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Biexcitonic effects in the nonperturbative regime of semiconductor microcavities

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Polarization-dependent transient pump-probe spectroscopy reveals important effects of biexcitonic interactions on coupled optical excitations in semiconductor microcavities. We show that exciton-to-biexciton transitions can result in a significant increase in normal mode splitting of cavity-polaritons, in sharp contrast to effects of nonlinear optical interactions such as band filling that reduce the splitting. A phenomenological model based on Maxwell-Bloch equations is developed to elucidate how biexcitonic effects contribute to cavity-polaritons. [S0163-1829(98)50120-8]

Planar semiconductor microcavities embedded with quantum wells (QW) have been used as a composite exciton-cavity system to investigate excitonic optical interactions in a nonperturbative regime where collective dipole coupling rates between the exciton and the cavity mode are large compared with relevant cavity decay rates and exciton dephasing rates. Resonant optical excitations of the composite system in this regime are characterized by coupled exciton-cavity modes, or cavity polaritons. Extensive studies of semiconductor microcavities have shown normal mode splitting (NMS) of cavity polaritons in emission and reflection spectra and normal mode oscillation in transient optical responses.[1,2] Motional narrowing of cavity polaritons in a disordered potential has also been investigated recently.[3]

Strong coupling between the exciton and the cavity mode in the nonperturbative regime also leads to unusual manifestations of excitonic nonlinear optical interactions. Since NMS between two cavity-polariton branches reflects the collective dipole coupling strength between the exciton and the cavity mode, nonlinear optical processes such as phase space filling that saturate the excitonic transition reduce the magnitude of NMS,[4] while processes such as excitation-induced dephasing (EID) primarily broaden the cavity-polariton resonance.[5]

Biexcitonic interactions are also expected to affect optical excitations in the nonperturbative regime. Biexcitonic effects were shown to be important in understanding coherent nonlinear optical processes such as four-wave mixing of cavity polaritons.[6] A clear physical understanding of how biexcitons contribute to coupled excitations in semiconductor microcavities, however, is still lacking in part because biexcitonic effects cannot be easily incorporated into the widely used semiconductor Bloch equations.[7]

In this paper we present experimental and theoretical investigations on unique manifestations of biexcitonic effects in the nonperturbative regime in a microcavity embedded with GaAs QWs. Using polarization-dependent transient pump-probe spectroscopy, we found that biexcitonic interactions can result in a significant *increase* in NMS of cavity polaritons, in sharp contrast to effects of nonlinear optical interactions such as phase space filling that reduce the NMS. We attribute the observed increase in NMS to coupled excitations associated with the exciton-to-biexciton transition. A phenomenological model based on Maxwell-Bloch equations is also developed to elucidate how biexcitonic effects contribute to optical excitations in semiconductor microcavities. Our result underscores the necessity to include biexcitonic interactions in theoretical descriptions of optical interactions in the nonperturbative regime in semiconductor microcavities and should also impact interpretations of many other nonlinear optical studies in semiconductor microcavities.

The microcavity structure used in our study contains four 13-nm $GaAs/Al_{0.3}Ga_{0.7}As$ QWs placed at the center (antinode) of a wavelength long cavity. The two Bragg reflectors of the cavity consist of 16 and 22 pairs of $Al_{0.11}Ga_{0.89}As/AlAs$, respectively. The heavy-hole (hh) exciton absorption linewidth is estimated to be 1 nm, indicating that excitons are inhomogeneously broadened. The empty cavity linewidth is 0.25 nm. Additional information on the sample can also be found in earlier studies on laser emission from semiconductor microcavities.[8] Transient pump-probe studies were performed in the reflection geometry and with output from a mode-locked Ti:Sapphire laser with a pulse duration of 150 fs and a repetition rate of 82 MHz. All measurements were carried out at 10 K.

Figure 1 shows as dashed-lines reflection spectra of the sample obtained at low excitation limit. The NMS observed is 2.6 nm. Note that the linewidth for the upper (higher-energy) cavity polariton is considerably greater than that for the lower cavity polariton even though the cavity is at or very near the hh exciton absorption line center. The asymmetric linewidth, which has also been observed in numerous earlier studies, is likely the result of an asymmetric inhomogeneous line shape (or motional narrowing) in a QW with interface disorders.[3] Effects of light-hole excitons may also play an important role.

