Keeping a spin qubit alive in natural silicon: Comparing optimal working points and dynamical decoupling

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There are two distinct techniques of proven effectiveness for extending the coherence lifetime of spin qubits in environments of other spins. One is dynamical decoupling, whereby the qubit is subjected to a carefully timed sequence of control pulses; the other is tuning the qubit towards ‘optimal working points’ (OWPs), which are sweet-spots for reduced decoherence in magnetic fields. By means of quantum many-body calculations, we investigate the effects of dynamical decoupling pulse sequences far from and near OWPs for a central donor qubit subject to decoherence from a nuclear spin bath. Key to understanding the behavior is to analyse the degree of suppression of the usually dominant contribution from independent pairs of flip-flopping spins within the many-body quantum bath. We find that to simulate recently measured Hahn echo decays at OWPs (lowest-order dynamical decoupling), one must consider clusters of three interacting spins, since independent pairs do not even give finite $T_2$ decay times. We show that while operating near OWPs, dynamical decoupling sequences require hundreds of pulses for a single order of magnitude enhancement of $T_2$, in contrast to regimes far from OWPs, where only about ten pulses are required.

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I. INTRODUCTION

Individual electronic and nuclear spins are promising candidates for realizing scalable quantum computing in solid state systems such as silicon. In addition, they offer a valuable test-bed for the experimental investigation of decoherence driven by quantum baths which typically comprise large numbers of spins surrounding a central spin qubit. Despite the large number of bath spins involved, accurate simulation of experimental coherence decays is computationally tractable since the bath can often be decomposed into independent contributions from many small sets or clusters of spins. In many cases, the dominant contribution to the decoherence dynamics arises from pairs of bath spins; in effect, from the magnetic noise due to the independent ‘flip-flopping’ of spin pairs. Contributions from larger clusters are usually only needed for high accuracy; however, there is also interest in cases where experimental evidence for the many-body or large-cluster nature of the bath might be most evident. For example, it was recently found that applying dynamical decoupling sequences of microwave pulses may enhance many-body correlations, relative to the independent spin-pair contributions.

A defining characteristic of a quantum bath is the back-action between the central system and environment, making the bath dynamics sensitive to the state of the central spin. A particularly striking example has been identified recently in spin systems with special coherence sweet-spots termed ‘optimal working points’ (OWPs) where theory and experiment found that coherence times can change by orders of magnitude with even small variations in applied magnetic field ($\sim 200$ G). A drastic change in back-action occurs via changes to the states of the central spin only: the change in external field has little direct effect on the bath spin-pair dynamics. However, it was found that usual cluster simulations using only pairs predicted infinite coherence times for the lowest order dynamical decoupling sequence (Hahn spin echo), at odds with experiment.

These findings motivate us to investigate here the interplay between dynamical decoupling and OWPs for a quantum bath system and in particular, to clarify where and to what extent, the independent spin-pair contribution dominates. Dynamical decoupling is one of the most established methods for extending coherence. It involves subjecting the qubit spin to a sequence of microwave or radio pulses. A wide variety of solid state spin qubits have been studied under dynamical decoupling control; these include Group V donors in silicon, nitrogen vacancy centres in diamond, GaAs quantum dots, rare-earth dopants in silicates, malonic acid crystals and adamantane.

Long-lived coherence is a key requirement for implementing fault-tolerant quantum computation as well as quantum memory. One way of extending coherence times $T_2$ in silicon is to use isotopically enriched samples in which the abundance of nuclear spin isotopes is significantly reduced. However, it is advantageous to retain nuclear spins for their potential use as long-lived quantum registers. It is thus also of practical importance to understand whether dynamical decoupling and OWP techniques may be advantageously combined for a quantum bath of nuclear spins. For donor electronic qubits in silicon, it is known that due to inhomogeneous broadening from naturally-occurring $^{29}$Si spin isotopes, there is a significant gap between the $T_2 \sim 100$ ms in natural silicon near an OWP and the $T_2 \sim 2$ s in the enriched samples.
isotopically enriched $^{28}\text{Si}$ with a low donor concentration at the same OWP\cite{13}. Also, dynamical decoupling may be useful when it is convenient to operate with the magnetic field close to but not exactly at the OWP.

It is well established that for dynamical decoupling to be effective, the pulse spacing $\tau = t/2N$ for a sequence of $N$ control pulses (where $t$ is the total evolution time) cannot exceed the correlation time of the bath noise. But the relevant correlation time, in turn, is an emergent property of the underlying microscopic quantum bath, comprised of typically $\sim 10^3 - 10^5$ significant clusters of spins of different coupling strengths, different sizes and subject to varying degrees of back-action from the central qubit. Therefore, to quantitatively simulate the response to dynamical decoupling, a realistic simulation of the combined system-bath dynamics at the microscopic level is important.

In this paper, we present quantum many-body simulations of the system-bath dynamics using the cluster correlation expansion (CCE)\cite{14,15} including contributions from clusters of up to 5 spins (CCE5) as illustrated in Fig. 1(b). We compare coherence decays at an OWP with regimes far from an OWP (denoted by ‘$\neq$ OWPs’) with a view to identify the optimal strategy for enhancing electronic spin coherence times of donor spins in natural silicon. Simple analytical expressions for the behavior of independent bath pairs coupled to the qubit aid understanding in all the regimes we consider.

In addition, our work fills an outstanding gap in understanding decoherence near OWPs. It was recently reported that near OWPs, numerical calculations of the Hahn spin echo ($N = 1$) involving only independent pair dynamics (CCE2) yield results in conflict with experiments: coherences decay initially, then after a short time, the decays stop\cite{14}. In the present work, we undertake the computationally more challenging CCE3 – 5 many-body calculations in order to clarify the origin of the measured coherence decays. We find that three-spin clusters suffice to give decays in good agreement with previously reported experimental results\cite{14}.

