Millisecond spin lifetimes in quantum dots at zero applied magnetic field due to strong electron-nuclear interaction

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A key to achieving ultra-long electron spin memory in quantum dots (QDs) at 0 T is the polarization of the nuclei, such that the electron spin is stabilized along the nuclear magnetic field. We demonstrate that spin-polarized electrons in n-doped QDs align the nuclear field via the hyperfine interaction. A feedback onto the electrons occurs, leading to stabilization of electron polarization. We suggest that the coupled electron-nuclear system forms a rigid nuclear spin polaron state as predicted by I.A. Merkulov [1], for which spin memory is retained over millisecond lifetimes.

New impetus for the use of QDs in quantum information processing has recently been gained by reports of long electron (e) spin relaxation times: coherence times $T_2$ of several $\mu$s [4, 5] and lifetimes $T_1$ of several ms [6] have been demonstrated. With techniques to charge QDs with spin polarized e’s, it appears that the e spin is a promising candidate for a qu-bit. Such a stable e-spin results from the suppression of spin relaxation mechanisms present in higher dimensionality structures. An interaction that does not become inefficient for QDs, however, and predicted to be the limiting factor for e-spin orientation, is the hyperfine interaction with the nuclei [11]. While only a partial dephasing of the e spins occurs over a $\mu$s timescale [12], total loss of e spin alignment eventually occurs due to fluctuations of the randomly oriented nuclei.

The spin dynamics in QDs are rather complex, as they are governed by multiple interactions, as sketched in Fig. 1(a). Confined e’s interact with the nuclei via the hyperfine interaction: An e spin exerts a magnetic field $b_e$ (called Knight field) onto the nuclei $\mathbf{b}_e$, which in combination act in turn on the e (called Overhauser effect) to give a magnetic field $\mathbf{B}_N = \sum_i^N \mathbf{b}_{Ni}$, where the sum is over all N nuclei in the QD. This hyperfine interaction has been shown to lead to transfer of angular momentum from the e to the nuclei via a spin “flip-flop”, a process which occurs over a typical timescale of $10^{-2}$ s [13]. The magnitude of the total nuclear alignment achieved, on the other hand, is influenced by the magnetic dipole-dipole interaction between the nuclei, the strongest depolarization process, acting on time scales of $10^{-4}$ s. This interaction may be switched off by applying an external magnetic field, such that the nuclear Zeeman splitting is larger than the dipole-dipole interaction [13]. The nuclear interaction with the lattice is the weakest of all, leading to a spin-lattice relaxation time of seconds or longer, and is therefore neglected in this paper.

Magnitude and direction of the nuclear field $\mathbf{B}_N$ depend on the nuclear polarization degree. For typically $\sim 10^5$ nuclei in a self-assembled QD, $B_N \gg b_e$, and therefore the nuclear precession frequency is orders of magnitude smaller than that of the e, so that over a timescale of $\sim \mu$s the e experiences a constant, ”frozen fluctuation” field $\mathbf{B}_N$ with a field strength of about 30 mT [14]. Due to the e precession, starting from an initial spin orientation $S_0$, the spin polarization retained is the projection $\langle S_z \rangle = S_0 \cos^2 \theta$ (see Fig. 1b) [11, 12, 15]. An ensemble measurement probes a distribution of $B_N$, and the e spin-phase between QDs is quickly lost, so that only an average spin $S_0/3$ is preserved [11]. The key to increasing the spin polarization therefore is to avoid this e spin precession: either a strong magnetic field needs to be applied [12] or the nuclear spins should be aligned, such that $S_0 \cos^2 \theta = S_0$.

Nuclear polarization may be achieved using the Overhauser effect [12, 16, 15], where the QD is constantly optically pumped with spin polarized excitons. The nuclei experience the Knight field from the e, and, as a result of a sequential multiple flip-flop processes, begin to align along the e spin polarization axis. Generally, an external magnetic field should be applied, leading to a splitting of the nuclear spin levels above the energy for the nuclear dipole-dipole interaction to occur. However, for QDs, the Knight field $b_e$ at the site of a nucleus is comparable with the field induced by a neighbouring nucleus [16]. Thus $b_e$ alone may induce a nuclear Zeeman splitting that effectively allows the hyperfine interaction to compete with the dipole-dipole interaction, so that nuclear polarization may occur [17]. However, if one introduces e’s by injecting neutral excitons, the Knight field vanishes with the radiative decay of the exciton.

