

Few-Electron Spin Qubits in Quantum Dots Probed Through Nuclear Spin Magnetism

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The knowledge of the energy spectrum completely defines the dynamics of a quantum system for a given initial state. This makes spectroscopy a key characterization technique when studying or designing qubits and complex quantum systems. In semiconductor quantum dots, the electronic quantum states can be probed through charge transport or charge sensing, but the technique is practically limited to gate-defined quantum dots. Epitaxial quantum dots benefit from excellent optical properties, but are usually incompatible with charge transport, while alternative spectroscopy techniques provide only limited information. Here we demonstrate a spectroscopy technique which utilizes nuclear spins as a non-invasive probe. By using the cotunneling-assisted transfer of spin angular momentum instead of the charge currents we achieve near-equilibrium probing. Experiments are conducted on low-strain GaAs/AlGaAs epitaxial dots, revealing energy spectra for charge configurations with up to seven electrons and with resolution limited only by the thermal broadening. Beyond the few-electron energy spectroscopy, we apply non-invasive probing via nuclear spins to reveal the subtle many-body physics of the quantized electronic states. We map out the magnetic field dependence of the electron spin lifetimes, revealing an unusual inverse quadratic dependence. The rich variety of further observations includes the long-lived spin-qubit states in the p shell, electron ground-state phase transitions, strong spin-orbit coupling regimes, and an anomalously fast nuclear spin diffusion process. Experiments are backed up by good agreement with the first-principles configuration interaction numerical modeling. Our work uncovers few-electron states as a new operating regime for optically active quantum dots. Accurate control and probing of many-body states offers a test-bed system for fundamental physics studies, while prospective technological applications include electron spin qubits with extended coherence and scalable electrical control.

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I. INTRODUCTION

Classical semiconductor devices, both digital and analogue, operate with macroscopic voltages and currents, where the discrete nature of the charges can usually be ignored. In the opposite limit, which became accessible through advances in semiconductor technology, one seeks to operate on just a single electron. Semiconductor quantum dots (QDs) are ideally suited for this role as they can trap a controllable number of individual charges in a potential well defined through morphology or electric gating. QDs are a promising platform for implementation of scalable quantum logic gates (qubits) that use the lowest-energy quantum states of individual electrons [1–3]. However, any real semiconductor system possesses a spectrum of excited quantum states that can be populated through real or virtual

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processes, resulting in complex dynamics of the qubit states. In QDs the complete Hilbert space includes the excited single-particle states as well as the multi-electron configurations. Few-electron quantum states have been a subject of intense research [4–6] both with a practical purpose of building quantum information processing devices, and in pursuit of understanding the fundamental physics of the many-body quantum interactions and correlations.

A variety of techniques have been developed to this end. In gate-defined QDs, typically operating at milli-Kelvin temperatures, the states of the few-electron configurations can be probed via electron transport [7,8] or charge sensing via quantum point contact (QPC) [9] and single electron transistors (SETs) [10]. On the other hand, epitaxial QDs can operate at higher (few-Kelvin) temperatures and offer the benefit of excellent quantum optical properties. However, charge transport or charge sensing in such structures is rather an exception, as it requires complex device fabrication [11–14], often incompatible with optical operation. Consequently, a range of alternative techniques has been applied to epitaxial QDs. Photoluminescence (PL) [15–19], resonance fluorescence [20,21] and PL excitation [22,23] spectroscopies are versatile techniques, but usually provide limited information due to the complex charging dynamics and unresolved contributions of the ground and optically excited states. Auger spectroscopy [24] circumvents many of these issues, but yields information only about single-particle states. The multi-electron states can be probed through capacitance-voltage spectroscopy [25–30], but the technique normally requires QD ensembles and therefore suffers from inhomogeneous broadening. Thus, understanding of the few-electron states in QDs remains limited.

Here we develop and implement a new approach for probing few-electron states in QDs. The method uses the spins of the crystal atom nuclei as a sensor. We measure spatial transfer of the spin angular momentum, which takes place without the actual net flow of charge carriers, but yields high resolution spectra akin to charge transport measurements. Historically, nuclear spins have been used to probe electronic states in a variety of applications, such as phase transitions in alkali fullerenes [31,32] and high temperature superconductors [33,34]. In GaAs-based structures, where all nuclear spins are non-zero, they have been used to probe ferromagnet [35] and canted antiferromagnet [36] phases as well as Skyrmion excitations [37] in two-dimensional electron systems. Due to their small magnetic moments and weak interactions, the nuclei probe the electronic states in a nearly non-invasive manner, which is difficult to achieve with optical and charge transport techniques.

We conduct our experiments on GaAs/AlGaAs QDs grown by *in situ* etching and infilling of nanoholes. This new generation of epitaxial structures is attracting

attention due to their excellent properties as quantum light emitters [38–40], electron [3] and nuclear [41] spin qubits, and quantum memories [42,43]. By measuring the nuclear spin relaxation rate as a function of the gate bias, we extract the ground state energies of different charge configurations with up to 7 electrons. Experiments reveal filling of the *s*, *p* and *d* shells in individual QDs of different sizes. The measurements are backed up by first-principles configuration interaction (CI) numerical modeling, which yields good agreement with experiments and offers insights such as prediction that small QDs can operate as spin qubits at elevated temperatures of up to ≈ 20 K. Beyond the energy state spectroscopy, our techniques reveal a wealth of nuanced information: this includes detection of the phase transition between spinless and spin-polarized four-electron ground states and observation of strong spin-orbit interaction in a five-electron configuration. Using nuclear spin sensing, we further demonstrate spin initialization, single-shot readout and long spin lifetimes exceeding ≈ 50 ms for unpaired electrons in the *s* and *p* shells. Using nuclear magnetic resonance (NMR) spectroscopy, we find a factor of ≈ 2 reduction in electron-nuclear spin interaction when going from the single-electron to three-electron configuration. These results suggest multi-electron states as a route for tailored spin coherence [44] and fast electrical control of spin-orbit qubits [45–47]. Furthermore, we find that a four-electron configuration gives rise to an unexpectedly efficient mechanism of nuclear spin diffusion, which is an important tool in hyperpolarization and signal enhancement in NMR spectroscopy [48–55].

II. RESULTS

A. Quantum dot device design and characterization

We study samples grown by *in situ* etching of nanoholes [56,57] in an AlGaAs layer, which are then infilled with GaAs to form the QDs. The structure is processed into a $p-i-n$ diode. The width of the tunnel barrier on the *n*-doped layer side (≈ 25 nm) is chosen to be much smaller than on the *p*-doped layer side (≈ 270 nm) so that an external gate bias V_G charges QDs deterministically with individual electrons, while preventing the tunneling of the holes. The band edge profiles are sketched in Fig. 1(a).

The sample is placed in a cryostat with a variable temperature $T \geq 4.2$ K. A static magnetic field B_z is applied in Faraday geometry along the growth axis *z*. Confocal microscopy configuration is used to excite and collect photoluminescence of individual QDs. PL is analyzed on a double grating spectrometer with a charge-coupled device (CCD) detector. Initial characterization of the QD charge state control is examined via bias dependent PL. The PL spectra shown in Fig. 1(b) reveal a series of excitonic features. The neutral exciton X^0 , consisting of one electron and one valence band hole, is

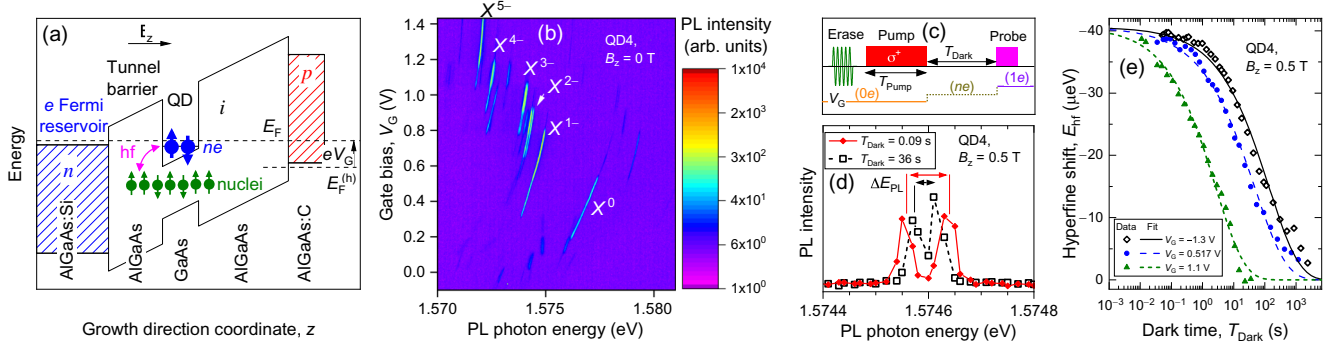


FIG. 1. Optical measurement of nuclear spin relaxation dynamics in GaAs QDs. (a) Simplified schematic of the band edge energies in a $p-i-n$ diode structure. The electron Fermi reservoir formed by a Si doped n -type layer is separated from the GaAs QDs by an AlGaAs tunnel barrier layer. The C doped p -type layer is separated by a thick AlGaAs layer which inhibits hole tunneling. The gate bias V_G is applied to shift the electron Fermi energy E_F with respect to the hole Fermi energy $E_F^{(h)}$ and tune the QD potential energy. A QD can trap an integer number n of electrons which interact with the nuclear spins of the crystal lattice via hyperfine (hf) coupling. (b) Photoluminescence (PL) spectra of a single QD excited with a ≈ 690 nm laser. Variation of the bias V_G results in PL dominated by a neutral exciton X^0 or excitons X^{n-} charged with n electrons. (c) Timing diagram of the nuclear spin relaxation (NSR) measurement. The cycle starts with a radio frequency (rf) burst to erase any remnant nuclear spin polarization, followed by nuclear spin polarization with a circularly polarized pump laser, next followed by the dark evolution delay T_{Dark} under arbitrary gate bias V_G , and completed by a short optical probe used to measure the PL spectrum. (d) Example probe PL spectra. Decay of nuclear spin polarization under increasing T_{Dark} results in reduction of the hyperfine shift E_{hf} , observed as a decrease of the X^{1-} PL doublet splitting ΔE_{PL} . (e) The changes in E_{hf} are measured as a function of T_{Dark} (symbols) under different biases V_G . Lines show stretched exponential fitting used to derive the nuclear spin half-decay times $T_{1,N}$, for which the E_{hf} reduces to 1/2 of its initial level.

identified from its fine structure splitting. The negatively charged trion X^{1-} (two electrons and one hole) is identified from spectroscopy of the forbidden transitions in an oblique magnetic field. The excitons charged with multiple electrons up to X^{5-} are identified by counting the bright PL lines appearing when V_G is increased.

A typical QD contains $N \approx 10^5$ atomic nuclei, each bearing a $3/2$ spin moment (in units of $\hbar$), coupled to the electron spins via the hyperfine (hf) magnetic interaction. The hf Hamiltonian is

$$\hat{\mathcal{H}}_{\text{hf}} = \sum_k a_k \hat{\mathbf{s}} \cdot \hat{\mathbf{I}}_k, \quad (1)$$

where a_k describes the coupling between the vector of spin operators $\hat{\mathbf{s}}$ of the resident electron and the k -th nuclear spin vector $\hat{\mathbf{I}}_k$. The hf interaction manifests in two ways. First, in addition to the bare Larmor frequency ν_N , each nucleus acquires a Knight shift $s_z a_k / (2\hbar)$, which is on the order of 100 kHz [58]. Second, the electron states with $s_z = \pm 1/2$ acquire the (Overhauser) hyperfine shifts $\pm E_{\text{hf}}/2$, resulting from the net polarization of the QD nuclear spins. The average hyperfine shift can be written as $E_{\text{hf}} = \sum_k a_k \langle \hat{I}_{z,k} \rangle$, where $\langle \dots \rangle$ denotes the expectation value.

B. Nuclear spin relaxation measurements

In order to use nuclear spin relaxation (NSR) measurements as a probe of the QD electronic state we implement an optical pump-probe experiment with a timing sequence

shown in Fig. 1(c). A radio frequency (rf) pulse is first applied to erase any remnant nuclear spin polarization [37,59] and then a reproducible nuclear spin state with polarization degree of $\approx 45\%$ is generated by a circularly polarized pump laser focused on a chosen individual QD [60–64]. A reverse bias is applied to empty the QD (0e configuration) during optical pumping of the nuclear spin. Once optical pumping is switched off, the gate bias V_G is set to an arbitrary desired level for a “dark time” T_{Dark} : this way evolution under an arbitrary QD charge state is studied for nominally identical initial nuclear spin polarizations. The cycle is completed by recording a PL spectrum under a short probe laser pulse. The changes in the splitting ΔE_{PL} of a negatively charged trion X^- , observed in the probe PL spectra [Fig. 1(d)], are used to derive the hyperfine (Overhauser) shift E_{hf} . This hf shift measures the average nuclear spin polarization degree [65] that remains in the QD following NSR during T_{Dark} . The hf shift E_{hf} is related to the total electron spin energy splitting $\Delta E_e = \mu_B g_e B_z + E_{\text{hf}}$, where the electron g -factor is negative and is typically $g_e \approx -0.1$ in the studied QDs [59]. The polarization of the pump laser is chosen to produce a negative E_{hf} , thus avoiding the electron-nuclear spin feedback at $\Delta E_e \approx 0$ where nonlinear electron-nuclear spin dynamics make PL probing difficult.

The measured dependencies of E_{hf} on T_{Dark} , shown by the symbols in Fig. 1(e), are fitted with stretched exponentials $E_{\text{hf}} = E_{\text{hf}}(0) 2^{-(T_{\text{Dark}}/T_{1,N})^\beta}$ to derive the characteristic NSR half-lifetimes $T_{1,N}$. Here, β is the stretching (compression) parameter for $\beta < 1$ ($\beta > 1$). Results shown

for three different sample biases V_G during the dark time reveal pronounced variations in the NSR dynamics. The detailed dependencies of $T_{1,N}$ and the corresponding NSR rate $\Gamma_N = 1/T_{1,N}$ are then measured systematically as a function of the gate bias V_G applied during NSR in the dark. The resulting $\Gamma_N(V_G)$ traces are shown in Fig. 2(a) for five different individual QDs and reveal a series of peaks, indicating accelerated nuclear spin relaxation at the biases comparable to those where PL switches between the different charge states [Fig. 1(b)]. We therefore ascribe the peaks in $\Gamma_N(V_G)$ to QD charging with individual electrons. While there is a considerable overlap of the charge states in the PL spectra (i.e. multiple charge states observed in PL at any given V_G), the $\Gamma_N(V_G)$ NSR dependence reveals the switching biases with high accuracy (see Fig. S6 in Supplemental Material for comparison [66]). These results demonstrate that NSR can be used as a spectroscopy tool to probe multi-electron states in QDs. Importantly, for the entire range of biases used in Fig. 2(a), the electric current through the QD semiconductor structure is negligible. Thus, NSR spectroscopy probes the few-electron states close to equilibrium.

