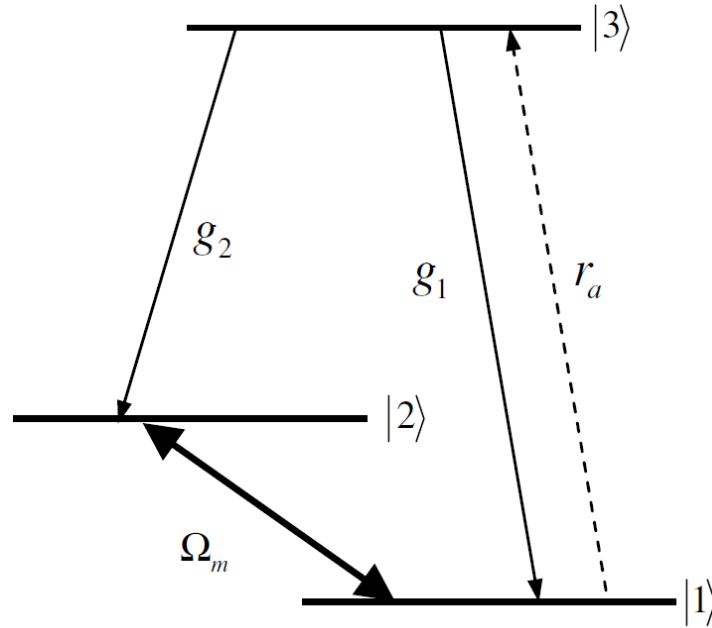


Figure 4.1: Schematic diagram of the three-level atomic system in a Λ configuration trapped in a doubly resonant cavity

4.1.1 Model and the Master Equation

Single three-level atom trapped in a doubly resonant cavity and the atoms are pumped in the upper level $|3\rangle$ by an incoherent pump at rate r_a . Two non-degenerate cavity modes, ν_1 and ν_2 , independently interact with the dipole allowed transitions $|3\rangle \leftrightarrow |1\rangle$ (with resonance frequencies ω_{32}) and $|3\rangle \leftrightarrow |2\rangle$ (with resonance frequencies ω_{32}), respectively, which are treated full quantum mechanically for the corresponding coupling constants g_1 and g_2 . Electric-dipole forbidden transition, $|2\rangle \leftrightarrow |1\rangle$, can be induced by applying strong external magnetic field for a magnetic-dipole allowed transition, which can be treated semiclassically in the orders of the Rabi frequency [39][40][41].

In the Schrödinger picture, ($\hbar = 1$)

$$\begin{aligned}
 H_0 &= \sum_{i=2,3} \omega_i |i\rangle \langle i| \\
 H_{int}^S &= -(\Omega_m e^{-i\omega_m t} |2\rangle \langle 1| + h.c.) + (g_1 a_1 |3\rangle \langle 1| + g_2 a_2 |3\rangle \langle 2| + h.c)
 \end{aligned} \tag{4.1}$$

turning into the interaction picture yields

$$\begin{aligned} |2\rangle &= c_2 = \tilde{c}_2 e^{-i\omega_m t} \\ \langle 1| &= c_1 = \tilde{c}_1 e^{i\omega_m t} \end{aligned}$$

and setting $\omega_m = \omega_2 - \omega_1$, we get the first term of the interaction Hamiltonian as

$$\begin{aligned} \Omega_m e^{-i\omega_m t} c_2 c_1^\dagger &= \Omega_m e^{-i(\omega_2 - \omega_1)t} c_2 e^{i\omega_2 t} c_1^\dagger e^{-i\omega_1 t} \\ &= \Omega_m c_2 c_1^\dagger \\ &= \Omega_m |2\rangle \langle 1| \end{aligned}$$

to use the same method we now write

$$\begin{aligned} a_1 &= \tilde{a}_1 e^{-i\nu_1 t} \\ |3\rangle &= c_3 = \tilde{c}_3 e^{-i\omega_3 t} \\ \langle 1| &= c_1 = \tilde{c}_1 e^{-i\omega_1 t} \end{aligned}$$

and the second term of the interaction Hamiltonian is

$$\begin{aligned} g_1 a_1 |3\rangle \langle 1| &= g_1 a_1 e^{-i\nu_1 t} c_3 e^{i\omega_3 t} c_1^\dagger e^{-i\omega_1 t} \\ &= g_1 a_1 e^{i(\nu_1 + \omega_1 - \omega_3)t} c_3 c_1^\dagger \\ &= g_1 a_1 |3\rangle \langle 1| \end{aligned}$$

and the third term can be found same as the others, so

$$g_2 a_2 |3\rangle \langle 2| \rightarrow g_2 a_2 |3\rangle \langle 2|$$

finally interaction Hamiltonian in the Rotating Wave Approximation takes the form

$$H_{int} = -(\Omega_m |2\rangle \langle 1| + h.c.) + (g_1 a_1 |3\rangle \langle 1| + g_2 a_2 |3\rangle \langle 2| + h.c.) \quad (4.2)$$

where *h.c.* means the Hermitian conjugate. Here state $|1\rangle$ is the ground state and for the sake of simplicity we assumed $\omega_1 = 0$. a_1 and a_2 are annihilation operators for the two non-degenerate modes of the cavity field, with frequencies ν_1 and ν_2 . g_1 and

g_2 are the atom-field coupling constants for the transitions $|3\rangle \leftrightarrow |1\rangle$ and $|3\rangle \leftrightarrow |2\rangle$, respectively. $\Omega_m = |\Omega_m|e^{-i\phi}$ denotes the Rabi frequency of the classical field with the frequency $\omega_m = \omega_2 - \omega_1$ at the resonance with the transition $|2\rangle \leftrightarrow |1\rangle$. Here ϕ is the phase of the applied classical field. Using the density matrix formalism, the reduced density equations of the cavity field $\hat{\rho}_f$ can be obtained by taking a trace over the atom

$$\dot{\rho}_f = -\frac{i}{\hbar} Tr_{atom}[H_{int}, \rho_{a-f}] \quad (4.3)$$

where

$$H_{int} = \begin{pmatrix} 0 & -\Omega_m^* & g_1 a_1^\dagger \\ -\Omega_m & 0 & g_2 a_2^\dagger \\ g_1 a_1 & g_2 a_2 & 0 \end{pmatrix} \quad (4.4)$$

$$\rho_{a-f} = \begin{pmatrix} \rho_{11} & \rho_{12} & \rho_{13} \\ \rho_{21} & \rho_{22} & \rho_{23} \\ \rho_{31} & \rho_{32} & \rho_{33} \end{pmatrix} \quad (4.5)$$

then calculating diagonal elements of the commutation relation between H_{int} and ρ_{a-f} and trace over the atom parameters yields,

