Exponential decay in a spin bath

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We show that the coherence of an electron spin interacting with a bath of nuclear spins can exhibit a well-defined purely exponential decay for special ("narrowed") bath initial conditions in the presence of a strong applied magnetic field. This is in contrast to the typical case, where spin-bath dynamics have been investigated in the non-Markovian limit, giving superexponential or power-law decay of correlation functions. We calculate the relevant decoherence time T_2 explicitly for free-induction decay and find a simple expression with dependence on bath polarization, magnetic field, shape of the electron wave function, dimensionality, total nuclear spin I, and isotopic concentration for experimentally relevant heteronuclear-spin systems.

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I. INTRODUCTION

There are many proposals to use the spin states of electrons in confined structures for coherent manipulation, leading to applications in quantum information processing and, ultimately, quantum computation.[1–5] A series of recent experiments on such spin states in quantum dots,[6,7] electrons bound to phosphorus donors in silicon,[8] nitrogen vacancy centers in diamond,[4,9,10] and molecular magnets[11,12] have shown that the hyperfine interaction between confined electron spins and nuclear spins in the surrounding material is the major obstacle to maintaining coherence in these systems.

Previous studies of this decoherence mechanism have pointed to the non-Markovian nature of a slow nuclear-spin environment, leading to nonexponential coherence decay.[13–25] These results suggest that it may be necessary to revise quantum error correction protocols to accommodate such a "nonstandard," but ubiquitous environment.[26] In the present work, we show that virtual flip-flops between electron and nuclear spins can lead to a well-defined Markovian dynamics, giving simple exponential decay in a large Zeeman field and for particular initial conditions (a "narrowed"[27] nuclear-spin state). Moreover, we calculate the decoherence time T_2, revealing the dependence on many external parameters for a general system.

The rest of this paper is organized as follows: In Sec. II, we introduce the Hamiltonian for the Fermi contact hyperfine interaction and derive an effective Hamiltonian for electron spin dynamics, which is valid in a strong magnetic field. In Sec. III, we present the Markov approximation and its range of validity, giving an analytical expression for the decoherence time T_2. We also give bounds for the non-Markovian corrections to our expression. Section IV gives a discussion of the decoherence rate for a homonuclear system, and in Sec. V we generalize these results for a heteronuclear-spin bath, providing explicit analytical expressions for T_2 within our Born-Markov approximation. We conclude in Sec. VI and present additional technical details in Appendixes A–D.

II. HAMILTONIAN

We begin from the Hamiltonian for the Fermi contact hyperfine interaction between a localized spin-1/2 $\mathbf{S}$ and an environment of nuclear spins,

$$H_{\mathrm{hf}} = bS^z + b\sum_k \gamma_k I_k^z + \mathbf{S}\cdot\mathbf{h}, \quad \mathbf{h} = \sum_k A_k \mathbf{I}_k. \tag{1}$$

Here, $\mathbf{I}_k$ is the nuclear-spin operator for the spin at site k with associated hyperfine coupling constant A_k, $b=g^*\mu_B B$ is the electron Zeeman splitting in an applied magnetic field B and γ_k is the nuclear gyromagnetic ratio in units of the electron gyromagnetic ratio (we set $\hbar=1$): $\gamma_k = g_{I_k}\mu_N/g^*\mu_B$. For an electron with an envelope wave function $\psi(\mathbf{r})$, we have $A_k = v_0 A^{i_k}|\psi(\mathbf{r}_k)|^2$, where A^{i_k} is the total coupling constant to a nuclear spin of species i_k at site k and v_0 is the volume of a unit cell containing one nucleus. For convenience, we define $A=\sqrt{\Sigma_i \nu_i (A^i)^2}$, where ν_i is the relative concentration of isotope i. The envelope function $\psi(\mathbf{r})$ of the bound electron has a finite extent, and, consequently, there will be a finite number $\sim N$ of nuclei with appreciable A_k. For typical quantum dots, $N\sim 10^4-10^6$, and for donor impurities or molecular magnets, $N\sim 10^2-10^3$. In Eq. (1), we have neglected the anisotropic hyperfine interaction, dipole-dipole interaction between nuclear spins, and nuclear quadrupolar splitting, which may be present for nuclear spin $I>1/2$. The anisotropic hyperfine interaction gives a small correction for electrons in a primarily s-type conduction band,[28] such as in III-V semiconductors or Si. Nuclear dipole-dipole coupling can give rise to dynamics in the spin bath, which can lead to electron-spin decay due to spectral diffusion on a time scale found to be $T_M \sim 10-100$ μs for GaAs quantum dots.[14,29,30] These times are 1 to 2 orders of magnitude longer than the T_2 we predict for a GaAs quantum dot carrying $N=10^5$ nuclei (see Fig. 3 below). For smaller systems, we expect the decay mechanism discussed here to dominate dipole-dipole effects substantially. The quadrupolar splitting has also been measured for nanostructures in GaAs, giving inverse coupling strengths on the order of 100 μs,[31] comparable to the dipole-dipole coupling strength, so quadrupolar effects should become relevant on comparable time scales.

