Observation of the Coupled Exciton-Photon Mode Splitting in a Semiconductor Quantum Microcavity

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The spectral response of a monolithic semiconductor quantum microcavity with quantum wells as the active medium displays mode splittings when the quantum wells and the optical cavity are in resonance. This effect can be seen as the Rabi vacuum-field splitting of the quantum-well excitons, or more classically as the normal-mode splitting of coupled oscillators, the excitons and the electromagnetic field of the microcavity. An exciton oscillator strength of 4×10^{12} cm^{-2} is deduced for 76-Å quantum wells.

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There is a present surge of activity in the experimental studies of atom-photon coupling in the context of the interaction between single or few atoms [1–3] and resonant optical media such as cavities [1–4] or structured materials for photonic gaps [5]. The interest is both fundamental, leading to a better understanding of atom-field coupling or stressing analogies between electrons and photons, and applied, as one expects optical systems with unsurpassed properties for applications such as single-mode, high-yield luminescence leading to the thresholdless laser [6].

So far, the studies in the fields of atomic physics and solid state physics (mostly semiconductors) have remained quite separate: The former activities are concentrated on atom-cavity interactions [1–3,7], and the latter on controlled spontaneous emission [4,8,9] and photonic-gap three-dimensional structures [5].

We present in this Letter a solid state QED effect which is a solid state analog of the vacuum-field Rabi splitting [10] so far carried out in atoms [2,3]. Besides its relying on a much simpler implementation—the solid state system is monolithic—the effect should lead to useful applications.

Let us first examine the semiconductor as an atomic system: The usual optical transitions due to the creation of electron-hole pairs yield a distributed oscillator strength over energy bands. Although the standard Rabi oscillations induced by strong optical fields have been observed in solids [11], it has been pointed out [12] that electron-hole pair transitions would not lead to vacuum-field Rabi oscillations, as the relaxation time of carriers is much shorter than the expected vacuum-field Rabi oscillation period. On the other hand, it is well known that sharp, atomiclike excitations do exist at low temperatures in many semiconductors, namely, the excitons, due to the electron-hole interaction, leading to a concentration of oscillator strength from the continuum of electron-hole unbound states into hydrogenlike bound levels [13]. In the ground state of 3D excitons, the oscillator strength per unit volume can be shown to be $\approx f/a_B$ [3], where f is an atomic oscillator strength and a_B is the exciton Bohr radius, therefore increasing by a large factor the Rabi frequency.

Rabi oscillations can be seen as a coupled-oscillator process, by which resonantly coupled atomic and field oscillators periodically exchange energy. In a mechanical oscillator description, the overall system response yields two split modes corresponding to the normal modes. In an atomic transition language, one considers the system as undergoing a coherent evolution with a photon being absorbed by an atom, which subsequently emits a photon with the same energy and wave vector **k**, that photon being reabsorbed, and so on. A simple criterion for this phenomenon to spontaneously occur in a cavity can be put as [3]

$$\alpha d \gg 1 - R \approx \pi/F , \quad (1)$$

where α is the absorption coefficient, d the absorbing medium length, R the cavity mirror reflection coefficient, and F the cavity finesse. This inequality states that before escaping the cavity, an emitted photon is reabsorbed. To be observable, such an effect also supposes that the atom interacts more strongly with the cavity mode than with either all other photon modes or other deexcitation channels [14].

At this point it should be recalled that the equivalent of vacuum-field Rabi oscillations have been proposed and seen long ago in solids, although in a different framework: Various types of polaritons, such as phonon polaritons [15] or excitonic polaritons [16], are just the coupled-mode oscillations of the vacuum field with either phonons or excitons which act as resonant two-level atomic systems. There is no cavity, as these traveling excitations possess both well-defined energy and wave vector, and therefore are only coupled to the one photon mode with the same energy and wave vector. Thus, irreversible coupling to the continuum of photon states is nonexistent, and only the scattering of polaritons by other material excitations can destroy the propagating coherent mode. Most direct evidence of polaritons has been provided by reflectivity [17] and light scattering experiments [18,19].

