Theory of magneto-optical properties of neutral and charged excitons in GaAs/AlGaAs quantum dots

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Detailed theoretical study of the magneto-optical properties of weakly confining GaAs/AlGaAs quantum dots is provided. We focus on the diamagnetic coefficient and the \( g \)-factor of the neutral and the charged excitonic states, respectively, and their evolution with various dot sizes for the magnetic fields applied along [001] direction. For the calculations we utilize the combination of \( \mathbf{k} \cdot \mathbf{p} \) and the configuration interaction methods. We decompose the theory into four levels of precision, i.e., (i) single-particle electron and hole states, (ii) single-particle excitons, (iii) multi-particle excitons without correlation, and (iv) that including the effect of correlation. The aforementioned approach allows us to pinpoint the dominant influence of various single- and multi-particle effects on the studied magneto-optical properties, allowing the characterization of experiments using models which are as simple as possible, yet retaining the detailed physical picture.

I. INTRODUCTION

Semiconductor III-V quantum dots (QDs) have been extensively studied in the past, owing to their properties stemming from the zero-dimensional nature of the quantum confinement. Those are, e.g., an almost \( \delta \)-function-like emission spectra, which lead to a number of appealing applications in semiconductor opto-electronics. Hence, such QDs are crucial for classical telecommunication devices as low threshold/high bandwidth semiconductor lasers and amplifiers\textsuperscript{13,15}, as sources of single and entangled photon pairs that might be used for the quantum communication\textsuperscript{16–17} or other quantum information technologies\textsuperscript{18–27}. However, the aforementioned applications are mostly based on the In(Ga)As QDs embedded in GaAs matrix. In that material system, the QDs are compressively strained due to the lattice mismatch between InAs and GaAs of \( \sim 7\%\textsuperscript{22} \). That in conjunction with the lack of inversion symmetry in the zincblende semiconductors, leads to considerable shear strain in and around the dots, causing among others the non-negligible fine-structure-splitting (FSS) of the QD ground-state exciton doublet\textsuperscript{14,17}. As a result, the emitted photons are distinguishable, hampering their use, e.g., as sources of single entangled states for quantum key distribution (QKD) protocols\textsuperscript{20,22,23}.

To overcome that drawback, recently GaAs QDs embedded in Al\textsubscript{0.4}Ga\textsubscript{0.6}As matrix were fabricated by the droplet-etching method\textsuperscript{31,33}. Since the lattice mismatch in that material system is only \( \sim 0.06\% \), they show very small FSS as was recently confirmed in Refs.\textsuperscript{34,35}. Hence, because of their favorable properties, the GaAs/Al\textsubscript{0.4}Ga\textsubscript{0.6}As QDs emerged as a promising source of non-classical states of light, such as single photons with a strongly suppressed multi-photon emission probability\textsuperscript{36}, highly indistinguishable photon states\textsuperscript{37,39}, and single polarization entangled photon-pairs with an almost near unity degree of entanglement\textsuperscript{37–41}.

Because of their well defined shape and size, the almost negligible built-in strain and alloy disorder, GaAs/Al\textsubscript{0.4}Ga\textsubscript{0.6}As QDs are excellent system for testing the current quantum mechanical theory of nanostructures. A preferred way of doing so is the comparison between the experimentally measured and theoretically predicted properties (emission energy, oscillator strength, polarization) for QDs under externally applied perturbations. Those might be strain, electric, or magnetic fields and we have recently shown\textsuperscript{42} the inadequacy of the single-particle model\textsuperscript{43–45} for the description of the latter. Further studies\textsuperscript{46,47} recently demonstrated the QD size dependence of the applied magnetic field response for excitons and charged trions. However, the detailed theoretical description of that is still missing and we fill that gap in this paper.

The paper is organized as follows: We start with the description of the theory model in section II, continue with discussion of the theory of the response of GaAs/AlGaAs QDs to externally applied magnetic field with particular emphasis on the diamagnetic shift and \( g \)-factor, and in section III we further compare our results with available experiments. Thereafter in section IV we focus on the magnetic field response of charged trions and we conclude in section V.

II. THEORY MODEL AND STUDIED QUANTUM DOT

We theoretically study the excitonic structure of the GaAs/Al\textsubscript{0.4}Ga\textsubscript{0.6}As QDs using the following methodology. It starts with the implementation of the 3D QD model structure (size, shape, chemical composition), see Fig.\textsuperscript{1}(a), and carries on with the calculation of the strain and the piezoelectricity. The resulting strain and polarization fields then enter the eight-band \( \mathbf{k} \cdot \mathbf{p} \) Hamiltonian\textsuperscript{48}. Thereafter, for the QD with applied magnetic field, the Hamiltonian introduced in Ref.\textsuperscript{49} with added
Pauli term describing the interaction between the magnetic field and the spin is solved using the Nextnano suite, yielding the electron and hole single-particle (SP) states. For the full list of material parameters used in this work see Ref. 52 (see, also, references 49, 53–57 therein). The Coulomb interaction between the quasiparticles and the correlation is accounted for by employing the configuration interaction (CI) method. Using the theory toolbox described thus far, we obtain the eigenenergies and eigenfunctions of various complexes like the neutral (X^0), the positively (X^+) and the negatively (X^-) charged exciton, see Fig. 1(b). See also Appendix I and Appendix II for details about the CI computation method and the evaluation of the corresponding results. Note, that since our theoretical description is supposed to be applicable for explanation of experiments, we simulate the structure for finite temperature of T = 8 K.

