Subsecond Spin Relaxation Times in Quantum Dots at Zero Applied Magnetic Field Due to a Strong Electron-Nuclear Interaction

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A key to ultralong electron spin memory in quantum dots (QDs) at zero magnetic field is the polarization of the nuclei, such that the electron spin is stabilized along the average nuclear magnetic field. We demonstrate that spin-polarized electrons in n-doped (In, Ga)As/GaAs QDs align the nuclear field via the hyperfine interaction. A feedback onto the electrons occurs, leading to stabilization of their polarization due to formation of a nuclear spin polaron [I. A. Merkulov, Phys. Solid State **40**, 930 (1998).]. Spin depolarization of both systems is consequently greatly reduced, and spin memory of the coupled electron-nuclear spin system is retained over 0.3 sec at temperature of 2 K.

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New impetus for the use of quantum dots (QDs) in quantum information processing [1,2] has recently been gained by reports of long electron (e) spin relaxation times, namely, coherence times T_2 of several μs [3,4] and lifetimes T_1 of several ms [5,6]. With techniques to charge QDs with spin-polarized e's [5–10], the e spin appears to be a promising qubit candidate. The most important e-spin dephasing mechanism in QDs is predicted to be the hyperfine interaction with the nuclei [11]. Precession around a randomly oriented nuclear field results in partial loss of e-spin orientation over a ns time scale [12], followed by a total loss of alignment over a μs time scale, due to fluctuations of the nuclear spins [11].

In this Letter we demonstrate for singly charged QDs a greatly increased spin lifetime of the coupled e-nuclear spin systems on the subsecond scale. We find experimentally that at zero magnetic field the e-nuclear spin system retains a memory of its optical orientation and the polarization vector (direction of orientation) on time scales far beyond the typical spin relaxation times of a nuclear system dominated by dipole-dipole interaction $T_{2,dd} \approx 10^{-4}$ s. The theoretical analysis shows that formation of a nuclear spin polaron (NSP) [13] controls the spin dynamics in singly charged QDs.

The samples studied contained 20 layers of (In, Ga)As/GaAs self-assembled QDs. Results from two samples A and B, thermally annealed at 945 and 900 °C, with ground state emission energies at 1.42 and 1.34 eV, respectively, are shown here. The structures were n-doped 20 nm below each dot layer with a dopant density about equal to the dot density, such that each dot was occupied with on average one e. The measurements were performed at temperatures $T = 2$ and 6.6 K in an optical cryostat, which was placed inside three pairs of Helmholtz coils, oriented mutually orthogonal to each other. These coils were used for compensating parasitic magnetic fields (e.g., the geomagnetic field) down to <5 μT, and also to apply fields up to 150 mT perpendicular to the optical axis (Voigt geometry). The QDs were excited with a mode-locked Ti:sapphire laser emitting pulses of 1.5 ps duration separated by 13.2 ns. Additionally, a combination of an acousto-optical and an electro-optical modulator was used to form pulse trains with duration from 110 μs to 500 ms of either σ^+ or σ^- circular polarization. The excitation beam was focused to a spot size of ~200 μm diameter. The QD photoluminescence (PL) dispersed by a monochromator was detected circular polarization-selective with a CCD camera or an avalanche Si-photodiode (APD).

Figure 1(a) shows the PL of QD Sample A after excitation with σ^+ polarized light in the wetting layer at 1.476 eV, detecting both copolarized ($\sigma^+\sigma^+$) and cross polarized ($\sigma^+\sigma^-$) to the excitation. The ground state PL polarization is negative, i.e., the QDs preferentially emit photons with the opposite polarity to the exciting light. The negative circular polarization (NCP) increases for higher excitation density, reaching saturation at -28% for ~20 W/cm^2 [see Fig. 1(b)].

