

Electron Spin Dephasing due to Hyperfine Interactions with a Nuclear Spin Bath

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We investigate pure dephasing decoherence (free induction decay and spin echo) of a spin qubit interacting with a nuclear spin bath. While for infinite magnetic field B the only decoherence mechanism is spectral diffusion due to dipolar flip-flops of nuclear spins, with decreasing B the hyperfine-mediated interactions between the nuclear spins become important. We give a theory of decoherence due to these interactions which takes advantage of their long-range nature. For a thermal uncorrelated bath we show that our theory is applicable down to $B \sim 10$ mT, allowing for comparison with recent experiments in GaAs quantum dots.

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Introduction.—How a localized electron in a solid loses its spin coherence is one of the oldest problems in condensed matter physics still attracting attention. It has taken on considerable recent significance due to the serious efforts in attempting to build a scalable quantum computer using electron spins in semiconductors as qubits [1]. It is therefore extremely important to understand all aspects of electron spin decoherence in solids qualitatively and quantitatively. The hyperfine (hf) interaction of the electron spin S with a bath of nuclear spins J_i (spin bath) is the main source of the decoherence of spin qubits at low temperatures, when relaxation effects due to phonons can be neglected. Many theoretical studies have been devoted to this problem [2–8], and the spin echo (SE) decoherence time T_{SE} has been recently measured in singlet-triplet [9] and single-spin qubits [10] based on gated GaAs quantum dots. The hf interaction alone can lead to spin decoherence, either through the direct electron-nuclear (e - n) spin flip [2] or through the effective (mediated by virtual e - n spin flips) intrabath interaction, which leads to fluctuations of the Overhauser field felt by the electron through the $S^z J_i^z$ part of the hf interaction [5–8]. These processes are suppressed at large magnetic fields B , and above a certain value of B one has to consider the decoherence due to the intrinsic bath dynamics caused by the dipolar interaction between the nuclear spins [3–6], the so-called spectral diffusion (SD).

Presently available theories assume the high magnetic field limit of $\Omega/\mathcal{A} \gg 1$, where \mathcal{A} is the total hf interaction energy (sum of the hf couplings with all the nuclei) and Ω is the electron spin splitting. This is fulfilled in SE experiments on electrons bound to the phosphorus donors in Si [11], where the SD theories have shown excellent agreement with observations [4,6,12]. However, in gated GaAs dots this limit corresponds to $B \gg 1$ T, while the experiments have been done at $B \approx 10$ – 100 mT and therefore the existing theories are of dubious validity in this case. The observed $T_{SE} \lesssim 1 \mu\text{s}$ in dots with $N \sim 10^6$ nuclei is at least an order of magnitude shorter than the

theoretical predictions for SD [4,5]. This shows that the SE decay in the low field regime in GaAs ($\Omega/\mathcal{A} < 1$) is *not* due to the spectral diffusion. The existing analytical theory addressing the SE decoherence due to hf interactions only [5] gives negligibly small decay at high B .

In this Letter we derive a theory of quantum dephasing due to the hf interaction, which applies at low magnetic fields. Although $\mathcal{A}/\Omega \ll 1$ is a sufficient condition for treating the e - n spin flip as a virtual process [13], one can expect that the necessary condition is weaker. In fact, it has been shown [2] that the smallness of the longitudinal spin decay is controlled by the parameter $\delta \equiv \mathcal{A}/\Omega\sqrt{N}$, and the condition of $\delta \ll 1$ is much less restrictive, especially for dots with many nuclear spins (i.e., large N). We argue here that this condition controls the convergence of our results at the relevant time scales of interest in GaAs spin qubits (as suggested in Ref. [5]), provided that the nuclear bath is thermal, uncorrelated, and unpolarized. Our theory can be used to calculate decoherence under any sequence of ideal π pulses driving the qubit (e.g., SE corresponds to $t/2$ - π - $t/2$ sequence), and we apply it to the case of free induction decay (FID) and SE. For FID we obtain results which at low fields differ qualitatively from the previously obtained ones [5,8]. For SE we identify the most relevant process contributing to its decay: the heteronuclear (i.e., involving nuclei of different species) flip-flop of a pair of nuclei. At high B this leads to a small oscillation superimposed on the SE signal due to the spectral diffusion, while at low B it leads to a decay of the SE in ~ 0.1 – $1 \mu\text{s}$ in gated GaAs dots. This result might explain the recent measurements of the SE decay on this time scale in GaAs [9,10].

