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Dynamical control of electron spin coherence in a quantum dot: A theoretical study

Wenxian Zhang,[1] V. V. Dobrovitski,[1] Lea F. Santos,[2] Lorenza Viola,[2] and B. N. Harmon[1]
[1]*Ames Laboratory, Iowa State University, Ames, Iowa 50011, USA*
[2]*Department of Physics and Astronomy, Dartmouth College, Hanover, New Hampshire 03755, USA*
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We investigate the performance of dynamical decoupling methods at suppressing electron spin decoherence from a low-temperature nuclear spin reservoir in a quantum dot. The controlled dynamics is studied through exact numerical simulation, with emphasis on realistic pulse delays and the long-time limit. Our results show that optimal performance for this system is attained by a periodic protocol exploiting concatenated design, with control rates substantially slower than expected from the upper spectral cutoff of the bath. For a known initial electron spin state, coherence can saturate at long times, signaling the creation of a stable "spin-locked" decoherence-free subspace. Analytical insight into saturation is obtained for a simple echo protocol, in good agreement with numerical results.

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Developing and benchmarking strategies for decoherence suppression in spin nanosystems is crucial for various areas of quantum physics, from quantum control theory to quantum device technologies. A single central spin 1/2 interacting with a bath of N external spins is a natural test bed for such studies,[1] showing a rich variety of decoherence regimes and paving the way to understanding more complex scenarios, such as decoherence of many-spin central systems. A prominent example is an electron spin localized in a GaAs quantum dot (QD): at experimentally relevant subkelvin temperatures and moderate (subtesla) magnetic fields, the hyperfine coupling with a bath of nuclear spins is the dominant decoherence channel. While electron spins in QDs have many potential applications in spintronics[2] and quantum information processing,[3] their coherence time is very short, $T_2^* \sim 10$ ns in a typical ($N \sim 10^6$) GaAs QD.[4] While suggestive proposals exist to increase T_2^* by achieving high bath spin polarization, by bath disentanglement, or by narrowing the nuclear spin distribution,[5] methods viable in a wider parameter range are still actively sought.

The distinctively non-Markovian behavior of the nuclear spin reservoir makes the electron spin in a QD an ideal candidate for dynamical decoupling (DD) techniques.[6,7] In double-QD systems, spin-singlet refocusing has been experimentally demonstrated.[4] For a single QD in a large external magnetic field ($B_0 \gtrsim 1$ T), where the nuclei simply dephase the electron spin, Hahn spin echoes and their Carr-Purcell-Meiboom-Gill extentions may increase T_2^* by more than an order of magnitude.[8,9] However, when the bias fields is not so large, the decoherence dynamics is much more complex, and since both dephasing and relaxation must be simultaneously eliminated, different DD protocols are demanded. Theoretical investigation of some DD protocols suitable for GaAs QDs at lower fields has begun very recently,[10,11] and several crucial questions remain unanswered. In particular, assessing the long-time performance of DD schemes under realistically large interpulse delays is among the most practically important, yet least studied, issues to date.

In this paper, we investigate quantitatively DD of electron spin decoherence in the experimentally relevant but very challenging situation of zero bias field. Using exact numerical simulations, we show that the very stringent formal limitations of DD methods may be relaxed and pulse delays *up to a factor* $\sqrt{N}$ *longer* than naively expected from analytical bounds can still extend the coherence time by 2–3 orders of magnitude. We study several DD protocols, focusing on the long-time limit, where error accumulation is crucial and neither the Magnus expansion (ME) nor the quasistatic approximation (QSA) is reliable *a priori*. Provided that the initial electron spin state is known, we demonstrate how nearly perfect coherence preservation is possible for indefinitely long times. Such a *saturation* is related to the creation of a *stable* decoherence free subspace[12] (DFS) and may allow one to stabilize the electron spin polarization in a QD.

