Coherent oscillations in semiconductor microcavities

Hailin Wang, Jagdeep Shah, T. C. Damen, W. Y.Jan, and J. E. Cunningham AT&T Bell Laboratories, Holmdel, New Jersey 07733

> M. Hong and J. P. Mannaerts AT&T Bell Laboratories, Murray Hill, New Jersey 07974 (Received 3 November 1994)

We present a coherent nonlinear optical study of composite exciton-cavity systems in the strongcoupling regime. Quantum oscillations in a transient four-wave-mixing response show the creation of a coherent superposition state between the two normal modes of the composite system and demonstrate in the time domain the coherent energy exchange between the exciton and cavity. The deep oscillation persists when the cavity resonance is far detuned from the exciton resonance. The result shows that dynamics of the nonlinear response can be in a nonperturbative regime while dynamics of the linear response remains in a perturbative regime.

The composite system of two-level atoms coupling to a single mode of a cavity has been used to investigate dynamics of open systems, where the interplay between coherent evolution and dissipation is essential and to explore quantum behaviors that have no classical counterpart.¹ In general, there are two distinct dynamical regimes for the atom-cavity interaction. In the weakcoupling (perturbative) regime, eFects of the coherent interaction between the atom and cavity are negligible and atomic spontaneous emission may be enhanced or suppressed.² In the strong-coupling (nonperturbative) regime, the coherent atom-cavity coupling becomes dominant compared with dissipative decay processes. A normal mode splitting in the linear response of the composite system can be observed.³⁻⁵ Manifestly quantum dynam ics is also expected in this regime.³ For example, excitation of higher lying states of the Jaynes-Cummings ladder can result in quantum revival and collapse of Rabi nutations, which has been observed in the microwave regime.

With the development of epitaxial growth technologies, it has become possible to investigate composite exciton-cavity systems by placing quantum wells (QW's) in a monolithic cavity consisting of two distributed Bragg refiectors (DBR). Modification of spontaneous emission of excitons in a microcavity has been reported.⁷ Normal mode splittings have also been observed in the linear response of the coupled exciton-cavity system. $8-10$ Semiconductor microcavities provide an alternative, but qualitatively different system from the atom-cavity system. Momentum conservation leads differences between emissions from an atom and an exciton. The nonlinear behavior of an exciton also differs
from that of an atom.¹¹ For independent two-level from that of an atom.¹¹ For independent two-leve atoms, the nonlinearity comes from saturation of the atomic transition, while excitonic nonlinearity can also result from exciton-exciton interactions. The nonlinear behavior of the strongly coupled exciton-cavity system can open up a new and versatile way of exploring quantum dynamics in meoscopic systems.

In this paper, we report on the coherent nonlinear optical studies of a composite exciton-cavity system in the strong-coupling regime. Coherent spectroscopy such as transient four-wave mixing (FWM) provides an ideal tool for probing directly the coherent dynamics of the composite system and can access processes that are inaccessible to linear measurements. By observing quantum oscillators in the spectrally resolved transient FWM response, we show the excitation of a coherent superposition state between the two normal modes of the composite system and demonstrate in time domain the coherent energy exchange between the exciton and cavity. Deep quantum oscillations with a period that varies with the normal mode splitting persist even when detuning between the cavity and exciton is much greater than the excitoncavity coupling rate. The result indicates surprisingly that the dynamics of the nonlinear response is in a nonperturbative regime even though dynamics of the linear response remain in a perturbative regime, leading to a unique situation where the linear and nonlinear responses remain in distinct dynamical regimes.

The QW microcavity is grown by molecular-beam epitaxy and consists of 15 and 20.5 stacks of $\lambda/4$ $\text{Al}_{0.11}\text{Ga}_{0.89}\text{As}/\text{AlAs}$ as the top and bottom DBR, respectively. Two 150-A GaAs QW's separated by a 100-A $\text{Al}_{0.3}\text{Ga}_{0.7}\text{As barrier are placed at the center (antinode) of }$ a λ cavity (approximately 2500 Å, corresponding to a round trip time of 5 fs), with a $Al_{0.3}Ga_{0.7}As$ spacer layer between the QW and DBR. Measuring cavity resonance far below the heavy-hole (hh) exciton gives a cold cavity linewidth (full width at half maximum) less than ¹ meV, reflecting the excellent quality of the cavity. The small variation in the thickness of the spacer layer (the sample is not rotated during the growth of spacer layers) allows us to tune the cavity resonance by moving the laser spot across the wafer. The exciton energy, however, remains nearly unchanged across the wafer. The linewidth for the hh exciton is estimated to be 2.4 meV from the reflectivity. The excitons are inhomogeneously broadened, since the growth of DBR degrades the inter-

face of the QW.

