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Uncoupled excitons in semiconductor microcavities detected in resonant Raman scattering

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We present an outgoing resonant Raman-scattering study of a GaAs/AlGaAs based microcavity embedded in a *p-i-n* junction. The *p-i-n* junction allows the vertical electric field to be varied, permitting control of exciton-photon detuning and quenching of photoluminescence which otherwise obscures the inelastic light scattering signals. Peaks corresponding to the upper and lower polariton branches are observed in the resonant Raman cross sections, along with a third peak at the energy of uncoupled excitons. This third peak, attributed to disorder activated Raman scattering, provides clear evidence for the existence of uncoupled exciton reservoir states in microcavities in the strong-coupling regime.

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The strong coupling between the cavity and exciton modes in semiconductor microcavities (MC's) has attracted a great deal of interest in recent years as a novel means to study and control the interaction between light and matter in semiconductors.[1,2] The resulting coupled photon-exciton modes, cavity polaritons, have been shown to exhibit a number of novel properties, including recently stimulated scattering arising from the bosonic character of the polariton quasiparticles.[3,4] It is now apparent that the polariton dynamics of MC's excited under conditions of nonresonant, high-energy excitation are governed largely by the dynamics of excitons in high k states ($k > \sim 10^6$ cm^{-1}), the so-called exciton reservoir. The density of these states is approximately 10^4 times higher than that of the states in the strongly-coupled polariton region, as a result of the large exciton mass of $\sim 0.2 m_e$ compared to the polariton mass of $\sim 10^{-4} m_e$. Direct evidence for states in the exciton reservoir is, nevertheless, to a large degree lacking largely, since they have k vectors outside the light cone ($k < 7.4 \times 10^4$ cm^{-1}) around the normal to the surface, and are thus not observable directly in optical measurements performed in the standard geometry in which light is incident on or exits through the Bragg mirrors of the cavity. Uncoupled exciton states have, however, been observed in photoluminescence (PL) detected from the edge of the cavity,[5,6] where coupling to the confined photon modes of the MC does not arise.

In this work, we probe the properties of an MC using resonant inelastic light scattering, and obtain strong evidence for the existence of uncoupled exciton states in experiments performed from the surface of the MC. The Raman photons are detected in the outgoing channel in which photons scattered by one LO phonon are resonant with exciton/polariton states of the system.[7–9] The detected photons are transmitted through one of the Bragg mirrors close to $k = 0$, wave vector being conserved in the inelastic light scattering probably by the disorder activated character of the resonant Raman process, which has been shown to be strong for excitons in quantum wells.[10–14] Reflectivity with a weak probe beam was measured simultaneously with Raman scattering to ensure that the exciton-photon coupling was unperturbed by the application of the laser probe. This was important in view of the marked contribution of uncoupled excitons to the Raman spectra, which would be expected to dominate the optical response if strong coupling were lost due to screening.[15] PL from lower branch polaritons, which would otherwise obscure the Raman signals was quenched by applying electric fields up to 30 kV/cm to sweep out photocreated electron-hole pairs.[16] Since Raman scattering is a virtual coherent process which occurs without the creation of real carriers or excitons, it is much more weakly affected by the relatively weak fields. In Refs. 7–9 the MC Raman signals were interpreted in terms of polariton mediation, as opposed to the exciton mediation observed here; the present results do not necessarily contradict those reported earlier, but do show that exciton mediation can nevertheless play an important role in the Raman process in MC's.

The sample studied was a $3\lambda/2$ GaAs cavity, incorporating two sets of three 100 Å $In_{0.06}Ga_{0.94}As$ quantum wells separated by 100-Å GaAs barriers, positioned at the antinodes of the optical field. The top (bottom) mirror consisted of 17 (20) repeats of $\lambda/4$ layers of AlAs and $Al_{0.18}Ga_{0.82}As$. Doping was introduced into the mirror regions to form a *p-i-n* junction, so that an electric field could be applied across the intrinsic cavity region. This allowed the quenching of PL discussed above, thus making it possible to detect the relatively weak outgoing Raman signal for both the lower and upper polariton branches, in contrast to other work, where the intensity of lower branch PL permitted study of the upper branch alone.[9] By selecting the strength of the electric field, it was also possible to tune the resonance conditions by varying the exciton energy due to the quantum confined Stark effect,[17] and as a result to confirm that uncoupled excitons contribute strongly to the Raman spectra. The experiments were performed on 400-μm diameter mesas with 200 μm apertures in the top metal contact for optical access.

