Long-term Dynamics of the Electron-nuclear Spin System of a Semiconductor Quantum Dot

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A quasi-classical theoretical description of polarization and relaxation of nuclear spins in a quantum dot with one resident electron is developed for arbitrary mechanisms of electron spin polarization. The dependence of the electron-nuclear spin dynamics on the correlation time $\tau_c$ of electron spin precession, with frequency $\Omega$, in the nuclear hyperfine field is analyzed. It is demonstrated that the highest nuclear polarization is achieved for a correlation time close to the period of electron spin precession in the nuclear field. For these and larger correlation times, the indirect hyperfine field, which acts on nuclear spins, also reaches a maximum. This maximum is of the order of the dipole-dipole magnetic field that nuclei create on each other. This value is non-zero even if the long-term average electron polarization vanishes. It is shown that the transition from short correlation time to $\Omega \tau_c \gtrsim 1$ does not affect the general structure of the equation for nuclear spin temperature and nuclear polarization in the Knight field, but changes the values of parameters, which now become functions of $\Omega \tau_c$. For correlation times larger than the precession time of nuclei in the electron hyperfine field, it is found that three thermodynamic potentials ($\chi, \xi, \varsigma$) characterize the polarized electron-nuclear spin system. The values of these potentials are calculated assuming a sharp transition from short to long correlation times, and the relaxation mechanisms of these potentials are discussed. The relaxation of the nuclear spin potential is simulated numerically showing that high nuclear polarization decreases relaxation rate.

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I. INTRODUCTION

The electron-nuclear spin system (ENSS) of a semiconductor quantum dot (QD) has been under intensive investigation in recent years.\cite{1,2,3}. This strong interest has been motivated by potential spintronics and quantum information applications, for which semiconductor quantum dots are promising.\cite{4,5,6}. The spin dynamics of this system is described by a variety of relaxation times which range from nanoseconds to seconds.

Optical orientation is a commonly used method to create and control the ENSS with a high degree of polarization. Nuclear polarization is caused by the Fermi hyperfine interaction\cite{7} between nuclear spins and photo oriented electrons. A simple theoretical description of the ENSS behavior is the short correlation time approximation (SCTA). (The electron correlation time, $\tau_c$, is the characteristic time of the free coherent undisturbed electron spin precession in the hyperfine field of the nuclei.) The SCTA is valid if the frequencies of electron spin precession ($\Omega$) in the local nuclear hyperfine field, and nuclear spin precession ($\omega$) in the electron hyperfine field are small enough: $\Omega \tau_c \ll 1$, and $\omega \tau_c \ll 1$. In a quantum dot, the electron interacts with a macroscopic number, $N$, of nuclei, i.e., $N \approx 10^5$ and $\Omega \gg \omega$. It follows that the SCTA can be used if $\Omega \tau_c \ll 1$. When $\Omega \tau_c \ll 1$ holds, frequencies of electron and nuclear spin precession are practically constant during the time $\tau_c$. Small deviations of these frequencies during $\tau_c$ is a small perturbations to the spin motion. This deviation is the mechanism for the slow transfer of spin polarization between electron and nuclei.

The SCTA is valid in many experimental scenarios. However, studying the problem beyond the SCTA stimulates experimental and theoretical investigations in the regime of intermediate correlation time, where $\Omega \tau_c \gtrsim 1$, and $\omega \tau_c \ll 1$, and in the regime of long correlation time, where $\omega \tau_c \gg 1$. These regimes occur at low temperature, and under constant wave (CW) light of low intensity or in the darkness, respectively. The intermediate regime may
be realized also by using circularly polarized light pulses. In this case, short periods of light illumination and high photoelectron concentration are alternated with long periods of darkness, when the ENSS motion is undisturbed.

In this paper we discuss the behavior of an ensemble of quantum dots, each containing a single resident electron. We consider the simplest experimental scenario, where in the first step some external action (e.g., circularly polarized photons) orients the resident electron which polarizes the QD nuclei, and in the second step the ENSS dynamics is determined only by interactions between quantum dot spins. The electron becomes polarized due to the spin exchange with optically oriented photo carriers. After switching off the light, a relaxation takes place. It is characterized by a long relaxation time \( T \gg \omega^{-1} \). The relaxation is a result of the dipole-dipole interaction between neighboring nuclei, and of the electron-phonon interaction.\(^6,7,8\)

In Section II we demonstrate (i) that the maximum rate of nuclear polarization by optically oriented electrons is reached for \( \Omega \tau_c \approx 1 \), (ii) that the nuclear polarization is a result of the nuclei cooling by spin oriented electrons in the Knight field (connected with the time averaged electron polarization), and (iii) that the spatial dependence of the hyperfine interaction decreases the photoinduced nuclear polarization. In the intermediate regime \( \Omega \tau_c \) mostly influences the nuclear spins relaxation times. In Section III we discuss the difference in the ENSS description between the intermediate and long correlation time approximations. We also present numerical results for the dipole-dipole relaxation. These calculations show an increasing relaxation time with increasing nuclear polarization. All cases are calculated for a Spherical Quantum Dot with Infinitely high Barrier (SQDIB), which allows us to ignore the exponentially small escape of the electron wave function out of the quantum dot. We limit the description of the ENSS interaction with the QD’s environment by introducing the leakage factor approximation, and we do not discuss the specifics of spin diffusion of the nuclear polarization outside the QD.\(^9,10,11\)

To model the spin system we use the quasi-classical approximation, which is valid for quantum dots with large numbers of nuclear spins.\(^12,13\) Finally, in section IV we summarize our main results.

II. POLARIZATION OF THE ELECTRON-NUCLEAR SPIN SYSTEM IN A QUANTUM DOT

In a semiconductor quantum dot with one resident electron the hyperfine interaction creates a localized electron-nuclear spin polaron.\(^14,15,16\) In this section we discuss this system’s behavior in the limit of short and intermediate correlation time, \( \omega \tau_c \ll 1 \). In these regimes the frequency of the nuclear spin precession in the electron hyperfine field is lower than the frequency of external perturbations of electron field orientation. We do not specify the character of the external interaction, but will simply suppose that this interaction can partially orient the electron spin. The exchange scattering of an optically polarized carrier is an example of such an external interaction.

