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Coherent control of exciton polaritons in a semiconductor microcavity

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We report on the dynamics of resonantly excited lower branch polariton states in a semiconductor quantum microcavity. We show that both the spin orientation and the density of polaritons can be coherently manipulated. The measurements of the optical dephasing time T_2 and the decay time T_1 of the radiant states as a function of the cavity detuning and the lattice temperature give further insights into the relaxation mechanisms of cavity polaritons. [S0163-1829(99)51104-1]

The optical properties of excitonic polaritons in semiconductor quantum microcavities have attracted great attention in recent years. Their potentialities for new all optical switching devices are particularly interesting.[1] The strong coupling between the exciton and the cavity mode has been studied with various techniques: reflectivity, absorption, photoluminescence, and four wave mixing (FWM) experiments.[2–5] The influence of the quantum well (QW) disorder on the optical properties, which is responsible for the *inhomogeneous broadening* of the spectral lines, has also been investigated. Due to the specific characteristics of the exciton-photon mixed states, i.e., low mass ($\sim 10^{-5} m_e$) and large size ($\sim 10^3$ nm), there is a significant averaging over the disorder potential and a subsequent ''motional narrowing'' of the spectral lines.[6,7] On the other hand, the *homogeneous broadening* (Γ_h) of the exciton-polariton states is of fundamental interest since it provides information on the interaction processes of the excited states,[8] such as elastic scattering by disorder induced potential, scattering by acoustical phonons, or mutual collision processes. The homogeneous broadening is usually determined in the time domain, through the measurement of the optical dephasing time T_2, both quantities being related by $\Gamma_h = 2\hbar/T_2$. Nonlinear coherent spectroscopy techniques such as FWM give access in principle to the optical dephasing time of excitons in bulk semiconductors or bare quantum wells (QW's) but also of exciton-polaritons in microcavity embedded QW's. Up to now, in this last case, the experiments have been performed with very short excitation pulses ($\sim$100 fs).[5] The corresponding spectral width ($\sim$20 meV), larger than the Rabi splitting Ω_R, leads to a coherent excitation of both the upper and lower branch mixed states, resulting in the observation of quantum beats. The optical dephasing time for an independently excited polariton branch cannot be measured in such conditions.

We have shown recently that the phase coherence loss of excitons in bare QW's can directly be monitored in a time-resolved secondary emission (SE) experiment using ps laser excitation.[9] In the present work, we apply this technique to the case of the exciton polaritons in microcavity embedded QW's. The ps experiment, characterized by a pulse spectral width smaller than Ω_R, allows us to excite *selectively* the lower branch (LB) polariton states. We show that the density, the alignment, and the spin orientation of microcavity polaritons can be coherently manipulated. As for excitons in bare QW's, the technique yields direct measurement of the optical dephasing time of the microcavity polariton. We measure both T_2 and the decay time T_1 of the radiant population as a function of the detuning $\delta \equiv E_c - E_e$, where E_c and E_e are the uncoupled cavity main mode and the exciton energies, respectively. Through the measurement of T_2 and T_1 as a function of the lattice temperature we show for zero cavity detuning, a quenching of the polariton scattering by acoustic phonons up to a temperature of 40 K. All these results are in agreement with theoretical predictions.[10,11]

The QW microcavity studied here is grown by molecular beam epitaxy and consists of four 12-nm $In_{0.14}Ga_{0.86}As$ wells located at the antinodes of the cavity photon mode. The top (bottom) Bragg mirror consists of 13 (22) pairs of GaAs and AlAs layers surrounding a $3\lambda/2$ GaAs microcavity. The sample is fabricated with a slight wedge, so that the effective thickness of the cavity L_c varies along the radial direction of the wafer. This allows us to tune the cavity resonance by moving the laser beam across the wafer. The Rabi splitting is equal to $\Omega_R = 6$ meV. The inhomogeneous broadening of exciton states and the cavity mode broadening, estimated at 12 K from cw-photoluminescence excitation (PLE) experiments performed, respectively, at large positive and negative cavity detuning, are $\Gamma_{\rm inh} \cong 3$ meV and $\Gamma_c \cong 0.4$ meV. At resonance ($\delta = 0$), the polariton broadening is $\Gamma = 1.4$ meV, which includes both homogeneous and inhomogeneous contributions.

