

Multiple-Pulse Coherence Enhancement of Solid State Spin Qubits

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We describe how the spin coherence time of a localized electron spin in solids, i.e., a solid state spin qubit, can be prolonged by applying designed electron spin resonance pulse sequences. In particular, the spin echo decay due to the spectral diffusion of the electron spin resonance frequency induced by the non-Markovian temporal fluctuations of the nuclear spin flip-flop dynamics can be strongly suppressed using multiple-pulse sequences akin to the Carr-Purcell-Meiboom-Gill pulse sequence in nuclear magnetic resonance. Spin coherence time can be enhanced by factors of 4–10 in GaAs quantum-dot and Si:P quantum computer architectures using composite sequences with an even number of pulses.

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Understanding spin coherence in solids is one of the oldest problems in condensed matter physics [1,2], going back to the seminal pioneering work of Hahn [3], Feher [4], and others. Recently, there has been a resurgence of widespread interest in this problem in the context of quantum computation using spin qubits in semiconductors. In particular, a necessary condition for quantum computation is long qubit coherence time so that quantum error correction can be meaningfully implemented. For various proposed electron spin qubits in semiconductor nanostructures, coherence times on the order of 0.1 ms (or longer) are required as a necessary condition; assuming typical gating times of 100 ps, this leads to a Q factor of $\sim 0.1 \text{ ms}/100 \text{ ps} = 10^6$, which satisfies the current quantum error correction constraint ($\sim 10^{-4}$ – 10^{-6}). In fact, the only reason for the current experimental and theoretical interest in spin-qubit based scalable solid state quantum computer architectures (as opposed to charge qubits, which are easier to manipulate and read out) is the expected long spin coherence times ($\geq \mu\text{s}$) compared with charge (i.e., orbital) coherence times ($\leq \text{ns}$) in solids, making it impossible (possible) for quantum error correction schemes to work for solid state charge (spin) qubits. Understanding all aspects of electron spin decoherence in the solid state nuclear spin environment is important so that effective strategies can be developed to restore and/or enhance coherence by, for example, efficient pulse engineering.

It is, therefore, highly desirable to enhance the coherence time of spin qubits in semiconductor structures, particularly since semiconductor-based spin quantum computer architectures have considerable advantages in terms of scalability and fabrication. It has been known for a long time that various refocusing techniques using electromagnetic pulses (e.g., Hahn spin echo) substantially enhance spin coherence compared with the free induction decay. In this Letter we describe techniques that substantially enhance coherence of solid state spin qubits by using various composite sequences going beyond the simple Hahn spin echo. Demonstrating sufficient spin coherence in such a way is a necessary step in the development of

spin-based fault tolerant quantum computing. This Letter has direct implications for quantum spin memory in semiconductors and offers potential for coherent quantum logic gate design.

A particular impetus for our theoretical study comes from the beautiful recent experimental work by Petta *et al.* [5] who measured Hahn spin echo decay of electron spin qubits in GaAs coupled quantum-dot architectures, finding a spin coherence time $T_2 \sim 1 \mu\text{s}$ (consistent with our spectral diffusion theory of spin decoherence in GaAs quantum dot spin qubits [6]), substantially enhancing (by a factor of 100) the inhomogeneous spin dephasing time of $T_2^* \sim 10 \text{ ns}$ in the same system. In the current Letter, we investigate the potential for further coherence time enhancement by using a more complex pulse sequence originally used by Carr and Purcell a long time ago [7] in the context of magnetic resonance studies. Further coherence enhancement in GaAs quantum dots has recently been achieved using the multiple-pulse sequence suggested in this Letter, and the experimental results are in good agreement with our predictions [8]. Earlier experimental work on the ensemble of P donor electronic states in Si, of great interest to Si quantum computer architectures, reported very long ($\geq \text{ms}$) spin echo T_2 coherence times [9,10], which can be further enhanced in Si through isotopic purification. We find that composite sequences beyond the simple Hahn echo sequence could lead to substantial spin coherence enhancement in Si spin qubits also.

