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Dynamical control of electron spin coherence in a quantum dot

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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 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 on 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 are of vital importance to various areas of quantum physics, from quantum control theory to quantum device technologies. A central spin 1/2 interacting with a bath of N external spins provides a natural testbed for detailed analysis. This conceptually simple system shows a rich variety of decoherence regimes, pitting the way to the understanding of more complex scenarios, such as decoherence of many-spin central systems. A prominent example is an electron localized in a quantum dot (QD) at experimentally relevant sub-Kelvin temperatures and moderate (sub-Tesla) magnetic fields, where the hyperfine coupling with a bath of nuclear spins is the dominant decoherence channel. Although electron spins in QDs have a broad range of potential applications in spintronics and scalable quantum information processing, the coherence time $T_2$ is very short, $T_2 \lesssim T_2^*$, where the free induction decay (FID) time $T_2 \sim 10$ ns in a typical GaAs QD. While suggestive proposals exist, to increase $T_2$ by achieving high bath spin polarization or bath disentanglement, or by narrowing the nuclear spin distribution, methods viable in a wider range of physical parameters are actively sought.

The long correlation time and distinctively non-Markovian behavior of the nuclear spin reservoir make the electron spin an ideal candidate for pulsed spin resonance and dynamical decoupling (DD) techniques. In double-QD devices, for instance, spin singlet refocusing has been experimentally demonstrated. For a single QD in a large external magnetic field, $B_0 \gtrsim 1$ Tesla, where the nuclei simply dephase the electron spin, Hahn spin echoes and their Carr-Purcell-Meiboom-Gill modifications are expected to enhance $T_2^*$ by at least an order of magnitude in GaAs QDs. For weaker bias fields, the nuclear spin coupling induces both dephasing and relaxation, and the use of higher-level DD schemes has been invoked recently. However, the formal limits of applicability of these analyses are very restrictive and extremely hard to meet in practice.

In this paper, we perform a quantitative study of the electron spin decoherence DD problem in regimes which are important for experimental DD implementations in QDs, yet have received little attention so far. Focusing on the challenging situation of zero external field, where dephasing and relaxation must be simultaneously eliminated, we investigate to what extent the very stringent formal limitations of DD methods may be relaxed. Using exact numerical simulations, we identify promising DD protocols, and show that for pulse delays up to a factor $\sqrt{N}$ longer than naively expected from analytical bounds, they are still capable of extending the coherence time by 2-3 orders of magnitude. Special emphasis is devoted to the asymptotic long-time limit, where error accumulation is crucial and neither intuition based on Magnus expansion (ME) nor the quasi-static approximation is reliable a priori. Provided that the initial electron spin state is known, nearly perfect coherence preservation may be achieved for indefinitely long times. Such a saturation is related to the creation of a stable decoherence-free subspace (DFS), and may be exploited for stabilizing 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 $H_S = H_0 S_z$ is the electron Zeeman energy in an external magnetic field $B_0$, $H_{SB} = \sum_{k=1}^{N} A_k S \cdot I_k$ the hyperfine contact interaction between the electron spin and the nuclei, and $H_B = \sum_{k=1}^{N} \sum_{l>k} \Gamma_{kl}(I_k \cdot I_l - 3 I_k^2 I_l^2)$ the intrabath dipolar coupling between nuclear spins. $S$ and $I_k$ denote the electron and the $k$-th bath spin operators, respectively. The nuclear spin value is set to $I_k = 1/2$. We focus on the limit of zero external field $B_0 = 0$ and assume that the bath is initially unpolarized. The FID time scale is $T_2^* = (N A^2/8)^{-1/2}$, where $A = (\sum_k A_k^2/N)^{1/2} \approx 10^{-4}$meV for typical GaAs QDs with $N = 10^9$. Time is measured in units of $1/A$.

Under ideal control assumptions, DD is implemented by subjecting the electron spin to sequences of instantaneous $\pi_n$ rotations along appropriate control axes $\hat{n}$, equally separated by the interval $\tau$. A variety of DD
protocols exist, based on both deterministic and randomized design. In cyclic DD, the control propaga-
tor is steered through a DD group of unitary operations \( G = \{ g_j \}, j = 0, 1, \ldots, |G| - 1 \), in a predetermined order, as opposed to randomized DD where the future control path is not known in advance. Changing \( g_j \) to \( g_{j'} \) requires the application of a DD pulse \( P_{i,j} = g_j g_{j'}^\dagger \). Thanks to the existence of a periodicity time scale \( T_c \), the analysis of cyclic DD has been mostly carried out within average Hamiltonian theory, upper performance bounds being determined by the dominant non-zero 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.

Periodic DD (PDD) is the simplest non-selective cyclic protocol, ensuring that the unwanted evolution is removed to first order in the ME at every \( T_n = n T_c \), \( n \in \mathbb{N} \), in the short \( T_c \) limit. For a single spin, PDD is based on the irreducible Pauli group \( G_p = \{ I, X, Y, Z \} \), which requires two-axis control sequences of the form \( C_1 = C_0 X C_0 Z C_0 X C_0 Z \), \( C_0 \) denoting a free evolution period. Improvement over PDD may be gained by symmetrized and/or concatenated design. Symmetric DD (SSD) guarantees that all odd terms in the ME are can-
celled, with \( T_c \) twice as long as PDD. Concatenated DD relies on a temporal recursive structure, so that at level \( \ell + 1 \) the protocol is \( C_{\ell+1} = C_1 C_2 Z C_2 X C_1 Z \). Here, we truncate the concatenation procedure at a certain level and repeat a periodic sequence, referred to as PCDD, after every \( 4^\ell \tau \). As repre-
sentatives among stochastic protocols, we consider naive random DD (NRD), which corresponds to uniformly random pulses over \( G \), and symmetric random path DD (SRPD), where a path to traverse \( G \) is chosen at ran-
dom and then symmetrized as in SDD.

