

Magneto optics of a doubly charged quantum dot - Observation of a negative diamagnetic shift

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We present magneto-optical studies of a self-assembled semiconductor quantum dot, concentrating specifically on the case in which the dot is doubly positively charged, studying this way the confined hole - hole exchange interaction. A simple harmonic potential model, which we extend to capture the influence of an externally applied magnetic field in Faraday configuration fully describe the observed polarization sensitive magneto-photoluminescence spectra. We deduce the effective composition of the quantum dot from its measured electronic g -factor. Using this value we determine the dot effective permittivity and quantitatively describe various measured excitonic transitions, their measured Zeeman splittings and their diamagnetic shifts. In particular, the model quantitatively accounts for an observed pronounced negative diamagnetic shift, which provides a direct measure for the hole-hole exchange interaction and its dependence on the externally applied magnetic field strength.

I. INTRODUCTION

Self-assembled Quantum Dots (QDs) in semiconductors form a well-known platform for quantum technologies. They have proven to be the best contemporary single-photon sources [1–4], while providing an excellent interface between anchored spin qubits and “flying” photon qubits. Much progress has been made in controlling confined-spin qubits in QDs [5–8] and entangling them with photons Sen[9–14], enabling deterministic generation of long strings of entangled photons [15–17]. In addition, QDs still provide a convenient platform for studying the many-body states of confined many carriers complexes. Interesting properties of such complexes include the relative interactions between the consisting particles, the form of their spatial wavefunctions, and their response to externally applied fields, to name a few. In particular, an externally applied magnetic field causes the associated optical transitions to energetically shift - an effect known as the *diamagnetic shift*. Modeling those shifts in confined systems is still an ongoing effort [18–22].

Here we present a magneto-optical study of semiconductor QDs in which we focus our attention on the optical properties of a doubly positively charged QD and its fundamental excitonic transition denoted by X^{+2} . The QD confined X^{+2} exciton contains three heavy holes and an electron. After radiative recombination of an electron-hole pair, the QD remains with two heavy-holes. The pairs of holes may form either three spin triplet states or one spin singlet state. Our work was spurred by noticing an anomaly in the diamagnetic shifts of the optical transitions into the singlet state, the $X_{S_0}^{+2}$, which we

found to be negative. In the effort to understand this phenomenon, we found that the X^{+2} excitonic transitions form an excellent platform for studying the hole-hole Coulomb exchange interaction and its dependence on externally applied magnetic field. We show, using a simple harmonic model for the QD spatial potential, that the measured diamagnetic shifts of the X^{+2} transitions and the measured electron and hole g -factors can be obtained using one free fitting parameter which describes the effective composition of the QD.

The paper is organized as follows. First, we present our measurements of the electron and hole g -factors by measuring the spectral Zeeman splittings of the bright and dark neutral excitons. From the values of the measured g -factors, we extract the composition of the QD as captured by the parameter x , defining the ratio of Indium and Gallium in the QD, $\text{In}_x\text{Ga}_{1-x}\text{As}$. Next, we present full polarization-sensitive magneto-PL measurements displaying the diamagnetic shifts of various optical transitions and their optical transition selection rules. We then concentrate on the anomalous negative shift of the doubly positively charged exciton, the $X_{S_0}^{+2}$, and fit its optical transition field dependence using no additional free parameters. Finally, we use a Hartree-Fock approximation to calculate the absolute values of the X^{+2} diamagnetic shifts in terms of the diamagnetic shift of the bright exciton, X_{BE}^0 .

II. EXPERIMENTAL SYSTEM

We studied a single $\text{In}_x\text{Ga}_{1-x}\text{As}$ self-assembled QD embedded in a planar microcavity grown along the [001] direction [23]. We used an Attocube closed-cycle cryostat to cool the sample down to 4 Kelvin. A built-in vector magnet enabled us to apply a magnetic field in any

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desired direction. The emitted photoluminescence (PL) was collected by a $\times 60$ objective. Its polarization was analyzed by pairs of liquid crystal variable retarders and polarizing beam splitters, enabling PL polarization projecting on any direction in the Poincaré sphere. The PL was then spectrally analyzed using an 80 cm double monochromator, providing a spectral resolution of $\sim 20 \mu\text{eV}$.

The QD was optically excited using an above band-gap CW red HeNe or a blue diode laser, emitting at 633 or 445 nm, respectively. The excitation colors induce the average charge state of the QD. While HeNe illumination results in positive charging blue excitation leads to negative charging [24].

We defined the symmetry axis of the QD and the optical beam direction as the z -axis of the experimental system. The x and y axes were defined along the polarization eigenstates of the QD's bright exciton, X_{BE}^0 . The X_{BE}^0 is an electron-hole pair which can be expressed in the spin basis $\{|+z\rangle = |\uparrow\downarrow\rangle, |-z\rangle = |\downarrow\uparrow\rangle\}$ with $\uparrow\downarrow$ and $\downarrow\uparrow$ denoting the spin projections of the heavy-hole and electron onto the z -axis. Since a heavy-hole and an electron have total angular momenta of $3/2$ and $1/2$, respectively, the angular momentum projection of a $|\uparrow\downarrow\rangle$ ($|\downarrow\uparrow\rangle$) pair along this axis is $+1$ (-1) [25]. Consequently, optical recombination of the $|\uparrow\downarrow\rangle$ and $|\downarrow\uparrow\rangle$ pairs results in a right-handed (R) and left-handed (L) circularly polarized photon emission, respectively. The anisotropic electron-hole exchange interaction in this QD lifts the degeneracy of the above basis by $\delta_1 \approx 30 \mu\text{eV}$ [26] thus forming new eigenstates, $\sqrt{2}|\pm x\rangle = |\uparrow\downarrow\rangle \pm |\downarrow\uparrow\rangle$. Recombination of those excitonic eigenstates results in either horizontal, $\sqrt{2}H = R + L$, or vertical $\sqrt{2}V = R - L$ rectilinear photon emission, enabling a one-to-one correspondence between the X_{BE}^0 's two-level system and the two-dimensional space of light polarization.

