Quantum stochastic theory of phonon scattering between polaritons

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Quantum stochastic operator equations are derived for inter-branch exciton and polariton processes caused by acoustic phonon scattering. The use of a fully quantum model combined with these recently developed techniques predicts the presence of “stimulated scattering” terms, and provides a sound basis for understanding the basis of the approximations used in generating the equations. The theory is applied to a model motivated by recent experiments where a stronger photoluminescence signal from the high energy microcavity polariton polariton is observed for low excitation powers, and also to an experimental scheme designed to show evidence of the stimulated scattering effect.

I. INTRODUCTION

The development of stochastic operator and wavefunction approaches to dissipative processes caused much interest in the field of quantum optics. My aim here is to present a derivation of equations for inter-branch phonon processes using one of these techniques, that of quantum stochastic differential equations (QSDE’s) [1]. The derivation of these equations relies on similar approximations to the well known master equation techniques [2, 3], and so has the advantage of being a systematic derivation in which the approximations are clearly stated and whose effects are generally well understood. Master equation techniques from a “quantum optical” viewpoint have already been applied to both excitonic optical bistability [4] and the semiconductor laser [5]. The stochastic wavefunction approach based on an “unravelling” of the master equation [2] has been applied to both electron-phonon scattering [6] and exciton-phonon scattering [7]. This exciton-phonon work was for a model involving a single quantum-dot exciton level, and the emphasis was on non-Markovian effects.

Here the QSDE technique is used to treat the phonon interaction as a coupling between two exciton or polariton branches and an infinite reservoir of phonon modes. Polaritons are formed in a semiconductor quantum microcavity, structures in which quantum wells are grown inside an optical cavity. The light field is confined by two Bragg mirrors, and the quantum wells create confined exciton states. With appropriate design, the photons in the optical cavity and the excitons couple together creating a quasi particle called a cavity polariton. The QSDE’s for the polariton system include factors accounting for its partly photonic nature. In addition, they contain extra “stimulated scattering” terms, also predicted by Pau et al [1], whose presence would not have been predicted by a phenomenological approach to modelling the system.

This paper was motivated by photoluminescence (PL) measurements of microcavity polaritons at low temperatures (1.7 – 50K) and a range of input laser intensity (0.4 – 10 Wcm$^{-2}$) [8]. Most investigations of these cavity polaritons use reflectivity measurements, with only a few considering the other optical emission properties. Contrary to expectations, it was the higher energy polariton that dominated the PL at the lowest laser power densities. The experimental results are due to the thermalisation of the free carriers down into the upper polariton branch and the dominance of the cavity decay rate over the scattering between the polariton branches. Free carrier scattering becomes insignificant at low carrier densities and LO phonon processes do not occur because their energies are greater than the polariton splitting. As a result, the photon intensity (PL signal) emitted from decay of the upper polariton is strong, while the signal from the lower polariton is restricted by the small influx of excitation due to the slow inter-branch acoustic phonon scattering. The equations for the theoretical model are solved in an appropriate limit to predict the severity of this bottleneck. The theory is also extended to a system where the upper polariton is coherently excited by a laser tuned to the polariton energy, and predicts a similar bottleneck effect. The results depend on the size of the photonic component of the polariton as well as its excitonic part.

Finally, the stimulated scattering term is investigated by constructing a model experimental system that could be used to measure its effects. This model also predicts the strength of the effect, which is proportional to the exciton-phonon coupling, the exciton-photon mixing, and the intensity of the stimulating field.

Note added (March 2002)

Stimulated emission of bulk polaritons was experimentally observed in II-VI sample in the late 1970’s [9], and microcavity polaritons in QW samples in 1997 [10]. Stimulated emission terms (i.e. a factor of $N + 1$) were inserted by hand into the theory of Tassone et. al. [11] on the bottleneck effects in microcavity polaritons, and then with more theoretical justification in by Tassone and Yamamoto [12] which included discussion of stimulated scattering. In these two papers the interest is restricted...
to generating a rate equation model, and the papers pay little theoretical attention to all the steps in a rigorous derivation such as I have attempted here, and certainly quantum noise terms are not considered at all.

The bottleneck effects and stimulated scattering have become of much recent interest – see for example some of the references brought to my attention by J. J. Baumberg [22, 23, 24]. It turns out the model I used when doing these early calculations at University of Sheffield in 1996 is far too simplistic, and so is not relevant to any likely experimental work. However, the theoretical method used to generate both the QSDE’s and model is still valid, and it would be a straightforward matter to recast the model to a more appropriate one and do some useful calculations.

II. THE PHONON INTERACTION

In this section I introduce a model Hamiltonian for the phonon scattering process. This does not specify the type of phonon explicitly, they could be either optical or acoustic (either longitudinal or transverse), as the phonons are treated as a reservoir of boson modes with particular properties, properties that are necessary for the QSDE technique to be valid. As long as the type of phonons considered have these properties, the results given here are applicable.

It is also relevant to consider here how we describe the exciton and cavity modes. For low excitation strengths the exciton can be described as a boson. This is because the resulting low density of excitons means that any phase space filling or exciton-exciton scattering processes are unlikely to occur. In addition, there is a continuum of exciton modes for different in-plane momenta. I will refer to this as an ‘exciton branch’, and the energies of these are given by the dispersion relations for the exciton. Similarly, a microcavity photon mode can also have a range of $k$ values, and can also be called a ‘photon branch’.

Consider an interaction Hamiltonian for quantum well excitons interacting with phonons. The two dimensional nature of each exciton branch is allowed for by a set of exciton modes indexed by the in-plane wave vector $k$, and these modes are associated with the operators $\hat{e}_i(k)$, $\hat{e}_i^\dagger(k)$. The phonon modes are indexed by in-plane wave vector $k$ as well as their growth direction wave vector $q$, with the operators $\hat{b}(k, q)$, $\hat{b}^\dagger(k, q)$. The coupling between exciton and phonons is denoted $\chi$. In the interaction picture, the operators all have an oscillatory time dependence given by their mode energies, and so $\hbar \omega \delta \hat{a}^\dagger \hat{a}$ type terms do not appear in the Hamiltonian. This time dependence is not explicitly included to avoid cluttering the notation. The general form of the interaction Hamiltonian for first order scattering involving any two exciton branches and one phonon branch is

$$\hat{H}_A = \sum_{ij} \sum_k \sum_{k'} \sum_{k''} \sum_q \chi_{ij}(k, k', k'', q) \left[ \hat{e}_i(k) + \hat{e}_i^\dagger(k) \right] \left[ \hat{e}_j(k') + \hat{e}_j^\dagger(k') \right] \left[ \hat{b}^\dagger(k'', q) + b(k'', q) \right]$$

(2.1)

Often the terms are written like $\left[ \hat{e}_i(k) + \hat{e}_i^\dagger(-k) \right]$, but this is simply a re-ordering of the same summation. The $i, j$ indexes span the exciton branches, the $k$ summations are over the in-plane wave vector of the excitons, and the $q$ sum is over the wave vector of the phonon in the growth direction. Note that with only minor modifications to the summations and indices the same expression can be used to describe phonon scattering in bulk or even 1 or 0 dimensional structures.

