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E Kozhemyakina[1], K Zhuravlev[1], A Amo[2,3] and L Viña[2]

[1] Institute of Semiconductor Physics, Pr. Lavrentieva, 13, 630090 Novosibirsk, Russia

[2] SEMICUAM. Dept. Física de Materiales, Universidad Autónoma de Madrid, E-28049 Madrid, Spain

[3] Present address: Laboratoire Kastler Brossel, Université Paris 6, CNRS – UPMC. F-75252 Paris, France

E-mail: kozhemyakina@thermo.isp.nsc.ru

Abstract. We present a new experimental method to obtain the exciton-formation time based on the detection of the zero-magnetic field energy splitting between excitons with spins +1 and −1 created by circularly polarized light. The splitting is proportional to the product of the exciton circular-polarization degree ρ and density n. The dynamics of the exciton density n can be therefore obtained from the values of splitting and ρ. We investigate the exciton formation dynamics in high purity bulk GaAs and $Al_{0.15}Ga_{0.85}As$ samples at excitation densities 6×10^{14} – 3×10^{17} cm^{-3} and obtain the exciton-formation time, which is found to vary from 70 ps to 360 ps.

1. Introduction

The question of how free electron-hole (*e-h*) pairs bind into excitons has been debated for more than ten years and is not clear yet. Excitons have been extensively studied via optical absorption, photoluminescence (PL), or nonlinear experiments at the semiconductor band gap [1]. In III-V semiconductors these techniques are only sensitive to a subset of excitons with center-of-mass momentum $K \approx 0$, revealing information about exciton thermalization rather than formation. Therefore the measurement of the exciton formation time τ_f is a quite difficult task. As a result, the reported experimental values of τ_f in quantum wells (QWs) range from less than 10 ps to about 1 ns [2–7] and only a few investigations of exciton formation in bulk have been carried out so far [8–11]. The dispersion of experimental values of τ_f is due to the difference in experimental conditions and measuring techniques. Moreover, theoretical calculations of τ_f range from 100 ps to more than 1 ns [12–14]. The investigation of exciton formation processes has recently received a new impulse with the appearance of a new terahertz absorption technique [6]. In this work we propose a novel way to determine the exciton formation time based on the analysis of the dynamics of the exciton spin splitting and use it to investigate the exciton formation in bulk GaAs and $Al_{0.15}Ga_{0.85}As$ high-quality samples.

According to theory [15], the interexcitonic exchange interaction produces a shift of the exciton energy levels, whose value depends on the density of excitons with spins +1 and −1 (n_+ and n_-). When

$n_+ \neq n_-$, the shift of the levels $+1$ and -1 is different and an energy splitting ensues in the absence of any magnetic field. Its value is proportional to $n_+ - n_-$ and is given in the 3D case by equation (1):

$$\Delta = 3.4 \,|\, E_x \,|\, (n_+ - n_-) \, a_B{}^d = 3.4 \,|\, E_x \,|\, \rho \, n \, a_B{}^3 \tag{1}$$

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 and ρ is the exciton circular-polarization degree.

The dynamics of the exciton density, n, can be obtained if the dynamics of the splitting, Δ, and the polarization, ρ, are measured.

2. Samples and experimental setup

We investigate high-quality GaAs and $Al_{0.15}Ga_{0.85}As$ layers of of 2.5 µm thickness, grown by molecular-beam epitaxy. The details of sample growth and preparation were reported in Ref. [16]. 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 $\pm1/2$ and $\pm3/2$ (light- and heavy-holes) caused by mechanical strain in the epitaxial film, which causes a sign reversal of the PL polarization (the polarization is negative) when a photoexcitation resonant with the light-hole exciton is used [17].

The samples are cooled down to 4.2 K in a cold finger cryostat and photoexcited with 2 ps-long pulses of a $Ti:Al_2O_3$ laser, circularly polarized with a quarter-wave plate. The PL in backscattering geometry is analyzed with a second 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.