Model and DD setting. The dynamics of a single electron spin S coupled to a bath B of N nuclear spins is described by a total Hamiltonian of the form $H=H_S+H_{SB}+H_B$, where $H_S=H_0S_z$ is the electron Zeeman energy, $H_{SB}=\Sigma_{k=1}^{N}A_k\mathbf{S}\cdot\mathbf{I}_k$ the hyperfine contact interaction between the electron spin and the nuclei, and $H_B=\Sigma_{k>l}^{N}\Gamma_{kl}(\mathbf{I}_k\cdot\mathbf{I}_l-3I_k^zI_l^z)$ the intrabath dipolar coupling between nuclear spins.[9,13–15] $\mathbf{S}$ and $\mathbf{I}_k$ denote the electron and the kth bath spin operators, respectively; the nuclear spin values $I_k=1/2$ are assumed.[16] We focus on the case $B_0=0$ and assume that, as in a standard experiment, temperature is much larger than the characteristic energy scales for nuclear spins. Thus, the bath's initial state is maximally mixed, $\rho_B(0)=2^{-N}\mathbf{1}_B$, $\mathbf{1}_B$ being the 2^N-dimensional identity matrix.[17] The free induction decay (FID) time is $T_2^*=(NA^2/8)^{-1/2}$, where $A=(\Sigma_kA_k^2/N)^{1/2} \approx 10^{-4}$ μeV for typical GaAs QDs with $N=10^6$ nuclear spins.[15] Below, time is measured in units of $1/A$.

Under ideal control assumptions, DD is implemented by subjecting the electron spin to sequences of instantaneous $\pi_{\hat{\mathbf{n}}}$ rotations along appropriate control axes $\hat{\mathbf{n}}$, equally separated by the interval τ. A variety of DD protocols exist, based on both deterministic[7,10] and randomized[18,19] designs. In *cyclic* DD, the control propagator is steered through a DD group of unitary operations $\mathcal{G}=\{g_j\}$, $j=0,1,\ldots,|\mathcal{G}|-1$, in a predetermined order, as opposed to *randomized* DD where the future control path is not known in advance. Changing g_i to g_j requires the application of a DD pulse $P_{i,j}=g_jg_i^\dagger$. Thanks to the existence of a periodicity time scale $T_c=|\mathcal{G}|\tau$, the analysis

of cyclic DD has been mostly carried out within average Hamiltonian theory,[6] upper performance bounds being determined by the dominant nonzero corrections in the ME for the time evolution operator. Average Hamiltonian theory no longer applies to randomized DD, where the evolution is most directly studied in a logical frame that follows the applied control.[18]

Periodic DD (PDD) is the simplest *nonselective* cyclic protocol, ensuring that the unwanted evolution is removed to first order in the ME at every $T_n=nT_c$, $n\in\mathbb{N}$, in the short T_c limit. For a single spin, PDD is based on the irreducible Pauli group $\mathcal{G}_P=\{I,X,Y,Z\}$, $X=\sigma_x$, and so on)[7] which requires two-axis control sequences of the form $C_1=C_0XC_0ZC_0XC_0Z$, C_0 denoting a free evolution period. Improvement over PDD may be gained by symmetrized and/or concatenated design. Symmetric DD (SDD) guarantees that all odd terms in the ME are canceled, with T_c twice as long as PDD. Concatenated DD[10] relies on a temporal recursive structure, so that at level $\ell+1$ the protocol is $C_{\ell+1}=C_\ell XC_\ell ZC_\ell XC_\ell Z$. Here, we *truncate* the concatenation procedure at a certain level and repeat a periodic sequence, referred to as PCDD, after every $4^\ell\tau$ (e.g., $\ell=2$ leads to PCDD_2). As representatives among stochastic protocols, we consider naive random DD (NRD), which corresponds to uniformly random pulses over $\mathcal{G}$, and symmetric random path DD (SRPD), where a path to traverse $\mathcal{G}$ is chosen at random and then symmetrized as in SDD.[19]

The use of control pulses may suit two purposes: (i) complete decoupling of the system from the bath, so that electron spin coherence is enhanced for an *arbitrary* initial state; (ii) preservation of a *specific* initial state, in which case the DD sequence may be tailored accordingly. Two performance metrics are then appropriate. For a fixed initial state $|\psi\rangle$, we use the input-output fidelity $F(T)=\mathrm{Tr}[\rho_S(T)\rho_S(0)]$, where $\rho_S(T)$ is the reduced density operator of S at time T starting from $|\psi\rangle$ and tracing out the bath. For an unknown initial state, we invoke minimum pure-state fidelity $F_m(T)=\min_{|\psi\rangle}F(T)$. Analytical bounds on the expected fidelity decay for various DD protocols have been obtained for short evolution times,[10,18,19] which calls for numerical analysis in the long-time regime. Simulations also make it possible to explore DD performance for values of T_c beyond the strict convergence domain of the ME, $\omega_cT_c\ll 1$, where the highest-frequency component $\omega_c\approx\Sigma_k|A_k|/4\sim NA/4$. Let σ denote the power spectrum width of the environmental coupling, $2\sigma\approx(\Sigma_kA_k^2)^{1/2}=\sqrt{N}A$.[20] We shall consider $\tau\sim 1/2\sigma$, and thus $T_c\geqslant 4\tau\sim\sqrt{N}\omega_c^{-1}$. To solve the time-dependent Schrödinger equation of the entire S plus B system, we apply the Chebyshev polynomial expansion method to the evolution operator[14,17] and choose $A_k>0$ as uniformly random numbers.