A. Electron spin precession in the nuclear hyperfine field

In GaAs-like semiconductors the electron and nuclear spins are coupled by the Fermi hyperfine interaction,

\[
\hat{H} = \frac{\pi}{3} \mu_B \sum_n \frac{\mu_n}{I_n} (s \cdot I_n) \delta(r - R_n),
\]

where \( \mu_B \) is the Bohr magneton, \( s \) and \( r \) are the spin and position of the electron, \( \mu_n \), \( I_n \), and \( R_n \) are the magnetic moment, spin and position of the \( n \)-th nucleus, respectively. The sum in Eq. (1) runs over all the nuclei inside the QD. The hyperfine energy has a maximum when \( s \) and \( I_n \) are parallel, and a minimum when they are antiparallel. In the following, and for simplicity, we will suppose that all nuclei have the same spins and magnetic moments \( I_n \equiv I \) and \( \mu_n \equiv \mu \).

A QD contains a macroscopic number, \( N \), of nuclear spins, \( N \approx 10^5 \gg 1 \). Therefore, the frequency of electron spin precession in the nuclear hyperfine field,

\[
\Omega = \sum_n \omega_n I_n,
\]

is distributed in a wide region from zero to \( \Omega_{\text{max}} \) given by

\[
\Omega_{\text{max}} = I \sum_n \omega_n.
\]

It is usual to separate \( \Omega \) in two parts, average and fluctuation: \( \Omega = \langle \Omega \rangle + \Delta \Omega \). In the following, average and fluctuation refer to the time evolution and time fluctuation of the nuclear spins. Because \( \langle \Omega \rangle \) and \( \Delta \Omega \) are not correlated, \( \langle \Omega^2 \rangle = \langle \Omega \rangle^2 + (\Delta \Omega^2) \).

\( \Omega \) is many orders of magnitude larger than the frequency of nuclear spin precession in the electron hyperfine field, \( \omega_n, \) i.e., \( \Omega \gg \Omega_{\text{flu}} \), where

\[
\Omega_{\text{flu}} = \sqrt{\left\| I \right\|^2 \sum_n \omega_n^2} = \left\| I \right\| \sqrt{\sum_n \omega_n^2} \gg \left\| s \right\| \sqrt{\sum_n \omega_n^2}.
\]

Here, \( \Omega_{\text{flu}} = \sqrt{\left\langle \Omega^2 \right\rangle} \big|_{\Delta \Omega = 0} \) is the characteristic value of the fluctuations of the electron spin precession frequency, \( \left\| I \right\| = \sqrt{I(I+1)} \), and \( \left\| s \right\| = \sqrt{s(s+1)} \), are the modulus of nuclear and electron spin, respectively,

\[
\omega_n = \frac{16\pi \mu_B \mu_n}{3I_n \hbar} \left\| \psi(R_n) \right\|^2,
\]

where \( \langle \Omega \rangle \) is the expectation value of the hyperfine interaction.
and \( \psi(R_n) \) is the electron wave-function on the \( n \)-th nucleus.

\( \Omega_{\text{max}} \) does not depend on the quantum dot’s volume because the electron wave function is normalized in this volume; it is determined solely by the chemical composition of the QD. For example, for a GaAs QD one can estimate \( \Omega_{\text{max}} \approx 10^{-11}\, \text{s}^{-1} \). Values of \( \Omega_{\text{fluc}} \propto \Omega_{\text{max}}/\sqrt{N} \) and \( \langle \omega \rangle \propto \Omega_{\text{max}}/N \) depend on the QD volume. For a quantum dot with \( N \approx 10^5 \) nuclei \( \Omega_{\text{fluc}} \approx 3.10^8\, \text{s}^{-1} \) and \( \langle \omega \rangle \approx 10^{-8}\, \text{s}^{-1} \). Characteristic frequencies and times are collected in Fig. I.

Under sample illumination, the photocarriers and photons scattering on a quantum dot, carrier capture and photon absorption in QD are the main mechanisms of the ENSS interaction with the environment. As we will show in section 1B the nuclear spin polarization and relaxation are a consequence of this perturbation of the electron and nuclear spin precession in the hyperfine field. To achieve the most effective collisions, the frequency of these collisions, \( \omega_{\text{coll}} \equiv \tau_c^{-1} \), should be about \( \Omega \gg \omega_n \). Between collisions nuclear spins change their directions by a very small angle of about \( \tau_c < \omega/2 \), which is much less than 1. In the zero-th order approximation, the spin, \( s(t) \), of the resident electron precesses in the constant (frozen) nuclear field:

\[
\begin{align*}
  s(t) &= \mathbf{s}_\Omega(t_0) + (s(t_0) - \mathbf{s}_\Omega(t_0)) \cos(\Omega \cdot \varsigma) \\
  &+ [\mathbf{e}_\Omega \times s(t_0)] \sin(\Omega \cdot \varsigma),
\end{align*}
\]

where

\[
\mathbf{s}_\Omega(t_0) = (s(t_0) \cdot \mathbf{e}_\Omega) \cdot \mathbf{e}_\Omega,
\]

\[
\mathbf{e}_\Omega = \Omega/\Omega, \quad s(t_0) \text{ is the initial spin, which is determined by collisions, } \delta t = t - t_0, \text{ and } t_0 \text{ is the time when the external action polarized the resident electron.}
\]

For the continuous-wave (CW) photoexcitation, the probability \( W(\delta t) \) of electron free (undisturbed) precession during time \( \delta t \) decreases exponentially with free precession time: \( W(\delta t) = \exp(-\delta t)/\tau_c \). The spin of localized electrons (averaged over initial polarization \( s(t_0) \) and \( \delta t \)) can be written as:

\[
\bar{s} = \frac{s_0 + [\Omega \times s_0] \tau_c + (\Omega \cdot s_0) \Omega \tau_c^2}{1 + (\Omega \tau_c)^2},
\]

where \( s_0 \) is the average value of the initial spin. For an excitation with a periodic train of short pulses \( W(t) \approx \delta(t - t_0 - \tau_c) \), and \( \bar{s} \) is given by Eq. (8), by substituting \( \cos((\Omega(t - t_0)) \rightarrow \sin(\Omega \tau_c))/(\Omega \tau_c) \) and \( \sin((\Omega(t - t_0)) \rightarrow (1 - \cos(\Omega \tau_c)))/(\Omega \tau_c) \). For our goal, the difference between the CW and the pulsed excitation regimes is only quantitative. In this paper we analyze (and present model calculations) only for the CW excitation regime.

In the short correlation time approximation \( \bar{s} \approx s_0 \), whereas in the opposite limit \( \Omega \tau_c \geq 1 \), the mean value of the electron spin depends on the angular distribution of frequencies, \( \Omega \). For a random distribution (unpolarized nuclear system) \( \langle \Omega \rangle = 0 \), \( \langle \Omega \cdot s_0 \rangle \cdot \Omega = \Omega^2 s_0/3 \), and \( \bar{s} \approx s_0/3 \). For a polarized nuclear spin system \( \langle \Omega \rangle \gg \Omega_{\text{fluc}}, \) and the mean value of the electron spin depends on the direction of \( \langle \Omega \rangle \). In the absence of external magnetic fields the ENSS has only one distinguished direction that is determined by the photo-electron polarization, \( s_0 \). For \( \langle \Omega \rangle \) along \( s_0 \) \( \langle \Omega \cdot s_0 \rangle \approx \Omega s_0 \), and \( \bar{s} \approx s_0 \).