We have performed coherent control experiments probed by time-resolved SE at $T = 12$ K. A sequence of two 1.5-ps laser pulses of defined polarizations (σ^1, σ^2) and equal intensities, produced by a Mach-Zender type interferometer, resonantly excites the heavy-hole lower branch (LB) polariton states at energy $E_{\rm LB}$. The temporal separation between the two pulses is controlled on two time scales: a coarse tuning sets the delay t_1 between the two pulses on a ps scale; a fine tuning adds the delay t_2 on a subfemtosecond scale. The exact delay between the two pulses is then $t_1 + t_2$ and it is convenient to calibrate the time scale so that t_1 is an exact multiple of $h/E_{\rm LB}$. The SE kinetics are recorded by using a two color up-conversion scheme with a synchronously pumped optical parametric oscillator (pulse width 1.4 ps).[9] The excitation beam is incident on the sample at an angle of

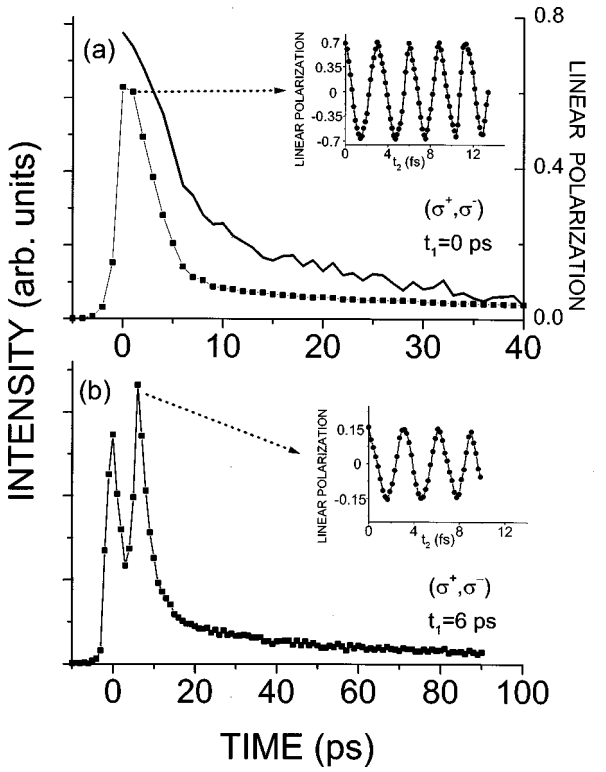

FIG. 1. The cavity detuning is $\delta=0$ meV; (a) Time evolution of the total SE (■) and the corresponding linear polarization (full line) after a (σ^+,σ^-) excitation sequence: $t_1=0$ ps and $t_2=0$. (b) Time evolution of the total SE for $t_1=6$ ps and $t_2=0$. Insets: Linear polarization P^1 as a function of the fine temporal separation t_2 between the two excitation pulses.