It is worth clarifying the physical meaning of the above-mentioned three-cluster result. It is not a matter of enlarging the quantum bath with additional nuclear spin clusters of the same size. As illustrated in Fig. 1(b), a three-spin cluster (blue) can be decomposed into three distinct flip-flopping pairs (each nuclear spin can contribute to more than one flip-flopping pair). Put simply, if all such three-clusters in a given, randomly generated set of impurities in a crystal are decomposed into the constituent flip-flopping pairs, an infinite decay time is obtained. If, however, the exact same configuration of spin impurities are aggregated into the ‘triangle’ structures illustrated in Fig. 1(b), the correct experimental behavior emerges. To our knowledge, there is no other example of a central spin system which so fully eliminates the pair-driven dynamics.

In contrast, for $\neq$ OWP regimes, Hahn decays for dipole-allowed transitions are well described by CCE2\cite{14,15}. However, for modest $N \lesssim 10$ pulse numbers there can be a large correction to CCE2 from clusters of 3 – 4 spins and for even $N$, but CCE2 still gives $T_2$ on the correct experimental timescale\cite{9}. For larger $N$, we find that CCE numerics including only independent pairs (CCE2) once again gives converged decays in all regimes whether in OWP or $\neq$ OWP regimes, so many-body calculations become progressively less important as $N \to \infty$.

The paper is organized as follows. In Section II we review the theory of decoherence and mixed donor qubits possessing OWPs. Our main results are presented in Section III, first for the Hahn spin echo, then for dynamical decoupling with moderate $N$. In Section IV, we provide an analysis on the suppression of the independent pairs contribution to decoherence as $N \to \infty$, as well as the magnetic field approaching the OWP: $B \to B_{\text{OWP}}$. Finally, we present large $N$ results in Section V and discuss important aspects of our results in Section VI.

For our dynamical decoupling calculations, we have chosen the Carr-Purcell-Meiboom-Gill (CPMG) sequence which applies a set of $N$ periodically spaced near-instantaneous pulses (CPMGN) as illustrated in Fig. 1(d)\cite{19,20}.

\section{II. DECOHERENCE AS QUBIT-BATH ENTANGLEMENT}

\subsection{A. Overlap of conditional bath evolutions}

For quantum baths, decoherence of a central spin system is understood in terms of entanglement between the system and the bath, as the combined system evolves under a total Hamiltonian given by:

$$\hat{H}_{\text{tot}} = \hat{H}_{\text{CS}} + \hat{H}_{\text{int}} + \hat{H}_{\text{bath}},$$

where $\hat{H}_{\text{CS}}$ is the central system Hamiltonian including all internal nuclear and electronic degrees of freedom, while $\hat{H}_{\text{bath}}$ is the bath Hamiltonian and $\hat{H}_{\text{int}}$ describes the system-bath interaction.

We start by placing the initial system state in a coherent superposition of an upper state $|i = u\rangle$ and a lower state $|i = l\rangle$ by applying a $\pi/2$ pulse: $|\Psi(t = 0)\rangle = \frac{1}{\sqrt{2}} (|u\rangle + |l\rangle) \otimes |\mathcal{B}(t = 0)\rangle$, where $|\mathcal{B}(t = 0)\rangle$ is a product state of the eigenstates of the non-interacting bath.

Under the joint system-bath dynamics, the initial state evolves into an entangled state:

$$\sqrt{2} |\Psi(t)\rangle = (e^{-i\hat{E}_{u,t} t} |u\rangle \otimes |\mathcal{B}_u(t)\rangle + e^{-i\hat{E}_{l,t} t} |l\rangle \otimes |\mathcal{B}_l(t)\rangle),$$

where $|\mathcal{B}_{u,l,t}(t)\rangle = \hat{T}_{u,l,t} |\mathcal{B}(t = 0)\rangle$ with unitaries $\hat{T}_{u,l,t}$ for each of the two system levels, and we have assumed no depolarization of the central states during the evolution. The measured temporal coherence decays $|\mathcal{L}(t)|$ can be simulated if one can accurately calculate the resultant overlap between the bath states correlated with the upper and lower qubit states:

$$|\mathcal{L}(t)| \propto |\langle \mathcal{B}_u(t)|\mathcal{B}_l(t)\rangle| = |\langle \mathcal{B}(0)|\hat{T}_{u,l}\hat{T}_{l,u}|\mathcal{B}(0)\rangle|.$$
Magnetic field, $B$ (T)

The spectra of donor spin systems such as arsenic, antimony or bismuth (pictured) are affected by strong mixing between the electron and host nuclear spin, at magnetic fields $B$ smaller or comparable to the hyperfine coupling $A$, allowing a richer behavior than unmixed electron spins. At particular field values termed optimal working points (OWPs), decoherence can be strongly suppressed; the arrows indicate the transitions with four of the most significant OWPs.

Since the initial bath states are usual trivial thermal spin states, the challenge is to obtain the corresponding unitaries $\hat{T}_u,l$ for extremely large baths ($\approx 10^4$ spins for convergent CCE).

OWPs are sweet-spots in $B$-field values where the two unitaries involving the upper and lower levels equalise: $\hat{T}_l \simeq \hat{T}_u$, occurring when $P_u \simeq P_l$\textsuperscript{[12]} This means that the state given by Eq. \textsuperscript{[2]} can be written as

$$\sqrt{2}\langle \Psi(t) \rangle = (e^{-iE_{u,t}|u\rangle} + e^{-iE_{l,t}|l\rangle}) \otimes \hat{T}_u(t)|B(0)\rangle,$$

with the product form preserved (i.e. the state is no longer entangled) and therefore decoherence is suppressed: $|\mathcal{L}(t)| \propto |\langle B(0)|\hat{T}_u(t)|B(0)\rangle| = |\langle B(0)|\hat{I}_z|B(0)\rangle| = 1$.