B. Nuclear spin precession in the electron hyperfine field

Let us now describe the mechanism of the nuclear polarization by the resident electron. We consider a slow nuclear spin precession that obeys the equation

\[
\frac{dI_n}{dt} = \omega_n [s(t) \times I_n] \\
\approx \omega_n [s \times I_n] + \omega_n \frac{ds}{dt} \times I_n \delta t + \omega_n s \times \frac{dI_n}{dt} \delta t.
\]

The average in this equation is done on the time region \( \Delta t \), such that \( \Omega \ll \Delta t \ll \omega \). The first term in the right-hand side of Eq. (11) gives the regular part of the nuclear spin precession in the mean electron hyperfine field, \( B_{K,n} = -\omega_n \bar{s} \), known as the Knight field

\[
\begin{array}{c|c|c|c}
\hline
\text{frequency} & \text{correlation} & \text{precession} & \text{times} \\
\tau_c & \Omega_{\text{fluc}} & \Omega & \text{inhomogeneous} \\
\hline
1.0 \times 10^{-10} & 1.0 \times 10^{-8} & 1.0 \times 10^{-6} & 0.0001 \\
0.01 & \text{frozen nuclear field} & \text{long correlation time} & \text{magneto-dipole field} \\
\hline
\end{array}
\]
second term describes the dynamical polarization of the nuclei. For small nuclear polarization, \( |I| \ll I \), and:

\[
\frac{\partial I_n}{\partial t} \bigg|_{dp} = -\omega_n \tau_c \left\langle \frac{[\Omega \times s_0] \times I_n}{1 + (\Omega \tau_c)^2} \right\rangle_I \approx \frac{\omega_n^2 \langle |I|^2 s_0 \rangle}{\langle \omega^2 \rangle \langle |s|^2 \rangle^2 T_{1c}},
\]

where

\[
T_{1c}(\Omega) = \frac{\Omega}{\langle \omega^2 \rangle} \left( \frac{2\langle |s|^2 \rangle^2 (\Omega \tau_c)}{3(1 + (\Omega \tau_c)^2)} \right)^{-1} \tag{10}
\]

is the characteristic time of the longitudinal nuclear spin relaxation, and \( \langle \omega^2 \rangle = \sum_n \omega_n^2 / N \). The third term in Eq. (9),

\[
\frac{\partial I_n}{\partial t} \bigg|_{I(t)} = \omega_n \sum_m \omega_m \left[ s(t) \times \int_0^2 [s(t_1) \times I_n] \, dt_1 \right], \tag{12}
\]

represents two processes: relaxation of nuclear polarization (averaged over initial electron spin direction),

\[
\frac{\partial I_n}{\partial t} \bigg|_{rel} = -\frac{\omega_n^2 I_n}{\langle \omega^2 \rangle T_{1c}(\Omega, \tau_c)} (I_n) + \frac{2 + (\Omega \tau_c)^2}{2} I_{n\perp}, \tag{13}
\]

and nuclear spin precession, \( \partial I_n / \partial t \bigg|_{ind} = [\eta_n \times I_n] \), in the indirect hyperfine field given by

\[
\eta_n = -\frac{\omega_n^2 \Omega \times I_n \tau_c}{\langle \omega^2 \rangle T_{1c}(\Omega, \tau_c)}. \tag{14}
\]

(For a derivation of Eqs. \(10\), \(13\) and \(14\) see the appendix.) Both of these processes, i.e., relaxation of nuclear polarization and nuclear spin precession, are determined by fluctuations of the electron hyperfine field. They are proportional to \( |s|^2 = 3/4 \) because \( T_{1c} \propto |s|^{-2} \). The relaxation time versus \( \Omega \tau_c \) is presented in Fig. 2. In the limit of short correlation time, all components of nuclear polarization relax at the same rate, \( T_{1c}(0, \tau_c)^{-1} = 2\langle |s|^2 \rangle \tau_c \cdot \langle \omega^2 \rangle / 3 \).

For intermediate correlation times, the relaxation time for the component of \( I, I_{||} \), longitudinal to \( \Omega \), increases as both \( \Omega \) and \( \tau_c \) increase, yielding \( T_{1c}(\Omega, \tau_c) = (1 + (\Omega \tau_c)^2)T_{1c}(0, \tau_c) \) (solid curves 1,3 in Fig. 2). On the other hand, the relaxation rate of the polarization component, \( I_{\perp} \), transverse to \( \Omega \), behaves different depending on whether \( \Omega \) is increase or \( \tau_c \) is increased. For \( \tau_c \) increasing, \( T_{1c,\perp} \) decreases monotonically (curve 2). As \( \Omega \) increases, \( T_{1c,\perp} \) increases saturating at \( T_{1c,\perp}(\infty, \tau_c) = 2T_{1c}(0, \tau_c) \) (curve 4).

The nuclear polarization, its relaxation time, and the indirect hyperfine field, are all proportional to \( \omega_n^2 \tau_c \propto |\psi(R_0)|^4 \), and all have a strong spatial dependence. They have a maximum in the center of the QD, and they decrease towards the border. In the limit of frozen nuclear field, i.e., short and intermediate correlation times, \( \omega_n \tau_c \) in Eqs. \(10\), \(13\) and \(14\) is much smaller than 1. In Eq. \(14\), for an intermediate time, this parameter may be rewritten as \( \omega_n / \Omega \), which is much less than 1, implying that characteristic rates for nuclear spin polarization, relaxation, and indirect hyperfine interaction are many times less then \( \omega_n \). These slow processes are relevant only if they change the system’s behavior, such as its nuclear polarization, Eq. \(10\), or its relaxation rate, Eq. \(12\).

By comparing the indirect field and the Knight field one can see that the former is significant only for \( s_0 \leq (\omega_n / \Omega) \cdot (\Omega \tau_c)^2 / (1 + (\Omega \tau_c)^2) \leq N^{-1/2} \ll 1 \). In the following context we suppose that the electron polarization is high enough, which allows us to ignore the indirect hyperfine interaction when it comes together with the Knight field. (The indirect hyperfine interaction between nuclei plays an important role in the relaxation of the electron polarization transversal to a strong external magnetic field.)

