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An Investigation of Giant Kerr Nonlinearity

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A thesis
submitted in partial fulfilment
of the requirements for the degree
of
Doctor of Philosophy
at the
University of Auckland

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Abstract

This thesis investigates the properties of an atomic system exhibiting a giant Kerr nonlinearity. The atomic energy level scheme involves four energy levels. A three level subsystem in the atom exhibits the effect of electromagnetically induced transparency (EIT), reducing the spontaneous emission noise. The fourth level leads to an ac–Stark shift of the ground state, which in turn leads to a giant, noiseless Kerr nonlinearity. Two different environments are explored. First, a system comprising of large number of atoms in an optical cavity is analysed. Detailed aspects of noise reduction in this system are investigated. In particular, strong squeezing in the quadrature in phase with the field driving the cavity mode is found, if the effective coupling of light to the atoms is strong. However, the linewidth of the predicted squeezing is found to be very narrow. This is attributed to a very steep linear susceptibility of the atomic medium. Since the widening of the squeezing window is possible only for weaker effective coupling, in turn reducing the squeezing level, a different environment is proposed. This involves a single four level atom, strongly coupled to the cavity mode. In such a strongly coupled system, the most appropriate approach is found to be that formulated in terms of polaritons – composite excitations of the ‘atom–cavity molecule’. Adopting the polariton approach, nonclassical correlations in the field leaving the cavity are investigated. Strong photon antibunching is found and the effect of photon blockade predicted and described. The photon blockade effect can also be found in a system comprised of a two level atom coupled to the cavity mode, if the external driving is tuned to one of the vacuum Rabi resonances. A comparison between the two schemes is performed, and it is found that the four level scheme exhibits much better photon blockade. The reason for this is quantum interference between secondary transitions in the dressed states picture. Destructive interference cancels the transitions that would otherwise introduce a second photon into the system, hence producing a more robust photon blockade. All of these results are valid in the regime where external driving is weak. If the external driving strength is increased, the photon statistics (as measured by the zero–delay second order correlation function) changes from strong antibunching to strong bunching, over a relatively narrow range of driving strengths. The occurrence of this change can again be attributed to quantum interference. It is shown that the interference effect prevents the excitation of the composite system by a second photon, but not excitation by a two–photon transition (following the first excitation). Therefore, the third excitation manifold is excited, which then decays back to the first manifold in a two photon cascade. This two photon cascade is the source of correlated photon pairs causing an increase in the second order correlation function.

The dynamics of forward scattering of light is presented, and nonclassical behaviour of the delay dependence of correlation function (‘overshoots’ and ‘undershoots’) is discussed. For the analytical treatment of this system, a method based on the polariton approach is devised, which includes the treatment of driving and damping. It is shown that this method is ideally suited to the analysis of strongly coupled systems, where only a few photons contribute to the dynamics.
Acknowledgments

There are many people who have helped to make this thesis possible, and it is sad that the person who initiated this work is no longer with us. Professor Dan Walls passed away when the work on this thesis was still at an early stage. Nevertheless, his impact was very valuable and is strongly felt throughout the thesis. I will always remember Dan for his warmth and enthusiasm, which always managed to get the best out of those around him. He is sadly missed by all of us who had the privilege of knowing him and working with him. After Dan’s passing, I was lucky enough to have two extremely able and friendly supervisors. I can’t thank Dr. Scott Parkins and Assoc. Prof. Sze Tan enough for all of the attention they have given me at all stages of research and drafting of this thesis. They are both very generous with the allocation of their time, and their friendly attitude and attention to detail have strongly contributed to make this thesis better. In the recent months they have both devoted a lot of attention into reading of the drafts of this thesis, making valuable suggestions and, in consequence, improving its content.

A collaboration with Prof. Ataç Imamoglu has thoroughly deepened my knowledge and understanding of the issues involved in this thesis. His involvement with my work over the last few years, and his readiness to plunge into a thoughtful discussion every time I would have a question (in person or via e-mail), are really appreciated. I would like to use this opportunity to thank him for his contribution. I would also like to thank him for his hospitality during my visit to the University of California in Santa Barbara. During this visit and afterwards via e-mail, I have also benefited from a fruitful exchange with Dr. Mike Werner, and I would like to extend my thanks to him.

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Of course, a big special thanks goes to all members of the Quantum Optics Group here in Auckland. Especially, I would like to acknowledge interactions with Assoc. Prof. Matthew Collett, Dr. Murray Olsen and Dr. Lev Plimak. I was blessed with an office that served to accommodate many visitors, and a few office mates. They are all too numerous to name, but some deserve a special mention (in chronological order). Dr. Mike Steel is acknowledged for his support and friendliness, Mr. Jevon Longdell for being a wonderful office mate. Most of all, Mr. Steve Clark, my current office mate, deserves a warmest thanks for taking an interest in my work, and proofreading of a thesis. This was a colossal undertaking, and it is my feeling that Steve’s input has improved immensely the readability of this thesis.

On a personal note, I would like to thank my wife Ljubica for her warmth, appreciation and support during the course of my research, and my son Alexander for changing my perspective on life (although, of course, he is blissfully unaware of it).

Finally, I would like to emphasise that the credit for any mistakes in this work, either mathematical, physical or typographical, belongs entirely to the author.
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Preface

It is obvious, however, that whatever side we take concerning the nature of light, many, indeed almost all the circumstances concerning it, are incomprehensible, and beyond the reach of human understanding.

*Encyclopaedia Britannica*, article on 'Light', 1792

The nature of light and its interaction with matter is a topic that has puzzled a huge number of scientists and philosophers throughout the entire history of human civilisation. The final resolution\(^1\) to this problem started to unravel about a century ago, with the advent of quantum mechanics. While there are still many unsolved problems, both deeply conceptual and purely technical, to be encountered by scientists in the future, it is my firm belief that the nature of light is *comprehensible* and *within* the reach of human understanding (contrary to the belief of our XVIII century forefathers). It is my hope that the work presented in this thesis contributes, however minutely, to the gigantic effort to gain a full understanding of this fascinating set of phenomena.

The aim of this thesis is to investigate the properties of a four level atomic system that has been predicted to generate a large optical Kerr nonlinearity. This system is appealing in its use of electromagnetically induced transparency (EIT) to produce a virtually noiseless nonlinearity, using destructive quantum interference to cancel the main contribution to the noise - spontaneous emission. Once the energy level scheme has been defined and a realistic atomic system, supporting the required energy level structure, has been identified, one obvious question arises. Which experimental environment should be used to examine this system? This thesis mainly examines two possible setups: (i) a many–atom environment, where a large number of atoms is assumed to be confined in an optical cavity, and (ii) a single atom trapped in the vicinity of a cavity field antinode in a microscopic–size optical cavity.

The thesis is divided into five main Parts. **Part I** is intended as an overview of the basic concepts and techniques used in the remainder of this thesis. Chapter 1 reviews the concepts and techniques used to study open quantum systems, with an emphasis on the master equation, stochastic differential equations and quantum trajectories. Chap-\(^1\) Or, to state it more carefully, what we consider to be a final resolution at our present level of understanding.
ter 2 overviews quantum coherence effects and introduces a four level atomic system that exhibits a giant Kerr nonlinearity.

**Part II** contains an analysis of a many atom cavity QED environment. The work presented here was in part published in


In Chapter 3 an analysis of noise fluctuations in the many atom system, such as quadrature squeezing and intensity squeezing of the light in the output from the cavity are discussed. It is shown that the photon blockade effect appears only within a very narrow band of frequencies centered at the cavity mode frequency. In Chapter 4 it is shown how this effect can be fully understood in classical terms, and a general description of linewidth reduction due to linear and/or nonlinear susceptibilities of arbitrary order is discussed.

From the practical point of view, the many atom setup has been found to be unsatisfactory for the realisation of the photon blockade effect. As an alternative to the atomic medium, in **Part III** a system consisting of a single four level atom in a high finesse microcavity is examined as a potential candidate for a photon blockade effect. This system will be focused on for the remainder of this thesis. Following Chapter 5, where the topic of single–atom cavity QED is briefly reviewed, Chapter 6 presents the numerical simulations of the single atom system. This analysis shows how the problems encountered in the many–atom case can be avoided by using the single–atom cavity QED arrangement. In Chapter 7 an effective model to explain the results obtained in Chapter 6 is developed. An analysis of the dressed states is presented and the concept of the polariton is introduced. The effective two level behaviour is identified and simulations performed that show Rabi oscillations between the two dressed states. Excerpts from this work were published as


In **Part IV** a two level atomic system coupled to a cavity mode, predicted to exhibit an effective two level behaviour is analysed (Chapter 8), and a comparison of the photon blockade in the two schemes presented (Chapter 9). It is shown that the four level scheme has much more flexibility and achieves better photon blockade than the two level scheme, even for comparable sets of parameters. This is attributed to destructive quantum interference between certain transition amplitudes in the dressed states basis. Article containing the work presented in this Part is in preparation for publication as


In **Part V**, the influence of the driving field on the photon statistics in the single–atom system is examined. In Chapter 10, a complete analytical solution for a single four level
atom strongly coupled to a cavity field mode and driven by external coherent laser fields is presented. Using this formalism, in Chapter 11 an analysis of the effective two level system, using the polariton approach is developed. The effect of dynamic Stark splitting of dressed states is found and the similarities with the analogous two level system are explored. The fluorescence spectrum is calculated numerically and the effective Hamiltonian is used to identify peaks and linewidths. Results of Chapters 10 and 11 are contained in


The photon statistics of light emitted from an EIT–Kerr system for a range of driving field strengths is studied in Chapter 12. A sudden change in the photon statistics of the light emitted from the cavity is found. Photon antibunching switches to photon bunching over a very narrow range of intracavity photon number. This effect, as well as nonclassical correlations at nonzero delay times, is studied in detail. The article containing the results of Chapter 12 is published as

Part I

Basic Concepts and Techniques
Chapter 1

Theory of Open Quantum Systems

This Chapter reviews the concepts and techniques used to study open quantum systems. In particular, we review three approaches developed to solve and analyse open quantum systems. These are based on (i) the master equation, (ii) stochastic differential equations and (iii) quantum trajectories. The purpose of this Chapter is to provide an overview of the methods that will be used later in this thesis to study systems that exhibit large Kerr nonlinearity.

1.1 Introduction

In quantum physics one is often confronted with a situation where a localised system (with a few degrees of freedom) interacts with several other systems (with very large numbers of degrees of freedom) whose exact states we do not know (and can not find out, even in principle). The collection of systems with large numbers of degrees of freedom is usually called an environment, or a reservoir, while the system with only a few degrees of freedom is called an open system. The point at which the division between the system and the environment is made is often based on a subjective choice, but there are some standard approaches. In this Chapter, we will consider a system coupled to a reservoir representing the loss mechanism, and introduce methods and techniques for the analysis of such a system.

There are two distinct ways of treating general quantum mechanics problems - the Schrödinger picture and the Heisenberg picture - which give rise to two approaches for treating system–reservoir interactions. The one based on the Schrödinger picture leads to a master equation. The other, based on the Heisenberg picture, leads to quantum stochastic differential equations. An alternative approach to the problem is provided by quantum trajectory methods, where a master equation is “unravelled” from being an equation for the density operator to being an equation for the evolution of a pure quantum state. This method provides additional insight into the intricacies of the quantum physics, and has also proven to be a powerful tool for performing numerical simulations of large (and otherwise intractable) systems.


1.2 Master Equation

All real physical processes have associated “loss mechanisms” which are due to coupling between the system and the environment. Some (but not all) of the losses that quantum systems are usually subjected to can be incorporated into the equations of motion for the probability amplitudes. We want to describe how losses may be incorporated into the quantum mechanical equations of motion. We outline the basis of the master equation formalism which provides a way of doing this [1].

Consider the case when the full state of the system is not known, but the probabilities \( p_i \) for having various state vectors \( |\phi_i\rangle \) are known. Such a system is described by a density operator \( \rho_s = \sum_i p_i |\phi_i\rangle \langle \phi_i| \), which may be written in an arbitrary orthogonal basis as

\[
\rho_s = \sum_{i,j} c_{ij} |\psi_i\rangle \langle \psi_j| , \tag{1.1}
\]

where \( \{|\psi_k\rangle\} \) represent an orthogonal basis set.

Consider a system described by the Hamiltonian \( \mathcal{H}_S \) coupled to a reservoir whose evolution is described by \( \mathcal{H}_R \). The weak interaction between the system and the reservoir is given by \( \mathcal{H}_{SR} \). The equation of motion for the total density operator of the system and reservoir \( \rho_{S+R} \) in the interaction picture is von Neumann’s equation:

\[
\dot{\rho}_{S+R}(t) = -\frac{i}{\hbar} [\mathcal{H}_{SR}(t), \rho_{S+R}(t)] . \tag{1.2}
\]

The solution of this equation can be found by iteration:

\[
\rho_{S+R}(t) = \rho_{S+R}(0) + \sum_{n=1}^{\infty} \left( -\frac{i}{\hbar} \right)^n \int_0^t dt_1 \int_0^{t_1} dt_2 \cdots \\
\times \int_0^{t_{n-1}} dt_n [\mathcal{H}_{SR}(t_1), [\mathcal{H}_{SR}(t_2), \ldots, [\mathcal{H}_{SR}(t_n), \rho_{S+R}(0)]]] . \tag{1.3}
\]

This equation contains more information than is needed for describing the system, since \( \rho_{S+R} \) is the density operator for the reservoir as well as the system. Hence, we define the reduced density operator \( \rho(t) \), describing the system alone by tracing over the reservoir variables:

\[
\rho(t) = \text{Tr}_R\{\rho_{S+R}\} . \tag{1.4}
\]

If the system and reservoir are initially uncorrelated, then \( \rho_{S+R}(0) = \rho(0) \otimes \rho_R \). Tracing (1.3) over reservoir variables gives

\[
\rho(t) = U(t)\rho(0) = (\mathbb{1} + U_1(t) + U_2(t) + \cdots)\rho(0) , \tag{1.5}
\]

\[
U_n(t)\rho(0) = \left( -\frac{i}{\hbar} \right)^n \int_0^t dt_1 \int_0^{t_1} dt_2 \cdots \\
\times \int_0^{t_{n-1}} dt_n \text{Tr}_R[\mathcal{H}_{SR}(t_1), [\mathcal{H}_{SR}(t_2), \ldots, [\mathcal{H}_{SR}(t_n), \rho_R \otimes \rho(0)]]] . \tag{1.6}
\]
1.2. MASTER EQUATION

Here, $U(t)$ is a unitary time evolution operator, given by a perturbation series expansion. A state (or another operator) to which the operator $U_n$ is applied is multiplied by $\rho_R$ from the left, as indicated. The von Neumann equation now becomes

$$\frac{d\rho}{dt} = \mathcal{L}\rho,$$

(1.7)

where $\mathcal{L}$ is the Liouvillian operator, acting as a generator of time evolution. The Liouvillian is given in terms of the unitary operators $U_n(t)$, given by (1.6), as

$$\mathcal{L} = \left(\dot{U}_1(t) + \dot{U}_2(t) + \cdots\right)U^{-1}(t).$$

(1.8)

We assume that $\mathcal{H}_{SR}(t)$ satisfies $\text{Tr}_R[\mathcal{H}_{SR}(t)\rho_R] = 0$ giving $U_1(t) = 0$. Furthermore, since it was assumed that $\mathcal{H}_{SR}(t)$ is weak, terms of order higher than second in the perturbation series may be dropped, giving $U^{-1}(t) \approx 1$, and

$$\mathcal{L} = \dot{U}_2(t).$$

(1.9)

Hence, the expression for the reduced density operator, given to the second order in the perturbation expansion, is

$$\frac{d\rho}{dt} = -\frac{1}{\hbar^2} \int_0^t dt_1 \text{Tr}_R[\mathcal{H}_{SR}(t), [\mathcal{H}_{SR}(t_1), \rho_R \otimes \rho(t)]].$$  

(1.10)

To proceed, we need to specify a particular system–reservoir interaction Hamiltonian, but before we do that, it is worthwhile emphasizing two approximations involved in the derivation of the master equation. Truncation of the perturbation expansion after the second order constitutes the Born approximation. Another approximation, the Markov approximation, is also hidden in the master equation (1.10). If we follow the derivation consistently, the density operator $\rho$ on the right–hand side should be evaluated at time $t_1$, rather than time $t$. In that case, the time development of the density operator $\rho(t)$ depends on its previous time development through the integration over $\rho(t_1)$. Physically, this is because past states of the system change the state of the reservoir, so that later time evolution of the system is affected by its past through the interaction with the reservoir. The Markov approximation assumes the reservoir remains in thermal equilibrium, and has a short correlation time compared to the dynamics of system. The past history of the system is thus “forgotten” and does not influence its future. Formally, this amounts to the replacement $\rho(t_1) \rightarrow \rho(t)$. An estimate of the reservoir correlation time in the context of the Markov approximation is given by Carmichael [2, §1.3.3]. Equation (1.10) is called the master equation in the Born–Markov approximation.

Often we are interested in the interaction of bosonic particles (such as photons) with a reservoir whose quanta are also bosons. Such a situation can in general be modelled by the damped harmonic oscillator. The interaction Hamiltonian for a simple harmonic
oscillator of frequency $\omega_0$ linearly damped via its interaction with the reservoir is given by

$$H_{SR}(t) = \hbar \left[ a \Gamma^\dagger(t) e^{-i\omega_0 t} + a^\dagger \Gamma(t) e^{i\omega_0 t} \right], \quad (1.11a)$$

$$\Gamma(t) = \sum_k g_k b_k e^{-i\omega_k t}, \quad (1.11b)$$

where the sum over $k$ is the sum over reservoir modes with annihilation operators $b_k$. This expression is rather general; for example, damping of an atomic two state transition $|i\rangle \rightarrow |j\rangle$ by a reservoir is described by a Hamiltonian of the same form, with the replacement $a \rightarrow \sigma_{ij}$, where $\sigma_{ij} = |j\rangle \langle i|$. It is now possible to derive the master equation starting from the Hamiltonian (1.11) and Equation (1.10). The sum over modes in the expression for the reservoir operators (1.11b) can be converted to a frequency space integral

$$\Gamma(t) = \int_0^\infty \frac{d\omega}{2\pi} D(\omega) g(\omega) b(\omega) e^{-i\omega t}, \quad (1.12)$$

where $D(\omega)$ denotes the density of modes in the reservoir.

The trace over the reservoir that appears in equation (1.10) gives rise to an average of products of field operators over the reservoir. If the reservoir is assumed to be thermal, the only nonvanishing averages will be

$$\langle b^\dagger(\omega_1) b(\omega_2) \rangle = 2\pi N(\omega_1) \delta(\omega_1 - \omega_2), \quad (1.13a)$$

$$\langle b(\omega_1) b^\dagger(\omega_2) \rangle = 2\pi [N(\omega_1) + 1] \delta(\omega_1 - \omega_2), \quad (1.13b)$$

where $N(\omega)$ is given by Planck’s thermal distribution:

$$N(\omega) = \frac{1}{e^{\hbar\omega/kT} - 1}. \quad (1.13c)$$

For a thermal reservoir, these are the only nonvanishing correlations. Some reservoirs, however, can also contain some phase dependent correlations, not included in the correlations above. For example, a squeezed reservoir has the additional correlations

$$\langle b(\omega_1) b(\omega_2) \rangle = 2\pi M(\omega_1) \delta(2\omega_0 - \omega_1 - \omega_2), \quad (1.13d)$$

$$\langle b^\dagger(\omega_1) b^\dagger(\omega_2) \rangle = 2\pi M(\omega_1)^* \delta(2\omega_0 - \omega_1 - \omega_2). \quad (1.13e)$$

Next, we assume that the functions $N(\omega), D(\omega)$ and $g(\omega)$ are slowly varying functions around $\omega = \omega_0$, where $\omega_0$ is very large. This assumption is consistent with the Markov approximation used to derive the equation (1.10), and is often called the first Markov approximation in the literature [3, 4, 5]. We define the damping rate $\kappa$ as

$$\kappa = D^2(\omega_0) g^2(\omega_0). \quad (1.14)$$
Putting all this together, we are able to evaluate the integral in Equation (1.10), and obtain the following equation in the interaction picture

\[ \frac{d\rho}{dt} = \kappa (N + 1) (2a^\dagger a - \rho a^\dagger a - a^\dagger a \rho) + \kappa N (2a^\dagger \rho a - \rho aa^\dagger - aa^\dagger \rho) + \kappa M (2a^\dagger \rho a^\dagger - \rho a^\dagger a^\dagger - a^\dagger a \rho) + \kappa M^* (2a^\dagger a - \rho a - aa^\dagger \rho). \]  

(1.15)

The positivity of the density operator requires

\[ |M|^2 \leq N(N + 1). \]  

(1.16)

This equation, describing the time evolution of the system's density operator, is called the master equation. In the Schrödinger picture, the equation becomes

\[ \frac{d\rho^{Sch}}{dt} = \frac{d\rho}{dt} - i\omega_0 [a^\dagger a, \rho], \]  

(1.17)

with \( \rho^{Sch} \) denoting the reduced density operator in the Schrödinger picture.

Equations of motion for the expectation values of system operators may be derived directly from the master equation. In general, for an operator \( \hat{O} \), not explicitly time dependent in the interaction picture, we have

\[ \frac{d}{dt} \langle \hat{O} \rangle = \text{Tr}\{\hat{O}\rho\}, \]  

(1.18a)

\[ \frac{d}{dt} \langle \hat{O} \rangle = \text{Tr}\left\{ \hat{O} \frac{d\rho}{dt} \right\}. \]  

(1.18b)

If the harmonic oscillator is perturbed by an additional interaction \( \mathcal{H}_I \), the master equation would contain an additional term, proportional to \( \mathcal{H}_I, \rho \).

### 1.3 Two Time Correlations

The calculation of two time correlation functions occupies a central place in quantum optics. For example, the first order correlation function of the field operator is needed to calculate the spectrum of the field, while the second order correlation function contains information about the photon statistics. The quantum regression theorem due to Lax [6] presents a relationship which enables us to calculate the two time correlations from the master equation for the reduced density operator.

Suppose that the following (complete) set of equations for the mean values of operators can be derived from the master equation

\[ \frac{\partial}{\partial t} \langle \hat{O}_i(t) \rangle = \sum_j g_{ij}(t) \langle \hat{O}_j(t) \rangle. \]  

(1.19)

\[ ^1 \text{As a consequence of the first Markov approximation, we can extend the upper range of integration in equation (1.10) to infinity. This means that the solutions will be valid for times which are long compared to the correlation time of the reservoir, but which may nevertheless be short compared with the decay time of the system.} \]
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We want to obtain a similar equation for the two time correlation function defined by

\[
\langle \hat{O}_i(t + \tau) \hat{O}_k(t) \rangle = \text{Tr}_S \{ \hat{O}_i e^{\mathcal{L} \tau} \hat{O}_k \rho(t) \},
\]

with \( \mathcal{L} \) being the Liouvillian operator of the corresponding master equation. If we formally set the initial condition (at \( t \)) of the master equation to

\[
\hat{\rho} = \hat{O}_k \rho(t),
\]

then the average values of \( \hat{O}_i \) computed over the solution \( \hat{\rho}(t + \tau) \) gives the two time correlation function. Using this, we can write the equation for the two time correlation function, by inspection from Equation (1.19), as

\[
\frac{\partial}{\partial \tau} \langle \hat{O}_i(t + \tau) \hat{O}_k(t) \rangle = \sum_j g_{ij}(t) \langle \hat{O}_j(t + \tau) \hat{O}_k(t) \rangle.
\]

This is the statement of the quantum regression theorem. In practice, two time correlations are calculated using this method. We first solve the master equation for the density operator \( \rho(t) \), or the set of equations (1.19). Then we formulate the new initial condition as (1.21), and solve the master equation again, now as a function of time delay \( \tau \). An arbitrary two time correlation function can be obtained in this way.

1.4 Quantum Langevin Equations

As mentioned earlier in this Chapter, there is another approach to the problem of damping and loss in quantum systems based on the Heisenberg picture. Now, we sketch some results and elaborate on the relationship between the two approaches.

Consider an arbitrary system interacting with a reservoir. The Hamiltonian for such a system may be written as

\[
\mathcal{H} = \mathcal{H}_{\text{sys}} + \mathcal{H}_{\text{reservoir}} + \mathcal{H}_{\text{int}},
\]

where \( \mathcal{H}_{\text{sys}} \) describes the system, \( \mathcal{H}_{\text{reservoir}} \) describes the reservoir, and \( \mathcal{H}_{\text{int}} \) describes the interaction of the system with the reservoir. For the sake of simplicity, we will consider only a single reservoir. The reservoir is characterised by the boson operator \( b(\omega) \), and it is taken to interact with the system via a coupling with the (unspecified) system operator \( c \). This coupling is taken to be linear

\[
\mathcal{H}_{\text{reservoir}} = \hbar \int_{-\infty}^{\infty} d\omega \, b^\dagger(\omega) b(\omega), \quad (1.24a)
\]

\[
\mathcal{H}_{\text{int}} = i\hbar \int_{-\infty}^{\infty} d\omega \, C(\omega) [b^\dagger(\omega) c - c^\dagger b(\omega)], \quad (1.24b)
\]

with

\[
[b(\omega), b^\dagger(\omega')] = \delta(\omega - \omega'). \quad (1.24c)
\]
1.4. QUANTUM LANGEVIN EQUATIONS

Note that the lower limit of integration is taken to be $-\infty$. In actual physical systems, this limit is zero. However, it proves to be convenient to work in a frame rotating with some characteristic frequency $\Omega$ (for example, the driving field frequency), which shifts this lower limit of integration to $-\Omega$. This characteristic frequency $\Omega$ is usually large compared to the frequency bandwidth of interest, allowing us to safely approximate the integration range by $(-\infty, \infty)$. Another approximation involved in this particular form of $H_{\text{int}}$ (1.24b) is the rotating wave approximation, which neglects the terms $(cb(\omega) + \text{H.c.})$ based on the assumption that these are rapidly oscillating terms, compared to those retained in Equation (1.24b), and can be averaged to zero.

The Heisenberg equation of motion for an arbitrary operator $\hat{O}$ is any arbitrary system operator $a$:

$$\frac{d}{dt} \hat{O} = -\frac{i}{\hbar} [\hat{O}, H].$$

We can derive the Heisenberg equations of motion for the reservoir operator $b(\omega)$, and

$$\dot{b}(\omega) = -i\omega b(\omega) + C(\omega)c,$$  

$$\dot{a} = -\frac{i}{\hbar} [a, H_{\text{sys}}] + \int_{-\infty}^{\infty} d\omega C(\omega) \{b^\dagger(\omega)[a, c] - [a, c^\dagger]b(\omega)\}.$$  

Equation (1.26) can be solved in terms of initial conditions at time $t_0 < t$ to give

$$b(\omega) = e^{-i\omega(t-t_0)}b_0(\omega) + C(\omega) \int_{t_0}^{t} e^{-i\omega(t-t')}c(t') \, dt',$$

where $b_0(\omega)$ is the value of $b(\omega)$ at $t = t_0$. Using this solution, equation (1.27) can be rewritten as

$$\dot{a} = -\frac{i}{\hbar} [a, H_{\text{sys}}] + \int_{-\infty}^{\infty} d\omega C(\omega) \{e^{i\omega(t-t_0)}b_0^\dagger(\omega)[a, c] - [a, c^\dagger]e^{-i\omega(t-t_0)}b_0(\omega)\}$$

$$+ \int_{-\infty}^{\infty} d\omega C^2(\omega) \int_{t_0}^{t} dt' \{e^{i\omega(t-t')}c^\dagger(t')[a, c] - [a, c^\dagger]e^{-i\omega(t-t')}c(t')\}.$$  

The first Markov approximation was introduced previously through the definition of the damping rate $\kappa$ (1.14). We now apply the same approximation to the system–reservoir coupling. Therefore, we assume that the coupling is independent of frequency

$$C^2(\omega) = \kappa/\pi.$$  

The constant $\kappa$ is called the decay constant, and is the same quantity obtained in the derivation of the master equation as the damping rate. Using the well-known relations

$$\int_{-\infty}^{\infty} \frac{d\omega}{2\pi} e^{-i\omega(t-t')} = \delta(t-t'),$$  

$$\int_{t_0}^{t} dt' c(t')\delta(t-t') = \frac{1}{2} c(t),$$

$^2$Note the difference in sign with von Neumann’s equation (1.2).
equation (1.29) can be put in a form
\[ \dot{a} = -\frac{i}{\hbar}[a, \mathcal{H}_{\text{sys}}] + \left\{ \left( \kappa c^\dagger + \sqrt{\kappa/\pi} \int d\omega e^{i\omega(t-t_0)} b_0^\dagger(\omega) \right)[a, c] - [a, c^\dagger] \left( \kappa c + \sqrt{\kappa/\pi} \int d\omega e^{-i\omega(t-t_0)} b_0(\omega) \right) \right\}. \quad (1.33) \]
This form of equation is called the quantum Langevin equation. Let us specify this equation for the case of the simple harmonic oscillator. Then, the system operator will be \( c = a \), and \( \mathcal{H}_{\text{sys}} = \hbar \omega_0 a^\dagger a \), giving:
\[ \dot{a} = -i\omega_0 a - \kappa a - \sqrt{\kappa/\pi} \int d\omega e^{-i\omega(t-t_0)} b_0(\omega). \quad (1.34) \]
This equation bears a similarity to the Langevin equation for Brownian motion [7], justifying the term quantum Langevin equation. It includes a damping term, \(-\kappa a\), and a noise term, dependent on the initial state of the reservoir. It is the average value and correlations of such terms that are usually of practical interest, rather than the exact form of the term itself. This corresponds to the ‘fluctuating force’ of the original Langevin treatment of Brownian motion. It is not necessary to make any particular assumption about the system’s initial state for the Langevin equation to be valid.

### 1.5 Input–Output Formalism

The equation (1.33) can be written in a more compact form if we define the input field operator [3, 4]
\[ b_{\text{in}}(t) = \int_{-\infty}^{\infty} \frac{d\omega}{\sqrt{2\pi}} e^{-i\omega(t-t_0)} b_0(\omega), \quad (1.35) \]
which satisfies the following commutation relation
\[ [b_{\text{in}}(t), b_{\text{in}}^\dagger(t')] = \delta(t-t'). \quad (1.36) \]
Equation (1.33) now takes the simplified form
\[ \dot{a} = -\frac{i}{\hbar}[a, \mathcal{H}_{\text{sys}}] + (\kappa c^\dagger + \sqrt{2\kappa} b_{\text{in}}^\dagger(t))[a, c] - [a, c^\dagger] (\kappa c + \sqrt{2\kappa} b_{\text{in}}(t)). \quad (1.37) \]
This is the quantum Langevin equation written in terms of initial conditions at the time \( t_0 < t \) (the input). We may wish to specify the equation in terms of final conditions at some time \( t_1 > t \) (the output). In that case, we may solve equation (1.26) to give
\[ b(\omega) = e^{-i\omega(t-t_1)} b_1(\omega) - C(\omega) \int_t^{t_1} e^{-i\omega(t-t')} c(t') \, dt', \quad (1.38) \]
1.6. QUANTUM NOISE

with \( b_1(\omega) \) being the value of \( b(\omega) \) at \( t = t_1 \), and define the output field operator as

\[
b_{\text{out}}(t) = \int_{-\infty}^{\infty} \frac{d\omega}{\sqrt{2\pi}} e^{-i\omega(t-t_1)} b_1(\omega) .
\] (1.39)

Following the same procedure as above produces the time reversed Langevin equation,

\[
a = -\frac{i}{\hbar} [a, \mathcal{H}_{\text{sys}}] + \{ (-\kappa c^\dagger + \sqrt{2\kappa b_{\text{out}}^\dagger}(t))[a, c] - [a, c^\dagger](-\kappa c + \sqrt{2\kappa b_{\text{out}}^\dagger}(t)) \} .
\] (1.40)

Obviously, this equation can be derived from equation (1.37) by substituting \( \text{in} \rightarrow \text{out} \) in the noise terms, and \( \kappa \rightarrow -\kappa \) in the damping terms.

A relation between the input, output, and system operators is readily found to be

\[
b_{\text{out}}(t) - b_{\text{in}}(t) = \sqrt{2\kappa} c(t) .
\] (1.41)

The commutators of system operators with the input and output field operators can be derived as

\[
[a(t), b_{\text{in}}(t')] = -\Theta(t - t') \sqrt{2\kappa} [a(t), c(t')] ,
\] (1.42a)

\[
[a(t), b_{\text{out}}(t')] = \Theta(t' - t) \sqrt{2\kappa} [a(t), c(t')] ,
\] (1.42b)

where \( \Theta(\tau) \) denotes the Heaviside step function:

\[
\Theta(\tau) = \begin{cases} 
0 & \text{for } \tau < 0 \\
\frac{1}{2} & \text{for } \tau = 0 \\
1 & \text{for } \tau > 0 
\end{cases} .
\] (1.43)

These commutation relations reflect the principle of causality, which requires that only the future motion of the system is affected by the present input, and only the future values of the output are affected by the present values of the system operators. Now we have a complete description of the system with inputs and outputs.

This \textit{input–output formalism} was first introduced into quantum optics by Gardiner and Collett [3, 4]. In a typical experiment, the input field interacts with the particular system of interest (such as the atomic medium). This system is confined inside an optical cavity, and the input field enters the cavity through one of its mirrors. Since no cavity is perfect, it allows leakage through one (or both) of its mirrors in the form of a measurable quantum field. This field provides the output. In other words, the input–output formalism describes the connection between the intracavity system and the outside world.

1.6 Quantum Noise

Technically, we may define noise as a time dependent random process with a short autocorrelation time. It does not carry the information of interest, but competes with the
signal and degrades the accuracy of information transfer. However, we are interested in the way in which the notion of noise enters into physics, quantum optics in particular.

Classical statistical mechanics was probably the first theory to encounter the presence of noise. In a typical system, the number of particles is of the order of Avogadro’s number, $N_A = 6.022 \times 10^{23}$ mol$^{-1}$. In phase space, each particle is described by at least two coordinates - position and momentum. In the framework of classical theory it would be possible in principle to determine the initial conditions and write down and solve an equation of motion for every single particle. In practice, of course, this is impossible, due to the enormous number of particles. In order to deal with such systems, one is forced to treat them in terms of statistical averages, thus ignoring the information believed not to be important for a particular case. In a full treatment, this (ignored) information is included as a noise.

Quantum physics goes further. Since the position and momentum of a particle satisfy the uncertainty principle of Heisenberg, it is not even in principle possible to obtain precise information about the initial conditions of the system. Therefore, in quantum systems, there is a certain intrinsic level of noise present which simply cannot be avoided.

There is another source of noise in physical systems, namely the one related to the dynamics. In classical physics, the example for this would be Brownian motion [7]. Since the nature of the interaction of pollen grains and giant molecules in a solution is not known, and the collisions between the two are random, Langevin postulated the notion of an unknown stochastic force (as a source of noise) to account for this phenomenon. This stochastic force represents the incessant impacts of the molecules of the liquid on the pollen grains. All that was known about it was the fact that it should be positive and negative with equal probability.

The first description of quantum noise was given by Wigner and Weisskopf in their theory of spontaneous decay of an atom. The inability to specify each of the infinite number of states of the electromagnetic field together with the intrinsic probabilistic aspect of quantum mechanics gives rise to the nonzero spectral linewidth. The fact that the spectral line is not sharp is due to the noise, although the concept was not originally used by Weisskopf and Wigner.

### 1.6.1 White Noise and the Wiener Process

We have mentioned that the noise may be defined as a time dependent random process with a short autocorrelation time, which is then assumed to be $\delta$—correlated in time. So, for a particular noise $\Gamma(t)$, the autocorrelation function becomes:

$$\langle \Gamma(t)\Gamma(t') \rangle = \delta(t - t').$$  \hspace{1cm} (1.44)

This kind of noise is called white noise. The spectrum of such noise can be found from the Wiener–Khinchin theorem [7]:

$$S(\omega) = \frac{1}{2\pi} \int_{-\infty}^{\infty} e^{-i\omega\tau} \langle \Gamma(\tau)\Gamma(0) \rangle \, d\tau.$$  \hspace{1cm} (1.45)
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Obviously, white noise has the flat frequency spectrum $S(\omega) = \text{const}$. In the case of light, the frequencies correspond to different colours of light. For white light, all the colours are represented in equal proportions (at least within the visible range) and hence the term white noise is applied to every source of noise having a flat frequency spectrum.

It is clear that the concept of white noise is an idealisation, since the energy in a field with a flat spectrum is infinite. However, in quantum optics, the systems of interest usually operate within some limited bandwidth. If the noise spectrum can be considered to be flat over this bandwidth, then the white noise approximation is valid.

Mathematically, white noise is usually taken to be the derivative of a continuous but non-differentiable random function $W(t)$. This function defines the Wiener process.

The probability distribution function for a Wiener process is a Gaussian [7]

$$ P(w, t|w_0, t_0) = [2\pi(t-t_0)]^{-1/2} e^{-\frac{(w-w_0)^2}{2(t-t_0)}}, \quad (1.46) $$

obtained by solving the appropriate differential equation with a delta function as an initial condition. The mean and mean square of a Wiener process are given by

$$ \langle W(t) \rangle = w_0, \quad (1.47) $$
$$ \langle [W(t) - w_0]^2 \rangle = t - t_0. \quad (1.48) $$

So, the Wiener process is characterised by spreading of an initially sharp distribution with increasing time $t - t_0$. If we define the increment for the stochastic integration as

$$ \Delta W_i = W(t_i) - W(t_{i-1}), \quad (1.49) $$

then, due to the Markovian nature of the Wiener process, the variables $\Delta W_i$ are independent of each other and of $W(t_0)$.

1.6.2 The Quantum Wiener Process

Classical white noise theory has been immensely successful in the description of time-dependent statistical problems where the driving fluctuations are spectrally broadband. The quantum theory of white noise is then a logical next step. It is formulated to treat quantum stochastic processes and deals with non-commuting operator variables.

Consider a system described by the Hamiltonian $\mathcal{H}_{\text{sys}}$. The nature of the input to the system is determined by the operator $b_{in}(t)$, defined by equation (1.35). Since this operator describes an ensemble of quantum states, the noise will be determined by the ensemble correlations, including the intrinsic (zero-point) quantum fluctuations. The expectation values of these operators for a broadband thermal input are

$$ \langle b_{in}(t) \rangle = \langle b_{in}^\dagger(t) \rangle = 0, \quad (1.50a) $$
$$ \langle b_{in}^\dagger(t)b_{in}(t') \rangle = N\delta(t - t'), \quad (1.50b) $$
$$ \langle b_{in}(t)b_{in}^\dagger(t') \rangle = (N + 1)\delta(t - t'). \quad (1.50c) $$
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It is important to note that the reservoir, strictly speaking, is not in a thermal initial state, since we assume that the number of photons in a particular mode is constant over a bandwidth of frequencies $N(\omega) = N$. $N$ is thus a number of quanta per unit bandwidth, while the genuine thermal ensemble obeys equation (1.13c).

On closer inspection, there seems to be a logical discrepancy in the definition of the Wiener process, since it was defined as a derivative of a non-differentiable function. This problem can be avoided by defining the Wiener process as an integral of white noise\(^3\). We define

$$B(t, t_0) = \int_{t_0}^{t} b_{in}(t') \, dt',$$

$$B^\dagger(t, t_0) = \int_{t_0}^{t} b_{in}^\dagger(t') \, dt'.$$\hspace{1cm} (1.51a)

These operators have the following properties:

$$\langle B^\dagger(t, t_0)B(t, t_0) \rangle = N(t - t_0),$$

$$\langle B(t, t_0)B^\dagger(t, t_0) \rangle = (N + 1)(t - t_0),$$\hspace{1cm} (1.52a)

$$\langle B^\dagger(t, t_0)B(t, t_0) \rangle = N dt,$$

$$\langle B(t, t_0)B^\dagger(t, t_0) \rangle = (N + 1) dt.$$\hspace{1cm} (1.52b)

and

$$[B(t, t_0), B^\dagger(t, t_0)] = t - t_0.$$\hspace{1cm} (1.52c)

The operators $B$ and $B^\dagger$ have a quantum Gaussian distribution, with a density operator of the form [5]

$$\rho(t, t_0) \propto e^{-\lambda B^\dagger(t, t_0)B(t, t_0)/(t-t_0)},$$\hspace{1cm} (1.53)

so these operators define the quantum analogue of a classical Wiener process.

Analogous to the increment (1.49), we now introduce stochastic differentials:

$$dB(t, t_0) = B(t + dt, t_0) - B(t, t_0) = \int_{t_0}^{t+dt} b_{in}(t') \, dt',$$\hspace{1cm} (1.54)

consistent with an integral definition of the Wiener process. The lower limit of integration, $t_+$, means that the integral is not evaluated at the time $t$, because in order to preserve causality, $dB(t)$ must be uncorrelated with the system variables for times $t' < t$. This definition lies at the heart of the Itô formalism, to be developed in the next section.

The definition of the noise increment and the statistics of the input field yield

$$dB(t, t_0)dB^\dagger(t, t_0) = (N + 1) dt,$$

$$dB^\dagger(t, t_0)dB(t, t_0) = N dt.$$\hspace{1cm} (1.55)

If the effect of the reservoir on the system is equivalent to driving the system with a quantum Wiener process as the input, then we have a quantum white noise theory.

\(^3\)In the classical case, this would turn the differential equation into an integral equation.
1.7 Itô Calculus

Consider some arbitrary function of time $G(t)$. If $B(t)$ is the (classical) Wiener process, we may ask what is the meaning of the integral

$$
\int_{t_0}^{t} G(t')dB(t').
$$

(1.57)

This is a stochastic integral and is defined in the manner of a Riemann–Stieltjes integral. The integration interval is divided into $n$ subintervals by means of the partition $t_0 \leq t_1 \leq t_2 \leq \cdots \leq t_{n-1} \leq t_n$, with intermediate points $\tau_i$ defined by $t_{i-1} \leq \tau_i \leq t_i$. The stochastic integral is defined as a limit of the partial sums:

$$
S_n = \sum_{i=1}^{n} G(\tau_i) \left[ B(t_i) - B(t_{i-1}) \right].
$$

(1.58)

It turns out that the choice of the intermediate point is not arbitrary, because different choices produce different results for the stochastic integral. In fact, if we also choose $G(\tau_i)$ to be the classical Wiener process, the mean value of the integral can be anything between zero and $t - t_0$, depending on the choice of intermediate points. There are two standard definitions for the intermediate points found in the literature:

**Itô definition** : $\tau_i = t_{i-1}$,

**Stratonovich definition** : $\tau_i = \frac{1}{2}(t_{i-1} + t_i)$.

The Itô definition leads to the Itô calculus, with rules different to the rules of ordinary calculus. The Itô stochastic integral is defined as

$$
\int_{t_0}^{t} G(t')dB(t') = \text{ms-lim}_{n \to \infty} \left\{ \sum_{i=1}^{n} G(\tau_i) \left[ B(t_i) - B(t_{i-1}) \right] \right\},
$$

(1.59)

where ms-lim denotes mean square limit [7]. This integral exists whenever $G(t')$ is a continuous and nonanticipating function$^4$ of $t'$.

It can be proved that $dB^2(t) = dt$, implying that the stochastic differential $dB(t)$ is of order $\sqrt{dt}$. Thus, when differentiating an arbitrary function of $B(t)$, all terms up to second order in $dB(t)$ must be kept. Accordingly, letting $(dt)^2 \to 0$, as well as $dt dB(t) \to 0$, we arrive at the following formula for the Itô stochastic differential of an arbitrary function $f[B(t), t]$:

$$
\text{d}f[B(t), t] = \frac{\partial f}{\partial t} dt + \frac{1}{2} \frac{\partial^2 f}{\partial t^2} (dt)^2 + \frac{\partial f}{\partial B} dB(t) + \frac{1}{2} \frac{\partial^2 f}{\partial B^2} [dB(t)]^2 + \cdots
$$

$$
= \left( \frac{\partial f}{\partial t} + \frac{1}{2} \frac{\partial^2 f}{\partial B^2} \right) dt + \frac{\partial f}{\partial B} dB(t).
$$

(1.60)

$^4$A function $G(t)$ is called a nonanticipating function of $t$ if for all $s$ and $t$ such that $t < s$, $G(t)$ is statistically independent of $B(s) - B(t)$. This means that $G(t)$ is independent of the behaviour of the Wiener process in the future of $t$. 

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
This formula is called the Itô formula. It gives the differentiation rule appropriate for the Itô calculus, which will be used extensively throughout this thesis. For two arbitrary non-commuting stochastic quantities $x$ and $y$, the second order differentiation rule is:

$$d(xy) = (dx)y + x(dy) + (dx)(dy).$$

Equipped with these results, we can now generalise the earlier results on quantum Langevin equations.

Let us mention briefly that the Stratonovich choice for the intermediate point leads to the theory in which the rules of standard calculus apply. Since the Stratonovich version of the stochastic calculus will not be used in this thesis, we will omit the discussion of its properties.

### 1.8 Quantum Stochastic Differential Equations

With the above results at our disposal, the quantum Langevin equation (1.37) can be written as an Itô quantum stochastic differential equation (Itô QSDE) for the arbitrary system operator $a$:

$$da = -\frac{i}{\hbar}[a, \mathcal{H}_{\text{sys}}]dt + \kappa(N + 1) \{[c^\dagger, a]c + c[a, c^\dagger] + \kappa N (c^\dagger c c^\dagger + c c^\dagger)\} dt + \sqrt{2\kappa} \{dB^\dagger(t)[a, c] - [a, c^\dagger]dB(t)\}. \quad (1.62)$$

It can be shown [5], that this equation is equivalent to the Langevin equation (1.37). To proceed further, we need to specify a particular interaction for the physical system of interest.

It is straightforward to establish a connection between this equation and the equivalent master equation. If the density operator at time $t = 0$ is given by $\rho(0)$ for the system and $\rho_0$ for the reservoir, then the system density operator at time $t$ becomes:

$$\rho(t) = \text{Tr}_R[U(t) \rho(0) \otimes \rho_0 U^\dagger(t)]. \quad (1.63)$$

This gives for the master equation:

$$\dot{\rho}(t) = -\frac{i}{\hbar} [\mathcal{H}_{\text{sys}}, \rho] + \kappa(N + 1) \{[c^\dagger, \rho(t)c^\dagger] + [c \rho(t), c^\dagger]\}$$

$$+ \kappa N \{[c^\dagger, \rho(t)] + [c^\dagger \rho(t), c]\}. \quad (1.64)$$

Note that the operators appearing in this equation are Schrödinger picture operators. These two approaches are equivalent if at all times both of them yield the same result for the expectation value of any system operator $\langle a(t) \rangle$. The proof of equivalence can be found in Gardiner and Zoller [5].
1.9 Quantum Trajectories Approach

The treatment of open quantum systems reviewed so far has been derived from the viewpoint of the theory of ensembles. While fundamentally this is a sound approach, since quantum mechanics is a theory of ensembles, the possibility to observe quantum jumps\(^5\) [11, 12, 13, 14] leads to the question of how to describe any single realisation of a system undergoing quantum jumps. In particular, a concept of conditional time evolution has been introduced to describe single experimental realisations of a quantum system. For example, the time evolution of a quantum system can be conditioned on whether a photon has been emitted by the system or detected by the detector. Quantum jumps occur randomly, collapsing the wave function of the system. Averaging over all possible random realisations of time evolution leads to an ensemble description of the system, obtained through the density matrix. To study the individual realisations, a description via wave functions is needed.

The concept of conditional time evolution, as one of infinitely many possible realisations of the quantum system, leads to the introduction of a conditional wave function and, consequently, conditional density operator. What is the relation of the conditioned density operator \(\rho_c\) to the density operator \(\rho\) which has been used so far? The master equation \(\dot{\rho} = \mathcal{L}\rho\) has the formal solution

\[
\rho(t) = e^{\mathcal{L}t}\rho(0). \tag{1.65}
\]

If we add and subtract an arbitrary superoperator \(\mathcal{S}\) from the Liouvillian superoperator \(\mathcal{L}\), the exponential in equation (1.65) can be expanded to give \([15]\)

\[
\rho(t) = e^{[(\mathcal{L} - \mathcal{S}) + \mathcal{S}]t}\rho(0)
= \sum_{k=0}^{\infty} \int_0^t dt_k \int_0^{t_k} dt_{k-1} \cdots \int_0^{t_2} dt_1 e^{(\mathcal{L} - \mathcal{S})(t-t_k)} \mathcal{S}
\times e^{(\mathcal{L} - \mathcal{S})(t_{k-1}-t_k)} \mathcal{S} \cdots \mathcal{S} e^{(\mathcal{L} - \mathcal{S})t_1}\rho(0). \tag{1.66}
\]

This equation can be interpreted as a general sum over all possible physical realisations that the system might follow between the time \(t = 0\) and time \(t\). In particular, if the system under consideration is a source of photons, then the possible physical realisations describe all the photon emission pathways that the source might follow in a given time. The time intervals \((t_{k-1}, t_k)\) are the intervals of the time evolution without photon emissions (or, in general, quantum jumps). These time evolutions are governed by the superoperator \((\mathcal{L} - \mathcal{S})\). Collapses at the times of the photon emission (quantum jumps) are governed by the superoperator \(\mathcal{S}\). The form of the superoperators \((\mathcal{L} - \mathcal{S})\) and \(\mathcal{S}\) is such that we can factorise the conditioned density operator as a pure state \(\rho_c(t) = |\psi_c(t)\rangle\langle\psi_c(t)|\). This insight determines the form of \(\mathcal{S}\), and we will give an illustrative example shortly.

\(^5\)For example, using the atomic shelving method, proposed by Dehmelt [8, 9] and elaborated by Cook and Kimble [10].

\(^6\)This statement is true only if \(\mathcal{S}\) is an operation i.e., a completely positive superoperator.
CHAPTER 1. THEORY OF OPEN QUANTUM SYSTEMS

Turning back to our original goal, it is now clear that the quantity inside the integrals on the right-hand side of Equation (1.66) is the unnormalised conditioned density operator \( \mathcal{P}_c(t) \) for an initial state \( \rho_c(0) \equiv \rho(0) \). The normalised conditioned density operator is then

\[
\rho_c(t) = \frac{\mathcal{P}_c(t)}{\text{Tr}\{\mathcal{P}_c(t)\}}, \quad (1.67)
\]

\[\mathcal{P}_c(t) = e^{(\mathcal{L} - \mathcal{S})t - t_k} \mathcal{S} \cdots \mathcal{S} e^{(\mathcal{L} - \mathcal{S})t_1} \rho(0). \quad (1.68)\]

This constitutes the decomposition of the quantum dynamics to an infinity of possible quantum paths, called the quantum trajectories. The measurement process holds a central role in the interpretation above. The times \( t_k \) are the times at which a photon is detected by a detector of unit efficiency, registering that the system has undergone a quantum jump, and the unravelling above separates the times at which a quantum jump occurs, from the intervals between subsequent quantum jumps, when the system evolves freely. Another key concept involved in this interpretation is the concept of continuous measurement just described, and a system undergoing continuous measurement is then an open system by definition.

To illustrate the concept of unravelling, consider the interaction of an optical cavity mode with some intracavity medium. Since this thesis will concentrate on the Kerr nonlinearity, we will consider a Kerr intracavity medium. As will become obvious, choice of the intracavity medium is rather irrelevant, and all the conclusions hold independently.

If the cavity has a decay constant \( \kappa \), the master equation is given by

\[
\dot{\rho} = -\frac{i}{\hbar} \left[ \mathcal{H}_0, \rho \right] + \kappa (2\alpha \rho a^\dagger - a^\dagger a \rho - \rho a^\dagger a), \quad (1.69)
\]

where \( \mathcal{H}_0 = \hbar \omega_{\text{cav}} a^\dagger a + \hbar \eta a^\dagger a a \). The superoperator \( \mathcal{S} \), describing the emission of photons is given by \( \mathcal{S} \rho = 2\kappa \alpha \rho a^\dagger \). The remainder of the right-hand side can then be written as

\[
(\mathcal{L} - \mathcal{S})\rho = -\frac{i}{\hbar} \left( \mathcal{H}_{\text{eff}} \rho - \rho \mathcal{H}_{\text{eff}}^\dagger \right) \quad (1.70)
\]

where the non-Hermitian operator \( \mathcal{H}_{\text{eff}} = \mathcal{H}_0 - i\hbar \kappa a^\dagger a \) describes the time evolution between the jumps. The superoperator \( \mathcal{S} \), can then be written as \( \mathcal{S} \rho_c = C \rho_c C^\dagger \), where operator \( C = \sqrt{2\kappa} a \) describes the quantum jump, and is called the collapse operator. The probability for a collapse to occur at time \( t_k \) is given by

\[
p_c(t_k) = \langle \psi_c(t_k)| C^\dagger C |\psi_c(t_k) \rangle \Delta t, \quad (1.71)
\]

where \( \Delta t \) is the discrete time step, while between the collapses the wave function evolves according to the effective Hamiltonian

\[
|\tilde{\psi}_c(t + \Delta t)\rangle = e^{-i/\hbar \mathcal{H}_{\text{eff}} \Delta t} |\tilde{\psi}_c(t)\rangle. \quad (1.72)
\]

The time evolution can now be simulated numerically. We evaluate the collapse probability \( p_c(t_k) \), generate a random number \( r \in [0,1] \), and compare the two. If \( p_c(t_k) \leq r \),
we calculate the wave function at a time $t_{k+1}$ as $\psi_c(t_{k+1}) = C |\psi_c(t_k)\rangle$, and renormalise it. If $p_c(t_k) > r$, the wave function is left to propagate according to $H_{\text{eff}}$, to give $\psi_c(t_{k+1}) = \exp\left[-(i/\hbar)H_{\text{eff}} \Delta t\right] |\psi_c(t_k)\rangle$, with $\Delta t = t_{k+1} - t_k$, and normalise it. This procedure gives one possible realisation of the time evolution. It is possible to repeat the procedure an arbitrary number of times, and average the results over all realisations. The result obtained that way will in the limit of an infinite number of trajectories asymptotically approach the result of the density matrix simulations.

