

Optical and Magnetic Response by Design in GaAs Quantum Dots

Christian Schimpf,^{1,*} Ailton J. Garcia, Jr.,² Zhe X. Koong,¹ Giang N. Nguyen,³ Lukas L. Niekamp,³ Martin Hayhurst Appel,¹ Ahmed Hassanen,^{1,4} James Waller,¹ Yusuf Karli,¹ Saimon Filipe Covra da Silva,^{2,5} Julian Ritzmann,⁶ Hans-Georg Babin,⁶ Andreas D. Wieck,⁶ Anton Pishchagin,⁷ William Hease,⁷ Nico Margaria,⁷ Ti-Huong Au,⁷ Sebastien Boissier,⁷ Martina Morassi,⁸ Aristide Lemaître,⁸ Pascale Senellart,⁸ Niccolo Somaschi,⁷ Arne Ludwig,⁶ Richard J. Warburton,³ Mete Atatüre,¹ Armando Rastelli,² Michał Gawelczyk,^{9,†} and Dorian A. Gangloff^{1,‡}

¹ Cavendish Laboratory, [University of Cambridge](#), J.J. Thomson Avenue, Cambridge CB3 0HE, United Kingdom

² Institute of Semiconductor and Solid State Physics, [Johannes Kepler University](#), Altenberger Str. 69, Linz, Austria

³ Department of Physics, [University of Basel](#), Klingelbergstrasse 82, 4056 Basel, Switzerland

⁴ Department of Engineering Science, [University of Oxford](#), Parks Road, OX1 3PJ United Kingdom

⁵ Instituto de Física Gleb Wataghin, [Universidade Estadual de Campinas](#), 13083-859 Campinas, Brazil

⁶ Lehrstuhl für Angewandte Festkörperphysik, [Ruhr-Universität Bochum](#), 44780 Bochum, Germany

⁷ [Quandela](#), 7 rue Leonard da Vinci, 91300 Massy, France

⁸ [Centre for Nanosciences and Nanotechnologies](#), CNRS, [Université Paris-Saclay](#), UMR 9001, 10 Boulevard Thomas Gobert, 91120 Palaiseau, France

⁹ Institute of Theoretical Physics, [Wrocław University of Science and Technology](#), Wrocław, Poland



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Quantum networking technologies use spin qubits and their interface to single photons as core components of a network node. This necessitates the ability to co-design the magnetic- and optical-dipole response of a quantum system—a capability that has been notably absent in solid-state platforms where spin-orbit coupling and the crystalline environment lead to inhomogeneity of electronic g -factors and optically active states. Here, we demonstrate the ability to design both the optical and magnetic response of a solid-state quantum emitter *a priori*. We show that GaAs quantum dots (QDs), obtained via local droplet etching epitaxy and already known as exceptionally coherent and efficient quantum light sources, also exhibit spin and optical properties that follow directly from assuming the highest possible system symmetry. Our measurements of electron and hole g -tensors—using a new sign-sensitive measurement protocol based on the hyperfine interaction—and of transition dipole moment orientations for charged excitons agree with our predictions from a multiband $\mathbf{k} \cdot \mathbf{p}$ simulation constrained only by a single atomic-force-microscopy reconstruction of QD morphology. This agreement is verified across multiple wavelength-specific growth runs at different facilities within the range of 730 to 790 nm for the exciton emission. Remarkably, our measurements and simulations track the in-plane electron g -factors through a zero-crossing from -0.1 to 0.3 and linear optical dipole moment orientations fully determined by an external magnetic field. The robustness and generality of these results establish a fundamentally new paradigm for solid-state spin-photon interfaces: one in which the properties of a spin qubit and its tunable optical interface can be designed—prior to growth—for a target magnetic and photonic environment, with direct applications to scalable and high-fidelity spin-photon entanglement.

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*Contact author: cs2273@cam.ac.uk

†Contact author: michal.gawelczyk@pwr.edu.pl

‡Contact author: dag50@cam.ac.uk

I. INTRODUCTION

Entanglement between matter-based and light-based qubits is the key resource in quantum communication and distributed quantum computation [1,2], and to deterministically generate resource states for measurement- and fusion-based quantum computation [3–6]. Amongst the variety of physical platforms being explored for this task, including atom-cavity systems [7], solid-state structures hold promise for compact and integrated systems that can be deployed at scale for useful applications. These include diamond or SiC color centers [8,9], rare-earth ions [10], and semiconductor quantum dots (QDs) [11,12] as promising candidates that satisfy the basic requirements of marrying a coherent spin-photon interface with a long-lived quantum memory. However, solid-state systems present unique challenges, including phonon coupling [13], charge noise, and nuclear-spin noise [14], and have so far lacked the precise control and reproducibility needed for scalable quantum devices.

Despite these challenges, semiconductor quantum dots have emerged as commercially viable quantum light sources, owing to their near-unity quantum efficiency, short radiative lifetimes, and high single-photon purity and photon indistinguishability [15]. Their light-matter coupling can be further enhanced via photonic structures such as integrated vertical cavities [16], circular Bragg resonators [17–19], photonic crystals [20], or open cavities [11]. For the latter, an impressive $>70\%$ photon collection efficiency into a single-mode fiber has now been demonstrated [21]. Recently, chiral interfaces between emitters and integrated photonic circuits were shown to benefit strongly from precise engineering of the photon polarization modes [22,23]. Going further, an exact alignment of the QD transition dipole moment to its photonic environment will be crucial.

On the spin side, proof-of-concept experiments with electron-, hole- and nuclear spin qubits in QDs [24–29] have highlighted their further potential for single-spin and spin-ensemble physics, with a prospect towards practical quantum memories [30], quantum computation [31,32], and the generation of multidimensional photonic cluster states [33]. The spin qubit in a QD is formed by splitting the discrete energy levels of a ground-state particle, often a single electron, via a static magnetic field. Spin control and read-out can be realized very efficiently by exploiting their coherent spin-selective optical transitions between a ground state spin and a charged exciton state (“trion”) [34–37].

Considering the vital role of light-matter-coupling in qubit control and photon extraction, it becomes clear that creating QD devices for integration within scalable photonic quantum technologies requires designing the spin-photon interface *a priori*, i.e., before device fabrication and integration. To do this, we require precise knowledge

and control over the energy-level structure of the spin qubit and the exciton states, via their g -tensor and the radiative transition dipole moments (TDMs) between them.

The spin qubit and transition dipoles are governed by the magnetic response of the particles involved, described by their effective g -tensors and the phase-relations between their wave functions [38,39]. In semiconductor QDs, the g -tensors are governed by a competition between the free-particle magnetic response and spin-correlated orbital currents (SCOCs) [40] (see Sec. II) resulting from spin-orbit interaction. Analogous to the classical magnetic moment of a current loop, the SCOCs depend on the size and the properties of the system in which they can “flow”. This leads to a strong sensitivity of the g -tensors and the TDMs to QD geometry, material composition, and strain [39,41–43], making the g -tensors notoriously difficult to predict and to reproduce. In InGaAs, the QD material system most studied to date for both photonic [44] and spin applications [26,45–47], the strain-driven growth leads to a deviation from a radially symmetric shape and alloy composition around the growth axis and to built-in strain. The reduced symmetry leads to complicated dependencies of the TDMs on the external magnetic field [38,39,43]. For substantial geometric imperfections or large strain, the TDMs are entirely pinned to a specific direction, independent of the magnetic field orientation [48,49], making alignment of TDMs to photonic structures very challenging [28]. Variations of shape and strain from QD to QD render the g -tensors and TDMs of a single InGaAs QD nearly impossible to predict and even difficult to fully determine experimentally [42,48].

In contrast to that, GaAs QDs obtained via the local Al droplet etching (LDE) technique [50,51], have demonstrated remarkable reproducibility and homogeneity, which led to the reported low variance in emission wavelengths, low fine structure splitting [51,52] and Fourier-limited linewidth [53]. Recently, electron spin coherence times beyond $100\ \mu\text{s}$ were demonstrated in such LDE GaAs QDs, outperforming other QD material systems by orders of magnitude and hinting towards nuclear-spin-memory times beyond $100\ \text{ms}$ [12,54–56]. This prior work on LDE GaAs QDs highlights their potential for modern quantum applications that require well-defined spin-photon interfaces and long spin coherence time. Despite impressive advances, the ability to predict and control the optical and magnetic dipole moments of GaAs QDs relative to their photonic and magnetic environments remains a key challenge.

