Excitation-induced coherence in a semiconductor microcavity

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We measure the dephasing times of high finesse semiconductor microcavities in the strong coupling regime using degenerate four-wave mixing. We find that under certain excitation conditions (near-normal incidence) the dephasing time of the lower polariton branch increases with the excitation intensity. This excitation induced coherence indicates that the lower polariton is acting as a Bose particle.

In most physical systems, as the density of excitations increases, their coherence time decreases due to scattering. Systems where the opposite occurs are rather unique systems such as superconductors, superfluids, Bose-Einstein condensates, and lasers. In each case, the increase in coherence is related to the bosonic nature of the excitations which stimulates scattering into the lowest energy state.

For direct gap semiconductor systems the lowest energy states are excitons and their coherence can be probed through optical experiments such as degenerate four-wave mixing (FWM). Excitons in quantum wells (QW's) have been studied in great detail and their dephasing mechanisms are well established and the following features are universally observed[1,2]: (1) the temporal behavior of the four-wave mixing signal is independent of incident angle, and (2) the dephasing rate increases with increasing excitation intensity.

When a quantum well is embedded in a high finesse microcavity, the interaction between the Fabry-Pérot mode and the exciton gives rise to an anticrossing behavior in energy as measured in reflectivity.[3] The coupled excitations, which we will refer to as cavity polaritons, have been the subject of much recent research.[4,5] We have studied the coherence of cavity polaritons with small in-plane momentum (i.e., $k_\parallel \simeq 0$) through FWM. We find that (a) the dephasing rate depends on the excitation angle; and (b) for small angles (very close to $k_\parallel = 0$) the dephasing rate decreases with increasing excitation intensity, i.e., we observe excitation induced coherence. Not only is the behavior very different to that of QW excitons, but the increase in coherence with excitation indicates that bosonic nonlinearities play an important role in this system.

The paper is laid out as follows. First we describe the microcavity sample, which is of exceptional quality; then we describe the FWM experiment and the variation of the dephasing time with angle and with intensity. Finally, we discuss these results and show how the difference between cavity-polaritons and quantum well excitons is related to their energy-momentum dispersion relations.

The sample used in this study is a strong coupling microcavity showing the well-known anticrossing behavior. The sample consists of 20 (front) and 23.5 (back) distributed Bragg reflector (DBR) pairs, AlAs/Al$_{0.1}$Ga$_{0.9}$As. Bounded by the DBR's is a $3\lambda/2$ cavity containing two 80-Å-thick In$_{.05}$Ga$_{.95}$As quantum wells, one placed at each antinode of the optical field inside the cavity. The wafer was not rotated

during the growth of the cavity layer which allows the detuning of the cavity mode across the sample.[6] All the experimental data were taken close to zero detuning, where $E_{cav} \simeq E_{exc}$. At zero detuning [see Fig. 1(a)], the sample shows a Rabi splitting of 4.7 meV and linewidths of approximately 0.4 and 0.2 meV for the upper polariton branch (UPB) and the lower polariton branch (LPB), respectively.

Dephasing times were measured using time integrated degenerate four-wave mixing in transmission geometry; see Fig. 1(b). An argon ion 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 t 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 measured FWM signal is the self-diffracted probe pulse from an intensity grating created by the interference of the two pulses. The intensity of the FWM signal depends on the strength of the χ^3 nonlinearity, and the coherence of the system can be tested by delaying the arrival time of the second pulse. In this setup, the diffracted signal is collected with an air-cooled photomultiplier tube, and time integrated using
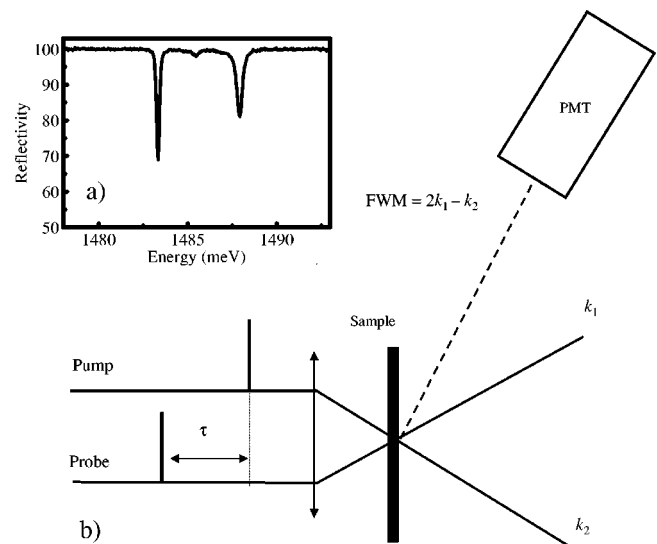

