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Spin Dynamics in Doped and Intrinsic GaAs Quantum Wells

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Abstract

We have determined the spin relaxation dynamics of electrons, holes and excitons in GaAs quantum wells by means of subpicosecond studies of the photoluminescence polarization. Our measurements demonstrate that the usual image of instantaneous hole-spin relaxation is not valid in 2D systems. For electrons, the spin relaxation rate is considerably larger than the corresponding value in p-doped bulk GaAs. The processes of spin relaxation of free excitons are far more complicated than those of free carriers: we find that many-body effects play an important role in the spin relaxation of excitons.

1. Introduction

The excitation of states with a preferential spin orientation, using selective optical excitation with circularly polarized light, is a very powerful technique for the investigation of relaxation processes in semiconductors [1, 2]. The optical orientation is revealed in a polarization of the photoluminescence (PL), whose sign and magnitude depend on the transition which is pumped and on the ratio between the lifetime and the spin relaxation time of the excited electronic states. This polarization provides crucial information about the symmetry of electron, hole and excitonic wave functions in semiconductors at the band edge. The spin dynamics of excitons and free carriers in bulk semiconductors has been extensively studied in the past, and different spin relaxation mechanisms have been identified [1, 3, 4]. Usually, the polarization of the emission is measured in steady state conditions pumping with circularly polarized light. Complementary information is obtained with the application of a magnetic field in the Voigt geometry (Hanle effect) [1] or in the Faraday configuration [5]. The measurement of the polarization decay of the luminescence, given a sufficient time resolution, provides an alternative and direct method to obtain the spin relaxation time.

In quantum well (QW) structures only a few studies have been addressed to polarization properties of excitons and free carriers. In the case of holes, since the fourfold degeneracy of the top of the valence band is lifted, a significant change in their spin relaxation is expected. The circular [6–11] and linear [12] polarization of the PL under cw conditions have been measured and various theories have been proposed to explain these results [13–19]. The studies about the dynamical behavior of the polarization are even more scarce. Time resolved spectroscopy of PL polarization has been reported recently for very high carrier densities [20], for intermediate densities [21], for low densities [22–25], and in the case of holes for variable densities by the application of an external electric field [26]. Time-resolved absorption measurements have also been used to investigate the spin relaxation kinetics of excitons at room temperature in multiple QW's [27, 28].

The mechanisms which determine the spin kinetics in QW's are not sufficiently understood: the influence of the formation dynamics of excitons and of many-body effects must be contemplated [24–25]; additionally, the differences in the behavior of holes, electrons and excitons need a careful experimental and theoretical investigation [17]. There is also a serious problem related to the sample quality. Localization effects can strongly modify the spin relaxation dynamics [11]. Since localization varies in different samples, this can lead to a wide diversity in the results [21]. Therefore it is important the use of high-quality samples to uncover the authentic nature of the spin-relaxation dynamics.

In this article we review our results on the spin dynamics of holes, electrons and excitons in GaAs QW's [25]. We have used n- and p-modulation doped QW's to investigate the hole and electron spin relaxation dynamics separately. We have found that the polarization of the holes decays with a time constant of ~ 4 ps, which demonstates that the usual hypothesis of instantaneous hole spin-relaxation is not valid in quasi-2D systems. The reduction of the hole spin-relaxation is interpreted in terms of the lifting of the valence-band degeneracy at $\mathbf{k}_{\parallel} = 0$. The electron spin-relaxation time in p-modulation-doped QW's is ~ 150 ps, approximately 4 times shorter than in comparably doped bulk GaAs. This increase in the relaxation rate is associated to an enhanced electron-hole exchange interaction in QW's. To study the spin relaxation of excitons we have used a, very high quality, intrinsic QW, which exhibits a nearly vanishing Stokes shift between the absorption and the emission and the line shape of the exciton is essentially homogeneously broadened [29]. We have found that the processes of spin relaxation of excitons are much more complicated than those of free carriers: their spin relaxation depends strongly on excitation density as well as in the process of excitonic formation. Many-body effects play a crucial role in the excitonic spin-relaxation.

The rest of the manuscript is organized as follows: the experimental details are given in Section 2. The hole and electron spin relaxation are presented in Sections 3 and 4, respectively. We discuss the exciton spin relaxation in Section 5. Finally, we summarize our results in Section 6.