We consider a single localized electron spin in a semiconductor (e.g., GaAs quantum-dot, Si:P) interacting with the surrounding nuclear spin bath. The electron's spin polarization is preserved by applying a magnetic field due to the very large (a factor of ~ 2000) difference between the electron and nuclear Zeeman energy splittings. Virtual electron spin-flip transitions allow hyperfine-mediated interactions between nuclei, but such processes cause only a small visibility decay of refocused echoes that is sufficiently small for fault tolerant quantum computing at fields above $B \sim 1 \text{ T}$ [11] in GaAs quantum-dots for

which the effect is particularly strong. Echo modulation due to anisotropic hyperfine coupling is also sufficiently suppressed at fields above $B \sim 9$ T [12] in Si:P for which the effect is particularly strong. In such a situation (i.e., an applied magnetic field and low $\lesssim 100$ mK operational temperatures), it is now well accepted that the decoherence of a solid state electron spin qubit is dominated by the spectral diffusion process, where the flip-flop dynamics of interacting nuclear spins creates a temporally random non-Markovian magnetic field at the electron spin location leading to decoherence. We have earlier theoretically studied spectral diffusion effects on the Hahn spin echo introducing a quantum cluster expansion technique [6,13] that provides a formally exact treatment of the non-Markovian quantum nuclear dynamics in the large applied magnetic field limit. In this Letter we apply this technique to multipulse echo sequences to investigate the possible enhancement of spin coherence. We find that carefully designed multipulse sequences could considerably enhance solid state qubit coherence with the spin decoherence time T_2 [14] increasing strongly with the number of pulses. Our work has obvious implications for the design and operation of spin quantum computer architectures in semiconductor nanostructures. In particular, we find that our non-Markovian treatment leads to low order symmetry related cancellations when an even number of pulses are applied. We also predict that the logarithm of the echo, as a function of interpulse time (i.e., τ), scales as the square of the number of applied pulses in stark contrast to linear scaling of the stochastic theory [15].

Spins of interest (e.g., spins representing qubits) can be refocused, partially reversing the effects of their local magnetic fields, by applying to them sequences of rotating pulses (e.g., via resonance). Our analysis treats these pulses as ideal, making the approximating assumption that they perform exact rotations of some desired spin in an infinitesimal time without affecting the rest of the system. We consider the application of the Carr-Purcell-Meiboom-Gill (CPMG) [7] pulse sequence (equivalent, for our purposes with ideal pulses, to the Carr-Purcell sequence) to a solid state electron spin qubit. We represent this sequence as $(\tau \rightarrow \pi \rightarrow \tau)^n$ by which we mean that the system evolves freely for a time τ , we apply a π -rotation pulse perpendicular to the applied magnetic field, evolve the system freely for time τ again, and repeat this process n times. The $n = 1$ case is equivalent to the Hahn echo. Except for comparison with the Hahn echo, we treat the $n = 2\nu$ case of an even number of pulses. We find analytically (within relevant approximations) that even pulses are enhanced. A very general cluster expansion technique was introduced in Ref. [13], where it was successfully applied to Hahn echoes of Si:P and was recently verified in an independently developed theory [16]. We apply this technique now to the CPMG sequence. This analysis begins by writing the exact expression for the echo of the $(\tau \rightarrow \pi \rightarrow \tau)^{2\nu}$ sequence:

$$v_{\text{CPMG}}(\tau) = \frac{1}{M} |\{[U(2\tau)]^\nu U(\tau)[U^\dagger(2\tau)]^\nu U^\dagger(\tau)\}|, \quad (1)$$

with $U(t) = \exp(-i\mathcal{H}_+t) \exp(-i\mathcal{H}_-t)$, $\mathcal{H}_\pm = \mathcal{H}_B \pm \sum_n A_n I_{nz}/2$, and $\mathcal{H}_B \approx \sum_{n \neq m} b_{nm} I_n + I_m - \sum_{n \neq m} 2b_{nm} I_{nz} I_{mz}$ where the first summation is restricted to pairs of like nuclei (so that Zeeman energy is preserved when they flip-flop). The Tr operation in Eq. (1) traces over the states of the nuclear bath and M is the number of such states. The $\{A_n\}$ are hyperfine coupling constants between the spins of the electron and nucleus n ; typically $\max(A_n) \sim 10^6$ s $^{-1}$ (with $\hbar = 1$ units). Nucleus n has spin operators denoted with I_n and its gyromagnetic constant is γ_n . The $\{b_{nm}\}$ are dipolar coupling constants between nuclei in the bath; typically, $\max(b_{nm}) \sim 10^2$ s $^{-1}$ so that $b_{nm} \ll A_n$ which is important for the convergence of our cluster expansion. Details are given in Ref. [6].

