Normal mode oscillation in the presence of inhomogeneous broadening

Hailin Wang, Young-Tak Chough, S. E. Palmer, and H. J. Carmichael

Department of Physics and Oregon Center for Optics, University of Oregon, Eugene, OR 97403

hailin@oregon.uoregon.edu

Abstract: We investigate effects of inhomogeneous broadening of excitons on normal mode oscillation in semiconductor microcavities using a coupled oscillator model. We show that inhomogeneous broadening can drastically alter the coherent oscillatory energy exchange process even in regimes where normal mode splitting remains nearly unchanged. The depth, frequency, and phase of normal mode oscillations of excitons at a given energy within the inhomogeneous distribution depend strongly on the energy separation between the exciton and the normal mode resonance. In addition, for an inhomogeneous broadened system, pronounced oscillations in the intensity of the optical field or the total induced optical polarization no longer imply a similar oscillatory coherent energy exchange between excitons and cavity photons.

OCIS codes: (270.1670) Coherent optical effect; (320.7130) Ultrafast processes in condensed matter, including semiconductors

References and links

1. C. Weisbuch, M. Nishioka, A. Ishikawa, and Y. Arakawa, "Observation of the coupled exciton-photon mode splitting in a semiconductor quantum microcavity," *Phys. Rev. Lett.* **69**, 3314 (1992).
2. T. B. Norris, J. K. Rhee, C. Y. Sung, Y. Arakawa, M. Nishioka, C. Weisbuch, "Time-resolved vacuum Rabi oscillations in a semiconductor quantum microcavity," *Phys. Rev. B* **50**, 14663 (1994).
3. J. Jacobson, S. Pau, H. Cao, G. Bjork, Y. Yamamoto, "Observation of exciton-polariton oscillating emission in a single-quantum-well semiconductor microcavity," *Phys. Rev. A* **51** 2542 (1995).
4. Hailin Wang, Jagdeep Shah, T. C. Damen, W. Y. Jan, J. E. Cunningham, M. H. Hong, and J. P. Mannaerts, "Coherent oscillations in semiconductor microcavities," *Phys. Rev. B* **51**, 14713 (1995).
5. D. Bogavarapu, D. McAlister, A. Anderson, M. Munroe, M. G. Raymer, G. Khitrova, and H. M. Gibbs, "Ultrafast photon statistics of normal mode coupling in a semiconductor microcavity," *Quantum Electronics and Laser Science Conference*, *OSA Technical Digest* **9**, 33 (1996).
6. S. Pau, G. Bjork, H. Cao, E. Hanamura, and Y. Yamamoto, "Theory of inhomogeneous microcavity polariton splitting," *Solid State Commun*. **98**, 781 (1996).
7. F. Jahnke, M. Ruopp, M. Kira, and S. W. Koch, "Ultrafast pulse propagation and excitonic nonlinearities in semiconductor microcavities," *Adv. in Solid State Phys.* **37**, accepted for publication (1997).
8. H. J. Carmichael , "Quantum fluctuations in absorptive bistability without adiabatic elimination," *Phys. Rev. A* **33**, 3262 (1986).
9. Y. Zhu, D. J. Gauthier, S. E. Morin, Q. Wu, H. J. Carmichael, and T. W. Mossberg, “Vacuum Rabi splitting as a feature of linear dispersion theory,” *Phys. Rev. Lett.* **64**, 2499 (1990).
10. M. Kira, F. Jahnke, S. W. Koch, "Ultrashort pulse propagation effects in semiconductor microcavities," *Solid State Commun*. **102**, 703 (1997).
11. R. Zimmermann and E. Runge, "Exciton lineshape in semiconductor quantum structures with interface roughness," *J. Lumin*. **60**&**61**, 320 (1994).
12. D.M. Whittaker, P. Kinsler, T.A. Fisher, M.S. Skolnick, A. Armitage, A.M. Afshar, M.D. Sturge, and J.S. Roberts, "Motional narrowing in semiconductor microcavities," *Phys. Rev. Lett.* **77**, 4792 (1996).
13. V. Savona, C. Piermarocchi, A. Quattropani, F. Tassone, and P. Schwendimann, "Microscopic theory of motional narrowing of microcavity polaritons in a disordered potential," *Phys. Rev. Lett.* **78**, 4470 (1997).
14. Different line width for lower and upper cavity-polaritons can also be obtained by using the simple coupled-oscillator model and an asymmetric inhomogeneous lineshape.