We carried out self-diffracted FWM in the reflection geometry. 100-fs pulses from a mode-locked Ti:Sapphire laser are centered near the hh exciton resonance. In this configuration, two incident laser pulses with wave vectors k_1 and k_2 interact in the sample (near normal incidence), producing a FWM signal in a direction near $2k'_2 - k'_1$, where \mathbf{k}'_1 and \mathbf{k}'_2 are wave vectors along the reflected directions of k_1 and k_2 . Because of multiple reflections inside the cavity, conceptually there is no difference between FWM signals from the refiection or the transmission geometry. The effective pulse duration inside the cavity is also limited by the cavity linewidth. All the measurements were performed at 10 K.

We first compare the linear and nonlinear spectral responses of the composite system. Figure ¹ shows spectra of the reflected laser pulse and the FWM emission at zero delay when the cavity is nearly resonant with the hh exciton. The incident photon flux for the FWM measurements is $10^{11}/\text{pulse}/\text{cm}^2$, assuming a laser spot of 0.1 mm². A normal-mode splitting is observed in the coherent FWM emission spectrum. A similar splitting is also observed in the reflected laser spectrum as shown in Fig. ¹ (obtained at a much lower intensity). With increasing intensities, the splitting in FWM spectra disappears rapidly. The same is observed in the corresponding reflectivity spectrum. At very high intensities, the linewidth of the reflectivity resonance approaches that of a cold cavity (not shown). These behaviors agree with the semiclassical treatment of normal-mode splittings, including saturation of the optical transition.¹² Physically bleaching of the excitonic transition significantly decreases the effective coupling strength between the exciton and cavity.

Figure 2 shows the dependence of FWM spectra on detuning between the cavity and exciton. The two incident pulses are cross-linearly polarized. Similar results are also obtained with the colinearly polarized geometry. To avoid the complication, due to light-hole (lh) excitons, $l³$ we keep the cavity resonance below the hh exciton energy. Again, two resonances are observed, corresponding to the two resonances in the reflectivity spectrum. The resonance at higher energy is excitonlike, while the reso-

FIG. 1. Spectra of the reflected laser pulse (obtained at much lower intensity) and FWM emission at zero delay.

FIG. 2. The FWM spectra at zero delay with various exciton-cavity detunings. The inset shows the position of the normal modes in the reflectivity spectra as a function of the detuning.

nance at lower energy is cavitylike. For large detunings, the FWM spectrum is completely dominated by the cavitylike resonance.

The inset of Fig. 2 shows the position of the two resonances in reflectivity spectra vs the cavity-exciton detuning. The detuning is derived from $\delta = \omega_c - \omega_x = \Omega_+ + \Omega_- - 2\omega_x$, where Ω_+ and Ω_- are positions of the two resonances in the reflectivity spectrum and ω_c and ω_x are the cavity and exciton frequency in the absence of the coupling, respectively. Solid lines in the inset of Fig. 2
are fits to $\Omega_{\pm} = \frac{1}{2}(\omega_x + \omega_c \pm \sqrt{\delta^2 + 4g^2})$, where g, the effective coupling rate between the cavity and exciton is 2. ¹ meV. The fit near zero detuning is surprisingly good given that we have ignored both damping and inhomogeneous broadening. Note that for $\delta > 0$ the coupling to lh excitons also affects the mode splitting show in the inset.

We can now discuss the transient nonlinear response of the composite system. Figure 3 shows decay of the FWM signal as a function of the delay between the two incident pulses. In this measurement, FWM signals are first sent through a spectrometer with a resolution better than 0.5 meV. The data in Fig. 3 are obtained at the energy of the corresponding cavitylike resonance in Fig. 2. Periodic oscillations are observed in all the temporal responses shown in Fig. 3. Similar oscillations with an identical phase are also present when we measure the FWM signal

Delay (ps)

FIG. 3. The FWM signal as a function of the delay between the two incident pulses. The data are obtained at the peak of the corresponding cavitylike resonance in Fig. 2. The period of the oscillation τ is also indicated in the figure.

at the energy of the excitonlike resonance. The period of the oscillation follows the energy separation between the excitonlike and the cavitylike resonances. Very deep oscillations persist when the corresponding FWM spectra in Fig. 2 show a negligible response at the excitonlike resonance and when we further increase the detuning (not shown). The persistence of deep oscillations for $|\delta| \gg g$ is very surprising, since in this case, the coupling between the cavity and exciton is very weak and effects of the coherent coupling are negligible for linear measurements. We also note that in the range of photon flux $(10^{10} - 10^{12}/\text{pulse/cm}^2)$ we have used, both the decay rate and the oscillation period are intensity independent, in contrast to measurements near zero detuning.

