Exciton dynamics and spin relaxation in unstrained and tensile-strained quantum wells

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We have investigated exciton dynamics and spin relaxation in a GaAs/AlAs multiple quantum well and in tensile-strained $GaAs_{1-x}P_x/Al_{0.35}Ga_{0.65}As$ quantum wells. The studies have been done by picosecond time-resolved photoluminescence spectroscopy. The strain introduced by the presence of phosphorous in the $GaAs_{1-x}P_x/Al_{0.35}Ga_{0.65}As$ quantum wells modifies the subband dispersions, permitting access to new structures that include the possibility of the light-hole exciton's being the first excited state of the system. Our studies have focused on the influence of the different subband dispersions on exciton dynamics and spin relaxation. We have found that the exciton formation and the spin relaxation times decrease for the quasi-degenerate heavy-hole–light-hole excitonic ground state and that the recombination time depends essentially on the character of the excitonic ground state.

1. INTRODUCTION

Exciton dynamics in semiconductor quantum wells (QW's) have been a subject of intense research in recent years. A large number of studies have focused on the dynamics of exciton formation and relaxation,[1–8] and nowadays it is accepted that the relaxation process that follows the optical creation of an electron–hole pair of the continuum may be separated into two steps.[2] First, in a time of the order of tens of picoseconds, the electron–hole pair relaxes its energy and forms an exciton with high kinetic energy. In the second step, which lasts hundreds of picoseconds, the excess energy is lost, within the 1*s* excitonic branch, in a slow process that brings the exciton to states with center-of-mass momentum (K) near zero, where it may recombine emitting a photon.

Despite this agreement in the fundamental ideas of exciton formation and relaxation, there are several aspects of these processes that are still controversial. Some authors have found that exciton dynamics in GaAs QW's are independent of excitation energy, even for excitation at the light-hole (lh) exciton energy, or for excitation at one LO phonon above the detection energy.[2] However, using similar systems, other researchers have shown a significant slowing down of the relaxation process when exciting at the lh exciton energy[5] and have even demonstrated an oscillation in the exciton luminescence rise time as a function of excitation energy.[6] This oscillation has been interpreted as the occurrence of a selective optical-phonon-assisted exciton formation, which suggests that the exciton formation process from free electrons and holes is sensitive to the wave vector of the electrons and the holes after the initial LO-phonon cascade.[6]

As in the case of exciton dynamics, exciton spin relaxation has attracted a lot of attention, and several theoretical and experimental studies have been dedicated to researching the different spin relaxation mechanisms.[8–16] In this respect Maialle *et al.* recently offered a theory of exciton spin relaxation driven by intraexcitonic exchange interaction,[14] and various experimental studies have found evidence that this is the leading spin relaxation mechanism at low temperatures.[8,15] It was shown in Ref. 14 that the spin relaxation rate that is due to the main part of exchange interaction, i.e., the long-range part, is proportional to the square of the exciton wave vector. This relationship reveals the dependence between exciton dynamics and spin relaxation, as well as the importance of the subband dispersion in the spin–flip processes.

In this paper we report on exciton dynamics and spin relaxation in tensile-strained $GaAs_{1-x}P_x/Al_{0.35}Ga_{0.65}As$ QW's grown on GaAs substrates. For comparison purposes we also present results obtained in a GaAs/AlAs multiple quantum well (MQW). In tensile-strained

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QW's, a proper choice of material parameters and QW dimensions permits tuning of the energy splitting between the heavy-hole (hh) and the lh states, allowing for access to a range of new subband structures that include the possibility of the lh exciton's being the first excited state of the system.[17] Therefore tensile-strained QW's represent a good choice for the investigation of a new aspect of exciton dynamics and spin relaxation, namely, the influence of the subband dispersion and of the character, heavy or light, of the ground excitonic state on these processes.

