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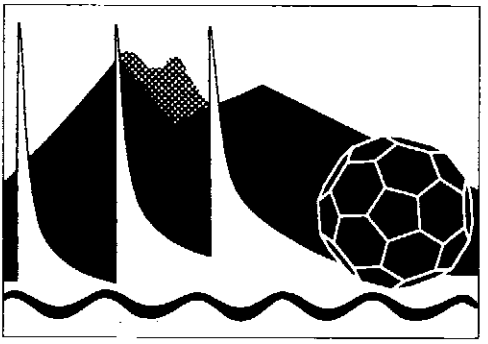

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DYNAMICS AND SPIN RELAXATION OF PHOTOCREATED CARRIERS IN INTRINSIC GaAs/AlAs QUANTUM WELLS

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ABSTRACT

Exciton dynamics and spin relaxation have been investigated in GaAs quantum wells by picosecond time-resolved photoluminescence spectroscopy. The studies have been done as a function of excitation energy, lattice temperature and carrier density. For resonantly created excitons, we have found that hole spin-relaxation is strongly dependent on excitonic localization. The excitation energy dependence of the polarization decay time indicates that exchange interaction is the leading spin relaxation mechanism.

We have investigated exciton dynamics and spin relaxation in a GaAs/AlAs multiquantum well, consisting of 50 periods of nominally 77 Å-wide GaAs wells and 72 Å-wide AlAs barriers. The sample presented, at 2 K and under very low excitation densities, a small Stokes shift of $\sim$2.5 meV. Time-resolved photoluminescence experiments were performed in a standard up-conversion spectrometer with a time resolution of 6 ps. The exciting light was circularly polarized by means of a $\lambda/4$ plate, and the photoluminescence (PL) was analyzed into its σ^+ and σ^- components using a second $\lambda/4$ plate before the non-linear crystal. A double-grating monochromator was used to disperse the upconverted signal.

We have found excitonic-dynamical properties analogous to the observed in very high quality samples with vanishing Stokes shifts.[1] Figure 1 shows, for different temperatures (T), time evolutions of the sum of the two components of the PL, exciting above the subband edge with an initial carrier density of 4×10^{10} cm^{-2}. The long rise times are due to the slow cooling of the excitons with large in-plane center-of-mass momentum ($\boldsymbol{K}_\perp$) created by electron-hole pairs of the continuum. We have fitted the time evolutions to a two-level dynamical model which include two characteristic times, the formation time of excitons with $\boldsymbol{K}_\perp=\boldsymbol{0}$, τ_f, and the exciton decay time, τ_d. The solid lines in the

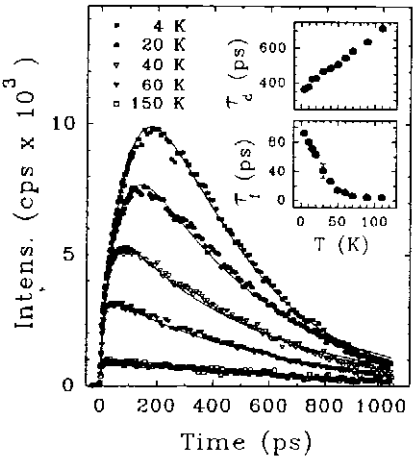

Figure 1. Time evolutions of the PL for different temperatures. The solid lines depict the fits to a dynamical model (see text). The insets show the T dependence of the decay and formation time.

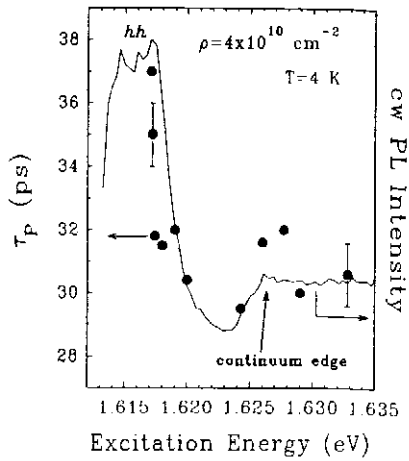

Figure 2. Dependence of the polarization decay time (filled circles) as a function of excitation energy. The solid line shows the PLE spectrum.

