Spectrum of an Electron Spin Coupled to an Unpolarized Bath of Nuclear Spins

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The main source of decoherence for an electron spin confined to a quantum dot is the hyperfine interaction with nuclear spins. To analyze this process theoretically we diagonalize the central spin Hamiltonian in the high magnetic B-field limit. Then we project the eigenstates onto an unpolarized state of the nuclear bath and find that the resulting density of states has Gaussian tails. The level spacing of the nuclear sublevels is exponentially small in the middle of each of the two electron Zeeman levels but increases superexponentially away from the center. This suggests to select states from the wings of the distribution when the system is projected on a single eigenstate by a measurement to reduce the noise of the nuclear spin bath. This theory is valid when the external magnetic field is larger than a typical Overhauser field at high nuclear spin temperature.

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Spin dynamics in semiconductor nanostructures has recently become a topic of great interest due to the possibility of using the spin degree of freedom instead of charge in electronic circuits [1] and equally important due to the proposal of using electron spin in a semiconductor quantum dot as a fundamental building block of the quantum computing device [2]. GaAs quantum dots are the main candidates in practical realizations of these proposals due to the well developed manufacturing technology. However, unavoidable inhomogeneous hyperfine interaction of electron spin with many nuclear spins of the host crystal acts as a noisy environment that is the main source of dephasing for the electron spin at low temperature when relaxation due to the phonons is ineffective.

The limit of fully polarized nuclear spin bath was analyzed exactly in [3], including spectral properties. However, it is rather hard to achieve a significant polarization dynamically, and thermodynamic polarization, requiring submilli Kelvin temperatures [4], is still out of reach for semiconductors. Currently, a more promising route is to actively reduce the distribution width of the nuclear Overhauser field by projective measurements [5–7]. This has been partially achieved in experiments leading to significantly longer decoherence times [8–10]. To further optimize projective measurement techniques it is essential to gain a better understanding of the spectral properties of the unpolarized system which, so far, have only been understood qualitatively.

In this paper we diagonalize the central spin Hamiltonian for a quantum dot in the high magnetic B-field limit using a $1/B$ expansion. Projecting the eigenstates on an unpolarized state of the nuclear spin bath we find that their density has Gaussian tails. Correspondingly the level spacing of the nuclear spin sublevels, which is exponentially small with the radius of the quantum dot in the middle of the two electron Zeeman levels, becomes superexponentially large with detuning away from the center; see Fig. 1. This suggests using a finite detuning from the bare electron Zeeman energy when one eliminates the effect of the nuclei by the projective measurement technique [5–10].

Our theory is applicable when the external magnetic field B is larger than a typical Overhauser field at high nuclear spin temperature due to fluctuations $B_{\mathrm{fluc}} = A\sqrt{S/\tilde{N}}/\mu$, where A/μ is the maximum Overhauser field, $\tilde{N}$ is the number of nuclei under the electron envelope wave function, and S is a number of degenerate hyperfine couplings. At low field $B < B_{\mathrm{fluc}}$, the spectrum can be obtained by a numerical solution of the Richardson equations [11] where the $1/B$ expansion of the present Letter can be used as a benchmark for complex numerical procedures.

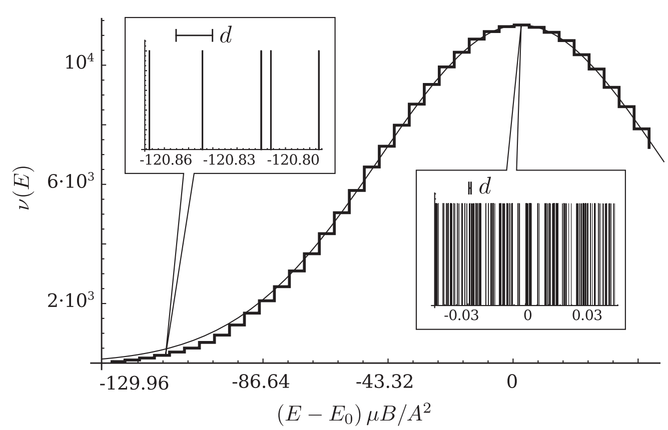

FIG. 1. Numerical evaluation of $\nu(E)$ using Eq. (2) on a course scale—thick line and Eq. (5)—thin line ($S_j = 1$, $r_0 = 8$, $N = 18$, a fixed external B field), A is a maximum Overhauser field, E_0 is a shift from Eq. (5). Insets show $\nu(E)$ on a fine scale in the middle of the upper electron Zeeman line and at a finite detuning, the average level spacing d was evaluated using Eq. (6).