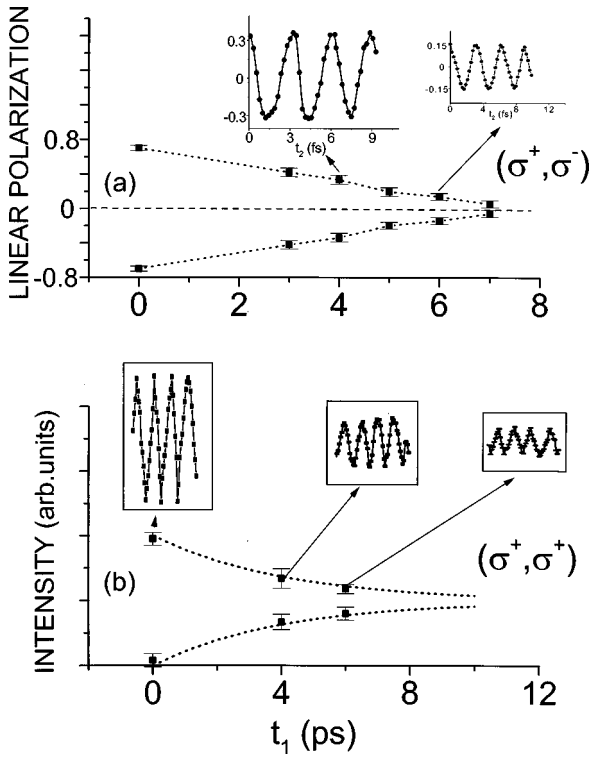

FIG. 2. The cavity detuning is $\delta=0$ meV; (a) The configuration is (σ^+,σ^-). The maxima and minima of the linear polarization oscillations as a function of t_1 (the dashed line is a guide for the eyes). (b) The configuration is (σ^+,σ^+). The maxima and minima of the total SE intensity oscillations for $t_1=0$, 4, and 6 ps. The period of the SE oscillations with respect to t_2 shown in the insets is $h/E_{\mathrm{LB}}\sim 3$ fs.

about 8° with respect to the growth direction (this corresponds to an in-plane wave vector $k_{\parallel,\mathrm{pump}}\sim 10^4\,\mathrm{cm}^{-1}$). The detection direction is set along the normal to the sample surface. The acceptance solid angle of the up-conversion system is about 10^{-3} steradians. All measurements are carried out for moderate excitation peak powers ($\sim$20 kW cm^{-2}). We have checked that the SE dynamics and the optical dephasing times are intensity independent in the range 5–80 kW cm^{-2}.

We consider a sequence of two optical pulses of opposite helicy (σ^+,σ^-) and excite resonantly the LB polariton for a cavity detuning $\delta=0$. First, the coarse delay between the two pulses is $t_1=0$ ps. The interference between the two laser pulses results in a linearly polarized optical excitation. Figure 1(a) displays the time dependence of the total SE and the corresponding linear polarization $P^1(t)=(I^X-I^Y)/(I^X+I^Y)$ where I^X and I^Y are the two linearly polarized SE components (we have carefully checked that the back-scattered laser light from the sample surface is negligible). P^1 decays with the transverse spin relaxation time $T_{s2}\sim 13$ ps. As reported previously by Sermage *et al.*, the SE signal exhibits a biexponential decay.[12] These authors attribute the fast initial decay time ($\sim$4 ps here) either to the immediate escape of the polariton from the cavity or to the absorption of phonons and scattering towards large nonradiant k states (the *reservoir*). The second slower decay is then attributed to the return from the reservoir towards the detected radiant states.[11] The inset in Fig. 1(a) presents the P^1 dependence versus the fine temporal separation t_2 between the two pulses. This recording, as all the similar data in this paper, is systematically taken just after the second pulse ($\sim$1 ps). The observed oscillations, with the pulsation $\omega=E_{\mathrm{LB}}/\hbar$, simply reflect the rotation of the excitation light polarization in the sample plane, driven by t_2.[9]

Figure 1(b) displays the time dependence of the total SE when the delay between the two excitation pulses (σ^+,σ^-) is $t_1=6$ ps. Now there is no temporal overlap between the two pulses. Nevertheless, the excitation with the second pulse results again in a linearly polarized SE. As shown in the inset of Fig. 1(b), this linear polarization oscillates again at the pulsation $\omega=E_{\mathrm{LB}}/\hbar$ as a function of the fine temporal delay t_2. These oscillations reflect the rotation of the orientation of the linear exciton-polaritons in the cavity plane. The oscillations amplitude decay time T_d is measured in Fig. 2(a), where we find $T_d=4\pm 1$ ps.