figure depict the results of the fits. The insets show the T dependence of τ_d and τ_f. The monotonic linear increase of τ_d is characteristic of free excitons,[2] and indicates that localization in our sample is relatively weak. The decrease of τ_f with increasing temperature, is a consequence of the spreading of the thermalized exciton distribution around $\boldsymbol{K}_\perp \approx \mathbf{0}$ as temperature increases.[1] Therefore, the initial distribution of large $\boldsymbol{K}_\perp$ excitons approaches faster the final $\boldsymbol{K}_\perp \approx \mathbf{0}$ distribution at high temperatures. We have also found a decrease in τ_f increasing carrier density, indicating that exciton-exciton collisions are important in the relaxation processes of large $\boldsymbol{K}_\perp$ excitons.[1]

In variance with the case of exciting above the subband-edge, setting the excitation energy at the heavy-hole (*hh*) exciton, the rise time of the PL decreases strongly and becomes of the order of our time resolution. We have found that this modification of exciton dynamics is revealed on spin relaxation processes. In fact, the polarization decay time (τ_p) increases in a $\sim$20% moving the excitation from the subband continuum to the peak of the *hh* exciton in the photoluminescence excitation spectrum (PLE). This is shown in Fig. 2, where the dependence of τ_p on excitation energy (E_{ex}) is displayed. We believe that this behavior is an indication of exchange interaction being the leading spin relaxation mechanism. Exciting at the subband continuum, large $\boldsymbol{K}_\perp$ excitons are created from uncorrelated electron-hole pairs, while exciting at the *hh* exciton peak in the PLE, excitons are created resonantly with $\boldsymbol{K}_\perp \approx \mathbf{0}$. As the long range part of the exchange interaction is proportional to the exciton wave vector,[3] the observed dependence of τ_p on E_{ex} should be expected if spin relaxation were due to exchange interaction.

Exciting at the *hh* exciton, a fast initial decay in the time evolution of the

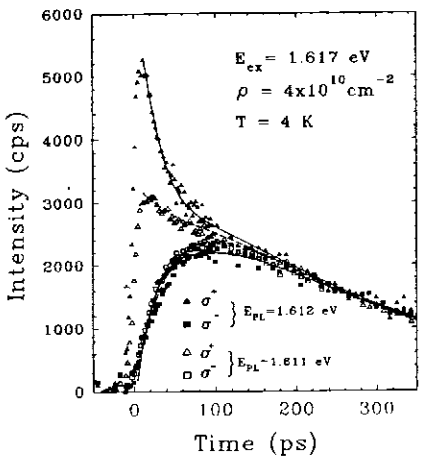

Figure 3. Evolutions of the σ^+ (triangles) and σ^- (squares) PL detecting at 1.612 eV (black symbols) and at 1.611 eV (hollowed symbols). The lines show the fit to a dynamical model (see text).

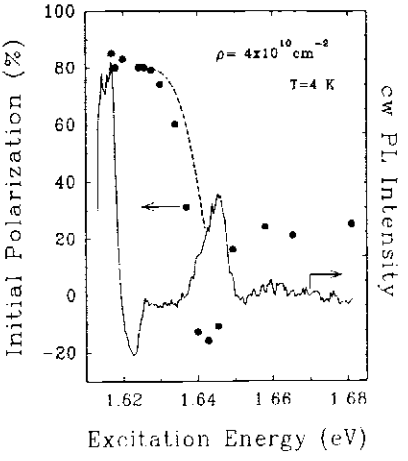

Figure 4. Dependence of the initial value of the polarization on excitation energy (circles). The dashed line shows the result of a calculation (see text). The solid line depicts the PLE spectrum.