Figure 1 shows a schematic of the experimental arrangement. The incident photons for the Raman scattering, performed at 10 K, were provided by a 1 W/cm^2 beam from a tuneable titanium-sapphire (Ti:S) laser, focused on to the sample by lens l_2 to a spot size of diameter $\sim$100 μm at an

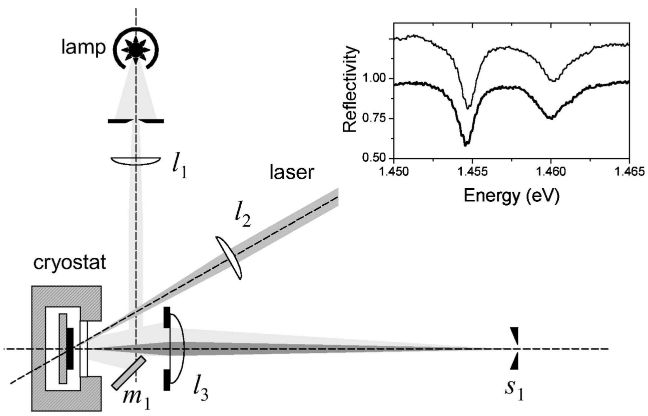

FIG. 1. Schematic of experiment. Inset shows white light reflectivity without laser excitation (top curve, vertically shifted for clarity) and under 1 W/cm^2 laser excitation (bottom curve).

angle of incidence $\theta=36°$. Plane parallel white light from a tungsten-halogen lamp was also incident on the sample close to $\theta=0$. Both the reflected white light, and the outgoing Raman signal were collected at $\theta=0$ by lens l_3, and magnified by ~15 to an image at the entrance slit (s_1) of a triple subtractive grating spectrometer. The large image size allowed the selection of the $\sim 50\times 50$ μm central region of the sample. The intensity of the Ti:S laser beam was more than a factor of 10 lower than the intensity at which any perturbation of the white light signal was detected (see Fig. 1 inset), thus ensuring that strong coupling was retained.

The incoming laser energy was tuned so that the outgoing Raman photon was close to resonance with the polariton modes at normal incidence, as shown in the schematic diagram of Fig. 2. For $\theta=36°$ this energy falls within the stop band of the Bragg mirrors, although its proximity to the strongly photonlike upper branch (see Fig. 2) permits an estimated ~1% transmission of photons into the cavity at energies falling with the exciton continuum of the quantum wells. Very similar results are obtained for conditions of double optical resonance, also shown schematically in Fig. 2, where both the incident[18] ($\theta\sim 51°$) and scattered photons are resonant with polariton modes. The single resonance conditions for $\theta=36°$ have the advantage that the transmission of the incident photon varies only slightly as a function of incident energy, whereas there is a huge variation in transmission for double resonance, which strongly distorts the shape of the resonant Raman profile.[19]