In this work, we show that GaAs QDs adhere closely to the behavior dictated by D_{2d} point group symmetry, making their optical and magnetic dipoles predictable and reliably controllable. To do so, we elucidate, both theoretically and experimentally, the material parameters, mechanisms, and symmetries governing the magnetic dipoles of the electron and the negatively charged trion in LDE

GaAs QDs and the optical transition dipoles between them. Using only atomic-force-microscopy (AFM) measurements of the droplet-etched nanoholes [51] as an input, we simulate the full g -tensors and TDMs in a framework of combined multiband $\mathbf{k} \cdot \mathbf{p}$ [57,58] and configuration-interaction [59] methods over a large parameter space. Using resonant spectroscopy and hyperfine-mediated measurements to fully characterize the magnetic and optical response of a QD, we obtain the g -tensors of the electron and the trion—including their sign—and the involved optical transition dipoles. We demonstrate excellent agreement with our simulations for LDE GaAs QDs from different molecular beam epitaxy growth runs and even different growth facilities over emission wavelengths from 730 to 790 nm, including those with an electron g -factor close to zero. Our work confirms that in modern LDE GaAs QDs the high in-plane symmetry, which leads to low fine structure splitting [52], also causes the TDMs to strongly adhere to those expected for pure D_{2d} symmetry. This makes the TDMs not only predictable but also fully tuneable by the applied magnetic field. Our work shows that, thanks to predictive theory and the exceptional uniformity and reproducibility of GaAs QDs grown by the liquid droplet etching method, designing the spin-photon interface of QD devices integrated with photonic structures is now possible and within reach. Unlike previous quantum emitter platforms—where spin and optical properties must be selected post-growth from a statistical spread—these properties are predictable and tuneable by design.

II. SPIN-PHOTON INTERFACE IN LDE GAAS QUANTUM DOTS

Figure 1(a) sketches an LDE GaAs QD, using the coordinate system of the crystal axes $\hat{x}, \hat{y}, \hat{z} := [100], [010], [001]$. A static magnetic field $\mathbf{B}$, applied at an angle χ relative to the optical axis $\hat{z}$ and an angle φ to $\hat{x}$, splits the spin of a confined electron into the two Zeeman-eigenstates $|e^\pm\rangle$. These states couple to the two negatively charged exciton (trion) states $|T^\pm\rangle$ via four possible optical transitions, E_i , $i = 1, 2, 3, 4$, with their respective TDMs described by the Stokes vectors $\mathbf{s}_i$. The electron has two spin projections $\pm 1/2$, while the trion is composed of two electrons in a spin-singlet configuration and a heavy hole (HH) with two spin projections $\pm 3/2$. The resulting optically active double-lambda system, shown in Fig. 1(b), is the workhorse for initializing, reading out, and controlling the electron spin-qubit in III–V semiconductor QDs [36]. For spin control and readout, two extremal configurations of χ are typically used, which are complementary in their use cases. In the *Faraday* configuration ($\chi = 0$), the forbidden diagonal transitions $E_{1,4}$ lead to a high cyclicity of the remaining transitions $E_{2,3}$, which enables non-destructive and single-shot readout [37]. In the *Voigt* configuration ($\chi = \pi/2$), all four transitions are equally allowed and enable coherent optical control of the spin-qubit via a two-color Raman scheme [36], but readout leads to an almost immediate destruction of the spin state (in an isotropic photonic environment). At oblique angles $0 < \chi < \pi/2$ all transitions become partially allowed [60].

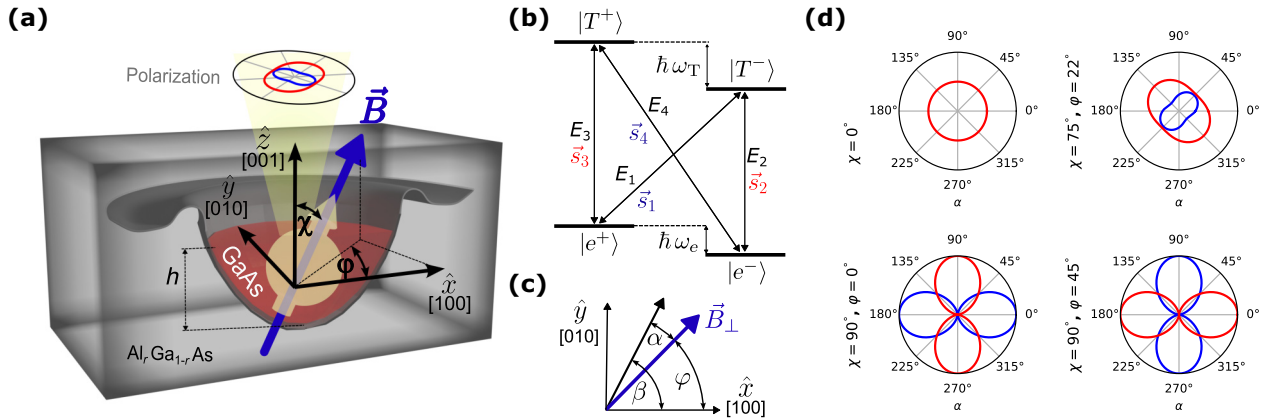


FIG. 1. (a) Schematic of a spin-photon interface in a GaAs QD obtained by local Al droplet etching. A single electron is confined in the QD consisting of pure GaAs with a filling height of h , buried in an $\text{Al}_r\text{Ga}_{1-r}\text{As}$ host matrix. (b) Energy-level structure of a GaAs QD in a static magnetic field $\mathbf{B}$ at a polar angle χ to the $[001]$ crystal axis ($\hat{z}$) (the growth- and optical axis) and an azimuthal angle φ relative to $[100]$ ($\hat{x}$). The ground-state electron and the negative trion split into the states $|e^\pm\rangle$ and $|T^\pm\rangle$, respectively. The four possible optical transitions E_{1-4} with their Stokes vectors $\mathbf{s}_{1-4}$ are ordered by their energies in ascending order. (c) In-plane magnetic field component $\mathbf{B}_\perp$ aligned at an angle φ relative to $\hat{x}$. An arbitrary angle of interest is either given as α relative to $\mathbf{B}_\perp$ or as β relative to $\hat{x}$. (d) Polar plots of the linear projection of the optical transition dipole moment for transitions $E_{1,4}$ (blue) and $E_{2,3}$ (red) for a system with D_{2d} symmetry and all positive g -factors vs α for different configurations of χ and φ . The angle $\alpha = 0^\circ$ corresponds to the direction of $\mathbf{B}_\perp$.

The high rotational symmetry of the cone-shaped LDE GaAs QDs around the $\hat{z}$ -axis and the low built-in strain [51] allow us to assume the D_{2d} point symmetry group as a first approximation. Although a conical shape, as opposed to a circular disk, formally reduces the symmetry to C_{2v} , this is limited to weak interfacial effects that, as we show, can be neglected. In this limit, we can neglect heavy hole-light hole (HH-LH) mixing. Given this assumption, $|e^\pm\rangle$ and $|T^\pm\rangle$ are separated by their respective Zeeman splittings,

$$\hbar\omega_{e,t} = \mu_B |\mathbf{B}| \sqrt{(g_z^{e,t} \cos \chi)^2 + (g_\perp^{e,t} \sin \chi)^2}, \quad (1)$$

with μ_B the Bohr magneton, $g_z^{e,t}$ the out-of-plane g -factors and $g_\perp^{e,t}$ the isotropic in-plane g -factors for the electron (e) and the trion (t), respectively. The four emerging transition energies are then given by $E_{1,2}/\hbar = \omega' + (-\omega_t \mp \omega_e)/2$ and $E_{3,4}/\hbar = \omega' + (\omega_t \mp \omega_e)/2$, with ω' the central transition frequency.