2. EXPERIMENTAL DETAILS

We report on exciton dynamics and spin relaxation in a GaAs/AlAs MQW and in tensile-strained $GaAs_{1-x}P_x/Al_{0.35}Ga_{0.65}As$ QW's. The GaAs/AlAs system, grown by molecular beam epitaxy, consists of 50 periods of nominally 77-Å-wide GaAs wells and 72-Å-wide AlAs barriers. The $GaAs_{1-x}P_x/Al_{0.35}Ga_{0.65}As$ QW's were grown by organometallic chemical vapor deposition on [001] GaAs substrates.[18] We investigated three GaAsP/AlGaAs samples that had phosphorous contents of 5, 8, and 12% (samples A, B, and C, respectively). The phosphorous compositions were determined by x-ray diffraction measurements.[19] In each GaAsP/AlGaAs sample there were several $GaAs_{1-x}P_x$ QW's with different thicknesses, separated by 400-Å $Al_{0.35}Ga_{0.65}As$ barriers. In this study we report the results of QW's of 80 and 120 Å, which, for samples A, B, and C, are referred to as A-80, A-120, B-80, B-120, C-80, and C-120, respectively.

We performed the experiments by using a cold finger cryostat, keeping the samples at 4 K and exciting with pulses from a Styryl 8 or a Pyridine 1 dye laser, tunable from 810 to 700 nm. The incident light was directed along the growth axis, and a backscattering geometry was used. The dye laser was synchronously pumped by the 532-nm line of a mode-locked Nd:YAG laser. From the power density of our excitation we estimate an initial carrier density of the order of 5×10^{10} cm^{-2}. The photoluminescence (PL) was time resolved in a standard upconversion spectrometer. The time resolution, which we obtained by overlapping the luminescence from the sample on a nonlinear crystal, $LiIO_3$, with a delayed pulse from the laser, is basically determined by the width of the pulse, which in our case is 6 ps. A double-grating monochromator was used to disperse the upconverted signal. The exciting light was circularly polarized by means of a $\lambda/4$ plate, and the PL was analyzed into its σ^+ and σ^- components with a second $\lambda/4$ plate placed before the nonlinear crystal.

3. EXCITON DYNAMICS

Figure 1 shows, for the GaAs/AlAs MQW, the time evolutions of the sum of the σ^+ and the σ^- components of the luminescence exciting with σ^+-polarized light at two different energies, 1.617 eV [curve (a), filled circles], and 1.630 eV [curve (b), open circles]. Both sets of time profiles were recorded detecting at the maximum of the PL band (1.612 eV). The inset of Fig. 1 shows the cw PL (thick curve) and the photoluminescence excitation (PLE) spectra (thin curve). The spectra were recorded at 2 K under very low excitation density (5 mW cm^{-2}). In the PLE spectrum the peaks corresponding to the hh and the lh excitons can clearly be seen at 1.617 eV and 1.645 eV, respectively. The onset of the hh subband continuum is also observed at 1.626 eV. The Stokes shift between the PL and the PLE spectra indicates that the PL originates from localized excitons. However, under the high-excitation levels used in our time-resolved experiments, the bound exciton contribution to the PL saturates, and the importance of the free exciton emission is enhanced.[15]

The large difference in the rise times of the two curves shown in Fig. 1 reflects the fact that curve (b) corresponds to the excitation of electron–hole pairs in the subband continuum (nonresonant excitation) and the fact that excess energy has to be lost before the exciton can recombine. In contrast, in the case of curve (a), the 1s–hh excitons have been created resonantly with $\mathbf{K} = 0$ with $\mathbf{K}$ being the center-of-mass wave vector, and they can directly couple to light. The fast initial decay observed in curve (a) is related to the filling of dark excitonic states because of hole spin flip and has been discussed in a previous paper (Ref. 15).