FIG. 1. (a) Reflectivity of the polariton showing a Rabi-splitting of 4.7 meV and linewidths of approximately 0.4 and 0.2 meV for the UPB and LPB respectively; (b): FWM setup.
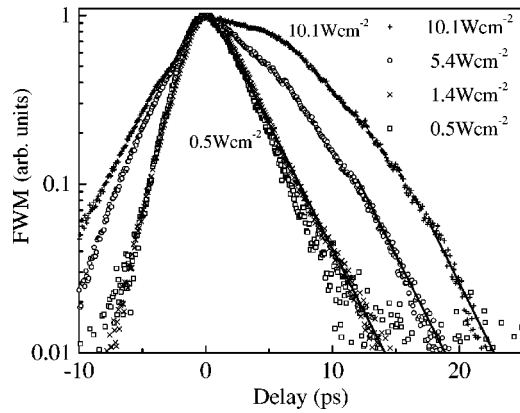
FIG. 2. FWM data taken for a range of optical intensities at an angle of 1.5° showing an increase in decay time with intensity. The second slope has a line for guidance.
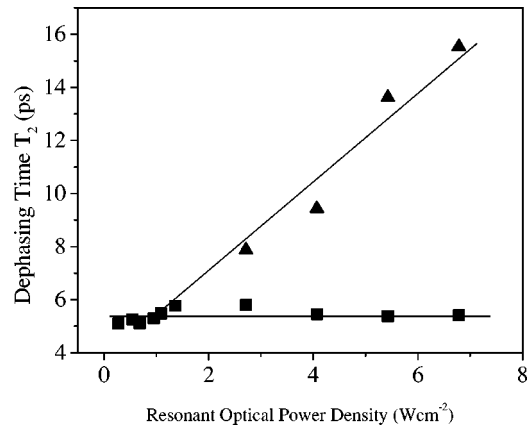

FIG. 3. Dephasing times versus optical power density. There is no significant change of the dephasing time of 5 ps up to an optical power density of 1.4 W cm^{-2}, whereupon the polariton shows two dephasing times. An initial long dephasing time (triangles) followed by a shorter dephasing time (dots). The shorter dephasing time remains constant, within experimental error, at 5 ps.

a dual lock-in detection system. The sample temperature was kept below 20 K in a closed cycle He cryostat.

We measured the FWM signal as a function of delay time between the two pulses for different angles between the two beams and for various excitation intensities. The dephasing times can be 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.

Figure 2 shows the FWM data taken at an angle of about 1.5°, for a range of optical intensities. Even at these narrow angles the diffracted signal is still well separated from the transmitted pulse beam with the geometry of our setup. Dephasing times of 5 ps are measured up to a resonant optical power density of 1.4 W cm^{-2}. Resonant optical power densities take into account the spectral overlap of the polariton and laser. At higher photon densities the decays no longer exhibit a single slope. The lower part of the decay stays constant with the excitation density, while the upper part becomes flatter with increasing excitation density. In order to give a quantitative analysis, we estimated dephasing times by fitting the first and last parts of the decay and these are shown in Fig. 3. At long times all the FWM data show the same decay time, 5 ps, while at short times the deduced dephasing times increase from 7 ps at 2.7 W cm^{-2} to 15 ps at 6.8 W cm^{-2}. The rise time of the FWM signal also lengthens along with the decays at higher intensities, as expected for a homogeneously broadened system.[2,7] These results show a clear decrease in the dephasing rate at *early* times and *high* intensities.

