SPIN RELAXATION DYNAMICS IN GaAs QUANTUM WELLS: FREE CARRIERS AND EXCITONS

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We present the results of an investigation of the spin relaxation dynamics of electrons, holes and excitons in GaAs quantum wells by means of subpicosecond spectroscopy of the photoluminescence polarization. The band to band luminescence was studied in *n* and *p* modulation-doped quantum wells. We show that the spin relaxation for electrons and holes in quasi-two-dimensional systems is quite different from that in bulk semiconductors. A very high quality $GaAs/Ga_{0.7}Al_{0.3}As$ intrinsic multiple quantum well was used to investigate the spin relaxation of excitons. We demonstrate that the processes of spin relaxation of excitons are far more complicated than those of free carriers. Many-body effects and the formation dynamics of the excitons strongly influence their spin relaxation dynamics.

1. Introduction

The polarization of interband optical transitions in semiconductors provides important information about the symmetry of electron, hole and excitonic wave functions at the band edge. Optical orientation methods, which allow the excitation of states with a preferential spin orientation, using selective optical excitation with circularly polarized light, are very powerful for investigating relaxation processes in semiconductors.[1] The optical orientation manifest itself in a polarization of the photoluminescence (PL). The degree of polarization, ρ, for one of the exciting helicities, is defined as the fractional difference of the PL intensities of two circular polarizations σ^+ and σ^- at a given energy, as a function of the excitation energy: i.e., for σ^+ excitation, $\rho=(I^+-I^-)/(I^++I^-)$. The sign and the magnitude of the polarization depend on the transition which is excited and on the ratio between the lifetime and the spin relaxation time. The spin relaxation of free carriers and excitons has been thoroughly studied in bulk semiconductors during the last years. The most commonly used technique involves the use of circularly-polarized exciting light and the measurement of the polarization of the emission in steady state conditions. The application of a magnetic field in the Voigt geometry (Hanle effect) facilitates the interpretation of the data.[1] These studies have led to the identification of different spin relaxation processes in bulk semiconductors.[1-3]

In quantum wells (QW's), cw measurements of linear[4] and circular polarization[5-9] have been reported and several theories have been proposed to explain these results.[10-16] It is believed that the spin-relaxation processes are significantly modified in two-dimensional systems, especially for holes, due to the removal of the valence-band degeneracy at zone center.[14,15] Provided the time resolution is sufficient, the measurement of the polarization decay of the luminescence gives the spin relaxation time in a direct way. Time resolved spectroscopy of PL polarization has been reported recently for very high carrier densities[17] ($\sim 10^{19}$ cm^{-3}), with 150 fs time resolution, for intermediate carrier densities,[18] and for low densities.[19-22] The spin relaxation kinetics of excitons at room temperature in multiple QW's has been also investigated by time-resolved polarization absorption measurements.[23,24]

Although a considerable effort is being dedicated to the studies of spin relaxation in QW's, the mechanisms which determine the spin kinectis are not fully understood. The influence of the formation dynamics of excitons and of many-body effects must be contemplated. Furthermore, the differences in the behavior of electrons, holes and excitons need a careful experimental investigation.[14] From a practical standpoint, there is an important problem

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related to the sample quality. Localization effects, in non-high quality samples, can strongly modify the spin relaxation dynamics. Since localization varies in different samples, this can lead to a wide diversity in the results,[18] and therefore to hide the authentic nature of the spin-relaxation dynamics.

