In- and out-going resonant Raman scattering from the cavity polaritons of semiconductor quantum microcavities

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We present a resonant Raman scattering study of an on-resonance semiconductor quantum microcavity. Clear evidence for polariton mediation of the resonant Raman process in a microcavity is obtained. Two resonant peaks are observed in the out-going channel, arising from the upper and lower resonantly coupled exciton-photon (polariton) modes of the system. The resonant enhancement from the upper polariton state is much weaker than that of the lower state, due to the shorter dephasing time of the former. In the in-going channel, by contrast, only a single resonant peak is observed. We interpret this as arising from the much larger carrier density created by the photoexcitation, which for the in-going case is resonant in energy with the polariton states. The high carrier density leads to screening of the excitons, and decoupling of the excitonic and photonic states. [S0163-1829(97)02644-1]

There is considerable contemporary interest in the physics of semiconductor quantum microcavities (QMC's) resulting from the ability to control both the electronic and photonic states in the same structure.[1–5] In QMC's the photonic states along the growth direction (z) are defined by a Fabry-Pérot resonator [formed from distributed Bragg reflectors (DBR's)], with quantum wells (QW's) embedded within the cavity to determine the lowest-energy excitonic behavior. Both exciton and photon states are quantized along z, with a quasi-continuum of wave-vector states occurring for photons and excitons within the planes of the QMC and QW's, respectively. The structures are designed to produce degeneracy between the excitonic and photonic states.[1–5] In the resonance regime, the exciton and photon states are strongly coupled for each in-plane wave vector ($k_{\|}$) (the vacuum-Rabi coupling), leading to the formation of so-called cavity polaritons.[1,3] On resonance, the upper and lower polariton states contain equal components of the exciton and photon states of the QW and cavity, respectively.

In this paper we present clear evidence for polariton mediation of the resonant Raman scattering (RRS) process in a QMC. Unlike a number of optical techniques used previously to study QMC's [such as reflectivity[1,2,4,5] and photoluminescence (PL) (Ref. 3)], resonant Raman scattering, the subject of the present work, is a coherent technique. It is sensitive to the damping of the resonant state, and as we show can be used to obtain information about polariton dephasing in a QMC. We observe RRS for both the out-going and in-going channels where the scattered and laser photons, respectively, are in resonance with the polaritons.

Raman scattering from semiconductors, particularly QW's, is conventionally considered as a third-order process in which a photon is absorbed by the crystal, an exciton is created, the exciton is scattered with the creation or annihilation of a phonon, and finally the exciton is annihilated, leading to photon emission.[6] The three interactions (two exciton-photon and one exciton-phonon) are treated within third-order perturbation theory, the dominant contribution to the Raman scattering efficiency under near-resonant conditions being given by

$$W \propto \sum_{u,v} \left| \frac{\langle 0|H_{\text{photon}}|v\rangle\langle v|H_{\text{phonon}}|u\rangle\langle u|H_{\text{photon}}|0\rangle}{(E_v-\hbar\omega_s+i\Gamma_v)(E_u-\hbar\omega_l+i\Gamma_u)} \right|^2 . \quad (1)$$

$|0\rangle$ is the ground state of the crystal, $|u\rangle$ and $|v\rangle$ are intermediate excitonic states, of energies E_u and E_v and homogeneous linewidths Γ_u and Γ_v, H_{photon} is the exciton-photon Hamiltonian, and H_{phonon} is the exciton-phonon Hamiltonian. $\hbar\omega_l$ and $\hbar\omega_s$ are the energies of the incident and scattered photons, respectively, which are separated by one phonon energy. Equation (1) exhibits resonances when $\hbar\omega_l = E_u$ or $\hbar\omega_s = E_v$ (in-going or out-going resonance), with resonant Raman efficiencies proportional to either Γ_u^{-2} or Γ_v^{-2} in the two cases.