We have observed that, for nonresonant excitation, exciton dynamics are independent of excitation energy, even for excitation at the lh exciton energy or for excitation at one LO phonon above the detection energy. These results are in agreement with the results of previous studies (Ref. 2), which have shown that the initial dynamics of nonresonantly excited luminescence is dominated by the slow relaxation process from excitons with large wave vectors to $\mathbf{K} = 0$ excitons. This slow relaxation process occurs as a consequence of exciton–exciton and exciton–phonon interactions.[2]

Although our results are in line with the main ideas of the current understanding of exciton relaxation, for excitation energies close to the subband edge we have observed a fast initial rise in the luminescence, which reveals the existence of a different relaxation process.

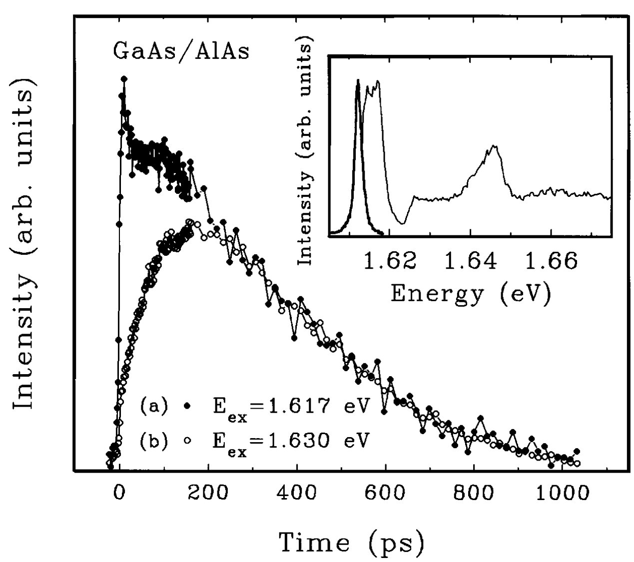

Fig. 1. Time evolution of the sum of the σ^+ and the σ^- components of the luminescence of the GaAs/AlAs MQW. Curve (a), excitation at 1.617 eV (filled circles). Curve (b), excitation at 1.630 eV (open circles). The inset shows the cw PL (thick curve), and the PLE spectra (thin curve) of the GaAs/AlAs sample.

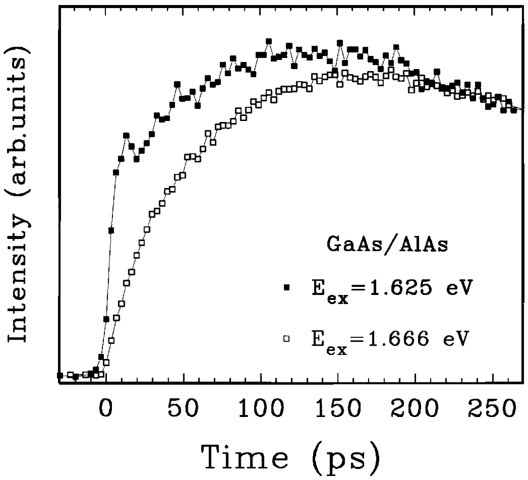

Fig. 2. Time evolution of the sum of the σ^+ and the σ^- components of the luminescence of the GaAs/AlAs MQW for two different excitation energies, 1.625 eV (filled squares) and 1.666 eV (open squares).

These results are shown in Fig. 2, in which the filled (open) squares depict the time evolution of the sum of the σ^+ and the σ^- components of the luminescence exciting with σ^+-polarized light at 1.625 eV (1.666 eV). The fast initial rise of the luminescence, for excitation close to the subband edge, indicates that a large number of excitons have reached $\mathbf{K} = 0$ states in a time of the order of our time resolution, revealing the existence of a fast relaxation process. Though further studies are necessary to determine the origin of this fast relaxation process, it could be related to the relaxation from ns $(n > 1)$ excitonic states to $1s$ states, inasmuch as it is observed only for excitation energies close to the subband edge.

