Long-term dynamics of the electron-nuclear spin system of a semiconductor quantum dot

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A quasiclassical 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 τ_c of electron-spin precession, with frequency Ω, 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 nonzero even if the 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 (χ, ξ, ς) 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.[1–3] This strong interest has been motivated by potential spintronics and quantum information applications, for which semiconductor quantum dots are promising.[2–5] The spin dynamics of this system is described by a variety of relaxation times that range from nanoseconds to seconds.

Optical orientation is a commonly used method to create and control[6] the ENSS with a high degree of polarization. Nuclear polarization is caused by the Fermi hyperfine interaction[7] between nuclear spins and photo-oriented electrons. A simple theoretical description of the ENSS behavior is the short correlation time approximation (SCTA).[1,6,8] (The electron correlation time, τ_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 (Ω) in the local nuclear hyperfine field, and nuclear-spin precession (ω) 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., $N \propto 10^5$ 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 τ_c. Small deviations of these frequencies during τ_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 continuous-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 or optically oriented charge carriers) 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. This scattering polarizes the resident electron and destroys its spin correlation with nuclear spins. 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.[9–11] We suppose that the quantum dot contains a macroscopically large numbers of nuclei $N \gg 1$. In this case the nuclear polarization and relaxation is connected to macroscopically large numbers of external actions. As we demonstrate in this paper, it opens the way for a classical description of nuclear polarization and nuclear relaxation of the resident electron.

In Sec. 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-spin relaxation times. In Sec. 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 QDs environment by introducing the leakage factor approximation, and we do not discuss the specifics of spin diffusion of the nuclear polarization outside the QD.[12–14]

To model the spin system we use the quasiclassical approximation, which is valid for quantum dots with large numbers of nuclear spins.[15,16] Finally, in Sec. 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.[17–19] 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, $\langle s(t_0)\rangle = s_0$. 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,[1]

$$\hat{H} = \frac{\pi}{3}\mu_B \sum_n \frac{\mu_n}{I_n}(\boldsymbol{s}\cdot\boldsymbol{I}_n)\,\delta(\boldsymbol{r}-\boldsymbol{R}_n), \tag{1}$$

where μ_B is the Bohr magneton, $\boldsymbol{s}$ and $\boldsymbol{r}$ are the spin and position of the electron, and μ_n, $\boldsymbol{I}_n$, and $\boldsymbol{R}_n$ are the magnetic moment, spin, and position of the nth nucleus, respectively. The sum in Eq. (1) runs over all the nuclei inside the QD. The hyperfine energy has a maximum when $\boldsymbol{s}$ and $\boldsymbol{I}_n$ are parallel and 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 $\mu_n \equiv \mu_I$.

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

$$\boldsymbol{\Omega} = \sum_n \omega_n \boldsymbol{I}_n, \tag{2}$$

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

$$\Omega_{\max} = I\sum_n \omega_n. \tag{3}$$

The nuclear spins have a quantum spin value $I_n \approx 1$ (in dimensionless units or $\hbar$ in physical units). Nevertheless the total nuclear hyperfine field on the electron is created by a macroscopically large number of nuclei and in our case it may be considered with high precision a classical field (see Appendix A). It is common to separate $\boldsymbol{\Omega}$ in two parts, average and fluctuation: $\boldsymbol{\Omega} = \langle\boldsymbol{\Omega}\rangle + \Delta\boldsymbol{\Omega}$ (see, for example, Ref. 15). For high nuclear-spin temperature (low polarization) the distribution of fluctuations does not depend on $\langle\boldsymbol{\Omega}\rangle$. In the following, average and fluctuation refer to the time evolution and time fluctuation of the nuclear spins. Because $\langle\boldsymbol{\Omega}\rangle$ and $\Delta\boldsymbol{\Omega}$ are not correlated, $\langle\Omega^2\rangle = \langle\Omega\rangle^2 + \langle(\Delta\Omega)^2\rangle$.

$\boldsymbol{\Omega}$ is many orders of magnitude larger than the frequency of nuclear-spin precession in the electron hyperfine field, $\omega_n \boldsymbol{s}$. At the same time, $\Omega \geq \Omega_{\text{fluc}}$, where

$$\Omega_{\text{fluc}} = \sqrt{\|I\|^2 \sum_n \omega_n^2} = \|I\|\sqrt{\langle\omega^2\rangle}\sqrt{N} \gg \|s\|\sqrt{\langle\omega^2\rangle}. \tag{4}$$

Here, $\Omega_{\text{fluc}} = \sqrt{\langle\Omega^2\rangle}|_{\Delta\Omega=0}$ is the characteristic value of the fluctuations of the electron-spin precession frequency, $\|I\| = \sqrt{I(I+1)}$ and $\|s\| = \sqrt{s(s+1)}$, are the modulus of nuclear and electron spin, respectively,[38]

$$\omega_n = \frac{16\pi\mu_B\mu_n}{3I_n\hbar}\|\psi(R_n)\|^2, \tag{5}$$

and $\psi(R_n)$ is the electron wave-function on the nth nucleus.[39]

$\Omega_{\max}$ 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[1] $\Omega_{\max} \approx 10^{11}$ s^{-1}. Values of $\Omega_{\text{fluc}} \propto \Omega_{\max}/\sqrt{N}$ and $\langle\omega\rangle \propto \Omega_{\max}/N$ depend on the QD volume. For a quantum dot

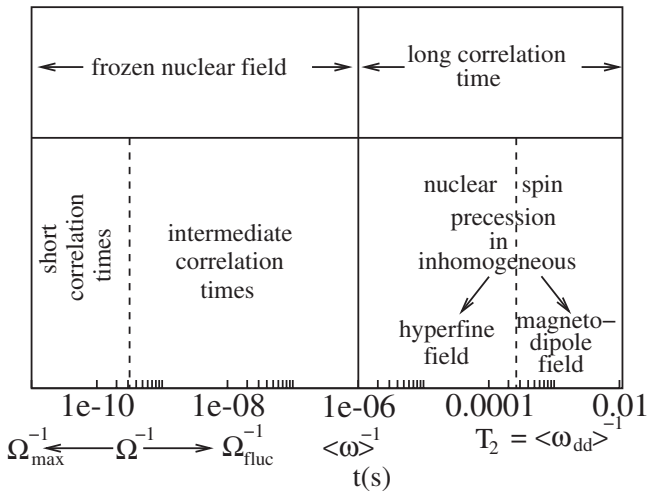

FIG. 1. Time scale for different regimes of hyperfine interaction in quantum dot. (1) Short correlation time regime ($\tau_c < \Omega^{-1}$). During τ_c the electron and nuclear spins rotate around a small angle. (2) Intermediate correlation time regime ($\Omega^{-1} < \tau_c < \langle\omega\rangle^{-1}$). During τ_c the electron spin rotates around a large angle with constant angular velocity. (3) Long correlation time regime ($\tau_c > \langle\omega\rangle^{-1}$). During τ_c the electron and nuclear spins change their direction. For $\langle\omega_{dd}\rangle^{-1} > \tau_c > \langle\omega\rangle^{-1}$ the total nuclear spin, I_Σ is conserved, but Ω changes direction as a result of the nuclear-spin precession in the nonuniform electron hyperfine field. $\langle\omega_{dd}\rangle$ is the frequency of nuclear-spin precession in the local magnetic field of the neighboring nuclei. For $\langle\omega_{dd}\rangle^{-1} < \tau_c$, the dipole-dipole interaction between spins of neighboring nuclei changes I_Σ.

with $N \propto 10^5$ nuclei $\Omega_{fluc} \propto 3 \times 10^8$ s^{-1} and $\langle\omega\rangle \propto 10^6$ s^{-1}. Characteristic frequencies and times are collected in Fig. 1.