For large b, we divide $H_{\rm hf}=H_0+V_{\rm ff}$ into an unperturbed part H_0 that preserves S^z and a term $V_{\rm ff}=\frac{1}{2}(S_+h_-+S_-h_+)$ that leads to energy nonconserving flip-flops between electron and nuclear spins.[15] We eliminate $V_{\rm ff}$ to leading order by performing a Schrieffer-Wolff-like transformation: $\bar{H}=e^S H_{\rm hf}e^{-S}\approx H=H_0+\frac{1}{2}[S,V_{\rm ff}]$, where $S=\frac{1}{\mathsf{L}_0}V_{\rm ff}$, and L_0 is the unperturbed Liouvillian, defined by $\mathsf{L}_0 O=[H_0,O]$. The resulting effective Hamiltonian is of the form[29,32] (see Appendix B)

$$H=(\omega+X)S^z+D. \tag{2}$$

The operators ω and D are diagonal with respect to a product-state basis of I_k^z eigenstates $\otimes_k|I_{i_k}m_k\rangle$, whereas the term X is purely off-diagonal in this basis, leading to correlations between different nuclei. We neglect corrections to the diagonal part of H of the order of $\sim A^2/Nb$, but retain the term of this size in the off-diagonal part X. This approximation is justified since, as we will show, the bath correlation time τ_c is much shorter than the time scale where these diagonal corrections become relevant for a sufficiently large Zeeman splitting $b\gg A$, where a Born-Markov approximation is valid: $\tau_c\sim N/A\ll Nb/A^2$. In addition, we ignore corrections to X that are smaller by the factors $A_k/b\sim A/Nb\ll 1$ and $\gamma_k\sim 10^{-3}$. Under these approximations, the various terms in Eq. (2) are given by (see also Appendix B),

$$\omega\simeq b+h^z,\quad D\simeq b\sum_k \gamma_k I_k^z, \tag{3}$$

$$X\simeq\frac{1}{2}\sum_{k\neq l}\frac{A_kA_l}{\omega}I_k^+I_l^+. \tag{4}$$

III. MARKOV APPROXIMATION

For large b, $H_{\rm hf}$ leads only to an incomplete decay of the longitudinal spin $\langle S_z\rangle_t$.[15] However, it is still possible for the transverse spin $\langle S_+\rangle_t$ to decay fully[16] through a pure dephasing process, which we now describe in detail. We assume that the electron and nuclear systems are initially unentangled with each other and that the nuclear-spin system is prepared in a narrowed state (an eigenstate of the operator ω: $\omega|n\rangle=\omega_n|n\rangle$) through a sequence of weak measurements,[27,33,34] polarization pumping,[35] frequency focusing under pulsed optical excitation,[36] or by any other means. For these initial conditions, dynamics of the transverse electron spin $\langle S_+\rangle_t$ are described by the exact equation of motion,[15]

$$\langle\dot{S}_+\rangle_t=i\omega_n\langle S_+\rangle_t-i\int_0^t dt'\Sigma(t-t')\langle S_+\rangle_{t'}, \tag{5}$$

$$\Sigma(t)=-i\,\mathrm{Tr}\,S_+\mathsf{L}e^{-i\mathsf{QL}t}\mathsf{QL}|n\rangle\langle n|S_-. \tag{6}$$

Here, L and Q are superoperators, defined by their action on an arbitrary operator O: $\mathsf{L}O=[H,O]$, $\mathsf{Q}O=(1-|n\rangle\langle n|\mathrm{Tr}_I)O$, where Tr_I indicates a partial trace over the nuclear-spin system.

To remove fast oscillations in $\langle S_+\rangle_t$, we transform to a rotating frame, in which we define the coherence factor $x_t=2\exp[-i(\omega_n+\Delta\omega)t]\langle S_+\rangle_t$ and associated memory kernel $\tilde{\Sigma}(t)=\exp[-i(\omega_n+\Delta\omega)t]\Sigma(t)$, with frequency shift determined self-consistently through $\Delta\omega=-\mathrm{Re}\int_0^\infty dt\tilde{\Sigma}(t)$. Additionally, we change integration variables to $\tau=t-t'$. The equation of motion for x_t then reads

$$\dot{x}_t=-i\int_0^t d\tau\tilde{\Sigma}(\tau)x_{t-\tau}. \tag{7}$$

If $\tilde{\Sigma}(\tau)$ decays to zero sufficiently quickly[44] on the time scale $\tau_c\ll T_2$, where T_2 is the decay time of x_t, we can approximate $x_{t-\tau}\approx x_t$ and extend the upper limit on the integral to $t\to\infty$ (Markov approximation), giving an exponential coherence decay with a small error $\epsilon(t)$,

$$x_t=\exp(-t/T_2)x_0+\epsilon(t),\quad \frac{1}{T_2}=-\mathrm{Im}\int_0^\infty dt\tilde{\Sigma}(t). \tag{8}$$

The non-Markovian correction $\epsilon(t)$ can be bounded precisely if $\tilde{\Sigma}(t)$ is known,[37]

$$|\epsilon(t)|\leq|\epsilon(t)|_{\max}=2\int_0^t dt'\left|\int_{t'}^\infty dt''\tilde{\Sigma}(t'')\right|. \tag{9}$$

Equation (9) gives a hard bound on the validity of the Markov approximation and, consequently, any corrections to the exponential decay formula. Figure 1 demonstrates an application of Eqs. (8) and (9) for decay in a homonuclear-spin system, which we discuss below.

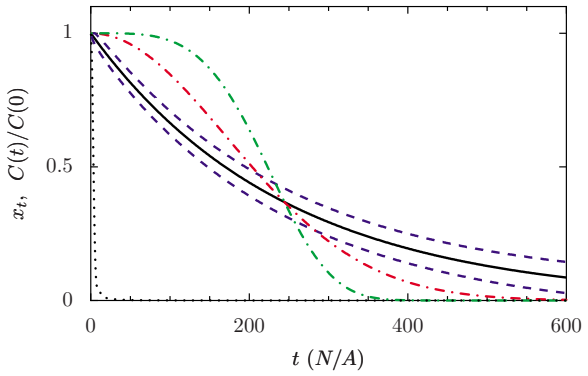

FIG. 1. (Color online) Exponential decay $x_t=\exp(-t/T_2)$ (solid line) and maximum error bounds $x_t\pm|\epsilon(t)|_{\max}$ (dashed lines), found by the numerical integration of Eq. (9) with parameters for a two-dimensional quantum dot [before Eq. (14)], $I=3/2$ and $A/b=1/20$. For comparison, we show the decay curves for superexponential forms $\exp[-(t/T_2)^2]$ and $\exp[-(t/T_2)^4]$ (dot-dashed lines) and rapidly decaying bath correlation function $C(t)/C(0)$ [dotted line, see Eqs. (10) and (11)].