Our cluster expansion is an approximation that is based upon the fact that we may exactly decompose Eq. (1) into a sum of all possible products of contributions from disjoint sets of nuclei:

$$v_{\text{CPMG}}(\tau) = \sum_{\substack{\{C_i\} \text{ disjoint.} \\ C_i \neq \emptyset}} \prod_i v'_{C_i}(\tau). \quad (2)$$

The full proof and necessary conditions for such a decomposition is provided in Ref. [6]. Each C_i denotes a set of nuclei and $v'_{C_i}(\tau)$ is called the ‘‘contribution’’ from this set. A contribution has the property that it can only be significant when interactions between nuclei in the set are significant and no part is isolated from the rest. We consider only local dipolar interactions in the current Letter, and thus contributions only arise when the nuclei in the set are spatially clustered together; hence we refer to these sets as clusters. Before we define the ‘‘cluster contribution,’’ $v'_{C_i}(\tau)$, we first define $v_C(\tau)$ as the solution to Eq. (1) when only considering nuclei contained in some set (or cluster) C . A cluster contribution may then be recursively defined by and computed using

$$v'_{C_i}(\tau) = v_C(\tau) - \sum_{\substack{\{C_i\} \text{ disjoint.} \\ C_i \neq \emptyset, C_i \subset C}} \prod_i v'_{C_i}(\tau), \quad (3)$$

subtracting from $v_C(\tau)$ the sum of all products of contributions from disjoint sets of clusters contained in C .

The decomposition of Eq. (2) is useful when making an approximation that assumes cluster contributions decrease with an increase in cluster size. We can then define the k th order of a cluster expansion by

$$v_{\text{CPMG}}^{(k)}(\tau) = \sum_{\substack{\{C_i\} \text{ disjoint.} \\ C_i \neq \emptyset, C_i \leq k}} \prod_i v'_{C_i}(\tau). \quad (4)$$

Because of the difficulty of iterating through all possible disjoint sets of clusters (below a given size), we make a further approximation by relaxing the constraint that they be disjoint sets. In this approximation, the logarithm of the

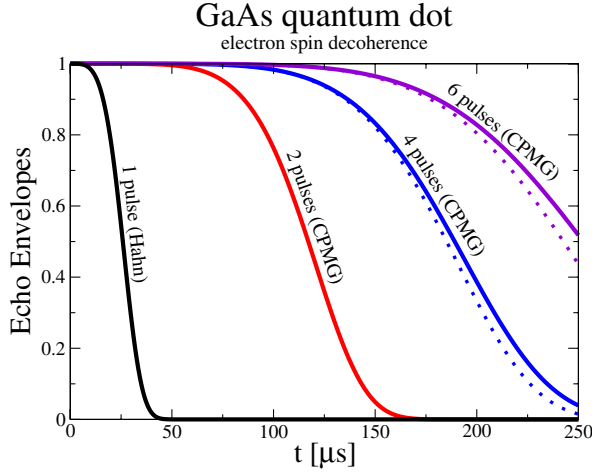


FIG. 1 (color online). Numerical results for echo envelopes of the Hahn and various CPMG pulse sequences (labeled by number of pulses, 2ν) for a quantum dot in GaAs with a 50 nm Fock-Darwin radius, 10 nm quantum well thickness, and applied magnetic field and thickness direction both along [001] in the lattice. Solid lines show convergent cluster expansion (exact) results while dotted lines show lowest order perturbative results (within the cluster expansion framework).

k th order result simply becomes the sum of all cluster contributions up to size k :

$$\ln[v_{\text{CPMG}}^{(k)}(\tau)] \approx \sum_{|C| \leq k} v'_C(\tau). \quad (5)$$

This approximation may be tested (or corrections made) as described in Ref. [6]. We wish to note the simplicity of implementing our approach using Eqs. (3) and (5) as compared to a very recent diagrammatic approach [17].

We show numerical results obtained by this method (up to visible convergence) in Figs. 1 and 2 for the coherence of a quantum-dot electron in GaAs and for the coherence of an electron bound to a donor in Si:P, respectively, comparing Hahn and CPMG echo envelopes. The effect of the coherence enhancement for even echoes is noticeable in both systems by comparing results of two-pulse CPMG versus the Hahn echo. These figures also show that coherence is indeed prolonged with an increase in the number of pulses. We note that these are plotted with respect to the total decay time, $t = 4\nu\tau$ (for the Hahn echo, $t = 2\tau$).