In this article we review our recent results on the spin dynamics of electrons, holes and excitons in GaAs QW's.[22] We have studied the spin relaxation of excitons in a, very high quality, intrinsic QW, in which the exciton shows a practically vanishing Stokes shift between the emission and the absorption and is nearly homogeneously broadened.[25] We have also used both n- and p-modulation doped QW's to investigate the hole and electron spin relaxation dynamics separately. We have performed the measurements as a function of temperature, excitation density and energy. In p-modulation-doped QW's, we have found that the electron spin-relaxation time is ~150 ps, a factor of 4 shorter than in comparably doped bulk GaAs. We have associated the increase in the relaxation rate to an enhanced electron-hole exchange interaction in QW's. Our measurements, in n-modulation-doped QW's, have shown that the usual hypothesis of instantaneous hole spin-relaxation is not valid in quasi-2D systems, since the polarization of the holes decays with a time constant of ~4 ps. We have interpreted the reduction of the hole spin-relaxation in terms of the lifting of the valence-band degeneracy at $k_{\parallel}=0$. In intrinsic QW's, we have found that the processes of spin relaxation of excitons are far more complicated than those of free carriers. Our experiments have shown that many-body effects play a very important role in the excitonic spin-relaxation: we have found a strong dependence of the spin-relaxation on excitation density as well as in the process of excitonic formation (resonant versus non-resonant). The polarized and unpolarized spectra present a conspicuous splitting that depends on time delay and excitation density. This splitting arises from many-body exchange interactions between excitons and contributes to an increase in the spin-relaxation time at higher densities.

The organization of this manuscript is the following: the experimental details are given in Sec. 2. The electron and hole spin relaxation are presented in Sec. 3 and 4, respectively. We discuss the exciton spin relaxation in Sec. 4. Finally, we summarize our results in Sec. 5.

2. Experimental details

We have studied three different GaAs/GaAlAs quantum wells. The intrinsic band to band luminescence was investigated in a high quality n (p) sample, consisting of 60 periods of $GaAs/Ga_{0.7}Al_{0.3}As$ with 50 Å (60 Å) wells and 280 Å barriers; the central 80 Å of the barriers were modulation doped with Si (Be). The doping densities, estimated from growth conditions, were $n_o=3 \times 10^{11}$ cm^{-2} and $p_o=4 \times 10^{11}$ cm^{-2}, respectively. The intrinsic sample was a $GaAs/Ga_{0.7}Al_{0.3}As$ multiple QW, consisting of 50 periods with 80 Å wells and 150 Å barriers. The photoluminescence FWHM for this sample obtained from cw measurements amounts to 0.65 meV at 10 K, and the Stokes shift between emission and absorption is almost negligible, indicating the high quality of the QW's. Previous studies on spin relaxation under resonant excitation[21] and on exciton formation and relaxation[25] have been published using the same sample.

The samples were mounted in a cold finger cryostat and excited with pulses from a synchronously pumped Styryl 8 dye laser, tunable from 810 nm to 720 nm. The width of the pulses was of 0.5 ps, except for excitation <20 meV from the detection energy, in which case the pulse width was increased to 1 ps. The spectral resolution was ~10 meV, limited primarily by the laser spectral width; the luminescence was time resolved using an up-conversion spectrometer.[26] The helicity of the exciting light was selected by means of a quarter-wave plate, while the emitted light was analyzed into its σ^+ component using a second quarter-wave plate, mounted in the detection arm of the spectrometer before the non-linear crystal used for the up-conversion, without altering the zero time of our setup. Identical results were obtained by keeping the polarization of the exciting light fixed and rotating the analyzer quarter-wave plate.

3. Electron spin relaxation

In the parabolic approximation of the dispersion relations, the conduction band states in QW's are eigenstates of the spin operator σ. The spin mixing appears in the anisotropic cubic term, which is the lowest correction to the parabolic dispersion law.[10,16] This leads to a mixing term which can be considered as a Zeeman term due to a magnetic field lying in the layer plane.[16] The D'yakonov-Perel' (DP) mechanism for the spin flip of the electrons is due to a precession of the electron spin in the effective magnetic field caused by the spin-orbit splitting of the conduction band in III-V semiconductors. The DP mechanism is believed to be the most important one to determine the spin relaxation kinetics of electrons at high temperatures.[27,28] Another efficient spin-relaxation mechanism has also been proposed by Elliot and Yafet[29] (EY) for materials with small direct gaps, large spin-orbit splittings, and high hole concentrations. In this mechanism the spin flip results from electron-phonon or electron-impurity interactions. In heavily doped materials, the exchange interaction between free carriers contributes an additional relaxation of the spin as proposed by Bir-Aranov-Pikus[29,30] (BAP).

