

Electron and Nuclear Spin Interactions in the Optical Spectra of Single GaAs Quantum Dots

D. Gammon, Al. L. Efros, T. A. Kennedy, M. Rosen, D. S. Katzer, and D. Park

Naval Research Laboratory, Washington, D.C. 20375

S. W. Brown

NIST, Gaithersburg, Maryland

V. L. Korenev and I. A. Merkulov

A. F. Ioffe Institute, St. Petersburg, Russia

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Fine and hyperfine splittings arising from electron, hole, and nuclear spin interactions in the magneto-optical spectra of individual localized excitons are studied. We explain the magnetic field dependence of the energy splitting through competition between Zeeman, exchange, and hyperfine interactions. An unexpectedly small hyperfine contribution to the splitting close to zero applied field is described well by the interplay between fluctuations of the hyperfine field experienced by the nuclear spin and nuclear dipole/dipole interactions.

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The spin of an electron in a 10 nm GaAs quantum dot (QD) interacts with $\sim 10^5$ nuclear spins. This hyperfine interaction, though relatively weak, may ultimately limit spin coherence of localized electrons in QDs or shallow impurities—a concern that strongly influences developing visions of information technologies based on spin [1–3]. Nevertheless, there may be ways around even this intrinsic scattering process; for example, by optically polarizing all nuclear spin and thereby dramatically reducing phase space [1]. Furthermore, one could imagine using the nuclear spin for information storage or to control the electronic spin [4]. However, it is necessary to develop a more precise understanding of spin interactions in nanostructures in the presence of external magnetic and optical fields before such creative ideas can be explored.

In this Letter, we present and analyze spectroscopic signatures of spin via fine and hyperfine structure splittings in the magneto-optical spectra of individual GaAs QDs under polarized and nearly resonant laser excitation. We find it necessary to consider the interaction of the electron spin with an external magnetic field (Zeeman interaction), exchange Coulomb interactions between the electron and hole, and hyperfine interactions between the electronic spin and the spins of the nuclei. Because of the hyperfine interaction, it is necessary to consider also the nuclear spin system. We are led then to consider, for the nuclei, the Zeeman interaction, dipole-dipole interactions between neighboring nuclear spins [5], and the hyperfine interaction. We quantify in experiment and theory how interactions manifest themselves in the spectral fine structure of a single exciton, discovering and explaining a remarkably complex dependence on magnetic field arising from competition between these various spin interactions.

We have studied QDs formed by monolayer-high interface islands in a 4.2 nm GaAs quantum well with 25 nm $\text{Al}_{0.3}\text{Ga}_{0.7}\text{As}$ barriers. The quantum wells were

grown using molecular beam epitaxy with two-minute growth interrupts at the interfaces to allow large interface islands to develop. Individual QDs were excited and detected through $\frac{1}{2}$ micron diameter apertures in an aluminum shadow mask patterned on the sample surface. A split-coil superconducting magnet was used in backscattering Faraday geometry. Previous studies have demonstrated that optical pumping could lead to large nuclear polarization [6–8]. For one experiment, a small Helmholtz coil was used to apply a transverse rf magnetic field as a controlled source of nuclear heating to measure the optical pumping rate of the nuclear spin system [9,10].

Shown in Fig. 1 are photoluminescence (PL) spectra obtained under applied magnetic field normal to the quantum well plane (z axis). Exciting light with circular polarization (σ^+) was tuned into the upper monolayer about 6 meV above the luminescing lines and with ~ 10 W/cm² to maximize the Overhauser effect [8]. At zero field, there is a doublet with a splitting of 24 μeV in which both components are linearly polarized along the $[110]/[1-10]$ axes [11–14]. Remarkably, the behavior is qualitatively different for the two signs of the applied field as shown in detail in Fig. 2. The splitting is nonlinear, nonmonotonic, and asymmetric in the sign of the external field.