The first simplification we can make is to apply conservation of in-plane crystal momentum $k$, which is not conserved because of the lack of translational symmetry in the growth direction. This reduces the number of summations in the above formula from three to two, and results in

$$\hat{H}_A = \sum_{ij} \sum_k \sum_{\Delta k} \sum_q \chi_{ij}(k, k - \Delta k, q) \left[ \hat{e}_i(k)\hat{e}_j^\dagger(k - \Delta k)\hat{b}(\Delta k, q) \right. + \hat{e}_i^\dagger(k)\hat{e}_j(k - \Delta k)\hat{b}(\Delta k, q) \\
\left. + \hat{e}_i(-k)\hat{e}_j^\dagger(k - \Delta k)\hat{b}(\Delta k, q) \right. + \hat{e}_i(-k)\hat{e}_j(k - \Delta k)\hat{b}(\Delta k, q) \right].$$

(2.2)

It is normal in quantum optics to make a rotating wave approximation to remove non-resonant (non energy conserving) terms because these off resonant terms oscillate at about twice the optical frequency. This is very much faster than any likely dynamical process in the system, and so their net effect is assumed to average to zero, in a kind of “coarse graining” approximation. In this system involving phonons, it is not obvious that this type of approximation can be made. Both the excitons and the phonons have a continuum of energies, and because the phonons can have frequencies near zero, the oscillations in the off resonant terms can easily be relatively slow, so the rotating wave approximation (RWA) cannot always be applied. A situation like this can be dealt with using non-rotating wave techniques (eg. [11]).

In the case of optical phonon scattering a RWA is possible. This is because optical phonons have a minimum frequency (about $10^{13}$ Hz), and the off resonant terms would be oscillating at about twice that rate. Also, acoustic phonon scattering could be treated between two sufficiently separated exciton branches, as again the off resonant terms would oscillate at twice the frequency of the separation of the two excitons. Alternatively, if for
some reason the exciton to phonon coupling vanished sufficiently fast for small phonon energies the exciton could only couple to relatively energetic phonons for which a RWA might be reasonable. Assuming well separated branches or optical phonons, the oscillations in the off resonant terms will occur on much shorter timescales than any other dynamical process in the system. This means we can apply the RWA, so the Hamiltonian becomes

\[
\hat{H}_A = \sum_{i \neq j} \sum_k \sum_{\Delta k} \chi_{ij}(k, k - \Delta k, q) \left[ \hat{e}_i(k) \hat{e}_j^\dagger(k - \Delta k) \hat{b}^\dagger(\Delta k, q) + \hat{e}_j^\dagger(k) \hat{e}_i(k - \Delta k) \hat{b}(\Delta k, q) \right]. \tag{2.3}
\]

A. The phonon QSDE

Following Gardiner [1], we can write down the Ito quantum stochastic differential equations (QSDE’s) for the above model. In this approach we treat the phonon modes as a reservoir or ‘heat bath’ with which the excitons can interact. At first sight, this QSDE method does not seem directly applicable to this problem. This is because there is only one reservoir of phonons in the system, whereas the standard application of the theory would assume that each scattering between excitons of different wave number is coupled to an independent reservoir of phonons. For this reason some correlations might persist (or perhaps form) between different exciton modes because they interact with this common reservoir. We might reasonably assume this to be an insignificant effect because the phonon reservoir is much bigger than the range of exciton states – the phonon reservoir is three dimensional, whereas the exciton states only fill two dimensions.

For example, compare this with the standard quantum optics case, where a single system mode interacts with a one dimensional continuum of reservoir modes – there are an infinite number of reservoir modes for the system mode to decay into. Similarly, the exciton phonon scattering model presented here also has sufficiently large reservoirs in which to “lose” any quantum correlations. Although plausible enough, this argument is not sufficient. Leaving the detailed calculations to a later work here I just make the extra assumption that any such correlations will decay away, and so the use of QSDE’s here is valid.

There are three properties this reservoir must have for the QSDE approach to be valid. Firstly, there must be a smooth, dense spectrum of phonons with which each exciton mode can interact. For acoustic phonons, which have an energy proportional to the magnitude of their wave vector, such a spectrum exists. For LO phonons, the phonon modes only exist above a certain energy. This can cause problems due to the effect of the edge of the LO phonon band, but if the inter-exciton transition is sufficiently larger than the lowest LO phonon energy, the scattering will not see the phonon band edge.

Secondly, the coupling between system and reservoir must be linear in the reservoir operators, in agreement with the first order interaction Hamiltonian used above.

Thirdly, the coupling constants between the reservoir and system must be smooth functions of frequency, which is holds for deformation potential coupling to phonons. This leads to the additional and related approximation usually called the First Markov Approximation. This requires the coupling parameter in the Hamiltonian between the excitons and the reservoir of phonons to be independent of frequency in the range of interest. This “range of interest” is where energy is very nearly conserved – where the phonon carries away (or supplies) the energy lost (or gained) by the conversion of the exciton. Note that the coupling is assumed to remain roughly constant over the width of the resonance. This is also a kind of “coarse graining”, as a flat frequency response implies a strongly peaked time response – so the coupling appears “flat” if the width of the time response is much shorter than the important dynamical processes occurring in the system. Also, the state of the reservoir must be unaffected by the interaction. In this case, the energy lost by the excitons in the form of phonons must not change the state of the reservoir – for example, by increasing its temperature. This is guaranteed for low excitation levels by the relative sizes of the system and reservoir.

The procedure has allowed us to eliminate the phonon modes from the model. It has given us a system in which the phonon scattering causes a loss of energy from the excitons and added a delta correlated “quantum noise”. This noise is the effect of the quantum uncertainty of the phonon reservoir feeding into the exciton system, and is described by new quantum noise operators \( d\hat{B}_P, d\hat{B}_P^\dagger \).

The Ito QSDE for some arbitrary system operator \( \hat{S} \) affected by phonon scattering between two different exciton branches is

\[
d\hat{S} = \sum_{i \neq j} \sum_{\Delta k} \frac{\chi_{ij}(k, k - \Delta k)}{2} \left\{ [N_P(\Delta k) + 1] \left[ 2\hat{e}_i^\dagger(k) \hat{e}_j(k - \Delta k) \hat{S} \hat{e}_i(k) \hat{e}_j^\dagger(k - \Delta k) - \hat{S} \hat{e}_i^\dagger(k) \hat{e}_j(k - \Delta k) \hat{e}_i(k) \hat{e}_j^\dagger(k - \Delta k) \right] + N_P(\Delta k) \left[ 2\hat{e}_i(k) \hat{e}_j^\dagger(k - \Delta k) \hat{S} \hat{e}_i^\dagger(k) \hat{e}_j(k - \Delta k) - \hat{S} \hat{e}_i(k) \hat{e}_j^\dagger(k - \Delta k) - \hat{e}_i(k) \hat{e}_j^\dagger(k - \Delta k) \hat{S} \hat{e}_i(k) \hat{e}_j^\dagger(k - \Delta k) \right] \right\} dt
\]

\[- \sum_{i, j} \sum_{\Delta k} \sqrt{\chi_{ij}(k, k - \Delta k)} \left( \hat{S} \hat{e}_i^\dagger(k) \hat{e}_j(k - \Delta k) \right) d\hat{B}_P \]
+ \sum_{i,j} \sqrt{\chi_{ij}(k, k - \Delta k)} dB^\dagger_P \left[ \hat{S}, \hat{e}_i(k) \hat{e}^\dagger_j(k - \Delta k) \right].

(2.4)

This operator \( \hat{S} \) could be any operator of interest – perhaps one of the mode operators, the intensity operator or even some kind of correlation function. The operators \( dB_P, dB^\dagger_P \) are quantum white noise operators, and \( N_P(\Delta k) \) is the average number of thermal phonons in each phonon mode. The summation over \( q \) has dropped out because the resonance condition allows us to calculate it in terms of the \( k \) terms and the dispersion curves. The noise operators \( dB_P, dB^\dagger_P \) are defined by their correlations, which are

\[
\langle dB_P(t) dB_P(t') \rangle = \langle [dB_P(t)^\dagger dB_P(t')] \rangle = 0,
\]

\[
\langle dB^\dagger_P(t) dB^\dagger_P(t') \rangle = (N_P + 1) \delta(t - t') dt,
\]

\[
\langle dB^\dagger_P(t) dB_P(t') \rangle = N_P \delta(t - t') dt.
\]

These operators cause “quantum white noise”, and correspond to a reservoir in which the number of thermal quanta is constant per unit bandwidth. This is not the same as a quantum thermal noise, which would have a Boltzmann distribution for its thermal quanta. Although using quantum white noise is not exact, it is a very convenient and widely used approximation. In particular, it would not be a good approximation for the low wavevector part of an acoustic phonon reservoir, where the large relative change in energy between nearby wavevectors means a proper thermal ensemble should be used [4]. However, since we have already introduced a rotating wave approximation bases on a large energy separation between exciton branches, and hence are not in the low energy part of the phonon spectrum, using quantum white noise does not cause any extra restriction.

