Collisional Broadening of Semiconductor Microcavity Polaritons

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We measure the effect of intensity on the homogeneous linewidth of two high finesse semiconductor microcavities in the strong coupling regime using degenerate four-wave mixing. We find that the collisional broadening due to polariton–polariton scattering shows a threshold behaviour with intensity. At low intensities, a strong suppression is seen due to the small density of states available. Above threshold, the number of accessible states dramatically increases, giving a large increase in the collisional broadening. We also investigate the effect of modifying the dispersion curve, by detuning, on the threshold behaviour.

Although the strong coupling of light–matter interaction is well established in atomic physics [1], it was not until 1992 that Weisbuch et al. [2] first saw strong coupling between an exciton and a cavity photon mode in a semiconductor. One major change when moving from atomic to semiconductor physics is that the structures become crystalline and periodic, which brings the concomitant property of dispersion. The modification of the excitonic dispersion [3, 4] when in a strongly coupled microcavity through the creation of new coupled states, cavity polaritons, has been a subject of much research [5–7]. The dispersion of the cavity polaritons has been found to strongly affect the efficiency of some scattering mechanisms [8].

The effect of the polariton dispersion curve is larger on scattering processes which do not conserve kinetic energy. Thus the dispersion curve only weakly affects scattering due to a static disorder potential, compared to acoustic phonon scattering and polariton–polariton scattering. Although polariton–polariton scattering, due to exciton Coulomb interaction, conserves the total kinetic energy, the kinetic energy of the individual polariton is not conserved. Scattering of the cavity polaritons by acoustic phonons has previously been investigated both theoretically [9, 10] and experimentally [11–13].

In this paper we look at the effect of the modified dispersion curve on the polariton–polariton scattering using time-integrated degenerate four-wave mixing (FWM) and experimentally demonstrate good agreement with the theoretical predictions of Ciuti et al. [14]. Previous work has been done on collisional broadening [15], but due to the high quality of the samples used in this study, the suppression seen in the samples is in better agreement with that theoretically predicted.

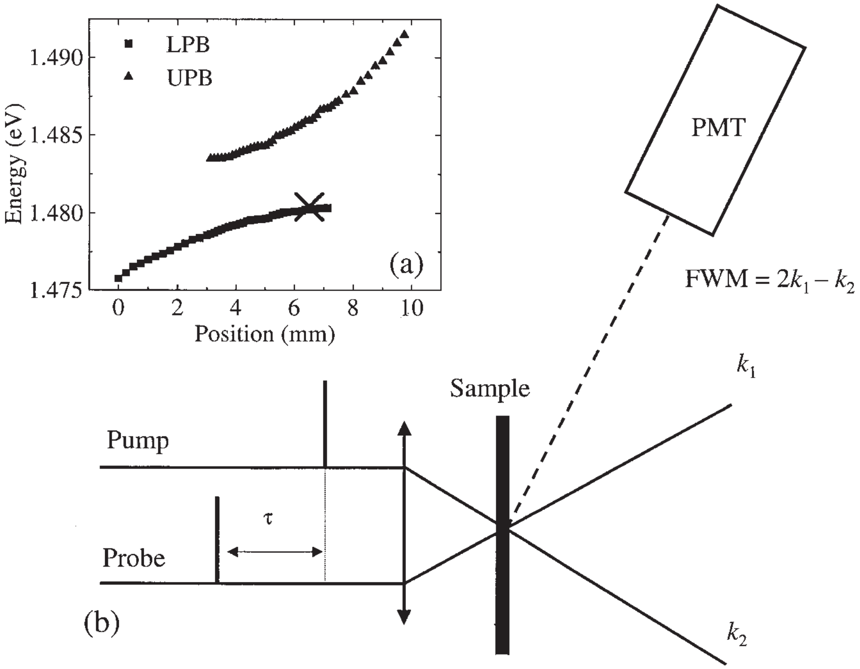

Fig. 1. a) Energy of the polaritons as a function of position on the two quantum well sample showing a Rabi splitting of 4.7 meV. X shows the position where the FWM data was taken. b) FWM set-up

The samples used in this study are strongly coupled microcavities showing the well known anti-crossing behaviour, see Fig. 1a. The two samples investigated consist of 20 front and 23.5 back distributed Bragg reflector (DBR) pairs, AlAs/$Al_{0.1}Ga_{0.9}As$. Bounded by the DBRs is a λ ($3\lambda/2$) cavity containing one (two) 80 Å thick $In_{0.05}Ga_{0.95}As$ quantum wells placed at the antinode(s) of the optical field inside the cavity. As is normal in MBE-grown samples, the wafers were not rotated during the growth of the cavity layer, which allows detuning of the cavity mode across the sample. The detuning across the sample is defined as $\Delta = E_{cav} - E_{exc}$, where E_{cav} is the energy of the cavity and E_{exc} is the energy of the exciton.