The advantages of the quantum trajectory approach are obvious. First, it reduces the size of a numerical problem from $N \times N$ for the density matrix approach, to only $N$. Second, it offers the possibility of studying individual realisations of possible conditional time evolutions. Third, it is fundamentally linked with the measurement process. In fact, the form of the collapse operator(s) will depend on the measurement scheme envisaged. In fact Carmichael [15] has shown that the unravelling of the master equation is in no way unique; it is determined by the detection scheme used to observe a particular system. The procedure that we have outlined in this Section, with our particular choice of collapse operator, is consistent with direct photodetection. For an example involving homodyne detection, see Carmichael [15, Chapter 9, § 9.2].

It is possible to simulate two time correlations using the quantum trajectories approach, and two different (but equivalent) methods are given by Mölmer et al. [16] and Gardiner and Zoller [5, 17]. An excellent review of the quantum jump method in quantum optics is given by Plenio and Knight [18].
Chapter 2

Giant Kerr Nonlinearity

This Chapter introduces a system that exhibits a giant Kerr nonlinearity. The relevant four level atomic system is described, and the significance of electromagnetically induced transparency (EIT) is explained. An overview is given of the photon blockade effect in terms of a dispersive Kerr nonlinearity.

2.1 Introduction

Achieving large noiseless optical nonlinearities has been the “holy grail” of nonlinear optics since its inception, but realistic nonlinear media have been plagued by a trade-off between the amount of noise and the strength of nonlinearity achieved. For example, the simplest nonlinear optical medium is one consisting of an assemblage of far-detuned two level atoms, and in this medium one can always increase the strength of the effective nonlinearity by tuning the probe laser closer to the atomic resonance. This, however, increases the excitation of the atoms and therefore the spontaneous emission rate, so that an increase in noise accompanies the increase in nonlinearity.

In recent years, however, the idea of utilising quantum coherence effects in multilevel atoms has offered an opportunity to reduce the noise while retaining the strength of nonlinearity. Atomic absorption can be reduced in carefully chosen systems via the effects of coherent population trapping (CPT) [19], electromagnetically induced transparency (EIT) [20, 21], lasing without inversion [22] and ghost transitions [23].

In this Chapter, a brief overview of how quantum coherence effects are used in multilevel atoms is presented. In particular, the effect of electromagnetically induced transparency is emphasized because of its importance in the scheme for generation of giant Kerr nonlinearity discussed in this thesis. A photon blockade effect will be introduced and discussed in the framework of pure optical Kerr nonlinearity.
Figure 2.1: Energy level scheme of a three level atom in \( \Lambda \)-configuration. Two transitions, \( |g_j\rangle \rightarrow |e\rangle \), are driven by external fields of Rabi frequencies \( \Omega_p \) and \( \Omega_s \), respectively.

### 2.2 Atomic Coherence Effects

This section reviews the concept of quantum coherence in multilevel (in particular, three level) atoms and its applications. The concept of coherence is widely and liberally used throughout physics, but a precise definition of it, strictly speaking, does not exist. The most common definition is related to the coherence of light, and laser light in particular. In general it is said that a process is coherent if it is characterised by the existence of some well-defined deterministic phase relationship, i.e. some phase is not subject to random noise [24]. While this definition offers some insight into the concept of coherence, it is not always transparent what the \textit{process} refers to, especially in the context of atomic coherence, i.e. coherence between atomic levels. Gheri [25] has adopted a specific definition for coherence between the atomic levels, which cannot be viewed in terms of “processes”. According to this definition, if superposition state(s) exist for an atomic system whose coefficients have a well defined phase relationship, then there is coherence between the levels out of which this state is formed. We will now review in brief some quantum coherence effects in quantum optics.

#### 2.2.1 Coherent Population Trapping

Atomic coherence effects were first found in a three level atomic system in a \( \Lambda \)-configuration, depicted in Fig. 2.1. An early demonstration was the experiment of Alzetta \textit{et al.} [26], who showed how coherence between the two states in a ground state doublet interacting with two near resonant coherent fields can lead to a complete cancellation of fluorescence from the atom. This explanation of the experimental results was given by Arimondo and Oriols [27], while Radmore and Knight [28] called this effect a \textit{dark resonance} or \textit{population trapping}. Atoms can spontaneously decay into a superposition of two ground states,
2.2. ATOMIC COHERENCE EFFECTS

which decouples from the fields $\Omega_{p,s}$. Since the ground state is now formed as a superposition of two doublet states $|g_{\pm}\rangle$, there are two probability paths for a transition to the excited state, and these interfere destructively. Therefore, the population is trapped in the lower states because the appropriate excitation mechanism is fully suppressed. The time dependent trapping state is found to be (see, for example, Scully and Zubairy [29])

$$|\psi(t)\rangle = \frac{\Omega_s(t)e^{-i\phi_s}|g_-\rangle + \Omega_p(t)e^{-i\phi_p}|g_+\rangle}{\sqrt{\Omega_p^2 + \Omega_s^2}}.$$  \hspace{1cm} (2.1)

Note that the important condition for CPT is that the atoms are initially prepared in this nonabsorbing state. The distribution of population between the two states $|g_{\pm}\rangle$ is determined by the ratio $\Omega_p/\Omega_s$.

An interesting application of such a trapping state is called adiabatic population transfer. Consider an atom initially in ground state $|g_-\rangle$, with $\Omega_p = 0$ and $\Omega_s$ finite. If the field $\Omega_p$ is turned on adiabatically, while the field $\Omega_s$ is turned off, the population of the atom will transfer into the state $|g_+\rangle$.

2.2.2 Lasing Without Inversion

The physics of lasing without inversion (LWI) has the same basic principles as that of population trapping. In fact, the only difference is in the initial state in which the atoms are prepared. Instead of having all the atoms prepared in an initial superposition state, imagine the atoms prepared in the excited state $|e\rangle$. This alone gives a standard lasing with inversion setup. If, however, the atomic medium simultaneously contains atoms prepared in the trapping state $|\psi(0)\rangle$ as per equation (2.1), lasing gain will still be achieved, regardless of the relative percentage of excited atoms in the mixture. So, lasing will occur even if the number of atoms in state $|e\rangle$ is minute. The reason can again be found in quantum coherence effects. The equation for the electric field amplitude can be written as [29]

$$\dot{E} = (a_e - a_+ - a_- + a_{+-} + a_{-+})E/2,$$  \hspace{1cm} (2.2)

where the term $a_e$ is a gain term and is proportional to the population of the excited state. Terms $a_+$ and $a_-$ are loss terms corresponding to absorption in transitions $|g_{\pm}\rangle \rightarrow |e\rangle$ and are proportional to the populations of the respective ground state levels. This is the familiar lasing with inversion: population in the excited state has to be larger than the combined population in the ground states in order to achieve gain. The terms $a_{+-}$ and $a_{-+}$ are phase dependent terms and are proportional to the coherence between the two ground states. With a particular choice of parameters, these can be chosen to cancel the $a_+$ and $a_-$ terms. Therefore, regardless of how small the fraction of atoms in the excited state is, it will produce gain in the laser. The LWI scheme described here was proposed and analyzed by Scully and coworkers [22, 30] and Kocharovskaya and coworkers [31, 32, 33, 34, 35].
2.2.3 Ghost Transitions

The concept of ghost transitions has been used by Gheri et al. [23] to circumvent a problem encountered in quantum nondemolition (QND) measurements via cross-phase modulation (by Grangier et al. [36]) in a two-photon cascade. Significant QND correlations can only be obtained with substantially increased atomic densities. The excess noise in such a system becomes more dominant as the density is increased, ultimately outweighing the improvement in performance and leading to total destruction of the QND correlations between the light fields.

The radical proposal of Gheri et al. [23] was to completely reconsider the setup and introduce a large degree of asymmetry into the system. Instead of a cascade system, they proposed the system of Figure 2.1. Asymmetry is introduced through the different detunings of the fields $\Omega_p$ and $\Omega_s$ from their respective atomic transitions. These transitions are also driven with very different strengths: strong signal $\Omega_s$ and much weaker probe $\Omega_p$. The effect of the strong signal is that almost all the population will be in the ground state of the probed atomic transition. Thus the signal will be applied to an essentially empty transition - the ghost transition - which reduces the signal absorption and spontaneous emission noise. Degradation of the signal is virtually negligible since the atomic medium will be effectively transparent.

This strong signal will produce large Rabi splittings in the relevant transition, offering the possibility of increasing correlation between the signal and the probe. In particular, tuning the probe as close as possible to one of the Rabi-split states, but still outside of its linewidth, increases the sensitivity of the probe to any changes in the signal intensity. This in turn provides a virtually noiseless way of detecting the QND correlations.

2.3 Electromagnetically Induced Transparency

We have remarked in Section 2.2.2 that the difference between CPT and LWI is the choice of the initial state in which the atoms are prepared, as well as the operating conditions. CPT, in particular, was achieved if all of the atoms were prepared in the non-absorbing state. The interesting aspect of CPT is that it may occur even if the atoms are not in a trapped state at $t = 0$. The continued action of electromagnetic fields and spontaneous emission can drive the atoms into the trapped state. The phenomenon of electromagnetically induced transparency (EIT) relies on this peculiar feature.

EIT is a technique for eliminating the effect of the medium on a propagating beam of electromagnetic radiation [21]. Classically, if one wishes to eliminate the effect of the medium, one would have to stop the electrons in the medium from oscillating at the frequencies of applied fields. This may occur if the electrons are driven by two oscillating forces of opposite phases, so that the total force vanishes. However, this classical explanation is unsatisfactory since the electrons in the atoms have to be treated quantum mechanically, and then the concepts of force and oscillation can be invoked only in discussing the averages or expectation values. The probability amplitude of the excited state $\ket{e}$ contains the two terms corresponding to the probability amplitudes for the transitions
2.3. ELECTROMAGNETICALLY INDUCED TRANSPARENCY

Figure 2.2: Coherence between the atomic levels \(|g_{-}\rangle\) and \(|e\rangle\) versus time for \(\Omega_p/\gamma = 1\) and \(\Omega_s/\gamma = 100\). The atom is initially in the ground state \(|g_{-}\rangle\).

to the ground states \(|g_{\pm}\rangle\), respectively. These can be tuned to have an equal amplitude and opposite phase, in which case they cancel to give a vanishing amplitude for the excited state. Cancellation requires that the atom is driven on two-photon resonance [37], i.e. the detunings of both \(\Omega_p\) and \(\Omega_s\) from the excited state have to be the same. In that case, the probe absorption coefficient will be proportional to the coherence \(\langle \sigma_{g_{-}e} \rangle\) of the atomic levels on the probe transition, for which the steady state value is found to be [38]

\[
\langle \sigma_{g_{-}e} \rangle_{ss} = -i \frac{\Omega_p/\gamma}{1 + (\Omega_s/\gamma)^2} \tag{2.3}
\]

for the resonant case of Figure 2.1. Note the important role that the field \(\Omega_s\) plays in this expression. It is customary to call this field the \textit{coupling field} in the framework of EIT, and we will adopt this convention from now on. Figure 2.2 shows the transient regime for the coherence, reaching the value of virtually zero in the steady state.

Another perspective on the EIT effect can be obtained by looking at the dressed states of the system. Under the described conditions, this system has three dressed states. One of them is resonant with the probe beam, while the other two are symmetrically detuned
from the resonance by an amount $\pm \sqrt{\Omega_p^2 + \Omega_s^2}$. The resonant state is the trapping state

$$|\psi\rangle = \frac{\Omega_s |g_\downarrow\rangle + \Omega_p |g_\uparrow\rangle}{\sqrt{\Omega_p^2 + \Omega_s^2}},$$

(2.4)

i.e. superposition of the two ground states. There is no component from the excited state and therefore absorption is suppressed, and the system is transparent.

Experimentally, EIT was first observed in the experiments of Boller et al. [20] and Field et al. [39]. The experiments of Xiao et al. [40] and Gea-Banacloche et al. [38] have shown that the ladder configuration of atomic levels can yield the same effect, reducing the total Doppler width of the two photon process. Kasapi [41] has applied EIT to isotope discrimination. By adjusting the intensity of a coupling laser, one isotope in a mixture of two is made resonantly opaque while the other is rendered transparent to a probe. Ling et al. [42] showed that the realistic experimental situation where an effective $\Lambda$ system is realised in multi-Zeeman-sublevel atoms (with e.g. 11 relevant atomic sublevels) is formally identical to the idealised situation of only three levels. EIT is also very appealing for a variety of applications due to its efficient noise reduction. Ling et al. [43] have proposed applying a standing wave coupling field to the atomic medium. Such a medium will be transparent to the weak probe at the antinodes of the coupling field and opaque at the nodes, creating what the authors called an electromagnetically induced grating. Scully [44] has shown that by including appropriate pumping, the refractive index can be increased at a point of vanishing absorption. Some of the most spectacular applications that have been achieved by this method are the reduction of the light velocity and the temporary storage of the light in an atomic EIT medium. Since these applications are related to the investigations presented in this thesis, they will be discussed later in comparison with our own work.

Finally, a short remark is in order about the use of a quantised cavity mode instead of a travelling wave as the probe field. In this case the basic conclusions of the previous discussion remain unchanged, and all the formulae are still valid provided we make the replacement $\Omega_p \rightarrow g\sqrt{n}$, where $g$ denotes the atom-field dipole coupling strength and $n$ is the number of photons in the cavity mode.

## 2.4 Generation of Giant Kerr Nonlinearity

Atomic coherence effects in general, and EIT in particular, can now be used to realise nonlinear optical processes free of spontaneous emission noise. The first glimpse of this exciting new possibility was offered by Harris et al. [45], who showed how a resonantly enhanced third order nonlinearity can be obtained while at the same time inducing transparency in the medium. Their analysis predicted improvements in conversion efficiency and parametric gain of many orders of magnitude. The predictions were indeed verified in the experiment of Hakuta et al. [46].

The system used to generate a giant Kerr nonlinearity, which is the main theme of this thesis, was proposed by Schmidt and Imamoglu [47]. It is a four level system with
2.4. GENERATION OF GIANT KERR NONLINEARITY

the energy level scheme shown in Figure 2.3. At the heart of this system lies an EIT scheme, consisting of levels |1⟩, |2⟩ and |3⟩. Levels |2⟩ and |3⟩ are coherently coupled by a classical laser field with Rabi frequency Ωc. An atomic system exhibits EIT if the probe beam (frequency ω_{12}) and the coupling beam of Rabi frequency Ωc are on two photon resonance with the transition |1⟩ → |3⟩. The state |3⟩ is assumed to be metastable so its decay rate can be neglected. In addition, a field of frequency ω_{34} is driving the atomic transition |3⟩ → |4⟩, and is detuned by Δ from the resonance. This is the key part of the scheme: coupling to the level |4⟩ induces an effective ac-Stark shift on the metastable ground state, thus inducing an effective third order nonlinearity.

Schmidt and Imamoglu [47] have solved the coupled amplitude equations for this system assuming the perturbative conditions Δ ≫ γ_3, γ_2, γ_3, Ω_c ≫ Ω_{12}, Ω_{34}, where Ω_{12} and Ω_{34} denote Rabi frequencies of the respective transitions. From the (nonlinear) coefficients, an effective third order nonlinearity is found as

\[ \chi^{(3)} = \frac{N|\mu_{13}|^2|\mu_{34}|^2}{2\epsilon_0\hbar^3} \frac{1 + i\gamma_3/\Delta}{\Omega_c^2\Delta}, \] (2.5)

where \( \mu_{ij} \) denotes the dipole matrix element for a given transition and \( N \) is the number of atoms. The size of the nonlinearity is obviously determined by the size of the ac-Stark shift \( |\mu_{34}|^2/\Delta \). In addition, the dependence \( \chi^{(3)} \propto \Omega_c^{-2} \) suggests that the loss in nonlinearity due to detuning \( \Delta \) can be compensated by reducing \( \Omega_c \). Note that

\[ \frac{\text{Im}[\chi^{(3)}]}{\text{Re}[\chi^{(3)]}} = \frac{\gamma_3}{\Delta}, \] (2.6)
i.e. nonlinear absorption is negligible and the nonlinearity is predominantly dispersive in nature.

This scheme can be viewed as a four level generalisation of a conventional cross–phase modulation (XPM) scheme, realised in a three level ladder configuration (Fig. 2.4). In the conventional scheme one can increase the nonlinearity by decreasing the detunings, especially the one from the intermediate level $|i\rangle$, but the effective nonlinearity in this case also decreases rapidly as $\sim 1/\Delta \omega_a^2 \Delta \omega_b$. Also, decrease in detuning $\Delta \omega_a$ causes an increase in the absorption of the incident wave at $\omega_a$. Another limit to the size of nonlinear effect comes from the decay rates which largely determine the values of both $\chi^{(3)}$ and $\text{Im}[\chi^{(1)}]$ for very small detunings. Finally, the conventional XPM scheme has a nonzero contribution to self–phase modulation that is not desirable if one is interested in the phase shifts of the wave at $\omega_a$, induced by the wave at $\omega_b$.

Comparison of the dispersive part of the nonlinearity in these two schemes shows an improvement of ten orders of magnitude obtained by the EIT–Kerr scheme as compared to conventional schemes. This shows that, in principle, one can obtain arbitrarily large XPM phase shifts in the probe field by arbitrarily weak signal fields. In particular, phase shifts of the order of $\pi$ should be achievable with single photons. An excellent review of this scheme is given by Parkins, Walls and Imamoglu [48] (see [49]).
2.5 Photon Blockade

Large single photon phase shifts are very appealing for their potential use in quantum computing and quantum communication, but their production is notoriously difficult because of the small sizes of conventional nonlinearities. The largest phase shift to date is the one achieved by Turchette et al. [50] with a single two level atom strongly coupled to a high finesse optical cavity. The maximum phase shift measured was $16\degree$ per intracavity photon. Clearly the possibility of phase shifts of order $\pi$ is very appealing in this context.

Imamoğlu et al. [51] have shown that, given a sufficiently large pure $\chi^{(3)}$ nonlinearity in an optical cavity, the effect of photon blockade occurs, as a consequence of large phase shifts. The envisaged experimental setup for demonstrating photon blockade is shown in Figure 2.5. The addition of a large nonlinearity in the cavity induces an anharmonic shift to the otherwise harmonic energy levels of the empty cavity. A first photon excites the cavity and detunes it via the $\chi^{(3)}$ interaction. If the shift is large enough, the driving field is sufficiently far from the new cavity resonance to preclude a second excitation until the first excitation decays via the emission of a phase shifted photon. The achievable phase shifts are proportional to the anharmonicity, which is in turn proportional to the nonlinearity.

Drummond and Walls [52] have presented a quantum treatment of the optical bistability of a $\chi^{(3)}$ medium. Although the operating regime for bistability does not coincide with the regime in which we expect to find photon blockade, the solutions they have obtained for the steady–state photon statistics are exact, and are thus valid independent of the particular regime of choice. Ignoring losses, a single–mode cavity with $\chi^{(3)}$ nonlinearity, driven externally by a classical coherent driving field $\mathcal{E}_p$, is described by the effective

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Figure 2.5: Envisaged experimental setup for the photon blockade. The coherent field $\mathcal{E}_p$ drives the cavity containing the $\chi^{(3)}$ nonlinearity exhibited by the four level atom. The additional coupling field $\Omega_c$ couples the appropriate atomic energy levels. The outgoing light is incident on a 50% – 50% beam splitter. Each arm of the beam splitter contains a high efficiency photodetector. The results of the photodetections are collected in a correlator, and the correlation function can be deduced.
Hamiltonian [52, 1]

\[ H = \hbar \omega_0 a^\dagger a + \hbar \eta (a^\dagger a)^2 + \hbar E_p (a - a^\dagger), \tag{2.7} \]

where \( \omega_0 \) denotes the frequency of the cavity mode, \( a \) is the mode annihilation operator and \( \eta \) is the anharmonicity parameter, closely related to the nonlinear susceptibility \( \chi^{(3)} \) (see Appendix B for a relation between anharmonicity and nonlinearity). Cavity losses can be included in this treatment by adding a non-Hermitian term to this Hamiltonian and considering the effective Hamiltonian \( \mathcal{H}_{\text{eff}} = \mathcal{H} - i\hbar \kappa a^\dagger a \), with \( \kappa \) denoting the cavity decay rate. Ignoring the driving term for a moment, the remainder of the Hamiltonian (2.7) is diagonal in the Fock basis for the intracavity field, and the energy of the \( n \)-th excitation is given as

\[ E_n = n\hbar \omega_0 + n(n - 1)\hbar \eta. \tag{2.8} \]

Hence the energy levels for \( n \geq 2 \) are shifted by an amount proportional to the anharmonicity \( \eta \). If this shift is larger than the linewidth of the corresponding states, photon blockade can be achieved. For a cavity characterised by damping constant \( \kappa \), Imamoglu et al. [51] have found that this condition amounts to \( \eta \gg \kappa, E_p \sqrt{\kappa} \).

The obvious way of looking at the photon blockade effect is that it is basically a system that converts the coherent (i.e. Poissonian) stream of photons entering the cavity from driving field \( E_p \) into a regular (sub-Poissonian) outgoing stream of photons, as shown in Fig. 2.5. If the photon blockade is successful, the outgoing light will be antibunched. The second order correlation function at zero time delay is then an appropriate measure for

![Figure 2.6: Plot of second order correlation function against anharmonicity parameter from the equation (2.9), with \( E_p/\kappa = 0.1 \).](image)
the photon blockade. For the pure $\chi^{(3)}$ optical nonlinearity, this correlation function has been calculated by Drummond and Walls \cite{52} to be

$$g^{(2)}(0) = \frac{\langle (a^\dagger a^2)^{ss} \rangle}{\langle (a^\dagger a)^{ss} \rangle^2} = \frac{|\kappa/\eta|^2 \, _2F_2(\kappa/\eta, \kappa/\eta, 2|\mathcal{E}_p/\eta|^2) \, _2F_2(\kappa/\eta + 2, \kappa/\eta + 2, 2|\mathcal{E}_p/\eta|^2)}{|\kappa/\eta + 1|^2 \, _2F_2(\kappa/\eta + 1, \kappa/\eta + 1, 2|\mathcal{E}_p/\eta|^2)}.$$  \hspace{1cm} (2.9)

where the quantity $\, _pF_q(a; b; c)$ is the generalised hypergeometric function. Its definition and properties can be found in standard references such as Abramowitz and Stegun \cite{53} and Gradshteyn and Ryzhik \cite{54}. We have also assumed that the anharmonicity $\eta$ is real. This is a necessary requirement, since the photon blockade is expected to work only if the intracavity medium is dispersive, rather than absorptive. Equation (2.6) shows that this requirement is satisfied to a good approximation. In Figure 2.6, the second order correlation function, Eq. (2.9), is plotted against the anharmonicity, with fixed $\mathcal{E}_p$. The plot shows that excellent antibunching is achieved for a sufficiently large anharmonicity.

The original proposal of Imamoglu et al. \cite{51} envisaged a four level atomic medium of density $\sim 10^{11}$ cm$^{-3}$ in a standard cavity QED environment. While appealing in its elegance, this setup was shown to be inappropriate for the realisation of photon blockade \cite{55, 56}. We now turn to the analysis of this system and propose an alternative setup containing a single four level atom in a high finesse optical cavity.
Part II

Many–Atom Cavity QED Environment
Chapter 3

Noise Reduction in the Many–Atom System

We will consider a four level atomic scheme that exhibits a giant Kerr nonlinearity as described in Chapter 2. At the heart of this scheme lies a three level \( \Lambda \)-configuration that is driven on Raman resonance. We analyse noise fluctuations in the many atom system. In particular, quadrature squeezing and intensity squeezing of the light in the output from the cavity are discussed.

3.1 Introduction

The proposal of Schmidt and Imamoğlu [47] for generation of giant Kerr nonlinearities, utilising electromagnetically induced transparency in an ensemble of four level atoms, has been reviewed in Chapter 2. In a continuation of their work, Imamoğlu et al. [51] derived an effective Hamiltonian (originally investigated by Drummond and Walls [52]) for an ideal \( \chi^{(3)} \) nonlinearity for the case in which the atomic variables are adiabatically eliminated and their quantum noise contribution is neglected. They found that strong antibunching of transmitted photons can be achieved, provided that the nonlinearity is large enough, and they predicted the photon blockade effect, a term chosen in analogy with the phenomenon of electron (Coulomb) blockade [57]. A full quantum analysis by Gheri, Walls and Marte [58] has also demonstrated how large effective \( \chi^{(3)} \) nonlinearity can be achieved in a three level \( \Lambda \)-system near its dark resonance.

The main objective of this Chapter is to investigate the validity of the effective Hamiltonian of reference [51], also given by equation (2.7). In particular, we want to explore the properties of the light coming out of the cavity containing the medium of four level atoms. We will then compare these properties with the properties of the light coming out of the cavity containing the pure Kerr nonlinearity, claimed to represent the adiabatic limit of the full atomic medium [51]. To that end, we compare the noise fluctuations of the light at the cavity output in both cases, described by their squeezing properties. We also comment on the significance to our results of an additional detuning \( \delta \) from energy level [2].
We consider a low density ensemble of \( N \) four level atoms in a cavity. The energy level scheme is very similar to the one in Figure 2.3. The differences are \( i \) the atoms are now placed in a cavity, with the cavity mode driving the transitions \( |1\rangle \rightarrow |2\rangle \) and \( |3\rangle \rightarrow |4\rangle \); and \( ii \) we now allow for an additional detuning from the excited state \( |2\rangle \) to the ground states, and \( \gamma_3 \) is the spontaneous decay rate of the state \( |4\rangle \). The driving field is at frequency \( \omega_{\text{cav}} \) and is resonant with the cavity, and \( \omega_c \) is the frequency of the coupling field.

3.2 Hamiltonian and the Equations of Motion

We consider a low density ensemble of \( N \) four level atoms in a cavity. The energy level scheme is very similar to the one in Figure 2.3. The differences are \( i \) the atoms are now placed in a cavity, with the cavity mode driving the transitions \( |1\rangle \rightarrow |2\rangle \) and \( |3\rangle \rightarrow |4\rangle \); and \( ii \) we now allow for an additional detuning from the excited level \( |2\rangle \), while keeping the two–photon resonance. The setup is shown in Figure 3.1.

The system Hamiltonian in the electric–dipole and rotating–wave approximations is

\[
\mathcal{H} = \mathcal{H}_1 + \mathcal{H}_2 + \mathcal{H}_3 + \mathcal{H}_4 + \mathcal{H}_{\text{baths}}. \tag{3.1a}
\]

\( \mathcal{H}_1 \) is the free part of the system Hamiltonian

\[
\mathcal{H}_1 = \sum_{\mu=1}^{N} \sum_{i=1}^{4} \hbar \nu_i \sigma_{ii}^\mu + \hbar \omega_{\text{cav}} a^\dagger a; \tag{3.1b}
\]
3.2. HAMILTONIAN AND THE EQUATIONS OF MOTION

$H_2$ describes the coupling of the atom to the travelling wave field mode

\[ H_2 = i \hbar g_1 \sum_{\mu=1}^{N} (a_\mu a_\mu e^{-i k_1 \cdot r^\mu} - a_\mu^\dagger e^{i k_1 \cdot r^\mu}) + i \hbar g_2 \sum_{\mu=1}^{N} (a_\mu b_\mu e^{-i k_1 \cdot r^\mu} - a_\mu^\dagger b^\dagger e^{i k_1 \cdot r^\mu}) + i \hbar \sum_{\mu=1}^{N} (\Omega_{\mu}^* a_\mu^\dagger e^{-i k_1 \cdot r^\mu} - \Omega_{\mu} a_\mu e^{i k_1 \cdot r^\mu}) , \] (3.1c)

$H_3$ describes damping of the cavity mode by the coupling with the field bath

\[ H_3 = i \hbar \int_{-\infty}^{+\infty} d\omega C(\omega) [b^\dagger(\omega) a - b(\omega) a^\dagger] ; \] (3.1d)

$H_4$ describes damping of the atoms by spontaneous emission

\[ H_4 = i \hbar \sum_{\mu=1}^{N} \int_{-\infty}^{+\infty} d\omega D_1(\omega) [\beta_1^\dagger(\omega) \sigma_1^\mu - \sigma_1^\mu \beta_1(\omega)] + i \hbar \sum_{\mu=1}^{N} \int_{-\infty}^{+\infty} d\omega D_2(\omega) [\beta_2^\dagger(\omega) \sigma_2^\mu - \sigma_2^\mu \beta_2(\omega)] + i \hbar \sum_{\mu=1}^{N} \int_{-\infty}^{+\infty} d\omega D_3(\omega) [\beta_3^\dagger(\omega) \sigma_3^\mu - \sigma_3^\mu \beta_3(\omega)] ; \] (3.1e)

$H_{bath}$ consists of the free Hamiltonians of the field and atomic baths

\[ H_{bath} = H_{bath}^{field} + H_{bath}^{atom} = \int_{-\infty}^{+\infty} d\omega \hbar \omega b^\dagger(\omega) b(\omega) + \sum_{j=1}^{3} \int_{-\infty}^{+\infty} d\omega \hbar \omega \beta_j^\dagger(\omega) \beta_j(\omega) . \] (3.1f)

The bath operators satisfy bosonic commutation relations

\[ [b_j(\omega), b_{j'}^\dagger(\omega')] = \delta_{jj'} \delta(\omega - \omega') , \] (3.2a)

\[ [\beta_j(\omega), \beta_{j'}^\dagger(\omega')] = \delta_{jj'} \delta(\omega - \omega') . \] (3.2b)

The definitions of quantities in these equations are given in Table 3.2

The coupling constants $g_j$ are related to the dipole matrix elements $\mu_j$ of equation (2.5) by $g_j = \sqrt{\omega_j/(2r_0 \hbar V_{cav})}\mu_j$, where $\omega_j$ denotes the atomic transition frequency, and $V_{cav}$ is the cavity volume. In the first Markov approximation one assumes that the couplings with the baths are constant over a broad band of frequencies. This means we can take $C^2(\omega) = 2\kappa/2\pi$, and $D_j^2(\omega) = \gamma_j/2\pi$. 

\[ C^2(\omega) = 2\kappa/2\pi, \] 
\[ D_j^2(\omega) = \gamma_j/2\pi. \] 

\[ B_j = \gamma_j. \]
CHAPTER 3. NOISE REDUCTION IN THE MANY-ATOM SYSTEM

### Quantity Definition

<table>
<thead>
<tr>
<th>Quantity</th>
<th>Definition</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\sigma_{kl}^{\mu}$</td>
<td>operators for the atomic populations (for $k = l$) and atomic transitions (for $k \neq l$) in the $\mu$-th atom</td>
</tr>
<tr>
<td>$a, a^\dagger$</td>
<td>boson operators for the cavity field mode</td>
</tr>
<tr>
<td>$h\nu_i$</td>
<td>energy of the $i$th atomic level</td>
</tr>
<tr>
<td>$\Omega_c$</td>
<td>Rabi frequency of the coupling field</td>
</tr>
<tr>
<td>$g_1, g_2$</td>
<td>respective coupling constants for the transitions $</td>
</tr>
<tr>
<td>$k_1$</td>
<td>wave vector of the cavity mode</td>
</tr>
<tr>
<td>$k_3$</td>
<td>wave vector of the coupling mode</td>
</tr>
<tr>
<td>$r^\mu$</td>
<td>position of the $\mu$-th atom</td>
</tr>
<tr>
<td>$b(\omega), b^\dagger(\omega)$</td>
<td>boson operators for the field bath</td>
</tr>
<tr>
<td>$\beta_j(\omega), \beta_j^\dagger(\omega)$</td>
<td>atomic bath operators for each transition ($j=1, 2, 3$)</td>
</tr>
<tr>
<td>$C(j)$</td>
<td>coupling coefficient between the photon bath and cavity mode</td>
</tr>
<tr>
<td>$D_j(\omega)$</td>
<td>coupling coefficient between the atomic bath and the atomic transitions ($j=1, 2, 3$)</td>
</tr>
<tr>
<td>$\kappa$</td>
<td>decay rate of the cavity mode</td>
</tr>
<tr>
<td>$\gamma_j$</td>
<td>atomic decay rate for the $j$-th transition</td>
</tr>
</tbody>
</table>

#### Table 3.1: Definitions of relevant quantities.

### 3.2.1 Equations of Motion

We shall take $a$ and $b(\omega)$ to be in a frame rotating at the frequency of the driving field. Since the driving field is assumed to be resonant with the cavity mode, the frame rotates at the frequency $\omega_{\text{cav}}$. Input fields are defined as [3, 4, 5]:

$$a^{\text{in}}(t) = \frac{1}{\sqrt{2\pi}} \int_{-\infty}^{+\infty} d\omega e^{-i\omega(t-t_0)} b^0(\omega) ;$$  \hspace{1cm} (3.3a)

$$\beta_j^{\text{in}}(t) = \frac{1}{\sqrt{2\pi}} \int_{-\infty}^{+\infty} d\omega e^{-i\omega(t-t_0)} \beta_j^0(\omega) ,$$  \hspace{1cm} (3.3b)

for $t > t_0$. Here, $b^0(\omega)$ is the value of $b(\omega)$ at $t = t_0$ and $\beta_j^0(\omega)$ is the value of $\beta_j(\omega)$ at $t = t_0$. The input to the cavity, $a^{\text{in}}(t)$, is related to the output from the cavity, $a^{\text{out}}(t)$, by the relation

$$a^{\text{out}}(t) - a^{\text{in}}(t) = \sqrt{2\kappa} a .$$  \hspace{1cm} (3.4)

We assume a vacuum reservoir for the cavity mode and the atomic noise, giving the only non-vanishing noise correlation functions

$$\langle a^{\text{in}}(t), a^{\text{in}}(t') \rangle = \delta(t-t') ,$$  \hspace{1cm} (3.5a)

$$\langle \beta_j^{\text{in}}(t), \beta_j^{\text{in}}(t') \rangle = \delta_{ij} \delta(t-t') .$$  \hspace{1cm} (3.5b)
The scaled atomic operators for the whole atomic system are defined as

\[ S_{ii} = \frac{1}{N} \sum_{\mu=1}^{N} \sigma_{ii}^{\mu}, \]  
\[ S_{12} = \frac{1}{N} \sum_{\mu=1}^{N} \sigma_{12}^{\mu} e^{-i k_{1} \cdot r^{\mu}}, \]  
\[ S_{23} = \frac{1}{N} \sum_{\mu=1}^{N} \sigma_{23}^{\mu} e^{i k_{3} \cdot r^{\mu}}, \]  
\[ S_{34} = \frac{1}{N} \sum_{\mu=1}^{N} \sigma_{34}^{\mu} e^{-i k_{1} \cdot r^{\mu}}, \]

with obvious definitions for the adjoint operators \( S_{21}, S_{32}, S_{43} \). Making the assumption that the atoms are independent, \( \sigma_{ij}^{\mu} \delta_{kl}^{\mu} = \delta_{jk}^{\mu} \sigma_{il}^{\mu} \), we obtain multiplication rules for the collective atomic operators:

\[ S_{ij} S_{kl} = \frac{1}{N} \delta_{jk} S_{il}. \]  

We make the scalings \( \tilde{a} = a/\sqrt{N} \), and \( \tilde{g}_{j} = g_{j} \sqrt{N} \), and define the (scaled) population difference operators as

\[ D_{1} = S_{22} - S_{11}, \]  
\[ D_{2} = S_{33} - S_{22}, \]  
\[ D_{3} = S_{44} - S_{33}. \]

The Langevin equation of motion (in a frame rotating at the driving field frequency \( \omega_{\text{cav}} \)) for the field mode is then easily derived as

\[ \dot{\Omega}_{d} = -\kappa (\Omega_{d} - C_{1} \gamma_{1} S_{12} - C_{12} \gamma_{3} S_{34}) - \sqrt{2\kappa} \Omega_{d}^{in}. \]  

We define the Rabi operator as \( \Omega_{d} = g_{1} a \) and choose to write equation (3.9) in terms of this, rather than the mode annihilation operator. Although the choice between the two forms is arbitrary, this way the equation is written in terms of cooperativities \( C_{j} \), which are commonly used for the description of many atom CQED systems. The parameter \( g_{2} \) could equally well have been chosen instead of \( g_{1} \), but since throughout our simulations we assume that \( g_{1} = g_{2} \), this makes no difference. Cooperativity parameters are defined as \( C_{1} = \tilde{g}_{1}^{2}/(\gamma_{1} \kappa), \) \( C_{2} = \tilde{g}_{2}^{2}/(\gamma_{3} \kappa) \) and \( C_{12} = \tilde{g}_{1} \tilde{g}_{2}/(\gamma \kappa) \), where \( \gamma = (\gamma_{1} + \gamma_{2})/2 \). The term cooperativity comes from laser theory \([59]\), where the necessary requirement for laser action is \( C > 1 \). Briefly, it is the ratio of coherent coupling to noise production in the system. The noise operator is consequently defined as \( \Omega_{d}^{in} = g_{1} a^{in} \). For simplicity, we have chosen to incorporate the coherent driving amplitude in this operator, i.e.

\[ \langle \Omega_{d}^{in} \rangle = -g_{1} \mathcal{E}_{p}/\sqrt{2\kappa}. \]
CHAPTER 3. NOISE REDUCTION IN THE MANY-ATOM SYSTEM

Note that the noise correlations (3.5a) remain the same (up to a numerical factor) regardless of their mean values (3.10a). Consistent with the assumption of vacuum atomic noise, we have

\[ \langle \beta_j(t) \rangle = 0. \]  

The equations for the population differences are obtained from the equations for the populations after a straightforward, if somewhat cumbersome, calculation:

\[ \dot{D}_1 = \frac{-2\gamma_1 + \gamma_2}{4} - \frac{2\gamma_1 + \gamma_2}{4} (D_1 - 2D_2 - D_3) - 2(S_{21}\Omega_d + \Omega_{d1}^T S_{12}) - (\Omega_{c}^* S_{32} + \Omega_{c} S_{23}) \]
\[ - 2\sqrt{\frac{\gamma_1}{N}} \sum_{\mu=1}^{N} (\sigma_{21}^{\mu} \beta_{1}^{\mu \dagger} + \beta_{1}^{\mu \dagger} \sigma_{12}^{\mu}) - \sqrt{\frac{\gamma_2}{N}} \sum_{\mu=1}^{N} (\sigma_{23}^{\mu} \beta_{2}^{\mu \dagger} + \beta_{2}^{\mu \dagger} \sigma_{32}^{\mu}), \]  

\[ \dot{D}_2 = \frac{\gamma_1 + 2\gamma_2 + \gamma_3}{4} + \frac{\gamma_1 + 2\gamma_2 + \gamma_3}{4} D_1 - \frac{\gamma_1 + 2\gamma_2 - \gamma_3}{2} D_2 - \frac{\gamma_1 + 2\gamma_2 - 3\gamma_3}{4} D_3 \]
\[ + (S_{21}\Omega_d + \Omega_{d1}^T S_{12}) + 2(\Omega_{c} S_{23} + \Omega_{c}^* S_{32}) + \frac{g_2}{g_1} (S_{43}\Omega_d + \Omega_{d1}^T S_{34}) \]
\[ + \sqrt{\frac{\gamma_1}{N}} \sum_{\mu=1}^{N} (\sigma_{21}^{\mu} \beta_{1}^{\mu \dagger} + \beta_{1}^{\mu \dagger} \sigma_{12}^{\mu}) + 2\sqrt{\frac{\gamma_2}{N}} \sum_{\mu=1}^{N} (\sigma_{23}^{\mu} \beta_{2}^{\mu \dagger} + \beta_{2}^{\mu \dagger} \sigma_{32}^{\mu}) \]
\[ + \sqrt{\frac{\gamma_3}{N}} \sum_{\mu=1}^{N} (\sigma_{43}^{\mu} \beta_{3}^{\mu \dagger} + \beta_{3}^{\mu \dagger} \sigma_{34}^{\mu}), \]  

\[ \dot{D}_3 = \frac{-\gamma_2 + 2\gamma_3}{4} - \frac{\gamma_2 + 2\gamma_3}{4} D_1 - \left( \gamma_3 - \frac{\gamma_2}{2} \right) D_2 - \frac{6\gamma_3 - \gamma_2}{4} D_3 \]
\[ - 2\frac{g_2}{g_1} (S_{43}\Omega_d + \Omega_{d1}^T S_{34}) - (\Omega_{c} S_{23} + \Omega_{c}^* S_{32}) \]
\[ - \sqrt{\frac{\gamma_2}{N}} \sum_{\mu=1}^{N} (\sigma_{23}^{\mu} \beta_{2}^{\mu \dagger} + \beta_{2}^{\mu \dagger} \sigma_{32}^{\mu}) - 2\sqrt{\frac{\gamma_3}{N}} \sum_{\mu=1}^{N} (\sigma_{43}^{\mu} \beta_{3}^{\mu \dagger} + \beta_{3}^{\mu \dagger} \sigma_{34}^{\mu}). \]

The factor \( \frac{g_2}{g_1} \) comes from the terms \( g_2 a \sigma_{ij}^{\mu} \) in the Hamiltonian and their adjoints. We could, in principle, define a second Rabi operator \( \Omega_2 = g_2 a \), but this would only introduce unnecessary complication into the equations. Note that the populations are related to the population differences through the following simple transformations:

\[ S_{11} = (1 - 3D_1 - 2D_2 - D_3)/4, \]  

\[ S_{22} = (1 + D_1 - 2D_2 - D_3)/4, \]  

\[ S_{33} = (1 + D_1 + 2D_2 - D_3)/4, \]  

\[ S_{44} = (1 + D_1 + 2D_2 + 3D_3)/4, \]  

and that the total population conservation condition, \( \sum_{j=1}^{4} S_{jj} = 1 \), is satisfied automatically. An interesting feature of the population difference equations are the free coefficients
on the right hand sides, which describe linear decay or growth of a particular population difference. Terms proportional to \(D_j\) in each equation describe the interplay of the population differences, which is best understood in terms of the individual level populations.

The population of level \(|2\rangle\) decays into level \(|1\rangle\) with rate \(\gamma_1\) and into level \(|3\rangle\) with rate \(\gamma_2\). The population of level \(|3\rangle\) is additionally enhanced by the decay of level \(|4\rangle\) with rate \(\gamma_3\).

The equations for the collective transition operators are

\[
\dot{S}_{12} = -\left(\frac{\gamma_1 + \gamma_2}{2} + i\Delta_{21}\right)S_{12} + D_1\Omega_d - \Omega_c S_{13} \\
+ \sqrt{\frac{\gamma_1}{N}} \sum_{\mu=1}^{N} (\sigma_{22}^\mu - \sigma_{11}^\mu)\beta_{1}^{\mu \text{in}} - \sqrt{\frac{\gamma_1}{N}} \sum_{\mu=1}^{N} \beta_{1}^{\mu \text{in}}\sigma_{13},
\]

(3.13a)

\[
\dot{S}_{23} = -\left(\frac{\gamma_1 + \gamma_2}{2} + i\Delta_{32}\right)S_{23} - \Omega_d^\dagger \left(S_{13} - \frac{g_2}{g_1}S_{24}\right) - \Omega_c^*D_2 \\
- \sqrt{\frac{\gamma_1}{N}} \sum_{\mu=1}^{N} \beta_{2}^{\mu \text{in}}\sigma_{13} + \sqrt{\frac{\gamma_2}{N}} \sum_{\mu=1}^{N} \sigma_{24}\beta_{2}^{\mu \text{in}} - \sqrt{\frac{\gamma_2}{N}} \sum_{\mu=1}^{N} (\sigma_{33}^\mu - \sigma_{22}^\mu)\beta_{3}^{\mu \text{in}},
\]

(3.13b)

\[
\dot{S}_{34} = -\frac{\gamma_3}{2} + i\Delta_{43}\right)S_{34} + \frac{g_2}{g_1}D_3\Omega_d + \Omega_c S_{24} \\
+ \sqrt{\frac{\gamma_2}{N}} \sum_{\mu=1}^{N} (\sigma_{44}^\mu - \sigma_{33}^\mu)\beta_{2}^{\mu \text{in}} + \sqrt{\frac{\gamma_3}{N}} \sum_{\mu=1}^{N} \beta_{3}^{\mu \text{in}}\sigma_{24},
\]

(3.13c)

\[
\dot{S}_{13} = -i\Delta_{31}S_{13} + S_{23}\Omega_d + \frac{g_2}{g_1}\Omega_d^\dagger S_{14} + \Omega_c^*S_{12} \\
+ \sqrt{\frac{\gamma_1}{N}} \sum_{\mu=1}^{N} \sigma_{23}\beta_{1}^{\mu \text{in}} + \sqrt{\frac{\gamma_2}{N}} \sum_{\mu=1}^{N} \beta_{1}^{\mu \text{in}}\sigma_{14} + \sqrt{\frac{\gamma_3}{N}} \sum_{\mu=1}^{N} \sigma_{12}\beta_{3}^{\mu \text{in}},
\]

(3.13d)

\[
\dot{S}_{24} = -\left(\frac{\gamma_1 + \gamma_2 + \gamma_3}{2} + i\Delta_{42}\right)S_{24} - \Omega_d^\dagger S_{14} - \frac{g_2}{g_1}S_{23}\Omega_d - \Omega_c^*S_{34} \\
- \sqrt{\frac{\gamma_1}{N}} \sum_{\mu=1}^{N} \beta_{1}^{\mu \text{in}}\sigma_{14} - \sqrt{\frac{\gamma_2}{N}} \sum_{\mu=1}^{N} \sigma_{23}\beta_{2}^{\mu \text{in}} - \sqrt{\frac{\gamma_3}{N}} \sum_{\mu=1}^{N} \sigma_{34}\beta_{3}^{\mu \text{in}},
\]

(3.13e)

\[
\dot{S}_{14} = -\frac{\gamma_3}{2} + i\Delta_{41}\right)S_{14} + \left(S_{24} - \frac{g_2}{g_1}S_{13}\right)\Omega_d \\
+ \sqrt{\frac{\gamma_2}{N}} \sum_{\mu=1}^{N} \beta_{2}^{\mu \text{in}}\sigma_{14} - \sqrt{\frac{\gamma_3}{N}} \sum_{\mu=1}^{N} \sigma_{13}\beta_{2}^{\mu \text{in}},
\]

(3.13f)

and the equations for the adjoint operators follow from these. The atomic detunings are defined consistently with Figure 3.1,

\[
\Delta_{21} = (\nu_2 - \nu_1) - \omega_{\text{cav}} = \delta,
\]

(3.14a)

\[
\Delta_{32} = (\nu_3 - \nu_2) + \omega_c = -\delta,
\]

(3.14b)
\[ \Delta_{31} = (\nu_3 - \nu_1) - (\omega_{\text{cav}} - \omega_c) = \Delta_{21} + \Delta_{32} = 0, \quad (3.14c) \]
\[ \Delta_{43} = (\nu_4 - \nu_3) - \omega_{\text{cav}} = \Delta, \quad (3.14d) \]
\[ \Delta_{42} = (\nu_4 - \nu_2) - (\omega_{\text{cav}} - \omega_c) = \Delta_{43} + \Delta_{32} = \Delta - \delta, \quad (3.14e) \]
\[ \Delta_{41} = (\nu_4 - \nu_1) - (2\omega_{\text{cav}} - \omega_c) = \Delta_{43} + \Delta_{32} + \Delta_{21} = \Delta_{43} + \Delta_{31} = \Delta. \quad (3.14f) \]

Note that these equations are quantum Langevin equations (QLE) for the system operators. We emphasize that the transition to the Itô calculus in this particular case is very simple - the Itô quantum stochastic equations (QSDE) have exactly the same form as the quantum Langevin equations. This follows from the fact that field variables couple to a vacuum bath, as do the atomic variables. Hence, the transition from the QLEs to the Itô QSDEs does not entail the addition of any new terms (for details, see Gardiner and Zoller [5, Section 5.3.7]).

### 3.3 Spectrum of Best Squeezing and Noise

At this point we will make an explicit assumption about the density of the atomic medium, an assumption that is already implicitly incorporated in the derivation of the equations of motion\(^1\). Specifically, we assume a low density medium, where the atoms are at least several wavelengths of the field apart. The introduction of the effective coupling constants (obtained by scaling \(g_j \rightarrow g_j \sqrt{N} \)) and the collective atomic operators produced equations of motion that have almost the same analytical appearance as the single atom equations, with only the summation in the noise terms revealing their collective nature. In particular, note that the collective operators, defined by (3.6), describe the net effect of the interaction averaged over the interaction volume.

The noise increments are linear combinations of the atomic coherence operators and bath operators [25, 5]. The introduction of collective atomic operators results in corresponding collective noise increments whose correlation coefficients are proportional to \(N^{-1} \), which is a consequence of the independent atoms assumption. One may use this to think of the individual noise increments as terms of order \(N^{-1/2} \), often referred to as the mesoscopic terms [60], which separate the terms in the time evolution equations into two groups with distinctively different sizes. This enables us to expand the system operators in an asymptotic power series and this is the essence of the system size expansion. We arrange all of the system operators in a vector
\[
\mathbf{v}(t) = (\Omega_d, \Omega_d^\dagger, S_{12}, S_{21}, S_{23}, S_{32}, S_{34}, S_{43}, S_{13}, S_{31}, S_{24}, S_{42}, S_{14}, S_{41}, D_1, D_2, D_3)^T. \quad (3.15)
\]

\(^1\)This assumption was critical in the derivation of noise terms, since it implies that each of the atoms may be considered as an independent dipole, decaying in its own private reservoir.
Then we expand this quantity into a power series to get

$$v(t) = \langle v(t) \rangle + \delta v(t) + O(1/N) ,$$  \hspace{1cm} (3.16)

where the term $\delta v(t)$ is of order $N^{-1/2}$. Truncation of the higher order terms $O(1/N)$ constitutes the linearised fluctuation approximation. In this expansion, $\langle v(t) \rangle$ is a $c$-number vector that describes the semiclassical behaviour of the system, while quantum features remain accounted for by the noise terms $\delta v(t)$.

Equations (3.9), (3.11) and (3.13) can now be rewritten as equations for the fluctuation operators in the linearised fluctuation approximation,

$$\delta \hat{v}(t) = -\hat{A}\delta v(t) + B(t) ,$$  \hspace{1cm} (3.17)

where $B(t)$ is the noise vector. The elements of the matrix $\hat{A}$ are components of the vector of deterministic solutions $\langle v(t) \rangle$. Note that the necessary condition that justifies the linearisation procedure is the existence of well-defined (nonzero) amplitudes $\langle v(t) \rangle$, such that the noise terms $\delta v(t)$ present only small fluctuations. In our case in particular, linearisation assumes that the cavity mode contains a sufficiently large number of photons that a field with a reasonably well-defined coherent amplitude is established in the cavity. While this is admittedly a departure from a strict photon blockade regime, it will enable the comparison of the full system with the adiabatic limit of Imamoglu et al. [51].

The independent atoms approximation enables us to calculate the noise correlations from the sums over atoms in $B(t)$ rather simply, giving

$$\langle B(t)B^T(s) \rangle = G(t - s) ,$$  \hspace{1cm} (3.18)

where $G$ denotes the correlation matrix. The equation (3.17) has the general solution

$$\delta v(t) = \delta v(0)e^{-\hat{A}t} + \int_0^t e^{-\hat{A}(t-t')}dB(t') .$$  \hspace{1cm} (3.19)

These expressions can now be used to develop a formalism for the analysis of quantum fluctuations.

Define the covariance matrix by

$$C(|t - s|) = \langle \delta v(t), \delta v^T(s) \rangle ,$$  \hspace{1cm} (3.20)

and the definition of the stationary covariance matrix follows as $C_0 = \langle \delta v(t), \delta v^T(t) \rangle$. Using equations (3.18) and (3.19), the following relations can be shown to be valid

$$G = A C_0 + C_0 A ,$$  \hspace{1cm} (3.21a)

$$C(|t - s|) = e^{-\hat{A}(t-s)}C_0 \Theta(t - s) + \Theta(s - t) C_0 e^{-\hat{A}^T(s-t)} ,$$  \hspace{1cm} (3.21b)

2Note that the linear fluctuation approximation reduces quantum electrodynamics to the stochastic electrodynamics [61, 62], where the field is treated classically, with added vacuum fluctuations.
where the step function $\Theta(\tau)$ is defined as

$$
\Theta(\tau) = \begin{cases} 
1 & \text{for } \tau > 0 \\
\frac{1}{2} & \text{for } \tau = 0 \\
0 & \text{for } \tau < 0
\end{cases}.
$$

The spectral matrix for the fluctuations in the cavity output field is calculated using the Wiener–Khinchin theorem [7] and equations (3.21), (3.4)

$$
S(\omega) = \frac{1}{2\pi} \int_{-\infty}^{\infty} d\tau \, e^{-i\omega\tau} \, 2\kappa \, C(\tau)
= \frac{\kappa}{\pi} (A + i\omega \cdot 1)^{-1} (A^{\dagger} - i\omega \cdot 1)^{-1}.
$$

Having obtained an expression for the spectral matrix, we would like to calculate the best squeezing spectrum as a measure of how strongly the fluctuations can be suppressed. We are interested in the best squeezing spectrum in the quadrature representation, i.e. to find that quadrature of light having the minimum fluctuations. We start by defining the amplitude and phase quadratures

$$
X = a + a^{\dagger}, \quad Y = -i(a - a^{\dagger}),
$$

which will be used as a basis in the quadrature space. A general quadrature is then defined as

$$
Q(\tau) = \cos \phi \, X(\tau) + \sin \phi \, Y(\tau),
$$

where the angle $\phi$ is called the quadrature angle. To examine the squeezing properties, we define the quadrature variance as $V_Q = \langle \delta Q(\tau), \delta Q(0) \rangle$, which can be expressed in terms of variances in amplitude and phase as

$$
V_Q = \cos^2 \phi \, V_X + \sin^2 \phi \, V_Y + \frac{1}{2} \sin 2\phi \, V_{XY},
$$

where now $V_X = \langle \delta X(\tau), \delta X(0) \rangle$, $V_Y = \langle \delta Y(\tau), \delta Y(0) \rangle$ and $V_{XY} = \langle \delta X(\tau), \delta Y(0) \rangle + \langle \delta Y(\tau), \delta X(0) \rangle$. The requirement $\partial V_Q / \partial \phi = 0$ defines an angle for which the variance takes on a minimum, giving the most favorable quadrature. This is readily calculated as

$$
\cos 2\phi = \frac{V_Y - V_X}{\sqrt{(V_Y - V_X)^2 + V_{XY}^2}}.
$$

Substituting this expression into (3.26) gives the variance of the most favorable quadrature

$$
V_Q^{(\phi)} = \frac{1}{2} \left[ V_X + V_Y - \left( [V_X - V_Y]^2 + V_{XY}^2 \right)^{1/2} \right].
$$
3.4. RESULTS

The variances $V_X$, $V_Y$ and $V_{XY}$ are readily obtained from the covariance matrix $C(\tau)$ as a simple linear combination of appropriate matrix elements. For example

$$V_X = \langle \delta X(\tau), \delta X(0) \rangle$$

$$= \langle \delta a(\tau), \delta a(0) \rangle + \langle \delta a^\dagger(\tau), \delta a^\dagger(0) \rangle + \langle \delta a^\dagger(\tau), \delta a(0) \rangle$$

$$= (C_{11} + C_{22} + C_{12} + C_{21})/g_1^2,$$ (3.28)

where $C_{ij}$ denotes element $(i, j)$ of matrix $C(\tau)$. Taking the Fourier transform, we find the spectrum of best squeezing in terms of the elements of the spectral matrix (3.23)

$$S^\text{best}_Q(\omega) = \frac{\kappa}{\pi} \left[ S_{12}(\omega) + S_{21}(\omega) - 2\sqrt{S_{11}(\omega)S_{22}(\omega)} \right],$$ (3.29a)

$$\phi(\omega) = \frac{1}{2} \cos^{-1} \left[ \frac{S_{11}(\omega) + S_{22}(\omega)}{2\sqrt{S_{11}(\omega)S_{22}(\omega)}} \right],$$ (3.29b)

where $S_{ij}$ denotes element $(i, j)$ of matrix $S(\omega)$. We have therefore expressed the spectrum of best squeezing in terms of drift and diffusion matrices $A$ and $G$, whose elements are composed of the elements of the mean value vector $\langle \nu \rangle$.