The interpretation is similar to the one given previously in the case of the coherent control of the heavy-hole excitons alignment in bare QW's.[9] As is well recognized now in inhomogeneously broadened QW systems, the SE detected just after a short excitation pulse is strongly dominated by resonant Rayleigh scattering (RRS).[13–15] For moderate disorder in the QW, the amplitude of the coherent electromagnetic field radiated within a small aperture in a nonspecular observation direction decays with the exciton optical dephasing time T_2, after an eventual much rapid decay phase (which is not resolved in ps experiments).[15] In two pulses experiments, the excitation by the second pulse results in coherent superposition of individual dipoles states, provided that the di-

poles excited by the first one are not dephased. For a (σ^+,σ^-) excitation sequence, the amplitude of the linear polarization oscillations when t_2 varies is directly proportional to the relative number of excitonic dipoles which have kept at time t_1 the phase memory of the first pulse. Because of the RRS nature of the detected signal, the amplitude decay of the linear polarization oscillations as a function of t_1 directly reflects the optical dephasing of the excitons.

In microcavities, the polarization oscillations observed in Fig. 1(b) demonstrate that coherent emission from $k_{\|}\cong 0$ exciton-polariton states can be obtained from an off-normal excitation direction. This fact was previously observed by Norris *et al.*[3] in interferometric pump-probe experiments performed in nonspecular directions. We interpret this as the result of the coupling by the disorder induced potential of $k_{\|,\mathrm{pump}}$ and $k_{\|}\cong 0$ polariton states through their excitonic component,[7] which leads to inhomogeneous broadening of polariton states and the possibility of RRS. We can thus conclude that, due to the RRS nature of the detected signal, the polarization oscillations decay time T_d observed in our experiments is the polariton dephasing time T_2. For microcavities with very low disorder, the energy conservation and dispersion effects should in principle lead to RRS emission directions located on the surface of a cone centered around the normal direction to the microcavity, the top angle of which is determined by the excitation light angle with respect to that normal. However, the energy dispersion between $k_{\|,\mathrm{pump}}\cong 10^4$ cm^{-1} and $k_{\|}\cong 0$ is, in all cases investigated in this work, smaller than the polariton states broadening,[16] so that elastic light scattering becomes possible out of the above defined conical surface.

A similar experiment has been performed with a sequence of two linearly cross-polarized laser pulses (σ^X,σ^Y). The excitation with the second pulse results in a circularly polarized SE.[9] This experiment, not reported here for briefness, yields also the measurement of T_2.

We evidence now the coherent control of the polariton density by performing the experiment with a sequence of two circularly copolarized (σ^+,σ^+) laser pulses. Constructive and destructive interferences between the second pulse and the coherent polarization surviving from the first one modulate now the polariton density. This is demonstrated by the oscillations of the total SE intensity as a function of t_2, shown for two typical delays $t_1=4$ ps and $t_1=6$ ps in Fig. 2(b).

We study now the cavity detuning dependence of the optical dephasing time. The energies of the lower $(E_-\equiv E_{\mathrm{LB}})$ and upper (E_+) polariton states, measured as a function of the cavity detuning δ, are displayed in Fig. 3(a). The solid lines are fits to $E_\pm=\frac{1}{2}[(E_e+E_c)\pm\sqrt{\delta^2+\Omega_R^2}]$. The decay of the oscillations amplitude, which yields the measurement of T_2, is displayed in Fig. 3(b) for three cavity detunings. We find $T_2=5\pm1$ ps, 4 ± 1 ps, and 3 ± 1 ps for $\delta=-3$, 0, and 3 meV, respectively.[17] The trend is in agreement with the result of the FWM experiment performed by Wang *et al.*,[5] who observed that the FWM signal decay was faster at resonance $(\delta=0)$ than for negative detuning. This trend can be interpreted from the simple expression.[8] $1/T_2=1/(2T_1)+1/T'$, where T' represents pure dephasing. The measured variation of the SE fast initial decay time T_1 as a function of the detuning is displayed in Fig. 3(c). T_1 varies between 2.5 and 10 ps when δ varies between -10 meV and $+10$ meV. The short T_1 value measured for negative detunings is very close to the calculated photon lifetime in this cavity (1.7 ps). For the negative detuning $\delta=-3$ meV, T_2 is very close to $2T_1$, which indicates that when the photon character of the polariton state dominates, the optical dephasing time is determined by the cavity photon lifetime. The contribution of pure dephasing processes is then negligible. In contrast, for the positive detuning $\delta=+3$ meV, the lifetime is no longer responsible for the loss in phase coherence since $T_1\sim 6$ ps and $T_2\sim 3$ ps. Clearly, when the polariton has a dominant exciton character, the optical dephasing is due to the pure dephasing processes represented by T'. In this case, the measured T_2 value is close to the values reported for heavy-hole excitons in bare QW's of equivalent quality.[18] Our measured dependence of T_2 on δ is in agreement with recent calculations of the homogeneous broadening Γ_h of the polariton modes by Savona and Piermarocchi.[10]