At zero detuning and a temperature of 8 K, the samples show a Rabi splitting of 3.5 meV (4.6 meV) for the one (two) quantum well sample and linewidths of 0.5 meV (0.6 meV) for the upper polariton branch (UPB) and 0.3 meV (0.4 meV) for the lower polariton branch (LPB), which illustrates the high quality of these samples.

Dephasing times were measured using time-integrated degenerate four-wave mixing in transmission geometry, see Fig. 1b. An Ar^+ pumped Ti:sapphire laser was mode-locked to give pulse widths of 1.1 ps and split into two pulses of equal power. At 1.1 ps these pulses provide enough spectral width to excite the LPB, but are narrow enough to avoid exciting the UPB. One pulse was delayed by a time τ with respect to the other pulse and the two beams were then focused onto the sample giving a spot of 40 μm in diameter. The angle between the two beams was less than 3° to ensure the excited states were at $k \approx 0$, where k is the in-plane wave vector. The measured FWM signal is the self-diffracted probe pulse from an intensity grating created by the interference of the two pulses. In this set-up, the diffracted signal is collected with an air-cooled photomultiplier tube, and time integrated using a dual lock-in detection system. The sample temperature was kept below 30 K in a closed cycle He-cryostat. The decay of the FWM signal is used to determine the linewidth Γ of the LPB. Dephasing times are derived from the decay of the FWM signal which follows $\exp(-2t/T_2)$ for a homogeneous system, where T_2 is the dephasing time. From this the homogeneous linewidth can be obtained using $\Gamma = 2\hbar/T_2$, where larger linewidths correspond to shorter dephasing times.

Figure 2a shows the FWM data taken on the two quantum well samples at a detuning of +4.9 meV. As the incident intensity is increased, initially there is no measurable change in the dephasing time up to an intensity of $7\ \mathrm{W\,cm^{-2}}$, where a decrease in dephasing time is seen. The corresponding change in linewidth $\Gamma - \Gamma_0$ with intensity is plotted in Fig. 2b, showing a clear threshold behaviour. Γ_0 is theoretically the linewidth with no incident intensity, but here it was taken to be the smallest linewidth measured. Hence, the increase in linewidth with intensity shows a threshold with intensity.

This threshold behaviour was predicted by Ciuti et al. in 1998 [14] and can be explained by looking at two factors which increase the polariton scattering efficiency: the density of states available for scattering into and the size of the exciton-like fraction of the polariton. The exciton is much more efficient at scattering than the cavity photon due to its effective mass $M_{\mathrm{cav}}/M_{\mathrm{exc}} = 10^{-4}$ at $k \approx 0$. At low intensities the collisional broadening is weak as there is only a small density of states for the polariton to scatter into near $k \approx 0$ and the polariton is not predominantly exciton-like. At higher intensities the collisional broadening increases allowing scattering to larger $|k|$ where there is a greater density of states and the states are more exciton-like. These factors dramatically increase the scattering efficiency leading to a threshold behaviour of the collisional

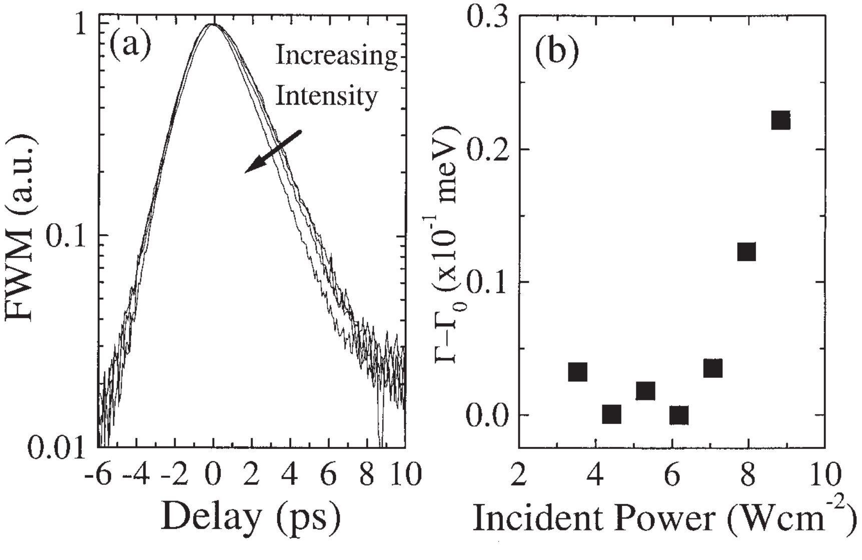

Fig. 2. a) FWM data for the two quantum well samples. b) Change in linewidth, showing a clear threshold behaviour with incident intensity

broadening. This threshold behaviour is measured by looking at the change in homogeneous linewidth due to the collisional broadening.

