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Femtosecond spectroscopy of carrier-spin relaxation in GaAs-$Al_xGa_{1-x}As$ quantum wells

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We investigate the spin relaxation of optically excited charge carriers in a variety of GaAs-$Al_xGa_{1-x}As$ quantum wells and superlattices at low temperatures by ultrafast time-resolved photoluminescence spectroscopy. Structures with uniform static optical properties show a markedly varied spin scattering in the time domain, thereby requiring a series of studies with systematically modified epitaxial growth conditions. A multiexponential spin relaxation is seen in quantum wells with a fast initial component of less than 1 ps and a subsequent relaxation within $\sim$150 ps. For the same growth conditions, the polarization decay time in superlattices approaches bulk values, indicating a dimensionality dependence of the spin scattering.

Polarization-dependent optical measurements have become an important tool to characterize electronic transitions in solids,[1] since the polarization state of the light directly reflects the symmetry properties of the carrier wave functions. While several different spin-relaxation mechanisms have been shown to be relevant in bulk semiconductors,[2] for quantum-well (QW) systems,[3] these dynamics are still poorly understood. Typically, spin-relaxation times τ_s are calculated from static optical pumping experiments, where the sample is excited with circularly polarized light of appropriate wavelength in order to generate spin polarized carriers. In the analysis it is generally assumed that the hole spin relaxation is instantaneous and the polarization ρ obeys the simple relation $\rho \sim (1+\tau/\tau_s)^{-1}$, where τ denotes the recombination time. However, the importance of the hole spin polarization has been pointed out in recent theoretical work, which shows that the confinement present in QW systems can lead to an incomplete hole spin relaxation.[4] In addition, the common assumption of a single spin-relaxation time in QW systems has not yet been directly verified experimentally. In order to gain further insight into spin scattering processes in quantum geometries it is important to directly investigate the spin dynamics by time-resolved optical pumping experiments as demonstrated recently for II-VI materials.[5]

Here we report on static and femtosecond time-resolved optical pumping experiments on GaAs-$Al_xGa_{1-x}As$ QW's and superlattices (SL's) grown by molecular-beam epitaxy. Surprisingly, our first time-resolved experiments on GaAs-$Al_xGa_{1-x}As$ multiple QW's (obtained from different sources) with uniform high-quality static photoluminescence (PL) spectra revealed a strong sample dependence of both the initial spin polarization and the spin-polarization decay time. This indicates that an analysis of static polarization spectra to determine spin-relaxation times is subject to large errors. Thus a series of QW and SL samples have been grown with systematically varied parameters. Under optimized growth conditions the QW systems reveal high initial polarizations in the time domain up to 100% and long-living polarization decays within about 150 ps. Furthermore, we observe interesting dynamical processes in the early time regime within the first few picoseconds. For the same growth conditions, a comparison of the polarization decay of SL-type samples and the QW's allows us to study the dimensionality dependence of the carrier spin-relaxation times.

The structures grown for these studies are GaAs-$Al_{0.3}Ga_{0.7}As$ single QW's, multiple QW's, and SL's. The layer sequence below these structures consists of a short period SL and a GaAs buffer layer, which is grown on a GaAs(100) substrate. During the epitaxy we introduce a combination of two additional processes. First, the growth is interrupted at every interface with interruption times between 20 and 120 s, allowing a redistribution of the surface atoms and thus a smoothening of the interface. Second, we periodically introduce GaAs monolayers within the $Al_xGa_{1-x}As$ barrier layers in order to reduce the roughness at the growth front.[6] Both procedures have an impact on the static polarization ratios of the ground-state excitonic transitions as illustrated in Fig. 1, which shows static optical pumping experiments in the wavelength range of the ground-state excitons. In GaAs the light-hole excitons reveal a negative polarization, which can be understood in terms of the high joint density of states of the light-hole excitonic transitions.[7] The most pronounced polarization ratios of about 30% and -10% for heavy holes and light holes, respectively, are observed when both additional processes are applied during the growth, the optimal growth-interruption time being 30 s. Therefore, interface scattering processes seem to play an important role for the spin relaxation in addition to phonon-scattering processes. It is interesting to note that long interruption times of 60 s or longer actually lead to additional carrier spin scattering and thus to poorer polarizations.