Unknown initial state. In Fig. 1 we compare $F_m(T)$ for the above-mentioned DD protocols. Because the characteristic time scale τ_D for nuclear dipolar dynamics due to H_B is (at least) two orders of magnitude slower than the one due to H_{SB} in typical QDs, setting $H_B=0$ is justified for practically relevant time regimes. All schemes lead to substantial enhancement of the electron spin coherence, PCDD_2 showing the most dramatic improvement. Although for this system

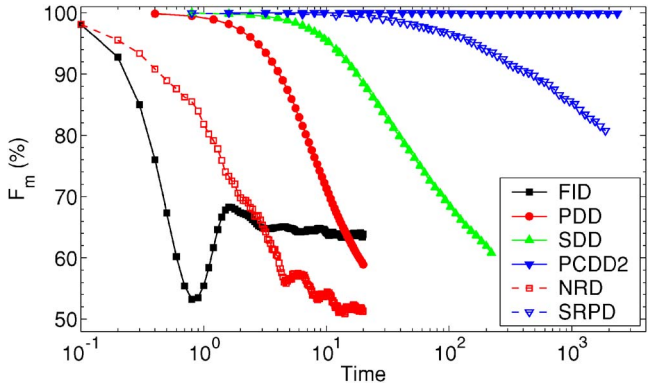

FIG. 1. (Color online) Minimum fidelity vs time in the logical frame with $\tau=0.1$. Hamiltonian parameters are $H_0=0$, $\Gamma_0=0$, and $N=15$. For deterministic DD, data points are acquired at the completion of each cycle, while for NRD and FID this is done after every τ and for SRPD after every 8τ. Random protocols are averaged over 10^2 control realizations.

both SDD and PCDD_2 remove H_{SB} to second order in the ME, the higher performance of PCDD_2 reflects its superiority in reducing coherent error accumulation. The poor performance of NRD is expected, since its advantages over deterministic DD emerge only when $\mathcal{G}$ is large. Contrary to the case of closed systems,[19] SRPD does not match PCDD_2 in the relevant parameter range, confirming the fact that irreducible DD groups and slow baths are predicted to be especially favorable for concatenated control.[10]

Motivated by the above results, we proceed with a more in-depth analysis of the PCDD protocol. Figure 2(a) compares the performance of two levels of concatenation, $\ell=2,4$, for different values of τ. As expected, the results deteriorate as τ increases but, interestingly, PCDD_4 may become worse than PCDD_2. Figure 2(b) illustrates, for each value of $2\sigma\tau$ and different N, the instant of time $T_{90\%}$ where

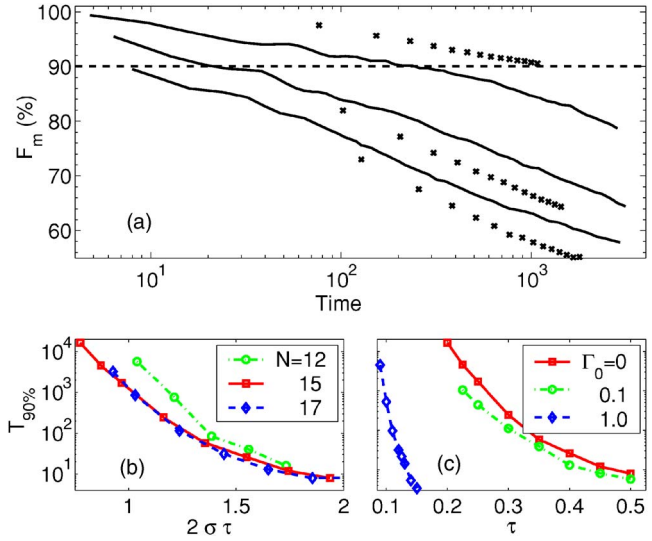

FIG. 2. (Color online) (a) PCDD_2 (solid lines) and PCDD_4 (crosses) for $\tau=0.3,0.4,0.5$, top to bottom. (b) and (c) $T_{90\%}$ vs τ for PCDD_2; different bath sizes (b) and intrabath interactions (c). $H_0=0$ in all panels, $N=15$ in (a) and (c).

