Observation of exciton-polariton oscillating emission in a single-quantum-well semiconductor microcavity

Joseph Jacobson,* Stanley Pau, Hui Cao, Gunnar Björk, and Yoshihisa Yamamoto†
ERATO Quantum Fluctuation Project, Edward L. Ginzton Laboratory, Stanford University, Stanford, California 94305
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We have observed direct time domain exciton-polariton oscillating emission from a single-quantum-well GaAs microcavity. Observed oscillation periods of 21.7 ps were in good agreement with the spectral splitting of 0.12 nm. Such observations are consistent with the view that recently observed thresholdless coherent emission is not resonant Rayleigh scattering but rather exciton-polariton emission.

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Over the past several years there has been great interest in atom-cavity coupled systems [1]. Recently these experiments have been extended to semiconductor systems in the form of semiconductor quantum-well microcavities [2]. Weisbuch *et al.* have observed a key signature of "atom-cavity" strong coupling, namely, normal mode splitting, in multiple-quantum-well systems. Such observations were made through modified cavity resonance [3]. This experimental technique may be termed "cavity spectroscopy" in which the reflectance or transmittance of a probe beam is measured and for which the natural physical interpretation is given in terms of a modified cavity resonance due to atomic dispersion [4]. We note that in the experiments to date the phase between the exciton and the photon modes has not been measured.

In this work we have observed direct time domain exciton-polariton oscillating emission in a single-quantum-well microcavity. In a quantum-well microcavity, the exciton-polariton is the normal mode of the system. By employing a zero of time marker we have made a measurement of the phase between the exciton and the photon modes which constitute the polariton. We employ a complimentary experimental technique which may be called "emission spectroscopy," in which an initial ensemble of atoms is prepared in a well defined quantum state and the emission from the coupled atom-cavity system is observed directly. The natural physical interpretation is given in terms of a modified atom resonance due to its strong coupling to a cavity vacuum field (dressed atom picture) [5]. In addition to representing a direct measurement of the cavity emission this technique has the advantage that a zero of time marker pulse reflected from the surface of the microcavity carries a temporal width tag making it clearly differentiable from the cavity emission.

Finally, we confirm that we are in the weak excitation regime by observing that the turn-on delay time between creation of the exciton ensemble and the emission peak of photons (corresponding to one-half the oscillation period) is invariant over a three order of magnitude variation in the pump intensity.

Figure 1(a) shows a typical exciton-polariton oscillating emission in the case of resonant excitation. The observed dynamics are independent of the pump intensity in the regime of weak excitation. Oscillations occur due to strong coupling which comes about by means of an enhanced dipole moment due to the spatial coherence of the resonantly excited excitons [6].

In contrast, Fig. 1(b) shows the cavity emission when excited nonresonantly with higher pump intensities. In this process the quantum well is excited above the band gap creating hot carriers which are thermalized by phonon emission to form $k \simeq 0$ electron-hole pairs. The delay between the pump pulse (narrow peak) and the emission is determined by the thermalization time and the turn-on delay time of the laser. As may be seen, the decay time of the emission is now intensity dependent, as expected in normal lasing governed by stimulated emission processes. The system now also has a considerably longer decay time (weakly coupled regime) due to the lack of spatial exciton coherence.

Just prior to the present experiments, thresholdless coherent emission was reported in an identical structure to the one used here [7]. Such thresholdless behavior requires coherent emission times which are faster than the relevant exciton dephasing times independent of the pump intensity. As discussed above, the generation of spatial exciton coherence as a result of resonant pumping leads to just such a fast emission process. Further, a measurement of the time delay between state preparation and emission (e.g., a measurement of the phase between the exciton and photon modes; see peaks *a* and *b* in Fig. 2 and text below) indicates a delay of approximately one-half a Rabi oscillation period. This delay distinguishes the present process from resonant Rayleigh scattering and makes such thresholdless coherent emission observations consistent with the picture that they are not resonant Rayleigh scattering (as has been contended) but rather exciton-polariton emission.

The GaAs quantum-well microcavity system may be modeled by coupled harmonic oscillators damped by their respective reservoirs. The microscopic Hamiltonian of a Wannier exciton coupled to the radiation field is given in Ref. [8]. Neglecting two-photon processes and adding

*FAX: (415) 723-5320.
Electronic address: jacobson@sierra.stanford.edu.
†Also at NTT Basic Research Laboratories, Atsugishi, Kanagawa, Japan.