For both σ^+ and σ^- excitation, we show as a function of field the average energy of the doublet in the inset of Fig. 2 and the energy splitting in Fig. 3. The average energy for both exciting polarizations is described well by a diamagnetic shift of 25 $\mu\text{eV}/\text{T}^2$. In contrast, the energy splitting is quite different for the two exciting polarizations. Except for a dip at the origin, both curves can be fit roughly by a linear dependence on the external field (with an exciton g factor $g_1 = 1.3$), but with a shift from the origin of ± 90 μeV , respectively. This shift of the energy splitting is a result of dynamic polarization of the nuclear spin arising from the hyperfine

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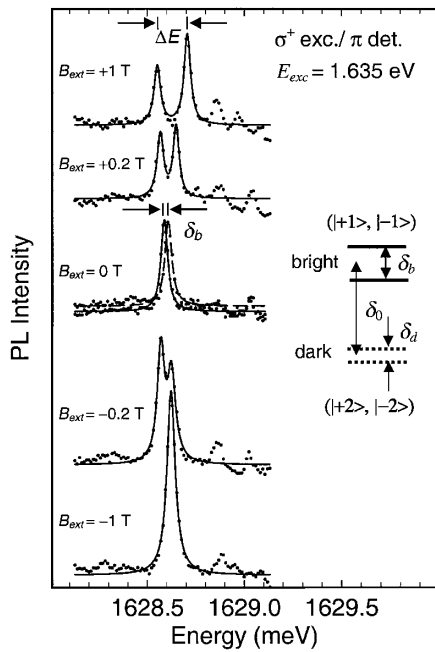


FIG. 1. Photoluminescence spectra as a function of external magnetic field. At nonzero fields, circular excitation (σ^+) and linear detection (π) were used. At $B_{\text{ext}} = 0$, both excitation and detection was with vertical (solid line) or horizontal (dashed line) linear polarization. Inset: Energy level diagram of the QD fine structure at zero field.

interaction (i.e., the Overhauser shift well-known in bulk [6]). The time measured for optical polarization of the nuclei in the QD is about 3 s [10]. The resulting effective magnetic field of the oriented nuclei, B_N , is independent of B_{ext} except around zero field where we see the narrow dip. The 90 μeV hyperfine contribution to the spin

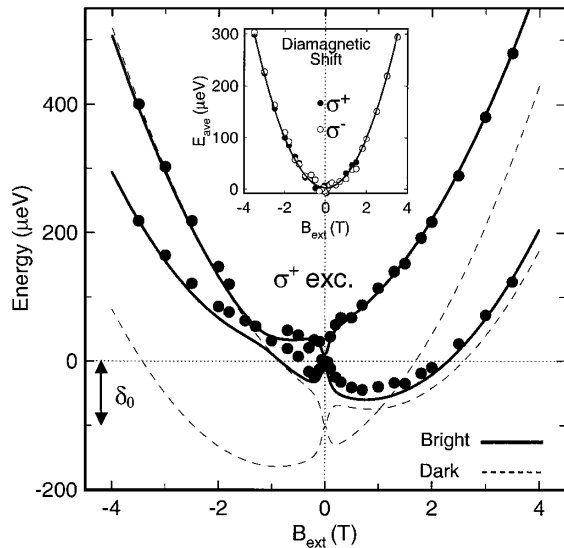


FIG. 2. Energies of the spectral lines for σ^+ excitation. The lines are calculated values of the bright (solid line) and dark (dashed line) energies. Inset: The average of the two components of the doublet, showing a quadratic dependence on B_{ext} .

splitting corresponds to a nuclear polarization of $\sim 65\%$ (at fields $B_{\text{ext}} > 0.2$ T) and to $B_N \sim 1.2$ T for a g factor of 1.3. At $B_{\text{ext}} = -B_N \sim \pm 1.2$ T, the fields compensate and a single peak is observed. Exchange splitting remains, but, because this measurement was made with linear polarization along [110], only one of the linear components was detected and the splitting was not measured.

There is a pronounced deviation from this simple picture at $B_{\text{ext}} \sim 0$ in the form of a strong dip in the energy splitting. This dip can be fit to a Lorentzian line shape with a half-width of 80 mT (Fig. 3: upper inset). Complete measurements have been made on several other QDs from the same sample with similar results. In bulk semiconductors, dipole-dipole interactions between nuclear spins prevent spin polarization at magnetic fields less than the field seen by a nuclear spin due to the dipole field of its neighbors ($\sim B_L \sim 0.15$ mT), and, as a result, a dip in the spin splitting similar in shape to that observed here is expected [15]. However, the width of the dip in ΔE that we observe is over 2 orders of magnitude larger.