The dark exciton (X_{DE}^0) is another electron-hole state, but with parallel spins $\sqrt{2}|\psi_{DE}\rangle_{1,2} = |\uparrow\uparrow\rangle \pm |\downarrow\downarrow\rangle$. In general, this state is optically inactive because the angular momentum is not conserved upon recombination [8]. However, small optical activity of the X_{DE}^0 was measured [27–29]. Zielinski et al. explained it by small mixing of the X_{DE}^0 and X_{BE}^0 eigenstates [28], which can be enhanced by applying an in-plane magnetic field perpendicular to the QD optical axis (Voigt configuration). For a magnetic field parallel to the symmetry axis (Faraday configuration) no additional mixing occurs, and the X_{DE}^0 barely emits [30].

III. RESULTS

A. Measuring single-carrier g -factors

The Zeeman interaction between an externally applied magnetic field and QD confined carriers' spin removes the Kramers' degeneracy between the confined carriers spin state which is parallel and anti-parallel to the field

direction. The Zeeman interaction linearly depends on the magnetic field magnitude. This dependence is most generally expressed in terms of a 3×3 g -factor tensor [31]. For simplicity we assume here that this tensor is diagonal and have only two different components: along the symmetry axis (g_e^z and g_h^z) and perpendicular to it ($g_e^\perp$ and $g_h^\perp$) [32].

In the first part of the experiment, we measured the confined electron and hole g -factors tensor components along the z -axis. This was done by measuring the Zeeman splitting of the X_{BE}^0 and X_{DE}^0 under B-field in the $\hat{z}$ direction. Assuming that the absolute magnitude of the g -factors of those transitions are given by the sum and difference of the absolute magnitudes of the single-carrier g -factors

$$|g_{BE(DE)}^z| = |g_e^z| \pm |g_h^z| \quad (1)$$

[33, 34], we were able to extract g_e^z and g_h^z from the measured g_{BE}^z and g_{DE}^z [30, 32, 35]. Eq. 1 is derived from the parallel and anti-parallel spin nature of the dark and bright excitons, using the sign convention given by the Zeeman Hamiltonian

$$\mathcal{H} = -\mu_B g_e^z B_z S_z + \frac{1}{3} \mu_B g_h^z B_z J_z. \quad (2)$$

Here, μ_B is the Bohr magneton, S_z and J_z are the angular momentum z -projections $\pm \frac{1}{2}$ and $\pm \frac{3}{2}$, and B_z is the magnetic field. We readily measured the Zeeman splitting of the X_{BE}^0 since its spectral doublet appears bright in the PL, as shown in figure 1. In contrary, the low optical activity of the X_{DE}^0 made its Zeeman splitting measurement more challenging. To overcome this problem, we added a 1.5T Voigt component to our measurement (in-plane B-field), enhancing the X_{DE}^0 emission to a measurable amount. Although the total B-field direction was no longer in the $\hat{z}$ direction, we found that the in-plane field effect on the measured $\hat{z}$ direction Zeeman splitting could be safely neglected. We verified it by measuring the influence of the in-plane field on the *bright* exciton splitting (X_{BE}^0), and found that it stayed unaffected within our experimental precision.

One can also notice in Figure 1 that the X_{DE}^0 cross-polarized doublet is not equally intense: at 0 Tesla, its horizontally (H) polarized component is much stronger than the vertically (V) polarized one, a phenomenon observed and explained in previous publications [36–38]. Adding magnetic field in Faraday configuration enhances the weaker component and gradually adds cross-circular polarization terms to the X_{DE}^0 doublet. However, up to the maximal field strength of 1.5 Tesla, the two X_{DE}^0 's components remain unequal. Nonetheless, we extracted the g -factors of the X_{BE}^0 and X_{DE}^0 by fitting their measured Zeeman splittings to the following expression:

$$\Delta E_{BE(DE)} = \sqrt{\delta_{1,2}^2 + (\mu_B g_{BE(DE)} B)^2} \quad (3)$$

, where $\delta_{1,2}$ are the fine-structure splittings of the X_{BE}^0 and X_{DE}^0 at 0 Tesla, respectively. We summarize the val-

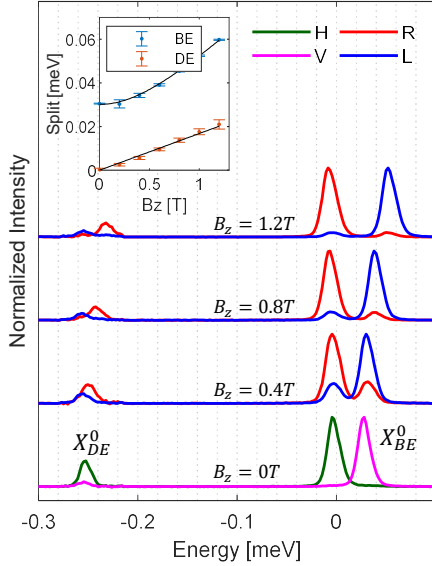


Figure 1. Polarization-sensitive magneto-PL of the bright exciton (X_{BE}^0) and dark exciton (X_{DE}^0), for various magnetic field strengths in the $\hat{z}$ direction. A constant, $\hat{x}$ -directional magnetic field of $1.5T$ was applied during all the measurements to allow the X_{DE}^0 optical transition. Inset: Zeeman splitting of the X_{BE}^0 and X_{DE}^0 versus B_z -field. The g -factors of the two transitions are extracted by fitting the measured splitting with $\Delta E = \sqrt{\delta^2 + (\mu_B g B)^2}$.