Although representing the fundamental electromagnetic excitations of 3D crystals, polaritons are not versatile and cannot be tailored at will. The new system (inset of Fig. 1) which we use here should prove much more important in that respect and is a monolithic solid state re-

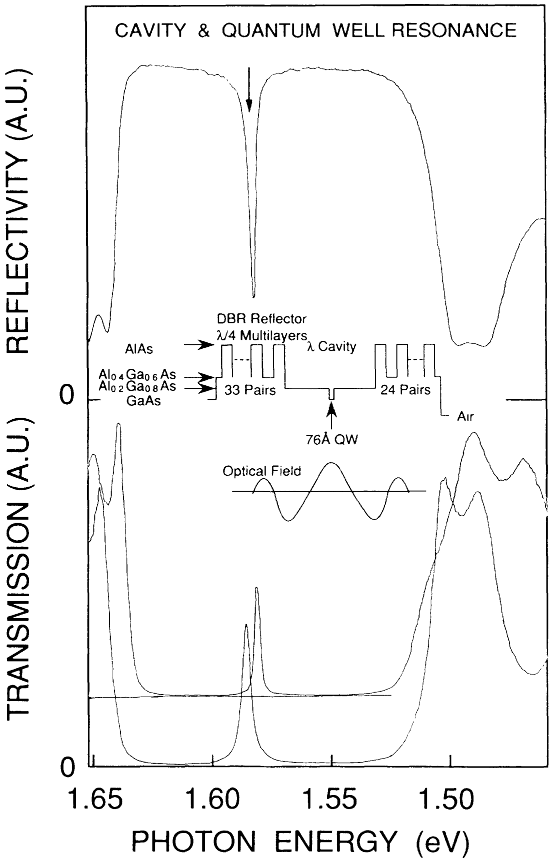

FIG. 1. 20-K reflectivity and transmission spectra of a single-quantum-well λ microcavity (structure shown in the middle of the figure). The bottom transmission spectrum corresponds to a detuned cavity with respect to QW exciton frequency while the two other spectra correspond to equal cavity and exciton frequencies, a situation reached by selecting the area on the wafer (see text).

plica of the usual atomic system [2,3]: The Fabry-Pérot (FP) cavity is made of an $L=\lambda$ GaAlAs layer with one or several GaAs quantum wells (QW's) imbedded between two distributed Bragg reflectors [20] (DBR's) made of quarter-wave stacks of GaAlAs/AlAs layers. This structure is very similar to that of the widely researched vertical cavity surface emitting lasers [21] (VCSEL's). The resonance frequency of the FP resonator is adjusted to the imbedded QW ground-state exciton energy. The distributed feedback (DFB) mirror center frequency is also adjusted at that value. The use of two-dimensional (2D) quantum wells as the optically active material, i.e., the one whose frequency will be brought in resonance with the FP cavity, leads to several properties: (i) As the QW layers are perpendicular to the cavity propagation axis, the excitons are blocked in each QW and cannot propagate in that direction. There is no exciton polariton like in the 3D semiconductor case [22]. (ii) For the cavity mode, the QW excitons therefore act as immobile excitations, atomlike, with an oscillator strength Nf per unit area per QW given by (in the exact quantum limit) [23]

$$Nf=8f/\pi a_B^2 \qquad (2)$$

(where f and a_B retain their 3D value), and with a coupling to the cavity optical mode which can be varied by changing the number of quantum wells. Now, of course, in contrast with bulk semiconductors, the cavity plays an essential role as it selects one photon mode with which QW excitons will be coupled preferentially to all other modes.

This is, however, a simplified picture, and we should stress here a major difference between this solid state experiment and atomic physics experiments: In these, the atomic beam resonance is narrow enough to select the one resonant cavity mode out of the continuum of oblique modes of a planar FP resonator. In our experiments, excitons can have in-plane wave vectors which allow excitons to couple to oblique FP modes. It therefore seems surprising that the excitons, which can interact with a continuum of photon modes, should display a Rabi oscillation evidencing the coupling to a single photon mode. However, because of the translational invariance of the crystal in the QW plane, the in-plane exciton wave vector is a good quantum number and must equal that of a photon in an optical transition. Therefore, an optically created exciton will have a well-defined in-plane wave vector, and will only interact with one cavity mode with the same transverse photon wave vector as long as the Rabi oscillation frequency is faster than the randomization time of the exciton wave vector (determined, for instance, from transient four-wave-mixing experiments [24]).