Reflection spectra when the sample is pre-excited by a resonant pump pulse are shown as solid lines in Fig. 1. When the pump-and-probe pulses have the same circular polarization, a large reduction in the NMS due to bleaching of the excitonic transition occurs as shown in Fig. 1(a). Significant broadening of the cavity-polariton resonance due to EID is

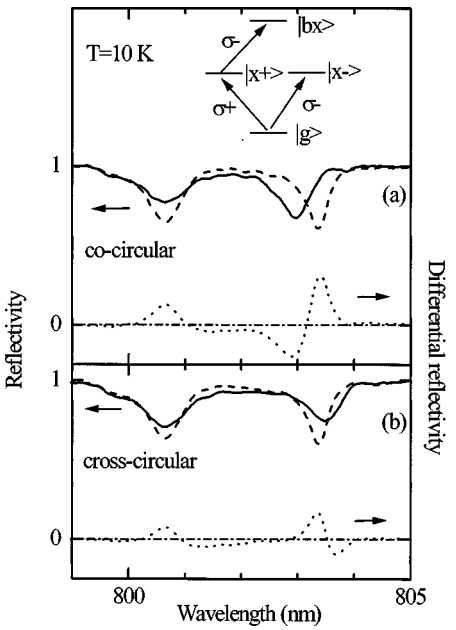

FIG. 1. Pump-probe reflection spectra. The dashed lines show as a reference spectra obtained in the absence of the pump beam. The solid lines are obtained when the sample is pre-excited with a resonant pump beam. The dotted lines represent the corresponding differential spectra. The pump-and-probe beams have the same and opposite circular polarization for (a) and (b), respectively. The inset shows the energy-level structure used to model the biexcitonic contribution.

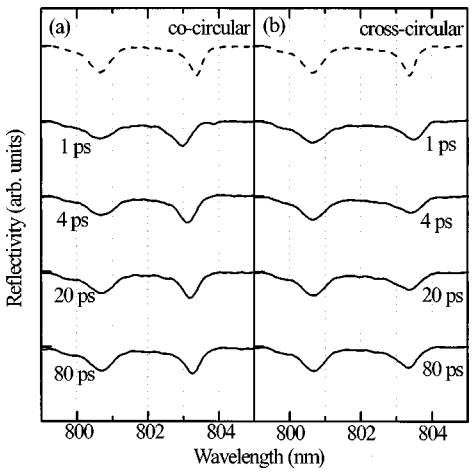

FIG. 2. Pump-probe reflection spectra as a function of the delay between the pump-and-probe beams as indicated in the figure. The dashed lines show as a reference reflection spectra obtained in the absence of the pump beam. The pump-and-probe beams have the same and opposite circular polarization for (a) and (b), respectively.

also evident. In comparison, when the pump-and-probe pulses have the *opposite* circular polarization (other experimental conditions remain the same) a significant *increase* of the NMS along with a broadening of the cavity-polariton resonance occurs, as shown in Fig. 1(b). Corresponding differential spectra plotted as dotted lines in Fig. 1 further confirm the above distinct polarization dependence. The average intensity of the pump beam used in these measurements is 30 W/cm^2, corresponding to an estimated exciton density of order 10^{10}/cm^2. The intensity of the probe beam used is less than 1% of that of the pump beam.

The observation of an increase in NMS is quite surprising since excitonic many body interactions are expected to lead to bleaching of the excitonic transition and consequently a decrease in NMS. An increase in NMS on the contrary corresponds to enhancement rather than bleaching of the underlying absorption process. Note that behaviors similar to that shown in Fig. 1(b) have also been observed at other pumping intensities where the NMS increases with the pumping intensity within the intensity range used in our measurements.

The polarization dependence of the pump-probe reflection spectra shown in Fig. 1 indicates that biexcitonic effects play an essential role.[9] The hh excitonic transition in a GaAs QW consists of both σ^+ and σ^- transitions associated with the excitation of σ^+ and σ^- excitons. Attractive interactions between two excitons with opposite spins (i.e., between σ^+ and σ^- excitons) can also lead to formation of biexcitons. The binding energy of biexcitons in GaAs QWs has been shown to range between 1 and 2 meV.[9] In the absence of spin relaxation, biexcitonic interactions become effective only when both σ^+ and σ^- excitons are excited.

When the microcavity structure is resonantly excited by a σ^+-polarized pulse, NMS of cavity polaritons associated with the σ^+ transition decreases due to absorption saturation. Cavity polaritons associated with the σ^- transition, however, can behave very differently. The presence of the σ^+ excitons does not lead to band filling for the σ^- excitonic transition. Furthermore, since biexcitons can be formed from two excitons with opposite spins, the presence of the σ^+ excitons induces an additional σ^- transition that results in creation of biexcitons (see the inset in Fig. 1). In the limit that the biexciton binding energy is comparable to the exciton inhomogeneous linewidth and an appreciable number of σ^+ excitons are excited, a significant increase in NMS associated with the σ^- transition is expected since the additional exciton-to-biexciton transition effectively increases the overall oscillator strength for the σ^- transition. In principle, strong coupling between the cavity mode and the additional exciton-to-biexciton transition can also lead to the formation of new cavity-polariton modes, as we will discuss in more detail later.