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 $n_0 = 3 \times 10^{11}\,cm^{-2}$ and $p_0 = 4 \times 10^{11}\,cm^{-2}$, 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 [24] and on exciton formation and relaxation [29] 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 measurements were performed as a function of temperature, excitation density and energy. 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 [30]. The polarization of the exciting light was selected by means of a quarter-wave plate. 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. The degree of polarization, P, 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, $P = (I^+ - I^-)/(I^+ + I^-)$.

3. n-modulation-doped QW's

The four-fold degeneracy of the heavy- and light-hole bands, in bulk III-V semiconductors, 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 [31]. 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 [17, 18]. 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 [17]. Ferreira and Bastard have calculated the hole spin-relaxation time due to the hole interaction with static scatterers [18]. These theories invalidate the interpretation of the PL polarization experiments in terms of an instantaneous hole depolarization and indicate that the hole spin relaxation is incomplete prior to radiative recombination.

The cw luminescence and photoluminescence excitation (PLE) spectra from the n-modulation-doped sample are depicted in Fig. 1. The power density was kept below $0.1\,W\,cm^{-2}$. The PL results from band-to-band recombination of photoexcited holes with electrons. A Stokes shift of 23 meV is observed between the PL maximum and the first peak of the PLE spectrum, which corresponds to the first band-to-band (electron to heavy-hole) transition at the Fermi wave vector k_F. The second peak in the PLE corresponds to an electron to light-hole transition. The degree of polarization is nearly zero in the whole spectral range, in contrast with the results of Ref. [26] where a clear dependence of P on photon energy is obtained, and is not shown in the figure.

The time evolution of I^+, I^-, and P are shown in Fig. 2 for excitation at 1.645 eV that generates electrons close to the Fermi energy and holes at ~3 meV. The polarization rises almost instantaneously reaching an initial value of about 30% and decays with a time constant of $\tau_{sf,h} = 4$ ps.

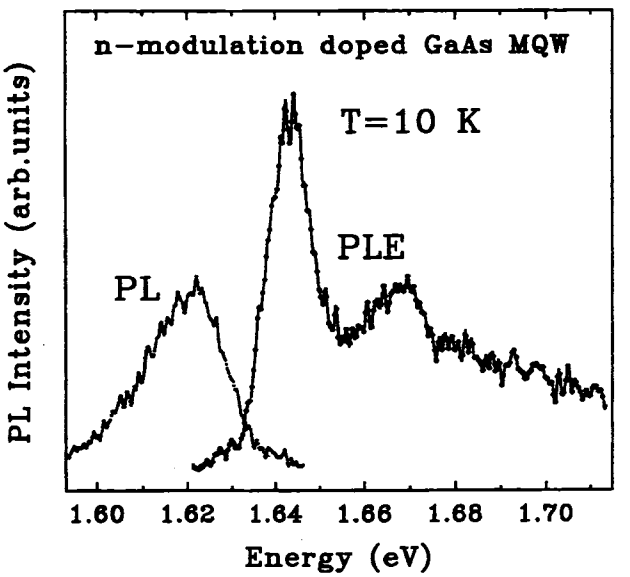

Fig. 1. CW photoluminescence (PL) and photoluminescence excitation (PLE) of the n-modulation doped, 50 Å, QW at 10 K.

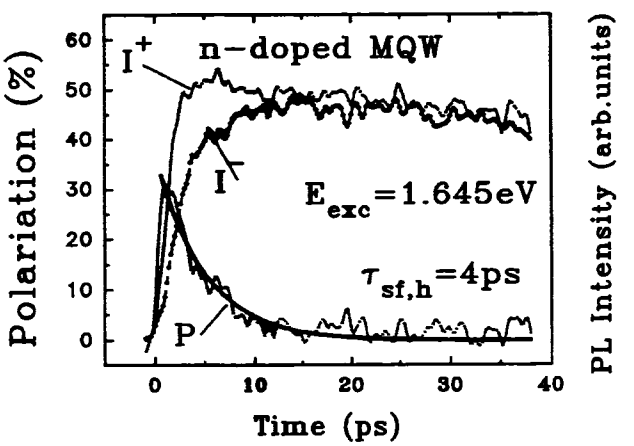

Fig. 2. Time evolution of the polarized (I^+) and unpolarized (I^-) PL intensities and polarization (P) for the n-modulation doped sample at $2 \times 10^{10}\,cm^{-2}$ exciting at 1.645 eV.