The excitation induced coherence is only observed at small angles. If the incident power is kept constant and the angle between the two beams is increased, then the dephasing rate becomes faster and the decay becomes monoexponential; see Fig. 4. For angles above 4° the dephasing rate always increases with excitation intensity, i.e., we get back to "normal" QW exciton behavior.

As mentioned above, dephasing rates normally increase with the excitation intensity due to increased collision rates. However, when there is a small angle between the pump and probe, we observe the opposite. Before attributing this behavior to an excitation induced coherence, the possibility that

this is an experimental artifact rather than a real change in dephasing dynamics needs to be ruled out. First we checked that the laser pulses were stable by replacing the sample with a second harmonic crystal. This showed that there was no double pulsing. Second, detector saturation was eliminated by checking the linearity of the detectors. Attenuating the second harmonic signal with neutral density filters, confirmed that the detector response was linear at up to ten times the maximum measured FWM signal.

A third possibility is that the decay of the FWM could be lengthened if the LPB shifted in energy with time. A shift away from the excitation laser wavelength would give a prolonged decay of the FWM signal, while a shift toward the laser could give an enhanced decay. The spectral width of the pulse is wide enough to compensate for some movement of the LPB. Any significant shift in the LPB at high intensities would show up as a change in the amount of the pulse re-
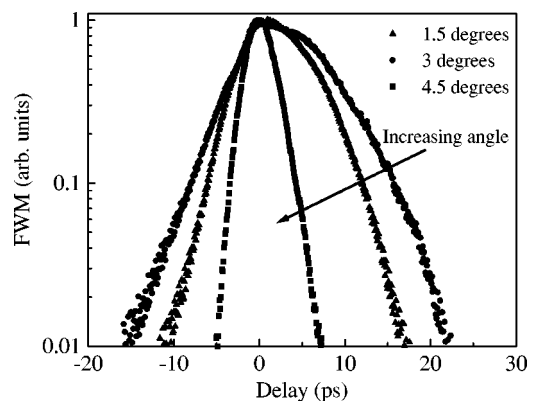

FIG. 4. FWM signal as a function of angle at an optical power density of 10.1 W cm^{-2}. At larger angles the FWM signal shows no anomalous behavior as the dephasing rate has decreased with excitation, displayed here it is no longer distinguishable from the pulse. In contrast, at the smallest angles the dephasing rate has steadily increased with excitation. The double slope can also be observed.

flected by the sample. The reflected light was measured as a function of incident power. However there was no change in the fraction of light reflected within experimental error.

This possibility was investigated further by measuring the reflectivity spectrum of the polariton as a function of incident power. This was done using the laser in the femtosecond mode; the increase in the spectral width enables the reflectivity of both polariton branches to be measured. We observed no shift of the polariton lines with intensity even up to the point where the polariton began to collapse, at a resonant optical power density of 28.3 W cm^{-2}.[8,9] Effects due to higher order chi terms and cascade processes have been ruled out for two reasons. First, to the authors' knowledge higher order terms would only result in faster dephasing rates; see, for example, Ref. 10. Second, although higher order terms may interfere with our FWM signal they would also give rise to angularly resolved signals, something we do not observe at the intensities used here. The detection system was maximized for the FWM signal and an aperture eliminated the straight through transmission and higher order χ processes (which already have a greater angular separation from the FWM signal than the through transmission). Having ruled out experimental artifacts, we conclude that the decreased dephasing rate at higher intensities is real.

Several important experimental observations should be noted. First, the long dephasing times only occur for small angles between the two beams, the angles at which excitation induced coherence is observed are less than 4.5° which is the angular linewidth of the LPB. Second, we are still in the strong coupling regime for the FWM data taken at the highest carrier density, as it is less than half the resonant optical density at which the polariton begins to collapse. Third, the same behavior with angle and intensity was observed in samples of similar quality but with one or three quantum wells instead of the two for the sample shown here.