The magnetic flux density (B) induces circulating current which leads to the magnetic momentum (μ) opposed to B. The interaction between B and μ causes, among others, the energy shift (∆E) of the spin degenerate state. In the first approximation one obtains

$$\Delta E = \gamma B^2,$$

where γ is the diamagnetic coefficient which is proportional to the spatial expansion of the wave function in the direction perpendicular to B. Hence, γ for a carrier in a semiconductor satisfies

$$\gamma \propto \langle r^2 \rangle / m^*,$$

where $\langle r^2 \rangle$ is the average expansion of the wave function in the direction perpendicular to B and $m^*$ is the effective mass of the charged carrier. Thus, e.g., by inspection of Fig. 1(b) one can anticipate larger γ for X^+ and X^- states compared to those of X^0.

The other dominant process which is observed is the Zeeman effect, which is due to the interaction of B with the projection of the spin momentum (S) to the direction parallel to B. In the case of B applied in the direction of QD growth (B_z), spin degeneracy of states is lifted. However, B applied in the plane of QD (B_x) also breaks the symmetry of the system and, thus, the coupling between different states is involved in that case, like, e.g., that between the dark with total angular momentum |±2⟩ and the bright with total angular momentum |±1⟩ states of X^0, respectively. The splitting of the energy levels

FIG. 1. (a) Side view of computed GaAs QD (black object) embedded in Al_{0.4}Ga_{0.6}As matrix. Horizontal and vertical lines mark the grid of the simulation space and the scale for (x) and (z) axis is given in units of nm; h and d label the height and the base diameter of QD, respectively. The shape of QD was inspired by Ref. 44, where it was chosen to fit the AFM measurements and optimized to match the emission energy and magneto-optical properties of X^0. (b) The isosurfaces of 90% of total probability density of X^0, X^-, and X^+ ground states for B_z = 0 T, respectively. The upper row in (b) shows the side, while the bottom row the top view of the density, respectively. The insets in (b) indicate the crystallographic orientation of the cuts. Notice the larger volume of the probability densities for X^- and X^+ in comparison with that for X^0.
depends on $B$ linearly in the first approximation, and the slope of that is commonly called the $g$-factor. We note, that in the following text we focus only on $B$ applied in the growth direction, i.e., we study the response to $B_z$.

The aforementioned effects are observable for a variety of quasiparticles like the holes, the excitons, or other complexes. To extract $\gamma$ and $g$-factor of computed (multi-)particle complexes we use the following model which is suitable also for evaluation of the experiments:[42]

$$E_{\uparrow/\downarrow} = E_0 + \gamma B^2 \pm \frac{1}{2} \sqrt{E_{\text{FSS}}^2 + g_0^2 \mu_B^2 B^2},$$

(3)

where $\uparrow / \downarrow$ labels the spin of the energy levels, $E_0$ and $\Delta E_{\text{FSS}}$ are the emission and FSS energies, respectively, of the corresponding state for $B = 0$ T; $g_0$ denotes the $g$-factor, and $\mu_B$ is the Bohr magneton. Note, that since the splitting is strongly linear in our calculations, we take into account only the zeroth term $g_0$ of $g$-factor and neglect the second order perturbation term $g_2$ introduced in Ref.[50].

### III. SIZE DEPENDENCE OF MAGNETO-OPTICAL PROPERTIES OF NEUTRAL EXCITON

In this section we study the size dependencies of the magneto-optical properties of $X^0$ ground state of the QDs shown in Fig. [1]. We mark the height and the diameter of QD base $h$ and $d$, respectively, see Fig. [1] (a), and we track $\gamma$, $g_0$, and the light-hole (LH) content for (i) QD with fixed $h$ and varying $d$, (ii) fixed $d$ and varying $h$, and (iii) for fixed $h/d$ ratio, thus, we track the variation of QD volume ($V$) in the latter case. The dependencies of $\gamma$ and $g$-factor are shown in Fig. [2]. We plot the results of CI calculation for the CI basis of 12 SP electron and 12 SP hole states marked as $12 \times 12$ (blue) and for the basis of 2 SP electron and 2 SP hole states marked as $2 \times 2$ (green). The comparison between those bases allows us to study the effects of correlation. To see the effect of the Coulomb and the exchange interaction, the dependencies of SP excitonic (red), electron (pink) and hole (orange) states are included in Fig. [2] as well.

The emission energy $\langle E_{X^0} \rangle$ of $X^0$ calculated in the $12 \times 12$ basis changes for all kinds of the studied size variations in the range of $\approx 1$ eV. To be able to quantitatively compare the computed dependencies, we define the parameter $b$ as

$$b = \frac{f(E_{X^0}^f) - f(E_{X^0}^i)}{E_{X^0}^f - E_{X^0}^i},$$

(4)

where $f(E_{X^0}^f)$ ($f(E_{X^0}^i)$) is the studied quantity (e.g. magneto-optical properties) for the final (initial) value of the size dependence of $E_{X^0}$. Note, that since the studied dependencies are not always linear and $E_{X^0}$ is not the quantity describing SP hole and electron states, $b$ provides only an estimation of the slope of the corresponding dependency.

### A. Diamagnetic coefficients

First, we study the size dependence of $\gamma$. From Eq. (3) we expect the sensitivity of $\gamma$ to variation of $d$. This is confirmed by the numerical calculation in Fig. [2] (a). We see that the absolute value of $\gamma$ for electrons ($\gamma_e$) is much larger than that for holes ($|\gamma_h|$). Since electrons have smaller effective mass, their states are much more sensitive to the change of QD shape or size than considerably heavier holes, which have in our calculation predominantly heavy-hole character. It follows, that $\gamma_e$ ($b = 117$) is more sensitive to variation of QD size and also its magnitude is larger than that of $|\gamma_h|$ ($b = 18$).

