Influence of the nuclear electric quadrupolar interaction on the coherence time of hole- and electron-spins confined in semiconductor quantum dots

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The real-time spin dynamics and the spin noise spectra are calculated for p and n-charged quantum dots within an anisotropic central spin model extended by additional nuclear electric quadrupolar interactions (QC) and augmented by experimental data studied using identical excitation conditions. Using realistic estimates for the distribution of coupling constants including an anisotropy parameter, we show that the characteristic long time scale is of the same order for electron and hole spins strongly determined by the QC even though the analytical form of the spin decay differs significantly consistent with our measurements. The low frequency part of the electron spin noise spectrum is approximately 1/3 smaller than those for hole spins as a consequence of the spectral sum rule and the different spectral shapes. This is confirmed by our experimental spectra measured on both types of quantum dot ensembles in the low power limit of the probe laser.

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Introduction: The promising perspective of combining traditional electronics with novel spintronics devices lead to intensive studies of the spin dynamics of a single electron (n) or hole (p) confined in a semiconductor quantum dot (QD) [1–4]. In contrast to defects in diamonds [5, 6], such QDs may be integrated into conventional semiconductor devices. While the strong confinement of the electronic wave function in QDs reduces the interaction with the environment and suppresses electronic decoherence mechanisms, it simultaneously enhances the hyperfine interaction between the confined electronic spin and the nuclear spin bath formed by the underlying lattice.

Generally it is believed [2, 4, 7, 8] that the hyperfine interaction dominates the spin relaxation in QDs. The s-wave character of the electron-wave function at the nuclei leads to an isotropic central spin model (CSM) [7] for describing the electron-nuclear hyperfine coupling, while for p-charged QDs, the couplings to the nuclear spins can be mapped onto an anisotropic CSM [4, 10]. Since the coupling constants for p-charged QDs are reduced compared to the n-charged QDs [4, 10], and additionally a large anisotropy factor $\Lambda > 1$ suppresses the spin decay of the $S_z$ component [4, 10], p-charged QDs have been considered as prime candidates for long lived spin excitations in spintronics applications.

Experimentally, however, there is evidence for comparable spin-decay times of the $S_z$ components [11, 15] in p- and n-charged QDs: hence the anisotropic CSM provides only an incomplete description of the relevant spin-relaxation processes in such systems.

In this paper, we resolve this puzzle by investigating the effect of an additional realistic nuclear electric quadrupolar interaction term (QC) [16] onto the spin decoherence. Most of the Ga and As isotopes have a nuclear spin $I = \frac{3}{2}$ which is subject to a quadrupolar splitting in electric field gradients that occur in self-assembled QDs by construction and couple to the quadrupole moment of the nuclei [16]. While previously simplified assumptions have been made [17, 19], or the problem has been mapped on an effective $I = \frac{1}{2}$ model in a random magnetic field [20] which does not capture the full dynamics, we have taken into account the proper independent anisotropy and realistic strain field orientations estimated by a recent microscopic calculation [21]. Although the short-time dynamics of p- and n-charged QDs are significantly different [4, 22], we show that the long time dynamics is governed by the same time scale set by the quadrupolar interactions in agreement with our experimental data presented below.

Over the last decade, an intuitive picture for the central spin dynamics interacting isotropically with a spin bath via hyperfine interaction has emerged. The separation of time scales [4] – a fast electronic precession around an effective nuclear magnetic field, and slow nuclear spin precessions around the fluctuating electronic spin – has motivated various semiclassical approximations [1, 7, 20, 23, 27] which describe the short-time dynamics of the central spin polarization very well. As can be shown rigorously [20] the CSM predicts a finite non-decaying spin polarization [7, 27] whose lower bound depends on the distribution function of the hyperfine couplings and is only limited to conservation laws. In semi-classical theories [7, 27], it is given by a third of the initial spin polarization leading to a large spectral weight at zero-frequency in the spin-noise spectrum. The absence of such a zero-frequency contribution in experiments [11, 29, 31] provides strong evidence that the CSM is incomplete and additional interactions such as QC play an important role in the decoherence mechanism.

In this work, we have employed a fully quantum mechanical approach, based on a Chebyshev polynomial
technique (CET)\cite{22,34}, to an extended anisotropic spin model. In order to include QC, we simulate \(I = 3/2\) nuclear spins. Within the CET method the largest accessible time scale or lowest frequency is linearly connected to the Chebyshev polynomial order. All technical details can be found in Refs. \cite{22,34}.

