Inhibition of exciton spin relaxation by longitudinal-optical phonon emission

A. Filoramo, R. Ferreira, and Ph. Roussignol
Laboratoire de Physique de la Matière Condensée, Ecole Normale Supérieure, 24 Rue Lhomond, F-75005 Paris, France

R. Planel and V. Thierry-Mieg
Laboratoire de Microélectronique et Microstructures-CNRS, 196 Avenue Henry Ravéra, F-92220 Bagneux, France

(Received 29 September 1997)

We study the interplay between a fast energy relaxation process and the spin depolarization for *excitons* in undoped double quantum wells. We show that the exciton depolarization is significantly inhibited when a fast channel exists for the energy relaxation after a nonresonant excitation. Specifically, we consider the favorable case of a strongly coupled asymmetric double quantum well where the first heavy and light exciton levels of the thin well are separated by a longitudinal-optical phonon energy. The relevance of different relaxation mechanisms is also discussed. [S0163-1829(98)08631-7]

INTRODUCTION

The electronic and optical properties of semiconductor quantum wells have been extensively studied. The relevance of the confinement on the electronic levels is well established theoretically and experimentally. The study of the spin-dependent ''fine'' structure of the electronic levels, on the contrary, is a less considered subject. In this work we are interested in the interplay between energy relaxation and spin depolarization in nonresonantly excited intrinsic quantum wells. On the one hand, the energy relaxation of photoexcited carriers assisted by acoustical and optical phonons is currently evidenced in both cw and time-resolved photoluminescence excitation (PLE) measurements. On the other hand, the reported spin dependent measurements concern mainly the evolution in time after a resonant or quasiresonant excitation and/or the study of the cw polarization resolved PLE spectra. Let us consider initially a few results in the literature.

Most of the reported time-resolved polarization measurements were done for resonant or quasiresonant excitation of the ground heavy-hole transition of a single quantum well.[1–6] In this case, the temporal dynamics involve an initial fast redistribution of the photocreated excitons followed by a slow thermalization of the resulting hot distribution in the fundamental dispersion. In this quasiresonant configuration, the evolution in time (at large times) of the polarization measured at the subband edge is due to the arrival of depolarized excitons from large K states.[6] Also, one assumes generally that the spin is conserved during the relaxation by emission of acoustical phonons, a reasonable assumption for the weakly spin-mixed low-energy exciton levels, and that the depolarization is related to processes that occur before relaxation and conserve the in-plane kinetic energy. Of course, the acoustical phonon assisted relaxation can be modified by varying the lattice temperature for a fixed energy detuning; however, the temperature also modifies the exciton distribution and so the energy relaxation rate cannot be modified in this way without affecting the spin depolarization rate (the latter increases with increasing mean kinetic energy for the population distribution).

Let us now consider the case of a nonresonant configuration. Information on the relationship between relaxation and depolarization has been extracted from both steady-state (measurement of the polarization spectrum[7]) and time-resolved (measurement of the initial polarization[8]) experiments. In the nonresonant configuration, the polarization is measured as a function of the energy detuning between excitation and detection inside a given subband at fixed (low) temperature. In this case, the total relaxation time for the population is expected to increase with increasing detuning. Thus, in a few works devoted to single wells, the depolarization was related to acoustical phonon assisted scattering processes, which simultaneously relax the energy and flip the spin.[9,10] However, a further effect, namely, the possibility of an energy-conserving depolarization mechanism, has also been discussed for independent carriers and shown to strongly depend on their energy.[11] We have to bear in mind also that in both cases (elastic and quasielastic scatterings) a depolarization is possible because of the heavy-light hole mixing of the valence states, which increases with increasing detuning and modifies the initial polarization *before* relaxation.[12] In conclusion, the interplay between relaxation and depolarization is somehow hindered by the existence of energy-conserving scatterings and by an energy-dependent initial polarization. A particular situation appears when the excitation is one optical phonon above the fundamental heavy-hole exciton. In this case, we would expect a faster relaxation and thus an important variation of the detected polarization for excitation energies around this particular detuning.

In this work the excitation energy is set around the first light-hole exciton transition and the detection is set at the ground heavy-hole one; the light and heavy levels under consideration are the fundamental ones of the thin well of an asymmetric double quantum well. The study of the unambiguous interplay between a fast energy relaxation process and the spin depolarization is possible in our sample for two main reasons: (i) The narrow well is coupled to a wider one by a thin tunnel barrier and (ii) the energy difference between the two exciton levels equals the energy of a longitudinal-optical (LO) phonon. The importance of these

two effects is discussed in the following. Our main result is that *the spin depolarization of excitons is strongly inhibited in the presence of a fast energy relaxation process*.

The paper is organized as follows. The sample used and the experimental setup are presented in Sec. I. The experimental results for both cw and time-resolved measurements are shown in Sec. II. Due to the particular resonance involving the heavy-light transition and an optical phonon energy, we were led to consider the relevance of a few mechanisms for the relaxation; this discussion is presented in Sec. III.