Spectral line	$\delta[\mu eV]$	g^z -factor	$\alpha[\frac{\mu eV}{T^2}]$
X_{BE}^0	31.0 ± 1.8	-0.81 ± 0.01	8.44 ± 0.14
X_{DE}^0	$1.4^* \pm 0.1$	-0.29 ± 0.02	7.0 ± 1.4
	g_e^z	-0.55 ± 0.02	
	g_h^z	-0.26 ± 0.02	
	$\Delta_0[\mu eV]$	270 ± 10	

Table I. Summary of the measured excitonic fine structure and Zeeman parameters. δ is the natural splitting at $B = 0$, and α is the diamagnetic shift coefficient capturing the quadratic dependence of the energy in B (αB^2). $g_{(e/h)}^z$ is the measured g -factor of the electron and hole in Faraday configuration, respectively. *The dark exciton (X_{DE}^0) splitting is too small to be directly observed in PL measurement, but can be measured using time-resolved spectroscopy [8]. For reference, we also added the $X_{DE}^0 - X_{BE}^0$ splitting denoted by Δ_0 .

ues of the measured excitonic and single-carrier g -factors in table I.

B. Estimating the QD effective composition from the measured electronic g -factor

The isotropic electronic g -factor in bulk semiconductors can be analytically calculated by the Roth's formula

[39]:

$$g_e = 2 - \frac{2}{3} \frac{E_p \Delta}{E_g(E_g + \Delta)} \quad (4)$$

where E_g is the band gap energy between the valence and conduction bands, Δ is the split-off gap (between the valence band and the spin-orbit band) at $k = 0$, and E_p is the Kane energy defined as $E_p \equiv \frac{2\hbar^2}{m} |\langle s | \partial_x | x \rangle|^2$, where $|s\rangle$ and $|x\rangle$ are the crystal Bloch functions of the electron in the conduction band and in one of the three p -like degenerate valence band, respectively.

QDs are not bulk semiconductors, and applying Roth's formula to them requires adjustment [40]. In principle, the confinement effect of the QD breaks the periodicity of the electronic wavefunctions, and the derivation of Roth's formula collapses. Nevertheless, as long as the confinement energy is much smaller than the parameters Δ , E_p and E_g , we expect Roth's formula to be a good approximation. Indeed, a typical separation between confinement energy levels in our QD is of order $10 - 30 meVs$ [41], much smaller than Δ , E_p and E_g (see table II).

Since our QD comprises two semiconductors, GaAs and InAs, we averaged the values of Δ and E_p over the two bulk materials. We introduced a weight parameter x to quantify the composition of the QD, $In_xGa_{1-x}As$, and define:

$$\Delta^x = x\Delta_{In} + (1-x)\Delta_{Ga}, \quad E_p^x = xE_{p(In)} + (1-x)E_{p(Ga)}$$

For the band gap, E_g , we preferred to use the directly measured value of the X_{BE}^0 emission, as it takes into account the confinement and lattice mismatch strain effects, omitted in the simple average [42]. We defined a corrected band gap $\widetilde{E}_g$ by adding $\sim 50 meV$ to the X_{BE}^0 emission energy, thus accounting for the binding energy of the exciton [25]. Combining all three parameters, we obtained an x -dependent Roth formula

$$g_e(x) = 2 - \frac{2}{3} \frac{E_p^x \Delta^x}{\widetilde{E}_g(\widetilde{E}_g + \Delta^x)} \quad (5)$$

that we can fit to the electronic g -factor as measured in the experiment. Fitting $g_e(x)$ to the measured value of -0.55 yields $x \approx 0.75$. We summarize the parameter values of that calculation in table II.

C. Measuring a negative diamagnetic shift for X^{+2}

In Figure 2, we present a full Magneto-PL measurement in Faraday configuration of the various spectral lines of our QD. We present it for two average charge states of the QD: negative and positive. The charge state is apparent in each case by considering the emission ratio between the positive and negative trions, X^+ and X^- . Many identified lines are marked in the PL following previous studies [24].