Our cavities, grown by metal organic chemical vapor deposition (MOCVD), involve 24 GaAlAs/GaAs stacks on the front side (air interface) and 33 stacks on the substrate side in order to balance the front and back side reflections at $\sim$98%. Despite this precaution, even well below the QW exciton energy when the QW does not play any role, we cannot obtain low reflectivity values at the FP resonance. This seems to be very general for all published data on microcavities [4,21]. In any event the good quality of the cavities is well attested by the expected and measured finesse $F\approx 100-300$. The sample is illuminated with a collimated white light source. Spatial selection is provided by imaging the sample on the entrance slit of an analyzing spectrometer. Taking into account the 2:1 enlargement ratio of the setup, the measured spot size on the sample is ≈ 0.5 mm high$\times$0.05 mm wide. We checked that the beam aperture does not induce any spectral broadening by repeating the same transmission or reflectivity experiments with a focused low-divergence Ti:sapphire laser. For the transmission measurements, the GaAs substrate has been etched down to the back mirror. Figure 1 shows a reflectivity and two transmission curves obtained at $T=20$ K for a single-quantum-well sample. As is usual, the thickness variation on the wafer is such that the FP wavelength varies ≈ 300 Å from wafer center to side, while the QW exciton frequency is hardly changed, its energy corresponding mainly to the GaAs band gap. To change the FP cavity frequency, one therefore only needs to probe different points on the wafer. The topmost curves in Fig. 1 correspond to equal resonant frequencies of the cavity and quantum-well exciton, while the bottom one corresponds to a cavity energy detuning above the exciton. Figure 2 shows various reflectivity curves for a sample with seven quantum wells in the Fabry-Pérot cavity.

A simple theoretical treatment consists in considering the DFB reflectors as simple, wideband mirrors of constant reflectivity and transmission coefficients. Then, the standard multibeam Fabry-Pérot analysis can be carried out, which yields the transmission coefficients as [3]

$$T(\nu)=\frac{T^2e^{-\alpha d}}{(1-Re^{-\alpha d})^2+4Re^{-\alpha d}\sin^2(\Phi/2)}\,, \tag{3}$$

where R and T are reflection and transmission coefficients of the front and back mirrors, assumed identical, α the absorption coefficient of the cavity QW's with total length d, and Φ the dephasing over the various materials of the cavity,

$$\Phi(\nu)=2\pi(\Delta-\Delta_{\rm res})/\Delta_{\rm FSR}+4\pi(n-n_b)d\nu/c\,, \tag{4}$$

where $\Delta=\nu-\nu_{\rm exc}$, $\Delta_{\rm res}=\nu_{\rm cav}-\nu_{\rm exc}$, $\Delta_{\rm FSR}=c/2L$ is the cavity free spectral range, and n and n_b are respectively the resonant and background index of refraction of the cavity quantum wells. We use for the exciton dielectric function a simple two-level approximation with Lorentzian linewidth $\delta=2$ meV. The peak absorption α is adjusted to the standard value of 3×10^4 cm^{-1}. Such a description, classical linear-dispersion model, of the system then yields two peak or dips in the transmission or reflectivity curves, respectively, separated by, when the oscillator frequencies are equal ($\nu_{\rm exc}=\nu_{\rm cav}$) [3],

$$\Omega=(\alpha d\delta\Delta_{\rm FSR}/\pi)^{1/2}. \tag{5}$$

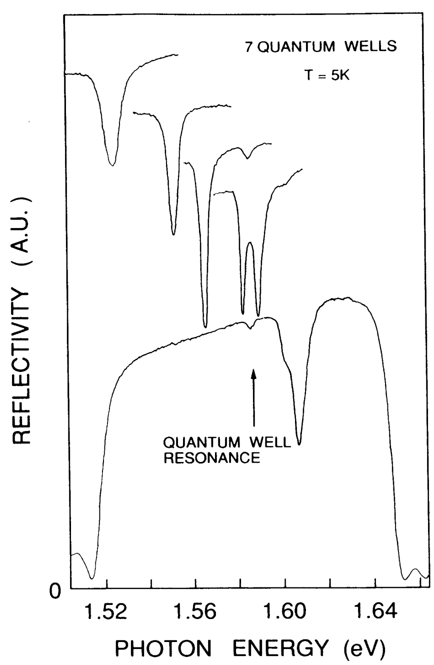

FIG. 2. 5-K reflectivity curves on a seven-QW microcavity structure. Various detuning conditions between cavity and QW exciton frequencies are obtained by choosing various points on the wafer, typically 0.5 mm apart. Note the line narrowing approaching and at resonance, the resonance mode splitting, and the indication of a light-hole exciton mode splitting around 1.605 eV for the lowest trace.