There are two physically distinct processes that can lead to oscillations in the transient FWM response.¹⁴ The first process results from interference of two fields with different frequencies in the detector (polarization beats). The second process results from two optical transitions sharing a common ground state (quantum beats). In this case, the interference takes place in the material instead of in the detector and is a direct consequence of the excitation of a coherent superposition state. Since we detect only one frequency component of the signal through spectral filtering, polarization beats do not contribute to the oscillations show in Fig. 3. The oscillations in Fig. 3 are quantum beats and are due to excitations of a coherent superposition state of the two normal modes.

The presence of quantum beats directly reflects the coherent energy exchange between the cavity and exciton. Using the two nearly degenerate eigenstates of the uncoupled exciton-cavity system $|g, 1\rangle$ and $|x, 0\rangle$, the wave function of the coherent superposition state can be written as

$$
|\psi\rangle = a(e^{-i\Omega_{-}t} + \alpha e^{-i\Omega_{+}t})|g,1\rangle
$$

+ $b(e^{-i\Omega_{-}t} + \beta e^{-i\Omega_{+}t})|x,0\rangle$, (1)

where a, b, α , β are constants. The wave function corresponds directly to a coherent oscillation of both the exciton and photon population in the cavity. We note that oscillations reported in linear measurements, such as transient reflectivity, are simply due to interference of fields with different frequencies in the detector and do not signal a coherent oscillation of the exciton population.⁹ In particular, in contrast to the transient FWM measurement, oscillations in linear measurements vanish when $|\delta| \gg g$.

When the cavity is far detuned from the exciton resonance, only a very small amount of the exciton wave function is mixed into the cavitylike mode and the coupled system is in an adiabatic regime. Dynamics of linear measurements in this limit is dominated by dissipative decay processes and is in a perturbative regime. In contrast, dynamics of the FWM response is still dominated by the coherent exciton-cavity coupling as shown in Fig. 3, because the nonlinear response results only from the excitonic part of the wave function. The extreme sensitivity of the FWM response to the coherent excitoncavity coupling leads to a unique situation where the linear and nonlinear responses remain in qualitatively different dynamical regimes.

To describe qualitatively the nonlinear response of the composite system we have, as a first attempt, adopted the semiclassical Maxwell-Bloch equations and have treated excitons phenomenologically as a two-level system. We treat the composite exciton-cavity system as a "molecule" and solve the interaction with the applied fields perturbatively. Effects of saturation of the excitonic transition on the coherent coupling rate g are ignored by assuming a weak excitation of the excitonic system. The dipole moments between the ground state and the two normal modes are denoted by μ_+ . We emphasize that only the excitonic part of the normal-mode wave function
contributes to the dipole moment and the contributes to the dipole moment and the nonlinear response of the composite system. Under an impulse excitation, the effective applied fields inside the cavity after multiple reflections may be described by $\epsilon_{+}f_{+}(t)exp(-i\Omega_{+}t)+\epsilon_{-}f_{-}(t)exp(-i\Omega_{-}t)$, where $f_+(t)$ is the normalized pulse shape in the cavity. The relative amplitude of ε_+ and ε_- depends on the amplitudes of the two resonance in reflectivity spectra.

To obtain analytical solutions for the Bloch equations, we take the pulse width in the cavity to be much shorter than the relevant decay time. The third-order nonlinear polarization at Ω_{-} inside the cavity is then given by

$$
P^{(3)}(\Omega_{-}) \propto \mu_{+}^{2} \mu_{-}^{2} \theta(\tau) \varepsilon_{-}^{3} e^{i(2k_{2} - k_{1}) \cdot \tau - i\Omega_{-} t - \gamma_{-} t}
$$

$$
\times (\eta^{2} e^{-\gamma_{-} \tau} + \kappa^{-2} e^{-i\Delta \tau} e^{-\gamma_{+} \tau}) , \qquad (2)
$$

where $\eta = \mu_-/\mu_+$, $\kappa = \varepsilon_-/\varepsilon_+$, $\Delta = \Omega_+ - \Omega_-$, $\tau = t_2 - t_1$, and γ_{\pm} is the dephasing rate for the two normal modes. Similar results are also obtained for the nonlinear polarization at Ω_{+} . Physically, the dipole moment of the coherent superposition state of the two normal modes oscillates with a frequency determined by the mode splitting, leading to oscillations in the FWM signal. The depth of the oscillation is determined by the relative amount of excitons excited in the two modes. We note that although only a small amount of the exciton wave function is mixed into the cavitylike mode when $|\delta| \gg g$ as discussed earlier, nearly the same amount of excitons are excited in both the cavitylike and excitonlike modes $[\eta_{\kappa} \approx 1 \text{ in Eq. (2) or } \beta \approx 1 \text{ in Eq. (1)}]$, since the cavitylike mode is excited much more strongly than the excitonlike mode, which explains the depth of the quantum beats shown in Fig. 3. The FWM signals at Ω_+ are very weak, due to spectral filtering effects of the cavity.