C. Few-electron quantum dot states

In order to interpret the NSR spectroscopy data we outline the background theory, starting from a basic description of the QD charging process. For n electrons in the dot the electrostatic energy is $n(n-1)E_C/2 - neV_G/\lambda$, where $E_C = e^2/C$ is the charging energy per electron, V_G is the sample gate bias, and the “lever arm” $\lambda > 1$ is approximately the total distance between the doped layers divided by the electron tunnel barrier thickness. This energy contribution is associated with the classical capacitance C of the QD [4,67,68]. In order to account for the quantum effects we further include the confinement energies of all the single-particle electron states, whose energies $E(\nu, m)$ are indexed by the quantum numbers ν and m . We further include the energy F_n arising from the quantum correlations between single particles in the n -electron system. The total ground state energy of the n -electron configuration is then:

$$U_n = n(n-1)E_C/2 - neV_G/\lambda + \sum_{i=1}^n E(\nu_i, m_i) + F_n, \quad (2)$$

The electrochemical potential of the QD is the difference of the energies of the adjacent (n and $n-1$) ground state charge configurations:

$$\begin{aligned} \mu_n &= U_n - U_{n-1} \\ &= (n-1)E_C - eV_G/\lambda + E_n + (F_n - F_{n-1}), \end{aligned} \quad (3)$$

where E_n is the energy of the topmost filled single-particle state (ν_n, m_n) . The potentials μ_n are tuned with the bias,

and at certain V_G , where μ_n equals the Fermi energy E_F of the electron Fermi reservoir, the dot has equal probability to be occupied by $n-1$ or n electrons. Under these conditions cotunneling is enhanced, so that an electron can transition back and forth between the QD and the Fermi reservoir of the n -type doped layer, carrying the nuclear spin polarization out of the QD [69,70]. Thus a peak in the NSR rate Γ_N observed at a resonant bias $V_G = V_R^{(ne)}$ [see Fig. 2(a)] corresponds to a cotunneling resonance $\mu_n = E_F$ for a certain n . No other peaks are observed when reducing the gate bias down to $V_G < -3$ V, hence we attribute the first peak (the one observed at the lowest V_G for each QD) to resonant injection of one electron into an empty QD. The plateaus in the valleys between the peaks correspond to Coulomb blockade regimes where a QD is occupied by a stable number n of electrons (e), as labeled for QD4 in Fig. 2(a). The data shown for five individual dots QD1–QD5 reveal a clear correlation between the tunneling peaks appearing at lower biases $V_R^{(1e)}$ and a lower ground state PL energy (E_{X^0}): the reduction in both quantities is attributed to a deeper confinement of the electrons in larger QDs.

At the peaks of the rate Γ_N , the NSR is dominated by the cotunneling-assisted transfer of the nuclear spin [69,70]. Under these conditions, the NSR rate does not depend on the instantaneous degree of the nuclear polarization, resulting in a nearly exponential decay with $\beta \approx 1$ (see details in Section 4D in Supplemental Material [66]). By contrast, away from the peaks (i.e. at the Coulomb blockade plateaus) the NSR is dominated by the weaker nuclear spin diffusion mechanism [59,60]. Spin diffusion proceeds through redistribution of the initially narrow spatial distribution of the nuclear spin polarization. Such broadening of the spatial distribution slows down further spin diffusion, resulting in decay dynamics characterized by a stretched-exponential decay with $\beta \approx 0.45$. The chain-like spatial transfer of angular momentum under spin diffusion does not require any electrons, but the charge carriers can affect the diffusion rate as discussed in Sec. III C.

Resonant electron cotunneling requires that: (i) the electron reservoir can provide electrons at energy μ_n to tunnel into the QD, and (ii) the reservoir has unfilled states at the same energy which can accept the electrons tunneling back from the QD. Thus the rate of the nuclear spin transfer can be modeled as $\propto f(1-f)$, where $f = 1/(1 + e^{(\mu_n - E_F)/k_B T})$ is the Fermi-Dirac distribution of the electrons in the Fermi reservoir, and k_B is the Boltzmann constant. By modeling each peak in this way [lines in Fig. 2(a), see details in Section 2F in Supplemental Material [66]] we find a very good agreement with experiments. The energy broadening of each peak is $k_B T$, which corresponds to $w = \lambda k_B T/e$ for the width of the peak measured as a function of the gate bias V_G .

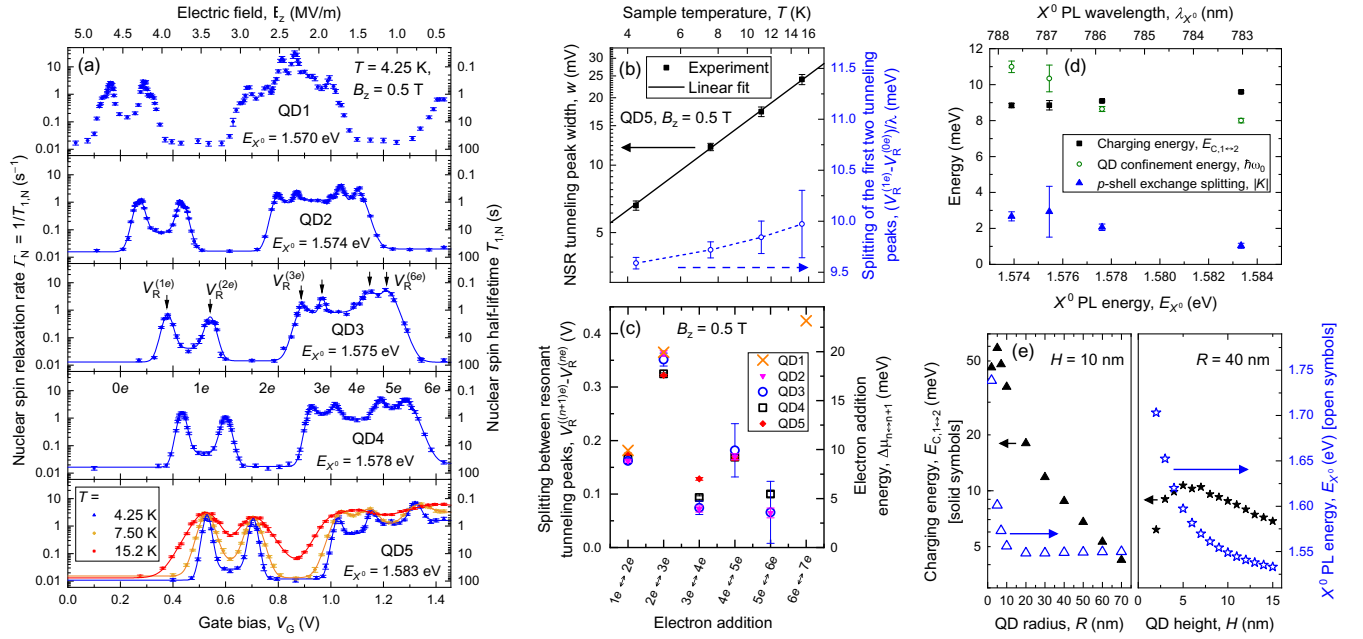


FIG. 2. Nuclear spin relaxation spectroscopy of the few-electron states in GaAs epitaxial QDs. (a) Bias dependence (symbols) of the nuclear spin half-decay time $T_{1,N}$ (right scale) and the corresponding nuclear spin relaxation rate $\Gamma_N = 1/T_{1,N}$ (left scale) measured in the dark. Results are obtained at sample temperature $T = 4.25$ K and are shown for individual dots QD1–QD5 (from top to bottom) with different ground state exciton emission energies E_{X^0} . Additional measurements on QD5 are shown for $T = 7.50$ K (diamonds) and $T = 15.2$ K (squares). Lines show model fitting. Top scale shows the calculated electric field E_z as a function of V_G . Vertical arrows highlight the resonant biases $V_R^{(n)}$ of the first six peaks ($n \leq 6$) of QD3. (b) Temperature dependence of the tunneling peak width (solid squares, left scale) and splitting of the first two peaks (open circles, right scale) for QD5. Solid line shows linear fitting of the peak widths. (c) Splittings of the adjacent peaks n and $n+1$ (left scale) and the corresponding electron addition energies $\Delta\mu_{n \leftrightarrow n+1}$ (right scale) for different quantum dots as a function of the electron number n . (d) Electron interaction energies measured for different QDs and plotted as a function of their ground state PL energy E_{X^0} . (e) QD energies calculated for a lens-shaped QD with different radii R at fixed full height $H = 10$ nm (left panel) or different H at fixed $R = 40$ nm (right panel). Solid black symbols (left scale) show the effective charging energy $E_{C,1 \leftrightarrow 2} = \Delta\mu_{1 \leftrightarrow 2}$ for adding the second electron to the dot. Open blue symbols (right scale) show the corresponding ground state PL energy E_{X^0} . All error bars in this figure are 95% confidence intervals.

For QD5 the dependencies $\Gamma_N(V_G)$ are measured at several temperatures, as shown in the bottom panel of Fig. 2(a). The dependence of the width of the first peak on the temperature T is shown by the squares in Fig. 2(b) (left scale): it is well described by a linear fit (straight line) and reveals the “lever arm” $\lambda \approx 18.3 \pm 0.6$, which we use to convert the applied gate bias V_G into resonant electrochemical potentials μ_n . Notably, λ is derived without any assumption and thus can be used to accurately estimate the geometry of the sample. In particular, knowing λ and the total thickness of the intrinsic region (≈ 300 nm) we can estimate the effective thickness of the tunnel barrier $300/18.3 \approx 16.4$ nm, which is otherwise difficult to measure since it is comparable to the height of the QD. We also find a small temperature-induced shift of the peaks, observed as repulsion of the first two peaks [circles in Fig. 2(b)]. These shifts are due to the twofold spin degeneracy, which leads to a difference in tunneling rates to and from the QD, leading in turn to an offset in the cotunneling peak positions from the exact $\mu_n = E_F$ condition [29,68,71].

D. Electron addition spectroscopy in epitaxial quantum dots

In order to characterize the few-electron interactions and correlations in epitaxial QDs, we further define the addition energies

$$\begin{aligned} \Delta\mu_{n \leftrightarrow n+1} &= \mu_{n+1} - \mu_n \\ &= E_C + (E_{n+1} - E_n) + (F_{n+1} + F_{n-1} - 2F_n), \end{aligned} \quad (4)$$

which can be derived from the separation of the adjacent cotunneling peaks in $\Gamma_N(V_G)$ dependencies [4,72] and have no explicit dependence on V_G . In the absence of quantum effects (i.e. in the classical limit of a large QD) the addition energies would all be equal to E_C . The addition energies measured at a small magnetic field $B_z = 0.5$ T are shown by the symbols in Fig. 2(c) and reveal a systematic dependence on the number of electrons already present in the QD. In order to interpret the addition energies, we use the two-dimensional parabolic-potential

model of Fock-Darwin, which is applicable since the electron confinement in the sample plane (xy) is weak compared to the out-of-plane confinement in the growth (z) direction [5]. The single-particle electron eigenenergies in the presence of magnetic field B_z are:

$$E(\nu, m) = \hbar(2\nu + |m| + 1)\sqrt{\omega_0^2 + \omega_c^2/4} + \hbar\omega_c m/2, \quad (5)$$

where $\nu = 0, 1, 2, \dots$ is the radial quantum number, $m = 0, \pm 1, \pm 2, \dots$ is the angular quantum number, $\hbar\omega_0$ is the confinement energy of the parabolic QD potential, $\omega_c = eB_z/m_e^*$ is the cyclotron angular frequency of the electron with an effective mass m_e^* (taken to be $m_e^* \approx 0.067m_e$ in GaAs [73]), and each state is twice degenerate due to the electron spin. The first electron occupies the lowest-energy state ($\nu = 0, m = 0$). When the second electron is added, it occupies the same single-particle state but with the opposite spin, forming a two-electron spin singlet $S = 0$. Thus for the first addition energy we have: $\Delta\mu_{1\leftrightarrow 2} = E_C + (F_2 + F_0 - 2F_1)$. Correlations require at least two particles, so that $F_0 = F_1 = 0$. Moreover, the two electrons occupying the same orbital ($\nu = 0, m = 0$) are restricted by the Pauli principle to have opposite spin projections. Therefore, we can interpret the F_2 term as pure Coulomb interaction and define the effective charging energy $E_{C,1\leftrightarrow 2} = \Delta\mu_{1\leftrightarrow 2} = (E_C + F_2)$. In this definition, we would like to set $B_z = 0$, so that $E_{C,1\leftrightarrow 2}$, as well as other parameters discussed below, describe the QD itself, rather than the externally applied fields. However, a NSR measurement in a QD requires a finite magnetic field of at least a few hundred mT. Luckily, the contribution of the finite cyclotron frequency $|\omega_c| > 0$ in Eq. (5) is relatively small even at $B_z = 0.5$ T. This allows us to use the low-field data, such as shown in Fig. 2(a), to estimate the charging energies at zero magnetic field. The results are plotted by the squares in Fig. 2(d), revealing $E_{C,1\leftrightarrow 2} \approx 9$ meV, consistent across all studied QDs.

Since the first two electrons in the $n = 2$ configuration form a closed s -shell singlet, the third electron has to occupy the next ($\nu = 0, m = \pm 1$) single-particle Fock-Darwin state, belonging to the higher-energy p -shell. As a result, the addition energy $\Delta\mu_{2\leftrightarrow 3}$ is considerably larger than $\Delta\mu_{1\leftrightarrow 2}$ [see Fig. 2(c)]. Neglecting the exchange interaction between the s -shell ($\nu = 0, m = 0$) and p -shell ($\nu = 0, m = \pm 1$) electrons (i.e. assuming $F_3 \approx 3F_2$), we use Eqs. (2), (3), (4) to estimate the single-particle confinement energy as $\hbar\omega_0 \approx E(0, 1) - E(0, \pm 1) \approx \Delta\mu_{2\leftrightarrow 3} - \Delta\mu_{1\leftrightarrow 2}$. The results derived from the data at $B_z = 0.5$ T are plotted in Fig. 2(d) by the circles and are within $\hbar\omega_0 \approx 9$ –11 meV, in reasonable agreement with $\hbar\omega_0 \approx 14$ meV obtained from Auger optical spectroscopy on a similar type of epitaxial GaAs QDs [24].

Noting that the separation of the fourth and fifth resonant peaks is larger than the separation of the third and fourth peaks, we follow Ref. [4] to estimate the exchange interaction energy within the p -shell as $|K| = (\Delta\mu_{4\leftrightarrow 5} - \Delta\mu_{3\leftrightarrow 4})/2 = (\mu_3 - 2\mu_4 + \mu_5)/2$. The results are shown by the triangles in Fig. 2(d), revealing exchange energies between $|K| \approx 1$ –3 meV for different individual QDs.