Here we take a different, all-optical approach, by which polarization of the e-nuclei system is achieved even without external magnetic field. A permanent field $b_e$ is maintained by resident e’s in the QDs. These e’s are oriented by circularly polarized laser excitation, for which we exploit an optical pumping mechanism unique for n-doped QDs [4, 5]. The nuclei are polarized through the hyperfine interaction, and as a result of a feedback by the nuclei, greatly stabilized e polarization is achieved. Both
spin systems are aligned such that the fluctuations of the interacting e-nuclear system may be low enough to allow the formation of a nuclear polaron state \( \sigma^+ \); the spin lifetime of this collective state is increased over orders of magnitude to the millisecond scale.

The studied samples contained 20 layers of (In,Ga)As/GaAs self-assembled QDs. A range of structures was addressed, results from two of which are presented here: Samples A and B, thermally annealed at 945 and 900°C, with ground state emission energies at 1.42 and 1.34 meV, respectively. 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. All measurements were performed at a temperature of \( T = 2 \) 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 \(< 1 \) μT, and also to apply fields in the Voigt/Faraday geometries. The QDs were excited with a mode-locked Ti:Sapphire laser emitting pulses of width \(< 1 \) ps separated by \(< 13 \) ns. The excitation beam was focused to a spot size of \(< 200\)µm diameter, the QD photoluminescence (PL) dispersed by a monochromator and resolved with a CCD camera, an avalanche Si-photodiode (APD) or a streak camera with time resolution 30 ps.

Fig. 1(c) shows PL of the QDs after excitation with \( \sigma^+ \) polarized light in the wetting layer at 1.476 eV, detecting both co- (\( \sigma^+ \sigma^+ \)) and cross-polarized (\( \sigma^+ \sigma^- \)) to the excitation with the CCD. The ground state PL polarization, at maximum 1.34 eV, is negative, i.e. the QDs preferentially emit photons with the opposite polarity to the exciting light. This negative circular polarization (NCP) is now a well-established phenomenon in n-doped QDs [7, 8, 9, 18].

The key feature of this phenomenon is that the resident e’s become polarized. While the polarization of these resident e’s in the ensemble may be low after a single pulse, several successive pulses allow for strong accumulation of the e spin polarization, as reflected in the NCP value. Its dependence on excitation power is given in Fig. 1(d); at low powers it increases, until saturation is reached at \(< 20 \) Wcm\(^{-2}\).

Under these conditions, the system is prepared for polarization of the nuclear system, for which we envisage the following scheme: After recombination of the trion (\(< 1 \) ns), and before the next laser pulse (\(< 13 \) ns), the QD contains a single polarized e, which may transfer angular momentum in form of a flip-flop process to the nucleus. At the same time the e maintains an internal magnetic field that produces a nuclear Zeeman splitting strong enough to suppress nuclear dipole-dipole dephasing. A following laser pulse then reorients the e spin, such that successive multiple pulses orient a significant number of nuclei and decrease the temperature of the nuclear system.

The polarization of the nuclei leads to a total field \( \mathbf{B}_N \) whose angle to the z-axis, \( \theta \) is small. The NCP measured, determined by \( \langle S_z \rangle = S_0 \cos^2 \theta \), should increase if there is a feedback onto the e. We first investigate whether the nuclei indeed affect the NCP, or whether it can be explained solely by the spin-polarized excitation of the e’s. The Knight field \( \mathbf{b}_K \) can be estimated to be of the order of 0.1 - 1 mT [10]. If relevant for creating a nuclear polarization, NCP should be strongly affected by external fields of comparable magnitude. Fig. 2(a) shows the influence of a much stronger field (100 mT range) applied in Voigt geometry perpendicular to the polarized e spin. The NCP reduces for field values \(< 80 \) mT due to several processes, such as the e precession about the external field (Hanle effect), and also the reduction of the anisotropic exchange [11].