Let us now concentrate on the results obtained for the $GaAs_{1-x}P_x/Al_{0.35}Ga_{0.65}As$ QW's. In this system the presence of phosphorous in the well layer decreases the magnitude of the lattice parameter, and the lattice mismatch between the GaAs and the $GaAs_{1-x}P_x$ layer is entirely accommodated in the $GaAs_{1-x}P_x$ layer as a biaxial tensile strain.[18] In response to the biaxial tension, the $GaAs_{1-x}P_x$ layer relaxes along the growth direction, giving rise to a uniaxial compression. The total strain reduces the energy gap and shifts the hh and the lh valence subbands to higher energies.[17] The amount of band-edge shift of the lh subband is larger than that of the hh one. Thus the inclusion of strain gives an extra degree of freedom in tuning the splitting of energy between the lh- and the hh-subband edges. Therefore new subband structures, including that of degenerate lh- and hh-subband edges or even that of the highest subband being lh-like can be achieved. This result is shown in Fig. 3, in which we depict the PLE spectra for the investigated $GaAs_{1-x}P_x/Al_{0.35}Ga_{0.65}As$ QW's. These QW's also present a Stokes shift of ~5 meV. The identification of the heavy or the light character of the excitons was obtained from previous PLE experiments in which circular polarization of the exciting light and analysis of the polarization of the emission was used.[20] The smallest peak in each spectrum corresponds to the lh exciton, whereas the largest one corresponds to the hh exciton. Those spectra that show only one structure correspond to a quasi-degenerate case. It can clearly be seen in the figure that the relative position of the hh and the lh excitonic states depends on well thickness (d) and on phosphorous composition (x). The A-80 QW presents the usual configuration, with the hh exciton being the ground excitonic state. The B-120 and the C-120 QW's show an inverse configuration, in which the ground state is the lh exciton, and the other three wells present a quasi-degenerate hh–lh exciton ground state.

Previous magneto-optical studies performed in these $GaAs_{1-x}P_x/Al_{0.35}Ga_{0.65}As$ samples yielded excitonic effective masses, binding energies, and lh–hh-subband-edge separations.[21] Those investigations demonstrated the flattening of the valence subbands, as the energy difference between the hh and the lh states is decreased, as a result of strain and quantum confinement effects.[21] Therefore, for these tensile-strained QW's, the study of the time evolution of the PL will provide information not only on the effect of the character of the ground excitonic state on exciton dynamics but also on the influence of the valence-band dispersion.

To study the dependence of exciton dynamics on the valence-band structure, we used the rise time (τ_{rise}) and the decay time (τ_{decay}) of the PL to characterize the relaxation time of the excitons to $\mathbf{K} = 0$ and the lifetime of the excitonic ground states, respectively. The values of τ_{rise} and τ_{decay}, obtained by excitation above the continuum edge for the different $GaAs_{1-x}P_x/Al_{0.35}Ga_{0.65}As$ QW's, are compiled in Table 1. The table also shows the values of Δ [$\Delta = E_g(\text{lh}) - E_g(\text{hh})$], the subband separation between hh and lh subbands, and the reduced hh and lh excitonic effective masses ($\mu_{\text{hh}}{}^*$ and $\mu_{\text{lh}}{}^*$, respectively) obtained from Ref. 21.

For the three QW's that correspond to the case of the nondegenerate excitonic ground state (A-80, B-120, and C-120), the PL rise times are of the order of 140 ps. This value is similar to the value of the rise time measured in the unstrained GaAs/AlAs system under nonresonant excitation. The long rise time indicates

