Parametric Processes in a Strong-Coupling Planar Microcavity

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I present a theoretical treatment of parametric scattering in strong coupling semiconductor microcavities to model experiments in which parametric oscillator behaviour has been observed. The model consists of a non-linear excitonic oscillator coupled to a cavity mode which is driven by the external fields, and predicts the output power, below threshold gain and spectral blue shifts of the parametric oscillator. The predictions are found to be in excellent agreement with the experimental data.

Recent experimental studies have demonstrated that very large optical non-linearities can be obtained in resonantly pumped strong-coupling microcavities. The excitations of these structures are polaritons, mixed modes which are part exciton, part cavity photon, and the non-linearity is due to interactions between the exciton components, which cause the polaritons to scatter off each other. This leads to a parametric process where a pair of pump polaritons scatter into non-degenerate signal and idler modes, while conserving energy and momentum. The scattering is particularly strong in microcavities because the unusual shape of the dispersion, shown in the inset to Fig. 1, makes it possible for pump, signal and idler all to be on resonance at the same time.

A further important property of planar microcavities is the correspondence between the in-plane momentum of each polariton mode and the direction of the external photon to which it couples. This makes it quite straightforward to investigate parametric scattering using measurements at different angles to access the various modes. Two types of process have been studied in this way: parametric amplification, where the scattering is stimulated by excitation of the signal mode with a weak probe field, and parametric oscillation, where there is no probe and a coherent population in the signal mode appears spontaneously.

Parametric amplification in microcavities was first observed by Savvidis et al., using ultrafast pump-probe measurements. The structure was pumped on the lower polariton branch at an incident angle of 16.5°. Narrow band gains of up to 70 were observed in the region of the polariton feature for a probe at 0°, along with idler emission at 35°. These are a set of angles for which the pair scattering resonance condition is satisfied. A related scattering process has been studied by Huang et al. using two pump beams at ±45° and a probe at normal incidence. The experimental results of Ref. 4 have been modelled by Ciuti et al. using a microscopic quantum treatment of polariton-polariton interactions.

Parametric oscillator behaviour has been observed Stevenson et al. and Baumberg et al., in CW experiments with the pump incident on the lower polariton branch at the ‘magic’ angle of about 16°. Above a threshold pump intensity, strong signal and idler beams were observed at about 0° and 35°, without any probe stimulation. The coherence of these beams was demonstrated by significant spectral narrowing, proving that they are due to a parametric process rather than resonantly enhanced incoherent photoluminescence. Houdré et al. have also observed a nonlinear emission at 0° for a structure pumped at 10°. However, in this experiment the pair scattering resonance condition is not satisfied, suggesting that different physics may be involved.

The purpose of this paper is to develop a simple classical model which provides a unified treatment of both amplifier and oscillator in the CW regime. This model is based on the textbook treatment of parametric phenomena in systems such as LiNbO3. Indeed, the microcavity behaviour has similar characteristics to a typical doubly-resonant parametric oscillator, where just the signal and idler modes are cavity resonances – the pump resonance is mainly important in enhancing the strength of the nonlinear effects. However, there are also significant novel aspects to the model: the microcavity operates in the strong coupling regime, where the modes are cavity polaritons not simple photons, and instead of a non-resonant \( \chi^{(2)} \) nonlinearity, the exciton provides a highly resonant \( \chi^{(3)} \) effect.

I. MODEL

The theoretical model is a classical treatment of a non-dispersive exciton mode, \( \psi(r) \), with energy \( \omega_p \), coupled to a cavity mode, \( \phi(r) \), with dispersion \( \omega_c(k) \), which is driven by the external fields. To account for broadening processes, \( \omega_s \) and \( \omega_i \) are taken to be complex energies, with imaginary parts \( \gamma_s \) and \( \gamma_i \) respectively. The exciton mode is non-linear, with potential energy \( V(\psi) = \frac{1}{2} \omega_x^2 \psi^2 + \frac{1}{4} \chi \psi^4 \).