On the other hand, the lateral confinement does not change in the case of the variation of $h$ for fixed $d$, see Fig. [2] (b). However, despite that, $\gamma$ grows slightly. First, we describe the height dependence for electrons and note that the value of $\gamma$ also depends on the effective mass, cf. Eq. (2). It was shown previously for InAs/GaAs QDs that the effective mass of electron and hole depends on QD height and base diameter.[52] The effective mass of the electron decreases with increasing $h$, what also explains the increase of $\gamma_e$ ($b = 34$) in our calculations. The case of the height dependence of $|\gamma_h|$ ($b = 7$) is, however, more complex. The effective mass of heavy-holes (HH) grows with increasing $h$.[60] On the other hand, increasing $h$ leads to admixture of [LH] states due to larger amount of $p_{z\gamma}$ Bloch waves, the contribution of which increases for higher QDs which might even consist of purely [LH] states.[53] Moreover, $|\gamma_h|$ varies very slowly, hence, we might assume that the two aforementioned effects nearly cancel each other.

Lastly, we fix the aspect ratio of QD and change both $d$ and $h$, $h/d = 3/13$. We observe the steepest change of $\gamma_e$ ($b = 176$). Here, both the reduction of the electron effective mass and the reduction of lateral confinement contribute to the increase of $\gamma_e$. In the case of $|\gamma_h|$ ($b = 10$) we can see combination of two opposing trends, discussed before. The reduction of lateral confinement caused by increasing $d$ leads to the increase of $|\gamma_h|$ while larger $h$ slightly reduces that. This results in a slower growing trend of $|\gamma_h|$ as we can see in Fig. [2] (c).

Using the SP approach we can write that $\gamma$ of $X^0$ is[50]

$$\gamma_{\text{SP}} = |\gamma_e| + |\gamma_h|.$$

(5)

The parameter $\gamma_{\text{SP}}$ is mostly influenced by the electronic part of $e^- - h^+$ pair SP transition which we mark as $X^0$, since we omit the effect of the Coulomb interaction. As we can see from Fig. [2] the presence of the direct and the exchange Coulomb interaction slightly reduces the values of $\gamma_{\text{CI}}$ computed by CI. The effect of correlations influences the excitonic $\gamma_{\text{CI}}$ rather weakly. In Figs. [2] (a) and (c) we observe that the deviation between $\gamma_{\text{SP}}$ and $\gamma_{\text{CI}}$ increases with growing size of QD. This means that the SP approximation describes rather well the response of small QDs with larger quantum confinement but fails for larger, weakly confined QDs, for
which it is necessary to include also the multiparticle corrections.

B. g-factors

We divide this section into three parts. Firstly, we discuss the electronic g-factor ($g_e$), we follow by the hole g-factor ($g_h$), and finally, the excitonic g-factor, computed both using SP approach ($g_{SP}$) and CI ($g_{CI}$), respectively, is considered.

The value of $g_e$ is +2 in the limit of infinite confinement, which is the result of the quenching of the angular momentum \[\hbar^2 J_0^2 / \hbar^2 L_0^2]^{1/2}. The observed size dependence of $g_e$ is caused by the admixture of Bloch states from the valence bands (VB), i.e. heavy-hole ($|\text{HH}\rangle$), light-hole ($|\text{LH}\rangle$), and split-off ($|\text{SO}\rangle$) Bloch states, into the ground state of electron and we proceed by discussing the reasons for that (see also Ref.59). In the eight-band $k \cdot p$ calculations we express each quantum state as a superposition of $|S\rangle$, $|\text{HH}\rangle$, $|\text{LH}\rangle$, and $|\text{SO}\rangle$ bulk Bloch components. The wave functions consist of the Bloch and the envelope parts with (total) angular orbital momenta $J$ and $L_E$, respectively, which are coupled due to the spin-orbit interaction. Since the $|S\rangle$ Bloch component has $L_E = 0$, it does not influence $g_e$ at all. However, the Bloch func-
show in Fig. 3 the size dependencies of $|\text{HH}\rangle$, $|\text{LH}\rangle$, and $|\text{SO}\rangle$ components (left column). In all cases of the studied size variations, the content of states from VB decreases with increasing size by $\sim 2\%$. The sum of all $L_{E_z}=\pm 1$ components decreases with increasing $d$ (Fig. 3 (a)) by $\sim 10$-times smaller rate compared to that for InAs/InP QDs. Interestingly, in the case of the variation of $h$ (Fig. 3 (b)) we observe even $\sim 24$-times reduced rate. Since the admixture of the VB components, which affect $g_e$, depends on the structural properties of QDs rather weakly we do not observe strong size dependence of $g_e$. For the completeness we also show the size dependence for the studied parameters in the case of fixed aspect ratio $\{\text{Fig. 3 (c)}\}$, even though we cannot compare to any similar study for InAs/InP QDs.

In the case of $g_h$ we observe a rather strong size dependence of that. Since in the case of holes, the analysis of $L_E$ is more complicated, we do not apply the approach which we discussed above for electrons. However, there exists a connection between the value of $g_h$ and HH-LH mixing in the hole ground state. The approach introduced in Ref. [3] utilizes 2D effective model and can be expressed as

$$g_h = 6\kappa + \frac{27}{2} q - 2\gamma_{lh},$$

where $\kappa$ and $q$ are the Luttinger parameters in the standard notation [39] which are for GaAs summarized in Table [1] and $\gamma_{lh}$ is given by the overlap of $|\text{HH}\rangle$ and $|\text{LH}\rangle$ states of the hole. It follows from the Eq. (6) that $g_h$ strongly depends on the bulk properties of the QD material and, furthermore, that the content of (LH) states in the hole reduces the magnitude of $g_h$, since $\gamma_{lh}$ attains positive values. The validity condition for the discussed 2D model is that $h/d \ll 1$, which is fulfilled for our QDs. Furthermore, it is assumed in the model that the mixing of (SO) states can be neglected since $E \ll \Delta_{\text{HH-LH}} \ll \Delta_{\text{SO}},$
\(|p_x\rangle\) and \(|p_y\rangle\) Bloch waves, which have predominantly HH character, increases as well. Consequently, the reduction of the amount of \(|LH\rangle\) is observed.