$$\begin{aligned} [H_{int}, \rho_{a-f}] &= \begin{pmatrix} -\rho_{12}\Omega_m + \rho_{13}g_1a_1 & - & - \\ - & -\rho_{21}\Omega_m^* + \rho_{23}g_2a_2 & - \\ - & - & \rho_{31}g_1a_1^\dagger + \rho_{32}g_2a_2^\dagger \end{pmatrix} \quad (4.6) \\ &= -\Omega_m^* \rho_{21} + g_1 a_1^\dagger \rho_{31} + \rho_{12} \Omega_m - \rho_{13} g_1 a_1 - \Omega_m \rho_{12} + g_2 a_2^\dagger \rho_{32} + \rho_{21} \Omega_m^* \\ &\quad - \rho_{23} g_2 a_2 + g_1 a_1 \rho_{13} + g_2 a_2 \rho_{23} - \rho_{31} g_1 a_1^\dagger - \rho_{32} g_2 a_2^\dagger \\ &= g_1 (a_1^\dagger \rho_{31} - \rho_{13} a_1 + a_1 \rho_{13} - \rho_{31} a_1^\dagger) + g_2 (a_2^\dagger \rho_{32} - \rho_{32} a_2 + a_2 \rho_{23} - \rho_{32} a_2^\dagger) \\ &= g_1 \{ [a_1^\dagger, \rho_{31}] + [a_1, \rho_{13}] \} + g_2 \{ [a_2^\dagger, \rho_{32}] + [a_2, \rho_{23}] \} \\ \dot{\rho}_f &= -ig_1 [a_1^\dagger, \rho_{31}] - ig_1 [a_2^\dagger, \rho_{32}] + h.c. \end{aligned}$$

where ρ_{a-f} is the full atom-field density operator $\rho_{31} = \langle 3 | \rho_{a-f} | 1 \rangle$, $\rho_{32} = \langle 3 | \rho_{a-f} | 2 \rangle$. According to the standard methods of laser theory in [22], to evaluate ρ_{31} and ρ_{32} , using the same expressions as above and the relaxation matrix $\langle n | \Gamma | m \rangle = \gamma_n \delta_{nm}$,

which is

$$\Gamma = \begin{pmatrix} \gamma_1 & 0 & 0 \\ 0 & \gamma_2 & 0 \\ 0 & 0 & \gamma_3 \end{pmatrix} \quad (4.7)$$

Then the Liouville equation takes the form of

$$\dot{\rho} = -\frac{i}{\hbar}[H_{int}, \rho] - \frac{1}{2}\{\Gamma, \rho\} \quad (4.8)$$

where $\{\Gamma, \rho\} = \Gamma\rho + \rho\Gamma$. Finally we find the matrix elements of the $[H_{int}, \rho]$ term

$$\begin{aligned} b_{11} &= -\Omega_m^* \rho_{21} + g_1 a_1^\dagger \rho_{31} + \rho_{12} \Omega_m - \rho_{13} g_1 a_1 \\ b_{12} &= -\Omega_m^* \rho_{22} + g_1 a_1^\dagger \rho_{32} + \rho_{11} \Omega_m^* - \rho_{13} g_2 a_2 \\ b_{13} &= -\Omega_m^* \rho_{23} + g_1 a_1^\dagger \rho_{33} + \rho_{11} g_1 a_1^\dagger - \rho_{12} g_2 a_2^\dagger \\ b_{21} &= -\Omega_m \rho_{11} + g_2 a_2^\dagger \rho_{31} + \rho_{22} \Omega_m - \rho_{23} g_1 a_1 \\ b_{22} &= -\Omega_m \rho_{12} + g_2 a_2^\dagger \rho_{32} + \rho_{21} \Omega_m^* - \rho_{23} g_2 a_2 \\ b_{23} &= -\Omega_m \rho_{13} + g_2 a_2^\dagger \rho_{33} - \rho_{21} g_1 a_1^\dagger - \rho_{22} g_2 a_2^\dagger \\ b_{31} &= g_1 a_1 \rho_{11} + g_2 a_2 \rho_{21} + \rho_{32} \Omega_m - \rho_{33} g_1 a_1 \\ b_{32} &= g_1 a_1 \rho_{12} + g_2 a_2 \rho_{22} + \rho_{32} \Omega_m^* - \rho_{33} g_2 a_2 \\ b_{33} &= g_1 a_1 \rho_{13} + g_2 a_2 \rho_{23} - \rho_{31} g_1 a_1^\dagger - \rho_{32} g_2 a_2^\dagger \end{aligned}$$

and the second term $\{\Gamma, \rho\}$ can be calculated as

$$\{\Gamma, \rho\} = \begin{pmatrix} 2\gamma_1 \rho_{11} & (\gamma_1 + \gamma_2) \rho_{12} & \gamma_1 \rho_{13} \\ (\gamma_1 + \gamma_2) \rho_{21} & \gamma_2 \rho_{22} & (\gamma_2 + \gamma_3) \rho_{23} \\ (\gamma_1 + \gamma_3) \rho_{31} & (\gamma_2 + \gamma_3) \rho_{32} & 2\gamma_3 \rho_{33} \end{pmatrix}.$$

Then using the Liouville equation we find the density matrices, diagonal terms

$$\begin{aligned} \dot{\rho}_{11} &= -\gamma_1 \rho_{11} + i\Omega_m^* \rho_{21} - i g_1 a_1^\dagger \rho_{31} - i\Omega_m \rho_{12} + i\rho_{13} g_1 a_1 \\ \dot{\rho}_{22} &= -\gamma_2 \rho_{22} + i\Omega_m \rho_{12} - i g_2 a_2^\dagger \rho_{32} - i\rho_{21} \Omega_m^* + i\rho_{23} g_2 a_2 \\ \dot{\rho}_{33} &= -\gamma_3 \rho_{33} - i g_1 a_1 \rho_{13} - i g_2 a_2 \rho_{23} + i\rho_{31} g_1 a_1^\dagger + i\rho_{32} g_2 a_2^\dagger \end{aligned}$$

and using the $\rho_{21}^* = \rho_{12}$, $\rho_{32}^* = \rho_{23}$, and $\rho_{31}^* = \rho_{13}$ relations we find the other terms.

Then we have the equations for density matrices in the following form

$$\begin{aligned}\dot{\rho}_{31} &= -\frac{\gamma_3}{2}\rho_{31} - ig_1 a_1 \rho_{11} - ig_2 a_2 \rho_{21} - i\rho_{32}\Omega_m + i\rho_{33}g_1 a_1 \\ \dot{\rho}_{32} &= -\frac{\gamma_2}{2}\rho_{32} - ig_1 a_1 \rho_{12} - ig_2 a_2 \rho_{22} - i\rho_{32}\Omega_m^* + i\rho_{33}g_2 a_2\end{aligned}$$

The terms including g_1 and g_2 atom-field coupling constants, relatively smaller than the Ω_m Rabi frequency terms, in this case setting the small terms as a zeroth-order density matrices as

$$\begin{aligned}\dot{\rho}_{31} &= -\frac{\gamma_3}{2}\rho_{31} - ig_1 a_1 \rho_{11}^{(0)} - ig_2 a_2 \rho_{21}^{(0)} - i\rho_{32}\Omega_m + i\rho_{33}^{(0)}g_1 a_1 \\ \dot{\rho}_{32} &= -\frac{\gamma_2}{2}\rho_{32} - ig_1 a_1 \rho_{12}^{(0)} - ig_2 a_2 \rho_{22}^{(0)} - i\rho_{32}\Omega_m^* + i\rho_{33}^{(0)}g_2 a_2\end{aligned}\quad (4.9)$$

and the corresponding zeroth-order equations, $\rho_{ij}^{(0)}$,

$$\begin{aligned}\dot{\rho}_{22}^{(0)} &= -\gamma_{32}\rho_{33}^{(0)} + i\Omega_m\rho_{12}^{(0)} - i\Omega_m^*\rho_{21}^{(0)} \\ \dot{\rho}_{33}^{(0)} &= -\gamma_3\rho_{33}^{(0)} + r_a\rho_f \\ \dot{\rho}_{21}^{(0)} &= i\Omega_m\rho_{11}^{(0)} - i\rho_{22}^{(0)}\Omega_m \\ \dot{\rho}_{11}^{(0)} &= (\gamma_3 - \gamma_{32})\rho_{33}^{(0)} + i\Omega_m^*\rho_{21}^{(0)} - i\Omega_m\rho_{12}^{(0)} - r_a\rho_f\end{aligned}\quad (4.10)$$