To model the parametric processes, the cavity is driven by harmonic plane waves, consisting of a pump with amplitude \( F_p \) at \( (\omega_p, k_p) \) and a probe with amplitude \( F_s \) at \( (\omega_s, k_s) \). The cavity and exciton modes are also expressed as a sum of plane waves at \( (\omega_p, k_p) \) and \( (\omega_s, k_s) \), plus an idler at \( (\omega_i = 2\omega_p - \omega_s, k_i = 2k_p - k_s) \). The cavity mode is linear, so the equations of motion for the pump, signal and idler mode separate out, giving

\[
\begin{align*}
(\omega_c(k_p)^2 - \omega_p^2)\phi_p + g\psi_p &= F_p \\
(\omega_c(k_s)^2 - \omega_s^2)\phi_s + g\psi_s &= F_s \\
(\omega_c(k_i)^2 - \omega_i^2)\phi_i + g\psi_i &= 0
\end{align*}
\]
where \( g \) is the strength of the coupling between the exciton and the cavity photon. The exciton equations are more complicated because the non-linearity generates many terms at different frequencies and wave-vectors. Only the terms at frequencies \( \omega_p, \omega_s, \omega_i \) are retained here: the others are at very different frequencies, such as \( 3\omega_p \), or are weak, less than \( O(\psi_p^2) \). This leaves

\[
(\omega_x^2 - \omega_p^2) \psi_p + g \phi_p + \kappa |\psi_p|^2 \psi_p + 2\kappa \psi_s \psi_i \psi_p^* = 0 \tag{2}
\]

\[
(\omega_x^2 - \omega_s^2) \psi_s + g \phi_s + 2\kappa |\psi_p|^2 \psi_s + \kappa \psi_p^2 \psi_s^* = 0
\]

\[
(\omega_x^2 - \omega_i^2) \psi_i + g \phi_i + 2\kappa |\psi_p|^2 \psi_i + \kappa \psi_s^2 \psi_i^* = 0
\]

These equations can be simplified by using Eqs.\([3]\) to eliminate the cavity photon fields \( \psi \) and write everything in terms of the exciton fields \( \psi \). It is also convenient to approximate \( \omega_x^2 - \omega_p^2 \approx 2\omega_x(\omega_x - \omega_p) \) etc and define

\( \Omega = g/\omega_x \), \( \kappa = \frac{1}{2} \kappa/\omega_x \) and \( f = \frac{1}{2} F/\omega_x \). Then Eqs.\([3]\) become

\[
(\omega_x + \kappa |\psi_p|^2 - \omega_p - \frac{(\Omega/2)}{\omega_x(k_p) - \omega_p}) \psi_p + 2\kappa \psi_s \psi_i \psi_p^* = -\Omega/2 \kappa \omega_x(k_p) - \omega_p \tag{3a}
\]

\[
(\omega_x + 2\kappa |\psi_p|^2 - \omega_s - \frac{(\Omega/2)}{\omega_x(k_s) - \omega_s}) \psi_s + \kappa \psi_p^2 \psi_s^* = -\Omega/2 \kappa \omega_x(k_s) - \omega_s \tag{3b}
\]

\[
(\omega_x + 2\kappa |\psi_p|^2 - \omega_i - \frac{(\Omega/2)}{\omega_x(k_i) - \omega_i}) \psi_i + \kappa \psi_s^2 \psi_i^* = 0 \tag{3c}
\]

Eqs.\([3]\) constitute the basic model for parametric processes in a microcavity. The terms in \( |\psi_p|^2 \) represent the renormalisation of the exciton energy due to the pump population. The other non-linear terms provide the scattering, which is the main interest here: \( \kappa \psi_p^2 \psi_s \psi_i \psi_p^* \) and \( \kappa \psi_p^2 \psi_s^2 \psi_i^* \) in \([3a,3b,3c]\) describe the build-up of the population in the signal and idler modes, while \( 2\kappa \psi_s \psi_i \psi_p \) in \([3a]\) represents the corresponding pump depletion.