The evolution of P in the same time interval, exciting at 1.7 eV, shown in Fig. 3(a), 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. 3(b)] yields a time constant of ~200 ps.

We identify the fast decay, $\tau_{sf,h} = 4$ ps, with the spin relaxation of holes. This small value of $\tau_{sf,h}$ originates from the fact that 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 $k = 0$ [17, 18]. These results show that the usual assumption of instantaneous hole spin relaxation is not valid in quasi-2D systems, and yield 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 $k_{\|} = 0$. This assignment is consistent with previous results in bulk GaAs under strain [31] and with theoretical predictions [17, 18]. 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 higher photon-energy excitation, 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 $\tau_{sf,e}$. The slow decay component of the polarization will be present only if there is a net polarization within the spectral width of detection, therefore it is absent for low-energy excitation for which the photoexcited electrons mix predominately with electrons near the Fermi edge (this spectral range is not accessible with our measurements). 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.

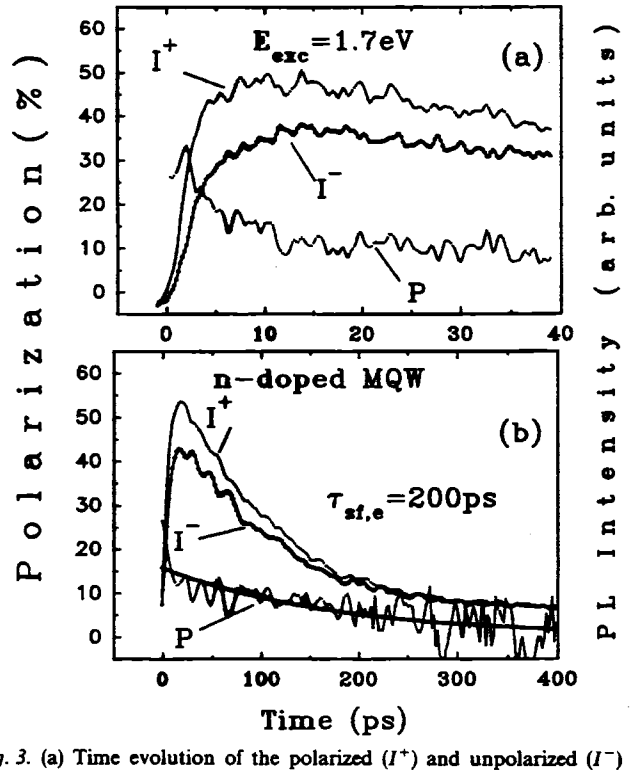

Fig. 3. (a) Time evolution of the polarized (I^+) and unpolarized (I^-) PL intensities and polarization (P) for the n-modulation doped sample at 2×10^{10} cm^{-2} exciting at 1.7 eV. (b) Same as upper panel in a longer time interval to observe the polarization decay due to the polarized electrons.

4. p-modulation-doped QW's

Three main mechanisms are believed to be responsible of the spin relaxation of electrons. The conduction band states in QW's, in the parabolic approximation of the dispersion relations, are eigenstates of the spin operator σ. However, a spin mixing appears in the lowest correction to the parabolic dispersion law, the anisotropic cubic term [13, 19]. This leads to a mixing term which can be considered as a Zeeman term due to a magnetic field lying in the layer plane [19]. 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 [32, 33]. Another efficient spin-relaxation mechanism has been proposed by Elliot and Yafet (EY) [34] 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 (BAP) [34, 35].

Figure 4 shows the cw PL, PLE for $\sigma^+\sigma^+$ (I^+) and $\sigma^-\sigma^+$ (I^-) configurations and luminescence polarization (P) as a function of photon energy for the p-modulation-doped sample. The emission, which presents a Stokes shift of ~17 meV, corrresponds to band-to-band recombination of photoexcited electrons with majority holes. The transitions which involve heavy- and light-holes are clearly seen in the I^- excitation spectrum. The polarization, measured near the peak of the PL, is large for excitation close to the 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 [17].