The magnetic response of an isolated particle in a vacuum is described by the Landé g -factor,

$$g_l = 2m_J \left(1 + \frac{J(J+1) + S(S+1) - L(L+1)}{2J(J+1)} \right), \quad (2)$$

such that, in analogy to Eq. (1), the total energy splitting in a magnetic field is $\mu_B |\mathbf{B}| g_l$ (i.e., we incorporate m_J into g_l to adhere to common experimental nomenclature), with L the orbital angular momentum, S the spin, $J = L + S$ the total angular momentum, and m_J the projection of J along $\mathbf{B}$. For an electron in a III-V semiconductor conduction band (CB), $S = 1/2$, $L = 0$, and $m_J = 1/2$ this results in $g_l = 2$, while for a valence band HH, $S = 1/2$, $L = 1$, and $m_J = 3/2$, this results in $g_l = 4$. In a bulk semiconductor with nonzero spin-orbit coupling, such as GaAs, this single-particle magnetic momentum has an added contribution from induced spin-correlated orbital currents (SCOCs) perpendicular to $\mathbf{B}$ [40]. The SCOCs depend on both the particle's envelope function and the lattice-periodic Bloch function [61]. For the electron, with a spherically symmetric Bloch function, the total magnetic response converges to the isotropic Γ -point bulk g -factor described by Roth's formula [62],

$$g_c = 2 - \frac{2E_p \Delta_{SO}}{3E_g (E_g + \Delta_{SO})}, \quad (3)$$

with E_p the interband coupling strength, E_g the band-gap energy and Δ_{SO} the valence-band spin-orbit splitting. For bulk GaAs at cryogenic temperatures $E_p = 28.8$ eV, $E_g = 1.519$ eV, and $\Delta_{SO} = 0.341$ eV [63,64], which results in $g_c = -0.44$, i.e., a negative contribution from the SCOCs dominates the bulk magnetic response.

In a QD, the confinement potential constrains the carrier motion to a region comprised of about 10^5 lattice sites [54],

which quenches the SCOCs, and hence their contribution to the g -factor. This can also be understood as the effect of quenching of electron angular momentum, and thus spin-orbit effects, at higher wavevector components enforced in QD states by spatially finite confinement as opposed to Γ -point bulk electrons [65]. We can, therefore, intuitively understand how the reported near-zero electron g -factors in LDE GaAs QDs [12,54,55] can emerge from a balance between the single particle response and the SCOCs. The trion's magnetic response is dominated by the HH, for which the SCOCs are more complex because of the lack of spherical symmetry of the p -like Bloch function [38,48,66]. But they still provide a competing contribution, i.e., one that is opposite to the free-particle magnetic moment. In the Faraday configuration, values of $g_z^t \gg g_z^e$ are reported [67], which means that the HH g -factor tends more closely towards the isolated-particle limit than the electron g -factor. In the Voigt configuration, the direct Zeeman coupling strength for HHs is zero due to the symmetry of the HH Bloch functions and the growth axis being distinguished. Instead, $g_\perp^t$ is governed by non-Zeeman interactions and mixing between HH and light-hole (LH) subbands [39,43,68], which we will discuss in more detail in Sec. III.

This qualitative insight into the magnetic response of the pure HHs already allows us to determine the orientation of the TDMs for an ideally D_{2d} -symmetric case, where the phase relations between $|e^\pm\rangle$ and $|T^\pm\rangle$ play a key role [69]. The in-plane angles relevant in the following are shown in Fig. 1(c), with φ the angle between the in-plane component of the magnetic field $\mathbf{B}_\perp$ and $\hat{x}$, α the angle between an arbitrary vector of interest and $\mathbf{B}_\perp$, and $\beta = \varphi + \alpha$. Figure 1(d) then depicts the resulting TDM polarizations for four different orientations of $\mathbf{B}$. In the Faraday configuration ($\chi = 0$), the angular momentum along the $\hat{z}$ -axis is conserved due to the radial symmetry so that only E_2 and E_3 are dipole-allowed if $S(g_z^e) = S(g_z^t)$, or only E_1 and E_4 if $S(g_z^e) \neq S(g_z^t)$, where $S(x)$ denotes the sign. In both cases, the polarizations are left- and right-circular, respectively [60]. In the Voigt configuration ($\chi = \pi/2$), E_{1-4} are equally dipole-allowed, where $s_1 = s_4$ are linear and orthogonal to $s_2 = s_3$. The angles $\beta_{1,2}$ between $s_{1,2}$ and $\hat{x}$ become [38,42,43]

$$\begin{aligned} \beta_1 &= -\varphi, & \beta_2 &= -\varphi + \frac{\pi}{2} & \text{for } S(g_\perp^e) = S(g_\perp^t), \\ \beta_1 &= -\varphi + \frac{\pi}{2}, & \beta_2 &= -\varphi & \text{for } S(g_\perp^e) \neq S(g_\perp^t). \end{aligned} \quad (4)$$

Interestingly, we can observe from Eq. (4) that s_1 or s_2 align with $\mathbf{B}$ for each turn of φ by $\pi/4$. As shown in Fig. 1(d), for $\varphi = 0$ the magnetic field $\mathbf{B}$ is aligned with $\hat{x}$ and $s_{1,4}$ are aligned with $\mathbf{B}$ (in the case of $S(g_\perp^e) = S(g_\perp^t)$). For other angles, the TDMs counter-rotate with respect to $\mathbf{B}$. For example, rotating $\mathbf{B}$ to the [110] crystallographic direction at $\varphi = \pi/4$ rotates the linear polarization

by $-\pi/4$, so that $s_{2,3}$ instead are aligned with $\mathbf{B}$. Note that the behavior in Eq. (4) can only hold for highly in-plane symmetric QDs with (atomistically) homogeneous QD material and negligible interfacial effects. Deformation, strain, material alloying in the QD, or defects can lead to strikingly different results, as clearly observed in InGaAs QDs [39,43,49]. For angles $0 < \chi < \pi/2$, the polarization becomes a superposition between the Faraday and the Voigt cases [69].

III. SIMULATION FROM ATOMIC FORCE MICROSCOPY DATA

The assumption of a global D_{2d} symmetry holds strictly for GaAs/AlGaAs quantum wells, where the GaAs layer extends out quasi-infinitely in the $\hat{x}$ and $\hat{y}$ directions. However, in the case of LDE GaAs QDs, even though the overall geometry has almost ideal rotational symmetry, the inclined sidewalls of the cone-like geometry combined with the tetrahedral crystal symmetry formally result in the C_{2v} symmetry due to the lack of inversion symmetry in

the $\hat{z}$ -direction. This effect is expected to be weaker than explicit QD in-plane shape anisotropy, as it exhibits only at the GaAs-AlGaAs interface [70]. In general, C_{2v} symmetry can lead to HH-LH mixing and anisotropy in the in-plane g -factors. To assess the importance of these deviations from the D_{2d} -symmetric case discussed in Sec. II and to calculate the g -tensor values, we perform simulations in the multiband $\mathbf{k} \cdot \mathbf{p}$ framework [57,58] combined with the configuration-interaction method [59] for trion states. The form of the Hamiltonian used and details of the magnetic field inclusion using the gauge-invariant discretization scheme [71] can be found in Ref. [72]. As an input, we only take the shape of a nanohole created during the growth process, as shown in the AFM image in Fig. 2(a), and parametrize the nominal GaAs filling height h and the Al fraction r of the $\text{Al}_r\text{Ga}_{1-r}\text{As}$ barrier. This allows us to calculate the wave-function envelopes of both the electron and the hole and further the eigenstates of the trion, including the admixtures of electron and hole excited levels present in the trion ground state. For all these states, we calculate the response to magnetic fields.

