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Annular resonant Rayleigh scattering in the picosecond dynamics of cavity polaritons

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We study the angular dependence of the emission from cavity polaritons resonantly excited by a picosecond laser pulse. We observe that, in a first stage, the initial excitation is rapidly redistributed by elastic scattering along a well-defined ring in the wave-vector space which results in an angular-dependent emission. This initial transfer, which conserves the polarization, is attributed mainly to resonant Rayleigh scattering of polaritons. We also study the width of this ring and show that it is detuning dependent, reflecting the energy dispersion of the polaritons. At longer delay, the emission is found to be isotropic and depolarized, in agreement with previous studies. [S0163-1829(99)52636-2]

For many years, resonant Rayleigh scattering (RRS) has been a subject of research in semiconductor physics. As evidenced in GaAs/$Al_xGa_{1-x}As$ quantum wells (QW's), when excitons are resonantly excited by a laser, it was interpreted as a consequence of the spatially fluctuating excitonic dielectric constant.[1] These fluctuations arise from the static disorder along the QW plane leading to the spatial variation of the resonance energy. The in-plane translation invariance breaking relaxes the in-plane wave vector $\mathbf{k}_\parallel$ conservation which governs the optical transitions in perfect systems. As a consequence, a resonantly excited exciton with a wave vector $\mathbf{k}_\parallel$ can emit in other directions than just the reflected and transmitted ones.

It was shown recently that, at low excitation density, RRS is the dominant process in generating the emission from resonantly excited excitons.[2,3] In this regime the excitonic polarization is elastically scattered by the disorder. This scattering mechanism preserves the phase coherence[3,4] and results in a quadratic rise of the emission in time.[2] On the other hand, for longer time delays the emission is usually termed photoluminescence (PL) and its decay time is governed by inelastic scatterings (such as exciton-phonon and exciton-exciton scatterings) and radiative processes.[5,6] In microcavities, new features have been evidenced due to the photon confinement.[7,8] In these structures, the photon and exciton states are both two dimensional. As a consequence, an exciton with an in-plane wave vector $\mathbf{k}_\parallel$ is coupled to one photon mode $(\mathbf{k}_\parallel, k_z)$, the perpendicular component k_z being fixed by the cavity length. These mixed exciton-photon modes or cavity polaritons are the eigenstates of the system. It is in this framework that Hayes *et al.* interpreted their femtosecond experiments in the Rayleigh scattering regime: the rise time of the RRS emission was found detuning dependent, reflecting the mixed state character of the polariton.[9]

On the other hand, time resolved PL experiments have been performed on polaritons resonantly excited by a picosecond laser pulse.[10] It was shown that, by tuning the energy cavity mode with respect to the exciton one, the first PL decay time of the lower polariton branch changes continuously from a pure photon lifetime at negative detuning (photonlike polariton) to a nonradiative bare exciton lifetime at positive detuning (excitonlike polariton). This behavior was qualitatively well described by a two level model. However, to understand more deeply the polariton dynamics, a rate equation model was then evolved, taking into account the polariton dispersion and scatterings with acoustic phonons.[11] The main assumption of this model concerns the initial stage of the dynamics. It is assumed that the polaritons excited with a well defined $\mathbf{k}_\parallel$ wave vector are rapidly elastically scattered in all directions with the same modulus $k_\parallel$. As a consequence, the populations at later times only depend on $k_\parallel$.

The purpose of this work is to check experimentally this hypothesis by measuring the $\mathbf{k}_\parallel$ dependence of the emission. Before describing our experimental results we explain briefly why such an hypothesis is reasonable. Figure 1 shows a schematic representation of the scattering process occurring after the resonant excitation of polaritons. Elastic and inelastic scatterings occur on two different time scales. The first one is fast[2,12] and not measurable within our time resolution whereas the second is in the range of 15–20 ps at low density.[10] Therefore the polaritons initially excited in a well defined $\mathbf{k}_\parallel$ state should be elastically scattered along a ring of radius $k_\parallel$ before being scattered outside, namely, in states of different energies. So we expect to observe, in the first stage, an intense emission for this ring in $\mathbf{k}_\parallel$ space, corresponding to the RRS part of the emission.