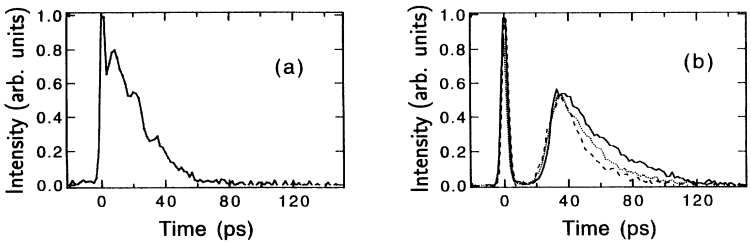

FIG. 1. (a) Typical exciton-polariton oscillating emission as a function of time from the resonantly excited quantum-well microcavity. The first peak corresponds to the reflected pump. (b) Emission from the microcavity when excited above the band for three different pump intensities: 50 mW (solid line), 100 mW (dotted line), 330 mW (dashed line). The first peak corresponds to the reflected pump.

reservoir damping yields the total system Hamiltonian

$$\hat{H} = \hbar\omega_{\rm ex}(\hat{B}^\dagger\hat{B} + \tfrac{1}{2}) + \hbar\omega_p(\hat{a}^\dagger\hat{a} + \tfrac{1}{2}) + \hbar\Omega(\hat{a}^\dagger\hat{B} + \hat{a}\hat{B}^\dagger) + \sum_k \hbar\omega_{{\rm ex},k}(\hat{B}_k^\dagger\hat{B}_k + \tfrac{1}{2}) + \sum_{k'} \hbar\omega_{p,k'}(\hat{a}_{k'}^\dagger\hat{a}_{k'} + \tfrac{1}{2}) + \sum_k \hbar g_k(\hat{B}_k^\dagger\hat{B} + \hat{B}^\dagger\hat{B}_k) + \sum_{k'} \hbar l_{k'}(\hat{a}_{k'}^\dagger\hat{a} + \hat{a}^\dagger\hat{a}_{k'}) \quad , \tag{1}$$

where $\omega_{\rm ex}$ and ω_p are the angular frequencies of the exciton and photon modes, respectively. We have used the rotating-wave approximation. The last four terms in the Hamiltonian correspond to the coupling and the self-energy of the reservoirs. At low densities the GaAs Wannier exciton obeys the boson commutation relations. Introducing the Markov approximation for the damping terms we may derive the Heisenberg equations of motion for the photon and exciton field operators $\hat{a}$ and $\hat{B}$ respectively. Solving the equations with the initial conditions $\langle\hat{B}\rangle = B_0$ and $\langle\hat{a}\rangle = 0$ yields the intensity of the photon field

$$I_p(t) = \langle\hat{a}^\dagger(t)\hat{a}(t)\rangle = \frac{B_0^2}{\Delta\Delta^*}\Omega^2 4\exp[-(\gamma_{\rm ex}+\gamma_p)t/2]\sin\left(\frac{\Delta t}{2}\right)\sin\left(\frac{\Delta^* t}{2}\right) \quad , \tag{2}$$

where $\Delta = \omega_+ - \omega_-$ is the difference between the two normal mode frequencies and

$$\omega_\pm = \frac{-[\omega_{\rm ex}+\omega_p - i(\gamma_p+\gamma_{\rm ex})/2] \pm \sqrt{[\omega_p - \omega_{\rm ex} - i(\gamma_p-\gamma_{\rm ex})/2]^2 + 4\Omega^2}}{2} \tag{3}$$

As mentioned previously the coupling factor Ω includes a cooperative enhancement similar to superradiance [6] due to the spatial coherence of resonantly excited excitons. Inhomogeneous broadening is incorporated by a suitable averaging of Eq. (2) with respect to $\omega_{\rm ex}$ using a Gaussian distribution of width $\Delta\lambda$ centered at the photon wavelength $\lambda_p = 2\pi c_0/\omega_p$.