Under sample illumination, the photocarriers and photon scattering and carrier capture and photon absorption are the main mechanisms of the ENSS interaction with the environment. These perturbations establish some initial value for the localized electron-spin polarization (mean electron spin $\boldsymbol{s}_0$) and destroy correlations between the electron and the nuclear spins. We will not discuss these details but will treat these perturbations as collisions with external particles. We will suppose that these collisions are distributed randomly in time. Then, the probability of unperturbed electron-spin precession during time t between two collisions is

$$W(t) = \exp\{-t/\tau_c\}/\bar{\tau}_c. \tag{6}$$

We will later consider τ_c as a correlation time for the electron spin. Between collisions nuclear spins change their directions by a very small angle. In the zeroth order of approximation, the spin, $s(t)$, of the resident electron precesses in the constant (frozen) classical nuclear field. For classical vector precession with angle frequency $\boldsymbol{\Omega}$, $\boldsymbol{s}(t)$ is described by the differential equation $d\boldsymbol{s}/dt = [\boldsymbol{\Omega} \times \boldsymbol{s}]$ and

$$\begin{aligned} \boldsymbol{s}(t) &= \hat{\Gamma}(\boldsymbol{\Omega}, \delta)\boldsymbol{s}(t_0) \\ &= \boldsymbol{s}_{\boldsymbol{\Omega}}(t_0) + (\boldsymbol{s}(t_0) - \boldsymbol{s}_{\Omega}(t_0))\cos(\Omega \cdot \delta t) \\ &\quad + [\boldsymbol{e}_{\boldsymbol{\Omega}} \times \boldsymbol{s}(t_0)]\sin(\Omega \cdot \delta t), \end{aligned} \tag{7}$$

where $\boldsymbol{s}(t_0)$ is the spin value at initial moment t_0,

$$\boldsymbol{s}_{\Omega} = (\boldsymbol{s} \cdot \boldsymbol{e}_{\Omega})\boldsymbol{e}_{\Omega}, \tag{8}$$

$\boldsymbol{e}_{\boldsymbol{\Omega}} = \boldsymbol{\Omega}/\Omega$, $\delta t = t - t_0$, and t_0 is the time when the external action polarized the resident electron. This linear equation is valid for the mean value of classical and quantum-mechanical spins. Quantum mechanically, the existence of electron-spin components rotating perpendicular to Ω implies that the electron state is mixed after a collision.

For the CW photoexcitation the collisions with external particles are randomly distributed on time, with probability given by Eq. (6), and the average over time of the spin value is described by the well-known formula:[1,6]

$$\bar{\boldsymbol{s}} = \frac{\boldsymbol{s}_0 + [\boldsymbol{\Omega} \times \boldsymbol{s}_0]\tau_c + (\boldsymbol{\Omega} \cdot \boldsymbol{s}_0)\boldsymbol{\Omega}\tau_c^2}{1 + (\Omega\tau_c)^2}, \tag{9}$$

where $\boldsymbol{s}_0$ is the average value of the initial spin. Consider the classical spin components (s_x, s_y, s_z) [Eq. (7)] their corresponding quantum operators $(\hat{s}_x, \hat{s}_y, \hat{s}_z)$ and the corresponding averages of these quantum operators. The equations describing the time evolution of any of these set of operators in a classical magnetic field is the same. The formal transition from the classical to the quantum-mechanical description is achieved by substituting (s_x, s_y, s_z) with $(\hat{s}_x, \hat{s}_y, \hat{s}_z)$. If the precession frequency Ω is directed along the Z axis, the Green's function for electron spin has the form:

$$\hat{\Gamma}(\boldsymbol{\Omega}, t) = \begin{pmatrix} \cos(\Omega t) & -\sin(\Omega t) & 0 \\ \sin(\Omega t) & \cos(\Omega t) & 0 \\ 0 & 0 & 1 \end{pmatrix}, \tag{10}$$

in both classical and quantum cases. From this it follows that the external classical magnetic field can be excluded from discussion by transitioning to a rotated frame.[9] In this section we investigate the influence of the quasiclassical nuclear field on the nuclear polarization and nuclear relaxation on electrons. The direct analogy between quantum and classical equations for electron-spin dynamics in a classical field allow us to perform this analysis using a classical description.

In the short correlation time approximation $\bar{\boldsymbol{s}} \approx \boldsymbol{s}_0$, whereas in the opposite limit ($\Omega\tau_c \geq 1$), the mean value of the electron spin depends on the angular distribution of frequencies, $\boldsymbol{\Omega}$. For a random distribution (unpolarized nuclear system) $\langle\boldsymbol{\Omega}\rangle = 0$, $(\boldsymbol{\Omega} \cdot \boldsymbol{s}_0) \cdot \boldsymbol{\Omega} = \Omega^2 \boldsymbol{s}_0/3$, and $\bar{\boldsymbol{s}} \approx \boldsymbol{s}_0/3$. For a polarized nuclear-spin system $\langle\boldsymbol{\Omega}\rangle \gg \boldsymbol{\Omega}_{fluc}$, 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 photoelectron polarization, $\boldsymbol{s}_0$. For $\langle\boldsymbol{\Omega}\rangle$ along $\boldsymbol{s}_0$ $(\boldsymbol{\Omega} \cdot \boldsymbol{s}_0) \approx \Omega s_0$ and $\bar{\boldsymbol{s}} \approx \boldsymbol{s}_0$.

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

$$\frac{d\boldsymbol{I}_n}{dt} = \omega_n[\boldsymbol{s}(t)\times\boldsymbol{I}_n(t)]$$
$$= \omega_n[\bar{\boldsymbol{s}}\times\boldsymbol{I}_n(t)] + \omega_n\left[(\boldsymbol{s}(t)-\bar{\boldsymbol{s}})\times\int_{t_0}^{t}\frac{d\boldsymbol{I}_n(t')}{dt'}dt'\right]$$
$$+ \omega_n\left[\int_{t0}^{t}\left(\hat{\Gamma}(\boldsymbol{\Omega},t-t')\frac{d\boldsymbol{s}(t')}{dt'}\right)dt'\times\boldsymbol{I}_n(t)\right] \quad (11)$$

where $\bar{\boldsymbol{s}}=\lim_{t\to\infty}\frac{1}{t}\int_{t_0}^{t}\boldsymbol{s}(t')dt'$ is the mean value of the electron spin. The difference between classical Eq. (11) and its quantum analog is only in that the quantum equation will have operators for the electron and nuclear spins.

There are a macroscopic number of nuclei in a quantum dot. Therefore, polarization and relaxation of ENSS is a result of many collisions with external particles. Averaging over these collisions gives

$$\frac{d\boldsymbol{I}_n}{dt} \approx \omega_n[\bar{\boldsymbol{s}}\times\boldsymbol{I}_n(t)]$$
$$+\frac{\omega_n}{\tau_c}\int_{t_0}^{\infty}dt e^{-t/\tau_c}\left\langle\omega_n\left\{\boldsymbol{s}(t)\times\int_{t0}^{t}[\boldsymbol{s}(t')\times\boldsymbol{I}(t)]dt'\right\}\right.$$
$$\left.+\sum_m\omega_m\left\{\int_{t0}^{t}\hat{\Gamma}(\boldsymbol{\Omega},t-t')[\boldsymbol{I}_m(t')\times\boldsymbol{s}(t')]dt'\right\}\times\boldsymbol{I}_n(t)\right\rangle_s, \quad (12)$$

where angle brackets $\langle\cdots\rangle$ indicate that the averaging is over initial sates of the electron spins oriented via a collision with an external particle, $\langle\boldsymbol{s}(t_0)\rangle=\boldsymbol{s}_0$, $\langle s_\alpha(t_0)s_\beta(t_0)+s_\beta(t_0)s_\alpha(t_0)\rangle_s/2=\delta_{\alpha,\beta}s^2/3=\delta_{\alpha,\beta}/4$, and t_0 is the moment of this collision. This equation is valid for classical spin with $s^2=3/4$ and $s_0^2\ll s^2$.

In the quantum-mechanical description the spin operators do not commute. Therefore, in the quantum-mechanical case, simple products $s_\alpha(t)s_\beta(t')$ corresponds to the symmetrized counterparts $[\hat{s}_\alpha(t)\hat{s}_\beta(t')]\equiv[\hat{s}_\alpha(t)\hat{s}_\beta(t')+\hat{s}_\beta(t')\hat{s}_\alpha(t)]/2$. As a result of this symmetrization the difference between the quantum mechanical and the classical descriptions again disappeared.