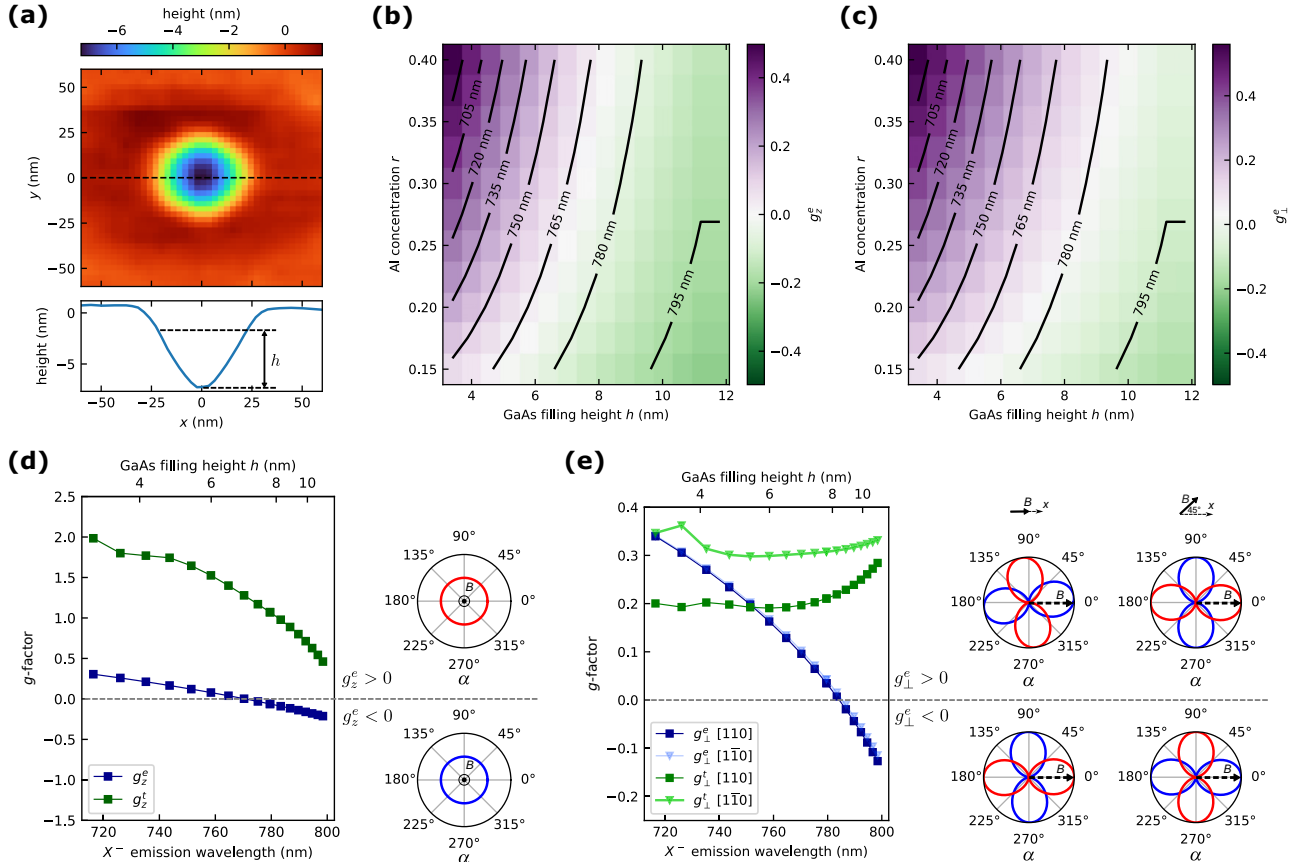


FIG. 2. (a) AFM image of an Al droplet etched nanohole, used to form a GaAs QD. The bottom graph shows a cross-section at $y = 0$. (b),(c) Simulated electron g -factors for $\mathbf{B}$ -fields in (b) [001] and (c) [110] directions as a function of GaAs filling height h and $\text{Al}_r\text{Ga}_{1-r}\text{As}$ barrier Al concentration r . The black lines indicate constant trion-to-electron (X^-) emission wavelength. (d),(e) Simulated electron- and trion g -factors in a GaAs QD in an $\text{Al}_{0.25}\text{Ga}_{0.75}\text{As}$ barrier in the (d) Faraday and (e) Voigt configurations as a function of the X^- emission wavelength. The polar plots show the associated transition dipole moment polarizations $s_{1,4}$ (blue) and $s_{2,3}$ (red).

Figures 2(b) and 2(c) show the calculated g_z^e and g_z^t , respectively, as a function of h and r . The solid curves highlight the constant trion-to-electron (X^-) emission wavelengths. The emission wavelength is strongly correlated with both h and r , as these two values mostly determine the discrete energy levels in the QD [51]. The two color maps can serve as a guideline for designing QDs to achieve the desired magnetic response. From here on, we will use the X^- emission wavelength, as tuned exclusively by h , as our control parameter.

Figure 2(d) shows the Faraday configuration response, i.e., g_z^e and g_z^t as a function of X^- emission wavelength at a typical value of $r = 0.25$. For g_z^e we observe a zero-crossing at about 770 nm, where the spin-orbit effects begin to dominate the magnetic response. Larger QDs have higher emission wavelengths and also more dominant SCOCs, pushing the g -factor towards the bulk limit of -0.44 . The trend for g_z^t is similar but with overall higher values compared to g_z^e . This behavior reflects both the higher bare particle Landé g -factor value in the valence band and quenched SCOCs for the HH due to its p -like Bloch functions, which make the g -factor tend more towards the isolated hole value of 4. The polarization of the two allowed transitions for the two scenarios $g_z^e > 0$, $g_z^t > 0$ (upper right) and $g_z^e < 0$, $g_z^t > 0$ (lower right), are almost perfectly circular and opposite. These almost perfectly circular polarizations again indicate the behavior expected from a fully D_{2d} -symmetric system free of LH effects.

The behavior of g -factors for the Voigt configuration is more complex, as shown in Fig. 2(e). The dependency of $g_\perp^e$ on the emission wavelength is very similar to the case of g_z^e , but the zero-crossing wavelength is offset by about 15 nm to 785 nm. The offset stems from the quenched SCOCs in the $\hat{z}$ -direction (the shortest QD dimension in the plane perpendicular to $\mathbf{B}$), which decreases the contribution of spin-correlated orbital momentum compared to the Faraday configuration. This quenching pushes the g -factor further up towards the free particle limit of 2. The simulation also reveals the existence of anisotropy in $g_\perp^e$ and $g_\perp^t$, with principal axes along the $[110]$ and the $[1\bar{1}0]$ crystallographic directions. These anisotropies originate from the global C_{2v} symmetry when considering the non-equivalence of $[110]$ and $[1\bar{1}0]$ directions in (Al)GaAs, in combination with the distinguished direction in the z -axis, and persist even in the absence of strain and for perfectly in-plane symmetric QDs [70]. Although the absolute anisotropy $\delta g_\perp^e \approx 0.005$ is small, its significant relative value $\delta g_\perp^e / \langle g_\perp^e \rangle$ in LDE GaAs QDs, with $\langle g_\perp^e \rangle$ the g -factors averaged over both in-plane directions, leads to a tuneable noncollinear electro-nuclear interaction, as observed experimentally [73]. This interaction allows for an electron-spin-dependent feedback loop to be established with nuclear spins, which we exploit in Sec. IV to determine the sign of the g -tensor components. The trion

exhibits an almost constant $\langle g_\perp^t \rangle \approx 0.25$ and a significant anisotropy $\delta g_\perp^t$ up to about 0.15.

