Controlled Exciton-Photon Interaction in Semiconductor Bulk Microcavities

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We report the first experimental investigation of the spectral response of a "bulk microcavity": a semiconductor system in which the same cavity material is the active medium. Despite the smaller oscillator strength of the bulk exciton, a Rabi splitting even clearer than that displayed by usual quantum-well microcavities is observed. The splitting increases with the cavity layer thickness and the exciton dispersion produces a satellite structure due to quantum confinement. We also show that control of the exciton-photon interaction can be realized by suitably tailoring the photonic wave function.

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In the last few years, the study of quantum microcavities has fascinated many researchers, mainly because, by altering the photonic density of states, these structures allow the investigation of fundamental quantum electrodynamics and are ideal candidates for new optical devices. The first experiments were performed in the field of atomic physics [1], but presently a great interest has surged in the semiconductor world because solid-state cavities with Bragg reflectors as mirrors can now be produced by standard growing techniques such as molecular beam epitaxy (MBE) or metalorganic chemical vapor deposition.

The interaction of quantum-well (QW) excitons with cavity modes has been the object of extensive experimental investigation, concentrating on the control (inhibition or enhancement) of spontaneous emission, and its consequences in the lasing performance [2–6]. Furthermore, the QW exciton *vacuum-field Rabi splitting* has been recently observed in a variety of structures both at low and room temperature [7–9]. This has stimulated a number of theoretical studies concerning the propagation of mixed light-exciton modes (namely, "cavity QW polaritons") in these new systems [10–15].

We have previously proposed a new "self-tuned" microcavity structure, in which the radiation cavity mode is in interaction with the exciton mode of the bulk [16–18]. Of course the fundamental electromagnetic excitations of bulk semiconductors are constituted by the excitonic polaritons, which are already stationary coupled exciton-photon modes [19,20]. In the optical properties, however, polaritonic effects are difficult to detect [21]. Our "bulk microcavity" structure can thus be used to actually "quantize" the bulk polaritons in both their photon and material components, much in the same way as the simpler and more familiar thin-layer system provides quantization of the exciton part only [22–24]. By tuning the optical cavity mode on the bulk exciton transition, we can use the entire body of the cavity as the active material. With respect to the usual QW cavities, we can take advantage of the larger useful thickness ($d \simeq 200$ nm) and of the smaller broadening ($dE \simeq 0.1$ meV) of the three-dimensional exciton, thus compensating for the relatively small oscillator strength. Theoretical calculations have been carried out, showing that these structures exhibit very strong and detailed Rabi splitting features and that enhancement or inhibition of the interaction with the photon can be obtained by controlling the reflection at the Bragg mirrors and the cavity thickness [16,17].

In this Letter we present the first experimental analysis of bulk microcavities in the *strong-coupling* regime, reporting the observation of two Rabi-split peaks in the reflectance as well as in the photoluminescence (PL) spectra. Differences with the QW case in both the magnitude of the splitting and the line shape of the peaks are found and discussed; a semicavity system (a regular cavity structure in which, however, only one of the two mirrors has a sufficiently high reflection coefficient to actually confine the photon) is also studied, to show the effect of the phase of the mirror reflectivity on the exciton-photon interaction.

The samples were grown by solid-source MBE on Si-doped GaAs(001) substrates. All structures contained n-type (Si-doped, $n = 1 \times 10^{18}$ cm^{-3}) 0.5-μm-thick GaAs(001) buffer layers grown at 600 °C. Four different GaAs cavities, tuned on the GaAs 1s exciton transition (1515 meV), were realized: two $\lambda/2$ semicavities and two full cavities ($\lambda/2$ and 2λ). In the full-cavity samples, the two mirrors, constituted by distributed Bragg reflectors (DBR's), were composed of 20 and 18 pairs of Al$_{0.18}$Ga$_{0.82}$As/AlAs $\lambda/4$ layers with nominal thickness of 595 and 696 Å, respectively. In both semicavities the two mirrors were composed of 22 and 1 pairs of Al$_{0.18}$Ga$_{0.82}$As/AlAs layers, respectively, the only difference being which layer (AlAs or AlGaAs) is at the GaAs interface. In order to achieve the required precision

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in tuning the Fabry-Pérot resonator thickness, sample rotation was stopped during its growth. This resulted in a wedgelike shape for the cavity layer, enabling us to change the frequency of the optical mode by simply moving the light spot on the sample. A 50-Å-thick GaAs cap layer concluded the growth of all structures.