We turn now to the behavior of the NCP for very low fields \(< 2 \) mT. As can be seen in Fig. 2 the NCP value drops from a value of \(< -28 \% \) to a value of \(< -23 \% \) with a half-width at half maximum of just 0.25 mT. Such low fields have a negligible direct effect on the carriers (for example, the Zeeman splitting induced is \(< 10 \) neV). The strong reduction of the NCP over this small field range is a consequence of the sensitivity of the nuclei to fields comparable to that of the Knight field.

This rather surprising effect results from a nuclear field, \( \mathbf{B}_N \), on the e that effectively multiplies the effect of the external field by several orders of magnitude, and may be explained as follows. When an external Voigt field is applied, the nuclei experience the vector addition of the Knight and the external fields, \( \mathbf{b}_F = \mathbf{b}_K + \mathbf{b}_T \), as shown schematically in the inset of Fig. 2(b). The nuclear field, \( \mathbf{B}_N \), is parallel to \( \mathbf{b}_T \). The strength of
B_N is several orders of magnitude larger than field b_T. As a consequence, optically oriented e’s precess around this field in a similar way to the behavior observed in Fig. 2(a), so that depolarization occurs. For b_x >> b_z, which is valid in our case for b_x > 1 mT, the precession direction will be very close to the x-axis. As one can see in Fig. 2(b), this NCP depolarization thus becomes saturated at b_x > 1 mT. A similar effect has been observed for e’s localised on donors in GaAs [16].

We therefore observe that, while at b_x = 0 T the co-aligned e-nuclear system leads to an increase in NCP, a strongly obliquely aligned nuclear field has a detrimental effect, causing a decrease in NCP to a value that is actually lower than if the nuclei were not polarized. Fig. 2(c) demonstrates this: a constant Voigt geometry field of b_z = 0.6 mT is applied, resulting in a strongly obliquely aligned nuclear field (see the value at 0.6 mT in Fig. 2(b)). A Faraday field b_z is then applied, and for values of b_z > b_x the polarization is restored to ~32%. From the experimental data at high values of b_z, the expected NCP is extrapolated to zero b_z field (dotted line): a value of ~ -24% is expected at b_z = 0 T. The experimental data deviates strongly from this behavior however, and, moreover, exhibits an asymmetry that is reversed upon reversal of the excitation helicity.

This asymmetry results from the partial cancellation of the Knight field, and therefore depolarization of the nuclei. At a certain point, the Faraday field b_z is equal and opposite to the Knight field b_z. The Knight field is necessary for overcoming the dipole-dipole interaction, and thus when it is cancelled the nuclei quickly depolarize. The result is in fact an increase in NCP as the nuclei depolarize: without the strong oblique field from the nuclei the e’s retain a greater polarization. Verification of this is gained by the reversal in asymmetry when exciting with σ^+ (solid black circles) or σ^- (open grey circles): reversal of helicity results in reversal of the Knight field. Note that a fairly broad resonance is observed: as each nucleus in the QD feels a different value of Knight field, the entire nuclear system does not become simultaneously depolarized. An estimate of the Knight field strength of ~1 mT may however be gained from the data, which agrees well with that observed in the similar system of e’s localized on donors in GaAs [16].

We observe from the results in Fig. 2 that the e and nuclear spin systems are highly co-dependent. I. A. Merkulov [1] makes the suggestion that in a strongly cooled nuclear system (a nuclear polaron) a giant increase of the nuclear spin relaxation lifetime should be observed. As the e polarization is strongly dependent on the feedback from the nuclei, its spin lifetime may also be increased. To investigate these spin lifetimes, we modulate the excitation over time scales for the hyperfine interaction to act efficiently. The sample is excited by trains of pulses (with a 13 ns pulse separation) which have duration from 20 μs to 12 ms. The modulation is done by either a photoelastic modulator at 50 kHz or by a mechanical chopper at 10 - 10^3 Hz. Fig. 3(a) shows the illumination scheme: the sample is excited for a time Δt by a first train of pulses (pump 1), and then by a second train (pump 2), with a dark period between the trains of the same duration, Δt.