The built-in biaxial strain due to lattice mismatch in QDs splits the HH and LH bands by Δ_{LH} . That strain in LDE GaAs QDs is very low (about 0.05 %) but not negligible and results in a minor HH-LH splitting of $\Delta_{\text{LH}}/2\pi \approx 750$ GHz [74] (two orders of magnitude lower compared to InGaAs QDs). The lack of $\hat{z}$ -axis symmetry enables the “same-spin” HH-LH mixing effect between HH with $m_J = 3/2$ and LH with $m_J = 1/2$ (and the same for flipped spins), typically referred to as the S matrix element in the Luttinger-Kohn Hamiltonian [57]. Additionally, any in-plane asymmetry of the overall confining potential enables the “opposite-spin” mixing, $\pm 3/2 \leftrightarrow \mp 1/2$ given by the R matrix element. Perturbatively, those effects lead to LH contributions to the hole ground state of magnitudes $|\langle S \rangle / \Delta_{\text{LH}}|^2$ and $|\langle R \rangle / \Delta_{\text{LH}}|^2$, respectively. Since Δ_{LH} is low, even weak mixing effects lead to an overall LH contribution of approximately equal to 7.6 % within the QD volume, according to our simulation. Despite this significant HH-LH mixing, the TDMs are almost perfectly circular in the Faraday configuration (99.7% degree of circular polarization [69]) and, in the Voigt configuration, also closely follow the D_{2d} symmetry prediction of Eq. (4). This behavior is evidenced by the $-\pi/2$ rotation of the linear polarization relative to $\mathbf{B}$ for a $\pi/4$ rotation of φ , as shown on the right side of Fig. 2(e). While the “same-spin” LH admixture does not add to the same optical transition as the HH, the “opposite-spin” admixture can contribute with a polarization orthogonal to the HH’s. To understand the robustness of the optical properties despite the presence of the “opposite-spin” LH admixture, we have to consider its spatial dependence. Close to the GaAs/AlGaAs interface, the bi-axial strain in the QD crosses zero and becomes compressive in the AlGaAs barrier [69]. For in-plane symmetric QDs, this leads to a high HH-LH admixture close to the QD border but a vanishing one at the QD center, where the electron resides. Additionally, the LH admixture envelope is predominantly odd, so its overlap with the electrons is close to vanishing. As a consequence, the HH-LH mixing contributes significantly to the energy shifts of the trion via g^t , but the effect on the TDM polarization remains close to negligible. These findings resolve the apparent contradiction between the presence of significant HH-LH mixing [75] and the near-perfect polarization-entangled photon pairs produced by LDE GaAs QDs [76,77]. We provide more detailed insight into HH-LH mixing and its constituents in the Supplemental Material [69].

IV. OPTICAL AND SPIN-POLARIZATION MEASUREMENTS

Equipped with detailed predictions on the g -tensors and TDMs of LDE GaAs QDs, we are now in a position to verify them experimentally. All measured QDs are embedded

in the intrinsic region of a p - i - n diode, as used in Ref. [54], to achieve charge control and to load the QD with a single electron [69]. The magnetic response in the Faraday configuration is well studied and thus not the focus of our experiments [67,78,79]. However, pinning down the exact values and signs of $g_{\perp}^e$ and $g_{\perp}^t$ experimentally has proven difficult in the past, as probing only the electron and trion Zeeman splittings and the polarization of the TDMs leaves the g -factor signs ambiguous [43,48]. This ambiguity can lead to assignments of g -factors to the wrong particle. To overcome this hurdle, we make use of an electron-specific magnetic response to disambiguate the g -tensor components: dynamic nuclear polarization (DNP) mediated by the Fermi-contact hyperfine interaction [24], in which an electron-spin-dependent nuclear polarization shifts the optical resonance frequencies of the QD. All our measurements, therefore, consist of two steps: first, we perform a polarization-resolved resonance-fluorescence (RF) scan of the transitions E_{1-4} to determine all energy splittings and the TDM orientation; second, we sense the electron-spin-dependent hyperfine-shift so that we can assign the correct g -tensor components and signs.

We will now demonstrate this two-step protocol for one example QD. We apply a magnetic field of 5.8 T in the $\hat{x}$ direction, resulting in the double-lambda system shown in Fig. 3(a) with four equally allowed dipole transitions and $s_1 = s_4$ and $s_2 = s_3$. The indicated level splittings in the sketch are only one possible example and not to scale, as we seek to determine the exact state configuration. In our first step, we excite the optical transitions between $|T^{\pm}\rangle$ and $|e^{\pm}\rangle$ resonantly using a frequency-tunable laser with a frequency $\omega_L/2\pi$ and a linewidth much smaller than the Fourier-limited linewidth $\Gamma_F/2\pi \approx 700$ MHz [53]. Our signal are the photons scattered by the excited trion state and collected through a polarizer oriented to filter out the laser excitation. In the measurements shown in Fig. 3(b) (left side), the laser is circularly polarized and thus couples to all four optical transitions with equal strength. The corresponding four bright features at low and high V_G are observed in the diode bias ranges over which fast electron spin reset through co-tunneling from and into the QD competes with optical spin pumping to maintain a bright steady-state fluorescence. The dark regions between each low-high voltage pair (visualized by black dotted lines) indicate slow co-tunneling and thus efficient optical spin-pumping into an electronic dark state [80]. The change of transition energy with varying V_G can be attributed to the changing electric field experienced by the QD in $\hat{z}$ -direction. In the context of this work, the effects of this variation on the electric and magnetic dipoles can safely be neglected [81] (more details, including simulations, can be found in the Supplemental Material [69]). To avoid the buildup of nuclear spin polarization during the measurement, we ramp V_G with a frequency of 1 kHz to periodically enter the co-tunneling regime, effectively

scrambling the electron- and the nuclear polarization [54]. The inhomogeneous linewidth extracted from this measurement is $\Gamma/2\pi = 0.72(4)$ GHz, which sets the resolution of the measurement close to Γ_F . On the right-hand side of Fig. 3(b), we see the same measurement, now with the laser polarized linearly along the direction of $\mathbf{B}$. In this case, the transitions E_2 and E_3 almost completely vanish and we can estimate $s_1^{(1)} = s_4^{(1)} = 0.86(10)$ and $s_2^{(1)} = s_3^{(1)} = -0.86(10)$, with $s^{(1)}$ the rectilinear parameter of the Stokes vector [69]. This means that to a good approximation $s_1^{(1)}, s_4^{(1)} \parallel \mathbf{B}$ and $s_2^{(1)}, s_3^{(1)} \perp \mathbf{B}$, as expected from Eq. (4).

In the second step, we determine the electron-spin orientation of each transition directly using DNP [82,83]. To explain the dynamics of this process, we assume a (predominantly) isotropic $g_{\perp}^e$ and switch into the basis of the spin quantization axis $\hat{x}$ with a magnetic field $\mathbf{B} = B_x \hat{x}$. The Hamiltonian of the electron interacting with N nuclei via the Fermi-contact hyperfine interaction is given by [24]

$$\mathcal{H}_{e,n} = g_{\perp}^e \mu_B B_x \hat{S}_x - \gamma_n B_x \sum_j^N \hat{I}_x^j + a \sum_j^N \hat{S}_x \hat{I}_x^j, \quad (5)$$

using the basis $|S_x, I_x\rangle$, with $\hat{S}_x$ the electron spin projection, $\hat{I}_x^j$ the spin-projection of the j th nucleus, γ_n the nuclear gyromagnetic ratio and a the average hyperfine constant [24]. The energy spacing between two states with an average $I_x = \langle \hat{I}_x \rangle$ differing by a single nuclear-spin flip $\delta I_x = \pm 1$ is then

$$E(S_x, I_x \pm 1) - E(S_x, I_x) = \pm (-\gamma_n B_x + a S_x). \quad (6)$$

From Eq. (6), we see that for the present case of $|\gamma_n| \ll |g_{\perp}^e \mu_B|$ and a nuclear Zeeman splitting $\hbar\omega_Z^n = |\gamma_n B_x| > a$, with $a > 0$, the states of the manifold $|\downarrow_x, I_x\rangle$ ($|\uparrow_x, I_x\rangle$) exhibit an energy splitting of ω_Z^n increased (decreased) by a , as depicted in Fig. 3(c). And, crucially, this difference of energy splittings only depends on the electron spin state S_x , not on the sign of the electron g -factor [67]. The trion is, to a good approximation, unaffected by the hyperfine interaction due to the electrons being in a singlet state and the hole's p -type Bloch function [84,85]. For a fixed change in nuclear polarization, the transition energy between the ground state electron and the trion thus increases or decreases depending on the electron spin state. Whether the optical transition energy increases or decreases with nuclear polarization thus acts as an absolute reference for defining the signs and relative magnitudes of $g_{\perp}^e$ and $g_{\perp}^t$. We measure this effect via optical resonance dragging [83], as follows.