Figure 1 shows the cw PL, photoluminescence excitation (PLE) for $\sigma^+\sigma^+$ (I^+) and $\sigma^-\sigma^+$ (I^-) configurations and luminescence polarization (ρ) as a function of photon energy for the p-modulation-doped sample. The emission corresponds to band-to-band recombination of photoexcited electrons with majority holes. The polarization, measured near the peak of the PL, is large for excitation close to the

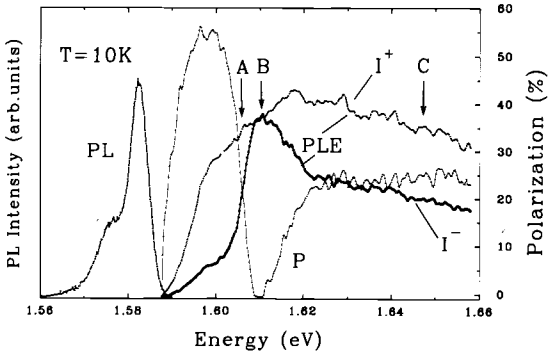

Fig. 1: CW photoluminescence (PL), photoluminescence excitation (PLE) and polarization (P) spectra of the p-modulation doped, 60 Å, QW at 10 K.

Fermi edge (A), exhibits a clear dip at the light-hole (*lh*) exciton energy (B) and recovers again at higher energies (C). This behavior is similar to that calculated by Uenoyama and Sham.[14]

The time evolution of I^+, I^- and $\wp$ at the peak of the PL are shown in Figs. 2 and 3 for excitation at A and at the *lh* exciton, respectively (the time evolution at C is similar to that at the Fermi edge). The spectra were recorded with an excitation density of ~$2x10^{10}$ cm^{-2}. For excitation energies different than that of the *lh* exciton, the polarization rises within our time resolution and then decays exponentially with a time constant of $\tau_{sf,e}$=150± 25 ps. On the other hand, we do not observe any significant polarization an any time for excitation at the *lh* exciton.

The results for the spin flip of the electrons are the simplest of the three cases investigated, and can be explained as follows. For the sake of simplicity, let us assume that the incident light is right-handed circularly polarized, σ^+ (similar arguments do also apply to left-handed polarized excitation). The promotion of electrons solely from the heavy-hole (*hh*) subband to the conduction band, which is obtained pumping at A, produces holes with spin +3/2 and electrons with spin -1/2. Since the number of photoexcited holes is insignificant compared to the doping level, the spin relaxation of holes is irrelevant and the measured decay of $\wp$ yields an electron-spin relaxation time of $\tau_{sf,e}$=150±25 ps for this sample.

The enhanced absorption of light holes at B, due to excitonic effects, and the increased spin admixture at large k in the *hh* wavefunction creates a nearly equal population of spin +1/2 and -1/2 electrons, resulting in a zero polarization at B at all times. The vanishing of the polarization, for excitation at the light-hole exciton, should be a general trend in all p-doped samples. For excitation above the light-hole threshold (C), the *hh* band to band absorption becomes stronger. This reduces the creation of spin +1/2 electrons as compared to excitation at B and therefore a positive polarization is again observed at C. The decrease

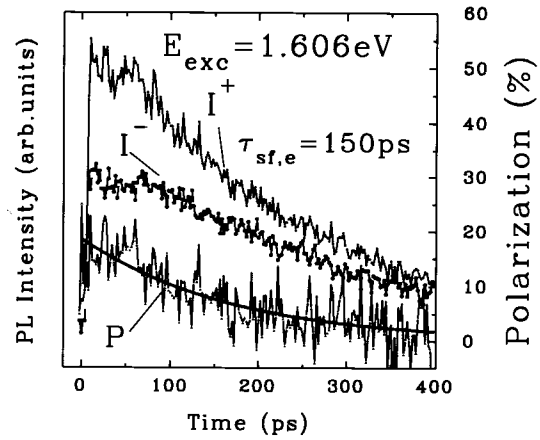

Fig. 2: Time evolution of I^+, I^-, and $\wp$ of the p-modulation doped sample for excitation at A (see Fig. 1) at 10 K. The excitation density is $2x10^{10}$ cm^{-2}.