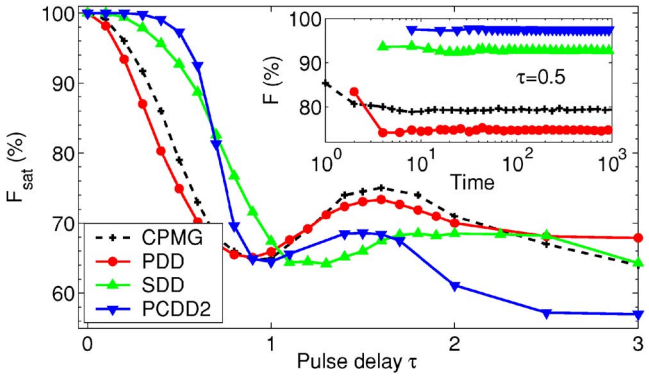

FIG. 3. (Color online) Fidelity saturation for CPMG, PDD, SDD, and $PCDD_2$ starting from an initial state along the half-cycle direction. Hamiltonian parameters as in Fig. 1. The inset shows how the asymptotic value F_{sat} is reached. In the main panel, a number of pulses sufficient to reach saturation and close the cycle of each protocol is chosen, $n_p \sim 50$.

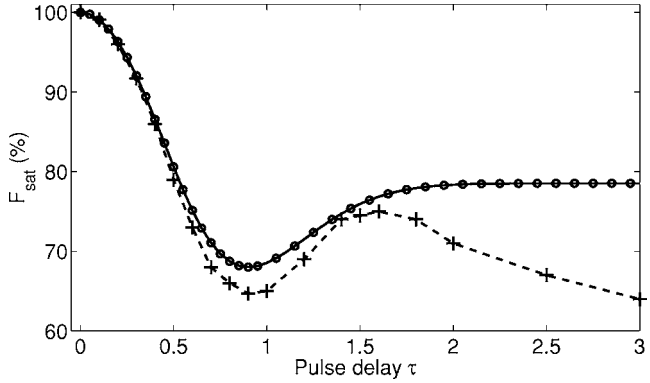

FIG. 4. Fidelity saturation vs pulse delay for CPMG from a known initial state. H as in Fig. 1. Circles, QSA results; solid line, classical random field model; dashed line with plus signs, exact numerical simulations.

$F_m(T)$ for $PCDD_2$ reaches 90%. The results are reasonably close to each other, particularly for larger N, supporting their applicability up to realistic situations with $N \sim 10^6$. Last, we analyze the effect of H_B, which becomes important once the coherence time is longer than τ_D. Let Γ_{kl} be uniformly random numbers in $[-\Gamma_0, \Gamma_0]$. To avoid demanding long-time simulations, we increase Γ_0 manually up to values comparable to A_k. The results are shown in Fig. 2(c), where a two-dimensional 3×5 QD with nearest-neighbor intrabath coupling is considered. $PCDD_2$ performance is significantly affected by a bath with fast dynamics. Although such a regime is not directly relevant to standard GaAs QDs, further investigation of randomized DD is necessary whenever H_B and H_{SB} compete.

Known initial state. If the electron spin is initially pointing along a known direction, *cyclic* DD protocols able to stabilize the input-output fidelity value for extremely long times may be used. This is shown in Fig. 3 (inset), where the curves $F(T)$ plateau after the application of a sufficient number n_p of pulses. While asymptotic saturation behavior has been reported for purely dephasing spin-boson models with *arbitrary* initial spin states,[7,21] the directional dependence observed here reflects the lack of a preferred direction in the error process generated by H_{SB}: a preferred direction only emerges through the "effective field" *created* by the control sequence, and long-time stability depends on proper alignment between such effective field and the initial state. In magnetic resonance language, the resulting saturation effect is closely related to the "pedestals" of the long-time magnetization signal in pulsed spin-locking experiments.[22] From a control standpoint, it indicates the dynamical generation of a stable one-dimensional DFS via DD.[12] Consider first a *selective* echo protocol—say, a single-axis PDD along the z direction, $\mathcal{G}_Z = \{I, Z\}$, with a corresponding (asymmetric) pulse sequence $C_Z = C_0 Z C_0 Z$—which we refer to as CPMG. For sufficiently small τ, symmetrization is enforced along the z axis, as described by a lowest-order Hamiltonian commuting with $\mathcal{G}_Z$ in the ME, and a corresponding effective field along z (Refs. 7 and 22): initial S_z eigenstates are (approximate) eigenstates of the decoupled evolution, whereas components perpendicular to the DFS are lost in the long-time regime. For nonselective DD protocols based on the irreducible group $\mathcal{G}_P$, all directions are approximately preserved for short times due to maximal averaging, yet long-time stability again occurs along the direction of the dominant term in the ME. Notice that the latter also coincides with the *half cycle* direction of the sequence: e.g., PDD may be obtained from concatenation of two CPMGs, $C_1 = C_X Y C_X Y = C_Y \circ C_X$, identifying the outer y direction as the stable one. Similarly, for SDD and $PCDD_2$, the saturated components are z and y, respectively.