3. Results and discussion

Figure 1 shows the time evolutions of the free exciton (FX) PL at different excitation densities for the $Al_{0.15}Ga_{0.85}As$ sample. The time for the excitonic PL to reach its maximum intensity (t_{max}) increases with increasing excitation density, the same behavior was found in GaAs. The t_{max} increase can be caused by increase of exciton dissociation and decrease of exciton scattering rates [18]. The initial polarization degree in the GaAs sample increases from 10 to 20% with increasing excitation density, and in the $Al_{0.15}Ga_{0.85}As$ sample it is almost constant and amounts to 30–35%.
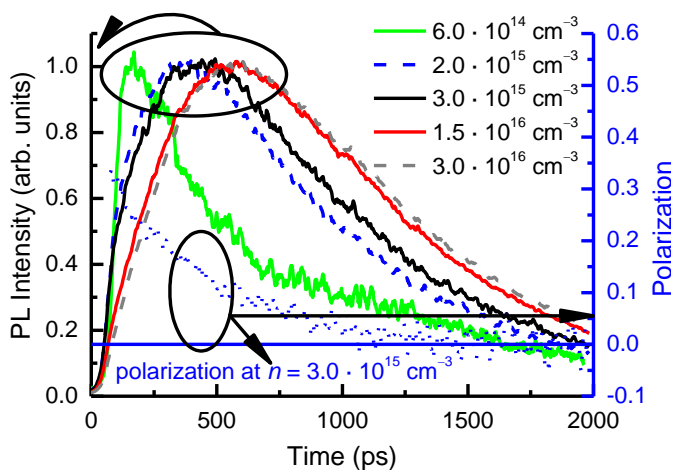
Figure 1. PL time traces at different excitation densities and polarization (at 3.0×10^{15} cm^{-3}) for the FX line of the $Al_{0.15}Ga_{0.85}As$ sample under excitation at 1.763 eV (32 meV above FX).
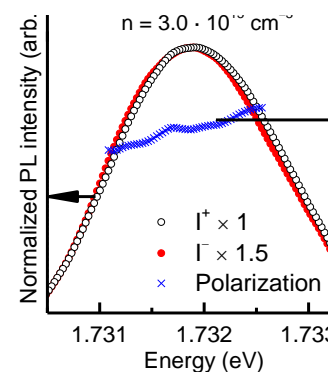
Figure 2. Co- and cross-polarized (I^+ and I^-) spectra in logarithmic scale and polarization for the $Al_{0.15}Ga_{0.85}As$ sample under excitation at 1.763 eV.

We fit the components of the FX line with Lorentzian line shapes, using a least-squares method, to determine the energy positions of the peaks. As an estimation of the error we 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 2 shows co- and cross-polarized spectra of the FX line for the $Al_{0.15}Ga_{0.85}As$ sample. As easily seen, the FX line is split: the cross-polarized component lies at slightly lower energy than the co-polarized 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.

Figure 3 shows the time evolution of the spin splitting in the $Al_{0.15}Ga_{0.85}As$ and GaAs samples at different excitation densities. The splitting has a non-monotonic dynamics: its value increases during 100 – 250 ps and then decays, reaching its maximum faster than the PL intensity. This dynamics is rather different from the experimental dependences obtained in 2D that exhibited a monotonic decay of the splitting [19, 20]. The non-monotonic dynamics of the splitting in our samples can be explained taking into account that its value depends on the exciton polarization ρ (which exponentially decays after the excitation) and concentration n (which firstly increases and then decreases). In the 2D case, the exciton formation time is probably faster and the fast polarization decay masks this effect.
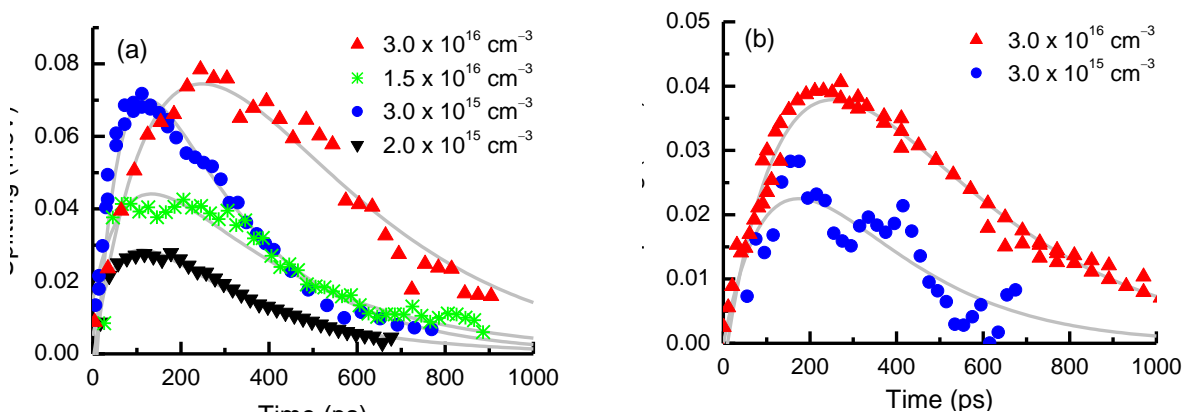
Figure 3. Time evolution of the spin splitting: (a) $Al_{0.15}Ga_{0.85}As$ sample under excitation at 1.763 eV (32 meV above FX energy); (b) GaAs sample, under excitation at 1.595 eV (80 meV above FX energy). The gray lines are guides for the eye.