The spin of an electron in a quantum dot couples to nuclear spins in the presence of an external B field as

$$H = \mu B S_0^z + \sum_{j=1}^{N} A_j \mathbf{S}_0 \cdot \mathbf{S}_j, \tag{1}$$

where $\mu = g\mu_B$ is the electron magneton (in the following we neglect the nuclear Zeeman splitting), S_0^z, $S_0^{\pm} = S_0^x \pm iS_0^y$ are electron spin-1/2 operators and $\mathbf{S}_j$ ($j \geq 1$) are spin operators of nuclear shell j with the maximum angular momentum $S_j \geq 1/2$ constructed out of $2S_j$ nuclei of spin-1/2 which have the same hyperfine coupling to the electron spin, $\mathbf{S}_j = \sum_{i=1}^{2S_j} \mathbf{I}_{ji}$, where i labels individual nuclei within the shell, $\mathbf{I}_{ji}$ are nuclear spin-1/2 operators, and N is the number of nuclear shells. Assuming harmonic confinement of the electron in all spatial directions the couplings are $A_j = A_0 \exp(-r_j^2/r_0^2)$, where A_0 is the coupling in the middle of the quantum dot, and r_0 and r_j are spatial size of the quantum dot and radius of jth shell in units of the lattice parameter.

In 1D only two nuclei have the same coupling ignoring the isotope effects and assuming equidistant lattice sites $r_j = j$; thus, the maximum total angular momentum is $S_j = S = 1$. In 2D degeneracy of the couplings gives $S_j = S = 4$ but the radii of the sequential shells are not equidistant because the number of nuclei grows linearly away from the center. We thus model the system as a set of concentric nuclear shells, $r_j = r + 4m/(\pi r)$ and also change the summation indices in Eq. (1), $\sum_{j=1}^{N} \rightarrow \sum_{r=1,m=1}^{N,\pi r/4}$ [12]. In 3D the degeneracy is larger than in 2D, $S_j = S = 12$, and the number of the nuclei grows quadratically away from the center, $r_j = r + 6m/(\pi r^2)$, $\sum_{j=1}^{N} \rightarrow \sum_{r=1,m=1}^{N,\pi r^2/6}$.

This model conserves the number of excitations $[H, J_z] = 0$, where $J_z = \sum_{j=0}^{N} S_j^z$, and the total angular momentum of each nuclear shell $[H, \mathbf{S}_j^2] = 0$. All of them also commute with each other, $[J_z, \mathbf{S}_j^2] = 0$ and $[\mathbf{S}_i^2, \mathbf{S}_j^2] = 0$. Thus the Hilbert space is partitioned into a set of disconnected subspaces labeled by the following quantum numbers: n is an eigenvalue of J_z and l_j [13] correspond to $\mathbf{S}_j^2$, $\mathbf{S}_j^2|\Psi\rangle = l_j(j_j + 1)|\Psi\rangle$. The latter becomes trivial when all of the nuclear spins have different couplings as for spin-1/2 operators $\mathbf{S}_j^2 = 3/4$ is a number but is nontrivial when $S_j > 1/2$.

The diagonalization in each subspace can be performed using degenerate perturbation theory when the B field is large. Splitting the Hamiltonian into the unperturbed part $H_0 = \mu B S_0^z$ and a perturbation $V = \sum_j A_j \mathbf{S}_0 \cdot \mathbf{S}_j$ defines two electron Zeeman levels, $E = \pm\mu B/2$ but leaves the nuclear spin sublevels hugely degenerate in the zeroth-order approximation. The latter degeneracy has to be lifted via a diagonalization of the perturbation V.