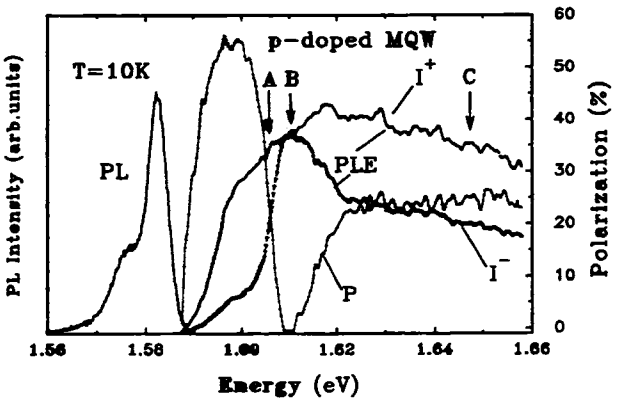

Fig. 4. CW photoluminescence (PL), photoluminescence excitation (PLE) and polarization (P) spectra of the p-modulation doped, 60 Å, QW at 10 K.

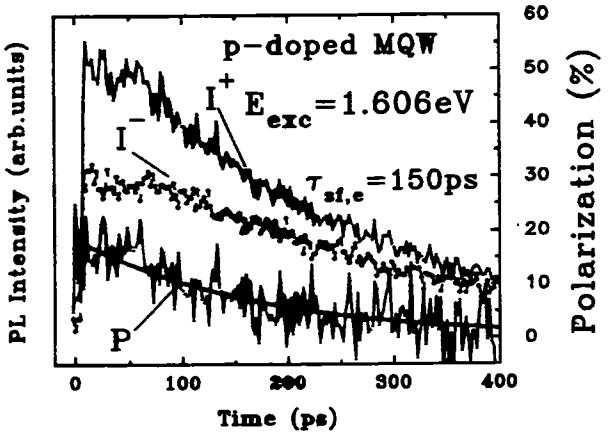

Fig. 5. Time evolution of I^+, I^- and P of the p-modulation doped sample for excitation at A (see Fig. 4) at 10K. The excitation density is $2 \times 10^{10}\,cm^{-2}$

The time evolution of I^+, I^- and P at the peak of the PL are shown in Figs 5 and 6 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 $\sim 2 \times 10^{10}\,cm^{-2}$. 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 $\tau_{sf,e} = 150 \pm 25$ ps. On the other hand, we do not observe any significant polarization at any time for excitation at the *lh* exciton (see Fig. 6).

The results for the spin flip of the electrons are the simplest of the three cases investigated, and can be interpreted as follows. Let us assume that the incident light is right-handed circularly polarized, σ^+ (similar arguments do also apply to left-handed polarized excitation). Pumping at A, only electrons from the heavy-hole (*hh*) subband are promoted to the conduction band, 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 P yields an electron-spin relaxation time of $\tau_{sf,e} = 150 \pm 25$ 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* wave function 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 obtained at C. The decrease 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. 17. 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.

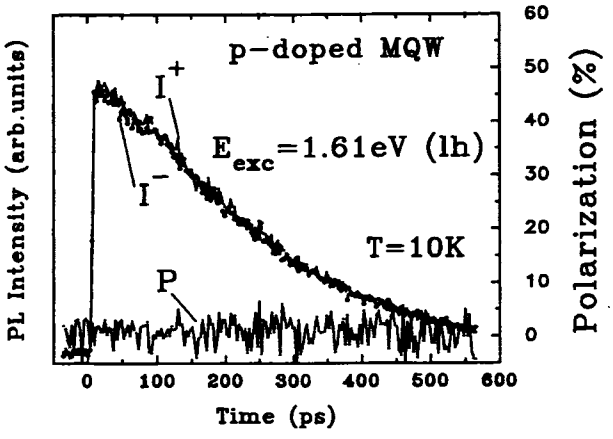

Fig. 6. Time evolution of I^+, I^-, and P of the p-modulation doped sample for excitation at the light hole exciton (B, see Fig. 4) at 10K. The excitation density is $2 \times 10^{10}\,cm^{-2}$.

