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Magnetic properties of charged excitons in self-assembled quantum dots

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We report on low temperature (4.2 K) magneto-luminescence measurements performed on charged tuneable GaInAs self-assembled quantum dots. We mapped the magnetic field dispersion (0–9 Tesla) of the exciton with excess electron charges set from 0 to 3. For the doubly and triply charged excitons the emission line shows a doublet corresponding to the singlet and triplet configuration of the quantum dot in the final state after photon emission. We map here for the first time the dispersion of the singlet state of the doubly charged exciton and show that the triply charged exciton undergoes a transition resulting from a magnetic field induced frustration of the Hund's rule.

1 Introduction Single dot spectroscopy of excitons confined in charge tuneable self-assembled quantum dots (QDs) has been recently reported [1–3]. Excitons that are doubly or triply charged show a splitting in the photoluminescence (PL) emission [1]. This splitting originates from the configuration of the final state of the charged QD after exciton recombination. Depending on the total spin of the final state, the configuration is in complete analogy with the He or Li atom singlet or triplet like. Since the total spin defines the configuration, it is desirable to investigate the effect of a magnetic field B on the PL of such excitonic complexes. Here we report the magnetic dependence of the PL of the doubly and triply charged exciton. In particular we map the magnetic properties of a single quantum dot when its excess charge is varied from 0 to 3. We found that the dispersion of the triply charged exciton differs dramatically from its lower charged counterpart. This new effect is interpreted in terms of a magnetic field induced frustration of Hund's rule that applies first to the final state of the triply charged exciton. We show here for the first time a direct optical manifestation of this effect, which was previous inferred from capacitance measurements [4].

2 Sample and experiment The sample is grown using solid source molecular beam epitaxy. A highly doped n^+ layer of GaAs is deposited and serves as a back contact. A 25 nm thin undoped GaAs layer is then grown. This layer will serve later as tunnel barrier for electrons. The self-assembled QDs are subsequently formed in a Stranski-Krastanov growth mode by depositing a 1.5 monolayer of InAs. At this

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stage, a thin InAs wetting layer dotted with lens shaped islands are formed. Then a 1 nm thin GaAs is deposited followed by a 1 min anneal step at growth temperature (520 °C) to allow QD shape and compositional changes [5]. In this way the obtained QDs become luminescent at around 950 nm wavelength range, which is detectable using Si based high quantum efficiency and low noise photodetectors. The QDs are covered with a 30 nm GaAs layer. A 116 nm short period AlAs/GaAs super lattice layer followed by a 4 nm GaAs cap is finally deposited preventing carrier tunnelling to the surface. In order to apply a gate voltage (V_g) between the surface and the back contact the sample is processed with a 5 nm thin semitransparent NiCr electrode. The final process step is made by forming apertures in a 150 nm Al layer deposited onto the sample. The apertures allow reducing the number of optically investigated QDs to a single or a few. For the PL measurements, we used a home made cryogenic scanning probe confocal microscope compatible for operation at high magnetic fields (0–9 T). The magnetic field is applied along the sample growth axis. Electron–hole pairs are optically excited at $\lambda = 820$ nm in the InAs wetting layer. The excitation power was chosen to be low enough to avoid bi-exciton generation in the QDs. The PL is collected with a NA 0.85 objective through the mask apertures and focused on the 5 μm core of a single mode fiber. The PL is then ported this way to the detection, dispersed on a 300 mm focal length grating spectrometer and detected with a cooled CCD camera. All measurements were performed at 4.2 K in a He exchange gas.

3 Results and discussion

3.1 Identification of charged excitons The excess charge in the QDs can be tuned from 0 up to 3 electrons [1]. We applied a gate voltage within the range of $V_g = -0.8$ V to +0.1 V. Figure 1a shows the PL measured on a single quantum dot as a function of V_g. For $V_g < -0.8$ V the PL is totally quenched because the electron tunnels out of the QD faster than exciton recombination life time [1]. At $V_g > -0.8$ V, the neutral exciton becomes radiative. Subsequently each time an electron is added electrostatically in the QD, the PL line red shifts due to Coulomb blockade effect [1–3]. The identification of the exciton charge is therefore obtained. Here, the emission lines are labelled by X^0, X^{1-}, X^{2-} and X^{3-} indicating the neutral, singly, doubly and triply charged exciton respectively. A further confirmation of the charge state identification is obtained by observing that in contrast to X^0 and X^{1-}, the X^{2-} and X^{3-} excitons are split into a singlet and triplet line [1]. This splitting energy is proportional to the exchange interaction between the electrons of the s and p states. In order to measure the magnetic field behaviour of the PL, the gate voltage is set at constant values corresponding to the X^0 ($V_g = -0.62$ V), X^{1-} ($V_g = -0.32$ V), X^{2-} ($V_g = -0.14$ V) and X^{3-} ($V_g = 0$ V).