We analyze the regime of short and intermediate correlation times $\omega_n\tau_c\ll 1$. Therefore, in Eq. (12) $\boldsymbol{I}_n(t)\approx\boldsymbol{I}_n(t')$, and we may rewrite the first part in angle brackets in the form:

$$\left\langle\left\{\boldsymbol{s}(t)\times\int_{t_0}^{t}[\boldsymbol{s}(t')\times\boldsymbol{I}_n(t)]dt'\right\}\right\rangle_s$$
$$\approx\left\langle\left\{[\boldsymbol{s}(t)\cdot\boldsymbol{I}_n(t)]\int_{t_0}^{t}\boldsymbol{s}(t')dt'\right\}\right.$$
$$\left.-\left\{\left[\boldsymbol{s}(t)\cdot\int_{t_0}^{t}\boldsymbol{s}(t')dt'\right]\right\}\boldsymbol{I}_n(t)\right\rangle_s. \quad (13)$$

After averaging over the electron-spin directions this part of Eq. (12) describes two well known processes: (i) the nuclear-spin relaxation

$$\left.\frac{\partial\boldsymbol{I}_n}{\partial t}\right|_{rel} = -\frac{\omega_n^2}{\langle\omega^2\rangle}\hat{T}_{1,e}^{-1}(\Omega\tau_c)\boldsymbol{I}_n, \quad (14)$$

and (ii) the nuclear-spin precession in the indirect hyperfine field created by all nuclei of the QD

$$\left.\frac{\partial\boldsymbol{I}_n}{\partial t}\right|_{I_n} = \frac{\omega_n^2}{\langle\omega^2\rangle}[\boldsymbol{\eta}(\Omega\tau_c)\times\boldsymbol{I}_n]. \quad (15)$$

In the coordinate system with Z axis parallel to $\boldsymbol{\Omega}$, the tensor of nuclear-spin-relaxation time $\hat{T}_{1,e}$ has a diagonal structure[40] $T_{1,e,xx}^{-1}(\Omega\tau_c)=T_{1,e,yy}^{-1}(\Omega\tau_c)=(T_e^{-1}(0)+T_e^{-1}(\Omega\tau_c))/2$, $T_{1,e,zz}^{-1}(\Omega\tau_c)=T_{1,e}^{-1}(\Omega\tau_c)$, where

$$T_{1e}^{-1}(\Omega\tau_c) = \frac{2\langle\omega^2\rangle\|s\|^2\tau_c}{3[1+(\Omega\tau_c)^2]} \quad (16)$$

and

$$\boldsymbol{\eta}_n(\Omega\tau_c) = \frac{\Omega\tau_c}{2T_e(\Omega,\tau_c)} \quad (17)$$

is a quasiclassical vector because the indirect hyperfine interaction is a result of interaction with a macroscopically large numbers of nuclei. In this case, the indirect hyperfine field gives a good example of the classical Weiss field.[20]

In the right-hand term of Eq. (12), the first part between angular brackets contains two nuclear and one electron spins. After averaging over many collisions with external particles (see Appendix B) it becomes proportional to the mean value of the electron spin and describes the nuclear dynamical polarization through the localized electron. At the same time, this result depends on the local value of the product of nuclear-spin components, $\{I_{n,\alpha}(t')I_{m,\beta}(t)\}\approx\{I_{n,\alpha}(t)I_{m,\beta}(t)\}$. The value of this product at a given time is unknown, but what matters are average values which are known. The time of nuclear-spin polarization and relaxation is usually many times larger than the characteristic time of the neighboring nuclear-spin interaction. Then, the local value of the product of nuclear-spin components can be substituted by its averaged value $\langle\{I_{n,\alpha}(t)I_{m,\beta}(t)\}\rangle_I\approx\delta_{n,m}\delta_{\alpha,\beta}\|I_n\|^2/3$, where the angle bracket $\langle\cdots\rangle_I$ denotes averaging over a random distribution of the nuclear-spin direction. In this approximation

$$\left\langle\left.\frac{\partial\boldsymbol{I}_n}{\partial t}\right|_{dp}\right\rangle_{I,s} \approx \frac{\omega_n^2\|\boldsymbol{I}_n\|^2\boldsymbol{s}_0}{\|\boldsymbol{s}\|^2\langle\omega^2\rangle T_e(\Omega\tau_c)}. \quad (18)$$

The approximation in this equation is valid for high enough nuclear polarization when $\boldsymbol{s}_0$, $\bar{\boldsymbol{s}}$, and $\boldsymbol{\Omega}$ are practically parallel to each other.

Results of classical calculations have no differences with respect to quantum-mechanical calculations for the polarization in an external magnetic field.[1,6] These results are also in agreement with quantum-mechanical calculations of the nuclear-spin relaxation on localized exitons for anisotropic quantum dots.[21] This calculation contains two parts—the electron precession in the total quasi classical nuclear field and the weak interaction between electron and one nucleus that transfers the polarization from electron to nucleus, and vice versa. The first one can be reduced by introducing a frame rotation and does not contain any quantum specific

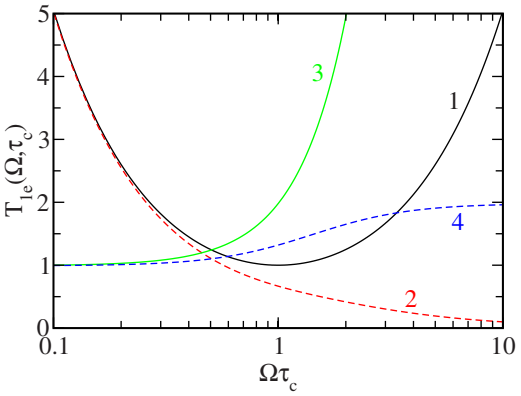

FIG. 2. (Color online) Nuclear-spin-relaxation time on the resident electron vs correlation time τ_c (curves 1 and 2) and *vs.* frequency Ω (curves 3 and 4) of the electron-spin precession in the nuclear hyperfine field. Solid curves 1 and 3 show the relaxation time for the nuclear polarization component along $\mathbf{\Omega}$. Dashed curves 2 and 3 show the relaxation time of the nuclear polarization component transversal to $\mathbf{\Omega}$. Dependences of relaxation time on τ_c (1 and 2) were calculated for a constant value of Ω. They are normalized to the value of $T_{1,e}$ at $\Omega\tau_c=1$. Dependences of relaxation time on Ω (3.4) were calculated for a constant value of τ_c, and are normalized to the value of $T_{1,e}$ at $\Omega=0$ The nuclear dynamic polarization is ineffective for $\Omega\tau_c \gg 1$ because in this region the value of the relaxation time for the nuclear-spin longitudinal component increases fast, and the leakage factor in the Eq. (20) decreases. The difference between times $T_{1,e}$ for longitudinal and transverse component of nuclear polarization in the region $\Omega\tau_c \gg 1$ increases ε in Eq. (20) but decreases nuclear polarization.

corrections. That there are no quantum corrections is also a result of the weak interaction between the electron and nuclear spins. After averaging over a random spin distribution, these processes contain only mean values of spins, and of correlations between spin components that depend only on the spin modulus. The quantum-mechanical values of these moduli are introduced in a classical description as a phenomenological parameter. At this level of precision there is no difference between classical and quantum-mechanical fluctuations because the measurable value of fluctuations in a high-temperature approximation is proportional to the spin modulus squared. [For a derivation of Eqs. (14), (17), and (18) see Appendix B.]

Relaxation of nuclear polarization and nuclear-spin precession is determined by square of the fluctuations of the electron hyperfine field. They are proportional to $\|s\|^2=3/4$ because $T_{1e}\propto\|s\|^{-2}$. 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,[6] $T_{1e}(0,\tau_c)^{-1}=2\|s\|^2\tau_c\cdot\langle\omega^2\rangle/3$.