IV. HOMONUCLEAR SYSTEM

If only one spin-carrying nuclear isotope is present, $\gamma_k=\gamma$, independent of the nuclear site. We then approximate $\Sigma(t)$ to leading order in the perturbation $V=XS^z$ (Born approximation, see Appendix C) by expanding Eq. (6) through the iteration of the Dyson identity: $e^{-i\mathsf{L}\mathsf{Q}t}$

$=e^{-i\mathsf{L}_0\mathsf{Q}t}-i\int_0^t dt' e^{-i\mathsf{L}_0\mathsf{Q}(t-t')}\mathsf{L}_V\mathsf{Q}e^{-i\mathsf{L}\mathsf{Q}t'}$, where $\mathsf{L}_V O=[V,O]$. Higher-order corrections to the Born approximation will be suppressed by the small parameter A/ω_n.[15] Inserting the result into Eq. (8), we find

$$\frac{1}{T_2}=\mathrm{Re}\int_0^\infty dt e^{-i\Delta\omega t}\langle X(t)X\rangle,\quad X(t)=e^{-i\omega t}Xe^{i\omega t}. \quad (10)$$

Here, $\langle\cdots\rangle=\langle n|\cdots|n\rangle$ denotes an expectation value with respect to the initial nuclear state. Equation (10) resembles the standard result for pure dephasing in a weak coupling expansion, where $X(t)$ would represent the bath operator in the interaction picture with an independent bath Hamiltonian. However, for the spin bath, there is no such weak coupling expansion, and $X(t)$ appears in the interaction picture with ω, the same operator that provides an effective level splitting for the system. Additionally, the general result for a heteronuclear system including interspecies flip-flops cannot be written in such a compact form.[38]

Previously, it has been shown that a Born-Markov approximation to *second* order in V_{ff} leads to no decay.[15] In contrast, a Born-Markov approximation applied to the effective Hamiltonian leads directly to a result that is *fourth* order in V_{ff} [Eq. (10)], describing the dynamics that become important at times longer than the second-order result. It is not *a priori* obvious that the effective Hamiltonian, evaluated only to second order in V_{ff}, can be used to accurately calculate rates to fourth order in V_{ff}. We have, however, verified that all results we present here are equivalent to a direct calculation expanded to fourth order in V_{ff} at leading order in $A/b\ll 1$.[38]

If the initial nuclear polarization is smooth on the scale of the electron wave function, the matrix elements of operators such as $I_k^{\pm}I_k^{\mp}$ can be replaced by average values. Neglecting corrections that are small in $A/Nb\ll 1$, this gives (see also Appendix D)

$$C(t)=\langle X(t)X(0)\rangle=\frac{c_+c_-}{4\omega_n^2}\sum_{k\neq l}A_k^2A_l^2e^{-i(A_k-A_l)t}. \quad (11)$$

Above, we have introduced the coefficients $c_{\pm}=I(I+1)-\langle\langle m(m\pm 1)\rangle\rangle$, and the double angle bracket indicates an average over I_k^z eigenvalues m.[15]

In the limit $N\gg 1$, we can include the term $k=l$ in Eq. (11) and perform the continuum limit $\Sigma_k\to\int dk$ with small corrections. For an isotropic electron wave function of the form $\psi(r)=\psi(0)\exp[-(r/r_0)^q/2]$ containing N nuclei within radius r_0 in d dimensions, the hyperfine coupling constants are distributed according to $A_k=A_0\exp[-(k/N)^{q/d}]$, where k is a non-negative index, and we choose A_0 to normalize A_k according to $A=\int_0^\infty dkA_k$ (Ref. 15) (see also Appendix A).

After performing the continuum limit, $C(t)$ will decay, with characteristic time τ_c given by the inverse bandwidth of nuclear flip-flop excitations $\tau_c\sim 1/A_0\sim N/A$. For large b, $1/T_2$ will be suppressed due to the smallness of X [see Eq. (4)], whereas τ_c remains fixed. At sufficiently large b, it will therefore be possible to reach the Markovian regime, where τ_c is short compared to T_2: $\tau_c/T_2\ll 1$. Evaluating the time integral in Eq. (10), we find the general result to leading order in A/ω_n (see Appendix D),

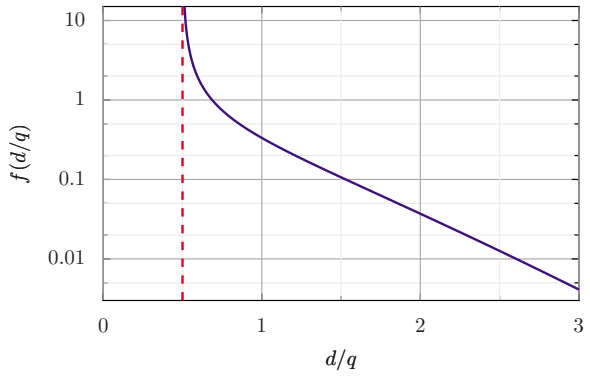

FIG. 2. (Color online) Geometrical factor $f(d/q)$ from Eq. (13), where $d=1,2,3$ is the dimension and q characterizes the electron envelope function $\psi(r)=\psi(0)\exp[-(r/r_0)^q/2]$.

$$\frac{1}{T_2}=\frac{\pi}{4}c_+c_-f\left(\frac{d}{q}\right)\left(\frac{A}{\omega_n}\right)^2\frac{A}{N}, \quad (12)$$

$$f(r)=\frac{1}{r}\left(\frac{1}{3}\right)^{2r-1}\frac{\Gamma(2r-1)}{[\Gamma(r)]^3},\quad r>1/2. \quad (13)$$