A 100 W xenon tungsten-wire lamp was used for reflectance measurements and a He-Ne laser as excitation source in the PL analysis. The signal was detected with a GaAs photomultiplier tube, a 1-m double monochromator, and a lock-in amplifier. The temperature was kept at 14 K using a closed-cycle cryostat.

We report in Fig. 1 the reflectance spectra of the $\lambda/2$ full-cavity sample. The various curves refer to different points of the samples, corresponding to different thickness of the Fabry-Pérot resonator. The typical anticrossing behavior of the coupled exciton-photon modes is clearly visible, and a Rabi splitting somewhat larger than 3 meV is detected. This is slightly less than what can be theoretically predicted for the same structure using an adapted transfer-matrix approach which features Pekar-Hopfield's additional boundary conditions to include the polaritonic spatial dispersion [17,25]. Many secondary peaks appear when the exciton and the photon are heavily mixed, owing to quantization of the exciton center-of-mass motion, similar to the case of thin layers [22].

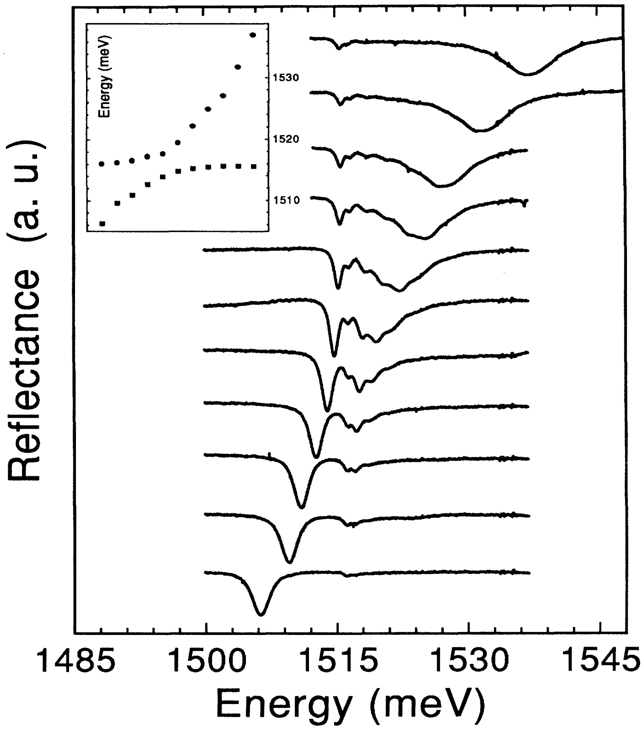

FIG. 1. Reflectance spectra of the $\lambda/2$ cavity; each curve refers to a different cavity thickness, with increasing thickness from top to bottom. The resonance condition ($\lambda_{\mathrm{exc}}/2 \simeq L_{\mathrm{cav}}$) is at about the seventh curve from the top. In the inset we report the energy position of the two main peaks for the various cavity thicknesses.

The PL spectra of Fig. 2 show the same general features. The center-of-mass quantized states are not apparent in this case, nevertheless their presence has strong consequences. In fact the separation between these states is sufficiently small to allow fast relaxation to the lowest level through acoustic phonon emission. Therefore almost no luminescence can be recorded from the quantized states. Additionally the high energy Rabi peak disappears when it starts to move away from the exciton resonance because many intermediate levels are present between the two Rabi states.

Comparing the spectra of Figs. 1 and 2 with those relative to QW cavities, one can note the better resolution of the peaks obtained in our system. This can be linked to the small broadening of the bulk exciton transition. We wish also to stress that no particular care had to be exercised in having a high-finesse cavity; in fact the Fabry-Pérot mode linewidth of our sample was several meV to be compared to the fractions of meV usually required by QW microcavities.