The studied sample is a wedge-shaped GaAs λ-microcavity surrounded by two distributed Bragg reflectors. Thus, scanning the exciting spot on the sample surface allows us to vary the detuning $\delta = E_{\mathrm{cav}} - E_{\mathrm{ex}}$ between the cavity mode and the exciton energy. The front (respectively, back) mirror consists of 17 (respectively, 27) periods of $Al_{0.1}Ga_{0.9}As$/AlAs $\lambda/4$ layers. Embedded in the center of the

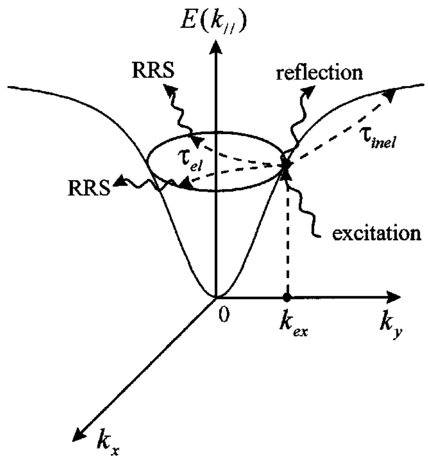

FIG. 1. Schematic representation of the elastic and inelastic scattering processes occurring after the resonant excitation of polaritons. τ_{el} (respectively, τ_{inel}) is an elastic scattering time (respectively, inelastic).

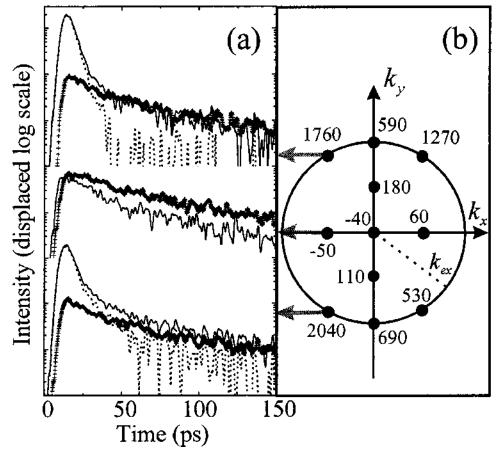

FIG. 2. (a) $\mathbf{I}_{\parallel}$, $\mathbf{I}_{\perp}$, and $\mathbf{I}_{\parallel}-\mathbf{I}_{\perp}$ normalized luminescence intensities for three different in-plane wave vectors $k_{\parallel}$. Full lines: $\mathbf{I}_{\parallel}$. Cross symbols: $\mathbf{I}_{\perp}$. Dotted lines: $\mathbf{I}_{\parallel}-\mathbf{I}_{\perp}$. The upper and lower spectra correspond to wave vectors belonging to the ring of radius k_{ex}. The middle spectra wave vector is $(-k_{\mathrm{ex}}/2,0)$. (b) Cartography of the $\mathbf{I}_{\parallel}-\mathbf{I}_{\perp}$ time integrated value in the $k_{\parallel}$ space. The in-plane excitation wave vector is $(k_{\mathrm{ex}},0)$ and the solid circles represent the detected wave vectors.

cavity is a single 8-nm $\mathrm{In}_{0.05}\mathrm{Ga}_{0.95}\mathrm{As}$ quantum well. We have determined from cw luminescence measurements a Rabi splitting of 3.4 meV.

The sample was maintained at 10 K by means of a closed-cycle helium refrigerator and excited by a mode-locked Ti-doped sapphire laser giving nearly Fourier-transform limited pulse in the range of 1–1.5 ps with a repetition rate of 82 MHz. The laser beam was incident on the sample at an angle of 8° with a TE polarization and focused on a 60-μm-diam spot. Its energy was tuned to the energy of the lower polariton branch so that, with this small external angle, we resonantly excited linearly polarized polaritons with an in-plane wave vector $k_{\mathrm{ex}}=10^4\,\mathrm{cm}^{-1}$. The emission emerging within a maximum angle of 11° from the sample normal is spectrally dispersed using a 32-cm monochromator. Next, temporal analysis is performed by a synchroscan streak camera. Finally, the signal is detected using a charge coupled device. The time resolution lies around 2.5 ps.

