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EXCITON DYNAMICS AND SPIN-FLIP IN TENSILE STRAINED QUANTUM WELLS

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Abstract—We have studied the exciton dynamics and spin–flip processes in tensile strained $GaAs_{1-y}P_y/Ga_{0.65}Al_{0.35}As$ quantum wells as a function of phosphorous composition and well width. The strain introduced by the presence of phosphorous modifies the valence band structure. This strongly affects the characteristic times of exciton formation and polarization decay. However, the recombination time is essentially determined by the light- or heavy-character of the excitonic ground state.

Exciton dynamics and spin flip processes have been extensively studied in GaAs quantum wells (QW's) which present a "normal" configuration, where the ground state in the valence band corresponds to the heavy hole exciton[1]. Tuning of the ground state between the heavy hole (hh) exciton, light hole (lh) exciton or a degenerated hh–lh exciton state can be achieved by the presence of strain in the quantum well[2].

Biaxial tension in a quantum well can be caused by the lattice mismatch between the materials forming the well and the barrier, as in $GaAs_{1-y}P_y/Al_xGa_{1-x}As$ QW's[3]. This tensile strain, introduced by the presence of phosphorous, shifts the hh and lh valence subbands to lower energies, leading to a reduction of the gap. The amount of band-edge shift of the lh subband is larger than that of the hh one. Thus, for certain combinations of phosphorous concentration and well widths, the lh subband can be at a lower energy than the hh subband, obtaining an "inverse" configuration of the band structure where the lh exciton is the ground state.

We present here the influence of the valence band structure on the exciton dynamics and spin flip processes in QW's. We have studied the initial degree of polarization of the photoluminescence (PL) and the characteristic times of exciton formation, recombination and polarization decay, as a function of the energy separation between the hh and the lh subbands, $\Delta = E_g(\mathrm{lh}) - E_g(\mathrm{hh})$. For this purpose, we have investigated a series of $GaAs_{1-y}P_y/Ga_{0.65}Al_{0.35}As$ QW's with different phosphorous compositions and well widths.

The samples, kept at 5 K, were excited in a back scattering geometry with pulses from a Styryl 8 or a Pyridine 1 dye laser, tunable from 810 to 700 nm, synchronously pumped by a mode-locked Nd:YAG laser. The PL was time resolved in an up-conversion spectrometer with a time resolution of ~5 ps. The exciting light was circularly polarized by means of a $\lambda/4$ plate. We estimate from the power density of our excitation an initial carrier density of the order of $5 \times 10^{10}\,\mathrm{cm}^{-2}$. In order to obtain the degree of polarization $\mathbf{P}(t) = (I^+(t)\text{-}I^-(t))/(I^+(t) + I^-(t))$, the PL was analyzed into its σ^+ and its σ^- components, using a second $\lambda/4$ plate. We have investigated six different QW's, grown by MOCVD[2], with phosphorous compositions of 5, 8 and 12%, well thicknesses of 80 Å and 120 Å, separated by 400-Å $Ga_{0.65}Al_{0/35}As$ barriers. An additional unstrained 120-Å GaAs QW, grown on the same samples, was used as a reference.

The samples have been investigated previously by means of cw photoluminescence excitation spectroscopy (PLE) in the presence of an external magnetic field applied in the Faraday configuration[4]. These studies have yielded the exciton binding energies, in-plane valence band effective masses and hh–lh subband separations (summarized in Table 1), which allow us to know the band structure for the different QW's.

Figure 1 compiles the PLE spectra for the seven different QW's. The smallest peak in each spectra corresponds to the light-hole exciton, while the largest one corresponds to the heavy-hole exciton. Those spectra which show only one structure, correspond to a quasi-degenerated case. It is clearly seen in the figure that the relative positions of the hh and lh excitonic states depend on well thickness (d) and phosphorous composition (y). The reference 120-Å GaAs QW and the ($d = 80$ Å, $y = 0.05$)-GaAsP QW present a "normal" configuration, with the hh exciton as the ground state. The (d = 120 Å,

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Table 1. Subband separation between hh- and lh-subbands, $\Delta = E_g(\mathrm{lh}) - E_g(\mathrm{hh})$, phosphorous composition, y, well width, d, hh and lh inverse masses, and characteristic times of the PL (τ_{rise}, τ_{decay}) and of the polarization decay, τ_p, for the different QW's used in the experiments. The values of the first five columns are obtained from Ref.[4]