Interestingly, even though the content of \(|LH\rangle\) Bloch waves decreases with growing \(d\) (see Fig. 4 (a)), we observe a slow decrease of \(g_h\) as well. Similar trend was previously observed for InAs pyramidal QDs in Ref.[99]. We assume that also other effect causes the reduction of \(g_h\), apart from those previously discussed. Since the decrease of \(g_h\) is the weakest of all the discussed cases, we assume that the effect which reduces \(g_h\) is similarly strong, when \(d\) grows, as the decrease of the content of \(|LH\rangle\) components. The parameter \(g_h\) also depends on the bulk material parameters, see Eq. [6]. As the lateral quantum confinement becomes weaker with increasing \(d\), the hole wave function moves towards the top of the QD. If hole wave function would partially leak out of the QD material, the change in \(g_h\) might have been affected also by the properties of the surrounding Al\(_{0.4}\)Ga\(_{0.6}\)As. In Table 1 we summarize the values of \(\kappa\) for GaAs and AlAs. Using the linear interpolation we can estimate \(\kappa\) of Al\(_{0.4}\)Ga\(_{0.6}\)As as \(\kappa = 1.08\). While the smaller value of \(\kappa\) would lead to the reduction of \(g_h\), an inspection of the probability density of hole ground state shows leakage of hole out of QD only for QDs with \(d > 30\) nm. Since, we observe the reduction of \(g_h\) also for smaller \(d\) we conclude that this effect is not strong enough to cause the decreasing trend of \(g_h\).

We observe strong dependence of \(g_h\) on \(V\) of QD for fixed QD aspect ratio. That effect is caused by the admixture of \(|LH\rangle\) Bloch waves, see Fig. 4 (c), similarly as in the previous case. Also \(E_X\) for which \(g_h\) crosses zero is similar for the cases of fixed aspect ratio and fixed \(h\).

We further fitted the \(B_z\) dependence of the difference of non-interacting SP neutral exciton (\(X^0\)) by Eq. [2] and obtained the slope of that which we mark as \(g_{SP}\). From the SP approach we find that \(g_{SP} = g_e + g_h\) of the bright state, similarly as in Refs. [98,49]. Resulting from that and already discussed properties of \(g_h\) and \(g_e\), it follows that the trend of \(g_{SP}\) is dominated by the hole part of \(X^0\). Since the value of \(g_e\) remains nearly constant with QD size change, it only causes the increase of the mean value of \(g\)-factor, leading to the zero-crossing of \(g_{SP}\) for smaller emission energies. To see how the HH-LH coupling affects \(g_{SP}\), we show in the last column of Fig. 2 the content of \(|LH\rangle\) Bloch wave in the exciton. The calculations were done for \(X^0\) and for \(X^0\) computed by CI with basis of \(2 \times 2\) and \(12 \times 12\) electron and hole SP states, respectively (see also Appendix II). Clearly, the HH-LH coupling is not influenced by the effect of correlation, however, it is weakly affected by the direct and the exchange Coulomb interactions. The connection between band mixing and \(g_{SP}\) is the same as that already discussed for holes.

The inclusion of the direct and the exchange Coulomb interaction \((2 \times 2\) basis\) causes the overall increase of the \(g\)-factor. Note, that the difference between \(g_{SP}\) and \(g_{CI}\) computed in \(2 \times 2\) basis grows with the size of QD. As a result of the exchange and the direct Coulomb interaction, \(g_{CI}\) does not cross zero for the range of considered sizes.

On the other hand, the effect of correlation causes the reduction of \(g_{CI}\). Hence, there are multi-particle effects.

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**TABLE I. Luttinger parameters for GaAs and Al\(_{0.4}\)Ga\(_{0.6}\)As**

|       |  \(\gamma_1\) |  \(\gamma_2\) |  \(\gamma_3\) | \(K\) | \(\kappa\) | \(g\) |
|-------|----------------|----------------|----------------|------|-------------|------|
| GaAs  |  6.98          |  2.06          |  2.93          |  1.2 |  0.04       |      |
| AlAs  |  3.76          |  0.82          |  1.42          |  0.12|  0.03       |      |
which affect the Zeeman splitting and $g$-factor in opposite ways. The exchange Coulomb interaction amplifies the Zeeman splitting, increasing $g_{CI}$, yet the correlations cause the reduction of that. The effect of multi-particle effects on the Zeeman splitting is sketched in Fig. 6 (d). To view the size dependence of the exchange interaction, we plot the direct consequence of that, $E_{FSS}$ for $12 \times 12$ basis, in Fig. 6 (a)–(c). Note, that $g_{CI}$ calculated in $12 \times 12$ basis shows similar trends as that for the exchange interaction.

Note, that for increasing $h$ of QD the difference between CI calculation without and with the effect of correlation grows. Surprisingly, for the calculations where only $d$ is varied, the deviation is not systematic, thus, it seems that the lateral size of QD does not influence the effect of correlation on $g_{CI}$. This observation is unexpected since the trend of increasing difference between calculations with and without the effect of correlation is stronger for varying $V$ with fixed aspect, what is the consequence of the faster decrease of the quantum confinement due to increasing of both $h$ and $d$.