In the basis of eigenstates of J_z, $|\Psi\rangle = |\pm, \{l_j, k_j\}\rangle$, $\langle\Psi|\Psi\rangle = 1$, V is a diagonal matrix within both of the electron spin subspaces where the spin-flip part of V that couples opposite electron levels can be neglected when the external field is very large [12]. Here $\pm$ refers to the "up" and "down" electron Zeeman levels and k_j are the numbers of nuclear spin excitations on each shell such that the quantum number $n = (1 \pm 1)/2 + \sum_{j=1}^{N} k_j$. The second order correction to the eigenenergies are due to the spin-flip part of V. Using the matrix elements of V in the basis of eigenstates of J_z we obtain

$$E = \pm\frac{\mu B}{2} \pm \sum_{j=1}^{N}\left[\frac{A_j(-l_j + k_j)}{2} + \frac{A_j^2(2l_j - k_j + \frac{1\mp1}{2})(k_j + \frac{1\pm1}{2})}{4\mu B}\right], \tag{2}$$

where the energy denominator in the last term was also expanded up to the leading order in $1/\mu B$. Including the first order corrections to the eigenfunctions we get

$$|\Psi\rangle = |\pm, \{l_j, k_j\}\rangle \pm \sum_{m=1}^{N} \frac{A_m}{\mu B} S_m^{\pm} |\mp, \{l_j, k_j\}\rangle. \tag{3}$$

The large magnetic field expansion has different conditions of applicability for the eigenenergies Eq. (2) and the eigenstates Eq. (3) in the subspaces of unpolarized nuclear spins $k_j \approx l_j$. The subleading terms in Eq. (2) are small in all subspaces when $B \gg B_{\rm fluc}$ where $B_{\rm fluc} = \sqrt{\sum_{j=1}^{N} A_j^2 S_j^2}/\mu$. But the next (second) subleading correction to Eq. (3) is small only when $B \gg B_{\rm max}$ where $B_{\rm max} = r_0^2 A_0/2\mu$ in 1D and 2D ($B_{\rm max} = r_0^3 A_0/\sqrt{8e}\mu$ in 3D) [12] is a much larger field than $B_{\rm fluc}$. The latter signals that the choice of the eigenfunctions, $|\Psi\rangle = |\pm, \{l_j, k_j\}\rangle$, is a poor zeroth-order approximation in the intermediate field regime, $B_{\rm fluc} \ll B \ll B_{\rm max}$. The correct approximation can be identified by merging the inner nuclear shells with different couplings up to the radius $\tilde{r} = r_0\sqrt{\ln(B_{\rm max}/B)}$ (in units of the lattice parameter) in 1D and 2D [$\tilde{r} = r_0(1 + \sqrt{\ln(B_{\rm max}/B)})/\sqrt{2}$ in 3D] [12] into a single shell with the same coupling A_0. Then, diagonalizing $H + V'$, where $V' = \sum_{j:r_j\leq\tilde{r}}(A_1 - A_j)\mathbf{S}_0 \cdot \mathbf{S}_j$, when $B_{\rm fluc} \ll B \ll B_{\rm max}$ instead of the original model H we obtain the same result as in Eqs. (2) and (3) but a different definition of nuclear shells $\tilde{S}_j$, where the first element is $\tilde{S}_1 = \sum_{j:r_j\leq\tilde{r}} S_j$, the middle elements are $\tilde{S}_j = 0$ for $1 < r_j \leq \tilde{r}$, and the outer elements, $r_j > \tilde{r}$, are $\tilde{S}_j = S_j$.

In two and three dimensions the parameter $B_{\rm max}$ is proportional to the measurable maximum Overhauser field $A = \sum_{j=1}^{N} S_j A_j$, A/μ is of the order of a few Tesla [14], with the numerical factor π^{-1} and $(2\pi^3 e)^{-1}$. In 1D, $B_{\rm max} = \tilde{N}A/(\pi\mu)$ is much larger than A, here

$\tilde{N} = \sum_{j=1}^{N} 2S_j A_j / A_0$. The parameter $B_{\rm fluc} = A\sqrt{S/\tilde{N}}/\mu$ scales with the number of nuclei under the electron envelop function in all dimensions.