I. SAMPLE AND EXPERIMENTAL SETUP

The sample consists in two GaAs quantum wells of different widths, 62 and 23 Å respectively, separated by a 27-Å-thick $Al_{0.3}Ga_{0.7}As$ barrier. Therefore, due to the small thickness of the $Al_{0.3}Ga_{0.7}As$ barrier, the two wells are strongly coupled. On both sides of this asymmetric double quantum well the $Al_{0.3}Ga_{0.7}As$ barriers are replaced by (8 ML)/(4 ML) GaAs/AlAs superlattices, 600 Å thick, in order to improve the quality of the interfaces. This structure is grown on an n^+-doped GaAs substrate. A 100-Å-thick GaAs layer caps the heterostructure.

Our experimental results were obtained by means of cw photoluminescence (PL) and PLE spectroscopies. Time-resolved photoluminescence and spin orientation experiments were also performed in order to investigate the dynamics of the LO phonon assisted energy relaxation processes and conservation of the spin polarization. All measurements have been done at 2 K, keeping the sample fully immersed in superfluid helium.

Continuous-wave measurements have been performed by standard lock-in techniques. The PL and PLE spectra were obtained with a cw Ar^+ laser pumped Ti-sapphire laser as excitation source. The excitation power was taken to be as weak as possible (≈ 0.5 W cm^{-2}) to allow a good signal-to-noise ratio with a 1-m double monochromator, a GaAs cooled photomultiplier tube, and a lock-in amplifier.

In time-resolved spectroscopy the excitation was provided by a cw mode-locked Tsunami Ti-sapphire laser, pumped by a cw Ar^+ laser, producing 2-ps pulses with 82-MHz repetition rate. The PL signal was detected by a Hamamatsu streak camera after a 32-cm Jobin-Yvon double monochromator. The spectral resolution was about 1 meV and the overall time resolution of the whole apparatus was of the order of 15 ps.

In spin orientation experiments the sample is excited by right-circularly polarized light σ^+. Just before focusing in the cryostat a linear polarizer checks the polarization of the excitation light and a quarter-wave plate is inserted in order to produce circular polarization. The right- and left-circularly polarized components of the PL signal, σ^+ and σ^-, respectively, are recorded by rotating a second quarter-wave plate located before the spectrometer. The $\lambda/4$ plate linearizes the σ^+ and σ^- components and the one of interest is selected with a second polarizer located before the spectrometer. Thus the light traveling inside the spectrometer always has the same polarization, avoiding any grating effect. The two signals σ^+ and σ^- are obtained by two successive acquisitions and the polarization $P=(\sigma^+-\sigma^-)/(\sigma^++\sigma^-)$ is then calculated.

Such a procedure may be questionable in time-resolved experiments. In order to eliminate any jitter effect between two successive accumulations, a temporal reference was inserted in the time profile recorded by the streak camera and part of the excitation beam, with the appropriate time delay, was sent directly to the streak camera. In this way we always had a reference laser pulse in each acquired image to set the origin of the time axis at the right position.

II. EXPERIMENTAL RESULTS

In this sample we have observed the presence of sharp peaks in the PL and PLE spectra of the 23-Å narrow well. These peaks are related to longitudinal-optical phonon emissions and we have studied the associated polarization properties. The situation is even more complicated in our sample where, unintentionally, the heavy-hole–light-hole (HH-LH) splitting (the energy difference between the ground light X'_{LH} and ground heavy X'_{HH} excitons of the narrow well) is about 36 meV and corresponds to the LO phonon energy. Different mechanisms can induce the occurrence of phonon related structures (such as hot excitonic luminescence, double resonant Raman scattering, and phonon replica) and will be discussed in Sec. III. We present first in Sec. II the results of our cw (Sec. II A) and time-resolved (Sec. II B) measurements.

A. cw experiments

In Fig. 1(a) we report some typical cw PL spectra of the narrow well recorded for various excitation energies (written on the left-hand side of the PL profile). The PL line is centered at 1.697 eV with a 6 meV spectral width. The Stokes shift is about 3 meV, a reasonable value for such a narrow well. For a 1.740-eV excitation energy a sharp structure starts to be observable at 1.705 eV on the high-energy side of the PL spectrum. In fact, two sharp peaks are clearly seen in the PL spectra obtained with 1.7355 and 1.7306-eV excitation energies. For excitation energies lower than 1.716 eV no sharp peaks are observable. This latter value for the excitation energy corresponds exactly to the LO phonon emission energy threshold. Of course, the intensity of the PL signal depends on the excitation wavelength (resonant or nonresonant excitation with the light-hole exciton) and for the sake of clarity, in Fig. 1(a), the PL profiles are normalized in order to have the same amplitude.

If we plot the energy of the two phonon peaks as a function of the excitation energy, we note that the main one always appears at $\approx$ 36 meV from the excitation, while the other one is at $\approx$ 33.5 meV [Fig. 1(b)]. We therefore assign these structures to the GaAs LO phonon and GaAs-type LO phonon of the $Al_xGa_{1-x}As$ barrier, respectively.[13]

These peaks are present in the cw PLE spectra of the narrow well and their energy shows the same dependence on the detection energy in the PL line. In Fig. 2 (lower part) we show a typical cw PLE spectrum with the detection set at 1.694 eV; the main phonon peak appears at 1.730 eV. Note that in our sample the energy difference between the ground light-hole exciton X'_{LH} and ground heavy-hole one X'_{HH} of the narrow well is of the order of 36 meV, so that the phonon peaks are always superimposed on the LH line.