To further visualize the importance of the correlation for the spin-resolved description of $X^0$ we show in Fig. 5 the content of hole and electron SP states in those of CI for a prototypical pyramid-shaped InAs/GaAs QD with the base width of 20 nm and height of 3 nm and GaAs/Al$_0.4$Ga$_{0.6}$As dot, shown in Fig. 1 with the base diameter 68 nm and height 16 nm. Here, the level of darkness identifies the content of each SP state, i.e., the darker the larger content. On the vertical axis the number of the CI eigenstate is shown. The states are ordered by energy, e.g. 1,2 is the ground state doublet. The numbers on the horizontal axis represent the numbers of the SP electron (green) or hole (red) states computed by Nextnano software. Here, numbers 11, 12 \{13, 14\} mark hole \{electron\} ground states. In the former case (InAs/GaAs QD), the first four CI ground states of $X^0$ (CI states 1–4 in Fig. 5) clearly show the dominant contribution of particular hole or electron SP state to a given CI state. That is smeared out for GaAs/Al$_0.4$Ga$_{0.6}$As QD. In the latter case, the correlation causes via the exchange interaction the mixing of the almost energy degenerate hole SP states, competing, thus, with the Zeeman interaction, as sketched in Fig. 6 (d). The details about the

FIG. 5. The contribution of SP states, used as the basis of the CI calculations, in $X^0$ without the application of external fields for InAs/GaAs pyramidal-shaped QD with base width of 20 nm and height of 3 nm (left) and GaAs/AlGaAs QD shown in Fig. 1 with $d = 68$ nm and $h = 16$ nm (right). The CI calculations were performed with the basis of 12 SP electron (green) and 12 SP hole (red) states and were carried out exactly the in the same manner. Note that the CI states are ordered from 1 to 29 according to their energy from lowest to highest.

FIG. 6. Dependencies of $E_{FSS}$. The calculations are shown as a function of QD size for (a) fixed height of $h = 9$ nm, (b) fixed base diameter of $d = 40$ nm, and (c) fixed aspect ratio of $h/d = 3/13$, where $V$ is the volume of QD. (d) Sketch of the effect of the direct and the exchange Coulomb interactions, respectively, and the correlation on the energy splitting of levels due to $B_z$.

InAs/GaAs

GaAs/AlGaAs
evaluation of the contents of SP states in the CI states are discussed in Appendix II.

C. Comparison with the experimental data

![Graph showing dependencies of X^0 ground state magneto-optical properties on the emission energy E_{X^0}. The experimental measurements were reprinted from Ref. 17 (grey).]

FIG. 7. Dependencies of X^0 ground state magneto-optical properties on the emission energy E_{X^0} (blue). The experimental measurements were reprinted from Ref. 17 (grey).

Finally, we compare our calculations with the measurements performed by M. C. Löbl and colleagues, see Fig. 7. The samples were grown by MBE on the (001) substrate. The measured QDs were, as well as those discussed in the previous sections, cone shaped and had also the same composition, i.e. pure GaAs QDs embedded in Al_{0.4}Ga_{0.6}As. The authors further assumed that all the Al droplets which etched the substrate had the same aspect ratio. Since the relation between Al droplet height h_d and QD height h is given by the phenomenological relation h \propto h_d^{0.67}, we assume that the aspect ratio of measured QDs was the same for all QDs and, thus, we compare with that our calculations where the aspect ratio of QDs is fixed as well. However, we show in our comparison all the size dependencies in order to indicate whether the aforementioned assumption is correct. For further details about the growth and measurements we refer the reader to Ref. 17.

For the comparison we use the magneto-optical properties of multi-particle X^0 where the effect of correlation is included. As we can see, the calculated trends reasonably fit the measurements. However, we observe larger disagreement with the size dependence of the measured magneto-optical properties for larger emission energies E_{X^0}, i.e. smaller QDs. The deviation between theory and experiment might be, e.g., attributed to the fact that we did not optimize the QD shape to fit the measurements more precisely and, thus, the aspect ratio or shape of the base of the calculated QDs can be slightly different than that for experiment.

Let us first compare theoretically and experimentally obtained \gamma. As we can see, almost all the calculated dependencies have similar non-linear trends. They decrease fast for smaller E_{X^0} and from certain value the decreases is approximately linear. The calculated dependencies resemble a combination of two linear trends with different slopes or a single exponential decrease. The linear trend shows also \gamma determined from the experiment. Generally, by comparing the steepness of the experimentally determined data and calculations, we deduce that measured QDs were slightly larger than those calculated.

In the case of g-factors we observe larger differences between calculations and experiment. Here, the slopes of calculated dependencies are unfortunately significantly larger than that of the measured data for smaller values of E_{X^0}. However, we note that a more favorable correspondence of the theoretical slope with that of the experiment might be observed for variation of d with fixed h and at the same time smaller values of E_{X^0}. However, to match the experiment, one would clearly need also the dot to have larger V.

IV. SIZE DEPENDENCE OF MAGNETO-OPTICAL PROPERTIES OF TRION STATES IN MAGNETIC FIELD

In this section we expand our analysis of the size dependence of magneto-optical properties to incorporate also the positively charged ground X^+ and excited X^+_3 and X^+_4 trions, respectively, and that for excitons with surplus electron, i.e., X^-.

The calculations are performed for QD shown in Fig. 1 with fixed aspect ratio of h/d = 3/13, similarly as that in section III. The B is again applied in the [001] growth direction, i.e. B_z, and the energies of the excitonic complexes are calculated by CI with the basis of 12 SP electron and 12 SP hole states. Studied size dependencies of magneto-optical properties are shown in Fig. 5.

The particular choice of X^+_3 and X^+_4 is motivated by their experimental observation in Ref. 13 and the larger contribution in the respective CI complexes which we show along with the contribution of SP states in CI complexes of X^- in Appendix III.