B. Intensity and amplitude operator equations

To get a clearer picture of what the phonon interaction does, I will give some examples relevant to the calculations in this paper. In the quasi-PL and coherent excitation models presented in section VI, equations for excitation number and mode amplitude are used; more specifically, these are derived from the QSDE’s for the mode intensity and mode amplitude operators. For simplicity, I write \( \hat{e}_i(k) \) as \( \hat{\varepsilon}_+ \), \( \hat{\varepsilon}_j(k - \Delta k) \) as \( \hat{\varepsilon}_- \), and \( \chi_{ij}(k, k - \Delta k) \) as \( \chi \). The intensity for the (+) mode is just \( \hat{I}_+ = \hat{\varepsilon}^\dagger_+ \hat{\varepsilon}_+ \), that for the (-) mode is \( \hat{I}_- = \hat{\varepsilon}^\dagger_- \hat{\varepsilon}_- \). The modes described by \( \hat{\varepsilon}_+ \) and \( \hat{\varepsilon}_- \) are different.

The QSDE equations for the intensities of the (+) and (-) modes due to the phonon scattering are

\[
d\hat{I}_+ = \chi \left[ -\hat{I}_+ (\hat{I}_- + 1) - N_P (\hat{I}_+ - \hat{I}_-) \right] dt
\]

\[
d\hat{I}_- = \chi \left[ \hat{I}_+ (\hat{I}_- + 1) + N_P (\hat{I}_+ - \hat{I}_-) \right] dt
\]

(2.6)

The noise terms cannot be rewritten in a form depending solely on \( \hat{I}_\pm \), so I leave them in terms of \( \hat{\varepsilon}_+ \), \( \hat{\varepsilon}_- \). However, we could define new operators \( \hat{J}_1 = \hat{\varepsilon}^\dagger_+ \hat{\varepsilon}_- \) and \( \hat{J}_2 = \hat{\varepsilon}^\dagger_- \hat{\varepsilon}_+ \), and from them we get extra equations which allow us to remove the \( \hat{\varepsilon} \) type terms. Note how the terms in the pair of equations balance each other to preserve excitation number, a consequence of the fact that the scattering transfers excitation between exciton branches while emitting (or absorbing) phonons. This contrasts with the usual loss models that use reservoirs which destroy excitations by removing them from the system.

The QSDE equations for the amplitudes of the (+) and (-) modes due to the phonon scattering are

\[
d\hat{\varepsilon}_+ = \chi \left[ -\hat{\varepsilon}_+ (\hat{\varepsilon}_- + 1) - N_P \hat{\varepsilon}_+ \right] dt + \sqrt{\chi} \hat{\varepsilon}_- dB_P,
\]

\[
d\hat{\varepsilon}_- = \chi \left[ \hat{\varepsilon}^\dagger_+ \hat{\varepsilon}_- - N_P \hat{\varepsilon}_- \right] dt + \sqrt{\chi} dB^\dagger_P \hat{\varepsilon}_+.
\]

(2.7)

These equations have an interesting feature – the presence of a “stimulated scattering” or “incoherent stimulated emission” term. These should up as the \( \hat{I}_+ \), \( \hat{I}_- \) and \( \hat{J}_1 \), \( \hat{J}_2 \) in the intensity equations. A naive consideration of the phonon scattering process would not suggest such terms, as the process is incoherent and is caused by loss. Hence it is natural to expect only simple loss terms or population transfer terms. The new terms are predicted by this quantum model because the two exciton modes are coupled together by the phonon interaction, and so their bosonic nature causes these stimulated effects. However, because the phonon scattering is Markovian, the stimulated emission loses its phase memory and becomes “stimulated scattering”. The appearance of these terms shows clearly the advantages of working from a more rigorous derivation, as opposed to a phenomenological approach based on simply writing down rate equations. They were also predicted by Pau et al [1] who started from the Heisenberg equations for this system.

III. AN IDEAL POLARITON

A polariton consists of an exciton coupled to a cavity mode. A full model would include a description of the different types of exciton, their excited states, as well as all the cavity modes and the couplings between these. However, in practise we know that excitons will only couple to a cavity mode if they have matching wavevectors. Also, for low excitation strengths only the lowest energy exciton is significant. This means I can consider an "ideal
polariton”, consisting of an idealised quantum well exciton, an idealised cavity mode, and the exciton to photon coupling term between the two. Further, I use the Coulomb gauge, the dipole approximation, and the rotating wave approximation to express the coupling term in its simplest form.

The result couples an exciton to a cavity mode, where both are modelled by quantised harmonic oscillators. I write \( \hat{e}^\dagger(k) \), \( \hat{e}(k) \) as the operators for the exciton mode, and \( \hat{a}^\dagger(k) \), \( \hat{a}(k) \) as the operators for the field in the cavity mode. The interaction Hamiltonian is

\[
\hat{H}_P = \hbar A(k) \left[ \hat{e}^\dagger(k) \hat{a}(k) + \hat{e}(k) \hat{a}^\dagger(k) \right] \tag{3.1}
\]

The parameter \( A(k) \) is the coupling between the cavity mode and the exciton. It is in units of frequency, and is equal to half the splitting between the pair of polaritons at resonance. I will denote the exciton energy as \( \hbar \Omega(k) \), and the cavity mode energy as \( \hbar \omega(k) \). This two mode system of the coupled exciton and cavity mode can be diagonalised, and then be written in terms of new polariton modes, which describe the two new polariton modes that can be used in place of the original exciton and photon modes. The two eigenvalues (or frequencies) of the eigenmodes (the polaritons) are

\[
2\epsilon_A(k) = \sqrt{\Omega(k) - \omega(k)} \\
\pm \sqrt{\Omega(k) - \omega(k)^2 + 4A(k)^2}. \tag{3.2}
\]

The polaritons are characterised by a label \( \pm \) denoting the “upper” or “lower” (in energy) polariton branch, and their wavenumber \( k \). If I introduce the operators \( \hat{p}^\dagger_\pm(k) \), \( \hat{p}_\pm(k) \) to describe the polaritons, the eigenvectors of the system describe how the cavity and exciton operators combine to form the polariton operators. The polariton operators for the coupled exciton - photon system are \( \hat{p}^\dagger_\pm(k) \), \( \hat{p}_\pm(k) \). The exciton and cavity modes have coupled together to form two new polariton modes.

The polariton operators are related to the exciton and cavity operators by the following expressions

\[
\hat{p}_+(k) = c_+(k) \hat{e}(k) + c_-(k) \hat{a}(k) \\
\hat{p}_-(k) = c_-(k) \hat{e}(k) + c_+(k) \hat{a}(k) \tag{3.3}
\]

where

\[
c_+(k) = -d_-(k) = \frac{1}{\sqrt{2}} \sqrt{1 + \sin \alpha(k)} \\
d_+(k) = c_-(k) = -\frac{1}{\sqrt{2}} \sqrt{1 - \sin \alpha(k)} \\
\tan \alpha(k) = \frac{\Omega(k) - \omega(k)}{2A(k)} \tag{3.4}
\]

Note that the equations for the polariton operators in terms of the exciton and cavity operators can be inverted. The results are

\[
\hat{e}(k) = c_+(k) \hat{p}_+(k) + c_-(k) \hat{p}_-(k) \\
\hat{a}(k) = d_+(k) \hat{p}_+(k) + d_-(k) \hat{p}_-(k). \tag{3.5}
\]

These last equations are the most useful for the work in this paper. They allow us to take an interaction involving only the exciton or photon modes and rewrite it in terms of the eigenstates of the coupled system, i.e. in terms of the polariton operators.

