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# SPIN SPLITTING OF EXCITONS IN GaAs QUANTUM WELLS AT ZERO MAGNETIC FIELD

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**Abstract**—We have observed an energy splitting between +1 and −1 excitons in GaAs/AlAs quantum wells at zero magnetic field. The studies, which have been done by picosecond time-resolved photoluminescence spectroscopy, show that exciton spin splitting depends strongly on excitonic density and on the dynamics of spin relaxation: the spin splitting increases, raising the initial carrier density, and the time evolution of the spin splitting follows the exponential decay of the degree of polarization of the photoluminescence. For large carrier densities and short time delays the observed exciton spin splitting is as large as 4 meV. We present theoretical results which support the interpretation of experimental spin splittings in terms of inter-excitonic exchange interactions.

## 1. INTRODUCTION

Exciton spin dynamics in semiconductors quantum wells (QWs) have been a subject of intense research during the last few years. Most of the works have focused on spin relaxation processes, and particularly, on the study of the different spin relaxation mechanisms. In this respect, Maialle *et al.* have recently given a theory of exciton spin relaxation driven by intra-excitonic exchange interaction[1], and several experimental works have found evidence that this is the leading spin relaxation mechanism at low temperatures[2,3].

A different and much less investigated facet of exciton spin dynamics is related to the appearance, at zero magnetic field, of an energy splitting between spin +1 and spin −1 excitons. The first experimental observation of this spin splitting was reported by Damen *et al.*, using time-resolved photoluminescence spectroscopy[4]. Later on, pump and probe experiments by Stark *et al.*[5], and, more recently, Amand *et al.*, with time-resolved photoluminescence[6,7], have confirmed the existence of an exciton spin splitting at high excitonic densities.

Pump and probe experiments performed in GaAs QWs have demonstrated the occurrence of a blue shift of excitonic transitions in the presence of a high density of excitons[8,9]. This blue shift, which is more evident in narrow QWs[9], is at a variance with the results found in bulk semiconductors, where it is well known that the exciton energy remains unchanged even at very high densities. The behavior observed in three-dimensional systems is attributed to an almost exact compensation between two many-body affects acting in opposite directions[9,10]: an attractive interparticle interaction, which at $T \approx 0$ takes the form of a van der Waals interaction between excitons, and a repulsive interaction having its origin in the Pauli exclusion principle acting on the Fermi particles forming the excitons. In two-dimensional systems, as a consequence of the relative unimportance of the screening the attractive component is strongly reduced, and the repulsive part becomes unbalanced[10], resulting in a blue shift of the excitonic energies. The magnitude of the exciton blue shift has been calculated by Schmitt-Rink *et al.*[10,11], and the theoretical results have been found compatible with the experimental values[9].

In this work we report new results on exciton spin splitting in GaAs QWs, and we present theoretical results which support the interpretation of the experimental spin splittings in terms of the same inter-excitonic exchange interaction which causes the blue shift observed in QWs at high excitonic densities.

## 2. EXPERIMENTAL DETAILS

We report on exciton spin splitting 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, although displaying at 2 K and very low carrier densities a Stokes shift of ~2.5 meV, exhibits excitonic dynamical properties analogous to those observed in very high quality samples with vanishing Stokes shifts[12]. The dynamical properties of this sample, as well as results concerning spin

relaxation, have been reported in a previous work[3]. The experiments were performed at 4 K, exciting with pulses from a Styryl 8 dye laser, tunable from 810 to 720 nm. The incident light was directed along the growth axis and a back-scattering geometry was used. The dye laser was synchronously pumped by the 532 nm line of a mode-locked Nd:YAG laser. The photoluminescence (PL) was time resolved in a standard up-conversion spectrometer. The time resolution, obtained by overlapping on a non-linear crystal, $LiIO_3$, the luminescence from the sample with a delayed pulse from the laser, is basically determined by the width of the pulse, which in our case is of 5 ps. A double-grating monochromator was used to disperse the upconverted signal. The exciting light was circularly polarized by means of a $\lambda/4$ plate, and the PL was analyzed into its $\sigma^+$ and $\sigma^-$ components using a second $\lambda/4$ plate before the non-linear crystal.

### 3. RESULTS AND DISCUSSION

Figure 1 depicts PL spectra obtained 6 ps after excitation with $\sigma^+$ polarized light at 1.630 eV. The excitation energy corresponds to the heavy-hole (hh) excitonic continuum, and it is below the light-hole (lh) excitonic peak; the initial carrier density was of $6.5 \times 10^{10}\,cm^{-2}$. The filled symbols show the $\sigma^+$ luminescence ($PL^{+/+}$ line), and the open symbols depict the $\sigma^-$ emission ($PL^{+/-}$ line). Both spectra are normalized to its maximum intensity, but the measured $\sigma^+$ PL is 3.5 times more intense than the $\sigma^-$ luminescence. A clear splitting, which amounts to