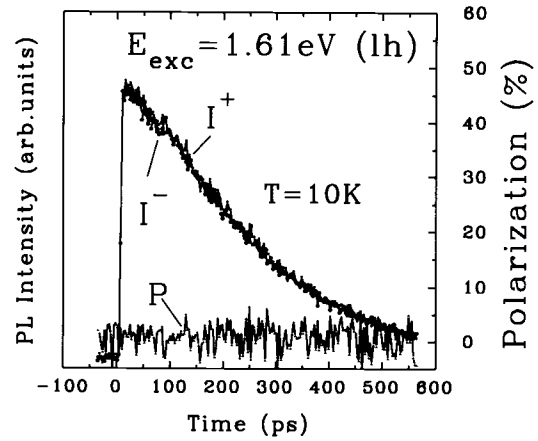

Fig. 3: Time evolution of I^+, I^-, and $\wp$ of the p-modulation doped sample for excitation at the light hole exciton (B, see Fig. 1) at 10 K. The excitation density is $2x10^{10}$ cm^{-2}.

in the cw polarization at C as compared with that at A is in good agreement with the reduction in $\tau_{sf,e}/(\tau_{sf,e} + \tau_{rec})$ averaged over the spectral bandwidth, where the spin-relaxation time, $\tau_{sf,e}$, and the recombination time, τ_{rec}, are taken from the time-resolved measurements. These features of the polarization spectrum are in good agreement with the theory of Ref. 14.

Our measured electron spin-relaxation time of ~150 ps in quantum wells, for comparable hole density ($6x10^{17}$ cm^{-3}) and temperature (10 K), is about a third to a quarter of that in p-doped bulk GaAs.[2] From the temperature and hole-density dependence of the data, it was concluded in Ref. 2 that the electron-hole scattering with simultaneous exchange (BAP) dominates over the DP mechanism in the spin flip processes of electrons. The enhancement, due to

exchange interaction, of the spin-relaxation rate in QW's compared to bulk materials is $3\pi/2k_FL$, where k_F is the Fermi vector for the 3D density and L the well width.[31] On the other hand, the enhancement due to the DP mechanism[32] is $4/(k_FL)^4$. For our sample, we obtain that the BAP enhancement is 3 while the corresponding to the DP mechanism is less than 1. Therefore the BAP is likely to be the dominant mechanism for the spin relaxation of electrons in p-doped quantum wells.

4. Hole spin relaxation

In bulk III-V semiconductors, the fact that the heavy- and light-hole bands are degenerate at k=0 leads to a rapid spin relaxation of holes. Actually, most of the work on bulk semiconductors assumes that the hole spin relaxation is instantaneous. The lifting of the degeneracy at the top of the valence band, as, for example, due to the application of an uniaxial stress, reduces the hole spin relaxation.[33] In QW's, the degeneracy is also removed due to confinement effects and a similar reduction should be expected. Two complementary theoretical approaches have been contemplated to predict the hole spin relaxation in QW's.[14,15] Since the spin components of a valence state away from k=0 are mixed, the spin flip will be determined by the scattering between different hole states during the process of energy and momentum relaxation. Uenoyama and Sham have considered the spin relaxation in n-doped QW's through momentum relaxation by acoustic-phonon interaction and by shakeup of the Fermi sea.[14] Ferreira and Bastard have calculated the hole spin-relaxation time due to the hole interaction with static scatterers.[15] Both theories indicate that the hole spin relaxation is incomplete prior to radiative recombination and invalidate the interpretation of the PL polarization experiments in terms of an instantaneous hole depolarization.