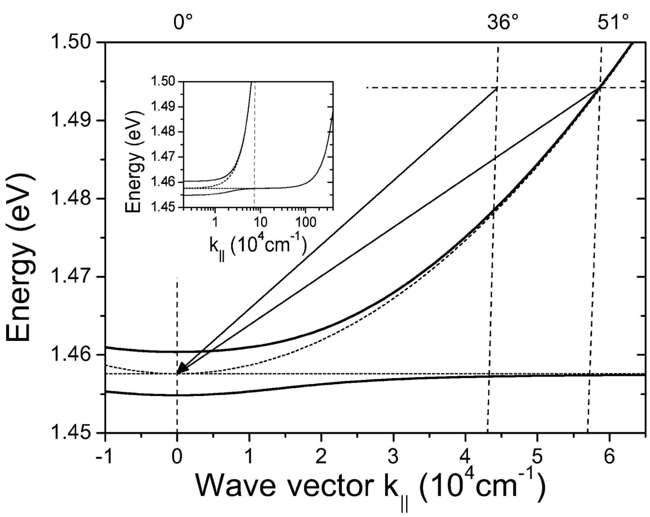

FIG. 2. Microcavity dispersion, showing polariton modes (solid lines), and uncoupled cavity and exciton modes (dashed lines). The conditions for doubly resonant Raman scattering are met at an angle of incidence of ~51°. The present experiments are performed at 36°. The scattering processes are indicated by arrows. The inset shows the polariton dispersion over a greater region of k space, highlighting the high density of states in the exciton reservoir. States to the right of the dashed vertical line fall outside the light cone and cannot couple directly to external light.

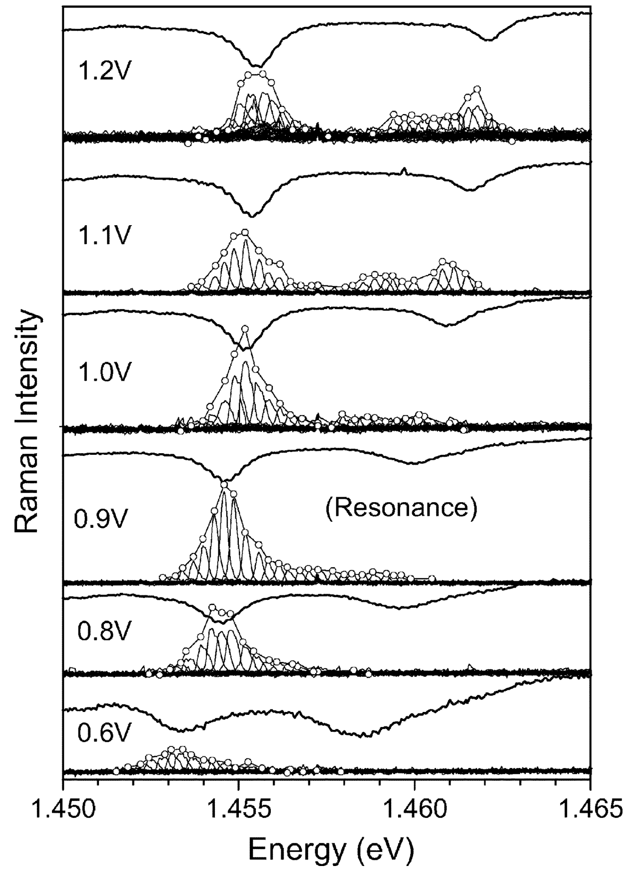

FIG. 3. Resonant Raman spectra as a function of outgoing photon energy are shown as a series of peaks for each bias. The Resonant Raman profiles corresponding to the integrated intensity of the Raman peaks are shown as points. Reflectivity spectra performed under the same conditions are shown at the top of each panel.

Near normal incidence reflectivity spectra, recorded under simultaneous Ti:S laser excitation, as a function of bias, are shown in Fig. 3. The spectra all have the same intensity scale, but are vertically shifted for clarity. Two well-resolved dips corresponding to the upper and lower polariton branches are observed with resonance being achieved at 0.9 V, separated by a well-defined region of reflectivity ~95% (~5% transmission into the sample) between the branches. A series of Raman spectra (white light switched off) are shown in Fig. 3, for the same set of bias voltages. For each successive Raman spectrum the energy of the incoming laser was incre-