Most of the successful theories of spin decoherence [4–6,14] involve an exponential resummation of the perturbation series for the decoherence time-evolution function $W(t) = |\rho_{+-}(t)|$ (where ρ_{+-} is the off-diagonal element of the electron spin reduced density matrix). We follow an approach along these lines. We take advantage of the essentially “infinite range” of the hf-mediated interactions

which couple effectively all the $N \sim 10^5$ – 10^6 nuclear spins in the bath (i.e., in the dot). We consider the ring diagrams, identified as the most important contributions for long-range interactions [6] in the perturbation expansion of $W(t)$. Corrections to these are suppressed as $1/N$. This approach is closely related to the $1/z$ (z being the coordination number) expansion of the partition function of the high-density Ising model [15], as well as recent calculations of the influence of the hf interactions on the electric dipole spin resonance in quantum dots [16]. Taking into account the ring diagrams only, we can sum all the terms in the cumulant expansion of $W(t)$.

Effective Hamiltonian.—The original Hamiltonian consists of Zeeman, hf, and dipolar interactions. The latter will be neglected below, since decoherence due to them can be calculated using well-tested methods [4]. The Zeeman energies are $\hat{H}_Z = \Omega S^z + \sum_i \omega_{\alpha[i]} J_i^z$ and hf interactions are given by $\hat{H}_{\text{hf}} = \sum_i A_i \mathbf{S} \cdot \mathbf{J}_i = \sum_i A_i S^z J_i^z + \hat{V}_{\text{sf}}$, where i labels the nuclear sites and $\alpha[i]$ labels the nuclear species at i th site (assigned randomly to sites in case of multiple isotopes, e.g., ^{69}Ga and ^{71}Ga). $\hat{V}_{\text{sf}} \sim S^\pm J^\mp$ is the spin-flip part of the hf interaction. The hf coupling A_i is proportional to its species' total hf energy $\mathcal{A}_{\alpha[i]}$ and the square of the envelope function $f_i = |\Psi(\mathbf{r}_i)|^2$: $A_i = \mathcal{A}_{\alpha[i]} f_i$ with normalization $\sum_i f_i = n_c$ (n_c being the number of nuclei in the unit cell, equal to 2 in GaAs). The number N of nuclei interacting appreciably with the electron is defined by $N \equiv$

$\sum_i f_i / \sum_i f_i^2$. For any α , $\sum_{i \in \alpha} A_i^2 = n_\alpha \mathcal{A}_\alpha^2 / N$, where n_α is the number of α nuclei per unit cell, and the maximal $A_{i \in \alpha} \approx \mathcal{A}_\alpha / N$. In calculations below we use a 2D Gaussian wave function (with the parameters for GaAs given in Fig. 1), but the results on time scales relevant for decay at low fields ($t \ll N / \mathcal{A}$) are determined only by N , not by detailed shape of $\Psi(\mathbf{r})$.

We derive the effective pure dephasing Hamiltonian \tilde{H} by removing the \hat{V}_{sf} interaction with a canonical transformation [5,8]. In the second order with respect to \hat{V}_{sf} we obtain the two-spin ($2s$) interaction considered in Refs. [5,8]: $\tilde{H}_{2s}^{(2)} = 2S^z \sum_{i \neq j} B_{i,j} J_i^+ J_j^-$ with $B_{i,j} = A_i A_j / 4 \Omega$. We will refer to this term as the S^z -conditioned term. These nonlocal hf-mediated flip-flop interactions are the most important at moderate fields and yield the effective Hamiltonian

$$\tilde{H}^{(2)} = \sum_i \omega_{\alpha[i]} J_i^z + \sum_i A_i S^z J_i^z + 2S^z \sum_{i \neq j} B_{i,j} J_i^+ J_j^- \quad (1)$$

The electron Zeeman term was discarded as being a constant of motion; the nuclear Zeeman terms, however, are important because $B_{i,j}$ can couple nuclei of differing species (this is crucial for SE). At higher orders we will have additional S^z -independent and multispin interactions which we address near the end of this Letter.