To explain this surprising observation, we consider the electronic and nuclear spin interactions in detail. The QD exciton arises from weak lateral confinement of a quantum well 2D exciton formed from the lowest electron ($S_z = \pm \frac{1}{2}$) and heavy hole ($J_z = \pm \frac{3}{2}$) subbands. The exciton thus consists of four degenerate substates: two optically allowed (bright), $|\pm 1\rangle$, and two optically forbidden (dark), $|\pm 2\rangle$ [16]. This degeneracy is split at zero field by the exchange interaction into two closely spaced doublets

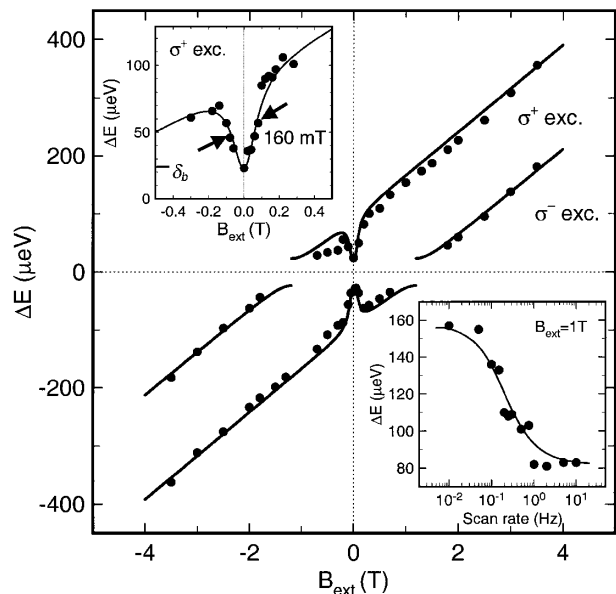


FIG. 3. Energy splitting for both σ^+ and σ^- excitation. Upper inset: Higher resolution data for the region around $B_{\text{ext}} = 0$, showing the dip in the energy splitting. Lower inset: Energy splitting at $B_{\text{ext}} = 1$ T as a function of the sweep rate of a transverse rf field through the nuclear spin resonances [10]. At a rate of $(3 \text{ s})^{-1}$, the optical alignment rate equals this external heating rate.

(Fig. 1: inset). The Hamiltonian describing the exchange interaction in an anisotropic QD is given by [16]

$$\hat{H}_{\text{ex}} = \frac{\delta_0}{2} \hat{\sigma}_z^e \hat{\sigma}_z^h + \frac{\delta_b}{4} (\hat{\sigma}_x^e \hat{\sigma}_x^h - \hat{\sigma}_y^e \hat{\sigma}_y^h) + \frac{\delta_d}{4} (\hat{\sigma}_x^e \hat{\sigma}_x^h + \hat{\sigma}_y^e \hat{\sigma}_y^h), \quad (1)$$

where we have outerproducts of the Pauli matrices ($\sigma_\alpha^{h,e}$) acting on electron and heavy-hole spin variables, respectively; and $\delta_{0,b,d}$ are the exchange interaction constants in anisotropic QDs (Fig. 1: inset). In this representation, the heavy-hole wave functions $|\pm \frac{3}{2}\rangle$ transform to pseudo-spin, $\mp \frac{1}{2}$. Diagonalization of Eq. (1) gives the zero field energy levels schematically shown in the inset of Fig. 1. The energy splitting between the bright and dark states is determined by δ_0 ($\sim 100 \mu\text{eV}$) [17,18]. The anisotropic exchange term ($\delta_b = 24 \mu\text{eV}$) leads to the observed fine structure splitting of the bright exciton and linear polarization of the PL at zero field [11,19]. The exchange splitting of the dark exciton, δ_d , is expected to be on the order of $1 \mu\text{eV}$ [20].

The origin of the complex dependence of the energy splitting on magnetic field arises from competition between the exchange, Zeeman, and hyperfine interactions: $\hat{H}_{\text{spin}}^{\text{exciton}} = \hat{H}_{\text{ex}} + \hat{H}_Z + \hat{H}_{\text{hf}}$. The Zeeman interaction term, with the external magnetic field directed along the z axis (B_{ext}), is

$$\hat{H}_Z = \frac{\mu_B}{2} (-g_h \hat{\sigma}_z^e \hat{1}^e + g_e \hat{\sigma}_z^e \hat{1}^h) B_{\text{ext}}, \quad (2)$$

where $\hat{1}^{e,h}$ are unit matrices, $g_{e,h}$ are the electron and hole g factors, and $\mu_B = 58 \mu\text{eV}/\text{T}$ is the Bohr magneton.