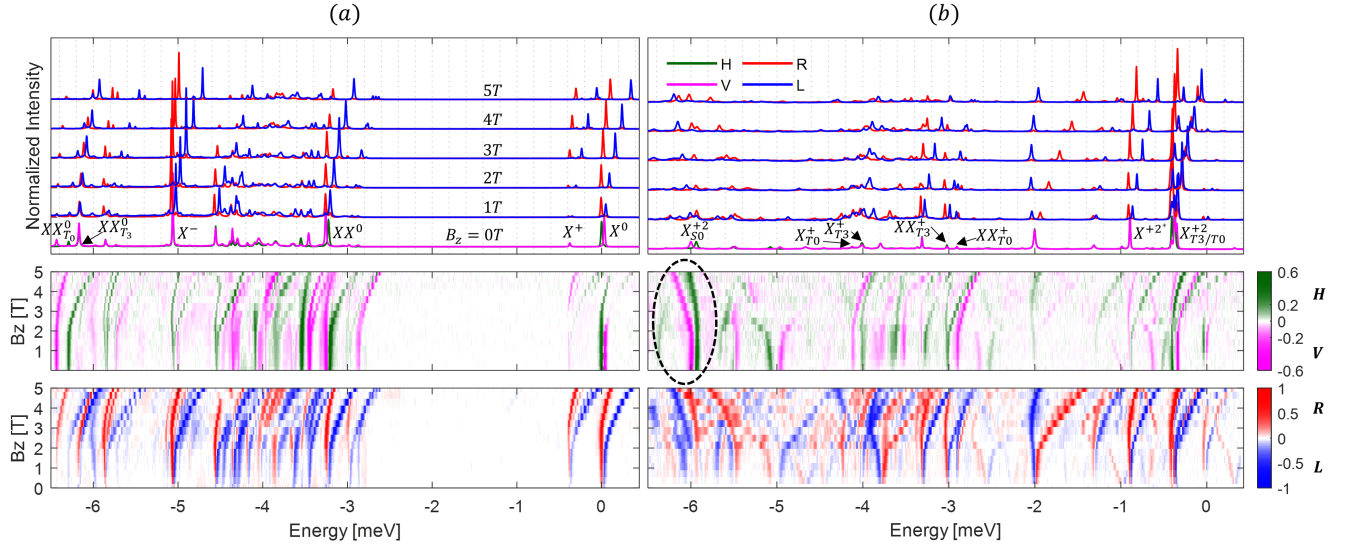


Figure 2. Polarization-sensitive magneto-PL spectra in Faraday configuration for various magnetic field strengths, for negatively (a) and positively (b) charged QD. The upper panel shows polarization-sensitive magneto-PL spectra (except at $B = 0T$, where the rectilinear polarization is shown). The panels below show the degree of circular and rectilinear polarizations (given by the color bars to the right) as a function of the photon energy and the externally applied magnetic field strength. The identified spectral lines are marked: X^0 - the exciton, XX^0 - the biexciton, X^+ - (X^-) positively (negatively) charged trion, $XX_{T_0(T_3)}^0$ metastable biexcitons with the two holes in $T_0(T_3)$ spin Triplet configurations. $X_{T_0(T_3)}^+$, and $XX_{T_0(T_3)}^+$ are similar positively charged excitons and biexcitons. The X^{+2} lines result from the recombination of the doubly positively charged exciton, leaving behind two holes which can form either a singlet S_0 or one of the triplets, $T_{\pm 3}$ or T_0 . Note the negative diamagnetic shift of the $X_{S_0}^{+2}$ (marked with an oval dash line). The energy scale is relative to the X_{BE}^0 spectral line at zero magnetic field.

	GaAs	InAs	In _{0.75} Ga _{0.25} As
E_p [eV]	28.8	22.2	23.85
$E_g(4K)$ [eV]	1.519	0.418	1.334*
Δ [eV]	0.341	0.371	0.363
g_e Calculated	-0.317	-14.65	-0.55
g_e Measured	-0.484	-14.9	-0.55

Table II. Comparison between measured electronic g -factors to calculated values using Roth's formula. The measured values for bulk GaAs and InAs semiconductors are taken from Ref. [43]. The parameters E_p and Δ for In_{0.75}Ga_{0.25}As are weighted averages of their values in GaAs and InAs. *The corrected band gap $\widetilde{E}_g$ (see text).

On top of the Zeeman splitting of the spectral lines, they undergo a quadratic-in- B diamagnetic shift, which we characterize by the coefficient α in the term αB^2 added to the Hamiltonian (Eq. 2). For each spectral line in Figure 2, the shift is attributed to its spectral “center of mass” (the spectral center of the doublet), which in most cases shifts towards higher energy (hence the terminology of “diamagnetic” versus “paramagnetic” shift). We explain this tendency by considering the areas of the initial and final states of each optical transition. It is a well-known result for quantum wells that the diamagnetic shift of a neutral exciton is proportional to its wavefunc-

tion area [43] in a plane which is normal to the direction of the magnetic field:

$$\alpha = \frac{e^2}{8\mu_{\parallel}c^2} \langle f | \hat{\rho}^2 | f \rangle = \frac{\pi}{4} \frac{e^2}{\mu_{\parallel}c^2} \int f^2(\rho) \rho^3 d\rho \quad (6)$$

Here, ρ is the relative in-plane coordinate between the electron and hole, $f(\rho)$ is the excitonic envelope wavefunction, $\mu_{\parallel} = m_e m_h / (m_e + m_h)$ is the in-plane reduced mass of the electron and hole, and e and c are the electron charge and the speed of light, respectively. The final state of the QD after the excitonic recombination is just the vacuum, possessing no magnetic dependence, so the overall diamagnetic shift is positive. Extending the area interpretation to other optical transitions, it seems that in most cases the radiative recombination results in a final configuration with a reduced area. As a result, most lines follow positive diamagnetic shift. Quantitatively, plugging in Eq. 6 $m_h = 0.25m_0$, $m_e = 0.065m_0$ (the effective masses of the hole and electron in the quantum dot [44] with m_0 the free electron mass), and the measured diamagnetic coefficient $\alpha = 8.44 \pm 0.14 \mu\text{eV}/T^2$, one calculates the exciton Bohr radius to be $\sim 3.7 \text{ nm}$ – a compatible result with the $\sim 30 \text{ nm}$ estimated diameter of our QD.

Figure 3 summarizes the diamagnetic shifts of several selected lines. One can see that many lines, including the X_{BE}^0 , XX^0 and the trions, X^- and X^+ , exhibit very similar diamagnetic shifts of $\sim 8 \mu\text{eV}/T^2$. We explain this

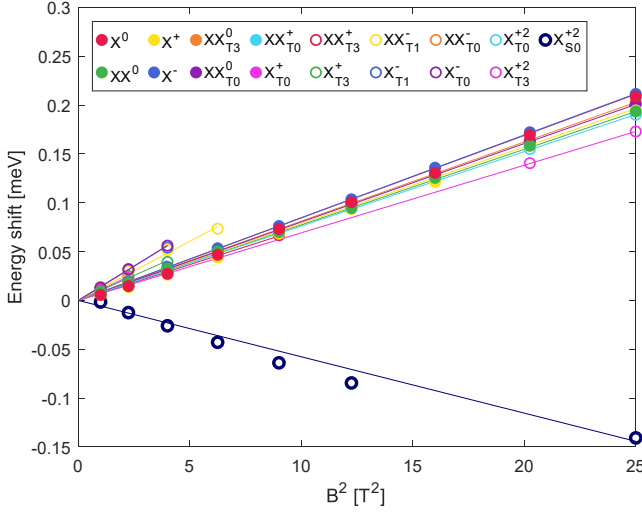


Figure 3. Measured energy shifts of various optical transitions as a function of B^2 . One spectral line is a prominent exception - the $X_{S_0}^{+2}$

similarity by arguing that in all those transitions both the initial and final states contain only charge carriers occupying the QD ground-level. We observe that when additional charge carriers occupy higher confined levels, the diamagnetic shift coefficients change (see for example $X_{T\pm 1}^-$, $X_{T\pm 3}^+$).

Interestingly enough, one prominent line that we attribute to the doubly charged exciton transition $X_{S_0}^{+2}$ exhibits a distinctive *negative* shift. In what follows, we will try to explain this observation in terms of the exchange interaction between the two heavy holes of the X^{+2} transitions' final states. Let us start by describing those transitions in detail.