In addition to the unique polarization dependence discussed above, biexcitonic contribution to coupled optical excitations in semiconductor microcavities also exhibits distinctive temporal behaviors. Figure 2 compares results of pump-probe measurements at various delays between the pump and probe pulses. NMS shown in Fig. 2(a), where both the pump and probe beams have the same circular polarization, *increases* with increasing delay and recovers gradually the splitting observed at the low excitation limit. The recovery time is expected to be determined by the exciton recombination time.[4] In comparison, when the pump-and-probe beams have the opposite circular polarization, the NMS *decreases* initially with increasing delay, as shown in Fig. 2(b). In this case, spin relaxation of excitons excited by a circularly polarized pump beam reduces biexcitonic effects and leads to increasing effects of band filling. At a delay of 20 ps, the NMS observed is nearly the same as that at the low excitation limit although significant broadening still persists [see Fig. 2(b)].

When the exciton spins are randomized or when a linearly polarized pump beam is used (not shown), the reduction in NMS due to absorption saturation exceeds the increase due to biexcitonic effects (which can also be concluded from the polarization-dependent pump-probe measurements shown in Fig. 1). The absence of an increase in NMS, however, does not imply that biexcitonic effects play no roles in these measurements. The biexcitonic effects to some extent compensate the reduction in NMS due to absorption saturation, which may also be partially responsible for the nearly constant energy position of cavity polaritons below an ionization density shown in earlier steady-state pump-probe studies of cavity polaritons.

A satisfactory model of biexcitonic effects in semiconductor microcavities would require us to extend the semiconductor Bloch equations beyond the Hartree-Fock limit or to extend the recently developed diagrammatic theory of nonlinear optical interactions in semiconductors to the nonperturbative regime of semiconductor microcavities.[10] Considerable physical insight, however, can still be gained by using phenomenological optical Bloch equations (OBE) based on few-level systems along with the Maxwell equation. Earlier use of the phenomenological OBE has led to improved physical understanding of many-body processes such as biexcitons and local field effects in coherent nonlinear optical processes in semiconductors.[9,11]

To include effects of bound biexcitonic states, we have used a four level system shown in the inset of Fig. 1.[9] At a given exciton density, the Maxwell-Bloch equation describing the coupling between a σ^+ polarized cavity mode and excitons as well as biexcitons is given by

$$\dot{\alpha}_+ = -(i\omega_c + \kappa)\alpha_+ + \Omega\sqrt{1-2N_+/N_0}\,\beta_{x+} + \Omega\sqrt{N_-/N_0}\,\beta_{b+} + \kappa\varepsilon(t), \quad (1)$$

$$\dot{\beta}_{x+} = -(i\omega_x + \gamma)\beta_{x+} - \Omega\sqrt{1-2N_+/N_0}\,\alpha_+, \quad (2)$$

$$\dot{\beta}_{b+} = -(i\omega_b + \gamma)\beta_{b+} - \Omega\sqrt{N_-/N_0}\,\alpha_+, \quad (3)$$

where α_+ is the expectation value of the annihilation field operator for the cavity mode at the position of the QW inside the cavity; β_{x+} and β_{b+} is proportional to the σ^+ polarized optical polarization associated with the excitonic transition and the exciton-to-biexciton transition, respectively; $\varepsilon(t)$ represents a normalized external driving field; κ is the cavity decay rate; ω_c, ω_x, and ω_b are the resonant frequency of the cavity, the excitonic transition, and the exciton-to-biexciton transition, respectively; Ω is the collective dipole coupling rate for the σ^+ excitonic transition at the low excitation limit; N_0 is the equivalent of the total available number of σ^+ excitons; and N_+ and N_- are the density of σ^+ and σ^- excitons, respectively. For simplicity, we have assumed the same dephasing rate (denoted by γ) and dipole moment for both excitonic and the exciton-to-biexciton transitions. Effects of EID can be included by assuming a density-dependent dephasing rate. Note that saturation of the biexcitonic transition has been ignored in the above equations. We also emphasize that the very simple model presented above is aimed to illustrate how biexcitonic effects contribute to coupled excitations in the nonperturbative regime rather than to have a direct comparison between theory and experiment.

