Dynamical decoupling based quantum sensing: Floquet spectroscopy

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Abstract

It is possible to sense the internal dynamics of individual clusters of nuclear spins by observing the coherence decay of a nearby electronic spin: the weak magnetic noise is amplified by a dynamical decoupling sequence of microwave pulses, though it remains challenging to relate experimental traces to underlying atomic-scale structure. For periodic dynamical decoupling control, we show that the Floquet eigenphases and eigenstates of the system provide the most natural framework for data analysis and fingerprinting of complex spin environments, offering more accuracy than the frequencies of either the static problem or of average Hamiltonian models. This approach is fully general for any temporally periodic protocol and sensor, but is here tested on NV centres and systems for which the quantization axis varies as a function of applied magnetic field; for instance, electron donors in silicon, such as arsenic and bismuth, exhibit regimes of high sensitivity of decoherence with respect to magnetic fields, so represent promising potential sensors.

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Central-spin systems are seen as key to the development of a set of new quantum technologies, not only as a source of versatile qubits for quantum information, but also because they underpin a new generation of quantum sensors, for magnetometry and atomic scale characterisation of the environment.

In particular, the electronic spin of single NV− centres in diamond exhibit long coherence times (even at room temperature) and spectral properties which render them uniquely suitable for both high-precision magnetometry and spin sensing. They have already generated a series of ground-breaking demonstrations of sensing and control at the single electronic and single nuclear spin level. Other types of defects in diamond such as silicon vacancies are also currently showing much promise. But in addition, it has also been shown that dynamical decoupling control of the central electron, applied through a train of microwave frequency pulses, provides a powerful tool for amplifying the detection of the weak signal from nearby spin clusters. Dynamical decoupling sequences have already led to several experimental detections of few or single proximate nuclear spins. The internal dynamics of a single pair of nuclear spins was recently experimentally detected and characterized using the electronic spin of a nitrogen vacancy centre in diamond. There is every prospect that such studies are test-beds which will lead in future to development of fully-fledged NMR on the atomic scale, including single molecule NMR.

However, it remains challenging to accurately and reliably extract experimental values for the interactions strengths, both internal to the cluster and between cluster spins and sensor spin and hence to relate these to the atomic scale structure (the spins’ positions on the crystal lattice). For both single nuclear spins and clusters, the measured decays of the sensor spin show sharp dips in the coherence at frequencies which must be related to the dynamics of the environment spins. For single spins or pairs the dynamics is essentially two-state and pseudo-spin models (see e.g. [11,12]) have proved very useful for quantitative simulation and modelling. However, extracting atomic-scale information accurately is still challenging. Furthermore, these methods do not scale to many-body clusters (in the sense of Fig. 1(b) where interactions between spins are non-negligible and affect the dip positions).

FIG. 1: (a) The coherence between “up” |u⟩ and “down” |d⟩ states of an electronic sensor spin is a sensitive probe of weak noise from nearby nuclear spins. This is amplified by dynamical decoupling control such as CPMG-N, leading to observed “dips” in coherence at well-defined frequencies. (b) The dynamics of the detected spins depends on u, d through the z projection cos βu,d of the sensor spin. For NV center sensors this arises from the effect of the crystal field; or for potential future sensors, from mixing with the host nuclear spin (purple arrow, shown) for Si:Bi or Si:As, or from spin-orbit effects for Si vacancies. For periodic control, instead of ωu,d, the frequencies of the static system, or from averaged Hamiltonians, the Floquet eigenphases provide more accurate, u, d independent, frequencies for analysis. (c) It is is challenging to differentiate between (i) independent pairs of spins and (ii) many-body effects from an equivalent interacting cluster. Unlike current geometric models, Floquet spectroscopy is not restricted to single spins or spin pairs and can be applied also to analysis of larger, correlated spin clusters.
Fortunately, many typical dynamical decoupling (DD) sequences involve repetition of a basic pulse sequence: in effect temporally periodic driving is applied. In these cases, Floquet’s theorem provides a canonical form for the solutions and has found wide applicability in various branches of quantum physics since 1962\cite{16} especially in light-matter interactions with continuous driving and multi-photon atomic physics. However, the Floquet formalism can equally be applied to trains of short pulses, interspersed by free evolution, such as the DD sequences. The basic idea is as follows: in order to understand quantum behavior in the presence of strong AC-driving, rather than to analyse the eigenstates of the static Hamiltonian which are appropriate only in the perturbative limit of weak driving, one should instead analyse the dynamics in terms of the eigenstates of the one period time-evolution propagator\cite{10} Floquet’s theorem is the temporal analogue of the well known Bloch theorem and applies to a periodic Hamiltonian for which $\hat{H}(t+T_P) = \hat{H}(t)$. Typically, this corresponds to a periodically driven system where:
\begin{equation}
\hat{H}(t) = \hat{H}_0 + V(t) \text{ where } V(t) = V(t + T_p),
\end{equation}
the potential varies periodically. In that case one may write solutions to the Schrödinger equation in the form:
\begin{equation}
|\Psi_i(t)\rangle = \exp(-i\epsilon_i t)|\Phi_i\rangle
\end{equation}
where $|\Phi_i(t)\rangle = |\Phi_i(t + T_p)\rangle$ has the same periodicity as the potential ($l = 1, ..., D$). These are the so called quasi-energy states (QES) and $\epsilon_i$ is the quasi energy. The QES form a powerful basis for the full solution of a wide range of driven quantum systems.