The static and time-resolved optical pumping experiments[8] are performed in an optical cryostat at 4.5 K. A cavity-dumped two-jet dye laser with Pyridine 2 laser dye is synchronously pumped by a frequency-doubled mode-locked Nd-doped yttrium-aluminum-garnet laser to produce optical pulse energies between 1.55 and 1.8 eV. The helicity of the light is adjusted in the excitation beam and analyzed in the detection beam by $\lambda/4$ plates, which are controlled by stepper motors. The static PL is analyzed by a 64-cm monochromator and a photomultiplier. In order to further reduce the dye-laser pulse width of 0.9 ps,

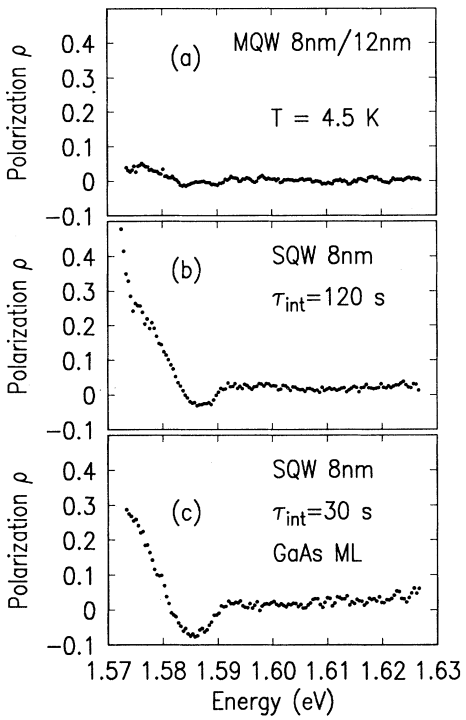

FIG. 1. Static polarizations $\rho=(\sigma^- - \sigma^+)/(\sigma^- + \sigma^+)$ obtained by excitation spectroscopy for (a) an 8-nm well-width and 12-nm barrier-width multiple QW grown with no additional growth processes, (b) an 8-nm well-width single QW grown with an interruption time at every interface τ_{int} of 120 s, and (c) an 8-nm well-width single QW with τ_{int} of 30 s and periodical insertion of GaAs monolayers (ML) within the $Al_xGa_{1-x}As$ barriers. The detection wavelength is set to the heavy-hole excitonic resonance. The average excitation intensity is 10 W/cm^2.

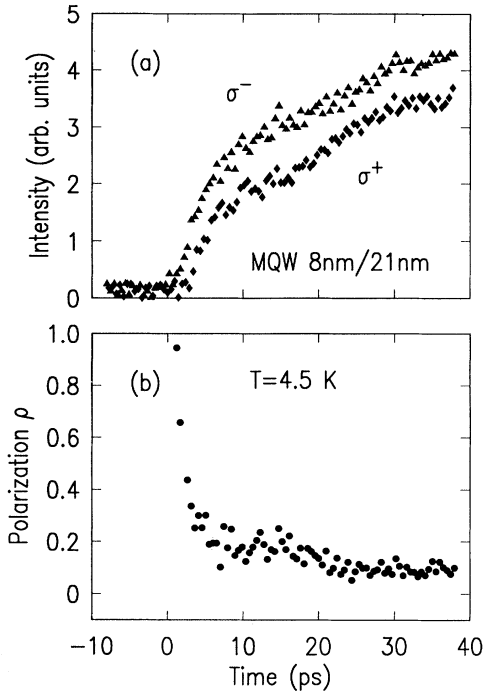

FIG. 2. (a) Time-resolved PL of a 10-period multiple QW sample with 8-nm well width and 21-nm barrier width grown under optimized conditions as explained in the text. The excitation energy is set 60 meV above the ground-state heavy-hole excitonic transition. The width of the optical pulse is 0.9 ps. The average excitation intensity is 30 W/cm^2. (b) Polarization $\rho=(\sigma^- - \sigma^+)/(\sigma^- + \sigma^+)$ obtained from (a).

the optical pulses are compressed in a fiber-prism arrangement to about 80 fs, which is determined by auto correlation assuming a sech^2 line shape. The time resolution of the PL is obtained by a pump and probe configuration using the sum-frequency generation optical gating technique.[9] The range of excitation wavelengths is limited by second-harmonic frequency generation, which dominates the PL signal if the detection and probe wavelengths differ by less than about 60 meV. The resulting signal is further analyzed by a monochromator and photon-counting techniques. The actual time resolution is limited to about 150 fs due to dispersion in the collection optics. The time zero position is determined by cross correlation of the probe pulse with pump light scattered from the sample. The time-dependent measurements are restricted to average excitation intensities below 100 W/cm^2, where excitonic effects are dominant. The linewidths of the excitonic transitions are, depending on the QW width, typically 5–10 meV and thus in the order of magnitude of the resolution of the detection system.