The hyperfine interaction between the electron and the nuclear spins is given by [6], chapter 2)

$$\hat{H}_{\text{hf}} = \frac{\nu_0}{2} \sum_j A^j |\psi(\mathbf{R}_j)|^2 \left(\hat{I}_z^j \hat{\sigma}_z^e + \frac{\hat{I}_+^j \hat{\sigma}_-^e + \hat{I}_-^j \hat{\sigma}_+^e}{2} \right), \quad (3)$$

where $\nu_0 = a_0^3$ is the volume of the unit cell, A^j is the constant of the hyperfine interaction, \mathbf{I}^j and \mathbf{R}_j are the spin and coordinate of the j th nucleus ($I = \frac{3}{2}$), $|\psi(\mathbf{r})|^2$ is the electron density at the j th nuclear site, and the sum goes over all nuclei. In the mean-field approximation, Eq. (3) describes the interaction of an electron with the effective magnetic field, B_N , of the nuclear spins: $\langle \hat{H}_{\text{hf}} \rangle_N = \frac{1}{2} A \langle I_z \rangle \hat{\sigma}_z^e = (\mu_B/2) g_e \hat{\sigma}_z^e B_N$, where $\langle I_z \rangle$ is the average spin of the polarized nuclei, $A = \sum_j A^j$, and the sum goes over all nuclei in the unit cell ($A \approx 90 \mu\text{eV}$ in GaAs [15]). If the nuclear spins are disordered, $B_N = 0$. Under conditions of optically induced nuclear orientation, their effective magnetic field leads to the additional splitting of the exciton sublevels known as the Overhauser shift [6,7,8].

Diagonalization of the spin Hamiltonian, $\langle \hat{H}_{\text{spin}}^{\text{exciton}} \rangle_N$, gives the fine structure of the exciton:

$$E_{\pm}^{(1)} = \frac{1}{2} (\delta_0 \pm \sqrt{h_1^2 + \delta_b^2}),$$

$$E_{\pm}^{(2)} = \frac{1}{2} (-\delta_0 \pm \sqrt{h_2^2 + \delta_d^2}), \quad (4)$$

where $h_n = \mu_B g_n B_{\text{ext}} + (-1)^n A \langle I_z \rangle$, $g_n = g_h + (-1)^n g_e$, and $n = 1$ or 2 . The splitting of both the bright ($n = 1$) and dark ($n = 2$) exciton doublet states, $\Delta E^{(1,2)} = E_+^{(1,2)} - E_-^{(1,2)}$, is determined by the sum ($h_{1,2}$) of an external magnetic field and the effective magnetic field of the nuclei, as well as the anisotropic exchange interaction, $\delta_{b,d}$.

To fully describe the electronic spectra, we must find now the average nuclear spin polarization, $\langle I_z \rangle$, which is determined by the balance between dynamical nuclear polarization and depolarization. These processes are governed by fluctuations of the electron and nuclear polarization from their average values.

Polarization of the nuclear spins by optically oriented excitons arises from the second part of Eq. (3), $\sim \sum_j A^j |\Psi(\mathbf{R}_j)|^2 (I_+^j \sigma_-^e + I_-^j \sigma_+^e)$. These ‘‘flip/flop’’ processes, which vanish in the mean-field approximation, involve the simultaneous spin flip of a nucleus and electron. The electronic spin flip causes the exciton to transform between bright and dark states and requires emission or absorption of energy (δ_0) because of the large mismatch in electronic and nuclear Zeeman energies. Therefore some additional ‘‘assisting’’ process is necessary to satisfy energy conservation (e.g., emission or absorption of a photon or a phonon). Transition rates for flip/flop transitions calculated in second order perturbation theory are δ_s^2/δ_0^2 smaller than those for the assisting processes with no spin flips, where $\delta_s^2 = \nu_0 \int |\psi(\mathbf{r})|^4 d^3 r \sum_j (A^j)^2 I^j (I^j + 1)$, and the sum goes over all nuclei in the unit cell. If the dark exciton lifetime is much longer than the bright exciton lifetime ($\tau_d \gg \tau_b$), nuclear polarization is determined by spin-flip assisted radiative recombination of the dark excitons [21]. We calculate the nuclear spin polarization rate to be $T_e^{-1} = (\delta_s^2/15N\delta_0^2)\tau_b^{-1} \approx (2.5 \text{ s})^{-1}$, where $N \sim 10^5$ nuclei in the QD [11] and $\tau_b \sim 0.1 \text{ ns}$ [22], in agreement with experiment [10].