However, in the case where the excitons and photons are strongly coupled to form polaritons, the use of perturbation theory to describe the exciton-photon interaction is no longer soundly based. Instead, in the strong-coupling limit, as discussed previously for bulk semiconductors in the late 1960s and 1970s,[7–9] the Raman process should instead be described as a first-order scattering process between the initial and final polariton states (ψ_i and ψ_f, respectively), with the Raman efficiency given by

$$W \propto \frac{T_i T_f}{v_g} \langle \psi_i | H^*_{\text{photon}} | \psi_f \rangle \rho_f , \quad (2)$$

where T_i is a transmission coefficient that describes the transformation of an external photon into an internal polariton and T_f describes the reverse process, H^*_{phonon} is the polariton-phonon Hamiltonian that couples the exciton fraction of the polariton mode to the phonons, v_g is the group velocity of the initial state polaritons, and ρ_f is the density of

polariton final states. The above treatment, although having a sounder physical basis than the third-order perturbation theory,[10] nevertheless has its limitations. In particular it treats the polaritons as eigenstates of the system (stationary states) that do not decay into other states, making the rigorous inclusion of dephasing or damping difficult. Although we present strong evidence for polariton mediation of outgoing RRS, where the scattered photon is in resonance with the polariton states, we find that the Raman cross sections are very different for the two polariton modes (since the dephasing is very different), a result that is not explicable within the first-order scattering theory of Eq. (2). For the in-going Raman channel we find no evidence for polariton, as opposed to exciton, mediation of RRS, probably because of the higher laser intensities in this case, which lead to screening of the exciton-photon interaction.

The structure studied was grown by metalorganic chemical vapor deposition, and consisted of a λ GaAs cavity, containing three 100-Å $In_{0.13}Ga_{0.87}As$ quantum wells.[5] The well width was designed to match the wavelength of the lowest-energy excitonic state with the cavity length at the experimental temperature of 1.7 K. The wells were positioned centrally at the antinode of the cavity photon field. The cavity was defined by two DBR's, each consisting of 20 periods of λ/4 AlAs and $Al_{0.13}Ga_{0.87}As$. These were doped *n* and *p* type, subjecting the cavity to a small in-built electric field of $\sim 10^4$ V cm^{-1}. Such small fields have only a limited influence on Raman scattering,[11] but were found to lead to significant quenching of the PL, by over one order of magnitude compared to PL intensities measured from contacted samples biased to flat band conditions. The relative quenching of the PL facilitates greatly the study of out-going RRS, where the Raman signals are degenerate with the PL. Resonant Raman, PL, and reflectivity measurements were performed with a tunable Ti-sapphire laser, optically pumped by an Ar^+ laser. The reflectivity was measured with a Si photodiode, while the other optical spectra were detected using a triple spectrometer and charge-coupled device camera. In all cases a backscattering geometry at normal incidence was employed, with an incident laser power density of approximately 100 W cm^{-2} for the Raman measurements.

Although the sample was not intentionally inhomogeneous, the small variation in growth parameters across the wafer led to a variation in the exciton-photon detuning, predominantly due to the change in energy of the cavity photon mode. Consequently, we were able to select a region for which resonance occurred very close to normal incidence, corresponding to zero in-plane $k_\parallel$. The on-resonance conditions for the region of the sample studied are confirmed by the lowest power reflectivity spectrum of Fig. 1(a), at 0.32 W cm^{-2}, where two reflectivity dips of near equal amplitude from the two $k_\parallel = 0$ polariton states are observed.[1,4,5]

In Figs. 2(a) and 2(b) we present a series of Raman spectra for the out-going and in-going resonant channels, respectively. In both cases a sharp Raman peak, 36.5 meV from the laser line, appears on the PL background. For in-going resonance the PL arises from an LO phonon satellite of selectively excited PL, and for the out-going resonance it arises from emission from the lower and upper polariton states, with peak separation of approximately 6 meV, the same as observed in reflectivity in Fig. 1(a). Figure 2 also shows the