In Eq. (12), A/N sets the scale for the maximum decay rate in the perturbative regime, the coefficients $c_{\pm}$ set the dependence on the initial nuclear polarization p [e.g., with $I=1/2$, we have $c_+c_-=(1-p^2)/4$], $A/\omega_n<1$ gives the small parameter, which controls the Born approximation, and $f(d/q)$ is a geometrical factor (plotted in Fig. 2). $f(d/q)$ is exponentially suppressed for $d/q>1$ $[f(r)\propto(1/3)^{2r-1}(1/r)^r, r>1]$, but $f(d/q)\to\infty$ for $d/q-1/2\to 0^+$. Due to this divergence, no Markov approximation is possible (within the Born approximation) for $d/q\leq 1/2$. We understand the divergence in $f(d/q)$ explicitly from the asymptotic dependence of $C(t)$ at long times: $C(t)\propto 1/t^{2d/q}, t\gg N/A, d/q<2$.[44] Surprisingly, there is a difference of nearly *two orders* of magnitude in $1/T_2$ going from a two-dimensional (2D) quantum dot with a Gaussian envelope function ($d=2$, $q=2$, and $d/q=1$) to a donor impurity with a hydrogenlike exponential wave function ($d=3$, $q=1$, and $d/q=3$) if all other parameters are fixed (see Fig. 2).

We now specialize to an initial uniform unpolarized spin bath, which is nevertheless narrowed: $\omega|n\rangle=b|n\rangle$, with equal populations of all nuclear Zeeman levels [i.e., $\langle\langle m\rangle\rangle=0$ and $\langle\langle m^2\rangle\rangle=\frac{1}{3}I(I+1)$]. For a 2D quantum dot with a Gaussian envelope function ($d=q=2$), we find the following from Eqs. (12) and (13):

$$\frac{1}{T_2}=\frac{\pi}{3}\left(\frac{I(I+1)A}{3b}\right)^2\frac{A}{N}. \quad (14)$$

There are two remarkable features of this surprisingly simple result. First, the condition for the validity of the Markov approximation, $T_2>\tau_c\sim N/A$ will be satisfied whenever $A/b<1$, which is the same condition that validates a Born approximation. Second, $1/T_2$ has a very strong dependence on the nuclear spin ($1/T_2\propto I^4$). Thus, systems with large-spin nuclei such as In ($I_{\mathrm{In}}=9/2$) will show relatively significant faster decay (see, e.g., Fig. 3).

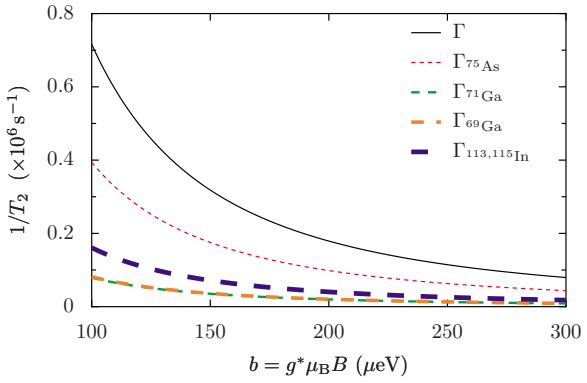

FIG. 3. (Color online) Decay rates for an $In_xGa_{1-x}As$ quantum dot with In doping $x=0.05$. Here, we have assumed $N=10^5$ and used values of ν_i and A^i for GaAs from Ref. 41: $A^{^{75}\mathrm{As}}=86$ μeV, $A^{^{69}\mathrm{Ga}}=74$ μeV, $A^{^{71}\mathrm{Ga}}=96$ μeV, $\nu_{^{75}\mathrm{As}}=0.5$, $\nu_{^{69}\mathrm{Ga}}=0.3(1-x)$, and $\nu_{^{71}\mathrm{Ga}}=0.2(1-x)$. The hyperfine coupling for In in InAs was taken from Ref. 42: $A^{^{113}\mathrm{In}}\approx A^{^{115}\mathrm{In}}\approx A^{\mathrm{In}}=170$ μeV, $\nu_{\mathrm{In}}=x/2$.

V. HETERONUCLEAR SYSTEM

For sufficiently large b ($|\gamma_k-\gamma_{k'}|b \gg |A_k-A_{k'}| \sim A/N$), heteronuclear flip-flops between two isotopic species with different γ_k are forbidden due to energy conservation. In this case, $1/T_2$ is given in terms of an incoherent sum, $1/T_2=\Gamma=\Sigma_i\Gamma_i$, where Γ_i is the contribution from flip-flops between nuclei of the common species i. Assuming a uniform distribution of all isotopes in a 2D quantum dot with a Gaussian envelope function, we find (see also Appendix D)

$$\Gamma_i = \frac{1}{T_2^i} = \nu_i^2 \frac{\pi}{3}\left(\frac{I_i(I_i+1)A^i}{3b}\right)^2 \frac{A^i}{N}. \tag{15}$$

The quadratic dependence on isotopic concentration ν_i is particularly striking. Due to this dependence, electron spins in GaAs, where Ga has two naturally occurring isotopic species, whereas As has only one, will show a decay predominantly due to flip-flops between As spins. This is in spite of the fact that all isotopes in GaAs have the same nuclear spin and nominally similar hyperfine coupling constants (see Fig. 3). Interestingly, we note that the relatively large flip-flop rates for In and As, due to large nuclear spin and isotopic concentration, respectively, may partly explain why only Ga (and not In or As) spins have been seen to contribute to coherent effects in experiments on electron transport through (In/Ga)As quantum dots.[39] The same effect may also explain why polarization appears to be transferred more efficiently from electrons to As (rather than Ga) in GaAs quantum dots.[40]

VI. CONCLUSIONS

We have shown that a single electron spin can exhibit a purely exponential decay for narrowed nuclear-spin-bath initial conditions and in the presence of a sufficiently large electron Zeeman splitting b. This work may be important for implementing existing quantum error correction schemes, which typically assume exponential decay of correlation functions due to a Markovian environment. In the limit of a large Zeeman splitting $b>A$, where a Born-Markov approximation is valid, we have found explicit analytical expressions for the decoherence time T_2, giving explicit dependences on the electron wave function, magnetic field, bath polarization, nuclear spin, and isotopic abundance for a general nuclear-spin bath. Moreover, within the Born-Markov approximation, we have found a divergence in the decoherence rate $1/T_2$ for a one-dimensional quantum dot, indicating a breakdown of the Markov approximation in this case.