A sketch showing the main mechanisms of the hyperfine interaction’s influence on the ENSS’s behavior under sample illumination is presented in Fig. 3. This figure shows three precessions (i) in the mean Knight field, created on the nuclei by a mean electron polarization, (ii) in...
the mean Overhauser field, created on the electron by the mean nuclear spin, and (iii) in the mean indirect Weiss field, created on the nuclei by the mean nuclear polarization. It also contains two dissipation processes: nuclear polarization by the oriented electron through the fluctuation of nuclear spins, and nuclear polarization relaxation on fluctuations of the electron hyperfine field.

When transitioning from short to intermediate correlation times, what changes most is the dependence of the $T_{1e}$ time on $\Omega \tau_c$. For $\Omega \tau_c \ll 1$, the time $T_{1e}$ is proportional to $\langle \omega^2 \rangle^{-1}$, and also to $N^2$. $T_{1e}$ is a decreasing function of $\tau_c$, and it reaches its minimum for $\Omega \tau_c = 1$. On the other hand, in the limit $\Omega \tau_c \gg 1$, $T_{1e}(\Omega) \approx \tau_c(3\Omega^2)/(2||s||^2\langle \omega^2 \rangle)$ is an increasing function of $\tau_c$. For a depolarized nuclear system, i.e., when $\Omega \approx \Omega_{fluc}$, the previous equation gives $T_{1e} \propto \tau_c N$, whereas for a polarized nuclear system $T_{1e} \propto \tau_c N^2$, as we saw before.

For a typical GaAs QD (with $N = 10^5$, $\langle \omega \rangle = 10^6 s^{-1}$, $\Omega_{fluc} \approx 3 \cdot 10^8 s^{-1}$), the shortest relaxation time for depolarized nuclei, calculated from Eq. (11) and condition $dT_{1e}/d\tau_c = 0$, is $T_{1e} \approx 10^{-3}s$. It is reached for $\tau_c^{(\text{min})} \approx 3 \cdot 10^{-9}s$. On the other hand, for 100% nuclear polarization, $T_{1e} \approx 10^{-1}s$. This relaxation time is reached when $\tau_c^{(\text{min})} = 10^{-11}s$. Since the short correlation time approximation is valid for $\tau_c \ll \tau_c^{(\text{min})}$, this region decreases by a factor of $\sqrt{N}$ for a highly polarized nuclear system.

The relaxation rate and precession frequency depend on the nuclear position in the quantum dot. The balance between nuclear dynamical polarization, Eq. (10), and relaxation, Eq. (13), gives:

$$\langle I_n \rangle = \frac{||I||^2 s_0}{||s||^2}.$$

Since the right-hand side of this equation does not contain $n$ or $\tau_c$, the average nuclear spin has the same value for all nuclei, i.e., $\langle I_n \rangle \equiv \langle I \rangle$. The previous statement is valid both for short and intermediate correlation times.

Equation (14) is correct if we take into account only one mechanism of nuclear spin relaxation, namely the hyperfine interaction with fluctuations of electron polarization. Additional channels of relaxation decrease the nuclear polarization. Often this decrease can be described by introducing in Eq. (15) the phenomenological leakage factor $f = T_{1l}/(T_{1e} + T_{1l}) \leq 1$. (Here $T_{1l}$ is the relaxation time for additional relaxation channels.) When $T_{1l}$ is finite, the leakage factor, $f$, and the average polarization, $\langle I \rangle$, are monotonously decreasing functions of $T_{1e}$, and have a maximum for $\Omega \tau_c \approx 1$.

C. Dipole-dipole interaction between nuclear spins. Nuclear spin temperature

The main additional channel of nuclear polarization relaxation is determined by the dipole-dipole interaction between neighboring nuclear spins. This interaction transfers nuclear angular momentum to the crystal lattice with a characteristic time $T_2 \approx 10^{-4}s$, which is much smaller than $T_{1e}$.

As a result, the steady state value of the quasi equilibrium nuclear polarization is $T_2/T_{1e}$ times less than that predicted by Eq. (13), and it has to be reached at a time $T_2$. Nevertheless, it is well known from many experiments and theoretical calculations, that the optically induced nuclear polarization cannot usually be ignored. This polarization is due to a decrease of the nuclear spin temperature, $\Theta$. The effect results from the balance of two energetic flows: cooling of nuclear spins by oriented electrons in an external magnetic field $B$: $J_{\text{cool}} \propto -\langle B \cdot \partial I/\partial |d_{\text{up}}\rangle$, and heating of nuclear spins by random fluctuations of electron polarization: $J_{\text{heat}} = \beta C$. Here $\beta = (k_B \Theta)^{-1}$ is the inverse spin temperature, and $C = dE/d\beta$ is the heat capacity of the nuclear spin system. In the short correlation time approximation, $(\Omega \tau_c \ll 1)$, and in a spatially uniform external magnetic field $B$, $\beta = f \frac{4I}{\mu_I B^2} \frac{(B \cdot s_0)}{\varepsilon B_L^2}$.
where $B_L^2$ is the characteristic value of the random local field squared, and $\varepsilon$ is a number of the order of one. $\beta$ has the opposite sign for parallel and anti-parallel orientations of $B$ and $s_0$. (It is not surprising that the spin temperature can be negative, since the energy of the spin system is limited both from above and below.) $\beta$ is positive if $s_0$ is parallel to $B$, in which case spin alignment decreases the nuclear spin energy.

In a general case, the field $B_L$ is a result of dipole-dipole and indirect hyperfine interactions between nuclei. For the special case of Eq. (14), the indirect interaction between two nuclei, $m$ and $n$, depends on $n$ and $m$ only through their product, $\omega_n\omega_m$, and has no influence on the local field part of Eq. (16): $\varepsilon B_L^2 \approx 3B_{dd}^2$. Here $B_{dd}^2$ is the square of the average dipole-dipole part of the local field. Eq. (10) is valid for high nuclear spin temperature, i.e., for $\beta\mu L^2B^2 + \varepsilon B_L^2 \ll 1$, and was derived for a spatially uniform $B$ and $T_1$. An average Knight field should be included in the regular external field, and the total magnetic field, $B + B_{K,n}^L$, depends on the nuclear position inside the QD.

The approximation that ignores the spatial dependence of the hyperfine interaction inside the dot is known as the “box model”. For the “box model” the nuclear spin temperature and nuclear polarization have the same value for all nuclei. In the real situation of a spatially inhomogeneous hyperfine interaction, the nuclear spin temperature and polarization cannot both be constant because $\langle I_n \rangle \propto \beta B_{K,n}^L$.