In the RF scheme shown in Fig. 3(c) the laser is in resonance with the electron-trion transition $\hbar\omega_0 = \hbar\omega' + aI_x^0$, with $\hbar\omega'$ the electron-trion transition energy without

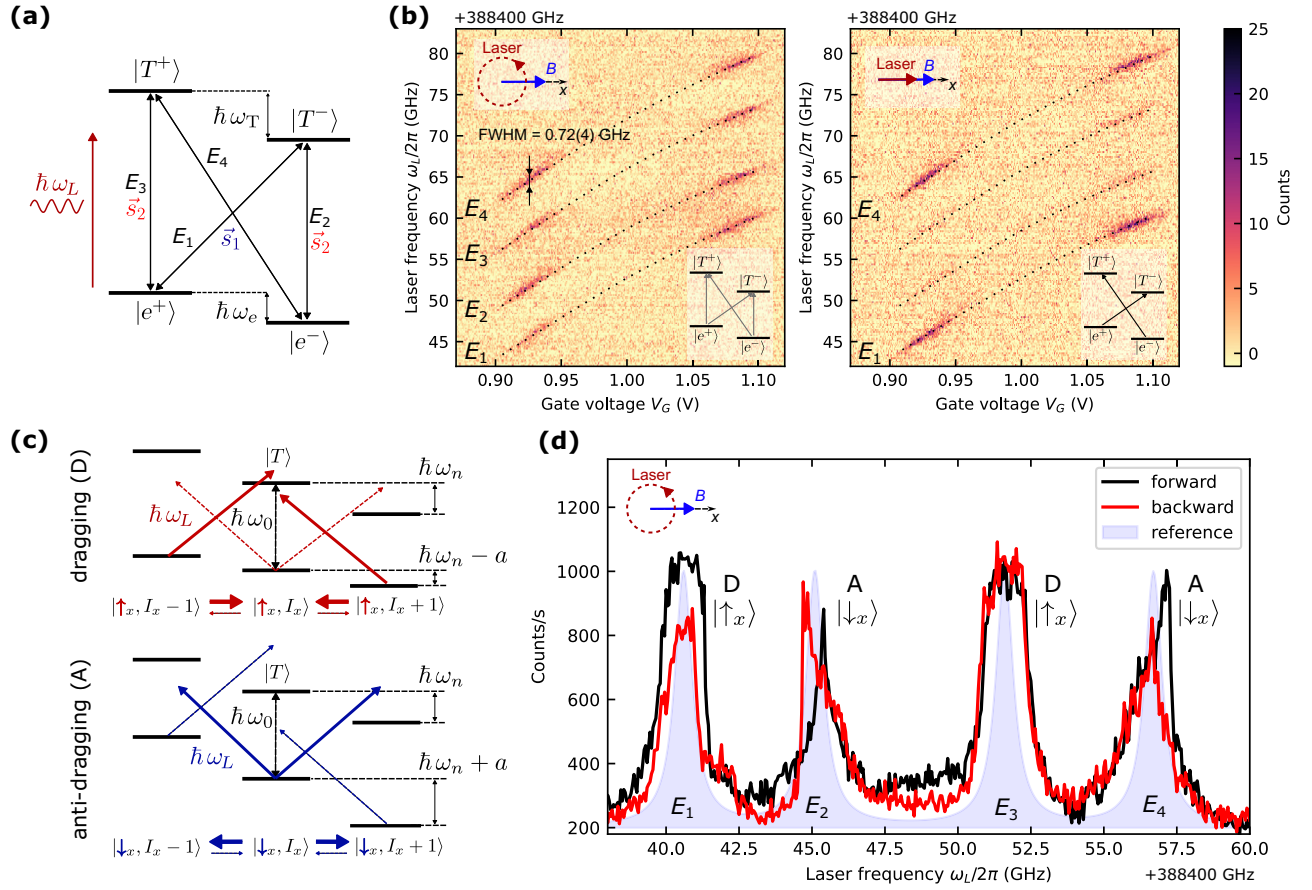


FIG. 3. (a) Scheme of the $|T^\pm\rangle$ and $|e^\pm\rangle$ energy levels in the Voigt configuration ($\chi = \pi/2$), driven by a narrow-band cw laser with frequency $\omega_L/2\pi$. In this configuration $s_1 = s_4$ and $s_2 = s_3$. (b) Resonance fluorescence (RF) emission intensity of the trion as a function of the gate voltage V_G and ω_L with $B_x = 5.7$ T. Left: a circularly polarized laser couples equally to E_{1-4} , resulting in four resonances. Right: a laser linearly polarized along $\mathbf{B}$ results in the predominant excitation of $E_{1,4}$, indicating $s_{1,4}^{(1)} \parallel \mathbf{B}$ and $s_{2,3}^{(1)} \perp \mathbf{B}$, with $s^{(1)}$ the rectilinear component of $\mathbf{s}$. The black dotted lines indicate the approximate resonance conditions for each transition. The transition linewidth extracted from this graph is $0.72(5)$ GHz. (c) Energy levels of the hyperfine-coupled electron-nuclear manifolds with the electron spin $|\uparrow_x\rangle$ (top) and $|\downarrow_x\rangle$ (bottom) drawn as the ground state. The diagonal arrows indicate electron-trion side-band transitions with a difference in I_x of $\delta I_x \pm 1$. In Ga and As the hyperfine constant $a > 0$, hence the $|\uparrow\rangle$ manifold tends to self-stabilize (“dragging”), while the $|\downarrow\rangle$ manifold avoids the resonance (“antidragging”). (d) Intensity of the trion emission at $\varphi = 45^\circ$ with a circular laser polarization at $V_G = 0.93$ V and a slowly varying ω_L from lower to higher frequencies (black) and reverse (red). The dragging transitions (D) show flat-top behavior, while the anti-dragging transitions (A) reveal a triangle-like shape. As a reference, the light blue filled area shows a theoretical pattern given by four Fourier limited transitions in the absence of dragging.

hyperfine interaction and I_x^0 the steady-state nuclear polarization. A small anisotropy in $g_\perp^e$ weakly allows sideband transitions with $\delta I_x = \pm 1$ to change the nuclear polarization [73]. When $S_x = +1/2$ and the optical transition energy decreases with nuclear polarization (upper panel), the side-band transitions involving a spin-flip towards I_x^0 are closer to resonance than sideband transitions involving a spin-flip away from I_x^0 . This leads to an inward flow, towards a stable locking-point I_x^0 . When ω_L changes slowly, the locking-point ω_0 is “dragged” by building up nuclear polarization such that the resonance condition $\omega_0 = \omega_L$ and thus the photon scattering level remains fixed [82,83]. At some point, the nuclear spin relaxation

outpaces the weak side-band transitions, and the dragging abruptly ends. The opposite behavior is observed for the scenario shown in the lower part of Fig. 3(c), where $S_x = -1/2$ and the optical transition energy increases with nuclear polarization. Here, the side-band transitions involving a spin-flip towards I_x^0 are farther from resonance than sideband transitions involving a spin-flip away from I_x^0 , which favors a flow away from I_x^0 . This leads to an “anti-dragging” behavior, where the nuclear polarization builds up in a way that avoids resonant scattering levels.

Figure 3(d) shows the measurement of these dragged and anti-dragged transitions. The laser frequency $\omega_L/2\pi$ is slowly varied under a constant $V_G = 0.93$ V, which

is in the center of the first co-tunneling region seen in Fig. 3(b). The resulting peaks marked with “D” are the dragged transitions associated with $S_x = +1/2$, showing the characteristic flat-top shape as the nuclear polarization builds up to follow the changing ω_0 . The peaks marked with “A” are the anti-dragged transitions associated with $S_x = -1/2$, identified by their more triangular (resonance-avoiding) shape and their often hysteretic behavior in the forward and backward frequency-scanning directions. This measurement unambiguously identifies that E_1 and E_3 (E_2 and E_4) are the transitions involving $|S_x\rangle = |\uparrow\rangle$ ($|\downarrow\rangle$).