#2660 - $10.00 US
Received September 12, 1997; Revised November 20, 1997

Strong coupling between excitons in a quantum well (QW) and a resonant or nearly resonant cavity mode can lead to the formation of a coupled exciton-cavity mode, or simply cavity-polaritons [1]. The excitation spectrum of this composite exciton-cavity system is characterized by two well-resolved cavity-polariton resonances (or normal mode resonances) if Ω>(γ, κ) where Ω is the collective dipole coupling rate between the excitonic system and the cavity mode and γ and κ are the exciton dephasing rate and the cavity field decay rate, respectively. In this limit, an impulsive excitation of the cavity mode can lead to a coherent oscillatory energy exchange (or normal mode oscillation) between excitons and cavity photons. Normal mode oscillations in semiconductor microcavities have been investigated extensively by using various transient optical techniques, including time-resolved reflection or emission, four wave mixing, and homodyne tomography [2-5]. Device application of coherent oscillation in the exciton population has also been suggested since the coherent energy exchange rate can be much faster than the exciton spontaneous emission rate.

Semiconductor heterostructures feature unavoidable interface fluctuations. For excitons confined in a QW, the energy of excitons depends strongly on the well width. At low temperature, potential fluctuations due to well-width variations can localize excitons at local energy minimum, leading to inhomogeneous broadening of the exciton absorption. Most if not all experimental studies of semiconductor microcavities at very low temperature were carried out in systems where excitons are inhomogeneously broadened. Theoretical calculations have shown that as long as the spectral linewidth of the inhomogeneous distribution is smaller than the normal mode splitting of a corresponding homogeneously broadened system, normal mode splitting remains nearly unchanged [6]. Effects of inhomogeneous broadening on bleaching of normal mode coupling have also been considered recently [7].

In this paper, we present a theoretical investigation on effects of inhomogeneous broadening on normal mode oscillation. We show that inhomogeneous broadening can drastically alter normal mode oscillation even in regimes where Γ_{inh}, HWHM of the inhomogeneous distribution, is considerably smaller than Ω and where inhomogeneous broadening results in no reduction in the normal mode splitting. Calculations based on a coupled oscillator model reveal that the frequency, phase, and depth of normal mode oscillation of localized excitons at a given energy within the inhomogeneous distribution depend strongly on the energy separation between the exciton and the normal mode resonance. Strongest normal mode oscillation occurs for excitons interacting with equal strength with optical fields at the two normal mode resonances, whereas only very weak oscillations are expected for excitons nearly resonant with a normal mode. In an inhomogeneous broadened system, pronounced oscillation in the intensity of the optical field or the total induced optical polarization no longer implies similar oscillations in the exciton population.

In the low excitation limit, a composite system of cavity and localized excitons can be described by a coupled-oscillator model that has also been used previously to describe composite atom-cavity systems [8, 9]. For an inhomogeneously broadened excitonic system, the cavity mode couples to excitons with a distribution of energies. The equation of motion for the coupled oscillator system is then given by:

$$\dot{\alpha} = -(i\omega_c + \kappa)\alpha + \Omega \int d\omega f(\omega)\beta(\omega) + \kappa\varepsilon(t) \;\; (1) \tag{1}$$

$$\dot{\beta}(\omega) = -(i\omega + \gamma)\beta(\omega) - \Omega\alpha \tag{2}$$

where α is the expectation value of the annihilation field operator for the cavity mode at the position of the QW inside the cavity, Ω is the collective dipole coupling rate determined by spectrally integrated oscillator strength, $\varepsilon(t)$ represents the external driving field, ω_c is the

#2660 - $10.00 US Received September 12, 1997; Revised November 20, 1997

resonant frequency of the cavity, and $f(\omega)$ is the normalized inhomogeneous distribution of the exciton energy. We have scaled $\beta(\omega)$ such that $P(\omega)=\mu\beta(\omega)/N^{1/2}$ represents the optical polarization for an exciton at frequency ω where N is the total number of excitons allowed within the inhomogeneous distribution and μ is the optical transition dipole moment. Note that for simplicity we have used as Eq. (1) a first order Maxwell equation that was originally derived for CW excitations. The first order Maxwell equation can also be used to describe transient processes with a time scale much longer than the cavity round trip time (which is a few fs for typical semiconductor microcavities), as shown in recent theoretical studies [10]. The population of excitons at frequency ω can be determined in the weak excitation limit from the following equation:

$$\dot{n}(\omega) = -\Gamma n(\omega) - \Omega[\alpha^* f(\omega)\beta(\omega) + c.c.] \tag{3}$$

where Γ is the exciton population decay rate. In-scattering terms due to exciton spectral diffusion are not included in Eq. (3) since we are mainly interested in excitons that are coherently excited by an external field. Note that a main limitation of the above model is that we have ignored lateral motion of excitons in the disordered potential. As a result, Eqs. (1-3) are identical to the Maxwell-Bloch equations used for describing composite atom-cavity systems in the weak excitation limit.