This set of equations can be solved, for example, by first writing in the matrix form,

$$\dot{R} = -MR + A \quad (4.11)$$

with

$$\begin{pmatrix} \dot{\rho}_{22}^{(0)} \\ \dot{\rho}_{33}^{(0)} \\ \dot{\rho}_{21}^{(0)} \\ \dot{\rho}_{12}^{(0)} \\ \dot{\rho}_{11}^{(0)} \end{pmatrix} = \begin{pmatrix} 0 & \gamma_{32} & (i\Omega)^* & (i\Omega) & 0 \\ 0 & -\gamma_3 & 0 & 0 & 0 \\ -(i\Omega) & 0 & 0 & 0 & (i\Omega) \\ (i\Omega)^* & 0 & 0 & 0 & (-i\Omega)^* \\ 0 & (\gamma_3 - \gamma_{32}) & (-i\Omega)^* & (-i\Omega) & 0 \end{pmatrix} \begin{pmatrix} \rho_{22}^{(0)} \\ \rho_{33}^{(0)} \\ \rho_{21}^{(0)} \\ \rho_{12}^{(0)} \\ \rho_{11}^{(0)} \end{pmatrix} \quad (4.12)$$

and then integrating

$$\begin{aligned}R(t) &= \int_{-\infty}^t e^{-M(t-t')} A dt' \\ &= M^{-1}A\end{aligned}\quad (4.13)$$

yield the solution such as

$$\begin{pmatrix} 0 \\ -\frac{r_a \rho_f}{\gamma_3} \\ 0 \\ \frac{r_a \rho_f i \Omega^*}{i \Omega (-i \Omega)^* + i \Omega i \Omega^*} + \frac{r_a \rho_f (\gamma_{32}((-i \Omega)^* + i \Omega^*) - \gamma_3 i \Omega^*)}{i \Omega \gamma_3 ((-i \Omega)^* + i \Omega^*)} \\ 0 \end{pmatrix} \quad (4.14)$$

then using in the Eq. (4.9), we get two coupled equation,

$$\dot{\rho}_{31} = -\frac{\gamma_3}{2} \rho_{31} - i g_1 a_1(0) - i g_2 a_2(0) - i \rho_{32} \Omega_m + i \left(\frac{r_a g_1 a_1 \rho_f}{\gamma_3} \right) g_1 a_1 \quad (4.15)$$

$$\begin{aligned} \dot{\rho}_{32} &= -\frac{\gamma_2}{2} \rho_{32} - i g_1 a_1 \left(\frac{-g_1 a_1 r_a \rho_f \gamma_{32}(\Omega)^*}{\gamma_3(\Omega)^* \Omega} \right. \\ &\quad \left. - \frac{i r_a \rho_f g_2 a_2}{\gamma_3} \right) - i g_2 a_2(0) - i \rho_{32} \Omega_m^* + i \left(\frac{r_a g_1 a_1 \rho_f}{\gamma_3} \right) g_2 a_2 \end{aligned} \quad (4.16)$$

using the same method for the solution, as we introduced as,

$$M^{-1}A = \begin{pmatrix} \frac{2\rho_f(a_1 g_1(\gamma_3 i r_a - 2i r_a \gamma_{32}) - 2i \Omega a_2 g_2 i r_a)}{\gamma_3^3 + 4i \Omega i \Omega^* \gamma_3} \\ \frac{2\rho_f(i \Omega a_2 g_2 \gamma_3 i r_a + a_1 g_1(i r_a \gamma_3 \gamma_{32} + 2i \Omega i r_a i \Omega^*))}{i \Omega \gamma_3(\gamma_3^2 + 4i \Omega i \Omega^*)} \end{pmatrix}$$

Finally, we get the expressions for the density matrices that we need to use in Eq. (4.6)

$$\dot{\rho}_{31} = \frac{2i \gamma_3 r_a \rho_f a_1 g_1 - 4i r_a \gamma_{32} \rho_f a_1 g_1 + 4\Omega r_a \rho_f a_2 g_2}{\gamma_3^2(\gamma_3 - 4|\Omega|^2)} \quad (4.17)$$

$$\dot{\rho}_{32} = \frac{2i \gamma_3 \Omega r_a \rho_f a_2 g_2 + 2r_a \gamma_3 \gamma_{32} \rho_f a_1 g_1 + 4i r_a |\Omega|^2 \rho_f a_1 g_1}{\Omega \gamma_3(\gamma_3^2 - 4|\Omega|^2)} \quad (4.18)$$

and

$$\dot{\rho}_{13} = \frac{-2i \gamma_3 r_a \rho_f a_1^\dagger g_1 + 4i r_a \gamma_{32} \rho_f a_1^\dagger g_1 + 4\Omega r_a \rho_f a_2^\dagger g_2}{\gamma_3^2(\gamma_3 - 4|\Omega|^2)} \quad (4.19)$$

$$\dot{\rho}_{23} = \frac{-2i \gamma_3 \Omega^* r_a \rho_f a_2^\dagger g_2 + 2r_a \gamma_3 \gamma_{32} \rho_f a_1^\dagger g_1 - 4i r_a |\Omega|^2 \rho_f a_1^\dagger g_1}{\Omega^* \gamma_3(\gamma_3^2 - 4|\Omega|^2)} \quad (4.20)$$

Finally, the equation of motion for the field density matrix ρ_f , master equation, can

be written as

$$\begin{aligned}
\dot{\rho}_f = & [\alpha_{11}(a_1^\dagger a_1 \rho_f - a_1 \rho_f a_1^\dagger) + \beta_{11}(a_1^\dagger \rho_f a_1 - \rho_f a_1 a_1^\dagger)] \\
& + \alpha_{22}(a_2^\dagger a_2 \rho_f - a_2 \rho_f a_2^\dagger) + \beta_{22}(a_2^\dagger \rho_f a_2 - \rho_f a_2 a_2^\dagger) \\
& + \alpha_{12}(a_1^\dagger a_2 \rho_f - a_2 \rho_f a_1^\dagger) + \alpha_{21}(a_2^\dagger a_1 \rho_f - a_1 \rho_f a_2^\dagger) \\
& + \beta_{12}(a_1^\dagger \rho_f a_2 - \rho_f a_2 a_1^\dagger) + \beta_{21}(a_2^\dagger \rho_f a_1 - \rho_f a_1 a_2^\dagger) + h.c.] \\
& - \kappa_1(a_1^\dagger a_1 \rho_f + \rho_f a_1^\dagger a_1 - 2a_1 \rho_f a_1^\dagger) \\
& - \kappa_2(a_2^\dagger a_2 \rho_f + \rho_f a_2^\dagger a_2 - 2a_2 \rho_f a_2^\dagger)
\end{aligned} \tag{4.21}$$

The coefficients α_{ij} and β_{ij} ($i, j = 1, 2$) are given in the Appendix A.