Note that the best squeezing spectrum determined by equations (3.29) gives information about the optimum squeezing that can be achieved for a particular frequency $\omega$. For two different frequencies the best squeezing does not normally occur in the same quadrature. Therefore the phase angle $\phi$ determining the most favourable quadrature is also a function of frequency $\omega$.

3.4 Results

The results of the previous Section will now be applied and the spectrum of best squeezing will be calculated from the equations (3.29). These results will be compared to the adiabatic case which is the case of pure optical $\chi^{(3)}$ nonlinearity, described by the following QLE

$$\dot{a} = -\kappa \left( 1 + \frac{2i\eta}{\kappa} a^\dagger a \right) a - \sqrt{2\kappa} a^{in},$$ (3.30)

Again, as with the equations for the full atomic system, this is a QLE, but its Itô counterpart has exactly the same form due to the vacuum noise input, assumed in equation (3.5a). The condition for the validity of the adiabatic elimination of atomic degrees of freedom is the condition which determines the validity of equation (3.30) for the description of the system dynamics, and has been identified as [55, 56, 63]

$$g_{eff} = \frac{g_1^2 N}{\Omega_e^2} \ll 1.$$ (3.31)

$^3$The factor $1/g_1^2$ is due to the fact that we have chosen to work with the Rabi operator $\Omega_d = g_1 a$, rather than the field annihilation operator $a$. 
We will return to this condition and its meaning later in this Chapter and in the following
Chapter. Assuming this condition is satisfied, we have shown in previous work [64, 65]
that it is indeed possible to choose such atomic parameters that minimize the remaining
atomic noise. In particular, the region of maximum noise reduction can be achieved by
simple adjustment of the atomic detunings $\Delta$ and $\delta$. We have obtained the result for the
anharmonicity parameter as
\[
\eta = g_{\text{eff}} \left( \frac{g_2^2 \Delta}{\gamma_3^2 + \Delta^2} - \frac{g_1^2 \delta}{(\gamma_1 + \gamma_2)^2 + \delta^2} \right), \tag{3.32}
\]
in the adiabatic limit, and we note that this result coincides with the result of Gheri et al. [63], who assumed $\delta = 0$ and therefore obtained only the first part in the brackets of
equation (3.32). The anharmonicity calculated by Imamoglu et al. [51] further assumes
that $\Delta \gg \gamma_3$. The additional difference between the anharmonicity of equation (3.32) (and
the result of Gheri et al.) on one side and the expression for the anharmonicity obtained
by Imamoglu et al. is the factor of 3, and the origin of this difference is explained in
Appendix B.

The condition (3.31) puts a very stringent limit on the required cavity parameters. In
particular, we note that the parameters envisaged in the original proposal for the photon
blockade effect [51] do not satisfy the adiabatic condition. It is worthwhile considering this
choice of parameters in some detail, and see how can one vary parameter $g_{\text{eff}}$ to pass
from the non–adiabatic limit to the adiabatic limit. Imamoglu et al. assumed a cavity
of volume $V_{\text{cav}} = 1 \times 10^{-4}$ cm$^3$, and length 2 cm, in which the atomic medium, of length
1 cm and density $n_{\text{at}} = 3 \times 10^{11}$ cm$^{-3}$, is placed. For a typical upper state decay rate
$\gamma_j = \gamma = 3 \times 10^7$ s$^{-1}$, the values of coupling constants and detunings can be deduced
as $\Omega_\omega = 15\gamma$, $\delta = 0$, $\Delta = 30\gamma$ and $g_j \sqrt{N} = 160\gamma$. If the cavity finesse is $\mathcal{F} = 10^4$, the
cavity decay rate can be calculated to give $\kappa = 0.1 \gamma$. These parameters combined give an
anharmonicity of $\eta/\kappa \approx 20$, which is the value we will use for a comparison. The adiabatic
parameter can be deduced as $g_{\text{eff}} \sim 10^2$, implying that the adiabatic approximation is
manifestly invalid for this particular set of parameters.

Figure 3.2 shows the results of the simulations for the full model and for the same
field quadrature in the adiabatic limit. The difference between the adiabatic model for
$\eta/\kappa = 20$ and the full model at $g_{\text{eff}} = 100$ is striking. While the noise reduction at
the center frequency is of comparable magnitude, the linewidth over which the effect is
appreciable is different by a factor of $\sim 50$. The reason for the linewidth narrowing is the
rapid variation of the linear susceptibility around the centre frequency which leads to a
reduction in the group velocity relative to the phase velocity which is equal to $c$ at the
centre frequency since $\chi^{(1)} = 0$ on resonance. If the adiabatic approximation is carried to
the time domain, i.e. within the equations of Section 3.2.1, no linear contribution to the
susceptibility will be found, since analysis in the time domain is restricted to the center
frequency. This was the case in the work of Imamoglu et al. [51], Rebić [64] and Dunstan

\[\text{In a single atom setup, detunings } \Delta \text{ and } \delta \text{ can be used to fine tune the quantum interference in the transitions between the dressed states. For details, see Chapter 9, Part IV.} \]
Figure 3.2: Spectrum of best squeezing for an optimal quadrature. The dashed line is the spectrum of the simple adiabatic model (3.30) with $\eta/\kappa = 20$. Solid lines correspond to the simulations of the full model. The parameters chosen are $\kappa/\gamma = 0.1$, $\Omega_c/\gamma = 15$, $\Delta/\gamma = 30$ and $\delta = 0$, and $g_{\text{eff}}$ was varied as denoted in the Figure. We assumed $\gamma_j = \gamma$ and $g_1 = g_2$. Cooperativity is then defined as $C = \Omega_c^2 g_{\text{eff}}/\kappa$.

et al. [65]. Gheri et al. [63] performed the adiabatic analysis in the frequency domain and found the linear contribution to the susceptibility. This term is contributed by the $\Lambda$–subsystem of the four level atomic system. Detailed explanation of linewidth narrowing due to linear dispersion is given in Chapter 4, where we present a quantitative analysis of cavity linewidth narrowing as well as the related effect of group velocity reduction.

The size of the linear susceptibility is determined by the parameter $g_{\text{eff}}$, and the condition (3.31) follows naturally. The size of anharmonicity (3.32) is also dependent on $g_{\text{eff}}$, so widening the linewidth comes at the expense of noise reduction. This is indeed shown in Fig. 3.2 for the cases $g_{\text{eff}} = 10$ and $g_{\text{eff}} = 1$. The spectral linewidth indeed widens, but the noise reduction is corrupted since the effective anharmonicity becomes too small ($\eta < \kappa$). It is interesting to note that the particular quadrature for which the best squeezing occurs is the phase quadrature (3.24b), since the calculation of the best
squeezing spectrum gives \( \phi = 1.57 \approx \pi/2 \) in equation (3.25) for all values of \( g_{\text{eff}}^5 \). This is not at all surprising. It is a well–known fact that light passing through a dispersive Kerr nonlinearity obtains a well–defined phase shift. This is exactly the message of the quadrature squeezing analysis. The noise is minimal in the phase quadrature, implying the existence of the well–defined phase of the light upon traversing the EIT–Kerr medium.

The linewidth behaviour depicted in Fig. 3.2 does not depend critically on any other parameter except \( g_{\text{eff}} \). We have varied the additional detuning up to the value of \( \delta = 10^4 \gamma \) and found that spectral properties remain the same. The detuning \( \Delta \) does not alter the linewidth properties but may corrupt the noise reduction if it is taken to be too large. In particular, the size of the ac–Stark detuning \( g_2^2/\Delta \) introduced by atomic level \( |4 \rangle \) is required to be large to observe the nonlinear effects.

### 3.5 Intensity Squeezing Spectrum

Having discussed the quadrature squeezing properties, we would like to examine the intensity squeezing properties of this system. Although we emphasize once again that the linearised treatment of intensity fluctuations depends critically on the assumption of a sufficiently large number of photons in the cavity mode, the linewidth narrowing, described in the previous Section, should still be visible.

The intensity representation described in this Section should be considered only informally. Strictly speaking, the transition to the intensity variables is not satisfactory in the mathematical sense, since it does not give a closed set of equations. The reason is the presence of the external driving in this system, and this issue will be further discussed later in this Section.

The intensity–representation variables are defined as

\[
I_d = \Omega^\dagger_d \Omega_d, \quad \Sigma_{12} = \Omega^\dagger_d S_{12}, \quad \Sigma_{23} = \Omega_c S_{23}, \quad \Sigma_{34} = \frac{g_2}{g_1} \Omega^\dagger_d S_{34}, \quad (3.33a)
\]

\[
\Sigma_{13} = \Omega_c \Omega^\dagger_d S_{13}, \quad \Sigma_{24} = \frac{g_2}{g_1} \Omega_c \Omega^\dagger_d S_{24}, \quad \Sigma_{14} = \frac{g_2}{g_1} \Omega_c (\Omega^\dagger_d)^2 S_{14}. \quad (3.33b)
\]

In addition, we define for simplicity,

\[
\gamma_{12} = \gamma + i\Delta_{21}, \quad \gamma_{23} = \gamma + i\Delta_{32}, \quad \gamma_{34} = \frac{\gamma_3}{2} + i\Delta_{43}, \quad (3.34a)
\]

\[
\gamma_{13} = i\Delta_{31}, \quad \gamma_{24} = \Gamma + i\Delta_{42}, \quad \gamma_{14} = \frac{\gamma_3}{2} + i\Delta_{41}, \quad (3.34b)
\]

where \( \gamma = (\gamma_1 + \gamma_2)/2 \) and \( \Gamma = (\gamma_1 + \gamma_2 + \gamma_3)/2 \). The equations for the intensity variables

---

5In addition, \( \Omega_d \) is completely real; this is the necessary additional requirement allowing the conclusion that the squeezing really occurs in the phase quadrature.
are

\[ \dot{I}_d = -2\kappa I_d + C_1 \gamma_1 \kappa (\Sigma_{12} + \Sigma_{21}) + C_1 \gamma_1 \kappa (\Sigma_{34} + \Sigma_{43}) + \beta_d, \]  
(3.35a)

\[ \dot{\Sigma}_{12} = \frac{C_1 \gamma_1 \kappa}{4} - (\kappa + \gamma_{12}) \Sigma_{12} - \Sigma_{13} \]
\[ + \left( I_d + \frac{C_1 \gamma_1 \kappa}{4} \right) D_1 - \frac{C_1 \gamma_1 \kappa}{4} (2D_2 + D_3) + \beta_{12}, \]  
(3.35b)

\[ \dot{\Sigma}_{23} = -\gamma_{23} \Sigma_{23} - \Sigma_{13} + \Sigma_{24} - I_c D_2 + \beta_{23}, \]  
(3.35c)

\[ \dot{\Sigma}_{34} = \frac{C_2 \gamma_2 \kappa}{4} - (\kappa + \gamma_{34}) \Sigma_{34} + \Sigma_{24} \]
\[ + \left( I_d + \frac{3C_2 \gamma_2 \kappa}{4} \right) D_3 + \frac{C_2 \gamma_2 \kappa}{4} (2D_2 + D_1) + \beta_{34}, \]  
(3.35d)

\[ \dot{\Sigma}_{13} = -(\kappa + \gamma_{13}) \Sigma_{13} + I_c \Sigma_{12} + \Sigma_{14} + (I_d + C_1 \gamma_1 \kappa) \Sigma_{23} + \beta_{13}, \]  
(3.35e)

\[ \dot{\Sigma}_{24} = -(\kappa + \gamma_{24}) \Sigma_{24} - I_d \Sigma_{23} - I_c \Sigma_{34} - \Sigma_{14} + (I_d - C_1 \gamma_1 \kappa) \Sigma_{23} + \beta_{24}, \]  
(3.35f)

\[ \dot{\Sigma}_{14} = -(2\kappa + \gamma_{14}) \Sigma_{14} - \Sigma_{13} + (I_d + 2C_1 \gamma_1 \kappa) \Sigma_{24} + \beta_{14}, \]  
(3.35g)

\[ \dot{D}_1 = \frac{-2\gamma_1 + \gamma_2}{4} D_1 + \frac{-2\gamma_1 + \gamma_2}{4} (D_1 - 2D_2 - D_3), \]
\[ -2(\Sigma_{21} + \Sigma_{12}) - (\Sigma_{23} + \Sigma_{32}) + \beta_1, \]  
(3.35h)

\[ \dot{D}_2 = \frac{2\gamma_2 + \gamma_1 + \gamma_3}{4} \]
\[ - \frac{\gamma_1 + 2\gamma_2 + \gamma_3}{4} D_1 + \frac{\gamma_3 - 2\gamma_2 - \gamma_3}{2} D_2 + \frac{3\gamma_3 - 2\gamma_2 - \gamma_3}{4} D_3 \]
\[ + (\Sigma_{21} + \Sigma_{12}) + 2(\Sigma_{23} + \Sigma_{32}) + (\Sigma_{34} + \Sigma_{43}) + \beta_2, \]  
(3.35i)

\[ \dot{D}_3 = \frac{-2\gamma_3 + \gamma_2}{4} \]
\[ - \frac{2\gamma_3 + \gamma_2}{4} D_1 + \frac{\gamma_2 - 2\gamma_3}{2} D_2 - \frac{6\gamma_3 + \gamma_2}{4} D_3 \]
\[ - 2(\Sigma_{43} + \Sigma_{34}) - (\Sigma_{23} + \Sigma_{32}) + \beta_3, \]  
(3.35j)

where \( \beta \)'s denote noise terms, given by equations (3.37), and \( I_c = |\Omega_c|^2 \). It can now be seen clearly that the intensity equations do not form a closed set of equations. An equation for the mean (semiclassical) intensity can be deduced from (3.35a)

\[ \langle \dot{I}_d \rangle = \Omega_p (\langle \Omega_d \rangle + (\Omega_d)^*) - 2\kappa \langle I_d \rangle \]
\[ + C_1 \gamma_1 \kappa (\langle \Sigma_{12} \rangle + (\Sigma_{21})) + C_1 \gamma_1 \kappa (\langle \Sigma_{34} \rangle + (\Sigma_{43})). \]  
(3.36a)

The first term of the right–hand side comes from the driving embedded into the cavity noise definition (3.10a), with \( \Omega_p = g_1 \mathcal{E}_p \), and involves semiclassical mean values of the Rabi operator. The equation for the fluctuation operator \( \delta I_d \) will also contain terms in the Rabi operators

\[ \delta \dot{I}_d = \Omega_p (\delta \Omega_d + \delta \Omega_d^*) - 2\kappa \delta I_d \]
\[ + C_1 \gamma_1 \kappa (\delta \Sigma_{12} + \delta \Sigma_{21}) + C_1 \gamma_1 \kappa (\delta \Sigma_{34} + \delta \Sigma_{43}). \]  
(3.36b)
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Formally, this means that the intensity representation is not well defined, since the intensity operators do not produce a closed set of equations. Informally, however, these equations can still be used in conjunction with the equations derived in Section 3.2.1. Following the formalism outlined in Section 3.3, we solve one huge system of simultaneous equations consisting of atomic and field equations as well as intensity equations and proceed to obtain the intensity squeezing spectrum. This huge system now forms the closed set and can be solved consistently.

The noise terms are given by the following equations

\[ \beta_d = -\sqrt{2\kappa} (\Omega_d^{in\dagger} \Omega_d + \Omega_d\Omega_d^{in}) , \]

\[ \beta_{12} = -\sqrt{2\kappa} \Omega_d^{in\dagger} S_{12} + \sqrt{\gamma_1} \sum_{\mu} \Omega_d^{\dagger} (\sigma_2^{\mu} - \sigma_1^{\mu}) \beta_{1}^{\mu in} \]

\[ -\sqrt{\gamma_2} \sum_{\mu} \beta_{12}^{\mu in} \Omega_d^{\dagger} \sigma_{13}^{\mu} , \]

\[ \beta_{23} = -\sqrt{\gamma_1} \sum_{\mu} \beta_{23}^{\mu in} \Omega_c \sigma_{13}^{\mu} + \sqrt{\gamma_2} \sum_{\mu} \Omega_c \sigma_{24}^{\mu} \beta_{2}^{\mu in} \]

\[ -\sqrt{\gamma_3} \sum_{\mu} \Omega_c (\sigma_{35}^{\mu} - \sigma_{22}^{\mu}) \beta_{3}^{\mu in} , \]

\[ \beta_{34} = -\sqrt{2\kappa} \Omega_d^{in\dagger} S_{34} + \sqrt{\gamma_2} \sum_{\mu} \Omega_d^{\dagger} (\sigma_{44}^{\mu} - \sigma_{33}^{\mu}) \beta_{2}^{\mu in} \]

\[ +\sqrt{\gamma_3} \sum_{\mu} \beta_{3}^{\mu in} \Omega_d^{\dagger} \sigma_{24}^{\mu} , \]

\[ \beta_{13} = -\sqrt{2\kappa} \Omega_d^{in\dagger} \Omega_c S_{13} + \sqrt{\gamma_1} \sum_{\mu} \Omega_d^{\dagger} \Omega_c \sigma_{23}^{\mu} \beta_{1}^{\mu in} \]

\[ +\sqrt{\gamma_2} \sum_{\mu} \beta_{23}^{\mu in} \Omega_c \sigma_{34}^{\mu} + \sqrt{\gamma_3} \sum_{\mu} \Omega_d^{\dagger} \Omega_c \sigma_{34}^{\mu} \beta_{23}^{\mu in} , \]

\[ \beta_{24} = -\sqrt{2\kappa} \Omega_d^{in\dagger} \Omega_c S_{24} - \sqrt{\gamma_1} \sum_{\mu} \beta_{13}^{\mu in} \Omega_d^{\dagger} \Omega_c \sigma_{45}^{\mu} \]

\[ -\sqrt{\gamma_2} \sum_{\mu} \Omega_d^{\dagger} \Omega_c \sigma_{23}^{\mu} \beta_{24}^{\mu in} - \sqrt{\gamma_3} \sum_{\mu} \Omega_d^{\dagger} \Omega_c \sigma_{24}^{\mu} \beta_{34}^{\mu in} , \]

\[ \beta_{14} = -2\sqrt{2\kappa} \Omega_d^{in\dagger} \Omega_c S_{14} + \sqrt{\gamma_1} \sum_{\mu} \Omega_c \Omega_d^{\dagger} \sigma_{34}^{\mu} \beta_{14}^{\mu in} \]

\[ -\sqrt{\gamma_2} \sum_{\mu} \Omega_c \sigma_{23}^{\mu} \Omega_d^{\dagger} \sigma_{13}^{\mu} \beta_{24}^{\mu in} , \]

\[ \beta_{1} = -2\sqrt{\gamma_1} \sum_{\mu} (\sigma_{21} \beta_{1}^{\mu in} + \beta_{1}^{\mu in\dagger} \sigma_{12}^{\mu}) \]

\[ -\sqrt{\gamma_2} \sum_{\mu} (\sigma_{23} \beta_{2}^{\mu in} + \beta_{2}^{\mu in\dagger} \sigma_{32}^{\mu}) , \]
3.5. INTENSITY SQUEEZING SPECTRUM

Note that there is no need to express the noise terms per se in the intensity representation, since their correlations naturally emerge in terms of intensity variables.

We use the same set of parameters as in Section 3.4, with $\delta = 0$, to obtain Fig. 3.3. The adiabatic limit is obtained by using the rules of Itô calculus to combine equation (3.30) with its adjoint to get the equation for $a^\dagger a$. The resulting equation is then linearised. The linewidth narrowing effect is still present although on a somewhat different scale than in the quadrature squeezing spectra. The reduction in linewidth is still $\sim 50$ compared to the adiabatic case, but both spectral lines are wider by a factor of approximately 2. This is not unexpected, since the decay rate of the intensity operator is $2\kappa$, compared to $\kappa$ for the field Rabi operator. Therefore, if the frequency axis in Fig. 3.3 would be scaled as $\omega/2\kappa$, the respective linewidths would be the same as in Fig. 3.2.
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3.6 Conclusion

In summary, we have presented the treatment of noise in the full many-atom system, and compared this with the case of pure Kerr nonlinearity. A quantum noise analysis has been obtained by linearisation of the Itô QSDEs around their mean semiclassical values. It was found that the optimum noise reduction occurs in the phase quadrature. The amount of noise reduction was comparable in both the full atomic system and pure Kerr nonlinearity, but the linewidths of the effect were vastly different. The condition was identified that has to be satisfied if the full atomic system is to be replaced with the pure Kerr nonlinearity as its adiabatic limit. This condition implies the minimisation of the linear contribution to the susceptibility which causes linewidth reduction. In the following Chapter we discuss the origin of linear susceptibility and give the quantitative treatment of linewidth reduction.
Chapter 4

Cavity Linewidth Reduction: Classical Treatment

It was shown in the previous Chapter how the photon blockade effect in a multiatom cavity QED environment appears only within a very narrow band of frequencies centered at the cavity mode frequency. This was attributed to the presence of a linear dispersion $\chi^{(1)}$, unaccounted for in the simple adiabatic model. In this Chapter it is shown how this effect can be fully understood in classical terms. A general description of linewidth reduction due to linear and/or nonlinear susceptibilities of arbitrary order is presented.

4.1 Introduction

The effect of linewidth narrowing for an optical cavity containing a nonabsorbing dispersive medium was predicted independently by Müller et al. [66] and Lukin et al. [67]. The experiment of Müller et al. [66] created the highly dispersive medium by using the phenomenon of coherent population trapping (CPT) [19]. The CPT medium contains only first-order dispersion $\text{Re}[\chi^{(1)}]$, and their measurement compared the linewidth of the empty cavity with the linewidth in the presence of the CPT medium. They found that the cavity linewidth in the presence of dispersion was reduced by a factor of $\sim 50$ compared to the empty cavity value. Basically the same effect was predicted by Lukin et al. [67] for an intracavity EIT medium. The setup of both CPT and EIT atomic media assumes classical fields driving two atomic transitions in a $\Lambda$-configuration. The excited level can decay into each of the ground states with a characteristic rate, and the coherence between the ground states is assumed to decay on a comparatively longer time scale.

Motivated by these results, we want to examine the effect of cavity linewidth reduction for the case of linear susceptibility or arbitrary order nonlinear susceptibility, and in particular explain the results obtained in Chapter 3 for the multiatom EIT–Kerr system.

A full quantum analysis would require a quantum multimode treatment, however we consider a classical analysis for the setup sketched in Fig. 4.1. We assume a classical input field $\mathcal{E}_m$, incident on a cavity mirror with reflectivity $r$ and transmittance $t$, so that $\mathcal{E}_1 = t\mathcal{E}_m + r\mathcal{E}_4$. The intracavity field $\mathcal{E}_1$ then passes through a $\chi$ medium to reach the
Figure 4.1: Envisaged setup for the calculation of the round-trip phase shift. Incident field $\mathcal{E}_{\text{in}}$ enters the cavity of length $L$ through a mirror of reflectivity $r$ and transmittance $t$. The total phase shift due to propagation through the empty cavity is denoted by $\varphi_0$. A medium of length $l$ having a susceptibility $\chi$, situated inside the cavity, contributes the additional phase shift $\Phi$.

second mirror as $\mathcal{E}_2 = \mathcal{E}_1 \exp(i\omega L/c) \exp(i\Phi/2)$. Field $\mathcal{E}_2$ is reflected from the perfect mirror at the right-hand side. The reflected field $\mathcal{E}_3$ again passes through the $\chi$ medium becoming $\mathcal{E}_4 = \mathcal{E}_3 \exp(i\omega L/c) \exp(i\Phi/2)$ at the input–output mirror. The output field is then $\mathcal{E}_\text{out} = t\mathcal{E}_4 + r\mathcal{E}_\text{in}$. Combining these equations we find the relation between the input and output fields to be

$$\frac{\mathcal{E}_\text{out}}{\mathcal{E}_\text{in}} = r + \frac{t^2 e^{i\varphi}}{1 - re^{i\varphi}}, \quad (4.1)$$

where $\varphi = \varphi_0 + \Phi$ denotes the total phase shift. Here, $\varphi_0 = 2\omega L/c$ is the phase shift due to free propagation, while $\Phi$ denotes a phase shift due to the $\chi$ medium, of length $l$. The ratio of intracavity to incident intensity is then given by

$$\frac{|\mathcal{E}_1|^2}{|\mathcal{E}_\text{in}|^2} = \frac{|t|^2}{1 + |r|^2 - 2\text{Re}[r \exp(i\varphi)]}. \quad (4.2)$$

This relation is important, since it connects the intracavity field used in subsequent analysis with the input field.
4.2 Solving Maxwell’s Equation

Assuming classical input and intracavity fields, the phase shift $\varphi$ can be obtained by solving Maxwell’s equation

$$\nabla^2 \mathcal{E}' - \frac{1}{c^2} \frac{\partial^2 \mathcal{E}'}{\partial t^2} = 4\pi \frac{N}{c^2} \frac{\partial^2 \mathcal{P}}{\partial t^2}.$$  \hspace{1cm} (4.3)

Here, $\mathcal{E}'$ denotes the field inside the $\chi$ medium, $N$ is the number of atoms per unit volume in the medium and $\mathcal{P}$ is the single atom polarisation expanded according to Bloembergen [68] as

$$\mathcal{P} = \mathcal{P}^{(1)} + \mathcal{P}^{(2)} + \mathcal{P}^{(3)} + \cdots = \chi^{(1)} \mathcal{E}_1 + \chi^{(2)} |\mathcal{E}_1|^2 + \chi^{(3)} |\mathcal{E}_1|^2 \mathcal{E}_1 + \cdots.$$  \hspace{1cm} (4.4)

Note that the expansion is written in terms of the intracavity field $\mathcal{E}_1$, and not the field inside the medium $\mathcal{E}'$. This simplification is called the *unperturbed field approximation* [69]. It is valid when the excited polarisation waves in the medium are weak compared to the external electromagnetic waves incident on the medium that create a linear or nonlinear polarisation in the medium. Defining $\delta \mathcal{E} = \mathcal{E} - \mathcal{E}_1$, the condition for the validity of this approximation is simply given by $\delta \mathcal{E} \ll \mathcal{E}_1$. When the susceptibility $\chi$ of the medium is nonlinear, analytical solutions of Maxwell’s equation are possible only in the simplest few cases. The unperturbed field approximation simplifies the equations considerably, since it is then the (unperturbed) incident electromagnetic field that yields a nonlinear polarisation. The corresponding Maxwell’s equation is a linear inhomogeneous partial differential equation, and the inhomogeneous term is the nonlinear polarization of the medium, taken, in general, as a given function of coordinates and time.

To simplify the notation, we define the susceptibility function $\chi$ as $\mathcal{P} = \chi \mathcal{E}_1$, where

$$\chi = \chi^{(1)} + \chi^{(2)} |\mathcal{E}_1|^2 + \chi^{(3)} |\mathcal{E}_1|^2 \mathcal{E}_1 + \cdots.$$  \hspace{1cm} (4.5)

Note that the requirement $d_i d_j / r^3 \ll d_i \mathcal{E}_1$ also encapsulates the essence of the independent atoms approximation.

It is beneficial to rewrite Maxwell’s equation (4.3) in terms of the variation of the field strength in the medium, $\delta \mathcal{E}$, as

$$\nabla^2 \delta \mathcal{E} + k^2 \delta \mathcal{E} = -4\pi N \hbar^2 \chi \mathcal{E}_1$$

$$= -4\pi N \hbar^2 \chi \mathcal{E}_\omega \exp (i \omega t - i \mathbf{k} \cdot \mathbf{r}),$$  \hspace{1cm} (4.7)

\footnote{This condition is, of course, derived for the case of dilute atomic gas weakly coupled to the cavity, i.e., having in mind the system considered in Chapter 3.}
where \( \mathbf{k} \) is a wave vector of the incident electric field \((k = |\mathbf{k}| = \omega/c)\), and \( \mathcal{E}_\omega \) is its Fourier amplitude. We now assume that the electromagnetic wave propagates in the \( z \) direction, and put a medium–vacuum interface at the \( z = 0 \) plane. The solution to the equation (4.7) that satisfies the boundary condition \( \delta \mathcal{E}(z = 0) = 0 \) (the absence of a polarisation wave at the interface) and ignores the reflected wave in comparison with \( \delta \mathcal{E} \) (and even more so compared with \( \mathcal{E}_1 \)), can be found straightforwardly as

\[
\delta \mathcal{E} = -2\pi i N \frac{\omega z}{c} \chi \mathcal{E}_\omega \exp (i \omega t - ikz). \tag{4.8}
\]

Note that the strength of the polarisation field grows linearly with \( z \) when light passes through the medium. The total field in the medium is then

\[
\mathcal{E}' = \left[ 1 - 2\pi i N \chi \mathbf{k} \cdot \mathbf{r} \right] \mathcal{E}_\omega \exp (i \omega t - i \mathbf{k} \cdot \mathbf{r}). \tag{4.9}
\]

Within the approximation (4.6), this expression can now be rewritten as

\[
\mathcal{E}' = \mathcal{E}_\omega \exp (i \omega t - i \mathbf{k}' \cdot \mathbf{r}), \tag{4.10}
\]

where the following notation has been introduced

\[
\mathbf{k}' = (1 + 2\pi l N \chi) \mathbf{k}. \tag{4.11}
\]

The first term in the brackets describes the free propagation, while the second term gives the correction due to the medium. This second term amounts to the desired phase shift due to the medium

\[
\frac{\Phi}{2} = \frac{2\pi \omega}{c} l N \chi. \tag{4.12}
\]

The total round-trip phase shift is then given by the following expression

\[
\varphi = \varphi_0 \left( 1 + \frac{2\pi l}{L} N \chi \right), \tag{4.13}
\]

where the first term \( \varphi_0 \) denotes the free propagation phase-shift, and the second term in the brackets describes the correction due to the presence of the dispersive medium. Note that this phase shift generally depends on the field \( \mathcal{E}_1 \) except in the simplest case of linear susceptibility \( \chi = \chi^{(1)} \).

### 4.3 Group Velocity and Cavity Linewidth

#### 4.3.1 Group Velocity

The phase correction (4.13) has some important consequences. We first evaluate the change in group velocity due to the presence of a medium. The group velocity is defined
4.3. GROUP VELOCITY AND CAVITY LINEWIDTH

as \( v_g = (dk'/d\omega)^{-1} \), and the change in group velocity in a cavity during a single round-trip is easily evaluated as

\[
v_g = \frac{2L}{d\phi/d\omega} = \frac{c}{1 + \frac{2\pi N}{L} \chi \frac{d\chi}{d\omega}}. \tag{4.14}
\]

Remembering that Maxwell’s equation was solved under the assumption \( N \ll 1 \), the above expression can be written

\[
v_g \simeq \frac{c}{1 + \frac{2\pi N L}{e \omega} \frac{d\chi}{d\omega}}. \tag{4.15}
\]

Therefore, provided that the effective susceptibility is sufficiently small to enable the application of the unperturbed field approximation, the correction to the group velocity comes from the gradient of the susceptibility close to the resonant frequency. Steep dispersion then implies a large reduction in group velocity. Note that we have rederived a well-known result for the group velocity in a dispersive medium [70]. For normal dispersion, \( \chi \) as well as \( d\chi/d\omega \) is positive, thus leading to a reduction in the speed of a wave packet.

In the cases of anomalous dispersion, \( d\chi/d\omega \) can be negative, leading to the apparently paradoxical result of velocity greater than that of light. There is no conflict here though, since the group velocity of the wave packet undergoing the anomalous dispersion is not given by \( v_g = (dk'/d\omega)^{-1} \). The behaviour of light in that case is much more involved and the concept of group velocity is simply not useful any more.

We further note that the group velocity reduction can also be viewed as the increase in the total round-trip time \( T = 2L/v_g \), given by

\[
T \approx T_0 \left(1 + \frac{2\pi N L}{e \omega} \frac{d\chi}{d\omega}\right), \tag{4.16}
\]

\( T_0 = 2L/c \) being the free-propagation round-trip time.

4.3.2 Cavity Linewidth

Having obtained a result for the reduction in group velocity, it is now possible to explain the cavity linewidth reduction in a straightforward manner. Intuitively, if the dispersion becomes steep close to the resonance frequency, the slightly off-resonant components of the input field will encounter a large index of refraction. Alternatively, it can be stated that a large reduction in group velocity (or increase in the round-trip time) causes light to remain in the cavity for a longer time before it emerges out through the leaky mirror, thus rescaling the cavity damping constant.

To quantify the above statement, note that the cavity damping constant is defined by\(^2\) \( \kappa = |v|^2/T \), given that the round-trip time \( T \) is very short, so that \( \kappa T \ll 1 \). Straight-
forward inclusion of equation (4.16) leads to
\[ \kappa \approx \frac{\kappa_0}{1 + \frac{2\pi N}{\lambda} \frac{dx}{d\lambda}}, \]  
(4.17)

with \( \kappa_0 = |t|^2 / T_0 \). Therefore the cavity damping (and hence the cavity linewidth) is reduced by the same factor as the group velocity.

4.4 Dispersion in Atomic Gases

We conclude this Chapter with a few applications of the above results. So far, we have made no assumptions about the precise nature of the \( \chi \) medium inside the cavity. In this Section, we use the results for the susceptibilities of gaseous media containing two level atoms and four level atoms, respectively, that we derived in Appendix B.

4.4.1 Linear Susceptibilities

The linear susceptibility of a two level atom is given by
\[ \chi_{2-\text{lev}}^{(1)}(\theta) = \frac{g^2}{\epsilon_0 \hbar} \frac{\theta + i\gamma/2}{(\gamma/2)^2 + \theta^2}, \]  
(4.18)

with \( \theta = \omega - \omega_0 \), where \( \omega_0 \) denotes the atomic resonant frequency, \( \gamma \) is the decay rate of the excited atomic state and \( g \) is the atom–field dipole coupling strength. The real part of this expression describes dispersion, while the imaginary part describes absorption. We are interested in the effects of dispersion, and therefore will neglect the absorptive part in further analysis. Also, for simplicity, we will assume that the atomic medium fills the cavity completely, so \( l = L \) in the formulae of the previous Section. The group velocity and cavity linewidth are then
\[ \frac{v_g}{c} = \frac{\kappa}{\kappa_0} = \frac{1}{1 + \frac{8N\omega_0 g^2}{\epsilon_0 \hbar} \frac{2\gamma}{\gamma^2}}. \]  
(4.19)

Note that these ratios depend critically on the quantity \( g/\gamma \), i.e. the ratio of coupling strength to spontaneous emission rate. In practice, it is very difficult to increase this ratio, although some spectacular results have been achieved in recent years (see, for example, Hood et al. [71, 72]). In general increasing the ratio \( g/\gamma \) requires an extremely small cavity mode volume, and its value is fixed for a given cavity. In other words, construction of the particular cavity fixes the value of\( g/\gamma \).

We will now show that the setup of the EIT–Kerr system opens up the possibility of flexible control over the ratios \( v_g/c \) and \( \kappa/\kappa_0 \). So far, our only concern was with the nonlinear contribution to the susceptibility of the four level atom and the resulting photon blockade effect. However, the EIT contribution, exhibited by the \( \Lambda \) subsystem, plays an important part in this scheme in more than one way. As well as minimising the
4.4. DISPERSION IN ATOMIC GASES

spontaneous emission noise, the $\Lambda$ system in EIT configuration contributes to the linear dispersion. The linear susceptibility for this system has already been calculated by Harris et al. [73], Grobe et al. [74], Xiao et al. [40] and Gea-Banacloche et al. [38], among others. In the context of the four level EIT–Kerr scheme, Gheri et al. [63] have performed a careful adiabatic elimination of the atomic variables, assuming $g_{12}^2 N / \Omega_c^2 \ll 1$. They have shown that the EIT subsystem still contributes a linear part to the total susceptibility, and they argued that this contribution is responsible for narrowing the linewidth of the photon blockade effect in the many–atom system we described in the Chapter 3. We will now show how this effect occurs. The first order susceptibility per atom is given by the expression

$$
\chi_{1-\text{lev}}^{(1)}(\omega) = \frac{1}{\epsilon_0 \hbar} \frac{g_{12}^2}{\Omega_c^2} \frac{\Omega_c^2 \tilde{\theta}}{\Omega_c^2 - \tilde{\theta}^2 - i(\gamma_1 + \gamma_2)\tilde{\theta}/2},
$$

where now $\tilde{\theta} = \omega - \omega_{12}$, $\omega_{12}$ being the frequency of the atomic transition $|1\rangle \rightarrow |2\rangle$. The argument of Gheri et al. [55, 63] becomes obvious. The linear contribution to the susceptibility becomes negligible for $g_{12}^2 N / \Omega_c^2 \ll 1$. It is in this limit that the adiabatic model of Imamoglu et al. [51] becomes valid and the third–order term becomes the leading contribution to the total susceptibility. This alone does not ensure photon blockade, since in addition the nonlinearity has to be large enough to produce a strong anharmonic term in the effective Hamiltonian [63], i.e. $\eta / \kappa > 1$, $\eta$ being the anharmonicity given by equation (B.12) \(^3\). The group velocity/cavity linewidth reduction is then given by

$$
\frac{v_g}{c} = \frac{\kappa}{\kappa_0} = \frac{1}{1 + \frac{2g_{12}^2}{\epsilon_0 \hbar} g_{12}^4 N / \Omega_c^2} \approx \frac{\epsilon_0 \hbar}{2 \omega_{12}} \frac{\Omega_c^2}{g_{12}^4 N}.
$$

For the set of parameters chosen by Imamoglu et al. [51] (also the parameters chosen in Chapter 3), we obtain $(\kappa_0 / \kappa)_{1-\text{lev}} \approx 50$, in agreement with the full model simulations. As a comparison, for the same set of parameters, equation (4.19) gives $(\kappa_0 / \kappa)_{2-\text{lev}} \approx 1.14$, i.e. negligible narrowing in the case of two level atoms.

Note that, unlike the two level case, this scheme allows tuning of the ratios $\frac{\omega}{c}$ and $\frac{\omega_{12}}{c_0}$ to just about arbitrary values, by simply tuning the coupling field Rabi frequency $\Omega_c$. Experimentally this is viable, since $\Omega_c$ is directly dependent on the coupling laser power. Indeed, a group velocity reduction to 17 m/s has been experimentally achieved by Han et al. [75] in a cloud of sodium atoms - a spectacular reduction of the speed of light by seven orders of magnitude. The cloud of about $10^{10}$ atoms was cooled to a temperature of 50 nK, forming a Bose–Einstein condensate (critical transition temperature $T_c = 435$ nK) to increase the density. In the experiment of Kash et al. [76] a reduction of group velocity to 90 m/s was observed in a hot gas of rubidium atoms at 360 K.

\(^3\)See the discussion on nonlinearity and anharmonicity in Appendix B
4.4.2 Nonlinear Susceptibilities

We will end this Chapter by briefly estimating the contribution of nonlinearities to the group velocity reduction and linewidth narrowing. Including only the third order term \( \chi = \chi^{(3)} |\mathcal{E}_1|^2 \) in the equations (4.15) and (4.17) brings into play the intracavity light intensity. We can now use equation (4.2) to express the intracavity intensity in terms of the driving (input) intensity. The total phase shift \( \varphi \) (4.13), where \( \varphi_0 = 2\omega L/c \), can be rewritten in terms of the cavity linewidth as

\[
\varphi = \frac{\varphi_0}{\kappa/\kappa_0}.
\]

Transmission and reflection coefficients are given by \( |t|^2 = 2L\kappa_0/c \) and \( |r| = \sqrt{1 - |t|^2} \). The resulting analysis is not as simple as in the linear case. Rather, we obtain a transcendental equation that has to be solved for \( \kappa \). The resulting transcendental equation for \( \kappa \) is

\[
\frac{\kappa_0 - \kappa}{2\pi N\omega (d\chi^{(3)}/d\omega) |\mathcal{E}_m|^2\kappa_0} = \frac{\kappa L}{c - \kappa_0 L - \sqrt{c(c - 2L\kappa_0)} \cos (\varphi_0\kappa_0/\kappa)}.
\]

This equation has been solved numerically to give an estimate for the contribution of third order nonlinearity to the linewidth narrowing.

The nonlinear susceptibilities per atom, calculated in Appendix B are

\[
\text{Re} \left[ \chi^{(3)}_{2\text{-lev}} \right] = \frac{1}{\epsilon_0\hbar^3} \frac{2g^2\theta}{(\gamma/2)^2 + \theta^2} \frac{g^2}{(\gamma/2)^2 + \theta^2 + g^2}
\]

\[
\text{Re} \left[ \chi^{(3)}_{4\text{-lev}} \right] = \frac{1}{2\epsilon_0\hbar^3} \left( \frac{g_1}{\Omega_c} \right)^2 \frac{g_5^2\Delta}{(\gamma_3/2)^2 + \Delta^2}.
\]

Note that the expression for the \( \text{Re} \left[ \chi^{(3)}_{4\text{-lev}} \right] \) is taken in the nonperturbative limit. As commented in Appendix B, this is the limit where the size of the nonlinearity in both models is largest and comparable in each. Therefore, it is in this limit where the largest corrections to the linear case can be expected. The parameters of Imamoglu \textit{et al.} [51] give the solution for the equation (4.23) relative to the empty cavity value to be

\[
\left. \left( \frac{\kappa_0}{\kappa} - 1 \right) \right|_{4\text{-lev}} \sim 10^{-3}.
\]

Clearly, the linewidth narrowing (and therefore group velocity reduction) due to third order nonlinearity is less than the linear dispersion by four orders of magnitude and therefore negligible. For the two level atom, the nonlinear correction is of the same order as for the four level atom. In this case, the linear contribution is much smaller and the relative contribution of the third order correction becomes an order of magnitude larger. Overall, however, we conclude that the linewidth narrowing and group velocity reduction in the two level medium is negligible.
4.5 Conclusion

In this Chapter we have presented a classical derivation for the effect of group velocity reduction and linewidth narrowing due to the presence of a dielectric material in an optical cavity. The derived theory is general, and can be applied to linear media as well as media of arbitrary orders of nonlinearity. We have compared two special cases of dielectric media, namely two level atoms and four level atoms in an EIT–Kerr configuration. The results obtained for the EIT–Kerr system are in agreement with the previous calculations (see Chapter 3 and Gheri et al. [63, 55]) and experimental results [75], and have been explained by the linear contribution to the susceptibility generated by the EIT subsystem. The effects are dramatic, ranging from reduction by a factor of 50 in linewidth, obtained in Chapter 3 and by Gheri et al. [63, 55], to the group velocity reduction by seven orders of magnitude, obtained by Hau et al. [75]. In contrast, the effects in the two level medium are found to be effectively negligible.
Part III

Large Kerr Nonlinearity with a Single Atom
Chapter 5

Single–Atom Cavity QED

In Part II, we analysed the noise properties of the atomic medium and found this setup to be unsatisfactory for the realisation of the photon blockade effect. As an alternative to the atomic medium, we may also consider using a single four level atom in a high finesse microcavity. This system will be focused on for the remainder of this thesis. In this Chapter, we review the topic of single-atom cavity QED.

5.1 Introduction

There has been huge interest lately in the physics of quantum computing and possible physical realisations of the required elements. Single photon control by utilizing large optical nonlinearities has emerged as a possible candidate for conditional quantum logic, i.e. optical quantum gates.

From this perspective, the photon blockade effect plays a very important role, since it offers a possibility of realising quantum logic elements. The analysis of Imamoglu et al. [51] assumed that the atoms can be adiabatically eliminated in the many-atom EIT–Kerr system. The necessary condition for the adiabatic approximation has been identified in equation (3.31). The other way of stating the adiabatic condition is to require that the response time of the medium is much shorter than the typical timescale of the cavity. Direct comparison of the cavity and atomic linewidths would suggest this to be the case. We have shown in Part II that this is not the case. Indeed the effective linewidth of the cavity containing the EIT–Kerr atomic medium is significantly smaller than that of the cavity containing a pure Kerr nonlinearity, i.e. the effective pure $\chi^{(3)}$ Hamiltonian can only be valid close to the central frequency. The total susceptibility of the medium has a linear, as well as a nonlinear, component. While the linear component vanishes at the central frequency, it can be very large at nearby frequencies. As a result, frequency components of the cavity mode other than the resonant component will experience a very strong dispersion.

To overcome this problem, we propose a scheme in which a single four-level atom is placed in a microcavity. This type of cavity offers the largest single photon dipole coupling strength achieved to date, exceeding, at the same time, the decoherence rates of...
the system due to atomic spontaneous emission and due to cavity loss. We show that this, combined with the effects of EIT, enables us to obtain a high $\chi^{(3)}$ nonlinearity which may act as a photon blockade. That is, when the first photon excites the cavity mode, the large nonlinearity switches the cavity off resonance with the incident light, thus preventing a second photon from further exciting the cavity mode.

5.2 Cavity Geometry and Parameters

A typical cavity geometry is shown in Figure 5.1. We wish to consider a single cavity field mode that interacts with a single atom in the cavity. Therefore, we would like to reduce the number of cavity modes within a given narrow frequency interval to just one. The number of cavity modes depends on the total cavity volume - a smaller cavity will support fewer modes. As an example, for a typical optical frequency of $3 \times 10^{14} \text{ s}^{-1}$, a completely enclosed optical resonator will possess $\sim 10^9$ modes within the frequency interval $\sim 10^{10} \text{ s}^{-1}$ per unit volume ($V_{\text{cav}} = 1 \text{ cm}^3$). In a closed cavity geometry, all of these modes would have approximately the same $Q$ factor. To avoid this situation and to
5.2. CAVITY GEOMETRY AND PARAMETERS

Figure 5.2: Spatial structure of a TEM$_{00}$ cavity mode for a cavity whose axis is oriented along the $z$-axis. Dependence of $\psi(\mathbf{r})$ on $z$ and $y$ is shown; the $x$ dependence is the same as the $y$ dependence.

discriminate against all but one frequency mode, an open cavity geometry has to be used. Thus, as shown in Fig. 5.1, a cavity is formed by aligning two mirrors along one axis. Such a cavity will discriminate heavily against all those modes whose energy propagates along directions other than the cavity axis, normal to the mirrors [77].

The cavity mode itself is a Gaussian standing wave TEM$_{00}$ mode, described by the mode function

$$\psi(\mathbf{r}) = \cos\left(\frac{2\pi z}{\lambda_{\text{cav}}}\right)e^{-(x^2+y^2)/w_0^2}.$$  \hspace{1cm} (5.1)

Here, we choose the coordinate system so that the cavity axis is along the $z$ direction, $\lambda_{\text{cav}}$ is the cavity mode wavelength and $w_0$ is the cavity waist. This spatial dependence is illustrated in Figure 5.2. This mode couples to the atom with a dipole coupling strength given by $g = g_0 \psi(\mathbf{r})$, with $g_0$ being its maximum magnitude. This quantity critically depends on the mode volume. Namely, if the photon is confined to a small volume $V_{\text{cav}}$, the electric field per photon, given by $E = \sqrt{\hbar\omega/(2\varepsilon_0 V_{\text{cav}})}$, will be large. In such an environment it is possible that a single photon saturates the atomic response. The energy of interaction between a photon and an atom is given by $\hbar g_0 = \mu E$, where $\mu$ is the atomic dipole moment. This gives for the maximum coupling strength

$$g_0 = \left(\frac{\omega}{2\hbar\varepsilon_0 V_{\text{cav}}}\right)^{1/2}\mu,$$  \hspace{1cm} (5.2)

with $\omega$ being the frequency of the atomic transition.
We now consider atomic and cavity losses in brief. Atomic losses are mainly due to spontaneous emission losses\(^1\), which occurs at the rate \(\gamma\). The atomic dipole moment can be expressed as \(\mu^2 = 3\pi\epsilon_0\hbar c^3\gamma/\omega^3\), so the expression (5.2) can be rewritten as

\[
g_0 = \left( \frac{3c\lambda^2\gamma}{8\pi V_{\text{cav}}} \right)^{1/2} \mu, \tag{5.3}
\]

where \(\lambda = 2\pi c/\omega\) is the wavelength of the atomic transition. So, not surprisingly, the atomic decay into the cavity mode enhances the effective coupling constant. The volume of the TEM\(_{00}\) mode of a Gaussian standing wave cavity can be calculated in terms of the cavity geometry as \(V_{\text{cav}} = w_0^2L\pi/4\) [78], where \(L\) denotes cavity length and \(w_0\) is the Gaussian waist given by

\[
w_0 = \frac{L\lambda}{\pi} \sqrt{q_1q_2(1-q_1q_2)} \left/ \frac{q_1 + q_2 - 2q_1q_2}{q_1 + q_2} \right., \tag{5.4}
\]

where \(q_j = 1 - L/R_j\), \(R_j\) being the radius of curvature of mirror \(j\). Therefore, the maximum coupling strength is determined by the known (geometric) quantities and the atomic quantities \(\lambda\) and \(\gamma\).

There are three contributions to the cavity losses: transmission, absorption and scattering, all related to cavity mirrors. Here, transmission is considered to be a good loss, since all the light in the cavity eventually has to leak out to be detected. Scattering and absorption are similar to spontaneous emission losses out the sides of the cavity and are considered bad. If we denote by \(\delta_0\) the total roundtrip losses due to the combination of intracavity medium, scattering and absorption at both mirrors, and define as \(\delta_{1,2}\) the power transmission coefficients of the respective mirrors, then the total cavity losses are \(\delta_{\text{cav}} = \delta_0 + \delta_1 + \delta_2\). The finesse of the cavity can be written as \(\mathcal{F} = 2\pi/\delta_{\text{cav}}\).

### 5.3 Loading the Atom

Having defined the cavity and atomic parameters, we will now describe briefly how the atom is loaded. In order to establish an efficient coupling of the optical beam to the dipole radiation pattern of the atom, we consider the atom to be located within the cavity mode volume. This means that it is relatively easy to probe the atom with beams of light along the \(z\) direction while minimising the loss of photons into the inaccessible dimensions \(x\) and \(y\). Almost exclusively, the experimental work in single-atom cavity QED, and most of the theoretical work so far, has involved two level atoms. We will illustrate basic concepts with the two level atomic systems without loss of generality, since the same conclusions remain qualitatively valid in a multilevel system as well.

We define the single-atom cooperativity as \(C = g^2/(\kappa\gamma)\). We also define two common regimes of single-atom CQED, namely the bad cavity regime defined as \(\kappa \gg g^2/\kappa \gg \gamma\) [79],

\(^1\)We will not consider collisional effects between the atoms or between the atom and a cavity mirror, since we are primarily interested in a dynamics of a single atom at a fixed position.
and the strong coupling regime, defined as \( g \gg (\kappa, \gamma) \). The bad cavity regime is of interest since the atom and cavity essentially retain their individual characters, i.e. the eigenstates of the coupled system split into an atom–like part and a photon–like part, while allowing for the efficient transfer of the electromagnetic fields from input to output channels (with rate \( \kappa \)). In this regime, \( C > 1 \), which means that the atom’s coherent coupling to the cavity mode dominates incoherent emission into free space (with rate \( \gamma \)). This regime effectively creates a one dimensional atom. The intracavity saturation photon number, \( n_s = 4\gamma^2/(3g^2) \), is obviously small. Indeed, Turchette et al. [80] measured the saturation of a one dimensional Cs atom for only 0.02 intracavity photons in a cavity, with parameters \((g_0, \gamma, \kappa)/2\pi = (20, 2.5, 75) \text{ MHz} \). The fraction of total spontaneous emission that is emitted into the cavity mode \( \beta = C/(1 + C) \) was \( \approx 0.7 \).

In the strong coupling regime, the atom and the cavity lose their individual characteristics to form an atom–cavity molecule [71, 72]. This will be the regime of interest for the remainder of this thesis, and we will discuss its properties later.