As mentioned above, we have also studied a 2λ cavity. In our bulk system a behavior opposite to that of QW cavities is expected. In the latter case, in fact, the role of the cavity is twofold: it allows the formation of a coherent traveling mixed mode (the cavity polariton) and in some sense quantizes its wave vector. Therefore the Rabi splitting is expected to increase with the active thickness d due to the increased superposition between material and photon wave functions. It is expected to decrease, however, with increasing cavity thickness L because of the deteriorated photon confinement. In fact, for a perfectly

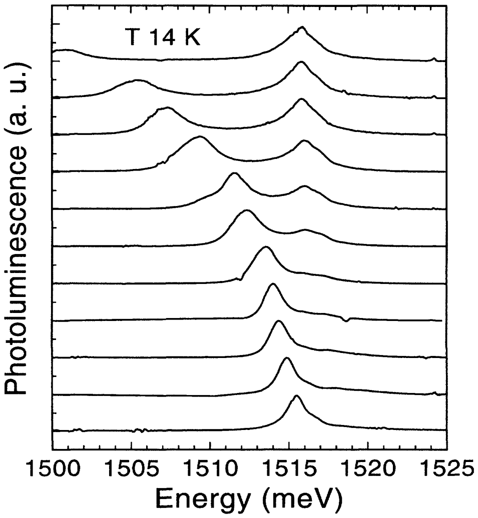

FIG. 2. Photoluminescence spectra of the $\lambda/2$ cavity; each curve refers to a different cavity thickness, from largest (top) to thinnest (bottom).

tuned Fabry-Pérot mode $\nu_{\rm cav} = \nu_{\rm exc}$, the Rabi splitting magnitude is given, for metallic mirrors, by [26]

$$\Omega = (\alpha d \delta c / 2L\pi)^{1/2}, \tag{1}$$

where α is the absorption coefficient, δ the linewidth of the transition, and c the speed of light. For a QW in a semiconductor cavity it can be calculated with [13]

$$\Omega = 2\sqrt{\omega_0 \omega_{\rm LT} (d/L_{\rm eff})}, \tag{2}$$

where ω_0 is the exciton resonance energy, $\omega_{\rm LT}$ the effective longitudinal-transverse splitting, and $L_{\rm eff}$ the effective cavity thickness, which accounts also for the field penetration in the DBR's. In our case $d = L$ and bulk polaritons propagate in the GaAs layer with the cavity simply quantizing their motion. Hence one should observe a splitting independent of the cavity thickness and always equal to the bulk polariton splitting at the cross point $2\sqrt{\omega_0 \omega_{\rm LT}/2}$ [27]. Note that even if (2) gives a splitting independent of the thickness in the case $d = L_{\rm eff}$, the result does not hold for the bulk because (2) is valid only in the limit $d \ll L$. Actually the penetration of the field in the DBR's makes $d = L < L_{\rm eff}$ also for the bulk and an increase of the Rabi splitting with increasing cavity thickness is obtained due to the larger ratio $d/L_{\rm eff}$ [17]. Although a square root dependence on $d/L_{\rm eff}$ is roughly correct also in this case, the exact value of the splitting is to be deduced from the calculated optical properties, as shown in detail in [17]. Indeed in our 2λ cavity we observed a Rabi splitting about 1 meV larger than that of the $\lambda/2$ sample (see Fig. 3); this qualitatively confirms our theoretical predictions, although the magnitude is less than what was calculated [25].

