Diamagnetic coefficient of excitonic complexes in GaAs/Al\(_{0.3}\)Ga\(_{0.7}\)As quantum dots

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Abstract. We present an experimental investigation of the magneto optical properties of neutral exciton, neutral biexciton and charged exciton confined in self-assembled GaAs/Al\(_{0.3}\)Ga\(_{0.7}\)As strain free quantum dots grown by droplet epitaxy. We measured the diamagnetic shift \(\gamma\) for several quantum dots spanning an interval of 200 meV of the exciton emission energy. The dependence of \(\gamma\) on quantum dot size and shape is discussed together with a comparison with Stranski Krastanov and fluctuation induced GaAs quantum dots, as well as with quantum well case.

1. Introduction
The radiative recombination of excitons confined in semiconductor quantum dots (QDs) has been proven to be a suitable process for the efficient realization solid state sources of single photon, entangled photon pairs [1] and indistinguishable photons [2]. For these purposes the comprehension and the control of the Coulomb interactions among carriers and their responses to a magnetic field [3, 4] is of the utmost importance.

A powerful tool for the investigation of the excitonic recombination (electron and heavy hole recombination) in QDs is the micro–photoluminescence (\(\mu\)PL) spectroscopy of single nanoemitters. The recombination commonly observed in the \(\mu\)PL spectra comes from neutral exciton (one e-h pair, X), neutral biexciton (two excitons, XX) and charged exciton or trion (one exciton and an additional spectator charge, T). The Coulomb exchange interaction that couples the spins of e and h [5] may influence the level degeneracy introducing the so-called fine structure splitting (FSS). Alternatively
the fine structure of QDs excitons can be evidenced by the interaction with an external magnetic field [4] (see Fig. 1). The exciton spin state splitting, at zero field, depends on the symmetry of the Hamiltonian which in turn is influenced by several effects such as piezoelectric field due to strain in the lattice structure, segregation and geometrical elongation of the QD shape [5] (see Fig. 1). The interaction between carrier spin and an external magnetic field is twofold: the X transition is split in two components which energy distance increases linearly with the applied field and at the same time increases the average energy of the doublet with a quadratic dependence on the field. This is described in the following formula for the neutral exciton X:

$$E_X(B) = E_X(0) \pm \mu_B g_X B + \gamma_X B^2$$

where $\mu_B = 57.9 \mu\text{eV/T}$ is the Bohr magneton, $g_X$ is the Landé factor, $\gamma_X$ is the diamagnetic constant and B is the applied magnetic field. Finally, when the interaction with nuclear spins becomes strong, the Overhauser shift of the excitons energy has to be considered [3]. In this paper we present a detailed investigation of the magneto–optical properties of the excitonic complexes (X, XX and T) in strain free GaAs QDs grown by droplet epitaxy (DE) [6, 7]. By studying the diamagnetic shift $\gamma$ we extrapolate information about the QDs lateral confinement [8]. It is worth noting that DE GaAs/AlGaAs QDs are lattice matched heterostructure and all the strain and piezoelectric effects which affect the conventional Stranski–Krastanov (SK) nanostructures are lacking.

2. Experimental
The experiments were performed on GaAs self-assembled QDs in an Al$_{0.3}$Ga$_{0.7}$As barrier, grown by droplet epitaxy in a conventional molecular beam epitaxy apparatus [6, 7]. The sample was then annealed at 680 C for one hour [6, 7]. The DE samples can be grown with a wide spread of surface density and for singles QD spectroscopic investigation the value of 6 - 10QD/μm$^2$ can be achieved. By far field PL measurement on the ensemble we find an emission energy spanning over 250 meV[7]. The sample growth and its optical characterization can be found in Ref. [6, 7]. For accessing the single QD we used a confocal μPL setup in excitation–collection configuration. The CW excitation was performed above the Al$_{0.3}$Ga$_{0.7}$As barrier energy and was provided by a HeNe laser emitting at 534 nm. Excitation polarization was set to be linear preventing the Overhauser effect. The PL was collected by the main collection microscopy objective (NA = 0.42) and focused in a confocal single mode optical fiber with a core diameter of 3.5 μm (lateral resolution ~1 μm). The sample was cooled down to 10 K with a low vibration, cold finger, liquid He cryostat. The PL was dispersed with a grating monochromator and detected with a Si–based CCD camera. The spectral resolution was 150 μeV expressed as full width at half maximum (FWHM). The magneto-optical investigation was performed by using a magnetic field in Faraday configuration (B || z where z is the growth direction). The magnetic field was
provided by a superconductive, He cooled coil surrounding the cryostat and was finely tuned up to 5 T. The circularly polarized PL, resulting from the recombination of the split energy levels corresponding to different spin configuration, was analyzed with a quarter wave plate and a linear polarizer between the two objectives.