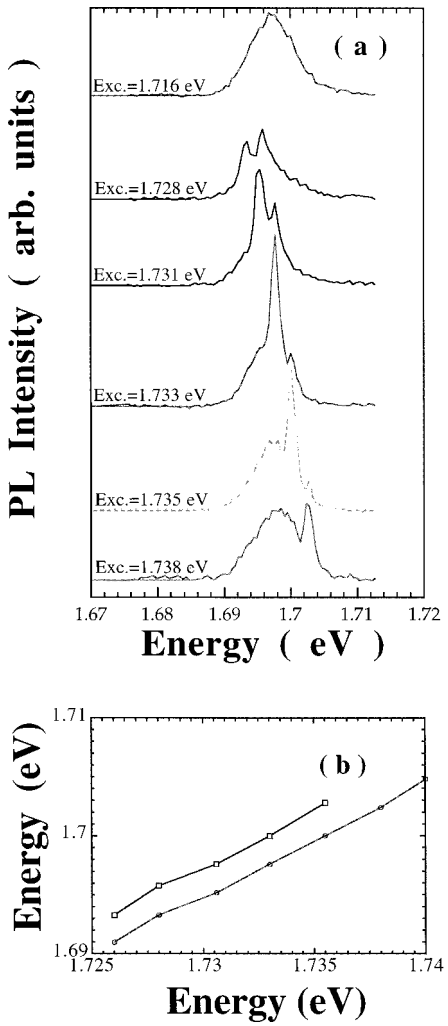

FIG. 1. (a) PL spectra of the narrow well for different excitation energies. (b) Energy position of the phonon replica observed in the narrow-well PL spectrum as a function of the excitation energy.

We have also performed PL spectra for incident powers varying between 0.5 and 13 W cm^{-2}. The corresponding photocreated carrier densities are estimated to 2.5×10^{9} and 6×10^{10} cm^{-2}. Typical results are shown in Fig. 3 for two different excitation energies: 1.733 eV for the left-hand side of the figure and 1.735 eV for the right-hand side. Note that the spectral resolution was smaller than in Fig. 1 because the experiment was performed under pulsed excitation and in this case the spectral resolution of the detection system is smaller (1 meV). At low excitation powers (a closed diamond, for 0.5 W cm^{-2} and a closed circle for 0.72 W cm^{-2}) the main phonon peak is still clearly observable, 36 meV below the excitation energy. When the excitation power is increased (an open triangle for 3.0 W cm^{-2} and an open circle for 4.3 W cm^{-2}) this peak becomes less and less observable. For the higher excitation powers (an open square for 10 W cm^{-2} and an open diamond for 13.3 W cm^{-2}), it has clearly disappeared.

Let us consider now the results of spin orientation measurements under cw illumination, which are reported in the upper part of Fig. 2. We have plotted the value of the polarization P^{cw} of the PL signal as a function of the excitation energy. As is usually observed,[7] for an excitation of the

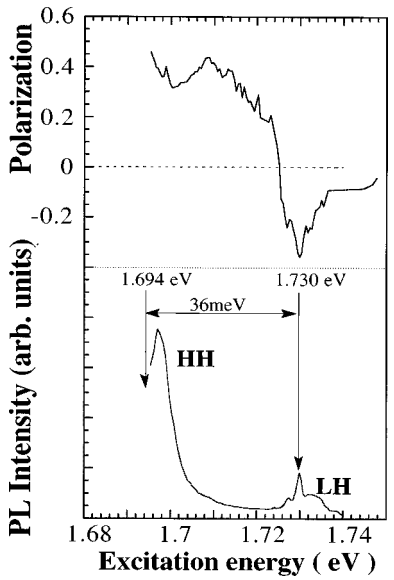

FIG. 2. cw polarization of the PL signal arising from the narrow well as a function of the excitation energy (upper panel). The corresponding PLE spectrum is also reported (lower panel).

heavy-hole subband the polarization is positive (+35% for a resonant excitation of X'_{HH}), whereas when the light-hole subband is involved it is negative (−20% for a resonant excitation of X'_{LH}). The lower degree of polarization (35% instead of the 100% expected) measured when exciting resonantly X'_{HH} is possibly due to two effects: (i) We do not detect free excitons and a depolarization may occur during the localization stage of the excitonic population (the influence of localization on the exciton polarization is studied in Refs. 14 and 15) and (ii) the structure is asymmetric and a further depolarization is expected for the double quantum well eigenstates (a detailed study of the role of the asymmetry on the exciton polarization is under way and will appear elsewhere[16]). The striking result of Fig. 2 is that, in correspondence with the sharp phonon peak present in the PLE spectrum, the polarization shows a clear enhancement and reaches a value of −38%. This enhancement of the polarization detected at the band edge when exciting 36 meV above will be discussed in Sec. III.