At $n = 6$ the p -shell originating from the ($\nu = 0, m = \pm 1$) orbitals is completely filled, so that the seventh electron has to occupy the single-particle orbital of the next d -shell (corresponding to $\nu = 1, m = 0$ and $\nu = 0, m = \pm 2$), resulting in an even larger addition energy. The onset of cotunneling of the seventh electron is observed only in QD1 [Fig. 2(a)], limited by the maximum bias V_G that can be applied to the sample. The seventh peak is indeed characterized by a large addition energy $\Delta\mu_{6\leftrightarrow 7} \gtrsim 23$ meV required to populate the d -shell. For this particular QD1, the NSR peaks exhibit additional structure, making model fitting impossible. The origin of the additional structure is not clear, but is likely related to electric field fluctuations arising from the charge traps adjacent to QD1.

This analysis shows how NSR spectroscopy can be used to quantify the key properties of the few-electron states in epitaxial QDs, including the single-particle confinement energy $\hbar\omega_0$, the charging energy $E_{C,1\leftrightarrow 2}$, and the exchange energy K .

III. APPLICATIONS OF NSR SPECTROSCOPY

Having discussed the principles and experimental examples of the nuclear spin relaxation spectroscopy, we now proceed to applications. Below we explore in more depth the physics of the different few-electron configurations. To this end, we exploit the core principle of non-invasive probing of the few-electron states via nuclear spins and develop specialized methods that provide insights into the many-body physics of each particular few-electron configuration.

A. Few-electron states in external magnetic field: experiment and configuration interaction model

We now extend NSR spectroscopy of multi-electron states to strong magnetic fields B_z . Application of external magnetic field modifies the electron state energies, especially in the p -shell, as observed from the $\Gamma_N(V_G)$ dependencies in Fig. 3(a). Figure 3(b) shows the magnetic field dependence of the resonant peak biases $V_R^{(ne)}$, which correspond to a situation where the electrochemical potential μ_n of the $n - 1 \leftrightarrow n$ transition matches the electron Fermi energy E_F . We observe a smooth increase (decrease) of $V_R^{(ne)}$ for $n = 1, 2$ ($n = 3, 4$) in the entire range of $B_z \leq 10$ T. By contrast, for $n = 5, 6$ the $V_R^{(5e)}(B_z)$

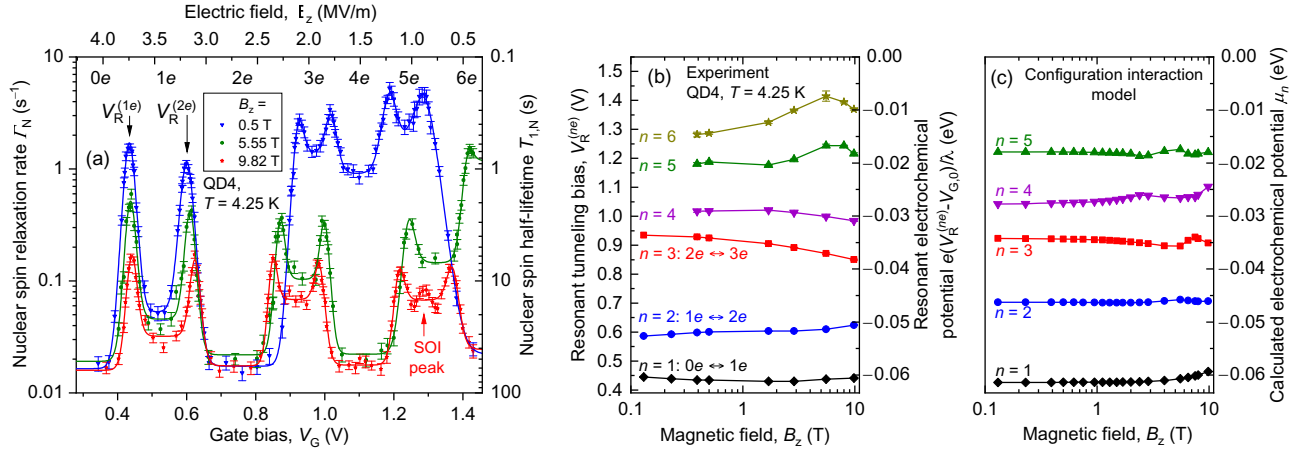


FIG. 3. Few-electron quantum dot states in external magnetic field. (a) Bias dependence (symbols) of the nuclear spin half-decay time $T_{1,N}$ (right scale) and the corresponding nuclear spin relaxation rate $\Gamma_N = 1/T_{1,N}$ (left scale) measured in QD4 at different magnetic fields. Vertical arrows highlight the resonant biases $V_R^{(ne)}$ of the first two peaks ($n = 1, 2$). Lines show model fitting. Top scale shows the calculated electric field as a function of V_G . An extra peak in the 5e valley, attributed to spin-orbit interaction (SOI), is highlighted by the arrow. (b) Resonant biases $V_R^{(ne)}$ derived from the measured positions of the cotunneling peaks in (a) and plotted as a function of magnetic field B_z . The right scale shows the corresponding QD electrochemical potentials derived as $e(V_R^{(ne)} - V_{G,0})/\lambda$, where $V_{G,0} = 1.55$ V is the gate bias corresponding to zero electric field at the QD. (c) Numerically calculated electrochemical potentials for a model QD based on a shape derived from atomic force microscopy (AFM). All error bars are 95% confidence intervals.

and $V_R^{(6e)}(B_z)$ dependencies are clearly non-monotonic. The cusps and kinks in magnetic field dependence of the resonant tunneling energies are indications of non-trivial electronic shell filling, including phase transitions [74–76], where the many-body ground state abruptly changes its angular momentum and spatial profile.

In order to explain the complex evolution of the few-electron ground state energies with magnetic field, we seek a description of the many-body electron problem that goes beyond the simple Fock-Darwin model. To this end, we model QDs with two types of geometries: either a spherical cap (lens) shape, or a shape derived from the atomic force microscopy (AFM) experiments conducted on a sample where growth was interrupted before the nanoholes were infilled with GaAs [59]. The spectrum of single-particle electron states for the chosen three-dimensional QD model is computed from the 8-band $k \cdot p$ model. The multi-electron basis states are then constructed from these single-particle basis orbitals as Slater determinants of appropriate symmetry. The multi-electron states are calculated using the CI method [77–79], taking into account electron-electron interactions and the applied electric and magnetic fields. Further details of the model can be found in Section 5 in Supplemental Material [66].

The calculated electrochemical potentials for an AFM-based QD model are shown in Fig. 3(c). The overall energy scales are well reproduced, within a few meV. The model predicts smooth dependencies of the electrochemical potentials on the external magnetic field for small electron numbers ($n = 1, 2$) with cusps and overall non-monotonic dependence for higher n , in general agreement with

experiment. However, the exact energies and positions of the cusps and kinks do not match the experiment, indicating the limitations of the model. The quantitative discrepancy is more pronounced for higher electron numbers $n \geq 3$. This is expected, since the many-body correlation effects become more pronounced with an increasing number of particles, and therefore more sensitive to the unknown nuances of the QD morphology and the single-electron spectrum. On the other hand, it is worth noting that the overall good agreement between the CI model and experimental few-electron spectra is achieved without any fitting. We thus conclude that the CI method is accurate enough to predict the key properties of the multi-electron energy spectrum where the accuracy within a few meV is sufficient.

Using the CI method, we explore the dependence of the few-electron interaction parameters for a wide range of QD morphologies that are not yet available in experiment. The effective charging energy $E_{C,1 \leftrightarrow 2}$ introduced above is an important parameter for operating the QD as an electron spin qubit, since the Coulomb blockade regime is only possible when $E_{C,1 \leftrightarrow 2} \gg k_B T$. We calculate $E_{C,1 \leftrightarrow 2}$ [solid symbols and left scale in Fig. 2(e)] together with the ground state exciton transition energy E_{X^0} [open symbols and right scale in Fig. 2(e)] for the lens-shaped (spherical cap) QDs characterized by the radius R of the cap base and the cap height H . We explore two options: a fixed $H = 10$ nm with variable R [left panel in Fig. 2(e)], or a fixed $R = 40$ nm with variable H [right panel in Fig. 2(e)]. At $R = 40$ nm and $H = 10$ nm, which corresponds approximately to the shape of the QD deduced from

AFM, the calculated charging energy is $E_{C,1\leftrightarrow 2} \approx 9$ meV, matching the experiment. When reducing R at fixed H [solid triangles in Fig. 2(e)] the charging energy increases, reaching the peak value ≈ 58 meV for a small radius of $R = 5$ nm, corresponding to a half-sphere QD shape. By contrast, the reduction of H at fixed $R = 10$ nm (solid stars) results in a pronounced blueshift of the exciton optical transition energy E_{X^0} , but has little effect on $E_{C,1\leftrightarrow 2}$. These results confirm that the magnitude of the electron-electron Coulomb interaction is dominated by the lateral confinement within the GaAs layer, whereas the optical transition energy is defined mainly by the confinement in the out-of-plane (sample growth) direction. Therefore, individual variation of the QD radius and height can be used to tune independently the optical energy and the thermal stability of the QD spin qubits. In particular, a factor of 5 increase in the charging energy (from the present ≈ 10 meV to the predicted ≈ 50 meV) may give a corresponding five-fold improvement in the QD spin qubit operating temperature from the present $T \approx 4.2$ K to $T \approx 20$ K, which would benefit deployability in applications including quantum communications and computing.

B. Phase transitions in the ground state of a four-electron configuration

Phase transitions of a multi-electron ground state were reported previously in mesa-etched and gate-defined QD structures [4,7] based on the observation of cusps and kinks in magnetic field dependence of the resonant charge carrier transport. Although such non-monotonic dependence, including cusps, is also observed in Fig. 3(b), we now demonstrate how nuclear spins can be used to probe the electronic state in a more direct way.

Rather than operating in the resonant cotunneling regime, where the QD electronic configuration is out of equilibrium, we now set the gate bias V_G either below the first resonant cotunneling peak in order to empty the QD ($n = 0$), or in the middle of the plateau between the two adjacent resonant peaks, which results in a stable number of electrons $n \geq 1$ in the QD. This Coulomb blockade regime has a significant advantage of allowing us to probe the few-electron state near equilibrium. We label the NSR rates measured in the n -electron Coulomb blockade as $\Gamma_N^{(ne)}$. The empty-QD rate $\Gamma_N^{(0e)} \approx 0.01$ s⁻¹ is dominated by nuclear spin diffusion [59]. As a result, $\Gamma_N^{(0e)}$ depends weakly on B_z , but is strongly affected by the nuclear spin pumping conditions (e.g. the duration and the power of the optical pump). Therefore $\Gamma_N^{(0e)}$ is used as a reference to plot the ratios $\Gamma_N^{(ne)}/\Gamma_N^{(0e)}$ as a function of B_z for different charge states $n = 1, 2, 4$, as shown in Fig. 4(a). The ratios $\Gamma_N^{(ne)}/\Gamma_N^{(0e)}$ are less sensitive to experimental parameters, but are sensitive to the spin state of the n -electron

configuration, which is of interest. (The absolute rates $\Gamma_N^{(ne)}$ are presented in Section 4B in Supplemental Material [66]).

We start with the simple cases of one and two electrons. The spin of a single unpaired electron [$n = 1$, circles in Fig. 4(a)] accelerates NSR ($\Gamma_N^{(1e)}/\Gamma_N^{(0e)} > 1$) in the entire available range of fields $B_z \leq 10$ T, which is explained by electron-mediated nuclear-nuclear interactions [59,69]. When two electrons ($2e$) occupy the lowest Fock-Darwin orbital ($\nu = 0, m = 0$) they form a spin singlet $S = 0$ which does not couple to the nuclear spins, and indeed we find $\Gamma_N^{(2e)}/\Gamma_N^{(0e)} \approx 1$ for the entire range $B_z \leq 10$ T.

We now focus on the four-electron ($n = 4$) configuration. While there is only a weak non-monotonic character in the $\mu_4(B_z)$ dependence, without any pronounced kinks, we find strong acceleration of NSR at low magnetic fields $B_z \leq 2$ T, with $\Gamma_N^{(4e)}/\Gamma_N^{(0e)}$ as large as ≈ 70 . By contrast, at high magnetic field $B_z \geq 5$ T we observe $\Gamma_N^{(4e)}/\Gamma_N^{(0e)} \approx 1$, indicating that the four-electron system transitions into a state akin to the two-electron spin singlet.

In order to explain this pronounced change in $\Gamma_N^{(4e)}/\Gamma_N^{(0e)}$ with magnetic field we recall that the first two electrons of a $4e$ configuration form a closed s -shell, while the remaining two electrons result in a half-filled p -shell. There are four single-particle states in the p -shell, arising from the two orbital states ($\nu = 0, m = \pm 1$), each two-fold degenerate due to spin. Thus there are six possible states of a half-filled p -shell: There are two singlet states $S = 0$ when both p -shell electrons have the same orbital quantum number m , and there are further four states spanned by a singlet $S = 0$ and a triplet $S = 1$ when the two p -shell electrons have unequal m [Fig. 4(b)]. At $B_z = 0$ the exchange interaction lifts the degeneracy, resulting in a triplet ground state, with the three singlets split off by the exchange energy K . When B_z is increased, the energies of the four-electron states evolve according to the quantum numbers m of the underlying Fock-Darwin single-particle orbitals. As a result, at some critical field $B_{z,cr}^{(4e)}$ the ($\nu = 0, m = -1$) singlet becomes the ground state as shown in the eigenenergy dependencies of Fig. 4(b), calculated according to Eq. (5). The nature of the ground state phase transition can be understood intuitively as follows. The low-field ground state is dominated by Coulomb repulsion, which forces the two p -shell electrons to occupy different orbitals ($m = -1$ and $m = +1$), resulting in a ground-state triplet [75]. At a sufficiently high magnetic field $B_z > B_{z,cr}^{(4e)}$ the energy cost of promoting an electron from the $m = -1$ to the $m = +1$ single-particle orbital exceeds the exchange energy K . As a result, it is energetically favorable for the two p -shell electrons to occupy the same $m = -1$ orbital, resulting in a ground-state singlet.