Figure 3 shows a plot of the peak positions observed when scanning through the resonance, evidencing equivalently a well-behaved level anticrossing, and normal-mode splitting or Rabi splitting. The theoretical fit is obtained from the above simplified analysis, or a standard multiple-interference analysis of the DBR-FP interferometer using a matrix formulation for optical propagation [20]. From the fit with $\alpha=3\times10^4$ cm^{-1} and $\delta=2$ meV, we can deduce $Nf=4\times10^{12}$ cm^{-2}, in excellent agreement with theoretical evaluations [25].

As mentioned previously the number n of quantum wells can be varied to provide various coupling strengths between excitons and cavity photons. As could be expected, a single well does not provide enough coupling to fulfill Eq. (1) (see, e.g., Fig. 1). At a number of wells ≥ 5 the splitting tends to saturate, possibly due to the noncentral position of outer wells in the cavity, collapse of the cavity finesse at such high absorption, or influence of other states (light-hole excitons, continuum states).

Several additional comments can be made at this preliminary stage: As is evident in Figs. 1 and 2, the cavity linewidth becomes smaller under resonant conditions. In atomic physics experiments, as the cavity finesse is usually extremely high, the coupled-mode linewidth can be smaller than the natural atom linewidth [2,14]. We are here in a converse situation where the cavity linewidth is narrowed by the interaction with QW's.

So far we used low temperatures (up to 77 K) to sharpen the features described here. We did not yet check at 300 K, as our structures are designed for mode splitting of the FP cavity with QW's at low temperatures, but we expect similar phenomena as the broadening should be

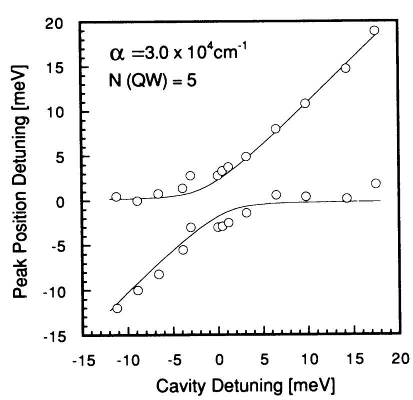

FIG. 3. Reflectivity peak positions as a function of cavity detuning for a five-quantum-well sample at $T=5$ K. The theoretical fit is obtained through a standard multiple-interference analysis of the DBR-Fabry-Pérot-quantum-well structure.

smaller or at most comparable to the splitting [23].

The present results should influence our design of several important optical devices. In VCSEL's, a main impact will only occur if laser action is based on exciton recombination. However, it is well known that, at least at room temperature, exciton dissociation and carrier interactions are much faster than the exciton radiative lifetime (in the ns range), so that excitons are usually wiped out into electron-hole pairs [23]. The existence of the rapid Rabi oscillation might drastically change this state of affairs, as the oscillation could be more rapid than the dissociation time, leading to efficient radiative recombination whenever a coupled exciton-photon mode escapes the cavity.

For those electro-optical or nonlinear optical devices which rely on unrelaxed excitations, excitons are still very important at room temperature [23] and are at the root of the unsurpassed performance of QW heterostructure-based systems [26]. So far, the structures involved many quantum wells as active materials, typically 30–100. The present experiments evidence that, due to coupled moding, the cavity optical properties are determined by as few as three QW's. This indicates that modification of the QW properties by the usual external actions such as an electric field (quantum confined Stark effect) or carrier-band filling can be exerted on much less material when in a strong coupled-moding situation (i.e., a high-Q cavity), leading to figures of merit of such devices improved by a factor 10–30.

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