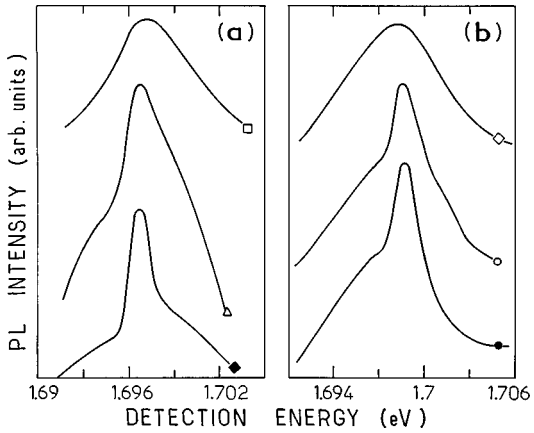

FIG. 3. Time-integrated PL spectra of the narrow well for different incident powers under pulsed excitation (see the text) and for two excitation energies: (a) 1.733 eV and (b) 1.735 eV.

Finally, we would like to stress that the presence of a phonon replica does not mean a poor quality of the sample but strongly depends on the barrier thickness.[17] Actually, we observe in the PLE spectrum of the wide well (not shown) that the PL intensities for resonant excitations of both the wide and the narrow wells are roughly the same, which confirms the strong tunnel coupling between the two wells in our structure.

B. Time-resolved measurements

Time- and polarization-resolved photoluminescence spectroscopy gives access to four parameters: the initial polarization $P(0)$, the spin relaxation time τ_S, the photoluminescence rise time τ_R, and the recombination (or decay) time τ_D. Let us consider a simple phenomenological three-level model in order to make clear the physical meanings of these four parameters. We make the following assumptions: The created population N in the high-energy level is entirely polarized (100%); the N population relaxes towards two degenerate excitonic levels (with n^+ the spin-up population and n^- the spin-down population), either conserving (towards n^- with the spin-conserving rise time τ_R^{SC}) or not (towards n^+ with the spin-flip rise time τ_R^{SF}) the polarization; the decay time τ_D is the same for both populations n^+ and n^-; and during the recombination process (and emission of the PL signal) spin relaxation (characterized by the spin relaxation time τ_S) between the spin-up and the spin-down populations may occur. Thus the N, n^+, and n^- populations obey the set of rate equations

$$\frac{dn^+}{dt}=\frac{N}{\tau_R^{\mathrm{SF}}}-\frac{n^+}{\tau_D}-\frac{n^+}{\tau_S}+\frac{n^-}{\tau_S},$$
$$\frac{dn^-}{dt}=\frac{N}{\tau_R^{\mathrm{SC}}}-\frac{n^-}{\tau_D}-\frac{n^-}{\tau_S}+\frac{n^+}{\tau_S}, \tag{1}$$

with $N=N_0\ \exp\{-t/\tau_R\}$ and $1/\tau_R=1/\tau_R^{\mathrm{SC}}+1/\tau_R^{\mathrm{SF}}$. This model leads to the expression for the initial polarization $P(0)$,

$$P(0)=\frac{\tau_R^{\mathrm{SC}}-\tau_R^{\mathrm{SF}}}{\tau_R^{\mathrm{SC}}+\tau_R^{\mathrm{SF}}}. \tag{2}$$

The creation rate appears in the expression of $P(0)$ only as a multiplicative factor so that considering a creation rate smaller than 100% decreases $P(0)$ by the same amount. The main point is that $P(0)$ is the only parameter reflecting directly the specificity of the relaxation process, spin conserving or not. It depends on the relative probability of these two types of energy relaxation processes. On the contrary, the time evolutions of n^+ and n^- are determined by τ_D, τ_S, and τ_R parameters, which are not sensitive to the nature, spin conserving or not, of the relaxation mechanism. The spin relaxation time τ_S is only affected by physical mechanisms occurring at the bottom of the band edge. By integrating Eq. (1) over time one may recover the expression of the cw polarization

$$P^{\mathrm{cw}}=P(0)\ \frac{\tau_S}{\tau_S+2\tau_D}. \tag{3}$$

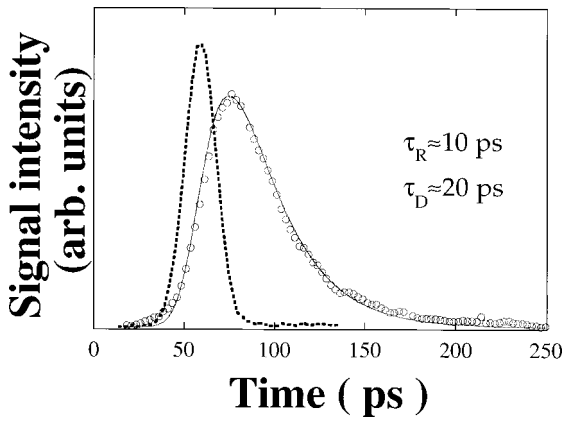

FIG. 4. Typical time evolution of the narrow-well PL intensity. Open circles, PL intensity; solid line, numerical fit; dashed line, laser pulse profile.