The exchange splitting can be estimated from the difference in the fourth and third addition energies [4,7]. The typical values for the studied QDs, shown by

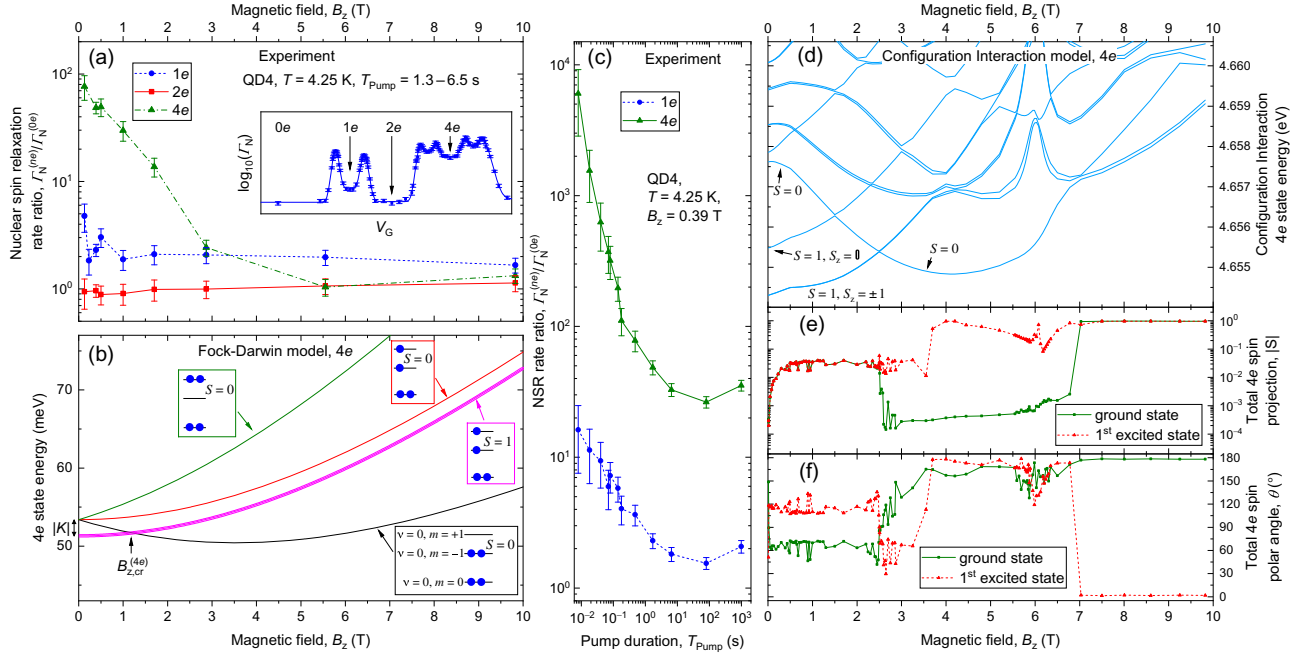


FIG. 4. Phase transitions in a four-electron quantum dot. (a) Magnetic field dependence of the NSR rates measured in a Coulomb blockade regime (see inset) where the QD is charged with n electrons. For each magnetic field the n -electron NSR rate $\Gamma_N^{(ne)}$ is normalized by the rate $\Gamma_N^{(0e)}$ measured in an empty QD. (b) Energies of the four lowest four-electron states as a function of magnetic field B_z , calculated from the Fock-Darwin model of Eq. (5), assuming exchange interaction energy $|K| \approx 2$ meV. Horizontal lines and balls depict the way the electrons fill up the single-particle orbitals for each eigenstate branch. (c) NSR rate ratios $\Gamma_N^{(ne)}/\Gamma_N^{(0e)}$ as a function of optical pumping time T_{pump} at constant $B_z = 0.39$ T. (d) Energies of the 4e states as a function of magnetic field derived from the configuration interaction (CI) numerical model. (e) Calculated total spin projection of the ground and first excited state of the 4e configuration. (f) Orientation of the spin projection calculated for the ground and first excited state of the 4e configuration, plotted as a polar angle between the spin projection and the z axis. All error bars in this figure are 95% confidence intervals.

the triangles in Fig. 2(e), are $|K| \approx 1\text{--}3$ meV, which corresponds to $B_{z,\text{cr}}^{(4e)} \approx 0.6\text{--}1.7$ T. For QD4, the estimated exchange interaction is $|K| \approx 2$ meV, which corresponds to $B_{z,\text{cr}}^{(4e)} \approx 1.2$ T. This estimate of the critical field, derived from a simple Fock-Darwin model, matches the range of magnetic fields where the measured NSR rate ratio $\Gamma_N^{(4e)}/\Gamma_N^{(0e)}$ depends strongly on B_z in QD4. Just as in case of kinks and cusps in the charge carrier transport spectra, the $\Gamma_N^{(4e)}(B_z)$ dependence gives a somewhat indirect evidence of the changes in the 4e ground state. Luckily, as we now show, nuclear spins allow for direct probing of the spin character of the few-electron ground state via NMR spectroscopy.

The NMR spectra are measured on individual QDs with the optical pump-rf-probe protocol, with the timing diagram shown in the inset of Fig. 5(a). Further details of the NMR spectroscopy techniques can be found in Section 3 in Supplemental Material [66]. By choosing the gate bias during the rf pulse, we measure NMR spectra of the QD nuclei subject to hyperfine (Knight) field of the different electronic configurations. Figure 5(a) shows NMR spectra of the ^{75}As spin-3/2 nuclei measured on a magnetic-dipole transition between the states with $I_z = \pm 1/2$ projections

of the nuclear spins. At high magnetic field ($B_z = 5.3$ T, top part of the graph) we find similarly narrow NMR spectra for an empty QD (0e) and for the 4e Coulomb blockade state. The NMR full width at half maximum of ≈ 1 kHz is limited by the second-order quadrupolar and the nuclear dipole-dipole spin interactions [41,43], indicating the absence of the Knight shifts and confirming the spin-0 phase of the four-electron ground state.

At low magnetic field [$B_z = 0.98$ T, bottom part of Fig. 5(a)] the 0e NMR spectrum remains narrow, whereas the 4e spectrum is significantly broadened, providing direct evidence of the hyperfine (Knight) field induced by the uncompensated electron spins. This low-field ground state is often referred to as polarized [4,74,80]. However, the Knight shifts observed in NMR are determined by both the ground state and the excited electronic states within the thermal energy $\propto k_B T$. This explains why the low-field 4e NMR spectrum stretches both into positive and negative frequency offsets, indicating that during the NMR rf pulse (which has a full width at half maximum of 40 μs for the 4e NMR measurement) the QD intermittently goes through the states with both positive and negative z -projections of the electron spin. The Knight shift broadening, characterized by the full width at half maximum of

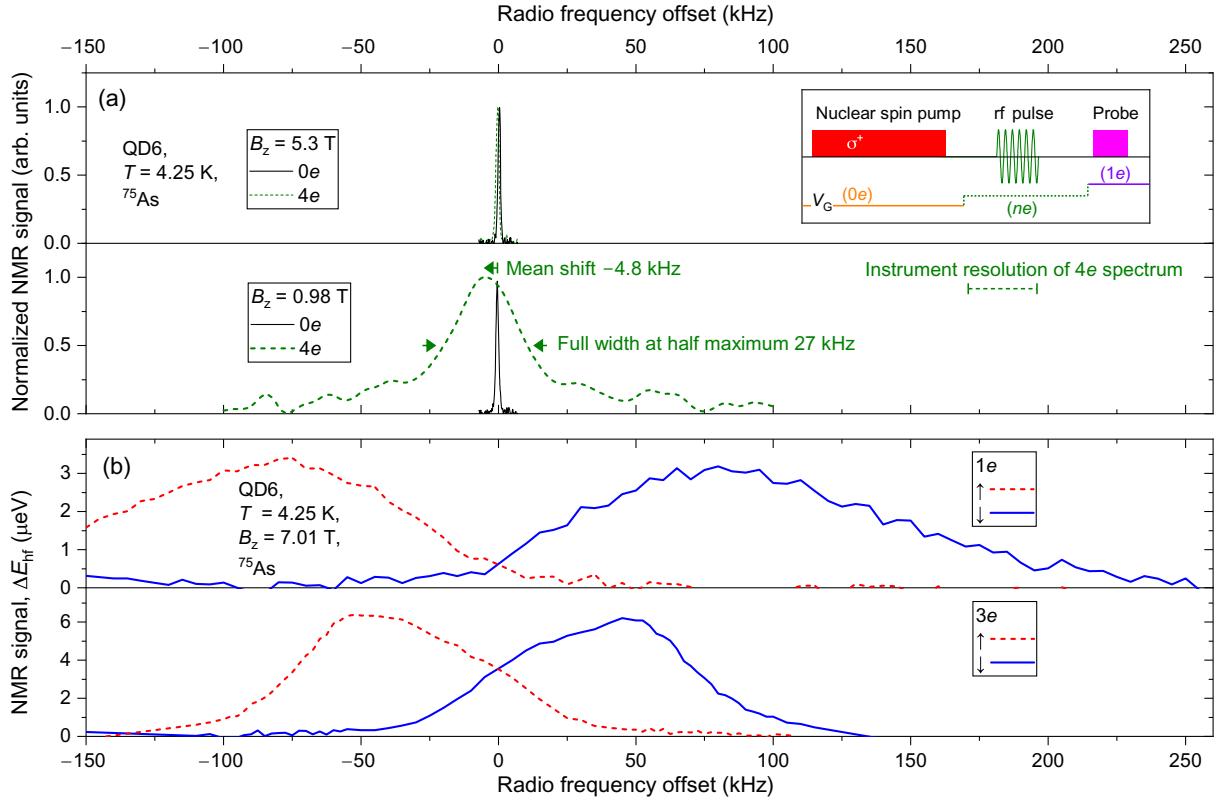


FIG. 5. Nuclear magnetic resonance spectra of individual quantum dots. (a) The inset shows the timing diagram of the NMR measurement cycle, including optical pumping of the nuclear spins, radiofrequency (rf) depolarization, optical probing, and the variation of the gate bias V_G during the cycle. The main graph shows NMR spectra of the $N \approx 10^5$ spin-3/2 ^{75}As nuclei in a single QD measured at high magnetic field ($B_z = 5.3$ T, top part) and low field ($B_z = 0.98$ T, bottom part). Solid (dashed) lines show spectra measured in an empty QD state $0e$ (QD in a $4e$ Coulomb blockade regime). The spectra are derived from Fourier transform free induction decay spectroscopy. The exception is the $4e$ spectrum at $B_z = 0.98$ T, recorded by varying the frequency offset of the rf pulse, in which case the spectral resolution is limited by the pulse duration and equals ≈ 25 kHz. See Section 3 in Supplemental Material [66] for details on the NMR techniques. (b) NMR spectra measured in a $1e$ Coulomb blockade regime (top part) and in a $3e$ Coulomb blockade regime (bottom part). These NMR spectra are measured in a single-shot regime, without averaging over multiple pump-rf-probe cycles. At finite rf offsets the two distinct values of E_{hf} are detected randomly, corresponding to spin-up (dashed lines) and spin-down (solid lines) states of the unpaired electron spin.

≈ 27 kHz, is accompanied by a ≈ -4.8 kHz mean shift of the NMR peak. The shift is a combined effect of the unequal (thermal) population of the ground and excited electronic states, and possibly the unequal electron spin projections of these states.

NMR spectroscopy gives the most direct confirmation that the $4e$ ground state undergoes a phase transition from a polarized phase (also referred to as maximum density droplet phase [74,75]) at low magnetic fields to a spin-0 phase (also referred to as Wigner molecule in the literature) at high magnetic fields. The NMR data also agree with the NSR data [Fig. 4(a)], which yields $\Gamma_N^{(4e)}/\Gamma_N^{(0e)} > 1$ in the polarized phase and $\Gamma_N^{(4e)}/\Gamma_N^{(0e)} \approx 1$ in the spin-0 phase. According to the NSR data, the critical field $B_{z,\text{cr}}^{(4e)}$ is between 3 and 5.5 T. On the other hand, the data of Fig. 4(a) show that below the critical field the NSR rate

$\Gamma_N^{(4e)}$ is not constant. Instead, $\Gamma_N^{(4e)}$ depends strongly on magnetic field B_z , suggesting that the four-electron ground state evolves with B_z even within the same (polarized) phase. We next discuss this strong dependence with the aid of CI model calculations.

C. Anomously fast nuclear spin relaxation and diffusion in the four-electron configuration

In order to explain fast NSR in the $4e$ configuration we first discuss the mechanisms of NSR. Nuclear spin diffusion is a process where the localized optically pumped nuclear spin polarization in a QD is transferred into the surrounding barriers through spin-exchange flip-flops between the adjacent nuclear spins. Spin diffusion happens both in the dark and under optical pumping of nuclear spins. Under long pumping, spin diffusion polarizes the

nuclei not only in the QD, but also in the surrounding material. As a result, subsequent spin diffusion in the dark (after optical pumping is switched off) is suppressed. Thus, the duration of optical pumping can be used as a tool to distinguish spin-diffusion and non-spin-diffusion NSR mechanisms. Figure 4(c) shows NSR rate ratios $\Gamma_N^{(ne)}/\Gamma_N^{(0e)}$ measured as a function of optical pumping time T_{Pump} for $1e$ ($n = 1$) and $4e$ ($n = 4$) Coulomb blockade charge configurations. Spin diffusion in the $1e$ configuration [circles in Fig. 4(c)] has been studied previously [59], so we discuss this effect only briefly. Acceleration of spin diffusion in the $1e$ configuration occurs because the electron spin can mediate spin exchange between distant nuclear spins, as opposed to direct dipole-dipole interaction that couples only the nearby nuclei. According to Fig. 4(c), in the limit of long pumping the electron has only a small effect on the NSR, characterized by $\Gamma_N^{(1e)}/\Gamma_N^{(0e)} \lesssim 2$. For short pumping, this increases to $\Gamma_N^{(1e)}/\Gamma_N^{(0e)} \gtrsim 15$. Thus the effect of the spin of a single electron on NSR is almost entirely through acceleration of nuclear spin diffusion.

The results for the spin-polarized $4e$ configuration (measured at $B_z = 0.39$ T, which is below the critical field) are quite different and more complicated. For the longest available pumping time [$T_{\text{Pump}} \approx 1000$ s in Fig. 4(c)] NSR in the $4e$ configuration is a factor of $\Gamma_N^{(4e)}/\Gamma_N^{(0e)} \approx 30$ faster than in an empty QD ($0e$). When the optical pumping is shortened ($T_{\text{Pump}} \approx 0.008$ s is the shortest available in experiment), NSR becomes even faster, characterized by $\Gamma_N^{(4e)}/\Gamma_N^{(0e)} \gtrsim 6000$. These results show that there are multiple NSR mechanisms induced by the $4e$ configuration: both the non-diffusion NSR (more pronounced at long T_{Pump}) as well as acceleration of spin diffusion (responsible for fast NSR at short T_{Pump}). In order to unravel these mechanisms we calculate the spectrum of the $4e$ eigenstates using the CI model, which reveals the detailed structure of the few-electron states that is not accessible in the simple Fock-Darwin model.