Our measured electron spin-relaxation time of ~ 150 ps in QW's is about a third to a quarter of that in p-doped bulk GaAs, for comparable hole density ($6 \times 10^{17}\,cm^{-3}$) and temperature (10K) [3]. From the temperature and hole-density dependence of the data, it was concluded in Ref. [3] 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 $3\pi/2k_F L$, where k_F is the Fermi vector for the 3D density and L the well width [36]. On the other hand, the enhancement due to the DP mechanism [37] is $4/(k_F L)^4$. 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.

5. Intrinsic QW's

In intrinsic QW's the optical spectra are dominated by excitonic effects. The spin relaxation rates of the photoexcited electrons and holes, which are bound by the Coulomb interaction, influence the rate of the exciton spin relaxation. The problem of exciton spin relaxation is clearly more complicated because we are dealing with a two-particle system. One can consider two extreme limits: the electron and hole will conserve their individual spin relaxation times if the electron-hole exchange interaction is weak. 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 limit, 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 [27] 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 PL, gave a similar value (50 ps) for resonant exci-

tation [24]. For non-resonant band-to-band excitation and low excitation density, the polarization was identically zero, within the streak camera resolution (~20 ps) [24]. Our up-conversion setup provides much better time resolution, nevertheless it requires higher excitation intensity and does not allow measurements for resonant excitation [30].

The time evolution of the polarization at the peak of the *hh* exciton, for excitation at 1.612 eV, is depicted in Fig. 7 for different incident powers. The spin-relaxation time increases from 50 ps to 150 ps as the excitation intensity is increased from 0.2 mW to 5 mW. This trend indicates the compatibility of the present data with the streak-camera results, obtained with lower excitation intensities [24].

Let us now examine why the spin-relaxation time, at the lowest intensity, is shorter for non-resonant excitation than for the resonant one. We propose that a combination of two ingredients, electron-hole exchange interaction and spin-orbit interaction on the hole, is required to flip the spin of the exciton from $|+1\rangle$ to $|-1\rangle$ or vice versa. The requirement 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 [24]. 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 [29]: It forms an exciton with a large center-of-mass momentum (**K**) 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 between the polarized (I^+) and unpolarized (I^-) spectra is observed at certain excitation intensities and time delays, as shown in Fig. 8 for 85 ps and 10 mW [38]. The depolarized spectrum (I^-) is always at lower energy, independently of the helicity of the exciting light. Figure 9 illustrates that the splitting decreases with delay: for 2 mW excitation, the splitting is ~0.5 mV at short times and it practically vanishes at ~300 ps. Moreover, this splitting depends strongly on excitation power: Figure 10 shows that the splitting is smaller than our experimental resolution for 0.1 mW, increases up to ~7 mW and then saturates.

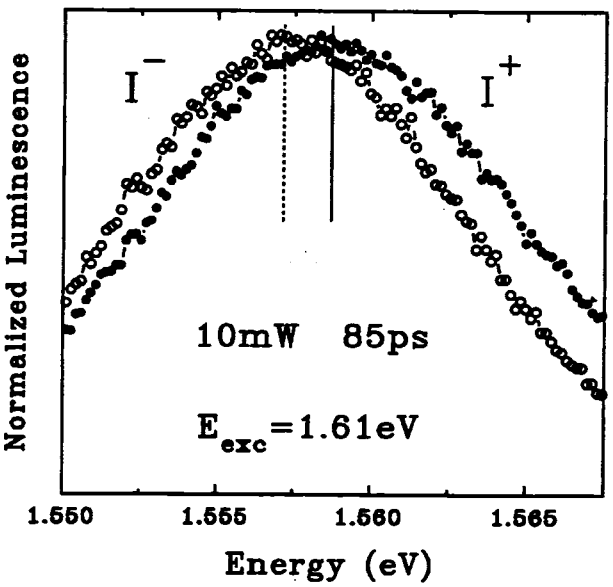

Fig. 8. Polarized (I^+) and depolarized (I^-) PL spectra of the intrinsic sample at a delay of 85 ps, with an excitation power of 10 mW and a laser energy of 1.61 eV, at 10 K. The vertical lines mark the peaks of the spectra.