B. Interaction and bath Hamiltonians

We consider the situation where the central spin interacts with a spin-1/2 bath (e.g. $^{29}$Si impurities) primarily through the contact hyperfine interaction:\textsuperscript{[2]}

$$\hat{H}_{\text{int}} = \sum_a J_a \hat{S} \cdot \hat{I}_a,$$

where $\hat{S}$ represents the central electron spin, $J_a$ is the strength of the contact hyperfine interaction and $a$ labels the bath spins $\hat{I}_a$. The bath Hamiltonian consists of nuclear Zeeman terms and dipolar coupling among bath spins:

$$\hat{H}_{\text{bath}} = \hat{H}_D + \hat{H}_{\text{NZ}},$$

$$\hat{H}_{\text{NZ}} = \sum_{a} \gamma_N \hat{B} \hat{I}_a^z,$$

$$\hat{H}_D = \sum_{a < b} \hat{I}_a \hat{D}(r_{ab}) \hat{I}_b,$$

where $\gamma_N$ is the gyromagnetic ratio of bath spins and $\hat{D}(r_{ab})$ is the dipolar tensor coupling bath spins $a$ and $b$ separated by $r_{ab}$, with components $D_{ij}(r) = \mu_0 h \gamma_a^2 (\delta_{ij}/r^3 - 3r_i r_j/r^5)$, where $i,j = \{x,y,z\}$, $\delta_{ij}$ is the Kronecker delta and $\mu_0 = 4\pi \times 10^{-7}$ N A$^{-2}$ is the magnetic constant divided by $4\pi$.\textsuperscript{[17]}

C. Example central system with OWPs

The dephasing properties of the central spin qubit and bath are extremely well studied for the case of a spin-1/2 qubit.\textsuperscript{[2],[13],[50]} Good agreement with experiment has been achieved by cluster-based methods such as the CCE,\textsuperscript{[50]} which decompose the bath dynamics into products of contributions from clusters of 2, 3, or more interacting bath spins as illustrated in Fig. 1(b). But simple spin-1/2 systems do not have OWPs.

For the donor systems, however, the central spin Hamiltonian $\hat{H}_{\text{CS}} \simeq \omega_0 \hat{S}_z + A \hat{I}_b \cdot \hat{S}$, contains the usual

FIG. 1: (color online) (a) The spectra of donor spin systems such as arsenic, antimony or bismuth (pictured) are affected by strong mixing between the electron and host nuclear spin, at magnetic fields $B$ smaller or comparable to the hyperfine coupling $A$, allowing a richer behavior than unmixed electron spins. At particular field values termed optimal working points (OWPs), decoherence can be strongly suppressed; the arrows indicate the transitions with four of the most significant OWPs. (b) Coherences of the central electronic spins are dephased primarily by a surrounding quantum bath of clusters of 2, 3, 4 or more nuclear spins for convergent CCE. (c) Effect of dynamical decoupling (CPMG with even pulse numbers $N$) as $N \rightarrow \infty$. Plots $T_2^{(N)}(t)/T_2^{(1)}$ showing enhancement of the electron spin coherence time $T_2$ as a function of pulse number $N$, relative to the $N = 1$ Hahn echo value. We find that while dynamical decoupling far from the OWP enhances $T_2$ by an order of magnitude with about 10 pulses, in contrast, behavior close to an OWP is insensitive to dynamical decoupling for low $N$. For high $N$, the behaviors near and far from OWPs become comparable. (d) Illustrates coherence enhancement as $B \rightarrow B_{\text{OWP}}$ (the Hahn spin echo time $T_2^{(1)}$ is plotted). The OWP is for a bismuth donor in natural silicon, investigated experimentally in Refs. 12,13. Inset of (d): The CPMG dynamical decoupling sequence consists of the initial $\pi/2$ pulse, followed by the $-$ $\tau$ $-$ $\pi$ $-$ $\tau$ echo sequence repeated $N$ times. The coherence times in (c) are when the CPMG decays in Fig. 3 and the fits to the decays in Fig. 4 have fallen to 1/e. OWP results are for the $|14\rangle \rightarrow |7\rangle$ transition for which $B_{\text{OWP}} = 799$ G. For the field value near the OWP ($B = 795$ G) in (c), $T_2^{(1)} \approx 96$ ms while $T_2^{(1)} \approx 0.79$ ms in the $\neq$ OWP regime ($B = 3200$ G). The OWP curve in (d) was calculated using the analytical formula Eq. (7).
Zeeman term arising from the external magnetic field \((\omega = \gamma_e B, \text{where } \gamma_e \text{ is the electronic gyromagnetic ratio})\) and also a significant hyperfine coupling \((A)\) of the host spin \(I_h\) to the electron. For example, for the bismuth donor, \(I_h = 9/2\) and \(A = 1.475\) GHz, thus the mixing between host nuclear and electronic spins becomes substantial for \(B \gtrsim 0 - 0.3\) T as seen in Fig. 1(a).

Details of the mixing of states and the corresponding energy levels and transition probabilities were obtained analytically in Refs. 10-13. As illustrated in Fig. 1(a), there are a total of \(2(2I_h + 1)\) quantum states (e.g. 20 levels for bismuth with \(I_h = 9/2, 8\) for arsenic which has \(I_h = 3/2\)). At high magnetic fields, the Zeeman states \(|i⟩ = |m_S, m_{I_h}\rangle, m_S = ±1/2, m_{I_h} = -I_h, -I_h + 1, \ldots, I_h\), provide good quantum numbers. At lower fields, a new adiabatic set of states \(|\pm, m⟩ = (|±, m⟩)\) must be employed, since \(m_S\) and \(m_{I_h}\) are not good quantum numbers, but \(m = m_S + m_{I_h}\) is. The relation between the Zeeman basis and the adiabatic basis is given by:

\[
|+, m⟩ = \cos \frac{\beta_m}{2} \frac{1}{2}, m - \frac{1}{2} \rangle + \sin \frac{\beta_m}{2} \frac{1}{2}, m + \frac{1}{2} \rangle \\
|−, m⟩ = -\sin \frac{\beta_m}{2} \frac{1}{2}, m - \frac{1}{2} \rangle + \cos \frac{\beta_m}{2} \frac{1}{2}, m + \frac{1}{2} \rangle,
\]