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APPENDIX A: CONTINUUM LIMIT

In this appendix, we describe how the dimensionality d and envelope wave function shape parameter q are defined. For further details on the definition of these quantities, see Ref. 15. For a homonuclear-spin system, the hyperfine coupling constants are given by

$$A_k = Av_0|\psi(\mathbf{r}_k)|^2, \tag{A1}$$

where A is the total hyperfine coupling constant, v_0 is the volume occupied by a single-nucleus unit cell, and $\psi(\mathbf{r})$ is the electron envelope wave function. We assume an isotropic electron envelope,

$$\psi(r_k) = \psi(0)\exp[-(r_k/r_0)^q/2], \tag{A2}$$

where r_0 is the effective Bohr radius, defined as the radial distance enclosing N nuclear spins, and r_k is the radial distance enclosing k spins. In d dimensions,

$$\frac{\mathrm{vol}(k\ \mathrm{spins})}{\mathrm{vol}(N\ \mathrm{spins})} = \frac{v_0 k}{v_0 N} = \left(\frac{r_k}{a_B}\right)^d. \tag{A3}$$

Inserting Eqs. (A3) and (A2) into Eq. (A1),

$$A_k = A_0 e^{-(k/N)^{q/d}}. \tag{A4}$$

To determine the $k=0$ coupling A_0, we enforce the normalization

$$\sum_k A_k = Av_0 \sum_k |\psi(r_k)|^2 \approx A\int d^3r|\psi(r)|^2 = A. \tag{A5}$$

This gives

$$A = A_0 \int_0^\infty dk e^{-(k/N)^{q/d}}. \tag{A6}$$

Making the change of variables $u=(\frac{k}{N})^{q/d}$, we immediately find

$$A = A_0 \frac{d}{q} N \int_0^\infty du u^{d/q-1} e^{-u} = A_0 N \frac{d}{q}\Gamma\left(\frac{d}{q}\right), \tag{A7}$$

which gives the final form for A_k,

$$A_k = \frac{A}{N\frac{d}{q}\Gamma\left(\frac{d}{q}\right)} e^{-(k/N)^{q/d}}. \tag{A8}$$

APPENDIX B: EFFECTIVE HAMILTONIAN

In this appendix, we give details leading to the derivation of the effective Hamiltonian, described by Eqs. (2)–(4) of the main text. Similar effective Hamiltonians have been derived previously in Refs. 32 and 29, but due to some differences in method and approximation, we give additional details here for the interested reader. We begin from the hyperfine Hamiltonian,

$$H_{\rm hf} = H_0 + V_{\rm ff}, \tag{B1}$$

$$H_0 = (b + h^z)S^z + b\sum_k \gamma_k I_k^z, \tag{B2}$$

$$V_{\rm ff} = \frac{1}{2}(S_+ h_- + S_- h_+), \tag{B3}$$

$$\mathbf{h} = \sum_k A_k \mathbf{I}_k. \tag{B4}$$

To find an effective Hamiltonian that eliminates the flip-flop term $V_{\rm ff}$ at leading order, we apply a unitary transformation,

$$\bar{H} = e^S H_{\rm hf} e^{-S}, \tag{B5}$$

where $S=-S^\dagger$ to ensure unitarity. We now expand Eq. (B5) in powers of S, retaining terms up to $\mathcal{O}(V_{\rm ff}^3)$, assuming $S \sim \mathcal{O}(V_{\rm ff})$,

$$\bar{H} = H_0 + V_{\rm ff} - [H_0, S] - [V_{\rm ff}, S] + \frac{1}{2}[S,[S,H_0]] + \mathcal{O}(V_{\rm ff}^3). \tag{B6}$$

To eliminate $V_{\rm ff}$ at leading order, we must choose S to satisfy $V_{\rm ff}-[H_0,S]=0$. The S that satisfies this relation is given by

$$S = \frac{1}{\mathsf{L}_0} V_{\rm ff}, \quad \mathsf{L}_0 O = [H_0, O], \tag{B7}$$

which is of order $V_{\rm ff}$, justifying our previous assumption: $S \sim \mathcal{O}(V_{\rm ff})$. Reinserting Eq. (B7) into Eq. (B6), we find the following up to corrections that are third or higher order in $V_{\rm ff}$:

$$\bar{H} = H + \mathcal{O}(V_{\rm ff}^3), \tag{B8}$$

$$H = H_0 + \frac{1}{2}[S, V_{\rm ff}]. \tag{B9}$$

Directly evaluating Eq. (B7) with H_0 defined in Eq. (B2) and $V_{\rm ff}$ defined in Eq. (B3) gives

$$S = \frac{1}{2}\sum_k A_k \left(\frac{1}{b + h^z + \frac{A_k}{2} - b\gamma_k} S^+ I_k^- - \frac{1}{b + h^z - \frac{A_k}{2} - b\gamma_k} S^- I_k^+ \right). \tag{B10}$$

