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Time-resolved vacuum Rabi oscillations in a semiconductor quantum microcavity

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We have used femtosecond optical spectroscopy to observe directly the vacuum Rabi oscillations associated with the coupled exciton-photon mode splitting in a semiconductor quantum microcavity. When the microcavity is impulsively excited by a coherent short optical pulse, the time-resolved emission from the cavity shows beats corresponding to the oscillation between cavity and exciton modes, with a decay corresponding to approximately twice the cavity lifetime. Interferometric pump-probe measurements clearly show the coherent evolution of the cavity polarization.

When an ideal two-level absorber (atom) is resonantly coupled to a single mode of the electromagnetic field, the field induces a periodic excitation and deexcitation of the atomic excited state given by the Rabi frequency $\Omega = e\langle r\rangle E/\hbar$, where $\langle r\rangle$ is the transition dipole moment and E the electric field. This Rabi flopping is manifested in the normal-mode spectrum as a splitting (or anticrossing) of the uncoupled atom and field modes.[1,2] The lowest excited states of the uncoupled system correspond to (i) one photon in the field mode with the atom in the ground state, and (ii) the field in the vacuum state (zero photons) with the atom in the excited state. These two states are mixed by the atom-field coupling, and are split by the so-called vacuum-field Rabi frequency.[3] Of course, this splitting is observable only if the atom and field damping rates are small enough compared to the Rabi flopping frequency; this is the strong-coupling regime of cavity QED. The atom-field coupling also modifies the dynamics of radiation. In the weak-coupling limit (strong damping), the effect of the cavity is only to increase or decrease the spontaneous-emission rate of the atom, depending on whether the cavity is on or off the atomic resonance, respectively.

The physics of this simple system has been investigated extensively within the framework of atomic cavity QED (Refs. 4 and 5) or linear dispersion theory,[6] where it has been possible to achieve a nearly ideal system of single cavity mode interacting with a single two-level atom or an absorber. It has also been pointed out[7,8] that essentially the same phenomenon appears in the interaction of light with excitons in a perfect crystal, namely, exciton polaritons. In this case, an exciton of wave vector $\bar{K}$ interacts with only a single mode of the field due to the translational symmetry of the crystal (momentum conservation), and the resulting polariton splitting is equivalent to the vacuum-field Rabi splitting. In fact, in his original paper on exciton polaritons,[7] Hopfield pointed out that this splitting corresponds to a time-domain oscillation between the exciton and photon modes.

More recently, with the development of semiconductor optical microcavities, it has been possible to achieve the strong-coupling regime of cavity QED in a monolithic solid-state system.[8] This was accomplished by situating a GaAs/Al$_x$Ga$_{1-x}$As multiple-quantum-well (MQW) structure in the center of a Fabry-Perot microcavity formed by two distributed Bragg reflectors (DBR's) separated by one wavelength (λ cavity). The sample structure was designed such that the MQW exciton absorption line can be made resonant with the Fabry-Perot cavity mode. On resonance the vacuum Rabi splitting may be expressed as $\hbar\Omega = e\hbar\sqrt{\frac{N}{2m\epsilon L_C}\left(\frac{f}{S}\right)}$, where we have used the electric field per photon $E = \sqrt{\hbar\omega/\epsilon V_C}$, and the exciton dipole moment has been written in terms of the oscillator strength. Here V_C is the cavity volume, L_C the effective cavity length,[9] $\frac{f}{S}$ the oscillator strength per unit area, and N the number of quantum wells. Taking $L_C = 3\lambda$ and $\frac{f}{S} = 6\times10^{-4}$ Å^{-2} for 76-Å quantum wells,[10] we find a splitting of about 5 meV. This is much larger than the exciton-polariton splitting of bulk GaAs (0.08 meV),[11] due to the enhancement of the exciton oscillator strength by the quantum confinement of the exciton, and to the enhancement of the vacuum-field strength by the cavity confinement of the photons. With such a structure, it is now possible to investigate the dynamics of strongly coupled cavity QED in a III-V semiconductor system.