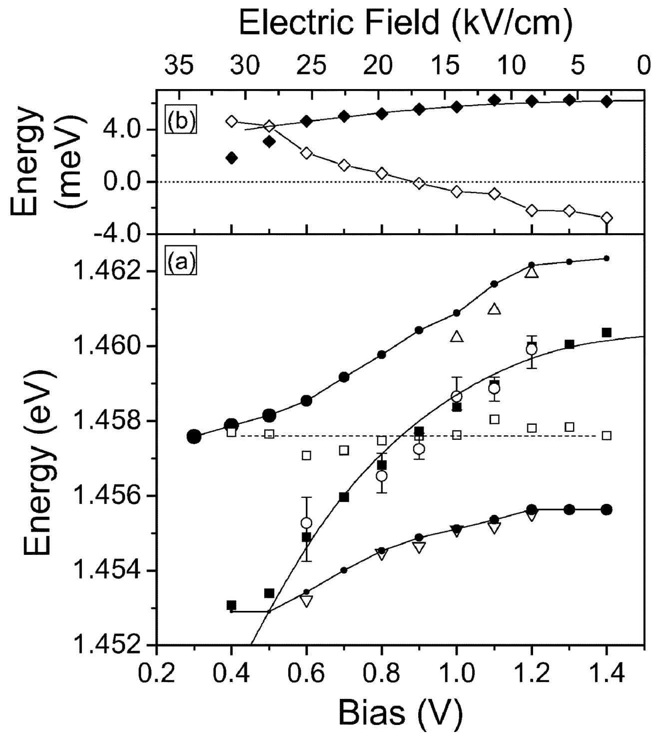

FIG. 4. (a) Measured energies of the polariton resonances in reflectivity (solid circles). The area of the circles is proportional to the intensity of the dip. The corresponding peaks in the RRP are shown as open triangles. Open and solid squares represent the deduced energies of the uncoupled photon and exciton modes, respectively. The maximum of the central peak in RRP is shown as open circles. The smooth full line shows the field dependence of the exciton energy calculated from solution of the Schrödinger equation for the QW excitons. (b) Measured Rabi splitting and cavity detuning, closed and open points. Smooth solid line—calculated field dependence of Rabi splitting.

mented in steps of ~0.1 meV, at an energy of 36.4 meV (the energy of GaAs LO phonons) above that of the scattered photons. The resolution limited (0.2 meV) Raman-scattered peak is well resolved in all spectra, with PL being suppressed by factors of ~100 for biases less than 1.2 V, due to the sweeping of photoexcited carriers from the depletion region. The integrated intensity of the resonant Raman peaks as a function of outgoing photon energy, the resonant Raman profile (RRP), is shown as points. For all biases, a peak in the RRP is seen corresponding to the energy of the lower polariton branch. A weaker peak corresponding to the upper branch is also observed for biases above 1.0 V. In addition to these two peaks, which have been observed previously,[8] significant scattering is observed at energies between the peaks, not resonant with any features in reflectivity. This is most clearly seen for biases 1.0–1.2 V, where a third peak is seen in the RRP, at an energy between the polariton branches.

The energies of all peaks in the RRP are plotted in Fig. 4(a) as a function of bias (open triangles for the outer two peaks and open circles for the central peak). The energies of the polariton modes determined from reflectivity are also plotted for comparison, with the area of each point proportional to the integrated area of the reflectivity dip. The upper and lower peaks in the RRP agree well in energy with those of the upper and lower polariton branches.[8]

In order to shed light on the origin on the central peak in the RRP, a two-level model for the exciton-cavity coupling is employed to calculate the energies of the uncoupled cavity and exciton modes.[2] These are calculated at each bias, together with the vacuum Rabi splitting (Δ_{VRS}), from the measured energies and relative intensities of the dips in reflectivity. The results are shown in Figs. 4(a)–(b). The energy of the uncoupled cavity mode [open squares, Fig. 4(a)] is found to be independent of field, as expected. The energy of the uncoupled exciton [filled squares, Fig. 4(a)] decreases with increasing field due to the quantum confined Stark effect, as does Δ_{VRS} [filled diamonds, Fig. 4(b)] due to the decrease of exciton oscillator strength.[20] The magnitude of the Stark shift in Fig. 4(a) and the variation of Δ_{VRS} are in good agreement with the results of solution of a Schrödinger equation for the quantum well (QW) excitons, shown by the smooth solid lines in Figs. 4(a) and 4(b).