This NCP is a well-established observation in QDs charged with a single e [7–10,14]. It is beyond the scope of this Letter to go into the details of the models explaining NCP. Instead, we note here only the facts needed for further discussion. NCP appears in the recombination of a trion singlet state, for which the total e spin is zero ($\mathbf{s}_1 + \mathbf{s}_2 = \mathbf{0}$). It is governed by the hole in the QD ground state, whose optical spin orientation is lost during relaxation, but which gains spin orientation while in the QD ground state via exchange with an optically oriented e [8]. Recombination of the hole with one of the trion e's results in a photon with polarization opposite to the excitation light. Furthermore, the polarization of the recombined hole is equal to the polarization of the resident e left in the dot after trion recombination. We will show below that this

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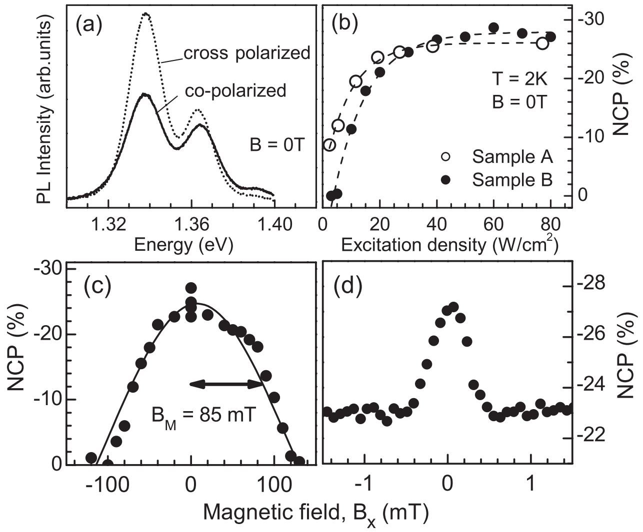

FIG. 1. (a) Polarization-selective PL spectra of sample B, with σ^+ polarized excitation. Detection was σ^+ or σ^-, measured with the CCD. (b) NCP at peak PL intensity vs excitation density for samples A (open circles) and B (closed circles). NCP for sample B at energy 1.34 eV as function of applied Voigt field B_x over (c) 100 mT and (d) 1 mT range. Lines in (b),(c) are guides to the eye. $T = 2$ K.

resident *e* transfers the polarization obtained from photoexcitation to the nuclear spin system. Thus, the hole has two roles. First, it is an important participant of the process which polarizes the *e* spin. Second, it serves as detector of the resident *e* polarization via the NCP measurement.

After trion recombination, the resident *e* spin loses 2/3 of its polarization due to precession about the randomly oriented fluctuations of the nuclear field [11]. However, polarization of the nuclei along the *e* spin direction reduces the effect of the fluctuations, which can be detected by an increased NCP value. This is confirmed by the Hanle effect experiments shown in Fig. 1. Figure 1(c) shows the dependence of NCP, measured at 1.34 eV with the APD, on a magnetic field oriented perpendicular to the polarized *e* spin. The circular polarization degree of the PL has a complex behavior, which is similar to the one reported for an *e* localized on a donor in bulk GaAs (see Fig. 5.7 in Ref. [15]). The polarization decreases sharply in weak magnetic fields $B \geq B_L \approx 0.3$ mT about equal to the local magnetic fields between neighboring nuclei B_L [Fig. 1(d)]. This depolarization is commonly attributed to amplification of the external magnetic field effect by the hyperfine field of polarized nuclei. The narrow Hanle curve is superimposed on a broader background with a half width at a half maximum of $B_M \approx 85$ mT [Fig. 1(c)]. This slow depolarization is due to a steady decrease of the nuclear hyperfine field $\mathbf{B}_N$. As a result, the total in-plane component of the magnetic field $\mathbf{B}_N + \mathbf{B}$, which controls the *e* precession, increases more slowly than the external field $\mathbf{B}$. It is worthwhile to note, that the hole spin in the QD ground state is not affected by the hyperfine nuclear field and only weakly influenced by the external magnetic field because of its small in-plane *g* factor. Therefore, Hanle depolarization curves reflect the resident *e*'s behavior.

We conclude from the results of Fig. 1 that the *e* and nuclear spin systems are highly codependent. Moreover, the nuclear polarization achieved by interaction with spin-oriented resident *e*'s can be detected via NCP. To investigate the dynamics of the coupled *e*-nuclear spin system in QDs at $B = 0$ T, we add additional modulation to the pulsed excitation over time scales for the hyperfine interaction to act efficiently. The sample is excited by trains of pulses, with train duration from 110 μs to 500 ms, i.e., each pump train contains between 10^4 and 10^7 pulses. The inset of Fig. 2(a) shows the illumination scheme: the sample is excited for a time Δt_{ill} by a first pulse train (pump 1), and then by a second train (pump 2), with a dark period between the trains of duration Δt_{dark}. The σ^+ helicity of pump 2 was kept constant, and its NCP monitored. The polarization of pump 1 was changed from σ^+ (copolarized) to σ^- (cross polarized).