There are two perturbation theories that help to explain cluster expansion convergence via diminishing cluster contributions with increasing cluster size: the “interbath perturbation” (previously called dipolar perturbation [6]) that treats the coupling of the nuclei to the electron, and the time (or τ) expansion. Using λ to generically denote the (small) perturbation parameter of either theory, it was shown [6] that a contribution from a cluster of size k is $O(\lambda^k)$. The interbath perturbation is most applicable to nuclei that are coupled strongly to the electron (e.g., those closer to the center), and the time expansion is most applicable to nuclei with a weak coupling to the electron

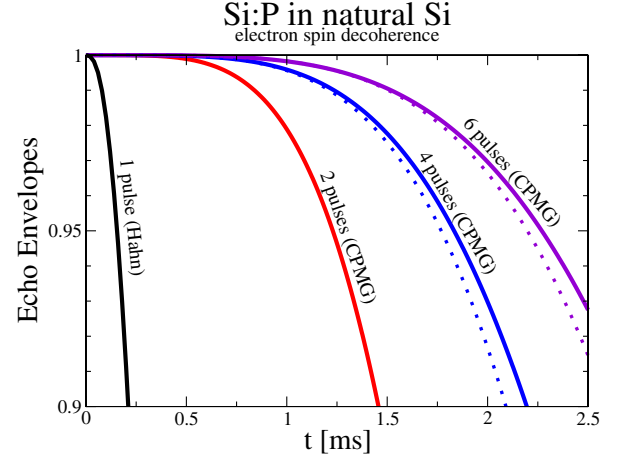


FIG. 2 (color online). Numerical results for echo envelopes of the Hahn and various CPMG pulse sequences (labeled by number of pulses, 2ν) for the Si:P donor electron in natural Si with the applied magnetic field along the [001] lattice direction. Solid lines show convergent cluster expansion (exact) results, while dotted lines show lowest order perturbative results (within the cluster expansion framework). The ordinate range is the first 10% (0.9 to 1) of echo decay where the cluster expansion is convergent for all pulse sequences shown.

(e.g., those further from the nuclei where lower energies result in slower evolution). The two theories work in tandem to produce overall convergence of the cluster expansion for a variety of physical systems.

By using the lowest order results of cluster contributions [Eq. (3)] with respect to either perturbation theory in the computation of the cluster expansion [Eq. (5)], we can see the important role played by these perturbation theories and gain useful insights. The lowest order result with respect to interbath perturbation explains the enhancement of even CPMG echoes relative to odd echoes as well as the scaling behavior with the number of pulses. Using $\lambda \sim b_{nm}/|A_n - A_m|$ as the perturbation parameter, the low order solution to Eq. (1) applied to a given cluster $v_C(\tau)$ behaves as $v_C(\tau) = 1 - O(\nu^2\lambda^4)$. For a cluster contribution [Eq. (3)] we then have $v'_C(\tau) = O(\nu^2\lambda^4)$, and from the approximate cluster expansion of Eq. (5),

$$v_{\text{CPMG}}(\tau) = \exp[-O(\nu^2\lambda^4)]. \quad (6)$$

The analogous expression for the Hahn echo has $O(\lambda^2)$; hence the extra symmetry of the time sequence in even-pulsed CPMG echoes leads to a cancellation of the lowest λ^2 order. For this reason, we predict an enhancement of even CPMG echoes over odd echoes when this lowest order result is valid (we find that it is generally valid as long as the cluster expansion itself is convergent as we observed and explained in our Hahn echo analysis [6]). Furthermore, Eq. (6) shows that the logarithm of the CPMG echo as a function of τ (but not t) scales with the number of pulses squared.

Similarly, the lowest order with respect to the time expansion gives $v_C(\tau) = 1 - O(\tau^6)$ so that

$v_{\text{CPMG}}(\tau) = \exp[-O(\tau^6)]$ for the short time behavior of the CPMG echoes. The analogous expression for the Hahn echo has $O(\tau^4)$ which is canceled [much like $O(\lambda^2)$ of the interbath perturbation] by symmetry in the time sequence of even-pulse CPMG echoes. This short time behavior is exhibited by the GaAs system but not the Si:P system (the reason relates to the shape of their electron wave functions and resulting cluster contribution statistics).