Decoherence function.—We write $\tilde{H} = \hat{H}_0 + 2S^z \hat{V}_1 + \hat{V}_2$, with the flip-flop interactions divided into S^z -conditioned \hat{V}_1 and S^z -independent \hat{V}_2 . We define the function $f(t; t')$ which encodes the sequence of n pulses applied to the qubit [17], $f(t; t') \equiv \sum_{k=1}^n (-1)^{n+k} \Theta(t' - t_k) \Theta(t_{k+1} - t')$, where t_k with $k = 1, \dots, n$ are the times at which the pulses are applied, and $t_0 = 0$, $t_{n+1} = t$, with t being the total evolution time. The decoherence function $W(t)$ can be written introducing the Keldysh contour [6,14,18]

$$W(t) = \left\langle \left\langle \mathcal{T}_C \exp \left[-i \int_C [c f(t; \tau) \mathcal{V}_1(\tau_c) + \mathcal{V}_2(\tau_c)] d\tau_c \right] \right\rangle \right\rangle, \quad (2)$$

where \mathcal{T}_C denotes the contour ordering of operators, $\langle \dots \rangle$ is the ensemble average, $\tau_c \equiv (\tau, c)$ is the time variable on the contour, with $c = +(-)$ on the upper (lower) branch of the contour, and $\mathcal{V}_{1,2}(\tau_c)$ are the interactions in which the spin operators are given by

$$J_j^\pm(\tau_c) = J_j^\pm e^{\pm i \omega_j \tau \pm i c \int_0^\tau f(t; t') (A_j/2) dt'} \equiv J_j^\pm d_j^\pm(\tau_c). \quad (3)$$

One remark is in order for the case of FID, in which the above formulas refer to the *single-spin* decoherence, defined in [5] where it was shown that in unpolarized thermal bath one can factor out the inhomogeneous broadening from $W(t)$ (this is equivalent to FID calculation using a “narrowed” state with no net nuclear polarization [8]).

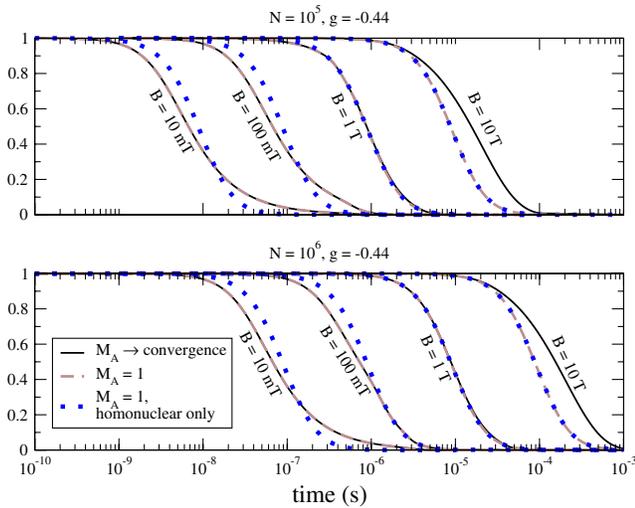


FIG. 1 (color online). $W(t)$ due to hf-mediated interactions for FID calculated for two GaAs dots of different sizes and various B . The solid lines are the results of the full \mathbf{T} -matrix calculations (with convergence achieved at matrix size $M \approx 100$), dashed lines are obtained with 3×3 \mathbf{T} matrix taking into account heteronuclear interactions, the dotted lines use Eq. (7). For GaAs we have used $n_\alpha = 0.604, 0.396, 1$, $\mathcal{A}_\alpha = 5.47, 6.99, 6.53 \times 10^{10} \text{ s}^{-1}$, and $\omega_\alpha/B = -6.42, -8.16, -4.58 \times 10^7 \text{ s}^{-1} \text{ T}^{-1}$ for nuclei of ^{69}Ga , ^{71}Ga , and ^{75}As , respectively. The electron spin splitting is $\Omega = 0.88 g_{\text{eff}} B [T] \times 10^{11} \text{ s}^{-1}$ with $g_{\text{eff}} = -0.44$ in a large quantum dot. At high B , the actual decay will be bounded by $\sim 10 \mu\text{s}$ due to spectral diffusion [5].