Figure 3 is a model of the dispersion curve for the two quantum well samples at at detuning of +4.9 meV. At low intensities (see Fig. 3a), Γ is extremely narrow allowing access to only a small density of states near $k \approx 0$. The small density of states due to the curvature of the LPB dispersion curve arises from the mixture of the flat dispersion curve of the uncoupled exciton and the parabolic cavity dispersion. As the intensity increases, the collisional broadening increases, Γ increases, but this increase is small due to the lack of states available to scatter into and the photonic-like part of the polariton. At a critical intensity (see Fig. 3b), the broadening becomes comparable to the energy difference between the LPB at $k = 0$ and the LPB at large $|k|$, where a large density of states is available for the polaritons to scatter into. These large $|k|$ states also have a greater exciton fraction which aids in the scattering process. Thus, at a critical intensity a rapid increase in the collisional broadening is seen.

It should be noted that the carrier density is well below that where the polaritons collapse. This was confirmed using femtosecond reflectivity where the polariton showed no shift in position or collapse at a carrier density of twice the maximum carrier density used here. Also the maximum intensity is within the carrier density where the polariton is predicted to collapse [16].

To further investigate the effect of the dispersion curve on the exciton–exciton scattering, we looked at three different detunings on the one quantum well sample. As the sample is scanned from positive to negative detuning, the LPB moves from being exci-

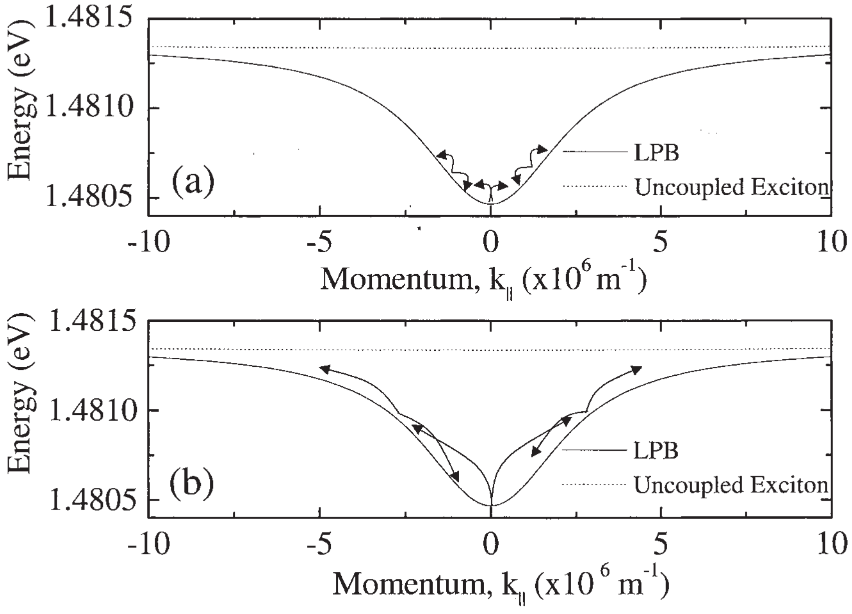

Fig. 3. Modelled dispersion curve at $\Delta = +4.9$ meV. The arrows are a pictorial representation of the polariton scattering. a) Below threshold the scattering is suppressed due to the low density of available states, but b) above threshold a larger density of states becomes available at high $|k|$ and the excitonic fraction of the lower polaritons increases. These effects give a sudden rise in the collisional broadening, i.e. a threshold