The energy of the central maximum in the RRP [the open circles in Fig. 4(a)] is seen to agree closely with the energy of the uncoupled exciton mode, providing strong evidence that the central maximum arises from Raman scattering mediated by excitons that are not coupled to the photon modes of the cavity. Studies of Raman-scattering resonant with exciton states in QWs[10–14] have shown that such scattering is defect or disorder activated, with the most probable scattering arising from a fourth-order process: the incoming photon makes a virtual transition to the exciton continuum (in our case at $\theta = 36°$, corresponding to $k = 4.3\times10^4$ cm^{-1} within the light cone to the sample normal, $k < 7.4\times10^4$ cm^{-1}), the exciton is scattered elastically to higher k by disorder, an LO phonon is emitted returning the exciton to $k\sim0$ from where the scattered photon is emitted. The involvement of the elastic scattering step to $k > \sim10^6$ cm^{-1}, of the order of $1/a_B$,[21] where a_B is the exciton Bohr radius, enables strong coupling of excitons to LO phonons,[22] since for $k\sim0$ the Fröhlich exciton-LO (X-LO) coupling is very weak due to cancellation of the contribution of the electron and hole polarization terms to the X-LO coupling.[13,23] Disorder activation of the Raman process thus enables coupling to the *high k* exciton reservoir, to normally dark states lying outside the light cone to the sample normal ($k < 7.4\times10^4$ cm^{-1}). The disorder most likely arises from In fluctuations in the InGaAs QW's, which is known to provide the dominant contribution to the ~1 meV linewidth of the QW excitons. The key role of disorder in giving rise to resonant Rayleigh scattering in microcavities has also been demonstrated recently.[24]

RRP's peaking at the exciton energy, rather than at the energy of the polariton modes have been reported in Refs. 7, 8, 25 although in these cases the cavity dispersion curves were not determined separately, thus raising the possibility that the cavities were in the weak-coupling regime due to screening by photogenerated excitons. Such possibilities are excluded in the present case, since the dispersion curves are probed simultaneously with the Raman scattering. The two outer peaks in the RRP which coincide with the energies of the polariton dips may arise from polariton mediation as reported previously, with the higher-energy peak weaker than the lower-energy one as a result of more rapid dephasing to states at high *k*.[8] However, on the basis of the present results an interpretation of the outer two features as arising from filtering of the exciton Raman signal by the MC acting as a filter cannot be excluded. Similar comments may also apply to the results of Refs. 7–9. The central peak in the RRS occurs by transmission of the uncoupled exciton signal out of the sample in the region of ~95% reflectivity between the polariton modes.

In conclusion, clear evidence for uncoupled exciton states in MC's otherwise in the strong-coupling limit has been reported. It is noteworthy, however, that very similar MC's have been shown to exhibit polariton quasiparticle effects characteristic of the strong-coupling regime in stimulated scattering experiments.[3,4] Performance of similar Raman experiments in direct comparison with studies of quantum wells outside an MC may permit semiquantitative estimates of the density of uncoupled exciton states in MC's to be obtained.

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[1] C. Weisbuch, M. Nishioka, A. Ishikawa, and Y. Arakawa, Phys. Rev. Lett. **69**, 3314 (1992).

[2] For reviews see M. S. Skolnick, T. A. Fisher, and D. M. Whittaker, Semicond. Sci. Technol. **13**, 645 (1998); A. I. Tartakovskii *et al.*, Adv. Mater. **13**, 1725 (2001); M. S. Skolnick *et al.*, Mater. Sci. Eng. C **19**, 407 (2002).

[3] P. G. Savvidis, J. J. Baumberg, R. M. Stevenson, M. S. Skolnick, and J. S. Roberts, Phys. Rev. Lett. **84**, 1547 (2000).