Inserting Eq. (B10) into Eq. (B9) gives

$$H = |\uparrow\rangle\langle\uparrow| H_\uparrow + |\downarrow\rangle\langle\downarrow| H_\downarrow, \tag{B11}$$

$$H_\uparrow = \frac{1}{2}(b + h^z) + b\sum_k \gamma_k I_k^z + h_\uparrow, \tag{B12}$$

$$H_\downarrow = -\frac{1}{2}(b + h^z) + b\sum_k \gamma_k I_k^z - h_\downarrow. \tag{B13}$$

Here, the contributions resulting from the second-order term in $V_{\rm ff}$ are given explicitly by

$$h_\uparrow = \frac{1}{8}\sum_{kl} A_k A_l \left(\frac{1}{b + h^z + A_k/2 - b\gamma_k} I_k^- I_l^+ + I_l^- \frac{1}{b + h^z - A_k/2 - b\gamma_k} I_k^+ \right), \tag{B14}$$

$$h_\downarrow = \frac{1}{8}\sum_{kl} A_k A_l \left(\frac{1}{b + h^z - A_k/2 - b\gamma_k} I_k^+ I_l^- + I_l^+ \frac{1}{b + h^z + A_k/2 - b\gamma_k} I_k^- \right). \tag{B15}$$

We can rewrite H in terms of spin operators using $|\uparrow\rangle\langle\uparrow| = \frac{1}{2} + S^z$ and $|\downarrow\rangle\langle\downarrow| = \frac{1}{2} - S^z$, which gives Eq. (2) from the main text,

$$H = (\omega + X)S^z + D, \tag{B16}$$

$$X = (1 - \mathsf{P}_d)(h_\uparrow + h_\downarrow), \tag{B17}$$

$$D = b\sum_k \gamma_k I_k^z + \frac{1}{2}(h_\uparrow - h_\downarrow), \tag{B18}$$

$$\omega = b + h^z + \mathsf{P}_d(h_\uparrow + h_\downarrow). \tag{B19}$$

In the above expressions, we have introduced the diagonal projection superoperator $\mathsf{P}_d O = \Sigma_l |l\rangle\langle l| O |l\rangle\langle l|$, where the index l runs over all nuclear-spin product states $|l\rangle = \otimes_k |I_k m_k^l\rangle$. We now apply the commutation relation $[I_k^+, I_l^-] = 2I_k^z \delta_{kl}$ and expand the prefactors in Eqs. (B14) and (B15) in terms of the smallness parameter $\frac{A_k}{b+h^z-b\gamma_k} \sim \frac{1}{N}\frac{A}{b} \ll 1$. At leading order in the expansion, we find $h_{\uparrow,\downarrow} \approx h_{\uparrow,\downarrow}^{(0)}$, where

$$h_\uparrow^{(0)} = \frac{1}{8}\sum_{kl} \frac{A_k A_l}{b + h^z - b\gamma_k} (I_k^- I_l^+ + I_l^- I_k^+), \tag{B20}$$

$$h_\downarrow^{(0)} = \frac{1}{8}\sum_{kl} \frac{A_k A_l}{b + h^z - b\gamma_k} (I_k^+ I_l^- + I_l^+ I_k^-). \tag{B21}$$

By commuting the nuclear-spin operators, Eqs. (B20) and (B21) can be rewritten to give

$$h_{\downarrow}^{(0)} = h_{\uparrow}^{(0)} + \frac{1}{2}\sum_k \frac{A_k^2}{b + h^z - b\gamma_k} I_k^z. \tag{B22}$$

This relation allows us to approximate the various terms in Eqs. (B17)–(B19),

$$X \approx (1 - \mathsf{P}_d)(2h_{\uparrow}^{(0)}) = \frac{1}{4}\sum_{k\neq l} \frac{A_k A_l}{b + h^z - b\gamma_k}(I_k^- I_l^+ + I_l^- I_k^+), \tag{B23}$$

$$D \approx \sum_k \left(b\gamma_k - \frac{A_k^2}{4(b + h^z - b\gamma_k)} \right) I_k^z, \tag{B24}$$

$$\omega \approx b + h^z + \mathsf{P}_d(2h_{\uparrow}^{(0)}) + \frac{1}{2}\sum_k \frac{A_k^2}{b + h^z - b\gamma_k} I_k^z, \tag{B25}$$

or

$$\omega \approx b + h^z + \frac{1}{2}\sum_k \frac{A_k^2}{b + h^z - b\gamma_k}[I_k(I_k + 1) - (I_k^z)^2]. \tag{B26}$$

Neglecting further corrections that are smaller by the factor $b\gamma_k/\omega \sim \gamma_k \sim 10^{-3}$ in Eq. (B23) and terms of order $\lesssim \Sigma_k \frac{A_k^2}{b+h^z-b\gamma_k} \sim \frac{A^2}{Nb}$ in Eqs. (B24) and (B26), we arrive immediately at Eqs. (3) and (4) of the main text. The terms of order $\sim A^2/Nb$ may become important on a time scale $\tau \sim Nb/A^2$. In our treatment, this time scale is long compared to the bath correlation time $\tau_c \sim N/A$ in the perturbative regime $A/b < 1$, and so neglecting these terms is justified.