For intermediate correlation times, the relaxation time for the component of I longitudinal to $\mathbf{\Omega}$, increases as both Ω and τ_c increase, yielding[9] $T_{1e}(\Omega,\tau_c)=(1+(\Omega\tau_c)^2)T_{1e}(0,\tau_c)$ (solid curves 1 and 3 in Fig. 2). On the other hand, the relaxation rate of the polarization component, $I_\perp$, transverse to $\mathbf{\Omega}$, behaves differently depending on whether Ω is increased or τ_c is increased. For τ_c increasing, $T_{1e,\perp}$ decreases monotonically (curve 2). As Ω increases, $T_{1e,\perp}$ increases saturating at $T_{1e\perp}(\infty,\tau_c)=2T_{1e}(0,\tau_c)$ (curve 4).[41]

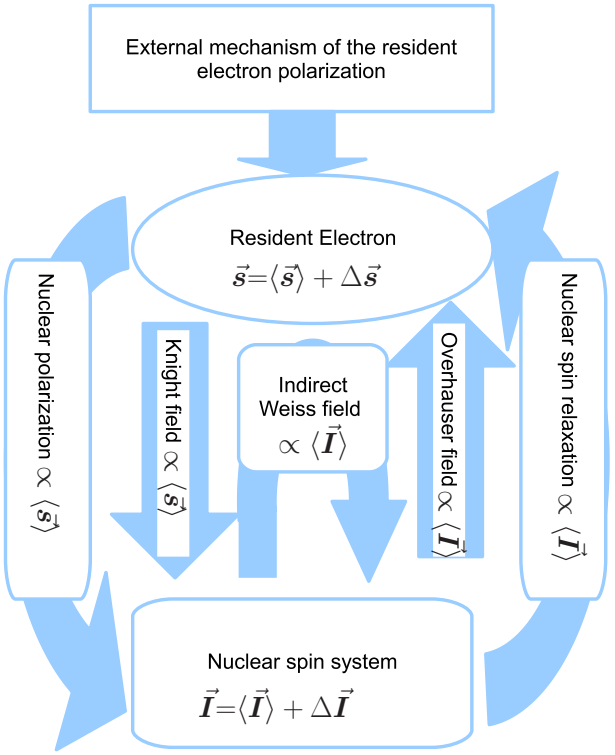

FIG. 3. (Color online) Different mechanisms for the hyperfine interaction between the resident electron and the nuclei. Nuclear spins precess in the mean Knight field and are polarized by the oriented resident electron. The rates of these effects are proportional to the average electron spin. Polarized nuclei create a mean Overhauser field on the electron, and a mean indirect Weiss field on each other. Their polarization also relaxes via interaction with the resident electron. The rates of these three mechanisms are proportional to the value of nuclear polarization.

The nuclear polarization, its relaxation rate, and the indirect hyperfine field are all proportional to $\omega_n^2\propto\|\psi(R_n)\|^4$, and all have a strong spatial dependence. They have a maximum in the center of the QD, and they decrease toward the border. In the limit of frozen nuclear field, i.e., short and intermediate correlation times, $\omega_n\tau_c$ in Eqs. (14), (17), and (18) is much smaller than 1. In Eq. (17), for an intermediate time, this parameter may be rewritten as ω_n/Ω, which is much less than 1, implying that characteristic rates for nuclear-spin polarization, relaxation, and indirect hyperfine interaction are many times less then ω_n. These slow processes are relevant only if they change the system's behavior, such as its nuclear polarization [Eq. (18)] or its relaxation rate [Eq. (14)].

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.[22–24])

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 τ_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 τ_c. For a depolarized nuclear system, i.e., when $\Omega \approx \Omega_{\text{fluc}}$, 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}, and $\Omega_{\text{fluc}} \approx 3\times 10^8$ s^{-1}), the shortest relaxation time for depolarized nuclei calculated from Eq. (16) and condition $dT_{1e}/d\tau_c=0$ is $T_{1e} \approx 10^{-3}$ s. It is reached for $\tau_c^{(\min)} \approx 3\times 10^{-9}$ s. On the other hand, for 100% nuclear polarization, $T_{1e} \approx 10^{-1}$ s. This relaxation time is reached when $\tau_c^{(\min)}=10^{-11}$ s. Since the short correlation time approximation is valid for $\tau_c \ll \tau_c^{(\min)}$, 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. (18)] and relaxation [Eq. (14)] gives

$$\langle \boldsymbol{I}_n \rangle = \frac{\|I\|^2 \boldsymbol{s}_0}{\|s\|^2}. \tag{19}$$

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

Equation (19) 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. (19) the phenomenological leakage factor[6] $f=T_{1l}/(T_{1e}+T_{1l}) \leq 1$. (Here T_{1l} is the relaxation time for additional relaxation channels.) When T_{1l} is finite, the leakage factor, f, and the average polarization, $\langle \boldsymbol{I} \rangle$, 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[6] 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. (19), and it has to be reached at a time T_2. Nevertheless, it is well known[1,6] from many experiments and theoretical calculations that the optically induced nuclear polarization cannot usually be ignored. This polarization is due to a decrease in the nuclear-spin temperature, Θ. The effect results from the balance of two energetic flows: cooling of nuclear spins by oriented electrons in an external magnetic field $\boldsymbol{B}$: $J_{\text{cool}} \propto -(\boldsymbol{B}\cdot\partial\boldsymbol{I}/\partial t|_{\text{dp}})$ and heating of nuclear spins by random fluctuations of electron polarization: $J_{heat}=\beta C$. 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[6]

$$\beta = f\frac{4I}{\mu_I}\frac{(\boldsymbol{B}\cdot\boldsymbol{s}_0)}{B^2+\varepsilon B_L^2}, \tag{20}$$

where B_L^2 is the characteristic value of the random local field squared and ε is a number of the order of one. β has the opposite sign for parallel and antiparallel orientations of $\boldsymbol{B}$ and $\boldsymbol{s}_0$. (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.[9,25]) β is positive if $\boldsymbol{s}_0$ is parallel to $\boldsymbol{B}$, 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.[6] For the special case of Eq. (17), the indirect interaction between two nuclei, n and m, depends on n and m only through their product, $\omega_n\omega_m$, and has no influence on the local field part of Eq. (20): $\varepsilon B_L^2 \approx 3B_{\text{dd}}^2$. Here B_{dd}^2 is the square of the average dipole-dipole part of the local field.

Equation (20) is valid for high nuclear-spin temperature, i.e., for $\beta\mu_I\sqrt{B^2+\varepsilon B_L^2} \ll 1$, and was derived for a spatially uniform $\boldsymbol{B}$ and T_{1e}. An average Knight field should be included in the regular external field, and the total magnetic field, $\boldsymbol{B}+\boldsymbol{B}_{K,n}$, depends on the nuclear position inside the QD.

The approximation that ignores the spatial dependence of the hyperfine interaction inside the dot is known[26,27] 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 $\langle \boldsymbol{I}_n \rangle \propto \beta \boldsymbol{B}_{k,n}$.

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 $\hbar\langle\omega\rangle s_0/N_R \approx \hbar\Omega_{\max}s_0(4/I^3N^4)^{1/3}$, 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 $\hbar 10^4$ s$^{-1} \approx \hbar\omega_{\text{dd}}$,

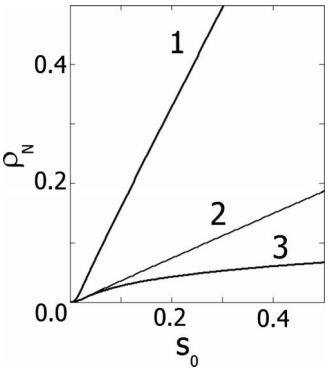

FIG. 4. Dependence of nuclear polarization on the mean spin of the resident electron. Here $T_{1e}(0)=T_{1l}$. Curve 1 was calculated using the box model with $\Omega_{\max}\tau_c \ll 1$, curve 2 using the SCIB model with $\Omega_{\max}\tau_c \ll 1$, and curve 3 using the SCIB model with $\Omega_{\max}\tau_c=10$.

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 β contains averaged values, i.e.,

$$\beta = -\frac{4}{\hbar} f \frac{\langle\omega^3\rangle s_0^2}{\langle\omega^4\rangle s_0^2 + \varepsilon\langle\omega_L^2\rangle\langle\omega^2\rangle}. \tag{21}$$

Here $\langle\omega^m\rangle=\Sigma_n \omega_n^m/N$ and $\langle\omega_L^2\rangle=\mu_I^2 B_L^2/\hbar^2$.