In terms of density of states the bare electron level acquires a finite smearing due to coupling to many degrees of freedom of unpolarized nuclear spins. When the quantum dot is empty the nuclei at different lattice sites are uncorrelated. After an electron, say with spin up, populates the quantum dot, the state of the combined system $|\Psi_0\rangle = S_0^+ \prod_{\{j,i\}} I_{ji}^+ |\Downarrow\rangle$ is not an eigenstate of the Hamiltonian Eq. (1), where $\{j, i\}$ labels a subset of nuclear lattice sites and $|\Downarrow\rangle$ is the all spins down (including the central spin) state. We analyze the distribution of the eigenenergies Eq. (2) using a projected density of states $\nu(E) = \sum_{\{l_j,k_j\}} P(\{l_j, k_j\}) \delta(E - E(\{l_j, k_j\}))$, where $P(\{l_j, k_j\}) = 1$ when $\langle \Psi_0 | \{l_j, k_j\}\rangle \neq 0$ and $P(\{l_j, k_j\}) = 0$ when $\langle \Psi_0 | \{l_j, k_j\}\rangle = 0$. Here the $\sum_{\{l_j,k_j\}}$ runs over all subspaces and all eigenstates within each subspace. Note that for any shell with $S_j > 1$ the complete set of the eigenstates includes l_j with multiplicities greater than one [13]. Only one of each l_j is kept since these multiplicities do not change $P(\{l_j, k_j\})$. We calculate the overlaps matrix elements only in the leading $1/\mu B$ order as the probability of measuring other eigenstates coming from subleading orders is at least as small as $A_j/\mu B$.

By representing the delta function as $\delta(x) = \int d\lambda e^{\imath x\lambda}/(2\pi)$, the Fourier transform of $\nu(E)$ can be written as a product of sums over each nuclear spin shell

$$\nu(\lambda) = \sum_{\{l_j,k_j\}} P(\{l_j, k_j\}) e^{-i\lambda E(l_j,k_j)} = \prod_{j=1}^{N} e^{-(i\lambda(p_j A_j - \mu B)/2)} \times \sum_{k=p_j(1+{\rm sgn} p_j)}^{p_j+\tilde{S}_j} e^{-(i\lambda A_j^2 (k-2p_j)(k+1)/4\mu B)}, \quad (4)$$

where $p_j = \langle \Psi_0 | S_j^z | \Psi_0 \rangle$, $|p_j| \leq l_j$, are polarizations of the shells given by the state of the system $|\Psi_0\rangle$.

Assuming that each shell is unpolarized $p_j \ll \tilde{S}_j$ and $\tilde{S}_j \gg 1$, the sum within a shell can be calculated as an integral, $I_j(\lambda) = \int_0^{\tilde{S}_j} dk e^{-ixk(k+1)} = \sqrt{\pi} e^{ix/4} [{\rm erf}((1 + 2\tilde{S}_j)\sqrt{ix}/2) - {\rm erf}(\sqrt{ix}/2)]/(2\sqrt{ix})$, $x = \lambda A_j^2/(4\mu B)$, which is an oscillating function of λ. Then the product of the oscillating functions can be approximated in the large-N limit by turning it into an exponential of a sum of logarithms, $\prod_{j=1}^{N} I_j(\lambda) = I_1(\lambda) \exp(\sum_{j:r_j>\tilde{r}}^{N} \log I_j(\lambda))$, and by expanding the exponent in λ, $\sum_{j:r_j>\tilde{r}}^{N} \log I_j(\lambda) \approx \sum_{j:r_j>\tilde{r}}^{N} [\log S_j - i(S_j/2 + S_j^2/3)\lambda A_j^2/(4\mu B) - (S_j^2/24 + S_j^3/12 + 2S_j^4/45)\lambda^2 A_j^4/(16\mu^2 B^2)]$.