As expected, \gamma of X^- and X^+ have very similar trend as that for X^0 (cf. Fig. 2 (c)). To see the differences among \gamma of the excitonic complexes we discuss again the parameter b defined in Eq. 4. Due to the dominant content of h^+, which has larger effective mass than e^-, we observe the smallest value of b for X^+ (b = 150). Interestingly, \gamma grows slightly more for X^0 (b = 168) than for X^- (b = 160). That results from previous investigation of size dependence of \gamma for X^0, since direct and exchange Coulomb interaction reduce the values of \gamma for larger QDs, what leads to the reduction of the parameter b. Moreover, in the case of X^- {X^+} the Coulomb interaction between the electron and the hole is doubled compared to X^0 and also that between electrons {holes} is included what also contributes to the reduction of b.

Due to the coupling of singlet (X^+_3) and triplet (X^+_3) states of the excited trions we observe the anomalous diamagnetic shift of X^+_3 transition. The parameter \gamma of X^+_3 is negative for all considered QDs and its absolute
value increases with size until it reaches the minimum for $V \approx 20 \cdot 10^3$ nm$^3$. On the other hand, $\gamma$ of $X_{4}^{+}$ grows monotonically with increasing $V$. Interestingly, we observe smaller absolute values of $\gamma(X_{3}^{+})$ and $\gamma(X_{4}^{+})$ for QDs with $h = 17$ nm and $d = 75$ nm (or $h = 18$ nm and $d = 78$ nm) considered in this section, than for QDs with $h = 15$ nm and $d = 70$ nm, see Tab. 2 in Ref. 44. We assume that this might be caused by different aspect ratio of QDs under consideration.

We now discuss the $g$-factors of $X^−$, $X^+$, $X_{3}^{+}$, and $X_{4}^{+}$. Since $g_0$ of trions is given by the SP state the energy of which is subtracted during the recombination, we observe significantly faster decrease for $X^+$ ($b = 19$) than for $X^−$ ($b = 9$). The size dependence of subtracted SP $e^−$ (pink curve) and $h^+$ (orange curve) states are shown in the second column in Fig. 8 (c). In the contrast with $X^0$, the $g$-factor of $X^+$ monotonically decreases towards negative values. The reason of smaller $g_0$ of $X^+$ is the larger content of $|\text{LH}\rangle$ Bloch states. At the same time, the $g$-factors of the excited trions increase with increasing volume and for $V = 11.7 \cdot 10^3$ nm$^3$ we observe a sudden decrease of those. In order to understand the aforementioned trends, we show in Fig. 9 the QD volume dependence of $g$-factors of non-recombined positive trion states, the final SP hole state, and the final emitted single photon state. The colours identify the non-recombined trion states and the markers the final SP hole states. As expected, we observe similar trends for final SP hole state and the emitted photons. On the other hand, the $g$-factors of non-recombined trion states have positive values and decrease in the whole considered range of QD volumes $V$.

![FIG. 8. Size dependence of (a) $\gamma$ and (b) $g$-factor of $X^-$ (light blue), $X^+$ (blue), $X_{4}^{+}$ (red), and $X_{3}^{+}$ (green), respectively.](image)

![FIG. 9. The QD volume dependence of the $g$-factor for (a) non-recombined positive trion states, (b) final SP hole states, and (c) emitted final single photon states. The colours identify the non-recombined trion states and the markers the final SP hole states.](image)
V. CONCLUSIONS

We have studied the size dependencies of the diamagnetic coefficient and $g$-factor of $X^0$, $X^-$, and $X^+$ ground states, and $X^+_3$ and $X^+_4$ excited positive trion states, respectively, of GaAs/AlGaAs cone shaped quantum dots. The magnetic field was applied in the [001] growth direction. The sizes of quantum dots were changed in three ways, i.e., for fixed height (and variable base diameter), for fixed base diameter (and variable height), and for fixed aspect ratio (and variable volume). To find the origin of the observed trends, we decompose the calculations into four levels of precision: dependencies for (i) single-particle electron and hole, (ii) single-particle exciton, (iii) multi-particle exciton without the effect of correlation and (iv) that with the inclusion of the effect of correlation. The calculated dependencies have reasonably well reproduced the experimental trends observed in Ref.\textsuperscript{22}

The diamagnetic coefficients of the correlated $X^0$ are found to be described sufficiently well by the single-particle approach for small dots. The increase of that with QD size is found to be mainly due to the single-particle electron states. The case of excitonic $g$-factor is more complex. Here, we find a decrease of that with size which is due to holes. However, the multi-particle effects for both, diamagnetic coefficient and $g$-factor, are found to be non-negligible in the case of large, weakly confined quantum dots. The exchange and direct Coulomb interactions increase the absolute value of the excitonic $g$-factor. At the same time, the effect of correlation decreases that. Furthermore, we find that also the slope of the dependencies is smaller when the exchange and direct Coulomb interactions are included.

We have also studied size dependencies of magneto-optical parameters for charged trions. Here, the dependencies of the diamagnetic shifts and $g$-factors of ground states of those complexes were found to have similar evolution with QD size as that for ground state $X^0$. Only for the latter ($g$-factor) we found the dependencies to be shifted in magnitude which we identified to be the result of the subtraction of the final single-particle state, electron for $X^-$ or hole for $X^+$. Strikingly, the excited positively charged exciton states, $X^+_3$ and $X^+_4$, show noticeably different behavior. Namely, the anomalous (enormous) diamagnetic shift in the case of the former (latter). Moreover, in both of the aforementioned complexes the $g$-factor has non-monotonic dependence with size. We interpret the former (diamagnetic shift) observation to be due to singlet-triplet mixing in large weakly confining GaAs dots. On the other hand, the latter phenomenon ($g$-factor) is again explained as a result of the subtraction of the final single-particle hole state.

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APPENDIX I.