In the limit of short correlation time, the difference between the result of Eq. (21) and the one obtained with the box model is only numerical. The dimensionless saturation value of the spin temperature, $\tilde{\beta}\equiv\hbar\langle\omega\rangle\beta$, is then $\langle\omega\rangle\langle\omega^3\rangle/\langle\omega^4\rangle$ times less than that predicted by Eq. (20). It reaches saturation if $s_0^2 \gg \varepsilon\langle\omega_{\rm dd}^2\rangle\langle\omega^2\rangle/\langle\omega^4\rangle$. [For a spherical quantum dot with infinite barrier (SCDIB), we have $\langle\omega\rangle^2/\langle\omega^2\rangle\approx 0.36$, $\langle\omega\rangle\langle\omega^3\rangle/\langle\omega^4\rangle\approx 0.22$, $\langle\omega^2\rangle^2/\langle\omega^4\rangle\approx 0.17$, and $\varepsilon\langle\omega_{\rm dd}^2\rangle\langle\omega^2\rangle/\langle\omega^4\rangle\approx 5\times 10^{-3}$.)

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

$$\langle \boldsymbol{I}_n\rangle = -\beta\frac{\|I\|^2}{3}(\hbar\omega_n)\boldsymbol{s}_0. \tag{22}$$

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 4–5 times smaller than the one predicted by the box model.

In Sec. II B, we saw that T_{1e} as function of τ_c has a minimum for $\tau_c=\Omega^{-1}$. For $\tau_c>\Omega^{-1}$, increasing the correlation time and the frequency $\boldsymbol{\Omega}$ increases T_{1e}. The nuclear polarization increases $\boldsymbol{\Omega}$, implying that if one wants to optimize the nuclear polarization by varying the correlation time, one has to start from a unpolarized nuclear system with $\Omega_{\rm fluc}\tau_c \ll 1$. At the same time, τ_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 SQDIB model (curves 2 and 3) are presented. All calculations are done for $T_{1e}(0)=T_{1l}$. Curves 1 and 2 are calculated in the short correlation time approximation, curve 3 for $\Omega_{\max}\tau_c=10$. One can see that the nuclear polarization for the SQDIB model is about five times less than for the box model. For $\Omega_{\max}\tau_c=10$, increasing the time T_{1e} by increasing the nuclear polarization additionally decreases the leakage factor and slows down an increase in 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 ineffective. We will consider only spin relaxation of an isolated ENSS in the darkness. In Sec. 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 Sec. 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 QDs environment.

A. Conservation laws and thermodynamic potentials of the ENSS

The τ_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.[1,5,9–11] 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.[28–30]

In the zeroth 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, $E=\hbar\Omega s_\Omega$, and total spin, $\boldsymbol{F}=\boldsymbol{I}_\Sigma+\boldsymbol{s}$. The total spin of the nuclei, $\boldsymbol{I}_\Sigma$, is many orders of magnitude larger than that of the electron. Therefore, with high precision $\boldsymbol{I}_\Sigma$ is also conserved. Moreover, as a result of the adiabatic approximation, $\Omega \gg \omega_n$, and the electron-spin projection, s_Ω, along $\boldsymbol{\Omega}$ is also conserved and must be quantized, i.e., $s_\Omega=\pm 1/2$. The conservation of Ω follows from the conservation of energy and s_Ω.

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 subensembles

(nuclear-spin polarons) with defined value of electron-spin projection, s_Ω, on the nuclear hyperfine field, i.e., $s_\Omega = \pm 1/2$ and energies $\pm\hbar\Omega/2$. The probability to find a quantum dot in one of these subensembles 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., $\langle I\rangle \gg 1/\sqrt{N}$. Considering the integrals of motion of the system, we can write the ENSS probability distribution Φ, as a function of three thermodynamic potentials: an electron-spin potential, ς, an inverse nuclear-spin temperature, χ_{s_Ω}, for each subensemble, and a nuclear-spin potential, $\boldsymbol{\xi}(\chi_{s_\Omega})$,

$$\Phi(\varsigma,\chi,\boldsymbol{\xi}) \approx \frac{\exp\{(\varsigma s_\Omega) - (\chi_{s_\Omega}\hbar\Omega s_\Omega) + (\boldsymbol{\xi}\cdot\boldsymbol{I}_\Sigma)\}}{(4\pi)^N(\exp\{\varsigma/2\} + \exp\{-\varsigma/2\})}. \quad (23)$$

At the last moment of illumination, the total nuclear-spin mean value, $\langle \boldsymbol{I}_\Sigma\rangle$, and the electron spin, $\boldsymbol{s}_0$, are directed along the Knight field, and the nuclear inverse spin temperature under illumination is β. Therefore, after switching off the light, the nuclear inverse spin temperature in the darkness is

$$\chi_{s_\Omega} \approx \beta\frac{s_0}{s_\Omega}. \quad (24)$$

Moreover, nuclear and electron-spin potentials are given by

$$\boldsymbol{\xi} = \frac{3\langle \boldsymbol{I}_\Sigma\rangle}{\langle \|\boldsymbol{I}_\Sigma\|^2\rangle} \quad (25)$$

and

$$\varsigma = \ln\frac{1+2s_0}{1-2s_0}. \quad (26)$$

In Eq. (24) we took into account that for a cooled QD nuclear-spin system $\langle \|\boldsymbol{I}_\Sigma\|^2\rangle \approx \|\boldsymbol{I}\|^2 N + \|\langle \boldsymbol{I}_\Sigma\rangle^2\|$. The first part of the right-hand side in this equation describes the fluctuation of the total nuclear spin, and the second part the square of the mean value of the total nuclear spin in the electron hyperfine field. Equations (24)–(26) completely determine the initial state of the system. From Eq. (24) one can see that $\chi_{+1/2} = -\chi_{-1/2}$. In the following we will consider only the subensemble with a positive spin temperature and omit χs subindex.

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

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

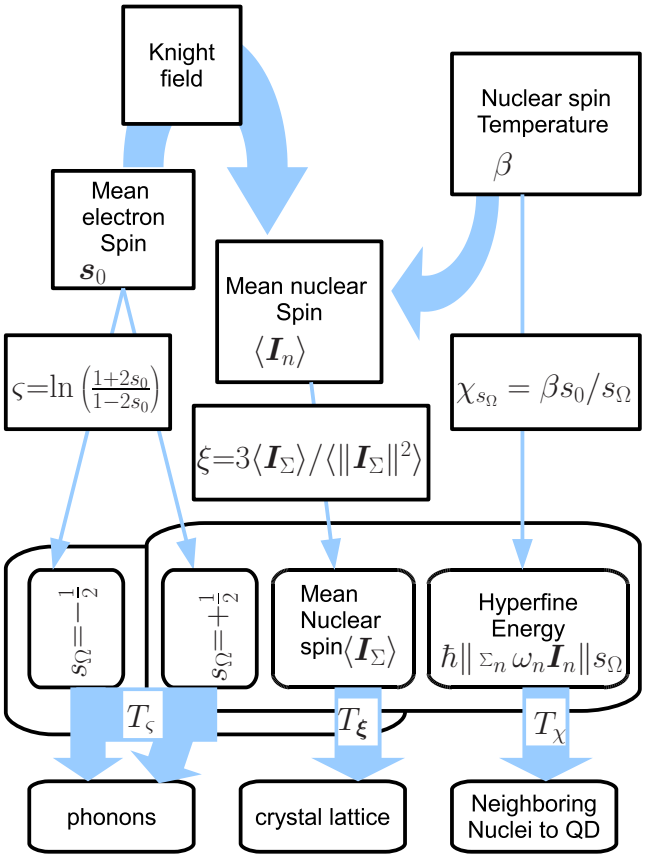

FIG. 5. (Color online) Relations between the ENSS's description under illumination and in the darkness. Under illumination the ENSS state is described by the average electron spin, s_0, and the inverse nuclear-spin temperature, β. The Knight field of the electron creates the mean polarization of the cooled nuclei. After a sharp transition to darkness, the ensemble of quantum dots splits in two subensembles with two different electron-spin projections, $s_\Omega = \pm 1/2$, on the nuclear hyperfine field. The ENSS is characterized by an electron potential, ς, by a nuclear potential, $\boldsymbol{\xi}$, and by a nuclear inverse spin temperature, χ_{s_Ω}. The relaxation of these thermodynamic potentials is due to the dipole-dipole interaction between nuclear spins and electron phonon interaction. This relaxation transfers nuclear spin to the crystalline lattice (on a time $T_{\boldsymbol{\xi}}$), energy diffusion from the quantum dot to the environment (on a time T_χ), and energy from the scattering of phonons (on a time T_ς).

