Observation of the zero-magnetic-field exciton spin splitting in high quality bulk GaAs and AlGaAs

E. V. Kozhemyakina,[1,2,a)] K. S. Zhuravlev,[1] A. Amo,[2,3] D. Ballarini,[2] and L. Viña[2]
[1]*Institute of Semiconductor Physics, 630090 Novosibirsk, Russia*
[2]*Departamento de Física de Materiales, Universidad Autónoma de Madrid, E-28049 Madrid, Spain*
[3]*Laboratoire Kastler Brossel, Université Paris 6, CNRS—UPMC, F-75252 Paris, France*

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We present an experimental study of the zero-magnetic-field exciton spin splitting measured by time-resolved photoluminescence in high purity bulk GaAs and AlGaAs samples. The dynamics of the splitting differs from that observed in two dimensions. Initially, the splitting increases during 100–250 ps and then decays. This initial increase is attributed to the fast rise of the density of excitons formed from noncorrelated *e-h* pairs. The splitting dynamics is used to determine the exciton formation time, which is found to vary with excitation density from 70 to 360 ps. © *2009 American Institute of Physics*. [doi:10.1063/1.3257369]

The investigations of charge carriers and excitons spin in semiconductors started in the late 1960s.[1,2] By now, there is a wide range of works devoted to the study of electron spin both in bulk and in low-dimensional structures. Spin polarization of excitons has been profusely studied in quantum wells (QWs).[3] During the last decade, particular attention has been paid to spin-related phenomena in quantum dots (QDs) due to their potential application in spintronic devices.[4] Nevertheless, some aspects have practically not been studied since the 1970s. The spin relaxation time of holes in bulk GaAs was measured only a few years ago[5] and theoretically investigated even later.[6,7] In recent years, the attention to exciton spin in bulk has also rised.[8] The revival of the interest to spin-related phenomena in bulk is caused by several reasons: (i) the advance in sample growth techniques has allowed producing samples of high quality where intrinsic excitonic effects are clearly observed, (ii) the knowledge of spin-related phenomena in bulk is also important for the understanding of the processes in low-dimensional structures. It can be used to distinguish between the effects that are due to the material properties and those of the structure, e.g., experimental results obtained in QDs are often compared to those in bulk. Moreover, charge carriers and excitons are captured in QDs from bulk barriers and, therefore, spin relaxation in bulk influence the resulting spin of the carriers in QDs.[9,10]

One of the most striking findings has been the discovery of an energy splitting between excitons with spins +1 and −1 created by circularly polarized light, in the absence of any magnetic field. This phenomenon was reported by Damen *et al.*[11] in GaAs QWs in time-resolved photoluminescence (TRPL) and later was confirmed by pump-and-probe experiments[12] and other TRPL studies.[13–15] The effect has also been observed in ultrathin InAs layers.[16] Fernández-Rossier *et al.*[17] showed that the interexcitonic exchange interaction produces a shift of the exciton energy levels. The value of the shift depends on the density of excitons with spins +1 and −1 (n_+ and n_-), thus when $n_+ \neq n_-$ the shift of the levels +1 and −1 is different and an energy splitting appears. It was predicted that the value of the splitting in two-dimensional (2D) case should be twice larger than that in bulk, but no experimental studies of this effect in bulk have been carried out so far.

This work presents an experimental study of the spin splitting of excitons in high quality GaAs and $Al_xGa_{1-x}As$ layers with x=0.05 and 0.15, and a thickness of 2.5 μm, grown by molecular-beam epitaxy (MBE) in a Riber-32P MBE system. The details of sample growth and preparation were reported in Ref. 18. All the samples have p-type conductivity with a hole concentration of 8×10^{14} cm^{-3} in GaAs and of $(1-5)\times10^{14}$ cm^{-3} in the $Al_xGa_{1-x}As$ layers. The low concentration of background impurities in these layers was confirmed by continuous wave photoluminescence (PL) experiments (not shown here, see Ref. 18): (i) the linewidths of the free exciton (FX) line in the spectra do not exceed the theoretically predicted values, and (ii) the ratio of the intensities of the FX and bound exciton lines is very high.

The $Al_{0.15}Ga_{0.85}As$ sample exhibits a splitting of a few meV between the subbands with hole angular momentum projections of ±1/2 and ±3/2 (light and heavy holes) caused by the mechanical strain in the epitaxial film that arises from the difference in the lattice constants of the film and the substrate (the light holes have a higher energy). The splitting causes a sign reversal of the PL polarization (the polarization is negative) when a resonant with the light-hole exciton photoexcitation is used.[19]

The samples are cooled down to 4.2 K in a cold finger cryostat and photoexcited with 2 ps-long pulses of a Ti:Sapphire laser, circularly polarized with a quarter-wave plate. The average laser power is varied from 2 μW to 1 mW. According to our estimation, 1 mW corresponds to an electron-hole (*e-h*) pair density $n_0 \approx 3\times10^{17}$ cm^{-3}. The PL in backscattering geometry is analyzed with an additional quarter-wave plate. The two PL components (I^+ and I^-, co- and cross-circularly polarized with the laser, respectively) are measured by rotating the polarizer and keeping the analyzer fixed. The PL is energy- and time-resolved by a synchroscan streak camera in conjunction with a spectrometer. The setup enables us to determine the energy positions of the co- and cross-polarized FX peaks separately. For that we fit the components of the FX line with Lorentzian line shapes using the least-squares method. As a value of the error, we

a)Electronic mail: kozhemyakina@thermo.isp.nsc.ru.