With the two measurements above presented in Figs. 3(b) and 3(d), we have all the information we need to fully identify $g_\perp^{e,t}$ of the QD. From Fig. 3(b), we concluded that $s_{1,4}^{(1)} \parallel \mathbf{B}$, which implies that $\mathcal{S}(g_\perp^e) = \mathcal{S}(g_\perp^t)$, according to Eq. (4). The ordering of D and A transitions in Fig. 3(d) requires that $|g_\perp^e| < |g_\perp^t|$ and pins down the g -factor signs to $g_\perp^e > 0$ and $g_\perp^t > 0$. Together with the measured energy splittings from Fig. 3(b), we can determine $g_\perp^e = 0.08(2)$ and $g_\perp^t = 0.13(2)$. The electron g -factor is in excellent agreement with simulated $\langle g_\perp^e \rangle = 0.09$ from Fig. 2(d) at an X^- emission wavelength of 772 nm. The hole g -factor agrees with the sign and is within a factor of 2 of the simulated value of $\langle g_\perp^t \rangle = 0.27$. Strikingly, the alignment of the TDMs follows the expectation from Eq. (4) for ideal D_{2d} symmetry.

The same measurement and analysis strategy can be applied to any QD system. Figure 4 summarizes our measurement protocol, showing all four possible configurations of $g_\perp^e$ and $g_\perp^t$ which can be deduced from the measurements above (for $|g_\perp^e| < |g_\perp^t|$; the patterns for $|g_\perp^e| > |g_\perp^t|$ follow straightforwardly).

V. APPLYING THE PROTOCOL TO A BROAD RANGE OF QDS

To verify the predictive capabilities of our simulations and to assess the reproducibility of LDE GaAs QDs, we characterize the in-plane g -factors $g_\perp^{e,t}$ and the TDMs of multiple LDE GaAs QDs across a wide range of transition wavelengths and samples coming from different growth facilities (see Supplemental Material for sample details [69]) using our measurement protocol (Sec. IV). Figure 5(a) shows the mean values and standard deviations of the TDM orientation (indicated by the blurring around the polar axis) and the measured dragging patterns for different φ and QD emission wavelengths. We observe a clear flip in both TDM orientation and dragging behavior between QDs with emission wavelengths lower and higher than 775 nm, which indicates the zero-crossing of $g_\perp^e$. The TDMs follow Eq. (4) and our simulations closely—the principal polarization axes are determined by the magnetic field orientation relative to the GaAs crystallographic axes—which confirms that in all measured

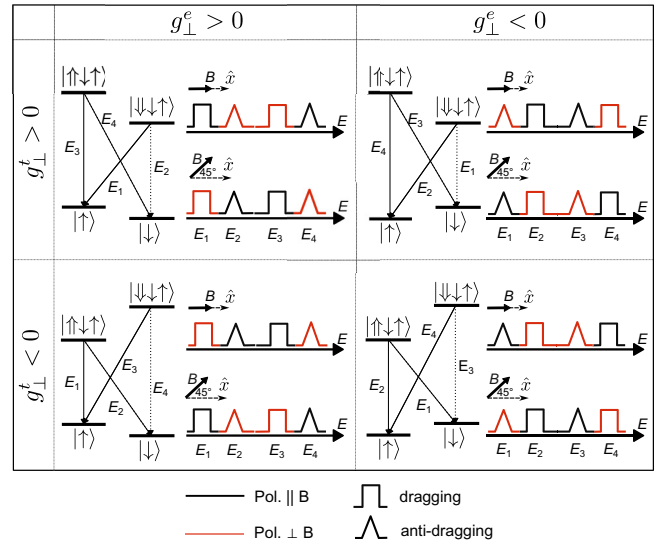


FIG. 4. Table of the four possible configurations of transition polarization and dragging patterns for $|g^t| > |g^e|$ in the Voigt configuration, which can be used to identify the signs of the g -factor elements. The spin configurations are labeling the energy levels in the basis along the magnetic field $\mathbf{B}$ to provide an intuition for the g -factor signs.

LDE GaAs QDs the D_{2d} -symmetric behavior dominates the orientation of the TDMs.

We can estimate the robustness of Eq. (4) against geometric imperfections by taking the fine structure splitting (FSS) of the neutral exciton as a measure of the in-plane anisotropy since a high FSS is an indicator for an ellipticity of the QD shape in the $\hat{x}$ - $\hat{y}$ plane [86]. The highest FSS value measured across all QDs in this work is 3 GHz (12 μ eV), which is an outlier among modern LDE GaAs QDs with typical values of $\leq 4 \mu$ eV [52,76,77]. In this outlier case, we see a deviation of the TDM orientation of about 20° away from $\mathbf{B}$, while Eq. (4) predicts the TDM to be aligned with $\mathbf{B}$ [69]. For the other QDs in this study with FSS values $\lesssim 1.2$ GHz (5 μ eV) we observe deviations of $\lesssim 10^\circ$. This relatively weak dependence on the FSS suggests the robustness of the TDMs against normal variations in QD shape and that the neutral exciton FSS can be used as a proxy for the expected correction to the ideal TDM angle.

Figure 5(b) summarizes the values of $g_\perp^e$ and $g_\perp^t$ as a function of X^- emission wavelength, with the correct signs deduced from the measured TDMs and dragging patterns. We note that the dragging patterns were measured for a subset of QDs with $g_\perp^e$ around zero, where the ambiguity of the g -factor is the highest. For QDs with wavelengths far away from the zero-crossing, it is reasonable to assume that the sign of $g_\perp^e$ is the same as that determined close to and on that side of the zero-crossing. Remarkably, most measured values of $g_\perp^e$ agree within confidence bounds with the trend predicted by our simulations (shaded blue region).