D. The doubly charged exciton X^{+2}

Figure 4 schematically describes the energy levels and the optical transitions associated with the doubly charged exciton, X^{+2} . This exciton comprises one electron in the ground-level $1e^1$, and three holes: two of them forming a singlet in the s-orbital ground-level, $1h^2$, and the third one occupies the first excited p-level $2h^1$. Here, np^m means: n - the energy level order, p - the particle type (e or h), and m - the number of particles occupying this level (either 1 or 2). The exchange interaction between the unpaired electron (in the 1st level) and hole (in the 2nd level) removes the degeneracy between the four possible two-carriers' spin configurations, forming four distinct eigenstates similar to the case of the neutral exciton (X^0). As such, we borrow the exciton “bright” and “dark” terminology to describe the eigenstates of the X^{+2} as well. States with anti-parallel e-h spins would be called “bright-like”, while states with parallel spins - “dark-like” (see Figure 4). We emphasize that the dark and bright

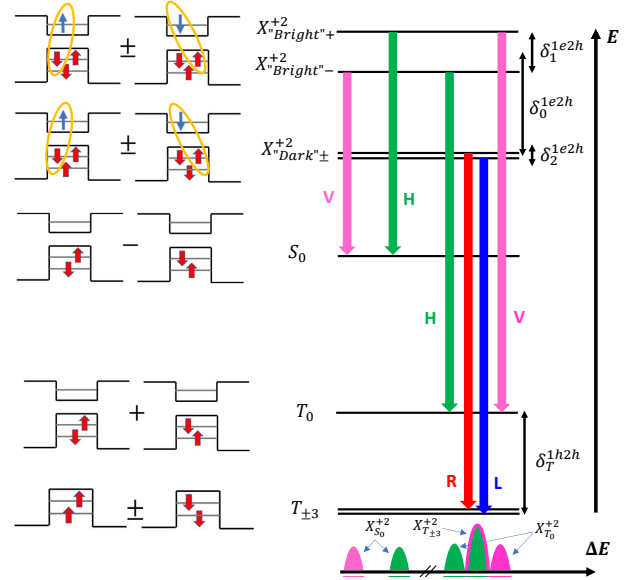


Figure 4. Schematic description of the energy levels and optical transitions associated with the doubly positively charged exciton X^{+2} . The configuration of each state is presented on the left, where thin blue arrows represent electrons with spin $\frac{1}{2}$, and thick arrows represent heavy-holes with spin $\frac{3}{2}$. The polarization selection rules are marked by colored downward arrows. H (V) marks the horizontal (vertical) rectilinear polarization, while R (L) marks right (left) circular polarization. A schematic description of the emitted PL is drawn at the bottom. The $X_{T_{\pm 3}}^{+2}$ spectral line is drawn in green with a pink edge, symbolizing that the H and V polarizations overlap such that the emission is unpolarized.

states are both optically active since the optical recombination occurs between the unpaired s -electron and one of the s -level singlet holes, rather than the unpaired p -hole.

The final states of the X^{+2} recombination contain two holes - one in the ground level and one in the first excited level. As identical particles, they form either one singlet spin state denoted by S_0^{1h2h} or three triplet states denoted by $\{T_0^{1h2h}, T_{\pm 3}^{1h2h}\}$, respectively. The two initial bright-like exciton states can only recombine to the singlet S_0^{1h2h} or triplet T_0^{1h2h} final states (but not to the $T_{\pm 3}^{1h2h}$), resulting in two pairs of cross-rectilinearly polarized doublets [45]; the dark-like states can only recombine to the $T_{\pm 3}^{1h2h}$ states. Since in the absence of external magnetic field ($B = 0$) both the dark-like and the $T_{\pm 3}^{1h2h}$ states are almost degenerate, the recombination results in a single, unpolarized, strong spectral line. We label the X^{+2} optical transitions by their final states, specified by the subscripts: $X_{T_0}^{+2}$, $X_{T_{\pm 3}}^{+2}$ and $X_{S_0}^{+2}$. The latter transition, $X_{S_0}^{+2}$, is the one exhibiting a negative diamagnetic shift. We note that in the absence of external field, the unpolarized $X_{T_{\pm 3}}^{+2}$ spectral line is positioned exactly in between the two cross linearly polarized components of the $X_{T_0}^{+2}$ line. This indicates that δ_0^{1e2h} , denoting the split-

Spectral line	$\delta_1 [\mu\text{eV}]$	g -factor	$\alpha [\frac{\mu\text{eV}}{T^2}]$	Model
X_{BE}^0	31.0 ± 1.8	-0.81 ± 0.01	8.44 ± 0.14	$g_{1e}^z + g_{1h}^z$
$X_{T_3}^{+2}$	0	-0.97 ± 0.05	6.90 ± 0.25	
$X_{T_0}^{+2}$	56.3 ± 2.3	0.55 ± 0.13	7.6 ± 0.5	$g_{1e}^z + g_{2h}^z$
$X_{S_0}^{+2}$	69.0 ± 6.8	0.65 ± 0.08	-5.8 ± 0.7	
X_{DE}^0	0	-0.29 ± 0.02	7.0 ± 1.4	$g_{1e}^z - g_{1h}^z$

Table III. Summary of the measured g -factors for the X^{+2} transitions, compared to those of the bright and dark excitons. The lines are classified by a simple model which assumes that the g -factor of a given transition can be decomposed to the sum of the comprising charge carrier g -factors of the initial and final states of that transition. $g_{n(e/h)}^z$ denotes the g -factor of the electron (hole) in the n energy level of the QD, where $n = 1$ is the ground state.

ting between the dark-like and bright-like X^{+2} states, is equal to δ_T^{1h2h} , the splitting between the holes' triplet states T_0^{1h2h} and $T_{\pm 3}^{1h2h}$. The reason why these two terms, one due to isotropic e-h exchange and the other due to h-h anisotropic exchange, are almost equal, remains an open question.