Ring diagrams.—We concentrate now on interactions from Eq. (1). Perturbation expansion of $W(t)$ is $W = 1 + \sum_{k=1} W^{(2k)}$, with $W^{(2k)} \sim \langle \mathcal{V}^{2k} \rangle$. We use here the thermal nuclear bath at temperature very high compared to the nuclear Zeeman energies, so that $\langle \cdots \rangle \sim \text{Tr}\{\cdots\}$, and inside the averages all the J_i^+ operators have to be paired up with J_i^- operators. Thus, $W^{(2k)}$ has the following structure:

$$W^{(2k)} \sim \sum_{i_1 \neq j_1} \cdots \sum_{i_{2k} \neq j_{2k}} \langle \mathcal{T}_C J_{i_1}^+ J_{j_1}^- \cdots J_{i_{2k}}^+ J_{j_{2k}}^- \rangle, \quad (4)$$

with at most $2k$ indices different. The full diagrammatic expansion is very cumbersome, since the commutation rule $[J_i^+, J_j^-] = 2\delta_{ij}J_i^z$ does not allow the standard form of Wick's theorem to work [6,14]. However, for “infinite” range interaction (all N spins coupled comparably with each other) we have $\sim N^{2k}$ terms in Eq. (4). From the sum over $2k$ indices we now take only the terms in which none of the indices is repeated. This amounts to introducing a $1/N$ error. The simplification is tremendous: now we only have one pair of J_i^+ and J_i^- for given i , and they effectively commute, as the averages containing J_i^z as the sole operator of the i th nuclei are zero in uncorrelated and unpolarized bath. We can then get rid of time-contour ordering, and classify all the terms in the perturbation series in terms of ring diagrams, which involve clusters of nuclei connected by two-spin interactions in a cyclical manner. Introducing the matrix T_{ij} we can write the ring diagram containing $2k$ nuclei as

$$R_{2k}(t) = \sum_{i_1 \neq i_2 \neq \cdots \neq i_{2k}} T_{i_1 i_2}(t) \cdots T_{i_{2k} i_1}(t) \simeq \text{Tr} \mathbf{T}(t)^{2k}, \quad (5)$$

where $T_{ij} \equiv (1 - \delta_{ij}) \sqrt{a_i a_j} B_{ij} \int_C c f(t; \tau) d_i^+(\tau_c) d_j^-(\tau_c) d\tau_c$, with $a_i \equiv \frac{2}{3} J_i (J_i + 1)$. With the same $1/N$ error, we may approximate all of the terms in the expansion of $W(t)$ as products of R_{2k} defined in Eq. (5), and $\ln W(t)$ is the sum of the linked terms [19], leading to

$$W(t) = \exp \left[\sum_{k=1}^{\infty} \frac{(-1)^k}{2k} R_{2k}(t) \right]. \quad (6)$$

Using Eqs. (5) and (6) we arrive at formula involving the eigenvalues λ_l of the \mathbf{T} matrix: $W(t) = \prod_l (1 + \lambda_l^2)^{-1/2}$. Since T_{ij} depends only on ω_i and A_i , we may approximate R_{2k} by considering continuous distribution $\rho(A)$ of A_i and then coarse graining it. With the relevant ranges of A_i divided into M_A slices and with N_J nuclear species, we have to deal with matrices of $M \times M$ size with $M = M_A N_J$. The calculation of $W(t)$ quickly converges as we increase M with the coarse-graining error negligible for $t \ll NM_A/\mathcal{A}$. In fact, we will show that $M = N_J$ (equal to 3 in the case of GaAs) gives a good approximation at experimentally relevant short times.

Free induction decay.—The main source of the FID decoherence is the interaction from Eq. (1) with only homonuclear flip-flops taken into account (as long as $\omega_{\alpha\beta} \equiv \omega_{\alpha} - \omega_{\beta} \gg \mathcal{A}/N$, which is fulfilled in GaAs

with $N \approx 10^6$ and $B \approx 0.1$ T). Then we can consider only ring diagrams involving only the nuclei of the same species, so that $T_{ij} \sim \sin(A_{ij}t/2)/A_{ij}$, with $A_{ij} = A_i - A_j$, and $R_{2k} = \sum_{\alpha} R_{2k}^{\alpha}$, so that $W_{\text{FID}}(t) = \prod_{\alpha} W_{\text{FID}}^{\alpha}(t)$. For short times, when $t \ll N/\mathcal{A}$ (i.e., $t \ll 20 \mu\text{s}$ for GaAs with $N = 10^6$), we obtain a very simple result: $R_{2k}^{\alpha} = (R_2^{\alpha})^k$ and $R_2^{\alpha} = \xi_{\alpha} t^2$, leading to the resummation of the exponentiated series in Eq. (6) giving us

$$W_{\text{FID}}^{\alpha}(t) = \frac{1}{\sqrt{1 + \xi_{\alpha} t^2}}, \quad \xi_{\alpha} = a_{\alpha}^2 \frac{n_{\alpha}^2 \mathcal{A}_{\alpha}^4}{4N^2 \Omega^2}, \quad (7)$$