In one dimension $I_j(\lambda)$ cannot be calculated as an integral since the degeneracy of the hyperfine couplings is two but the explicit evaluation of the sum of only two terms within each shell and the small-λ expansion yields a similar expression, $\sum_{j:r_j>\tilde{r}}^{N} \log I_j(\lambda) \approx \sum_{j:r_j>\tilde{r}}^{N} [\log 2 - i\lambda A_j^2/(4\mu B) - \lambda^2 A_j^4/(\sqrt{2}4\mu B)^2]$. Strictly speaking, the small-λ expansion is good when $\lambda \ll 16\mu B/A_{\tilde{r}}^2$ but the resulting Gaussian is also quite a good approximation for a large λ since the original product of many oscillating functions is zero due to random phases of $I_j(\lambda)$ when $\lambda \geq 4\mu B/A_{\tilde{r}}^2$, provided that the couplings A_j have a non regular distribution.

By evaluating the inverse Fourier transform $\nu(E) = \int d\lambda \nu(\lambda) \exp(-iE\lambda)$ in the limit $B \gg B_{\rm max}$ we obtain

$$\nu(E) = \frac{\tilde{S}_1 \prod_{j:r_j>\tilde{r}}^{N} S_j}{\sqrt{\pi}\sigma} \exp\left[-\frac{(E - E_0)^2}{\sigma^2}\right], \quad (5)$$

where $E_0 = \sum_{j=1}^{N} p_j A_j/2 - \mu B/2$ is a shift of the bare electron level that depends on the momentary state of the nuclei and a finite linewidth $\sigma = \sqrt{\sum_{j:r_j>\tilde{r}}^{N} (S_j^2/96 + S_j^3/48 + S_j^4/90) A_j^4/(\mu B)}$ that is common for all unpolarized nuclear states. In the intermediate regime $B_{\rm fluc} \ll B \ll B_{\rm max}$ Eq. (5) is valid when $E \geq \tilde{S}_1^2 A_1^2/(4\mu B)$. The contribution of the inner shells can be approximated as $I_1(\lambda) = \tilde{S}_1$ when, due to the fast oscillating exponential, the main contribution to the inverse Fourier transform comes from $\lambda \leq 4\mu B/(\tilde{S}_1 A_1)^2$.

In 1D, the Gaussian result agrees precisely with the spectroscopically measurable line shape when $B \gg B_{\rm max}$. As the degeneracy of hyperfine couplings is 2 for all shells, all projections [13] are the overlap of the singlet (or triplet) and two nuclear spin states which give $1/\sqrt{2}$ and the calculation of the line shape gives Eq. (5). When the degeneracy is larger than 2 the two calculations are different. It is also worth noting that the state $|\Psi(0)\rangle$ is an eigenstate of the model Eq. (1) with $S_j = 1/2$ in the high B-field $B \gg B_{\rm max}$.

Rediscretization of Eq. (5) recovers the average level spacing of the nuclear spin levels. From the definition of the density of states, $d = 1/\nu(E)$ is an energy range that contains only one state. But, as the prefactor in $\nu(E)$ increases to infinity when more and more outer shells are taken into account, the level spacing becomes zero. On the other hand the coupling strengths of the outer shells become superexponentially small which make the splitting of the inner shells' levels into sublevels due to the outer shells very narrow. Thus, by selecting an effective number of the significantly coupled nuclear shells $r_j < 4r_0$, we find

$$d(E) = d(E_0) \exp[(E - E_0)^2/\sigma^2], \quad (6)$$

where $d(E_0) = \sqrt{\pi}\sigma/(\tilde{S}_1 \prod_{j:\tilde{r}<r_j<4r_0} S_j)$ is exponentially small, $d(E_0) \simeq \tilde{S}_1 \exp(-\tilde{N}/S)$. Thereby, $d(E_0)$ is a tiny level spacing in the middle of the upper electron Zeeman line but $d(E)$ increases superexponentially at a finite detuning $E \neq E_0$ on a characteristic energy scale σ when $B \gg B_{\max}$ and $\tilde{S}_1^2 A_1^2/(4\mu B)$ when $B_{\text{fluc}} \ll B \ll B_{\max}$.