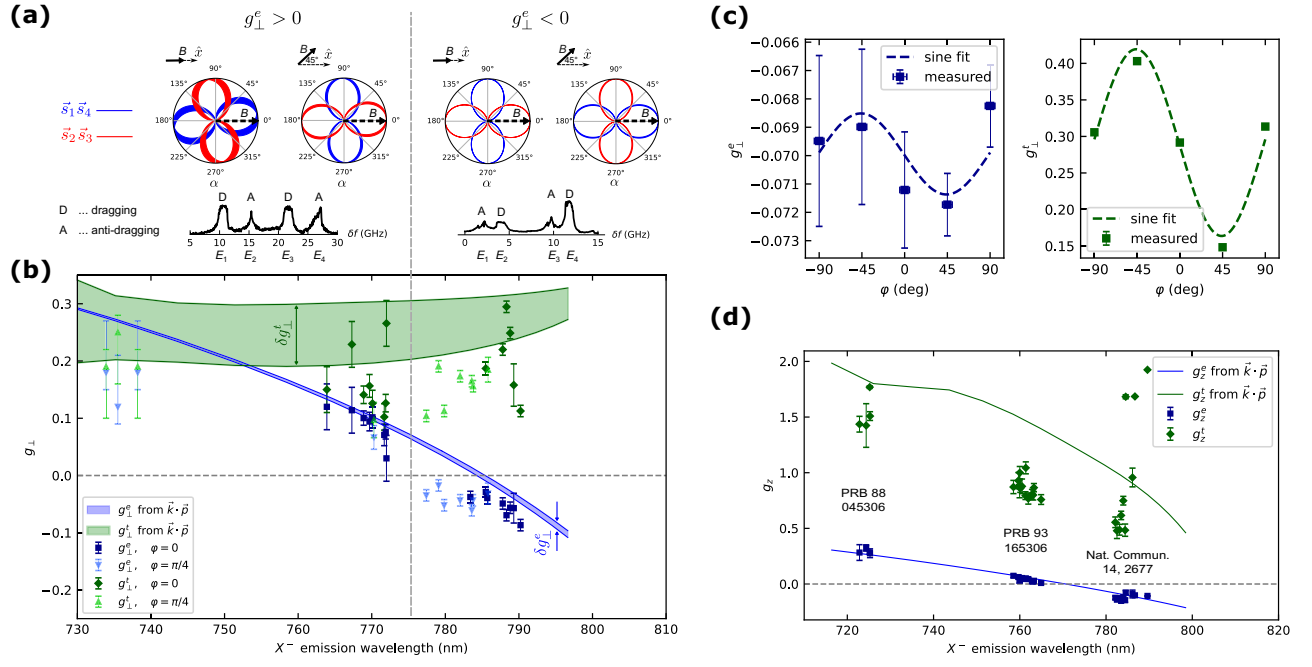


FIG. 5. (a) Orientation of the optical transition dipole moments and dragging patterns for different ϕ and X^- emission wavelengths. The polar plots show an average of all measured $s_{1,4}$ (blue) and $s_{2,3}$ (red) oriented relative to $\mathbf{B}$ (0° reference). The dragging patterns below are measured at emission wavelengths of 770 nm (left) and 785 nm (right). (b) g -factors $g_{\perp}^e$ and $g_{\perp}^t$ of different LDE GaAs QDs across different samples measured in this work as a function of X^- emission wavelength. The shaded areas show the range of values simulated via $\mathbf{k} \cdot \mathbf{p}$ across all in-plane directions, with the anisotropies indicated by $\delta g_{\perp}^e$ and $\delta g_{\perp}^t$. (c) In-plane g -factors of one QD emitting at 785 nm measured for different ϕ . (d) Compilation of Faraday g -factors g_z^e and g_z^t from literature [67,78,79] together with our numerical predictions.

For the trions, $g_{\perp}^t$ follows the prediction from the simulation of a positive value that is approximately independent of wavelength. While this value is larger by approximately 50% in our simulation compared to our measurement of $\langle g_{\perp}^t \rangle \approx 0.18$, this remains a useful predictor given the complex nature of the in-plane trion g -factor and the range of values it could have conceivably acquired (see Sec. II). The green shaded region depicts the range of simulated values obtained as a function of magnetic field angle relative to crystallographic axes and thus represents a significant anisotropy of $g_{\perp}^t$. Our measurements do not distinguish between the $[110]$ and the $[1\bar{1}0]$ (which is possible in principle, by observing patterns on the back of the unpolished wafer substrates or by keeping track of the crystal orientation during wafer processing), which may contribute to the scatter of our experimental data.

To get a better measurement of both electron and trion anisotropies we picked one QD with an emission wavelength of 785 nm and measured $g_{\perp}^e$ and $g_{\perp}^t$ for different ϕ , as shown in Fig. 5(c). We observe that for the electron (left) the anisotropy $\delta g_{\perp}^e = 0.003(2)$ is barely resolvable in our measurements, but non-negligible, as expected from our simulation in Sec. III and from previous studies [73]. The trion (right) shows a more pronounced anisotropy of $\delta g_{\perp}^t = 0.25(3)$, with the minima and maxima clearly

aligned with the $[110]$ ($\phi = 45^\circ$) and $[1\bar{1}0]$ ($\phi = 135^\circ$) axes, as expected from the symmetry of the zinc-blende crystal structure [70] and from our simulations. While the measured value of $\delta g_{\perp}^t \sim 0.25$ is higher than the 0.15 expected from our simulations, the prediction of the magnetic properties of the trion within less than a factor of 2 remains a key result.

To complete the picture, we show a compilation of $g_z^{e,t}$ values from different LDE GaAs QDs in the literature [67,78,79] in Fig. 5(d). These values were measured via the magnetic field-dependent splittings of the neutral exciton emission lines (see Ref. [67] for details) and also follow our simulations closely. The visible kink in the low-wavelength part of the theoretical curve for the trion g -factors can be attributed to inaccuracies that arise from difficulties with reaching full convergence of the many-body calculation in large-volume QDs [69], while the offset with respect to experimental points can result from the difference in methodology (trion vs exciton magnetic response) but also from a simulation offset caused by a single material parameter, κ , responsible for the isotropic part of the valence-band Zeeman term [see Supplemental Material [69] Eq. (S2)]. The Faraday response strongly depends on this parameter, and a small change in it could be used to compensate for the offset without altering the

Voigt configuration results. We decided to keep within the range of literature values [87], while noting that κ may deserve experimental revisiting.

VI. CONCLUSION

Our numerical and experimental studies on GaAs QDs obtained by local droplet etching epitaxy (LDE GaAs QDs) show that, for the electrons with spherically symmetric *s*-like Bloch functions, the *g*-tensor can be well described by the balance between the intrinsic magnetic moment and spin-correlated orbital currents (SCOCs), which are defined by the wave-function envelope [40]. We predict numerically and verify experimentally that the electron *g*-factor crosses zero at an emission wavelength of about 770 nm in the Faraday configuration (out-of-plane magnetic field) and about 780 nm in the Voigt configuration (in-plane magnetic field). Our polarization- and electron-spin-sensitive resonance fluorescence measurements over wavelengths from 730 to 790 nm remove any remaining ambiguity about *g*-factor signs, even at values very close to zero. The measured electron- and trion-*g*-factors and the associated optical transition dipole moments (TDMs) follow our predictions from the multiband $\mathbf{k} \cdot \mathbf{p}$ -configuration-interaction framework, which only uses the AFM-measured shape of one Al droplet-etched nanohole as input.

The more complex behavior of trions emerges from the *p*-like Bloch function of the heavy hole (HH) and the distinction of the growth axis in zinc-blende heterostructures. In the Voigt configuration, the Zeeman interaction vanishes for the HH, and the magnetic response and the relative spin phase of the trion wave function are dominated by higher-order corrections—likely a combination of a non-Zeeman term and LH admixture [69]. This leads to a non-trivial dependence of the orientation of the TDMs on the in-plane component of the external magnetic field [38, 39, 43]. However, high geometrical symmetry, QD material homogeneity, and low built-in strain in LDE GaAs QDs [51, 76, 77] make the TDMs predictable and tunable with magnetic field by assuming D_{2d} symmetry [39, 43], despite the intersection of the underlying zinc blende crystal symmetry and the conical shape symmetry being formally a lower C_{2v} symmetry. With the fine structure splitting (FSS) indicative of the degree of in-plane asymmetry [52], we can estimate that the D_{2d} approximation holds for QDs with an FSS at least up to about 2.5 GHz (10 μ eV), well above typical for those QDs. The reduced C_{2v} symmetry is revealed mainly as a broken in-plane symmetry in the vicinity of the GaAs-AlGaAs interface [70], which leads to the observed anisotropies of the in-plane *g*-factors [88] and to heavy hole-light hole mixing (HH-LH mixing), even in the absence of strain or geometric imperfections. We find that in unstrained LDE GaAs QDs, this HH-LH mixing is mostly confined to the region close to the GaAs/AlGaAs

interface, where the electron-hole wave function overlap is small, and thus the impact of HH-LH mixing on the optical properties is almost negligible as evidenced by 99.7% degree of circular polarization of the trion emission without the magnetic field.

In conclusion, we have demonstrated that the spin-photon interface in LDE GaAs QDs can be predicted and designed before QD growth and then realized by choosing the appropriate GaAs filling height and Al concentration in the barrier. This is made possible by the strain-free and reproducible growth mode, which yields highly in-plane symmetric and homogeneous QDs with a spread of emission wavelengths <5 nm across the wafer [51]. The *g*-tensors governing the relevant energy splittings and the TDMs can be measured without ambiguity, even for values close to zero, following the protocol introduced in this work. The well-defined dependency of TDMs and *g*-factors on the external magnetic field vector can be utilized to tune, *in-situ*, the QD polarization to match the modes of anisotropic photonic structures, such as waveguides [28], photonic crystals and birefringent cavities [11]. It also allows varying the cyclicity of the optical transitions for optimized spin readout and spin-photon entanglement [37] and the strength of the electron-nuclear coupling for optimized quantum memory protocols [73]. We believe that our findings will vastly facilitate the design and exploitation of high-quality spin-qubits in LDE GaAs QDs for highly integrated and scalable quantum technology.

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DATA AVAILABILITY

The data that support the findings of this article are openly available [89].

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