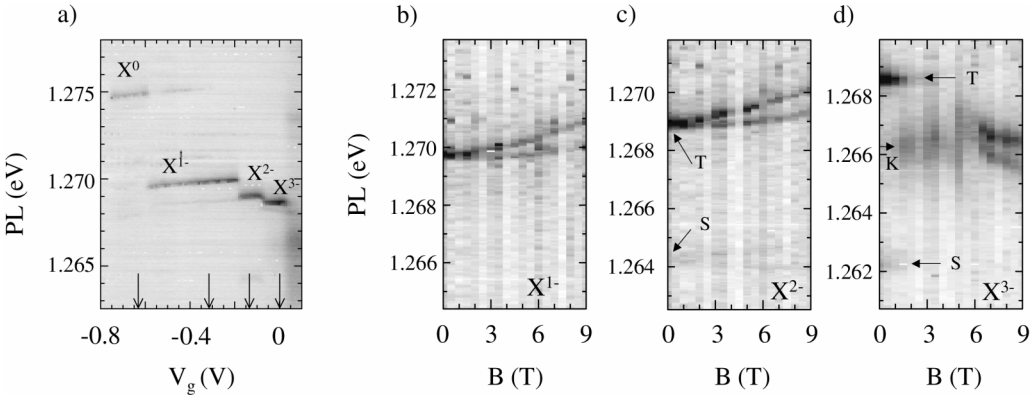

Fig. 1 a) PL measurement performed on a single quantum dot, as a function of gate voltage, as a function of the magnetic field for the b) singly, c) doubly and d) triply charged excitons. The arrows in a) indicate the gate voltage at which the magnetic field measurements were performed. The singlet (S) and triplet (T) state configurations of the X^{2-} and X^{3-} excitons as well as the new line (K) are indicated.

3.2 Magnetic field measurements Figures 1b to d show the measured PL as a function of B for the X^{1-}, X^{2-} and X^{3-} excitons performed on the same single quantum dot. The dispersion of the PL is obtained by plotting the maximum intensity of the emission line as a function of B as shown in Fig. 2. For all measured charged excitons we observe a characteristic Zeeman spin splitting independent of the dot charge [6, 7]. For the X^0, X^{1-} and the X^{2-} triplet exciton we also observed a diamagnetic shift. The data fit the quadratic form $E = E_0 + sg_{ex}\mu_B B + \alpha B^2$ where E_0, s, g_{ex}, μ_B and α represents the PL emission energy at zero magnetic field, the spin quantum number (+1/2 or –1/2), the effective excitonic g-factor, Bohr's magneton and the diamagnetic coefficient respectively. We obtained a diamagnetic shift of $\alpha = +9.78$ μeV/T^2 and an excitonic g-factor of $g_{ex} = 1.63$ for the X^0 exciton. Similar values are found for the X^{1-} ($\alpha = 9.44$ μeV/T^2, $g_{ex} = 1.70$) and the X^{2-} triplet ($\alpha = 9.50$ μeV/T^2, $g_{ex} = 1.63$) charged excitons. The fits in Fig. 2a show clearly that for this quantum dot, the diamagnetic shift as well as the spin splitting in the PL do not depend on the exciton charge [6, 7].

3.3 Suppression of the diamagnetic shift in the X^{2-} singlet configuration In contrast to the quadratic diamagnetic shift measured on the X^0, X^{1-} and X^{2-} triplet excitons, we find a completely different behaviour for the X^{2-} singlet exciton. Figure 2b illustrates this. Here although the spin splitting of the PL line remains unaffected, the quadratic diamagnetic contribution disappeared. We propose the following tentative interpretation based on a parabolic QD confinement model [6, 7]. The X^{2-} triplet has a total angular momentum $S = 1$, the spin wave function is symmetric while the spatial component of the wave function is antisymmetric. Therefore the electrons are well separated reducing coulomb interaction. The opposite is true for the singlet state ($S = 0$), so in this case electron–electron interactions are enhanced. We have shown previously that the effect of an enhanced coulomb interaction leads to the reduction of the excitonic diamagnetic shift [7]. The calculated effect is found however to be much weaker than the almost complete suppression of the diamagnetic shift measured here. This aspect is still not understood.