Applying the lowest order interbath perturbation represented by Eq. (6) to the GaAs quantum-dot system of Fig. 1 and fitting to these numerical results yields $\ln[v_{\text{CPMG}}(\tau)] \approx -\nu^2(\tau/55 \mu\text{s})^6 - \nu^2(\tau/31 \mu\text{s})^6$ where the first (second) term result from clusters of size two (three). Interestingly, 3-cluster contributions dominate as a consequence of the low order λ^2 cancellation. This approximation agrees with the $O(\tau^6)$ short time behavior that we anticipated. It also agrees with the exact results shown in Fig. 1 except as shown by the dotted lines which deviate, in a conservative way (predicting overly quick decoherence), from their corresponding exact results.

For the GaAs system of Fig. 1, the two-pulse CPMG echo prediction of $T_2 = 120 \mu\text{s}$ [14] is more than 4 times the Hahn echo $T_2 = 28 \mu\text{s}$, demonstrating the enhancement of even echoes. More generally, in the conservative perturbation approximation, $\nu^2\tau^6$ factor implies that τ effectively scales as $\nu^{-1/3}$; this means that the time between pulses, 2τ , must be shortened as one increases the number of pulses, 2ν , to yield the same degree of coherence. However, since the total pulse sequence time, t , is given by $t = 4\nu\tau$, we have $T_2 = \nu^{2/3} \times 120 \mu\text{s}$. Therefore, coherence is enhanced by applying multiple CPMG pulses, but one is experimentally limited by the minimum time allowed between pulses which is in turn limited by the time needed to apply each pulse.

Applying the lowest order interbath perturbation represented by Eq. (6) to the Si:P system of Fig. 2 (with $B \parallel [100]$) and fitting to these numerical results yields $\ln[v_{\text{CPMG}}(\tau)] \approx -\nu^2 f^2(\tau/230 \mu\text{s})^{4.3} - \nu^2 f^3(\tau/74 \mu\text{s})^{4.3}$ where f represents the fraction of Si that are the ^{28}Si isotope, the only isotope of Si with a net spin that can contribute to the spectral diffusion. The f^2 (f^3) dependent term comes from 2-cluster (3-cluster) contributions. Note that isotopic purification will make the most impact when 3-cluster contributions dominate over 2-cluster contributions because of their relative scaling with f . These two contributions become comparable when $f \approx 0.7\%$. This may be an important consideration for a cost-benefit analysis of isotopic purification of Si.

For natural Si ($f = 4.67\%$), $\ln[v_{\text{CPMG}}(\tau)] \approx -\nu^2(\tau/0.63 \text{ ms})^{4.3}$. This agrees with the exact results shown in Fig. 2 except as shown by the dotted lines which deviate, in a conservative way (predicting overly quick decoherence), from their corresponding exact results. For the two-pulse CPMG echo ($\nu = 1$) of natural Si, $T_2 = 2.5 \text{ ms}$, more than 4 times longer than the Hahn echo $T_2 = 0.6 \text{ ms}$, demonstrating, again, the coherence enhancement

of even echoes over odd echoes. In general, the above $\nu^2\tau^{4.3}$ factor implies that τ effective scales as $\nu^{-0.47}$; since $t = 4\nu\tau$, T_2 scales as $\nu^{0.53}$. This is comparable to, but not quite as good as, the $\nu^{0.67}$ scaling of T_2 for GaAs. For the natural Si system of Fig. 2, $T_2 = \nu^{0.53} \times 2.5 \text{ ms}$, in this conservative perturbation approximation.

We have developed a cluster expansion theory for calculating the enhancement of spin coherence in the context of multiple-pulse spin echoes in the limit of large applied magnetic fields. We find considerable enhancement of spin coherence in semiconductor qubits when even CPMG pulses are employed; T_2 increases fourfold in both GaAs and Si:P systems in comparing 1- (Hahn) and 2-pulse sequences, and increases with additional pulses. Using a conservative approximation for these GaAs and Si:P systems, T_2 scales with the number of pulses to the power of 0.67 and 0.53, respectively. We also report a crossover in the scaling of even CPMG echoes as a function of isotopic purification of Si in Si:P marking a transition between 3-cluster and 2-cluster dominance. This is a further consequence of the interesting symmetry related cancellation responsible for our important result that even-numbered pulse sequences are substantially more effective in extending or restoring spin coherence than odd sequences.

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