The cavity can be loaded with atoms that were previously cooled in a magneto–optical trap (MOT). This particular method of loading the cavity has been employed by the Caltech group in a series of spectacular experiments [71, 72, 81, 82]. The atoms in the MOT are cooled by a standard six–beam optical molasses configuration, and the MOT coils are arranged to be parallel with the cavity mirrors. In this way, cold atoms released from the trap fall from a perpendicular direction into the cavity. Atoms are distributed according to Poisson statistics in a beam released from the trap. The presence of the atom in the cavity is detected by observing a dip in the cavity transmission spectrum. Mabuchi et al. [81] and Hood et al. [71] have reported real–time detection and manipulation of individual atoms falling through the cavity. It was shown that an individual atom can be trapped by the field of a single photon and its motion followed in real time [72]. We will review the trapping mechanism in this and other setups in the next Section. A thorough and detailed observation of the dynamics of a single atom is given by Mabuchi et al. [82].

In the strong coupling limit, where an atom and a cavity form a coupled system, it is of paramount importance to know the number of atoms in the cavity mode volume. Given the random arrival times of the atoms, and the Gaussian profile of the cavity mode seen by falling atoms (see Fig. 5.2 and Eq. (5.1)), an important question is how many atoms effectively contribute to the intracavity dynamics. Experimentalists in cavity QED have a good control over atomic density \( \sigma \), but not directly over the number of atoms. Carmichael and Sanders [83] have analysed the case of two level atoms in some detail and defined the one atom and many atom regimes of cavity QED. It is beneficial to review their findings to establish clearly what is meant by the single–atom regime.

Let’s assume that the atomic beam extends from \( z = -M\lambda/4 \) to \( z = M\lambda/4 \) along the cavity axis \( z \) (\( M \) being an odd integer), and for a distance \( \gg w_0 \) perpendicular to the cavity axis. Taking into account the Poissonian distribution of the atomic arrival times, the distribution of the collective dipole coupling strength \( G \) is given by [83]

\[
P(G) = \frac{G}{\pi} \int_{-\infty}^{\infty} ds \ e^{-isG^2} Q(s) , \tag{5.5}\]
where

\[ Q(s) = \exp \left\{ 2N_{\text{eff}} \frac{4}{\pi} \int_0^1 d\xi \frac{e^{-i\xi^2} - 1}{\xi \cos^{-1} \xi} \right\}. \tag{5.6} \]

Here, the parameter \( N_{\text{eff}} = \sigma (M\lambda \pi w_0^2/8) \) introduces the beam density \( \sigma \) in the distribution. The conditions \( N_{\text{eff}} \gg 1 \) and \( N_{\text{eff}} \ll 1 \) identify the many–atom and one–atom regimes of cavity QED, respectively. An analytical approximation to the distribution (5.5) is found by an expansion in a power series, which is truncated to an appropriate number of terms depending on the number of interacting atoms. Comparison with numerical simulations show that a strict definition of the number of interacting atoms is impossible. It follows that it is impossible to define the intermediate regime between single–atom and many–atom cases. This is due to the probabilistic character of the atomic beam and the fact that the individual atoms interact with the cavity mode even when they are far into the exponential tail (along, say, the \( y \)-direction in Fig. 5.2). In other words, it is impossible to tell exactly where the cavity mode starts and where it ends, implying that it is impossible to tell exactly how many atoms from the beam interact with the cavity mode in any given time interval. Carmichael and Sanders have found that the spatial volume within which the atom interaction with the cavity mode cannot be neglected is determined by \( 3w_0 \) (\( w_0 \) being the cavity waste), and not \( w_0 \) as the conventional wisdom implies. Experiments considering single–atom CQED must therefore strictly satisfy the condition \( N_{\text{eff}} \ll 1 \). An analogous analysis of the atomic beam properties has been performed by Carmichael et al. [84] in the context of photon correlation spectroscopy - a spectroscopic technique for observing the excited state resonances in the Jaynes–Cummings model.

An alternative setup has been proposed by Münsterman et al. [85], which involves an atomic fountain as a source of atoms. Atoms of \(^{85}\text{Rb} \) are launched from a MOT with a moving optical molasses and detected 25 cm above the cavity. Varying the launch velocity of the cloud, the atomic velocity at the entrance to the cavity can be tuned from a few meters per second to almost zero. In addition, adjusting the loading time of the MOT gives control over the atom flux in a fountain burst. As in the Caltech experiment, the passage of a single atom through the cavity can be detected by a sudden drop in transmission from the cavity.

### 5.4 Trapping the Atom

Once the atom has been loaded into the cavity, its interaction with the cavity mode can be studied in a way that goes beyond just measuring a dip in the transmission spectrum. In order to keep the atom in the cavity for long enough to study the effects of its interaction, some trapping mechanism has to be employed. In this Section, we will review two successful trapping mechanisms.

In the experiment of Hood et al. [71], a cavity of finesse \( \mathcal{F} = 180 \, 000 \), waist \( w_0 = 15 \mu\text{m} \) and length 10.1 \( \mu\text{m} \) was used and had the following parameters: \( (g_0, \kappa, \gamma, 1/T)/2\pi = (120, 40, 2.6, 0.002) \text{ MHz} \), where \( T \) denotes the atom–cavity interaction time \( (T \gg \kappa^{-1}) \).
This is the strong coupling regime of cavity QED, and one atom is expected to change significantly the optical properties of a weakly driven cavity. Indeed, the usual Fabry–Perot peak of an empty cavity splits into two Rabi peaks detuned by \( \pm g \) from the resonance. We have described in the previous Section how the value of \( g \) depends on the atom’s position in the cavity. It is therefore possible to observe in real time the motion of the atom as it goes through the cavity by looking at the transmission of a weak probe beam. When this probe beam is detuned from the cavity resonance, a significant asymmetry between the red and blue detunings is observed. In the red detuned case, more atoms achieve a large value of \( g \) than in the blue detuned case. This was attributed to a light induced force at the single photon level which channels the atoms into the antinodes of the cavity mode, thus increasing the value of \( g \) close to its maximum value \( g_0 \). A blue detuned photon causes channelling into the nodes. The spatial dependence of the energy, \( \hbar g \), represents a pseudopotential well that can be selectively populated by appropriate choices of the strength and frequency of an external driving field, therefore enabling an atom with kinetic energy \( E_k \ll \hbar g_0 \) to be trapped. Hood et al. called this effect the \textit{photon–covalent bonding of atom and cavity}, since the atom sees an attractive potential due to the presence of a red detuned photon, just as one atom can be attracted to the other through sharing of an electron pair. This interpretation has motivated the introduction of the term \textit{atom–cavity molecule} describing the atom strongly coupled to the cavity mode.

So, the trapping procedure works in two steps:

1. A probe beam close to the atom–cavity resonance detects atoms entering the edge\(^2\) of the cavity mode.

2. A driving field of strength \( \sim 1 \) photon, detuned by \( -g_0 \) (lower Rabi resonance) is then switched on. The system is driven to the lower bound state and the atom is effectively trapped at the antinode.

This scheme opens the possibility of reconstruction of the individual atom trajectories from the record of cavity transmission. In the later experiment of Hood et al. [72], an \textit{atom–cavity microscope} was realised that traced a single atom bound in orbit by the mechanical forces of a single photon. The optical cavity in that experiment was formed by two mirrors of diameter 1 mm and radius of curvature 10 cm. The finesse of the cavity was \( \mathcal{F} = 480\,000 \). For the measured cavity length of 10.9 \( \mu \)m and waist of 14 \( \mu \)m, the following parameters were established: \( (g_0, \kappa, \gamma)/2\pi = (110, 14.2, 2.6) \) MHz. A 2 \( \mu \)m spatial resolution in a 10 \( \mu \)s time interval of the microscope was reported, which is near the standard quantum limit for sensing the motion of a Cs atom. Further technical details about the cavity are given in Hood et al. [86].

Similar observations of atomic motion in a cavity have been reported by Pinkse \textit{et al.} [87], though it is unclear whether the effect reported there is the same as the single–

\(^2\)As we saw in the previous Section, the “edge” is not well defined. In an experiment, an edge is defined as the point at which the transmission starts to decrease.
A detailed comparison of the two experiments, supplemented by theoretical simulations, has been performed by Doherty et al. [88]. A different trapping mechanism has been reported by Ye et al. [89], who used a far-off-resonance optical trap (FORT) [90] to provide a confining potential to trap the atom within the cavity mode. In this experiment, $10^8$ Cs atoms were accumulated in the first MOT, cooled to $3 \mu$K, and then transferred with an efficiency of 10% to a second, lower lying MOT. The second MOT is situated in an ultra high vacuum chamber, where the atoms are further cooled to $2 \mu$K and then dropped into the cavity from 5 mm above, with a velocity $\sim 5 \text{ cm/s}$, and kinetic energy $E_k \approx 30 \mu$K. The FORT beam is selected to be two longitudinal mode orders below the cavity QED mode, so the standing wave patterns of the two modes have approximately coincident antinodes near the center and at the ends of the cavity. This forms a blue-detuned optical dipole trap version of FORT, as described by Lee et al. [91]. For details and a theoretical simulation of the trapping mechanism, see van Enk et al. [92]. An optical cavity of length 44.6 mm and waist 20 mm was used, with finesse $F = 420,000$. This gives cavity parameters $(g_0, \kappa, \gamma)/2\pi = (32, 4, 2.6)$ MHz. The trap lifetime was reported to be $\tau = 28 \pm 6$ ms, and is limited by fluctuations in the intensity of the intracavity trapping field (FORT). It is anticipated that the trap lifetime can be made longer, since Miller et al. [90] reported confinement times of 200 ms in free space, and Lee et al. [91] reported lifetimes up to 50 s, also in free space. We note, however, that it may be difficult to achieve such long times inside an optical cavity. Nevertheless, the trapping time of 28 ms compares favourably to mean trapping times of $< 1$ ms in the experiments of Hood et al. [72] and Pinkse et al. [87]. In addition, the presence of an external trapping potential and the cavity QED field adds flexibility in predetermining where, and to what degree, atoms will be trapped and cooled.

A recent experiment by Guthohrlein et al. [93] achieved a very precise positioning of a single calcium ion, trapped by a radio-frequency trap in an optical cavity. A single atom is confined in the trap. The trap is then surrounded with a pair of highly reflective concave mirrors to form an optical cavity. Two mirrors, with a radius of curvature 10 mm, are positioned 6 mm apart. A calcium ion ($^{40}\text{Ca}^+$), sensitive to radiation close to the resonance line $4^2\text{S}_{1/2} - 4^2\text{P}_{1/2}$ at a wavelength of $\lambda = 397 \text{ nm}$, is probed by a laser beam of this wavelength. Throughout the experiment, probe laser is kept at a power of a few hundred nanowatts. The fluorescent light emitted by the ion is detected with a photomultiplier tube. The three-dimensional spatial structure of the first four $\text{TEM}_{0n}$ optical field modes inside the cavity is measured with a spatial resolution as small as 60 nm, and the modes were scanned over a range of up to 100 $\mu$m. More important from the viewpoint of cavity QED and the work presented in this thesis, the localisation of the ion in the optical field was measured as $\Delta x \approx 16 \text{ nm} = \lambda/25$. It is important to emphasise that this excellent spatial resolution was achieved without the material support structures, so the strong localisation and position control demonstrated in this remarkable experiment is an important advance as compared to the other techniques outlined in this Chapter. In addition, the linear geometry of the trap renders this setup an attractive candidate for simultaneous trapping of two or more ions along the axis. In such a configuration, the cavity field may be used to entangle two (or more) ions, and eventually perform quantum
operations on an adjacent pair of trapped ions in a long chain.

5.5 Conclusion

In conclusion, in this Chapter we have reviewed the physics of single–atom cavity QED to establish an experimental basis for the theoretical models we develop from now on. The field of single–atom cavity QED provides a realisable experimental environment for the analysis and description of quantum phenomena in a strong coupling limit.
Chapter 6

Model and Numerical Simulations

In this Chapter we will formulate the problem of a single-atom in a microcavity and present the methods used and results obtained from the numerical simulations of the system. This will show how the problems encountered in the many-atom case can be avoided by using the single-atom cavity QED arrangement.

6.1 Introduction

The many-atom version of the EIT-Kerr system failed to produce a satisfactory environment for the photon blockade effect, as was described in Chapters 3 and 4. Instead of a many-atom environment, one can also envisage a single atom with the same energy level structure placed in a microcavity of the type described in the previous Chapter. Provided the atom-cavity coupling strengths are sufficiently large, there is no reason why this system should not yield a photon blockade. In addition, having control over the parameters of the system, particularly the Rabi frequency $\Omega_c$, can provide us with an opportunity to avoid linewidth narrowing of a magnitude that would prevent successful implementation of photon blockade, described earlier in Chapter 4.

In Chapter 3 we reviewed some basic properties of the single-atom regime in a microcavity QED environment and its physical implementation. Such an environment is routinely implemented in at least two laboratories (Caltech in the USA and the Max Planck Institute in Germany), whose efforts and successes have been reviewed. These have encouraged us to consider the single-atom environment to be realistic for the experimental implementation of the scheme. In this Chapter we outline the model and present the results of simulations which demonstrate that a successful photon blockade can indeed be achieved in such a system, and hint at the parameter regimes for which the effect will occur.

In Chapter 7 we interpret results presented here in terms of dressed states, discuss the dynamics, and derive an effective Hamiltonian in terms of quasiparticles called polaritons.
6.2 Hamiltonian and the Master Equation

The atomic energy level scheme is the same as shown in Fig. 3.1. The cavity mode of frequency $\omega_{\text{cap}}$ couples the $|1\rangle \rightarrow |2\rangle$ and $|3\rangle \rightarrow |4\rangle$ transitions with coupling constants $g_1$ and $g_2$. The cavity is driven on resonance by a (weak) external probe field, while the transition $|3\rangle \rightarrow |2\rangle$ is driven by a classical coupling field with Rabi frequency $\Omega_c$, facilitating EIT of the probe field on the $|1\rangle \rightarrow |2\rangle$ transition. The condition for EIT in $\Lambda$-type three-level atoms is two-photon resonance between ground states. In addition, we assume a common detuning $\delta$ of the fields from level $|2\rangle$. The rates $\gamma_j$ are spontaneous emission rates for decay from the excited atomic levels via their respective channels.

Note that we assume that the coupling strengths $g_1$ and $g_2$ are constant and maximal, which amounts to the assumption that the atom is confined inside the cavity close to an antinode of the standing-wave field. In this context, the trapping mechanism realised by Ye et al. [89] is the preferred choice of the two mechanisms described in Section 5.4. This is because their trapping is achieved with an external mechanism, and the cavity mode interacting with the atoms remains free to be varied in the experiment. For example, a change in the external driving of the cavity mode will not cause change in the trapping potential, nor the position at which the atom is trapped.

In the electric dipole and rotating wave approximations, the Hamiltonian can be written as $H = H_{\text{system}} + H_{\text{damping}} + H_{\text{pump}}$, where $H_{\text{pump}} = i\hbar \mathcal{E}_p(a - a^\dagger)$ and $H_{\text{system}}$, in the interaction picture, is

\[
H_{\text{system}} = \hbar \delta \sigma_{22} + \hbar \Omega_\sigma_{44} + i\hbar \left[ g_1(a^\dagger \sigma_{12} - \sigma_{21}a) + g_2(a^\dagger \sigma_{34} - \sigma_{43}a) + (\Omega_c^* \sigma_{32} - \sigma_{23} \Omega_c) \right].
\]

(6.1)

Here, $\sigma_{ij}$ are the atomic raising or lowering operators (for $i \neq j$) and energy population operators (for $i = j$), and $\mathcal{E}_p$ is the strength of the incident (probe) field. $H_{\text{damping}}$ describes the coupling of the system to reservoirs mediating cavity decay and spontaneous emission. This term takes the usual form, which is the single-atom counterpart of the Hamiltonians (3.1d) and (3.1e),

\[
H_{\text{damping}} = i\hbar \int_{-\infty}^{+\infty} d\omega C(\omega)[b^\dagger(\omega)a - b(\omega)a^\dagger]
+ i\hbar \int_{-\infty}^{+\infty} d\omega D_1(\omega)[\beta^\dagger_1(\omega)\sigma_{12} - \sigma_{21}\beta_1(\omega)]
+ i\hbar \int_{-\infty}^{+\infty} d\omega D_2(\omega)[\beta^\dagger_2(\omega)\sigma_{32} - \sigma_{23}\beta_2(\omega)]
+ i\hbar \int_{-\infty}^{+\infty} d\omega D_3(\omega)[\beta^\dagger_3(\omega)\sigma_{34} - \sigma_{43}\beta_3(\omega)],
\]

(6.2)

with \{b(\omega), \beta_j(\omega)\} defined as the reservoir annihilation operators at frequency $\omega$, and \{C(\omega), D_j(\omega)\} coupling coefficients. As before, we employ the first Markov approximation, implying that \{C(\omega), D_j(\omega)\} are taken to be constants around the frequencies of
6.2. HAMILTONIAN AND THE MASTER EQUATION

interest, i.e. $C^2(\omega) = \kappa/\pi$ and $D^2_j(\omega) = \gamma_j/\pi$. We choose to derive a master equation instead of stochastic differential equations, which have served as the basis for the analysis of the many-atom case. The reason is that, having only one atom, the dimensions of the Hilbert space are $4 \times N$, where $N$ is now the truncation order for the dimension of the cavity mode subspace. In the regime in which the photon blockade is expected to work, the number of photons populating the intracavity mode is very small at any given moment, so the truncation $N = 10$ is "safe". The total dimension of the Hilbert space is then 40, which is numerically tractable. The master equation for the system density operator is derived by standard means [1], and we choose to write it in the form

$$\dot{\rho}_{sys} = \rho_{sys} + L \rho,$$

where $\rho_{sys}$ is the cavity decay rate and the detunings $\Delta_{ij}$ are defined as

$$\Delta_{21} = \omega_{21} - \omega_{cav} = \delta,$$  

$$\Delta_{31} = \omega_{31} - (\omega_{cav} - \omega_c) = 0,$$  

$$\Delta_{41} = \omega_{41} - (2\omega_{cav} - \omega_c) = \Delta,$$

according to Fig. 3.1, and where $\omega_{ij}$ denote the frequencies of the atomic transitions. Condition (6.4b) is the Raman resonance condition for the two-photon transition, chosen to preserve the effect of EIT.

Following Schmidt and Imamoglu [47] and Imamoglu et al. [51], one can derive an expression for the effective photon-photon interaction strength in an adiabatic approximation. Briefly, this derivation goes as follows [64]. First, a system of stochastic differential equations for the field and atomic operators is derived. Then the atomic operators are adiabatically eliminated, leaving only the equation for the field mode. From this equation, a term describing the effective photon-photon interaction strength can be identified, with an anharmonicity parameter

$$\eta = \left( \frac{g_1}{\Omega_c} \right)^2 \left( \frac{g_2^2 \Delta}{\gamma_1^2 + \Delta^2} - \frac{g_1^2 \delta}{(\gamma_1 + \gamma_2)^2 + \delta^2} \right),$$

in correspondence with the term $H_{Kerr} = \hbar \eta (a^\dagger a)^2$ in the effective Hamiltonian for the pure $\chi^{(3)}$ model of Ref. [51] (see also equation (2.7)). This expression will be used only as a guide for the choice of parameters, as we will show that the straightforward adiabatic approximation is not always justified in the configuration being considered.
6.3 Numerical Simulations and Discussion

The master equation (6.3) has been solved numerically. As a measure of how successfully the system works as a photon blockade, we choose to calculate the steady-state second order correlation function of the intracavity mode defined by

$$g^{(2)\tau} = \lim_{t \to \infty} \frac{\langle a^\dagger(t)a^\dagger(t+\tau)a(t+\tau)a(t) \rangle}{\langle a^\dagger(t)a(t) \rangle^2},$$

(6.6)

which we calculate via the quantum regression theorem [1]. The quantum regression theorem enables us to calculate two-time correlations by a proper choice of initial condition. To calculate $g^{(2)\tau}$, as defined by equation (6.6), we first find the steady-state solution $\rho_{ss}$ to the master equation (6.3). The denominator is then simply calculated by $\langle a^\dagger a \rangle_{ss} = \text{Tr}\{a^\dagger a \rho_{ss}\}$. To calculate the two-time second order correlation in the numerator, we solve the master equation as a function of time with an initial condition $\rho_{init} = a\rho_{ss}a^\dagger$. The solution is then a function of the time delay $\dot{\rho}(\tau)$, and $\langle a^\dagger a^\dagger(\tau)a(\tau)a \rangle_{ss} = \text{Tr}\{a^\dagger a \dot{\rho}(\tau)\}$. The numerical solution has been performed using the computational toolbox developed by S. M. Tan [94]. The concept of the toolbox and details of some of its routines are presented in [95].

For the cavity configuration, we assume that the mirror through which the cavity is driven by the probe field $E_p$ is almost perfect, while the other mirror contributes dominantly to the cavity mode decay rate $\kappa$ (see the setup in Fig 2.5). Then $g^{(2)\tau}$ measures the joint photocount probability of detecting a photon at time $t$, and another photon at time $t+\tau$, in the light field output through the ‘leaky’ mirror (i.e., in the field transmitted through the cavity). Ideal photon blockade then requires $g^{(2)\tau}(0) \approx 0$, followed by a rise of $g^{(2)\tau}$ to a value close to one on a timescale of the order of $\kappa^{-1}$. This is basically a requirement of photon antibunching in the transmitted field [1].

We choose parameters consistent with those realised in the recent experiments of Hood et al. [72] and Ye et al. [89]. For simplicity, we assume $g_1 = g_2 = g$ and $\gamma_1 = \gamma_2 = \gamma_3 = \gamma$. In the experiment of Hood et al. these values are $(g, \kappa, \gamma)/2\pi = (110, 14.2, 2.6)$ MHz, or

$$g = 7.75\kappa,$$

(6.7a)

$$\gamma \approx 0.18\kappa.$$ 

(6.7b)

The experiment of Ye et al. had the parameters $(g, \kappa, \gamma)/2\pi = (32, 4, 2.6)$ MHz, or

$$g = 8\kappa,$$

(6.8a)

$$\gamma \approx 0.65\kappa.$$ 

(6.8b)

We use values for $g$, $\kappa$ and $\gamma$ close to these two cases for all of the simulations. We vary $\Omega_c, E_p$ and $g$ (within reasonable limits) to give, according to equation (6.5), large effective nonlinear susceptibilities of order $\eta = 20\kappa$ or greater, and compare our results with those for an ideal $\chi^{(3)}$ nonlinearity.

Figure 6.1 shows several examples of the steady–state second order correlation function for the case of pure $\chi^{(3)}$ nonlinearity. The anharmonicity parameter is chosen to be...
6.3. NUMERICAL SIMULATIONS AND DISCUSSION

Figure 6.1: Second order correlation function for a pure nonlinearity vs. delay time, for different values of driving. Dot–dashed line is for $E_p = 0.5\kappa$, solid line for $E_p = 2.5\kappa$ and dashed line for $E_p = 5.0\kappa$. The inset shows the mean intracavity photon number for $E_p = 0.5\kappa$, calculated in the presence of damping (solid) and without damping (bold). Dotted line shows variance $\Delta n$ in the photon number for the undamped case.

$\eta/\kappa = 20$ for all cases. Dependence on the size of anharmonicity was already shown in Fig. 2.6. The field correlations are as would be typically expected in a two–level system undergoing resonance fluorescence [1]. There is a threshold value of driving $E_p = \kappa/2$ below which the solutions are not oscillatory and above which oscillations with frequency $E_p/(2\kappa)$ are present. The inset shows the mean value of the intracavity photon number for $E_p = 0.5\kappa$. While in the presence of damping, $\langle n \rangle$ quickly reaches its steady state, it is illuminating to look at its values in the absence of damping. It is seen that its value oscillates between zero and one, i.e. there is never more than one photon in the cavity, while the variance vanishes at $\langle n \rangle = 1$. This is the signature of photon blockade.

Figure 6.2 shows the results of simulations of equation (6.3). These results verify that strong antibunching can indeed be achieved, and the response time of the system is comparable to the adiabatic case of Imamoglu et al. [51] (dot–dashed curve). The response time in question is the key quantity for comparison with the Fourier–transformed analysis.
of the many atom system. The narrow frequency linewidth exhibited in the intensity squeezing spectrum will transform into a second order correlation function that shows slow recovery from zero. It is shown that by careful variation of parameters, this signature of the response time can be brought under control. Note in particular that the response time gets slower with increasing ratio \( g/\Omega_c \) (see Fig. 7.3 and discussion thereof). A quantitative explanation of this feature will be given in the following Chapter, where we construct an effective model to explain these simulations. Therefore, the single–atom system, unlike the many–atom medium \([55, 56]\), can in principle produce a photon blockade fast enough to be effective as a switch in a quantum logic environment. The small oscillations superimposed on the curves represent Rabi oscillations between the ground levels \( |1\rangle \) and \( |3\rangle \) and the excited levels \( |2\rangle \) and \( |4\rangle \), respectively, due to a small but non–zero population of the excited states.

In Figures 6.3 (a, c) the mean value of the intracavity field operator, \( \langle a \rangle \), and the
atomic coherences, $\langle \sigma_{12} \rangle$ and $\langle \sigma_{13} \rangle$, are plotted in the transient regime. The large coherence between the ground states, $\langle \sigma_{13} \rangle$, compared with the polarisation $\langle \sigma_{12} \rangle$, indicates EIT in the lower $\Lambda$—subsystem. The field $\langle a \rangle$ is plotted to indicate the time scale of the field oscillations and to compare it to the time scales for the atomic coherences. These are practically the same, implying that the adiabatic approximation is not justified in this system\(^1\). The adiabatic approximation is a procedure that relies on the existence of vastly different time scales in the system. A necessary condition for the elimination of the atomic degrees of freedom would be the existence of a fast atomic time scale, so that these variables approach their steady—state values much earlier than the cavity field variable(s). In that case, all of the atomic variables can be replaced with their steady—state values in the appropriate equations of motion.

Figs. 6.3 (b, d) show the mean values of the energy—level populations in the atom. It

\(^1\)It is important to say that this conclusion was not reached based on Fig. 6.3 only; rather, simulations performed for vast range of parameters all lead to the same conclusion.
can be seen that the total population is mostly distributed between the two ground states, with only a small fraction in the excited states. Due to EIT, the population of level \(j\) reaches a smaller value in its steady-state than the population of \(j_2\). This behaviour remains the same even for the case when \(\delta = 0\) and \(\Delta\) is large (see Chapter 2 and in particular Li and Xiao [96] for a detailed account of EIT dynamics).

We have also explored the possibility of varying the atomic detunings \((\delta, \Delta)\). For a chosen range of values between 0 and 10\(\kappa\), the only effect was slight deterioration in antibunching, i.e. slight increase in \(g^{(2)}(0)\), as could be expected from the equation (6.5).

Finally, while we have concentrated on the case of a continuous probe field \(\mathcal{E}_p\), it is clear that one can also consider the situation in which a ’\(\pi\)’ pulse, rather than continuous driving, is applied to the system. This will switch the system from the state with no photons in the cavity to the state with one photon in the cavity, in a single discrete operation. We will show in Chapter 7 that the advantage of this system over a two–level atom in a cavity lies in the flexibility of the effective parameters, such as the decay rate of the excited state, which can be adjusted to demand using the external field \(\Omega_c\).

### 6.4 Conclusion

The possibility of realising a photon blockade is exciting from many perspectives. For one, it offers a possibility of creating quantum gates and a single photon quantum control [97] environment. However, it also offers the possibility of studying fundamental properties of quantum systems, and quantum fields in particular. A successful realisation of the photon turnstile device would enable routine creation and study of single–photon Fock states. These are notoriously difficult to create, and even more difficult to detect unconditionally. Recent experiments have been performed by Brattke et al. [98] and Lvovsky et al. [99].

In this Chapter, we have presented the results of the numerical simulations of the single–atom cavity QED realisation of the EIT–Kerr system, and the photon blockade effect in particular. We have found that it is possible to realise a photon blockade effect in the single–atom system, and still have some control over the response time behaviour. We will present an effective model that explains these results in the next Chapter.
Chapter 7

Polariton Interpretation

In this Chapter we will develop an effective model to explain the results obtained in the previous Chapter. An analysis of the dressed states is presented and the concept of the polariton is introduced. An effective Hamiltonian is derived in the polariton basis.

7.1 Introduction

The parameters chosen for the simulations presented in the previous Chapter clearly fall into the strong coupling limit of cavity QED. This means that the atom and the photon from the field mode that couples to the atom necessarily lose some of their individual characteristics. As a result, the coupled system will exhibit some new features, which cannot be explained in terms of the properties of its constituents. This property of strongly coupled systems rests at the heart of quantum mechanics. It was as early as 1935 that Erwin Schrödinger stated this in his famous cat paradox paper [100]. There he states very succinctly

“Maximal knowledge of a total system does not necessarily include total knowledge of all of its parts, not even when these are fully separated from each other and at the moment are not influencing each other at all.”

In this sentence, we can recognise two main ideas applicable to the strongly coupled atom–cavity system:¹

1. Our knowledge (or total information) about the system is limited.

2. The total information of a composite system is not necessarily fully contained in its individual constituents.

There is also a third point, namely that these statements are independent of the actual space–time arrangements of the individual constituents and/or measurements performed

¹Of course, Schrödinger’s argument was not limited to strongly coupled systems, but is concerned with any coupled system in quantum mechanics. In practice, at this stage of technological development, strongly coupled systems are the only observable systems showing this peculiar behaviour.
CHAPTER 7. POLARITON INTERPRETATION

upon them. This point, however, is not relevant for our analysis (we are assuming a continuously monitored, spatially confined setup); it relates to the discussions of Einstein–Podolsky–Rosen correlations and Bell inequalities [101].

The concept of dressed states embodies the essence of the ideas mentioned above. Dressed states are defined as eigenstates of the Hamiltonian describing the system and its interactions. They are usually expressed as a particular linear combination of bare states, i.e. the states of the individual constituents of the coupled system. The coefficients of the linear combination depend on the nature of the interaction and paint a physical picture of the coupled system. It is therefore of fundamental importance to perform a dressed states analysis of the atom–cavity molecule.

7.2 Dressed States Analysis

We now investigate the coupled atom–field system in the dressed states picture. As well as being fundamentally sound, this approach will help us to identify the conditions under which the system described by $H_{\text{system}}$ (6.1) realises a good approximation to the pure $\chi^{(3)}$ nonlinearity as described by the effective Kerr Hamiltonian

$$H_{\text{Kerr}} = \hbar \eta \left( a^\dagger \right)^2 a^2.$$  
(7.1)

We will mainly concentrate on the analysis of $H_{\text{system}}$, which dominates the dynamics of the system, because the damping and pumping (by the probe field) are assumed to be virtually negligible on the scale of the dressed state structure. We will nevertheless obtain expressions for the effective pumping term and cavity damping.

Using the notation $|\text{cavity photon number, atomic state}\rangle$ for the bare states, the general dressed state can be written as

$$|\Psi\rangle = \sum_{n=0}^{\infty} \sum_{k=1}^{4} c_{nk} |n, k\rangle.$$  
(7.2)

The coefficients $c_{nk}$ are obtained by diagonalising the Hamiltonian (6.1). The ground state of the system is obviously $|e_0\rangle = |0, 1\rangle$. In the absence of driving, the total Hamiltonian naturally separates into different manifolds, according to the level of excitation of the coupled atom–cavity system. For example, a single photon excites the system into the first manifold; $n$ photons excite it into the $n$–th manifold. The bare states $|1, 1\rangle$, $|0, 2\rangle$ and $|0, 3\rangle$ are coupled by the Hamiltonian $H_{\text{system}}$ into the first manifold (in the interaction picture) to form the dressed states

$$|e_2\rangle = \frac{|1, 1\rangle + g_1/\Omega_c |0, 3\rangle}{\sqrt{1 + (g_1/\Omega_c)^2}},$$  
(7.3a)

$$|e_{1,3}\rangle = -\frac{(g_1/\Omega_c) |1, 1\rangle + i (\epsilon_{1,3}/\hbar \Omega_c) |0, 2\rangle - |0, 3\rangle}{\sqrt{1 + (g_1/\Omega_c)^2 + (\epsilon_{1,3}/\hbar \Omega_c)^2}},$$  
(7.3b)
Figure 7.1: Energy eigenstates of the empty cavity and the Hamiltonian $H_{\text{system}}$. Empty cavity modes are separated by $\hbar \omega_{\text{cav}}$, and dressed state structure is denoted with $|e_j\rangle$. The energy separation of the empty cavity modes is much larger than the energy separation within each group of dressed states ($\hbar \omega_{\text{cav}} \gg \epsilon_j$).
with corresponding energy eigenvalues \( E_j = \hbar \omega_{\text{cav}} + \epsilon_j \), where

\[
\begin{align*}
\epsilon_2 &= 0, \\
\epsilon_{1,3} &= \frac{\hbar \delta}{2} \mp \sqrt{\left(\frac{\hbar \delta}{2}\right)^2 + \left(\hbar \Omega_c\right)^2 \left(1 + \frac{g_j^2}{\Omega_c^2}\right)}.
\end{align*}
\]

(7.4a)

(7.4b)

Note that we take \( \Omega_c \) to be real. These states describe the excitations of the combined atom-cavity mode system, given that only a single photon (at most) excites the cavity mode.

For the case of the second excitation of the cavity mode, the Hamiltonian \( H_{\text{system}} \) couples bare states \( |2, 1\rangle, |1, 2\rangle, |1, 3\rangle \) and \( |0, 4\rangle \) to give

\[
|e_j\rangle = \alpha_j |2, 1\rangle + \beta_j |1, 2\rangle + \mu_j |1, 3\rangle + \nu_j |0, 4\rangle; \quad j = 4, \ldots, 7,
\]

(7.5)

with corresponding energy eigenvalues \( E_j = 2\hbar \omega_{\text{cav}} + \epsilon_j \). To calculate these, we keep the atomic detunings small (\( \delta, \Delta \ll g_j \)), enabling us to employ a perturbation expansion. The energy shifts \( \epsilon_j \) are calculated perturbatively in Appendix A (see also [102]).

The dressed states energy structure for the ground state and the lowest two manifolds is shown in Fig. 7.1. Arrows denote the transitions that contribute most strongly to the dynamics for the chosen parameters. The transition \( |e_0\rangle \rightarrow |e_2\rangle \) dominates, since in our scheme it is resonantly excited by the probe field. Two transitions \( |e_2\rangle \rightarrow |e_{5,6}\rangle \) are off-resonant, but have finite transition probabilities, and require the transfer of a second excitation to the atom–cavity system from the probe driving field. For the typical set of parameters used in our simulations, the values \( \epsilon_{5,6} \sim \mp \kappa \) are the detunings from the bare cavity resonance for these transitions. Such detunings enable the transitions \( |e_2\rangle \rightarrow |e_{5,6}\rangle \) to be suppressed, making possible an efficient photon blockade. The size of the detunings \( \epsilon_{5,6} \) of the states \( |e_{5,6}\rangle \) can be increased by increasing the atom–field coupling strength. Fig. 7.2 shows the coupling dependence of the energy detunings from the cavity resonance of the dressed states belonging to the second manifold. Increasing the coupling strength increases the splitting between the states \( |e_{5,6}\rangle \). This is not surprising, since the size of the anharmonicity (6.5) depends on the coupling strength. In other words, since the atom is the source of the nonlinearity, photons have to couple strongly to the atom in order to shift the harmonic energy levels away from the empty cavity resonance.

### 7.3 Polaritons and Dressed States

To define the dressed states picture consistently, and obtain another useful perspective on the dynamics of this system, we introduce the concept of the polariton. In general, a polariton is defined as a quasiparticle arising from the interaction of a photon with the polarisation quantum of the medium. If the interaction is weak, both the photon and the polarisation quantum largely retain their individual properties. If the interaction is strong, however, the photon and the polarisation quantum exist rather as a single quantity which can be interpreted as a new elementary excitation – a polariton. In the
7.3. POLARITONS AND DRESSED STATES

Figure 7.2: Energies of the dressed states $\epsilon_j = E_j - 2\hbar \omega_{\text{cav}}$ plotted against $g/\Omega_c$. Parameter $g$ was varied, while keeping $\Omega_c = 5\kappa$.

In solid state physics, a polarisation quantum can be a transverse optical phonon for the polarisation of the ion lattice, or an exciton for the polarisation of the electron system.
corresponding to the other polariton states \( |e_j \rangle \). For example, the other two polariton operators for the first manifold would be

\[
p_{1,3}^\dagger = - \frac{(g_1/\Omega_c) a^\dagger + i (\epsilon_{1,3}/\hbar \Omega_c) \sigma_{21} - \sigma_{31}}{\sqrt{1 + (g_1/\Omega_c)^2 + (\epsilon_{1,3}/\hbar \Omega_c)^2}}. \tag{7.6b}
\]

It is interesting to observe that polaritons are neither bosons nor fermions. The commutation relations satisfied by the operators \( p_j \) and \( p_{j}^\dagger \) are

\[
[p_2, p_2^\dagger] = \frac{1 - (g_1/\Omega_c)^2 (\sigma_{33} - \sigma_{11})}{1 + (g_1/\Omega_c)^2}, \tag{7.7a}
\]

\[
[p_{1,3}, p_{1,3}^\dagger] = \frac{(g_1/\Omega_c)^2 + (\epsilon_{1,3}/\hbar \Omega_c)^2 (\sigma_{22} - \sigma_{11}) - (\sigma_{33} - \sigma_{11})}{1 + (g_1/\Omega_c)^2 + (\epsilon_{1,3}/\hbar \Omega_c)^2}, \tag{7.7b}
\]

with all the other commutators vanishing. Therefore, strong coupling of bosons and fermions yields an excitation (or a quasiparticle) of mixed statistics.

We can define two limits in which the polaritons become dominated by the contribution of their constituents, according to the ratio \( g_1/\Omega_c \). Note that this ratio is closely related to the single–atom version of the parameter \( g_{\text{eff}} \), defined in Chapter 3, equation (3.31). When \( g_1/\Omega_c \ll 1 \), polariton \( p_2 \), and its corresponding eigenstate \( |e_2 \rangle \), become dominated by their photonic contribution, while polaritons \( p_{1,3} \) and corresponding eigenstates become dominated by their atomic contribution. So, different polaritons become either photon–like or atom–like. The opposite situation occurs for \( g_1/\Omega_c \gg 1 \). The region \( g_1/\Omega_c \sim 1 \) is a “no–mans land”, where photons and atoms contribute comparably to the composition of polaritons.

### 7.4 Effective Hamiltonian

Provided that the detunings of the states \( |e_{1,3} \rangle \) and \( |e_{5,6} \rangle \) are large, the EIT–Kerr system becomes an effective two–level system. In that case an approximate form of the Hamiltonian (6.1) can be derived in terms of the polariton operators involving the ground state \( |e_0 \rangle \) and resonant state \( |e_2 \rangle \).

To accomplish this, we will first assume that the detuning of level \( |4 \rangle \) is sufficiently large \( (\Delta \gg \gamma_3) \), so that its population is negligible and we can adiabatically eliminate it. The resulting Hamiltonian describes a three level system in the EIT configuration, with an additional term that describes the ac–Stark shift to the level \( |3 \rangle \),

\[
\mathcal{H}_{\text{res}} = \mathcal{H}_{\text{Stark}} + \mathcal{H}_{\text{EIT}}, \tag{7.8a}
\]

\[
\mathcal{H}_{\text{Stark}} = \hbar \Delta \frac{g_2^2}{\gamma_3^2 + \Delta^2} a^\dagger a \sigma_{33}, \tag{7.8b}
\]

\[
\mathcal{H}_{\text{EIT}} = \hbar \delta \sigma_{22} + i \hbar [g_1 (a^\dagger \sigma_{12} - \sigma_{21} a) + (\Omega_c^* \sigma_{32} - \sigma_{23} \Omega_c)] \tag{7.8c}
\]
The remaining operators can be transferred to the polariton basis by using the projector approach and defining

\[ a^\dag \sigma_{12} = |1, 1 \rangle \langle 0, 2|, \quad (7.9a) \]
\[ \sigma_{23} = |0, 2 \rangle \langle 0, 3|, \quad (7.9b) \]
\[ \sigma_{22} = |0, 2 \rangle \langle 0, 2|, \quad (7.9c) \]
\[ \sigma_{33} = |0, 3 \rangle \langle 0, 3|, \quad (7.9d) \]

with their adjoints following naturally. Note that we have concentrated on the ground and first manifold states, as a consequence of the large detunings from resonance with the higher lying polaritons, which makes them unlikely to be excited. In general, the definitions (7.9) are only the first terms in the expansion over the excitation number, i.e. the order of the manifold. The bare states on the right-hand sides of the equations above can be expressed in terms of polariton states. We define the vectors containing bare and dressed states as

\[ b = \left( |1, 1 \rangle, |0, 2 \rangle, |0, 3 \rangle \right)^T, \quad (7.10a) \]
\[ d = \left( |e_1 \rangle, |e_2 \rangle, |e_3 \rangle \right)^T. \quad (7.10b) \]

The transformation is then \( b = Td \), with the transformation matrix

\[ T = \left( \begin{array}{ccc}
-\frac{g_1/\Omega}{N_1} & \frac{1}{N_2} & -\frac{g_2/\Omega}{N_3} \\
\frac{\epsilon_1/\Omega}{N_1} & 0 & \frac{\epsilon_2/\Omega}{N_3} \\
\frac{1}{N_2} & \frac{g_1/\Omega}{N_3} & \frac{1}{N_3}
\end{array} \right), \quad (7.11) \]

where

\[ N_2 = \sqrt{1 + \left( \frac{g_1}{\Omega_c} \right)^2}, \quad (7.12a) \]
\[ N_{1,3} = \sqrt{1 + \left( \frac{\epsilon_{1,3}}{\Omega_c} \right)^2 + \left( \frac{g_1}{\Omega_c} \right)^2}. \quad (7.12b) \]

The polariton operators \( p_j \) (equations (7.6a) and (7.6b)) can be written as operators in the dressed basis as \( p_j^\dag e^{i\epsilon_j} = |e_j \rangle \langle e_0| \). Note the phase term in the definition of the operators. It is due to the energy separation of the different polariton states. For the large detunings \( \epsilon_{1,3} \), an equivalent of the rotating wave approximation can be introduced, i.e. the terms containing the operators \( p_{1,3} \) (and their adjoints) will oscillate with much higher frequency than the terms containing only \( p_2 \) (and its adjoint). The faster rotating terms will average to zero on the time scale of \( p_2 \) and can be neglected. The resulting effective Hamiltonian, retaining only the dominant terms, takes the simple form

\[ H_{\text{Kerr}} = \hbar \epsilon_1 p_1^\dag p_1 + \hbar \epsilon_3 p_3^\dag p_3 + \hbar \tilde{\gamma} (p_2^\dag p_2)^2, \quad (7.13) \]

\(^3\text{We could have started from the Hamiltonian not written in the rotating frame, and these phase factors would follow naturally.}\)
with the anharmonicity being

\[ \tilde{\eta} = \frac{\eta}{1 + \eta_1^2 / \Omega_c^2} \]  

(7.14)

where \( \eta \) is given by (6.5). Therefore, the effective Kerr nonlinearity is not purely optical in nature but involves the polariton operator \( p_1^\dagger \). It is the effective polariton nonlinearity.

The two terms proportional to \( \epsilon_{1,3} \) represent the detunings of the other two polaritons in the first manifold.

A somewhat less general result can be obtained by assuming a sufficiently large detuning \( \delta \gg (\gamma_1 + \gamma_2)/2 \), and adiabatically eliminating level \( |2\)\. This leaves the system described just by the operators \( \{a, \sigma_{13}, \sigma_{33}, \sigma_{11}\} \) (and their adjoints), with, for example, terms of the form \( a^\dagger a \sigma_{ii} \) \((i = 1, 3)\). A simplified version of the procedure above can be performed, by noting that \( \sigma_{11} \) and \( \sigma_{33} \) can be expressed in terms of polariton operators \( p_2 \) and \( p_1^\dagger \) only.

The pump term can be transformed into the polariton basis by noting that

\[ a \approx |0, 1\rangle \langle 1, 1| \]  

(7.15)

and repeating the procedure outlined above. In the rotating wave approximation, the polariton pump term is then

\[ \mathcal{H}_{\text{pump}} = i \frac{\hbar \Omega_1}{2} (p_1 - p_1^\dagger) + i \frac{\hbar \Omega_R}{2} (p_2 - p_2^\dagger) + i \frac{\hbar \Omega_3}{2} (p_3 - p_3^\dagger), \]  

(7.16)

with effective Rabi frequencies for driving the respective transitions

\begin{align*}
\Omega_1 &= 2 \mathcal{E}_p T_{11} = -\frac{2 \mathcal{E}_p g_1 / \Omega_c}{N_1} \\
&= -2 \mathcal{E}_p \frac{g_1 / \Omega_c}{\sqrt{1 + (\epsilon_1 / \hbar \Omega_c)^2 + (g_1 / \Omega_c)^2}}, \quad (7.17a) \\
\Omega_R &= 2 \mathcal{E}_p T_{12} = \frac{2 \mathcal{E}_p}{\sqrt{1 + (g_1 / \Omega_c)^2}}, \quad (7.17b) \\
\Omega_3 &= 2 \mathcal{E}_p T_{13} = -\frac{2 \mathcal{E}_p g_1 / \Omega_c}{N_3} \\
&= -2 \mathcal{E}_p \frac{g_1 / \Omega_c}{\sqrt{1 + (\epsilon_3 / \hbar \Omega_c)^2 + (g_1 / \Omega_c)^2}}, \quad (7.17c)
\end{align*}

where \( T_{ij} \) denote respective elements of matrix \( T \), given by equation (7.11). If we follow the procedure of eliminating atomic level \( |2\)\, only the term driving the resonant transition will be obtained, i.e. \( \mathcal{H}_{\text{pump}} \approx i \hbar \Omega R / 2 (p_2 - p_2^\dagger) \).

There are two (basically equivalent) ways of obtaining the effective cavity damping. One may add the term \(-i \hbar \mathcal{E}_p a^\dagger a\) to the Hamiltonian, and perform the same analysis as with the other terms, or one can include it into the diagonalisation procedure. Since the
Figure 7.3: Probabilities for having zero or one photon \((a,b)\) and polariton \((c,d)\) in a cavity, for parameters as in dotted \((a,c)\) and dashed \((b,d)\) curve in Fig. 6.2. In all Figures, solid curves describe the full system, while dashed curves describe the system with no damping. The time scale of Rabi oscillations in the absence of damping is determined by \(\Omega_R\).

Hamiltonian with this effective damping term is non-Hermitian, its eigenvalues will be complex. The imaginary part gives effective linewidths associated with the appropriate polaritons. Since state \(|e_2\rangle\) is resonant with the cavity, its linewidth is the effective cavity damping. This way is more straightforward and yields

\[
\Gamma_2 = \frac{\kappa}{1 + \left(\frac{g_1}{\Omega_c}\right)^2}.
\]  

The effective damping term then becomes \(-i\hbar(\Gamma_1p_1^\dagger p_1 + \Gamma_2p_2^\dagger p_2 + \Gamma_3p_3^\dagger p_3)\), where the linewidths of the other two polariton states are given to a good approximation by \(\Gamma_{1,3} \approx \kappa/2\).

As remarked earlier, the polariton operator (7.6a) can be regarded in two limits as photon-like or atom-like, depending on whether it is dominated by the contribution from
the atomic or field operator. It is now clear that the case of optical $\chi^{(3)}$ nonlinearity emerges in the photon–like case when $g_1^2/\Omega_c^2 \ll 1$. In this limit, the effective Hamiltonian becomes

$$
\hbar \tilde{\eta} \left( p_2^\dagger \right)^2 p_2^2 + \frac{i \hbar \Omega_R}{2} \left( p_2 - p_2^\dagger \right) \rightarrow \hbar \eta (a^\dagger)^2 a^2 + i \mathcal{E}_p (a - a^\dagger).
$$

Note that this is the single–atom version of the limit $g_{\text{eff}} \ll 1$ defined in Chapter 3, equation (3.31). The polariton analysis presented above offers an explanation for this limit. When the polaritons formed by coupling the cavity field mode to the atom(s) are dominated by their photonic contribution, then it is justified to perform the adiabatic elimination and arrive at the purely optical effective nonlinearity.

### 7.5 More On the Polariton Structure

To illustrate the polariton structure, we numerically calculate the probabilities for intracavity photons and polaritons. In Figs. 7.3 (a, b), the probabilities for finding the system with zero and one photon in the cavity are calculated as $\text{Tr}\{(|0\rangle \langle 0| \otimes \mathbb{1}_{\text{atom}}) \rho\}$ and $\text{Tr}\{(|1\rangle \langle 1| \otimes \mathbb{1}_{\text{atom}}) \rho\}$ and shown as functions of time. These probabilities very nearly add to one at all times, indicating that there is basically never more than one photon in the cavity at a time, as expected for a system exhibiting a photon blockade. In the absence of damping, there are coherent oscillations in the probabilities, but these are not of unit amplitude. The reason for this can be seen in Figs. 7.3 (c, d) where we now plot the probabilities for finding the system in the dressed states $|e_0\rangle$ and $|e_2\rangle$, calculated as $\text{Tr}\{(|e_0\rangle \langle e_0|) \rho\}$ and $\text{Tr}\{(|e_2\rangle \langle e_2|) \rho\} = \text{Tr}\{(p_2^\dagger p_2) \rho\}$. The oscillations of near unit amplitude confirm that this is effectively a two–state system undergoing Rabi oscillations. The frequency of the Rabi oscillation was calculated earlier and is given by equation (7.17b).

For the parameters of (a) and (c) (as for the dotted curve in Fig. 6.2), the polariton state $|e_2\rangle$ is primarily composed of the state $|0,3\rangle$ rather than $|1,1\rangle$, so that the excitation is stored mainly in the atom rather than in the cavity mode. On the other hand, for the parameters of (b) and (d) (as for the dashed curve in Fig. 6.2), the state $|e_2\rangle$ is primarily composed of $|1,1\rangle$ so that the excitation is stored mainly in the cavity mode.

The fast modulations appearing in these Figures as well as in the correlation functions in Figures 6.2 and 6.3 in Chapter 6 are the signature of the states $|e_{1,3}\rangle$, and occur with the frequency $\epsilon_1 \approx \epsilon_3$. These modulations are described with the first two terms in the effective Hamiltonian (7.13).

We will now explore the transitions $|e_2\rangle \rightarrow |e_{5,6}\rangle$ in more detail. These transitions did not contribute strongly in the simulations of Figs. 7.3 (c, d), but did result in a small difference from unity for the maximum value of the population of $|e_2\rangle$.

We calculate the relevant probabilities numerically and plot them to see their contribution. We choose the photon–like limit of $g_1/\Omega_c = 3/5$, as given by the dashed curve in Fig. 6.2 and in Figs. 7.3 (b, d). Fig. 7.4 summarises the results. In Fig. 7.4 (a), we
Figure 7.4: Probabilities for (a) polaritons $|e_0\rangle$ and $|e_2\rangle$, (b) polaritons $|e_5\rangle$ and $|e_6\rangle$, and (c) sum of polaritons from (a) and (b). Parameters are as in Figs 7.3 (b, d).

show the same plot as in Fig. 7.3 (d). In Fig. 7.4 (b), we plot the populations of the polariton states $|e_5\rangle$ and $|e_6\rangle$. Again, we show damped and undamped cases for each of the polaritons. In Fig. 7.4 (c), we sum the probabilities for $|e_2\rangle$, $|e_5\rangle$ and $|e_6\rangle$ of Figs. 7.4 (a) and (b). This sum oscillates between zero and unity exactly, indicating that we have now incorporated all of the most important states involved in the dynamics. It is this contribution from the second manifold polaritons that accounts for the small departure from zero of the correlation functions $g^{(2)}(0)$ shown in Fig. 6.2. A probability for ground state $|e_0\rangle$ is also shown for the comparison.

We have considered if it is possible to choose parameters to better suppress the contribution of the states $|e_5,6\rangle$, i.e. if it is possible to choose parameters so as to detune the states $|e_{5,6}\rangle$ further off the bare cavity resonance. The limit $g_1/\Omega_c \ll 1$ is also the limit of a photon-like polariton $|e_2\rangle$, but in order to achieve sufficient detuning, we would have to choose a ratio that would reduce the size of nonlinearity (as given by (6.5)) below the value required for successful photon blockade. The limit $g_1/\Omega_c \gg 1$ is desirable from the
viewpoint of strong nonlinearity, but it is also the limit of an atom-like polariton $|e_2\rangle$. In this case, the excitation is largely stored in the atom, thus slowing down the response time of the system. Unfortunately, varying the atomic detunings does not help either. In the region of interest, this only induces relatively small shifts in the values of the energy level detunings. So, in order to minimise the effects of transitions to the second excited state manifold, it would seem that values of $g_1/\Omega_c$ close to those already considered in the earlier calculations offer perhaps the best compromise.

7.6 Conclusion

In conclusion, we have proposed a scheme for a photon blockade, based on EIT in a single four level atom in a microcavity. The small size of the microcavity enhances the atom-field coupling, while high finesse mirrors and EIT keep decoherence and absorption to a minimum. A thorough analysis in the dressed states/polariton picture was performed, which enabled us to understand the dynamics of the system and identify the best operating regime. A full master equation was numerically simulated and it was shown that such a system can indeed produce large Kerr nonlinearities of the type proposed by Imamoglu et al. [51], avoiding the difficulties encountered in the many-atom case [55, 56].

The effective two level behaviour was identified and simulations performed that show Rabi oscillations between the two polariton states $|e_0\rangle$ and $|e_2\rangle$. The polariton approach and the dressed states analysis will be further developed in Chapter 10.
Part IV

Photon–Photon Interactions in Cavity QED
Chapter 8

Photon Blockade with Two Level Atoms

We have shown in the previous Chapter how a single–atom EIT–Kerr system exhibits a photon blockade in the limit of weak driving. In this Chapter we show how Kerr nonlinearity and the photon blockade effect can be realised using a two level atom.

8.1 Introduction

In the previous Chapter, we demonstrated that an EIT–Kerr scheme based on a single atom could enable the observation of photon blockade using state–of–the–art cavity QED configurations [72]. A dressed states analysis and the polariton interpretation enabled a full understanding of the numerical simulations. It is also of interest to compare the performance of the EIT–Kerr scheme with that of other schemes that have been predicted to yield photon blockade.