Coupling of neighboring nuclear spins through the dipole-dipole interaction leads to nuclear spin depolarization. In a magnetic field larger than the dipole field, $B_{\text{ext}} > B_L \sim 0.15 \text{ mT}$, the energy of the nuclear dipole-dipole interaction is not enough to drive the transition between two nuclear spin sublevels split by the Zeeman energy ($\mu_I B_L/I \ll \mu_I B_{\text{ext}}/I$, where μ_I is the magnetic moment of the nucleus), and this mechanism should be negligible. However, fluctuations of the z component of the electron polarization can provide the necessary energy, leading to depolarization

of the total nuclear spin, even in a relatively strong magnetic field. Specifically, our calculations show that the nuclear dipole-dipole interaction weakly mixes the wave functions of different nuclear spin projection states, of order B_L/B_{ext} . Transitions between these mixed states are induced by the hyperfine magnetic field of the electrons, $\nu_0 |\psi(\mathbf{R}_j)|^2 \sigma_z^e A^j / (2\mu_I/I)$, acting on the nucleus, j , during the exciton lifetime. The dark exciton plays the main role in this process because $\tau_d \gg \tau_b$. This leads to a nuclear spin depolarization with a rate calculated in second order perturbation theory to be $T_{e\text{-dip}}^{-1} \approx 0.06\tau_d(\delta_s/\hbar)^2(B_L/B_{\text{ext}})^2N^{-1}$.

The average nuclear polarization $\langle I_z \rangle$ is given by the rate equation [5],

$$\frac{d\langle I_z \rangle}{dt} = -\frac{f_d}{T_e} [\langle I_z \rangle - \langle S_z^{(d)} \rangle Q] - \frac{f_d}{T_{e\text{-dip}}} \langle I_z \rangle, \quad (5)$$

where $\langle S_z^{(d)} \rangle$ is the time-averaged electron spin in the dark exciton state, f_d is the fraction of time that the QD contains a dark exciton, and $Q = I(I+1)/S(S+1) = 5$. The first term describes the probability of nuclear polarization due to the flip-flop hyperfine transitions, and the second is the rate of nuclear spin depolarization due to nuclear dipole-dipole interactions in the fluctuating hyperfine field of the electron.

Substituting for $1/T_{e\text{-dip}}$ and $1/T_e$ in Eq. (5), we obtain the steady state average nuclear spin:

$$\langle I_z \rangle = Q \langle S_z^{(d)} \rangle \frac{B_{\text{ext}}^2}{B_{\text{ext}}^2 + \tilde{\xi} B_L^2}, \quad (6)$$

where $\tilde{\xi} \approx (\delta_0 \sqrt{\tau_d \tau_b} / \hbar)^2$. Using Eq. (6) for the nuclear polarization, the exciton energies were calculated from Eq. (4) and plotted in Figs. 2 and 3, showing good agreement with the data using reasonable fitting values of $\langle S_z^{(d)} \rangle = 0.2$ and $\sqrt{\tau_d \tau_b} = 3.5$ ns. Thus, the nuclear spin polarization tracks the electron spin polarization of the dark exciton except in the strongly depolarized region around zero applied field where it follows a Lorentzian line shape with width, $\sqrt{\tilde{\xi}} B_L$, that is 300 times wider than in bulk GaAs [15].

Direct measurements of fine and hyperfine structure in single GaAs QDs are described well if both nuclear polarization and depolarization processes are included. The measurements show that, although a large fraction of the nuclei have been optically polarized (65%), the rate of nuclear polarization is strongly suppressed by the electron/hole exchange interaction in quantum dots. The depolarization of the nuclei is a result of their heating by temporal fluctuations of the hyperfine field of optically created electrons. To suppress this depolarization, an applied magnetic field much larger than in the bulk is required. This field increases with the magnitude of the exchange interaction and with the lifetimes, and therefore should in-

crease in smaller and more strongly confined QDs such as the self-assembled QDs [12–14].

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