In this paper, we discuss experiments which directly time-resolve the dynamics of resonantly excited semiconductor microcavities. There are a number of motivations for investigating the dynamical behavior of the coupled exciton-cavity system. First, a direct observation of the vacuum Rabi oscillation is desired in a semiconductor system. It is important to know to what extent the MQW-exciton–planar-DBR-microcavity system approaches the ideal case of a two-level absorber–single-mode cavity. Recently, a number of researchers have be-

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gun to study theoretically QED dynamics in DBR and other photonic-band-gap dielectric structures,[12,13] and complementary time-domain experimental investigations are needed. In addition, time-domain experiments provide a way of probing the effect of the in-plane two-dimensional polariton on the system (an effect clearly not present in the atomic cavity-QED system). Second, it is interesting not only to observe oscillatory exciton-photon dynamics, but also to probe directly the coherence of the coupled-mode polarization, as will be shown below. Finally, the dynamics of the semiconductor microcavity system is important for potential applications which rely on the modification of the spontaneous-emission rate.[14] In this paper, we also discuss the emission rate for the case of resonant pumping.

The samples used in these experiments are the same as those of Ref. 8. The front and back DBR's have reflectivities of approximately 98%, leading to a cavity finesse of about 150. Five 76-Å quantum wells were situated in the center of the λ cavity, where the electric-field amplitude of the cavity mode is maximum. Due to a slight spatial variation in growth rate, the cavity resonance wavelength shifts monotonically as one scans across the sample in one direction. The quantum-well resonance, however, is only a weak function of position. Hence, it is possible to tune arbitrarily the cavity in and out of resonance with the exciton absorption simply by optically probing the appropriate position on the sample.

The cw reflectivity spectra of our samples have been reported in Ref. 8. They clearly indicate an anticrossing of the coupled exciton-photon normal modes as the cavity is tuned through the exciton resonance. The normal mode-splitting is 6 meV at resonance. This splitting should correspond to a vacuum-Rabi-oscillation period of 680 fs.

In our experiments investigating the dynamics of the resonantly excited microcavity system, the system is impulsively excited by a coherent optical pulse whose spectral bandwidth exceeds the normal-mode splitting. The initial state of the system is the linear superposition of normal modes corresponding to the photon or cavity mode. As the system evolves in time, the character of the excitation oscillates between the cavity and exciton modes at the vacuum Rabi frequency. The excitation pulse peak power is kept low enough that the Rabi frequency due to the pump-pulse electric field is always much less than the vacuum Rabi frequency; we estimate that the pump-pulse-Rabi-oscillation period was approximately 7 ps for the experiments reported here. (At lower pump powers, the dynamics were independent of intensity as expected, although much higher powers resulted in saturation of the excitonic absorption and a reduction of the vacuum Rabi splitting due to the reduced oscillator strength.) We have performed two experiments to monitor the evolution of the system. First, we have time-resolved the light emitted from the cavity following impulsive excitation. Second, we have performed a pump-probe experiment to observe the time-dependent polarization in the cavity. A Ti:sapphire laser producing 80-fs pulses at a 76-MHz repetition rate was used for both experiments. The sample temperature was 12 K.

A standard upconversion technique was used to time-resolve the cavity emission.[15] The Ti:sapphire laser was split into two beams. One beam (the pump) was used to excite the sample. The pump was focussed to a spot size of 160-μm diameter, with an incidence angle of about 3°. The cavity emission was collected by a lens and imaged onto a thin (0.2 mm) beta-barium metaborate (BBO) crystal. The second beam was passed through a variable-delay line and also focussed onto the BBO crystal. The resulting sum-frequency radiation in the phase-matched direction gave the amplitude of the cavity emission as a function of the pulse delay. It should be noted that for resonant excitation, the radiation from the cavity is coherent in the direction of the reflected pump beam. This is because the exciton dephasing time T_2 at low temperature is expected to be approximately 1 ps,[16] which is longer than both the cavity lifetime (τ_c =140 fs) and the vacuum Rabi frequency (680 fs). Therefore, due to momentum conservation, the polarization excited in the cavity by the pump pulse will coherently radiate in the direction of the reflected pump with a decay time $2\tau_c$. (This decay time is a manifestation of the linewidth averaging effect.[5,6]) Since $T_2 \gg \tau_c$, most of the energy will be radiated away coherently, and almost no luminescence is observed in other directions.