Δ (meV)	y	d (Å)	$(m_{\mathrm{hh}})^{-1}$	$(m_{\mathrm{lh}})^{-1}$	τ_{rise} (ps)	τ_{decay} (ps)	τ_p (ps)
−16.9	0.12	120	4.2	3.6	160 ± 10	900 ± 50	—
−12.4	0.08	120	2.5	1.5	140 ± 10	950 ± 80	—
−0.9	0.08	80	3.4	−2.7	40 ± 10	1020 ± 50	30 ± 10
−0.7	0.05	120	−0.8	−3.4	75 ± 8	1050 ± 100	32 ± 8
3.2	0.12	80	4.3	−1.1	58 ± 8	900 ± 100	38 ± 10
16.5	0.05	80	5.9	9.1	126 ± 8	570 ± 50	100 ± 30
	0	120			160 ± 10	440 ± 50	200 ± 50

$y = 0.08$) and ($d = 120$ Å, $y = 0.12$) QW's show an "inverse" configuration, where the ground state is the lh exciton, and the other three wells present a hh–lh quasi-degenerated excitonic ground state.

Figure 2 shows the time evolution of the total PL ($\sigma^+ + \sigma^-$ emission) for the ($d = 80$ Å, $y = 0.08$) QW, with a quasi-degenerated lh excitonic ground state (broken line) and for the ($d = 80$ Å, $y = 0.05$) QW, which presents a hh excitonic ground state (solid line). The intensities have been normalized to their maximum value. However, the intensity of the broken trace is approximately three times smaller than the solid one, as it corresponds to its light-hole character. Both wells were excited above the continuum edge with the same excess excitation energy (ΔE), measured from the ground state, of 60 meV. It is easily appreciated that the rise time (decay time) of the PL is shorter (longer) for the QW with a quasi-degenerated lh ground state than for the QW with a hh ground state.

The current knowledge of the exciton dynamics, after optical excitation, separates the relaxation process, which follows the creation of an electron–hole pair of the continuum, in two steps[5]. First, the electron–hole pair relaxes its energy and forms an exciton with non-zero kinetic energy in a time scale of 10 picoseconds. Secondly, the hot exciton loses its energy cooling down towards the exciton band-edge, forming a $\mathbf{K} = 0$ exciton, which can recombine emitting a photon.

In order to study the dependence of exciton dynamics on the hh–lh separation, we use the rise time of the PL (τ_{rise}) and the decay time of the PL (τ_{decay}) to characterize the formation time of $\mathbf{K} = 0$ excitons and the lifetime of the excitonic ground state, respectively. The values of τ_{rise} and τ_{decay}, exciting above the continuum edge, for the different QW's are compiled in Table 1. We have found that τ_{rise} depends strongly on the band structure. The rise time for the QW's with a quasi-degenerated excitonic ground state is much shorter (~50 ps) than that of the QW's with a hh or lh excitonic ground state (~130 ps). The shortest value of τ_{rise}, 40 ps, appears for the ($d = 80$ Å, $y = 0.08$) QW, which has $\Delta \cong 0$ and a quasi-degenerated lh excitonic ground state. These results can be explained in terms of the effective masses. Table 1 shows that the hh and lh effective masses become larger for Δ close to zero. This also implies very large *total* masses of the hh and lh excitons. Thus, the hh and lh excitonic subbands become very flat and close to each other, which

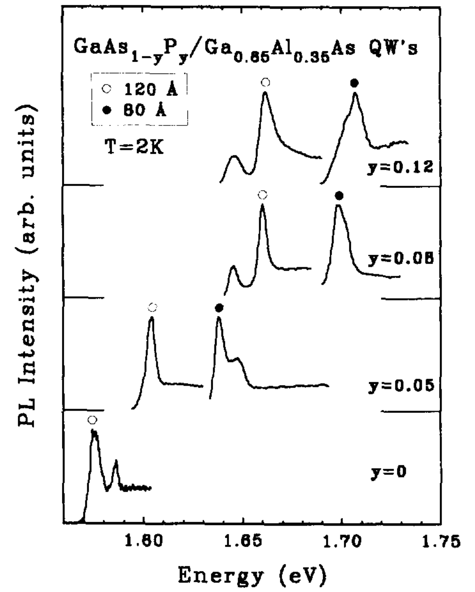

Fig. 1. Low temperature (2 K) PLE spectra of $GaAs_{1-y}P_y/Ga_{0.65}Al_{0.35}As$ QW's of 80 Å (●) and 120 Å (○) widths and different P concentrations, y. The measurements were performed exciting with σ^+ polarized light and detecting the σ^- emission.