In particular, Tian and Carmichael [103] have predicted that under conditions of strong dipole coupling of an optical cavity to a single two level atom, an effective two level behaviour can be expected if this system is excited near one of the vacuum Rabi resonances. This is essentially a photon blockade, since, under these conditions, the possibility of a second excitation of the coupled system is strongly suppressed due to the lack of resonant energy level(s) in the excited manifold(s). An effective $\chi^{(3)}$ nonlinearity is exhibited by this system, and it was shown by Werner and Imamoğlu [104] that the maximum nonperturbative Kerr nonlinearity of the EIT–Kerr scheme is actually very close to that of the two level scheme.

In this Chapter we will outline the physics of photon blockade with two level atoms. The single–atom and many–atom cases will be reviewed and it will be shown that the addition of more atoms reduces the effectiveness of the photon blockade. These results are then compared in Chapter 9 with the EIT–Kerr model. In Appendix B, we derive the expressions for the effective nonlinearities in both systems and compare their sizes in the perturbative (weak coupling) and nonperturbative (strong coupling) limits.
CHAPTER 8. PHOTON BLOCKADE WITH TWO LEVEL ATOMS

Figure 8.1: Schematics of a two level atom. Ground and excited states are denoted by $|\rangle$ and $|\rangle + i\rangle$ respectively. The field mode couples to the atomic transition of frequency $\omega_0$ with strength $g$, and may be detuned from the atomic transition by $\delta_{JC}$. The excited state $|\rangle + i\rangle$ has a spontaneous decay rate $\gamma$.

8.2 The Extended Jaynes–Cummings Model

The canonical model for study of two level atomic systems is the Jaynes–Cummings (JC) model [105]. It describes the interaction of a single two level atom with a single mode of the quantised electromagnetic field. A strong coupling limit is assumed in this model, and damping of the field mode and atomic spontaneous emission are neglected.

Tavis and Cummings [106] have generalised this model to the case of $N$ identical atoms interacting with the single quantised radiation field mode in what is sometimes called the Tavis–Cummings model. This is a natural extension of the single-atom model, but does not include the dissipation terms either.

To include damping and pump terms in the analysis, we define the extended JC model. By the extended JC model we assume a collection of two-level atoms interacting with a single mode of a quantized cavity field, where the interaction of each atom with the field mode can be described by the well-known JC Hamiltonian [105], extended to include dissipative coupling. The Tavis–Cummings part of the Hamiltonian of the extended JC model [105] (omitting dissipative couplings) is given by

$$\mathcal{H}^{JC} = \mathcal{H}_0^{JC} + \mathcal{H}_{\text{pump}} + \mathcal{H}_{\text{int}}^{JC},$$  \hspace{1cm} (8.1)
8.3. DRESSED STATES AND THE PHOTON BLOCKADE

where

\[
\mathcal{H}_{0}^{JC} = \frac{\hbar \omega_0}{2} \sum_{j=1}^{N} \sigma_j^z + \hbar \omega_{cav} a^\dagger a, \tag{8.2a}
\]

\[
\mathcal{H}_{pump} = i \hbar \mathcal{E}_p (a e^{-i \omega_L t} - a^\dagger e^{i \omega_L t}), \tag{8.2b}
\]

\[
\mathcal{H}_{int}^{JC} = i \hbar \sum_{j=1}^{N} (g_j a^\dagger \sigma_j^+ - g^*_j \sigma_j^- a). \tag{8.2c}
\]

Here, \(\mathcal{H}_{0}^{JC}\) is the free Hamiltonian of the atoms and field and \(\mathcal{H}_{int}^{JC}\) describes the interaction of the two level atoms with the quantized field. The operators \(a\) and \(a^\dagger\) are the annihilation and creation operators for the single cavity field mode, while \(\sigma_j^z\) and \(\sigma_j^\pm\) are the inversion, raising and lowering operators for the \(j\)-th atom. The driving of the cavity mode by a classical field is described by the Hamiltonian \(\mathcal{H}_{pump}\). \(N\) is the number of atoms in the cavity, \(\omega_0\) and \(\omega_{cav}\) are, respectively, the atomic and cavity resonance frequencies, and \(\omega_L\) is the frequency of the external driving field. \(\mathcal{E}_p\) is the coupling strength of the driving field with the cavity mode, taken to be real. The coherent coupling \(g_j\) between an individual atom and the cavity mode in general depends on the position of the atom; we will assume \(g_j = g\) for simplicity. Including the dissipation leads to the effective Hamiltonian

\[
\mathcal{H}_{eff}^{JC} = \mathcal{H}^{JC} - i \hbar \kappa a^\dagger a - i \hbar \sum_{j=1}^{N} \gamma_j \sigma_j^+ \sigma_j^-, \tag{8.2d}
\]

where \(\kappa\) and \(\gamma_j\) are the cavity and atomic spontaneous emission decoherence rates. Naturally, \(\mathcal{H}_{eff}^{JC}\) has to be combined with a gedanken measurement process in order to obtain the complete dynamics of the system. This means that the effective Hamiltonian (8.2d) has to be complemented by the appropriate collapse operators [15], which in the case of direct measurement are

\[
C_a = \sqrt{\kappa} a, \tag{8.3a}
\]

\[
C_j = \sqrt{\gamma_j} \sigma_j^-. \tag{8.3b}
\]

Fig. 8.1 illustrates the system commonly described by the JC model.

8.3 Dressed States and the Photon Blockade

As in the case of the EIT–Kerr system, the emergence of photon blockade can be best understood in terms of dressed states. This is again a strongly coupled system, and the dressed states again emerge as a natural representation for the analysis. We differentiate the single–atom case from the case of more than one atom to simplify the comparison with the EIT–Kerr system, where this distinction becomes even more important.
Figure 8.2: Dressed states of the single two level atom coupled to the cavity mode. We assume that the cavity mode is resonant with the atomic transition $\omega_{cav} = \omega_0$. Detunings from the resonance are given by Eq. (8.4a) taken in the strong coupling limit. The ground state is denoted by $|0\rangle$.

8.3.1 Single Atom Case

We diagonalise the Hamiltonian (8.2d) for the single–atom in the absence of driving and obtain

$$\epsilon_{\pm}^{(n)} = \pm g \sqrt{n - \left(\frac{\gamma - \kappa}{2}\right)^2},$$

$$\Gamma_{\pm}^{(n)} = \frac{\gamma + (2n - 1)\kappa}{2},$$

where $\Gamma_{\pm}^{(n)}$ are the linewidths, and $E_{\pm}^{(n)} = \hbar(\omega_{cav} + \epsilon_{\pm}^{(n)})$ are the energies of the Rabi split states

$$|\epsilon_{\pm}^{(n)}\rangle = \frac{1}{\sqrt{2}} \left\{ |n - 1, +\rangle + \left[ i \frac{\gamma - \kappa}{2g\sqrt{n}} \pm \sqrt{1 - \left(\frac{\gamma - \kappa}{2g\sqrt{n}}\right)^2} \right] |n, -\rangle \right\}.$$ 

In the limit of strong coupling, $g\sqrt{n} \gg (\gamma - \kappa)/2$, these states assume the familiar form

$$|\epsilon_{\pm}^{(n)}\rangle = \left( |n - 1, +\rangle \pm |n, -\rangle \right)/\sqrt{2}.$$ 

Figure 8.2 shows the first two excited manifolds relative to the frequency of the cavity mode. The states in the Figure are also shown in the strong coupling limit.
8.3. DRESSED STATES AND THE PHOTON BLOCKADE

Tian and Carmichael [103] have shown that if this system is weakly driven on the lower Rabi resonance\(^1\), the system behaves essentially as a two level system. This situation is described by the Hamiltonian, written in the frame rotating at \(\omega_{\text{cav}} = \omega_0\),

\[
\mathcal{H} = \hbar \delta_{JC} (a^\dagger a + \sigma_z/2) + i\hbar g (a^\dagger \sigma_- - \sigma_+ a) + \hbar \xi_p (a - a^\dagger) - i\hbar \gamma \sigma_+ \sigma_-,
\]

where the detuning \(\delta_{JC} = \omega_{\text{cav}} - \omega_L = g\) is the common detuning of both the atom and the cavity mode from the driving field. Note that the cavity mode is resonant with the atom. The detuning \(\delta_{JC}\) does not change the relative position of the dressed states, and contributes only a trivial shift \(\delta_{JC}(n - 1/2)\) to the entire ladder. The linewidths are also unchanged.

Photon blockade can occur in a familiar way. After the first photon excites the system, a second incident photon is detuned by a frequency

\[
\tilde{\epsilon}_{N=1} = (2 - \sqrt{2}) g
\]

from resonance with a second excitation. In the next Section, we will show that this detuning critically depends on the number of atoms \(N\).

It is possible to follow the same reasoning as in Section 7.4 and obtain the effective Hamiltonian for this system. We define the raising operators, \(q_\pm^\dagger\) such that \(|e^{(1)}_\pm\rangle = q_\pm^\dagger |0, -\rangle\), as

\[
q_\pm \exp (\pm i e^{(1)}_\pm) = \frac{1}{\sqrt{2}} (\sigma_- \pm a).
\]

The commutation relations \([q_\pm, q_\pm^\dagger] = (1 + \sigma_z)/2\) again show excitations with mixed statistics, but now there is no simple limit in which these become predominantly photonic or atomic in nature. Substituting the operators (8.8) in the Hamiltonian (8.6), transforming the Hamiltonian to a frame rotating at laser frequency \(\omega_L = \omega_{\text{cav}} - \delta_{JC}\), and performing the rotating wave approximation, we arrive at the effective Hamiltonian

\[
\mathcal{H}_{\text{eff}} = 2\hbar \delta_{JC} q_+^\dagger q_+ + i \frac{\hbar \xi_p}{\sqrt{2}} (q_- q_+^\dagger + q_+ q_-^\dagger)
\]

\[
- i\hbar \frac{\gamma + \kappa}{2} (q_- q_-^\dagger + q_+ q_+^\dagger).
\]

This Hamiltonian contains the effective Hamiltonian of Tian and Carmichael [103], plus two more terms, proportional to \(q_+^\dagger q_+\). Figure 8.3 shows the second order correlation function for two different values of driving. We note three key features:

1. The degree of applicability of the effective model is marked by the value of \(g^{(2)}(0)\). Ideally, it should be exactly zero, and it indeed approaches this value for large \(g\).

\(^1\)The same argument remains valid for driving of the upper Rabi resonance.
CHAPTER 8. PHOTON BLOCKADE WITH TWO LEVEL ATOMS

Figure 8.3: Second order correlation function for a single two level atom in a cavity driven on the lower Rabi resonance by the external field $E_p$. The atomic transition is resonant with the cavity mode. The detuning of the driving is $\delta_{JC} = g$, where $g = 6\kappa$. The spontaneous emission rate is taken to be $\gamma = 0.1\kappa$.

2. Large amplitude oscillations of frequency $\sqrt{2}E_p$ are predicted by the effective model Hamiltonian to occur for $E_p > (\kappa + \gamma)/2$.

3. Small amplitude modulations of frequency $2\delta_{JC} = 2g$ occur as a signature of the upper Rabi resonance. These are also predicted by the effective Hamiltonian.

The last feature points at the failure of the effective two level model. In contrast, the Hamiltonian (8.9) represents an effective two manifold model, reducing the dynamics to transitions between the ground and first excited manifolds. This is the source of terms proportional to $q_-^+ q_+^-$, and accounts for the modulation, where the effective two level model of Tian and Carmichael fails. The dependence on driving will be treated in Part V.
8.3. DRESSED STATES AND THE PHOTON BLOCKADE

\[ N = 2 \quad N \gg 1 \]

\[ \hbar \omega_0 \]

\[ \pm g \sqrt{6} \quad \pm 2g \sqrt{N} \]

\[ \hbar \omega_0 \]

\[ \pm g \sqrt{2} \quad \pm g \sqrt{N} \]

\[ \ket{0} \quad \ket{0} \]

Figure 8.4: Dressed states for two and many atoms coupled to the cavity mode (energy spacing is not up to scale). Ground state is denoted by \( \ket{0} \).

8.3.2 Dependence on the Number of Atoms

Introducing more than one atom in the cavity necessarily jeopardises the photon blockade. Figure 8.4 shows the dressed states for the case of two atoms and for the limiting case of \( N \gg 1 \) atoms. The distinctive feature of these sets of states is the emergence of a third state in the second (and higher) manifolds, resonant with the cavity mode. On the other hand, the splitting between the Rabi resonances widens by a factor of \( \sqrt{N} \). Driving the system containing two atoms on the lower Rabi resonance gives a detuning for the second excitation of

\[ \tilde{\epsilon}_{N=2} = (2\sqrt{2} - \sqrt{6}) \hbar g < \tilde{\epsilon}_{N=1} \]

from the nearest resonance.

For a large number of atoms \( (N \gg 1) \), the manifold of the \( n = 1 \) excitation contains two states detuned by \( \pm g \sqrt{N} \) from the resonance, while the manifold \( n = 2 \) contains three dressed states. One of them is resonant, while the other two are detuned by \( \pm 2g \sqrt{N} \) from the resonance. Therefore, for \( N > 1 \) atoms in a cavity, detuning of the second photon asymptotically decreases to zero in the \( N \gg 1 \) limit.
Another feature of the extended JC model is the dependence of the position of the Rabi resonances on the number of interacting atoms. We have mentioned in Section 5.3 the conceptual difficulties related to the definition of the number of interacting atoms, when the individual atoms are loaded into the cavity via the traversing atomic beam. Clearly in this case the presence of an additional atom somewhere in the outskirts of the cavity mode changes the position of the Rabi resonance, and “driving on the lower Rabi resonance” becomes essentially an ill-defined concept. In the following Chapter we compare the photon blockade in the two level system with the photon blockade in the EIT–Kerr system, and we will elaborate further on this property.

8.4 Conclusion

In this Chapter we have presented the photon blockade exhibited by two level atoms. We have analysed the single atom case as well as the many atom case. The photon blockade effect deteriorates with the introduction of additional atoms into the cavity. The effective model derived for one atom is the effective two manifold model, describing the transitions between the ground state and the first excited states. It was found that in the limit of strong coupling and weak driving, this model describes successfully all of the significant features of the numerical simulations.
Chapter 9

Photon Blockade: A Comparison Between Two Schemes

Having reviewed the photon blockade exhibited by two level atoms in the previous Chapter, in this Chapter we compare the two schemes presented so far. We show that the EIT–Kerr scheme has much more flexibility and achieves better photon blockade than the extended JC scheme, even for comparable sets of parameters.

9.1 Introduction

In Chapter 8 we described the fundamental properties of two level systems in a high finesse optical cavity, from the perspective of the photon blockade effect. In this Chapter, we compare the Kerr nonlinearities of the EIT–Kerr and the extended JC schemes. We will show that the strength of the photon blockade effect as measured by $g^{(2)}(\tau = 0)$ is up to two orders of magnitude larger in the EIT–Kerr scheme than in the extended JC scheme. The advantage of the EIT–Kerr scheme lies in the destructive quantum interference of the probability amplitudes between the two transitions which would otherwise introduce a second excitation into the atom–cavity system.

The decoherence rate of the effective two level system can also be much smaller in the EIT–Kerr scheme. In other words, the EIT–Kerr scheme enables us to eliminate the quantum noise associated with the atomic dissipation and suppress the cavity dissipation. Finally, we show that the dependence of the photon blockade effect on the number of atoms $N$ involved in the interaction is qualitatively different for the two schemes. For the EIT–Kerr scheme, the photon blockade vanishes if $N > 1$, but can be restored by an appropriate choice of atomic detuning. In the JC scheme the strength of the nonlinearity disappears gradually with increasing $N$, and cannot be restored.

The EIT–Kerr scheme involves four level atoms in the configuration shown in Fig. 3.1. We write the Hamiltonian of this model for comparison with the Hamiltonian for the
extended JC model (8.2) as $\mathcal{H}^{EIT} = \mathcal{H}_0^{EIT} + \mathcal{H}_{pump} + \mathcal{H}_{int}^{EIT}$, with

$$\mathcal{H}_0^{EIT} = \hbar\omega_{\text{cav}} a^\dagger a + \hbar \sum_{j=1}^{N} (\omega_{21}\sigma_{22}^{(j)} + \omega_{34}\sigma_{33}^{(j)} + \omega_{41}\sigma_{44}^{(j)}) ,$$  \hspace{3cm} (9.1a)

$$\mathcal{H}_{pump}^{EIT} = i\hbar \mathcal{E}_p (ae^{-i\omega_L t} - a^\dagger e^{i\omega_L t}) ;$$ \hspace{3cm} (9.1b)

$$\mathcal{H}_{int}^{EIT}^{(j)} = i\hbar \sum_{j=1}^{N} \left[ g_1^{(j)} (a^\dagger \sigma_{12}^{(j)} - \sigma_{21}^{(j)} a) + (\Omega_c^* \sigma_{32}^{(j)} - \sigma_{23}^{(j)} \Omega_c^*) ight]$$

$$+ g_2^{(j)} (a^\dagger \sigma_{34}^{(j)} - \sigma_{43}^{(j)} a) ,$$ \hspace{3cm} (9.1c)

where $\sigma_{\mu\nu}^{(j)}$ are the operators describing the atomic coherence (for $\mu \neq \nu$) and energy level population (for $\mu = \nu$) for the $j$-th atom and $\Omega_c$ is the Rabi frequency of the (classical) coupling field. Again, dissipation can be included to give the following effective Hamiltonian

$$\mathcal{H}_{\text{eff}}^{EIT} = \mathcal{H}^{EIT} - i\hbar \kappa a^\dagger a - i\hbar \sum_{j=1}^{N} \left[ (\gamma_1 + \gamma_2) \sigma_{22}^{(j)} + \gamma_3 \sigma_{44}^{(j)} \right].$$ \hspace{3cm} (9.1d)

Again, this effective Hamiltonian has to be complemented by the appropriate collapse operators

$$C_a = \sqrt{\kappa} a, \hspace{3cm} (9.2a)$$

$$C_1 = \sqrt{\gamma_1} \sigma_{12}^{(j)}, \hspace{3cm} (9.2b)$$

$$C_2 = \sqrt{\gamma_2} \sigma_{32}^{(j)}, \hspace{3cm} (9.2c)$$

$$C_3 = \sqrt{\gamma_3} \sigma_{34}^{(j)}, \hspace{3cm} (9.2d)$$

associated with the gedanken measurement process. Note that we have assumed identical spontaneous decay rates for all atoms in the system.

### 9.2 Photon Blockade: Single–Atom Case

We now turn to investigating the photon blockade effect in the case of a single atom in a high finesse optical cavity. The atom is strongly coupled to the cavity field mode, and therefore the system has to be described in the dressed states (polariton) picture developed in Chapters 7 and 8.

It was shown in the previous Chapter that a single two level atom strongly coupled to the cavity mode behaves as a two state system\(^1\) when excited near one of its vacuum Rabi resonances. If the laser field is tuned to the lower vacuum Rabi resonance $\delta_{JC} = g$, then photon blockade occurs. After the first photon excites the system, the second transition is detuned and cannot occur before the first one deexcites.

\(^1\)In fact, as we have seen, as a two manifold system.
9.2. PHOTON BLOCKADE: SINGLE-ATOM CASE

A dressed states analysis for a single atom in the EIT–Kerr configuration has been carried out by Rebić et al. [102] (elaborated upon in Chapter 7) and Werner and Imamoglu [104]. An interesting investigation of the photon blockade regimes of implementation, also from the viewpoint of dressed states, has been given by Greentree et al. [107]. To recap the dressed states, there are three states in the $n = 1$ manifold, one of which is resonant with the cavity mode. The second manifold contains four states. Two of these are far detuned from the resonance and therefore their contribution to the system dynamics is negligible. The other two are also detuned, but lie closer to the resonance. They approach the other two states in the limit $g_2 / \kappa \gg 1$. It is the detuning of these two dressed states that determines the efficiency of this system for photon blockade.

We perform a Monte–Carlo simulation of the effective Hamiltonians (8.2d) and (9.1d) to compare the two schemes in the single-atom case. Figure 9.1 shows the second-order correlation function in the JC scheme as compared to two different cases in the EIT–Kerr

![Figure 9.1: Second order correlation functions in Jaynes-Cummings and EIT-Kerr schemes. Parameters for the JC scheme are $g = 6\kappa$, $\gamma = 0.1\kappa$, $\delta_{JC} = g$ and $\mathcal{E}_p = 0.1\kappa$. For the EIT–Kerr scheme, parameters are $\gamma_j = 0.1\kappa$, $g_j = 6\kappa$, $\Delta = 0.1\kappa$, $\delta = 0.2\kappa$, $\mathcal{E}_p = 0.1\kappa$ and $g_j / \Omega_c = 1$ (for EIT$_1$), $g_j / \Omega_c = 10$ (for EIT$_2$).]
scheme. In the extended JC scheme, the driving of the system was tuned to the lower dressed state of the \( n = 1 \) manifold. This gives significant antibunching, as measured by \( g^{(2)}(\tau = 0) \). It is also clear that this regime approximates the two level behaviour well (assuming weak driving). The other two curves simulate the EIT–Kerr system for the same range of parameters as in Chapter 6, but with different values of \( \Omega_c \), and therefore a different ratio \( g_1/\Omega_c \). They exhibit essentially the same values at the origin, which are now very close to zero. The fast modulation apparent in the correlation functions were explained previously - these are signatures of the other states in the first excited manifold. In the extended JC case, the modulation frequency is associated with the frequency difference between the upper and lower Rabi resonances, namely \( 2\delta_{JC} = 2g \).

In both of the EIT–Kerr cases it is the signature of the other two states in the first manifold, and the frequency is \( \epsilon_1 \approx \epsilon_3 \approx \sqrt{g_1^2 + \Omega_c^2} \), an expression that becomes exact for \( \delta \ll g_1, \Omega_c \).

We note the improvement in the correlation \( g^{(2)}(0) \) by a factor of 40 of the EIT–Kerr scheme over the extended JC scheme. Such a large difference cannot be simply attributed to the ability to eliminate the dissipation and retain the strength of the nonlinearity. The nonlinearities are roughly comparable (see Appendix B), while the detunings and linewidths of the higher lying states show that both models operate far off resonance for the transition from the first to the second manifold.

We now show that this striking difference arises as a consequence of quantum interference between the transition amplitudes for the dressed state transitions from the first to the second manifold. To prove this assertion, we calculate the transition amplitudes between the eigenstates in the first manifold \( |e_{1,2,3}\rangle \) (dark state) and the two eigenstates in the second manifold closest to the cavity resonance \( |e_{5,6}\rangle \). For a graphical depiction, see the dressed states ladder in Figure 7.1. These states have been already calculated in Chapter 7 to be

\[
|e_{1,3}\rangle = \frac{-(g_1/\Omega_c) |1, 1\rangle + i(\epsilon_{1,3}/\hbar\Omega_c) |0, 2\rangle - |0, 3\rangle}{\sqrt{1 + (g_1/\Omega_c)^2 + (\epsilon_{1,3}/\hbar\Omega_c)^2}},
\]

\[
|e_2\rangle = \frac{|1, 1\rangle + (g_1/\Omega_c)|0, 3\rangle}{\sqrt{1 + (g_1/\Omega_c)^2}},
\]

\[
|e_{5,6}\rangle = \alpha_{5,6}|2, 1\rangle + \beta_{5,6}|1, 2\rangle + \mu_{5,6}|1, 3\rangle + \nu_{5,6}|0, 4\rangle.
\]

The expressions for coefficients in \( |e_{5,6}\rangle \) are given in Appendix A, but there we have neglected the contribution of dissipation. To prove the existence of quantum interference, we must include dissipation in the analysis. In the absence of damping, the cancellation of the two transition amplitudes is straightforward and it is analogous to the cancellation of the contribution to the index of refraction that occurs between any two resonances. It is dissipative terms (i.e. the linewidths of these states) that are ultimately responsible for the breakdown of the two level (two manifold) behaviour, and it is these states that need
9.2. PHOTON BLOCKADE: SINGLE-ATOM CASE

to be investigated. This amounts to the analysis of the following Hamiltonian

$$\mathcal{H}_{\text{sys}} = \begin{pmatrix} -2i\hbar \kappa & -i\hbar g_1 \sqrt{2} h[\delta - i(\kappa + \gamma_1 + \gamma_2)] & 0 & 0 \\ ihg_1 \sqrt{2} h & -i\hbar \Omega_c & 0 & 0 \\ 0 & -i\hbar \kappa & -i\hbar g_2 & h(\Delta - i\gamma_3) \\ 0 & 0 & ihg_2 & h(\Delta - i\gamma_3) \end{pmatrix}, \quad (9.4)$$

valid for the second manifold states. The coefficients in the expansion (9.3c) are then easily calculated to be

$$\alpha_{5,6} = -i \frac{g_1 g_2 \sqrt{2}}{\Omega_c (\epsilon_{5,6} + 2i\kappa)} \left[ 1 - \frac{(\epsilon_{5,6} + i\kappa)(\epsilon_{5,6} - \Delta + i\gamma_3)}{g_2^2} \right] \nu_{5,6}, \quad (9.5a)$$

$$\beta_{5,6} = \frac{g_2}{\Omega_c} \left[ 1 - \frac{(\epsilon_{5,6} + i\kappa)(\epsilon_{5,6} - \Delta + i\gamma_3)}{g_2^2} \right] \nu_{5,6}, \quad (9.5b)$$

$$\mu_{5,6} = -i \frac{\epsilon_{5,6} - \Delta + i\gamma_3}{g_2} \nu_{5,6}, \quad (9.5c)$$

$$\nu_{5,6} = \left\{ \left( \frac{\epsilon_{5,6} - \Delta + 2i\gamma_3}{g_2} \right)^2 + \left( \frac{g_2}{\Omega_c} \right)^2 \left( 1 + \frac{2g_2^2}{\epsilon_{5,6} + \kappa^2} \right) \right\}^{-1/2} \left( \frac{\epsilon_{5,6}(\kappa + \gamma_3)}{g_2^2} \right)^2 + \left( 1 + \frac{\kappa \gamma_3 - \epsilon_{5,6}(\epsilon_{5,6} - \Delta)}{g_2^2} \right)^2 \right\}^{-1/2}. \quad (9.5d)$$

The states (9.3) have energies given by $E_{1,3} = \hbar(\omega_{\text{cav}} + \gamma_3)$, $E_2 = \hbar\omega_{\text{cav}}$ and $E_{5,6} = 2\hbar\omega_{\text{cav}} + \epsilon_{5,6}$.

The only nonvanishing contribution to the transition amplitudes comes from the term $\mathcal{H}_{\text{pump}} = i\hbar \mathcal{E}_p (a - a^\dagger)$, since it is the only term that connects the adjacent manifolds. Note that from now on we use the form for $\mathcal{H}_{\text{pump}}$ written in the frame rotating at frequency $\omega_L$. We obtain

$$A_{1,\pm} = \langle \epsilon_{5,6} | \mathcal{H}_{\text{pump}} | \epsilon_1 \rangle \frac{i \hbar \mathcal{E}_p}{\sqrt{1 + (g_1 / \Omega_c)^2 + (\epsilon_1 / \Omega_c)^2}} \left( \alpha_{5,6}^* \sqrt{2} \frac{g_1}{\Omega_c} + \frac{i \epsilon_1}{\Omega_c} \beta_{5,6}^* - \mu_{5,6}^* \right), \quad (9.6a)$$

$$A_{2,\pm} = \langle \epsilon_{5,6} | \mathcal{H}_{\text{pump}} | \epsilon_2 \rangle \frac{-i \hbar \mathcal{E}_p}{\sqrt{1 + (g_1 / \Omega_c)^2}} \left( \alpha_{5,6}^* \sqrt{2} + \frac{g_1}{\Omega_c} \mu_{5,6}^* \right), \quad (9.6b)$$

$$A_{3,\pm} = \langle \epsilon_{5,6} | \mathcal{H}_{\text{pump}} | \epsilon_3 \rangle \frac{i \hbar \mathcal{E}_p}{\sqrt{1 + (g_1 / \Omega_c)^2 + (\epsilon_3 / \Omega_c)^2}} \left( \alpha_{5,6}^* \sqrt{2} \frac{g_1}{\Omega_c} + \frac{i \epsilon_3}{\Omega_c} \beta_{5,6}^* - \mu_{5,6}^* \right). \quad (9.6c)$$
Taking the explicit expressions for $\alpha_{5,6}, \beta_{5,6}$ and $\mu_{5,6}$, we find

\begin{align}
A_{1,\pm} &= \frac{-\hbar \mathcal{E}_p}{\sqrt{1 + (g_1/\Omega_c)^2 + (\epsilon_1/\Omega_c)^2}} \left\{ \frac{g_2}{\Omega_c^2} \left( \epsilon_1 + \frac{2g_1^2}{\epsilon_{5,6} + 2i\kappa} \right) \right.
\bigg. + \frac{\epsilon_{5,6} - \Delta + i\gamma_3}{g_2} \left[ 1 + \frac{\epsilon_{5,6} + i\kappa}{g_2} \left( \epsilon_1 + \frac{2g_1^2}{\epsilon_{5,6} + 2i\kappa} \right) \right] \bigg. \right\}, \quad (9.7a) \\
A_{2,\pm} &= \hbar \mathcal{E}_p \left\{ \frac{(\epsilon_{5,6} - \Delta)(\epsilon_{5,6}^2 - 2\kappa^2) + 2\epsilon_{5,6}(g_2^2 + \kappa\gamma_3)}{(\epsilon_{5,6}^2 + 4\kappa^2) \sqrt{1 + (g_1/\Omega_c)^2}} \right.
\bigg. + \frac{i[2\kappa(\epsilon_{5,6}^2 - \epsilon_{5,6}\Delta - 2g_2^2 - \gamma_3(\epsilon_{5,6}^2 - 2\kappa^2)]}{(\epsilon_{5,6}^2 + 4\kappa^2) \sqrt{1 + (g_1/\Omega_c)^2}} \nu_{5,6}, \quad (9.7b) \\
A_{3,\pm} &= \frac{-\hbar \mathcal{E}_p}{\sqrt{1 + (g_1/\Omega_c)^2 + (\epsilon_3/\Omega_c)^2}} \left\{ \frac{g_2}{\Omega_c^2} \left( \epsilon_3 + \frac{2g_1^2}{\epsilon_{5,6} + 2i\kappa} \right) \right.
\bigg. + \frac{\epsilon_{5,6} - \Delta + i\gamma_3}{g_2} \left[ 1 + \frac{\epsilon_{5,6} + i\kappa}{g_2} \left( \epsilon_3 + \frac{2g_1^2}{\epsilon_{5,6} + 2i\kappa} \right) \right] \bigg. \right\}. \quad (9.7c)
\end{align}

These expressions are valid in the weak driving limit, where the contribution of $\mathcal{H}_{\text{pump}}$ to the dressed states structure can be neglected. The transition rate can now be calculated using Fermi’s Golden Rule. We use the approach outlined in Sakurai [108, Sec. 5.6] to obtain

\[ W_{1\rightarrow 2} = \frac{2\pi}{\hbar} \left| \sum_{j=1,2,3} (A_{j,+} + A_{j,-}) \right|^2, \quad (9.8) \]

where it is understood that this expression emerges from a more conventional expression for Fermi’s golden rule upon the integration with $\int dE_n \rho(E_n)$, where $\rho(E_n)$ is the density of final states. Figures 9.2 to 9.5 show the transition rate from the first to the second manifold as a function of the atomic detunings. The parameter $\Omega_c$ (and therefore $g_1/\Omega_c$) has been varied to reach the different operational regimes of the EIT–Kerr system. Although the transitions from all of the first manifold states have been taken into account, the dominant contribution obviously comes from the resonant state $|e_2\rangle$. The ratio $g_1/\Omega_c$ then controls whether the polariton is atom–like for small values of $\Omega_c$, or photon–like for large values of $\Omega_c$. Not surprisingly, the transition rates are larger in the atom–like limit, and the sensitivity on atomic detunings is more pronounced.

To further illustrate our findings we plot the correlation function for two different values of atomic detunings in Figure 9.6. Note that the dashed line now has superimposed modulations of two distinct frequencies, caused by the two different values of $\epsilon_1$ and $\epsilon_3$.

It is therefore possible to achieve near perfect cancellation with a suitable choice of atomic detunings. This is the central result of this Chapter; the benefits of the EIT–Kerr system over the extended JC system rely on two cases of quantum interference. The first constitutes the standard EIT and suppresses the atomic noise. The second reduces the complex dressed states structure to an effective two–level system. In Chapter 10, we will derive the absorption rate from the first to the second manifold using a full polariton picture developed there.
Having established the quantum interference effect, another difference in favour of the EIT–Kerr system can be established. In the extended JC model, one sees $g^{(2)}(0) < 1$ for $g^2 > \kappa \gamma$ [79]. Since the effect of quantum interference at the cavity resonance does not depend on the relative sizes of atom–field coupling constants and atomic and cavity decay rates, one may expect that the EIT–Kerr system would not have the same constraint for
Achieving the values $g^{(2)}(0) < 1$. Figure 9.7 shows that this is indeed the case; as soon as the atom-field interaction is nonzero, $g^{(2)}(0)$ starts decreasing asymptotically towards zero. This feature can be assigned entirely to the effect of quantum interference, since the analysis performed only in terms of detunings and linewidths cannot account for this feature.

We now discuss the possibility of controlling the lifetime of the effective excited state (9.3b). For larger values of $g_1/\Omega_c$, such as the one depicted in curve EIT$_2$ in Fig. 9.1,
Figure 9.4: Transition amplitudes from first to second manifold states for $\Omega_c = 6\kappa$ and $\Omega_c = 8\kappa$. The other parameters are the same as in Figure 9.2.

another decisive feature is noticed, namely the slow recovery of the correlation function from the origin. This is a consequence of the fact that the excitation is predominantly atomic in nature and not in the cavity field mode, thus reducing the contribution of the cavity decay to the total dissipation. In fact, the coherence time of the effective two level system can be adjusted to any prescribed value by changing the ratio $g_1/\Omega_c$. This is not surprising, since in the weak driving regime, the excited state of the effective two state system is the dark state $|e_2\rangle = (\Omega_c |1,1\rangle + g_1 |0,3\rangle)/\sqrt{g_1^2 + \Omega_c^2}$. Fleischhauer et al. [109]
have shown that if the system is in such a state, the round trip time (and therefore the effective cavity lifetime) is enhanced by a factor of $1 + (g_1 / \Omega_c)^2$. The physical explanation for this effect is that the polariton state $|e_2\rangle$ is dominated by its atomic contribution in the limit of large $g_1 / \Omega_c$. The storage of the photon in the atom increases the coherence time of the atom–cavity molecule. The storage of light in atoms using this procedure has been proposed by Fleischhauer and Lukin [110]. Their idea is to populate the col-
9.2. PHOTON BLOCKADE: SINGLE–ATOM CASE

Figure 9.6: Quantum interference at work – correlation function for two different values of detuning. The case of Figure 9.5, $\Omega_c = 12\kappa$ is chosen with $\Delta = 0$ and the values of detuning $\delta$ as indicated. The inset shows an enlargement of the origin.

Collective polariton state counterparts of $|e_2\rangle$ in an atomic vapour, and then adiabatically turn off the coupling field $\Omega_c$. These actions have the effect of projecting the polariton excitations onto their atomic component - thus effectively storing the excitation in the atomic system. If the coupling field is then turned back on adiabatically, the excitation is released back from the atoms in the form of a light pulse of essentially the same profile as the one that was stored. This effect has been experimentally demonstrated by Liu et al. [111] using sodium vapour cooled to 0.9 $\mu$K (just above the critical temperature for Bose–Einstein condensation). This experiment has also shown that the strict adiabatic condition for turning the coupling field off and back on is not required. Crude switching of the coupling laser produces the same effect. Phillips et al. [112] used hot rubidium vapour at temperatures $\sim$ 70–90 °C to demonstrate the same effect, thus showing that it is not limited to an extremely cold medium. Fleischhauer and Lukin [113] have also shown that in principle an arbitrary quantum state can be stored and retrieved using this technique, and Lukin et al. [114] proposed entangling atomic ensembles by trapping correlated photon states - this concept is also based on the idea of dark state polaritons.
CHAPTER 9. PHOTON BLOCKADE: A COMPARISON

Figure 9.7: Correlation function at zero delay, plotted against the single-atom cooperativity parameter $C = g^2/\kappa\gamma$, for the EIT-Kerr model. We vary atom-field coupling $g_1 = g_2 = g$, and keep $\gamma_j = \gamma = 0.1\kappa$, $\Omega_c = 2\kappa$ constant. Driving strength is $\mathcal{E}_p = 0.1\kappa$.

and EIT.

All of the described effects can be implemented using the EIT–Kerr system as well, with the additional benefit of giant Kerr nonlinearity, not exhibited by the three level $\Lambda$–system\(^2\). If the cavity supports two field modes (maybe two polarisations), the atomic transition $|1\rangle \rightarrow |4\rangle$ can be driven, and the resulting system will exhibit $\chi^{(2)}$ nonlinearity. J. Longdell [116] has shown, using the cascaded cavities approach [117, 118, 119], that this system can be configured to implement a photon doubler. The first cavity would generate single photon pulses [120], that will drive the second cavity containing the $\chi^{(2)}$ system. Matching the impedances of the two cavities in the right way can suppress one photon decay and enhance two photon decay of the second cavity, making this essentially a very effective parametric amplifier.

\(^2\)We note, however, the recent experimental demonstration of enhanced Kerr nonlinearity in a three level atomic system by Wang et al. [115]. The size of the nonlinearity is not so dramatic as in the EIT–Kerr system, and the $\Lambda$–system was driven slightly off-resonance. A similar idea to the one realised experimentally was proposed by Gheri et al. [58].


## 9.3 Photon Blockade: Many–Atom Case

Having analysed the single–atom case in some detail, we now turn to the analysis of the many–atom system. The notation of this Section will be slightly changed from that used so far in order to emphasize the different setups and differentiate between the distinct schemes.

Assume \( N \) atoms in the cavity. The \( n = 1 \) manifold, where a single photon excitation is shared among the \( N \) atoms, contains three energy levels \( \hbar(\omega_{\text{cav}} + \epsilon_i) \) whose energy shifts and widths, neglecting spontaneous emission, are approximately given by

\[
\epsilon_0^{n=1} \approx \frac{-i\kappa}{1 + g_{\text{eff}}}, \\
\epsilon_{\pm}^{n=1} \approx -\frac{i\kappa}{2} \pm \Omega_c \sqrt{1 + g_{\text{eff}} - (\kappa/2\Omega_c)^2},
\]

provided we assume that all atoms couple identically to the cavity mode. Here, we return to the many–atom notation of Chapter 3, and denote \( g_{\text{eff}} = g_2^2 / N \Omega_c^2 \). Note that the eigenstate associated with the eigenvalue \( \epsilon_0^{n=1} \) has a width which decreases with the number of atoms \( N \) while the splitting of the other two dressed states increases with \( N \). This is the linewidth narrowing predicted in Chapters 3 and 4, and by Grangier et al. [55] for this system, and independently predicted for a general cavity–EIT system by Müller et al. [66] and Lukin et al. [67]. The eigenstate corresponding to \( \epsilon_0^{n=1} \) is the \( N \)-atom extension of (9.3b)

\[
|\phi_0^{n=1}\rangle = \frac{1}{\sqrt{1 + g_{\text{eff}}}} \left[a_+ - \frac{g_1}{\Omega_c} \sum_{i=1}^N \sigma_{21}^{(i)}\right]|0\rangle.
\]

When \( N > 1 \), the \( n = 2 \) manifold contains six energy eigenstates provided the contribution from level 4 can be treated perturbatively. For \( g_2 = 0 \), one of these eigenstates,

\[
|\phi^{n=2}\rangle = \frac{1}{\sqrt{2(1 + g_{\text{eff}})}} \left[a_+ - \frac{g_1}{\Omega_c} \sum_{i=1}^N \sigma_{21}^{(i)}\right]^2 |0\rangle,
\]

has energy \( 2\hbar\omega_{\text{cav}} \), yielding a harmonic energy level diagram if probed by a weak external field. The state |\( \phi^{n=2}\rangle \) corresponds to two cavity–EIT trapping state excitations and has a linewidth which decreases with \( 1 + g_{\text{eff}} \). For \( g_2 \neq 0 \), this energy eigenstate experiences an ac–Stark shift. The level shift of |\( \phi^{n=2}\rangle \) caused by the perturbation due to level 4 is

\[
\epsilon^{n=2} = -2\eta \frac{g_{\text{eff}}}{(1 + g_{\text{eff}})^2},
\]

where the perturbing Hamiltonian is \( H_{\text{pert}} = -\hbar\mu a \sum_i \sigma_{33}^{(i)} \) in the limit \( |\Delta| \gg g_2, \gamma_3 \) and \( \eta = g_2^2 / \Delta \). The atom–cavity molecule has an anharmonic response to the driving field if the condition \( |\epsilon^{n=2}| > 2\kappa/(1 + g_{\text{eff}}) \) holds. We note that the (nonlinear) splitting of the eigenstate corresponding to \( \epsilon^{n=2} \) initially increases with the number of atoms \( N \) in the
cavity as would be expected from a nonlinear optical system. However, with increasing dispersion ($g_{\text{eff}} \approx 1$), this increase in the splittings saturates. Further increasing the dispersion ($g_{\text{eff}} \gg 1$) causes a decrease in the splitting of the state $e^{n=2}$ and its width, under the assumption that all $N$ atoms are positioned to have the maximum possible interaction.

It is instructive to calculate the exact energy eigenvalues $\hbar \epsilon_i$ for the $n = 2$ manifold eigenstates using an electric dipole interaction term for the $|3\rangle \leftrightarrow |4\rangle$ coupling. This gives
the Hamiltonian

\[
\begin{pmatrix}
-2i\kappa & 0 & -2ig_1 \\
0 & -i\kappa & -i\Omega_c \\
2ig_1 & i\Omega_c & \delta - i(\kappa + \gamma_1 + \gamma_2) \\
0 & 0 & 0 \\
0 & 0 & 0 \\
0 & 0 & 0 \\
2i\Omega_c & 0 & 0 \\
0 & 0 & 0 \\
0 & 0 & 0 \\
0 & 0 & 0 \\
\end{pmatrix},
\]

(9.13)

obtained by assuming a system state vector \(|\psi\rangle\) formed by a linear superposition of basis states generated by the following set of operators acting on the ground state: \(a^\dagger\), \(a^\dagger\sum_3 \sigma_3, a^\dagger\sum_3 \sigma_2, \sum_3 \sigma_3 \sigma_3 \sigma_3 \), \(\sum_3 \sigma_2 \sigma_2 \sigma_2 \), \(\sum_3 \sigma_2 \sigma_2 \sigma_2 \), \(\sigma_2 \sigma_2 \sigma_2 \). When \(g_2 = 0\) there are eigenvalues \(\Gamma_{12} = (\gamma_1 + \gamma_2) + i\delta\) and \(\Gamma_3 = \gamma_3 + i\Delta\), which imply states close to the resonance can be moved out of resonance when necessary. In addition, if \(\delta = 0\) other eigenvalues are given by \(\pm 2\Omega_c \sqrt{1 + g_{eff}}\) and \(\pm \Omega_c \sqrt{1 + g_{eff}}\) for \(N \sim N - 1\).

The eigenvalues are plotted in Fig. 9.8 as a function of \(g\) for three atoms. Although the energy shift for the near resonant state is larger for smaller values of \(g\) and can be increased further by reducing \(\delta\), the linewidth would be too large to observe the desired significant photon antibunching effects.

When the \(|3\rangle \leftrightarrow |4\rangle\) coupling can be treated perturbatively, the Hamiltonian and the associated eigenvalue problem reduces to

\[
\begin{pmatrix}
-2i\kappa & 0 & -2ig_1 \\
0 & \eta - i\kappa & -i\Omega_c \\
2ig_1 & i\Omega_c & \delta - i(\kappa + \gamma_1 + \gamma_2) \\
0 & 0 & 0 \\
0 & 0 & 0 \\
0 & 0 & 0 \\
2i\Omega_c & 0 & 0 \\
0 & 0 & 0 \\
0 & 0 & 0 \\
0 & 0 & 0 \\
\end{pmatrix},
\]

(9.14)

where \(\eta = g_2^2/\Delta\). In the special case of doubly resonant EIT (\(\delta = 0\)) with \(N > 1\), one of the eigenstates

\[
|\psi^{n=2}\rangle = \frac{1}{\sqrt{6}} \left[ a^\dagger + \frac{1}{N} \sum_{i \neq j} \sigma_2^{(i)} \sigma_2^{(j)} + \frac{1}{N} \sum_{i \neq j} \sigma_3^{(i)} \sigma_3^{(j)} \right] |0\rangle
\]

(9.15)

has energy \(2\hbar \omega_{cav}\) and a linewidth that is independent of \(g_{eff}\) (and therefore \(N\)). This state remains unshifted by \(g_2\) and has a nonzero transition rate from \(|\phi^{n=1}\rangle\) that decreases with \(g_{eff}\). Since \(|\phi^{n=2}\rangle\) and \(|\psi^{n=2}\rangle\) are not orthogonal we choose an orthogonal basis \(|\psi^{n=2}\rangle, |\varphi^{n=2}\rangle\) where \(|\varphi^{n=2}\rangle = \sqrt{2} |\psi^{n=2}\rangle - \sqrt{3/2} |\phi^{n=2}\rangle\) with a level shift \(3\varepsilon_n/2\). The transition rate from the \(n = 1\) manifold EIT state to this shifted eigenstate increases with the number of atoms \(N\), provided the conditions \(g_{eff} > 1\) and \(\eta > g_{eff} \kappa\) are satisfied.
Increasing the number of atoms therefore leads to a reduction in the magnitude of the photon–photon interaction. The presence of these two states implies that photon blockade is small for the special case of $\delta = 0$, when one would naively expect nonlinearity to be the strongest. Note also that the detrimental effect of the degenerate state $|\psi^{n=2}\rangle$ inhibiting the nonlinearity implies that the adiabatic elimination of the atomic degrees of freedom is not justified even in the low dispersion limit where it is normally assumed to be valid.

The photon blockade can be restored by increasing $\delta$. In particular, when $|\epsilon^{n=2}|$, $\kappa < |\delta| < \Omega_c \sqrt{1 + g_{\text{eff}}}$, the degeneracy in the $n = 2$ manifold is removed and the dominant contribution for the transition from the $n = 1$ to the $n = 2$ manifold is determined by $|\phi^{n=2}\rangle$. In this case the transition probability becomes independent of $g_{\text{eff}}$ for $g_{\text{eff}} \gg 1$ and approaches the single-atom result. Therefore photon blockade can be achieved in
9.3. PHOTON BLOCKADE: MANY-ATOM CASE

Figure 9.10: Restoration of photon blockade with increasing the atomic detuning in the EIT-Kerr system. Parameters are the same as in Figure 9.9, except that the detuning $\delta$ is increased for cases with two (solid) and three (dashed) atoms. The curve were obtained using the quantum trajectory approach, by averaging over 10 000 trajectories.

The main points of the previous discussion are illustrated in Figure 9.9 where we plot the values of $g^{(2)}(0)$ for up to three atoms. The correlation shows a dramatic increase as we move from $N = 1$ to $N = 2$ and $N = 3$. Figure 9.10 illustrates how the photon blockade can be improved by increasing $\delta$.

We now show that the EIT-Kerr and JC systems show vastly different behaviour when the number of atoms in the cavity becomes larger than one. The dressed states structure for the JC model is well-known (see Chapter 8 or the article by Kimble [78] in Berman [121]). In the case of two atoms, the $n = 1$ manifold contains two states detuned...
from the atomic resonance frequency by $g\sqrt{2}$. The $n = 2$ manifold contains three states, one of which is on the atomic resonance and the other two are detuned by $g\sqrt{6}$.

This behaviour is again illustrated in Figure 9.9, where we see gradual (but not dramatic) increase in $g^{(2)}(0)$ for up to three atoms. Comparing the extended JC scheme with the EIT–Kerr scheme, the latter clearly wins in the single–atom case, with $g^{(2)}(0)$ being roughly 40 times smaller than in the extended JC scheme. For $N > 1$, however, the sudden jump in $g^{(2)}(0)$ for the EIT–Kerr scheme can be avoided by appropriately increasing $\delta$. The increase in the JC scheme cannot be counteracted.

9.4 Conclusion

In conclusion, we have analyzed the Kerr nonlinearities of the EIT–Kerr and JC models, and compared the strength of the photon blockade effect. It was shown that destructive quantum interference between certain transition amplitudes yields much stronger antibunching for the EIT–Kerr scheme than for the extended JC scheme in the nonperturbative regime. In addition, the EIT–Kerr scheme offers the possibility of realizing an effective two level system with ultralong coherence times. The dependence of the nonlinearity on the number of intracavity atoms also exhibits marked differences in the two schemes. It was shown that photon blockade in the EIT–Kerr system can be restored for $N > 1$ by introducing an additional atomic detuning.
Part V

Photon Bunching in the EIT–Kerr System
Chapter 10

Full Polariton Solution

In this Chapter, we present a complete analytical solution for a single four level atom strongly coupled to a cavity field mode and driven by external coherent laser fields. The solution is presented in terms of dressed states and polariton operators. Driving and damping are treated separately, and it is shown how to treat quantum jump terms in the master equation.

10.1 Introduction

From the treatment presented so far in this thesis, it is obvious that analyzing the single atom EIT–Kerr system theoretically is not in general a straightforward task. In the bad cavity regime or the good cavity regime, approximate solutions are possible, based on the relative sizes of the atom–field coupling constant and the decay rates. In particular, it is possible to adiabatically eliminate either the cavity or the atomic degrees of freedom, respectively. In the strong coupling case, neither of these simplifications is possible. The ‘atom–field molecule’ must be truly regarded as a fundamental entity, which exhibits features that cannot be explained in terms of individual properties of its constituents. The natural basis for analysis of such a system is the polariton basis. In this Chapter we perform a polariton analysis of the strongly coupled atom–cavity system. Although we concentrate on an EIT–Kerr atomic configuration, the underlying method is general and could be applied to any strongly coupled system.

Polariton analysis has been used extensively of late to study the dynamics of EIT systems [109, 110, 113], but these analyses have concentrated on the semiclassical case of an atomic gas driven by laser light; in particular, on the dynamics of ‘slow’ light. Juzeliunas and Carmichael [122] have refined the analysis of the corresponding ‘slow polaritons’, and showed that it is possible to reverse a stopped polariton by reversing the control beam. However, none of the treatments so far have dealt with the coupled atom–cavity system.

We diagonalise the interaction Hamiltonian exactly to find a set of basis states for subsequent analysis. The driving term and damping terms are then expressed in terms of the new basis set, and the effective Hamiltonian in the polariton representation is found.
The Chapter concludes with an outlook on how the polariton method can be generalised and used in an arbitrary strongly coupled system.

10.2 Bare Model

We proceed by formulating the Hamiltonian in the bare state basis. Although this is essentially the same Hamiltonian as has been used so far, it is beneficial to present it again for completeness. The atom is assumed to be coupled to a single cavity field mode and this cavity is driven through one of its mirrors by a coherent laser field. The interaction picture Hamiltonian describing the system in the rotating wave and electric dipole approximations is $\mathcal{H} = \mathcal{H}_0 + \mathcal{H}_d$, where

$$
\begin{align*}
\mathcal{H}_0 &= \hbar \delta \sigma_{22} + \hbar \Delta \sigma_{44} + i\hbar g_1 (a_1^\dagger \sigma_{12} - \sigma_{21} a) \\
&\quad + i\hbar \Omega_c (\sigma_{23} - \sigma_{32}) + i\hbar g_2 (a_1^\dagger \sigma_{34} - \sigma_{43} a), \\
\mathcal{H}_d &= i\hbar \mathcal{E}_p (a - a^\dagger).
\end{align*}
$$

(10.1a) \hspace{1cm} (10.1b)

Here, $\sigma_{ij}$ represent atomic raising and lowering operators (for $i \neq j$), and energy level population operators (for $i = j$); $a$ ($a^\dagger$) is the cavity field creation (annihilation) operator. Detunings $\delta$ and $\Delta$ are defined from the relevant atomic energy levels; $g_{1,2}$ are atom-field coupling constants for the respective transitions, and $\Omega_c$ is the coupling field Rabi frequency. The cavity driving field is introduced through the parameter $\mathcal{E}_p$, given by

$$
\mathcal{E}_p = \sqrt{\frac{P \kappa T^2}{4\hbar \omega_{cav}}}.
$$

(10.2)

In this expression, $T$ is the cavity mirror transmission coefficient, $\kappa$ is the cavity decay rate, $P$ is the power output of the driving laser, and $\omega_{cav}$ is the frequency of the cavity mode. Damping due to cavity decay and spontaneous emission is discussed below.

Assume that the cavity mode subspace has been truncated at some finite size $N$. Together with the four atomic levels, these span a Hilbert space of dimension $4 \times N$. In the absence of driving (or in the limit where term (10.1b) becomes negligible), Hamiltonian (10.1a) takes a block-diagonal form, with $N$ blocks on the main diagonal. Each block represents a manifold of eigenstates associated with the appropriate term in the Fock expansion. The ground, first and second manifolds have been analyzed from the viewpoint of photon blockade in Parts III and IV (see also [102, 104, 107]), where this truncation approach was found to be very useful. Addition of the driving term (10.1b) significantly complicates the analysis. This term couples the different manifolds, and the Hamiltonian matrix loses its block-diagonal form. Therefore, it is not practical to perform a simple analytical diagonalization of the Hamiltonian (10.1), given the large size of the $4N$ by $4N$ matrix.