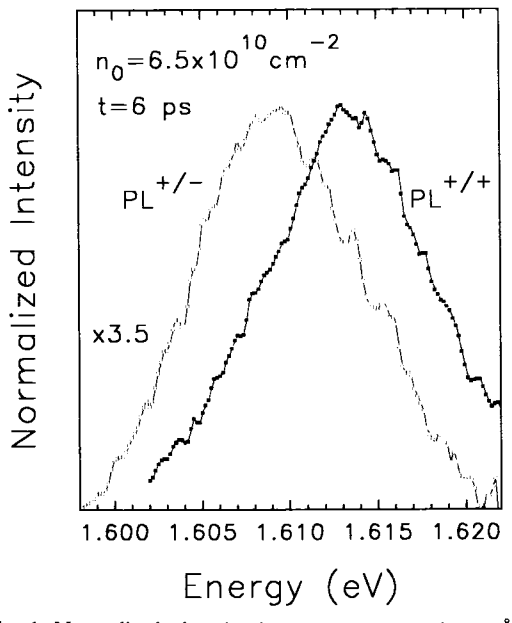


Fig. 1. Normalized photoluminescence spectra of a 77 Å-wide GaAs/AlAs QW obtained 6 ps after excitation with $\sigma^+$ polarized light at 1.630 eV. The initial carrier density was $6.5 \times 10^{10}\,cm^{-2}$. The filled symbols show the $\sigma^+$ luminescence ($PL^{+/+}$ line), and the open symbols depict the $\sigma^-$ emission ($PL^{+/-}$ line).

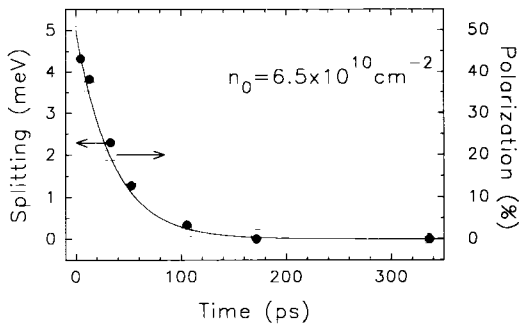


Fig. 2. Time evolutions of the splitting between +1 and −1 excitons (filled circles), and of the degree of polarization (open squares), for an initial carrier density of $6.5 \times 10^{10}\,cm^{-2}$. The solid line corresponds to an exponential decay with a characteristic time of 35 ps.

~4 meV (almost half the exciton binding energy), is observed in the figure between both spectra.

Exciting with $\sigma^+$ light at 1.630 eV corresponds to the creation of a distribution of spin −1/2 electrons and spin +3/2 heavy holes. In the absence of spin relaxation processes, these carriers form spin +1 excitons that will recombine, emitting $\sigma^+$ polarized photons ($PL^{+/+}$ line). However, due to different spin relaxation mechanisms, free carriers, as well as excitons, may flip their spins, resulting in ±2 and −1 spin excitons. The ±2 excitons cannot couple to light, and therefore will not be considered in this work. On the other hand, −1 excitons will recombine emitting $\sigma^-$ photons ($PL^{+/-}$ line). The spectra presented in Fig. 1 show results concerning two different aspects of exciton spin dynamics. First, the intensity of the $\sigma^+$ luminescence is higher than that of the $\sigma^-$ emission, indicating that at $t = 6$ ps most of the excitons have +1 spin. This is due to the fact that, in this sample, the spin relaxation time for this carrier density is of 35 ps[3]. The second aspect of exciton spin dynamics is related to the energy difference between $\sigma^+$ and $\sigma^-$ spectra, that reveals the existence of an energy splitting between +1 and −1 excitons. The origin of this spin splitting will be discussed below. We have checked that, independently of the helicity of the exciting pulse, the spectrum corresponding to the majority type of excitons lies always at higher energies.

Figure 2 shows the time evolutions of the splitting between +1 and −1 excitons (filled circles), and of the degree of polarization (open squares), for the same excitation energy and the same initial carrier density as in Fig. 1. The solid line corresponds to an exponential decay with a characteristic time of 35 ps. The degree of polarization that has been obtained as the difference between the spectrally integrated $\sigma^+$ and $\sigma^-$ luminescence normalized to the sum of both values, provides information about the difference between the densities of +1 and −1 excitons ($n_{+1}$ and $n_{-1}$, respectively). These two densities tend to equilibrate each other due to various spin relaxation

mechanisms, resulting in an exponential decay of the degree of polarization. The results of Fig. 2 indicate that exciton spin splitting is strongly correlated to the difference between $n_{+1}$ and $n_{-1}$ densities. In fact, the time evolutions of the exciton spin splitting and of the degree of polarization can be fitted with the same exponential decay (solid line in Fig. 2).

Figure 3 shows the dependence of exciton spin splitting on initial carrier density ($n_0$). The results have been obtained exciting at 1.630 eV for a time delay of 20 ps. Two different carrier density regimes are observed: for carrier densities smaller than $5 \times 10^{10}\,\mathrm{cm}^{-2}$, the exciton spin splitting exhibits a linear dependence on $n_0$, for higher carrier densities the spin splitting is approximately independent of $n_0$. We think that the high density behavior could be due to a saturation of absorption produced by phase-space filling and exchange interaction[10].