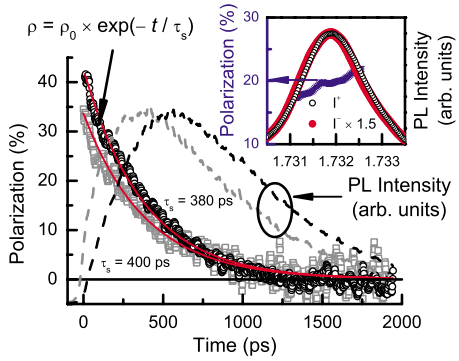

FIG. 1. (Color online) PL and polarization degree time traces of the FX line for the $Al_{0.15}Ga_{0.85}As$ sample. Solid lines are exponential fits of polarization decay. The excitation is done at 1.763 eV (32 meV above FX energy), with $n_0=3.0\times10^{15}$ cm^{-3} (gray line and squares) and 1.5×10^{16} cm^{-3} (black line and circles). The inset shows co- and cross-polarized (I^+ and I^-, respectively) spectra and polarization degree at a delay of 330 ps (integrated from 305 to 355 ps) for $n_0=3.0\times10^{15}$ cm^{-3}.

take the value of the energy when the mean square deviation is increased by 20% (the difference between the experimental data and the fit could be seen by eye). The value of the splitting is obtained as the difference between the energy positions of the co- and cross-polarized FX peaks, with an error of ±0.02 meV.

Figure 1 shows the time evolutions of the PL intensity and the polarization of the FX line in the $Al_{0.15}Ga_{0.85}As$ sample. The degree of circular polarization, ρ, decays exponentially. In the inset, co- and cross-polarized spectra of the FX line and the corresponding degree of polarization are shown for the same sample. As one can see, the FX line is split: the cross-polarized component lies at a slightly lower energy than the copolarized one. The spin splitting leads to a spectral dependence of the polarization across the FX line: the high-energy part of the line has a higher polarization. The nonmonotonous polarization of the FX line in bulk GaAs has been discussed in detail in Ref. 20. The maximum value of the splitting is observed in the $Al_{0.15}Ga_{0.85}As$ sample, amounting to 0.07 meV.

In Fig. 2 the dynamics of the splitting for the $Al_{0.15}Ga_{0.85}As$ sample is shown for different excitation powers. As it is readily seen, it is nonmonotonous: the value of the splitting rises during $t<t_{max}$, reaching its maximum faster than the PL intensity, and then decays. The t_{max} increases with excitation density from 100 ps at

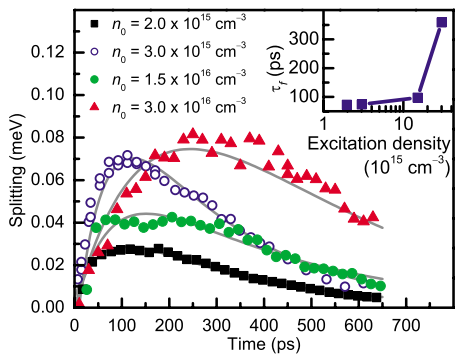

FIG. 2. (Color online) Time evolutions of the splitting for the $Al_{0.15}Ga_{0.85}As$ sample, excited at 1.763 eV. The gray lines are double exponential fits (described in the text). The inset shows the dependence of the exciton formation time on excitation density.

2×10^{15} cm^{-3} to 250 ps at 3×10^{16} cm^{-3}. This dynamics is rather different from the experimental dependence obtained in 2D that exhibited an exponential decay of the splitting.

The theory predicts that the splitting is proportional to the difference between excitons with spins +1 and −1 and is given by Eq. (1):[17]

$$\Delta = k|E_x|(n_+ - n_-)a_B^d = k|E_x|\rho n a_B^d, \quad (1)$$

where n_+ and n_- are the densities of excitons with spins +1 and −1, respectively, E_x is the exciton binding energy, a_B is the Bohr radius, n is the exciton density, ρ is the exciton degree of polarization, d is the dimension of space, and k is a coefficient that in three-dimensional (3D) equals to 3.4.