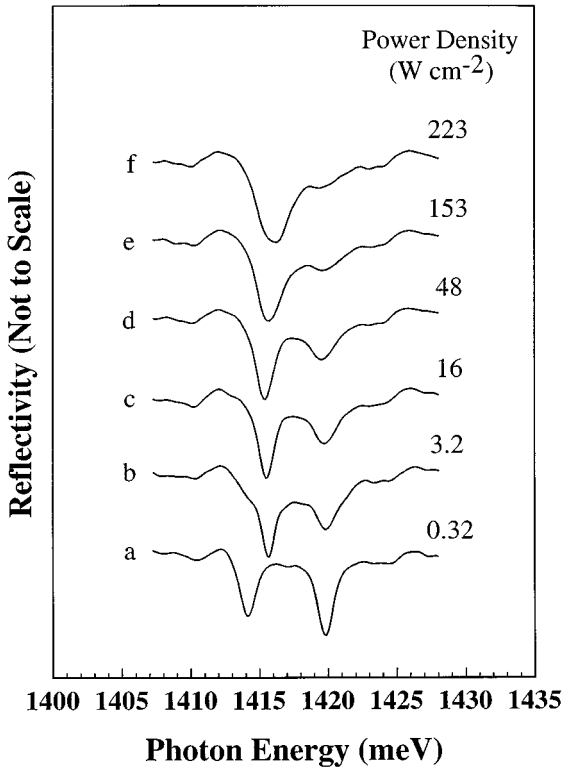

FIG. 1. Power-dependent reflectivity measurements. The power densities are approximate, with an estimated ±20% error. With increasing power the vacuum Rabi splitting between the two polariton branches decreases, until for powers greater than ~100 W cm^{-2} only one peak is visible, due to exciton screening at high power density.

resonant Raman profiles (RRP's), where the intensity of the Raman peak is plotted as a function of the scattered photon energy. The horizontal scales for the in-going and out-going measurements are offset by exactly one LO phonon energy (36.5 meV) so that they can be compared directly. In a previous report of Raman scattering from a QMC (Ref. 12) a number of phonons were observed at energies below 36.5 meV due to modes with nonzero in-plane wave vector. In the present experimental geometry $k_\parallel = 0$, so that only the zone center GaAs-like LO vibration is observed.[13]

Considering first the measurements in the outgoing channel, the laser is tuned to one phonon energy above the polariton energies. Resonant enhancement of the RRP is observed at the energies of both the upper and lower polaritons, but with the resonance of the upper state a factor of ~9 weaker than that of the lower state.[14] The observation of such a two-peak structure in the RRP, with the same peak energies as observed in the low power reflectivity spectrum of Fig. 1 and in the PL spectra of Fig. 2(a), immediately provides evidence for the involvement of polariton properties in the Raman process. However, since the out-going transmission factor T_f of Eq. (2) will exhibit two peaks in the polariton region, corresponding to the two polariton dips seen in reflectivity, it is difficult to conclude whether the probability for Raman scattering within the QMC, given by the $\langle \psi_i | H^*_{\text{phonon}} | \psi_f \rangle \rho_f$ term of Eq. (2), is also two peaked, as would be expected for polariton mediation of RRS. However, the strongly differing intensities of the Raman cross