Here we give the description of the CI method. Let us consider the exciton complex |M⟩ consisting of \( N_c \) electrons and \( N_h \) holes. The CI method uses as a basis the Slater determinants (SDs) consisting of \( n_e \) SP electron and \( n_h \) SP hole states which are determined using the envelope function method based on \( \mathbf{k} \cdot \mathbf{p} \) approximation. Obtained SP states read

\[
\Psi_{\alpha_i}(\mathbf{r}) = \sum_{\nu \in \{s,x,y,z\} \otimes \uparrow, \downarrow} \chi_{\alpha_i,\nu}(\mathbf{r}) u_{\nu}^{\Gamma}, \tag{8}
\]

where \( u_{\nu}^{\Gamma} \) is the Bloch wave-function of s-like conduction band or p-like valence band at the center of the Brillouin zone, \( \uparrow/\downarrow \) mark the spin and \( \chi_{\alpha_i,\nu} \) is the envelope function, where \( \alpha_i \in \{e_j, h_k\} \).

The trial function of considered exciton complex reads

\[
|M\rangle = \sum_{m=1}^{n_{SD}} \eta_m |D^M_m\rangle, \tag{9}
\]

where \( n_{SD} \) is the number of SDs \( |D^M_m\rangle \) and \( \eta_m \) is the constant which is looked for using the variational method. The \( m \)-th SD is given by

\[
|D^M_m\rangle = \frac{1}{\sqrt{N!}} \sum_{\tau \in S_N} \text{sgn}(\tau) \phi_{\tau\{i_1\}}(\mathbf{r}_1) \phi_{\tau\{i_2\}}(\mathbf{r}_2) \cdots \phi_{\tau\{i_N\}}(\mathbf{r}_N). \tag{10}
\]

Here, we sum over all permutations of \( N := N_c + N_h \) elements over the symmetric group \( S_N \). For the sake of notation convenience, we joined the electron and hole wave functions from which the SD is composed of, in the unique set \( \{\phi_{1}, \ldots, \phi_{N}\}_m := \{\Psi_{e_1}, \ldots, \Psi_{e_{N-c}}; \Psi_{h_1}, \ldots, \Psi_{h_{N-h}}\} \), where \( j \in \{1, \ldots, n_c\} \) and \( k \in \{1, \ldots, n_h\} \). In similar fashion we join the positional vectors of electrons and holes \( \{r_{1}, \ldots, r_{N}\} := \{\mathbf{r}_{e_1}, \ldots, \mathbf{r}_{e_{N-c}}; \mathbf{r}_{h_1}, \ldots, \mathbf{r}_{h_{N-h}}\} \).

Further, we solve the Schrödinger equation

\[
H^M |M\rangle = E^M |M\rangle, \tag{11}
\]

where \( E^M \) is the energien of excitonic state \( |M\rangle \) and \( H^M \) is the CI Hamiltonian which reads

\[
H^M = H^M_0 + \sum_{k \neq l} \frac{e_k e_l}{4\pi\epsilon_0 |\mathbf{r}_k - \mathbf{r}_l|}, \tag{12}
\]

where \( H^M_0 \) represents SP Hamiltonian and the second term on the right-hand side is the Coulomb interaction between SP states. Here \( e_l \) and \( e_k \) label the elementary charge \( |e| \) of either electron, i.e., \(-e\), or hole i.e. \( e \), and \( e(\mathbf{r}_k, \mathbf{r}_l) \) is spatially dependent dielectric function. Note, that the Coulomb interaction is treated as a perturbation.

APPENDIX II.

To visualize the contents of SP states computed in multi-particle complexes calculated by CI, we need to transform the results of CI calculations to the basis of SP states instead of that of SDs. In this appendix we describe that method.

During the set-up of SDs, we create the matrix \( \hat{A} \) with rank \( n_{SD} \times N \), where \( m \)-th row consists of SP states used in the corresponding SD

\[
A_m = \left( \Psi_{e_j}, \ldots, \Psi_{e_{j+N_c-1}}; \Psi_{h_k}, \ldots, \Psi_{h_{k+N_h-1}} \right). \tag{13}
\]

As a result of CI calculation we get \( n_{SD} \) eigenvectors (CI states) with \( n_{SD} \) components

\[
|M^l\rangle = (\eta_1^l, \ldots, \eta_{n_{SD}}^l)^T, \tag{14}
\]

where index \( l \) identifies the eigenvector. Now let us consider a particular SP state \( \Psi_{e_j} \{\Psi_{h_k}\} \). We choose those values of \( \eta_m^l \) which corresponds to the \( A_m \) consisting of \( \Psi_{e_j} \{\Psi_{h_k}\} \), sum the squares of the absolute values

\[
c_{e_j} = \sum_{m} \sum_{j'} |\eta_m^l(j')|^2 \delta_{jj'}, \tag{15}
\]

\[
\tag{16}
\]

and obtain the vector

\[
\left(c_{e_1}^l, \ldots, c_{e_{N-c}}^l; c_{h_1}^l, \ldots, c_{h_{N-h}}^l \right)^T. \tag{17}
\]

The values \( c_{e_j} \) and \( c_{h_k} \) are then normalized by imposing that \( \sum_j c_{e_j}^l + \sum_k c_{h_k}^l = 1 \). Since \( |\eta_m^l|^2 \) describes the weight of the corresponding SD in CI eigenvector, we
look for the weights of individual SP electron or hole states. The example of the result is shown for $X^0$ in InAs/GaAs and GaAs/AlGaAs QD in Fig. 5 or for $X^+$ in GaAs/AlGaAs QD in Fig. 10.

The procedure described thus far allows us to study also other excitonic properties, such as the influence of multi-particle effects on band mixing or visualising the probability density of the studied excitonic complexes.