The angular analysis of the emission was done by means of an iris diaphragm which can be precisely moved in a parallel beam section of the detection path. To keep a good signal/noise ratio the diaphragm aperture was fixed to 2.5 mm corresponding to a range of $2.5\times10^3\,\mathrm{cm}^{-1}$ in the in-plane wave-vector amplitude. We detected the emission through a Glan-Thompson polarizer and record $\mathbf{I}_{\parallel}$ (respectively, $\mathbf{I}_{\perp}$), the emission polarized parallel to the incident beam (respectively, perpendicular). Special care was taken to reduce the zero-point jitter along the time scale. In each measurement a directly detected laser pulse served as a common time reference. In order to minimize polariton-polariton scattering effects,[9,13] the density was kept around $10^9\,\mathrm{cm}^{-2}$ which is the minimum density we can study in this experimental configuration.

We have performed the emission cartography in $\mathbf{k}_{\parallel}$ space for both polarizations. We define by (k_x,k_y) the in-plane coordinates of $\mathbf{k}_{\parallel}$, deduced from the diaphragm position. Initially the polaritons are excited in the state $(k_{\mathrm{ex}},0)$ with a TE polarization [Fig. 2(b)]. For example, Fig. 2(a) shows the temporal evolution of $\mathbf{I}_{\parallel}$, $\mathbf{I}_{\perp}$ and $\mathbf{I}_{\parallel}-\mathbf{I}_{\perp}$ at resonance ($\delta=0$) for three wave vectors $\mathbf{k}_{\parallel}$. Two of them belong to the ring of radius k_{ex} and are symmetrical with respect to the axis k_x, the other lying out of the ring. The striking difference between these spectra is the presence of an intense initial polarized spike from polaritons in ring states. During this early time the emission is polarized to more than 85% and the $\mathbf{I}_{\parallel}-\mathbf{I}_{\perp}$ decay time is about 2.7–3.5 ps. In a second stage, approximately 40 ps after the laser pulse, the emission is found to be depolarized (compare the $\mathbf{I}_{\parallel}$ and $\mathbf{I}_{\perp}$ intensity) in agreement with a previous study.[6] The initial spike of polarized emission is not observed for polaritons in the state $(-k_{\mathrm{ex}}/2,0)$ which is out of the ring. In this case the $\mathbf{I}_{\parallel}$ and $\mathbf{I}_{\perp}$ intensities have approximately the same temporal shape and the early time emission is depolarized. Moreover, the temporal evolution of $\mathbf{I}_{\perp}$ is approximately the same as in the previous cases and looks independent of the wave-vector modulus.

We can assume that the RRS emission contributes only to $\mathbf{I}_{\parallel}-\mathbf{I}_{\perp}$.[14] However, depending on the exciton spin relaxation time,[15] inelastic processes generating PL could also contribute to this polarized signal. Nevertheless, RRS is the fastest process and should dominate at the early time. Therefore, we will focus in the following on $\mathbf{I}_{\parallel}-\mathbf{I}_{\perp}$ which, under these assumptions, should be mainly due to RRS.

We have performed these measurements for several other $\mathbf{k}_{\parallel}$. Figure 2(b) gives some values of the time integrated intensity of $\mathbf{I}_{\parallel}-\mathbf{I}_{\perp}$ as a function of (k_x,k_y). The emission coming out from polaritons in $\mathbf{k}_{\parallel}$ states well out of the ring is depolarized and gives a negligible contribution to the overall polarization. Therefore, the important feature coming out of these results is that only the early time dynamics of $\mathbf{I}_{\parallel}$ is strongly wave-vector dependent and that $\mathbf{I}_{\parallel}-\mathbf{I}_{\perp}$ is maximum for polaritons belonging to the ring of radius k_{ex}. We checked that in all cases the residual scattered light on the sample surface was negligible compared to the resonant emission (the off resonance scattered intensity is several orders of magnitude smaller than the on resonance one).

