Effect of magnetic field on the electron-nuclear spin dynamics in quantum dots

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Abstract. We report on the theoretical study of the joint effect of the hyperfine interaction and an external magnetic field on the electron-nuclear spin dynamics in a quantum dot. The study is based on the quantum-mechanical description of the hyperfine interaction of an electron spin with a finite number of nuclear spins in the frame of the graded box model. This approach clearly demonstrates electron-spin depolarization in the nuclear spin fluctuations and its suppression by the external magnetic field and dynamic nuclear spin polarization.

1. Introduction
The dynamics of nuclear spin polarization in quantum dots (QDs) has been intensively investigated in recent years [1]. In the typical scenario of experiments, a series of pumping pulses of one helicity creates spin polarization of localized electrons. This polarization is a source of angular momentum, which can be transferred to the nuclear spin bath via electron-nuclear hyperfine interaction. Usually, the dynamic nuclear-spin polarization (DNP) can be observed in the Zeeman splitting of ground state optical transition in single-QD spectroscopy [2, 3, 4, 5]. In these experiments, one can measure the steady-state value of DNP. However, this information is not complete because there always appear also the nuclear spin fluctuations (NSF), the interaction with which leads to dephasing and decoherence of the electron spin [6, 7]. The dynamics of DNP also strongly depends on the NSF because the NSF determine what fraction of electron spin polarization can be stored. Therefore, a key point to understand nuclear spin dynamics is to understand the NSF.

Here, we theoretically study the effect of magnetic field on the electron-nuclear spin system of a QD. For this purpose, we extend the graded box model approach [8] such that it allows us to simulate jointly the hyperfine interaction of the electron spin with a nuclear spin bath as well as the effect of a magnetic field on this coupled system. The model allows us to qualitatively compare theoretical results with experimental measurements of electron spin polarization in (In,Ga)As/GaAs QDs.

2. Model
We consider a system comprising $N$ nuclear spins $\hat{I}_j$ coupled to one localized-electron spin $\hat{S}$. In the presence of an external magnetic field $\mathbf{B}$ oriented along $z$ axis of the Cartesian coordinate system, the
Hamiltonian of electron-nuclear interaction can be written as

$$\hat{H} = \mu_B g_e B \hat{S}_z - \hbar \sum_{j=1}^{N} \gamma_j B \hat{I}_z^j + \sum_{j=1}^{N} A_j \left( \hat{S} \cdot \hat{I}_j \right)$$  \hspace{1cm} (1)

where $\mu_B$ and $g_e$ are the Bohr magneton and electron Landé $g$ factor, $\gamma_j$ and $A_j$ are the nuclear gyromagnetic ratio and the hyperfine coupling of electron spin to $j$th nucleus. $A_j = v_0 |\psi(R_j)|^2$, and $A$ determines the hyperfine interaction strength, respectively. We consider a special regime of excitation, in which at the time-moments $T_m$, the electron spin is initialized in the pure quantum mechanical state characterized by the electron-spin density matrix $ho_e = |\uparrow_z\rangle \langle \uparrow_z|$. We assume $T_{m+1} - T_m$ to be much larger than the period of electron spin precession about the effective nuclear field. In this regime, the electron and nuclear spins coherently interact during their relatively long correlation time, $\tau_c$. One can describe the electron-nuclear spin motion using the density matrix formalism. In this case, the electron-nuclear density matrix is

$$\rho(T_m + t) = \exp[-\frac{i}{\hbar} \hat{H}t] \rho(T_m + 0) \exp[+\frac{i}{\hbar} \hat{H}t].$$  \hspace{1cm} (2)

The coherent evolution of the electron-nuclear spin system initiated by the preceding pulse is interrupted by the next electron spin initialization pulse at time $T_{m+1}$. This external action affects the electron spin but does not affect the nuclei, so that the nuclear density matrix, $\rho_n$, is continuous and can be extracted from the total density matrix by partially tracing over the electron-spin states: $\rho_n(T_{m+1}) = \text{Tr}_e \rho(T_{m+1}) |0\rangle \langle 0|$. Immediately after the pulse, the total density matrix is $\rho(T_{m+1} + 0) = \rho_n(T_{m+1}) \otimes \rho_e$.

We further assume that the nuclear spin state can only slightly vary during $\tau_c$, which is small in comparison with the precession period of nuclear spins about external magnetic field. Hereafter, we ignore in Eq. (1) this slow precession.

To calculate Eq. (2) we use the graded box model approach [8], in which the nuclei can be divided in different groups, in each of which they are considered to be interacting with the same strength with the electron spin, i.e., we can introduce averaged hyperfine constants, $A(k)$, for each group. In this

![Figure 1](image-url)

**Figure 1.** (a) Time-dependencies of electron spin polarization after first initialization for different external magnetic fields: dephasing in frozen NSF. (b) Magnetic field dependence of mean electron spin polarization.
approximation, Eq. (1) can be rewritten in the basis of collective spin momenta $\mathbf{j}_k = \sum_{j \in (k)} \mathbf{I}_j$ of the $k$th group of nuclei. Then for a number $n \ll N$ of nuclear groups, the Hamiltonian matrix is block-diagonal with the size of each block drastically smaller than the size of the total Hamiltonian matrix.