The effect of collision broadening on the homogeneous linewidth of the LPB was theoretically investigated by Ciuti et al.[11] They predicted that, due to the modified dispersion, the effect of collisional broadening should be much weaker for the LPB exciton than for the QW exciton, up until a threshold intensity after which LPB collisional broadening is enhanced. This is indeed what was observed in our lab and elsewhere.[12,13] However, an increase in coherence with intensity is totally unexpected. It should be noted that the results presented here are taken near resonance where no collisional broadening is predicted below the saturation density of the polariton[11].

The dephasing of excitons has been theoretically treated with various levels of complexity ranging from the optical Bloch equations for a two level system through to the semiconductor Bloch equations in the Hartree-Foch limit and beyond. A simple interpretation does not exist in any these approaches because there are no low order terms which add to the coherence of the system. Instead we start from the two-level approximation, which is often used at low excitation intensities.[14] The decay of the coherence of a two level system is governed by the decay of the population T_1 and pure dephasing mechanisms T_2^* such as scattering. The net coherence lifetime T_2 is thus given by
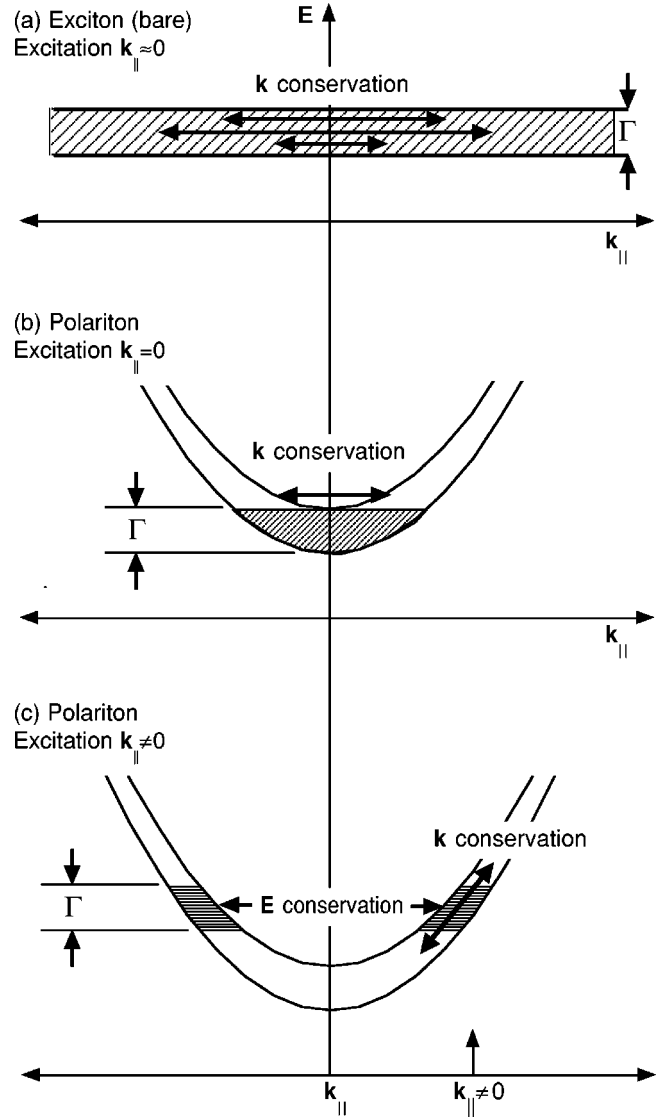

FIG. 5. (a) The energy-momentum dispersion of the uncoupled exciton line. Γ is the linewidth of the polariton. Backscattering from $k_\parallel \neq 0$ will be small, due to the flat dispersion curve. (b) The energy-momentum dispersion of the lower cavity polariton branch. The polaritons population when excited at $k_\parallel = 0$ is the area shaded in. The steeper dispersion results in almost no k-space for scattering whilst maintaining energy conservation. (c) The energy momentum of the lower cavity polariton branch showing the polariton population which has scattered to $k_\parallel \neq 0$. These polaritons may scatter back to $k_\parallel = 0$ due due to stimulation by occupation, a bosonic behavior.