The sample is that of Ref. [9]. It consists of a single 200-Å GaAs quantum well embedded in a distributed Bragg reflector λ microcavity. The device was cooled to 4 K and excited on resonance (813 nm) with a 200-fs mode-locked Ti:sapphire laser at an angle of 4° from the normal. The zero of time was set by means of the scattered reflection of the pump pulse from the front surface of the microcavity.

Figure 2 shows the emission output into the normal direction as a function of time as seen on a streak camera. The pump power was 380 μW. Peak a is the reflected pump pulse giving the resolution of the instrument which is 1.6 ps and acting as the zero of time marker corresponding to the initial creation of excitons. The turn-on delay time for the first photon emission, peak b, corresponds to one-half an exciton-polariton oscillation period and was measured to be 8.3 ps. The next photon emission, peak c, occurred 21.7 ps later. The fact that the oscillation period is greater than a simple factor of 2 times the turn-on delay is due to the finite decay of the coherent exciton dipole and photon field and to inhomogeneous broadening in the sample. The solid line is the theoretical curve of photon emission using $\Omega = 0.13$ ps^{-1} (0.086 meV),

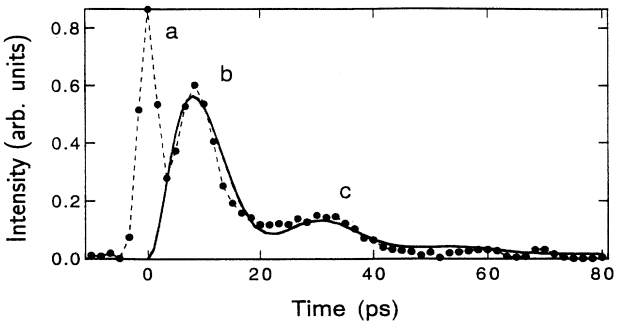

FIG. 2. Exciton-polariton microcavity emission as a function of time. The dots are data connected by a dashed line to guide the eye. Peak a is the reflected pump which sets the zero of time (time of exciton ensemble creation). Peaks b and c are the peaks of the photon emission. The solid line is the theoretical curve of photon emission using $\Omega = 0.13$ ps^{-1}, $\gamma_{\rm ex} = 0.02$ ps^{-1}, and $\gamma_p = 0.10$ ps^{-1} and the inhomogeneous line width is $\Delta\lambda = 2$ nm.

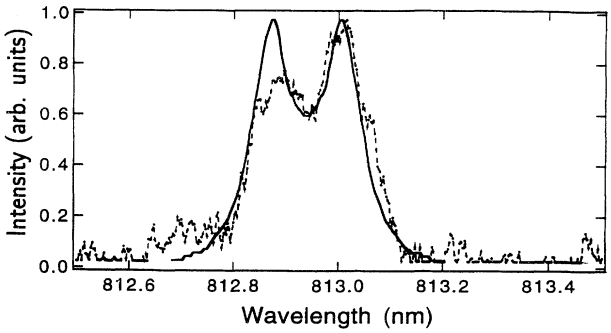

FIG. 3. Exciton-polariton microcavity emission as a function of wavelength. The broken line is experimental data and the solid line is the Fourier transform of the theoretical curve shown in Fig. 2.

$\gamma_{ex} = 0.02$ ps^{-1} (0.013 meV), $\gamma_p = 0.10$ ps^{-1} (0.066 meV) [10], and an inhomogeneous linewidth of $\Delta\lambda = 2$ nm. As is evident, the theoretical fit is quite good. The fit yields an oscillator strength per unit area per quantum well of $f = 2.1 \times 10^{-5}$ Å^{-2}. This is about a factor of 20 smaller than that reported in Ref. [3]. The discrepancy is partly attributable to the difference in quantum-well thickness in the two cases and partly due to sample growth variability.

Figure 3 shows the corresponding simultaneously measured emission spectrum. The observed splitting is 0.12 nm, in close agreement with the temporal data. The solid line is the Fourier transform of the theoretical curve of Fig. 2.