Quantitative results on the dependence of the saturation value upon control parameters are given in Fig. 3. Interestingly, a crossing between SDD and $PCDD_2$ occurs at $\tau \sim 0.8$. In particular, the CPMG sequence, which is not a maximal DD scheme for the Hamiltonian in question, leads to saturation values comparable to the other protocols; thus, it may be useful in settings where accurate control along two axes may not be available. Moreover, its simplicity allows for a direct analytical study of the saturation effect. Within the QSA,[9,15] let $A_k = A$, $\mathbf{I} = \Sigma_k \mathbf{I}_k$, and $M = I_z$. After n CPMG cycles, the survival probability of the initial state $|\Psi(0)\rangle = |\uparrow\rangle \otimes |I, M\rangle$ is given by $|\langle \Psi(0)|\Psi(2n\tau)\rangle|^2 = 1 - (C^2/B^2)\tan^2\theta \cos^2 2n\theta$, where $C = A\sqrt{(I-M)(I+M+1)}$, $B = A(M+1/2)$, $\tan\theta = d/\sqrt{1-d^2}$, $d = -(B/\Omega)\sin(\Omega\tau/2)$, and $\Omega^2 = B^2 + C^2$. For the maximally mixed bath state, $\rho_B(0) = 2^{-N} 1_B$, and for large n and N, $F \to F_{sat} = 1 - (1/2)\int dI dM P(I,M)\ (C^2/B^2)\tan^2\theta$, where $P(I,M) \simeq (I/D\sqrt{2\pi D}) e^{-I^2/2D}$ and $D = N/4$.[20] In the limit of small τ, we obtain $F_{sat} = 1 - (1/16)\tau^2 A^2 N = 1 - \tau^2/2T_2^{*2}$. For randomly distributed A_k, $A^2 N \mapsto \Sigma_k A_k^2$.

Figure 4 compares the above analytical result with the saturation value predicted by a semiclassical approximation, which treats the nuclear Overhauser field as an effective random magnetic field with zero average, but finite variance.[23] The two curves superimpose, consistent with the fact that for large N the Overhauser field induced by $\mathbf{I}$ indeed approaches a classical field. Also shown are data from exact numerical simulations of a quantum spin bath with randomly distributed A_k. Remarkably, for short pulse delays the exact and

QSA results are in good agreement, in spite of the QSA being well known to be only valid for times comparable to T_2^*. Thus, (i) DD effectively extends the region of validity of QSA and (ii) saturation is entered before the QSA becomes invalid, allowing the QSA to accurately predict F_{sat} for short τ.

In summary, we have quantitatively characterized DD of an electron spin coupled to a nuclear spin bath, with emphasis on long-time behavior. We find that DD can significantly enhance the coherence time for an arbitrary initial state, actual performance depending on both control and physical parameters. For a known initial state, the possibility of long-time saturation has been established numerically and analytically, which may provide a way for preserving the electron spin polarization without the need of a strong permanent magnetic field. While from a practical standpoint the estimated control time scales ($\sim$1 ns) are roughly an order of magnitude away from current pulsing capabilities in GaAs QDs, experimental progress is steady. In particular, single-electron spin rotations have been demonstrated both in gate-defined GaAs and self-assembled QDs.[24] In addition, multipulse CPMG-DD has also been realized, not only in standard nuclear magnetic and electron spin resonance experiments, but also in single solid-state centers,[25] which share many relevant features with electron spins in QDs. These advances support the hope that the experimental implementation of more complex protocols will be achievable in the near future.

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