Dissipation can be added to the model by adding an anti–Hermitian term to the Hamiltonian (10.1). This term results from coupling to reservoir modes, and is obtained...
by tracing the system over these modes. In this approach we identify collapse operators, each of them corresponding to one decay channel [15]. In the EIT-Kerr case, there are the following four collapse operators

\[ C_1 = \sqrt{\gamma_1} \sigma_{12}, \quad C_2 = \sqrt{\gamma_2} \sigma_{32}, \]
\[ C_3 = \sqrt{\gamma_3} \sigma_{34}, \quad C_4 = \sqrt{\kappa} a, \]

(10.3)

where \( \gamma_k \) denote spontaneous emission rates into each of the decay channels, and \( \kappa \) denotes the cavity intensity decay rate. The effective non-Hermitian Hamiltonian takes the form

\[ \mathcal{H}_{\text{eff}} = \mathcal{H} - i\hbar \sum_{k=1}^{4} C_k^\dagger C_k, \]

(10.4)

\( \mathcal{H} \) being given by (10.1).

### 10.3 Dressed States Analysis

In this Section we solve the eigenvalue problem exactly for the Hamiltonian \( \mathcal{H}_0 \) given by Equation (10.1a), and obtain a basis for further calculations. In a strongly coupled system such as the one under analysis, dressed states [123] represent the natural basis for analysis, since the system under consideration should be viewed as an ‘atom–cavity molecule’, rather than the mere sum of its constituent parts (atom + cavity mode in this case). We have already remarked in Section 10.2 on the complexity of the problem of finding the exact (with coherent driving included) dressed states. Alsing et al. [124] succeeded in obtaining the exact solution for the case of a two level atom when the driving field is resonant with the cavity mode. They recognised that the eigenstates can be expressed as a direct product of field and atomic states, where the field states are displaced squeezed states, thus simplifying the calculation. The driven EIT–Kerr system does not have a solution for the field states with similarly convenient properties, so the method of Ref. [124] cannot be consistently applied. Instead we opt for an alternative approach which will be outlined in Section 10.4.

#### 10.3.1 Ground and First Manifold States

We use the notation \( |\text{number of photons in cavity mode, atomic energy level}\rangle \) to denote the bare states. The ground state is

\[ |e_{0}^{(0)}\rangle = |0, 1\rangle, \]

(10.5)

and has energy \( E_{0}^{(0)} = 0 \). Dressed state \( j \) belonging to the manifold \( n \) is denoted as \( |e_{j}^{(n)}\rangle \).

There are three first manifold states, one of them resonant with the cavity mode, the other two nonresonant

\[ |e_{0}^{(1)}\rangle = \alpha_{0}^{(1)}|1, 1\rangle + \mu_{0}^{(1)}|0, 3\rangle, \]
\[ |e_{\pm}^{(1)}\rangle = \alpha_{\pm}^{(1)}|1, 1\rangle + \beta_{\pm}^{(1)}|0, 2\rangle + \mu_{\pm}^{(1)}|0, 3\rangle, \]

(10.6a, 10.6b)
where the coefficients of the bare states are given by

\[
\alpha_0^{(1)} = \frac{1}{\sqrt{1 + (g_1/\Omega_c)^2}}, \quad \mu_0^{(1)} = \frac{g_1/\Omega_c}{\sqrt{1 + (g_1/\Omega_c)^2}} \quad (10.7a)
\]

\[
\alpha_\pm^{(1)} = \frac{g_1/\Omega_c}{\sqrt{1 + (g_1/\Omega_c)^2 + (\epsilon_\pm^{(1)}/\Omega_c)^2}}, \\
\beta_\pm^{(1)} = \frac{i\epsilon_\pm^{(1)}/\Omega_c}{\sqrt{1 + (g_1/\Omega_c)^2 + (\epsilon_\pm^{(1)}/\Omega_c)^2}}, \\
\mu_\pm^{(1)} = \frac{1}{\sqrt{1 + (g_1/\Omega_c)^2 + (\epsilon_\pm^{(1)}/\Omega_c)^2}}. \quad (10.7b)
\]

The energies of these eigenstates are given by \( E_j^{(1)} = \hbar(\omega_{cav} + \epsilon_j^{(1)}) \), where

\[
\epsilon_0^{(1)} = 0, \quad (10.8a)
\]

\[
\epsilon_\pm^{(1)} = \frac{\delta}{2} \pm \sqrt{\frac{\delta}{2}^2 + \Omega_c^2 \left(1 + \frac{g_1^2}{\Omega_c^2}\right)} \quad (10.8b)
\]

Note that \( \sum_{i=\pm0} \epsilon_i^{(1)} = \delta \), reflecting the fact that the cavity mode can be detuned from a one–photon excitation of the atom.

### 10.3.2 Second and Higher Manifold States

The second and higher manifold states can be written in a generic form,

\[
|\epsilon_k^{(n)}\rangle = \alpha_k^{(n)}|n, 1\rangle + \beta_k^{(n)}|n - 1, 2\rangle \\
+ \mu_k^{(n)}|n - 1, 3\rangle + \nu_k^{(n)}|n - 2, 4\rangle, \quad (10.9)
\]

with \( n \geq 2 \) being the manifold label.

There are four states in each manifold, with energies \( E_k^{(n)} = \hbar(n\omega_{cav} + \epsilon_k^{(n)}) \). The
coefficients of these states are

\[
\alpha_k^{(n)} = -i \frac{g_1 g_2 \sqrt{n(n-1)}}{\epsilon_k^{(n)} \Omega_c} \left[ 1 - \frac{\epsilon_k^{(n)} (\epsilon_k^{(n)} - \Delta)}{g_2^2(n-1)} \right] \nu_k^{(n)},
\]
(10.10a)

\[
\beta_k^{(n)} = \frac{g_2 \sqrt{n-1}}{\Omega_c} \left[ 1 - \frac{\epsilon_k^{(n)} (\epsilon_k^{(n)} - \Delta)}{g_2^2(n-1)} \right] \nu_k^{(n)},
\]
(10.10b)

\[
\mu_k^{(n)} = -i \frac{\epsilon_k^{(n)} - \Delta}{g_2 \sqrt{n-1}} \nu_k^{(n)},
\]
(10.10c)

\[
\nu_k^{(n)} = \left\{ 1 + \left( \frac{\epsilon_k^{(n)} - \Delta}{g_2 \sqrt{n-1}} \right)^2 + \left( \frac{g_2 \sqrt{n-1}}{\Omega_c} \right)^2 \right. \\
\left. \times \left[ 1 + n \left( \frac{g_1}{\epsilon_k^{(n)}} \right)^2 \right] \left[ 1 - \frac{\epsilon_k^{(n)} (\epsilon_k^{(n)} - \Delta)}{g_2^2(n-1)} \right]^2 \right\}^{-1/2}.
\]
(10.10d)

The exact energies of the four states within a given manifold are found to be, in increasing order,

\[
\epsilon_{1,2}^{(n)} = \frac{C}{4} - \frac{1}{2} \sqrt{\frac{C^2}{4} - \frac{2A}{3} + D}
\]
\[
\mp \frac{1}{2} \sqrt{\frac{C^2}{4} - \frac{4A}{3} - D + \frac{2B + AC + C^2/4}{\sqrt{C^2/4 - 2A/3 + D}}},
\]
(10.11a)

\[
\epsilon_{3,4}^{(n)} = \frac{C}{4} + \frac{1}{2} \sqrt{\frac{C^2}{4} - \frac{2A}{3} + D}
\]
\[
\mp \frac{1}{2} \sqrt{\frac{C^2}{4} - \frac{4A}{3} - D - \frac{2B + AC + C^2/4}{\sqrt{C^2/4 - 2A/3 + D}}},
\]
(10.11b)

where the following abbreviations have been used:

\[
A = \Delta \delta - g_1^2 n - g_2^2(n-1) - \Omega_c^2,
\]
(10.12a)

\[
B = \Delta [g_1^2 n + \Omega_c^2] + \delta g_2^2(n-1),
\]
(10.12b)

\[
C = \Delta + \delta,
\]
(10.12c)

\[
G^2 = (g_1 g_2)^2 n(n-1),
\]
(10.12d)

\[
X_1 = 2A^3 + 9A(BC - G^2) + 27(B^2 - C^2G^2),
\]
(10.12e)

\[
X_2 = A^2 + 3BC + 12G^2,
\]
(10.12f)

\[
X = \sqrt{X_1 + \sqrt{X_1^2 - 4X_2^3}},
\]
(10.12g)

\[
Y = X_2/X,
\]
(10.12h)

\[
D = \frac{(2^{1/3}Y + 2^{-1/3}X)/3}{2}. \]
(10.12i)

Note that for the \( n \)-th \( (n \geq 2) \) manifold, \( \sum_{i=1}^{4} \epsilon_i^{(n)} = \Delta + \delta \), which is the two-photon detuning of the atom from the cavity resonance. These equations are the exact eigenstates
of Hamiltonian (10.1a), and can be rewritten using the polariton operators as

\[ H_0 = \hbar \epsilon_0^{(1)} \hat{p}_-^{(1)} \hat{p}_-^{(1)} + \hbar \epsilon_0^{(2)} \hat{p}_+^{(1)} \hat{p}_+^{(1)} + \sum_{n=2}^{4} \sum_{j=1}^{4} \hbar \epsilon_j^{(n)} \hat{p}_{k_j}^{(n)} \hat{p}_{k_j}^{(n)}, \]  

(10.13)

where the polariton operators are defined as \( p_{ij}^{(n)} = |e_i^{(n-1)}\rangle \langle e_j^{(n)}| \). Index \( k \) in the second row of Equation (10.13) is a dummy index, since \( p_{k_j}^{(n)} = |e_j^{(n)}\rangle \langle e_j^{(n)}| \). Polariton operators \( p_{0, \pm}^{(1)} = |e_0^{(0)}\rangle \langle e_0^{(1)}| \) can be written in a relatively simple form. These expressions, as well as a short discussion on the statistical properties of polaritons, were given in Section 7.3.

The Hamiltonian (10.13) is written in a frame rotating at the cavity frequency \( \omega_{\text{cav}} \). The energy level structure obtained has a form as shown in Figure 10.1: dressed states with the same detuning relative to the cavity resonance will be formally degenerate. There are only two degenerate eigenstates: ground state \( |\epsilon_0^{(0)}\rangle \) and the first manifold state \( |\epsilon_0^{(1)}\rangle \).

It can be expected that driving this transition coherently yields dynamic Stark splitting. This indeed happens, and will be elaborated upon in Section 11.2, but next we turn to the problem of how to include driving and damping into our polariton model.

### 10.4 Driving and Damping Terms

The simplest way to include off-diagonal terms (as associated with driving and damping) into the effective Hamiltonian, written in the basis of states calculated in Section 10.3, is to express the atomic and field operators in terms of operators describing the transitions between these states. Here again, we make a distinction between first and higher manifolds, a distinction imposed by the very different structure of these two groups of manifolds.
The first manifold dressed states can be expressed in terms of bare states as \( \mathbf{b}_1 = M_1 \mathbf{d}_1 \), where \( \mathbf{b}_1 = ([1, 1], [0, 2], [0, 3])^T \) and \( \mathbf{d}_1 = ([e_1^{(1)}], [e_0^{(1)}], [e_+^{(1)}])^T \). The transformation matrix is

\[
M_1 = \begin{pmatrix}
-\frac{g_1/\Omega_c}{N_+} & \frac{1}{N_0} & -\frac{g_1/\Omega_c}{N_+} \\
\frac{i e_0^{(1)}/\Omega_c}{N_+} & 0 & \frac{i e_+^{(1)}/\Omega_c}{N_+} \\
\frac{1}{N_+} & \frac{g_1/\Omega_c}{N_0} & \frac{1}{N_+}
\end{pmatrix}, \tag{10.14}
\]

where

\[
N_0 = \sqrt{1 + (g_1/\Omega_c)^2}, \tag{10.15a}
\]
\[
N_\pm = \sqrt{1 + (\epsilon_\pm/\Omega_c)^2 + (g_1/\Omega_c)^2}. \tag{10.15b}
\]

Note that the Equations (10.14) and (10.15) are identical to the Equations (7.11) and (7.12) of Section 7.4. Similarly, we can write for the higher manifolds \( \mathbf{b}_n = M_n \mathbf{d}_n \), \( n \geq 2 \), where \( \mathbf{b}_n = ([n, 1], [n - 1, 2], [n - 1, 3], [n - 2, 4])^T \) and \( \mathbf{d}_n = ([e_1^{(n)}], [e_2^{(n)}], [e_3^{(n)}], [e_4^{(n)}])^T \), and

\[
M_n = \begin{pmatrix}
\alpha_1^{(n)*} & \alpha_2^{(n)*} & \alpha_3^{(n)*} & \alpha_4^{(n)*} \\
\beta_1^{(n)*} & \beta_2^{(n)*} & \beta_3^{(n)*} & \beta_4^{(n)*} \\
\mu_1^{(n)*} & \mu_2^{(n)*} & \mu_3^{(n)*} & \mu_4^{(n)*} \\
\nu_1^{(n)*} & \nu_2^{(n)*} & \nu_3^{(n)*} & \nu_4^{(n)*}
\end{pmatrix}, \tag{10.16}
\]

with the coefficients given by Eqs. (10.10). These expressions provide all of the information needed for the subsequent calculations.

### 10.4.1 External Driving

Strongly coupled systems are very sensitive to the number of photons. In fact, the most interesting regimes include one or a few photons. In the system under investigation, the effect of photon blockade occurs when the dynamics is limited to the exchange of excitation between the ground state and the first manifold. It is therefore natural to express the field annihilation operator \( a \) in terms of the transitions it produces between two adjacent manifolds. In general, deexcitation from manifold \( n \) to manifold \( n - 1 \) occurs via the operator \( a^{(n)} \), expressed in terms of the bare states as

\[
a^{(1)} = |0, 1\rangle\langle 1, 1| \tag{10.17a}
\]
\[
a^{(2)} = \sqrt{2}|1, 1\rangle\langle 2, 1| + |0, 2\rangle\langle 1, 2| + |0, 3\rangle\langle 1, 3| \tag{10.17b}
\]
\[
a^{(n)} = \sqrt{n}|n - 1, 1\rangle\langle n, 1| + \sqrt{n - 1}(|n - 2, 2\rangle\langle n - 1, 2| + |n - 2, 3\rangle\langle n - 1, 3|) + \sqrt{n - 2}|n - 3, 4\rangle\langle n - 2, 4|, \tag{10.17c}
\]

and the full annihilation operator would then be given by a sum over all manifolds, \( a = \sum_{n=1}^{\infty} a^{(n)} \). For the transition from the first excited state to the ground state we
obtain

\[ E_p \alpha^{(1)} = \Omega_{-}^{(1,0)} p_-^{(1)} + \Omega_0^{(1,0)} p_0^{(1)} + \Omega_+^{(1,0)} p_+^{(1)}, \tag{10.18a} \]

with the polariton operators defined by

\[ |e_0^{(0)}\rangle = p_j^{(1)} |e_j^{(1)}\rangle, \quad j = 0, \pm. \]

The effective Rabi frequencies \( \Omega_j^{(0,1)} \) can be calculated from the matrix (10.14) as

\[ \Omega_0^{(1,0)} = \frac{E_p}{\sqrt{1 + (g_1/\Omega_c)^2}}, \tag{10.18b} \]

\[ \Omega_\pm^{(1,0)} = -\frac{E_p g_1}{\sqrt{g_1^2 + \Omega_c^2 + (\epsilon_j^{(1)})^2}} \tag{10.18c} \]

The three terms in the expansion (10.18a) correspond to the three transitions between the ground state and the three states excited by a single photon. Each transition has an associated effective Rabi frequency \( \Omega_j^{(1,0)} \). Note that the negative sign in (10.18c) means the driving of the off-resonant states is out of phase with the driving of the resonant state (see Figure 10.2 (a)).

There are twelve possible transitions between the first and the second manifolds, driven with the effective Rabi frequencies \( \Omega_{ij}^{(2,1)} \), where

\[ \Omega_{ij}^{(2,1)} = E_p \left[ \sqrt{2} \alpha_i^{(1)*} \alpha_j^{(2)} + \beta_i^{(1)*} \beta_j^{(2)} + \mu_i^{(1)*} \mu_j^{(2)} \right], \tag{10.19} \]

with \( i = 0, \pm; j = 1, \ldots, 4 \) and it follows from the Equation (10.6a) that \( \beta_0^{(1)} = 0 \) (see Figure 10.2 (b)). Since the second and subsequent manifolds have four states each, there are sixteen transitions between the adjacent manifolds, with effective Rabi frequencies of driving

\[ \Omega_{ij}^{(n,n-1)} = E_p \left[ \sqrt{n} \alpha_i^{(n-1)*} \alpha_j^{(n)} + \sqrt{n-1} \left( \beta_i^{(n-1)*} \beta_j^{(n)} + \mu_i^{(n-1)*} \mu_j^{(n)} \right) \right], \tag{10.20} \]

with \( n > 2 \) and \( i, j = 1, \ldots, 4 \) (see Figure 10.2 (c)). The coefficients in this expression are given in Eqs (10.10).

Note that the Rabi frequencies \( \Omega_{ij}^{(n,n-1)} \) can also be obtained from

\[ i \hbar \Omega_{ij}^{(n,n-1)} = \langle e_i^{(n-1)} | \mathcal{H}_d | e_j^{(n)} \rangle, \]

with \( \mathcal{H}_d \) given by Equation (10.1b). However, the expansion of the operator \( a \) in terms of contributions to different transitions, Equation (10.17), offers a clearer physical picture of the processes involved in the dynamics. The driving Hamiltonian can therefore be written
Figure 10.2: Transitions between the polaritons in adjacent manifolds. The cavity resonance is located at the center of each manifold. Figures represent: (a) Transitions between the ground state and first manifold states; (b) Transitions between first and second manifold states; (c) Transitions between polaritons in manifolds \((n-1)\) and \(n\) for \(n \geq 3\).

in terms of the polariton operators as

\[
\mathcal{H}_d = i\hbar \mathcal{E}_p (a - a^\dagger) \\
= i\hbar \sum_{i=\pm,0} \Omega_i^{(1,0)} \left( p_i^{(1)} - p_i^{(1)\dagger} \right) \\
+ i\hbar \sum_{i=\pm,0} \sum_{j=1}^4 \Omega_{ij}^{(2,1)} \left( p_{ij}^{(2)} - p_{ij}^{(2)\dagger} \right) \\
+ i\hbar \sum_{n=3,1} \sum_{i,j=1}^4 \Omega_{ij}^{(n,n-1)} \left( p_{ij}^{(n)} - p_{ij}^{(n)\dagger} \right) .
\]

Expression (10.21) is the expansion of the driving Hamiltonian in terms of the transitions that are permitted to occur between any two dressed states, as shown in Figure 10.2. The obvious advantage of this expansion over the original form of driving is in the strong coupling/low photon number regime. In this regime, the expansion (10.21) can be truncated at the order justified by the problem, while still retaining all (but not more!) of the relevant contributions from the external coherent driving. We will illustrate this assertion in Section 11.2.
10.4.2 Damping by Reservoir Modes

The remaining part of the dynamics to be expressed in the polariton representation is damping by the reservoir modes. In Section 10.2 it was explained how the damping enters into the effective Hamiltonian; in particular after a trace has been performed over the reservoir variables. The resulting Hamiltonian operator is anti-Hermitian and has the form

\[ H_{\text{res}} = -i\hbar k a^\dagger a - i\hbar (\gamma_1 + \gamma_2) \sigma_{22} - i\hbar \gamma_3 \sigma_{44}. \]  

(10.22)

We follow the reasoning of the previous Section and expand the relevant field and atomic operators in terms of the contributions from the individual manifolds:

\[ a^\dagger a = \sum_{n=1}^{\infty} (a^{(n)}^\dagger a^{(n)}) \]  

(10.23a)

\[ = |1, 1\rangle\langle 1, 1| + \sum_{n=2}^{\infty} \left[ |n, 1\rangle\langle n, 1| + (n - 1) |n - 1, 2\rangle\langle n - 1, 2| \right. \]

\[ + |n - 1, 3\rangle\langle n - 1, 3| + (n - 2) |n - 2, 4\rangle\langle n - 2, 4| \]  

diagonal elements of \( H_{\text{res}} \). The operator \( H_{\text{res}} \) clearly takes a block-diagonal form in the dressed state representation, as the operator expansion (10.23) includes terms containing every possible dressed level within a given manifold. The diagonal terms correspond to damping of the dressed states due to their decay straight into the reservoir. Off-diagonal terms couple two different dressed levels in a given manifold. This coupling arises due to couplings of both levels to the same reservoir. If each of these diagonal blocks is again diagonalised, in the presence of damping we get shifts appearing on each level. The complex eigenvalues add their real part to the energy shift and the imaginary part becomes the damping rate. Energies and damping rates calculated in this manner will coincide with the experimentally observed ones (in the absence of driving). It was pointed out by Harris [125] and Imamoglu [126] that these cross terms can be essential in creating destructive interference between the transition amplitudes of the appropriate transitions (see also Li and Xiao [96]).

The contribution of the off-diagonal terms to the eigenenergies and damping rates (diagonal elements of \( H_{\text{res}} \)) is very small, and we will ignore their contribution to the eigenvalues in the rest of this Chapter for simplicity, though we leave them in a general expression for the damping Hamiltonian.
We write the damping Hamiltonian in a form that emphasizes the diagonal and off-diagonal contributions,

\[ \mathcal{H}_{\text{res}} = -i\hbar \sum_{i=\pm,0} \Gamma_i^{(1)} p_i^{(1)\dagger} p_i^{(1)} - i\hbar \sum_{j \neq k=\pm,0} \Gamma_{jk}^{(1)} p_j^{(1)\dagger} p_k^{(1)} \]

\[ -i\hbar \sum_{n=2}^{\infty} \sum_{j=1}^{4} \Gamma_{jj}^{(n)} p_{ij}^{(n)\dagger} p_{ij}^{(n)} - i\hbar \sum_{n=2}^{\infty} \sum_{j \neq k} \Gamma_{jk}^{(n)} p_{ij}^{(n)\dagger} p_{ik}^{(n)} . \]  

(10.24a)

where

\[ \Gamma_{jk}^{(n)} = n \kappa \alpha_j^{(n)\ast} \alpha_k^{(n)} + [(n - 1) \kappa + \gamma_1 + \gamma_2] \rho_j^{(n)\ast} \rho_k^{(n)} \]

\[ + (n - 1) \kappa \mu_j^{(n)\ast} \mu_k^{(n)} + [(n - 2) \kappa + \gamma_3] \nu_j^{(n)\ast} \nu_k^{(n)} , \]  

(10.24b)

For each manifold \((n)\), matrix \(\Gamma_{jk}^{(n)}\) is a positive definite matrix, so we can write [127, 128]

\[ \Gamma_{jk}^{(n)} = \cos \theta_{jk} \sqrt{\Gamma_{jj}^{(n)} \Gamma_{kk}^{(n)}}, \]  

(10.25a)

\[ \cos \theta_{jk} = \frac{\mu_j \cdot \mu_k}{|\mu_j| |\mu_k|}, \]  

(10.25b)

where \(\mu_{j,k}\) can be thought of as the effective dipole moments of the transitions between dressed states contributing the off-diagonal terms. These can be calculated from the dipole moments associated with the atomic transitions. Assume that all of the atomic dipole moments are the same and equal to \(\mu_{ad}\). If a state \(j\) from the manifold \(n\) couples to the state \(l\) from the manifold \(n - 1\), then the effective dipole moment for that transition can be calculated as \(\mu_j \sim (\epsilon_j^{(n)} |\mu_{ad}| \epsilon_i^{(n-1)})\). Equation (10.25) assumes that both states \(j\) and \(k\) couple to the same lower state \(l\).

Furthermore, we note that the diagonal matrix elements belonging to the first manifold can be written in a simple closed form as

\[ \Gamma_0^{(1)} = \frac{\kappa}{1 + (g_1/\Omega_c)^2}, \]  

(10.26a)

\[ \Gamma_{\pm}^{(1)} = \frac{\kappa g_1^2 + (\gamma_1 + \gamma_2) \left(\epsilon_{\pm}^{(1)}\right)^2}{g_1^2 + \Omega_c^2 + \left(\epsilon_{\pm}^{(1)}\right)^2}. \]  

(10.26b)

As before, all of the \(\Gamma\)’s could have also been calculated from \(-i\hbar \Gamma_{jj}^{(n)} = \langle \epsilon_j^{(n)} |\mathcal{H}_{\text{res}}| \epsilon_j^{(n)} \rangle\) and \(-i\hbar \Gamma_{jk}^{(n)} = \langle \epsilon_j^{(n)} |\mathcal{H}_{\text{res}}| \epsilon_k^{(n)} \rangle\), but again, the outlined procedure offers a deeper physical insight.

### 10.4.3 Quantum Jumps

The effective Hamiltonian (10.4) describes the time evolution of the quantum system between successive jumps. The effect of quantum jumps is not included, and the proper
way to include these is the subject of the quantum trajectories approach [15]. Here, we briefly describe the transformation of collapses into the dressed state basis.

Quantum jumps are included in the master equation for the time evolution of the density matrix $\rho$ via terms of the form $C_j\rho C_j^\dagger$, where $C_j$ denotes a collapse operator from the set (10.3). Each of the collapse operators can then be expressed in terms of polariton operators, and a new set of collapse operators $S_{ij}^{(n)} = \sqrt{\Gamma_{jj}^{(n)}} p_{ij}^{(n)}$ can be obtained. Operator $a$, as given by Equation (10.17), can be substituted directly into term $apa^\dagger$, and the resulting jump terms in the dressed states basis obtained. The remaining collapse operators can be transformed into the new basis according to the correspondences

$$\sigma_{12} = \sum_n |n, 1\rangle \langle n, 2|,$$

$$\sigma_{32} = \sum_n |n, 3\rangle \langle n, 2|,$$

$$\sigma_{34} = \sum_n |n, 3\rangle \langle n, 4|.$$  

Note that the effective master equation resulting from the polariton expansion will contain cross terms in the collapse operators, giving damping terms of the form

$$\Gamma_{jk}^{(n)} \left(2S_{ij}^{(n)} \rho S_{ik}^{(n)\dagger} - S_{ik}^{(n)\dagger} S_{ij}^{(n)} \rho - \rho S_{ik}^{(n)\dagger} S_{ij}^{(n)}\right).$$

These cross terms have a very important role in modifying the emission rate from the polariton states to the reservoir. Such terms have been studied and well understood for the case of the modification of spontaneous emission in multilevel atoms [129, 130]. An example of the effective model, including jump terms, is given in Appendix D, related to the model developed in Chapter 12.

### 10.5 Quantum Interference Involving Dressed States

In this Section, we use the polariton formalism developed in this Chapter to show how the effect of quantum interference, analysed in Section 9.2, can be restated in terms of the dressed states.

We calculate the absorption rate for transitions from the resonant eigenstate in the first manifold $|e_0^{(1)}\rangle$ (dark state) to the two eigenstates in the second manifold closest to the cavity resonance $|e_{\pm}^{(2)}\rangle$. The effective Rabi frequencies and damping rates have been calculated exactly. Here, we present the expressions approximated from the exact ones under the condition $\delta, \Delta \ll g_j, \Omega_c$, an approximation that is valid for all of the single atom simulations presented in this thesis. In this approximation, effective Rabi frequencies and damping rates for the transitions $|e_0^{(1)}\rangle \rightarrow |e_{\pm}^{(2)}\rangle$ are calculated to give

$$\Omega_{\pm} \approx \pm \frac{E_p}{\sqrt{2 g_2}} \frac{g_1^2 + \Omega_c^2 - g_2^2}{\sqrt{g_1^2 + \Omega_c^2}},$$

$$\Gamma_+^{(2)} \approx \Gamma_-^{(2)} = \Gamma \approx \frac{\kappa + \gamma_1 + \gamma_2 + \gamma_3}{2}. $$
Additional damping $\Gamma_r$ arises from the coupling of the dressed states due to their interaction with the same reservoir. Applying the same approximation as above, the additional damping coefficient is found to be $\Gamma_r \approx \Gamma/\sqrt{2}$. With these results, the absorption rate can be calculated using standard methods [37, 125] to give

$$w_{ab} = 4|\Omega_{\pm}|^2 \frac{\Delta_p^2 (\Gamma + \Gamma_r)}{\Delta_p^2 (\Gamma^2 + \Gamma_r^2) + \epsilon^2}.$$  \hspace{1cm} (10.30)

In this expression, detuning $\Delta_p = \omega_p - \omega_{\text{cav}}$ denotes the detuning of the driving (probe) laser from the cavity resonance, and $\epsilon^{(2)}_+ = -\epsilon^{(2)}_- = \epsilon$ is the detuning of the second manifold states from the cavity resonance frequency. It should be emphasised that the validity of this conclusion is not limited to the particular approximation on the atomic detunings used to derive Equation (10.30). The absorption rate vanishes for arbitrary value of detunings, only in this approximation it reduces to the simple form (10.30), enabling direct comparison with the EIT–type absorption rate.

Note that the polariton approach simplifies considerably the analysis of the quantum interference effect, compared to the techniques and results of Section 9.2.

### 10.6 Conclusion

In this Chapter, we have presented an exact solution to the eigenvalue problem of the Hamiltonian for a four level atom strongly coupled to a cavity mode. The regime of strong coupling CQED presents a difficult problem for analytical calculation, as well as for the understanding of the physics involved. We have shown that consistent application of the polariton approach can offer a significant insight and even enable a relatively simple analytical treatment of the physical problem. In particular, the problem of an externally driven atom/quantum field system has been reduced to the problem of composite excitations, transitions between which are effectively driven by classical fields of Rabi frequencies $\Omega_{ij}^{(n,n-1)}$, which were calculated exactly.

The polariton approach can be interpreted as a change of basis in Hilbert space. It is obvious that such a change can in general simplify the analysis of the problem. The reason for this is that in the dressed state basis the number of degrees of freedom can be significantly reduced, compared to the treatment in terms of the bare states. For example, if the atom has $N_a$ levels (degrees of freedom), and the quantum field mode can be safely truncated at some number $N_c$, the problem in the bare state basis has a dimension of at least $N_a \times N_c$. In the dressed state basis, we can identify which $N_p$ dressed states (and associated polaritons) participate in the dynamics, effectively reducing the dimension of the problem to $N_p \leq N_a \times N_c$. In other words, from all of the (infinite number of) dimensions of Hilbert space, the polariton approach lets us pinpoint those few dimensions that are predominantly involved in the system dynamics.

Of course, this approach does not guarantee that the reduced problem will be analytically solvable. There are some general limits on solvability in the dressed state basis.
For example, if the number of atomic levels $N_a > 4$, the diagonalisation of the interaction Hamiltonian is impossible \textit{in principle}, except perhaps in some special cases, since $N_a$ determines the order of the polynomial of the eigenvalue problem. Also, the size of the reduced problem $N_p$ can still be impractically large. While nothing much can be done about the first problem, for the second one, we foresee ways to simplify the involved numerics. In particular, the coupled amplitudes approach and the effective master equation look promising. The following Chapter on the dynamic Stark splitting provides a simple but extremely successful example of this method.
Chapter 11

Dynamic Stark Splitting

In this Chapter we present an analysis of the effective two level system for the EIT–Kerr configuration, using the polariton approach developed in Chapter 10. We find the effect of dynamic Stark splitting of dressed states and explore the similarities with the corresponding two level system formed by a two level atom in an optical cavity.

11.1 Introduction

The formalism developed in Chapter 10 is very powerful for small photon number. This regime usually coincides with the weak driving, and the simplest case for consideration is the case when only two dressed states suffice to explain the main features of the system dynamics. The polariton approach enables us to incorporate a driving field in the analysis through an effective (classical) Rabi frequency.

In this Chapter we apply the polariton approach to predict the dynamic Stark effect and obtain expressions for the Stark splitting and the spectrum of weak excitations in the effective two level system. As well as energy levels, the effective damping rates are calculated. We then compare these results with the results obtained for the two level atom.

Moreover, we calculate the fluorescence spectrum and illustrate how to use the effective Hamiltonian to identify peaks and linewidths in the fluorescence spectrum for the light exiting the cavity mode. The dominant part of the spectrum is found to have a familiar three–peaked Mollow shape, but additional peaks are also present. The central peak in the Mollow spectrum corresponds to the resonant transitions between the ground state and the excited state. We find that the sidebands appear as a consequence of Stark splitting at some threshold value of effective driving. The polariton model enables the calculation of this threshold value of driving field. In addition, using the effective Hamiltonian, we identify the transitions that produce the additional peaks.
11.2 Analysis of the Dynamic Stark Splitting

It was proven in Parts III and IV of this thesis (see also [102, 104]) that the EIT-Kerr system can behave as an effective two level system, with the states $|e_0^{(0)}\rangle$ and $|e_0^{(1)}\rangle$ being its ground and excited states. The two level approximation is best for large values of effective dipole coupling, i.e. $(g_1/\Omega_c)^2 \gg 1$ and $g_2 \gg \kappa$. The two states of the effective model are coupled by the external field, with the Rabi frequency of the coupling, $\Omega_0^{(0,1)}$, given by Equation (10.18b), and the decay rate of the excited state, $\Gamma_0^{(1)}$, given by Equation (10.26a). It is therefore expected that this system exhibits a dynamic Stark splitting, characteristic of every driven two state system. We now explore this effect in more detail, using the polariton model.

Recall that the effective Hamiltonian (10.4) is non-Hermitian. We truncate the expansion over manifolds of the polariton Hamiltonian after the first manifold. Moreover, we concentrate on the ground and excited state of the effective two level system and write its polariton Hamiltonian in the reduced form

$$ H_{\text{red}} = i\hbar \Omega_0^{(1,0)} \left( p_0^{(1)} - p_0^{(1)\dagger} \right) - i\hbar \Gamma_0^{(1)} p_0^{(1)} p_0^{(1)\dagger}. $$

The eigenvalues of such an effective Hamiltonian are complex:

$$ \varepsilon_\pm = \tilde{\varepsilon}_\pm - i\tilde{\Gamma}_\pm $$

$$ = i \frac{\Gamma_0^{(1)}}{2} \pm \sqrt{\Omega_0^{(1,0)^2} - \left( \frac{\Gamma_0^{(1)}}{2} \right)^2}. $$

Figure 11.1: Graphical representation of the dynamic Stark splitting of the effective two level system.
The real parts $\tilde{\epsilon}_\pm$ of these eigenvalues represent the energies of the dressed states, while the imaginary parts $\tilde{\Gamma}_\pm$ represent their associated decay rates. We identify two operating regimes, depending on the size of $\Omega_0^{(1,0)}$, i.e. the size of $\mathcal{E}_p$:

- **Regime 1**: $\Omega_0^{(1,0)} < \Gamma_0^{(1)}/2$

\[
\begin{align*}
\tilde{\epsilon}_\pm & = 0, \\
\tilde{\Gamma}_\pm & = \frac{\Gamma_0^{(1)}}{2} \pm \sqrt{\left(\frac{\Gamma_0^{(1)}}{2}\right)^2 - \Omega_0^{(1,0)^2}}. 
\end{align*}
\]  
(11.3a)

- **Regime 2**: $\Omega_0^{(1,0)} > \Gamma_0^{(1)}/2$

\[
\begin{align*}
\tilde{\epsilon}_\pm & = \pm \sqrt{\Omega_0^{(1,0)^2} - \left(\frac{\Gamma_0^{(1)}}{2}\right)^2}, \\
\tilde{\Gamma}_\pm & = \frac{\Gamma_0^{(1)}}{2}. 
\end{align*}
\]  
(11.3b)

The eigenstates corresponding to the Stark–split states are

\[
|\psi_{\pm}^{(0,1)}\rangle = \frac{1}{\sqrt{2}} \left( |e_0^{(0)}\rangle \pm |e_0^{(1)}\rangle \right) \\
= \frac{1}{\sqrt{2}} \left( |0, 1\rangle \pm \frac{|1, 1\rangle + g_1/\Omega_c |0, 3\rangle}{\sqrt{1 + (g_1/\Omega_c)^2}} \right).  
\]  
(11.4)

The transition between the two regimes happens at $\Omega_0^{(1,0)} = \Gamma_0^{(1)}/2$, or, in terms of the original parameters, at

\[
\mathcal{E}_p = \frac{\kappa/2}{\sqrt{1 + (g_1/\Omega_c)^2}}. 
\]  
(11.5)

Note that there is no contribution of the atomic decay rates in this simple model. This absence occurs because the excited state in the effective model is a dark state with respect to atomic spontaneous emission. The dynamic Stark splitting effect is shown schematically in Figure 11.1.

Another effective two level system was predicted by Tian and Carmichael [103], who studied the case of a two level atom in the cavity, driven on the lower Rabi resonance. They also predicted Stark splitting [131] comparable to that presented in this paper.

For further comparison, the analysis of a two level atom with spontaneous emission rate $\gamma$, coupled with strength $g$ to the vacuum cavity mode, yields results identical to those of Eqs. (11.3), with the replacements $\Omega_0^{(1,0)} \rightarrow g$ and $\Gamma_0^{(1)} \rightarrow \gamma + \kappa$. Thus, the dynamic Stark splitting found in the EIT–Kerr system can be thought of as the exact counterpart
Figure 11.2: Numerical (dashed lines) eigenenergies for the full system (cavity mode subspace truncated at 40) compared with the analytical solution of Equation (11.2) (solid lines). The parameters are \( g_j/\kappa = 6, \gamma_j/\kappa = 0.1, \Omega_c/\kappa = 2 \).

to the vacuum Rabi splitting characteristic for a two level atom coupled to the cavity mode. The distinction of the EIT–Kerr system is in the fact that the parameters \( (1,0) \) and \( (1)0 \) can be adjusted by a simple adjustment of the coupling laser Rabi frequency \( \Omega_c \).

How well can these results describe the full EIT–Kerr system, including damping terms, as described by Hamiltonian (10.4)? Figure 11.2 compares the two solutions. There is very good agreement between numerical solution and analytical approximation, which breaks down only for large values of \( \mathcal{E}_p \), where truncation after the first manifold is not justified any more, since the contribution of states from higher manifolds cannot be ignored.

One additional feature can be seen by looking at the eigenenergies in Figure 11.2. Notice that the Stark splitting of eigenenergies does not start at \( \mathcal{E}_p = 0^+ \) but at some small, finite value of \( \mathcal{E}_p \). This behaviour and the related behaviour of the decay rates for weak excitation is shown in Figure 11.3. In the limit \( \mathcal{E}_p = 0 \), the ground and excited states of the effective two level system are uncoupled, so the decay rates separate accordingly to the decay rates of ground state (which is zero) and the excited state \( \Gamma_0^{(1)} \). Increasing the
driving strength mixes these two states so that their decay rates become approximately equal. Once $\mathcal{E}_p$ exceeds the value given by Equation (11.5), the energy levels shift in opposite directions, giving rise to the Stark splitting. For the parameters of Figure 11.3, this happens at $\mathcal{E}_p \cong 0.16\kappa$.

In the weak driving regime (Regime 1 above), $\mathcal{E}_p < (\kappa/2)/\sqrt{1 + (g_1/\Omega_c)^2}$, and the EIT–Kerr system truly behaves as an effective two level system due to the absence of normal mode splitting. This is also the ideal photon blockade regime, and will be called a weak driving regime. The case $\mathcal{E}_p > (\kappa/2)/\sqrt{1 + (g_1/\Omega_c)^2}$ (Regime 2) then includes the intermediate and strong driving regimes, which will be studied in Chapter 12. Once again, we emphasise the similarity of the effective two level system coupled to the driving field and the two level atom coupled to a vacuum cavity mode. There is an equivalence in the behaviour of the Rabi–split states in the latter [80, 78] to the behaviour of the Stark–split states in the former.

The analysis of the dynamics of the Stark splitting in the dressed state basis offers a simple example of the convenience of the polariton approach. Once the Hamiltonian is expressed in terms of the polaritons and the reduced effective Hamiltonian is identified, the subsequent analysis is considerably simplified.

11.3 Two Level Atom

In Section 8.3.1, we analysed the extended JC system from the viewpoint of photon blockade. As a basis for the analysis, we obtained the dressed states for the single–atom in the absence of driving, i.e. we obtained the expressions for the vacuum Rabi splittings and their linewidths, given in equations (8.4). The complex eigenvalues for the first manifold ($n = 1$) Rabi splittings are given by $\hbar \varepsilon^{JC}_{\pm}$, where

$$\varepsilon^{JC}_{\pm} = \epsilon_{\pm} - i\Gamma_{\pm}$$

$$= \frac{i(\gamma + \kappa)}{2} \pm \sqrt{g^2 - \left(\frac{\gamma - \kappa}{2}\right)^2}. \quad (11.6)$$

We can identify two regimes here, depending on the size of the coupling $g$, that will determine whether the expression under the square root is positive or negative, giving:

- **Regime 1**: $g < |\gamma - \kappa|/2$

  $$\epsilon_{\pm} = 0,$$

  $$\Gamma_{\pm} = \frac{\kappa + \gamma}{2} \pm \sqrt{\left(\frac{\gamma - \kappa}{2}\right)^2 - g^2}, \quad (11.7a)$$
Figure 11.3: Rabi splittings and linewidths for the extended Jaynes–Cummings system (left) and the EIT–Kerr system (right). The extended JC system was analysed for $\gamma = 0.1\kappa$, while the EIT–Kerr system parameters are $g_j = 6\kappa$, $\Omega_c = 2\kappa$.

- Regime 2: $g > |\gamma - \kappa|/2$

$$
\begin{align*}
\epsilon_\pm &= \pm g \sqrt{1 - \left(\frac{\gamma - \kappa}{2g}\right)^2}, \\
\Gamma_\pm &= \frac{\gamma + \kappa}{2},
\end{align*}
$$

(11.7b)

where $\epsilon_\pm$ denote the dressed state energies and $\Gamma_\pm$ denote the respective decay rates. The transition between the different regimes happens at $g = |\gamma - \kappa|/2$. Both regimes are shown in Figure 11.3. This structure of the Rabi splittings is well understood (see, for example,
Kimble [78] and Turchette et al. [50]). For $g = 0$, the atom and the cavity are uncoupled, and consequently the decay rates separate to the pure atomic branch $\gamma$ and pure cavity branch $\kappa$. As the coupling of cavity mode to the atom increases, the two branches start mixing, until they reach a limiting value of $(\gamma + \kappa)/2$.

The first regime includes the bad cavity regime $\gamma \ll g^2/\kappa \ll \kappa$. In this regime, we recognise two decay branches of equation (11.7a) as (i) enhanced atomic decay at rate

$$\Gamma_+ \approx \gamma + \frac{g^2}{\kappa} = \gamma(1 + C),$$

(11.8)

where $C = g^2/(\kappa \gamma)$ denotes the single-atom cooperativity parameter, and (ii) cavity decay at a somewhat reduced rate

$$\Gamma_- \approx \kappa - \frac{g^2}{\kappa} = \kappa \left(1 - \frac{\gamma C}{\kappa}\right).$$

(11.9)

In the strict bad cavity limit, we have $\gamma/\kappa \to 0$, $C$ constant and $\Gamma_- \to \kappa$.

The second regime is the strong coupling regime $g \gg \kappa, \gamma$, where we find Rabi splitting of the excited state, with each of the split states having the same decay rate.

### 11.4 Fluorescence Spectrum

The prediction of dynamic Stark splitting in Section 11.2 leads us naturally to an examination of the fluorescence spectrum of the light emitted by the 'atom–cavity molecule'. We solve the master equation of the problem numerically to obtain the spectrum, and interpret the result using the insight provided by the polariton analysis.

The master equation of the full atom/cavity system may be written in the bare form as

$$\dot{\rho} = -i\hbar \left(\mathcal{H}_{\text{eff}} \rho - \rho \mathcal{H}_{\text{eff}}^\dagger\right) + 2 \sum_{i=1}^{4} C_i \rho C_i^\dagger,$$

(11.10)

where $\rho$ is the density matrix of the system, $\mathcal{H}_{\text{eff}}$ is given by Equation (10.4) and $C_i$ denote the four collapse operators of Equation (10.3). Using the quantum regression theorem [1], we solve the master equation and calculate the steady-state fluorescence spectrum,

$$S_F(\omega) = \text{Re} \left[ \lim_{t \to \infty} \int_0^\infty \text{d} \tau \langle a^\dagger(t), a(t + \tau) \rangle e^{i\omega \tau} \right].$$

(11.11)

Results, for different values of driving $E_p$, are shown in Figs. 11.4 and 11.5.

Figure 11.4 shows how the central peak (at the frequency of driving) splits into a Mollow triplet [132]. The particular values of parameters are given in dimensionless units, and the small value of $\kappa$ is chosen to produce narrow, well-resolved peaks. For this set of parameters, Equation (11.5) predicts a threshold value for the appearance of
Figure 11.4: Emergence of the Mollow triplet with increasing driving field. Parameters are $\kappa = 0.25$, $\gamma_j = 0.1$, $g_j = 6$, $\Omega_c = 2$, $\delta = 0$ and $\Delta = 0.1$. Arbitrary units are used, as explained in the main text.

Mollow sidebands at $\mathcal{E}_p = 0.0395$. The central peak is the result of the two transitions $|\psi_-^{(0)}\rangle \leftrightarrow |\psi_+^{(0)}\rangle$ and $|\psi_-^{(1)}\rangle \leftrightarrow |\psi_+^{(1)}\rangle$ between the Stark doublet states in the ground and excited states (see Figure 11.1). Transitions $|\psi_-^{(0)}\rangle \leftrightarrow |\psi_+^{(1)}\rangle$ and $|\psi_-^{(1)}\rangle \leftrightarrow |\psi_+^{(0)}\rangle$ cause the appearance of the sidebands at frequencies $\omega = \omega_{cav} + (\tilde{\varepsilon}_+ - \tilde{\varepsilon}_-)$ and $\omega = \omega_{cav} + (\tilde{\varepsilon}_- - \tilde{\varepsilon}_+)$, respectively, where $\tilde{\varepsilon}_\pm$ are given by Equation (11.3b).

The linewidths of these peaks can also be calculated. It is straightforward to write the master equation in the polariton picture as

$$\dot{\rho} = -\frac{i}{\hbar}\left(\mathcal{H}_{red}\rho - \rho\mathcal{H}_{red}^\dagger\right) + 2\Gamma_0^{(1)} P_0^{(1)} \rho P_0^{(1)\dagger},$$

where $\mathcal{H}_{red}$ is given by Equation (11.1). Equations for the density matrix elements in the basis spanned by Stark states $|\psi_\pm\rangle$ of Equation (11.4) can be derived by performing a
rotation of basis in the Hilbert subspace (see Figure 11.6), to give

$$\rho_{++} = -\Gamma_0^{(1)} \rho_{++} + \frac{\Gamma_0^{(1)}}{2},$$

(11.13a)

$$\rho_{+-} = -\left(\frac{3\Gamma_0^{(1)}}{2} - i2\Omega_0^{(1,0)}\right)\rho_{+-}$$

$$-\frac{\Gamma_0^{(1)}}{2}\rho_{-+} - \Gamma_0^{(1)}.$$  

(11.13b)

From these equations, it is easy to read the spectral linewidths of the Mollow spectrum. The central peak will have linewidth $\Gamma_0^{(1)}$, while the sidebands have linewidth $3\Gamma_0^{(1)}/2$. This is consistent with the results for resonance fluorescence [1].

Given the complex energy level structure of the atom–cavity molecule, it can be expected that transitions other than those producing the Mollow spectrum will be seen in the fluorescence spectrum. This is indeed true, and Figure 11.5 shows the additional sidebands. These peaks are relatively small ($\sim 10^{-3}$), so the associated transitions are not
expected to contribute significantly to the dynamics. They do, however, cause a departure from the ideal two level behaviour.

The transitions responsible for the sidebands are identified in Figure 11.7, where the relevant energy level structure is shown. Contributions of the transitions up to the third manifold states can be seen. We find peaks at the following frequencies: $\pm \Delta_1 \approx \pm 2.3$, $\pm \Delta_2 \approx \pm 5.7$, $\pm \Delta_3 \approx \pm 6.05$ and $\pm \Delta_4 \approx \pm 6.3$. The transitions corresponding to these peaks can be identified from the energy eigenvalues as $\Delta_1 = \epsilon_3^{(3)} - \epsilon_2^{(2)}$, $-\Delta_1 = \epsilon_2^{(3)} - \epsilon_2^{(2)}$, $\Delta_2 = \epsilon_3^{(2)} - \epsilon_+^{(1)}$, $-\Delta_2 = \epsilon_2^{(2)} - \epsilon_-^{(1)}$, $\Delta_3 = \epsilon_3^{(2)} - \epsilon_-^{(1)}$, $-\Delta_3 = \epsilon_2^{(2)} - \epsilon_+^{(1)}$, $\Delta_4 = \epsilon_+^{(1)} - \epsilon_0^{(0)}$. The tiny asymmetry between the positions of the positive frequency peaks and negative frequency peaks arises from the fact that the polariton states are asymmetrically detuned from the cavity resonance. The source of this asymmetry can be traced to the nonzero values of atomic detunings $\delta$ and $\Delta$. The linewidths of these sidebands can also be computed using the same method that produced the Equation (11.13), but the results would be hard to check, given the large degree of overlap between the adjacent peaks, seen in Figure 11.5. We thus have a complete explanation of the fluorescence spectrum.

11.5 Conclusion

In this Chapter we have presented an application of the methods developed in Chapter 10 to an effective two level system. The dynamic Stark splitting of the dressed states has

Figure 11.6: Change of basis in the Hilbert subspace.
Figure 11.7: Schematic depiction of the relevant transitions for Figure 11.5. Full lines denote the energy levels involved, dashed lines denote positions of the cavity resonances. $\Delta_j$'s are explained in the main text.
been analysed and discussed. The fluorescence spectrum of light exiting the cavity mode has been calculated, and peaks and linewidths in the fluorescence spectrum have been identified, using the effective Hamiltonian and the equations of motion for the Stark-split states.
Chapter 12

Analysis of Photon Statistics for the Driven EIT–Kerr System

In this Chapter, we explore the photon statistics of light emitted from an EIT–Kerr system, for a range of driving field strengths. We find a sudden change in the photon statistics of the light emitted from the cavity for intermediate values of driving strengths. Photon antibunching switches to photon bunching over a very narrow range of intracavity photon number. This effect, as well as nonclassical correlations at nonzero delay times, is studied in detail.

12.1 Introduction

The validity of the results so far presented in this thesis depends on the assumption of weak driving. It is of interest to explore how robust the effect of photon blockade is as the driving field strength is increased. The formalism of Chapter 10 is ideally suited for this analysis, since it enables the truncation of the driving field contribution according to the relevant polariton structure.

We perform an analysis of the driving field dependence in the EIT–Kerr system, and briefly compared the results with those obtained for the extended Jaynes–Cummings model. A surprising feature is found in the EIT–Kerr system, where a sudden change in photon statistics (i.e. of $g^{(2)}(0)$) of light emerging from the cavity occurs at intermediate driving strengths. If plotted against the mean intracavity photon number, $g^{(2)}(0)$ is seen to undergo a sudden transition, switching rapidly between antibunching and strong bunching. The exact position of the threshold depends on the characteristic system parameters, namely the atom–field interaction strength and the (EIT) coupling field Rabi frequency.

For the numerical simulations of the photon statistics, we choose the method of quantum trajectories, which enables the large system to be solved and, later on, compared with the solution of the effective model. We prove that the sudden change in photon statistics occurs due to the existence of robust quantum interference between the dressed states of the atom–cavity system. Furthermore, it is shown that the strong photon bunch-
ing is a nonclassical effect for certain values of driving field strength, violating classical inequalities for field correlations.

12.2 Model

12.2.1 Hamiltonian

The Hamiltonian describing the system in the rotating wave and electric dipole approximations is $\mathcal{H} = \mathcal{H}_0 + \mathcal{H}_d$, where, in the interaction picture,

$$\mathcal{H}_0 = \hbar \delta \sigma_{22} + \hbar \Delta \sigma_{44} + i \hbar g_1 (a^\dagger \sigma_{12} - \sigma_{21} a)$$

$$+ i \hbar \Omega_c (\sigma_{33} - \sigma_{33}) + i \hbar g_2 (a^\dagger \sigma_{34} - \sigma_{43} a),$$

$$\mathcal{H}_d = i \hbar \mathcal{E}_p (a - a^\dagger).$$

The notation in these equations is the same as in the remainder of the thesis. The cavity driving field is characterised through the parameter $\mathcal{E}_p$, related to the power output of the driving laser $\mathcal{P}$ via

$$\mathcal{E}_p = \sqrt{\frac{P \kappa T^2}{4 \hbar \omega_{cav}}}.$$ 

where $T$ is the cavity mirror transmission coefficient, $\kappa$ is the cavity decay rate, and $\omega_{cav}$ is the cavity mode frequency. The envisaged setup is shown in Figure 12.1 (a). Damping due to cavity decay and spontaneous emission is introduced below in the context of the quantum trajectory approach [118].

Four atomic levels plus the cavity mode span a Hilbert space of dimension $4 \times N$, where $N$ denotes the truncation order in a Fock state expansion of the cavity field subspace. In the absence of driving, Hamiltonian $\mathcal{H}_0$, given by Equation (12.1a), takes a block-diagonal form, with $N$ blocks on the main diagonal. Each block represents a manifold of eigenstates associated with the appropriate term in the Fock expansion. The ground, first and second manifolds have been analyzed from the viewpoint of photon blockade in Parts III and IV (also [102, 104, 107]). The general structure of the dressed states in an arbitrary $n^{th}$ manifold has been discussed in Chapter 10 and [133].

Addition of the driving term (12.1b) complicates the analysis, since the Hamiltonian matrix does not retain the block-diagonal form. It is possible, however, to re-express the driving Hamiltonian in terms of polariton operators and effective Rabi frequencies for transitions between dressed states. This has been done in Chapter 10, and in the remainder of this Chapter we will draw on these results.