The dipole-dipole interaction between nuclear spins produces an energy flow from the region with high spin temperature to the region with low spin temperature. It is commonly assumed that the nuclear spin diffusion inside the area of electron localization is suppressed by the strong gradient of the Knight field. (If the difference in hyperfine splitting of the nearest nuclear spin levels is larger than their dipole-dipole broadening, the flip-flop process between nearest nuclear spins is suppressed by the energy conservation law.) But one can show that the typical difference in the splitting of the nearest nuclear spin levels for a spherical QD is about $h\langle \omega \rangle s_0/N_R \approx h\Omega_{\text{max}} s_0 (4/3)^{1/3}$, where $N_R \approx (N/4)^{1/3}$ is the number of nuclei along the QD radius. For a GaAs QD with $N = 10^5$ this difference is about or less than $h10^4\tau_c^{-1} \approx h\varepsilon_{dd}$, and, therefore, there are no reasons for a strong suppression of the spin diffusion.

In the limit of efficient spin diffusion, the nuclear spin temperature should have the same value for all nuclei, and the equation for $\beta$ contains averaged values, i.e.,

\[
\beta = -\frac{4}{\hbar} \int \langle \omega^3 \rangle s_0^2 + \varepsilon \langle \omega^2 \rangle^2 / \langle \omega^2 \rangle.
\]

Here $\langle \omega^m \rangle = \sum \omega^m_n / N$ and $\langle \omega^2 \rangle = \mu_s^2 B_L^2 / \hbar^2$.

In the limit of short correlation time, the difference between the result of Eq. (17) and the one obtained with the “box model” is only numerical. The dimensionless saturation value of the spin temperature, $\beta \equiv \hbar(\omega)\beta$, is then $\langle \omega \rangle / \langle \omega^3 \rangle$ times less than that predicted by Eq. (10). It reaches saturation if $s_0^2 \gg \varepsilon \langle \omega^2_{dd} \rangle / \langle \omega^2 \rangle$. For a spherical quantum dot with infinite barrier (SCDIB), we have: $\langle \omega \rangle / \langle \omega^3 \rangle \approx 0.36$, $\langle \omega \rangle / \langle \omega^2 \rangle \approx 0.22$, $\langle \omega^2 \rangle / \langle \omega^4 \rangle \approx 0.17$, and $\varepsilon \langle \omega^2_{dd} \rangle / \langle \omega^2 \rangle \approx 5 \cdot 10^{-3}$.

In equilibrium, the mean value of the nuclear spin in this field is

\[
\langle I_n \rangle = -\beta \frac{\parallel I \parallel^2}{3} (\hbar \omega_n) s_0. \tag{18}
\]

When there is a single global nuclear spin temperature for all points in the QD, the maximum average nuclear polarization within the SCDIB model is 4 to 5 times smaller than the one predicted by the “box model”.

In section II B we saw that $T_{1\text{e}}$ as a function of $\tau_c$ has a minimum for $\tau_c = \Omega^{-1}$. For $\tau_c > \Omega^{-1}$, increasing the correlation time and the frequency $\Omega$ increases $T_{1\text{e}}$. The nuclear polarization increases $\Omega$, implying that if one wants to optimize the nuclear polarization by varying the correlation time, one has to start from an unpolarized nuclear system with $\Omega_{\text{enc}}\tau_c \ll 1$. At the same time, $\tau_c$ should be long enough to achieve the condition $\Omega \tau_c = 1$, and to maximize the leakage factor in the final polarized state. Then, the best regime for generating high nuclear polarization is on the border between short and intermediate correlation times.

For $\Omega \tau_c \gg 1$, the anisotropy of the nuclear spin relaxation on the electron also renormalizes the parameter $\varepsilon \approx 3 + (\Omega \tau_c)^2$. We can neglect this effect for $\Omega \tau_c \leq 1$. In Fig. 4 results of the nuclear spin polarization calculations for the “box model” (curve 1) and

![FIG. 4: Dependence of nuclear polarization on the mean spin of the resident electron. Here $T_{1\text{e}}(0) = T_{1\text{H}}$. Curve 1 was calculated using the “box model” with $\Omega_{\text{max}}\tau_c \ll 1$, curve 2 using the SCIB model with $\Omega_{\text{max}}\tau_c \ll 1$, and curve 3 using the SCIB model with $\Omega_{\text{max}}\tau_c = 10$.](image-url)
SQDIB model (curves 2,3) are presented. All calculations are done for $T_{1e}(0) = T_{1h}$. Curves 1 and 2 are calculated in the short correlation time approximation, curve 3 for $\Omega_{\text{max}}\tau_c = 10$. One can see that the nuclear polarization for the SQDIB model is about 5 times less than for the “box model”. For $\Omega_{\text{max}}\tau_c = 10$, increasing the time $T_{1e}$ by increasing the nuclear polarization additionally decreases the leakage factor and slows down an increase of the nuclear polarization.

III. ELECTRON-NUCLEAR SPIN SYSTEM IN THE LIMIT OF LONG CORRELATION TIME (IN DARKNESS)

In this section we consider the relaxation of an isolated ENSS in the limit of long correlation time. The nuclear polarization by spin-oriented carriers is extremely inefficient. We will consider only relaxation of an isolated ENSS in the darkness. In section III A we discuss the main thermodynamic potentials that characterizes the isolated spin system. We connect these potentials to the relaxation of ENSS parameters under illumination and discuss the relaxation mechanism. In section III B we present the results of a numerical simulation of the QD spin relaxation due to the dipole-dipole interaction that transfers nuclear spin into the crystalline lattice angular momentum. This process is controlled only by the state of the QD ENSS, and is independent of the QD’s environment.

A. Conservation laws and thermodynamic potentials of the ENSS

The $\tau_c$ of the ENSS largely increases in the darkness. At liquid helium temperatures the characteristic time of spin relaxation for electrons on phonons is about seconds, whereas for nuclei it ranges from days to years. The direct transfer of spin angular momentum to the crystal by the dipole-dipole interaction between nuclear spins is the main mechanism for the relaxation of the spin polarization. This interaction is also responsible for the energy diffusion from the quantum dot to the neighboring nuclei.

In the zero-th order approximation, we keep only the hyperfine interaction and switch off all other interactions, which makes it easier to determine some integrals of motion. The Fermi interaction conserves energy, $E = \hbar \Omega s_0$, and total spin, $F = I_S + s$. The total spin of the nuclei, $I_S$, is many orders of magnitude larger than that of the electron. Therefore, with high precision $I_S$ is also conserved. Moreover, as a result of the adiabatic approximation, $\Omega \gg \omega_n$, and the electron spin projection, $s_0$, along $\Omega$ is also conserved and must be quantized, i.e., $s_0 = \pm 1/2$. The conservation of $\Omega$ follows from the conservation of energy and $s_0$.