APPENDIX C: BORN APPROXIMATION

In this appendix, we give further details on the Born approximation. We begin from the equation of motion for the transverse spin in the rotating frame x_t after applying the Markov approximation, neglecting the correction $\epsilon(t)$ [following Eq. (7)],

$$\dot{x}_t = -i\int_0^\infty d\tau \tilde{\Sigma}(\tau) x_t, \tag{C1}$$

$$\tilde{\Sigma}(t) = e^{-i(\omega_n + \Delta\omega)t}\Sigma(t), \tag{C2}$$

$$\Sigma(t) = -i\,\mathrm{Tr}\, S_+ \mathsf{LQ} e^{-i\mathsf{LQ}t}\mathsf{LQ}|n\rangle\langle n|S_-. \tag{C3}$$

In general, it is not simple to find the exact form of the self-energy (memory kernel) $\Sigma(t)$. Fortunately, it is possible to generate a systematic expansion in the perturbation $V = XS^z \propto 1/b$, valid for a sufficiently large Zeeman splitting $b > A$,[15]

$$\Sigma(t) = \Sigma^{(2)}(t) + \Sigma^{(4)}(t) + \cdots, \tag{C4}$$

where $\Sigma^{(n)}(t)$ indicates a term of order $\sim \mathcal{O}(V^n) \sim \mathcal{O}[(\frac{A}{b})^n]$. The expansion is performed most conveniently in terms of the Laplace-transformed variable,

$$\Sigma(s) = \mathcal{L}[\Sigma(t)] = \int_0^\infty dt e^{-st}\Sigma(t). \tag{C5}$$

We expand the propagator $\mathcal{L}[e^{-i\mathsf{LQ}t}] = \frac{1}{s+i\mathsf{LQ}}$ by dividing the full Liouvillian into unperturbed and perturbed parts: $\mathsf{L} = \mathsf{L}_0 + \mathsf{L}_V$, where L_0 and L_V are defined by their action on an arbitrary operator O through $\mathsf{L}_0 O = [H_0, O]$ and $\mathsf{L}_V O = [V, O]$. To obtain an expansion in terms of the perturbation L_V, we now iterate the Dyson identity in Laplace space,

$$\frac{1}{s + i\mathsf{LQ}} = \frac{1}{s + i\mathsf{L}_0\mathsf{Q}} - i\frac{1}{s + i\mathsf{L}_0\mathsf{Q}}\mathsf{L}_V\mathsf{Q}\frac{1}{s + i\mathsf{L}_0\mathsf{Q}} + \mathcal{O}(\mathsf{L}_V^2). \tag{C6}$$

Inserting the iterated expression [Eq. (C6)] into the Laplace-transformed version of Eq. (C3), we find that the self-energy in the Born approximation (to second order in V) is

$$\Sigma^{(2)}(s) = -i\,\mathrm{Tr}\left[S_+ \left(1 - i\mathsf{L}_0\mathsf{Q}\frac{1}{s + i\mathsf{L}_0} \right) \mathsf{L}_V \frac{1}{s + i\mathsf{L}_0}\mathsf{L}_V |n\rangle\langle n| S_- \right]. \tag{C7}$$

We have simplified the above expression using the following identities for the projection superoperators $\mathsf{Q} = 1 - |n\rangle\langle n|\mathrm{Tr}_I$ and $\mathsf{P} = 1 - \mathsf{Q}$:

$$\mathsf{P}\mathsf{L}_0\mathsf{P} = \mathsf{L}_0\mathsf{P}, \tag{C8}$$

$$\mathsf{P}\mathsf{L}_V|n\rangle\langle n| = 0, \tag{C9}$$

$$\mathsf{Q}\mathsf{L}_0\mathsf{Q} = \mathsf{Q}\mathsf{L}_0, \tag{C10}$$

which can be proven directly. To further reduce the above expression, we evaluate the action of L_0 and L_V on the electron-spin operator S_-,

$$\mathsf{L}_V S_- = -\frac{1}{2}\mathsf{L}_X^+ S_-, \tag{C11}$$

$$\mathsf{L}_0 S_- = \left(-\frac{1}{2}\mathsf{L}_\omega^+ + \mathsf{L}_D \right) S_-, \tag{C12}$$

where

$$\mathsf{L}_X^+ O = [X, O]_+, \tag{C13}$$

$$L_\omega^+ O = [\omega, O]_+, \tag{C14}$$

$$L_D O = [D, O], \tag{C15}$$

and here we denote anticommutators with a $+$ subscript: $[A,B]_+ = AB + BA$. This leads to

$$\Sigma^{(2)}(s) = -\frac{i}{4}\mathrm{Tr}_I\left[\left(1 + \frac{i}{2}\mathsf{L}_\omega^+\mathsf{Q}\frac{1}{s - \frac{i}{2}\mathsf{L}_\omega^+} \right) \times \mathsf{L}_X^+ \frac{1}{s + i\left(\mathsf{L}_D - \frac{1}{2}\mathsf{L}_\omega^+ \right)} \mathsf{L}_X^+ |n\rangle\langle n| \right]. \tag{C16}$$

Now, noting that

$$\mathsf{Q}|n\rangle\langle n| = 0, \tag{C17}$$

$$\mathsf{Q}|k\rangle\langle k| = |k\rangle\langle k| - |n\rangle\langle n|, \tag{C18}$$

we can evaluate Eq. (C16) directly, giving

$$\Sigma^{(2)}(s+i\omega_n) = -\frac{i}{2}\sum_k |X_{kn}|^2 \left(\frac{s+\frac{i}{2}\delta\omega_{nk}}{s+i\delta\omega_{nk}}\right) \times \left(\frac{1}{s+i\left(\delta D_{kn}+\frac{1}{2}\delta\omega_{nk}\right)} + \frac{1}{s-i\left(\delta D_{kn}-\frac{1}{2}\delta\omega_{nk}\right)}\right), \tag{C19}$$

where $\delta D_{kn}=D_k-D_n$, $\delta\omega_{nk}=\omega_n-\omega_k$, and ω_k and D_k are the eigenvalues associated with eigenstate $|k\rangle$: $\omega|k\rangle=\omega_k|k\rangle$ and $D|k\rangle=D_k|k\rangle$. Additionally, we have denoted $X_{kn}=\langle k|X|n\rangle$.