Modelling a quantum dot: The dynamics of a single p- and n-charged QD is described by the Hamiltonian \(H\) consisting of three contributions:

\[
H = \frac{g \mu_B B}{\hbar} S^z + H_{\text{CSM}} + H_{\text{QC}}. \tag{1}
\]

The first term represents an external magnetic field of strength \(B\) applied along the growth direction of the QD, which is defined along the \(z\)-direction. Furthermore, \(\mu_B\) denotes Bohr’s magneton, and the occurring \(g\)-factor depends on the geometry of the dots and is different for electrons and holes \cite{11}.

The coupling of the central electron or hole spin \(\vec{S}\) to the nuclear spin bath can be casted \cite{10} into the anisotropic CSM Hamiltonian \(H_{\text{CSM}}\)

\[
H_{\text{CSM}} = \sum_{k=1}^{N} A_k \left( S^z I^z_k + \frac{1}{\lambda} (S^z I^x_k + S^y I^y_k) \right). \tag{2}
\]

\(I^z_k\) denotes the nuclear spin of the \(k\)-th nucleus, and \(N\) is the total number of nuclear spins. The anisotropy parameter \(\lambda\) of the spin-flip term \cite{10} distinguishes between electron \((\lambda = \|1\rangle\) and hole spins, where \(1 < \lambda < \infty\) applies depending on the mixture between light and heavy holes. Due to the enlarged Hilbert space of \(2^{2N+1}\) for \(I = 3/2\), we have restricted ourselves to \(N = 10\) in the numerics. This, however, reproduces the previous results \cite{22} for \(N = 20\) nuclear spins with \(I = 1/2\) in the absence of the QC term.

The energy scale \(A_s = \sum_k A_k\) is expected to be of \(O(10)\) \(\mu eV\) for electrons and approximately one order of magnitude smaller for holes \cite{10}. The coupling constants \(A_k\) are proportional to the squared absolute value of the electron or hole envelope-wave function at the \(k\)-th nucleus – for details concerning a realistic modelling of the considered set of \(A_k\) entering our numerics see Ref. \cite{22}.

The additional quadrupolar term \cite{10} in Eq. \cite{11}

\[
H_{\text{QC}} = \sum_{k=1}^{N} q_k \left[ \left( \vec{I}_k \cdot \vec{n}^z_k \right)^2 - \frac{(I(I+1))}{3} \right] + \frac{q_k \eta}{3} \left[ \left( \vec{I}_k \cdot \vec{n}^x_k \right)^2 - \left( \vec{I}_k \cdot \vec{n}^y_k \right)^2 \right]. \tag{3}
\]

originates from electric field gradients in self-assembled QDs that couple to the nuclear electric quadrupole moment and are of crucial importance for the long-time dynamics of the central spin. The coupling constant \(q_k\) is mainly governed by the second order derivative of the strain induced electric potential \(V\) \cite{10}. The local \(z\)-direction at the \(k\)-th nucleus is denoted by the normalized orientation vector \(\vec{n}^z_k\) which refers to the eigenvector corresponding to the largest eigenvalue of the quadrupolar electric interaction tensor. The unit vectors \(\vec{n}^x/y/k\) complete the local orthonormal basis.

The asymmetry parameter \(\eta = (V_{xx} - V_{yy})/V_{zz}\) is commonly neglected in the literature \cite{17,18,20,35}. A recent microscopic calculation of the nuclear electric quadrupolar couplings \cite{21} in self-assembled InGaAs QDs, however, has found values up to \(\eta \approx 0.5\) depending on the In concentration in the QD. Therefore, we have included a finite \(\eta = 0.5\) in our calculations.

The individual coupling constants \(q_k\) are expected to be up to \(O(1)\) \(\mu eV\) \cite{21}, but only those \(q_k\) are relevant for the central spin dynamics where simultaneously \(A_k\) is of the same order of magnitude or larger. We define \(A_q = \sum_k q_k\) as a measure of relevant total quadrupolar coupling strength which is expected to be in the range of \(1 - 100\) \(\mu eV\) restricting the largest \(q_k\) to \(q_{\text{max}}\). The ratio \(Q_r = A_q/A_s\) determines the relative strength of the QC.

For our simulations, we generate random orientation vectors \(\vec{n}^z_k\) for each nucleus in our calculation whose deviation angles are restricted to \(\theta_{\text{q}} \leq 35^\circ\) in accordance to the average deviation angle \(\vec{I}_z \approx 25^\circ\) between the growth direction of the dot and the orientation vectors \(\vec{n}^z_k\) for InGaAs found by Bulutay \cite{21}. The coupling constants \(q_k\) have been generated randomly from a uniform distribution \(q_k/q_{\text{max}} \in [0.5 : 1]\).

For \(\eta = 0\), \(H_{\text{QC}}\) partially lifts fourfold degenerate nuclear spin states. Pinning \(\vec{n}^z_k\) to the growth direction, decoherence of the central spin would be suppressed with increasing \(q_k\). A distribution of \(\vec{n}^z_k\) due to the inhomogeneous strain fields \cite{21} favors the decoherence. Including a finite \(\eta\) further enhances the decoherence due to the \((S^+)^2 + (S^-)^2\) term.