$$\frac{1}{T_2} = \frac{1}{2T_1} + \frac{1}{T_2^*}, \tag{1}$$

where the factor of 2 comes from the loss in intensity rather than the amplitude. The shorter of these two decay mechanisms will dominate. The population decay T_1 is probably similar to the cavity decay time which is about 10 ps in these high finesse microcavity samples.[15] At low intensities we measure dephasing times of 5 ps, implying a T_2^* of about 6–7 ps which is typical of QW excitons.[16,17] Thus the

dephasing time is primarily governed by scattering (pure dephasing) rather than radiative recombination (population decay). Experimentally, as the intensity increases the measured dephasing time increases. This change cannot be due to an increase in T_1; instead only an increase in T_2^* will have an appreciable effect on the dephasing rate. We conclude that the effect of increasing intensity is to either inhibit scattering or to compensate out-scattering with in-scattering.

Consider a coherent population of excitons created at $k_\parallel = 0$ [see Fig. 5(a)]. Exciton-exciton collisions will scatter the excitons away from $k_\parallel = 0$. The energy-momentum dispersion is so flat that the $k_\parallel = 0$ excitons occupy only a small part of the available k space, so that backscattering from $k \neq 0$ will be small and these excitons will be dephased with respect to the original population. This type of scattering decreases the coherence as measured in FWM experiments, and increases the linewidth as observed in cw experiments. For cavity polaritons the dispersion[18] [see Fig. 5(b)] is 10^4 steeper than that of bare excitons, with the result that the polaritons have almost no k space for scattering while maintaining energy conservation. This reduces the effects of collisional broadening. (There will still be some scattering away from $k_\parallel = 0$, e.g., guided modes.) Now there are two possibilities. (1) Nonbosonic behavior. The increase in density leads to more collisions, exchange, and screening, all of which decrease the dephasing time with intensity. (2) Bosonic behavior. As the density increases the scattering away from $k_\parallel = 0$ can be compensated for by stimulated scattering back to the $k_\parallel = 0$ state. The stimulation term depends on the number of states already at $k_\parallel = 0$; see Fig. 5. Eventually a condition will be reached where the stimulated scattering dominates over the collisional (spontaneous) scattering and the coherence time of the system should increase dramatically. If the initial states are at $k_\parallel = k_i \neq 0$, then the stimulated scattering will cause a relaxation to $k_\parallel = 0$ and an increase in the dephasing rate as measured at k_i. The mechanisms of both stimulated scattering[19–21] and parametric amplification have already been observed in microcavity systems,[22,23] lending support to our interpretation. However, it has been shown that this interpretation of FWM is over simplified when looking at polaritons,[24] and we encourage theorists to extend the recent body of work[25,26] with a view to giving a fuller interpretation of these results.

FWM through χ^3 is a probe of the coherence of polaritons[27] rather than the photon field, and this enables us to distinguish between coherence due to stimulated photon emission (i.e., lasing) and coherence in the polariton population. The FWM setup was in the degenerate configuration, as described above, and although not directly incident we feel the necessary condition $k_\parallel = 0$ was satisfied at an angle around $1.5°$. The necessary conservation was broken at larger angles, $k_\parallel \neq 0$, where no anomalous effects were seen. Indeed, the coherence of the polaritons behaves similarly to the coherence of the photon field in a laser as one approaches threshold, i.e., it increases dramatically. Nondegenerate four-wave mixing at the so-called magic angle might give similar results. At the magic angle, energy and k conservation can still be maintained in a polariton-polariton scattering process,[22] but, as Giacobino pointed out, $k = 0$ is also a "magic" angle.

In conclusion we have observed an intensity dependent increase in the coherence time of the LPB using FWM. We have demonstrated that the excitation induced coherence is not due to any experimental artifact or a shifting of the LPB energy. For composite particles such as excitons and cavity polaritons the underlying particles are fermions and at high densities their fermionic nature dominates. However, our observations indicate that for cavity polaritons at least one can reach densities where a dramatic increase of the coherence time with intensity occurs, which is typical of composite bosons. We actively encourage theoretical work to investigate this effect. It remains to be seen whether condensation can be observed in this system.

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[7] The data also shows an anomalous bump at high intensities near -5 ps; $+5$ ps is where the slope changes and it may be that these two effects are linked.

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