It is often useful to make the simplification of considering a situation where the pump, signal and idler energies are all close to the corresponding polariton resonance values, and the broadenings are small compared to the Rabi splitting \( \Omega \). Then it is a good approximation to replace the polariton response by a single Lorentzian function at each \( k \), with strength \( |\kappa|^2 \), where \( x \) is the exciton amplitude (Hopfield coefficient) for the mode. The driving terms on the right hand side of Eqs.\([3]\) can similarly be approximated by \( (c/x) f \), where \( c \) is the photon amplitude. Then, Eqs.\([3]\) reduce to

\[
\frac{1}{|x_p|^2} (\omega_p^2 + i\gamma_p - \omega_p) \psi_p + 2\kappa |\psi_s|^2 \psi_i \psi_p^* = \frac{c_p}{x_p} f \tag{4a}
\]

\[
\frac{1}{|x_s|^2} (\omega_s^2 + i\gamma_s - \omega_s) \psi_s + \kappa |\psi_p|^2 \psi_s^* = \frac{c_s}{x_s} f \tag{4b}
\]

\[
\frac{1}{|x_i|^2} (\omega_i^2 + i\gamma_i - \omega_i) \psi_i + \kappa |\psi_s|^2 \psi_i^* = 0 \tag{4c}
\]

where \( \omega_p^0, \omega_s^0, \omega_i^0 \) are the polariton resonance frequencies, and \( \gamma_p, \gamma_s, \gamma_i \) the corresponding widths. In writing the equations in this form, the exciton renormalisation is effectively ignored, though it can be considered to be included as a renormalisation of the polariton frequencies: at each point on the dispersion, it leads to a blue shift of approximately \( \frac{2\Omega}{|x|^2} |\psi_p|^2 \). For low pump powers, where the blue shift is small compared to the pump polariton width, and when \( \omega_p = \omega_p^0, \psi_p \) can be approximated, using Eq.\((4a)\), by \( \psi_p \approx -i c_p x_p^* f_p / \gamma_p \), and the blue shift is

\[
\delta \omega_p \approx \frac{2\Omega}{|x|^2} \frac{|c_p|^2}{\gamma_p} \frac{|x_p|^2}{f_p} = \frac{I_p}{I_0} \tag{5}
\]

where \( I_p = |f_p|^2 \) is the pump intensity.

It is interesting to compare the present classical model with the treatment in Ref.\([3]\), which gives a good fit to the pump-probe parametric amplifier experiments of Ref.\([3]\). This treatment was based on a quantum mechanical picture of the exciton-exciton scattering process. However, with the approximations that were made, Eqs.(1-3) of Ref.\([3]\) contain essentially the same physics as Eqs.\([3]\) here. Of course, using a microscopic model gives a value for the non-linearity \( \kappa \). However, \( \kappa \) only imposes a scale on the problem: rescaling all the fields so \( \psi \rightarrow \psi/\sqrt{\kappa} \), \( f \rightarrow f/\sqrt{\kappa} \), effectively makes \( \kappa = 1 \).

## II. PARAMETRIC AMPLIFIER

In the parametric amplifier, both \( f_p \) and \( f_s \) are non-zero. It is also helpful to assume that \( f_s \ll f_p \), so that \( \psi_s \) and \( \psi_i \) are small, and the pump depletion term in Eqs.\([3a,3b,3c]\) can be neglected. Consider first the situation when the probe, idler and pump satisfy the triple resonance condition, so Eqs.\([3]\) can be used with \( \omega_p = \omega_p^0, \omega_s = \omega_s^0 \) and \( \omega_i = \omega_i^0 \). Without the pump depletion term, these equations are solved by eliminating \( \psi_p \) and \( \psi_i \) using \([3a]\) and then \([3c]\), to get

\[
\psi_s = \frac{-i c_s x_s^* f_s / \gamma_s}{1 - \frac{1}{\pi^2} |x|^2 |x_p|^2 |x_s|^2 [c_p]^2 / |f_p|^4} \tag{6}
\]