Figure 2(a) shows the time-resolved PL of a multiple QW sample for the two helicities σ^+ and σ^-. The sample was grown under optimized growth conditions with growth interruption at each interface and insertion of additional GaAs monolayers in the barriers. The optical excitation is performed with σ^- polarized light 60 meV above the ground-state heavy-hole excitonic transition, where the signal is detected. Within the first 100 ps, the PL signal increases monotonically, which reflects the filling of the heavy-hole ground state due to the energy relaxation of the excited carriers. The behavior of the spin polarization $\rho=(\sigma^- - \sigma^+)/(\sigma^- + \sigma^+)$ is plotted in Fig. 2(b). The time-dependence of the polarization reveals several interesting features, which are consistently observed for all QW samples grown under optimized conditions. (i) The initial spin polarization is very high, between 90% and 100%. (ii) The decay of the polarization is multiexponential. The initial decay occurs within time scales less than 1 ps, where the polarization drops to values between 20% and 30%. From investigations with longer time delays we find that the remaining polarization subsequently decays within about 150 ps. For other nonoptimized growth conditions, we find widely differing results.[10] The initial polarizations are lower and less reproducible, the fast initial decay is not clearly observed and the subsequent longer decay turns out to be more rapid (100 ps or less). The rather strong dependence of the time-resolved spin polarization on the growth conditions suggests that interface scattering processes play an important role for the understanding of these results. As a general trend, we observe longer polarization decay times for smoother interfaces, consistent with the stronger polarizations observed in static optical pumping experiments. The

time dependence of the spin polarization is a very sensitive indicator of the detailed sample structure, more sensitive than the commonly considered static optical properties such as the PL linewidth. In particular, the PL lifetimes, which are about 570 ps, appear to be relatively insensitive to the different growth conditions.

In order to explore the effects of dimensionality we compare, for the same optimized growth conditions, the results of the quasi-two-dimensional QW samples with corresponding SL structures representing quasi-three-dimensional systems. Figure 3(a) shows the time-resolved PL of a SL for the two different helicities σ^+ and σ^-, together with the corresponding time dependence of the polarization [Fig. 3(b)]. The experimental conditions are chosen as in the case of the QW samples. When comparing the time-resolved polarizations of the SL samples with the results obtained for the QW samples, the following differences become apparent. (i) The initial polarization reaches only one-half of the corresponding QW value. (ii) The decay of the polarization is again multiexponential but shows larger relaxation times. The initial polarization decay occurs within 5–10 ps, which is considerably longer than for the QW systems. From investigations with longer time delays we find that the subsequent polarization decay occurs within about 300 ps.

Due to miniband formation, the wave functions in a SL are extended throughout all SL layers. In particular, depending on the miniband overlap, the heavy- and light-hole states are mixed at the Γ point similar to the situation of the heavy-hole–light-hole degeneracy in bulk systems. As a consequence, the electrons and holes cannot be fully polarized by the optical excitation, which results in lower observed initial spin polarizations. The multiexponential spin relaxation is observed for all investigated quantum structures and suggests that the assumption of a single spin-relaxation decay does not hold. Moreover, no strong polarization change occurs after the fast initial spin-polarization decay, indicating that a quasiequilibrium condition has been reached. The polarization-decay times in the SL systems are at least a factor of 2 longer as compared to QW systems and approach corresponding times found for bulk GaAs,[11] suggesting that the carrier spin scattering is enhanced for lower-dimensional structures. These results may be explained in terms of the enhancement of the electron-hole correlation induced by the quantum confinement, which is also responsible for the enhancement of exciton-binding energies and oscillator strengths. Provided that the hole spin relaxation in QW and SL systems is must faster than the electron-spin relaxation, which has not yet been directly verified experimentally, the electron-spin relaxation can be considerably shortened by the stronger electron-hole exchange in QW's.[5]

Figure 4 shows time-resolved PL measurements on a multiple QW sample in the early time regime within the first 3 ps for the two different helicities σ^+ and σ^- [Fig. 4(a)], and the corresponding polarization decay curve [Fig. 4(b)]. Unexpectedly, the PL reveals a sharp maximum in particular for σ^- emission. This signal rapidly decays, depending on the QW sample, within about