### IV. COHERENT DRIVING, LOSSES, AND THERMAL NOISE

Of course it is not only the phonon scattering which is main topic of this paper that needs to be considered. Any realistic model needs to include losses due to exciton recombination and the imperfect cavity mirrors. Also, sources of excitons or polaritons need to be included – these can originate either from the combination of hot free carriers to generate a thermal population of excitons, or by coherent excitation from a laser beam. To do this I use the quantum stochastic differential equation (QSDE) for a polariton with coherent driving, linear losses and thermal noise. When describing a realistic polariton, a sum over in-plane wave number \( k \) is needed to account for the dispersion. However, since each \( k \) mode is independent in these interactions, I do not need to include the sum over states here. The standard driving and loss terms for a boson mode described by operators \( \hat{f}, \hat{f}^\dagger \) have an interaction Hamiltonian like

\[
\hat{H}_D = i \left[ \epsilon^* \hat{f} - \epsilon \hat{f}^\dagger \right] + \sum_i \gamma_i \left[ \hat{f} \hat{\Gamma}_i^\dagger + \hat{f}^\dagger \hat{\Gamma}_i \right]. \tag{4.1}
\]

The first term describes a coherent driving process, which can be imagined as being due to a classical driving field, but is actually a coherent state driving term. The second term couples the mode to a reservoir consisting of an infinite number of boson modes \( \hat{\Gamma}_1, \hat{\Gamma}_1 \). The loss from the system is modelled by irreversible transfer of excitation from the system into this reservoir. The amount of loss is described by a decay rate \( \gamma \) that is related to the \( \gamma_i \), but includes the sum over the density of states of the reservoir.

In a polariton system the excitons are not the eigenstates. The eigenstates are the polaritons, and so I need to replace the exciton operators with their equivalent in terms of polariton operators, defined by eqn (3.3). I will now use \( f_\pm \) to represent \( c_\pm \) or \( d_\pm \), depending on whether I am considering interaction with the cavity mode (\( \hat{f} = \hat{a} \)) or the exciton (\( \hat{f} = \hat{e} \)). In polariton operators, the interaction Hamiltonian can be rewritten

\[
\hat{H}_D = i \left\{ \epsilon^* f_+ \hat{p}_+(t) - [\epsilon^* f_+^* \hat{p}_+^\dagger(t) \right\}
\]
of the equation, with the quantum noise terms averaged to zero. Replacing the expectation value with a number \( I_\pm = \langle I_\pm \rangle \), the equation can be rewritten as

\[
\frac{d}{dt} I_\pm = \gamma f_\pm^* f_\pm [-I_\pm + N].
\]

Now I consider the effect of a coherent driving term as well as losses. The QSDE for how the operator \( \hat{p}_\pm \) of the mode changes due to the losses and the associated noise is

\[
d\hat{p}_\pm = \left[ f_\pm^* \epsilon - f_\pm^* f_\pm \frac{\gamma}{2} \hat{p}_\pm \right] dt + \sqrt{\gamma} f_\pm^* d\tilde{B}. \tag{4.6}
\]

A complex number equation for the mode amplitude can be produced from this QSDE easily. We can take the expectation values of the equation, and assume the mode is in a coherent state \( |\alpha\rangle \). The quantum noise term then averages to zero, and we get an equation in terms of the coherent amplitude \( \alpha \)

While this approach might seem fine if the mode is in a coherent state, what if it is not? Fortunately, the basic idea can still be used. Any quantum state can be expanded over a basis of coherent states \([1] \), by using either the Glauber-Sudarshan P representation \([14, 15] \), or by the subsequent generalisations \([16] \). Here the systematic procedure is to convert the QSDE into the equivalent master equation form and then apply the representations in the standard way. In practise this means that the state of the mode is not specified by a single complex number \( \alpha \), but by an appropriately distributed ensemble, each member of which follows the equation

\[
\frac{d}{dt} \alpha = f_\pm^* \epsilon - f_\pm^* f_\pm \frac{\gamma}{2} \alpha. \tag{4.7}
\]

In these coherent state representations the non thermal part of the quantum noise terms vanish, as they are absorbed into the coherent state basis. The effect of thermal noise on the system is to spread the ensemble out – this does not affect its average amplitude, but will change its intensity by some amount. If thermal noise can be neglected, and the system starts or is being coherently driven, then it can be adequately described by a single complex number. However, if there are additional interactions, such as strong phonon scattering, then the system might well depart strongly from a coherent state.

V. THE POLARITON-PHONON INTERACTION

In section II I described an interaction Hamiltonian for excitons scattering off phonons. To convert it into one relevant to a polariton system, I employ the same procedure as for the coherent driving and decay interactions. Since the eigenstates of the system are the polaritons, and so I need to replace the exciton operators with their
equivalent in terms of polariton operators, as defined by eqn 3.3. A single exciton and cavity mode branch gives us a two polariton system, so the interaction Hamiltonian in the polariton operators is

\[
\hat{H} = \sum_k \sum_{\Delta k} \sum_q \chi(k, k - \Delta k; q) \left[ \{ d_+(k) \hat{p}_{+}(k; t) + d_-(k) \hat{p}_{-}(k; t) \} \right.
\]

\[
\left. + \sum_{k} \left[ d_+^*(k - \Delta k) \hat{p}_{+}^\dagger(k - \Delta k; t) + d_-^*(k - \Delta k) \hat{p}_{-}^\dagger(k - \Delta k; t) \right] \hat{b}(\Delta k; q) \right].
\]  

(5.1)

When this is expanded out, there are four types of important term. Two are intra- polariton terms, involving phonon scattering along the upper or lower polariton branches, and the other two are inter- polariton scatterings. In addition to these, there are non-resonant terms which do not conserve energy such as those that create or destroy two polaritons.

How do all these processes fit together? First consider the dispersion curves for the polaritons and the acoustic phonons. This is a 2D problem – the polariton is confined in the growth direction by the microcavity DBR mirrors, and the quantum well. Because both polaritons only have in-plane freedom, only phonons that conserve in-plane momentum can be emitted. The dispersion relations for the polaritons are bowl-like surfaces, and the phonon dispersion is like a solid cone. Acoustic phonons carry very little energy for their momentum, so this cone is very shallow compared to the other dispersions. The ‘cone’ is solid because the phonons can have a non zero growth direction momentum – the internal parts of the cone correspond to when some of the phonon momentum is in the growth direction.

Figure 1 is a schematic diagram showing the dispersion curves and allowed acoustic phonon scattering processes. The intra-branch process mainly transfers population from large \(k\) to small \(k\) along the same branch, although the opposite can also occur in the presence of thermal phonons. The inter-branch processes transfers population between the two branches, predominantly from the upper (+) to the lower (−) branch. Again, the reverse process is possible in the presence of thermal phonons. However, note that the upwards thermal processes use a phonon dispersion with its apex on the lower initial state. Similarly, figure 2 shows the dispersion curves and allowed optical phonon scattering processes. This follows the same principle as the acoustic case, but with the optical phonon dispersion curve and so includes a minimum phonon energy.