The time-resolved cavity emission for the case of resonant pumping is shown in Fig. 1(a). The reflected pump spectrum is displayed in the inset; the two minima correspond to the normal modes of the coupled system. The pump laser has been tuned so that both modes are excited with nearly equal amplitudes. The time-resolved signal has a strong peak at zero delay, corresponding to the reflected pump pulse from the cavity. At later times, the signal shows a fast decay corresponding to twice the cavity lifetime, and strong beats with a period of 600 fs, close to the expected vacuum Rabi oscillation period. The data shown in Fig. 1(b) correspond to the same experimental conditions as for Fig. 1(a), except that the sample was translated so that the cavity mode and exciton absorption are slightly off resonance; the reflected pump spectrum (inset) shows a normal mode splitting of 11 meV, which should correspond to a vacuum Rabi oscillation period of 370 fs. The time-resolved data show a rapid decay of the radiated signal after the pump pulse, with weak beats with a period of about 350 fs. The small amplitude of the oscillations and faster decay arise from the weaker coupling of the cavity and exciton modes off resonance.

In order to determine the time-dependent cavity emission and population more thoroughly, we performed a pump-probe experiment. In this experiment, the pump-pulse excited the cavity mode at time zero, as in the previous experiment. The second (probe) pulse was then delayed (in increments of 0.67 fs), and overlapped at focus on the sample with the pump beam. The probe beam was also at near-normal incidence to the sample (about 3°), so that the pump and probe were interacting as much as possible with the same cavity mode. The pump beam was chopped and the resulting modulated reflected probe was detected with a photodiode and lock-in amplifier. There are two contributions to the signal in this experiment. One is the change in probe reflectivity induced by the pump-generated exciton population in the quantum wells. The other is light scattered into the probe beam direction by small inhomogeneities in the sample structure; fluctuations in layer thickness and alloy composition of epitaxially grown samples give rise to spatial fluctuations of dielectric constant which act as elastic scattering centers.[17] If scattered light is phase coherent with the probe pulse, then the two pulses interfere on the detector, and fringes are observed as the delay is varied by a fraction of a wavelength. We observe strong fringes when the angle between pump and probe beams is small; this allows us to probe the coherence of the light emitted by the sample.

In Fig. 2, we show the result of a pump-probe experiment in which the microcavity sample is replaced by a