(7)

for all states except the two states with \(|m| = I_h + S\) (i.e. states 10 and 20 in Fig. 1(a)) which remain unmixed at all fields. All other states mix with one another, forming doublets of constant \(m\). The transformation between the Zeeman basis and adiabatic basis is given by:

\[
|±, m⟩ \equiv |±, m⟩ = |±, m⟩ = (|±, m⟩) \rangle + (|±, m⟩) \rangle,
\]

which is the expectation value of the electron spin z-projection; it is no longer fixed at either \(m_S = 1/2\) or \(m_S = -1/2\) as for an unmixed qubit, but is instead a strongly field-dependent quantity \(P_1(B) \in [-1/2 : 1/2]\).

OWPs correspond to operating the qubit at particular \(B\) values where \(P_1(B) \simeq P_0(B)\). They provide a highly effective method of mitigating decoherence as illustrated in Fig. 1(d). Recently, their importance has been recognised for certain donors in silicon such as arsenic or bismuth. They were investigated theoretically in Refs. 10-12, and also experimentally13,14 extending the electronic spin coherence time from 0.5 ms to 100 ms for an ensemble of donors in natural silicon. OWP's have been extensively investigated for classical field noise,14,15,65 and in Ref. 65 dynamical decoupling was also studied.

III. RESULTS: LOW-ORDER DYNAMICAL DECOUPLING

A. Hahn spin echo (CPMG1)

Understanding of decoherence for such mixed systems nevertheless remains incomplete. In Refs. 11,12,59...
CCE2 calculations were carried out to obtain coherence times for allowed and forbidden electron spin resonance (ESR) transitions coupling different pairs of states \( |u\rangle \rightarrow |l\rangle \). These CCE2 calculations gave excellent agreement with experiment over most regimes. However, in the vicinity of the OWPs (where \( P_u \approx P_l \)), the CCE2 Hahn echo decay failed to converge and no decay was obtained other than initially, for a short time. Single-cluster spin free induction decay (FID), in contrast, gave finite decays at all magnetic fields.

In Ref. [12] an analytical expression estimating \( T_2 \) as a function of \( B \) was obtained, by inspection of the short time behavior of the form of the FID decays (which can be given analytically for each pair cluster):

\[
T_2(B) \approx C(\theta) \frac{\langle |P_u| + |P_l| \rangle}{|P_u - P_l|}.
\]  

(9)

The magnetic field dependence is wholly contained in the \( \langle |P_u| + |P_l| \rangle \) envelope, while the prefactor \( C(\theta) \) depends only on magnetic field orientation, the density of nuclear spins and their gyromagnetic ratio, but is independent of the strength of \( B^2 \).

This simple closed-form equation gave remarkable and accurate quantitative agreement with experiment in all regimes, spanning orders of magnitude changes in \( T_2 \), whether in the unmixed limit of a spin-1/2, or for certain transitions which are ESR forbidden at high fields, or at OWPs. The universal validity of Eq. (9) is worthy of discussion. Farther than about 100 G from the OWP, and where CCE is converged at CCE2, there is little difference between single-spin FID and Hahn echo decays; thus, it is not surprising that an equation obtained by considering the independent spin-pair contribution to FID can accurately model the Hahn echo experiments. Its validity within the OWP regions, however, is not yet fully understood. In particular, it remains unclear why a single \( C(\theta) \) prefactor suffices to accurately estimate experimental \( T_2 \), whether very far or very close to OWPs; and to describe different OWP regions (of which there are 16 for the bismuth system, with \( P_u,l \) values varying by close to an order of magnitude).

In the present work however, Eq. (9) is employed simply to simulate the expected experimental behavior. In Fig. 2 we present converged CCE calculations for the Hahn spin echo (CPMG1) near the OWP. We show that including three-spin clusters (CCE3) give converged results while qubit-bath correlations from only spin pairs (CCE2) give little decay (red line) except at short timescales. However, it can also be seen that all orders have similar short time behavior and that the inclusion of the three-clusters in effect recovers the short time behavior of the pair decays. Hence, the converged CCE agrees well with Eq. (9), which was derived from the early time decay of correlations from pairs and hence accounts for the experimental behavior observed in Ref. [13]. This is one of the key results of this work. Details of the CCE calculations are given in Appendix B.

**B. CPMGN with few pulses \( N \approx 2 - 20 \)**

In Fig. 3 we present comparisons of the response to dynamical decoupling near and away from an OWP by means of converged CCE calculations in both cases for \( N \) up to 16. One notable feature of the comparison is the insensitivity of OWP behavior to low numbers of pulses, in sharp contrast to the \( \neq \text{OWP} \) regime where there is a factor of 3 “jump” in \( T_2 \) from CPMG1 to CPMG2; while for larger \( N \), we find \( T_2 \sim N \) as seen in Fig. 1(c). However, OWPs are extremely effective at suppressing decoherence: for the point shown near the OWP, \( T_2 \approx 100 \text{ ms} \) already at CPMG1, while away from the OWP, to obtain comparable values, \( N \approx 100 \text{ pulses} \) are required.
as shown in Section V.

Previous studies, including a recent study of the ESR dynamics of a phosphorus donor at X-band frequencies (a system without OWPs for electron qubit decoherence)\[\text{[54x105]}\] observed a sharp increase in the coherence time between CPMG1 and CPMG2. The spin pair contributions were also suppressed, requiring many-body correlations for convergence and thus exposing the latter.\[\text{[54x116]}\] Nevertheless, CCE2 was shown to still give a reasonable approximation to the magnitude of the observed $T_2$ time, for both CPMG1 and CPMG2. In the case away from an OWP, the FID is very similar to CPMG1. This is in contrast to the OWP, where CCE2 gives no decay at all, while the FID gave decay curves comparable to converged CCE3 (and Eq. (9)). Thus, there is a drastic change from FID to CPMG1 at OWPs; in contrast, for regimes away from an OWP, there is little change between FID and CPMG1, but a strong enhancement for CPMGN with $N > 1$.