The fluctuations of the transversal and longitudinal component of the unpolarized nuclear spin bath, referred to as Overhauser field, defines the time scale \(T^* = \lambda/\sqrt{\sum_{k=1}^{N} A^2_k}\). The short-time evolution of the central spin \(\vec{I} = \vec{I}^x + \vec{I}^y\) in the absence of \(H_{\text{QC}}\) has been used to define the dimensionless Hamiltonian \(\vec{H} = \vec{T}^*H\). Two factors in the definition of \(T^*\) suggest a longer lifetime for hole spin coherence than for electron spins: (i) the coupling constants \(A_k\) for holes are typically one order of magnitude smaller \cite{10} than for electrons, and (ii) the parameter \(\lambda \geq 1\) to larger values suppresses flips of the central spin. Both factors enter the time scale linearly, yielding an expected lifetime increase of a factor \(\sim 10\) for holes compared to electrons. However, when the spin-flip term in \(H_{\text{CSM}}\) becomes of the order of \(H_{\text{QC}}\), this argument fails and the long time decay rate will be strongly influenced by the QC for p-doped QDs as we will demonstrate below.

Definition of the spin-noise function: The Fourier transformation \(S(\omega)\) of the fluctuation function
larized central spin interacting with an unpolarized density operator has been used in all numerical calculations. Then the spin auto-correlation function is given by the sum-rule
\[ \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} S(\omega) = S(0) = \langle (S^z)^2 \rangle - \langle S^2 \rangle^2 \]
for the spin-noise spectrum. In the absence of an external magnetic field, its value is fixed to 1/4 for a QD filled with a single spin.

Since all experiments are performed in the high-temperature limit, the inverse temperature \( \beta = 0 \), and a constant density operator has been used in all numerical calculations. Then the spin auto-correlation function \( S(t) \) also describes the spin-decay of an initially fully polarized central spin interacting with an unpolarized nuclear spin bath, i.e., \( S(t) = \frac{1}{2} \langle (S^z(t))^2 \rangle / 2 \).

Results: For various relative QC strengths \( Q_r \), Fig. (a) shows \( S(t) \) for electron spins (\( \lambda = 1 \)). The two-stage spin dynamics is clearly visible: The initial short-time decay on the scale \( T^* \) to a plateau of approximately \( S(0)/3 \) is only governed by the Overhauser field and not influenced by QC. Here, we have used the time scale of \( T^* \approx 1 \text{ ns} \), see, for example, Ref. [15]. The second stage of the spin-decay is independent of the first for small values of \( Q_r \), and the decay is governed by QC. The shape of our curves agree remarkably with the data of Bechtold et. al. [15]: \( Q_r \approx 0.06 \div 0.1 \) seems to be an adequate choice for electrons confined in those InGaAs QDs.

We have defined a second time scale \( T_H \) at which \( S(t) \) has dropped to the value \( S(0)/6 \) indicated by the black dashed line in Fig. (a) (half the plateau) and have plotted the dependency of the lifetime \( T_H \) on \( Q_r \) in the inset. \( T_H(Q_r) \) approximately obeys a power law \( \propto Q_r^{-3/2} \).

Fig. (b) shows the spin-noise spectra \( S(\omega) \) for undoped QDs for various \( Q_r \). The peak at around 100 MHz reflects the short time behavior of \( S(t) \) up to 10 ns and it is only slightly influenced by the variation of \( Q_r \). Since this peak contains approximately 2/3 of the total spectral weight of \( S(\omega) \), the signal of the long time decay for electrons is expected to be a factor of 3 smaller than for holes. As demonstrated in Fig. (a) the QC mainly impacts the low frequency peak corresponding to the long time decay: an increase of \( Q_r \) broadens the peak width and induces a change of the gradient of \( S(\omega) \) at intermediate frequencies.
Now we focus on p-charged QDs. Since the overall QC strength $A_q$ does not depend on the doping of the QD while $A_s$ is decreasing by one order of magnitude when turning from electrons to holes, the ratio $Q_r$ is increasing by one order of magnitude at fixed $A_q$. Thus, we expect $T_H$ to decrease by a factor of $\sim 32$ when turning from electrons to holes. At the same time $T^*$ is increasing by a factor $10\lambda$, i.e., we expect the lifetime $T_H$ to be of the same order of magnitude for electrons and holes.