To explain the nonmonotonous dynamics of the splitting we propose a simple model. The exciton polarization, ρ, decreases exponentially on a time scale τ_s (see Fig. 1). To simplify the calculations, we assume that the time evolution of n can be described with the exciton formation and recombination times (τ_f and τ_R, respectively): $n(t)=A\times[-\exp(-t/\tau_f)+\exp(-t/\tau_R)]$, A being a constant. The value of the splitting can be therefore fitted with $\Delta(t)=k|E_x|a_B^3\rho_0\times[\exp(-t/\tau_s)n(t)]$ (Fig. 2, gray lines). The exciton formation times τ_f in the $Al_{0.15}Ga_{0.85}As$ sample obtained from the fit are shown in the inset of the Fig. 2. τ_f equals to ~70 ps at an excitation density $n_0\approx2\times10^{15}$ cm^{-3} and then increases up to 360 ps at 3×10^{16} cm^{-3}. The topic of exciton formation has been debated for several decades and it is still not clear. There are only a few works studying exciton formation in bulk semiconductors[21,22] compared with the numerous studies of this process in QWs. Theoretical values of τ_f range from 100 ps in QWs (Refs. 23 and 24) to more than 1 ns in quantum wires and 2D systems (Ref. 25) and experimental ones range in QWs from less than 10 ps to about 1 ns.[26–30] The dispersion of the experimental values is due to the different experimental conditions and measuring techniques. An exciton formation time of a few hundred picoseconds is in a good agreement with the most recent studies in QWs.[29,30] However, the bimolecular exciton formation time is expected to decrease with excitation density ($\tau_f^b=1/Cn_0$, where C is the bimolecular formation coefficient and n_0 is the excitation density), as was experimentally observed in Ref. 29. On the other hand, an increase of τ_f with excitation density has also been reported.[28] The coefficient C decreases with excitation density due to heating effects of the carrier distribution,[24,25] but more slowly than n_0 increases. The discrepancy between our data and the expected behavior can be explained taking into account that τ_f in our model includes both τ_f^b and the time of exciton dissociation to e-h plasma τ^d: $1/\tau_f=1/\tau_f^b-1/\tau^d$, and the latter increases with excitation density.

According to Eq. (1), the value of the splitting depends on $\rho\times n$. In our experiments the laser intensity and, therefore, n_0 is varied. We have found that the initial degree of circular polarization, ρ_0, depends on n_0, excitation energy and Al concentration of the sample and varied from −8% to more than 25%. To summarize all the results in one graph, we plot in Fig. 3 the dependence of the value of the splitting for $t=t_{max}$ on $\Delta n=\rho\times n_0$ where n_0 is the initial e-h pair density and ρ is the degree of polarization for $t=t_{max}$ (this gives, therefore, an upper estimation of Δn). The line represents the theoretical splitting calculated for $E_x=4.2$ meV, $a_B=15$ nm, and $k=3.4$. As one can see from Fig. 3, the experimental value of the splitting is in good agreement with

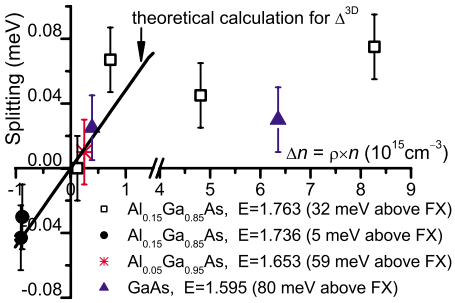

FIG. 3. (Color online) Dependence of the value of the splitting at $t=t_{max}$ on Δn for the GaAs and AlGaAs samples (points) and model prediction (line).

the theory. The sign of the splitting depends on the sign of ρ: when ρ is negative (under light-hole exciton resonant-excitation conditions for the $Al_{0.15}Ga_{0.85}As$ sample as aforementioned) the splitting is also negative (the cross-polarized component lies at a higher energy than the co-polarized one). The splitting grows with increasing Δn and saturates at $\Delta n \approx 10^{15}$ cm^{-3} which corresponds to an *e-h* pair density $n_0 \approx 4 \times 10^{15}$ cm^{-3}. It should be noted that a similar saturation was also observed in intrinsic QWs.[11,12] This saturation at high densities can be understood taking into account that the experimental points are plotted against *e-h* pair density, while the theory depends on the density of excitons, whose fraction decreases at excitation densities near the Mott transition.

In conclusion, we have investigated the exciton spin splitting at zero-magnetic-field in bulk GaAs and AlGaAs. The maximum value of the splitting increases with increasing excitation density and saturates at a density of $n_0 \approx 4 \times 10^{15}$ cm^{-3}. The saturation can originate from the decrease of the fraction of excitons formed from *e-h* pairs at high densities. The maximum value of the splitting of 0.07 meV is observed in the $Al_{0.15}Ga_{0.85}As$ sample. We have also found that, in contrast to the results obtained in low-dimensional systems, in 3D the dynamics of the splitting is nonmonotonous: it increases during $t < t_{max}$ and then decays. The initial increase of the splitting is due to the fast rise of the density of excitons formed from noncorrelated *e-h* pairs. The investigation of the dynamics of the splitting can therefore be used to determine exciton formation time, which was found to vary from 70 to 360 ps.

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