The CI spectrum of the $4e$ eigenenergies as a function of the external magnetic field B_z is shown in Fig. 4(d). For each eigenstate with wavefunction ψ we calculate the magnitude of the total electron spin projection $|S| = \sqrt{\langle \psi | \hat{S}_x | \psi \rangle^2 + \langle \psi | \hat{S}_y | \psi \rangle^2 + \langle \psi | \hat{S}_z | \psi \rangle^2}$, shown in Fig. 4(e). We also define the polar angle of the total spin quantization axis $\theta = \arctan \left(\sqrt{\langle \psi | \hat{S}_x | \psi \rangle^2 + \langle \psi | \hat{S}_y | \psi \rangle^2} / \langle \psi | \hat{S}_z | \psi \rangle \right)$, shown in Fig. 4(f). Further details can be found in Section 5C in Supplemental Material [66]. The lowest energy manifold predicted by the CI is a doublet of states originating from the triplet ($S = 1$) states with z -projections $S_z = \pm 1$. Exchange interaction and electron-electron correlations in the anisotropic quantum dot potential shift the $S_z = 0$ projection state to higher energies and further

mix the $S_z = \pm 1$ states. The resulting eigenstates are characterized by reduced but non-zero spin projections $|S| \approx 3 \times 10^{-2}$. Moreover the spins of these two ground states are quantized along the axis that is nearly orthogonal to the external magnetic field ($\theta \approx 70^\circ$ and $\theta \approx 110^\circ$). Such $4e$ electronic states are very different both from the $2e$ configuration, where the spin of the singlet state is very small $|S| < 10^{-4}$, and the $3e$ and $5e$ configuration where the spin predicted by CI is close to $|S| \approx 1/2$ and is quantized approximately along the external magnetic field z axis (see details in Section 5C in Supplemental Material [66]). We ascribe the anomalously fast NSR to this peculiar spin character of the $4e$ ground state doublet, which arises from the $S = 1$ triplet. Next, we discuss how the structure of the $4e$ spectrum affects the non-diffusion and spin diffusion mechanisms.

We start with the non-diffusion mechanism. The smaller spin projections $|S|$ of the $4e$ polarized states predicted in CI are corroborated by the NMR spectra, which show considerably smaller hyperfine shifts (narrower NMR spectrum) experienced by the nuclei in the $4e$ configuration, when compared to the $1e$ and $3e$ configurations (Fig. 5). Although $|S|$ is small, the deviation of the $4e$ electron spin quantization axis from the external magnetic field is what leads to the non-diffusion contribution toward the anomalously fast NSR. In order to understand the NSR mechanism, we first note that the nuclear spins are quantized essentially along the external magnetic field (due to the relatively small hyperfine interaction and small electric quadrupolar effects in lattice-matched GaAs/AlGaAs structures). If the electron spin is also quantized along the external field, as is the case with good accuracy for the $1e$, $3e$, and $5e$ ground states, then the energy mismatch between the electron and nuclear spin Zeeman splittings precludes electron-nuclear spin flip-flops. In this “collinear” case, the strong magnetic field effectively truncates the hyperfine Hamiltonian (Eq. (1)) leaving only the $\propto \hat{s}_z \hat{I}_{z,k}$ terms, under which the electron can change the nuclear spin splitting (via Knight shift) but cannot flip the nuclear spin from one $\hat{I}_z$ eigenstate to the other. The situation is radically different if the electron spin is quantized along a tilted axis, as is the case for the $4e$ ground state according to the CI model calculations. In this “noncollinear” case the electron spin can cause nuclear spin precession. If the electron spin undergoes spin jumps, it generates an effective stochastic transverse magnetic field that acts on the nuclei, leading to NSR. Indeed, from NMR measurements (see Sec. III E), the electron spin lifetime is found to be shorter than the duration of the $40 \mu\text{s}$ rf pulse, suggesting that the jumps in the $4e$ configuration occur on a timescale of tens of μs , or shorter. Such telegraph-noise-like [58] random magnetic field contains a sufficiently strong spectral component that is resonant with the nuclear Larmor frequency and enables coupling of the nuclear spins to the lattice phonons (via the

electron spin) resulting in NSR without nuclear spin diffusion and without cotunneling. The measured non-diffusion NSR rate driven by the $4e$ configuration is on the order of $\Gamma_N^{(4e)} \approx 0.12 \text{ s}^{-1}$ [at $B_z = 0.39 \text{ T}$, $T_{\text{pump}} \approx 1000 \text{ s}$, see Fig. S5(c) in Supplemental Material [66]], which is much faster than the NSR rate $\Gamma_N^{(0e)} \approx 0.0033 \text{ s}^{-1}$ in an empty ($0e$) QD under the same conditions. The measured Γ_N agrees with results of numerical modeling on a simple system of one nucleus coupled to the electron spin. This simple model also confirms that the relaxation mechanism requires both the non-collinearity (i.e. the tilt of the electron quantization axis) and the fast electron spin jumps. We note a similarity with noncollinear hyperfine interaction in Stranski-Krastanov QDs [69,81], except that the roles of the nuclei and electrons are reversed there: the large strain-induced quadrupolar effects tilt the effective quantization for the nuclei in Stranski-Krastanov QDs [82], while the electron is quantized approximately along the magnetic field.

We now turn to the spin-diffusion component of the anomalously fast NSR induced by the polarized $4e$ configuration. For magnetic fields exceeding tens of mT, the direct electron-nuclear spin flip-flops are suppressed by the energy mismatch. However, the electron spin can mediate a flip-flop of two nuclear spins. For two nuclei j and k with hyperfine constants a_j, a_k the effective flip-flop coupling can be derived as a second-order perturbation [55,83] and scales as $\propto a_j a_k / \Delta E_e$, where ΔE_e is the energy splitting of the electron spin levels. This interaction plays an important role in nuclear spin diffusion [59,84–86], since it allows spin transfer between the distant nuclei, whose direct dipole-dipole coupling is otherwise negligible. Electron-mediated spin diffusion can be derived for various configurations, including a two-electron two-nucleus system as demonstrated recently [55]. However, such derivations are complex, meaning that rigorous treatment of spin-diffusion in a $4e$ configuration is outside the scope of this work. Instead, we gain insights from numerical modeling of a simple system of two nuclei quantized along the z axis and coupled via hyperfine interaction [Eq. (1)] to a common electron whose quantization axis is tilted away from the z axis. Unlike with direct (non-diffusion) NSR discussed above, we find that the tilt of the electron quantization axis has no significant impact on spin-diffusion NSR. By contrast, the short electron spin lifetime of the $4e$ configuration plays an important role in allowing the flip-flops between the two nuclei even if they have slightly different effective Zeeman energies (non-resonant nuclei). The fluctuating hyperfine (Knight) field of a rapidly flipping electron spin effectively broadens the nuclear spin energy levels. Such broadening bridges the energy gap of the nuclei that are otherwise made non-resonant by the inhomogeneous strain-induced quadrupolar shifts and chemical shifts. Furthermore, the small spin projection $|S|$ of the $4e$ configuration reduces

the ΔE_e denominator, while the large spatial extent of the $4e$ wavefunction provides electron-mediated coupling for a larger number of nuclear spins. This interpretation agrees with our experimental results, which reveal very pronounced acceleration of nuclear spin diffusion in the $4e$ configuration [at least a factor of ≈ 300 faster than spin diffusion driven by a single electron in the $1e$ configuration, see Fig. 4(c)]. Interestingly, the measured absolute NSR rates in the $4e$ diffusion regime ($\Gamma_N^{(4e)} \gtrsim 270 \text{ s}^{-1}$ at $B_z = 0.39 \text{ T}$, $T_{\text{pump}} \approx 0.008 \text{ s}$, see Fig. S5(c) in Supplemental Material [66]) are comparable to the NSR rates at zero magnetic field $\Gamma_N \approx 10^3 \text{ s}^{-1}$, where the dipole-dipole interactions do not conserve spin and NSR is very fast.

In summary, the CI model provides an accurate qualitative description of the subtle many-body phenomena such as reduced spin projection in the $4e$ triplet state, and the resulting anomalously fast NSR and nuclear spin diffusion. Spin diffusion is an important tool in NMR signal enhancement and structural analysis [48–54]. Experimental results on the $4e$ triplet give an example of how spin diffusion can be engineered through control of the few-electron configuration. On the other hand, the CI numerical model is inevitably limited by a variety of factors, including the accuracy of the material parameters and approximate knowledge of QD morphology. This leads to quantitative discrepancies, such as prediction of the second phase transition of the $4e$ ground state at $B_z \approx 7 \text{ T}$, which is not observed in experiments up to $B_z \approx 10 \text{ T}$. Further work would be needed to examine in more detail the mechanisms of NSR and spin diffusion in presence of few-electron configurations.

D. Spin orbit interaction in the five-electron configuration

Experimental data in Fig. 3(a) exhibit an additional peak in the NSR rate in the middle of the $5e$ Coulomb blockade valley (at $V_G \approx 1.3 \text{ V}$) but only at the strongest magnetic field $B_z = 9.82 \text{ T}$ available in this work. Such extra NSR rate peak has been predicted previously to arise from the spin-orbit interaction (SOI), but to our knowledge has not been observed experimentally. In the previous theoretical work, conducted in the context of the gate-defined QDs [87,88], the appearance of an extra peak was classified as a weak SOI regime. In the present study on epitaxial QDs, this regime turns out to be the strongest achievable SOI because no other additional peaks are observed in the middle of the Coulomb blockade plateaus. Observation of the peak in the $5e$ plateau and its absence for other configurations with an unpaired electron spin ($1e$ and $3e$) agree with the general expectation that the spin orbit interactions are enhanced by the many-body effects. In the same manner, the expected increase of the spin orbit effects with magnetic field [89,90] explains why the extra

NSR peak is seen only at the highest B_z and already becomes indistinct when the field is reduced to $B_z = 8$ T (see additional data in Section 4 A in Supplemental Material [66]). Experimentally, the additional peak is observed only at one value of the external field, hence our attribution of this peak to SOI is tentative. Further experimental studies would be needed and would benefit from stronger magnetic fields, lower temperatures, and higher electron addition energies, which would facilitate observation of the peculiar peak-in-the-plateau structure.

E. Few-electron spin qubits and electron spin lifetimes

The few-electron configurations, observed in this work via nuclear spin relaxation spectroscopy, offer a variety of electronic states that can be used as spin qubits. We demonstrate this by performing initialization and readout of the resident electron spin in different shells. The timing diagram of the relevant experiment is shown in Fig. 6(a). In addition to optical nuclear spin pumping, a second optical pulse (typically 100 μ s long) is applied optionally in order to polarize the electron spin after the QD is loaded with n electrons. Adapting the techniques developed recently on InGaAs QDs [91], we use nuclear spins for single-shot readout of the electron spin [58]. An electron spin up $\uparrow$ (spin down $\downarrow$) shifts the NMR to lower (higher) frequencies via hyperfine (Knight) shift. Thus, an rf π pulse detuned to lower frequencies rotates (does not rotate) the nuclear spins if the electron is in the $\uparrow$ ($\downarrow$) state. Following the detuned rf pulse, which performs quantum non-demolition measurement of the electron spin s_z projection, the nuclear hyperfine shift E_{hf} is probed via photoluminescence spectroscopy. Importantly, the measurement is conducted in a single-shot mode, without averaging over multiple pump-rf-probe cycles. A large variation ΔE_{hf} of the hyperfine shift E_{hf} (a small variation of E_{hf}) produced by the detuned rf pulse heralds the detection of a $\uparrow$ ($\downarrow$) electron spin state. In the experiment, a large number of pump-rf-probe cycles is measured. The resulting individual single-shot NMR signals are then plotted as histograms in Figs. 6(b) and 6(c), confirming that ΔE_{hf} indeed takes on two distinct values, observed as a bimodal statistical distribution. This method of electron spin readout has been applied recently to the s -shell electron spin qubit in the $1e$ configuration [58]. Here we apply this technique to the $3e$ configuration by choosing the gate bias V_G during the rf pulse to be at the center of the $3e$ Coulomb blockade plateau. The results are shown in Figs. 6(c) and 6(e). The two distinct modes in the histogram of the single-shot NMR signals are attributed to the spin up ($\uparrow$) and spin down ($\downarrow$) states of the unpaired p -shell electron, demonstrating that high-fidelity single-shot readout can be performed on the qubit states of the few-electron configurations.

When the electron spin pumping pulse is omitted, the $\uparrow$ and $\downarrow$ readouts have nearly the same probabilities, as

demonstrated in the histograms of Figs. 6(b) and 6(c) measured with rf detunings optimized for the spin readout in the $1e$ and $3e$ configurations, respectively. By activating the electron spin optical pumping pulse and tuning its wavelength we are able to prepare a non-equilibrium electronic state either with a positive or a negative spin polarization. This is shown in Figs. 6(d) and 6(e) (for $1e$ and $3e$ configurations, respectively), where one finds a pronounced difference in the probabilities to detect the two possible ΔE_{hf} values. For simplicity we use the same photon energy (typically tuned ≈ 10 meV above the ground state trion recombination energy) both for the nuclear and electron spin pumping. Such quasiresonant optical pumping limits the electron spin polarization degrees to $|P_e| \lesssim 0.8$, but is sufficient for a proof-of-principle demonstration that s and p shell electrons can be initialized optically. Further improvement in electron spin preparation fidelity can be achieved using the well-known resonant optical spin pumping techniques [92].

Next we apply the single-shot electron spin readout protocol to perform NMR spectroscopy and probe the electron-nuclear interactions. To this end we measure the histograms of the single shot hyperfine shifts ΔE_{hf} , such as shown in Figs. 6(b)–6(e), but at variable frequency detunings of the rf pulse. In these experiments the electron spin pumping pulse is omitted, so that the histograms include the contributions from both electron spin projection states $\uparrow$ and $\downarrow$. For each rf frequency offset we find the centers of gravity of the two histogram modes and plot them as shown in Fig. 5(b). The traces obtained from the two histogram modes correspond to the two branches of the NMR spectra, measured selectively for the $\uparrow$ (dashed lines) and $\downarrow$ (solid lines) states of the unpaired electron spin. The method is applicable both to the $1e$ [top part of Fig. 5(b)] and $3e$ [bottom part of Fig. 5(b)] charge configurations. The NMR spectra of Fig. 5(b) show that the average Knight shifts produced by an unpaired p -shell electron of the $3e$ configuration are a factor of ≈ 2 smaller than those of the s -shell electron in the $1e$ configuration. This observation is consistent with the expected larger spatial extent of the envelope wavefunction of the more delocalized p -shell electron. The smaller Knight shift indicates that the p -shell electron envelope wavefunction is spread over a ≈ 2 times larger number N of the nuclei of the crystal lattice. An even more pronounced increase in N would be expected for higher electron numbers n . Since the electron spin qubit dephasing time scales as $N^{-1/2}$ the use of the higher-shell electrons as spin qubits [44] can be a route to an improved qubit coherence.