3. Results and discussion

Typical PL spectra of a single DE GaAs QD without applied magnetic field are shown in the bottom panel of Fig. 2 a). The spectra in the high excitation regime show several components that have already been attributed to neutral exciton X, positively charged exciton T and neutral biexciton XX, trough FSS and power dependence analysis [5]. However the resolution of the experimental setup for magneto–optical measurements doesn’t allow to resolve the small FSS observed in DE QDs [5], and the two circularly polarized contributions to the QD PL are identical within the experimental error. When the magnetic field B is applied each emission line splits into an oppositely circularly polarized doublet as shown in central and top panel of Fig. 2 a). By fitting the PL spectra with Gaussian profiles we can evaluate the evolution of each PL line with the magnetic field. An example of the result obtained for the T line is shown in the top panel of Fig. 2 b). This kind of analysis allows us to evaluate the diamagnetic shift $\gamma$ by fitting the average position of the two split lines ($\sigma^+$ and $\sigma^-$) with a quadratic dependence on the applied field as described in the following formula:

$$\left( E^\sigma_+ (B) + E^\sigma_- (B) \right) / 2 = \gamma \alpha B^2 $$

(2)

where $\alpha = X, T, XX$. In the case of the QD shown in Fig. 2 the values of $\gamma_\alpha$ are: $\gamma_X = 5.8 \pm 0.2 \mu eV/T^2$, $\gamma_T = 5.8 \pm 0.1 \mu eV/T^2$ and $\gamma_{XX} = 5.9 \pm 0.8 \mu eV/T^2$. The diamagnetic shift of the excitonic transition X reflects the spatial confinement acted on the exciton wavefunction and a measure of the lateral size of the QD can be obtained in the hypothesis of weak magnetic field [$a_0/L_B \ll 1$ where $a_0$ is the exciton Bohr radius and $L_B = \sqrt{\hbar/eB}$ is the magnetic length]. According to the following formula

$$\gamma_X = \frac{e^2 \langle \rho_X^2 \rangle}{8\mu}$$

(3)

where $\rho_X$ is the excitonic radius and $\mu$ is the exciton reduced mass (in GaAs $\mu = 0.06m_0$ where $m_0$ is the electron mass), we obtain an exciton extent of about $\sim 8$ nm for the QD shown in Fig. 2.
The resume of the measured diamagnetic shift $\gamma_\alpha$ is shown in Fig. 3. The values of $\gamma_\alpha$ for X, T and XX are quite similar even if strong fluctuation are found. The average value of $\gamma_X$ is $\sim 5.5 \, \mu\text{eV/T}^2$ which is remarkably smaller than the $25 \, \mu\text{eV/T}^2$ reported by Gammon and coworkers for fluctuation induced GaAs/AlGaAs QDs [10]. This is not surprising since their QDs provide a much smaller lateral confinement on the exciton wavefunction with respect to our DE QDs. In the case of QWs, where the lateral confinement of carriers is determined by the Coulomb interaction between electron and hole, larger values of $\gamma_X$ were found (up to hundreds of $\mu\text{eV/T}^2$ [9]). On the other side diamagnetic shift as small as 3 to 10 $\mu\text{eV/T}^2$ was already reported in literature for InAs/InP and In(Ga)As/GaAs QDs, which are quite similar to our result [11, 12, 13].

From the same set of data is possible to measure the Zeeman splitting and the corresponding Landé g factor. This analysis is presented in the bottom panel of Fig. 2 b) where the difference of the emission energy of the two split components of the T line are shown. By fitting the experimental data with the following formula

$$E^{\sigma+}_{\alpha}(B) - E^{\sigma-}_{\alpha}(B) = g_\alpha \mu_B B \tag{4}$$

we obtain $g_X = 0.9 \pm 0.02$, $g_T = 1.02 \pm 0.04$, $g_{XX} = 1.05 \pm 0.03$. As expected the values of $g_\alpha$ are quite similar for X, T and XX. The average value of the Landé factor for our DE QDs is about $\sim -1.25$ which is in good agreement with the value -1.3 found in fluctation induced GaAs/AlGaAs QDs.

4. Conclusions
In conclusion we have studied the magnetophotoluminescence of X T and XX in DE GaAs QDs by means of micro PL measurements. We measured Landé g factor and diamagnetic shift $\gamma$ for X, T and XX and we compared our results with QW counterpart and with SK QDs. We showed that the non–monotonic trend of the measured diamagnetic shift is in qualitative agreement with theoretical simulation and reflects the high geometrical aspect–ratio of DE QDs.

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