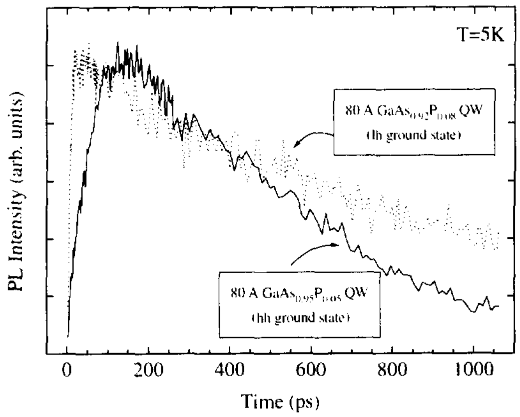

Fig. 2. Normalized time evolution of the total PL ($\sigma^+ + \sigma^-$ emission) at 5 K for the 80-Å $GaAs_{0.92}P_{0.08}$ QW (broken line) and the 80-Å $GaAs_{0.95}P_{0.05}$ QW (solid line). The excitation was done above the continuum edge with σ^+ polarized light.

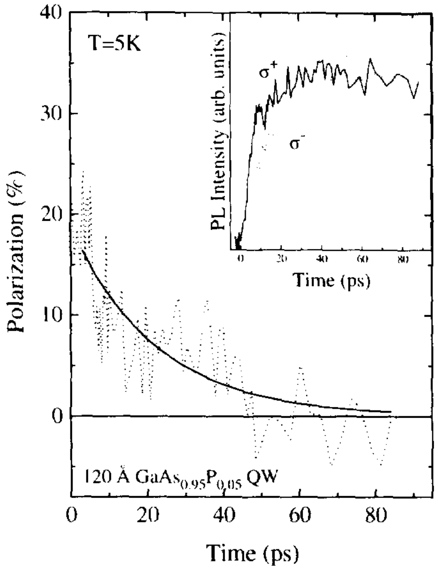

Fig. 3. Time evolution of the degree of polarization, P(t), (dotted line) for the 120-Å $GaAs_{0.95}P_{0.05}$ QW excited above the continuum edge with σ^+ polarized light. The solid line depicts a fit to a mono-exponential decay. The inset shows the initial time evolution of the σ^+ (solid line) and σ^- (dotted line) components of the PL. The measurements were performed at 5 K.

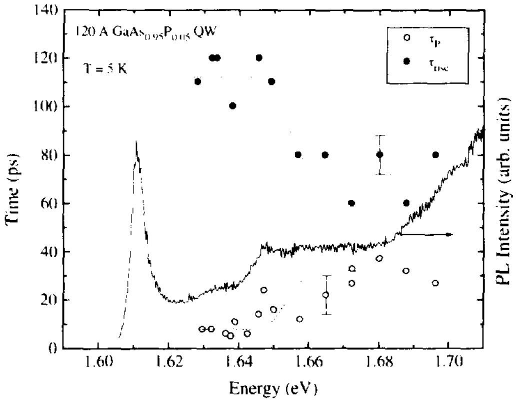

Fig. 4. Dependence of τ_P (open circles) and τ_{rise} (solid circles) on excitation energy for the 120-Å $GaAs_{0.95}P_{0.05}$QW. The two dashed lines are guides to the eye to show the sharp decrease (increase) of τ_{rise} (τ_P) for ΔE larger than ~36 meV. The corresponding PLE spectrum for this QW is included.

increases the transition probability between them and, therefore, decreases τ_{rise}.

On the contrary, τ_{decay} does not depend on Δ but only on the hh- or lh-character of the ground state. QW's having a hh ground state present a recombination time of ~500 ps, while the value is doubled to ~1 ns for QW's having a lh ground state. This dependence is in agreement with the larger value of the oscillator strength of the hh transitions than that of the lh ones[6].

We have also measured the time evolution of the degree of polarization, P(t), in order to study the spin flip processes in our tensile strained QW's. The inset in Fig. 3 depicts the initial time evolution of the polarized (σ^+, solid line) and unpolarized (σ^-, dotted line) PL for the ($d = 120$ Å, $y = 0.05$) QW excited with σ^+ polarized light above the continuum edge. It is clearly observed that the evolution of the unpolarized PL lags behind that of the polarized one, giving rise to a net degree of polarization. The corresponding time evolution of P(t) is shown in Fig. 3, dotted line, together with a fit to a mono-exponential decay, solid line. The exponential fit of P(t) yields a decay time, τ_P of ~30 ps and an initial degree of polarization, P(t_0), of ~17%. P(t) evolves from positive values for the ($d = 80$ Å, $y = 0.05$) and the ($d = 120$ Å, $y = 0.05$) QW's, which have hh and a quasi-degenerated hh excitonic ground states, respectively, to negative values for the ($d = 80$ Å, $y = 0.12$) and the ($d = 80$ Å, $y = 0.08$) QW's, which have lh and a quasi-degenerated lh excitonic ground states, respectively, and it is negligible for the ($d = 120$ Å, $y = 0.08$) and the ($d = 120$ Å, $y = 0.12$) QW's, which present a lh excitonic ground state. In the case of negative values of P(t), the initial degree of polarization can be as large as 20%.