The cw luminescence and excitation spectra from the n-modulation-doped sample are depicted in Fig. 4. In this case the PL results from band-to-band recombination of photoexcited holes with electrons. The degree of polarization is nearly zero in the whole spectral range, and is not shown in the figure. The time evolution of I^+, I^-, and ρ are shown in Fig. 5 for excitation at 1.645 eV that generates electrons close to the Fermi energy and holes at ~3 meV. The initial polarization of about 30% decays with a time constant of $\tau_{sf,h}$=4 ps. The evolution of ρ in the same time interval, exciting at 1.7 eV, shown in Fig. 6, reveals the presence of an additional slow component. At this energy, holes from both the *hh* and *lh* subbands are created; a fitting to an exponential decay in a longer time interval (Fig. 7) obtains a time constant of ~200 ps. Using arguments similar to those for the p-sample, we identify

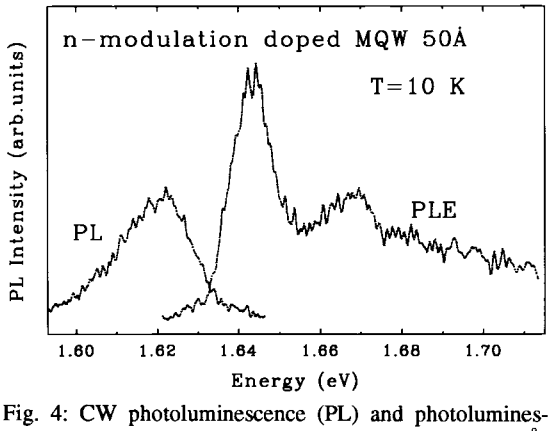

Fig. 4: CW photoluminescence (PL) and photoluminescence excitation (PLE) of the n-modulation doped, 50 Å, QW at 10 K.

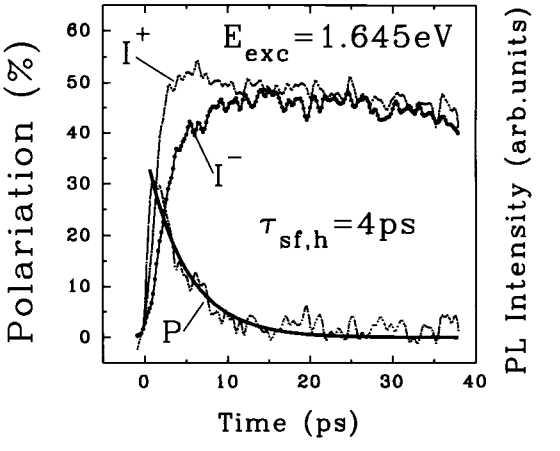

Fig. 5: Time evolution of the luminescence intensities and polarization for the n-modulation doped sample at $2x10^{10}$ cm^{-2} exciting at 1.645 eV.

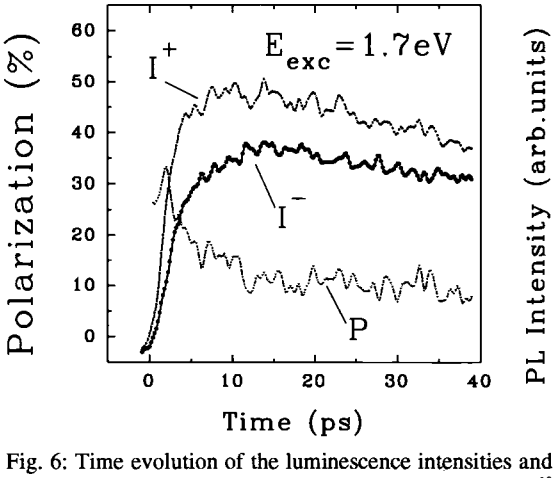

Fig. 6: Time evolution of the luminescence intensities and polarization for the n-modulation doped sample at $2x10^{10}$ cm^{-2} exciting at 1.7 eV.