In order to determine experimentally the times τ_D, τ_S, and τ_R it appears more convenient to consider the sum $(n^+ + n^-)$ and the difference $(n^+ - n^-)$:

$$n^+ + n^- = N_0\ \frac{\tau_D}{\tau_R-\tau_D}\ (e^{-t/\tau_R}-e^{-t/\tau_D}),$$
$$n^+ - n^- = N_0\ \frac{\tau_R^{\mathrm{SC}}-\tau_R^{\mathrm{SF}}}{\tau_R^{\mathrm{SC}}+\tau_R^{\mathrm{SF}}}\ \frac{T}{\tau_R-T}\ (e^{-t/\tau_R}-e^{-t/T}), \tag{4}$$

with $1/T=1/\tau_D+2/\tau_S$. The time dependence of both quantities depends only on two parameters. Thus the fitting procedure is easier and much more reliable. The characteristic times are determined by comparing the experimental curves to the convolution of Eqs. (4) with the experimental time response of the setup.

We find out that the rise and the decay times of the PL signal of the narrow well are always short: $\tau_R\approx 10$ ps and $\tau_D\approx 20$ ps (see Fig. 4 in comparison with the laser reference pulse provided by the delay line). The spin relaxation time τ_S is always in the range 190–220 ps. No dependence is observed for these times on either the excitation or detection energy. In accordance with our three-level picture, the initial polarization $P(0)$ is the only energy-dependent parameter. It is clearly enhanced in correspondence with the phonon related structure, as shown in Fig. 5. In this figure we show the time dependence of the σ^+ (light solid lines) and σ^- (heavy solid lines) components of the PL signal for two excitation energies. The corresponding time evolution of the polarization is also reported (dashed lines). When the energy difference between excitation and detection equals the GaAs LO phonon energy the initial polarization reaches a value of -0.75 (see the lower panel). A slight shift of the excitation energy decreases significantly the initial polarization (-0.2 in the upper panel). For both cases the polarization decay times are roughly the same.

It is possible to check further the consistency of our model as follows. Reported in Fig. 6 are the experimental values (closed circles) of the cw polarization P^{cw} of the PL spectrum of the narrow well. By using Eq. (3) and taking the experimental values of $P(0)$, τ_D, and τ_S, P^{cw} can be calculated (open circles in Fig. 6). Considering the simplicity of the three-level approach, the agreement is rather good.

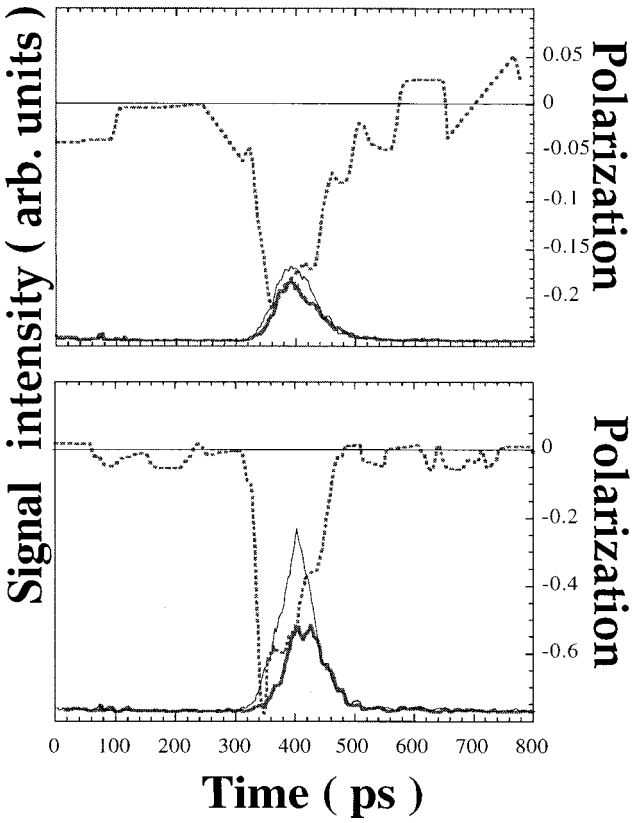

FIG. 5. Time evolution of the σ^+ and σ^- narrow-well PL intensities (heavy and light solid lines) and of the resulting polarization (dashed lines) after a σ^+ excitation. The energy difference between excitation and detection is equal to (lower panel) or different from (upper panel) the GaAs LO phonon energy.

III. DISCUSSION

In the following we distinguish among various possible processes that could lead to the appearance of these sharp peaks in the PL and PLE spectra: (i) hot excitonic luminescence (HEL), (ii) enhancement of the PL signal due to double resonant Raman scattering (DRRS), (iii) a ''direct'' phonon replica due to an energy relaxation from the light-hole exciton towards the heavy one assisted by LO phonons, and (iv) an ''indirect'' phonon replica due to a two-step process where an elastic or quasielastic scattering converts the light-hole exciton into a heavy one and is followed by an optical phonon assisted energy relaxation towards the edge of the X'_{HH} dispersion.