Figure 4 shows the dependence of τ_{rise} and τ_P on excitation energy for the ($d = 120$ Å, $y = 0.05$) QW. The rise time decreases from ~110 ps to ~75 ps and, concomitantly, τ_P increases from our time resolution, ~5 ps, to ~30 ps as the excitation energy becomes larger. For excitation energies above the continuum edge, τ_P is ~30 ps for all the QW's which present a non-vanishing P(t), except for those which present a "normal" situation, ($d = 80$ Å, $y = 0.05$) QW and the reference GaAs QW, which have longer values of τ_P, ~100 ps and ~200 ps, respectively (see Table 1).

Various mechanisms have been invoked in the literature to explain the spin-flip processes in QW's[7,8]. Some of them refer to single particle spin-flip and others to the simultaneous spin-flip of the two particles, electron and hole, which form the exciton. The spin-flip of electrons has been attributed to D'yakonov-Perel (DP)[9] and Bir-Aranov-Pikus (exchange)[10] mechanisms, while that of the holes is believed to be related with valence band mixing[11,12]. Exciton spin-flip can occur either via the consecutive spin-flip of the electron and hole or via the simultaneous spin-flip of the two particles by means of the exchange interaction[13]. The spin-flip times resulting from the exchange and DP mechanisms are inversely proportional to the scattering times[8]. The spin-flip processes of individual electrons and/or holes give rise to an initial rapid decay of the PL due to the creation of non-optically active excitons[14]. The exchange driven mechanism is believed to be the most efficient one to produce spin-flip processes between optically active excitons. This mechanism is strongly enhanced for excitons with large values of their center of mass momentum, K[13].

The polarization decay time, τ_P, gives an estimation of the effective spin–flip time involving the different spin–flip processes which occur during the exciton relaxation. The short value of τ_P (~ 30 ps), found for all the QW's which present a quasi-degenerated valence band edge, can be also explained in terms of the band structure. These QW's present values of Δ close to zero which means, as previously mentioned, very flat subbands. Exciting with the same excess energy, ΔE, excitons with larger **K** are created when the dispersion relations are flatter. Since the probability of exciton spin–flip processes is enhanced for large **K** excitons, a shorter spin–flip time is expected for those QW's with small values of Δ, as born out from our experiments. Moreover, the probability of spin–flip, due to the short-range exchange interaction, is inversely proportional to Δ[13]. This also decreases τ_P for QW's having small values of Δ. On the other hand, Fig. 4 shows that τ_P increases from ~ 5 ps to ~ 30 ps with increasing excess excitation energy, ΔE. The sharp decrease of τ_{rise} for ΔE larger than ~ 36 meV can be attributed to the onset of the LO phonon scattering contribution in the process of excitonic cooling. The decrease of τ_P to ~ 5 ps when ΔE is lower than 36 meV is directly related to the larger value of τ_{rise} for this energy range. A larger value of τ_{rise} implies that the excitons spend a longer time with $\mathbf{K} \neq 0$, which means a higher probability of spin–flip processes by means of the exchange interaction mechanism.

Finally, the longer values of τ_P for the reference GaAs QW and the "normal" GaAsP QW, as compared with those obtained in high quality GaAs QW's[1,7,14], are compatible with the fact that the present samples are grown by MOCVD. Since their quality is worse, the scattering times with impurities and defects are shorter, and therefore, if the spin–flip mechanism is either DP or exchange, an increase of τ_P follows. Alloy scattering in the ternary QW should also decrease the scattering times.

In summary, we have studied the spin–flip relaxation, the exciton formation and its recombination dynamics in tensile-strained quantum wells by means of time-resolved photoluminescence spectroscopy. The recombination times depends essentially only on the heavy- or light-character of the excitonic ground state, being approximately a factor of two larger for those quantum wells with a hh excitonic ground state. However, the formation and spin–flip times show a strong dependence on the details of the valence band structure. For degenerated hh and lh subbands, the rise time and spin–flip time are considerably smaller than those obtained in the case of non-degenerated subbands.

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