4.1.2 Quantum Entanglement Analysis of the System

Before studying the entanglement property of the two field modes based on our model [47], we recall that formally a system is entangled if it is nonseparable; i.e. the density operator for the state ρ_f cannot be written as a convex combination of product states

$$\rho_f = \sum_j p_j \rho_j^{(1)} \otimes \rho_j^{(2)} \tag{4.22}$$

with $p_j \geq 0$ and $\sum_j p_j = 1$. $\rho_j^{(1)}$ and $\rho_j^{(2)}$ are the normalized states of two field modes, respectively. Here, we use the simple inseparability criterion proposed in [46] for continuous variable states. The criterion is based on the calculation of the total variance of a pair of Einstein-Podolsky-Rosen (EPR) type operators. A maximally entangled continuous variable state can be expressed as a co-eigenstate of a pair of EPR type operators, such as $\hat{x}_1 + \hat{x}_2$ and $\hat{p}_1 - \hat{p}_2$. To be more general, we consider the following type of EPR-like operators:

$$\hat{u} = |c| \hat{x}_1 + \frac{1}{c} \hat{x}_2 \tag{4.23}$$

$$\hat{v} = |c| \hat{p}_1 - \frac{1}{c} \hat{p}_2 \tag{4.24}$$

where we assume a is an arbitrary (nonzero) real number. For any separable state, the total variance of a pair of EPR-like operators with the commutators $[\hat{x}_j, \hat{p}_{j'}] = i\delta_{jj'}$

$(j, j' = 1, 2)$ satisfies the the inequality

$$\langle(\Delta\hat{u})^2\rangle_\rho + \langle(\Delta\hat{v})^2\rangle_\rho \geq c^2 + \frac{1}{c^2} \quad (4.25)$$

to proof this theorem, we can directly calculate the total variance of the \hat{u} and \hat{v} operators using the decomposition of the density operator ρ , ($\rho = \sum_i p_i \rho_{i1}^{(1)} \otimes \rho_{i2}^{(2)}$), and finally get the following expression,

$$\begin{aligned} \langle(\Delta\hat{u})^2\rangle_\rho + \langle(\Delta\hat{v})^2\rangle_\rho &= \sum_i p_i (\langle\hat{u}^2\rangle_i + \langle\hat{v}^2\rangle_i) - \langle\hat{u}\rangle_\rho^2 - \langle\hat{v}\rangle_\rho^2 \\ &= \sum_i p_i (c^2 \langle\hat{x}_1^2\rangle_i + \frac{1}{c^2} \langle\hat{x}_2^2\rangle_i + c^2 \langle\hat{p}_1^2\rangle_i + \frac{1}{c^2} \langle\hat{p}_2^2\rangle_i) \\ &\quad + 2\frac{c}{|c|} (\sum_i p_i \langle\hat{x}_1\rangle_i \langle\hat{x}_2\rangle_i - \sum_i p_i \langle\hat{p}_1\rangle_i \langle\hat{p}_2\rangle_i) - \langle\hat{u}\rangle_\rho^2 - \langle\hat{v}\rangle_\rho^2 \\ &= \sum_i p_i (c^2 \langle(\Delta\hat{x}_1)^2\rangle_i + \frac{1}{c^2} \langle(\Delta\hat{x}_2)^2\rangle_i + c^2 \langle(\Delta\hat{p}_1)^2\rangle_i + \frac{1}{c^2} \langle(\Delta\hat{p}_2)^2\rangle_i) \\ &\quad + \sum_i p_i \langle\hat{u}\rangle_i^2 - (\sum_i p_i \langle\hat{u}\rangle_i)^2 + \sum_i p_i \langle\hat{v}\rangle_i^2 - (\sum_i p_i \langle\hat{v}\rangle_i)^2 \end{aligned} \quad (4.26)$$

According to this criterion, a state of the system is entangled if the sum of the quantum fluctuations of two Einstein-Podolsy-Rosen (EPR)-like operators \hat{u} and \hat{v} of the two modes satisfy the inequality

$$\langle(\Delta\hat{u})^2\rangle + \langle(\Delta\hat{v})^2\rangle < 2 \quad (4.27)$$

here

$$\hat{u} = \hat{x}_1 + \hat{x}_2 \quad (4.28)$$

$$\hat{v} = \hat{p}_1 - \hat{p}_2$$

Now we define inseparability parameter

$$\lambda = 2(c^2 \langle a_1^\dagger a_1 \rangle + \langle a_2^\dagger a_2 \rangle / c^2 + \text{sign}(c) \langle a_1 a_2 + a_1^\dagger a_2^\dagger \rangle) - \langle u \rangle^2 - \langle v \rangle^2 \quad (4.29)$$

where

$$c^2 = [(\langle a_2^\dagger a_2 \rangle - |\langle a_2 \rangle|^2) / (\langle a_1^\dagger a_1 \rangle - |\langle a_1 \rangle|^2)]^{\frac{1}{2}}$$

$$\text{sgn}(c) = \text{sgn}[\text{Re}\{\langle a_1 a_2 \rangle\} - \text{Re}\{\langle a_1 \rangle\} - \text{Re}\{\langle a_2 \rangle\} + \text{Im}\{\langle a_1 \rangle\} + \text{Im}\{\langle a_2 \rangle\}]$$

with $\hat{x}_j = (a_j + a_j^\dagger/\sqrt{2})$ and $\hat{p}_j = (a_j - a_j^\dagger/\sqrt{2}i)$ ($j = 1, 2$) are the quadrature operators for the two modes 1 and 2. For a general state, this is a sufficient criterion for entanglement for two-mode continuous-variable Gaussian states. Finally we can express the total variance of the operators \hat{u} and \hat{v} in terms of the operators a_j and a_j^\dagger and get

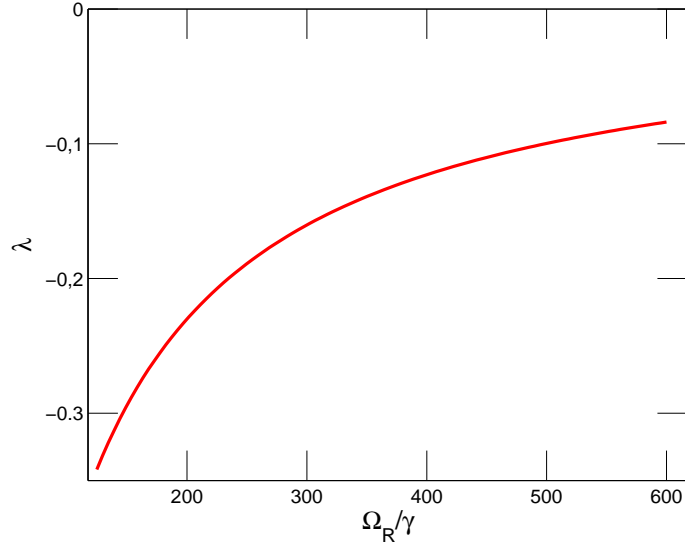
$$\begin{aligned} \langle(\Delta\hat{u})^2\rangle + \langle(\Delta\hat{v})^2\rangle &= 2[1 + \langle a_1^\dagger a_1 \rangle + \langle a_2^\dagger a_2 \rangle + \langle a_1^\dagger a_2^\dagger \rangle + \langle a_1 a_2 \rangle \\ &\quad - \langle a_1 \rangle \langle a_1^\dagger \rangle - \langle a_2 \rangle \langle a_2^\dagger \rangle - \langle a_1 \rangle \langle a_2 \rangle - \langle a_1^\dagger \rangle \langle a_2^\dagger \rangle] \end{aligned} \quad (4.30)$$