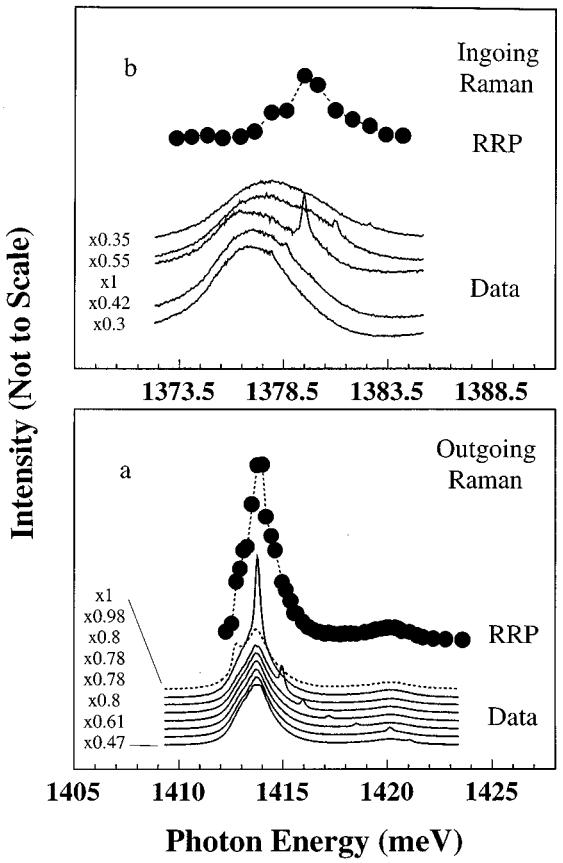

FIG. 2. Individual Raman measurements, and the resonant Raman profiles (RRP's) for both the out-going (a) and in-going (b) resonances. In both cases the sharp Raman peaks are superimposed on a PL background. Both RRP's are plotted as a function of the scattered photon energy. However, the horizontal scales for the two cases are rigidly shifted by the LO phonon energy of 36.5 meV to allow direct comparison. The individual Raman measurements have been scaled for clarity of presentation.

sections at the energies of the two polariton modes does provide conclusive evidence for polariton mediation of the Raman process. In reflectivity the two modes have equal strength and thus the T_f factor must be the same for the two modes. Hence if the resonant Raman process involved only uncoupled excitonic states and was simply modulated by the transmission factor, two peaks of equal strength would be expected in the RRP.

The much weaker intensity of the upper branch in the RRP instead finds a natural explanation if polariton mediation of the Raman process within the QMC is important. We ascribe the weak Raman cross section for the upper branch relative to that for the lower branch to the more rapid dephasing of polaritons in the upper branch compared to those in the lower branch. Raman scattering being a coherent process is sensitive to the dephasing (scattering) times of polaritons in the final or initial states. Such dephasing leads to loss of coherence and reduction of the Raman signal. The dephasing time is expected to be shorter for the upper branch since it is resonant with higher bound and continuum excitonic states, as well as with higher $k_\parallel$ states from the lower branch, thus permitting disorder-induced elastic scattering processes for the upper branch. By contrast such scattering cannot occur for the near $k_\parallel = 0$ lower branch polaritons. However, as remarked on earlier, such dephasing or damping processes cannot be included rigorously within the first-order treatment of Eq. (2). It is worth commenting that the third-order treatment of Eq. (1) does include initial- and final-state damping, although at the expense of treating the excitons and photons as decoupled, which as discussed above is unjustified in the strong-coupling regime; the homogeneous linewidths $\Gamma_{u,v}$ in the denominator are inversely proportional to the dephasing time, with a short dephasing time corresponding to a small Raman cross section for the upper branch.

Clearly a complete theory of RRS for cavity polaritons, including the effects of final state dephasing, is required in order to account fully for the present results.[15] However, in the absence of such a description, we emphasize that a polariton description including final-state dephasing does capture the essential physics of the process, and accounts qualitatively for the relative intensities of the two features in the RRP. We note also that the influence of disorder-induced scattering, predominantly from the upper polariton branch to excitonic excited or continuum states,[2] or to finite $k_\parallel$ lower branch polariton states[16] has been proposed in the literature to account for the larger linewidth of the on-resonance reflectivity dip for the upper relative to the lower polariton state.[2] Our coherent Raman measurements support this conclusion.

Turning now to the in-going resonance, the most notable feature of Fig. 2(b) is that the in-going RRP displays only a single peak, which after correction for the phonon energy lies between the two peaks observed in the out-going case. The observation of the single peak is unexpected since the mechanisms for the in- and out-going resonances are expected to be closely related.

We believe that the difference between the Raman profiles in the two cases arises from the very different transmission factors of the cavity for incident photons in the in-going and out-going resonant conditions. For the out-going resonance, the incident photons are at an energy for which the cavity reflectivity is extremely high ($\sim$99.5%; see Ref. 17), so that only a small fraction of the incident photons enter the cavity, resulting in a very small photoexcited carrier density ($<10^9$ cm^{-2} for a polariton lifetime of 100 ps). For in-going resonance, however, the incident photon energy corresponds to allowed optical states of the cavity (with an experimentally measured transmission of around 50%), leading to a much larger photoexcited carrier density ($\sim 10^{11}$ cm^{-2}—see below). Houdré *et al.*[18] have reported PL measurements that demonstrate that at carrier densities sufficiently high to screen the excitons, the exciton-photon interaction is significantly reduced. This reduction occurs because the interaction, expressed in terms of the vacuum-Rabi splitting between the polariton branches, is proportional to $(f_{\rm osc})^{1/2}$, where $f_{\rm osc}$ is the exciton oscillator strength.[1,5,18] Beyond a saturation density (estimated in Ref. 18 to be 4×10^{10} cm^{-2}), exciton screening, and the consequent sharp reduction in $f_{\rm osc}$, leads to decoupled excitons and photons, characterized by a crossing (as opposed to an anticrossing) between the exciton and photon dispersion curves.