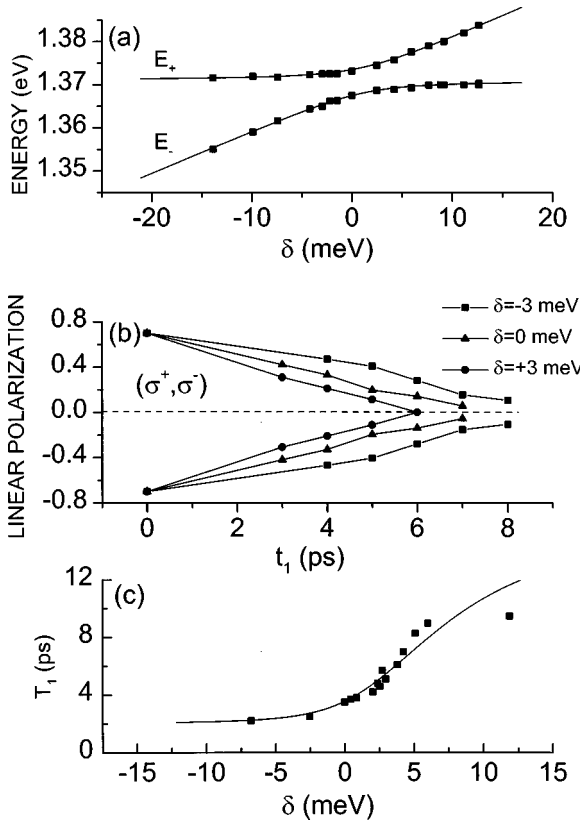

FIG. 3. (a) Energies of the polariton modes, measured in a cw photoluminescence experiment, as a function of the cavity detuning δ (the same excitation geometry as the time-resolved experiments is used). (b) The cavity detuning dependence of T_2. The minima and maxima of the linear polarization oscillations as a function of t_1 for $\delta=-3$, 0, $+3$ meV, respectively. (c) Initial SE decay time T_1 (■) as a function of the cavity detuning. The full line is a fit to $1/T_1=|C_e|^2/\tau_e+|C_c|^2/\tau_c$ with the parameters $\tau_c=2$ ps and $\tau_e=16$ ps.

Finally we investigate the influence of polariton-acoustic phonon scattering on both the optical dephasing time T_2 and the lifetime T_1. This is particularly attractive since we expect drastic changes of the phonon scattering efficiencies due to the specificity of the polariton dispersion curves.[10,19] The pronounced decay of the exciton optical dephasing time in bare QW's for increased lattice temperature is well known: this T_2 decrease in the temperature range 1–80 K