4.1.2.1 Entanglement of the Cavity Field

Here we calculate the moments with the help of the master equation and the fact that $\frac{d}{dt}\langle a(t) \rangle = \text{Tr}(\dot{\rho}_f a)$, the following sets of coupled equations for various moments are obtained as,

$$\begin{aligned} \frac{d\langle a_1 \rangle}{dt} &= (A_1 - \kappa)\langle a_1 \rangle - D_1\langle a_2^\dagger \rangle \\ \frac{d\langle a_2 \rangle}{dt} &= -(B_1 + \kappa)\langle a_2 \rangle + C_1\langle a_1^\dagger \rangle \\ \frac{d\langle a_1^\dagger a_1 \rangle}{dt} &= 2(A_1 - \kappa)\langle a_1^\dagger a_1 \rangle - D_1\langle a_1^\dagger a_2^\dagger \rangle - D_1^* \langle a_1 a_2 \rangle + 2A_1 \\ \frac{d\langle a_2^\dagger a_2 \rangle}{dt} &= -2(B_1 + \kappa)\langle a_2^\dagger a_2 \rangle + C_1\langle a_1^\dagger a_2^\dagger \rangle + C_1^* \langle a_1 a_2 \rangle \\ \frac{d\langle a_1 a_2 \rangle}{dt} &= C_1\langle a_1^\dagger a_1 \rangle - D_1\langle a_2^\dagger a_2 \rangle + (A_1 - B_1 - 2\kappa)\langle a_1 a_2 \rangle + C_1 \end{aligned}$$

where $\langle N_1 \rangle = \langle a_1^\dagger a_1 \rangle$ and $\langle N_2 \rangle = \langle a_2^\dagger a_2 \rangle$ are average photon numbers in modes 1 and 2. The steady-state solutions of this set of equations can be found by setting $\frac{d}{dt} = 0$. Then we have $\langle a_1 \rangle = \langle a_2 \rangle = 0$ and the second order solutions are obtained by using the fact that $\alpha_{ij}^* = -\alpha_{ij}$, $\beta_{ij}^* = -\beta_{ij}$, $\alpha_{ii}^* = -\alpha_{ii}$ and $\beta_{ii}^* = -\beta_{ii}$. Other expressions

Figure 4.2: Plot of λ versus Rabi Frequency of the classical field

for the moments can be calculated again using the same method in (4.11)

$$\begin{aligned} \langle a_1^\dagger a_1 \rangle &= \frac{\kappa C_1 D_1}{(-A_1 + B_1 + 2\kappa)(-A_1 B_1 + C_1 D_1 - \kappa A_1 + \kappa B_1 + \kappa^2)} \\ &+ \frac{\kappa(B_1 + \kappa)}{-A_1 + C_1 D_1 \kappa A_1 + \kappa B_1 + \kappa^2} - 1 \\ \langle a_2^\dagger a_2 \rangle &= \frac{\kappa C_1 C_1}{(A_1 + B_1 + 2\kappa)(-A_1 B_1 + C_1 D_1 - \kappa A_1 + \kappa B_1 + \kappa^2)} \\ \langle a_1 a_2 \rangle &= \frac{\kappa C_1 (B_1 + \kappa)}{(-A_1 + B_1 + 2\kappa)(-A_1 B_1 + C_1 D_1 - \kappa A_1 + \kappa B_1 + \kappa^2)} \end{aligned}$$

where the coefficients given by

$$\begin{aligned} A_1 &= \frac{3g_1'^2 r_a \Omega'^2}{(1 + \Omega'^2)(4 + \Omega'^2)} \\ B_1 &= \frac{g_2'^2 r_a}{(1 + \Omega'^2)} \\ C_1 &= \frac{ig_1'^2 g_2'^2 r_a \Omega' (\Omega'^2 - 2) e^{-i\phi}}{(1 + \Omega'^2)(4 + \Omega'^2)} \\ D_1 &= \frac{ig_1'^2 g_2'^2 r_a \Omega' e^{-i\phi}}{(1 + \Omega'^2)} \end{aligned}$$

In Figure 4.2 we have plotted the quantity λ versus the dimensionless quantity Rabi frequency, for pump phase $\phi = -\frac{\pi}{2}$, $r_a = 5$ kHz, $g_1 = g_2 = g = 43$ kHz, $\kappa_1 = \kappa_2 = \kappa = 3.85$ kHz, $\gamma = 20$ kHz. The parameter values used from the micromaser experiments given in [48]. It is clear that entanglement criterion is satisfied and two mode cavity field evolves into an entangled state. Reference [49] has showed that the bright two-color CV entanglement can be generated in the experiment which opens new potential applications in the quantum information science.

4.2 Numerical Method to Solve the Master Equation

The recent interest in quantum optics involving only a few photons and atoms in cavity QED systems can be solved numerical simulations. Here we discuss numerical method to solve equations of motion for a quantum optical systems starting from the Hamiltonian of the system and the coupling to the reservoirs. Given the density matrix ρ finding the expectation value of some operator a involves calculation of

$$\langle a \rangle = Tr(a\rho); \quad (4.31)$$

which is a linear functional acting on ρ to produce a number. Similarly, given a state ket $|\psi\rangle$; the expectation value of a is given by

$$\langle a \rangle = Tr(a|\psi\rangle\langle\psi|) = \langle\psi|a|\psi\rangle \quad (4.32)$$

Here we consider a cavity with resonant frequency ω_c and cavity rate κ containing a two-level atom with transition frequency ω_0 , field coupling strength g and spontaneous emission rate λ . The cavity is driven by a coherent (classical) field E . The Hamiltonian of the system is as given above,

$$H = (\omega_0 - \omega_L)\sigma_+\sigma_- + (\omega_0 - \omega_L)a^\dagger a + ig(a^\dagger\sigma_- - \sigma_+a) + E(a^\dagger + a) \quad (4.33)$$

and there are two collapse operators

$$\begin{aligned} C_1 &= \sqrt{2\kappa}a \\ C_2 &= \sqrt{\gamma}\sigma_- \end{aligned} \quad (4.34)$$

corresponding to leakage from the cavity and spontaneous emission from the atom respectively. In this case The Liouvillian has the standard Linblad form

$$L = -i(H\rho - \rho H) + \sum_{j=1} 2C_j\rho C_j^\dagger - \frac{1}{2}(C_j^\dagger C_j\rho + \rho C_j^\dagger C_j) \quad (4.35)$$

After writing proper Liouvillian, we seek a steady-state solution for ρ using the $L\rho = 0$

4.2.1 Cyclic Transition Model For Efficient Generation of Pure Single Photon Modes

The applications such as quantum cryptography require a single-photon source with high efficiency and reduce two-photon emission events. In addition, the dynamics should be deterministic. The emission of the photon then occurs on demand, in response to trigger pulse. The single-photon pulses should therefore occupy a well-defined. A large number of experimental attempts have been made to build an efficient single-photon source. While producing antibunched light, these systems had problems with a combination of low efficiency. To increase the collection efficiency of single photons from single photon sources such as quantum dots and ions, microcavities have been employed. We propose here that single molecule, strongly coupled to an optical cavity, can be used to produce single-photon pulses with high efficiency. In contrast to atoms, single molecules can be readily localized in the cavity on a scale far below their resonance wavelength [54]. With single molecules, efficient and truly continuous emission of single photons is possible, without the problems associated with non-stationary atoms in an optical cavity.