For the outgoing resonance condition, the reflectivity curve of Fig. 1(a) (measured under similar excitation

conditions[17]), and the two-peaked PL background of Figure 2(a), confirm that the excitons and photons are strongly coupled, as expected at the low carrier density of $<10^9$ cm^{-2}. For in-going conditions, however, the carrier density exceeds the saturation value, and the resultant crossing between the excitons and photons produces only a single resonant peak, at an energy between the two in the out-going case. Assuming a polariton lifetime of 100 ps,[19] a carrier density for the in-going case of 10^{11} cm^{-2} is estimated, consistent with this explanation. Further support for this interpretation is obtained from the power-dependent reflectivity measurements of Figs. 1(b)–1(f). If the excitons and photons decouple, a transition from the distinctive two dip reflectivity at low laser powers to a single dip at an intermediate energy at high laser powers is expected. This behavior is seen clearly in Figs. 1(c)–1(f), with a transition to a single dominant peak occurring for power densities between 48 and 153 W cm^{-2}, comparable to that used for the ingoing Raman measurements. Furthermore the energy of the single dip correlates closely with the energy of the ingoing RRP peak, further substantiating our interpretation of the form of the RRP.

In conclusion, we have performed resonant Raman measurements on an on-resonance semiconductor QMC, and have obtained clear evidence for polariton mediation of the Raman process in the out-going channel. The weak intensity of the upper polariton branch in the coherent Raman measurements provides clear evidence for more rapid polariton dephasing in the upper state. In the in-going channel, only a single resonance peak is observed. This behavior is attributed to screening of the excitons due to the larger photoexcited carrier density for ingoing resonance.

Finally we note that since this paper was submitted, another report of RRS from cavity polaritons has been published.[20] Measurements in the in-going channel only were reported with only one resonance feature being observed in the Raman profile, in agreement with the present work. This was tentatively attributed to possible contributions from weakly coupled excitons, damping, or $k_\parallel$ partial nonconservation.

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[9] Polariton mediation of resonant light scattering is also reviewed by C. Weisbuch and R. G. Ulbrich, in *Light Scattering in Solids*, edited by M. Cardona and G. Guntherodt (Springer, Berlin, 1982), Vol. 3, p. 207.

[10] In semiconductor QW's exciton decay into a one-dimensional continuum of photon states can occur due to the lack of translational symmetry along the growth direction (z) [see, e.g., L. C. Andreani, F. Tassone, and F. Bassani, Solid State Commun. **77**, 641 (1991)]. As a result the coupled mode polaritons are no longer the eigenstates of the system and the third-order perturbation treatment is applicable. For a QMC, the photon states are also quantized along z and the polariton picture is restored.

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[13] For $In_{0.13}Ga_{0.87}As$ grown on GaAs, the redshift of the GaAs-like LO phonon energy due to alloying is approximately balanced by the blueshift due to strain [see F. Iikawa *et al.*, Solid State Commun. **68**, 211 (1988)]. Therefore the GaAs-like LO phonon is not confined to the quantum wells. As the polariton states also extend throughout the cavity, the Raman scattering does not take place solely in the quantum wells, though it is still dominated by scattering near the center of the cavity due to the maximum in the photon field.

[14] For different positions on the sample, all of which correspond to exciton-photon resonance in reflectivity, we see a variation of the ratio of the Raman intensities of the lower to upper polariton branches from 4 to >10. This is very likely related to variation of sample disorder, and hence polariton scattering rates with position.

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