We believe that the energy splitting and the increase on the exciton spin-relaxation time with increasing excitation

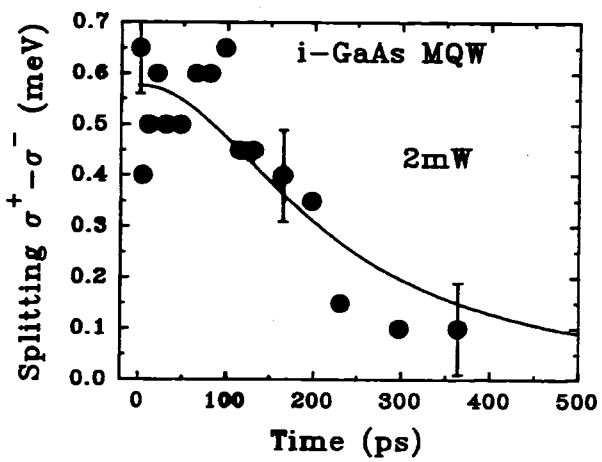

Fig. 9. Time dependence of the splitting between I^+ and I^- in the intrinsic sample at 2 mW. The line is a guide to the eye.

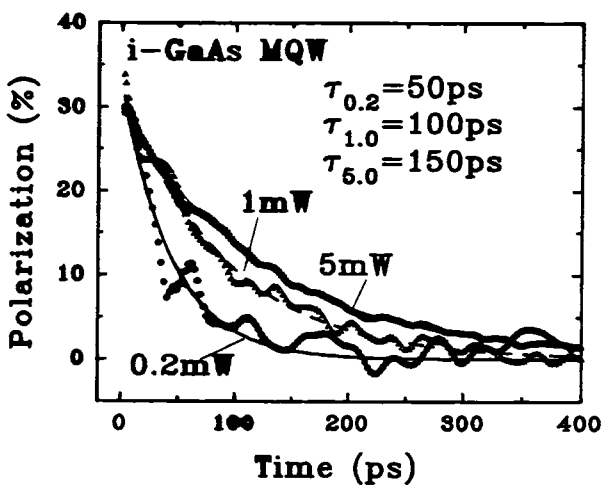

Fig. 7. Time evolution of the polarization of the intrinsic, 80 Å, QW at 10 K for different excitation powers.

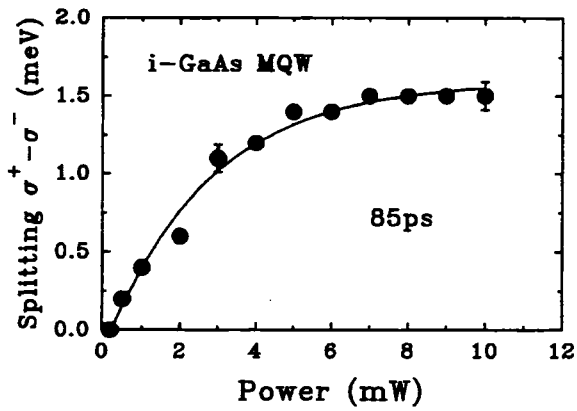

Fig. 10. Excitation density dependence of the splitting between I^+ and I^- in the intrinsic sample at a delay of 85 ps. The line is a guide to the eye.

intensity have a common origin, namely a reduction of the electron-hole exchange interaction. Let us consider again for the sake of clarity excitation with σ^+ light, which predominately creates $|+1\rangle$ 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. Making an analogy with the formation of antibonding and bonding states of a hydrogen molecule, the $|+1\rangle$ excitons will suffer a repulsion and consequently will have a larger energy than the $|-1\rangle$ 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 9 and 10). 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. The longer spin-relaxation times at higher excitation intensities displayed in Fig. 7 are induced by these many-body effects.

6. Summary

We have determined the hole, electron and exciton spin-relaxation times in high-quality QW's using time-resolved luminescence spectroscopy with ~ 0.5 ps time resolution. The spin-relaxation time for holes is not instantaneous, contrary to common assumptions in bulk semiconductors. The electron spin-relaxation time is considerably faster than in bulk materials, as a result of an enhanced electron-hole exchange interaction in QW's. We have shown that the spin dynamics in intrinsic samples is extremely complicated: the exciton spin relaxation is influenced by the processes of exciton formation as well as by many-body effects. 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.

Acknowledgements

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38. We should note that the FWHM of the PL is determined by the laser spectral width, the up-conversion process in the non-lineal crystal and the spectrometer slits. The cw FWHM of the PL is only 0.65 meV.