Let us now examine the phase effect of the Bragg mirror reflection, making use of the semicavity structure, which avoids concentration of the oscillator strength in the two main Rabi peaks, allowing a broad range of photon frequencies around the resonance to be examined. There are two ways of constructing a dielectric mirror. Let n_1 and n_2 be the refraction indices of the two materials which constitute the $\lambda/4$ layers. Assuming the mirror starts with a n_1 layer and ends with a n_2 layer, we have two cases: (i) $n_1 > n_2$, (ii) $n_1 < n_2$. At the center of the stop band of a Bragg mirror, the reflection coefficient is [28]

$$r|_{\omega_0} = \frac{1 - (n_{\rm sub}/n_{\rm cav})(n_1/n_2)^{2N}}{1 + (n_{\rm sub}/n_{\rm cav})(n_1/n_2)^{2N}}, \tag{3}$$

where N is the total number of $\lambda/4$ layers, $n_{\rm sub}$ the refractive index of the substrate, and $n_{\rm cav}$ that of the cavity. Clearly, for large N, $r = 1$ if $n_1 < n_2$ and $r = -1$ if $n_1 > n_2$. This means a π-phase shift for $n_1 > n_2$ reflectors and a 0-phase shift for $n_1 < n_2$ reflectors. Now if a bare cavity layer is grown on one of the two Bragg mirrors, the photon field will be modulated strongly depending on the phase matching with the supporting mirror. This can be used as a way to control the exciton-photon interaction. We show in Fig. 4 the reflectance spectra measured for two $\lambda/2$ semicavities having the two configurations of the dielectric mirrors described above. A structure with many peaks due to the quantization of the exciton center-of-mass motion clearly appears in the $n_1 < n_2$ case. In the other case only the resonance peak can be detected and the higher energy features are completely wiped out [16,17]. In fact the transition probability P is proportional to the square of the overlap between the photon field wave function $\phi_\nu(z)$ and the function which describes the exciton center-of-mass motion $\psi_{\rm ex}(z)$:

$$P \propto \left| \int \phi_\nu(z) \psi_{\rm ex}(z)\, dz \right|^2. \tag{4}$$

The optical waves in the cavity layer have an antinode or a node at the mirror interface depending on whether

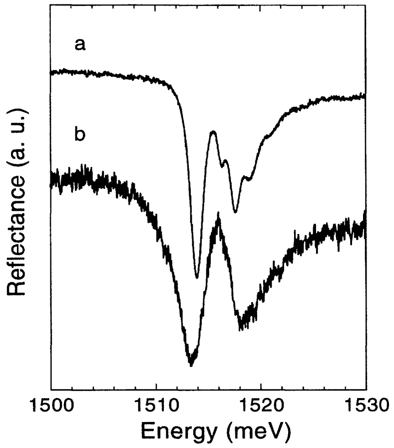

FIG. 3. (a) Reflectance spectrum of the $\lambda/2$ cavity near the polariton cross point; (b) the same for the 2λ sample.

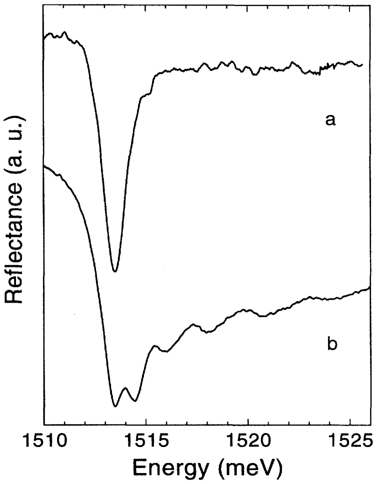

FIG. 4. (a) Reflectance spectrum of a $\lambda/2$ semicavity with the Bragg mirror having $n_1 > n_2$; (b) the same for a $\lambda/2$ semicavity with the Bragg mirror having $n_1 < n_2$.

the reflectivity of the DBR is $r = 1$ or $r = -1$. On the contrary the exciton wave function must always go to zero at the surfaces. It follows from (4) that, for a $\lambda/2$ semicavity, all the exciton states with odd parity with respect to the center of the layer are optically active if $r = 1$, while only the first even state has a nonvanishing overlap with the photon if $r = -1$.

In summary, we have obtained the first direct experimental evidence of the exciton Rabi splitting in a bulk semiconductor, showing the dramatic effects that the polaritonic behavior can have in the optical properties of a bulk microcavity. The advantages related to the large active thickness and to the small broadening of the three-dimensional transition have been discussed in relation to the usual QW cavity system. The phase effect of the Bragg mirror reflection was also analyzed, demonstrating a new way to control the exciton-photon coupling, achievable tailoring of the structure of the DBR's.

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