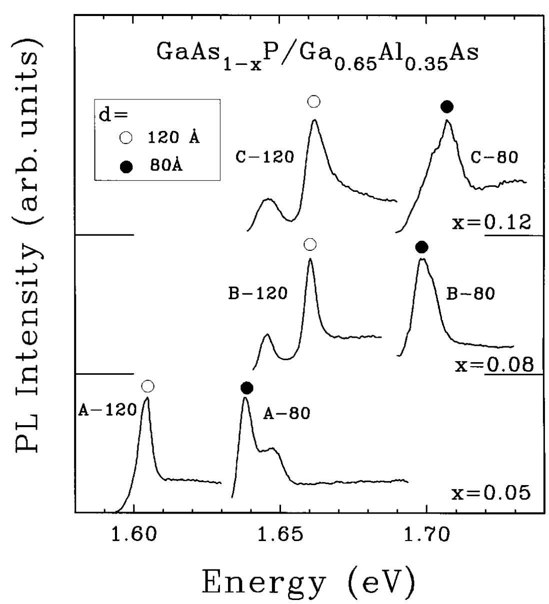

Fig. 3. PLE spectra of $GaAs_{1-x}P_x/Al_{0.35}Ga_{0.65}As$ QW's of 80- and 120-Å widths (filled and open circles, respectively), and of different phosphorous contents, x. The measurements were performed at 2 K, with excitation by σ^+-polarized light and detection of the σ^+ emission.

Table 1. Values Obtained by Excitation above the Continuum Edge[a]

QW	P (%)	d (Å)	Δ (meV)	μ_{hh}^* (m_0)	μ_{lh}^* (m_0)	τ_{rise} (ps)	τ_{decay} (ps)	$\tau_{\mathcal{P}}$ (ps)
A-80	5	80	16.5	0.051	0.044	126±8	570±50	100±30
A-120	5	120	−0.7	0.077	0.096	75±8	1050±100	32±8
B-80	8	80	−0.9	0.060	0.094	40±10	1020±50	30±8
B-120	8	120	−12.4	0.063	0.068	140±10	950±80	–
C-80	12	80	3.2	0.052	0.087	58±8	900±100	38±10
C-120	12	120	−16.9	0.060	0.060	160±10	900±50	–

[a]Phosphorous content (P); QW thickness (d); splittings between the hh and the lh subbands [$\Delta = E_g(\mathrm{lh}) - E_g(\mathrm{hh})$]; reduced effective masses of the hh and the lh excitations (μ_{hh}^* and μ_{lh}^*, respectively); rise and decay times of the luminescence (τ_{rise} and τ_{decay}, respectively); and polarization decay times ($\tau_{\mathcal{P}}$) for the six $GaAs_{1-x}P_x/Al_{0.35}Ga_{0.65}As$ QW's. The values of Δ, μ_{hh}^*, and μ_{lh}^* are obtained from Ref. 21.

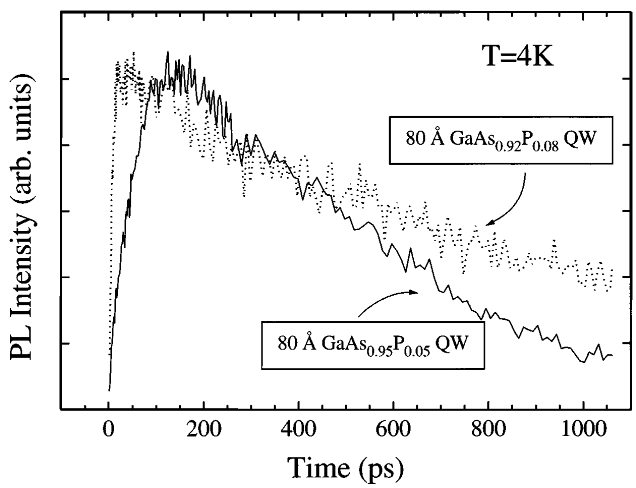

Fig. 4. Normalized time evolution of the sum of the σ^+ and the σ^- components of the PL for the 80-Å wide WQ's with P contents of 8% (dotted curve) and 5% (solid curve). In both cases the excitation was done above the continuum edge.

that exciton emission is dominated by the slow relaxation process from large-**K** excitons to **K** = 0 excitons. However, for the three wells that present a quasi-degenerate hh–lh excitonic ground state (A-120, B-80, and C-80), we observed a faster exciton relaxation, with rise times of the order of 60 ps. We also found that τ_{decay} depends on the hh or the lh character of the excitonic ground state. QW's that have an hh ground state present a decay time of ~500 ps, whereas the value is doubled to ~1 ns for QW's that have an lh state. This behavior is in agreement with the larger value of the oscillator strength of the hh transitions compared with that of the lh ones.