The quantum numerics do evidence a clear dependence of the pair contribution on pulse number $N$. For example, in the inset of Fig. 3(a), we have shown that, for a given field $B$ in the vicinity of the OWP, as $N$ increases to $N \approx 16$, the pair contribution once again gives significant decay. To suppress decay for $N = 16$ one must choose a value of $B$ even closer to the OWP. In fact this is one of the main findings of the present work: whether at OWPs or far from OWPs, our comparisons establish, two-state analytical pseudospin models of the qubit-bath dynamics.

### IV. ANALYSIS: PSEUDOSPIN MODELS

We now proceed to analyse correlations from independent pairs in order to obtain insight on the effect of dynamical decoupling near and far from OWPs. As described above, assuming pure dephasing justified by $\omega_0 \gg J_0$, for the case of a pair of bath spins, the joint system-bath dynamics reduces to a simplified two-state form for each of the two qubit states (upper and lower) and is governed by effective Hamiltonians:

\[
\hat{h}_{u,l} = \frac{1}{4}(\Delta_{u,l} \sigma_z + C_{12} \sigma_x),
\]

where $\Delta_{u,l}$ is a system-dependent detuning and $C_{12}$ is the strength of the intrabath secular dipolar interaction. The Pauli matrices are written in the non-interacting bath basis $\{|\uparrow\uparrow\rangle, |\downarrow\downarrow\rangle\}$, where $\uparrow$ ($\downarrow$) denotes spin up (down) for each of the two bath spins and we have ignored the trivial states $|\uparrow\downarrow\rangle$ and $|\downarrow\uparrow\rangle$ which cannot flip-flop. The detunings in our case are $\Delta_{u,l} = P_{u,l}(J_1 - J_2)$, where $J_1$ and $J_2$ are the hyperfine coupling strengths for the two bath spins. The system-bath evolution is then obtainable analytically from the eigenvalues $\omega_{u,l} = \frac{1}{4} \sqrt{(\Delta_{u,l})^2 + (C_{12})^2}$ and eigenvectors of the above effective Hamiltonians.

After preparing the initial qubit superposition, the CPMG pulse sequence can be summarized as $(T(\tau) - \pi - T(\tau))^N$, with final evolution time $t = N\tau/2$. The unitaries $T(\tau)$ represents free evolution and $\pi$ denotes the refocusing pulse which flips between $|u\rangle$ and $|l\rangle$: $|u\rangle |l\rangle + |l\rangle |u\rangle$ but leaves all other central states unperturbed. In the so-called pair-correlation approximation, the coherence decay is simply given by $\mathcal{L}(t) = \prod_k |\mathcal{L}_k^{(N)}(t)|$, where $|\mathcal{L}_k^{(N)}|$ is the decay contribution from the $k$-th spin pair and the product is over all spin pairs in the bath.

For CPMG1, if

\[
|B_{u,l}(t)\rangle = \tilde{T}_{u,l}^{(N)} |B(0)\rangle,
\]

we can write for the Hahn spin echo case (i.e. CPMG1):

\[
\tilde{T}_{u,l}^{(1)} = A_0 \hat{\sigma}_z - iA_{u,l} \cdot \hat{\sigma},
\]

where $A_u = (A_x, A_y, A_z)$ and $\hat{\sigma}$ is the vector of Pauli matrices in the bath basis: $\{|\uparrow\rangle, |\downarrow\rangle\}$. The $A_{u,l}$ components depend on time and can easily be given explicitly in terms of the pseudospin parameters; for example: $A_0(t) = \cos \omega_u t \cos \omega_l t - \sin \omega_u t \sin \omega_l t \cos (\theta_u - \theta_l)$; and $A_{u,l}(t) = -\sin \omega_u t \sin \omega_l t \cos (\theta_u - \theta_l)$, where $\theta_{u,l} = \tan^{-1}(C_{12}/\Delta_{u,l})$. In fact, the only term which is not invariant w.r.t. $u \leftrightarrow l$ is $A_y$ and thus $A_u = (A_x, -A_y, A_z)$.

The coherences $|\mathcal{L}^{(N)}(t)| \propto |\langle B(0) | \tilde{T}_{u,l}^{(N)} | \tilde{T}_{l}^{(N)} | B(0) \rangle|$ are obtained simply from $\tilde{L}^{(N)}(t) \equiv \tilde{T}_{u,l}^{(N)} \tilde{T}_{l}^{(N)}$.

For both CPMG1 and CPMG2, the unitarity of the evolution of upper relative to lower states is broken by a term $\propto A_y$. For CPMG1,

\[
\hat{L}^{(1)}(t) = \hat{1} - 2i A_y \hat{\sigma}_y \tilde{T}_{u,l}^{(1)}.
\]

We can consider higher sequences; since $\tilde{T}_{u,l}^{(2)} = \tilde{T}_{u,l}^{(1)} \tilde{T}_{l}^{(1)}$ and $\tilde{T}_{l}^{(2)} = \tilde{T}_{l}^{(1)} \tilde{T}_{u,l}^{(1)}$, we obtain for CPMG2

\[
\hat{L}^{(2)}(t) = \hat{1} - 4i A_y (A_x \hat{\sigma}_x - A_z \hat{\sigma}_z) \tilde{T}_{u,l}^{(2)}.
\]
We can easily obtain the coherence decay envelopes for CPMG1 in general, assuming pulse interval $\tau$:

$$\mathcal{L}^{(1)}(t = 2\tau)^2 = 1 - 4A_y^2A_0^2,$$

where we drop the $k$ label for convenience and $A_0 \equiv A_0(\tau)$, $A_y \equiv A_y(\tau)$. For arbitrary even numbers of pulses, CPMGN such that $N/2$ is an integer,

$$\mathcal{L}^{(N)}(t = 2N\tau) = 1 - \frac{2A_y^2}{A_y^2 + A_0^2} \sin^2 \left[ \frac{N\phi(\tau)}{2} \right],$$

where $\cos\phi(\tau) = A_0(2\tau)$. An equivalent expression was obtained in Ref. 9. Both expressions Eq. (16) and Eq. (17) are equally valid for both regimes (OWP and $\neq$OWP).