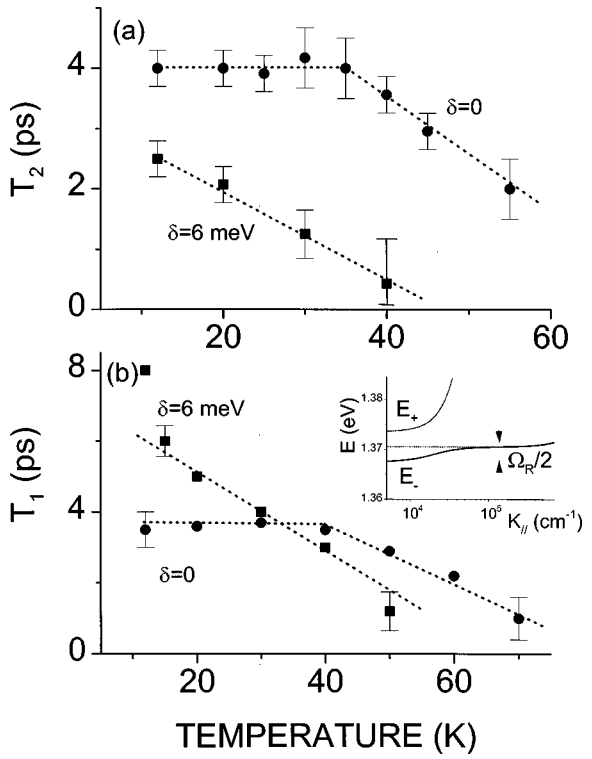

FIG. 4. Temperature dependence of (a) the optical dephasing time T_2 and (b) the population lifetime T_1 for two cavity detunings (●) $\delta=0$ and (■) $\delta=+6$ meV. Inset: dispersion curve of the LB polariton for $\delta=0$ as a function of the in-plane wave vector.

is attributed to the linear increase of the scattering rate of excitons with thermal acoustic phonons.[18] Figure 4(a) shows the measured T_2 of the LB polariton as a function of the lattice temperature for two cavity detunings $\delta=0$ and $\delta=+6$ meV. For the positive detuning, i.e., when the excitonic character of the polariton state is dominant (for $\delta=+6$ meV, we calculate the weight of the exciton component to be $|C_e|^2\sim 0.85$), we find a linear decrease of T_2 when the temperature increases. This behavior is similar to that of excitons in bare QW's.[18] At resonance ($\delta=0$), the striking feature is that T_2 is independent of the temperature up to 40 K. The decrease above 40 K shows approximately the same slope than the one measured for $\delta=+6$ meV. This stability of the homogeneous broadening at low temperature was predicted by Savona and Piermarocchi.[10] These authors have shown that there is almost no contribution of acoustic phonons to the LB polariton homogeneous broadening for low temperatures. This is the consequence of the very low mass in the bottleneck region. When $k_BT<\Omega_R/2$, only this bottleneck region, where the density of state is low, is available for acoustic phonon scattering [the polariton dispersion calculated for $\delta=0$ is shown in the inset of Fig. 4(b)]. Above 40 K for $\delta=0$ meV, we attribute the decrease of T_2 to the scattering by acoustic phonons, a process which is again efficient since the states beyond the bottleneck region become attainable ($k_BT>\Omega_R/2$).

In order to clarify the role played by acoustic phonons in the relaxation dynamics of exciton polaritons, we have investigated simultaneously the variation of the lifetime T_1 as a function of the temperature for the two cavity detunings $\delta=0$ and $+6$ meV. Figure 4(b) shows that the dependence of T_1 and T_2 are very similar. Note that this behavior of T_1 with temperature was predicted by Bloch and Marzin.[11] When the excitonic character of the polariton is strong ($\delta=+6$ meV), the acoustic phonon scattering determines both T_2 and T_1 in the explored temperature range. On the contrary, at resonance ($\delta=0$) and low temperature ($T<40$ K), T_1 is controlled by the cavity escape time.

In summary, we have shown that coherent control experiments can directly probe the dynamics of exciton polaritons in the strong coupling regime. By using a sequence of two phase-controlled optical excitation pulses, the density, the alignment, and the spin orientation of exciton polaritons can be coherently controlled and directly observed in a time-resolved SE experiment. The technique, which relies on the linear response of the material, allows a simultaneous measurement of both the lifetime T_1 and the optical dephasing time T_2 of the exciton-photon mixed modes. For zero detuning, the temperature dependence of both T_2 and T_1 clearly shows a quenching of the polariton-acoustic phonon scattering up to a temperature of the order of $\Omega_R/2k_B$.

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[17] Note that the dephasing resulting from the inhomogeneous broadening [as could be observed, e.g., in time-resolved reflectivity experiments] (Ref. 3) would result in a shorter decay time than observed experimentally here: for $\delta=0(+3$ meV), we should find $T_d\cong 2.6$ ps (1.6 ps), corresponding to $\Gamma_{\mathrm{inh}}=1.4$ meV (2.3 meV).
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