The measured diamagnetic shifts and g -factors of the X^{+2} transitions are summarized in table III. To qualitatively explain the measured g -factors, we assume that a g -factor of a state can be deduced by summing up its individual single carrier component's g -factors, and that the total g -factor of a transition results from the difference between its initial and final states. By further assuming that charge carriers in the well-defined symmetry configurations S_0 and T_0 do not exhibit Zeeman splitting, we conclude that $X_{T_3}^{+2}$'s g -factor behaves as the bright excitation's (X_{BE}^0) factor, while the g -factors of $X_{T_0}^{+2}$ and $X_{S_0}^{+2}$ depend on the *excited* hole's g -factor, g_{2h}^z . More measurements justifying the above classification as a general result can be found in the authors' theses [46, 47]. It is interesting to note that plugging into the $g_{1e}^z + g_{2h}^z$ sum the measured g_{1e}^z -factor (~ -0.55), and using for the sum an averaged value of 0.6 ± 0.1 (see table III), one finds that $g_{2h}^z = 1.15 \pm 0.10$. This value is opposite in sign compared to the ground state g -factors of the hole and the electron (-0.26 and -0.55 , respectively, according to Table I).

A detailed polarization-sensitive magneto-PL spectra of the X^{+2} spectral lines are presented in figure 5. One can see that while the triplet lines shift towards higher energy with increasing B-field, the singlet lines shift towards lower energy. Since the initial states of the $X_{S_0}^{+2}$ and $X_{T_0}^{+2}$ transitions are the same (the bright-like exciton states), we conclude that the difference in the sign of the diamagnetic shift between the two transitions stems

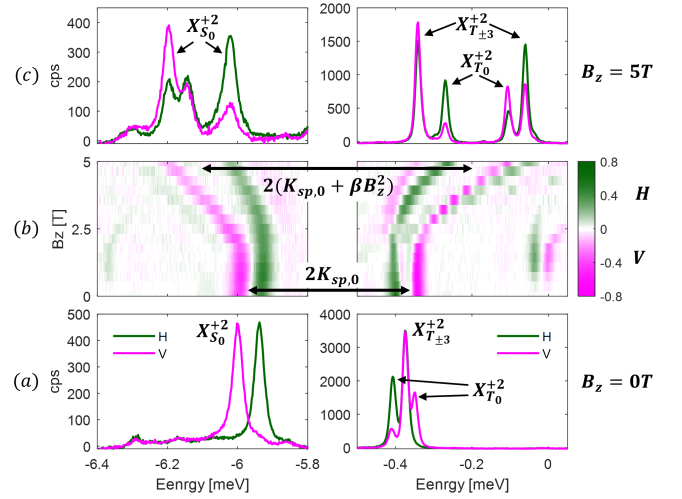


Figure 5. Rectilinear polarization-sensitive PL spectra of the X^{+2} spectral lines relative to the neutral exciton state a) at zero magnetic field, b) as function of the externally applied field in Faraday configuration, and c) in magnetic field of 5T. The transitions are marked by their final spin configurations ($S_0, T_0, T_{\pm 3}$). The energy difference between the $X_{T_0}^{+2}$ and the $X_{S_0}^{+2}$ doublets (marked) equals twice the hole-hole exchange interaction.

from the different influence that the external magnetic field has on the final states. The h-h singlet final state *rises* in energy faster than the initial state such that the overall spectral shift is negative. On the other hand, the h-h triplet state rises in energy slower than the initial state, and thus the total spectral shift is positive.

E. Magnetic field dependence of the exchange integral

We now use a simple harmonic oscillator model to quantitatively describe how the hole-hole exchange interaction is affected by the magnetic field. The exchange integrals between various states confined by a 2D harmonic potential with circular symmetry were calculated in Ref [48]. For two identical particles, one in a s-shell and one in a p-shell the exchange energy is:

$$K_{sp,0} = \frac{1}{4} \sqrt{\frac{\pi}{2}} \frac{e^2}{4\pi\epsilon_0\epsilon_r} \frac{1}{l_0} \quad (7)$$

where e is the electron charge, ϵ_0 is the vacuum permittivity and ϵ_r is the relative permittivity of the QD material. The effective length l_0 characterizes the extent of the harmonic potential and is equal to:

$$l_0 = \sqrt{\frac{\hbar}{m\omega_0}} \quad (8)$$

where m is the in-plane effective mass of the charge carriers, in our case holes, and ω_0 is the harmonic frequency of

the confining potential. To include the effect of the magnetic field, we replace ω_0 with $\omega \equiv \sqrt{\omega_0^2 + \frac{e^2 B^2}{4m^2}}$, obtained by adding a magnetic field hamiltonian to the harmonic oscillator one and solving for the eigenenergies (harmonic spectrum + Landau levels spectrum). The expression for the effective length then becomes:

$$l = \frac{l_0}{\left[1 + \left(\frac{eB}{2m\omega_0}\right)^2\right]^{1/4}} \quad (9)$$

Inserting Eq. 9 into Eq. 7 yields an expression for the field dependence of the exchange energy:

$$K_{sp}(B) = K_{sp,0} \left[1 + \left(\frac{eB}{2m\omega_0}\right)^2\right]^{1/4} \quad (10)$$

Furthermore, $m\omega_0$ can be expressed in terms of $K_{sp,0}$ by using Eq. 7 and 8:

$$m\omega_0 = \frac{512}{e^4} \pi \hbar \epsilon_0^2 \epsilon_r^2 (K_{sp,0})^2 \quad (11)$$

Inserting this expression into Eq. 10, we obtain an expression for the field dependence of the hole-hole exchange energy

$$K_{sp}(B) = K_{sp,0} \left[1 + \left(\frac{e^5 B}{1024 \pi \hbar \epsilon_0^2 \epsilon_r^2 (K_{sp,0})^2}\right)^2\right]^{1/4} \quad (12)$$