Let us discuss now the origin of exciton spin splitting and its connection with exciton spin relaxation. The strong parallelism observed in Fig. 2 between the time evolutions of the degree of polarization and of the exciton spin splitting seems to indicate that the spin splitting and the spin relaxation have the same physical origin. However, previous results obtained in this sample have shown that the spin flip processes which tend to equilibrate $n_{+1}$ and $n_{-1}$ densities are mainly controlled by *intra*-excitonic exchange interaction[3], a mechanism which is unable to describe the large spin splittings observed in this work[1]. As we will address below, we think that exciton spin splitting is due to *inter*-excitonic interactions, which turn out to be also dominated by exchange.

To investigate the origin of the exciton spin splitting, we have extended to the spin-dependent case the Generalized-Hartee–Fock approach previously developed by Haug and Schmitt-Rink[11], including screening in RPA. To the lowest order in $n_{+1}$ and $n_{-1}$, the spin splitting $\Delta E$ for excitons at rest is given by

$$\Delta E = \alpha(1-f)(n_{+1}-n_{-1})R_y^*,$$

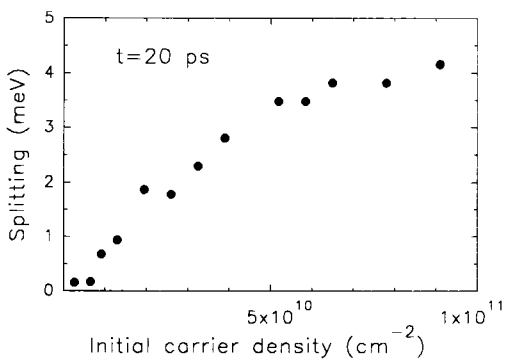


Fig. 3. Dependence of exciton spin splitting on initial carrier density ($n_0$). The results have been obtained for a time delay of 20 ps exciting at 1.630 eV.

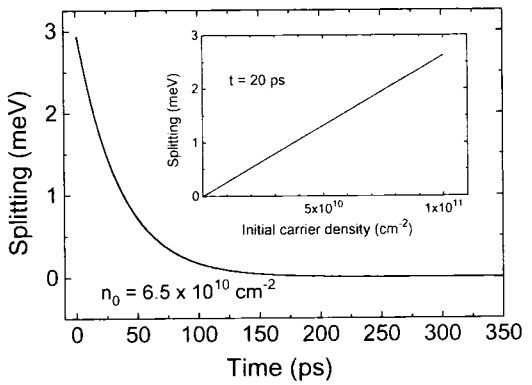


Fig. 4. Calculated splitting between $+1$ and $-1$ excitons as a function of time for an initial carrier density ($n_0$) of $6.5 \times 10^{10}\,\mathrm{cm}^{-2}$. Inset: calculated splitting vs $n_0$ at 20 ps.

where $\alpha$ and $f$ are spin-conserving and spin–flip *inter*-excitonic exchange interactions, respectively, and $R_y^*$ is the binding energy of the exciton. Since we are working in the lowest order of perturbation, $\alpha$ and $f$ are completely determined by the wave-functions of non-interacting excitons. The spin relaxation time associated with $f$ is much larger than the spin–flip time due to *intra*-excitonic exchange interaction. therefore, despite the large spin splittings produced by *inter*-excitonic exchange interaction (as the results of Fig. 4 will show), the spin relaxation process that tends to equilibrate $n_{+1}$ and $n_{-1}$ densities is controlled by *intra*-excitonic exchange interaction.

Figure 4 shows the theoretical time evolution and carrier density dependence (inset) of the exciton spin splitting produced by *inter*-excitonic exchange interaction. The time evolution and the carrier density dependence have been calculated for an initial carrier density of $6.5 \times 10^{10}\,\mathrm{cm}^{-2}$ and a time delay of 20 ps, respectively. In both cases a spin relaxation time of 35 ps has been assumed[3]. Since the calculations do not include the effects of absorption saturation[10], the results shown in the inset of Fig. 4 do not reproduce the experimentally observed bending of the spin-splitting vs carrier density (Fig. 3). However, the theoretical spin splittings are in good agreement with the experimental values. The splitting increases linearly with carrier density and it decays exponentially with time, following the evolution of the difference between $n_{+1}$ and $n_{-1}$ densities. This results in the idea that exciton spin splitting is due to *inter*-excitonic exchange interaction.

## 4. SUMMARY

We have investigated the spin splitting between $+1$ and $-1$ excitons in GaAs QWs by time-resolved photoluminescence spectroscopy. Our results show that the time evolution of the spin splitting follows the evolution of the difference between the densities of $+1$ and $-1$ excitons, and that the spin splitting

increases with initial carrier density. Theoretical results have been presented which reproduce the main features of the experimental spin splittings, supporting the idea that exciton spin splitting is due to inter-excitonic exchange interaction.

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