Pumps 1 and 2 are in addition delayed by 3 ns with respect to each other during the 13 ns laser repetition rate, allowing the PL from each to be separated using a streak camera. NCP was analysed from the time-resolved PL spectra after the initial PL risetime. The helicity of Pump 2 was kept at σ+ and its NCP monitored as the polarization of Pump 1 is changed from σ+ (co-polarized) to σ− (cross-polarized). Due to the long Δt, each pump train contains between 10^3 and 10^6 individual pulses. Without nuclear effects, we presume that the resident e spin polarization in the ensemble reaches a steady state value well before the end of the pulse train (remember NCP saturation as function of pump power). However, due to the hyperfine interaction induced nuclear polarization occurring over timescale of 10^{-2} s, influencing the back-coupling to the e, we expect a strong dependence of NCP on illumination time Δt. For co-polarized excitation, both pumps reinforce each others’ orientation to allow maximum polarization, whereas in the cross-polarized case the two pumps act in competition.

Fig. 3(b) shows the NCP values as function of modulation time Δt. Let us first consider two limiting cases: The dotted line indicates the NCP value (~ -17%) obtained when Pumps 1 and 2 are cross-polarized, and modulated at 50 kHz. During this Δt (~ 1000 pulses) the nuclei experience a field b_e that fluctuates over the timescale.
of 20 µs, so that the nuclei should not become polarized. Instead, the e’s experience a randomly polarized nuclear field of ~ 30 mT. In contrast, the dashed line indicates the NCP achieved without dark time and co-circular polarization, exploiting the hyperfine interaction to its full degree. This value gives the maximum NCP achievable of ~28% at B = 0 T. Thus, the e polarization is almost doubled by the nuclear polarization.

Let us now consider the NCP achieved for intermediate modulation times Δt. The squares in Fig. 3(b) show the NCP for co-polarized pumps. The polarization increases from ~ −17%, to ~ −23%. As Δt ranges from below to above the time scale for hyperfine effects (10−2 s), this NCP increase is expected. At the beginning of illumination the spin-polarized e’s begin to polarize the nuclei through flip-flop processes. The longer the illumination lasts, the more nuclear polarization will accumulate, which in turn stabilizes the e polarization, determining also the NCP.

During the dark time, however, some nuclear spin polarization is lost. If full nuclear polarization were retained during the dark time, it would accumulate over the pump pulse cycles in the co-polarized case, and reach the un-modulated value of ~ −28%. If on the other hand no polarization memory remained from the last pump, the NCP achieved would be identical for both co and cross-pumping protocols. That some nuclear polarization is maintained can be seen in Fig. 3(b) from the comparison of the data for co- and cross-polarized excitation. A difference in NCP is evident even for Δt = 20 ms. From the decrease of the difference in NCP for both protocols a spin memory time in the ms-range may be evaluated.

To investigate this long spin memory, let us consider the spin dynamics during the dark time. If there were no nuclear polarization, the e in each dot would precess about the random nuclear field, which fluctuates on a µs scale; on a ms timescale all e polarization would be lost. Due to the presence of finite nuclear polarization along the optical propagation direction, the influence of the random field is reduced, and the e spin is stabilized. This is in turn important for the nuclei: an e spin depolarization would immediately reduce the nuclear polarization, as has been demonstrated in Fig. 3. The spin lifetime lies therefore for both e and nuclei in the ms-range due to mutual stabilization. This is orders of magnitude longer than for either decoupled system.

Under conditions of a cooled nuclear spin system, spin fluctuations of both nuclei and e should be greatly reduced. Theoretical estimates suggest that after nuclear cooling by optical pumping, a nuclear spin polaron may be formed for localized e’s at crystal temperatures of 5 K and e spin polarization > 20%, close to the situation in our experiment. We suggest therefore, that a nuclear polaron is formed between the e and the nuclei in the QD due to their strong interaction, which is, however quite fragile: A small external perturbation to this system results in strong depolarization of the e and nuclei. It appears therefore that the predicted "giant increase of the nuclear spin relaxation time" suggested in Ref. is responsible for the millisecond NCP lifetimes measured, and is therefore an extremely good indication that a nuclear spin polaron is formed in this system.

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