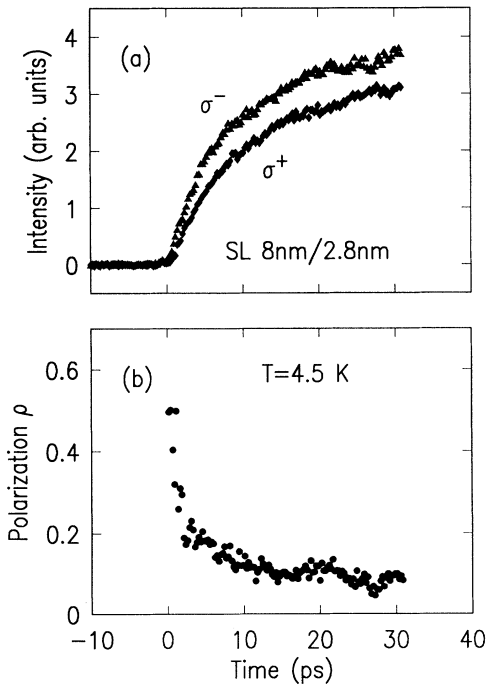

FIG. 3. (a) Time-resolved PL of a 10-period SL sample with 8-nm well width and 2.8-nm barrier width grown under optimized conditions as explained in the text. The experimental conditions are similar as for Fig. 2. (b) Polarization $\rho = (\sigma^- - \sigma^+)/(\sigma^- + \sigma^+)$ obtained from (a).

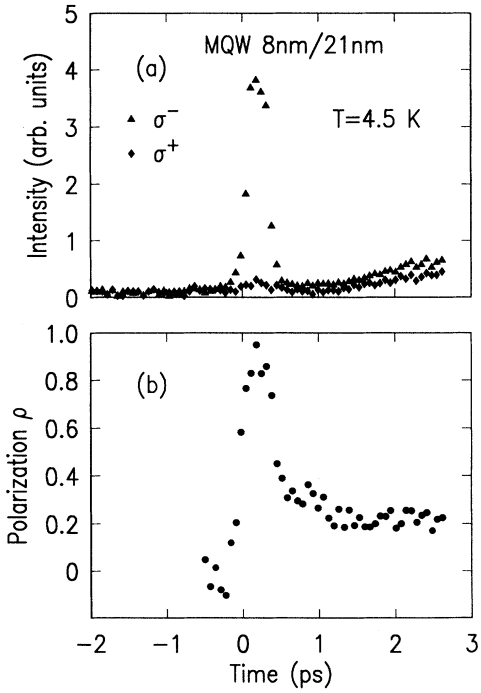

FIG. 4. (a) Time-resolved PL of the multiple QW sample investigated in Fig. 2 for the early time regime. The width of the optical pulse is 80 fs. The average excitation intensity is 100 W/cm^2. (b) Polarization $\rho = (\sigma^- - \sigma^+)/(\sigma^- + \sigma^+)$ obtained from (a).

300–500 fs. The corresponding polarization curve shows the same features already discussed above, namely, the high initial polarization and the rapid initial polarization decay. The origin of the sharp PL maximum, which is not observed in SL-type samples, is not understood so far. Note that, unlike typical pump-probe experiments, no coherent artifact can be generated using this experimental technique. The fast initial polarization decay seems to be related to the relaxation dynamics of the heavy-hole and light-hole excitons. Since the excitation wavelength is about 60 meV above the ground-state heavy-hole exciton transition, we simultaneously excite in all our measurements both heavy-hole and light-hole excitons. Despite this fact, our experiment reveals initial polarizations close to 100%, which are independent of the helicity in the excitation beam. Since at the moment of photoexcitation mainly excitons of heavy-hole type are observed, the rapid initial polarization decay could reflect the slower relaxation of light-hole-type excitons, which have an opposite spin orientation, thus causing the observed reduction of the spin polarization to about 20%–30%. However, presently we cannot draw any conclusion about the importance of hole spin-relaxation processes in this time frame, which could also be partly responsible for the fast initial polarization decay. This problem is especially interesting, since the hole spin relaxation is theoretically predicted to be considerably longer in QW systems.[4] A direct way to test the role of the hole spin relaxation is to perform a time-resolved optical pumping experiment by exciting the samples near resonance at the heavy-hole transition energetically below the light-hole transition. This experiment requires a two-laser technique in order to avoid second-harmonic frequency generation,[12] but with femtosecond time resolution.

In conclusion, we have performed femtosecond time-resolved optical experiments to study the carrier spin relaxation in QW and SL structures at low temperatures. In contrast to commonly considered static optical properties, the observed carrier spin scattering is very sensitive to the detailed, growth-determined structure of the samples. Only for QW samples grown under optimized conditions, the ground-state heavy-hole excitonic transition reveals high initial polarizations of almost 100% and long polarization-decay times of about 150 ps. These results are consistent with strongly pronounced polarizations, about 30% for heavy hole and about -10% for light-hole excitons as observed in static optical pumping experiments. In contrast to common assumptions, the polarization decays are found to be multiexponential with a short initial component of less than 1 ps. The optimized SL-type samples reflect, with initial polarizations of only 50% and polarization-decay times of about 300 ps, a more bulk-like behavior, indicating the importance of dimensionality for the carrier spin relaxation.

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