From Eqs. (C1), (C2), and (C5), the electron-spin decoherence rate within a Born-Markov approximation will now be given by

$$\frac{1}{T_2} = -\,\mathrm{Im}\,\Sigma^{(2)}[s=i(\omega_n+\Delta\omega)+0^+], \tag{C20}$$

where 0^+ denotes a positive infinitesimal. Our goal here is to find the leading-order dependence of $1/T_2$ on $1/b$ for a large Zeeman splitting: $b>A$. We therefore set $\Delta\omega=-\mathrm{Re}\,\Sigma^{(2)}[s=i(\omega_n+\Delta\omega)+0^+]\sim\mathcal{O}(\frac{A}{N}(\frac{A}{b})^2)\approx 0$ since this term will lead to higher-order corrections in $1/b$ within the perturbative regime. Additionally, noting that the matrix element X_{kn} induces a flip-flop for spins at two sites $k_{1,2}$, we find $|\delta D_{kn}|=|b(\gamma_{k_1}-\gamma_{k_2})|$ and $|\delta\omega_{kn}|=|A_{k_1}-A_{k_2}|$. In the case of a homonuclear system $\gamma_{k_1}=\gamma_{k_2}$, we can set $\delta D_{kn}=0$ in Eq. (C19). Otherwise, in a sufficiently large magnetic field $|b(\gamma_{k_1}-\gamma_{k_2})|>|A_{k_1}-A_{k_2}|$, we find a negligible contribution to the decoherence rate for terms from two different isotopic species (where $\gamma_{k_1}\neq\gamma_{k_2}$); i.e., heteronuclear flip-flops no longer conserve energy, although homonuclear flip-flops (for which $\gamma_{k_1}=\gamma_{k_2}$) will still occur. Restricting the sum to homonuclear flip-flops and setting $\delta D_{nk}=0$ in this regime gives

$$\Sigma^{(2)}(s+i\omega_n) = -i\sum_j\sum_k |X^j_{kn}|^2\frac{1}{s+i\delta\omega_{nk}}, \tag{C21}$$

where $X^j_{kn}=\langle k|X^j|n\rangle$ and X^j is restricted to run over flip-flops between nuclei of the common species j at sites denoted by the indices k_j and l_j;

$$X^j = \frac{1}{2}\sum_{k_j\neq l_j}\frac{A^j_{k_j}A^j_{l_j}}{\omega}I^-_{k_j}I^+_{l_j}. \tag{C22}$$

Inserting Eq. (C21) for a homonuclear system (one isotopic species j) into Eq. (C20) and inverting the Laplace transform leads directly to Eq. (10) of the main text.

APPENDIX D: DECOHERENCE RATE

Applying Eq. (C20) (setting $\Delta\omega\approx 0$) with Eq. (C21) gives the rate

$$\frac{1}{T_2} = \pi\sum_j\sum_k |X^j_{kn}|^2\delta(\delta\omega_{kn}), \tag{D1}$$

which can be found directly from the formula

$$\frac{1}{x\pm i0^+} = \mathcal{P}\frac{1}{x}\mp i\pi\delta(x), \tag{D2}$$

where $\mathcal{P}$ indicates that the principal value should be taken in any integral over x. Rewriting Eq. (D1) using the definition of X^j given in Eq. (C22),

$$\frac{1}{T_2} = \frac{\pi}{4}\sum_j\sum_{k_j\neq l_j}\frac{c^{jk_j}_- c^{jl_j}_+}{\omega_k\omega_n}(A^j_{k_j})^2(A^j_{l_j})^2\delta(A^j_{k_j}-A^j_{l_j}), \tag{D3}$$

where k_j and l_j are restricted to run over sites occupied by isotopic species j. The coefficients $c^{jk_j}_\pm$ give the expectation value of the operator $I^\mp_{k_j}I^\pm_{k_j}$ with respect to the initial state,

$$c^{jk_j}_\pm = \langle n|I^\mp_{k_j}I^\pm_{k_j}|n\rangle, \tag{D4}$$

$$= I^j(I^j+1) - \langle n|I^z_{k_j}(I^z_{k_j}\pm 1)|n\rangle. \tag{D5}$$

With small corrections of order $A/Nb\ll 1$, we can replace $\omega_k\simeq\omega_n$ in the denominator of Eq. (D3). If the various nuclear isotopes are uniformly distributed with isotopic concentrations ν_j, we allow the sum over k_j and l_j to extend over all sites k and l at the expense of a weight factor ν_j for each index,

$$\sum_{k_j\neq l_j}\approx\nu_j^2\sum_{k\neq l}. \tag{D6}$$

Additionally, we assume that the system is uniformly polarized on the scale of variation of the hyperfine coupling constants so that the coefficients $c^{jk}_\pm$ can be replaced by average values $c^j_\pm=\langle\langle c^{jk}_\pm\rangle\rangle$ (double angle brackets indicate an average over all sites) and taken out of the sum. Finally, we change the sums over sites to a double integral using the prescription and coupling constants described in Appendix A, neglecting the small $\mathcal{O}(1/N)$ correction due to the requirement $k\neq l$,

$$\sum_{k\neq l}\to\int_0^\infty dk\int_0^\infty dl. \tag{D7}$$

These approximations give

$$\frac{1}{T_2} = \frac{\pi}{4\omega_n^2} \sum_j \nu_j^2 c_-^j c_+^j \int_0^\infty dk \int_0^\infty dl (A_k^j)^2 (A_l^j)^2 \delta(A_k^j - A_l^j). \tag{D8}$$

Inserting the coupling constants defined by Eq. (A8) and evaluating the integrals gives

$$\frac{1}{T_2} = \frac{\pi}{4} f\left(\frac{d}{q}\right) \sum_j \nu_j^2 c_-^j c_+^j \frac{A^j}{N} \left(\frac{A^j}{\omega_n}\right)^2, \tag{D9}$$

with the geometrical factor $f(d/q)$ given by Eq. (13) of the main text. Equation (D9) reduces to Eqs. (12), (14), and (15) of the main text in the special cases discussed there.

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