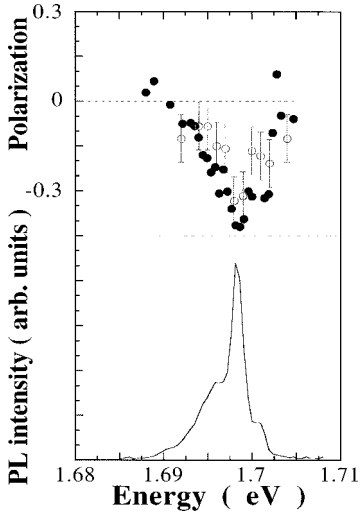

FIG. 6. Upper panel, measured (●) and evaluated ○ cw polarizations of the narrow-well luminescence; lower panel, PL spectrum of the narrow well.

A. Hot excitonic luminescence

Hot excitonic luminescence is the signal arising from the indirect population of an exciton state through the simultaneous absorption of a photon and emission (or absorption) of a LO phonon when the incident photon has the ''appropriate'' energy. The HEL processes are widely seen in II-VI material where the exciton-phonon coupling is large.[18–20] Generally speaking, after this indirect absorption the exciton is created at a large in-plane wave vector K and therefore has to relax down to reach $K\approx 0$ where it can radiatively recombine. When this exciton is created at the energy $E_{\mathrm{dect}}+n\hbar\omega_{\mathrm{LO}}$ it can rapidly relax through the emission of n LO phonons (LO phonon cascade) and be detected (hot excitonic luminescence) before thermalization. For the excitons created at $E\neq E_{\mathrm{dect}}+n\hbar\omega_{\mathrm{LO}}$ a cascade of LO phonons takes place with a final energy close to E_{dect} but to reach this latter value and be detected in the PL and PLE spectra they should relax by emitting acoustical phonons or via the exciton-exciton interaction. When these final relaxation processes are less efficient than the non-radiative ones, there is no trace of such excitons in the spectra.

In our sample there are two reasons to exclude a hot luminescence process.

(i) The negative polarization measured at the energy of the phonon peak is enhanced. If we were in the presence of a hot excitonic population it should be a heavy one and therefore have a positive polarization (or zero polarization if we consider the phonon cascade as a possible depolarizing process) but not a negative one, even if we consider the mixed nature of the heavy-hole states for large wave vectors K.

(ii) On the one hand, the indirect creation of an exciton with large K assisted by the *emission* of a LO is forbidden for E_{exc} strictly equal to $E_{\mathrm{dect}}+\hbar\omega_{\mathrm{LO}}$.[17,18] This is exactly the selected range for the excitation energy in our sample. Actually, when $E_{\mathrm{exc}}=E_{\mathrm{dect}}+\hbar\omega_{\mathrm{LO}}+\Delta$ ($\Delta\ll\hbar\omega_{\mathrm{LO}}$) the excitonic population with $K\approx 0$ (and little excess energy Δ) can be created by indirect absorption assisted by LO phonon emission. In this case, however, the polarization should be definitively positive or zero as for heavy-hole quasiresonant excitation. On the other hand, the indirect creation of excitons with large K assisted by *absorption* (and not emission) of a LO phonon is still possible (X'_{HH} can then be reached after a two-LO-phonon cascade), but the probability that it happens is negligible at low temperatures and for weakly polar III-V semiconductor based heterostructures.

B. Double resonant Raman scattering

Let us consider now the possibility of DRRS. It occurs when both the incoming and the outgoing photons are resonant with an electronic state of the quantum well and when the energies of these two states differ by the energy of one

longitudinal optical phonon.[21] This is the case of the excitonic levels in the narrow well (Fig. 2). The experimental results, however, allow us to exclude that this phenomenon plays an important role. The first insight is given by the time-resolved measurements. When the detection is set on the phonon peak a short but finite rise time is measured (see Fig. 4). DDRS is an instantaneous process and the rise time should be given by the excitation pulse itself. Another reason is the power dependence of the phonon line reported in Fig. 3, which cannot be explained in terms of a resonant Raman process. The expected behavior for such a process is not so drastic. For example, experimental variations of the intensities of the PL and DRRS lines as a function of the incident power I_p are reported in the literature. They behave like $(I_p)^{\alpha}$, with $\alpha=1.4$ for the PL line and $\alpha=0.93$ for the DRRS peak.[22] The reported value for the DDRS line is very close to the expected one. The value of 1.4 for the PL line is coherent with excitonic recombination in the presence of nonradiative recombination. In fact, this factor is expected to vary between 1 and 2.[23] In any case, in our experiments, even considering $\alpha=2$ for the PL line, it is not possible to reproduce the fast disappearance of the phonon structure in the PL spectra.