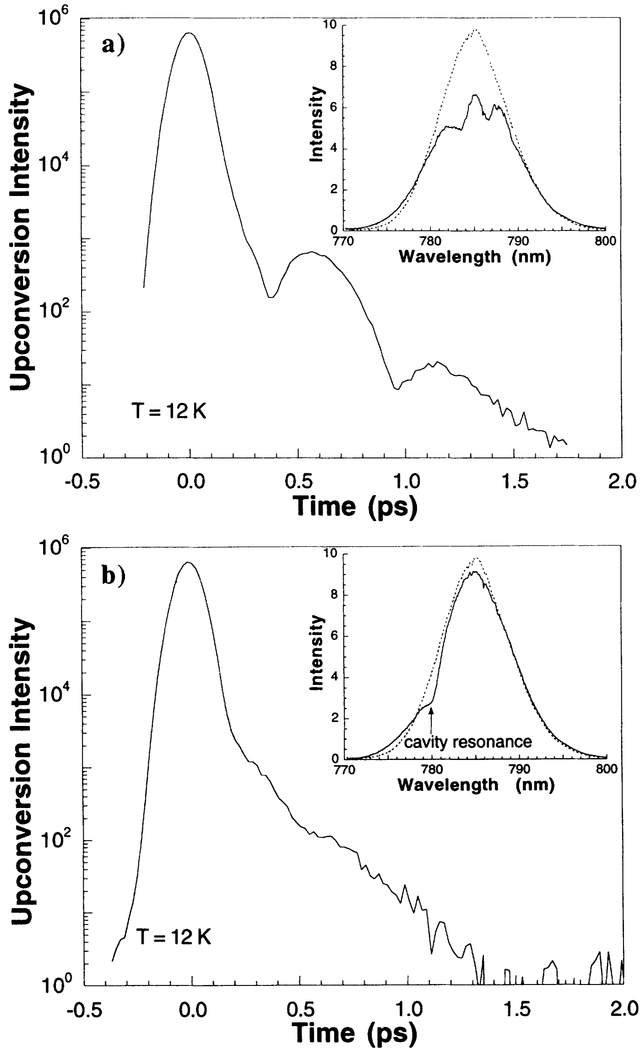

FIG. 1. Time-resolved emission intensity (in arbitrary units) from the impulsively excited cavity. In the insets, the dotted line shows the incident pump spectrum, and the solid line the reflected pump spectrum. The two dips in the reflected pump spectrum correspond to the normal modes of the system. In (a), the cavity and exciton modes are near resonance. In (b), the cavity is detuned from the exciton resonance.

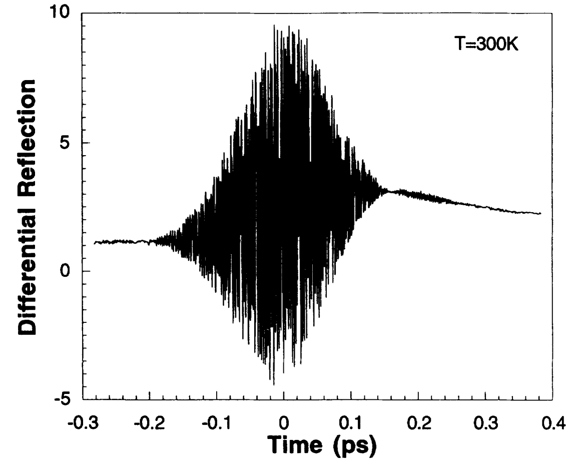

FIG. 2. Interferometric time-resolved pump-probe result for a multiple-quantum-well sample without the top DBR mirror (i.e., no cavity). The laser was tuned to the exciton resonance, and the sample was at room temperature. The ordinate axis is in arbitrary units.