These results are illustrated in Fig. 4, in which the normalized time evolution of the sum of the σ^+ and the σ^- components of the PL is presented for the B-80 QW (dotted curve), which presents a quasi-degenerate hh–lh excitonic ground state, and for the A-80 QW (solid curve), which displays an hh excitonic ground state. Both time evolutions were recorded exciting above the continuum edge with the same excess energy of 60 meV. It can easily be appreciated from Fig. 4 that the rise time (decay time) of the PL is shorter (longer) for the QW with a quasi-degenerate hh–lh excitonic ground state than for the QW with an hh excitonic ground state.

4. SPIN RELAXATION

To investigate exciton spin relaxation we measured the time evolution of the degree of circular polarization of the PL, $\mathcal{P}$. The degree of circular polarization is defined, for one of the exciting helicities, as the fractional difference of the PL intensities of the two circular polarizations, σ^+ and σ^-, at a given energy, i.e., for σ^+ excitation, $\mathcal{P} = (I^+ - I^-)/(I^+ + I^-)$. We found that, for all the samples, the time evolution of $\mathcal{P}$ can be fitted to a single exponential decay. Therefore we used the decay time of $\mathcal{P}$, $\tau_{\mathcal{P}}$, as an effective spin relaxation time.[15]

In the case of GaAs/AlAs MQW, we observed that exciton dynamics and its dependence on excitation energy (see Fig. 1) strongly influence the spin relaxation processes. The dependence of $\tau_{\mathcal{P}}$ on excitation energy is shown in Fig. 5 (filled circles) together with the cw PLE spectrum (curve). The inset shows the time evolution of the polarization (open circles) and the best fit to a monoexponential decay (line), for which $\tau_{\mathcal{P}} = 30$ ps, exciting at 1.63 eV. The ~20% larger value of $\tau_{\mathcal{P}}$ at the hh excitonic peak compared with that at the subband edge demonstrates the influence of the exciton wave vector on spin relaxation, and, moreover, it evidences that exchange interaction is the leading spin relaxation mechanism. With excitation at the subband continuum, large-**K** excitons are created from uncorrelated electron–hole pairs, whereas with excitation at the hh exciton peak in the PLE excitons are

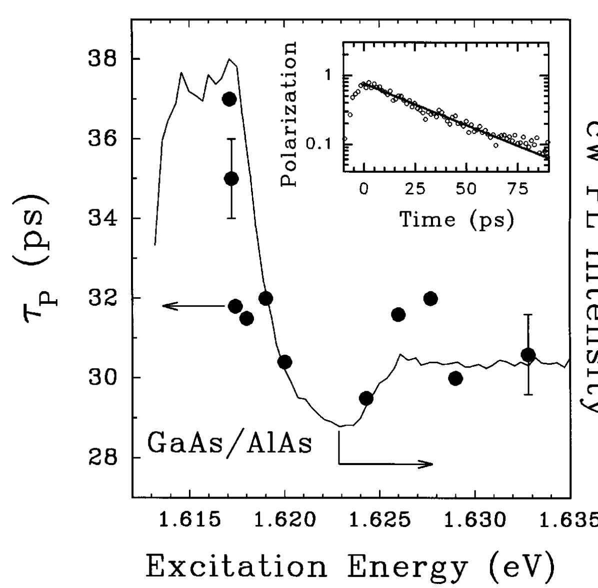

Fig. 5. Dependence of the polarization decay time (filled circles) as a function of excitation energy for the GaAs/AlAs MQW. The solid curve depicts the PLE spectrum. The inset shows the time evolution of the polarization (open circles) and the best fit to a monoexponential decay (line) exciting at 1.63 eV.

created resonantly with $\mathbf{K} = 0$. As the long-range part of the exchange interaction is proportional to the exciton wave vector,[14] the observed dependence of $\tau_{\mathcal{P}}$ on excitation energy should be expected if spin relaxation were due to exchange interaction. The decrease of $\tau_{\mathcal{P}}$ in the high-energy tail of the hh excitonic peak can also be linked to an increase in the long-range exchange interaction, inasmuch as it has been proposed that the high-energy tail in the pseudoabsorption arises from a partial violation of the **K**-conserving rule, which permits the creation of large-**K** excitons.[22] The influence of exciton localization on the spin relaxation processes was discussed in detail in Ref. 15.