When the magnetic energy is much smaller than the zero-field exchange energy, we can expand this expression to the first non-vanishing order in B:

$$K_{sp}(B) \approx K_{sp,0} + \beta B^2 \quad (13)$$

where:

$$\beta_{(theory)} = \frac{e^{10}}{2^{22} \pi^2 \hbar^2 \epsilon_0^4 \epsilon_r^4 (K_{sp,0})^3} \quad (14)$$

To compare this result to the measured diamagnetic shift coefficients, we further extract $K_{sp,0}$ and ϵ_r from our measurements: $K_{sp,0}$ equals half of the energy separation between the $X_{S_0}^{+2}$ and $X_{T_0}^{+2}$ optical transitions at $B = 0$ (see Figure 5). From the magneto-PL in Figure 5, we obtain $K_{sp,0} = 2.79(1) meV$. To estimate ϵ_r , we average its value over the InAs and GaAs constituents of the QD, $\epsilon_r^x = x\epsilon_r^{(In)} + (1-x)\epsilon_r^{(Ga)}$, like we did for the Δ and E_p parameters in Section III B. Using $x \approx 0.75$, as we found by fitting Roth formula to the measured g -factor, we obtain $\epsilon_r^{0.75} \approx 14.25$. Combining those values, we conclude $\beta_{theory} \approx 6.6 \frac{\mu eV}{T^2}$.

In the experiment, β is directly measured as half the difference between the diamagnetic shifts of the $X_{T_0}^{+2}$ and $X_{S_0}^{+2}$ spectral lines. To see this, note that the initial states of $X_{T_0}^{+2}$ and $X_{S_0}^{+2}$ transitions are the same and thus cancel

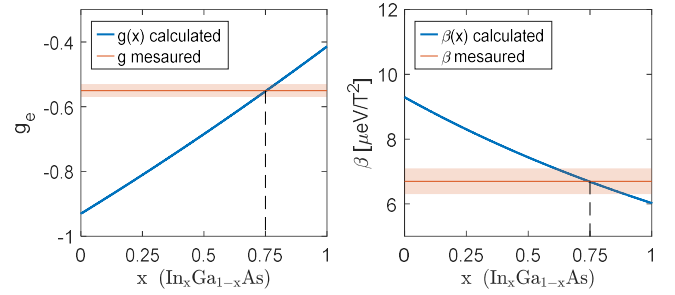


Figure 6. The calculated electronic g -factor $g_e(x)$ and the calculated relative diamagnetic shift $\beta(x)$ between the X^{+2} singlet and triplet transitions as function of the composition ratio x . Note that the calculation of the g -factor uses constant band gap energy as measured in the experiment, and thus at $x = 0, 1$ it does not reproduce the values for pure InAs and GaAs bulk materials.

out upon subtraction. The only contribution, then, is the final hole-hole state which is either a singlet or a triplet. We observe:

$$\begin{aligned} \beta_{measured} &= \frac{\alpha_{X_{T_0}^{+2}} - \alpha_{X_{S_0}^{+2}}}{2} \\ &= \frac{7.6 \frac{\mu eV}{T^2} - (-5.8 \frac{\mu eV}{T^2})}{2} = 6.7(4) \frac{\mu eV}{T^2} \end{aligned} \quad (15)$$

The calculated and the measured values of β agree up to $0.1 \frac{\mu eV}{T^2}$, well within our experimental error. We see this compliance as a strong validation of the hole-hole exchange interaction model. For further conviction, we test the sensitivity of our result to the value of x . This is shown in Figure 6. One can see that the dependencies $g(x)$ and $\beta(x)$ are close to linear, and that a deviation of x by more than 0.02 would cause those parameters to miss the measured values.

F. Diamagnetic shifts of the singlet and triplet lines

After explaining the *relative* diamagnetic shift between the $X_{S_0}^{+2}$ and $X_{T_0}^{+2}$ transitions, we proceed by calculating the absolute values of those shifts. We find that we can figure those values using the measured emission energy of the X^{+2} spectral lines relative to the neutral exciton X^0 , and its diamagnetic shift coefficient. In those calculations, we use the Hartree-Fock approximation to evaluate the energies of the optical transitions' initial and final many-body states. The energy of a many-body state is calculated, within this approximation, by summing over its individual-particle confining energies and the interactions between all the particle pairs involved.

Under this approximation, the energy of the ground state neutral exciton is:

$$E_{X^0} = E_s^e + E_s^h - J_{ss}^{eh} \quad (16)$$

where $E_s^{e(h)} = \frac{1}{2}\hbar\omega_{e(h)}$ is the energy of the electron (hole) in the s -level and J_{ss}^{eh} is the direct Coulomb interaction between the electron and the hole. This expression describes the exciton center-of-splitting (including at $B \neq 0$) as it does not include the e-h exchange Coulomb interaction. In the following derivation, we use $J_{n_1 n_2}^{p_1 p_2}$ ($K_{n_1 n_2}^{p_1 p_2}$) to describe the direct (exchange) Coulomb interaction between orbitals n_1 and n_2 of the charge carriers p_1 and p_2 , the latter representing either holes (h) or electrons (e).

The emission energies of the $X_{S_0}^{+2}$ and $X_{T_0}^{+2}$ optical transitions are given by the difference between the energies of the final and initial states (see figure 4).

$$\begin{aligned} E_{X_{T_0(S_0)}^{+2}}^{initial} &= E_s^e + 2E_s^h + E_p^h + J_{ss}^{hh} + 2J_{sp}^{hh} - 2J_{ss}^{eh} - J_{sp}^{eh} \\ E_{X_{T_0(S_0)}^{+2}}^{final} &= E_s^h + E_p^h + J_{sp}^{hh} \mp K_{sp}^{hh} \end{aligned} \quad (17)$$