C. Energy and polarization relaxation due to phonon replica

On the contrary, this power dependence is well explained in the case of LO phonon assisted energy relaxation from the light-hole exciton X'_{LH} towards the heavy one X'_{HH} of the narrow well as follows. The presence of these replicas is ultimately related to a short carrier lifetime. During a PLE spectrum carriers created with excess energy have to relax down to their respective band edge before they can recombine. As it is well known, the PLE spectrum can be rather different from the absorption one (related only to the density of states) and can show structures related to the energy relaxation process. Any kind of escape mechanism is in competition with the required energy relaxation and an enhancement in the spectrum is expected any time the energy relaxation time is faster than the escape one. If the escape time is always long compared to the relaxation time no enhancement is observed. The escape in our sample is not dominated by nonradiative processes (impurity or defects scattering), as it is generally the case in low-quality samples, but to a short tunneling time from the narrow well towards the wide one. The presence of the wide well allows us to observe in the PLE spectrum the contribution of fast intra-narrow-well energy relaxation channels as the LO phonons ones. The tunneling towards the wide well decreases the photoluminescence intensity so that the wide well acts as a non-radiative sink for the narrow one (this has been pointed out by Clerot *et al.*,[17] who studied a series of asymmetric coupled quantum wells with different intermediate barrier thicknesses). Note that when the density of photocreated pairs (or the excitation power) increases, the escape process tends to saturate[24–26] and, principally, other intrawell relaxation processes (as the one due to carrier-carrier interaction) become more probable, decreasing the importance of phonon-related ones. Therefore, the intensity of the PL line increases more than the one of the replica and the latter one is finally embodied in the PL line and disappears.

In conclusion, we associate the existence of phonon-related structures in the PL and PLE spectra of our sample to phonons replica, that is, to an initial effective photocreation of light-hole excitons followed by a relaxation towards the ground heavy-hole exciton level. In principle, the relaxation can be either a ''direct'' or an ''indirect'' process. We can, however, discard the contribution of a direct process since the observed fast energy relaxation (Sec. II) cannot be explained by a direct emission of an optical phonon because a diagonal Fröhlich interaction cannot couple the initial light and the final heavy $K=0$ states.

We then have to consider indirect (or two-step) processes where the light exciton initially ''escapes'' into other states with the same energy but from which a phonon emission towards the ground heavy exciton is possible. This escape process is possible only because of the heavy-light mixing introduced by the complicated nature of the valence states. It can be due to *assisted* scatterings and/or to an *intrinsic* coupling. Actually, a fast escape for the light excitons is expected with the help of a Fano-like coupling with the continuum of dissociated heavy excitons.[27–29] The latter affects only the exciton relative motion and couples light and heavy states with different in-plane angular symmetries, e.g., the $K=0$ $1S$-like light state is coupled to the continuum of $K=0$ three-dimensional-like dissociated heavy states. An energy relaxation towards the states on the ground heavy dispersion is then possible by a subsequent emission of one optical phonon. This emission is, however, governed by a matrix element that vanishes at resonance ($K_f=K_i=0$), while the luminescence intensity displays a clear resonant behavior (see Fig. 2). Thus the solely intrinsic coupling is unable to account for the experimental data. The observed resonant behavior, on the other hand, is consistent with a partial relaxation of either the wave vector K and/or of the angular symmetry, as provided by assisted elastic and quasielastic scatterings. Note that two different processes are in principle possible: (i) an intrinsic, Fano related, escape followed by an elastic scattering and (ii) an escape governed by the scatterings. In any case, our results clearly show the role played by elastic and quasielastic scatterings in the first part of the relaxation process between the photocreated light and the final heavy excitons of the narrow well.

In conclusion, we analyzed various processes and retained a two-step mechanism for the population relaxation; it is important to point out that the latter is consistent with our results for the polarization. Actually, an enhancement of the negative polarization in correspondence with the phonon replica is expected within a very simple picture, namely, any possible mechanism ''has no time to depolarize'' whenever the energy relaxation is fast enough. A quantitative analysis should account for the fact that, as is usual in semiconductors, any attempt to interpret the spin-related effects observed in real samples involves a detailed description of the relevant electronic levels and the consideration of the broadening of these levels.[30] This is beyond the scope of this work. We aimed here only to stress the interplay between a fast energy relaxation process and the conservation of the *exciton* spin: We note that an enhancement of the spin polarization in correspondence with an optical phonon assisted energy relaxation was observed for hot *electrons* in *p*-doped bulk materials.[30] For excitons in intrinsic quantum wells, to our

knowledge, this is the first clear evidence of the fact that a fast relaxation inhibits the spin depolarization.

CONCLUSIONS

We presented and discussed time-resolved experiments that clearly show the interplay between a fast energy relaxation process and the spin depolarization for *excitons* in undoped double quantum wells. More specifically, we showed that the exciton depolarization is significantly inhibited when a fast channel exists for the energy relaxation after a nonresonant excitation. To this end, we studied the favorable case of a strongly coupled asymmetric double quantum well where the first heavy and light exciton levels of the thin well are separated by a LO phonon energy. Taking into account the resonant configuration for the heavy-light exciton separation and LO phonon energies, the relevance of different relaxation mechanisms was also discussed. Our results as a function of the excitation intensity and the observed marked polarization behavior allowed us to conclude that the exciton relaxation led to phonon replica and that the absorption, relaxation, and final radiative recombination should be regarded as independent processes.

ACKNOWLEDGMENTS

The authors would like to thank C. Delalande, J.-Y. Marzin, and G. Bastard for various fruitful discussions. The Laboratoire de Physique de la Matière Condensée is ''Laboratoire Associé au CNRS (URA 1437) et aux Universités Paris 6 et Paris 7.'' A.F. was supported by the European Community (TMR Contract No. ERBFMBICT950388).