Let us now discuss the results concerning exciton spin relaxation in the tensile-strained $GaAs_{1-x}P_x/Al_{0.35}Ga_{0.65}As$ QW's. In these systems the PL signal is much less intense than in the GaAs/AlAs MQW, so we have not been able to perform quasi-resonant experiments.[23] However, as is addressed below, we also found indications on the influence of the exciton wave vector on spin relaxation. The measured values of $\tau_{\mathcal{P}}$ for excitation above the continuum edge are reported in Table 1. For the B-120 and the C-120 QW's these values are missing, as we found that the degree of polarization is negligible at all times.

For the A-80 QW, which presents a normal hh–lh configuration, the polarization decay time is ~100 ps, much longer than for the GaAs/AlAs system (~30 ps). This increase in the polarization decay time should be expected as a consequence of the relation between spin relaxation time and carrier scattering time. For exchange-driven spin relaxation events a shorter carrier scattering time implies a longer spin–flip time.[14] Alloy scattering in the ternary QW's decreases the scattering times, so spin relaxation times are expected to be longer in the $GaAs_{1-x}P_x/Al_{0.35}Ga_{0.65}As$ system than in the GaAs/AlAs MQW, in agreement with the result for the A-80 QW. However, for the three QW's that exhibit a quasi-degenerate hh–lh excitonic ground state (A-120, B-80, and C-80), the measured spin relaxation times are ~35 ps. This reduction in the spin–flip time can be attributed to the influence of band structure on spin relaxation. As was shown in Ref. 21, with decreasing hh–lh subband separation the hh– and the lh-subband effective masses increase. Therefore the three QW's with quasi-degenerate hh–lh excitonic ground state have large excitonic masses, which implies that excitons created from uncorrelated electron–hole pairs have a larger center-of-mass wave vector **K**. The long-range part of the exchange interaction is proportional to the exciton wave vector,[14] so shorter spin-relaxation times are expected for the QW's with larger excitonic masses. Moreover, the probability of spin relaxation resulting from the short-range part of the exchange interaction is inversely proportional to energy splitting between the hh and the lh excitons.[14] Therefore those QW's that present a quasi-degenerate hh–lh excitonic ground state should have shorter spin relaxation times, as is borne out by our experiments.

Finally, it should be noted that, at the excitation densities used in our experiments ($\sim 5 \times 10^{10}\ \mathrm{cm}^{-2}$), many-body effect, especially exciton–exciton interaction, influence the dynamics of exciton and spin relaxation. A splitting of energy between spin-up and spin-down excitons, with as much as half the exciton binding energy, has been observed[11,24] and has been attributed to interexcitonic exchange interaction.[24]

5. SUMMARY

We have investigated the influence of subband structure on exciton dynamics and spin relaxation. The studies have been done by picosecond time-resolved photoluminescence spectroscopy in a GaAs/AlAs MQW and in tensile-strained $GaAs_{1-x}P_x/Al_{0.35}Ga_{0.65}As$ QW's. We have found that the exciton relaxation time decreases for the quasi-degenerate hh–lh excitonic ground state and that the recombination time depends essentially on the character of the excitonic ground state, being larger for those QW's with an lh excitonic ground state. Our experimental results show that the spin relaxation time decreases for the QW's with larger excitonic masses; it also decreases to increase the excitation energy from the hh excitonic peak to the subband edge. Both results are in agreement with the theoretical predictions for the exchange-driven spin relaxation time; the theory anticipates that the spin–flip times become shorter when the exciton wave vector is larger.

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