The level structure of single molecules as well as the relevant system parameters are different from their atomic counterparts. Here, we present a theoretical model of single-photon generation with a single molecules, providing a realistic description of a molecule coupled to a single-cavity mode. In our model energy is delivered to the system by means of a pulsed pump laser on the resonance transition, which also serves as a trigger. A single photon is created from the pump beam, yielding a photon in the cavity mode. In this no additional photon is created in the pump cycle.

In the next section, the master equation of the driven system is solved to numer-

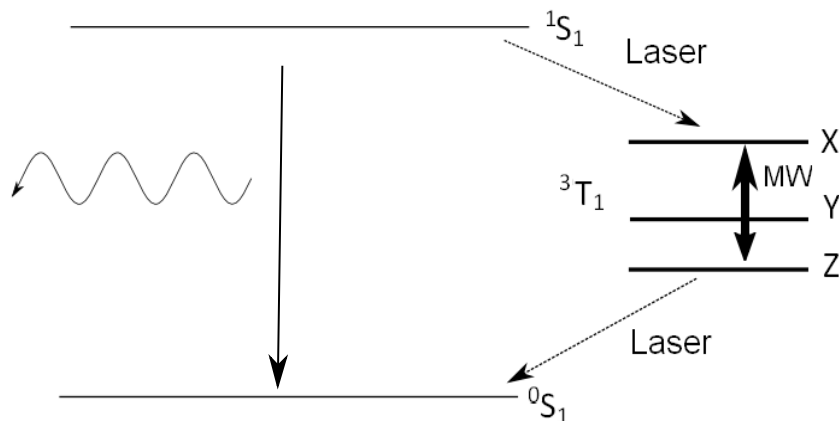


Figure 4.3: Energy level scheme of the single molecule's atomic levels coupled by the trigger MW pulse, the cavity and possible repumping pulses

ically investigate the process of single-photon generation.

4.2.1.1 Single Molecule as a Single Photon Source in a Cavity

As fluorescence light of a single emitter is antibunched for timescale on the order of the excited state radiative lifetime such systems can simply produce single photons on demand [50][51][52].

In order to understand the basic principles of single photon generation by single molecules we have to consider the role of lowest excited triplet state as in Figure (4.3). Energy level scheme shows the singlet ground state S_0 , the first excited state S_1 and the lowest excited triplet state T_1 . During the excitation emission cycle the molecule occasionally crosses from the diamagnetic singlet state to the longer lived nonemissive triplet state with total spin $S = 1$. Thus fluorescence cycle is interrupted by dark periods when molecule is in the triplet state. The time spent in this state depends on which of the $2s + 1 = 3$ spin sublevels is populated. To emit more than one photon at a time, a molecule has to undergo a full excitation, emission and reexcitation cycle within the same excitation pulse. The probability of this occurrence is extremely small when the pulse duration is much shorter than the excited state lifetime. Following

the experimental analysis of references [53][54] we estimate second order correlation function to prove indistinguishability. As shown in Figure (4.3) we consider a five level atom trapped in a doubly resonant cavity as a single photon source as discussed as a single molecule in ref [54]. The atoms pumped in the upper level $|4\rangle$ at a pump rate r_a . The transitions between triplet states T_1 , say $|3\rangle \leftrightarrow |3\rangle$ induced by a strong magnetic field with the Rabi frequency Ω_m and transition between S_1 and T_1 , $|4\rangle \leftrightarrow |3\rangle$ induced by laser with Ω_n , while ω_l applied between S_0 and T_1 , $|3\rangle \leftrightarrow |1\rangle$.

In the Schrödinger picture, under the rotating-wave approximation, the interaction Hamiltonian of the such a system can be written as (with the assumption of $\hbar = 1$)

$$H_{int} = (\Omega_p|4\rangle\langle 3| + \Omega_m|3\rangle\langle 2| + \Omega_l|2\rangle\langle 1| + h.c.) + (g_1 a_1|4\rangle\langle 1| + h.c.) \quad (4.36)$$

where h.c. means Hermitian conjugate and a_1 and a_2 are the annihilation operators for the field modes 1 and 2, and g_1 is the corresponding atom-field coupling constants. $\Omega_m = |\Omega_m| \exp(-i\Phi)$ denotes the Rabi frequency (Φ is the phase of the classical field). When we include the effects of spontaneous emission, the master equation in the interaction picture is

$$\frac{d}{dt}\hat{\rho} = -i[\hat{H}, \hat{\rho}] + \sum_{k \in \{a,b,c\}} (\hat{C}_k \hat{\rho} \hat{C}_k^\dagger - \frac{1}{2} \hat{C}_k \hat{C}_k^\dagger \hat{\rho} - \hat{\rho} \frac{1}{2} \hat{C}_k \hat{C}_k^\dagger) \quad (4.37)$$

where the 'collapse operators'

$$\hat{C}_1 = \sqrt{\gamma_1}|4\rangle\langle 3| \quad \hat{C}_2 = \sqrt{\gamma_2}|3\rangle\langle 2| \quad \hat{C}_3 = \sqrt{\gamma_3}|2\rangle\langle 1| \quad \hat{C}_4 = \sqrt{\gamma_4}|4\rangle\langle 1| \quad (4.38)$$

we also introduce cavity decay rate

$$\hat{D}_1 = \sqrt{2\kappa_1}a_1 \quad \hat{D}_2 = \sqrt{2\kappa_2}a_2 \quad (4.39)$$

Instead of seeking steady state solution, our numerical approach, depends on iterations of Liouville equation using the generic form of a master equation

$$\frac{d}{dt}\rho = L\rho \quad (4.40)$$

where ρ is the density matrix and the Liouvillian L is a superoperator.

4.2.2 Quantum Trajectory Methods to Calculate Correlation Functions

The description of open quantum systems is a central issue in numerous areas in physics. To study the stochastic dynamics of individual open quantum systems conditioned on the time history of photon emission events, a quantum stochastic approach is essential. A quantum trajectory represents a single history of the state vector of an open quantum system conditioned on the outcomes of measurement performed on the environment.

Einstein, in his classical paper on the A and B coefficients for spontaneous and stimulated emission, assumed the existence of quantum jumps, which greatly stimulated quantum mechanics [42]. However, quantum jumps played practically no role in various theories coupling radiation and matter, and these types of interactions are well described by the Schrödinger wavefunction describing the properties of an ensemble, rather than individual systems. Now, considering the systems coupled to reservoirs, we start from the Liouville equation for the system coupled to the reservoir, then we trace over the reservoir variables and after Born, Markov and Rotating Wave Approximations, end up with a usually Lindblad form of master equation [43].

In contrast to the Optical Bloch Equation, (OBE), which provides an ensemble description of the irreversible dynamics of the single photon source, each quantum trajectory generated in a simulation run can be visualized as corresponding to a single realization of the source evolution of the spontaneously emitted photons. The average over a large ensemble of such trajectories reproduces the reduced density matrix of the quantum source determined from the master equation.