[1] R. J. Seymour and R. R. Alfano, Appl. Phys. Lett. **37**, 231 (1980).
[2] A. Tackeuchi, S. Muto, T. Inata, and T. Fujii, Appl. Phys. Lett. **56**, 2213 (1990).
[3] T. C. Damen, L. Viña, G. E. Cunnigham, J. Shah, and L. J. Sham, Phys. Rev. Lett. **67**, 3432 (1991).
[4] M. Kohl, M. R. Freeman, D. D. Awschalom, and J. M. Hong, Phys. Rev. B **44**, 5923 (1991).
[5] S. Bar-Ad and I. Bar-Joseph, Phys. Rev. Lett. **68**, 349 (1992).
[6] A. Vinattieri, J. Shah, T. C. Damen, D. S. Kim, L. N. Pfeiffer, M. Z. Maialle, and L. J. Sham, Phys. Rev. B **50**, 10 868 (1994); A. Vinattieri, J. Shah, T. C. Damen, D. S. Kim, L. N. Pfeiffer, and L. J. Sham, Solid State Commun. **88**, 189 (1993).
[7] R. C. Miller and D. A. Kleinman, J. Lumin. **30**, 520 (1985).
[8] Ph. Roussignol, P. Rolland, R. Ferreira, C. Delalande, G. Bastard, A. Vinattieri, L. Carraresi, M. Colocci, and B. Etienne, Surf. Sci. **267**, 360 (1992).
[9] T. Uenoyama and L. J. Sham, Phys. Rev. B **42**, 7114 (1990).
[10] Ph. Roussignol, R. Ferreira, C. Delalande, G. Bastard, A. Vinattieri, J. Martinez-Pastor, L. Carraresi, M. Colocci, J.-F. Palmier, and B. Etienne, Surf. Sci. **305**, 263 (1994).
[11] R. Ferreira and G. Bastard, Phys. Rev. B **43**, 9687 (1991); Solid-State Electron. **37**, 851 (1994).
[12] A. Twardowski and C. Hermann, Phys. Rev. B **35**, 8144 (1987).
[13] G. Abstreiter, in *Proceedings of the NATO Advanced Study Institute on Molecular Beam Epitaxy and Heterostructures*, edited by L. L. Chang and K. Ploog (Nijhoff, Dordrecht, 1985).
[14] A. Frommer, A. Ron, E. Cohen, J. A. Kash, and L. N. Pfeiffer, Phys. Rev. B **50**, 11 833 (1994).
[15] L. Muñoz, E. Pérez, L. Viña, and K. Ploog, Phys. Rev. B **51**, 4247 (1995).
[16] D. Larousserie, A. Filoramo, Ph. Roussignol, and R. Ferreira (unpublished).
[17] F. Clerot, B. Deveaud, A. Chomette, A. Regreny, and B. Sermage, Phys. Rev. B **41**, 5756 (1990).
[18] R. Planel, A. Bonnot, and C. Benoît à la Guillaume, Phys. Status Solidi B **58**, 251 (1973).
[19] N. Pelekanos, J. Ding, Q. Fu, A. V. Nurmikko, S. M. Durbin, M. Kobayashi, and R. L. Gunshor, Phys. Rev. B **43**, 9354 (1991).
[20] R. P. Stanley, J. Hegarty, R. Fisher, J. Feldmann, E. O. Göbel, R. D. Feldman, and R. F. Austin, Phys. Rev. Lett. **67**, 128 (1991).
[21] Y. R. Shen, *The Principles of Non Linear Optics* (Wiley, New York, 1984).
[22] D. A. Kleinman, R. C. Miller, and A. C. Gossard, Phys. Rev. B **35**, 664 (1987).
[23] J. E. Fouquet and A. E. Siegman, Appl. Phys. Lett. **46**, 280 (1985).
[24] R. Sauer, K. Thonke, and W. T. Tsang, Phys. Rev. Lett. **61**, 609 (1988).
[25] Ph. Roussignol, A. Vinattieri, L. Carraresi, M. Colocci, and A. Fasolino, Phys. Rev. B **44**, 8873 (1991).
[26] R. Ferreira, P. Rolland, Ph. Roussignol, C. Delalande, A. Vinattieri, L. Carraresi, M. Colocci, N. Roy, B. Sermage, J.-F. Palmier, and B. Etienne, Phys. Rev. B **45**, 11 782 (1992).
[27] D. A. Broido, and L. J. Sham, Phys. Rev. B **34**, 3917 (1986).
[28] G. E. W. Bauer and T. Ando, Phys. Rev. B **38**, 6015 (1988).
[29] L. C. Andreani and A. Pasquarello, Phys. Rev. B **42,** 8928 (1990).
[30] See, for instance, G. E. Pikus and A. N. Titkov, in *Modern Problems in Condensed Matter Sciences: Optical Orientation*, edited by F. Meyer and B. P. Zakharchenya (North-Holland, Amsterdam, 1984).