Figures 4(a) and 4(b) show the turn-on delay (exciton-polariton oscillation half period) and the normal mode splitting as a function of pump intensity. The solid line corresponds to the theoretical delay for $\Omega = 0.1$ ps^{-1} [Fig. 4(a)] and to a splitting of 0.12 nm in [Fig. 4(b)]. The turn-on delay and the normal mode splitting are nearly constant over a three order of magnitude variation in the pump intensity. At the highest pump intensities the Wannier exciton is expected to deviate from a purely bosonic particle (in its internal degree of freedom) due to Coulomb-Coulomb interaction. The decrease in turn-on delay time and the increase in normal mode splitting at higher pump intensities may be due to this effect. Note that the data in Fig. 4(a) were taken at a slightly different position on the microcavity from that of Figs. 2 and 3, giving the slight difference in turn-on delay time as compared to those figures.

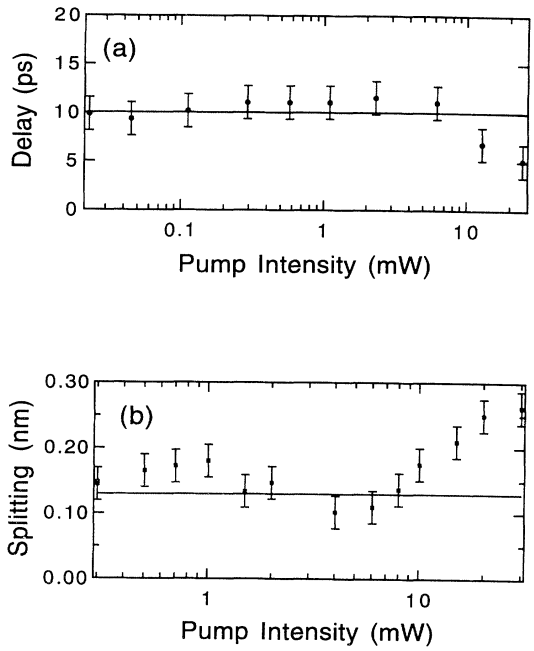

FIG. 4. (a) Turn-on delay (exciton-polariton oscillation half period) and (b) normal mode splitting as a function of pump power.

In conclusion we have directly observed exciton-polariton oscillating emission in a single-quantum-well system. These findings support the view that recently observed thresholdless coherent emission [7] is not resonant Rayleigh scattering but rather exciton-polariton emission. We expect that these experiments will find an interesting application to novel coherent light sources.

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[1] For a recent review on atom-cavity quantum electrodynamics see S. Haroche and D. Kleppner, Phys. Today **42** (1), 24 (1989).

[2] For a recent review on semiconductor cavity quantum electrodynamics see Y. Yamamoto and R. Slusher, Phys. Today **46** (6), 66 (1993).

[3] C. Weisbuch M. Nishioka, A. Ishikawa, and Y. Arakawa, Phys. Rev. Lett. **69**, 3314 (1992); T. B. Norris, J.-K. Rhee, C. Y. Sung, Y. Arakawa, M. Nishioka, and C. Weisbuch, in *QELS Conference Proceedings, Baltimore, 1993* (Optical Society of America, Washington, DC, 1993).

[4] Y. Zhu, D. J. Gauthier, S. E. Morin, Q. Wu, H. J. Carmichael, and T. W. Mossberg, Phys. Rev. Lett. **64**, 2499 (1990); A. Tredicucci, Y. Chen, F. Bassani, J. Masseis, G. Neu, and C. Deparis, Phys. Rev. B **47**, 10 438 (1993); Y. Chen, A. Tredicucci, and F. Bassani, J. Phys. IV **3**, 453 (1993).

[5] C. Cohen-Tannoudji, J. Dupont-Roc, and G. Grynberg, *Atom-Photon Interactions* (Wiley, New York, 1993).

[6] E. Hanamura, Phys. Rev. B **38**, 1228 (1988).

[7] Y. Yamamoto, F. Matinaga, S. Machida, A. Karlsson, J. Jacobson, G. Björk, and T. Mukai, J. Phys. II **3**, 39 (1993).

[8] F. Bassani, F. Ruggiero, and A. Quattropani, Nuovo Cimento D **7**, 700 (1986).

[9] G. Björk, H. Heitmann, and Y. Yamamoto, Phys. Rev. A **47**, 4451 (1993).

[10] H. Heitmann, Y. Kadota, T. Kawakami, and Y. Yamamoto, Jpn. J. Appl. Phys. **32**, L1141 (1993).