PL with the same polarization as the exciting laser appears. Using a model which takes into account spin flip of the exciton, as well as spin flip of the electrons and holes individually, this behavior has been attributed to the removal of excitons from optically active states (spin ± 1) to dark states (spin ± 2), as a consequence of the spin flip of the holes.[4] We have fitted our data with the same model, and we have found that hole spin-relaxation is strongly dependent on detection energy. These findings are compiled in Fig. 3, where we show the time evolution of the σ^+ (triangles) and σ^- (squares) photoluminescence for two different detection energies, 1.612 eV -the maximum of the PL spectrum- (black symbols) and 1.611 eV (hollowed symbols). The black-time profiles are well fitted (solid lines) using a spin-flip time for the holes (electrons) of $\tau_h = 25 \pm 8$ ps ($\tau_e = 200 \pm 100$ ps) and an exciton spin-flip time of $\tau_x = 35 \pm 10$ ps. For the profiles at 1.611 eV, the values from the fit (dashed lines) are $\tau_h = 5 \pm 5$ ps, $\tau_x = 20 \pm 10$ ps, and the rest of the times the same as those used detecting at 1.612 eV.

We believe that the strong dependence of τ_h on detection energy is a consequence of the influence of excitonic localization on the spin-flip time of holes correlated to electrons. Bastard et al have calculated that, for localized excitons on interface defects with the same depth, a change in lateral size of $\sim$ 100 Å produces a variation of $-$ 1 meV in their binding energy.[5] Therefore, changing the detection energy from 1.612 eV to 1.611 eV, we are sampling stronger localized excitons with larger uncertainties in the in-plane center-of-mass momentum, $\Delta K_\perp$. As $K_\perp = k_{e\perp} + k_{h\perp}$, where $k_{e\perp}$ and $k_{h\perp}$ are electron and hole in-plane wave vectors, the uncertainty in momentum of correlated electrons and holes depends on the degree of excitonic localization. Hole-spin relaxation, which is due to valence band mixing, shows a strong dependence on $k_{h\perp}$; the relaxation rate increases with increasing the wavevector.[6] Therefore, for stronger localized excitons,

which have a larger $\Delta k_{h\perp}$, a shorter spin-flip time of the holes is expected.

We have found that the initial degree of polarization, $\mathcal{P}(t_0)$, is strongly dependent on excitation energy. $\mathcal{P}(t_0)$ is defined as the value of the polarization at time $t_0 = 3$ ps (half the width of the laser pulse). Figure 4 shows that exciting below the light-hole (*lh*) exciton, $\mathcal{P}(t_0)$ increases as the excitation energy decreases. For E_{ex} corresponding to the *lh* exciton, $\mathcal{P}(t_0)$ becomes negative and reaches values of about -20%. This negative value of the polarization is due to excitonic effects, which enhance the creation of light holes and electrons with spin +1/2. To observe the luminescence at the *hh* exciton, holes have to relax to ±3/2 spin states. Since the spin-flip time of the electrons is considerably larger than t_0,[7] a significant σ^- emission involving +1/2 and -3/2 holes is obtained.

The dependence of $\mathcal{P}(t_0)$ on E_{ex}, exciting below the *lh* exciton, is due to band mixing in the valence band, which causes that for σ^+ excitation, electrons from the first *hh* subband are promoted not only to -1/2 spin conduction-band states, but also to +1/2 spin states. We have calculated the relative initial populations of +1/2 and -1/2 spin conduction electrons, following the absorption of a σ^+ pulse, in the envelope function approximation taking valence band mixing into account.[6] To obtained $\mathcal{P}(t_0)$, we assumed, for simplicity, a complete hole-spin relaxation and neglected electron spin relaxation; an exciton spin-relaxation time of 30 ps justifies that at the subband edge, where band mixing is negligible, the maximum value of $\mathcal{P}(t_0)$ is only 80%. The calculated degree of polarization, taken as the fractional difference between the populations of +1 and -1 excitons, is plotted as a dashed line in the figure.

In summary, we have found that localization, even being not strong enough to change exciton dynamics, modifies markedly hole spin relaxation. We have obtained indications of exchange interaction being the leading spin-relaxation mechanism, and we have proved that valence band mixing determines the initial degree of polarization.

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