Our analysis of the driven ‘atom–cavity molecule’ will proceed in two complementary directions. First, using the method of quantum trajectories [118], we obtain numerical data. Then, using the formalism of Chapter 10, we construct an effective Hamiltonian in the polariton basis, which encapsulates the physics sufficiently to explain the numerical results.
12.2. MODEL

(a)

Driving $E_p$ 

$\kappa$

$\Omega_c$

$g^{(2)}(\tau)$$

(b)

$|4\rangle$

$\Delta$

$|2\rangle$

$g_2; \omega_{\text{cav}}$

$\gamma_3$

$\gamma_2$

$\Omega_c$

$g_1; \omega_{\text{cav}}$

$\gamma_1$

$|3\rangle$

$|1\rangle$

Figure 12.1: (a) Envisaged experimental setup. A single four level atom is trapped in an optical cavity with the decay constant $\kappa$. The cavity is driven by a coherent laser field which couples to the cavity mode with strength $E_p$. An additional laser with Rabi frequency $\Omega_c$ directly couples to the atomic transition. (b) Atomic energy level scheme. The cavity mode drives transitions $|1\rangle \rightarrow |2\rangle$ and $|3\rangle \rightarrow |4\rangle$, with the respective coupling strengths $g_1$ and $g_2$. The transition $|2\rangle \leftrightarrow |3\rangle$ is coupled by a classical field of frequency $\omega_c$ and Rabi frequency $\Omega_c$. Spontaneous emission rates are denoted by $\gamma_j$. Detunings $\delta$ and $\Delta$ are positive in the shown configuration.

12.2.2 Damping: Quantum Trajectories

In the quantum trajectories approach, damping enters the model through collapse operators, with each of these corresponding to one decay channel. In the case under consideration we require the following four collapse operators,

$$
C_1 = \sqrt{\gamma_1} \sigma_{12}, \quad C_2 = \sqrt{\gamma_2} \sigma_{32}, \\
C_3 = \sqrt{\gamma_3} \sigma_{34}, \quad C_4 = \sqrt{\kappa} a,
$$

(12.3)
where $\gamma_k$ denote spontaneous emission rates into each of the decay channels, and $\kappa$ is the cavity field decay rate. The effective Hamiltonian used in the trajectories approach is non–Hermitian and takes the form

$$\mathcal{H}_{\text{eff}} = \mathcal{H} - i \sum_{k=1}^{4} C_k^{\dagger} C_k,$$

with $\mathcal{H}$ given by (12.1).

In deciding on the truncation for the cavity mode Hilbert space, special care must be taken to include a sufficient number of states to capture all the relevant dynamics. If an empty cavity would be driven by an external coherent field corresponding to the parameter $\mathcal{E}_p$, the amplitude of the intracavity coherent field would be

$$\alpha = i \mathcal{E}_p/\kappa,$$

and the expected mean intracavity photon number $\langle n \rangle = |\alpha|^2$. Our simulations include driving strengths of up to $\mathcal{E}_p = 3\kappa$, so the truncation is set at $N = 40$. The inset in Figure 12.2 shows that the actual mean intracavity photon number stays well below its empty cavity counterpart for the given range of driving, thus justifying the chosen truncation.

### 12.3 Numerical Simulations of Photon Statistics

In this section we present the results of numerical simulations using the method of quantum trajectories [118], with results averaged over 10000 trajectories. Values of parameters chosen for the simulations are consistent with recent experiments [72], so the scheme presented in this thesis is experimentally viable. The simulations performed here are, of course, stochastic simulations. The results obtained from these carry a certain degree of error, observed as the fluctuations visible on the curves. It should be noted here that in quantum trajectory approach, this is the consequence of finite number of trajectories involved in simulations. In the limit of infinite number of trajectories, curves would be smooth, identical to the solution obtained from the equivalent master equation.

We evaluate the second–order correlation function $g^{(2)}(0)$ as a function of driving strength $\mathcal{E}_p$. It was established earlier [51, 102] that this function is a good measure of photon blockade; perfect photon blockade yields perfectly antibunched photons. The steady–state second–order correlation function is given by

$$g^{(2)}(\tau) = \lim_{t \to \infty} \frac{\langle a^{\dagger}(t)a^{\dagger}(t+\tau)a(t+\tau)a(t) \rangle}{\langle a^{\dagger}(t)a(t) \rangle \langle a^{\dagger}(t+\tau)a(t+\tau) \rangle}.$$  

(12.6)

Perfect antibunching/photon blockade corresponds to $g^{(2)}(\tau = 0) = 0$; for a coherent field $g^{(2)}(\tau = 0) = 1$, and for a bunched field $g^{(2)}(\tau = 0) > 1$ [1].

Figure 12.2 shows $g^{(2)}(\tau = 0)$ as a function of the (scaled) driving parameter for a single–atom EIT–Kerr system, compared with the same quantity calculated for the
12.3. NUMERICAL SIMULATIONS OF PHOTON STATISTICS

Figure 12.2: Second order correlations at zero time–delay against the (scaled) driving parameter. The inset shows mean intracavity photon number in the steady state. The dotted line denotes the extended Jaynes–Cummings scheme for which $\gamma = 0.1\kappa$, $g = 6\kappa$. Other curves show the single–atom EIT–Kerr model with $\gamma_j = 0.1\kappa$, $g_l = 6\kappa$, $\delta = -0.2\kappa$, $\Delta = 0.1\kappa$. The solid line represents the case $\Omega_c = 2\kappa$; dot–dashed line $\Omega_c = 6\kappa$; dashed line $\Omega_c = 18\kappa$. All curves are obtained by averaging over $10^4$ trajectories.

extended Jaynes–Cummings model. The extended JC model is driven on the lower Rabi resonance, as envisaged by Tian and Carmichael [103]. In the weak driving regime, simulation confirms earlier results – stronger photon blockade in the EIT–Kerr system. As the driving increases, the extended Jaynes–Cummings model gradually saturates, and the field correlation asymptotically tends to one. Naively, one would expect qualitatively similar behavior in the EIT–Kerr model. Our simulation, however, shows a vastly different result. Over a narrow range of driving, the statistics of the field changes from strongly antibunched to strongly bunched, and the coherent state value $g^{(2)}(0) = 1$ is approached asymptotically from above as $\mathcal{E}_p$ is increased further.

Note that the quantity being increased here is the scaled driving parameter, so one may argue that it is the ratio that determines this behavior, i.e. we may either increase the driving strength or decrease the cavity decay rate to obtain the same result. However, to clarify this issue as related to the photon statistics, in Figure 12.3 we show a parametric plot of second–order correlation function against the expectation value of intracavity photon number.
Figure 12.3: Second order correlations at zero time–delay against the mean steady–state intracavity photon number for a single–atom EIT–Kerr system. Parameters are the same as in Figure 12.2. Figure (a) shows the normalised correlation function with main figure showing the range where the sudden change can be seen in detail, while the inset shows the whole range. Figure (b) shows the unnormalised second order correlations.

photon number, \(\langle a^\dagger a \rangle_{ss} \). Both quantities are now unscaled by any external parameter, and the sudden nature of the switch becomes even more obvious, showing a phase–transition–like behaviour. This can be related back to Figure 12.2. In particular, the transition from antibunching to bunching happens over a range of driving for which the intracavity photon number stays practically constant (see inset of Figure 12.2), leading to the suddenness of the transition seen in Figure 12.3, and the concentration of points around the threshold region. To further emphasise this, in Figure 12.3 (b) we plot the ‘pure’ second–order correlation function \(\langle a^\dagger a^\dagger aa \rangle_{ss} \) which also exhibits a threshold–like behaviour.

One other feature of the numerical results is noted. The bunching–antibunching transition is sharper and the subsequent bunching stronger for smaller \(\Omega_c\). In fact, it is the increasing ratio \(g_1/\Omega_c\) that really matters. The sharper transitions also occur at smaller values of \(\langle a^\dagger a \rangle_{ss} \). For a decreasing ratio \(g_1/\Omega_c\), \(g^{(2)}(0)\) approaches the behaviour of the
12.4 Density Matrix Treatment

We proceed to determine which eigenstates of the strongly coupled quantum system contribute significantly to its dynamics. The total density matrix of the system in the steady state can be written in most general terms as

\[ \rho = \sum_{a,b} c_{ab} |a\rangle \langle b|, \tag{12.7} \]

where \( a, b \) belong to the set of all possible states of the system in an arbitrary basis. The natural basis for the simulation is the one of bare states. The cavity mode subspace is truncated at 40, making the size of \( \rho \) equal to \( 160 \times 160 \). Let \( T \) be the transformation that diagonalises Hamiltonian (12.1), i.e. the full Hamiltonian, with driving included. The density matrix can be transformed into a new basis as \( \rho_{\text{diag}} = T \rho T^{-1} \), and we can look for the nonzero elements of this matrix. Diagonal elements of the matrix correspond to populations of the dressed states, while off–diagonal elements correspond to coherences between the dressed states. The results depend on the size of \( \mathcal{E}_p \), i.e. we expect the number of non–zero elements to increase as \( \mathcal{E}_p \) is increased. For a large part of the \( \mathcal{E}_p \) range considered, however, the density matrix is dominated by elements corresponding to two particular states.

Figure 12.4 shows the nonzero matrix elements of density matrix \( \rho_{\text{diag}} \). Diagonal states are sorted in increasing order; the state with largest negative detuning is at \((i, j) = (1, 1)\), while the state with largest positive detuning is at \((i, j) = (160, 160)\). States with the smallest detuning (i.e. closest to the cavity resonance) are at the centre of matrix, at positions 80 and 81 along the main diagonal. Two diagonal elements dominate the matrix, and we identify these as being the populations of the Stark–split states \( |\psi_{\pm}\rangle \) (see Section 11.2). Stark–split states are therefore found at the positions 80 and 81 on the main diagonal of the density matrix. At the positions 79 and 82 are two states from the second manifold (two–photon excitations), closest to the resonance. At the positions 78 and 83 are the two remaining states from the first manifold; at the positions 77 and 84 are two states from the third manifold (three–photon excitations), closest to the resonance, and so on. The states we have just identified suffice to indicate the dynamics of the system. Off–diagonal elements are coherences between the appropriate dressed states.

One striking feature can be noted immediately from Figure 12.4. Namely, the first ‘square’ of elements (4 elements on each side) encircling the centre square (Stark states, \( 2 \times 2 \)) remains much smaller than all of the other accessible states. In terms of dressed states, this means that the second manifold states remain unpopulated and the coherences between these and all the other states vanish as well. Furthermore, as the driving increases, the states whose coherences with the Stark states increase to a noticable size are the third manifold states (see Figure 12.4 for \( \mathcal{E}_p = 0.09\kappa \)). As the driving increases further, the remaining two first manifold states (their populations and coherences with
CHAPTER 12. ANALYSIS OF PHOTON STATISTICS

Figure 12.4: Relevant density matrix elements at the different values of driving. Elements of the central submatrix (dimension 21 \times 21) of the total density matrix (160 \times 160) are shown. In the basis which diagonalises the Hamiltonian, the chosen submatrix contains elements corresponding to the dressed states closest to the cavity resonance.

From the above analysis of the density matrix elements, we can deduce the solution to the ‘photon statistics puzzle’ of Figs. 12.2 and 12.3. Namely, strong photon antibunching is the consequence of the effective two level system, and has been well understood. The sharp rise in \(g^{(2)}(0)\) can be attributed to the two–photon transitions needed to populate third manifold states from the first manifold, and then decay back in cascade to the first manifold. These two–photon decays cause the sharp increase in the output field correlations. But, what causes the system to skip second manifold states? Again, the answer can be deduced from the density matrix. Strong coherence between the first and third manifold states, together with the missing population in the second manifold states (and vanishing coherences associated with these states) uncovers the effect of EIT–
12.4. DENSITY MATRIX TREATMENT

Figure 12.5: Semilogarithmic plots of the density matrix elements. (a) Plot of the coherences between the first manifold and third manifold states. Coherence $\rho_{14}^{(+)}$ is the coherence between the upper Stark state and lower third manifold state. Coherence $\rho_{15}^{(-)}$ is the coherence between the lower Stark state and upper third manifold state. (b) Plot of the populations of the dressed states. $2^{\text{nd}}$ denotes a sum of populations in the two relevant states of the second manifold, $1^{\text{st}}$ denotes a sum of populations in the two far detuned states in the first manifold and $3^{\text{rd}}$ denotes a sum of populations in the two relevant states of the third manifold. Parameters are as in Figure 12.2, solid line.

type quantum interference between the dressed states. This is not surprising, since the quantum interference between the transitions from first to second manifold have been already discussed in Ref. [134].

These features are shown in Figure 12.5, where the relevant populations and coherences are shown. Note that the combined population of the two inner second manifold states (Figure 12.5 (b), solid line) vanishes for a large interval of driving, since the value of $\sim 10^{-16}$ is at the numerical precision value, and fluctuations are numerical, not physical in nature. These populations become nonzero at the value of driving strength at which $g^{(2)}(0)$ of Figure 12.2 (solid line) peaks. These plots further justify the discussion of the preceding paragraphs.
Figure 12.6: Schematic depiction of the six states used to formulate the effective model. States $|e_0\rangle$ and $|e_1\rangle$ are the effective two level system from Chapter 11. States $|e_2\rangle$ and $|e_3\rangle$ are two second manifold states closest to the resonance, and $|e_4\rangle$ and $|e_5\rangle$ are two third manifold states closest to the resonance. Arrows represent effective driving.

12.5 Effective Model

In the formulation of an effective model, we rely on the formalism developed in Chapter 10. This formalism was very successful in the development of an effective two level theory, and in explaining the fluorescence spectrum. Now we extend the model and include a total of six dressed states in the effective model. These states are shown in Figure 12.6.

It should be noted, however, that the dressed states shown in Figure 12.6 do not correspond exactly to the dressed states discussed in Section 12.4. Namely, the dressed states of Section 12.4 are often referred to as doubly dressed states, since they diagonalise the Hamiltonian with driving contributions included. In this Section, we will treat driving separately (see Chapter 10 and [133]), and let the dressed states represent the eigenstates of the interaction Hamiltonian (12.1a) alone. Driving can then be included through effective Rabi frequencies $\Omega_{ij}$, coupling dressed states $|e_i\rangle$ and $|e_j\rangle$.

The relevant effective non-Hermitian Hamiltonian (including driving and damping) is
thus

\[
\mathcal{H}_{\text{eff}} = \hbar e_2 p_2^+ p_2 + \hbar e_3 p_3^+ p_3 + \hbar e_4 p_4^+ p_4 + \hbar e_5 p_{15}^+ p_{15} \\
+ i\hbar \Omega_{01} \left( p_1 - p_1^\dagger \right) + i\hbar \left( \Omega_{12}^\dagger p_2 - \Omega_{12} p_2^\dagger \right) + i\hbar \left( \Omega_{13}^\dagger p_3 - \Omega_{13} p_3^\dagger \right) \\
+ i\hbar \Omega_{24} \left( p_{24} - p_{24}^\dagger \right) + i\hbar \Omega_{25} \left( p_{25} - p_{25}^\dagger \right) \\
+ i\hbar \Omega_{34} \left( p_{34} - p_{34}^\dagger \right) + i\hbar \Omega_{35} \left( p_{35} - p_{35}^\dagger \right) \\
- i\hbar \Gamma_{e_1} p_1^\dagger p_1 - i\hbar \Gamma_{22} p_2^\dagger p_2 - i\hbar \Gamma_{33} p_3^\dagger p_3 - i\hbar \Gamma_{44} p_4^\dagger p_4 - i\hbar \Gamma_{55} p_{15}^\dagger p_{15} \\
- i\hbar \Gamma_{23} p_{23}^\dagger p_{23} - i\hbar \Gamma_{33} p_{33}^\dagger p_{33} - i\hbar \Gamma_{45} p_{45}^\dagger p_{45} - i\hbar \Gamma_{54} p_{54}^\dagger p_{54}.
\]  

(12.8)

Operators \( p_{jk} \) are polariton operators defined by \( |e_k\rangle = p_{jk}^\dagger |e_j\rangle \). For the states \( |e_k\rangle \) which are accessible from only one lower state, the notation has been abbreviated, so, for example, \( p_2 \equiv p_{12} \). Note also that the indices \( j \) are dummy indices, i.e., \( p_{jk}^\dagger p_{jl} = |e_k\rangle \langle e_l| \). Rabi frequencies \( \Omega_{12} \) and \( \Omega_{13} \) have a phase term making them purely imaginary, while all the other Rabi frequencies are real. Damping terms were discussed in detail in Chapter 10. It is now possible to formalise the distinction between the two dressed states bases used in this Section and Section 12.4. The basis in which the Hamiltonian (12.8) is written is defined in Chapter 10 and depicted in Figure 12.6. The discussion of Section 12.4 is based upon diagonalizing the total Hamiltonian (including all of the 160 states used for the numerical simulation).

With the effective Hamiltonian (12.8), we can also write the master equation for an effective density matrix as

\[
\dot{\rho}_{\text{eff}} = -\frac{i}{\hbar} \left( \mathcal{H}_{\text{eff}} \rho_{\text{eff}} - \rho_{\text{eff}} \mathcal{H}_{\text{eff}}^\dagger \right) + 2 \sum_{i,j} S_i \rho_{\text{eff}} S_j^\dagger,
\]  

(12.9)

where \( S_k \) now denote the polariton collapse operators. The effective density matrix \( \rho_{\text{eff}} \) has the dimension \( 6 \times 6 \) - a significant reduction from \( 160 \times 160 \) used to obtain the results in Section 12.4. Equations of motion for the elements of \( \rho_{\text{eff}} \) are given in Appendix D.2.

From the equations of motion, we can uncover terms which lead to the effect of quantum interference. For example, equations for the populations of the second manifold states, \( \rho_{22} \) and \( \rho_{33} \) depend on the populations \( \rho_{44} \) and \( \rho_{55} \) of the third manifold states and the coherences between these states \( \rho_{45} \) and \( \rho_{54} \); the latter with rate \( \Gamma_{45} \). At the same time, equations for \( \rho_{44} \) and \( \rho_{55} \) do not depend on the second manifold states, nor their mutual coherence. The same holds for the coherences \( \rho_{23} \) and \( \rho_{45} \) and their adjoints; the equation for \( \rho_{23} \) depends on \( \rho_{44}, \rho_{55}, \rho_{45} \) and \( \rho_{54} \), but not vice versa. Population of and coherence between the second manifold states is linked to the population of and coherence between the third manifold states. If this dependence is removed from the equations of motion, i.e., terms dependent on \( \rho_{44}, \rho_{55}, \rho_{45} \) and \( \rho_{54} \) are removed from the equations for \( \rho_{22}, \rho_{33}, \rho_{23} \) and \( \rho_{32} \), cancellation of the population in the second manifold ceases to occur.
The 35 equations of the effective model can, in principle, be solved analytically in the steady state. However, the resulting expressions are complicated and do not offer significant physical insight, so we have opted to perform numerical solutions of the equations given in Appendix D.2 and check for validity of the effective model. Having the solutions for the populations and coherences, the second order correlation function for zero time delay can be calculated as the ratio of

\[
\langle a^\dagger a^\dagger a a \rangle = |w_{01}|^2 |w_{12}|^2 \rho_{22} + |w_{01}|^2 |w_{13}|^2 \rho_{33} + \left[ |w_{12}|^2 |w_{24}|^2 + |w_{13}|^2 |w_{34}|^2 + (w_{12}^* w_{24}^* w_{13} w_{34} + \text{c.c.}) \right] \rho_{44} + \left[ |w_{12}|^2 |w_{25}|^2 + |w_{13}|^2 |w_{35}|^2 + (w_{13}^* w_{35}^* w_{12} w_{25} + \text{c.c.}) \right] \rho_{55} + \left( |w_{01}|^2 w_{12} w_{13} \rho_{34} + \text{c.c.} \right) + \left[ (|w_{12}|^2 |w_{24} w_{25}^* + |w_{13}|^2 w_{34} w_{35}^* + w_{12}^* w_{25}^* w_{13} w_{34} + w_{12} w_{24} w_{13}^* w_{35}^*) \rho_{45} + \text{c.c.} \right] ,
\]

(12.10a)

and the square of

\[
\langle a^\dagger a \rangle = |w_{01}|^2 \rho_{11} + |w_{12}|^2 \rho_{22} + |w_{13}|^2 \rho_{33} + \left( |w_{24}|^2 + |w_{34}|^2 \right) \rho_{44} + \left( |w_{25}|^2 + |w_{35}|^2 \right) \rho_{55} + \left( w_{12} w_{13}^* \rho_{23} + (w_{24} w_{25}^* + w_{34} w_{35}^*) \rho_{45} + \text{c.c.} \right)
\]

(12.10b)

where \(w_{ij} = \Omega_{ij}/\mathcal{E}_p\), and c.c. stands for complex conjugate.

The results are displayed in Figure 12.7. We have compared the numerical solutions of the effective model with the results of Section 12.3 and found a very good qualitative agreement. We do find strong bunching and threshold behaviour occurring at the same values of \(\mathcal{E}_p\) and \(\langle a^\dagger a \rangle_{ss}\). However, the effective model differs from the full simulations in the size of \(g^{(2)}(0)\) at its peak, by approximately a factor of 2. Including more states in the effective model would yield full agreement with the numerical data. In particular, we found that including two states closest to the resonance from up to eight manifolds reproduces the numerical data exactly. The reason is that the two–photon cascade decay responsible for the behaviour of \(g^{(2)}(0)\) can result from the decay of the higher states to third manifold states first, i.e. two–photon cascade can, in the manner of speaking, be driven “from below” and “from above”. Naturally, the decay of higher lying states introduces more single–photon transitions as well. Therefore, adding one manifold at a time to an effective model reveals that the increase in maximum value of \(g^{(2)}(0)\) gradually diminishes with new manifolds added, settling at its maximum value after the inclusion of the eighth manifold. On the other end, the effective model of Figure 12.6 is the smallest possible model which (at least qualitatively) reproduces the strong bunching effect in this system.

In Appendix C, we present the calculation of the second order perturbative corrections to the dressed states, with respect to the driving field. The results show the dependence of eigenenergies on the driving. These results show that the change in dressed state energies due to the contribution of driving cannot account for the change in photon statistics.
12.6 Dynamics of the Forward Scattering of Light

To obtain a different and useful perspective on the physical processes involved in the changing nature of the statistics of light emitted by the coupled atom–cavity system, we can split the field operator into contributions from a coherent mean amplitude and from an incoherent part [135],

\[ a = \alpha + \Delta a, \quad (12.11a) \]

where \( \alpha \equiv \langle a \rangle \) denotes the coherent amplitude component of the intracavity field, while \( \Delta a \) denotes the incoherent amplitude component \( \langle \Delta a \rangle = 0 \), whose emergence results from scattering of the cavity field by the atom. Using this decomposition, one can rewrite the

Figure 12.7: Comparison of the numerical results obtained from the effective model including three manifolds with the results of numerical simulations including up to eight manifolds. Details are described in the text. The parameters are the same as for the solid line in Figure 12.2.
expression for \( g^{(2)}(0) \) as

\[
g^{(2)}(0) - 1 = \frac{\langle a^\dagger a^\dagger a a \rangle_{ss}}{\langle a^\dagger a \rangle_{ss}^2} - 1 = \frac{\langle a^\dagger (\alpha^* \Delta a + \alpha \Delta a^\dagger)^2 \rangle_{ss}}{\langle |\alpha|^2 + \langle a^\dagger a a \rangle_{ss} \rangle^2}
\]

\[
+ \frac{4|\alpha| \Re \left[ \langle \Delta a^2 \Delta a \rangle_{ss} \right]}{\langle |\alpha|^2 + \langle a^\dagger a a \rangle_{ss} \rangle^2}
\]

\[
+ \frac{\langle \Delta a^2 \Delta a^2 \rangle_{ss} - \langle \Delta a^\dagger \Delta a \rangle_{ss}^2}{\langle |\alpha|^2 + \langle a^\dagger a a \rangle_{ss} \rangle^2}
\]

\[= S(\Delta a) + T(\Delta a) + V(\Delta a), \tag{12.11b}\]

where \( \langle a^\dagger a \rangle_{ss} = |\alpha|^2 + \langle a^\dagger a a \rangle_{ss} \). The three terms in this expansion, denoted by \( S, T \) and \( V \), have been identified by Carmichael [135] for the case of a two level atom. The decomposition (12.11) shows how the behaviour of \( g^{(2)}(0) \) for different values of driving field can be interpreted as the effect of self–homodyning between the coherent and incoherent components of the intracavity field [136]. From this viewpoint, it is easy to identify \( S(\Delta a) \) as a term describing the squeezing in the field quadrature in phase with the driving field, \( V(\Delta a) \) gives the variance in the incoherent component, and \( T(\Delta a) \) describes intensity–amplitude correlations in the incoherent component. Both \( V(\Delta a) \) and \( T(\Delta a) \) are determined by the correlations in the intensity, so the departure from the coherent value of the correlation function can be assigned to the effects of squeezing and the effects of intensity correlations.

The contributions from the incoherent component of the intracavity field are shown in Figure 12.8. The squeezing and intensity correlation parts are shown separately. The antibunching for weak fields comes from the squeezed fluctuations, which reduce the contribution from the coherent scattering. As the driving increases, the squeezing decreases, but the variance in intensity fluctuations becomes negative, so the remaining antibunching comes from the sub–Poissonian intensity fluctuations in the incoherent component of the field. For \( \mathcal{E}_p \sim \kappa \), the squeezing contribution effectively vanishes, while \( T \) and \( V \) become positive, and antibunching disappears. Strong bunching clearly originates in the super–Poissonian intensity correlations, and the correlation function is dominated by \( V \).

It is interesting to compare these findings with the case of resonance fluorescence, i.e. the case of a single four level atom in free space, driven by a coherent laser field instead of cavity mode. Full treatment of this case, including the calculation of the second order correlation function and the analysis of incoherent scattering, is presented in Appendix E. It is shown that antibunching in two cases arises from a different mechanism.

\[\text{1This becomes obvious if a similar decomposition is performed for finite delay time } \tau. \text{ Then, term } T \text{ is proportional to } \langle \Delta a^\dagger (\tau) \Delta a^\dagger \Delta a(0) + \Delta a^\dagger \Delta a(\tau) \Delta a(0) \rangle_{ss}. \text{ Taking the limit } \tau \to 0 \text{ recovers the expression (12.11b).}\]
12.7 Nonclassical Behaviour of the Correlation Function

The presence of nonclassical effects in a driven atom–cavity system is a topic that has received much attention for many years. The system usually studied has been the canonical system of quantum optics – a single two level atom confined in an optical cavity. Photon statistics in the bad–cavity limit was thoroughly studied by Rice and Carmichael [79], who analyzed the sub–Poissonian statistics and photon antibunching in the cavity transmission, for the case of weak driving. Their analysis was extended by Carmichael et al. [137] to a system containing $N$ two level atoms. This analysis was further refined by Brecha et al. [138]. Clemens and Rice [139] have extended the consideration involving single atom to include arbitrary driving field strength and dephasing. In their analysis, Clemens and Rice pay special attention to nonclassical effects known as ‘undershoots’ and ‘overshoots’. These are related to the violation of inequalities that hold for classical correlations, in particular violations that occur not in the value of $g^{(2)}(\tau = 0)$, but for certain time delays $\tau > 0$. The explanation for the undershoots has been given by Carmichael et al. [137] in terms of quantum interference of probability amplitudes and collapse of the wavefunction.

The requirements for the classicality of the field correlations can be derived from the
Cauchy–Schwartz inequality (see [138] and references therein), and expressed in terms of
the second order correlation function as

$$|g^{(2)}(\tau) - 1| \leq |g^{(2)}(0) - 1|.$$  \hspace{1cm} (12.12)

Values in excess of those allowed classically are called overshoots, while values below are
called undershoots. Overshoots have been observed recently by Mielke et al. [140].

In this context, it is of interest to see if the overshoots and/or undershoots can be found
in the single–atom EIT–Kerr system under consideration. Photon antibunching, as an ex-
ample of nonclassical photon statistics, has already been predicted (Chapter 6 and [102]),
and the effective two level behaviour analyzed (Parts III and IV). In Section 12.6, we
have shown how the effects of self–homodyning of squeezed dipole radiation yields photon
antibunching in the low to moderate driving limit. We have also shown (Sections 12.4
and 12.5) that quantum interference between the probability amplitudes contributes to
both strong antibunching and strong bunching, for weak and strong driving fields, re-
spectively. Given this range of behaviours, we might therefore expect undershoots and
overshoots to also occur in the single–atom EIT–Kerr system under suitable conditions.
Figure 12.9 shows correlation functions for several values of driving field strength. The values of driving have been chosen where nonclassical behaviour is expected to be found. For weak driving, where the antibunching is strong, the delay–time dependence of the correlation function is well-understood in terms of the effective two level system. The interesting region is for those values of driving for which $g^{(2)}(0)$ increases through 1, the value for a coherent field. This is also the region in which the dynamics is well-described in terms of quantum interference and increased incidence of two-photon emissions.

Not surprisingly, this is also the range of parameters where largest violations of the classical inequalities occur. Since the increase in the value of $g^{(2)}(0)$ is due to the purely quantum effect of interference between probability amplitudes, non-classical behaviour of the correlations can be expected. The explanation of these effects given by Rice and Carmichael [79] and Carmichael et al [137], although in a different context, still holds. As shown in Section 12.6, self-homodyning of squeezed dipole radiation with the driving field occurs in the EIT–Kerr system in a similar manner to that for a two level atom. An alternative explanation in terms of quantum interference of the driving field with the atomic polarization after the collapse of the wave function upon a photon detection event offers even more insight. This is best understood in the context of quantum trajectory theory.

This explanation is illustrated in Figure 12.10, where single trajectory realizations for the intracavity field and photon number are plotted. We see that the collapses tend to occur in succession before the system returns to a (quasi–) steady state (which for this region of parameters happens after time $\gamma_j^{-1}$). Once it returns into a steady state, a few Rabi cycles pass before the next set of collapses occur. The value of $g^{(2)}(0)$ is determined by the ratio of number jumps upwards to number of jumps downwards in photon number, where jump upwards suggests that the detection of a photon increases the probability of detecting a second photon immediately afterwards. Naturally, at the value of driving where $g^{(2)}(0)$ peaks (see Figure 12.2), collapses are almost exclusively upwards, as illustrated in Figure 12.11. Undershoots appear as the consequence of a change in sign that the amplitude undergoes at the collapse [137]. Polarization in the collapsed state becomes close in magnitude and opposite in sign to the driving field, for a near–zero mean intracavity field, leading to the reduced detection probability for a second photon.

The overshoots can be explained in terms of the collapses of the wave function. The detection of the first photon, emitted from the steady state situation, collapses the wave function of the system. The subsequent time evolution as the system returns to the steady state determines the photon correlations. For weak driving field, the probability for the second collapse to occur before the system returns to the steady state is extremely small, since it is proportional to the mean intracavity photon number. For stronger driving fields the probability for subsequent collapses increases, specifically due to the large correlations between first and third manifold states, as shown in Figure 12.5. Therefore, the probability for a second photon detection after some time $\tau < \gamma^{-1}$ increases as well, causing the correlation function overshoot. In experiment, such event pairs are the source of delayed coincidence counts. A third collapse is also likely to occur before the steady state is
reached. However, the overshoot disappears (or significantly decreases) for all time delays \( \tau \) after the first peak. This is expected, since the exact form of the wave function after the second collapse depends on the delay time between the second and third photon. Averaging over all possible realizations washes out the nonclassical effects due to different possible evolutions following the second collapse.

A stronger driving field causes more subsequent collapses to occur, and nonclassical correlations are completely washed out. We find that overshoots and undershoots vanish at driving strength \( \mathcal{E}_p \approx 1.14\kappa \).

### 12.8 Conclusion

We have presented an analysis of the properties of the photon statistics of light emitted by a single atom intracavity EIT–Kerr system. It was found that the statistics change abruptly as the driving field strength increases. Specifically, strong photon antibunching...
12.8. CONCLUSION

Figure 12.11: Intracavity photon number in a typical realization of a single quantum trajectory, for $\mathcal{E}_p/\kappa = 1.75$.

ing, dominant in the weak to moderate driving regimes, is replaced by a strong photon bunching in the output field for the stronger driving. We have identified the effect of quantum interference between the dressed states to be responsible for this sudden change, and presented an effective model explaining qualitative features of this behaviour.

Furthermore, we have analyzed contributions from the incoherent scattering to the system dynamics and found that the strong photon antibunching can be explained in terms of the self-homodyning of the incoherent intracavity component with the coherent component for the weak driving, and in terms of reduced intensity fluctuations for moderate driving strengths. Strong bunching is the signature of super-Poissonian intensity fluctuations.

Finally, nonclassical behaviour of the correlation function was found, and the effects of undershoots and overshoots analyzed.
Conclusion

This last Chapter contains a summary of the work presented in this thesis and is intended to provide a synthesis of the conclusions of each individual Chapter into one coherent unit. The summary is followed by an outlook on future research which is suggested by the results obtained in this thesis.

Summary

The work in this thesis has been devoted to a systematic theoretical analysis of a giant Kerr nonlinearity in an atomic medium. The atomic energy level scheme is assumed to have four energy levels, interacting with a two light fields. One field (the “coupling laser”) is treated classically, while the other field (the “driving field”) couples to one mode of the optical cavity (treated quantum mechanically) containing the atomic medium. The output of the cavity mode was analysed by means of its photon statistics. In that respect, two extreme cases were considered in detail. First, a setup containing a large number of atoms ($10^9$) in a conventional optical cavity, and second, a setup involving only a single atom in a high finesse optical microcavity.

The system containing a large number of atoms was analysed in Chapter 3, from the viewpoint of quadrature squeezing. In particular, the quadrature squeezing spectrum of the full system (including both field and atoms) was analysed using the linearised fluctuation approximation. The results were compared with those obtained for an ideal dispersive $\chi^{(3)}$ nonlinearity in a cavity. While the maximum degree of squeezing was found to be comparable, the linewidth of the effect was found to be vastly different, being about 50 times narrower in the full atom–cavity system. This is an important conclusion, pointing at a general feature of systems which use quantum coherence effects (such as EIT in this case) as a means to reduce or eliminate unwanted noise contributions. The adiabatic approximation, usually used to reduce the system containing a large number of atomic and field degrees of freedom to a simpler system containing only field (or atomic) degrees of freedom, is not always the appropriate procedure in nonlinear systems exhibiting noise reduction. The exact condition for the validity of the adiabatic approximation has been found. Based on this insight, in Chapter 4 the linewidth narrowing and related effects were discussed in some detail. Using the expression for linear susceptibility, it was found that the condition enabling the adiabatic elimination of the atoms correspondingly reduces the steep linear dispersion, but also reduces the size of the effective optical nonlinearity.
To resolve these problems, an alternative setup was proposed, consisting of a single four level atom strongly coupled to a single mode of an optical cavity. The physical background of single atom cavity QED was reviewed in Chapter 5, establishing the theoretical formulation of a single atom limit and reviewing experimental achievements in this field. In Chapter 6 the problem of a single four level atom strongly coupled to a cavity mode was formulated, and full numeric solutions of the master equation were presented for a set of parameters consistent with those experimentally available. These numerical solutions show that it is indeed possible to realise a photon blockade with time response (i.e., linewidth) comparable to that of an ideal, pure $\chi^{(3)}$ nonlinearity. It was also inferred from the numerical results that the time scale of atomic and field variables is approximately the same, thus showing explicitly that the adiabatic approximation is not appropriate in this regime. Following the numerical analysis, in Chapter 7 an effective model was used to explain the numerical results. A dressed states analysis of the first and second excited manifolds of the atom–cavity system was performed, and the effective Hamiltonian formulated in terms of operators switching between the dressed states (polariton operators). An interesting effect emerged from the effective Hamiltonian, namely a reduction of effective (polariton) nonlinearity by a group velocity reduction factor. This effect provides a continuous passage from the slow–light regime \cite{75} to stored–light \cite{111, 112} case: latter is (ideally) a “frozen” state where a concept of the effective nonlinearity is obsolete.

Armed with these results, it was possible to perform a thorough comparison between photon blockade in a four level EIT–Kerr model and photon blockade exhibited by a single two level atom in a similar cavity QED environment, as reviewed in Chapter 8. In particular, a polariton model, equivalent to the one developed in Chapter 7, was formulated for the two level atom case. It was found that a careful description in a photon blockade regime requires a formulation in terms of an effective two manifold model (as opposed to a more naïve two level model), which then permits analysis of the second order correlation function. Following this review, Chapter 9 compares the effectiveness of a two level scheme as a photon blockade mechanism with the four level scheme. It was found that the benefits of a four level scheme over the two level scheme are based on two cases of quantum interference effects. The first is customary EIT, leading to the cancellation of spontaneous emission from the excited state, while the second effect occurs between dressed–state transitions, eliminating the excitation probability from the first to the second excitation manifold. This second case of interference is a central finding of this thesis emphasizing that the most appropriate way of thinking about such strongly coupled systems is in terms of eigenstates of the interaction Hamiltonian. These are necessarily composite states consisting of both atomic and field contributions. This way of thinking also clarifies when it is appropriate to use the adiabatic approximation - for example, when one of the components (say, field) dominates the dressed states, it is appropriate to regard the other (say, atoms) as perturbing the properties of the first.

Finally, in Chapter 9 it was also shown how the introduction of additional atoms to the cavity reduces the effectiveness of photon blockade in both models. However, while nothing can be done about this in the two level case, it is found that increasing a particular atomic detuning in the EIT–Kerr case restores the effect.
Chapter 10 continues with the emphasis on dressed states, with the formulation of the complete analytical solution (for arbitrary manifold) in terms of polariton operators. The simplest example of the usefulness of this solution was then given in Chapter 11, through the analysis of dynamic Stark splitting of dressed states due to the external driving. The influence of external driving was further analysed in Chapter 12, where it was found that a sudden change in photon statistics occurs for a certain value of intracavity intensity. Qualitative analysis of this effect was given in terms of another effective model based on the polariton picture, and quantum interference between polariton transitions was identified as being responsible for the switch in behaviour.

Outlook

Giant Kerr nonlinearity has emerged in recent years as an extremely desirable “tool”, especially in the field of quantum information processing. This is the field where most of the work presented in this thesis is envisaged to have its application. A list of possible applications follows.

Quantum Phase Gates and Conditional Quantum Dynamics. A useful visualisation of the photon blockade effect is through the energy shift that Kerr nonlinearity induces in the second manifold states. It is explained in this thesis how the harmonic spectrum of the empty cavity acquires an anharmonic term due to the presence of nonlinearity. This anharmonicity causes the blockade of any photons subsequent to the first one exciting the atom–cavity system. Only after the first photon leaves can the cavity be re-excited by a subsequent photon. The presence of the anharmonic term is manifested in the form of the phase shift of the photon leaving the cavity, a phase shift that is proportional to the size of the anharmonicity, (i.e., nonlinearity). Given the giant size of the nonlinearity, phase shifts of order $\pi$ are expected to be possible. To realise the conditional dynamics, two orthogonal modes $a$ and $b$ can be used. The goal is to implement particular dynamics for the quantum outputs conditioned on the quantum inputs to the system. In the case of two orthogonal modes, the key is to obtain a cross–Kerr nonlinearity, i.e. effective behaviour corresponding to an $a^\dagger ab^\dagger b$ interaction term in the Hamiltonian. Preliminary calculations have shown that it is indeed possible to obtain this type of effective interaction using the EIT–Kerr system.

Quantum State Engineering. If the system is driven by an optical pulse of appropriately chosen area, the photon blockade effect can be used to prepare a state of the cavity field mode that is an arbitrary superposition of the Fock states $|n = 0\rangle$ and $|n = 1\rangle$. Furthermore, it should be possible to extend this idea to produce arbitrary predefined superpositions of the cavity field mode. The idea is to use a classical driving that is composed of many frequencies, each frequency chosen to match a given transition between the photon number states (i.e., manifolds) $|n – 1\rangle$ and $|n\rangle$.

Generation of Schrödinger Cat States. Yurke and Stoler [141] have proposed to generate Schrödinger Cat States using the intracavity dispersive Kerr nonlinearity. It was shown that if the coherent state $|\alpha\rangle$ is considered at the input, the output state will oscil-
late in time between $|\alpha\rangle$ and $|-\alpha\rangle$, and at certain times will have the form of $|\alpha\rangle + e^{i\phi} |-\alpha\rangle$. To distinguish this cat state from the statistical mixture, a scheme was proposed involving homodyne detection, to look for interference fringes in the quadrature phase distribution. Given the flexibility offered by the EIT–Kerr system, it is of interest to consider how (and if) the cat states could be produced and measured in the environment described in this thesis. The EIT–Kerr system is especially suitable for such considerations, given that the major obstacle for successful production of cat states lies in the interaction with the dissipative environment causing the destruction of quantum coherence (see Gerry and Knight [142] for an excellent review).

Quantum Teleportation. A direct application of giant Kerr nonlinearity can be found in realisations of the quantum teleportation protocol [143, 144]. The protocol involves Bell state measurement which projects the unknown state of the combined system into one of the four pure states, distinguished from the original state by a simple unitary transformation. In order to obtain an exact copy of the unknown state, the experimenter then has to perform the appropriate unitary operation on the received state, which can be performed using conventional linear optical elements. The Bell state measurement itself, however, requires large Kerr nonlinearity. Thus the large Kerr nonlinearity becomes a necessary element for the successful realisation of quantum teleportation protocol.
Part VI

Appendices
Appendix A

Dressed States

In this Appendix we present details of the eigenstate analysis of the Hamiltonian (6.1).

For the bare states, we use the notation

$$|j; i, n, 1, 2, 3, 4_i, j = 1, 2, 3, 4, 4, \ldots; 7\rangle$$

The bare states $$|n+1, 1, 2, 3, \ldots, n, 4\rangle$$ are coupled by the Hamiltonian $$H_{\text{system}}$$ which may be diagonalised in each submanifold $$n$$ (in the interaction picture) to give the relevant dressed states. The matrix representation of $$H_{\text{system}}$$ for a given submanifold is

$$H_{\text{sys}} = \begin{pmatrix}
0 & -ihg_1 \sqrt{n} & 0 & 0 \\
ihg_1 \sqrt{n} & h \delta & -ih \Omega_c & 0 \\
0 & ih \Omega_c & 0 & -ihg_2 \sqrt{n} - 1 \\
0 & 0 & ihg_2 \sqrt{n} - 1 & h \Delta
\end{pmatrix}. \quad (A.1)$$

For $$n = 1$$, dressed states can be calculated exactly by diagonalising the $$3 \times 3$$ matrix, obtained by ignoring the lowest row and last column of the matrix (A.1). This reflects the fact that starting with only one photon in the cavity, one cannot reach the atomic energy level $$|4_i$$, and therefore the matrix element $$h \Delta$$ does not come into consideration. The result is given by Eqs. (7.3a, 7.3b) and (7.4a, 7.4b).

For $$n = 2$$, eigenstates are given by

$$|e_j\rangle = \alpha_j |2, 1\rangle + \beta_j |1, 2\rangle + \mu_j |1, 3\rangle + \nu_j |0, 4\rangle; \quad j = 4, \ldots, 7, \quad (A.2)$$

Energy shifts $$\epsilon_j$$ are calculated in second order perturbation theory with the assumption of small detunings compared to $$\Omega_c$$. This is a reasonable assumption, given that Eq. (6.5) yields large values for nonlinearity only for small atomic detunings. For the range of parameters used in our simulations, it is reasonable to truncate the perturbation series after second order. The energy eigenvalues follow as

$$E_j = 2h \omega_{av} + \epsilon_j; \quad (A.3)$$

$$\epsilon_j \approx \epsilon_j^{(0)} + c_{jj} \sum_{k \neq j} \frac{|c_{kj}|^2}{\Delta \epsilon_{j,k}}, \quad j, k = 4, \ldots, 7 \quad (A.4)$$
where

\[
\epsilon_{4,5}^{(0)} = -\frac{\hbar \Omega_c}{\sqrt{2}} \sqrt{1 + \left( \frac{g_1}{\Omega_c} \right)^2 + \left( \frac{g_2}{\Omega_c} \right)^2 \pm \sqrt{\left( 1 + \frac{g_1^2}{\Omega_c^2} + \frac{g_2^2}{\Omega_c^2} \right)^2 - \left( 2 \frac{g_1 \cdot g_2}{\Omega_c^2} \right)^2}}, \quad (A.5a)
\]

\[
\epsilon_{6,7}^{(0)} = -\epsilon_{5,4}^{(0)}. \quad (A.5b)
\]

The coefficients \( c_{kj} \) are defined as usual in perturbation theory,

\[
c_{kj} = \langle e_k^{(0)} | H_{\text{per}} | e_j^{(0)} \rangle = \hbar \Delta \left\{ 1 + \frac{\delta}{\Delta} \left( \frac{g_2}{\Omega_c} \right)^2 \left[ 1 - \left( \frac{\epsilon_k^{(0)}}{\hbar g_2} \right)^2 \right] \left[ 1 - \left( \frac{\epsilon_j^{(0)}}{\hbar g_1} \right)^2 \right] \right\}, \quad (A.6)
\]

\[
\epsilon_j^{(0)} = \left\{ 1 + \left( \frac{\epsilon_j^{(0)}}{\hbar g_2} \right)^2 + \frac{\hbar g_1}{\epsilon_j^{(0)}} \left[ 1 + \left( \frac{\epsilon_j^{(0)}}{\hbar g_1} \right)^2 \right] \right\}^{-\frac{1}{2}} \quad (A.7)
\]

where

\[
c_j^{(0)} = \left\{ 1 + \left( \frac{\epsilon_j^{(0)}}{\hbar g_2} \right)^2 + \frac{\hbar g_1}{\epsilon_j^{(0)}} \left[ 1 + \left( \frac{\epsilon_j^{(0)}}{\hbar g_1} \right)^2 \right] \right\}^{-\frac{1}{2}} \quad (A.8)
\]

In the above expression, \( |e_k^{(0)} \rangle \) represent unperturbed eigenkets. The energy level structure is shown in Fig. 7.1.

The coefficients of the bare states in \( |e_j \rangle \) can be calculated straightforwardly from (A.1) to give

\[
\alpha_j = -i \frac{g_1 \cdot g_2 \sqrt{2}}{\epsilon_j \Omega_c} \left( 1 - \frac{\epsilon_j (\epsilon_j - \hbar \Delta)}{\hbar g_2^2} \right) \nu_j, \quad (A.9a)
\]

\[
\beta_j = \frac{g_2}{\Omega_c} \left( 1 - \frac{\epsilon_j (\epsilon_j - \hbar \Delta)}{\hbar g_2^2} \right) \nu_j, \quad (A.9b)
\]

\[
\mu_j = -i \frac{\epsilon_j - \hbar \Delta}{\hbar g_2} \nu_j, \quad (A.9c)
\]

\[
\nu_j = \left\{ 1 + \left( \frac{\epsilon_j - \hbar \Delta}{\hbar g_2} \right)^2 + \frac{\hbar g_1}{\epsilon_j \Omega_c} \left[ 1 + 2 \left( \frac{\hbar g_1}{\epsilon_j \Omega_c} \right)^2 \right] \right\}^{-\frac{1}{2}} \quad (A.9d)
\]

The above expressions were used to produce Fig. 7.2.
Appendix B
Nonlinearity and Anharmonicity

B.1 Comparison of Nonlinearities

We compare the magnitude of the Kerr nonlinearity of the two level atoms and four level atoms in the perturbative and nonperturbative regimes (see Figure B.1). To that end, we will replace the cavity mode with a travelling classical laser field with Rabi frequency $g$ ($g_1$ and $g_2$ in the case of EIT scheme). First we calculate the general expression for the nonlinearity in the JC model. Defining $w = 2\langle \sigma_z \rangle$, $u = \pm \text{Im}[\langle \sigma_\pm \rangle]$ and $v = \text{Re}[\langle \sigma_\pm \rangle]$, the following set of equations is found (in the low driving limit)

\begin{align}
\dot{w} &= -\gamma(w + 1) - 4gu, \\
\dot{u} &= -\frac{\gamma}{2}u - \theta v + gw, \\
\dot{v} &= -\frac{\gamma}{2}v + \theta u.
\end{align}

(B.1a, B.1b, B.1c)

Here, $\theta = \omega_0 - \omega_L$ is the detuning of the laser field from the atomic resonance. These equations are then solved in the steady state, using the Laplace transform method. The nonlinear susceptibility is proportional to $v - v_{lin}$

\begin{align}
v &= \frac{-g^2\theta}{(\gamma/2)^2 + \theta^2 + 2g^2}, \\
v_{lin} &= \frac{-g^2\theta}{(\gamma/2)^2 + \theta^2}.
\end{align}

(B.2, B.3)

Here, $v$ is the steady state polarisation obtained from the equations (B.1), and $v_{lin}$ is the linear part of the polarization obtained by applying first–order perturbation theory. We find the general expression for the nonlinearity in the JC scheme,

\[ \text{Re}[\chi^{NL}] = \frac{N}{\epsilon_0 h^3} \frac{2g^2\theta}{(\gamma/2)^2 + \theta^2} \frac{g^2}{(\gamma/2)^2 + \theta^2 + g^2}. \]

(B.4)

In the perturbative limit ($g \ll \gamma, \theta$) the nonlinearity given by the expression (B.4) has a strong dependence on the detuning. In order to suppress linear absorption, or equivalently,
APPENDIX B. NONLINEARITY AND ANHARMONICITY

atomic spontaneous emission, the two level atom has to be driven off-resonance, but this detuning in turn reduces the size of nonlinearity as $\theta^3$. The linear absorption on the other hand decreases as $\theta^2$.

Taking $\delta = 0$ in the EIT-Kerr scheme, a perturbative result for the third-order nonlinear susceptibility has been calculated by Schmidt and Imamoğlu [47]

$$\text{Re} \left[ \chi^{(3)} \right] = N \frac{g_1^2 g_2^2}{2\epsilon_0 \hbar^3} \frac{1}{\Omega_c^2 \Delta}.$$ (B.5)

In the ideal EIT-Kerr scheme where the dephasing rate of the $|2\rangle \rightarrow |1\rangle$ (hyperfine) transition ($\gamma_{21}$) vanishes, there is no linear absorption. The finite two-photon detuning $\Delta$ from state $|4\rangle$ leads to two-photon absorption; however this loss mechanism, which scales as $\Delta^{-2}$, can be minimized by increasing $\Delta$. In principle, we are free to reduce $\Omega_c$ to achieve a large nonlinear susceptibility, provided that $\Omega_c^2 \gg \gamma_3 \gamma_{21}$. Therefore, in the perturbative limit, one can obtain ultralarge values of nonlinearity with the EIT-Kerr scheme while keeping the loss negligible; recent experiments by Hau et al. have demonstrated a $10^6$ improvement in Kerr nonlinearity using this scheme.

To explore the nonperturbative regime, one takes the limit $g \gg \gamma, \theta$ in the Eq. (B.4): the nonlinearity obtained in this case is given by\(^1\)

$$\text{Re} \left[ \chi^{NL} \right] \propto \frac{g^2}{\theta}.$$ (B.6)

Obviously large nonlinearities can be obtained in this limit as compared to the perturbative case. The nonperturbative nonlinearity in the EIT-Kerr scheme can be calculated from the coupled amplitude equations (assuming $\delta = 0$ and in the low driving limit)

\begin{align*}
\dot{a}_1 &= ig_1 a_3, \\
\dot{a}_2 &= i\Omega_c a_3 + ig_2 a_2, \\
\dot{a}_3 &= -i\frac{\gamma_2}{2} a_3 + ig_1 a_1 + i\Omega_c a_2, \\
\dot{a}_4 &= -i \left( \frac{\gamma_3}{2} + i\Delta \right) a_4 + ig_2 a_2.
\end{align*} \quad (B.7a-7d)

The nonlinearity is proportional to $v_{EIT} = (a_1^* a_2 + a_2^* a_4)/2$, giving

$$\text{Re} \left[ \chi^{(3)} \right] = N \frac{g_1^2 g_2^2}{2\epsilon_0 \hbar^3} \frac{g_1^2}{\Omega_c^2 + g_1^2}.$$ (B.8)

Comparing Eq. (B.8) with expression (B.6), we conclude that, in the nonperturbative limit, the nonlinearities of the two schemes are similar. We conclude that the EIT-Kerr scheme exhibits giant nonlinearities (as compared to conventional schemes such as the JC scheme) only in the perturbative limit where the atom-field coupling is much smaller than all the other relevant energy scales.

\(^1\)Maximum nonlinearity in this regime becomes $\propto 2g/\gamma$. 

The Hamiltonian describing a single-mode quantum field interacting with a dispersive Kerr medium will be that of a harmonic oscillator with the added anharmonic term $\hbar \eta (a^\dagger)^2 a^2$, where the parameter $\eta$ is called the anharmonicity parameter. We now establish the connection between nonlinearity, as calculated above, and anharmonicity, which is the parameter of interest from the viewpoint of photon blockade.

**B.2 Anharmonicity**

There are two established ways of calculating the anharmonicity. One is to calculate the effective optical nonlinearity $\chi^{(3)}$ (as above), and then evaluate the Hamiltonian density as

$$\mathcal{H} = \frac{1}{2} \mathbf{D} \cdot \mathbf{E},$$

where

$$\mathbf{D} = \epsilon_0 \mathbf{E} + \hat{\mathbf{P}},$$

and $\hat{\mathbf{P}}$ is a polarization operator expansion [68], where we assume vanishing $\chi^{(2)}$ contributions. $\mathbf{E}$ is the electric field operator. This is the procedure used in [51] to obtain the expression for anharmonicity, and we will use this EIT–Kerr system calculation as an illustration. The relationship between nonlinearity and anharmonicity is given by

$$\eta = \frac{3}{2} \left( \frac{\omega^2}{V} \right) \text{Re} [\chi^{(3)}],$$

$$= 3 \frac{g_1^2 g_2^2}{\Omega_0^2 \Delta} N.$$  

(B.11)

The underlying assumption of this approach is that for all practical purposes atoms contribute to the total dynamics of a cavity field mode by effectively adding the anharmonic term to the otherwise harmonic Hamiltonian of the mode. The total energy of the cavity mode can then be described by this Hamiltonian operator.