4.2.2.1 Two-time Correlation Functions

The density matrix of an open system evolves according to the master equation, which can be written in the standard Lindblad form

$$\frac{d}{dt}\rho_S = \frac{1}{i\hbar}[H(t), \rho_S] + \sum_j \left(-\frac{1}{2}L_j^\dagger L_j \rho_S - \frac{1}{2}\rho_S L_j^\dagger L_j + L_j \rho_S L_j^\dagger \right) \quad (4.41)$$

where H is the Hamiltonian of the system, and the Lindblad operators L_j represent the effect of the reservoir on the system in the Markov approximation [44]. In general an open system can be represented as a system S interacting with the reservoir R . We may write the second order correlation function of the system operators A and B in this way

$$C(t, t') = \langle A(t)B(t') \rangle = Tr_{SR}[A(t)B(t')\rho(0)] \quad (4.42)$$

where $A(t)$ and $B(t)$ are in the Heisenberg picture and $\rho(0) = \rho_S(0) \otimes \rho_R(0)$ is the initial density operator. At time $t = 0$ the system and the reservoir are decoupled; $A(0) = A_0 \otimes 1_R$ and $B(0) = B_0 \otimes 1_R$ act only on the system. Since the unitary time evolution operator $U(0, t)$ of the composite system $S + R$ creates entanglement, for $t > 0$, $A(t)$ and $B(t)$ act both on the reservoir and the system considered. The reduced dynamics of the system is given in the Schrödinger picture by

$$\rho_S(t) = T(t, 0)\rho_S(0) = Tr_R[U(t, 0)\rho(0)U^\dagger(t, 0)] \quad (4.43)$$

Thus the correlation function Eq. (4.42) reads

$$\begin{aligned} C(t, t') &= Tr_{SR}[U^\dagger(t, 0)A(0)U(t, 0)U^\dagger(t', 0)B(0)U(t', 0)\rho(0)] \quad (4.44) \\ &= Tr_{SR}[[U(t', t)(U(t, 0)\rho(0)U^\dagger(t, 0))A_0U^\dagger(t', t)]B(0)] \\ &= Tr_S[\{T(t', t)[(T(t, 0)\rho_0)A_0]\}B_0] \quad (t \leq t') \end{aligned}$$

where $\rho_0 = \rho_S(0)$ denotes the density operator of the system at initial time $t = 0$ and we fix the time ordering to be $t \leq t'$. A similar equation holds for the reverse case

$$\langle A(t)B(t') \rangle = Tr_S[[T(t, t')(B_0T(t', 0)\rho_0)]A_0](t \geq t') \quad (4.45)$$

For any operator X acting on the system, one can define the operator $X(t', t)$ by requiring that

$$Tr[X(t', t)Y] = Tr[XT(t', t)Y] \quad (4.46)$$

stands for any system operator Y . Here we may now omit the index S from the trace over the system. Since $X(t', t)$ is linear in X , we can define the adjoint of the time evolution superoperator $T^a(t', t)$ as

$$T^a(t', t)X = X(t', t) \quad (4.47)$$

By substituting this definition into Eq. (4.46) we obtain that

$$Tr[(T^a(t', t)X)Y] = Tr[XT(t', t)Y] \quad (4.48)$$

The time evolution of $T(t', t)$ in the parameter t' is governed by the master equation

$$\frac{d}{dt'}T(t', t)X = \frac{1}{i\hbar}[H(t'), T(t', t)X] + \sum_j \left(-\frac{1}{2}L_j^\dagger L_j T(t', t)X - \frac{1}{2}T(t', t)X L_j^\dagger L_j + L_j T(t', t)X L_j^\dagger\right). \quad (4.49)$$

As an example of second order correlation function calculation using trajectory method, we start with

$$\begin{aligned} C(t, t') &= Tr_{SE}[A(t)B(t')C(t')D(t)\rho_0] \quad t \leq t' \quad (4.50) \\ &= Tr_{SE}[U^\dagger(t, 0)A(0)U(t, 0)U^\dagger(t', 0)B(0)U(t', 0)U^\dagger(t', 0) \\ &\quad \times C(0)U(t', 0)U^\dagger(t, 0)D(0)U(t, 0)\rho(0)] \\ &= Tr_{SE}[U^\dagger(t, 0)A(0)U(t, t')B(0)C(0)U(t, t')D(0)U(t, 0)\rho(0)] \\ &= Tr_{SE}[U(t, 0)\rho(0)U^\dagger(t, 0)A(0)U(t, t')B(0)C(0)U(t', t)D(0)] \end{aligned}$$

and using resolution of identity after $U(t, 0)$

$$\begin{aligned} C(t, t') &= Tr_{SE}[U(t, 0)U^\dagger(t, 0)U(t, 0)\rho(0)U^\dagger(t, 0)A(0)U(t, t')B(0)C(0)U(t', t)D(0)] \\ &= Tr_{SE}[U(t, t)U(t, 0)\rho(0)U^\dagger(t, 0)A(0)U(t, t')B(0)C(0)U(t', t)D(0)] \end{aligned}$$

and due to fact that $U^\dagger(t, 0) = U(0, t)$ and $U(t, 0)U(0, t) = 1$

$$\begin{aligned} C(t, t') &= Tr_{SE}[T(t, 0)\rho_s(0)A(0)U(t, t')B(0)C(0)U(t', t)D(0)] \\ &= Tr_{SE}[T(t, 0)\rho_s(0)A_0\{T(t, t')B_0C_0\}D_0] \end{aligned}$$

where we used $U(t, 0)\rho(0)U^\dagger(t, 0) = T(t, 0)\rho(0)$ and finally we get an expression for the second-order correlation function

$$C(t, t') = Tr_S[(T(t, 0)\rho_0)A_0(T(t, t')B_0C_0)D_0] \quad t \leq t' \quad (4.51)$$

The time evolution of $T(t', t)$ in the parameter t' is governed by the master equation given in the Eq. (4.49). We first iterated $T(t, 0)\rho_0$ then multiplied with A_0 from the

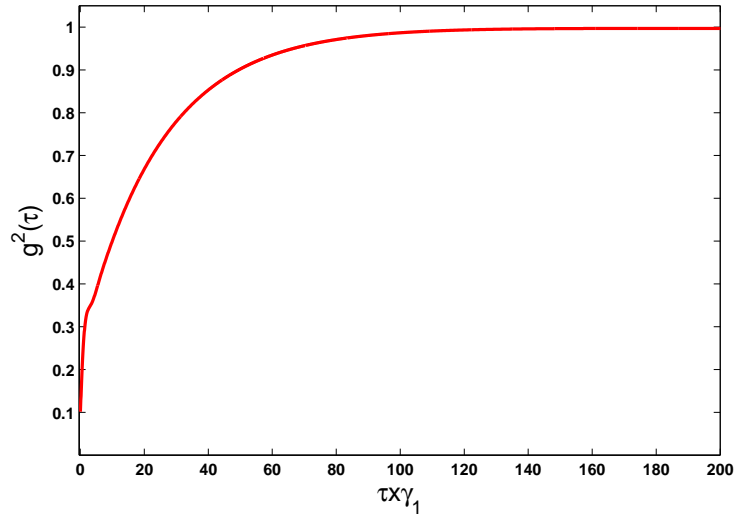


Figure 4.4: Second order correlation function

right hand side and we used initial density matrix, ρ_0 , from the previous discussions. Then we use second iteration for the $T(t, t') * B_0 * C_0$ and multiply with D_0 from right hand side. Finally we combine results of first and second iteration to generate second order correlation function. In our case, for the photon mode 1,

$$g^{(2)}(\tau) = Tr(a_2^\dagger(t)a_1^\dagger(t')a_1(t')a_1(t)) \quad (4.52)$$

In Figure 4.4 we show the second order correlation function of the single molecule single photon source is $g(\tau = 0) = 0.1$ which is ideally zero. As we can see here antibunching implies that fluorescent photons tend to be separated in time. In this case, there can be no photon emission immediately after the first photon detection for the ideal single photon sources.