There is also a finite temperature smearing. To average the hyperfine shift E_0 over all possible nuclear spin configurations at a high temperature, $\nu_0(E) = \sum_{\{p_j\}} \delta(E - E_0)$, we use the same approach as in the calculation of $\nu(E)$ and obtain the Gaussian distribution of levels with a width $\sigma_0 = \sqrt{\sum_{j=1}^N S_j^2 A_j^2/6}$ and an average level spacing $d_0 = \exp[(E - \mu B/2)^2/\sigma_0^2]\tilde{S}_1 \prod_{j:\tilde{r}<r_j<4r_0} 2S_j/(\sqrt{\pi}\sigma_0)$. This implies that if the nuclear spin state is not prepared in a specific way but is a thermal state, there are two energy scales in a projective measurement to narrow the nuclear spin bath [8–10] in order to suppress fluctuations of the Overhauser field [5–7]. A measurement in the coarse resolution of d_0 will select a single specific nuclear spin configuration suppressing only thermal fluctuations and a measurement in the fine resolution of d will project the system on an eigenstate within a given nuclear bath state.

Using the eigenstates and the spectrum in Eqs. (2) and (3) one can evaluate the time-dependent density matrix of the electron with an unpolarized state of the nuclei, $|\Psi(0)\rangle = (1 + S_0^+)\prod_{\{j,i\}} I_{ji}^+ |\Downarrow\rangle/\sqrt{2}$ such that $\langle\Psi(0)|J_z|\Psi(0)\rangle = 0$, as an initial condition. As a result the diagonal matrix elements do not decay in time in the leading $1/\mu B$ order, $T_1 = \infty$. When the degeneracy of the hyperfine couplings is only 2 (1D case and $B \gg B_{\max}$) the off-diagonal matrix elements have a slow Gaussian envelop with decay time $T_2 = 1/\sigma$ on top of the fast electron spin Rabi oscillations with frequency μB. Note that one obtains the Gaussian decay assuming a phenomenological model of a quasistatic ensemble of nuclear magnetic fields [6]. At a high temperature, averaging over different $|\Psi(0)\rangle$, one also obtains the Gaussian decay due to thermal fluctuations with $T_2 = 1/\sigma_0$ [15] which is much faster than $1/\sigma$.

When the degeneracy of the hyperfine couplings is larger than 2 (2D and 3D cases and $B_{\text{fluc}} \ll B \ll B_{\max}$ in 1D) we establish a bound on the shortest decay time assuming that all Clebsch-Gordan coefficients in the overlaps between the initial state $|\Psi(0)\rangle$ and the eigenstates Eq. (3) are equal and neglecting degeneracies of l_j [13]. This simplification gives a Gaussian decay with decay time $T_2 = 1/\sigma$. A more accurate calculation would give a spectroscopic line shape, see discussion after Eq. (5), which is narrower than the distribution of the eigenenergies thus giving a longer decay time.

The eigenenergies Eq. (2) are a good benchmark for numerical studies of Richardson equations [16]. The spectrum of the model Eq. (1) can be found at arbitrary field and for any quantum number n by solving a set of coupled nonlinear equations [11],

$$\sum_{j=1}^{N} \frac{2l_j A_j/2}{E_\nu + A_j/2} + 1 - \frac{\mu B}{E_\nu} + \sum_{k=1\neq\nu}^{n} \frac{2E_k}{E_\nu - E_k} = 0, \qquad (7)$$

as $E = \sum_{\nu=1}^n E_\nu + \sum_{j=1}^N l_j A_j/2 - \mu B/2$. At an infinitely large magnetic field solutions of these equations are sets of numbers E_ν which are close either to $-A_j/2$ or μB. At a finite magnetic field a $1/B$ expansion of the Eqs. (7) at these values of E_ν recovers the $1/B$ expansion in Eq. (2) and a $1/B$ expansion of the Gaudin states [11] recovers Eq. (3).

In conclusion we have diagonalized the central spin Hamiltonian in the high B-field limit. Projecting the eigenstates on an unpolarized state of the nuclear bath we have shown that the level spacing of the nuclear sublevels, which is exponentially small in the middle of the bare electron level, becomes superexponentially large with detuning away from the middle. This suggests to select states from the wings of the distribution when one attempts to eliminate the decohering effect of the nuclei by projective measurement techniques. This theory is valid when the external B field is larger than typical Overhauser fields.

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