The other approach is the adiabatic elimination of the atomic degrees of freedom, provided that the adiabatic condition [55, 56, 63] is satisfied. In that approach, we first derive the Itô quantum stochastic differential equations (QSDE) of motion for the field and atomic variables, and then adiabatically eliminate atomic degrees of freedom. This leaves us with the QSDE for the field operator alone. From this equation, we can deduce the effective adiabatic Hamiltonian, which has the same anharmonic structure as the one obtained by the first method\(^2\). This has been done by Rebić et al. [102] and Gheri et

\(^2\)Equivalently, one can also derive an Itô QSDE from the Hamiltonian obtained by the first method and compare anharmonic terms. We also omit the discussion of an additional linear dispersion found by the adiabatic method – see [63].
al. [63] who both obtained the same result for anharmonicity (valid for $\gamma_3 \ll \Delta, \delta = 0$),

\[ \eta = \frac{g_1^2 g_2^2}{\Omega^2 \Delta} N, \]  

(B.12)

smaller by factor of three than the one obtained by the first method (B.11). Which result do we believe?

The effective Hamiltonian obtained by the first method is the energy operator for the cavity field mode in the presence of a dispersive optical Kerr nonlinearity, as given by the expressions calculated above. Therefore, the energy eigenstates and eigenvalues of such a system will clearly be shifted by the amounts corresponding to this anharmonicity. In that respect, this is the true anharmonicity, measuring the amount by which the Hamiltonian becomes anharmonic if the cavity mode interacts with the Kerr nonlinearity. However, this approach neglects every other contribution that a realistic atomic system might make.

The adiabatic method leads to an effective Hamiltonian that cannot be interpreted as an energy operator, but rather as a tool for calculating the equations of motion. These equations contain, provided that the adiabatic assumption is fully satisfied, all information on the manner in which an atomic medium adiabatically affects the cavity mode, such as the effective contribution of atomic noise, changes in photon statistics and additional harmonic and/or anharmonic corrections. These equations should be used for calculation of quantities such as field correlations and waiting time distributions.
B.2. ANHARMONICITY

Figure B.1: Energy level schemes in (a) Jaynes-Cummings and (b) EIT-Kerr schemes. $g$, $g_1$, $g_2$ are atom-field coupling constants, $\theta$ is the detuning from the atomic transition in the JC scheme, while $\delta$ and $\Delta$ are the detunings of the field modes from the atomic resonance. $\Omega_c$ is the Rabi frequency of the classical coupling field, and the $\gamma_j$’s are spontaneous emission decay rates for the given decay channel.
Appendix C

Second–Order Perturbation Corrections

In this Appendix, we calculate perturbative corrections to the eigenenergies due to the presence of the driving field. In the limit $\mathcal{E}_p \to 0$, we recover two resonant dressed states. As discussed in Chapter 11, resonant coupling via the driving field leads to the dynamic Stark splitting, i.e. the removal of degeneracy requires the replacement of the two resonant polariton states by their symmetric and antisymmetric linear combinations. Which of these two pairs of states then constitutes the “proper” basis for the perturbation expansion? Taking the limit $\mathcal{E}_p \to 0$ restores the degeneracy in energy of the Rabi split states, but the eigenstates remain coupled. Thus, the proper basis for the analysis of the driving field dependence is the basis of Rabi split states, given by Equation (11.4). As a consequence, the polariton states basis remains largely the same, with the exception of the subspace spanned by the two resonantly coupled eigenstates. The basis of that subspace is therefore replaced with the new basis

$$\left\{ \left| \psi^{(0)}_+ \right\rangle, \left| \psi^{(0)}_- \right\rangle \right\} \rightarrow \left\{ \left| \psi^{(0)}_+ \right\rangle, \left| \psi^{(0)}_- \right\rangle \right\} ,$$

obtained from the old one by a simple rotation, as shown in Figure 11.6.

Since the driving field couples states from the adjacent manifolds, first order perturbative corrections vanish. Second order corrections to the Stark states are given by

$$\tilde{\epsilon}_\pm = -\left| \frac{\langle e^{(1)}_+ | \mathcal{H}_d | \psi^{(0)}_\pm \rangle}{\hbar\epsilon^{(1)}_\pm} \right|^2 - \left| \frac{\langle e^{(1)}_- | \mathcal{H}_d | \psi^{(0)}_\pm \rangle}{\hbar\epsilon^{(1)}_-} \right|^2$$

$$- \sum_{k=1}^4 \left| \frac{\langle e^{(2)}_k | \mathcal{H}_d | \psi^{(0)}_\pm \rangle}{\hbar\epsilon^{(2)}_k} \right|^2 ,$$

so the total energy of the Stark–split states to second order in perturbation is

$$\epsilon_\pm \approx \epsilon^{(0)}_\pm + \tilde{\epsilon}_\pm .$$

Note that we denote perturbative corrections by $\tilde{\epsilon}$. 
APPENDIX C. SECOND-ORDER PERTURBATION CORRECTIONS

Required matrix elements can be easily calculated as

\[
\langle e_{\pm}^{(1)} | \mathcal{H}_d | \psi_{\pm}^{(0)} \rangle = -i\hbar \mathcal{E}_p \frac{\alpha_{\pm}^{(1)}}{\sqrt{2}}, \quad (C.4a)
\]

\[
\langle e_{-}^{(1)} | \mathcal{H}_d | \psi_{\pm}^{(0)} \rangle = -i\hbar \mathcal{E}_p \frac{\alpha_{-}^{(1)}}{\sqrt{2}}, \quad (C.4b)
\]

\[
\langle e_{k}^{(2)} | \mathcal{H}_d | \psi_{\pm}^{(0)} \rangle = \mp i\hbar \mathcal{E}_p \left( \alpha_{k}^{(2)*} \alpha_{0}^{(1)} + \frac{1}{\sqrt{2}} \mu_{k}^{(2)*} \mu_{0}^{(1)} \right). \quad (C.4c)
\]

and

\[
\langle e_{\pm}^{(1)} | \mathcal{H}_d | e_{k}^{(2)} \rangle = i\hbar \mathcal{E}_p \left( \sqrt{2} \alpha_{\pm}^{(1)*} \alpha_{k}^{(2)} + \beta_{\pm}^{(1)*} \beta_{k}^{(2)} + \mu_{\pm}^{(1)*} \mu_{k}^{(2)} \right) \quad (C.5a)
\]

\[
\langle e_{j}^{(3)} | \mathcal{H}_d | e_{k}^{(2)} \rangle = -i\hbar \mathcal{E}_p \left[ \sqrt{3} \alpha_{j}^{(3)*} \alpha_{k}^{(2)} + \sqrt{2} \right. \\
\left. \times \left( \beta_{j}^{(3)*} \beta_{k}^{(2)} + \mu_{j}^{(3)*} \mu_{k}^{(2)} \right) + \nu_{j}^{(3)*} \nu_{k}^{(2)} \right]. \quad (C.5b)
\]

With the appropriate expressions in Equation (C.2) evaluated, this gives

\[
\epsilon_{\pm} \approx \epsilon_{\pm}^{(0)} - \frac{\mathcal{E}_p^2}{2} \left[ \frac{(\alpha_{\pm}^{(1)})^2}{\epsilon_{\pm}^{(1)}} + \frac{(\alpha_{-}^{(1)})^2}{\epsilon_{-}^{(1)}} + \sum_{k=1}^{4} \frac{(\alpha_{k}^{(2)*}) \alpha_{0}^{(1)} \sqrt{2} + \mu_{k}^{(2)*} \mu_{0}^{(1)}}{\epsilon_{k}^{(2)}} \right]^2. \quad (C.6)
\]

Corrections to the other dressed states can be calculated in a similar manner. Since these states are all non-degenerate, the first order in the perturbation expansion vanishes, and the second order gives the lowest corrections for the energy levels. The remaining first manifold energies are then given by \( \epsilon_{\pm}^{(1)} + \epsilon_{\pm}^{(2)} \), and second manifold energies by \( \epsilon_{k}^{(2)} + \epsilon_{k}^{(2)} \).

Here

\[
\hbar \epsilon_{\pm}^{(2)} = \frac{\left| \langle \psi_{\pm}^{(0)} | \mathcal{H}_d | e_{\pm}^{(1)} \rangle \right|^2 + \left| \langle \psi_{\pm}^{(0)} | \mathcal{H}_d | e_{\mp}^{(1)} \rangle \right|^2}{\hbar \epsilon_{\pm}^{(1)}} \\
+ \sum_{k=1}^{4} \left( \frac{\left| \langle e_{k}^{(2)} | \mathcal{H}_d | \psi_{\pm}^{(0)} \rangle \right|^2}{\hbar \epsilon_{\pm}^{(1)} - \hbar \epsilon_{k}^{(2)}} + \frac{\left| \langle e_{k}^{(2)} | \mathcal{H}_d | \psi_{\mp}^{(0)} \rangle \right|^2}{\hbar \epsilon_{\mp}^{(1)} - \hbar \epsilon_{k}^{(2)}} \right) \\
= \mp \hbar \mathcal{E}_p^2 \left[ \frac{(\alpha_{\pm}^{(1)})^2}{\epsilon_{\pm}^{(1)}} + \frac{(\alpha_{-}^{(1)})^2}{\epsilon_{-}^{(1)}} + \sum_{k=1}^{4} \frac{(\alpha_{k}^{(2)*}) \alpha_{0}^{(1)} \sqrt{2} + \mu_{k}^{(2)*} \mu_{0}^{(1)}}{\epsilon_{k}^{(2)}} \right]^2 \\
\times \left( \frac{1}{\epsilon_{\pm}^{(1)} - \epsilon_{k}^{(2)}} + \frac{1}{\epsilon_{-}^{(1)} - \epsilon_{k}^{(2)}} \right). \quad (C.7a)
\]
Before we compare the perturbative with the numerical solutions, it is necessary to establish the range of validity of the perturbation series. This is not always an easy task, and ours is one such case. The perturbation procedure relies on the existence of the “small” parameter determining the size of perturbation relative to the size of the unperturbed Hamiltonian. It is usually reasonable to assume that the condition $|V_{ij}/(\epsilon_i - \epsilon_j)| < 1$ ensures the convergence of the perturbation series [108]. Here, $V_{ij}$ is the matrix element of the perturbation, taken between two unperturbed states $|i\rangle$ and $|j\rangle$, and $\epsilon_{i,j}$ are energies of these states. However, from the viewpoint of accuracy of the result, the convergence requirement can be somewhat relaxed. Dirac claims in his seminal book [145, Sec. 42] that “Even when the [perturbation] series does not converge, the first approximation obtained by means of it is usually fairly accurate.” This means that the good agreement between the perturbative and numerical result cannot in general be taken as a hint on the convergence of perturbation series. In turn, even if the perturbation series does not converge, we anticipate that the results in the lowest orders can still be taken to be reasonably accurate. A sufficient condition for convergence can be formulated by requiring that every successive order in the perturbation series be smaller than the previous one. This condition is indeed satisfied in our range of interest, i.e. for $0 < \epsilon_p/\kappa < 3$. How well does the perturbative treatment compare with the numerical results?

We have compared the results from perturbation theory with the numerical results for the energy splittings of the Rabi states $|\psi_{\pm}\rangle$. The perturbative results do not show a notable improvement to Figure 11.2. As the highest order in the perturbation series is second order, this result can be expected. Second order perturbation only accounts for the corrections involving states from the neighbouring manifolds. Based on the results of Chapter 12, the corrections to the Rabi states can come in the lowest order from third manifold states, therefore requiring third order perturbative corrections.

Figure C.1 compares the perturbative corrections to the (a) off-resonant states in the first manifold and (b) second manifold, with the numerical solutions. Perturbative corrections again include terms up to the second order in $E_p$. These quadratic corrections show good qualitative agreement with the numerical solutions. For a better quantitative agreement, higher order perturbations are again needed. Denoting the matrix element of $\mathcal{H}_d$ taken between any two unperturbed states as $V_{ji} = \langle e_j | \mathcal{H}_d | e_i \rangle$, we can write the expression for the third order perturbative corrections to the energy eigenvalue $\hbar\epsilon_n$ as

$$
\hbar\epsilon_n^{(3)} = \sum_{k \neq n} \sum_{m \neq n} \frac{V_{nm}V_{mk}V_{kn}}{(\hbar\epsilon_k - \hbar\epsilon_n)(\hbar\epsilon_m - \hbar\epsilon_n)} - V_{nn} \sum_{m \neq n} \left( \frac{|V_{nm}|}{\hbar\epsilon_m - \hbar\epsilon_n} \right)^2. \quad (C.8)
$$

It can be determined by inspection that this expression vanishes because the perturbation
Figure C.1: Comparison of numerical results (dashed) with the perturbation theory results (solid) given by Eqs. (C.7). Figure (a) shows off–resonant states in the first manifold and Figure (b) shows the entire second manifold.
couples adjacent manifolds only: this cannot be accommodated with the cyclic chain of indices in the first term, while $V_{nm} \equiv 0$ makes the second term disappear.

The pattern of dependence of the dressed states splittings on the driving field can be seen from the presented results. As the driving field increases, splittings decrease and ultimately for large driving tend to zero. This behaviour comes as the atom saturates, and the energy levels probed by the external driving become bare cavity (harmonic) energy levels.
APPENDIX C. SECOND-ORDER PERTURBATION CORRECTIONS
Appendix D

Quantum Jumps and Density Matrix Elements

In this Appendix, we derive jump terms for the effective master equation (12.9), and the equations of motion for density matrix elements.

D.1 Jump Terms

We follow the notation of Carmichael [118] and rewrite the effective master equation (1.10) in a Lindblad form as \( \dot{\rho}_{\text{eff}} = \mathcal{L}_{\text{eff}} \rho_{\text{eff}} \), where the Liouvillian superoperator can be divided into a part describing the free evolution between the jumps, \( \mathcal{L}_{\text{eff}} = \mathcal{S}_{\text{eff}} \), and a part describing the jumps, \( \mathcal{S}_{\text{eff}} \). Jump term \( \mathcal{S}_{\text{eff}} \rho_{\text{eff}} \) arises from the equivalent jump term in the full master equation,

\[
\mathcal{S}_{\rho} = 2 \kappa a \rho a^\dagger + 2 \gamma_1 \sigma_{12} \rho \sigma_{21} + 2 \gamma_2 \sigma_{32} \rho \sigma_{23} + 2 \gamma_3 \sigma_{34} \rho \sigma_{43} .
\]

(D.1)

Using the methods of Ref. [133], and consistent with the truncation of the dressed states space (as in Figure 12.6), we can write field and atomic collapse operators in terms of the bare states as

\[
a = |0, 1\rangle \langle 1, 1| + |0, 3\rangle \langle 1, 3| + |0, 4\rangle \langle 1, 4| + \sqrt{2} (|1, 1\rangle \langle 2, 1| + |1, 2\rangle \langle 2, 2| + |1, 3\rangle \langle 2, 3|)
\]

+ \sqrt{3} |2, 1\rangle \langle 3, 1| ,

(D.2a)

\[
\sigma_{12} = |1, 1\rangle \langle 1, 2| + |2, 1\rangle \langle 2, 2| ,
\]

(D.2b)

\[
\sigma_{32} = |1, 3\rangle \langle 1, 2| + |2, 3\rangle \langle 2, 2| ,
\]

(D.2c)

\[
\sigma_{34} = |0, 3\rangle \langle 0, 4| + |1, 3\rangle \langle 1, 4| .
\]

(D.2d)

It can be deduced from the bare states that the collapse operators can be expressed as a linear combinations of the following polariton operators: \( a \propto p_1, p_2, p_3, p_{24}, p_{34}, p_{25}, p_{35} \);
\( \sigma_{12}, \sigma_{34} \propto p_2, p_3, p_{24}, p_{34}, p_{25}, p_{35} \) and \( \sigma_{32} \propto p_{2}^\dagger p_2, p_{3}^\dagger p_3, p_{4}^\dagger p_4, p_{5}^\dagger p_5, p_{2}^\dagger p_{23}, p_{3}^\dagger p_{34}, p_{4}^\dagger p_{45}, p_{5}^\dagger p_{54} \). Note that the polariton jump terms arising from the atomic jumps associated with the operator \( \sigma_{32} \) (i.e. proportional to \( \gamma_3 \)) couple dressed states within the same manifold. This is possible since the associated atomic transition is not coupled to the cavity mode, but to the classical field \( \Omega_c \), and therefore jumps occurring in this atomic decay channel do not switch between the adjacent manifolds.

Using the correspondences above, one can write all of the jump terms in the polariton basis, thus making the transition \( \mathcal{S} \rho \rightarrow \mathcal{S}_{ef} \rho_{ef} \). This transformation can be viewed as a rotation of a truncated basis of a superoperator. The coefficients \( \Gamma_{ijkl} \) of the jump terms are given in Table D.1.

### D.2 Equations of Motion

The equations of motion for the density matrix elements in the basis schematically shown in Figure 12.6 can be found from the master equation (1.10). There is a total of 35 equations, since population conservation can be used to eliminate one of them. The equations for populations are

\[
\begin{align*}
\dot{\rho}_{00} &= 2\Gamma_0 \rho_{11} + \Omega_0 (\rho_{01} + \rho_{10}), \\
\dot{\rho}_{11} &= -2\Gamma_0 \rho_{11} + \Gamma_{1222} \rho_{22} + \Gamma_{1333} \rho_{33} - \Omega_0 (\rho_{01} + \rho_{10}) + \Omega_{12} \rho_{12} + \Omega_{12}^* \rho_{21} + \Omega_{13} \rho_{13} + \Omega_{13}^* \rho_{31}, \\
\dot{\rho}_{22} &= -2(\Gamma_{22} + \Gamma_{2222}) \rho_{22} - \Gamma_{23} (\rho_{32} + \rho_{23}) + 2\Gamma_{45} (\rho_{45} + \rho_{54}) - \Omega_{12} \rho_{12} - \Omega_{12}^* \rho_{21} + \Gamma_{2233} \rho_{33} + \Gamma_{2244} \rho_{44} + \Gamma_{2255} \rho_{55} + \Omega_{24} (\rho_{24} + \rho_{42}) + \Omega_{25} (\rho_{25} + \rho_{52}), \\
\dot{\rho}_{33} &= -2(\Gamma_{33} + \Gamma_{3333}) \rho_{33} - \Gamma_{23} (\rho_{32} + \rho_{23}) + 2\Gamma_{45} (\rho_{45} + \rho_{54}) - \Omega_{13} \rho_{13} - \Omega_{13}^* \rho_{31} + \Gamma_{3322} \rho_{22} + \Gamma_{3344} \rho_{44} + \Gamma_{3355} \rho_{55} + \Omega_{34} (\rho_{34} + \rho_{43}) + \Omega_{35} (\rho_{35} + \rho_{53}), \\
\dot{\rho}_{44} &= -2(\Gamma_{44} + \Gamma_{4444}) \rho_{44} + \Gamma_{4555} \rho_{55} - 2\Gamma_{45} (\rho_{45} + \rho_{54}) - \Omega_{24} (\rho_{24} + \rho_{42}) - \Omega_{34} (\rho_{34} + \rho_{43}), \\
\dot{\rho}_{55} &= -2(\Gamma_{55} + \Gamma_{5555}) \rho_{55} + \Gamma_{5544} \rho_{44} - 2\Gamma_{45} (\rho_{45} + \rho_{54}) - \Omega_{25} (\rho_{25} + \rho_{52}) - \Omega_{35} (\rho_{35} + \rho_{53}).
\end{align*}
\]

The equations for coherences are

\[
\begin{align*}
\dot{\rho}_{01} &= -\Gamma_0 \rho_{01} + 2\Gamma_{02} \rho_{12} + 2\Gamma_{03} \rho_{13} + \Omega_0 (\rho_{11} - \rho_{00}) + \Omega_{12} \rho_{02} + \Omega_{13} \rho_{03}, \\
\dot{\rho}_{02} &= - (\Gamma_{22} + \Gamma_{2222} - i\epsilon_2) \rho_{02} + \Gamma_{23} \rho_{03} + \Gamma_{0214} \rho_{14} + \Gamma_{0215} \rho_{15} + \Omega_{0} \rho_{12} - \Omega_{12}^* \rho_{01} + \Omega_{24} \rho_{04} + \Omega_{25} \rho_{05}, \\
\dot{\rho}_{03} &= - (\Gamma_{33} + \Gamma_{3333} - i\epsilon_3) \rho_{03} - \Gamma_{23} \rho_{02} + \Gamma_{0314} \rho_{14} + \Gamma_{0315} \rho_{15} + \Omega_{0} \rho_{13} - \Omega_{13}^* \rho_{01} + \Omega_{34} \rho_{04} + \Omega_{35} \rho_{05}.
\end{align*}
\]
\[\begin{align*}
\dot{\rho}_{04} &= - (\Gamma_{44} + \Gamma_{4444} + i\epsilon_4) \rho_{04} - \Gamma_{45} \rho_{05} - \Omega_{24} \rho_{02} - \Omega_{34} \rho_{03} + \Omega_0 \rho_{14} & (D.4d) \\
\dot{\rho}_{05} &= - (\Gamma_{55} + \Gamma_{5555} + i\epsilon_5) \rho_{05} - \Gamma_{56} \rho_{06} - \Omega_{25} \rho_{02} - \Omega_{35} \rho_{03} + \Omega_0 \rho_{12} & (D.4e) \\
\dot{\rho}_{12} &= - (\Gamma_0 + \Gamma_{22} + \Gamma_{2222} - i\epsilon_2) \rho_{12} - \Gamma_{23} \rho_{13} \\
&+ \Gamma_{1224} \rho_{24} + \Gamma_{1225} \rho_{25} + \Gamma_{1234} \rho_{34} + \Gamma_{1235} \rho_{35} \\
&+ \Omega_1^{*} (\rho_{22} - \rho_{11}) - \Omega_0 \rho_{02} + \Omega_1^{*} \rho_{32} + \Omega_{24} \rho_{14} + \Omega_{25} \rho_{15} & (D.4f) \\
\dot{\rho}_{13} &= - (\Gamma_0 + \Gamma_{33} + \Gamma_{3333} - i\epsilon_3) \rho_{13} - \Gamma_{23} \rho_{12} \\
&+ \Gamma_{1324} \rho_{24} + \Gamma_{1325} \rho_{25} + \Gamma_{1334} \rho_{34} + \Gamma_{1335} \rho_{35} \\
&+ \Omega_1^{*} (\rho_{33} - \rho_{11}) - \Omega_0 \rho_{03} + \Omega_1^{*} \rho_{32} + \Omega_{34} \rho_{14} + \Omega_{35} \rho_{15} & (D.4g) \\
\dot{\rho}_{14} &= - (\Gamma_0 + \Gamma_{44} + \Gamma_{4444} - i\epsilon_4) \rho_{14} - \Gamma_{45} \rho_{15} \\
&- \Omega_0 \rho_{04} + \Omega_{24} \rho_{12} + \Omega_{34} \rho_{13} + \Omega_1^{*} \rho_{24} + \Omega_1^{*} \rho_{34} & (D.4h) \\
\dot{\rho}_{15} &= - (\Gamma_0 + \Gamma_{55} + \Gamma_{5555} - i\epsilon_5) \rho_{15} - \Gamma_{45} \rho_{14} \\
&- \Omega_0 \rho_{05} + \Omega_{25} \rho_{12} + \Omega_{35} \rho_{13} + \Omega_1^{*} \rho_{25} + \Omega_1^{*} \rho_{35} & (D.4i) \\
\dot{\rho}_{23} &= - [\Gamma_{22} + \Gamma_{2222} + \Gamma_{33} + \Gamma_{3333} + \Gamma_{2233} - i(\epsilon_2 + \epsilon_3)] \rho_{23} \\
&- \Gamma_{23} (\rho_{22} + \rho_{33}) + \Gamma_{2344} \rho_{44} + \Gamma_{2355} \rho_{55} + \Gamma_{2332} \rho_{32} + \Gamma_{2345} \rho_{45} + \Gamma_{2354} \rho_{54} \\
&+ \Omega_1^{*} \rho_{13} + \Omega_1^{*} \rho_{21} + \Omega_3^{*} \rho_{24} + \Omega_3^{*} \rho_{25} + \Omega_{24} \rho_{43} + \Omega_{25} \rho_{53} & (D.4j) \\
\dot{\rho}_{24} &= - [\Gamma_{22} + \Gamma_{2222} + \Gamma_{44} + \Gamma_{4444} + \Gamma_{2244} - i(\epsilon_2 + \epsilon_4)] \rho_{24} \\
&- \Gamma_{23} \rho_{34} - \Gamma_{45} \rho_{25} + \Gamma_{2435} \rho_{35} \\
&- \Omega_{24} (\rho_{44} - \rho_{22}) - \Omega_{12} \rho_{14} - \Omega_{34} \rho_{23} + \Omega_{25} \rho_{54} & (D.4k) \\
\dot{\rho}_{25} &= - [\Gamma_{22} + \Gamma_{2222} + \Gamma_{55} + \Gamma_{5555} + \Gamma_{2255} - i(\epsilon_2 + \epsilon_5)] \rho_{25} \\
&- \Gamma_{23} \rho_{35} - \Gamma_{45} \rho_{24} + \Gamma_{2534} \rho_{34} \\
&- \Omega_{25} (\rho_{55} - \rho_{22}) - \Omega_{13} \rho_{15} - \Omega_{35} \rho_{23} + \Omega_{24} \rho_{45} & (D.4l) \\
\dot{\rho}_{34} &= - [\Gamma_{33} + \Gamma_{3333} + \Gamma_{44} + \Gamma_{4444} + \Gamma_{3344} - i(\epsilon_3 + \epsilon_4)] \rho_{34} \\
&- \Gamma_{23} \rho_{24} - \Gamma_{45} \rho_{35} + \Gamma_{3425} \rho_{25} \\
&- \Omega_{34} (\rho_{44} - \rho_{33}) - \Omega_{13} \rho_{14} + \Omega_{35} \rho_{54} - \Omega_{24} \rho_{32} & (D.4m) \\
\dot{\rho}_{35} &= - [\Gamma_{33} + \Gamma_{3333} + \Gamma_{55} + \Gamma_{5555} + \Gamma_{3355} - i(\epsilon_3 + \epsilon_5)] \rho_{35} \\
&- \Gamma_{23} \rho_{25} - \Gamma_{45} \rho_{34} + \Gamma_{3524} \rho_{24} \\
&- \Omega_{35} (\rho_{55} - \rho_{33}) - \Omega_{13} \rho_{15} + \Omega_{34} \rho_{45} - \Omega_{25} \rho_{32} & (D.4n) \\
\dot{\rho}_{45} &= - [\Gamma_{44} + \Gamma_{4444} + \Gamma_{55} + \Gamma_{5555} + \Gamma_{4455} - i(\epsilon_4 + \epsilon_5)] \rho_{45} \\
&- \Gamma_{45} (\rho_{44} + \rho_{55}) + \Gamma_{4554} \rho_{55} \\
&- \Omega_{24} \rho_{25} - \Omega_{34} \rho_{35} - \Omega_{25} \rho_{42} - \Omega_{35} \rho_{43} & (D.4o)
\end{align*}\]

Damping coefficients $\Gamma_0$ and $\Gamma_{ij}$ have been evaluated in general in Chapter 10. Coefficients $\Gamma_{ijkl}$ are related to the jump operators and are given in Table D.1. The source of the different terms in these equations should be apparent from the earlier discussion. For example, damping emerging from jump terms is denoted as $\Gamma_{ijkl}$, while damping coming from the effective Hamiltonian (12.8) is denoted as $\Gamma_0$ and $\Gamma_{ij}$. 
It is straightforward to prove that the following equalities hold:

\begin{align}
2 (\Gamma_{22} + \Gamma_{2222}) &= \Gamma_{122} + \Gamma_{3322} , \\
2 (\Gamma_{33} + \Gamma_{3333}) &= \Gamma_{1133} + \Gamma_{2233} , \\
2 (\Gamma_{44} + \Gamma_{4444}) &= \Gamma_{2244} + \Gamma_{3344} + \Gamma_{5544} , \\
2 (\Gamma_{55} + \Gamma_{5555}) &= \Gamma_{2255} + \Gamma_{3355} + \Gamma_{4455} ,
\end{align}

(D.5a) (D.5b) (D.5c) (D.5d)

preserving the population conservation condition \( \sum_j \rho_{jj} = 1 \), i.e. \( \sum_j \dot{\rho}_{jj} = 0 \).
Table D.1: Expressions for the 'jump' coefficients. Here, \( w_{ij} = \Omega_{ij}/\mathcal{E}_p \), and the parameters \( \alpha, \beta, \mu \) and \( \nu \) are given in Section 10.3.
Appendix E

Resonance Fluorescence of a Four Level Atom in Free Space

This Appendix briefly reviews the main features of a driven four level atom in free space, with the driving fields assumed to be classical and coherent. The purpose of this review is to emphasise the difference between the intracavity and free space situations.

We discuss how the four level atomic scheme behaves without the presence of a cavity, i.e., we replace the cavity mode with a travelling classical laser field of Rabi frequency $\Omega_{1,2}$, with indices 1 and 2 denoting the two atomic transitions which were in the previous arrangement assumed to be coupled to the cavity.

An appealing feature of this resonance fluorescence setup is that the equations of motion are linear and can, at least in principle, be solved analytically. Unfortunately, the analytical solutions are very complex and do not give much physical insight. However, the general expression for the correlation functions derived here are useful for highlighting the differences between the atom-cavity and free–atom cases.

E.1 Master Equation and Bloch Equations

The energy level configuration under study is the same as presented in Fig. 2.3, and Rabi frequencies associated with coupling of the probe field and atomic transitions of frequencies $\omega_{12}$ and $\omega_{34}$ are $\Omega_1$ and $\Omega_2$, respectively. Detunings from the atomic transitions are defined as throughout the thesis: $\Delta$ is the detuning of transition $|3\rangle \rightarrow |4\rangle$, and $\delta$ is the common detuning of the transitions$^{1}$ $|1\rangle \rightarrow |2\rangle$ and $|3\rangle \rightarrow |2\rangle$. The master equation for the reduced density operator of the atom in the interaction picture is

$$\dot{\rho} = -i\Delta [\sigma_{44}, \rho]_+ - i\delta [\sigma_{22}, \rho]_- - i\Omega_1 [\sigma_{12} + \sigma_{21}, \rho]_+ - i\Omega_2 [\sigma_{34} + \sigma_{43}, \rho]_- - i\Omega_c [\sigma_{23} + \sigma_{32}, \rho]_- + 2\gamma_1 \sigma_{12}\rho\sigma_{21} + 2\gamma_2 \sigma_{32}\rho\sigma_{23} + 2\gamma_3 \sigma_{34}\rho\sigma_{43} - (\gamma_1 + \gamma_2) [\sigma_{22}, \rho]_+ - \gamma_3 [\sigma_{44}, \rho]_+ , \quad \text{(E.1)}$$

$^{1}$Remember that the condition for EIT is the two–photon resonance for the transition $|1\rangle \rightarrow |3\rangle$. 

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where \([A, B]_{\pm} := AB \pm BA\) denote commutator (+) and anti-commutator (-).

We assume for simplicity that the Rabi frequencies are real. From the master equation, the equations of motion for the expectation values of atomic populations and coherences can be derived using standard methods [1] to give

\[
\begin{align*}
\langle \dot{\sigma}_{12} \rangle &= - (\gamma_1 + \gamma_2 + i\delta) \langle \sigma_{12} \rangle + i\Omega_1 (\langle \sigma_{22} \rangle - \langle \sigma_{11} \rangle) - i\Omega_c \langle \sigma_{13} \rangle, \quad (E.2a) \\
\langle \dot{\sigma}_{23} \rangle &= - (\gamma_1 + \gamma_2 - i\delta) \langle \sigma_{23} \rangle + i\Omega_1 (\langle \sigma_{13} \rangle - \langle \sigma_{24} \rangle) + i\Omega_c (\langle \sigma_{33} \rangle - \langle \sigma_{22} \rangle), \quad (E.2b) \\
\langle \dot{\sigma}_{34} \rangle &= - (\gamma_3 + i\Delta) \langle \sigma_{34} \rangle + i\Omega_2 (\langle \sigma_{34} \rangle - \langle \sigma_{33} \rangle) + i\Omega_c \langle \sigma_{24} \rangle, \quad (E.2c) \\
\langle \dot{\sigma}_{13} \rangle &= i(\Omega_1 \langle \sigma_{23} \rangle - \Omega_2 \langle \sigma_{14} \rangle) - i\Omega_c \langle \sigma_{12} \rangle, \quad (E.2d) \\
\langle \dot{\sigma}_{24} \rangle &= - [\gamma_1 + \gamma_2 + \gamma_3 + i(\Delta - \delta)] \langle \sigma_{24} \rangle - i(\Omega_2 \langle \sigma_{23} \rangle - \Omega_1 \langle \sigma_{14} \rangle) + i\Omega_c \langle \sigma_{34} \rangle, \quad (E.2e) \\
\langle \dot{\sigma}_{14} \rangle &= - (\gamma_3 + i\Delta) \langle \sigma_{14} \rangle + i(\Omega_1 \langle \sigma_{24} \rangle - \Omega_2 \langle \sigma_{13} \rangle), \quad (E.2f) \\
\langle \dot{D}_{21} \rangle &= \frac{-2\gamma_1 + \gamma_2}{2} \langle D_{21} \rangle + \frac{2\gamma_1 + \gamma_2}{2} \langle D_{43} \rangle + (2\gamma_1 + \gamma_2) \langle D_{32} \rangle - i\Omega_c (\langle \sigma_{23} \rangle - \langle \sigma_{32} \rangle) + 2i\Omega_1 (\langle \sigma_{12} \rangle - \langle \sigma_{21} \rangle), \quad (E.2g) \\
\langle \dot{D}_{32} \rangle &= \frac{-\gamma_1 + 2\gamma_2 + \gamma_3}{2} \langle D_{32} \rangle - \frac{\gamma_1 + 2\gamma_2 - 3\gamma_3}{2} \langle D_{43} \rangle + \frac{\gamma_1 + 2\gamma_2 + \gamma_3}{2} \langle D_{21} \rangle + 2i\Omega_c (\langle \sigma_{23} \rangle - \langle \sigma_{32} \rangle) - i\Omega_1 (\langle \sigma_{12} \rangle - \langle \sigma_{21} \rangle) - i\Omega_2 (\langle \sigma_{34} \rangle - \langle \sigma_{43} \rangle), \quad (E.2h) \\
\langle \dot{D}_{43} \rangle &= \frac{-2\gamma_2 + \gamma_3}{2} \langle D_{43} \rangle - \frac{6\gamma_3 - \gamma_2}{2} \langle D_{43} \rangle + \frac{2\gamma_3 + \gamma_2}{2} \langle D_{21} \rangle - (2\gamma_3 - \gamma_2) \langle D_{32} \rangle - i\Omega_c (\langle \sigma_{23} \rangle - \langle \sigma_{32} \rangle) + 2i\Omega_2 (\langle \sigma_{34} \rangle - \langle \sigma_{43} \rangle), \quad (E.2i)
\end{align*}
\]

where the population differences are defined as \(D_{ij} = \sigma_{ii} - \sigma_{jj}\). The representation involving the population differences allows us to eliminate one of the equations for the atomic population, since \(\sum_{j=1}^{4} \sigma_{jj} = 1\).

We now proceed to solve this set of equations in general. Defining a vector containing the coherences and population differences as

\[
\mathbf{v} = \left( \langle \sigma_{12} \rangle, \langle \sigma_{21} \rangle, \langle \sigma_{23} \rangle, \langle \sigma_{32} \rangle, \langle \sigma_{34} \rangle, \langle \sigma_{43} \rangle, \langle \sigma_{13} \rangle, \langle \sigma_{31} \rangle, \langle \sigma_{24} \rangle, \langle \sigma_{42} \rangle, \langle \sigma_{14} \rangle, \langle \sigma_{41} \rangle, \langle D_{21} \rangle, \langle D_{32} \rangle, \langle D_{43} \rangle \right)^T, \quad (E.3)
\]

the set of equations (E.2) can be written as a single nonhomogeneous linear first–order equation

\[
\dot{\mathbf{v}} = A\mathbf{v} + \mathbf{C}, \quad (E.4)
\]

with the vector of coefficients not involving components of \(\mathbf{v}\) being

\[
\mathbf{C} = (0, \ldots, 0, -\gamma_1 - \gamma_2/2, \gamma_2 + (\gamma_1 + \gamma_3)/2, -\gamma_3 - \gamma_2/2)^T. \quad (E.5)
\]
E.2. FIELD CORRELATIONS

The steady state is given by \( \mathbf{v} = -\mathbf{A}^{-1}\mathbf{C} \), and with the definition \( \mathbf{\sigma} = \mathbf{v} - \mathbf{v}_{ss} \), the equation (E.4) can be rewritten in a homogeneous form as

\[
\dot{\mathbf{\sigma}} = \mathbf{A}\mathbf{\sigma},
\]

and solved using standard techniques.

Defining the matrix exponential \( \mathbf{E}(t) = \exp(\mathbf{A}t) \), the solution for the time evolution of \( \mathbf{v} \) is written as

\[
\mathbf{v}(t) = [\mathbf{1} - \mathbf{E}(t)] \mathbf{v}_{ss} + \mathbf{E}(t) \mathbf{v}_0,
\]

where \( \mathbf{v}_0 \) contains the values of coherences and population differences at time \( t = 0 \).

Having obtained the general solution for the time evolution of the system variables, we turn to the calculation of the correlations in the scattered field.

### E.2 Field Correlations

In this Section, we show how to calculate two time correlation functions of light. In particular, we will be interested in comparing the second order correlation function, defined as

\[
g_{ss}^{(2)}(\tau) = \frac{G_{ss}^{(2)}(\tau)}{|G_{ss}^{(1)}(\tau)|^2} = \frac{G_{ss}^{(2)}(\tau)}{\lim_{\tau \to -\infty} G_{ss}^{(2)}(\tau)},
\]

with the results obtained earlier for the intracavity case. The above equation is the definition of the normalised correlation function, given in terms of its unnormalised counterparts

\[
G_{ss}^{(1)}(\tau) = \lim_{t \to -\infty} \langle \mathcal{E}^{(-)}(\mathbf{r}, t) \mathcal{E}^{(+)}(\mathbf{r}, t+\tau) \rangle,
\]

\[
G_{ss}^{(2)}(\tau) = \lim_{t \to -\infty} \langle \mathcal{E}^{(-)}(\mathbf{r}, t) \mathcal{E}^{(-)}(\mathbf{r}, t+\tau) \mathcal{E}^{(+)}(\mathbf{r}, t) \mathcal{E}^{(+)}(\mathbf{r}, t) \rangle,
\]

where \( G_{ss}^{(j)}(\tau) \) is the \( j \)-th order steady-state correlation function calculated at the delay time \( \tau \) and

\[
\mathcal{E}^{(+)}(\mathbf{r}, t) = i \sum_{\mathbf{k},\lambda} \sqrt{\frac{\hbar \omega_k}{2 \epsilon_0 V}} \hat{e}_{k,\lambda} s_{k,\lambda}(t) \exp(i \mathbf{k} \cdot \mathbf{r}),
\]

\[
\mathcal{E}^{(-)}(\mathbf{r}, t) = (\mathcal{E}^{(+)}(\mathbf{r}, t))^\dagger,
\]

is the electromagnetic field operator. In this expression, \( \mathbf{k} \) is a wavevector and \( \lambda \) is a polarisation state of the electromagnetic field mode, \( \omega_k \) is its frequency, \( \hat{e}_{k,\lambda} \) is the unit polarisation vector and \( V \) is the quantisation volume. Operator \( s_{k,\lambda} \) is the reservoir mode.
Following standard methods [2], the electric field operator (E.10) at the position \( \mathbf{r} \) of an idealised point-like detector at a time \( t \) can be expressed in terms of atomic operators. The interaction of the four level atom with a reservoir in the electric dipole and rotating wave approximations is described by the Hamiltonian

\[
\mathcal{H}_{\text{AR}} = \sum_{k,\lambda} \hbar \mathbf{s}_{k,\lambda}^\dagger \left( \kappa_{k,\lambda}^{(12)*} \sigma_{12} + \kappa_{k,\lambda}^{(34)*} \sigma_{34} \right) + \text{H.c.},
\]

where \( \text{H.c.} \) denotes Hermitian conjugate. The Heisenberg equation of motion for the reservoir operator is then

\[
\dot{\mathbf{s}}_{k,\lambda} = -i \omega_{k,\lambda} \mathbf{s}_{k,\lambda} - i \left( \kappa_{k,\lambda}^{(12)*} \sigma_{12} + \kappa_{k,\lambda}^{(34)*} \sigma_{34} \right).
\]

Solving for the reservoir operator, the scattered electromagnetic field can be expressed in terms of atomic operators evaluated at retarded time \( t - r/c \),

\[
\mathcal{E}^{(+)}(\mathbf{r}, t) = \phi_{12}(\mathbf{r}) \sigma_{12}(t - r/c) + \phi_{34}(\mathbf{r}) \sigma_{34}(t - r/c),
\]

\[
\phi_{ij}(\mathbf{r}) = \frac{-\omega_{ij}^2 \mathbf{\mu}_{ij} \times \mathbf{\hat{r}} \times \mathbf{\hat{r}}}{4\pi\varepsilon_0 c^2 r}.
\]

Here, \( \mathbf{\hat{r}} = \mathbf{r}/|\mathbf{r}| \) is the unit vector in the direction of \( \mathbf{r} \), and \( \mathbf{\mu}_{ij} \) is the dipole moment of the atomic transition \(|i\rangle \rightarrow |j\rangle\).

Using these, we find that

\[
G^{(1)}(t + r/c, t + r/c + \tau) =
\]

\[
= f_{12}^4 \langle \sigma_{21}(t)\sigma_{12}(t + \tau) \rangle + f_{34}^4 \langle \sigma_{34}(t)\sigma_{34}(t + \tau) \rangle
\]

\[
+ f_{12}f_{34} \{ \langle \sigma_{21}(t)\sigma_{34}(t + \tau) \rangle + \langle \sigma_{34}(t)\sigma_{12}(t + \tau) \rangle \},
\]

and

\[
G^{(2)}(t + r/c, t + r/c + \tau) =
\]

\[
= f_{12}^4 \langle \sigma_{21}(t)\sigma_{22}(t + \tau)\sigma_{12}(t) \rangle + f_{34}^4 \langle \sigma_{34}(t)\sigma_{44}(t + \tau)\sigma_{34}(t) \rangle
\]

\[
+ (f_{12}f_{34})^2 \{ \langle \sigma_{21}(t)\sigma_{44}(t + \tau)\sigma_{12}(t) \rangle + \langle \sigma_{34}(t)\sigma_{22}(t + \tau)\sigma_{34}(t) \rangle 
\]

\[
+ f_{12}f_{34} \{ \langle \sigma_{21}(t)\sigma_{22}(t + \tau)\sigma_{34}(t) \rangle + \langle \sigma_{34}(t)\sigma_{34}(t + \tau)\sigma_{12}(t) \rangle \}
\]

\[
+ f_{12}f_{34}^3 \{ \langle \sigma_{21}(t)\sigma_{44}(t + \tau)\sigma_{34}(t) \rangle + \langle \sigma_{34}(t)\sigma_{44}(t + \tau)\sigma_{12}(t) \rangle \},
\]

where the coefficients are defined as

\[
f_{ij} = \frac{\omega_{ij}^2 \mathbf{\mu}_{ij} \sin \theta_{ij}}{4\pi\varepsilon_0 c^2 r},
\]

and \( \theta_{ij} = \angle(\mathbf{r}, \mathbf{\mu}_{ij}) \). From now on, we assume that \( \theta_{12} \equiv \theta_{34} = \theta \). Taking the limits \( t \to \infty \) in the correlation functions of Equations (E.14) enables us to obtain the expression for
the normalised correlation functions \( g_{ss}^{(1)}(\tau) \) and \( g_{ss}^{(2)}(\tau) \). We need to use the quantum regression theorem (QRT) \([1, 6]\) and Equation (E.4) to calculate the mean values

\[
\frac{d}{dt} \langle v(t) \rangle = A \langle v(t) \rangle + C. \tag{E.16a}
\]

The two–time correlations then satisfy the differential equation

\[
\frac{d}{d\tau} \langle \sigma_{ij} v(\tau) \rangle = A \langle \sigma_{ij} v(\tau) \rangle + \langle \sigma_{ij} \rangle_{ss} C, \tag{E.16b}
\]

\[
\langle \sigma_{ij} v \rangle_{ss} = -\langle \sigma_{ij} \rangle_{ss} A^{-1} C = \langle \sigma_{ij} \rangle_{ss} v_{ss}. \tag{E.16c}
\]

Equation (E.16b) can be solved with the initial condition \( \langle \sigma_{ij} v(0) \rangle = \langle \sigma_{ij} \rangle_{ss} \) to give

\[
\langle \sigma_{ij} v(\tau) \rangle = I \langle \sigma_{ij} \rangle_{ss} v_{ss} + E(\tau) (\langle \sigma_{ij} v \rangle_{ss} - \langle \sigma_{ij} \rangle_{ss} v_{ss}). \tag{E.16d}
\]

Note that the \((r, \theta)\) dependence from the Equation (E.15) cancels through the normalisation to give

\[
g_{ss}^{(1)}(\tau) = \frac{g_{12}^{2} \langle \sigma_{21} \sigma_{12}(\tau) \rangle + g_{34}^{2} \langle \sigma_{43} \sigma_{34}(\tau) \rangle + g_{12} g_{34} (\langle \sigma_{21} \sigma_{43}(\tau) \rangle + \langle \sigma_{34} \sigma_{12}(\tau) \rangle)}{g_{12}^{2} \langle \sigma_{22} \rangle_{ss} + g_{34}^{2} \langle \sigma_{44} \rangle_{ss}}, \tag{E.17}
\]

and \( g_{ij} = \omega_{ij}^{2}/(4\pi\epsilon_{0}c^{2}) \) are atom–field coupling constants.

The second order correlation function of Equation (E.14b) can be evaluated to give

\[
G_{ss}^{(2)}(\tau) = f_{12}^{4} \langle \sigma_{21}(0) \sigma_{22}(\tau) \sigma_{12}(0) \rangle + f_{34}^{4} \langle \sigma_{43}(0) \sigma_{44}(\tau) \sigma_{34}(0) \rangle
\]

\[
+ (f_{12} f_{34})^{2} \left[ \langle \sigma_{21}(0) \sigma_{43}(\tau) \sigma_{12}(0) \rangle + \langle \sigma_{34}(0) \sigma_{22}(\tau) \sigma_{34}(0) \rangle \right], \tag{E.18}
\]

and we can again use the QRT to find

\[
\langle \sigma_{21}(0) v_{ss} \sigma_{12}(0) \rangle = \left[ I - E(\tau) \right] \langle \sigma_{22} \rangle_{ss} v_{ss} + E(\tau) \langle \sigma_{21} v_{12} \rangle_{ss}, \tag{E.19a}
\]

\[
\langle \sigma_{43}(0) v_{ss} \sigma_{34}(0) \rangle = \left[ I - E(\tau) \right] \langle \sigma_{44} \rangle_{ss} v_{ss} + E(\tau) \langle \sigma_{43} v_{34} \rangle_{ss}. \tag{E.19b}
\]

Also, since \( \lim_{\tau \to \infty} \langle \sigma_{ij}(0) v_{ss} \sigma_{kl}(0) \rangle_{ss} = \langle \sigma_{il} \rangle_{ss} v_{ss} \delta_{jk} \), we find that

\[
\lim_{\tau \to \infty} G_{ss}^{(2)}(\tau) = \left( f_{12}^{2} \langle \sigma_{22} \rangle_{ss} + f_{34}^{2} \langle \sigma_{44} \rangle_{ss} \right)^{2} \equiv \lim_{\tau \to \infty} G_{ss}^{(1)}(\tau)^{2}. \tag{E.20}
\]

We now have the analytical expression for the field correlation functions, and we will use them to briefly discuss the difference between this setup and the setup involving the optical cavity.

### E.3 Results and Discussion

The results for the second order correlation function calculated in the previous Section are presented here. For reason of better comparison, we assume the parameters to be of comparable size to those used with the cavity setup. Note that this is not an impractical
APPENDIX E. RESONANCE FLUORESCENCE OF A FOUR LEVEL ATOM

Figure E.1: Second order correlation function for resonance fluorescence involving a four level atom. Parameters are $\Omega_j/\gamma = 6$, $\Omega_c/\gamma = 2$, $(\Delta, \delta)/\gamma = (0.1, -0.2)$.

idealisation, since the size of the Rabi frequencies (which are the counterparts of the atom–cavity couplings), can be adjusted by adjusting the power of the driving laser.

Figure E.1 shows the result for the correlation function, calculated by first solving the set of equations (E.2) for steady state and time dependent cases, and then proceeding using the methods outlined in Section E.2. One feature is immediately obvious. Just like the case of resonance fluorescence involving a two level atom, the second order correlation function at zero delay time vanishes. This is not surprising, since the mechanism responsible for the zero correlation in a two level system is still in place here. Consider a four level atom, excited to the level $|4\rangle$. Assume that it emits a photon, decaying to the level $|3\rangle$ (since decay to the level $|2\rangle$ is forbidden). It takes some time for the atom to be reexcited into either level $|4\rangle$ or level $|2\rangle$, before it is able to emit a second photon. Another feature is noted. Namely, the oscillations modulating the correlation function are composed of contributions with three different frequencies. The frequency of large oscillations is related to the frequency of driving. They occur for $\Omega_j > \gamma_j/2$, at the frequency $\sqrt{\Omega^2 - (\gamma/2)^2}$, since we have taken $\Omega_1 = \Omega_2 = \Omega$ and $\gamma_1 = \gamma_3 = \gamma$. The other two frequencies could be understood in the same way as for the intracavity case – in terms of dressed states. The dressed states of this system will have a ground state, coinciding
with the ground state of the atom, and an excited state which splits into four, just like
the higher manifold states of the intracavity system. The energies of these four states will
again be given by Equations (10.11), with the substitutions $g_{1,2} \sqrt{n} \rightarrow \Omega_{1,2}$. In the limit
of vanishing atomic detunings, we can write

$$\epsilon = \pm \sqrt{\frac{1}{2} (\Omega_1^2 + \Omega_2^2 + \Omega_c^2) \pm \sqrt{\frac{1}{4} (\Omega_1^2 + \Omega_2^2 + \Omega_c^2)^2 - (\Omega_1 \Omega_2)^2}}, \quad (E.21)$$

for the eigenenergies. Obviously, these are symmetrically spaced around the resonance,
so one modulation frequency will represent each pair of states. This remains true (to a
good approximation) for small atomic detunings, such as those used in the simulation.
In Figure E.2 the second–order spectrum is plotted, clearly showing the four sidebands,
originating from the four dressed states. The second–order spectrum $S_2(\omega)$ is defined as
Fourier transform of $g_{ss}^{(2)}(\tau) - 1$.

Our formalism for calculating the correlation functions reveals that, as in the two level
case, the value of $g^{(2)}(0)$ always vanishes, regardless of the parameters in the system. This
can be seen from the expression for $G^{(2)}(\tau)$ in Equation (E.18). Since atomic operators must satisfy the relation $\sigma_{jk} \sigma_{il} = \delta_{jk} \sigma_{il}$, expression (E.18) vanishes at $\tau = 0$ by default. This is a stark difference from the intracavity case, where we have shown a sharp change in statistics, as represented by $g^{(2)}(0)$, occurring for an increasing driving field.

Another useful perspective can be obtained by considering the form of the field operator in the two different environments. In the resonance fluorescence case here, it is obvious that the electromagnetic field (not including the component scattered in the forward direction) is described in terms of atomic operators as

$$a_{\text{free}} \sim \Omega_1 \sigma_{12} + \Omega_2 \sigma_{34},$$  \hspace{1cm} (E.22)

while the intracavity field operator contains an additional contribution from the coherent driving

$$a_{\text{cav}} \sim E_p/\kappa + (g_1/\kappa) \sigma_{12} + (g_2/\kappa) \sigma_{34}.$$  \hspace{1cm} (E.23)

The contribution of the coherent driving changes the photon statistics significantly, through both its independent contribution and through the interference with the part incoherently
scattered from the atom. This can be seen from the expression for the second order correlations

$$\langle a_{\text{can}}^\dagger a_{\text{can}}^2 \rangle \sim (E_p/\kappa)^4 + 4 (E_p/\kappa)^2 \left[ (g_1/\kappa)^2 \langle \sigma_{22} \rangle + (g_2/\kappa)^2 \langle \sigma_{44} \rangle \right] + 4 (E_p/\kappa)^3 \Re \left[ (g_1/\kappa) \langle \sigma_{12} \rangle + (g_2/\kappa) \langle \sigma_{34} \rangle \right].$$  \hspace{1cm} (E.24)

For the case of resonance fluorescence, the highest possible nonvanishing moment is the mean photon number $$\langle a_{\text{free}}^\dagger a_{\text{free}}^2 \rangle \sim \Omega_1^2 \langle \sigma_{22} \rangle + \Omega_2^2 \langle \sigma_{44} \rangle.$$ Equation (E.24) shows the two contributions to the zero–delay correlation function mentioned above. The term $$(E_p/\kappa)^4$$ comes from the driving field amplitude alone, while the other terms represent the interference of the incoherently scattered field and the coherent amplitude.

To make a full comparison, in Figure E.3 we plot the contributions from the incoherent amplitude component, as in Section 12.6 (see Equation (12.11) and Figure 12.8). To recap, term $S$ describes the squeezing in the field quadrature in phase with the driving field, $V$ denotes variance in the incoherent component and $T$ denotes intensity–amplitude correlations in the incoherent component. Again, both $V$ and $T$ are determined by the correlations in the intensity. The difference to the intracavity case is rather drastic, showing that the sub-Poissonian field statistics and photon antibunching arise solely from the sub-Poissonian intensity fluctuations.

In conclusion, we have presented a brief review of a different setup to that studied in the main body of this thesis, namely the free space resonance fluorescence case. We have calculated an exact expression for the second order correlation function, which enabled us to study the cause behind the different photon statistics exhibited by the different arrangements.

We have explained the main characteristics of the second order correlation function in terms of dressed states and introduced a simplified qualitative model enabling us to understand the difference arising from the coherent amplitude component present in the cavity case, but absent in the resonance fluorescence case.
Bibliography


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