Here I am primarily concerned with the downward inter-branch process, from the upper to the lower branch with the production of a phonon. This means a RWA can be justified for sufficiently large branch separation as the non resonant terms will oscillate at a rate given by twice this – much faster than the speed of the important system dynamics. The resonant upper to lower phonon branch interaction term is then contained in

\[
\hat{H} = \sum_k \sum_{\Delta k} \sum_q \chi(k, k - \Delta k; q) \left[ d_+^*(k) \hat{p}_{+}(k; t) d_-^*(k - \Delta k) \hat{p}_{-}^\dagger(k - \Delta k; t) \hat{b}(\Delta k; q) \right] + \sum_k d_+^*(k) \hat{p}_{+}^\dagger(k; t) d_-^*(k - \Delta k) \hat{p}_{-}(k - \Delta k; t) \hat{b}(\Delta k; q) \right].
\]  

(5.2)

Note that this is the same as the exciton phonon Hamiltonian if we replace \(d_+^*(k) \hat{p}_{+}(k)\) with \(\hat{e}(k)\) and \(d_-^*(k - \Delta k) \hat{p}_{-}^\dagger(k - \Delta k)\) with \(\hat{e}(k - \Delta k)\).

### A. The polariton-phonon QSDE

The justification for the validity of the QSDE method for this system is identical to that used in the initial exciton scattering section, so I do not repeat it here. The only thing that has changed is the introduction of the coefficients for the component of exciton in the polariton, and the particular allowed scattering transitions between branches. The Itô QSDE for a general system operator \(\hat{S}\) in a two polariton system undergoing (+) to (−) phonon scattering is
LO phonon scattering

\[ d \hat{S} = \sum_{\Delta k} \frac{\chi(k, k - \Delta k)}{2} |d_+(k)d^*_+(k - \Delta k)|^2 \]
\[ \{ (N_p(\Delta k) + 1) \]
\[ \left[ 2 \hat{p}_+(k) \hat{p}_-(k - \Delta k) \hat{S} + \hat{p}_+(\Delta k) \hat{p}_-(k - \Delta k) \hat{S} \right] \]
\[ - \hat{S} \hat{p}_+(\Delta k) \hat{p}_-(k - \Delta k) \hat{S} \]
\[ + N_p(\Delta k) \left[ 2 \hat{p}_+(k) \hat{p}_+(k - \Delta k) \hat{S} + \hat{p}_+(\Delta k) \hat{p}_+(k - \Delta k) \hat{S} \right] \]
\[ - \hat{S} \hat{p}_+(\Delta k) \hat{p}_-(k - \Delta k) \hat{S} \]
\[ - \sum_{\Delta k} \sqrt{\chi(k, k - \Delta k)} [d_+(k)d^*_+(k - \Delta k)] \]
\[ \left[ \hat{S}, \hat{p}_+(k) \hat{p}_-(k - \Delta k) \right] d\hat{B} \]
\[ + \sum_{\Delta k} \sqrt{\chi(k, k - \Delta k)} |d_+(k)d_-(k - \Delta k)| \]
\[ d\hat{B} \left[ \hat{S}, \hat{p}_+(k) \hat{p}_-(k - \Delta k) \right]. \] (5.3)

The operators \( d\hat{B}_p, d\hat{B}^+_p \) are quantum noise operators which replace the infinity of phonon operators \( \hat{b} \) and \( \hat{b}^\dagger \), and \( N_p(\Delta k) \) is the average number of thermal phonons in each phonon mode. The summation over \( q \) has dropped out because the resonance condition allows us to calculate it in terms of the \( k \) terms and the dispersion curves. The noise operators \( d\hat{B}_p, d\hat{B}^+_p \) are defined in the same way as in eqn (2.7).

B. Intensity and amplitude operator equations

Similarly to subsection II.B, I calculate some relevant examples of these QSDE’s, and convert them into rate and complex number equations. For simplicity, I write \( \hat{p}_+ \) as \( \hat{p}_+ \) and \( \hat{p}_- \) as \( \hat{p}_- \). The intensity for the \((+)\) mode is just \( \hat{I}_+ = \hat{p}_+^\dagger \hat{p}_+ \), that for the \((-)\) mode is \( \hat{I}_- = \hat{p}_-^\dagger \hat{p}_- \). The polariton coefficients I will also simplify, writing \( d_+(k) \) as \( d_+ \), and \( d_-(k - \Delta k) \) as \( d_- \), and \( \chi(k, k - \Delta k) \) as simply \( \chi \).

The QSDE equations for the intensity of the \((+)\) and \((-)\) modes due to the phonon scattering are

\[ d\hat{I}_+ = \chi |d_+ d_+^\ast|^2 \left[ -\hat{I}_+ (I_+ + 1) + N_p (I_- - I_+) \right] dt \]
\[ -\sqrt{\chi} [d_+ d_-^\ast \hat{p}_- \hat{B}_p + d_+^\ast d_- \hat{B}^+_p \hat{p}_-^\dagger] \]
\[ d\hat{I}_- = \chi |d_+ d_-^\ast|^2 \left[ \hat{I}_+ (I_+ + 1) - N_p (I_- - I_+) \right] dt \]
\[ + \sqrt{\chi} [d_+ d_-^\ast \hat{p}_+ \hat{B}_p + d_+^\ast d_- \hat{B}^+_p \hat{p}_+^\dagger]. \] (5.4)

As in the exciton case, the noise terms cannot be rewritten in a form depending solely on \( I_\pm \), so I leave them in terms of the \( \hat{p}_+, \hat{p}_- \). Again the terms in the pair of equations balance each other to preserve excitation number. Using the same procedure as in IV.A, these have a rate equation form

\[ \frac{d}{dt} \hat{I}_+ = \chi |d_+ d_+^\ast|^2 \left[ -\hat{I}_+ (I_+ + 1) + N_p (I_- - I_+) \right] \]
\[ \frac{d}{dt} \hat{I}_- = \chi |d_+ d_-^\ast|^2 \left[ \hat{I}_+ (I_+ + 1) - N_p (I_- - I_+) \right]. \] (5.5)

Similarly, the QSDE equations for the amplitude of the \((+)\) and \((-)\) modes due to the phonon scattering are

\[ d\hat{p}_+ = \chi |d_+ d_+^\ast|^2 \hat{p}_- \hat{p}_+^\dagger \hat{B}_p \]
\[ + \sqrt{\chi} d_+ d_-^\ast \hat{B}_p \hat{p}_-^\dagger \]
\[ d\hat{p}_- = \chi |d_+ d_-^\ast|^2 \hat{p}_- \hat{p}_+^\dagger \hat{B}_p \]
\[ + \sqrt{\chi} d_+ d_-^\ast \hat{B}_p \hat{p}_+^\dagger. \] (5.6)

Using the procedure referred to in section IV.A, these have a complex number form for the coherent amplitude, which is

\[ d\alpha_+ = \chi |d_+ d_+^\ast|^2 \left[ -\alpha_+ (\alpha_+^\dagger \alpha_- + 1) - N_p \alpha_+ \right] dt \]
\[ + \sqrt{2} \chi d_+ d_- N_p \alpha_+ \alpha_- dW_A \]
These polariton phonon scattering equations have the same features as the exciton equations derived in section II.B. This means the same comments apply here too—notably those about stimulated scattering. The new terms $dW$ are white noise increments, $dW_A$ and $dW_B$ are uncorrelated real white noises, and $dW_2$ is a complex white noise, where ($i = \{A, B, 2\}$)

$$\langle dW_i(t)dW_j(t')^* \rangle = \delta_{ij} \delta(t-t').$$

(5.8)

VI. POLARITON PHONON MODELS

Here I consider two models—one of a photoluminescence-like process and the other involving coherent excitation. Both models involve a pair of polariton branches that result from the coupling of a single exciton and a cavity. The polaritons are described by boson operators $\hat{p}_\pm(k)$, $\hat{p}_\pm^\dagger(k)$, indexed by in-plane wave vector. The splitting between the higher (+) and lower (−) energy polaritons is assumed to be less than the minimum optical phonon energy, so only acoustic phonons need to be included. The coupling to acoustic phonons is much weaker than that to optical phonons, and this causes the bottleneck effect.