Resident *e*'s are polarized on a time scale much shorter than the train duration, but nuclear polarization occurs over

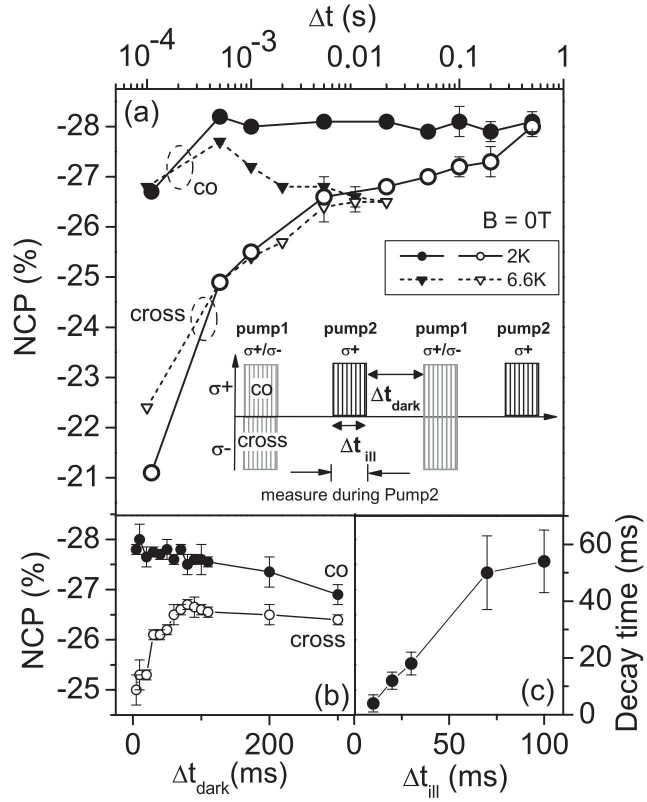

FIG. 2. (a) NCP of sample B vs modulation time $\Delta t = \Delta t_{\text{ill}} = \Delta t_{\text{dark}}$ for pumps 1 and 2 copolarized (full symbols) and cross polarized (open symbols) at 2 (circles) and 6.6 K (triangles). Power density $\sim$50 W/cm^2. Inset: Excitation scheme for co- and cross-polarized pumps 1 and 2. NCP is measured during pump 2. (b) NCP as a function of dark time Δt_{dark}, for illumination time $\Delta t_{\text{ill}} = 100$ ms. (c) Decay time of NCP memory [NCP(co)—NCP(cross)] as function of Δt_{ill}.

comparable times. Therefore, for copolarized excitation, both pumps reinforce each other to allow maximum nuclear polarization, whereas in the cross-polarized case the two pumps compete, reversing each other's nuclear polarization. The polarization established during illumination decays in the dark periods between trains and depends on both $\Delta t_{\rm ill}$ and $\Delta t_{\rm dark}$.

Figure 2(a) shows NCP values at $T = 2$ K (circles), for the case $\Delta t_{\rm ill} = \Delta t_{\rm dark} = \Delta t$. For the copolarized protocol, the NCP is saturated at $\sim -28\%$ for $\Delta t > 500$ μs. The illumination time is long enough, and the nuclear spin decay slow enough, to allow nuclear spin to accumulate between pulse trains and reach saturation. For the cross-polarized protocol, however, the nuclear polarization from pump 1 acts detrimentally on pump 2. A longer illumination time is then required to fully invert the nuclear system and reach saturation, as evidenced by the increase in NCP from -21 to -28% over the time scale $10^{-4} < \Delta t < 1$ s.

7A significant difference between the NCP achieved for copolarized and cross-polarized pumps 1 and 2 is seen for all values of Δt up to 0.2 s, which exceeds the nuclear dipole-dipole interaction time, $T_{2,dd} \approx 10^{-4}$ s, by 3 orders of magnitude. This difference is a clear indication of spin memory in the system: the polarization of pump 2 does not change in either protocol, therefore any observed change must be due to spin memory of the polarization sign of pump 1, which has persisted during the dark time $\Delta t_{\rm dark}$. An increase in lattice temperature induces a shortening of the spin memory times. Figure 2(a) (triangles) shows data taken at $T = 6.6$ K. The difference between the co and cross protocols is now only present up to ~2 ms: a reduction in time by 2 orders of magnitude compared to 2 K is observed.