The state with $\chi \approx 0$ and $\varsigma \neq 0$ can be identified as a fluctuating nuclear-spin polaron.[42] In the fluctuation polaron state the electron-spin "remembers" its angle with $\boldsymbol{\Omega}$ 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 ς 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.[9–11] One can see the direct connection among times T_{ξ}, T_{χ}, T_{ς}, and T_2 for nuclear transverse polarization relaxation and T_1 for nuclear and electrons spin energy relaxation. A sketch of connections between ENSS parameters and their relaxations is presented in Fig. 5.

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

B. Dipole-dipole relaxation of an isolated quantum dot

The dipole-dipole relaxation of the nuclear-spin system is commonly[9] characterized by a time $T_2 \propto (\sqrt{\langle \omega_{\mathrm{dd}}^2 \rangle})^{-1}$. For GaAs (Ref. 6) $T_2 \approx 10^{-4}$ s. The estimations in Refs. 1, 31, and 32 demonstrate that, for a QD composed of an electron and polarized nuclei (nuclear-spin polaron), the relaxation time of spin polarization should increase as

$$T_{\xi}(N, \rho_{\Omega}) \approx T_2 N \rho_{\Omega}^2, \tag{27}$$

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

We now compare the estimation of Eq. (27) with the results of our numerical simulations of the ENSS spin dynamics. For this we calculate the nuclear-spin correlator,

$$G(t, \rho_{\Omega}) = \frac{\int \boldsymbol{I}_{\Sigma}(t') \boldsymbol{I}_{\Sigma}(t'+t) dt}{\int \boldsymbol{I}_{\Sigma}(t') \cdot \boldsymbol{I}_{\Sigma}(t') dt}. \tag{28}$$

Our numerical model[43] considers a spherical quantum dot containing N_{mod} nuclei located on a cubic crystal lattice. To make the calculation feasible we take N_{mod} to be of the order of hundreds and, therefore, much less than the number of nuclei in a real quantum dot. However, $N_{\mathrm{mod}} \gg 1$, and the adiabatic approximation may be used to model the system. Therefore, the electron spin has a constant projection, s_{Ω}, on the total nuclear field given by $s_{\Omega} = \pm 1/2$, whereas the nuclear-spin precesses around the hyperfine electron field (directed along $\boldsymbol{\Omega} = \Sigma_n \omega_n \boldsymbol{I}_n$) with frequency $\omega_n/2$.

As the frequency unit we take the mean frequency of nuclear-spin precession, $\langle \omega \rangle / 2 \equiv 1$. 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:

$$\left. \frac{\partial \boldsymbol{I}_n}{\partial t} \right|_{\mathrm{dd}} = \gamma \sum_{m \neq n} \frac{\left[\left(\frac{1}{3} \boldsymbol{I}_m - \boldsymbol{e}(\boldsymbol{e} \boldsymbol{I}_m) \right) \times \boldsymbol{I}_n \right]}{d_{nm}^3}. \tag{29}$$

Here $\boldsymbol{e}$ is the unit vector joining sites n with spin m, d_{nm} is the distance between these sites, $\gamma = \omega_D / \langle \omega \rangle \approx 5.7 \times 10^{-2}$, and $\omega_D = \mu_n^2 / (\hbar r_0^3)$ is the characteristic frequency of the nuclear-spin precession in the field of nearest-neighbor dipoles.

The sign of the electron spin affects only the direction of the nuclear hyperfine precession but has no influence on the dipole relaxation. For this reason, we simulated only the subensemble with positive spin temperature. The initial distribution of nuclear spins is determined by the inverse temperature, χ, with natural units $\hbar \langle \omega \rangle$. This initial spin distribution was generated by a random process using the Boltzmann distribution function. In order to decrease the effects of crystal magnetic anisotropy in the initial state,[44] Ω was directed along the [111] axis.

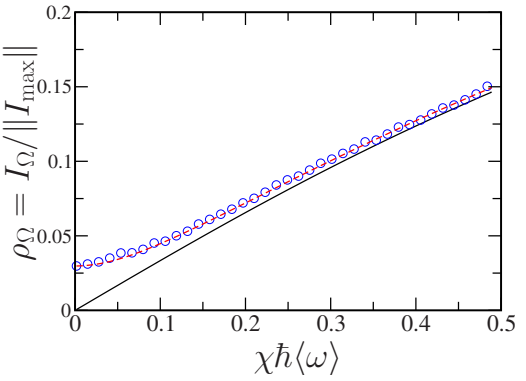

FIG. 6. (Color online) Nuclear-spin polarization as a function of the inverse nuclear-spin temperature for a spherical quantum dot with infinite wall. The solid curve is the averaged Langevin distribution [Eq. (31)] for infinite numbers of nuclei, the dashed curve, and the calculation for the SQDIB model [Eq. (30)] with $N=489$. Circles are the Monte Carlo simulation, also with $N=489$.

The calculated dependence of ρ_{Ω} on the inverse spin temperature, χ, is presented in Fig. 6. The numerical results are in a good agreement with the simple theoretical equation,

$$\langle \rho_{\Omega} \rangle \approx \sqrt{\langle L \rangle^2 + \langle \omega \rangle^2 / (N \langle \omega^2 \rangle)}. \tag{30}$$

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

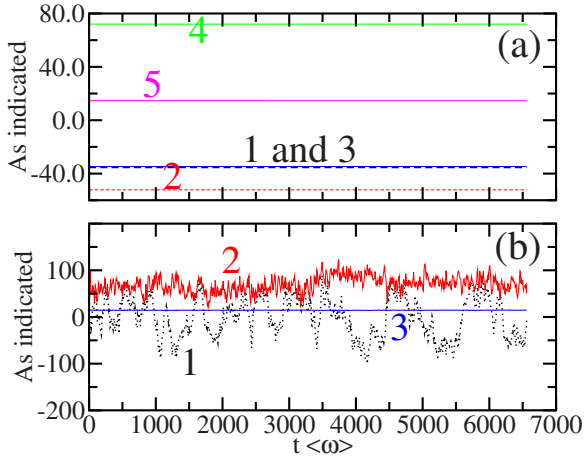

FIG. 7. (Color online) Conservation of the quantum dot's parameters. (a) With the dipole-dipole interaction switched off, the spin components $I_{\Sigma,x}$, $I_{\Sigma,y}$, and $I_{\Sigma,z}$, given by curves 1, 2, and 3, respectively, the spin modulus, I_{Σ}, given by curve 4, and the energy of the 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.