Employing the spin preparation and readout techniques we vary the delay T_{Dark} between the electron spin optical pump and the rf spin detection pulse to measure the decay of the electron spin polarization. The results are averaged over many pump-delay-rf-probe cycles and are shown by the symbols in Fig. 6(f) revealing a gradual decay of the

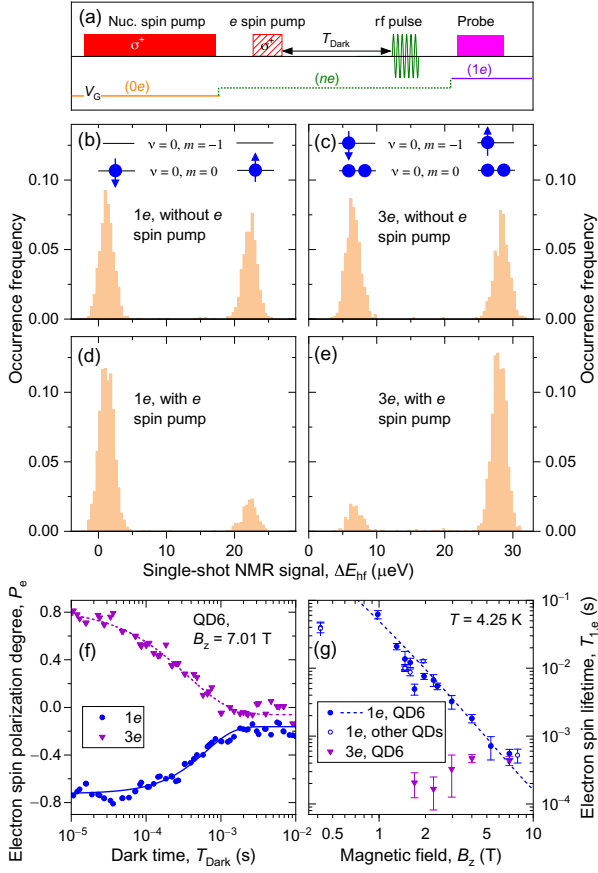


FIG. 6. Quantum dot few-electron spin qubits. (a) Timing diagram of the experiment to probe electron and nuclear spins. The cycle starts with a quasiresonant circularly polarized laser pulse to polarize the nuclear spins in an empty ($0e$) QD. After the bias is changed to load QD with n electrons, an optional second pulse of the same laser can be applied to polarize the unpaired electron spin. Following the dark evolution delay T_{Dark} , a detuned rf π rotation pulse is applied to convert electron spin into a nuclear spin hyperfine shift E_{hf} , which is then read out by measuring the X^1- PL spectrum under a nonresonant optical probe [58,91]. (b) Histogram of the hyperfine shift variation values ΔE_{hf} measured at an optimal rf offset in $1e$ charge configuration without electron spin pumping. Two distinct histogram modes correspond to spin-up ($\uparrow$) and spin-down ($\downarrow$) states of an unpaired s -shell electron (orbital state ($\nu = 0, m = 0$)), depicted with balls and arrows. (c) Same as (b) but measured in a $3e$ charge configuration with the two histogram modes corresponding to spin-up and spin-down states of an unpaired p -shell electron (orbital state ($\nu = 0, m = -1$)). (d) Same as (b) but including the electron spin pumping pulse. The delay time is $T_{\text{Dark}} \approx 10 \mu\text{s}$. The balance of peaks reveals predominant population of the spin-down state. (e) same as (c) but with electron spin pumping. Here, the wavelength is tuned to populate preferentially the spin-up state of the unpaired p -shell electron. (f) Decay of the s -shell (circles) and p -shell (triangles) electron spin polarization measured at $B_z = 7.01 \text{ T}$ by varying T_{Dark} . Lines show compressed exponential fitting yielding the characteristic $1/e$ spin lifetimes $T_{1,e}$. (g) Magnetic field dependence of $T_{1,e}$. Error bars are 95% confidence intervals.

optically pumped electron spin polarization. Using exponential fitting (lines) we derive the characteristic ($1/e$) electron spin lifetimes $T_{1,e}$, which are plotted in Fig. 6(g) as a function of magnetic field B_z for 6 different QDs with neutral exciton energies between $E_{X^0} = 1.590$ – 1.606 eV . Electron spin relaxation is known to be dominated by the phonon-related mechanism, and in the high-temperature regime $\Delta E_e \gg k_B T$ it leads to the following electron spin lifetime scaling [90,93,94]:

$$T_{1,e} \propto (k_B T)^{-1} B_z^{-2} \Delta E_e^{-2} \quad (6)$$

For InGaAs QDs, Zeeman splitting dominates over the nuclear hyperfine (Overhauser) field $|\mu_B g_e B_z| \gg |E_{\text{hf}}|$ in a wide range of magnetic fields. As a result, the total electron spin splitting is dominated by the Zeeman effect $\Delta E_e \propto B_z$. This leads to the well-known $T_{1,e} \propto B_z^{-4}$ (high temperature regime [70]) or $T_{1,e} \propto B_z^{-5}$ dependence (low temperature regime [94,95]). By contrast, in the GaAs epitaxial QDs studied here the electron g -factor is small [59,96] ($g_e \approx -0.04$ in QD6). With the typical $E_{\text{hf}} \approx -50 \mu\text{eV}$ in our electron spin relaxation experiments, the hyperfine shift dominates over the Zeeman effect ($|E_{\text{hf}}| > |\mu_B g_e B_z|$) in the entire experimental range of magnetic fields $B_z \lesssim 10 \text{ T}$. This results in an unusual field dependence close to $T_{1,e} \propto B_z^{-2}$. Using a factor of proportionality as the only fitting parameter, we plot Eq. (6) by the dashed line in Fig. 6(g) to find good agreement with the experiment on the s -shell electron spin ($1e$). At low magnetic fields $B_z \lesssim 1 \text{ T}$, the measured lifetimes saturate at $T_{1,e} \approx 50 \text{ ms}$, indicating that the field-independent cotunneling-related mechanism [70] of electron spin relaxation becomes dominant. The rate of the cotunneling-related electron spin relaxation is controlled by the thickness of the tunnel barrier between the QD and the electron Fermi reservoir and is therefore specific to the studied semiconductor device. By contrast the phonon-related electron spin relaxation rate has a nearly universal dependence on electron spin energy splitting, temperature, and a rather weak dependence on the size of the QD. Therefore, the $T_{1,e}$ relaxation times shown in Fig. 6(g) for the $1e$ configuration above $B_z \gtrsim 1 \text{ T}$ can be used as a practical calibration curve: the spin lifetime $T_{1,e}$ is an important parameter since it sets an upper limit for the electron spin coherence time $T_{2,e}$. Our results show that for the s -shell electrons in GaAs/AlGaAs QDs the lifetimes range from $T_{1,e} \approx 50 \text{ ms}$ to $\approx 0.5 \text{ ms}$ when magnetic field is changed between $B_z \gtrsim 1 \text{ T}$ and 8 T . Even longer $T_{1,e}$ can be achieved in principle at low magnetic fields by increasing the thickness of the tunnel barrier, although this comes at the expense of a more prominent Auger recombination, with some unwanted consequences for optical manipulation of the electron spin qubit [70].

The spin lifetimes of the unpaired p -shell electron in the $3e$ configuration [triangles in Fig. 6(g)] show a rather different trend with a moderate increase in $T_{1,e}$ with the applied magnetic field. The relaxation of the p -shell electron is likely dominated by the field-independent cotunneling coupling to the electron Fermi reservoir in the entire range of the studied magnetic fields $B_z < 8$ T. This explanation is consistent with the smaller width of the $3e$ Coulomb blockade valley [Fig. 3(a)], which becomes even narrower at low magnetic fields, enhancing both the NSR and the electron spin decay. We expect that the p -shell electron spin lifetimes can be increased significantly by effectively widening the $3e$ Coulomb blockade plateau. This in turn can be achieved by a moderate reduction of the base temperature (from $T = 4.25$ K to $T \approx 2$ K) or by reducing the lateral size of the QD in order to increase the electron charging energy [see Fig. 2(e)]. With these improvements, a more in-depth investigation of the few-electron spin lifetimes should be possible, opening the way to full coherent control and investigation of spin coherence times in the few-electron spin qubits in epitaxial QDs.

We also conduct single-shot NMR experiments in the spin-polarized (low-field) phase of the $4e$ configuration. NMR is measured at $B_z = 0.98$ T with $T_{\text{rf}} = 80$ μ s rf pulses (full width at half maximum is 40 μ s) but, unlike for $1e$ and $3e$ configurations, no bimodal distribution can be observed in the histogram of the single-shot NMR signals. Such lack of bimodality can be easily explained by assuming that the electron spin lifetime of the $4e$ polarized state is shorter than 40 μ s. In that case the electron spin polarization goes through the opposite states during nearly each rf pulse, so that the NMR measures the time-averaged Knight shifts rather than the eigenstate values. While the rf pulse can be made much shorter than 40 μ s, this leads to spectral broadening of the rf pulse in excess of the Knight shift splitting, which is small due to the small spin projections of the $4e$ polarized states. As a result the opposite $4e$ spin states cannot be resolved in NMR even with short rf pulses. However, NMR spectroscopy is sufficient to conclude that the spin lifetimes of the polarized $4e$ phase states are very short, at least two orders of magnitude shorter than in the $1e$ spin qubit. The short $T_{1,e}$ qualitatively agrees with the CI calculations, which indicate strong mixing of the spin and orbital degrees of freedom as well as tilting of the quantization axis of the $4e$ ground states with respect to the external magnetic field. Further investigation of the fast $4e$ electron spin dynamics can be attempted using optical techniques for spin pumping and detection [70,92,95] and would shed more light on the nature of the $4e$ states and the physics of the ground state phase transitions.

IV. DISCUSSION AND OUTLOOK

The results presented in this work demonstrate the rich variety of the few-electron states that can be constructed in

optically active epitaxial GaAs QDs. Such n -electron states may be used to build qubits with favorable properties. The large spin-orbit effects observed with the increasing number of the electrons in a QD offer a route to spin-orbit qubits [45,46] where coherent control can be implemented with oscillating electric fields, to achieve scalability and high speeds [47] that are not attainable with oscillating magnetic fields [97]. Such an electric dipole spin resonance (EDSR) control, previously achieved only in the milli-Kelvin regime [45,46,98], could thus be adapted to epitaxial QDs where higher operating temperatures (few-Kelvin) and efficient spin-optical interfaces offer advantages for implementation of quantum devices.

Bias-dependent nuclear spin relaxation (NSR) spectroscopy demonstrated here is a versatile probe of both the energy spectrum and the spin properties of the few-electron states. The technique does not rely on electron transport or charge sensing, which are usually unavailable in the optically active semiconductor nanostructures, and can thus be extended to a wide range of semiconductor nanostructures. The absence of electric currents is itself an advantage, as it permits probing of the few-electron quantum states near equilibrium. The measurements presented here have been taken with waiting times as long as $T_{\text{Dark}} \approx 3000$ s, which allows for in depth understanding of the NSR dynamics. However, if one is interested only in the few-electron energy spectrum, the experiment can be run considerably faster. The pumping time can be reduced to $T_{\text{Pump}} \approx 0.1$ –1 s, which accelerates the NSR dynamics (at the expense of a reduced contrast between the cotunneling peaks and the charging plateaus). Accurate bias-NSR spectra can then be derived even from partial relaxation, by taking just 4–5 data points with $T_{\text{Dark}} \lesssim 30$ s and performing a simple linear (rather than full exponential) regression to estimate the NSR rates Γ_N . The high-quality NSR spectra, such as shown in Fig. 2(a), can be collected within a few hours of measurements. With further optimization, e.g. by measuring a single data point with a fixed $T_{\text{Dark}} \lesssim 10$ –30 s at each gate bias V_G , the acquisition of the bias-NSR spectrum can in principle be reduced to tens of minutes.

Although the hyperfine coupling of the valence band carriers is small compared to the conduction band electrons, we expect the NSR technique to be applicable to spectroscopy of the few-hole quantum dot states, where even more rich physics can be expected, due to the interplay of Coulomb, exchange, and heavy hole—light hole coupling contributions. Future work will investigate the effect of structural parameters (such as size, shape and chemical composition of the QD) on the multi-electronic energy spectrum and the qubit performance characteristics, such as spin lifetimes, spin coherence and the possibility of the electric spin control.

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S. M., S. F. C. S and A. R. developed, grew and processed the quantum dot samples. P. M.-H. and E. A. C. conducted multi-electron spectroscopy experiments. P. K. conducted configuration interaction modeling. H. E. D. and G. G. conducted electron spin lifetime and NMR experiments. E. A. C. drafted the manuscript with input from all authors. E. A. C. coordinated the project.

DATA AVAILABILITY

Source data are provided with this paper in the Source Data file SourceData.zip. Additional information and data related to this study are available from the corresponding authors upon request.

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- [1] T. Nakajima, A. Noiri, J. Yoneda, M. R. Delbecq, P. Stano, T. Otsuka, K. Takeda, S. Amaha, G. Allison, K. Kawasaki, A. Ludwig, A. D. Wieck, D. Loss, and S. Tarucha, *Quantum non-demolition measurement of an electron spin qubit*, *Nat. Nanotechnol.* **14**, 555 (2019).
 - [2] P.-A. Mortemousque, E. Chanrion, B. Jadot, H. Flentje, A. Ludwig, A. D. Wieck, M. Urdampilleta, C. Bäuerle, and T. Meunier, *Coherent control of individual electron spins in a two-dimensional quantum dot array*, *Nat. Nanotechnol.* **16**, 296 (2021).