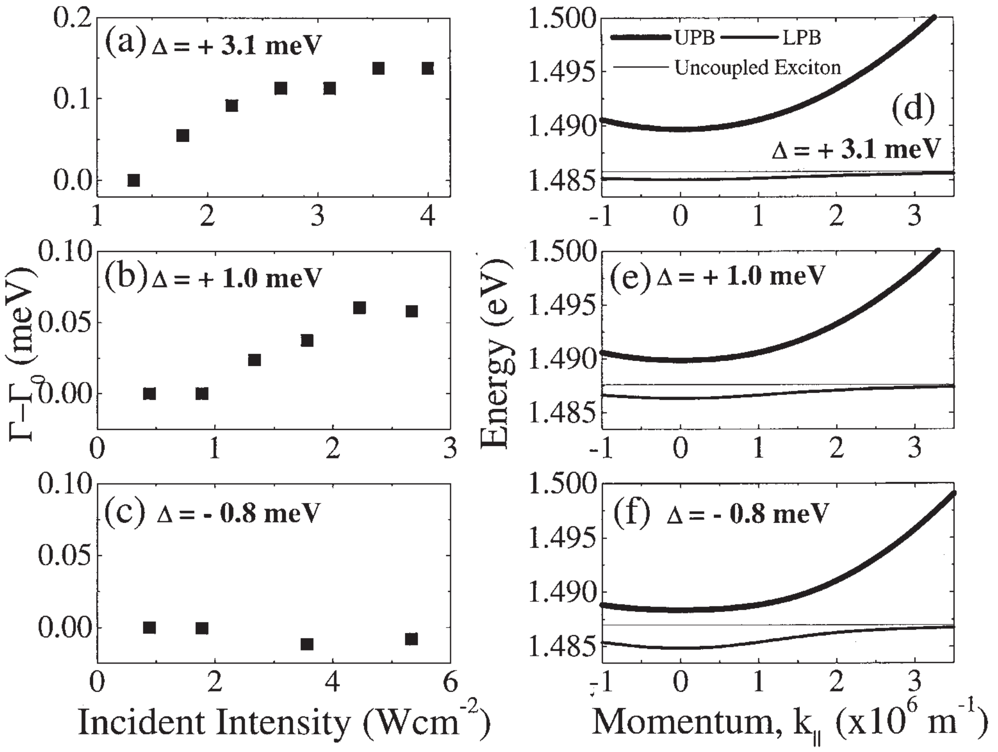

Fig. 4. Change in linewidth and dispersion curves for different detunings across the one quantum well sample. The threshold steadily increases as the sample is moved towards larger negative detunings. This can be explained by looking at the dispersion curves. Moving from (d) through (e) to (f), the energy difference between the LPB at $k \approx 0$ and the large $|k|$ states increases, as does the curvature of the LPB at $k \approx 0$, causing the threshold of the collisional broadening to increase

ton-like to cavity-like and the energy gap from the LPB at $k = 0$ to large $|k|$ increases (see Figs. 4d, e, and f). As we move towards more negative detunings, the threshold intensity increases, as illustrated in Fig. 4.

At a positive detuning of +3.1 meV (Figs. 4a and d), the threshold intensity is below that obtainable with this experimental set-up, so no threshold effect is seen. The low threshold is due to the LPB at $k \approx 0$ being close in energy to the large density of states at larger $|k|$. The linewidth of 0.7 meV of the LPB will allow access to the more plentiful larger $|k|$ states. In addition to this, at far negative detunings the lower polariton is very exciton-like and has a very flat dispersion curve, increasing the scattering efficiency. At high intensities, the linewidth can no longer be distinguished from the pulse and so the change in linewidth remains constant, i.e. the FWM is effectively measuring the decay of the pulse.

At a detuning of +1.0 meV (Figs. 4b and e), the energy gap between the LPB at $k = 0$ and large $|k|$ has increased, the threshold intensity has increased, and the threshold behaviour of the collisional broadening can be seen clearly. However, at a detuning of −0.8 meV (Figs. 4c and f), no threshold is seen before pumping at intensities where we can no longer assume the polariton is in the strongly coupled regime [16].

(Although the incident intensity is less than that of the data taken at positive detunings on the two quantum well sample (Fig. 2b), the absorption is much larger. The absorption of the LPB branch decreases as it is detuned away from resonance.) This threshold increase is due to the steeper dispersion curve of the LPB and the increased cavity-like behaviour of the lower polariton at negative detunings, decreasing the scattering efficiency.

Comparing the two samples, it is important to note that although the polariton linewidths are similar in both, the splitting of the one quantum well sample, 3.5 meV, is smaller than that of the two quantum well sample, 4.6 meV. This implies that the two quantum well sample has a larger coupling strength, for a fixed detuning the exciton–photon coupling is larger. At a fixed positive detuning, the LPB of the two quantum well sample would have a larger photon-like fraction causing the carrier density needed to observe threshold behaviour to be greater than the LPB in the one quantum well sample. The LPB in the one quantum well sample would have a smaller photon-like fraction and is therefore more exciton-like causing threshold to occur at smaller carrier densities. This behaviour was seen in our samples; at a detuning of +4.9 meV a threshold effect could still be observed in the two quantum well sample while at a detuning of +3 meV in the one quantum well sample threshold behaviour could not be seen as data could not be recorded at the low carrier densities where the threshold behaviour was happening.

In conclusion we have seen a strong inhibition of polariton–polariton scattering below a threshold intensity at positive detunings in two strongly coupled microcavity samples. We have seen a clear threshold. At negative detunings it is no longer possible to see the threshold behaviour as the threshold intensity is above the intensity at which the strong coupling collapses.

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