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 \( F(T) = \text{Tr}[\rho_S(T)\rho_S(0)] \), where \( \rho_S(T) \) is the reduced den-
sity operator of \( S \) at time \( T \) starting from \( |\psi\rangle \) and tracking out the bath. For an unknown initial state, we in-
voke minimum pure-state fidelity \( F_m(T) = \min_i F(T) \). Analytical bounds on the expected fidelity decay for various DD protocols have been obtained for short evolution times, which calls for numerical analysis in the long-time regime. Simulations also make it possible to ex-
plor DD performance for values of \( T_c \) beyond the strict convergence domain of the ME, \( \omega_c T_c \ll 1 \), where the highest frequency component \( \omega_c \approx \sum_k |A_k|/|4 \sim NA/4 \). Let \( \sigma \) denote the power spectrum width of the environmental coupling, \( 2\sigma \approx (\sum_k A_k^2)^{1/2} \approx N \omega_c^{1/2} \). We shall consider \( \tau \sim 1/2\sigma \), thus \( T_c \gtrsim 4\tau \sim \sqrt{N} \omega_c^{-1} \). 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, 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 character-

istic time scale \( \tau_D \) 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 justi-
fied for practically relevant time regimes. All schemes lead to substantial enhancement of the electron spin co-
herence, PCDD2 showing the most dramatic improve-
ment. Although, for this system both SDD and PCDD2 remove \( H_{SB} \) to second order in the ME, the higher performance of PCDD2 reflects its superiority in reducing coherent error accumulation. The poor performance of NRD is expected, since its advantages over deterministic DD emerge only when \( G \) is large. Contrary to the case of closed systems, SRPD does not match PCDD2 in the relevant parameter range, confirming the fact that irre
ducible DD groups and slow baths are predicted to be especially favorable for concatenated control.

Motivated by the above results, we proceed with a more in-depth analysis of the PCDD protocol. Fig. 2(a) com-
pared the performance of two levels of concatenation, \( \ell = 2, 4 \), for different values of \( \tau \). As expected, the results deteriorate as \( \tau \) increases but, interestingly, PCDD4 becomes worse than PCDD2. Also interesting is the exponen-
tial fidelity decay of both protocols at long times. Fig. 2(b) illustrates, for each value of 2\( \sigma \) and different \( N \), the instant of time \( T_{90\%} \) where \( F_m(T) \) for PCDD2 reaches 90\%. The results are reasonably close to each other, par-
ticularly for larger \( N \), supporting their applicability up to realistic situations with \( N \sim 10^5 \). Lastly, we analyze the effect of \( H_B \), which becomes important once the co-
herence time is longer than \( \tau_D \). Let \( \Gamma_{kl} \) be uniformly random numbers in \([-\Gamma_0, \Gamma_0] \). To avoid demanding long time simulations, we increase \( \Gamma_0 \) manually up to values comparable to \( A_k \). The results are shown in Fig. 2(c), where a two dimensional 3 \times 5 QD with nearest neighbor
intrabath coupling is considered. PCDD₂ 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, 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. From a control standpoint, it indicates the difficulty again occurs along the direction of the dominant \( Z \) which we refer to as CPMG. For sufficiently large \( \tau \) and \( N \), PCDD₂ performance is 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, \( C_1 = C_X Y C_X Y = C_Y C_X \), identifying the outer \( y \) direction as the stable one. Similarly, for SDD and PCDD₂, 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 PCDD₂ 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 quasistatic bath approximation (QSA), let \( A_k = A, I = \sum_k I_k \), and \( M = I_2 \). After \( n \) CPMG cycles, the survival probability of the initial state \( |\Psi(0)\rangle = |I\rangle \otimes |I,M\rangle \) is given by

\[
\langle \Psi(0)|\Psi(2n\tau)\rangle^2 = 1 - \left( N^2 C^2 + B^2 \right) \tan^2 \theta \cos^2 2\theta, \tag{5} \]

where

\[
C = A \sqrt{|I - M|} (I + M + 1), \quad B = A (M + 1/2), \quad \tan \theta = d/\sqrt{1 - d^2}, \quad d = (B/\Omega) \sin(\sigma), \quad \Omega = B^2 + C^2. \]

Averaging over the nuclear spin bath and taking the limit of large \( n \) and \( N \), \( F \to F_{sat} = 1 - (1/2) \int dI dM P(I, M) (C^2/B^2) \tan^2 \theta \) with \( P(I, M) \sim (I/D \sqrt{2\pi D})e^{-I^2/(2D)} \) and \( D = N/4 \) for an unpolarized bath. In the limit of small \( \tau \), we obtain \( F_{sat} = 1 - (1/16)\tau^2 A^2 N = 1 - \tau^2/2T_z^2 \). For randomly distributed \( A_k \), \( A^2 N \to \sum_k A_k^2 \).

Fig. 3 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. The two curves superimpose, consistent with the fact that for large \( N \) the Overhauser field induced by \( I \) 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 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 QSA to accurately predict $F_{sat}$ for short $\tau$.

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, multipulse CPMG-DD has also been realized, not only in standard NMR and ESR experiments, but also in single solid-state centers\(^{22}\), 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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\begin{enumerate}
  \item J. M. Taylor \textit{et al.}, eprint cond-mat/0602470.
  \item J. Bergli and L. Glazman, eprint cond-mat/0609490.
  \item Up to appropriate parameter renormalization, the analysis is not qualitatively dependent upon this choice.
\end{enumerate}