A. OWP limit

The only important difference between these regimes is that $\theta_u \rightarrow \theta_l$ for the approach to an OWP and $\theta_u = \pi - \theta_l$ for the spin away from the OWP. Alternatively, from the explicit expressions for the components of $A_{u,l}$, we see that the OWP condition is $|A_y| \rightarrow 0$. Thus, the suppression of qubit-bath correlations from pairs for OWPs is of the same order for CPMG1, CPMG2 or any other even-pulsed CPMG; for all bath spin pairs equally, the decay due to correlations from an independent pair uniformly tends to zero as $(A_{y,k})^2 \rightarrow 0$ for the $k$-th pair as $B \rightarrow B_{\text{OWP}}$.

The dependence on $N$ is entirely contained in the $\sin^2 N\phi(\tau)/2$ term. If $N\phi(\tau) \ll 1$ then increasing $N$ has a strong amplifying effect on the signal, while if $N\phi(\tau) \gg 1$, increasing $N$ simply results in oscillatory behavior. Near OWPs, from the expression for $A_0(2\tau)$, we see that if $\theta_u = \theta_l$, $\phi(\tau)/2 \approx (\omega_u + \omega_l)\tau$. Hence we only expect a response to dynamical decoupling if $\tau$ is sufficiently small (i.e. if $\tau \lesssim (\omega_u + \omega_l)^{-1}$).

B. $\neq$OWP limit

In contrast, for CPMG away from an OWP, the $A_y^2$ prefactor is still there, but is not small. The origin of the suppression of correlations from independent pairs for small numbers of pulses is more subtle to analyse with the pseudospin model. For CPMG2 ($\neq$OWP limit), we obtain:

$$\mathcal{L}^{(2)}(t)^2 = 1 - 64A_y^2A_0^2A_t^2.$$  

The large jump in $T_2$ from CPMG1 to CPMG2 was also analysed in Ref. 9. In the notation of Ref. 9, we see that for CPMG1, the decay envelope is of order $n_x^2$, while for CPMG2 it is of order $n_x^2n_z^2$, where $n_x = \sin\theta_u = \sin\theta_l$ while $n_z = \cos\theta_u = -\cos\theta_l$. Since the bath spans all angles $|\theta_{u,l}| = [0, \pi/2]$ one cannot a priori assume $\sin\theta_{u,l}$ is small. However, previous numerical studies support the idea that those spin pairs which have $|J_1 - J_2| \gg |C_{12}|$ (i.e. are strongly coupled to the central system) and therefore small pseudospin angles, dominate the Hahn echo contribution. For CPMG2, such strong-coupled spin pairs are strongly suppressed, and so $T_2$ becomes dominated by more weakly coupled spin pairs which are less effective in decohering the qubit.

V. RESULTS: CPMG$N$ WITH LARGE $N$

For large $N$, decays from independent pairs only (CCE2) are restored as well as the sensitivity to dynamical decoupling at OWPs. Even for $N = 16$ (Fig. 3) we see that the initial period of no decay $\mathcal{L}(t) \sim 1$ is prolonged. For larger $N$ (Fig. 4), the enhancement of coherence even at OWPs is clear, but however, the decays become extremely noisy. The noise can be attributed to the timescales of individual nearby spin clusters and the time interval between pulses. For these long coherence times ($\sim 1$s) there are very large numbers of resonances.
The CPMG sequence provides a means of amplifying noise from nearby clusters whenever pulse intervals become resonant with the characteristic cluster frequency. While this makes the CPMG a valuable technique for spin detection, large numbers of such resonances are undesirable if the aim is to protect qubit coherence. In contrast, far from OWPs, the decays for high \( N \) remain relatively smooth. While the noise at OWPs can be mitigated by ensemble averaging, this is likely introduce a considerable disadvantage in terms of single-shot operation of a single qubit.

VI. DISCUSSION

In sum, we have seen that a key difference between OWP and \( \neq \) OWP behaviors arises from the \( A_y^2 \propto \sin^2(\theta_u - \theta_l) \) prefactor which globally suppresses all independent pair contributions on the approach to an OWP, and accounts for the drastic effect at OWPs, but which is independent of \( N \) and has little effect far from OWPs. However, to analyse decays resulting from dynamical decoupling one must consider the remainder of the expression in Eq. (17), which reflects the dependence on \( N \). The ineffectiveness of dynamical decoupling near OWPs for small \( N \) can also be understood with an intuitive picture considering the relevant timescales of the system. For dynamical decoupling to be effective, the time interval between pulses \( (t/2N) \) must be shorter than to the correlation time of the bath \( \tau_c \). Since typical intra-bath interactions are at most a few kHz, \( \tau_c \sim 1 \) ms. Near the OWP, \( \omega^u \simeq \omega^l \) and \( \theta_u \simeq \theta_l \), so the frequency of the bath noise spectrum \( (\sim \omega_{u,l}) \) is appreciably higher than \( 1/\tau_c \) and thus dynamical decoupling becomes ineffective in extending the coherence time \( T_2 \gg \tau_c \). At short times and for high \( N \) however \( (t/2N < \tau_c) \), dynamical decoupling does protect the central system as evidenced for CPMG16 in Fig. 3(a) and higher \( N \) in Fig. 4. In contrast, dynamical decoupling is far more effective in extending \( T_2 \) away from the OWP and for relatively small \( N \) (Fig. 3(b)); although the pseudospin frequencies are comparable, the pseudospin fields are in opposing directions \( (\theta_u \simeq \pi - \theta_l) \), thus, the frequency of noise is much slower and becomes comparable to \( 1/\tau_c \sim 1/T_2 \).