4.2.3 Mandel Q Parameter

Photon antibunching and bunching is usually connected with sub- and super-Poissonian statistics in the probability distribution of photon numbers detected in a given time

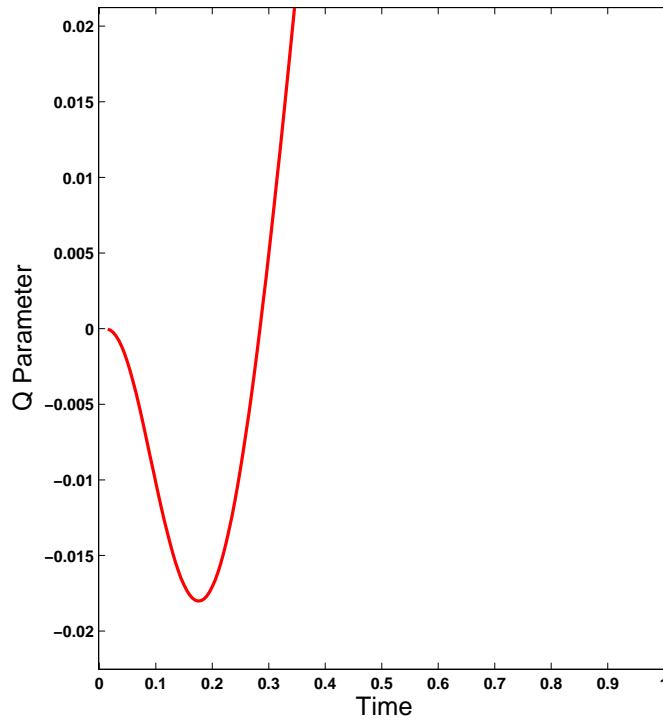


Figure 4.5: Mandel Q parameter

interval. The occurrence of sub-Poissonian statistics is a second criterion of a non-classical radiation field.

Emitted single photon from the single-molecule source is also reflected in the sub-Poissonian character of the probability density for photon number counts. The fluctuation in the number of photons emitted by the single molecule single photon source is weaker than the coherent light that follows Poissonian statistics. The sub-Poissonian character of the photon counting distribution is usually called Mandel Q parameter

$$Q = \frac{\langle n^2 \rangle - \langle n \rangle^2}{\langle n \rangle - 1} \quad (4.53)$$

in the long measurement limit. In order to reveal quantum nature of light we check Mandel Q parameter. For a coherent light source, we have $Q = 0$ and for the ideal

single photon source while photons arrive at regularly we have $Q = -1$. Figure 4.5 shows the Mandel Q parameter for certain single molecule. In the previous sections the single event photon statistics measurement of a single photon source based on the HBT experiment is studied. It is shown that the imperfect detectors, the imperfect beam-splitter and the unbalanced linear propagation efficiencies will reduce the single event Mandel Q parameter.

Chapter 5

CONCLUSION

In this thesis we present the work related to the realisation and the statistical characterisation of single photon sources using single molecules as single emitters. For this purpose we have developed a model, describing the interaction between a single molecule and an optical cavity. Using the atomic levels including all relevant decay terms, we have calculated the master equation of the system and evaluated the time-dependent density matrix to estimate the cavity output as a function of time. We can therefore accurately predict the properties of the photon pulses emitted from the cavity.

We used standard stochastic methods and a numerical solution of the master equation for a quantitative treatment in the weak-excitation regime. We first investigated λ -type three level atomic system both analytically and numerically. We calculated quantum entanglement criteria which is clearly less than 0 that satisfies entanglement criterion. Thus the two-mode cavity field evolves into an entangled state.

We have realized an efficient triggered single photon source relying on the control of a single molecule fluorescence. Single photon emission by the molecule occurs at each excitation pulse after a random time delay related to the molecules excited state lifetime. In our model we used MW driven single molecule in a two-mode optical cavity to model more efficient single photon emitter.

We calculated the second order correlation function $g^2(\tau)$ using quantum trajectory method to estimate coincidence detection of single photons like HBT interferometry. We also investigated time-dependant Mandel parameter $Q(T)$ which has negative values to prove non-classical behaviours of single photons. With expected application to quantum cryptography, more efficient of single photon sources exhibit

advantages over the other pulse sources such as attenuated laser pulses. In recent experiments for the fluorescence of a single emitter observed a significant increase in efficiency of the emitted photons. All these works are promising realization of an efficient single photon source well-suited for secure open-air quantum key distribution.

Appendix A

COEFFICIENTS

The coefficients α_{ij} and β_{ij} (i,j=1,2) in Eq. (4.21) are given by the following:

$$\begin{aligned}
\alpha_{11} &= -g_1^2 \left[\frac{\gamma_3}{(\gamma_3/2 + 2|\Omega|^2/\gamma_3)(r_a + 2\gamma_3)} + \frac{2r_a\gamma_{32}}{\gamma_3(\gamma_3/2 + 2|\Omega|^2/\gamma_3)(r_a + 2\gamma_3)} \right] \\
\beta_{11} &= g_1^2 \frac{r_a}{(\gamma_3/2 + 2|\Omega|^2/\gamma_3)(r_a + 2\gamma_3)} \\
\alpha_{22} &= -g_2^2 \left[\frac{\gamma_3}{(\gamma_3/2 + 2|\Omega|^2/\gamma_3)(r_a + 2\gamma_3)} - \frac{2r_a\gamma_{32}}{\gamma_3(\gamma_3/2 + 2|\Omega|^2/\gamma_3)(r_a + 2\gamma_3)} \right] \\
\beta_{22} &= g_2^2 \frac{r_a}{(\gamma_3/2 + 2|\Omega|^2/\gamma_3)(r_a + 2\gamma_3)} \\
\alpha_{12} &= g_1g_2 \left[\frac{2i\Omega}{(\gamma_3/2 + 2|\Omega|^2/\gamma_3)(r_a + 2\gamma_3)} + \frac{i\gamma_{32}r_a}{\Omega^*(\gamma_3/2 + 2|\Omega|^2/\gamma_3)(r_a + 2\gamma_3)} \right] \\
\alpha_{21} &= g_2g_1 \left[\frac{2i\Omega^*}{(\gamma_3/2 + 2|\Omega|^2/\gamma_3)(r_a + 2\gamma_3)} + \frac{-i\gamma_{32}r_a}{\Omega(\gamma_3/2 + 2|\Omega|^2/\gamma_3)(r_a + 2\gamma_3)} \right] \\
\beta_{21} &= g_1g_2 \frac{-2i\Omega r_a}{\gamma_3(\gamma_3/2 + 2|\Omega|^2/\gamma_3)(r_a + 2\gamma_3)} \\
\beta_{12} &= g_2g_1 \frac{-2i\Omega^* r_a}{\gamma_3(\gamma_3/2 + 2|\Omega|^2/\gamma_3)(r_a + 2\gamma_3)}
\end{aligned}$$

VITA

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