We now briefly discuss decay of the cavitylike mode. Figure 3 shows that the decay time of the FWM signal for the cavitylike mode decreases from 1.6 to 0.5 ps with decreasing detuning, which is unexpected since the intrinsic dephasing rate of the localized exciton is longer than 10 ps, 15 much longer than the cavity lifetime. In a simple picture, decay of the normal mode is determined by the average of the decay rates of the exciton and photon when $\delta = 0$ and approaches that of the uncoupled mode when $|\delta| \gg g$. The inhomogeneous broadening of the exciton, however, can considerably complicate the problem near $\delta = 0$. For example, in the strong coupling limit, the exciton-photon interaction can mix nearly degenerate localized exciton states and as a result prevents rephasing of the FWM signal. A detailed analysis is

- ¹For a recent review, see, Cavity Quantum Electrodynamics, edited by P. R. Berman (Academic, Boston, 1994).
- ²For a review, see S. Haroche, in Fundamental Systems of Quantum Optics, edited by J. Dalibard, J. M. Raymond, and J. Zinn-Justin (Elsevier, Amsterdam, 1992).
- 3 For a recent review, see, H. J. Kimble, in Cavity Quantum Electrodynamics, edited by P. R. Herman (Academic, Boston, 1994).
- 4J. J. Sanchez-Mondragon, N. B. Narazhny, and J. H. Eberly, Phys. Rev. Lett. 51, 550 (1983); G. S. Agarawal, ibid. 53, 1732 $(1984).$
- 5 H. J. Carmichael et al., Phys. Rev. A 40, 5516 (1989); Y. Zhu et al., Phys. Rev. Lett. 64, 2499 (1990).
- ⁶G. Rempe, H. Walter, and N. Klein, Phys. Rev. Lett. 58, 353 $(1987).$
- 7Y. Yamamoto, S. Machidwa, and G. Bjork, in Coherence, Amplification and Quantum Effects in Semiconductor Lasers, edited by Y. Yamamoto (Wiley, New York, 1991).

beyond the scope of this paper.

It should be noted that as pointed out previously, normal-mode splittings in an exciton-cavity system are similar to the exciton-polariton splitting, but with different boundary conditions.⁸ The nonlinear studies discussed in this paper reveal very different behaviors from resonant nonlinear studies of exciton polaritons in $Cu₂O$, where propagation effects also lead to quantum beats with a time-dependent period.¹⁶ We also point out that although the simple theory presented here describes qualitatively the main features of current nonlinear measurements, full quantum treatment is essential for further studies of manifestly quantum dynamics. In addition, effects of exciton-exciton interactions on the coupled modes also remain to be explored.

In conclusion, nonlinear optical measurements directly probe the coherent dynamics of the composite excitoncavity system in the strong-coupling regime. The quantum oscillation observed in the spectrally resolved transient FWM response shows the creation of a coherent superposition state between the two normal modes and demonstrates in time domain the coherent energy exchange between the exciton and photon in the composite system. The persistence of the quantum beats when $|\delta| \gg g$ indicates that dynamics of the nonlinear response can be in a nonperturbative regime, while dynamics of the linear response remain in a perturbative regime. These results lay the groundwork for further studies of quantum dynamics in the nonlinear response of semiconductor microcavities.

We are grateful to R. E. Slusher for a critical reading of the manuscript.

- $8C$. Weisbuch et al., Phys. Rev. Lett. 69, 3314 (1992); R. Houdre et al., ibid. 73, 2043 (1994); Phys. Rev. B 49, 16761 (1994).
- ⁹T. B. Norris et al., Phys. Rev. B 50, 14 663 (1994).
- 10 J. Jackbson et al. (unpublished).
- $11H$. Haug and S. W. Koch, Quantum Theory of the Optical and Electronic Properties of Semiconductors (World Scientific, Singapore, 1993).
- ¹²J. J. Childs, K. An, R. R. Dasari, and M. S. Feld, in Cavity Quantum Electrodynamics (Ref. il.
- 13 Since the lh exciton is only 8 meV from the hh exciton, the transient FWM response can exhibit oscillations that involve lh excitons complicating interpretations of the result.
- ¹⁴M. Koch et al., Phys. Rev. Lett. 69, 3613 (1992).
- ¹⁵See, for example, H. Wang and D. G. Steel, Appl. Phys. A 33, 514 (1991).
- ^{16}D . Frohlich *et al.*, Phys. Rev. Lett. 67, 2343 (1991).