The photon component of the polaritons can decay due to cavity losses, which I model by coupling to optical reservoirs. These represent the field modes outside the cavity. The cavity decay rate is $\gamma_L(k)$, with thermal photon number $N_L(k)$, and quantum noise operators $dB_L(k)$, $dB_L^\dagger(k)$. The exciton component of polaritons on either branch can recombine and emit photons, which I model by coupling each exciton mode to independent photon reservoirs. This recombination rate is $\gamma_E(k)$, with thermal photon number $N_E(k)$, and quantum noise operators $dB_E(k)$, $dB_E^\dagger(k)$.

The acoustic phonon process couples the (+) modes to all allowed (−) modes. The presence of thermal phonons is described using a thermal phonon number $N_P(k)$. Experiments done by Fainstein et al. \cite{13} for Raman scattering but in an otherwise similar situation show no power dependence in the spectra. This indicates that the effects of thermal phonons are likely to be negligible, so we can set $N_P(k) = 0$. Only the downwards $(+) \rightarrow (-)$ inter-polariton phonon scattering process is included here.

I will simplify both models with some physical considerations. Both the recombination reservoirs and the cavity decay reservoirs have very low thermal excitations ($N_L(k), N_E(k) \approx 0$), because the exciton and cavity mode energies are much greater than the thermal energy. Further, the recombination decay $\gamma_E(k)$ is typically a much slower process than the cavity decay rate $\gamma_L(k) \ll \gamma_L(k)$, and so can usually be ignored when the polariton has appreciable fractions of both cavity mode and exciton.

In the following equations, I will suppress the $k$ argument when $k = 0$ for brevity, so that $\hat{p}_\pm = \hat{p}_\pm(0)$, $d_\pm = d_\pm(0)$, $\gamma_L = \gamma_L(0)$, $\gamma_E = \gamma_E(0)$, $N_L = N_L(0)$, $N_E = N_E(0)$, and I also write $\chi(0, k)$ as $\chi(k)$.

A. Photoluminescence

The photoluminescence model is one in which the polariton is pumped as the free carriers excited by the driving field can combine in concert with the cavity mode to form polaritons. Exciton formation in PL is a complicated process, with free carriers combining to form hot excitons (with $k \gg 0$), and excited excitons (e.g. those with extra orbital momentum). These decay by LO or acoustic phonon emission or other scattering processes into the “cold” exciton states that can decay radiatively, and can therefore be seen optically. The situation with polaritons is similar.

A full description of this process is beyond the scope of this paper, so here I use an approximate model. I suppose that the recombination processes preferentially fill the upper polariton branch, something which could occur because the upper branch has higher energy than the lower branch for the same $k$. This agrees with the results that originally motivated this paper \cite{8} in which the upper branch is preferentially populated. Further, I assume the statistical properties of the newly created polaritons are influenced most strongly by the thermal nature of the free carriers they originated from. We suppose that the predominant effect of the optical pumping is that each mode (k value) of the upper polariton branch is driven by an independent thermal population, modelled by coupling to a set of reservoir modes which cause a weak loss $\gamma_C(k) \approx 0$, but have a high thermal occupancy $N_C(k)$ so there is significant thermal driving strength $M_C(k) = \gamma_C(k)N_C(k)$. The free carrier decay rate is small because the polaritons are unlikely to be scattered into free carriers.

I make the further simplifying assumption that the finite $k$ higher polariton states decay rapidly down into the $k = 0$ state due to optical and acoustic phonon scattering. As a result only the $k = 0$ upper polariton state is relevant, and I couple a single $k = 0$ upper polariton to a reservoir described by the parameters $\gamma_C$, $M_C$, and the quantum noise operators $dB_P$, $dB_P^\dagger$.

The complete equations for $\hat{I}_+(t) = \hat{I}_+(0)$, $\hat{I}_-(k)$ can then be constructed. Firstly, however, I can simplify them with the physical considerations given above. For the $(+)$ mode the sources of thermal noise are swamped by the free carrier component, since $M_C = \gamma_C N_C \gg$
Replacing the expectation value with a number \( I_k = \langle \hat{I}_k \rangle \), I can describe the system using rate equations –

\[
\frac{d}{dt} I_+ \approx \left[ -\gamma_L(0) |c_+|^2 I_+ + M_C \right],
\]

\[
\frac{d}{dt} I_- \approx \left[ -\gamma_L(k) |c_-|^2 I_- (k) + \chi(k) |d_+ d_-|^2 I_+ [I_-(k) + 1] \right].
\]

The exciton modes have steady state populations of

\[
I_+ = \frac{M_C}{|c_+|^2 \gamma_L},
\]

\[
I_- \approx \frac{\chi(k) |d_+ d_-|^2 I_+}{|c_-|^2 \gamma_L |\gamma_L(k)},
\]

where the equation for \( I_- (k) \) has been further simplified by reusing the fact that the phonon scattering is a weak process, with \( |c_-|^2 \gamma_L(k) \gg \sum_k \chi(k) |d_+ d_-|^2 \) and \( I_+ \) small. Each polariton mode emits photons at a rate given by the product of its decay rate \( |c_\pm(k)|^2 \gamma_L(k) \) and the occupation \( I_\pm \). For the upper \((+\) mode this is just \( M_C \). Using the \((+)\) mode population, the \((-)\) modes have populations of

\[
I_- = \frac{\chi(k) |d_+ d_-|^2 M_C}{|c_-|^2 \gamma_L |\gamma_L(k)}.
\]

The ratio of cavity decay intensities between all \((-)\) modes and the \( k = 0 \) \((+)\) mode is

\[
R = \sum_k \frac{\chi(k) |d_+ d_-|^2}{|c_+|^2 \gamma_L}.
\]

From this expression we can see that the relative strengths of the upper and lower polariton emission peaks is controlled largely by the strength of the phonon scattering between the two branches. As a result, the upper peak is larger than the lower peak. In contrast, a model that coupled the free carriers to both branches would populate them according to their Boltzmann factors, and would have the lower exciton peak as the larger because of its greater thermal population.

**B. Coherent excitation**

Now I will now consider a system where the higher energy polariton \((+)\) is coherently excited by a laser beam tuned to \( k = 0 \). The laser field is assumed to be strong enough so as to be unaffected by the energy lost in creating polaritons. This does not necessarily imply a 'high power' experiment, since even low intensity beams have
FIG. 4: The coherent excitation model. The thin curves are the polariton branches. The triangular shaded area gives the allowed energies and in-plane momenta of the acoustic phonons. The overlap of these two is the set of possible final states given that the initial state was at $k = 0$. The left hand side dashed arrow represents the coherent excitation $E = |\epsilon_+|^2$ of the upper $k = 0$ state, and the two right hand side arrows are the light emitted by decay from the polaritons through the cavity mode.

many photons, and the coupling between field and polaritons is weak. The rate that new polaritons are created is $\epsilon_+$. The (+) mode is dominated by its coherent driving, and so I write down the equation for the evolution of its amplitude. In contrast, the (−) mode is populated solely through the incoherent phonon scattering, and so has no coherent amplitude. As a result I use an equation for its intensity, which is non zero.