Thus, the polariton dynamics appears to be governed by two kinds of scattering occurring in two different time scales. In the early times, polaritons are mostly scattered elastically as attested by the fast redistribution in the ring states. This process conserves both the wave-vector modulus and the polarization and no energy relaxation occurs since all the ring states are at the same energy. Most of these polaritons escape the cavity (due to their finite radiative lifetime) before being inelastically scattered. Thus, during this time, the emission is essentially due to RRS. As we detect wave vectors out of the ring, the emission changes progressively from RRS to PL. Indeed, due to the strong polariton dispersion these states are nonresonant with the excitation one and can only be populated by inelastic processes. This can explain the depolarization.

The RRS contribution to the emission disappears very rapidly as evidenced by the fast decay time of $\mathbf{I}_{\parallel}-\mathbf{I}_{\perp}$ [Fig. 2(a)]. We have found experimentally that it strongly depends on the polariton density. When the density is raised, the decay time of $\mathbf{I}_{\parallel}-\mathbf{I}_{\perp}$ shortens and the RRS contribution tends to disappear. Indeed, the lowest density allowing this angular analysis ($\approx 10^9\,\mathrm{cm}^{-2}$) is close to the onset of polariton-polariton scattering evidenced in Ref. 9. Therefore, in the range of density used in these experiments, this decay time results probably from a fast dephasing process due to this scattering. Recent studies have already shown its important contribution to the dynamics of resonantly excited excitons[16,17] but, to our knowledge, none has been performed in microcavities.

We have performed the same angular- and time-resolved measurements for several detunings. Figure 3(a) shows the $\mathbf{I}_{\parallel}-\mathbf{I}_{\perp}$ time integrated intensity for various states $(0,k_{\parallel})$, lying on a diameter ring, and for three different detunings. As previously, the laser creates polaritons in the state $(k_{\mathrm{ex}},0)$. In all cases, the RRS emission is maximum for the states $(0,\pm k_{\mathrm{ex}})$ belonging to the ring. This emission, as well as the intensity contrast of RRS over PL, depends on the detuning and is much more intense for a photonlike polariton ($\delta<0$) than for an excitonlike polariton ($\delta>0$). This feature can be explained by the mixed state character of the polariton. When the polaritons are photonlike, the escape time decreases and inelastic processes are less efficient because they arise from the exciton part of the polariton.[11,13] Both features enhance the RRS intensity with respect to PL.

We now focus on the $k_{\parallel}$ width of the elastic ring and on its detuning dependence. From Fig. 3(a) we define $\Delta k_{\parallel}$ as the half width at half maximum of the $\mathbf{I}_{\parallel}-\mathbf{I}_{\perp}$ time integrated intensity, which represents the RRS part of the overall emission. Despite our poor angular resolution a clear tendency appears. As the polariton becomes photonlike the ring becomes more intense and well defined in the $\mathbf{k}_{\parallel}$ space with a strong decrease of $\Delta k_{\parallel}$. On the other hand, for positive detuning, $\Delta k_{\parallel}$ increases rapidly and the RRS emission becomes less wave-vector dependent.

In Fig. 3(b), we have plotted $\Delta k_{\parallel}$ as a function of $(dE/dk_{\parallel})^{-1}$, the inverse slope of the polariton dispersion curve calculated at $k_{\parallel}=k_{\mathrm{ex}}$, to reveal the relationship between the angular width of the RRS emission and the polariton homogeneous energy broadening Γ. In fact, this finite energy broadening allows the nonconservation of the initial excitation wave vector k_{ex} in the RRS process. To illustrate this idea, Fig. 3(c) shows two calculated dispersion curves of the lower polariton branch for $\delta=-2.7$ and $+1.6$. Then it is