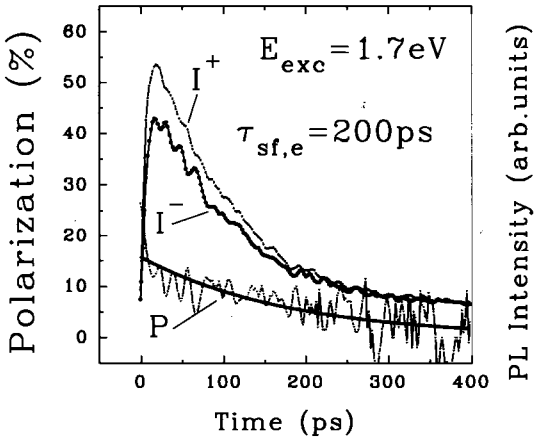

Fig. 7: Same as Fig.6 in a longer time interval to observe the polarization decay due to the polarized electrons.

the fast decay, $\tau_{sf,h}$=4 ps, with the spin relaxation of holes. As we have mentioned above, any energy and momentum relaxation process leads to the spin relaxation of the holes, because the hole states are an admixture of various spin states away from k=0.[14,15]

Our measurements show that the usual assumption of instantaneous hole spin relaxation is not valid in quasi-2D systems, and constitutes the first direct measurement of the hole spin-relaxation rate in a semiconductor. We attribute the increased hole spin-relaxation time to the lifting of the valence band degeneracy at $k_{\parallel}$=0. This is consistent with previous results in bulk GaAs under strain[33] and with theoretical predictions.[14,15] Unfortunately, a quantitative comparison with theory is not possible at present because our experiments measure the spin relaxation of a thermalized, hot-hole distribution, which is achieved in less than 1 ps. Furthermore, different scattering mechanisms, such as acoustic phonon, impurity, defect or carrier-carrier scattering, contribute to the hole relaxation and their relative importance has not yet been established.

For excitation at higher energies, the electrons are created with energies comparable to or larger than the electron Fermi energy (~10 meV). Electron-electron collisions mix the electrons and can give the Fermi sea a net spin polarization that decays with $\tau_{sf,e}$. Since this slow decay component of the polarization will be present only if there is a net polarization within the spectral width of detection, it is absent for low-energy excitation for which the photoexcited electrons mix predominately with electrons near the Fermi edge. We find, for high energy excitation, that the ratio of the slow to the fast component of the polarization decay increases with power density as expected. A small, net polarization of the Fermi sea in the p-doped QW should be present at very short times (<4 ps); however, its effects are probably hidden by the larger polarization of the photoexcited electrons.

5. Exciton spin relaxation

The problem of exciton spin relaxation is clearly more complicated because we are dealing with a two-particle system. The spin relaxation rates of the electron and hole which are bound by the Coulomb interaction influence the rate of the exciton spin relaxation. One can consider two extreme limits: if the electron-hole exchange interaction is weak, the electron and hole will conserve their individual spin relaxation times. Spin-flip of both particles is required to observe an optically active exciton ($|+1\rangle$ or $|-1\rangle$), therefore the particle with slower spin relaxation will determine the exciton spin-relaxation time. On the other hand, if the exchange interaction is strong, both electrons and holes will flip the spin simultaneously, giving an average time constant between those of the slower and the faster particle. In a real semiconductor, it is possible that an intermediate situation between the two limits is found.