Under illumination every nucleus is acted on by an average Knight field, whose value and direction is determined by the average electron spin, whereas in the darkness the ensemble of quantum dots decomposes in two sub-ensembles (nuclear spin polaron) with defined value of electron spin projection, $s_0$, on the nuclear hyperfine field, i.e., $s_0 = \pm 1/2$, and energies $\pm \hbar \Omega/2$. The probability to find a quantum dot in one of these sub-ensembles is given by the conservation of energy, frequency, and total nuclear spin.

We will assume that the transition from illumination to darkness is sharp, and that in the initial state the total nuclear polarization is larger than its fluctuations, i.e., $(I) \gg 1/\sqrt{N}$. Considering the integrals of motion of the system, we can write the ENSS probability distribution $\Phi$, as a function of three thermodynamic potentials: an electron spin potential, $\varsigma$, an inverse nuclear spin temperature, $\chi_{s_0}$, for each sub-ensemble, and a nuclear spin potential, $\xi(\chi_{s_0})$.

$$\Phi(\varsigma, \chi, \xi) \approx \frac{\exp \{\varsigma s_0 - (\chi_{s_0} \hbar \Omega s_0) + (\varsigma \cdot I_S)\}}{(4\pi)^N (\exp(\varsigma/2) + \exp(-\varsigma/2))}$$  \hspace{1cm} (19)

At the last moment of illumination, the total nuclear spin mean value, $\langle I_S \rangle$, and the electron spin, $s_0$, are directed along the Knight field, and the nuclear inverse spin temperature under illumination is $\beta$. Therefore, after switching off the light, the nuclear inverse spin temperature in the darkness is

$$\chi_{s_0} \approx \frac{\beta s_0}{s_0}.$$  \hspace{1cm} (20)

Moreover, nuclear and electron spin potentials are given by

$$\varsigma = \frac{3\langle I_S \rangle}{\langle ||I_S||^2 \rangle},$$  \hspace{1cm} (21)

and

$$\xi = \ln \frac{1 + 2s_0}{1 - 2s_0}.$$  \hspace{1cm} (22)

In Eq. (20) we took into account that for a cooled QD nuclear part of the spin system $\langle ||I_S||^2 \rangle \approx ||I||^2 N + ||I_S||^2$. The first part of the right-hand side in this equation describes the fluctuation of the total nuclear spin, and the second part is the square of the mean value of the total nuclear spin in the electron hyperfine field. Equations (20), (21), and (22) completely determine the initial state of the system. From Eq. (20) one can see that $\chi_{s_0} = -\chi_{s_0}$. In the following we will consider only the sub-ensemble with a positive spin temperature, and omit $\chi$’s sub-index.

In the darkness, the relaxation of the potentials $\varsigma$, $\chi$, and $\xi$, is due to the dipole-dipole interaction between neighboring nuclei, and to phonon scattering. Each relaxation potential has its own relaxation time, for $\varsigma$, $T_\varsigma$; for $\chi$, $T_\chi$; and for $\xi$, $T_\xi$. The dipole-dipole interaction does not conserve total spin, $I_S$. As a result, the angular distribution of $I_S$ tends to the isotropic distribution,
and $\xi$ tends to zero. But the dipole-dipole interaction conserves total energy, and the modulus of $I_\Sigma$ fluctuates around its average value $\langle I_\Sigma(\chi) \rangle = \chi \hbar \langle \omega \rangle N \langle I \rangle^2 / 6$. In section 11-13 we demonstrate that the rate at which $\xi$ relaxes depends on the value of $I_\Sigma$, or, in other words, that $T_\xi$ is a function of $\chi$.

The relaxation of $I_\Sigma$, and of the frequency, $\langle \Omega \rangle$, of the quantum dot, is the result of the nuclear spin energy flow out of the quantum dots provided by spin-spin interactions between neighboring nuclei. As a result of this process, $\chi$ tends to 0, and the frequency of electron spin precession relaxes from its initial value to an asymptotic fluctuation value given by $\Omega_{\text{fluc}} = \sqrt{\langle I \rangle^2 N \langle \omega^2 \rangle}$, which is non zero for a finite system. The rate of this relaxation process depends on the value and sign of the nuclear spin temperature around the QD.

The state with $\chi \approx 0$ and $\varsigma \neq 0$ can be identified as a fluctuating nuclear spin polaron. In the fluctuation polaron state the electron “remembers” its spin direction after total nuclear spin relaxation takes place. To put it another way, the potential $\varsigma \neq 0$ describes not electron polarization but correlation between electron spin and nuclear field. The relaxation of $\varsigma$ is caused by the flipping of the electron spin. The energy of this transition, $\hbar \Omega$, cannot be taken from the nuclear spin system as $\Omega \gg \omega_L$, but is due to phonon-scattering instead $^{6,7,8}$. A sketch of connections between ENSS parameters and their relaxations is presented in Fig. 5.

In short, the relaxation of $\varsigma$ and $\chi$ is due to the open character of the QD ENSS, i.e., it depends on the environment. We will not discuss it later.

### B. The dipole-dipole relaxation of an isolated quantum dot

The dipole-dipole relaxation of the nuclear spin system is commonly characterized by a time $T_2 \propto \sqrt{\langle \omega_n^2 \rangle}$. For GaAs $T_2 \approx 10^{-8}$ s. The estimations in Refs. 1,26,27 demonstrate that, for a QD composed of an electron and polarized nuclei (nuclear spin polaron), the relaxation time of spin polarization should increase as

$$T_\xi(N, \rho_\Omega) \approx T_2 N \rho_\Omega^2,$$

where $\rho_\Omega^2 = \langle I_\Omega^2 \rangle / N^2 \langle I \rangle^2$ is determined by the nuclear spin temperature, $\chi$. $I_\Omega$ is the projection of the total nuclear spin on the direction of the nuclear hyperfine field.

We now compare the estimation of Eq. (23) with the results of our numerical simulations of the ENSS spin dynamics. For this we calculate the nuclear spin correlator,

$$G(t, \rho_\Omega) = \frac{\int I_\Sigma(t') I_\Sigma(t' + t) \cdot dt}{\int I_\Sigma(t') \cdot I_\Sigma(t') dt}.$$  

Our numerical model considers a spherical quantum dot containing $N_{\text{mod}}$ nuclei located on a cubic crystal lattice. To make the calculation feasible we take $N_{\text{mod}}$ to be of the order of hundreds, and, therefore, much less than the number of nuclei in a real quantum dot. However, $N_{\text{mod}} \gg 1$, and the adiabatic approximation may be used to model the system. Therefore, the electron spin has a constant projection, $s_\parallel$, on the total nuclear field given by $s_\Omega = \pm 1/2$, whereas the nuclear spin processes around the hyperfine electron field (directed along $\Omega = \sum_n \omega_n I_n$) with frequency $\omega_n / 2$.