Figure 2(a) shows $S(t)$ for p-charged InGaAs QDs for three sets of parameters $\lambda$ and $Q_r$. For the conversion from the model parameters to the absolute time scale, we have assumed a reduction of $A_s$ by a factor of 10 compared to the n-charged case. For fixed absolute QC parameters $q_k$, $Q_r$ simultaneously increases also by 10, and, therefore, the absolute values $q_k$ are comparable to those used in Fig. 1. The initial decay due to the Overhauser field is suppressed in p-charged QDs by two effects that both decrease spin flips of the central spin on short time scales: (i) the increase of the asymmetry parameter $\lambda$ and (ii) the introduced energy splitting to the nuclei due to QC. Due to the lack of the short-time spin decay for hole spins, we define $T_H$ as $S(T_H) = S(0)/e$, indicated by the black dashed line in Fig. 2(a). For $\lambda = 4$ and $Q_r = 1.0$ we have determined the lifetime $T_H = 176$ ns which matches the finding $T_H = 188$ ns for electron spins at $Q_r = 0.1$ extremely well. For the other parameter sets, the lifetimes of 400 ns ($Q_r = 0.8$) and 740 ns ($Q_r = 0.6$) are found, which are slightly larger than corresponding electron decay times ($T_H(Q_r) \propto Q_r^{-3/2}$), but still of the same order of magnitude.

The spin-noise spectrum $S(\omega)$ is shown in Fig. 2(b) for various external longitudinal magnetic field strengths $B$, $\lambda = 4$ and $Q_r = 1.0$. The calculated $S(\omega)$ corresponds to recent measurements 30 and a nice agreement between our theory and the experiments is found: for increasing $B$ the spectral weight, fulfilling the sum rule 40, is shifted from large to small frequencies. As a consequence the gradient of $S(\omega)$ in the intermediate frequency regime $\omega \sim O(0.1)$ MHz is increasing, which is referred to as a shift from an approximately Lorentzian lineshape for $B = 0$ to a $1/f$ noise with increasing $B$ as reported in Ref. 30. Unfortunately, the resolution of our numerical investigations is limited to $\sim 0.1$ MHz for this parameter regime requiring already 6000 Chebychev polynomials. The linewidth of the added Lorentzian (dotted dashed line) at half width half maximum is 0.9 MHz, corresponding to the observed lifetime $T_H = 176$ ns. Note that for the parameter set $\lambda = 6$ and $Q_r = 0.8$ the corresponding linewidth is 400 kHz, which matches the experimental findings of Ref. 30.

For further comparison with our calculation, Fig. 3 shows experimentally measured spin noise spectra at a temperature of 5 K. The experiments were performed on (In,Ga)As quantum dot ensembles of similar dot density, in one case on average doped by a single electron per dot, in the other case by a single hole 11,31. The samples were studied using identical excitation conditions. The linearly polarized light beam of a single frequency laser was tuned to the ground state transition energy maximum 11. The laser power was reduced to 4 mW focused into a spot of 100 µm diameter, giving a good signal to noise ratio in 10-20 minutes of accumulation time, while simultaneously minimizing the laser excitation impact 30. The noise spectra are taken by a real time FFT using a FPGA module 11 and the spin-component is retrieved from the noise background by interlacing the data at zero and 250 mT magnetic field applied in Voigt direction. At 250 mT the peaked contribution to the noise due to spin precession is shifted out of the measured spectral range.

The comparison of the electron and hole spin noise spectra in Figs. 3(a) and (b) with the calculations reveals that the theory qualitatively correctly predicts the shape and widths of the spin-noise spectra. In particular, the following features are worth noting: (i) The electron spin noise shows an additional peak around 100 MHz unveiling the electron’s precession in the frozen Overhauser field 7, as also present in Fig. 1(b). (ii) Since $S(\omega)$ must obey the sum-rule 5, the low-frequency spectral weight of $S(\omega)$ for n-charged QD is only about 1/3 of those for holes. A Lorentzian fit to the low frequency components ($f < 35$ MHz) of the experimental data confirms this difference in the amplitudes. (iii) a spin correlation time of the same order of magnitude in the long-time range for electrons and hole spins, as predicted by the theory. In the experiment this time is on the order of 400 ns, as estimated from the peak width at low frequencies.

Summary: We have compared the impact of the hyperfine interaction on the spin coherence in n- and p-charged QDs, including the nuclear quadrupolar electric...
interaction generated by the strain fields, which provides an additional decoherence mechanism acting equally for n- and p-charged QDs. This mechanism is sufficient to explain the very similar long-time decay time $T_H$ of n- and p-charged QDs. On the other hand, the different coupling of electron and hole spins in the central spin part of the Hamiltonian leads to significant deviations in the short term dynamics, most prominently evidenced by the electron spin precession about the nuclear magnetic field.

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For the other two sets of parameters depicted in Fig. 2(a) qualitatively the same results are found.