3.4 Magnetic dispersion of the X^{3-} exciton Figure 1d shows that the X^{3-} exciton behaves in magnetic field in a way drastically different from its lower charged excitons counterparts. First at $B = 0$, we see both the X^{3-} exciton singlet and triplet lines, which are $\Delta E_{ST} = 6$ meV apart. Beyond a critical field of about 1.5 T both lines fade, and finally disappear at about 3 T. At the same time, a new line (K-Peak) appears at 2 meV below the triplet and 4 meV above the singlet line. This new peak has a width of about 1 meV. This is in contrast to the X^{3-} triplet line, which is limited by the resolution of our spectrometer. At higher magnetic fields the K-peak shows a well-defined anticrossing. The behaviour of the K-line at high magnetic field is presently understood and will be reported elsewhere. We focus now on the field region where the triplet and singlet line disappears and a new line grows. We associate this effect with a

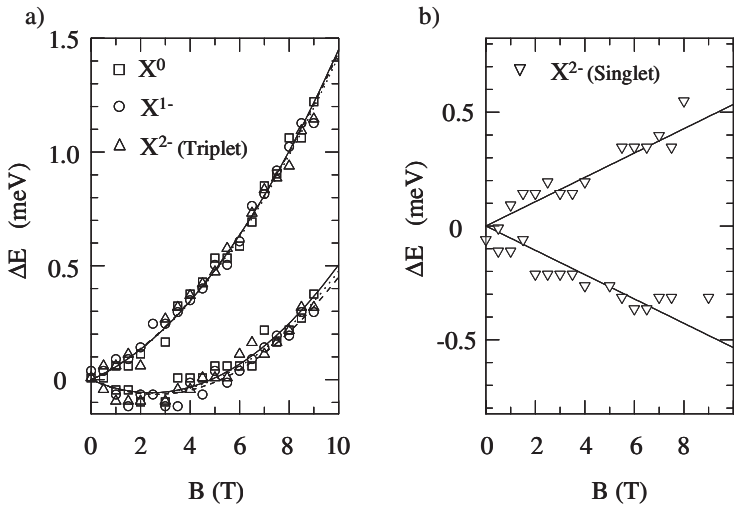

Fig. 2 a) Comparison of parabolic magnetic dispersion for X^0, X^{1-} and X^{2-} (Triplet) excitons. b) The X^2 (Singlet) shows non diamagnetic behaviour ($\alpha \sim 0$, $g_{ex} = 1.85$).

magnetic field induced change in the electronic configuration of the X^{3-} exciton. At $B = 0$, both p-level orbitals of the QD are occupied with one electron each, and both electrons have parallel spins. This configuration is driven by the exchange energy between the p-level electrons. When a magnetic field is applied, one of the p-orbital shifts proportionally to B to higher energy while the other orbital shifts downward. Beyond a critical field the energy separation between the two orbitals exceeds the exchange energy and both electron transfer to the lowest energy (paramagnetic) p-orbital. The new line (K-peak) is the PL emission of the X^{3-} for which the exchange interaction has been suppressed. This interpretation is confirmed by the fact that the K-peak energy position appears precisely at 1/3 ΔE_{ST} bellow the triplet line and 2/3 ΔE_{ST} above the singlet line. This is the expected energy position when the exchange interaction in the final state is suppressed.

4 Conclusion We measured the magnetic field dispersion of charge tuneable excitons confined in a single QD. To within our experimental resolution, we found that the excitonic *g*-factor is charge independent. The dispersion of the X^0, X^{1-}, and X^{2-} triplet lines exhibits a regular diamagnetic shift quadratic in B. In contrast the X^{2-} singlet exciton line shows a suppressed diamagnetic behaviour possibly due to electron–electron interaction. The X^{3-} exciton dispersion shows a dramatically different behaviour. There, Hund's rules are frustrated beyond a critical magnetic field, leading to a suppression of the singlet-triplet splitting into a new broad PL line. At higher magnetic field this new line shows anti crossings with the Landau levels of the wetting layer continuum.

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