It is a remarkable experimental result that at zero magnetic field the coupled e-nuclear spin system sustains on a subsecond time scale not only the memory about its exposure to circularly polarized light (which leads to a decrease of the nuclear spin temperature relaxing with the energy relaxation time T_1), but also about the sign of the light polarization. This means that the transverse relaxation time T_2 of the e-nuclear spin system polarization vector in a QD should be in the subsecond range.

A more direct measure of the spin memory time is gained by keeping $\Delta t_{\rm ill}$ constant, and varying $\Delta t_{\rm dark}$. Figure 2(b) shows that with increasing $\Delta t_{\rm dark}$ the copolarized NCP decreases and the cross-polarized NCP increases. While the exact dynamics require further investigation, one may phenomenologically fit an exponential decay to the difference [NCP(co)-NCP(cross)]. Figure 2(c) shows this decay time as a function of illumination time varied from 10 to 100 ms. A clear monotonic increase of the decay time from ~4 to 50 ms is observed as $\Delta t_{\rm ill}$ is increased. In fact, the spin dynamics also contains a long-lived component of at least 0.3 sec, seen in Fig. 2(b) as difference between the co and cross protocols at $\Delta t_{\rm dark} = 300$ ms. Let us now analyze the experimental findings:

(1) Deceleration of e spin relaxation in crystals exposed to circularly polarized light is well established [16], and originates from mutual interaction of e and nuclear spins. It is well known that the e spin is transferred to the nuclear spin system by the hyperfine interaction (see Ch. 2 in Refs. [15,17]):

$$\langle \mathbf{I} \rangle = f \frac{I(I+1)}{s(s+1)} \frac{(\langle \mathbf{s} \rangle \cdot \mathbf{B}_e)\mathbf{B}_e}{B_e^2 + \tilde{B}_L^2}. \quad (1)$$

Here $\langle \mathbf{I} \rangle$ and $\langle \mathbf{s} \rangle$ are the mean spin polarization vectors of the optically oriented nuclei and e's, with spins I and s, respectively, ($I_{\rm As} = I_{\rm Ga} = 3/2, I_{\rm In} = 9/2, s = 1/2$). $\mathbf{B}_e = b_e \langle \mathbf{s} \rangle$ is the mean e hyperfine field, and $\tilde{B}_L$ is a parameter which is of the same order of magnitude as the nuclear fluctuation field $B_L \approx 0.3$ mT, caused by dipole-dipole interactions. $f \leq 1$ is a phenomenological leakage factor accounting for additional channels of nuclear spin temperature relaxation. b_e depends on the dot size and for our case can be estimated as 10–30 mT [17]. $\langle s \rangle \approx 0.28 \cdot s$ can be taken from the experimental value for the NCP saturation level of 28%. Therefore, $B_e \gg \tilde{B}_L$ and, according to Eq. (1), the e field does not affect the nuclear polarization $\rho = \langle \mathbf{I} \rangle / I$. We estimate the nuclear polarization for Ga and As as $\rho_{3/2} \approx f \times 0.47$ and for In as $\rho_{9/2} \approx f \times 1$.

The leakage factor may be evaluated from the linewidth of the Hanle curve $B_M = 85$ mT in Fig. 1(c). It follows from section 2.3 of Ref. [15], that for a relatively high nuclear polarization, B_M is about equal to the geometrical mean of the e and nuclear fields:

$$B_M^2 \approx \frac{4}{3} f \langle s_0 \rangle^2 \left| b_e \sum_i b_{N,i}(I_i + 1) \right|. \quad (2)$$

Here $b_{N,i}$ is the nuclear hyperfine field acting on the e for 100% polarization of type i nuclei. $\langle s_0 \rangle$ is the spin of the oriented e's at $B = 0$ T. The evaluation for $In_{0.5}Ga_{0.5}As$ QDs with $b_{N,{\rm In}50\%} \approx -4.3$ T [18], $b_{N,{\rm As}} \approx -2.76$ T [19], and $b_{N,{\rm Ga}50\%} \approx -1.26$ T gives $\sum b_{N,i}(I_i + 1) \approx -33.7$ T, from which we obtain $f \approx 0.5$, which is of the order of unity and, therefore, the nuclear polarization is very high [20]. It exceeds the fluctuations of the hyperfine nuclear field by 2 orders of magnitude and therefore should suppress the e spin relaxation by these fluctuations.