As the frequency unit we take the mean frequency of nuclear spin precession, $\omega / 2 \equiv 1$. As the length unit we take the distance between nearest nuclei, $r_0 \equiv 1$. We use the following equation to calculate the precession of the nuclear spin at site $n$ in the magnetic field created by neighboring spins,

$$\varsigma = \ln \left( \frac{1 + 2s_n}{1 - 2s_n} \right) \quad \xi = 3 \langle I \rangle / \langle I \rangle^2,$$

where $\varsigma = \beta s_0 / s_\parallel$.
The time dependence of the spin correlator can be approximated by a Gaussian distribution:

\[ G(t, |I|, N) \approx \exp \left\{ -\frac{t^2}{T_2(\beta, N)} \right\} . \]  

This equation allows us to quantitatively compare the result of our numerical experiment with the estimation of Eq. (24). Figure 9 shows our numerically calculated value for \( T_2(\beta, N) \) for 4 different spherical quantum dots, with \( N \) equal to 251, 485, 895 and 1365 nuclear spins. There \( T_2(\beta, N) \langle \omega \rangle / N \) vs. \( \rho_1^0(\beta) \) is presented; the polarization \( \rho_1^0(\beta) \) was directed along the [111] axis.

The calculated dependence of \( \rho_1^0 \) on the inverse spin temperature, \( \chi \), is presented in Fig. 6. The numerical results are in a good agreement with the simple theoretical equation,

\[ \langle \rho_1 \rangle \approx \sqrt{\langle L \rangle^2 + \langle \omega \rangle^2 / \langle \omega^2 \rangle^2}. \]  

Here \( \langle L \rangle \) is the Langevin function, averaged on the QD volume, and is given by

\[ \langle L(x) \rangle = 3 \int_0^\infty \left[ e^{x \sin^2(r)} + e^{-x \sin^2(r)} - \frac{r^2}{x \cdot \sin^2(r)} \right] r^2 dr, \]  

where \( x = \chi \omega_0 \), and the index 0 indicates the nucleus at the center of the QD. The second term under the square root in Eq. (20) describes the nuclear spin fluctuation, which is important in the limit of extremely high temperature, when \( \chi \omega_0 \) is equal to 1/\( \sqrt{N} \).

To control the numerical precision, we checked the conservation law for the energy and the total nuclear spin of the system without dipole-dipole interaction (see Fig. 7(a)). After introducing the dipole-dipole interaction (Fig. 7(b)) the total energy is still conserved (curve 3), but now all three components of the total spin have a random behavior (see, for example, curve 1 for \( I_{\Sigma, x}(t) \)). The total nuclear spin, \( \| I_{\Sigma}(t) \| \), fluctuates around its mean nonzero value that is related to the total hyperfine energy, \( \langle \omega \rangle |I| \approx \langle \Omega \rangle \propto E \). The fluctuations of hyperfine energy are negligibly small because they reflect very weak transfers of energy from hyperfine to dipole-dipole reservoirs and vice versa.

In Fig. 8 is the calculated correlator \( G(t, |I|, N) \), for various nuclear spin temperatures, as indicated. One can see that an increase of the nuclear polarization, from curve 4 with \( \rho_1^0 = 0.04 \), to curve 1 with \( \rho_1^0 = 0.152 \), decreases the spin relaxation rate. On a short time scale, the time dependence of the spin correlator can be approximated by a Gaussian distribution:

\[ G(t, |I|, N) \approx \exp \left\{ -\frac{t^2}{T_2(\beta, N)} \right\} . \]

FIG. 6: Nuclear spin polarization as a function of the inverse nuclear spin temperature for a spherical quantum dot with infinite wall. The solid curve is the averaged Langevin distribution, Eq. (24), for infinite numbers of nuclei, the dashed curve, the calculation for the SQDIB model, Eq. (26), with \( N = 489 \). Circles are the Monte Carlo simulation, also with \( N = 489 \).

FIG. 7: Conservation of the quantum dot’s parameters. (a) With the dipole-dipole interaction switched off, the spin components \( I_{\Sigma, x}, I_{\Sigma, y}, \) and \( I_{\Sigma, z} \), given by curves 1, 2, and 3, respectively, the spin modulus, \( I_{\Sigma} \), given by curve 4, and the energy of the system, \( E \), given by curve 5, are all conserved. (b) When the dipole-dipole interaction is switched on, the spin components are no longer conserved, and change their values chaotically instead (curves 1, 2, z component is now shown). But the spin modulus, curve 3, is still conserved within numerical error.
FIG. 8: Spin correlator, $G$, vs. time for different nuclear temperatures, as indicated, on a quantum dot with $N = 1365$ nuclei. Solid lines are for the numerical simulation, whereas dashed lines are a Gauss approximation with fitting parameter $T_\xi$ following Eq. (23). (1) $\beta \hbar / \omega \approx 3.5$, $\rho_0 = 0.410$, $T_\xi (\omega) = 820 \pm 15$, (2) $\beta \hbar / \omega \approx 0.18$, $\rho_0 = 0.051$, $T_\xi (\omega) = 160 \pm 15$, (3) $\beta \hbar / \omega \approx 0.078$, $\rho_0 = 0.036$, $T_\xi (\omega) = 90 \pm 15$, (4) $\beta \hbar / \omega \approx 0.05$, $\rho_0 = 0.027$, $T_\xi (\omega) = 60 \pm 15$.

FIG. 9: Dependence of nuclear spin relaxation time on nuclear spin polarization. Circles are for $N = 251$, squares for $N = 485$, diamonds for $N = 895$, and triangles for $N = 1365$. All points coincide well with the straight line calculated from Eq. (24), with slope $T_2 \gamma \approx 3$.

IV. SUMMARY

(1) We demonstrated that for the short correlation time limit the rate of nuclear relaxation on electrons is proportional to the correlation time, i.e., $T_{1e} \propto \tau_c^{-1}$. In the opposite limit of long correlation time $(\Omega \tau_c \gg 1)$ we found that $T_{1e} \propto \tau_c$. The maximal rate of nuclear polarization by the QD’s electron is reached for an intermediate value of correlation time, $\Omega \tau_c \approx 1$. In this case the leakage factor reaches its maximum. Nuclear polarization in the long correlation time regime is not efficient, which follows directly from the general equations in Ref. [28], that connect nuclear polarization and relaxation rates with electron spin correlator.