Figure 4 shows time evolutions of the PL intensity, polarization and spin splitting in the $Al_{0.15}Ga_{0.85}As$ sample under excitation of only 5 meV above the FX energy (under light-hole exciton resonant-excitation conditions as aforementioned). The splitting has the same sign as the polarization (both are negative).
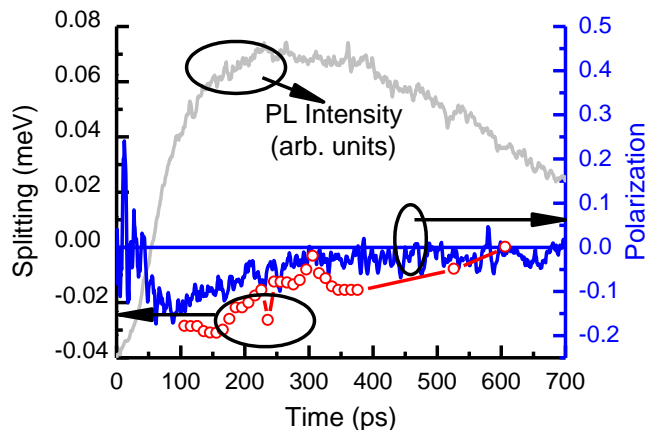
Figure 4. Time evolution of the PL intensity, polarization degree and spin splitting (circles) in the $Al_{0.15}Ga_{0.85}As$ sample under excitation at 1.736 eV (5 meV above FX energy) and $n = 1.2\times10^{16}$ cm^{-3}.

We use equation (1) to obtain the exciton density dynamics from $\rho(t)$ and $\Delta(t)$. We assume that the time evolution of n can be described with 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 exciton formation time τ_f equals to 70 ps in the $Al_{0.15}Ga_{0.85}As$ sample at an excitation density $n = 2\times10^{15}$ cm^{-3}. At higher excitation densities the exciton dissociation rates increase and the scattering rates decrease, which causes an increase of τ_f (150 ps at an excitation density $n = 1.5\times10^{16}$ cm^{-3} and 360 ps at 3×10^{16} cm^{-3} in the $Al_{0.15}Ga_{0.85}As$ sample).

4. Conclusion

We have investigated the exciton spin splitting in bulk GaAs and $Al_{0.15}Ga_{0.85}As$. In contrast to the results obtained in low-dimensional systems, in 3D the dynamics of the splitting is non-monotonous: it increases during 100 – 250 ps and then decays, reaching its maximum faster than the PL intensity. The increase of the splitting is due to the fast rise of the density of excitons formed from non-correlated *e-h* pairs. We used the exciton splitting dynamics to determine the exciton density $n(t)$. The exciton formation time was found from a fitting of $n(t)$, yielding $\tau_f = 70$ ps at an excitation density of 2×10^{15} and $\tau_f = 360$ ps at 3×10^{16} cm^{-3} in the $Al_{0.15}Ga_{0.85}As$ sample.

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