- [3] L. Zaporiski, N. Shofer, J. H. Bodey, S. Manna, G. Gillard, M. H. Appel, C. Schimpf, S. F. Covre da Silva, J. Jarman, G. Delamare, G. Park, U. Haeusler, E. A. Chekhovich, A. Rastelli, D. A. Gangloff, M. Atatüre, and C. Le Gall, *Ideal refocusing of an optically active spin qubit under strong hyperfine interactions*, *Nat. Nanotechnol.* **18**, 257 (2023).
- [4] L. P. Kouwenhoven, D. G. Austing, and S. Tarucha, *Few-electron quantum dots*, *Rep. Prog. Phys.* **64**, 701 (2001).
- [5] S. M. Reimann and M. Manninen, *Electronic structure of quantum dots*, *Rev. Mod. Phys.* **74**, 1283 (2002).
- [6] R. Hanson, L. P. Kouwenhoven, J. R. Petta, S. Tarucha, and L. M. K. Vandersypen, *Spins in few-electron quantum dots*, *Rev. Mod. Phys.* **79**, 1217 (2007).
- [7] S. Tarucha, D. G. Austing, T. Honda, R. J. van der Hage, and L. P. Kouwenhoven, *Shell filling and spin effects in a few electron quantum dot*, *Phys. Rev. Lett.* **77**, 3613 (1996).
- [8] L. P. Kouwenhoven, T. H. Oosterkamp, M. W. S. Danoesastro, M. Eto, D. G. Austing, T. Honda, and S. Tarucha, *Excitation spectra of circular, few-electron quantum dots*, *Science* **278**, 1788 (1997).
- [9] J. M. Elzerman, R. Hanson, L. H. Willems van Beveren, B. Witkamp, L. M. K. Vandersypen, and L. P. Kouwenhoven, *Single-shot read-out of an individual electron spin in a quantum dot*, *Nature (London)* **430**, 431 (2004).
- [10] R. J. Schoelkopf, P. Wahlgren, A. A. Kozhevnikov, P. Delsing, and D. E. Prober, *The radio-frequency single-electron transistor (rf-set): A fast and ultrasensitive electrometer*, *Science* **280**, 1238 (1998).
- [11] T. Ota, K. Ono, M. Stopa, T. Hatano, S. Tarucha, H. Z. Song, Y. Nakata, T. Miyazawa, T. Ohshima, and N. Yokoyama, *Single-dot spectroscopy via elastic single-electron tunneling through a pair of coupled quantum dots*, *Phys. Rev. Lett.* **93**, 066801 (2004).
- [12] M. Jung, T. Machida, K. Hirakawa, S. Komiyama, T. Nakaoka, S. Ishida, and Y. Arakawa, *Shell structures in self-assembled InAs quantum dots probed by lateral electron tunneling structures*, *Appl. Phys. Lett.* **87**, 203109 (2005).
- [13] S. Amaha, T. Hatano, S. Teraoka, A. Shibatomi, S. Tarucha, Y. Nakata, T. Miyazawa, T. Oshima, T. Usuki, and N. Yokoyama, *Laterally coupled self-assembled InAs quantum dots embedded in resonant tunnel diode with multigate electrodes*, *Appl. Phys. Lett.* **92**, 202109 (2008).
- [14] Y. Kanai, R. S. Deacon, S. Takahashi, A. Oiwa, K. Yoshida, K. Shibata, K. Hirakawa, Y. Tokura, and S. Tarucha, *Electrically tuned spin-orbit interaction in an InAs self-assembled quantum dot*, *Nat. Nanotechnol.* **6**, 511 (2011).
- [15] S. Seidl, M. Kroner, P. A. Dalgarno, A. Högele, J. M. Smith, M. Ediger, B. D. Gerardot, J. M. Garcia, P. M. Petroff, K. Karrai, and R. J. Warburton, *Absorption and photoluminescence spectroscopy on a single self-assembled charge-tunable quantum dot*, *Phys. Rev. B* **72**, 195339 (2005).
- [16] P. A. Dalgarno, J. M. Smith, J. McFarlane, B. D. Gerardot, K. Karrai, A. Badolato, P. M. Petroff, and R. J. Warburton, *Coulomb interactions in single charged self-assembled quantum dots: Radiative lifetime and recombination energy*, *Phys. Rev. B* **77**, 245311 (2008).

- [17] V. Mlinar, M. Bozkurt, J. M. Ulloa, M. Ediger, G. Bester, A. Badolato, P. M. Koenraad, R. J. Warburton, and A. Zunger, *Structure of quantum dots as seen by excitonic spectroscopy versus structural characterization: Using theory to close the loop*, *Phys. Rev. B* **80**, 165425 (2009).
- [18] M. Holtkemper, D. E. Reiter, and T. Kuhn, *Influence of the quantum dot geometry on p-shell transitions in differently charged quantum dots*, *Phys. Rev. B* **97**, 075308 (2018).
- [19] D. Huber, B. U. Lehner, D. Csontosová, M. Reindl, S. Schuler, S. F. Covre da Silva, P. Klenovský, and A. Rastelli, *Single-particle-picture breakdown in laterally weakly confining GaAs quantum dots*, *Phys. Rev. B* **100**, 235425 (2019).
- [20] A. Nick Vamivakas, Y. Zhao, C.-Y. Lu, and M. Atatüre, *Spin-resolved quantum-dot resonance fluorescence*, *Nat. Phys.* **5**, 198 (2009).
- [21] C. Nawrath, H. Vural, J. Fischer, R. Schaber, S. L. Portalupi, M. Jetter, and P. Michler, *Resonance fluorescence of single In(Ga)As quantum dots emitting in the telecom C-band*, *Appl. Phys. Lett.* **118**, 244002 (2021).
- [22] P. Hawrylak, G. A. Narvaez, M. Bayer, and A. Forchel, *Excitonic absorption in a quantum dot*, *Phys. Rev. Lett.* **85**, 389 (2000).
- [23] M. E. Ware, E. A. Stinaff, D. Gammon, M. F. Doty, A. S. Bracker, D. Gershoni, V. L. Korenev, Ş. C. Bădescu, Y. Lyanda-Geller, and T. L. Reinecke, *Polarized fine structure in the photoluminescence excitation spectrum of a negatively charged quantum dot*, *Phys. Rev. Lett.* **95**, 177403 (2005).
- [24] M. C. Löbl, C. Spinnler, A. Javadi, L. Zhai, G. N. Nguyen, J. Ritzmann, L. Midolo, P. Lodahl, A. D. Wieck, A. Ludwig, and R. J. Warburton, *Radiative Auger process in the single-photon limit*, *Nat. Nanotechnol.* **15**, 558 (2020).
- [25] H. Drexler, D. Leonard, W. Hansen, J. P. Kotthaus, and P. M. Petroff, *Spectroscopy of quantum levels in charge-tunable InGaAs quantum dots*, *Phys. Rev. Lett.* **73**, 2252 (1994).
- [26] B. T. Miller, W. Hansen, S. Manus, R. J. Luyken, A. Lorke, J. P. Kotthaus, S. Huan, G. Medeiros-Ribeiro, and P. M. Petroff, *Few-electron ground states of charge-tunable self-assembled quantum dots*, *Phys. Rev. B* **56**, 6764 (1997).
- [27] R. Wetzler, A. Wacker, E. Schöll, C. M. A. Kapteyn, R. Heitz, and D. Bimberg, *Capacitance-voltage characteristics of InAs/GaAs quantum dots embedded in a pn structure*, *Appl. Phys. Lett.* **77**, 1671 (2000).
- [28] D. Reuter, R. Roescu, M. Mehta, M. Richter, and A. D. Wieck, *Capacitance-voltage spectroscopy of post-growth annealed InAs quantum dots*, *Phys. E* **40**, 1961 (2008).
- [29] F. Brinks, A. D. Wieck, and A. Ludwig, *Thermal shift of the resonance between an electron gas and quantum dots: What is the origin?*, *New J. Phys.* **18**, 123019 (2016).
- [30] N. F. Brosda, P. J. Badura, i. d. I. Bölükbaş, i. d. I. Engin, P. Lindner, S. R. Valentin, A. D. Wieck, B. Sothmann, and A. Ludwig, *Towards quantitative understanding of quantum dot ensemble capacitance-voltage spectroscopy*, *Phys. Rev. B* **112**, 165201 (2025).
- [31] R. Tycko, G. Dabbagh, M. J. Rosseinsky, D. W. Murphy, R. M. Fleming, A. P. Ramirez, and J. C. Tully, *^{13}C NMR spectroscopy of K_xC_{60} : Phase separation, molecular dynamics, and metallic properties*, *Science* **253**, 884 (1991).
- [32] R. Tycko, *Electronic properties and phase transitions of alkali fullerenes: Investigations by nuclear magnetic resonance spectroscopy*, *J. Phys. Chem. Solids* **54**, 1713 (1993).
- [33] R. E. Walstedt and W. W. Warren, Jr., *Nuclear resonance properties of $\text{YBa}_2\text{Cu}_3\text{O}_{6+x}$ superconductors*, *Science* **248**, 1082 (1990).
- [34] J. A. Martindale, S. E. Barrett, C. A. Klug, K. E. O'Hara, S. M. DeSoto, C. P. Slichter, T. A. Friedmann, and D. M. Ginsberg, *Anisotropy and magnetic field dependence of the planar copper NMR spin-lattice relaxation rate in the superconducting state of $\text{YBa}_2\text{Cu}_3\text{O}_7$* , *Phys. Rev. Lett.* **68**, 702 (1992).
- [35] J. H. Smet, R. A. Deutschmann, F. Ertl, W. Wegscheider, G. Abstreiter, and K. von Klitzing, *Gate-voltage control of spin interactions between electrons and nuclei in a semiconductor*, *Nature (London)* **415**, 281 (2002).
- [36] N. Kumada, K. Muraki, and Y. Hirayama, *Low-frequency spin dynamics in a canted antiferromagnet*, *Science* **313**, 329 (2006).
- [37] S. E. Barrett, G. Dabbagh, L. N. Pfeiffer, K. W. West, and R. Tycko, *Optically pumped NMR evidence for finite-size skyrmions in GaAs quantum wells near Landau level filling $\nu = 1$* , *Phys. Rev. Lett.* **74**, 5112 (1995).
- [38] J. Liu, R. Su, Y. Wei, B. Yao, S. F. Cda Silva, Y. Yu, J. Iles-Smith, K. Srinivasan, A. Rastelli, J. Li, and X. Wang, *A solid-state source of strongly entangled photon pairs with high brightness and indistinguishability*, *Nat. Nanotechnol.* **14**, 586 (2019).
- [39] L. Zhai, M. C. Löbl, G. N. Nguyen, J. Ritzmann, A. Javadi, C. Spinnler, A. D. Wieck, A. Ludwig, and R. J. Warburton, *Low-noise GaAs quantum dots for quantum photonics*, *Nat. Commun.* **11**, 4745 (2020).
- [40] N. Tamm, A. Javadi, N. O. Antoniadis, D. Najer, M. C. Löbl, A. R. Korsch, R. Schott, S. R. Valentin, A. D. Wieck, A. Ludwig, and R. J. Warburton, *A bright and fast source of coherent single photons*, *Nat. Nanotechnol.* **16**, 399 (2021).
- [41] E. A. Chekhovich, S. F. C. da Silva, and A. Rastelli, *Nuclear spin quantum register in an optically active semiconductor quantum dot*, *Nat. Nanotechnol.* **15**, 999 (2020).
- [42] M. H. Appel, A. Ghorbal, N. Shofer, L. Zaporski, S. Manna, S. F. C. da Silva, U. Haeusler, C. Le Gall, A. Rastelli, D. A. Gangloff, and M. Atatüre, *A many-body quantum register for a spin qubit*, *Nat. Phys.* **21**, 368 (2025).
- [43] H. E. Dyte, S. Manna, S. F. Covre da Silva, A. Rastelli, and E. A. Chekhovich, *Storing quantum coherence in a quantum dot nuclear spin ensemble for over 100 milliseconds*, *Nat. Commun.* **17**, 239 (2025).
- [44] R. C. C. Leon, C. H. Yang, J. C. C. Hwang, J. C. Lemyre, T. Tanttu, W. Huang, K. W. Chan, K. Y. Tan, F. E. Hudson, K. M. Itoh, A. Morello, A. Laucht, M. Pioro-Ladrière, A. Saraiva, and A. S. Dzurak, *Coherent spin control of s-, p-, d- and f-electrons in a silicon quantum dot*, *Nat. Commun.* **11**, 797 (2020).