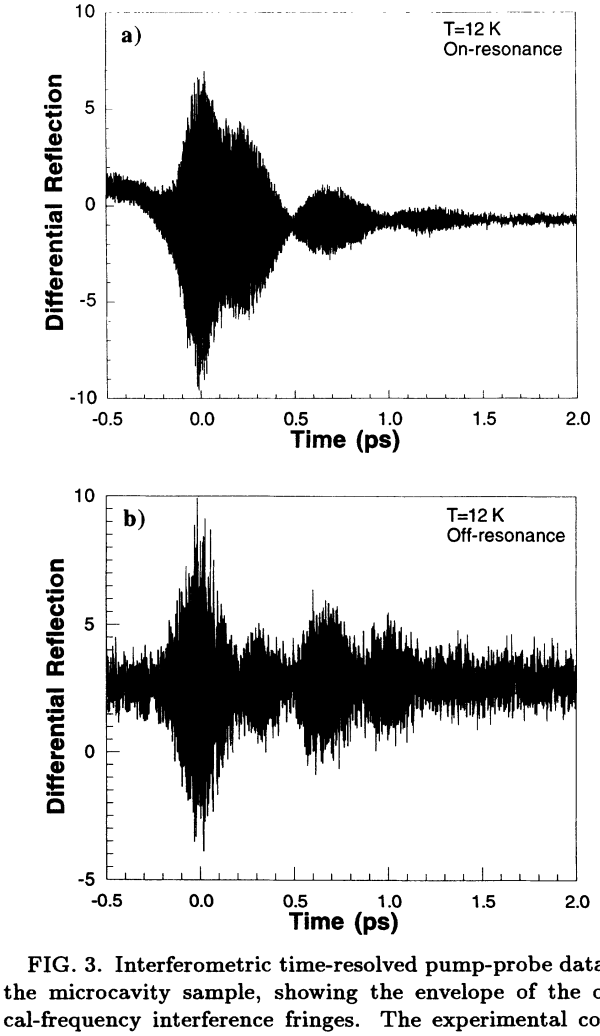

FIG. 3. Interferometric time-resolved pump-probe data for the microcavity sample, showing the envelope of the optical-frequency interference fringes. The experimental conditions are essentially the same as for Fig. 1: (a) resonance, and (b) cavity tuned off resonance. The ordinate axis is in arbitrary units.

sample consisting of a MQW on top of a DBR layer,[19] i.e., there is no resonant cavity. The laser was tuned to the exciton absorption peak. The signal shows a long-lived increase in the probe reflectance induced by the pump, and a coherent signal around zero time delay showing strong interference fringes. The main contribution to the coherent signal is just the scattered pump pulse, followed by a small contribution from the scattered excitonic coherent emission.[18]

In Fig. 3(a) we show the results of the pump-probe experiment with the microcavity on resonance, with essentially the same experimental conditions as for the emission experiment discussed above. The signal shows strong interference fringes at zero time delay, when the pump and probe pulses are coincident on the sample. However, unlike the signal from the sample without a cavity, the fringes persist and show an oscillation with a period of 500–600 fs. The fringes after time zero are an unambiguous signature of the coherence of the polarization remaining in the cavity following the pump pulse. Our interpretation of the signal after $t = 0$ is as follows. Because the "filling time"[20] of the cavity is longer than the pulse width, the field builds up in the cavity with the integral of the pump pulse. Thus, the maximum cavity field occurs within about 80 fs after $t = 0$. The system then undergoes a damped vacuum Rabi oscillation, with the excitation oscillating between cavity and exciton modes. The phase shift of the off-resonance portions of the reflected pump spectrum gives rise to the destructive interference in the signal around 100-fs delay.

When the cavity is translated so that the cavity and exciton modes are off resonance (again with essentially the same experimental conditions as for the emission experiment), the signal changes dramatically, as shown in Fig. 3(b). The coherent spike is still apparent at zero time delay, however, the fringes have greatly reduced amplitude due to the reduced cavity-exciton coupling. The vacuum Rabi oscillation period is reduced to about 330 fs.

To summarize, when a MQW is situated at the field maximum of a Fabry-Perot microcavity such that the exciton absorption is resonant with the cavity mode, the normal modes of the system are linear superpositions of the exciton and cavity modes. These coupled modes may be excited by a short coherent pulse such that initially only the cavity mode is excited. The excitation then oscillates in character between cavity and exciton modes at the vacuum Rabi frequency, maintaining its phase coherence. Since the cavity lifetime is even shorter than the exciton dephasing time, the system coherently radiates in the direction of the reflected pump beam with a lifetime of approximately twice the cavity lifetime. (This should be contrasted with excitonic emission without a cavity. In that case, the lifetime is given by the exciton radiative lifetime, about 10 ps,[21] which is much longer than the lifetime in the strongly coupled microcavity system.) Emission from the cavity is possible when the system excitation is in the cavity mode (that is, a photon in the cavity has a nonzero probability to leak out). Thus the vacuum Rabi oscillation is manifested by oscillatory cavity emission; this was observed by upconverting the emission with a short pulse. The phase coherence of the oscillating polarization was directly confirmed by an interferometric pump-probe experiment.

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