Dividing by the value of \( \psi_s \) without the pump, i.e. with \( f_p = 0 \), gives the internal gain for the probe:

\[
\alpha_s = \frac{1}{1 - \frac{I_p^2}{I_0^2}} \tag{7}
\]

where

\[
I_0 = \frac{\gamma_p^2 \sqrt{\gamma_i \gamma_s}}{\pi |c_p|^2 |x_p|^2 |x_s||x_i|} \tag{8}
\]

The gain increases from unity at \( I_p = 0 \) to become singular at \( I_p = I_0 \). This suggests that \( I_0 \) represents the threshold pump intensity for oscillation, which will indeed be shown to be the case in the next section.

The previous discussion was limited to the case where all the fields are on resonance. If this condition is not satisfied, it is still possible to obtain an analytic solution
when the exciton renormalisation is neglected. Solving Eqs.\((3)\) with the same weak probe approximation gives

\[
\psi_s = \frac{-\left(\Omega/2\right) f_s / \Lambda_s}{1 - \pi^2 \left(\omega_s(k_s) - \omega_s(k_p)\right) \left(\omega_s(k_i) - \omega_s(k_p)\right) \left(\Omega^2/4\right)^4 \left|\Lambda_p\right|^4 \left|f_p\right|^4}
\]

where \(\Lambda_p = (\omega_s - \omega_p)(\omega_s(k_p) - \omega_p) - (\Omega/2)^2\), and \(\Lambda_s, \Lambda_i\) are similarly defined.

Fig.1 shows the signal response at normal incidence, calculated using Eq.\((3)\), for different pump angles. The pump energy is varied to be on the polariton resonance for each angle. The spectra show the two polariton features at normal incidence, with clear gain on the lower branch for pump angles in the region of \(16^\circ\): for this pump angle, the pair scattering resonance condition is satisfied when the probe angle is \(0^\circ\).

In Fig.2 the signal response is mapped out as a function of energy and angle with the pump kept on resonance at \(16^\circ\). There is an enhanced probe response when either the signal or the idler is on resonance – these single resonance energies are indicated by the dashed lines on the figure. The response is much stronger for the signal resonance, because the probe couples to the signal directly, but to the idler only via parametric scattering. The strongest response occurs at the double resonances, where the dashed lines intersect (at \(0^\circ, 16^\circ\) and \(33.5^\circ\)), but there is a long segment of the dispersion very close to double resonance, where the gain remains high.

III. PARAMETRIC OSCILLATOR

In the parametric oscillator regime, there is no probe, so \(f_s = 0\), but solutions can still be found with finite signal and idler fields. Again, it is simplest to start with the triply resonant case. Focusing on Eqs.\((4a, b, c)\), taking the complex conjugate of one of them, and treating \(\psi_p\) as a parameter, there are non-zero solutions for \(\psi_s, \psi_i\) only if the determinant of the coefficients is zero, that is:

\[
\pi |\psi_p|^2 |x_s||x_i| = \sqrt{\gamma_s \gamma_i}
\]

The physical interpretation of this condition is obvious – for a steady state solution, the generation rate of polaritons in the signal and idler directions, on the left hand side, must equal the (geometric) mean of the loss rates, \(\sqrt{\gamma_s \gamma_i}\). This is only possible with the value of \(\psi_p\) in Eq.\((10)\). For a given external driving field \(f_p\), the required value of \(\psi_p\) is attained by the depletion of the pump polariton field due to the stimulated scattering term.

The resulting signal intensity is calculated by using Eq.\((11)\) to write \(\psi_i\) in terms of \(\psi_s\), then substituting in Eq.\((10)\) to obtain

\[
|\psi_s|^2 = \frac{\gamma_i}{2\pi^2 |\psi_p|^4 |x_i|^2 |X_p|} \left|f_p - \frac{|\psi_p|}{|X_p|} \gamma_p\right| |\psi_p| |X_p| \gamma_p
\]

where \(|\psi_p|\) is now just a constant given by Eq.\((11)\). Since the emitted signal intensity, \(I_s\), is proportional to \(|\psi_s|^2\), this relationship is of the form

\[
I_s \propto \sqrt{T_p} - \sqrt{T_0},
\]

where \(T_0\) is the same threshold intensity as in Eq.\((8)\). Of course \(I_s \geq 0\), so this solution only exists when \(T_p \geq T_0\).