(2) The nuclear spin diffusion inside a QD increases the average nuclear spin temperature and decreases nuclear polarization. The diffusion is a result of the spatial dependence of the spin relaxation rate and of the Knight field. Nuclear polarization diffuses from the QD’s center to its periphery. In the vicinity of the barrier, the Knight field is comparable with the local dipole field, and strong dipole-dipole relaxation destroys nuclear polarization. This effect decreases the mean value of the nuclear polarization by a factor of more than 4. One should take this effect into account when experimentally describing the value of the nuclear polarization.

(3) The indirect hyperfine field is contributed by a macroscopically high number of QD nuclei. The strength of the indirect hyperfine interaction between nuclei increases for longer $\tau_c$, and reaches a maximum for $\Omega \tau_c \geq 1$. This maximum is about $h \omega_n^2 / \Omega$. It is inversely proportional to nuclear polarization. Usually the field is less than the Knight field, $h \omega_n \Omega$. The indirect field plays an important role in the problem of electron spin dephasing. It may also be important in the realization of dynamic nuclear self-polarization, where electron polarization and Knight field are equal to zero.

(4) In the regime of long correlation time, the state of the quantum dot is characterized by three thermodynamic potentials: $\varsigma$, $\xi$ and $\chi$. These potentials directly affect the average electron spin, the average nuclear spin, and the nuclear spin temperature, respectively. The relaxation of the nuclear spin potential $\xi$ by the dipole-dipole interaction affects the transfer of angular momentum to the crystal lattice. The diffusion of energy to the QD’s environment is the main mechanism for the relaxation of the inverse nuclear spin temperature, $\chi$. The relaxation of the electron spin potential $\varsigma$ is connected with phonon scattering.

(5) Our numerical simulation showed the ENSS’s behavior for the simple case of a pure hyperfine interaction between a resident electron and nuclei, and for the real case of an additional dipole-dipole interaction between nearest nuclei. They demonstrated a suppression of the $\xi$ relaxation that was caused by a decrease in the nuclear spin temperature. The dipole-dipole relaxation time was found to be proportional to the number of nuclei, and to the nuclear polarization squared. These results are in good agreement with the analytical expression introduced in Refs. [20,27].
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APPENDIX: DERIVATION OF NUCLEAR SPIN PRECESSION EQUATIONS

To derive Eq. (10) we introduce Eq. (10) in Eq. (9)

\[
\frac{ds}{dt} = s_0 - \frac{(\Omega \tau_c)^2 s_0 - [\Omega \times s_0] \cdot \tau_c - (\Omega \cdot s_0) \cdot \tau_c^2}{1 + (\Omega \tau_c)^2}
\]

(A.1)

For small nuclear polarization \(||I|| \ll \|I\||\), and the vector product in the right-hand side of Eq. (A.1) plays the main role in the average rate of nuclear polarization, i.e., \((\langle I \rangle)^2 [\Omega \times I_\parallel] \tau_c^2 \rangle \ll \langle [\Omega \times s_0] [\Omega \times I] \tau_c^2 \rangle \ll \langle [\Omega \times s_0] I_\parallel \tau_c \rangle \ll \frac{\omega_n \tau_c \|I\|^2 s_0}{2}.\ Here \(\langle \cdots \rangle\) represents an average over the nuclear spin direction. Eq. (10) directly follows from this.

It follows from (Eq. 12) that

\[
\langle \frac{dI_n}{dt} \rangle_s = \frac{\omega_n^2 \tau_c}{4} \left\{ \begin{array}{l} 2I_n^\parallel + I_n^\perp (2 + (\Omega \tau_c)^2) + \\
\Omega \times I_\parallel \tau_c \end{array} \right\}.
\]

(A.2)

where \(\langle \cdots \rangle_s\) represents the average over the initial spin direction. The time dependence of electron spin is given by Eq. (10). Eq. (A.2) is an odd function of the electron spin, and to linear approximation it does not depend on polarization. For a random initial electron spin distribution \((s_\alpha(0)s_\beta(0)) = \delta_{\alpha,\beta}\|s\|^3 = \delta_{\alpha,\beta}/4\), yielding

\[
\frac{1}{\tau_e} \left\langle \int_0^\infty \left( s(t) \cdot I_n \right) \int_0^t s(\tau) d\tau \right\rangle \exp \left\{ -\frac{t}{\tau_e} \right\}_s
\]

\[
= \frac{\tau_e}{4} \left( \frac{I_n^\parallel}{1 + (\Omega \tau_c)^2} - \frac{I_n^\perp}{1 + (\Omega \tau_c)^2} \right),
\]

(A.3)

and finally

\[
\langle \frac{dI_n}{dt} \rangle_s = -\frac{\omega_n^2 \tau_e}{4} \left\{ \begin{array}{l} 2I_n^\parallel + I_n^\perp (2 + (\Omega \tau_c)^2) + \\
\Omega \times I_\parallel \tau_c \end{array} \right\}.
\]

(A.5)

The first term in the right-hand side of Eq. (A.5) contains the relaxation of the nuclear polarization components both parallel \((I_n^\parallel)\) to and transverse \((I_n^\perp)\) to \(\Omega\), see Eq. (12). Its last term describes the nuclear spin precession in the indirect hyperfine field, see Eq. (13).

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In this paper the field is given in energy units. The negative sign of the Knight field implies that the hyperfine energy has a minimum when $I$ and $s_0$ are parallel.

We neglect the part of Eq. (9) proportional to the square of the mean electron spin.

This saturation value is determined by the fluctuation of the electron spin along $\Omega$. The average values of nuclear spins, $\langle I \rangle$, and frequency, $\langle \Omega \rangle$, are parallel to each other, and the relaxation rate for the polarized nuclei, $I_\parallel \gg \| I \| / \sqrt{N}$, decreases as $(1 + (\Omega \tau_c)^2)^{-1}$. For states with low polarization the fluctuation of the nuclear spin directions are more important than the fluctuation of the modulus. For these states, increasing the correlation time decreases relaxation rate by a factor of 2.

A fluctuating magnetic polaron was investigated in the Raman spectroscopy of diluted magnetic semiconductors in Refs. 21,30,31,32.

As is well known from magnetism theory, the energy of magnetic anisotropy for a cubic spin lattice is proportional to $H_{\Sigma, x}^2 + H_{\Sigma, y}^2 + H_{\Sigma, z}^2 + O(H_0^2)$. It is important only for high enough nuclear polarization.