- [45] K. C. Nowack, F. H. L. Koppens, Y. V. Nazarov, and L. M. K. Vandersypen, *Coherent control of a single electron spin with electric fields*, *Science* **318**, 1430 (2007).
- [46] S. Nadj-Perge, S. M. Frolov, E. P. A. M. Bakkers, and L. P. Kouwenhoven, *Spin-orbit qubit in a semiconductor nanowire*, *Nature (London)* **468**, 1084 (2010).
- [47] J. Yoneda, T. Otsuka, T. Nakajima, T. Takakura, T. Obata, M. Pioro-Ladrière, H. Lu, C. J. Palmstrøm, A. C. Gossard, and S. Tarucha, *Fast electrical control of single electron spins in quantum dots with vanishing influence from nuclear spins*, *Phys. Rev. Lett.* **113**, 267601 (2014).
- [48] K. Schmidt-Rohr, J. Clauss, and H. W. Spiess, *Correlation of structure, mobility, and morphological information in heterogeneous polymer materials by two-dimensional wideline-separation NMR spectroscopy*, *Macromolecules* **25**, 3273 (1992).
- [49] D. E. Demco, A. Johansson, and J. Tegenfeldt, *Proton spin diffusion for spatial heterogeneity and morphology investigations of polymers*, *Solid State Nucl. Magn. Reson.* **4**, 13 (1995).
- [50] D. A. Hall, D. C. Maus, G. J. Gerfen, S. J. Inati, L. R. Becerra, F. W. Dahlquist, and R. G. Griffin, *Polarization-enhanced NMR spectroscopy of biomolecules in frozen solution*, *Science* **276**, 930 (1997).
- [51] P. C. A. van der Wel, K.-N. Hu, J. Lewandowski, and R. G. Griffin, *Dynamic nuclear polarization of amyloidogenic peptide nanocrystals: GNNQQNY, a core segment of the yeast prion protein Sup35p*, *J. Am. Chem. Soc.* **128**, 10840 (2006).
- [52] T. Manolikas, T. Herrmann, and B. H. Meier, *Protein structure determination from ^{13}C spin-diffusion solid-state NMR spectroscopy*, *J. Am. Chem. Soc.* **130**, 3959 (2008).
- [53] A. J. Rossini, C. M. Widdifield, A. Zagdoun, M. Lelli, M. Schwarzwald, C. Coperet, A. Lesage, and L. Emsley, *Dynamic nuclear polarization enhanced NMR spectroscopy for pharmaceutical formulations*, *J. Am. Chem. Soc.* **136**, 2324 (2014).
- [54] J. Viger-Gravel, A. Schantz, A. C. Pinon, A. J. Rossini, S. Schantz, and L. Emsley, *Structure of lipid nanoparticles containing siRNA or mRNA by dynamic nuclear polarization-enhanced NMR spectroscopy*, *J. Phys. Chem. B* **122**, 2073 (2018).
- [55] G. von Witte, S. Kozerke, and M. Ernst, *Two-electron two-nucleus effective Hamiltonian and the spin diffusion barrier*, *Sci. Adv.* **11**, eadr7168 (2025).
- [56] C. Heyn, A. Stemmann, T. Koppen, C. Strelow, T. Kipp, M. Grave, S. Mendach, and W. Hansen, *Highly uniform and strain-free GaAs quantum dots fabricated by filling of self-assembled nanoholes*, *Appl. Phys. Lett.* **94**, 183113 (2009).
- [57] P. Atkinson, E. Zallo, and O. G. Schmidt, *Independent wavelength and density control of uniform GaAs/AlGaAs quantum dots grown by infilling self-assembled nanoholes*, *J. Appl. Phys.* **112**, 054303 (2012).
- [58] H. E. Dyte, G. Gillard, S. Manna, S. F. Covre da Silva, A. Rastelli, and E. A. Chekhovich, *Is wave function collapse necessary? Explaining quantum nondemolition measurement of a spin qubit within linear evolution*, *Phys. Rev. Lett.* **132**, 160804 (2024).
- [59] P. Millington-Hotze, S. Manna, S. F. Covre da Silva, A. Rastelli, and E. A. Chekhovich, *Nuclear spin diffusion in the central spin system of a GaAs/AlGaAs quantum dot*, *Nat. Commun.* **14**, 2677 (2023).
- [60] D. Paget, *Optical detection of NMR in high-purity GaAs: Direct study of the relaxation of nuclei close to shallow donors*, *Phys. Rev. B* **25**, 4444 (1982).
- [61] R. Tycko, S. E. Barrett, G. Dabbagh, L. N. Pfeiffer, and K. W. West, *Electronic states in gallium arsenide quantum wells probed by optically pumped NMR*, *Science* **268**, 1460 (1995).
- [62] H. Hayashi, K. M. Itoh, and L. S. Vlasenko, *Nuclear magnetic resonance linewidth and spin diffusion in ^{29}Si isotopically controlled silicon*, *Phys. Rev. B* **78**, 153201 (2008).
- [63] A. E. Nikolaenko, E. A. Chekhovich, M. N. Makhonin, I. W. Drouzas, A. B. Van'kov, J. Skiba-Szymanska, M. S. Skolnick, P. Senellart, D. Martrou, A. Lemaître, and A. I. Tartakovskii, *Suppression of nuclear spin diffusion at a GaAs/Al_xGa_{1-x}As interface measured with a single quantum-dot nanoprobe*, *Phys. Rev. B* **79**, 081303 (2009).
- [64] P. Millington-Hotze, H. E. Dyte, S. Manna, S. F. Covre da Silva, A. Rastelli, and E. A. Chekhovich, *Approaching a fully-polarized state of nuclear spins in a solid*, *Nat. Commun.* **15**, 985 (2024).
- [65] B. Urbaszek, X. Marie, T. Amand, O. Krebs, P. Voisin, P. Maletinsky, A. Högele, and A. Imamoglu, *Nuclear spin physics in quantum dots: An optical investigation*, *Rev. Mod. Phys.* **85**, 79 (2013).
- [66] See Supplemental Material <http://link.aps.org/supplemental/10.1103/j8xw-w72c> for further details, which includes Refs. [99–113].
- [67] D. V. Averin, A. N. Korotkov, and K. K. Likharev, *Theory of single-electron charging of quantum wells and dots*, *Phys. Rev. B* **44**, 6199 (1991).
- [68] C. W. J. Beenakker, *Theory of Coulomb-blockade oscillations in the conductance of a quantum dot*, *Phys. Rev. B* **44**, 1646 (1991).
- [69] C. Latta, A. Srivastava, and A. Imamoglu, *Hyperfine interaction-dominated dynamics of nuclear spins in self-assembled InGaAs quantum dots*, *Phys. Rev. Lett.* **107**, 167401 (2011).
- [70] G. Gillard, I. M. Griffiths, G. Ragunathan, A. Ulhaq, C. McEwan, E. Clarke, and E. A. Chekhovich, *Fundamental limits of electron and nuclear spin qubit lifetimes in an isolated self-assembled quantum dot*, *npj Quantum Inf.* **7**, 43 (2021).
- [71] L. Cockins, Y. Miyahara, S. D. Bennett, A. A. Clerk, S. Studenikin, P. Poole, A. Sachrajda, and P. Grutter, *Energy levels of few-electron quantum dots imaged and characterized by atomic force microscopy*, *Proc. Natl. Acad. Sci. USA* **107**, 9496 (2010).
- [72] A. Franceschetti and A. Zunger, *Pseudopotential calculations of electron and hole addition spectra of InAs, InP, and Si quantum dots*, *Phys. Rev. B* **62**, 2614 (2000).
- [73] I. Vurgaftman, J. R. Meyer, and L. R. Ram-Mohan, *Band parameters for III–V compound semiconductors and their alloys*, *J. Appl. Phys.* **89**, 5815 (2001).
- [74] H.-M. Müller and S. E. Koonin, *Phase transitions in quantum dots*, *Phys. Rev. B* **54**, 14532 (1996).

- [75] M. B. Tavernier, E. Anisimovas, F. M. Peeters, B. Szafran, J. Adamowski, and S. Bednarek, *Four-electron quantum dot in a magnetic field*, *Phys. Rev. B* **68**, 205305 (2003).
- [76] Y. Nishi, Y. Tokura, J. Gupta, G. Austing, and S. Tarucha, *Ground-state transitions beyond the singlet-triplet transition for a two-electron quantum dot*, *Phys. Rev. B* **75**, 121301 (2007).
- [77] P. Klenovský, P. Steindl, and D. Geffroy, *Excitonic structure and pumping power dependent emission blue-shift of type-II quantum dots*, *Sci. Rep.* **7**, 45568 (2017).
- [78] H. Huang, D. Csontosová, S. Manna, Y. Huo, R. Trotta, A. Rastelli, and P. Klenovský, *Electric field induced tuning of electronic correlation in weakly confining quantum dots*, *Phys. Rev. B* **104**, 165401 (2021).
- [79] P. Klenovský, *Coulomb correlated multiparticle states of weakly confining GaAs quantum dots*, *Phys. Rev. B* **113**, 125410 (2026).
- [80] A. H. MacDonald, S. R. Eric Yang, and M. D. Johnson, *Quantum dots in strong magnetic fields: Stability criteria for the maximum density droplet*, *Aust. J. Phys.* **46**, 345 (1993).
- [81] C.-W. Huang and X. Hu, *Theoretical study of nuclear spin polarization and depolarization in self-assembled quantum dots*, *Phys. Rev. B* **81**, 205304 (2010).
- [82] C. Bulutay, *Quadrupolar spectra of nuclear spins in strained $\text{In}_x\text{Ga}_{1-x}\text{As}$ quantum dots*, *Phys. Rev. B* **85**, 115313 (2012).
- [83] W. A. Coish, J. Fischer, and D. Loss, *Exponential decay in a spin bath*, *Phys. Rev. B* **77**, 125329 (2008).
- [84] D. Klauser, W. A. Coish, and D. Loss, *Nuclear spin dynamics and Zeno effect in quantum dots and defect centers*, *Phys. Rev. B* **78**, 205301 (2008).
- [85] D. J. Reilly, J. M. Taylor, J. R. Petta, C. M. Marcus, M. P. Hanson, and A. C. Gossard, *Exchange control of nuclear spin diffusion in a double quantum dot*, *Phys. Rev. Lett.* **104**, 236802 (2010).
- [86] Z.-X. Gong, Z.-Q. Yin, and L.-M. Duan, *Dynamics of the Overhauser field under nuclear spin diffusion in a quantum dot*, *New J. Phys.* **13**, 033036 (2011).
- [87] Y. B. Lyanda-Geller, I. L. Aleiner, and B. L. Altshuler, *Coulomb “blockade” of nuclear spin relaxation in quantum dots*, *Phys. Rev. Lett.* **89**, 107602 (2002).
- [88] Y. B. Lyanda-Geller, I. L. Aleiner, and B. L. Altshuler, *Charging effects on nuclear spin relaxation in quantum dots*, *J. Supercond.* **16**, 751 (2003).
- [89] F. Baruffa, P. Stano, and J. Fabian, *Theory of anisotropic exchange in laterally coupled quantum dots*, *Phys. Rev. Lett.* **104**, 126401 (2010).
- [90] L. C. Camenzind, L. Yu, P. Stano, J. D. Zimmerman, A. C. Gossard, D. Loss, and D. M. Zumbühl, *Hyperfine-phonon spin relaxation in a single-electron GaAs quantum dot*, *Nat. Commun.* **9**, 3454 (2018).
- [91] G. Gillard, E. Clarke, and E. A. Chekhovich, *Harnessing many-body spin environment for long coherence storage and high-fidelity single-shot qubit readout*, *Nat. Commun.* **13**, 4048 (2022).
- [92] M. Atatüre, J. Dreiser, A. Badolato, A. Högele, K. Karrai, and A. Imamoglu, *Quantum-dot spin-state preparation with near-unity fidelity*, *Science* **312**, 551 (2006).
- [93] A. V. Khaetskii and Y. V. Nazarov, *Spin-flip transitions between Zeeman sublevels in semiconductor quantum dots*, *Phys. Rev. B* **64**, 125316 (2001).
- [94] M. Kroutvar, Y. Ducommun, D. Heiss, M. Bichler, D. Schuh, G. Abstreiter, and J. J. Finley, *Optically programmable electron spin memory using semiconductor quantum dots*, *Nature (London)* **432**, 81 (2004).
- [95] C.-Y. Lu, Y. Zhao, A. N. Vamivakas, C. Matthiesen, S. Fält, A. Badolato, and M. Atatüre, *Direct measurement of spin dynamics in InAs/GaAs quantum dots using time-resolved resonance fluorescence*, *Phys. Rev. B* **81**, 035332 (2010).
- [96] A. Ulhaq, Q. Duan, E. Zallo, F. Ding, O. G. Schmidt, A. I. Tartakovskii, M. S. Skolnick, and E. A. Chekhovich, *Vanishing electron g factor and long-lived nuclear spin polarization in weakly strained nanohole-filled $\text{GaAs}/\text{AlGaAs}$ quantum dots*, *Phys. Rev. B* **93**, 165306 (2016).
- [97] F. H. L. Koppens, C. Buizert, K. J. Tielrooij, I. T. Vink, K. C. Nowack, T. Meunier, L. P. Kouwenhoven, and L. M. K. Vandersypen, *Driven coherent oscillations of a single electron spin in a quantum dot*, *Nature (London)* **442**, 766 (2006).
- [98] M. Shafiei, K. C. Nowack, C. Reichl, W. Wegscheider, and L. M. K. Vandersypen, *Resolving spin-orbit- and hyperfine-mediated electric dipole spin resonance in a quantum dot*, *Phys. Rev. Lett.* **110**, 107601 (2013).
- [99] A. Oshiyama and S. Ohnishi, *DX center: Crossover of deep and shallow states in $\text{Si-doped Al}_x\text{Ga}_{1-x}\text{As}$* , *Phys. Rev. B* **33**, 4320 (1986).
- [100] P. M. Mooney, *Deep donor levels (DX centers) in III–V semiconductors*, *J. Appl. Phys.* **67**, R1 (1990).
- [101] F. Bloch, *Nuclear induction*, *Phys. Rev.* **70**, 460 (1946).
- [102] E. A. Chekhovich, M. M. Glazov, A. B. Krysa, M. Hopkinson, P. Senellart, A. Lemaître, M. S. Skolnick, and A. I. Tartakovskii, *Element-sensitive measurement of the hole-nuclear spin interaction in quantum dots*, *Nat. Phys.* **9**, 74 (2013).
- [103] D. Gammon, A. L. Efros, T. A. Kennedy, M. Rosen, D. S. Katzer, D. Park, S. W. Brown, V. L. Korenev, and I. A. Merkulov, *Electron and nuclear spin interactions in the optical spectra of single GaAs quantum dots*, *Phys. Rev. Lett.* **86**, 5176 (2001).
- [104] B. Eble, O. Krebs, A. Lemaître, K. Kowalik, A. Kudelski, P. Voisin, B. Urbaszek, X. Marie, and T. Amand, *Dynamic nuclear polarization of a single charge-tunable InAs/GaAs quantum dot*, *Phys. Rev. B* **74**, 081306 (2006).
- [105] J. Skiba-Szymanska, E. A. Chekhovich, A. E. Nikolaenko, A. I. Tartakovskii, M. N. Makhonin, I. Drouzas, M. S. Skolnick, and A. B. Krysa, *Overhauser effect in individual $\text{InP}/\text{Ga}_x\text{In}_{1-x}\text{P}$ dots*, *Phys. Rev. B* **77**, 165338 (2008).
- [106] G. Ragnathan, J. Kobak, G. Gillard, W. Pacuski, K. Sobczak, J. Borysiuk, M. S. Skolnick, and E. A. Chekhovich, *Direct measurement of hyperfine shifts and radio frequency manipulation of nuclear spins in individual CdTe/ZnTe quantum dots*, *Phys. Rev. Lett.* **122**, 096801 (2019).
- [107] E. A. Chekhovich, A. Ulhaq, E. Zallo, F. Ding, O. G. Schmidt, and M. S. Skolnick, *Measurement of the spin temperature of optically cooled nuclei and GaAs hyperfine*

- constants in GaAs/AlGaAs quantum dots, *Nat. Mater.* **16**, 982 (2017).
- [108] Y. Wang and K. Takeda, *Speedup of nuclear spin diffusion in hyperpolarized solids*, *New J. Phys.* **23**, 073015 (2021).
- [109] E. A. Chekhovich, K. V. Kavokin, J. Puebla, A. B. Krysa, M. Hopkinson, A. D. Andreev, A. M. Sanchez, R. Beanland, M. S. Skolnick, and A. I. Tartakovskii, *Structural analysis of strained quantum dots using nuclear magnetic resonance*, *Nat. Nanotechnol.* **7**, 646 (2012).
- [110] E. A. Chekhovich, I. M. Griffiths, M. S. Skolnick, H. Huang, S. F. Covre da Silva, X. Yuan, and A. Rastelli, *Cross calibration of deformation potentials and gradient-elastic tensors of GaAs using photoluminescence and nuclear magnetic resonance spectroscopy in GaAs/AlGaAs quantum dot structures*, *Phys. Rev. B* **97**, 235311 (2018).
- [111] S. Birner, T. Zibold, T. Andlauer, T. Kubis, M. Sabathil, A. Trellakis, and P. Vogl, *Nextnano: General purpose 3-D simulations*, *IEEE Trans. Electron Devices* **54**, 2137 (2007).
- [112] A. Schliwa, M. Winkelkemper, and D. Bimberg, *Few-particle energies versus geometry and composition of $\text{In}_x\text{Ga}_{1-x}\text{As}/\text{GaAs}$ self-organized quantum dots*, *Phys. Rev. B* **79**, 075443 (2009).
- [113] O. Stier, *Electronic and optical properties of quantum dots and wires*, Ph.D. thesis, Technische Universität Berlin, 2000.