Note that the hole singlet $E_{X_{S_0}^{+2}}^{final}$ is higher in energy than the triplet $E_{X_{T_0}^{+2}}^{final}$ ($K_{sp}^{hh} > 0$) due to the different symmetries of the associated spatial wavefunctions upon particle exchange. Thus, for the optical transition we have:

$$\begin{aligned} E_{X_{T_0(S_0)}^{+2}} &= E_{X_{T_0(S_0)}^{+2}}^{final} - E_{X_{T_0(S_0)}^{+2}}^{initial} \\ &= E_s^e + E_s^h + J_{ss}^{hh} + J_{sp}^{hh} - 2J_{ss}^{eh} - J_{sp}^{eh} \pm K_{sp}^{hh} \end{aligned} \quad (18)$$

We can cast this in terms of the Hartree-Fock neutral exciton transition energy, $E_{X^0} = E_s^e + E_s^h - J_{ss}^{eh}$, as:

$$E_{X_{T_0(S_0)}^{+2}} = E_{X^0} + J_{ss}^{hh} + J_{sp}^{hh} - J_{ss}^{eh} - J_{sp}^{eh} \pm K_{sp}^{hh} \quad (19)$$

All the elements in this equation can be expressed by the effective length of the electron ($l_e = \sqrt{\frac{\hbar}{m_e\omega_e}}$) and hole ($l_h = \sqrt{\frac{\hbar}{m_h\omega_h}}$), using Ref [48]. Summing them up gives the following expression:

$$E_{X_{T_0(S_0)}^{+2}} = E_{X^0} + (a \pm 1) K_{sp}^{hh} \quad (20)$$

where we defined the constant a as

$$a \equiv 7 - 4 \left(\frac{2}{1 + \gamma^2} \right)^{1/2} - (1 + 2\gamma^2) \left(\frac{2}{1 + \gamma^2} \right)^{3/2} \quad (21)$$

and $\gamma \equiv \frac{l_e}{l_h}$ is the ratio between the effective lengths. As K_{sp}^{hh} is the exchange energy between two holes, we have already found in the previous section that it can be expressed as $K_{sp}^{hh}(B) \approx K_{sp,0} + \beta B^2$. Using this, Eq. 20 becomes:

$$E_{X_{T_0(S_0)}^{+2}}(B) = E_{X^0}(B) + (a \pm 1) K_{sp,0} + \beta(a \pm 1) B^2 \quad (22)$$

The ratio γ can not be determined directly, as we lack the knowledge about the ratio between the effective in-plane masses of the electron and hole, or the ratio between their related harmonic confinement frequencies ω_e

and ω_h . However, we can extract γ from a , since it is a function of directly measured quantities. At zero magnetic field:

$$\begin{aligned} E_{X_{T_0}^{+2}} &= E_{X^0} + (a + 1) K_{sp,0} \Rightarrow a = \frac{E_{X_{T_0}^{+2}} - E_{X^0}}{K_{sp,0}} - 1 \\ E_{X_{S_0}^{+2}} &= E_{X^0} + (a - 1) K_{sp,0} \Rightarrow a = \frac{E_{X_{S_0}^{+2}} - E_{X^0}}{K_{sp,0}} + 1 \end{aligned} \quad (23)$$

$E_{X_{T_0}^{+2}} - E_{X^0}$ and $E_{X_{S_0}^{+2}} - E_{X^0}$ are the energies of the X^{+2} transitions relative to the neutral exciton, which at zero magnetic field, according to figure 5, equal respectively $-0.38(1)meV$ and $-5.95(1)meV$. Using these values and the previously obtained $K_{sp,0}$, both expressions in 23 yield $a \approx -1.13$, which in turn implies $\gamma \approx 0.17$.

Finally, we are ready to calculate the absolute diamagnetic shifts of the hole-hole triplet and singlet lines. According to Eq. 22, the diamagnetic coefficients of the X^{+2} transitions are:

$$\alpha_{X_{T_0(S_0)}^{+2}} = \alpha_{X^0} + \beta(a \pm 1) \quad (24)$$

where $\alpha_{X^0} = 8.4(2) \frac{\mu eV}{T^2}$ is the diamagnetic shift coefficient of the exciton, and $\beta = 6.7(4) \frac{\mu eV}{T^2}$ is the relative X^{+2} diamagnetic shift calculated in the previous section. We find:

$$\begin{aligned} \alpha_{X_{T_0}^{+2}} &= \alpha_{X^0} - 0.13\beta = 7.5(2) \frac{\mu eV}{T^2} \\ \alpha_{X_{S_0}^{+2}} &= \alpha_{X^0} - 2.13\beta = -5.9(8) \frac{\mu eV}{T^2} \end{aligned} \quad (25)$$

which agree with our measured values $7.6(4) \frac{\mu eV}{T^2}$ and $-5.8(7) \frac{\mu eV}{T^2}$, respectively.

IV. SUMMARY

We performed magneto - PL spectroscopy on a well-characterized $\text{In}_x\text{Ga}_{1-x}\text{As}/\text{GaAs}$ QD in Faraday configuration. From the measurements we extracted the g -factors and the diamagnetic shifts of many excitonic transitions. In particular, we observed an anomalous negative diamagnetic shift of spectral lines resulting from the radiative recombination of a doubly charged exciton ($X_{S_0}^{+2}$). Our results are explained using simple models for the Zeeman interaction and for the measured diamagnetic shifts. For both interactions we use one free parameter: x , the effective relative Indium content of the ternary QD. We use this parameter to linearly interpolate the QD electronic g -factor and permittivity, from those of its binary components GaAs and InAs.

By analysis of the measured g -factors of various optical transitions we show that while the g -factors of the electron in the first and second level have the same sign, the g -factors of the hole in these levels are opposite in sign.

We explain the difference between the diamagnetic shifts of the optical transitions of the doubly positively charged exciton which result in the remaining holes in a singlet ($X_{S_0}^{+2}$) and that in which they form a triplet ($X_{T_0}^{+2}$) using a simple circular harmonic potential model. The model describes in analytical form the hole-hole exchange interaction including the influence of the externally applied magnetic field. Finally, using the Hartree-Fock approximation we calculate the *absolute* diamagnetic shifts of these spectral lines using the measured diamagnetic

shift of the neutral exciton (X^0).

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