so that the characteristic decay time is $T_{\text{FID}} \sim N\Omega/\mathcal{A}^2$ as in Ref. [5]. In Fig. 1 we show that this formula describes very well the FID decoherence in GaAs for $B < 1$ T. Let us also note that for InGaAs with $N = 10^5$, $g_{\text{eff}} = 0.5$, and at $B = 6$ T from Eq. (7) we obtain an estimate $T_{\text{FID}} \sim 0.6 \mu\text{s}$ which is close to the recently observed value of $3 \mu\text{s}$ in an optically selected “narrowed” state [20]. Equation (7) should be compared with the “pair approximation” result of Ref. [5], which in our notation reads $W^{\alpha} = \exp(-\frac{1}{2}R_2^{\alpha})$. This agrees with Eq. (7) for $R_2 \ll 1$, when both formulas give $W \approx 1 - \frac{1}{2}R_2$, but at times at which R_2 becomes comparable to 1 only Eq. (7) correctly takes into account the correlations of groups of spins larger than pairs. Interestingly, for long times ($t \gg N/\mathcal{A}$), the ring diagrams with more than two spins are suppressed by oscillations in T_{ij} , and the only remaining term is then $R_2 \sim t$ at this time scale. Thus we recover the long-time Markovian behavior discussed previously [5,8].

Spin echo.—If we use only the S^z -conditioned interactions and allow only for homonuclear flip-flops, there is no SE decoherence [5], i.e., $W_{\text{SE}}(t) = 1$. The SE decay can come then either from S^z -independent interactions or from considering the heteronuclear flip-flops in Eq. (1). The latter lead, for $t \ll N/\mathcal{A}$ and $\omega_{\alpha\beta} \gg \mathcal{A}/N$, to

$$T_{k \in \alpha, l \in \beta}^{\text{SE}} = (1 - \delta_{\alpha\beta}) \frac{A_k A_l}{4 \Omega} \sqrt{a_{\alpha} a_{\beta}} \frac{8i e^{i\omega_{\alpha\beta} t/2}}{\omega_{\alpha\beta}} \sin^2 \frac{\omega_{\alpha\beta} t}{4}. \quad (8)$$

The multiplicative dependence of the above matrix on A_k and A_l allows us to define a matrix $\tilde{T}_{\alpha\beta}$ of $N_J \times N_J$ dimension, in which A_k is replaced by $\sqrt{n_{\alpha}/N} \mathcal{A}_{\alpha[k]}$. Then we have $R_{2k}(t) = \sum_l \tilde{\lambda}_l^{2k}$ in terms of N_J eigenvalues of $\tilde{T}_{\alpha\beta}$. In GaAs we have $N_J = 3$, and we get $\tilde{\lambda}_i = 0, \pm \tilde{\lambda}$, so that $W_{\text{SE}}(t) = [1 + \frac{1}{2}R_2(t)]^{-1}$, where $R_2 = 2(|\tilde{T}_{12}|^2 + |\tilde{T}_{13}|^2 + |\tilde{T}_{23}|^2)$ and

$$|\tilde{T}_{\alpha\beta}|^2 = \frac{4 \mathcal{A}_{\alpha}^2 \mathcal{A}_{\beta}^2}{N^2 \Omega^2 \omega_{\alpha\beta}^2} n_{\alpha} n_{\beta} a_{\alpha} a_{\beta} \sin^4 \frac{\omega_{\alpha\beta} t}{4}. \quad (9)$$

The SE decoherence is shown in Fig. 2. At high B when $|\tilde{T}_{\alpha\beta}|^2 \ll 1$ we obtain a small oscillation which in experiments will be superimposed on the SD decay. At low B , which for $N = 10^5(10^6)$ in GaAs corresponds to $\Omega < \Omega_c \sim \sqrt{r} \mathcal{A}/\sqrt{N} \approx 250(75)$ mT (where $r \sim 10^3$ is the ra-