For intrinsic GaAs QW's, time-resolved pump and probe absorption measurements have obtained a spin relaxation time of the order of 30 ps[23] for the ground state of the heavy-hole exciton. Previous results on our high-quality sample, using a streak camera to measure the decay of the polarized luminescence, gave a similar value (50 ps) for resonant excitation.[21] For non-resonant band-to-band excitation, the polarization for low excitation density was identically zero, within the streak camera resolution (~20 ps).[21] In our experiments we detect the PL at the *hh* exciton; the up-conversion setup provides much better time resolution, however it requires higher excitation intensity and does not allow measurements for resonant excitation.[26]

We show in Fig. 8 the time evolution of the polarization at the peak of the *hh* exciton, for excitation at

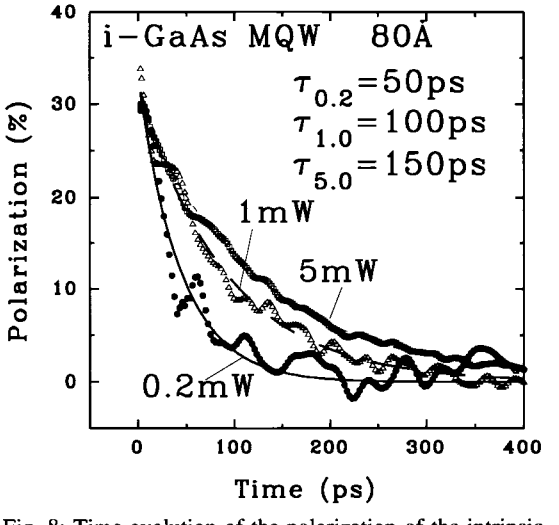

Fig. 8: Time evolution of the polarization of the intrinsic, 80 Å, QW for different excitation powers at 10 K.

1.612 eV, for different incident powers. The spin-relaxation time decreases from 150 ps to 50 ps as the excitation intensity is decreased from 5 mW to 0.2 mW. This trend indicates the compatibility of the present data with the streak-camera results, obtained with lower excitation intensities.[21] Let us now discuss why the spin-relaxation time, at the lowest intensity, is shorter for non-resonant excitation than for the resonant one. We suggest that a combination of the electron-hole exchange interaction and the spin-orbit interaction on the hole is required to flip the spin of the exciton from $|+1>$ to $|-1>$ or vice versa. The necessity of the spin-orbit interaction is due to the fact that exchange conserves the total angular momentum, thus it cannot flip the total spin by itself. The experiments under resonant excitation show that the exciton spin-flip time, $\tau_{sf,x}$, is ~50 ps.[21]

For non-resonant excitation, the increase in the spin-orbit mixing during the formation of the exciton can lead to a shorter spin-flip time. In this case, the electron-hole pair relaxes in two steps:[25] It forms an exciton with a large center-of-mass momentum in less than 20 ps and then, in over 300 ps, the exciton loses its total momentum. The hole, being the heavier particle, acquires the major share of the total momentum during the first stage. This is the cause for the increase in the spin-orbit mixing, which, with the aid of the exchange, enhances the spin flip compared to that in the exciton ground state.

Another striking experimental feature is that a splitting in energy between the polarized (I^+) and unpolarized (I^-) spectra is observed at certain excitation intensities and time delays, as shown in Fig. 9 for 85 ps and 10 mW. The depolarized spectrum is always at lower energy, independently of the helicity of the exciting light. We should also note that the FWHM of the PL is determined by the laser spectral width and the spectrometers slits; the cw FWHM of the PL is only 0.65 meV. Figure 10 demonstrates that the splitting decreases with delay: for 2 mW excitation, the splitting is ~0.5 meV at short times and it vanishes at ~300 ps. Furthermore, this splitting depends strongly on excitation power: it is smaller than our experimental resolution for 0.1 mW, increases up to ~7 mW and then saturates (see Fig. 11).

We believe that the energy splitting and the increase on the exciton spin-relaxation time with increasing excitation intensity have a common origin, namely a