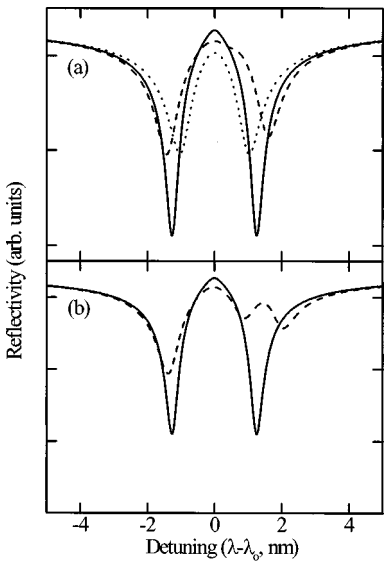

FIG. 3. Reflection spectra calculated based on the model discussed in the text. The solid lines show the reflection spectra in the low excitation limit. The dotted and dashed lines are reflection spectra at a given density of σ^+-excitons and are for σ^+ and σ^--polarized probes, respectively. The biexciton binding energy is assumed to be 1.2 meV in (a) and 3 meV in (b). λ_0 is the resonant wavelength of the cavity.

Equation (2) and the term proportional to β_{x+} in Eq. (1) describe coupling between the excitonic transition and the cavity mode and can lead to the formation of coupled exciton-photon modes. Equation (3) and the term proportional to β_{b+} in Eq. (1) describes coupling between the exciton-to-biexciton transition and the cavity mode and can lead to the formation of coupled excitations associated with the exciton-to-biexciton transition. This later coupling becomes effective only when there is a significant number of σ^- excitons present since the collective dipole coupling rate for the process is given by $\Omega\sqrt{N_-/N_0}$.

The calculated reflection spectrum at low excitation limit is shown as the solid line in Fig. 3 where we have assumed $\omega_c = \omega_x$, $\kappa = 0.75\ \mathrm{ps}^{-1}$, $\gamma = \kappa/2$, and $\Omega = 10\kappa$. Reflection spectra at a given density of σ^+ excitons are shown in Fig. 3(a) as the dotted line for a σ^+-polarized probe and the dashed line for a σ^--polarized probe where we have used an exciton dephasing rate of $1.2\ \mathrm{ps}^{-1}$ and an exciton density such that the collective dipole coupling rate is reduced to 0.8Ω for the σ^+ transition. For the σ^+-polarized probe, biexcitonic effects are absent and one expects a reduction in NMS and a broadening of the cavity-polariton resonance as shown in Fig. 3(a).

The σ^--polarized probe, however, is sensitive to the biexcitonic contribution. Along with a broadening of the cavity-polariton resonance, a significant increase in NMS is clearly shown in Fig. 3(a) where a biexciton binding energy of 1.2 meV is assumed. This increase in the NMS results directly from an increase in the overall absorption strength for the σ^- transition due to the induced exciton-to-biexcitons transition, as we have discussed earlier. The cavity-polariton resonance now involves both the excitonic and the exciton-to-biexciton transition. Note that the energy shift of the two cavity-polariton resonances is asymmetric. For the upper cavity-polariton resonance, a blueshift induced by the increase in

the oscillator strength is partially compensated by effects of a redshift in the center frequency of the overall σ^- transition.

In the limit that the biexciton binding energy is large compared with the exciton and cavity linewidth, new coupled excitations can arise from a strong coupling between the cavity mode and the exciton-to-biexciton transition. As a result, an additional cavity-polariton resonance can emerge in the reflection spectrum as shown in Fig. 3(b) where we have used a biexciton binding energy of 3 meV. It should be noted that in this case the strong coupling between the cavity mode and the exciton-to-biexciton transition affects strongly all cavity-polariton resonances.

Finally, we note that although Eqs. (1)–(3) can also be extended to include inhomogeneous broadening induced by interface disorders, as done in an earlier study,[12] a satisfactory description of the asymmetric cavity-polariton linewidth and especially the highly asymmetric reduction in NMS shown in Fig. 1(a) will require a microscopic understanding of effects of motional narrowing in disordered potentials on excitonic nonlinear optical processes.

In conclusion, using polarization-dependent pump-probe spectroscopy, we have shown important contribution of biexcitonic effects to coupled excitations in semiconductor microcavities. Biexcitonic interactions can lead to an increase in NMS and in the limit that the biexciton binding energy is large compared with relevant linewidth of the composite system can also result in an additional polariton resonance. The experimental results and the phenomenological model presented in this paper should stimulate further experimental and theoretical efforts in understanding optical interactions in the nonperturbative regime in semiconductors.

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