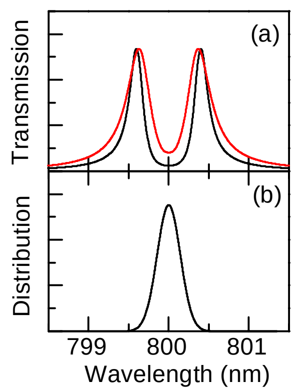

Fig. 1. (a) Transmission spectra (normalized) for composite exciton-cavity systems. All parameters are normalized to Ω (taken to be 1 ps^{-1} or 3.4 Å). For the black curve (inhomogeneously broadened), $\Gamma_{inh}=\kappa=0.5$ and $\gamma=0.05$. For the red curve (homogeneously broadened), $\gamma=\kappa=0.5$ and $\Gamma_{inh}=0.01$. (b) The inhomogeneous distribution used in calculating the black curve in (a).

Equations (1-3) are solved numerically after an impulsive excitation of the cavity mode. Figure 1a compares the transmission spectrum of a microcavity with homogeneously broadened excitons with that with inhomogeneously broadened excitons. The same cavity decay rates and collective excitonic dipole coupling rates are used for both microcavities. For simplicity, the inhomogeneous distribution is taken to be a Gaussian with its center ω_0 at ω_c. As shown in Fig. 1a, the magnitude of normal mode splitting is nearly the same for both microcavities in agreement with earlier studies [6]. The spectral linewidth for the inhomogeneously broadened system is actually smaller than that for the homogeneously broadened system. The inhomogeneous distribution used is also plotted in Fig. 1b as a reference.

Figure 2 shows the dynamical behavior of the inhomogeneously broadened system after an impulsive excitation of the cavity mode. Deep oscillations in the intensity of the

#2660 - $10.00 US
Received September 12, 1997; Revised November 20, 1997

optical field and the total induced optical polarization can be clearly observed in Fig. 2a. These oscillations are also expected from the well-resolved normal mode splitting shown in the spectral domain response in Fig. 1.

The inhomogeneous broadening, however, significantly weakens the coherent energy exchange process between the exciton and cavity as evidenced in Fig. 2b. The oscillation in the total exciton population is much weaker compared with that in the optical field or the polarization. In this case, deep oscillations in the intensity of the optical field or total optical polarization do not imply a similar coherent energy exchange process between excitons and cavity photons even though the inhomogeneous broadening results in no reduction in the normal mode splitting.

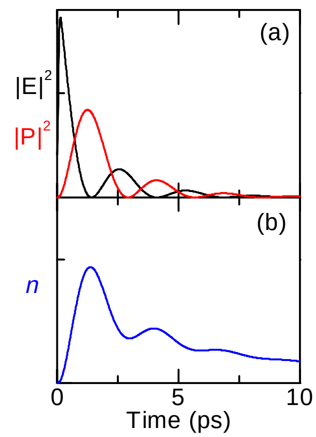

Fig. 2. (a) Normal mode oscillation for an inhomogeneously broadened system in the intensity of the field (black curve) and polarization (red curve). (b) Normal mode oscillation in the total exciton population. $\Gamma=2\gamma$ and other parameters used are the same as in Fig. 1.

To understand the much weaker normal mode oscillation in the total exciton population, we plot in Fig. 3 normal mode oscillations in the induced optical polarization and exciton population for individual excitons. As shown in Fig. 3, for an exciton at a given energy within the inhomogeneous distribution, the normal mode oscillation in the exciton population essentially follows that in the corresponding optical polarization. Detailed behaviors of the oscillation, including depth, frequency, and phase of the oscillation, however, vary greatly as we vary the energy of excitons within the inhomogeneous distribution. The oscillation is strongest for excitons at the center of the distribution and becomes much weaker for excitons away from the center. In fact for excitons at or near the frequency of a given normal mode, only very weak oscillations are observed. In addition, the frequency of the normal mode oscillation for excitons at the center of the inhomogeneous distribution is actually only half that of the normal mode oscillation in the intensity of the optical field or the total optical polarization.

In a microcavity with an inhomogeneously broadened excitonic system, the cavity mode couples to excitons with many different resonant frequencies. The microcavity should thus be considered as a coupled many-oscillator instead of coupled two-oscillator system. Normal mode splitting observed in the transmission or reflection spectrum can still be considered as resulting from the interaction between the cavity mode and a "super"-oscillator that represents the whole excitonic system. The actual coherent energy exchange process, however, is more complicated.