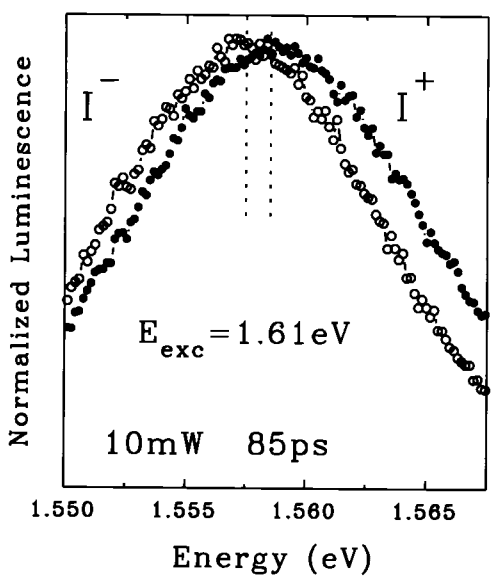

Fig. 9: Polarized (I^+) and depolarized (I^-) photoluminescence spectra of the intrinsic sample at a delay of 85 ps, with an excitation power of 10 mW and a laser energy of 1.61 eV, at 10 K. The peaks of the spectra are marked by dotted lines.

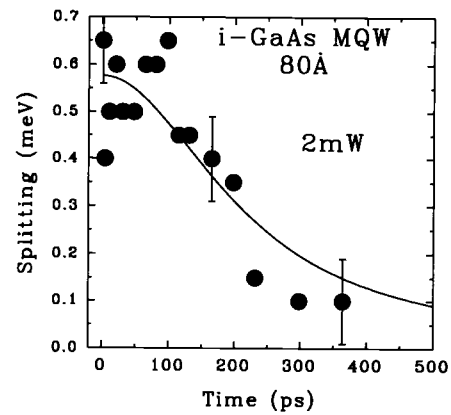

Fig. 10: Time dependence of the splitting between I^+ and I^- in the intrinsic sample at 2 mW. The line is a guide to the eye.

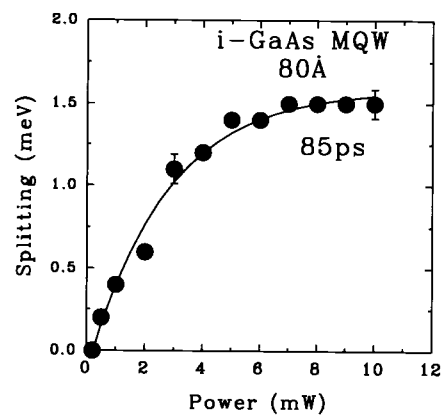

Fig. 11: Excitation density dependence of the splitting between I^+ and I^- in the intrinsic sample at a delay of 85 ps. The line is a guide to the eye.

reduction of the electron-hole exchange interaction. Let us consider again for the sake of clarity excitation with σ^+ light, which predominately creates $|+1>$ excitons. Due to the Pauli exclusion principle, which acts separately on the electrons and holes, the excitons will have an energy distribution if the density is high enough. Similarly to the formation of anti-bonding and bonding states of a hydrogen molecule, the $|+1>$ excitons will suffer a repulsion and consequently will have a larger energy than the $|-1>$ excitons, which experiment an attraction. The splitting depends on the density of excitons, and vanishes as the densities of the two spin populations approach each other. Both of these expectations are borne out by our experiments (see Figs. 10 and 11). For a larger intensity of excitation, the closer approach of the excitons also enlarges the electron-hole relative distance and thus lowers the exchange interaction. These many-body effects induce the longer spin-relaxation times at higher excitation intensities shown in Fig. 8.

5. Summary

We have determined the electron, hole and exciton spin-relaxation times in high-quality quantum wells using time-resolved luminescence spectroscopy with ~0.5 ps time resolution. The spin-relaxation time for holes is not instantaneous, contrary to common assumptions. On the other hand, the electron spin-relaxation time is considerably faster than in bulk materials, as a result of an enhanced electron-hole exchange interaction in quantum wells. We have clearly demonstrated that the spin dynamics in intrinsic samples is extremely complicated: the exciton spin relaxation is influenced by many-body effects as well as by the processes of exciton formation. The lack of resonant pumping experiments with sub-picosecond time resolution does not allow us to conclude whether the exciton spin relaxation takes place in one step, or, on the contrary, the electron and hole spin relax independently. We believe that this is an open issue, which needs further investigation.

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