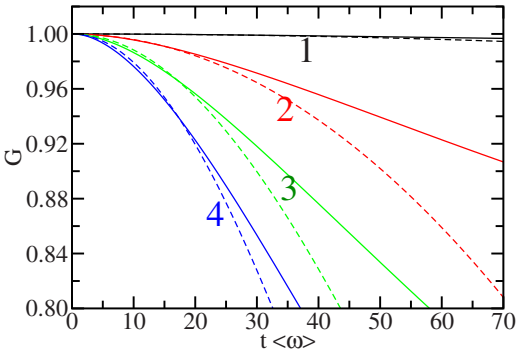

FIG. 8. (Color online) Spin correlator, G, vs time for different nuclear temperatures, as indicated, on a quantum dot with $N=1365$ nuclei. Solid lines are for the numerical simulation, whereas dashed lines are a Gauss approximation with fitting parameter T_{ξ} following Eq. (32). (1) $\beta\hbar\langle\omega\rangle\approx 3.5$, $\rho_{\Omega}=0.410$, $T_{\xi}\langle\omega\rangle=820\pm 15$, (2) $\beta\hbar\langle\omega\rangle\approx 0.18$ ($\rho_{\Omega}\approx 0.051$, $T_{\xi}\langle\omega\rangle=160\pm 15$, (3) $\beta\hbar\langle\omega\rangle\approx 0.078$, $\rho_{\Omega}=0.036$, $T_{\xi}\langle\omega\rangle=90\pm 15$, (4) $\beta\hbar\langle\omega\rangle\approx 0.05$ $\rho_{\Omega}=0.027$, $T_{\xi}\langle\omega\rangle=60\pm 15$.

$$\langle L(x)\rangle = 3\int_0^{\pi}\left[\frac{e^{x\sin^2(r)/r^2}+e^{-x\sin^2(r)/r^2}}{e^{x\sin^2(r)/r^2}-e^{-x\sin^2(r)/r^2}}-\frac{r^2}{x\sin^2(r)}\right]r^2dr, \tag{31}$$

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. (30) describes the nuclear-spin fluctuation, which is important in the limit of extremely high temperature, when $\chi\hbar\langle\omega\rangle\leq 1/\sqrt{N}$.

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, $\langle\omega\rangle\langle I\rangle\approx\langle\Omega\rangle\propto E$. 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.

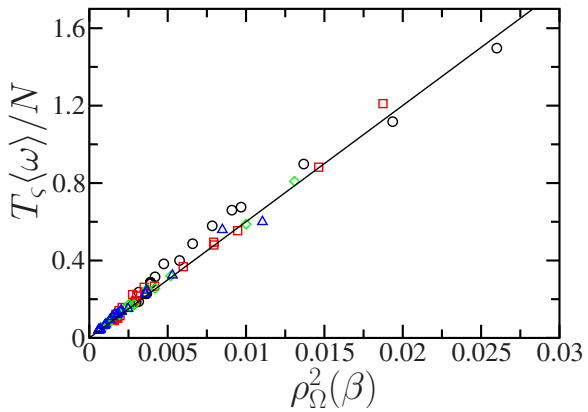

FIG. 9. (Color online) Dependence of nuclear-spin-relaxation time on nuclear-spin polarization. Circles are for $N=251$, squares for $N=485$, diamonds for $N=895$, and triangles for $N=1365$. All points coincide well with the straight line calculated from Eq. (28), with slope $T_2\gamma\approx 3$.

In Fig. 8 is the calculated correlator $G(t)$ [Eq. (28)] for various nuclear-spin temperatures as indicated. One can see that an increase in the nuclear polarization, from curve 4 with $\rho_{\Omega}=0.04$ to curve 1 with $\rho_{\Omega}=0.152$, decreases the spin-relaxation rate. On a short time scale, the time dependence of the spin correlator can be approximated by a Gaussian distribution:

$$G(t,\langle I\rangle,N)\approx\exp\left\{-\frac{t^2}{T_{\xi}^2(\beta,N)}\right\}\approx 1-\frac{t^2}{T_{\xi}^2(\beta,N)}. \tag{32}$$

This equation allows us to quantitatively compare the result of our numerical experiment with the estimation of Eq. (27). Figure 9 shows our numerically calculated value for $T_{\xi}(\beta,N)$ for four different spherical quantum dots, with N equal to 251, 485, 895, and 1365 nuclear spins. There $T_{\xi}(\beta,N)\langle\omega\rangle/N$ vs $\rho_{\Omega}^2(\beta)$ is presented; the polarization relaxation rate decreases fast with increasing nuclear polarization. $T_{\xi}(\beta,N)\langle\omega\rangle/N$ has approximately the same value for different N and calculated points for different system sizes are grouped around a straight line. As follows from Eq. (27), the slope of this line gives the characteristic time, which in this case is $T_2\langle\omega\rangle\approx 60$ or $T_2\gamma\approx 3$. These results are also in good agreement with Eq. (27); they demonstrate the universal character of the predicted dependence of the dipole-dipole relaxation time for the nuclear-spin potential ξ on $\rho_{\Omega}(\beta)$ and system size, N.

IV. SUMMARY

(1) For a quantum dot with a macroscopically large numbers of nuclei, quantum-mechanical and classical calculations of the nuclear dynamical polarization give the same result.

(2) 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 QDs 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. 8, that connect nuclear polarization and relaxation rates with electron-spin correlator.

(3) 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 QDs 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 four. One should take this effect into account when experi-

mentally describing the value of the nuclear polarization.

(4) 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 τ_c and reaches a maximum for $\Omega\tau_c \geq 1$. This maximum is about $\hbar\omega_n^2/\Omega$. It is inversely proportional to nuclear polarization. Usually the field is less than the Knight field, $\hbar\omega_n s_0$. The indirect field plays an important role in the problem of electron-spin dephasing.[22–24] It may also be important in the realization of dynamic nuclear self-polarization,[33] where electron polarization and Knight field are equal to zero.

(5) In the regime of long correlation time, the state of the quantum dot is characterized by three thermodynamic potentials: $\mathfrak{s}$, ξ, and χ. 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 ξ by the dipole-dipole interaction affects the transfer of angular momentum to the crystal lattice. The diffusion of energy to the QDs environment is the main mechanism for the relaxation of the inverse nuclear-spin temperature, χ. The relaxation of the electron-spin potential $\mathfrak{s}$ is connected with phonon scattering.

(6) Our numerical simulation showed the ENSSs 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 ξ 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. 31 and 32.

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APPENDIX A: CLASSICAL APPROXIMATION FOR THE TOTAL NUCLEAR SPIN I_Σ AND HYPERFINE FIELD

One can treat the electron-spin precession in the hyperfine nuclear field as a precession in a classical magnetic field. This is true when the electron-spin feedback influences the hyperfine nuclear field a negligibly small way, and the quantum uncertainty in the field direction and value is less than the classical one.

The total spin of the quantum dot nuclei has a macroscopically large value $I_\Sigma \geq \|I\|\sqrt{N}$, where N is the number of nuclei in the quantum dot. (For the model case $I=3/2$, $N=10^5$, and $I_\Sigma > 600$.) The quantum uncertainty of the hyperfine nuclear field direction is about $\sqrt{I_\Sigma(I_\Sigma+1)-I_\Sigma^2}/I_\Sigma = I_\Sigma^{-1/2} \leq 0.05$. For a small number of nuclei the quasi classical approach gives only a rough estimation and a numerical simulation of quantum-mechanical spin cluster dynamics must be used (see, for example, Ref. 34).

If the total nuclear-spin operator commutes with the Hamiltonian of the system then all three spin components keep their values (known or unknown) in time. The previous statement is not exactly true for the hyperfine interaction; $\boldsymbol{I}_\Sigma$ does not commute with Hamiltonian Eq. (1). But the operator corresponding to the total spin of the quantum dot ENSS $\hat{\boldsymbol{F}}=\hat{\boldsymbol{s}}+\hat{\boldsymbol{I}}_\Sigma$ commutes with this Hamiltonian, implying that all three components of $\boldsymbol{F}$ keep their values. The fluctuations of $\boldsymbol{I}_\Sigma$ around $\boldsymbol{F}$ are determined by electron-spin variation, which in turn cannot be larger than 1. Therefore, as a result of the hyperfine interaction, the direction of the total nuclear-spin fluctuates on a very small value of about $1/I_\Sigma \approx 0.002$.

If for the classical calculation one has to have well-defined total nuclear-spin direction, the precision of such a calculation is still limited by quantum mechanics and has to be less than 0.05. But if the classical calculation is made for a random distribution $\boldsymbol{I}_\Sigma$, and it is important that this spin keeps only its value and direction, the precision of the result is about 0.002.

Over a short time $\omega t \ll 1$, the spins of every nucleus also keep their directions, both in the classical and quantum cases. The total nuclear field (and $\boldsymbol{\Omega}$) keeps its value and direction over the time $\Omega^{-1} \leq t \ll \omega^{-1}$, with precision $\langle\omega\rangle/(2\Omega)$, because the total nuclear spin change cannot be larger than 1. Then the influence of the electron on the total hyperfine field is small, and the electron-spin precession in this field over a time $t\omega \ll 1$ can be described as a precession in a constant field.