The complete equations for $\hat{P}_+ = \hat{P}_+(0)$, $\hat{I}_-(k)$ can then be constructed. Firstly, however, I can simplify them with the physical considerations given above by removing the weakest decay terms and the small thermal contributions. The phonon scattering itself is very weak. Because of this, we also have $\gamma_L(0) \gg \sum_k \chi(k) |d_+d^*_k(k)|^2$. In the typical situation where $\hat{I}_-(k)$ remains small because the lower polariton is only populated by this scattering process, this means that the effect of the phonon scattering terms in the equation for $\hat{P}_+$ is negligible.

Figure 4 shows a diagram of the model. The QSDE’s for it are

$$d\hat{P}_+ \approx \left\{ \epsilon_+ - \frac{1}{2} \gamma_L c_+^* c_+^{\dagger} \hat{P}_+ \right\} dt + \sqrt{\gamma_L c_+^* d\hat{B}_L}(0)$$

$$d\hat{I}_-(k) \approx \left\{ -\gamma_L(k) c_+^* c_-^{\dagger} \hat{I}_- + \chi(k) |d_+d^*_k(k)|^2 \hat{P}_+^{\dagger} \hat{I}_-(k) + 1 \right\} dt + \sqrt{\gamma_L(k) c_-^* d_-^{\dagger}(k) d\hat{B}_L}(k)$$

Since we do not need the quantum mechanical noise correlations, we can approximate them by taking the expectation values and factorising the different intensity moments (see section IV.A). Using $I_- = \langle \hat{I}_- \rangle$ and $\alpha_+ = \langle \hat{P}_+ \rangle$, I can rewrite the equations as complex amplitude and rate equations

$$\frac{d}{dt} \alpha_+ \approx \frac{\epsilon_+ - \frac{\gamma_L}{2} |c_+|^2 \alpha_+}{},$$

$$\frac{d}{dt} I_-(k) \approx \left[-\gamma_L(k) |c_-^{\dagger}(k)|^2 I_-(k) + \chi(k) |d_+d_-^{\dagger}(k)|^2 |\alpha_+|^2 [I_-(k) + 1] \right] (6.8)$$

The exciton modes have steady state values of

$$\alpha_+ = \frac{2\epsilon_+}{\gamma_L |c_+|^2},$$

$$I_-(k) \approx \frac{\chi(k) |d_+d_-^{\dagger}(k)|^2 |\alpha_+|^2}{|c_-^{\dagger}(k)|^2 \gamma_L(k)}, \quad (6.9)$$

where the equation for $I_-(k)$ has been further simplified by reusing the fact that the phonon scattering is a weak process, with $|c_-^{\dagger}(k)|^2 \gamma_L(k) \gg \sum_k \chi(k) |d_+d_-^{\dagger}(k)|^2$ and small $|\alpha|^2$. Each polariton mode emits photons at a rate given by the product of its decay rate $|c_\pm|^2 \gamma_L(k)$ and the occupation $|\alpha_+|^2$ or $I_-$. For the upper (+) mode this is just $|c_+(k)|^2 \gamma_L(k) |\alpha_+|^2$. Using the (+) mode population, the (−) modes have populations of

$$I_-(k) = \frac{\chi(k) |d_+d_-^{\dagger}(k)|^2 |\alpha_+|^2}{|c_-|^2 \gamma_L(k)} \approx \frac{\chi(k) |d_+d_-^{\dagger}(k)|^2 |\alpha_+|^2}{|c_-|^2 \gamma_L(k)} \cdot (6.10)$$

The ratio of cavity decay intensities between all (−) modes and the $k = 0$ (+) mode is

$$R = \sum_k \frac{\chi(k) |d_+d_-^{\dagger}(k)|^2}{|c_-|^2 \gamma_L(k)}. \quad (6.11)$$

From this expression we can see that the relative strengths of the upper and lower exciton emission peaks is controlled largely by the strength of the phonon scattering between the two exciton branches. As a result, the upper exciton peak is larger than the lower exciton peak. Note that in the limits considered here, this result is identical to that for the PL system.

VII. STIMULATED SCATTERING EXPERIMENT

The stimulated scattering is perhaps the most interesting effect to be predicted by this quantum model of phonon scattering. Given this, how would it be possible
The stimulated scattering experiment. The thin curves are the polariton branches. The triangular shaded area gives the allowed energies and in-plane momenta of the acoustic phonons. The overlap of these two is the set of possible final states given that the initial state was at $k = 0$. The left hand side dashed arrow represents the coherent excitation of the polariton states. The upper arrow is a continuous driving field $E_1 = |c_+|^2$ exciting the upper $k = 0$ state, and the lower is the pulse train that periodically excites the chosen lower state at $k = k_0$. The two right hand side arrows are the light emitted by decay from the polaritons through the cavity mode. The upper level emission has a series of dips caused by the stimulated scattering induced by the periodic excitation of the lower polariton level.

FIG. 5: The stimulated scattering experiment. The thin curves are the polariton branches. The triangular shaded area gives the allowed energies and in-plane momenta of the acoustic phonons. The overlap of these two is the set of possible final states given that the initial state was at $k = 0$. The left hand side dashed arrow represents the coherent excitation of the polariton states. The upper arrow is a continuous driving field $E_1 = |c_+|^2$ exciting the upper $k = 0$ state, and the lower is the pulse train that periodically excites the chosen lower state at $k = k_0$. The two right hand side arrows are the light emitted by decay from the polaritons through the cavity mode. The upper level emission has a series of dips caused by the stimulated scattering induced by the periodic excitation of the lower polariton level.

The coherent driving terms mean that it is most convenient to use the c-number amplitude equations. The equations are ($N_p = 0$)

$$\frac{d}{dt} \alpha_+ = \epsilon_+ - \frac{\gamma_+}{2} \alpha_+ + \frac{\chi'}{2} \alpha_+ (\alpha_- \alpha_+ + 1) + i \sqrt{\chi \alpha_+ \alpha_-} W_2,$$

$$\frac{d}{dt} \alpha_- = \epsilon_- (t) - \frac{\gamma_-}{2} \alpha_- + \frac{\chi'}{2} \alpha_- \alpha_+ \alpha_+ + \sqrt{\chi \alpha_+ \alpha_-} W_2 + i \sqrt{\chi \alpha_+ \alpha_-} W_2$$

The equations can be explicitly converted into those for a polariton system by setting $\alpha_+ \to \alpha_+(0)$, $\alpha_- \to \alpha_-(k_0)$, $\gamma_+ = \gamma_L c_+(0)^* c_+ (0)$, $\gamma_- = \gamma_L c_-(k_0)^* c_-(k_0)$, $\epsilon_+ = \epsilon_L c_+(0)^*$, $\epsilon_- = \epsilon_L c_-(k_0)^*$. It is also possible to do this kind of experiment in a two branch exciton system by using angled and appropriately tuned laser beams for direct excitation of the desired point on the exciton branch. Although in a PL setup a steady source of excitation might be supplied by the combination of free carriers into polaritons, this is neither $k$ specific, nor can it be rapidly modulated, nor can the individual branches be selectively excited.