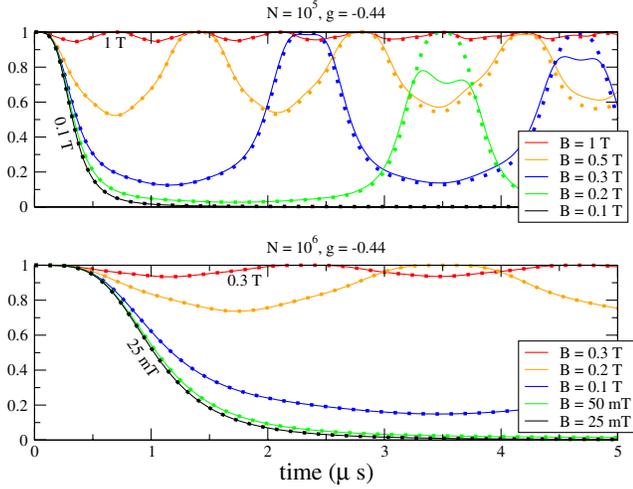


FIG. 2 (color online). Spin echo decoherence $W^{\text{SE}}(t)$ in GaAs due to heteronuclear processes. The dots are obtained in the $\mathcal{A}/N \ll \omega_{\alpha\beta}$, $1/t$ limit, when $W(t) = [1 + \frac{1}{2}R_2(t)]^{-1}$, while the solid lines are the results of the calculation with the full \mathbf{T} matrix. The differences between the two approaches on a microsecond time scale are visible for the smaller dot (upper panel), but are negligible for the larger one (lower panel). The decay time due to spectral diffusion (calculated as in Ref. [4]) is $>10 \mu\text{s}$ in both cases. The GaAs parameters used are the same as in Fig. 1.

ratio of electron g factor to the typical difference of nuclear g factors), the coherence decays to zero in characteristic time $T_{\text{SE}} \approx r/\Omega_c \approx 0.25(0.75) \mu\text{s}$. This is close to $T_{\text{SE}} \approx 0.3\text{--}0.4 \mu\text{s}$ observed in [10] at $B \approx 50\text{--}70$ mT. The B dependence of decay is visible in a narrow range of fields when the decay is substantial but not complete.

S^z -independent two-spin term.—The contribution of $\tilde{H}_{2s}^{(3)} \sim \sum_{i,j} C_{ij} J_i^+ J_j^-$ with $C_{ij} \propto A_i^2 A_j / \Omega^2$ to FID and SE is calculated in the analogous way, and it turns out to be negligible for $B \geq 10$ mT and $N \geq 10^5$.

Influence of multispin interactions.—The most important multispin terms are the ones involving n nuclei in the n th order in expansion of \tilde{H} , e.g., $\tilde{H}_{3s}^{(3)} \sim S^z \sum_{i,j,k} C_{ijk} J_i^+ J_j^- J_k^z$ with $C_{ijk} \propto (\mathcal{A}/N)^3 / \Omega^2$. Applying the operator norm $\|\hat{A}\| \equiv \max_{\langle\Phi|\Phi\rangle=1} |\langle\Phi|\hat{A}|\Phi\rangle|$ to these terms, one gets $\|\tilde{H}^{(n)}\| \approx \mathcal{A} (\mathcal{A}/\Omega)^{n-1}$, so that it may seem that the expansion is convergent only when $\mathcal{A}/\Omega < 1$. However, the state $|\Phi\rangle$ maximizing the norm is a highly entangled state of nuclear spins. Here we are interested in the evolution of the nuclear spin system averaged over an uncorrelated thermal ensemble, and therefore such massively entangled nuclear states do not appear in the calculations.

We calculate the lowest order term in the expansion of $W_{3s}(t) \approx 1 + W_{3s}^{(2)} + \dots$ in powers of three-spin (3s) interaction. For both FID and SE we get that for $t \ll N/\mathcal{A}$ [when $W(t)$ decays practically to zero due to the two-spin

interactions] we have $W_{3s}^{(2)}/R_2 \sim \delta^2$. Influence of each higher-order multispin term in \tilde{H} comes with a higher power of the small parameter δ , and our neglect of these terms is a well-controlled approximation at the time scale of interest. We therefore believe that the approach used in this work remains valid at relevant times down to $B \approx 10$ mT for GaAs dots with $N \approx 10^5\text{--}10^6$ nuclei.

Conclusions.—We have presented a theory of quantum decoherence due to hf interaction of a central spin with a thermal nuclear bath. For both FID and SE we obtain analytical formulas for nonexponential time dependence of decoherence. For SE we obtain a good agreement with the measurements in gated GaAs dots, and predict the appearance of characteristic oscillations in the signal at magnetic fields slightly higher than the ones used in experiments.

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