For an impulsive excitation and after a few cavity round trips, the power spectrum of photons inside the cavity should be characterized by two normal mode resonances in the

#2660 - $10.00 US Received September 12, 1997; Revised November 20, 1997

transmission spectrum (such as that shown in Fig. 1a). For a given exciton within the inhomogeneous distribution, details of the optical interaction depend strongly on the detuning between the exciton and the two normal mode resonances. Excitons at or near ω_0 (assuming ω_0=ω_c) will interact with equal strength with optical fields at both normal mode resonances, leading to a strong oscillation in the excitonic polarization as well as the exciton population. In comparison, excitons near a given normal mode resonance will couple much more strongly to optical fields at the given normal mode resonance than to optical fields at the other normal mode resonance. Only very weak oscillations are expected for these excitons. The oscillation in the total exciton population is thus much weaker than that of the corresponding homogeneously broadened systems. Oscillation in the total optical polarization, however, remains pronounced because of interference between polarizations arising from excitons at different energies.

It should be noted that lateral motion of localized excitons ignored in the above coupled-oscillator model can lead to motional narrowing effects such as asymmetric inhomogeneous lineshapes [11]. These effects can result in different linewidth for upper and lower cavity-polaritons as shown in recent studies, but are not expected to affect the qualitative conclusions drawn above [12-14].

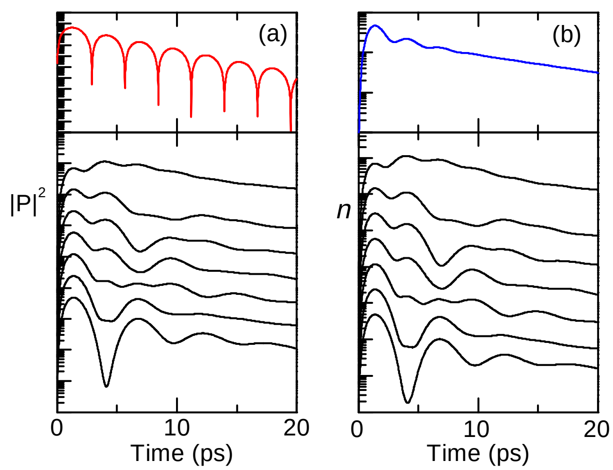

Fig. 3. Normal mode oscillation in the intensity of the polarization (a) and in the exciton population (b). The top figures show the oscillation in the total polarization and population shown in Fig. 2. The bottom figures are for an exciton within the inhomogeneous distribution. The wavelength used (from bottom to top) is 800, 800.03, 800.07, 800.1, 800.14, 800.17, and 800.27 nm, respectively. Successive curves are displaced by half a decade in the log plot.

Inhomogeneous broadening also affects the phase difference between intensities of the field and polarization in the normal mode oscillation. From the node position of the intensity of the total polarization shown in Fig. 3a, we determine the phase difference to be 0.88π, deviating significantly from the π phase difference expected for a homogeneously broadened system and also reflecting an incomplete energy exchange between the field and the polarization.

Effects of inhomogeneous broadening are expected to diminish when the inhomogeneous linewidth becomes small compared with the normal mode splitting. Fig. 4 shows normal mode oscillation in the total exciton population for microcavities with various inhomogeneous linewidth. As expected, the oscillation in the total exciton population becomes stronger with decreasing inhomogeneous linewidth.

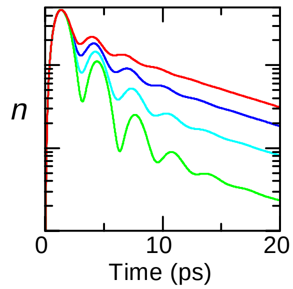

Fig. 4. Normal mode oscillation in the total exciton population in microcavities with κ=0.5 and γ =0.05 and with Γ_{inh}=0.2, 0.3, 0.4, 0.5 (from green to red).

In conclusion, using a coupled oscillator model we have investigated theoretically effects of inhomogeneous broadening on normal mode oscillations. We show that inhomogeneous broadening can drastically alter coherent energy oscillations even in regimes where the inhomogeneous linewidth is still considerably smaller than the normal mode splitting and where there is no apparent reduction in the normal mode splitting. For excitons at a given energy within the inhomogeneous distribution, the depth, frequency, and phase of normal mode oscillations in the induced optical polarization and the exciton population depend strongly on the detuning between the exciton and the normal mode resonances. While it is difficult to probe experimentally population dynamics of excitons at a given energy in a microcavity, theoretical results discussed above clearly indicate that in an inhomogeneously broadened system, pronounced oscillation in the intensity of the optical field or total induced optical polarization no longer implies a similar oscillatory coherent energy exchange between excitons and cavity photons.

H. Wang wishes to acknowledge many helpful and stimulating discussions with Jagdeep Shah and Erich Ippen. This work is supported in part by AFOSR and NSF.