I now expand equations (7.1) about their relaxed state, and write

$$\alpha_+ (t) = \alpha_{+0} + \beta_+ (t),$$

$$\alpha_- (t) = \alpha_{-0} + \beta_- (t),$$

where $\alpha_{\pm 0}$ are the 'relaxed states' of the two levels. The depletion $\beta_+$ of the upper level can be treated as a small perturbation, since it will be small compared to its level of excitation $\alpha_{+0}$. The relaxed state is like that for the bottleneck calculations so it has $\alpha_{+0} = 0$ because there is no coherent driving and the excitation due to phonon scattering is incoherent. The intensity $I_{-0} = \alpha_{-0}^2 \alpha_{-0} \approx \chi''/|\alpha_{+0}|^2/\gamma_+$ would be non zero because of the $W_2$ noise terms. Because the excitation of the lower level is very small so the change in its excitation $\beta_-$ cannot be treated as a perturbation. Inserting equations (7.2) into the equations above (7.1) gives us

$$\frac{d}{dt} \beta_+ = \frac{1}{2} \frac{\gamma_+}{\gamma_+} \left[ -\beta_+ - \frac{\chi'}{\gamma_+} \beta_+ (\alpha_{-0}^* \alpha_- - \alpha_{-0} \alpha_{-0}^* + \alpha_{-0}^* \beta_- + \beta_+ \beta_-) \right]$$

$$\frac{d}{dt} \beta_- = \epsilon_- (t) - \frac{\gamma_-}{2} \frac{\gamma_+}{\gamma_+} \left[ 1 - \frac{\chi'}{\gamma_+ \gamma_-} (\alpha_{+0}^* \alpha_{+0} + \alpha_{+0} \alpha_{+0}^* + \alpha_{+0}^* \beta_+ + \beta_+ \beta_-) \right] \beta_-.$$
the small parameter \( q = \chi'/\gamma_+ \). I have not defined new scaled variables, but just collected the factor together in an appropriate way.

The truncation to first order in \( \chi'/\gamma_+ \) is easy if \( \gamma_-/\gamma_+ \sim 1 \), and \( \beta_+^* \beta_- \sim 1 \). The condition on \( \beta_- \) still allows the lower level driving pulses to be non perturbative, but ensures that the depletion they cause in the upper level is perturbative. Further, since the depletion depends on the coupling, I assume \( \beta_+ \) is at most of order \( \chi'/\gamma_+ \). This assumption is self consistent with the resulting solutions.

Note that these conditions are needed for a simple theoretical solution for this problem, but the character of the results for more general conditions is unchanged. Truncating the equations to first order in \( \chi'/\gamma_+ \) involves neglecting any terms including two of \( \chi'/\gamma_+ \), \( \beta_+ \), \( \alpha_- \). The equations then can be easily simplified down to

\[
\begin{align*}
\frac{d}{dt} \beta_+ &= -\frac{\gamma}{2} \left[ \beta_+ - \frac{\chi'}{\gamma_+} \alpha_0 \beta_- \right], \\
\frac{d}{dt} \beta_- &= \epsilon_-(t) - \frac{\gamma_-}{2} \beta_-.
\end{align*}
\tag{7.4}
\]

I have introduced an effective decay rate \( \gamma_- = \gamma_+ - 2\chi' \alpha_0 \alpha_0 \).

Now I have set up the model, I can investigate the effect of the lower level driving pulses. Imagine one of the \( \epsilon_- \) pulses arrives at \( t = 0 \). For simplicity, I assume that the width of the pulse is much shorter than \( 1/\gamma_- \), so it can be treated as a delta function. Laplace transform methods then quickly give the simple exponential solution for \( \beta_- \) as

\[
\beta_-(t) = a_- \exp \left( -\gamma_- t/2 \right). \tag{7.5}
\]

The equation for \( \beta_+ \) is now completely defined, and is

\[
\frac{d}{dt} \beta_+ = -\frac{\gamma}{2} \beta_+ - \frac{\chi'}{2} \alpha_0 |a_-|^2 \exp(-\gamma_- t). \tag{7.6}
\]

This can also be solved by Laplace transform methods, to give the pair of solutions

\[
\begin{align*}
\beta_+ &= \frac{\chi' \alpha_0 |a_-|^2}{2\gamma} \exp(-\gamma t/2) \left[ 1 - \exp(-\gamma t) \right], \\
\gamma &= 2 \gamma_- - \gamma_+ \neq 0 \\
\beta_+ &= \frac{\chi \alpha_0 |a_-|^2}{2} \exp(-\gamma t/2), \\
\gamma &= 2 \gamma_- - \gamma_+ = 0.
\end{align*}
\tag{7.7}
\]

So the result is a dip in the excitation of the upper level, with a duration of about \( 1/\gamma_+ \) and a relative depth proportional to \( \chi' |a_-|^2 \) – note that this is consistent with the assumptions that \( \beta_+ \) was of order \( \chi'/\gamma_+ \) which was used in the truncation to first order. The size of the dips is proportional to the pulse intensity (and not its amplitude). A train of these input pulses exciting the lower level will stimulate phonon scattering in bursts and produce a corresponding series of dips in the upper level amplitude. These dips will show up in the optical decay from the upper mode, and could be detected by a peak in the output spectrum at a frequency given by their repetition rate. A single dip would be hard to detect, because it would be easily masked by noise. These dips show up in simulations of the equations – figures 6 and 7 show the behaviour of the outputs as a function of time for both the simple noiseless case as well as the case with quantum noise included.

Lastly, it is necessary to consider the effect of the other phonon scattering processes in a real system, since up to now we have restricted it to a simple two mode model, when in reality there will be scattering to other parts of the lower branch as well, and so the excitation in the chosen lower level \( k \) value will depend on re-scattered excitation as well as the driving pulses and the scattering from the upper level. However, these processes will merely provide a nearly constant background – mostly due to the excitation cascading down along the lower

FIG. 6: Graphs of (a) \( \alpha_+ \) and (b) \( \alpha_- \) as functions of time for the model without the quantum noise terms. The spectra for \( \alpha_+ \) and \( \alpha_- \) show the sidebands caused by the stimulated scattering.

FIG. 7: Graphs of (a) \( \alpha_+ \) and (b) \( \alpha_- \) as functions of time for the model with the quantum noise terms. The spectra for \( \alpha_+ \) and \( \alpha_- \) show the sidebands caused by the stimulated scattering.
branch through \( k = k_0 \) down to \( k = 0 \). The only way they will change the signal because of the pulse train is through second order processes. The effect of the reduced excitation has to scatter down into the lower level (one factor of \( \chi' \), then back up (another factor of \( \chi' \)). These types of processes already occur in the simple model, but were neglected due to their small size, and so will have negligible effect on the measurements.

Light could also scatter from the pulse train directly into the \( k = 0 \) detector. The strength of this could be measured by turning off the CW upper level driving field and retaining the pulse train. Also, any instability in the driving laser at a frequency about that of the pulse repetition should be reduced as much as possible to avoid contaminating the depletion signal. This should be minimised by stabilising the laser, or by adjusting the repetition rate to fall in a quiet part or the laser spectrum.

\[ k \]

VIII. CONCLUSION

This paper has presented a derivation of equations for phonon processes using the technique of quantum stochastic differential equations (QSDE’s). The procedure had the advantage of being a systematic derivation in which the approximations are clearly stated and whose effects are generally well understood. The technique was used to treat the phonon interaction as a coupling between the two polariton branches and an infinite reservoir of phonon modes, and properly accounted for the partly photonic nature of the polariton. The basic theoretical models of experimental situations used here in conjunction with the weak nature of acoustic phonon scattering predicts a “bottleneck” effect like that seen in recent low power polariton experiments. In addition, the rigourous quantum approach to the phonon scattering interaction revealed some new “stimulated scattering” terms whose existence would not have been obvious from a more phenomenological approach.

Further experimental investigations of this effect might take one of two paths. Firstly, the detuning of exciton and photon might be varied to check the change in strength of the bottleneck as the fractions of exciton and photon in the polaritons vary. The change in strength should mean that the ratio of the intensity peak areas should change in a corresponding way. The second path, and the one with more potential is to explore the use of coherent excitation. With appropriate tuning of the driving field and perhaps even control of its angle to the perpendicular, specific parts of the polariton branch could be excited and the resulting angular dependence of the luminescence measured.

An experiment was proposed to try to observe the “stimulated scattering” predicted by the quantum model used in this paper. However, whether it is possible to separate this effect out from the “background” of other processes is debatable. Theoretical avenues would involve treating the intra-branch phonon scattering properly, and doing more detailed modelling of particular experiments.

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