[4] R. M. Stevenson, V. N. Astratov, M. S. Skolnick, M. Emam-Ismail, A. I. Tartakovskii, P. G. Savvidis, J. J. Baumberg, and J. S. Roberts, Phys. Rev. Lett. **85**, 3680 (2000).

[5] R. P. Stanley, R. Houdré, C. Weisbuch, U. Oesterle, and M. Ilegems, Phys. Rev. B **53**, 10 995 (1996).

[6] M. Emam-Ismail, V. N. Astratov, M. S. Skolnick, and J. S. Roberts, Phys. Rev. B **62**, 1552 (2000).

[7] A. Fainstein, B. Jusserand, and V. Thierry-Mieg, Phys. Rev. Lett. **78**, 1576 (1997).

[8] W. R. Tribe, D. Baxter, M. S. Skolnick, T. A. Fisher, D. J. Mowbray, and J. S. Roberts, Phys. Rev. B **56**, 12 429 (1997).

[9] A. Fainstein, B. Jusserand, and R. André, Phys. Rev. B **57**, R9439 (1998).

[10] W. Kauschke, A. K. Sood, M. Cardona, and K. Ploog, Phys. Rev. B **36**, 1612 (1987).

[11] C. Tejedor, J. M. Calleja, L. Brey, L. Viña, E. E. Mendez, W. I. Wang, M. Staines, and M. Cardona, Phys. Rev. B **36**, 6054 (1987).

[12] A. J. Shields, M. Cardona, R. Nötzel, and K. Ploog, Phys. Rev. B **46**, 10 490 (1992).

[13] A. J. Shields, C. Trallero-Giner, M. Cardona, H. T. Grahn, K. Ploog, V. A. Haisler, D. A. Tenne, N. T. Moshegov, and A. I. Toropov, Phys. Rev. B **46**, 6990 (1992).

[14] D. W. Peggs, P. E. Simmonds, M. S. Skolnick *et al.*, Superlattices Microstruct. **15**, 317 (1994).

[15] See, e.g., R. Butté, G. Delalleau, A. I. Tartakovskii, M. S. Skolnick, V. N. Astratov *et al.*, Phys. Rev. B **65**, 205310 (2002).

[16] In our previous work in Ref. 8, performed on a 1λ cavity with *p,n* Bragg mirrors but without control of electric field, residual built-in fields led to partial suppression of photoluminescence.

[17] D. A. B. Miller, D. S. Chemla, T. C. Damen, A. C. Gossard, W. Wiegmann, and T. H. Wood, Phys. Rev. B **32**, 1043 (1985).

[18] A. Fainstein, B. Jusserand, and V. Thierry-Mieg, Phys. Rev. Lett. **75**, 3764 (1995).

[19] Correcting for the strongly energy-dependent transmission factor for double resonance nevertheless leads to Raman cross sections very similar to those presented here.

[20] T. A. Fisher *et al.*, Phys. Rev. B **51**, 2600 (1995). In this reference resonance was maintained by temperature tuning, enabling Δ_{VRS} to be determined directly at each field.

[21] It is estimated in Ref. 13 that the maximum coupling occurs for in-plane wave vectors $4/a_B \sim 2\times10^6$ cm^{-1}.

[22] The dispersion in LO phonon energy over this range of *k* is ~0.1 meV, less than the experimental resolution, see, e.g., Fasol *et al.*, Phys. Rev. B **38**, 6056 (1988).

[23] A. J. Shields, M. Cardona, and K. Eberl, Phys. Rev. Lett. **72**, 412 (1994).

[24] T. Freixanet, B. Sermage, J. Bloch, J. Y. Marzin, and R. Planel, Phys. Rev. B **60**, R8509 (1999); R. Houdré, C. Weisbuch, R. P. Stanley, U. Oesterle, and M. Ilegems, *ibid.* **61**, R13 333 (2000); D. M. Whittaker, *ibid.* **61**, R2433 (2000).

[25] In Ref. 8 the peak at the exciton energy was observed only for incoming resonance and not for the outgoing process.