In the case of band mixing we multiply the contents of $\{|S\rangle, |HH\rangle, |LH\rangle, |SO\rangle\}$ of the particular SP state by the corresponding coefficient from Eq. (17). Hence, we get the matrix with rank $(n_e + n_h) \times 4$ for each $l$ and we sum separately all $|S\rangle$, $|HH\rangle$, $|LH\rangle$ and $|SO\rangle$ contents in that matrix to get the four corresponding values for each CI state. Finally, we normalize the contents in the same fashion as for Eq. (17).

On the other hand, for visualizing the probability density of an eigenstate of the complex $|M\rangle$ with wavefunction $\Phi_M(r)$, we calculate

$$|\Phi_M(r)|^2 = \sum_j |c_{ej} \Psi_{ej}(r)|^2 + \sum_k |c_{k-h} \Psi_{h}(r)|^2. \quad (18)$$

Similarly as before, the probability density is finally normalized, i.e., $\langle M \mid M \rangle = 1$. The example of the calculated probability density of $X^0$, $X^+$, and $X^-$ is shown in Figs. 1(b).

APPENDIX III.

Now we briefly describe the construction of the excited positive trion states. First, we introduce the single-particle approach considering the complex consisting of the electron in the ground state $(\uparrow_s/\downarrow_s)$, the heavy hole in the ground state $(\uparrow_p/\downarrow_p)$ and the heavy hole in the first excited state $(\uparrow_p/\downarrow_p)$, where the total angular momentum of electron is $J = \pm 1/2$ and that of hole is $J = \pm 3/2$. Due to the exchange interaction, holes in the complex split into singlet and triplet states and, thus, the excited positive trions read

$$|X_{2}^+\rangle = \begin{cases} \uparrow_s \uparrow_p \uparrow_p & J_z = +1/2 \\ \downarrow_s \downarrow_p \uparrow_p & J_z = -1/2 \end{cases}, \quad (19)$$

$$|X_{3}^+\rangle = \begin{cases} \uparrow_s \uparrow_p \uparrow_p & J_z = +1/2 \\ \downarrow_s \downarrow_p \uparrow_p & J_z = -1/2 \end{cases}, \quad (20)$$

$$|X_{4}^+\rangle = \begin{cases} \uparrow_s \uparrow_p \uparrow_p & J_z = +5/2 \\ \downarrow_s \downarrow_p \uparrow_p & J_z = -5/2 \end{cases}, \quad (21)$$

$$|X_{4}^++\rangle = \begin{cases} \uparrow_s \uparrow_p \uparrow_p & J_z = +7/2 \\ \downarrow_s \downarrow_p \uparrow_p & J_z = -7/2 \end{cases}, \quad (22)$$

where $J_z$ marks the projection of total angular momentum of the excited trions into the direction of $B_z$. The singlet ($X_{4}^+$) and the triplet ($X_{2}^+$ and $X_{3}^+$) states emit single photon when s-shell electron and s-shell hole recombines. Hence, we get the energy of the emitted single photon when we subtract the energy of SP p-shell hole corresponding to the three SP electron and SP hole states where the s-shell $e^- - h^+$ pair recombines. Note, that $X_{4}^+$ is the dark state due to the dipole selection rules.

Since each excitonic state calculated by CI consists of different amount of $n_e$ SP electron and $n_h$ SP hole states, we subtract the energy of the excited SP hole state which has the largest contribution in the considered CI state. That procedure allows us to reproduce the measured results. In Fig. 10 we show SP hole states, the energies of which were subtracted in our investigation of $X^+, X_{3}^+, X_{4}^+$ transitions. As can be seen, the largest contribution in the first and the second excited trion eigenstates (corresponding to numbers 3, 4 and 5, 6, and we mark them as $X_{b}^+$ and $X_{c}^+$, respectively) relates to the fourth excited SP hole state (that corresponds to the numbers 3, 4 and we mark that as $h_4$). Hence, the energies of the emitted single photons $E$ which are the results of such transitions are

$$E_4 (X_{4}^+) = \begin{cases} E^{X^+}_{c} \{u\} - E_4 \{\uparrow\} \\ E^{X^+}_{c} \{d\} - E_4 \{\downarrow\} \end{cases}, \quad (23)$$

$$E_4 (X_{3}^+) = \begin{cases} E^{X^+}_{b} \{u\} - E_4 \{\uparrow\} \\ E^{X^+}_{b} \{d\} - E_4 \{\downarrow\} \end{cases}, \quad (24)$$

$$E_0 (X_{0}^+) = \begin{cases} E_{a}^{X^+} \{u\} - E_0 \{\uparrow\} \\ E_{a}^{X^+} \{d\} - E_0 \{\downarrow\} \end{cases}, \quad (25)$$

where subscripts $a$, $b$, $c$ mark the Kramers doublet of the trion state before recombination ($a$ identifies the lowest energy). Furthermore, subscripts 0 and 4 label the Kramers doublet of the final SP hole state (0 identifies the energy of the ground state), and $u \{d\}$ denotes the higher \{lower\} energy of considered trion doublet. For further details see also Ref.13.
FIG. 10. The contribution of SP states used for the basis of the CI calculations of QD with $d = 68$ nm and $h = 16$ nm, in the non-recombined $X^+$ (left) and $X^-$ (right) trion states for $B = 0$ T. The CI calculations were performed with the basis of 12 SP electron (green) and 12 SP hole (red) states. The states in blue, green, and red rectangles in the left panel were used as the final SP states in constructing the studied emission energies of $X^+$, $X^+_{3^+}$, and $X^+_{4^+}$, respectively, and that in light blue for the right panel corresponds to the final SP state of $X^-$. The colors correspond to those in Fig. 8. Note that the CI states are ordered from 1 to 8 by their energy from lowest to highest.