Finally, it is important to note that for direct quantitative comparisons between our dynamical decoupling calculations and experimental ensemble measurements, inhomogeneous broadening due to \( ^{29}\text{Si} \) nuclei might also have to be factored in (see Appendix C).

VII. CONCLUSIONS

Understanding the interplay between optimal working points and dynamical decoupling involves understanding of the quantum behavior as a function of the two limits \( B \to B_{\text{OWP}} \) and \( N \to \infty \) corresponding to approaching an OWP and simultaneously increasing the number of dynamical decoupling pulses. An underlying question of physical interest is when decoherence is the result of the magnetic noise from independently flip-flopping pairs of spins and when consideration of the many-body nature of the quantum bath is important. The answer is of practical importance. For one, if decoherence is due to flip-flopping pairs, there are widely used models (such as the analytical pseudospin expressions in Section IV) which can be used to accurately calculate decays. Otherwise, more complex full many-body numerics become essential to simulate and fully understand experimental behaviors. The clear answer is that for low order dynamical decoupling, the elimination of correlations from independent pairs is so drastic at OWPs, that many-body numerics is almost indispensable for full understanding and accuracy. Even away from OWPs, it was shown that many-body effects make a large contribution for \( N \lesssim 10 \). However, once \( N \to \infty \) we find little difference between independent-pair and many-body results.

For practical applications, by solving for the many-body qubit-bath dynamics to calculate coherence times, one can hope to identify the best strategy for enhancing the coherence of donor qubits whilst still keeping the nuclear spin bath of naturally occurring silicon for its potential technological use. By operating near OWPs without dynamical decoupling, the maximum achievable \( T_2 \) is 0.1 s due to inhomogeneous broadening from the environmental nuclei. For isotopically purified samples in which the nuclear spin bath is nearly eliminated, \( T_2 \) at the OWP was measured to be about 1 s and is limited by decoherence mechanisms involving donor-donor interactions. Therefore, to bridge this single order of magnitude difference in \( T_2 \) at OWPs without resorting to isotopic purification, dynamical decoupling should be applied with at least a few hundred pulses. The effect of dynamical decoupling in extending coherence times near an OWP is marginal with a moderate number of pulses (up to \( N \sim 16 \)) in contrast to the usual regimes far from OWPs. For high donor concentrations, the timescale of donor-donor decoherence is comparable to the \( T_2 \) obtained in a nuclear spin bath, hence one might also want investigate suppressing those mechanisms with dynamical decoupling.

However, combining dynamical decoupling with OWPs is not without its drawbacks. As \( T_2, N \to \infty \), potentially many spins in a silicon bath may become resonant with the dynamical decoupling pulse spacing, resulting in very noisy decays in single central spin realisations. Although ensemble measurements are unaffected by this noise, this means that for single-qubit operations, if OWPs can be exploited, their extraordinary potential for coherence suppression may be sufficient.

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Appendix A: OWPs vs. clock transitions

The OWPs we consider correspond to suppression of decoherence in quantum spin environments. Similar points of interest are clock transitions or $df/dB = 0$ points, where decoherence arising from classical field noise is suppressed. For donor spin systems, the OWP is close to but not exactly at the $df/dB = 0$ point, and not all $df/dB = 0$ points are OWPs as shown in Ref. [12]. This is because the donor is coupled to the nuclear spin bath primarily via the electron, and the couplings between the host donor nucleus and bath nuclei are negligible compared to the electron-bath hyperfine interaction. It is easy to show that the fields at which $df/dB = 0$ satisfy

$$0 = P_u - P_I + \frac{\Delta m}{1 + \delta}, \quad (A1)$$

where $\delta = 10^{-4}$ is the ratio of the electronic to host nuclear gyromagnetic ratios and $\Delta m$ is the difference in quantum number $m$ between the upper and lower levels. Since $\delta \ll 1$, the OWP (where $P_u = P_I$) and $df/dB = 0$ points nearly coincide.

It is important to note that all of the transitions with OWPs couple two neighbouring avoided crossings. Selection rules were detailed in Ref. [14] but all such transitions have $\Delta m = \pm 1$ which implies that $\langle u | S_z | l \rangle = 0$. Thus, magnetic field fluctuations represent pure dephasing noise. One might also consider the possibility of creating a superposition of two states $|u\rangle$ and $|l\rangle$ at a single avoided crossing; for example, the superposition $|11\rangle + |9\rangle$ in Fig. [1]a), at the avoided crossing between these states at $B = 0.21T$. Although the $|11\rangle \rightarrow |9\rangle$ transition is never allowed, such a superposition might be created by a two pulse excitation from level $|10\rangle$. Both states are at zero energy gradient ($dE_{u,l}/dB = 0$) so coherences are to first order insensitive to dephasing noise; however, as shown in Ref. [14] in that case $\langle u | S_z | l \rangle \neq 0$ so magnetic fluctuations couple the states in the superposition and thus coherence is vulnerable to depolarisation by magnetic noise.

Appendix B: Cluster correlation expansion simulations

The CCE is a well-established method for accurately calculating the coherence decay $\mathcal{L}(t)$ of a central spin system in a quantum spin bath. In the CCE formulation, the spin bath is decomposed into groups of spins or “clusters” and the closed evolutions of clusters interacting with the central system are combined to approximate the exact coherence involving the entire spin bath.