(2) In general it would not be surprising that illumination with fixed polarization results in an e polarization different from that for alternating polarization. It is known that an oscillating e field caused by a change of the sign of light polarization leads to heating of the nuclear spin system and decreases the nuclear polarization [15,21]. But in our experiments $\Delta t_{\rm dark} \gg T_{2,dd}$ and therefore the cooling efficiency of the nuclear system is the same for copolarized and cross-polarized illumination. In addition, the heating of the nuclear spin system, caused by the fast changes of the e fields at the fronts of the pump trains, should be independent of polarization sign if the dark intervals exceed $T_{2,dd}$.

The experimental observation of a preserved polarization vector for times greatly exceeding the dipole relaxation time of the nuclear system $T_{2,dd} \approx 10^{-4}$ s is very unlikely from the viewpoint of the classical theory of optical orientation of the e-nuclear spin system. Such long times are characteristic of energy relaxation T_1, responsible for equalizing an inverted nuclear spin temperature β [15,22]. But for nuclear polarization by the e field $\beta \propto (\langle s \rangle \cdot B_e) \propto \langle s \rangle^2$ is proportional to the square of the mean e spin and therefore does not hold information about the sign of the light polarization, i.e., about the direction of the polarization vector.

(3) We explain the giant deceleration of the e-nuclear spin relaxation dynamics as the formation of a nuclear spin polaron [13] (in analogy with magnetic polarons in diluted magnetic semiconductors [23]). From our estimation of the nuclear hyperfine field follows a splitting of the e spin levels in this field of about 2 K. This means that the oriented nuclei must induce a considerable quasiequilibrium polarization of the resident e in a QD, which in turn will prevent the nuclear polarization from relaxation. A self-consistent e-nuclear spin complex, known as a nuclear spin polaron, should be formed.

The NSP loses memory of the initial orientation direction of its macroscopically large spin over times considerably exceeding $T_{2,dd} \approx 10^{-4}$ s. The total NSP spin $\mathbf{J}_\Sigma \propto \langle \mathbf{I} \rangle N$ greatly exceeds the fluctuation of nuclear spins $J_{\rm Fl} \propto I\sqrt{N}$ in the QD. Here $N \approx 10^5$ is the number of nuclei in the e-nuclear spin complex. In dark the J_Σ value is retained by the local hyperfine field of the resident e in a QD, and changes of its orientation are provided by magnetodipole interactions among the nuclei. During the $T_{2,dd}$ time, $\mathbf{J}_\Sigma$ rotates by an arbitrary angle $\delta\varphi \propto J_{\rm Fl}/J_\Sigma$, and, averaged over all events, the projection of $\mathbf{J}_\Sigma$ onto its initial direction is decreased by $\langle \delta \mathbf{J}_\Sigma \rangle \approx -\langle \mathbf{J}_\Sigma \rangle (J_{\rm Fl}^2/2J_\Sigma^2)$. Therefore, the directional relaxation time of the NSP can be evaluated as [13,23]

$$T_2^{\rm (NSP)} \approx f^2 \frac{6I(I+1)\langle s \rangle^2}{s^2(s+1)^2} N T_{2,dd}. \tag{3}$$

Keeping in mind the macroscopically large N value and substituting in Eq. (3) the minimal possible value of the nuclear spin we get $T_2^{\rm (NSP)}/T_{2,dd} > 10^4$. Therefore, the time of dipole relaxation of the NSP polarization lies in the seconds range, which is in good agreement with experiment and, in turn, substantiates the validity of the applied model. The validity of the NSP model is additionally supported by the strong temperature dependence of the spin memory time shown in Fig. 2(a). A temperature increase up to 6.6 K erases the long-lived memory, which allows us to dismiss a possible explanation of the effect by quadrupole splittings of nuclear spin levels in alloys [15].

The estimations performed are based on equations derived in the high-temperature approximation, for which a short correlation time of e and nuclear spins is assumed. We conclude a very strong polarization of the nuclear spin system and the presence of a strongly correlated regime in the e-nuclear spin system. A proper accounting of all details of this regime with long correlation times may change the estimated values, but does not change the qualitative conclusion about a huge deceleration of the directional NSP spin relaxation.

To conclude, we have shown experimentally and theoretically the formation and stability of a nuclear spin polaron in a QD charged with a single electron. The polaron formation occurs in zero external magnetic field and results in a subsecond electron spin memory.

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