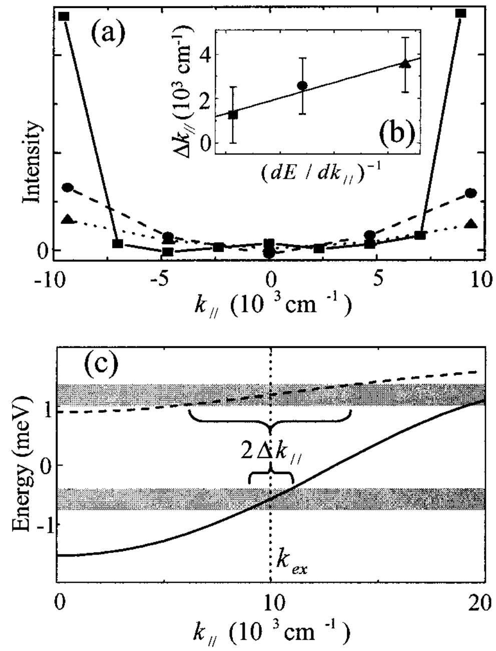

FIG. 3. (a) $\mathbf{I}_{\parallel}-\mathbf{I}_{\perp}$ time integrated intensity as a function of $k_{\parallel}$ for the states $(0,k_{\parallel})$ and for three detunings. (■): $\delta=-2.7$. (●): $\delta=0$. (▲): $\delta=1.6$. (b) The half width $\Delta k_{\parallel}$ of the former distribution as a function of $(dE/dk_{\parallel})^{-1}$ calculated at $k_{\parallel}=k_{\mathrm{ex}}$. The linear fit (full line) gives a homogeneous linewidth of $\Gamma=0.28$ meV. (c) Polariton dispersion curves for $\delta=-2.7$ (full line) and $\delta=1.6$ (dotted line). The shadowed area defines the Γ-broadened polariton region around $k_{\parallel}=k_{\mathrm{ex}}$ where the elastic processes take place.

clear that, assuming a constant Γ, the angular width of the RRS emission is mainly determined by the inverse slope of the dispersion curve. Indeed, the linear fit in Fig. 3(b) has been obtained with $\Gamma=0.28$ meV. This is consistent with our measurements of the RRS decay times (2.7–3.5 ps) since the RRS emission is expected to decay with a time proportional to the inverse of the full homogeneous polariton linewidth.[9,14] Introducing the group velocity $v_g=(dE/dk_{\parallel})/\hbar$ and the lifetime $\tau=\hbar/\Gamma$, then the angular width of the RRS emission appears to be simply related to the polariton mean free path l by

$$2\Delta k_{\parallel}=1/(v_g\tau)=1/l, \tag{1}$$

the numerical factor 2 arising from our definition of $\Delta k_{\parallel}$.

Finally, let us underline that this annular emission is also expected classically for the transmission of monochromatic light through a Fabry-Perot resonator[18] with a diffusive plate set along the optical axis. Light will be scattered in all directions, but only light in the direction resonant to the cavity mode will be transmitted through the cavity. The resulting ring in the transmission has to be attributed to the cavity mode dispersion. In the case of strong coupling with excitons, the specific effect is that the states probed by the scattering processes are the polariton states instead of the bare cavity modes.

In conclusion, we have shown experimentally the existence of an annular distribution of the polariton emission in the early times of their dynamics. This emission is strongly polarized parallel to the laser and arises mainly from polariton states belonging to a ring whose radius is the modulus of the in-plane excitation wave vector. In addition the angular width of this emission strongly depends on the detuning in a way which can be related to the polariton mean free path. All this gives us strong arguments to attribute this emission to polariton mediated RRS. On the other hand, the emission at long delays is unpolarized and weakly depends on the in-plane wave vector: it is attributed to incoherent PL. Therefore, due to their strong in-plane dispersion, RRS and incoherent PL emission from cavity polaritons can be angularly well discriminated. To get more insight on the coherent nature of this emission, interferometric and four wave mixing experiments would be of great interest.

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