The expansion is given by

$$\mathcal{L}(t) = \prod_{\mathcal{K}} \hat{\mathcal{L}}_{\mathcal{K}}(t),$$

$$\hat{\mathcal{L}}_{\mathcal{K}}(t) = \frac{\mathcal{L}_{\mathcal{K}}(t)}{\prod_{\mathcal{R} \subset \mathcal{K}} \mathcal{L}_{\mathcal{R}}(t)}, \quad (B1)$$

where $\mathcal{K}$ is a subset of the bath and the first product is over all subsets of the bath. The irreducible or “true” correlation term for cluster $\mathcal{K}$, $\mathcal{L}_{\mathcal{K}}(t)$, is recursively defined as follows. First, the coherence is calculated by exactly solving for the combined qubit-bath dynamics governed by the total Hamiltonian in Eq. [1], but including only those bath spins contained in $\mathcal{K}$. Second, the resulting $\mathcal{L}_{\mathcal{K}}(t)$ is divided by all correlation terms formed out of the proper subsets $\mathcal{R}$ of $\mathcal{K}$. The usefulness of the CCE becomes clear when truncating the expansion,

$$\mathcal{L}_{|\mathcal{K}|}(t) = \prod_{|\mathcal{K}| \leq k} \hat{\mathcal{L}}_{\mathcal{K}}(t), \quad (B2)$$

in which only clusters containing a maximum of $k$ bath spins are included. The lowest non-trivial order for nuclear spin diffusion is the pairwise correlation approximation ($k = 2$ or CCE2), given by Eq. [15], and we calculate up to $\mathcal{L}_{|\mathcal{K}|=5}(t)$. The coherence decay is often considered converged when $|\mathcal{L}_{|\mathcal{K}|}(t) - \mathcal{L}_{|\mathcal{K}|+1}(t)| \ll 1, \forall t$ and for our case this condition is satisfied for up to $k' = 3$.

Note that in the CCE method, the pure dephasing approximation is not required, and the interaction Hamiltonian in general includes terms which depolarize the states of the central system. Our CCE calculations include the $\hat{S}^- \hat{I}^- + \hat{S}^+ \hat{I}^+$ terms in the hyperfine interaction Hamiltonian Eq. [5], but we find that these give small corrections to the case when only $\hat{S}^z \hat{I}^z$ terms are included. This was expected due to the large mismatch between electronic and nuclear gyromagnetic ratios.

For our simulations, crystal sites of a cubic silicon superlattice were uniformly populated with $^{29}$Si nuclei ($I = 1/2$) with the natural fractional abundance of 0.0467 and with equal probability of spin-up and spin-down for the initial states. To calculate the electron-bath hyperfine couplings, we use the Kohn-Luttinger electronic wavefunction for the bismuth donor in silicon with an ionization energy of 0.069 eV. The total size of the spin bath is dictated by the spatial extent of the wavefunction which decays exponentially with distance from the donor site, and a superlattice of side length 160 Å with 10$^4$ impurities gave convergent coherence decays. Due to cubic decay of the dipolar interaction as the distance between a pair of $^{29}$Si spins is increased, it is not necessary to include all spin clusters in the calculation. At the lowest non-trivial CCE order (CCE2), spins separated by at most the 4-th nearest neighbor distance in silicon ($\sqrt{4} a_0/4$, where $a_0 = 5.43$ Å) gave convergent decays away from OWPs. To choose 3-clusters (i.e. including three bath spins), we loop over all sites in the crystal and add to each 2-cluster only those spins that are at
most separated by $\sqrt{11}a_0/4$ from any of the two spins in the 2-cluster. The same procedure was applied to choose higher-order clusters, by adding spins to clusters one order down. In Fig. 2 and Fig. 3, the numbers of 2, 3, 4, and 5-clusters were each of order $\approx 10^4$.

**FIG. 5:** (color online) Sharp $B$-field dependence of $T_2$ for various CPMG orders near an OWP. Inhomogeneous broadening from $^{29}$Si nuclei can be incorporated by convolving the decays with a Gaussian $B$-field distribution centred about $B$ (here centred about 797 G) and with standard deviation $w \approx 2$ G (dashed line). For a donor concentration of $3 \times 10^{15}$ cm$^{-3}$, $T_2$ is limited by donor-donor processes at about 300 ms.\(^{29}\) The $T_2$ lines were calculated for bismuth donors using the CCE up to 3rd order and for $B \parallel [110]$. The OWP under investigation is shown in red at 799 G.

**Appendix C: Effect of inhomogeneous broadening**

For direct quantitative comparisons between our dynamical decoupling calculations and experimental ensemble measurements, inhomogeneous broadening might also have to be considered. As shown in Fig. 5 for various orders of CPMG, $T_2$ varies sharply with magnetic field over a few G near an OWP. Inhomogeneous broadening of $B$ due to $^{29}$Si impurities has a FWHM of about 4 G in natural silicon and may therefore need to be included in the calculation in order to predict the shape and rate of experimental decays near OWPs. The broadening can be simulated by convolving the decays $\mathcal{L}_B(t)$ with a Gaussian magnetic field distribution with standard deviation $w \approx 2$ G:

$$D_B(t) = \frac{1}{w\sqrt{2\pi}} \int e^{-\frac{(B-B')^2}{2w^2}} \mathcal{L}_B(t) dB'.$$

(C1)

Depending on the donor concentration, donor-donor processes may also need to be included. For example, for a donor concentration of $3 \times 10^{15}$ cm$^{-3}$, $T_2$ near an OWP is limited by direct flip-flops of the central donor with other donors in the ensemble.\(^{29}\) The measured $T_2$ in an isotopically purified sample ranges from $0.2 - 2$ s ($T_2$ for a donor concentration of $3 \times 10^{15}$ cm$^{-3}$ is 300 ms). Therefore, care should be taken to include donor-donor processes very near the OWP (within about 1 G), where nuclear spin diffusion coherence times are comparable to those of donor-donor processes.
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