3. Results

The model above allows us to solve numerically Eq. (2) and to obtain the time dependence of density matrix $\rho(t)$. The time dependencies of electron-spin polarization, $\langle \hat{S}_z(t) \rangle = \text{Tr}[\rho(t) \cdot \hat{S}_z]$, calculated for a model with $N = 12$ nuclear spins with $I = 3/2$ arranged in two nuclear groups are shown in Fig. 1(a) for different magnetic fields aligned along $z$ axis. The panel gives the dynamics of electron spin initially polarized in the state $|\uparrow_z\rangle$ and interacting with unpolarized nuclear spins, whose initial state is characterized by a high temperature density matrix ($\rho_n(0) \propto 1$).

The time evolution obtained in these calculations agrees well with the phenomenological approach proposed by Merkulov et al [6]. In particular, the electron spin polarization rapidly relaxes to its minimum and then saturates at a steady-state value, which depends on the magnetic field magnitude. At zero magnetic field, the dephasing time, $T_\Delta = \hbar/(\mu_B g_e \Delta_B)$, depends on the statistics of frozen NSF, $w(B_N)$, which can be described by a Gaussian with variance $\Delta_B = \sqrt{\frac{2}{3} \sum_j I_j (I_j + 1) A_j^2 / (\mu_B g_e)^2}$ for a large number of unpolarized nuclei [5].

The magnetic field dependence of steady-state electron spin polarization, $\bar{S}_z = 1/\tau \int_0^\tau \langle \hat{S}_z(t) \rangle dt$, is shown in Fig. 1(b). As one can see, the longitudinal magnetic field stabilizes the electron spin along the axis of initialization. It is interesting, that the half width of the dip in $\bar{S}_z$ near zero magnetic field is of the order of the NSF variance $\Delta_B$. This result has been obtained earlier also in phenomenological calculations [6, 9].

To understand how the DNP affects the electron spin polarization, we have calculated similar magnetic field dependencies of $\bar{S}_z$ when the nuclear spins have been polarized by a sequence of electron-spin initializations with different orientation along $z$ or $-z$ axis. To suppress some artefacts in the calculations of this nuclear spin training [8], a Poisson statistics has been used for each next excitation.

Figure 2. (a) Magnetic field dependencies of electron spin polarization for the cases of unpolarized nuclear spins (solid line) and polarized nuclear spin (symbols) for different signs of electron spin initialization. (b) Magnetic field dependencies of negative PL polarization measured for (In,Ga)As/GaAs QD ensembles. The curves are measured for alternatingly modulated excitation polarization ($\sigma^-/\sigma^+$) when no DNP has developed (solid line) as well as for constant circular polarization ($\sigma^-$ or $\sigma^+$) creating DNP (circles and triangles, respectively).
pulse with average pump repetition period $\langle \Delta T \rangle = 8T_\Delta$. The results of these calculations are shown in Fig. 2(a). The appearance of DNP leads to the decrease of dip depth which illustrates the effect of electron spin stabilization. Besides, some small shift of the dip from the zero value of external magnetic field can be seen. This effect is due to the appearance of a regular DNP field which is subtracted from or added to the external magnetic field. One can also see an asymmetry of the dips in Fig. 2(a) calculated for trained nuclear spins. The reason for this asymmetry is that the nuclear polarization rate strongly depends on the applied magnetic field as confirmed by additional numerical studies. There are also small peculiarities of these curves, which appear, most probably, because of the small number of nuclei in our model.

We compare these calculated curves with data measured experimentally [see Fig. 2(b)]. These experimental dependencies have been obtained by studying the photoluminescence (PL) of singly negatively charged (In,Ga)As/GaAs QDs annealed at 900 °C. The PL polarization is negative and reflects the spin polarization of resident electrons in these QDs. Further details can be found in Ref. [10]. As one can see, the theoretical modeling qualitatively reproduces the observed experimental behavior of electron-nuclear spin system. In particular, when the nuclei are unpolarized, the dip has the largest depth and is of symmetrical shape. When the nuclei become polarized, the experimental curves reveal a decrease of the dip depth and its shift from zero magnetic field, i.e., both effects obtained theoretically. However, there is also a quantitative difference between theory and experiment. In particular, the width of dip observed experimentally for unpolarized nuclei is much smaller than that obtained theoretically (in units of the dip shift when the nuclei are polarized). The reason for that is clear, as in the calculations only a small number of nuclear spins is considered. Therefore, the NSF, which are responsible for the dip, are drastically larger than those in real QDs and, correspondingly, give rise to the large dip width. Besides, the calculations show decreasing the dip width with pumping [see Fig. 2(a)], which is caused by NSF narrowing as it is already discussed in Ref. [8]. However, in contrast to the calculations, the experiment shows large broadening the dip at the permanent helicity of excitation polarization [see Fig. 2(b)]. Possible explanation for this difference is the large spread of DNP build-up from dot to dot in a QD ensemble as it is already discussed in Ref. [10]. This ensemble-related spread exceeds the theoretically predicted narrowing the NSF variance.

In conclusion, we have modeled the dynamics of electron-nuclear spin system in longitudinal magnetic field. A dip is observed in magnetic field dependence of electron spin polarization near zero field. This dip is shifted from zero field when nuclear spin system is polarized by the optical pumping. The predicted behavior of electron-nuclear spin system is supported by experiment. The narrowing of dip with pumping, however, is not observed experimentally, probably due to large spread of nuclear fields in the QD ensemble studied.

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