The treatment can be extended to the case where the signal direction is such that the signal and idler are not both on resonance, that is the mismatch \(\Delta = 2\omega_p - \omega_s^0 - \omega_i^0 \neq 0\). The steady state condition, Eq.\((10)\) becomes
\[\pi |\psi_p|^2 |x_s||x_i| = [\omega_s + i\gamma_s - \omega_s - (\omega_i - i\gamma_i)]^{1/2}, \quad (13)\]

with, once again, \(\omega_i = 2\omega_p - \omega_s\). A solution is only possible if the right hand side of Eq. (13) is real, which requires \(\omega_s - \omega_s^0 = \Delta \gamma_s/(\gamma_s + \gamma_i)\), and correspondingly \(\omega_i - \omega_i^0 = \Delta \gamma_i/(\gamma_s + \gamma_i)\). The physical significance of this requirement can be seen by looking at \(\psi_s\) and \(\psi_i\), for the allowed value of \(\omega_s\), \(\gamma_s|\psi_s|^2/|x_s|^2 = \gamma_i|\psi_i|^2/|x_i|^2\), that is, the loss rates through the signal and idler modes are identical. This is the Manley-Rowe relation for the parametric oscillator.

Continuing the solution for the signal intensity, for resonant pumping the effect of the finite mismatch \(\Delta\) is to shift the threshold, so

\[I_0(\Delta) = I_0(0) \left(1 + \frac{\Delta^2}{(\gamma_s + \gamma_i)^2}\right)^{1/2}, \quad (14)\]

where \(I_0(0)\) is the value given in Eq. (8). The threshold is lowest when \(\Delta = 0\) and pump, signal and idler are all on resonance.

These results show that the signal intensity is determined by the pump depletion, which produces the value of \(|\psi_p|^2\) required by Eqs. (10,13). For \(I_p > I_0\), \(|\psi_p|^2\) remains unchanged, just as the population inversion in a conventional laser is clamped at its threshold level. Since the actual value of \(|\psi_p|^2\) is unique to a particular pair of signal and idler directions, in equilibrium there can only be one finite signal amplitude. It is easy to see what will happen in an out of equilibrium situation when there is more than one signal. A signal whose loss rate exceeds its generation rate will decay, while one for which the generation rate exceeds the loss will grow. So, in a process akin to mode selection in a laser, the signal with the lowest loss rate will dominate, depleting the pump until only it survives.

**IV. DISCUSSION**

This model of the parametric oscillator makes two simple predictions which can be checked against experiment: the \(\sqrt{I_p}\) power dependence in Eq. (2), and the clamping of the pump polariton amplitude, \(\psi_p\), to the value given in Eq. (10). The latter effect should be observable as a saturation, above threshold, of the blue shift of the polariton dispersion. Below the threshold the shift is roughly linear with pump power, and it saturates at the value given in Eq. (2) with \(I_p = I_0\). For the signal mode the saturated shift is thus \(\delta \omega_s^0 \approx 2\sqrt{\gamma_s \gamma_i} |x_i|/|x_s|\). Of course, there are other effects, not included in the model, which can cause energy shifts. These include the exciton renormalisation due to the signal field, and at higher powers the break down of the strong coupling regime. The former effect can be estimated and is small: the signal field will give an energy shift of \(\pi |x_s|^2 |\psi_s|^2\), which is about 10% of the saturated \(\delta \omega_s^0\) when \(I_p = 2I_0\), using the same structural parameters as in Fig. 4.

**Fig. 3.** Theoretical fits to the experimental data (points) for (a) signal power and (b) blue shift from Ref. 5.
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