For high nuclear-spin temperature the nuclear spins are weakly correlated, and the characteristic value of the nuclear field fluctuations is described by

$$\langle\Omega_{\mathrm{fluc},x}^2\rangle = \langle\Omega_{\mathrm{fluc},y}^2\rangle = \langle\Omega_{\mathrm{fluc},z}^2\rangle = \sqrt{\sum_n (\omega_n \|I_n^2\|)}/3, \quad \text{(A1)}$$

which is the same in a classical or a quantum description. The difference between these descriptions is in the definition of the spin modulus. In classical physics it is a phenomenological parameter of the nuclear-spin system. In quantum mechanics it can be determined from first principles $\|I_n^2\| = I_n(I_n+1)$, where I_n is again a phenomenological parameter with quantized value. The equality between averaged values of nuclear field fluctuation in all directions follows from symmetry considerations and does not depend on the description being classical or quantum. One can conclude that there are no differences between classical and quantum mechanics both in the description of electron-spin precession and in the description of the high-temperature distribution of nuclear field fluctuations. This should not be a surprise because for high temperatures the classical description of a large system usually gives precise results (Curie law for paramagnetic susceptibility, Drude model for semiconductors

conductivity, etc.). However, for a state with high nuclear polarization $|\langle \boldsymbol{I}_n\rangle| - I_n \ll 1$ or with strong correlation between nearest nuclear spins, the classical approach gives only an estimation and quantum mechanics or quantum field theory are indeed needed.

APPENDIX B: DERIVATION OF NUCLEAR-SPIN PRECESSION EQUATIONS

For calculation of integrals we introduce in Eq. (13) the dependence of electron spin from time (7). We suppose that for the initial state of the electron in the moment $t_0=\langle\{s_\alpha(t_0)s_\beta(t_0)\}\rangle_s=\delta_{\alpha,\beta}/4$, which is always true for quantum spin $\frac{1}{2}$. It is also true for a classical vector with $s^2=\frac{3}{4}$ in the case of low spin polarization (mean value of the vector and quadruple moment much less than one). In this limit

$$\frac{1}{\tau_c}\left\langle \int_0^\infty \left(\boldsymbol{s}(t)\cdot\int_0^t \boldsymbol{s}(\tau)d\tau\right)\exp\left\{-\frac{t}{\tau_c}\right\}d\tau_c\right\rangle_s = \frac{\tau_c}{4}\left(1+\frac{2}{1+(\Omega\tau_c)^2}\right), \tag{B1}$$

$$\frac{1}{\tau_c}\left\langle \int_0^\infty \left\{(\boldsymbol{s}(t)\cdot\boldsymbol{I}_n)\int_0^t \boldsymbol{s}(\tau)d\tau\right\}\exp\{-t\tau_c\}dt\right\rangle_s = \frac{\tau_c}{4}\left\{\left(\boldsymbol{I}_{n\|}+\frac{\boldsymbol{I}_{n\perp}2}{1+(\Omega\tau_c)^2}\right)-\frac{[\boldsymbol{\Omega}\times\boldsymbol{I}_n]\tau}{1+(\Omega\tau_c)^2}\right\}, \tag{B2}$$

and the mean value of the first correction to nuclear-spin precession in the mean Knight field is

$$\left\langle\frac{d\boldsymbol{I}_n}{dt}\right\rangle_s = -\frac{\omega_n^2\tau_c}{4}\left\{\frac{2\boldsymbol{I}_{n\|}+\boldsymbol{I}_{n\perp}(2+(\Omega\tau_c)^2)}{1+(\Omega\tau_c)^2}+\frac{[\boldsymbol{\Omega}\times\boldsymbol{I}_n]\tau_c}{1+(\Omega\tau_c)^2}\right\}. \tag{B3}$$

The first term in the right-hand side of Eq. (B3) contains the relaxation of the nuclear polarization components both parallel ($\boldsymbol{I}_{n\|}$) to and transverse ($\boldsymbol{I}_{n\perp}$) to $\boldsymbol{\Omega}$, see Eqs. (14) and (16). Its last term describes the nuclear-spin precession in the indirect hyperfine field, see Eq. (14). See Eqs. (14) and (16). Its last term describes the nuclear-spin precession in the indirect hyperfine field, see Eq. (15).

For intermediate value of the correlation time $(\Omega^{-1}<\tau_c\ll\langle\omega\rangle^{-1})$ the mean nuclear dynamical polarization rate has a strong dependence on the angle between $\boldsymbol{\Omega}$ and $\boldsymbol{s}_0$. In the steady-state regime without external magnetic field $\langle\boldsymbol{\Omega}\rangle$ is parallel to $\boldsymbol{s_0}$ and $\boldsymbol{s_0}=\bar{\boldsymbol{s}}(\langle\boldsymbol{\Omega}\rangle)$. We suppose that $\langle\Omega\rangle\gg\Omega_{\text{fluc}}$ but $\langle I\rangle^2\ll\|\boldsymbol{I}\|^2$. These two conditions can be realized at the same time for dots with large number of nuclei. In this case nuclei are polarized along $\langle\boldsymbol{\Omega}\rangle$ is parallel to $\boldsymbol{s_0}$ and $\boldsymbol{s_0}=\bar{s}(\langle\boldsymbol{\Omega}\rangle)$. Here we calculate only the nuclear polarization rate for this direction which we denote as z.

As a result of the electron-spin interaction with fluctuation of nuclei hyperfine field

$$\langle\hat{\Gamma}(t-t')[[\boldsymbol{\Omega}\times\boldsymbol{s}(t')]\boldsymbol{I}_n]\rangle_{s,I}=\boldsymbol{s}_0\omega_n\frac{2\|\boldsymbol{I}\|^2}{3}\cos(\Omega(t-t')). \tag{B4}$$

Here we utilized that $\langle\boldsymbol{s}(t_0)\rangle_s=\boldsymbol{s}_0$ and for low polarized nuclei spin system $\langle I_{n,\alpha}\cdot I_{m,\beta}=\delta_{n,m}\delta_{\alpha,\beta}\|\boldsymbol{I}\|^2/3$. After integration of Eq. (B4) on t' and averaging this integral at the t the final result for nuclei polarization rate is given by Eq. (18).

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[38] These quantum-mechanical equations for the square of the spins modulus enter in our classical calculations as phenomenological parameters. Fluctuations where the square is proportional to I^2 are considered statistical. Fluctuations where the square is proportional to I are usually considered quantum (Ref. 25).
[39] $\|\psi(R_n)\|^2$ and ω_n depend on the type of nucleus and on its position in the crystal cells and QD.
[40] Here we are talking not about the difference between longitudinal (T_1) and transversal (T_2) times of nuclear-spin relaxation related to nuclear-spin precession in an external magnetic field. We discuss the anisotropy of longitudinal time (T_1) related to electron-spin precession in the Overhauser field.
[41] This saturation value is determined by the fluctuation of the electron spin along $\mathbf{\Omega}$. The average values of nuclear spins, $\langle \boldsymbol{I} \rangle$, and frequency, $\langle \mathbf{\Omega} \rangle$, are parallel to each other, and the relaxation rate for the polarized nuclei, $I_{\|} \gg \|I\| / \sqrt{N}$, 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 two.
[42] A fluctuating magnetic polaron was investigated in the Raman spectroscopy of diluted magnetic semiconductors in Refs. 26 and 35–37.
[43] Usually for an unpolarized system of nuclei we have only an estimation of the characteristic time for this relaxation $T_2(0)$. For this calculation the short correlation time approximation is invalid. Here we are interested in the qualitative effect—the increase in the nuclear magnetodipole relaxation time with an increase in the total nuclear spin—(with nuclear polaron formation) $T_2(\beta)/T_2(0)$. For this reason we will do here only a classical simulation of the nuclear-spin system behavior, which is significantly easier than a quantum-mechanical simulation.
[44] As is well known from magnetism theory, the energy of magnetic anisotropy for a cubic spin lattice is proportional to $I^4_{\Sigma,x}+I^4_{\Sigma,y}+I^4_{\Sigma,z}+O(I^6_{\Sigma})$. It is important only for high enough nuclear polarization.