However, for problems (such as our present study) where we require only “stroboscopic” knowledge of our system (i.e. read-out once every period $T_P$), the solution is even simpler. We can obtain Floquet phases/modes simply as the eigenvalues/eigenstates of the one-period unitary evolution operator $\hat{T}(T_P,0)$. The Floquet modes obey the relation:
\begin{equation}
|\Phi_iF(t = T_P)\rangle = \exp(-iE_{\ell}(T_P))|\Phi_i\rangle
\end{equation}
where now $E_{\ell}(T_P)$ is the eigenphase (the Floquet phase).

In our case specifically, we can obtain Floquet phases/modes simply as the eigenvalues/eigenstates of $T_{u,d}^{(2)}$ (the basic CPMG-2 periodic sequence, for which $T_P = 4\tau$, see Appendix A) and for instance:
\begin{equation}
T_{u}^{(2)}|\Phi_{u}^{\ell}\rangle = e^{-iE_{\ell}}|\Phi_{u}^{\ell}\rangle
\end{equation}
where $i = u, d$ denotes the state of the sensor spin. These eigenphases characterise the time-evolution of the system. One notable point is that the Floquet phases are not dependent on $t$. Since:
\begin{equation}
T_{u}^{(2)} = T_{u}^{(1)}T_{d}^{(1)}; \quad T_{d}^{(2)} = T_{d}^{(1)}T_{u}^{(1)}
\end{equation}
the two operators can be constructed from products of the same matrices, it is a textbook result to prove they have the same eigenvalues but not the same eigenstates.

For the static approach, one obtains the eigenvalues of $H_{tot}$; however, a standard approximation, given that the energy-scale of the sensor spin is large compared with the bath is to define state dependent Hamiltonians $H_i \equiv \langle i|H_{tot}|i\rangle$ with eigenvalues $\omega_u, \omega_d$. The independence of $E_{\ell}$ on the spin sensor state is an advantage although one should note that the Floquet phases $E_{\ell}(\tau)$ depend on $\tau$, whereas the $\omega_u, \omega_d$ do not.

Once the Floquet phases and modes are obtained for $T_{u,d}^{(2)}$, one can immediately obtain the dynamical evolution for arbitrary numbers of pulse pairs $N_p = 2N$. Projecting the initial bath states onto the eigenmode basis $|B(0)\rangle = \sum c_{j|l}|\Phi_{j|l}\rangle$ where $c_{jl} = \langle \Phi_{j|l}|B(0)\rangle$ for $j = u, d$ and thus:
\begin{equation}
|B_j(t = N_P4\tau)\rangle = \sum_l e^{-iN_P\epsilon_l}c_{jl}|\Phi_{j|l}\rangle.
\end{equation}
Hence time-propagation of a quantum state becomes analogous to the static case, but with a characteristic frequency $|\epsilon_l| = |E_{\ell}(\tau)|/4\tau$, which is equivalent to the quasi-energy.

![Figure 2](image-url)

**FIG. 2:** (a) usual **Geometric** approach: under CPMG-N control ((b)) the detected spins represent two-state systems which precess about effective magnetic field, depending on the “up” $|u\rangle$ and “down” $|d\rangle$ states of the probe spin. The coherence dips are understood by following the divergence and growing angle between these spins with increasing $N$. (c) **Spectroscopic** picture. The dips in coherence occur at avoided crossings of the Floquet eigenstates. Both the position and contrast of the decoherence dip is related to the curvature of the crossing. This is characterised by the splitting between the states $\delta$ and the deviation from the early $\tau \rightarrow 0$ linear evolution. The early time evolution (the $\epsilon_{\ell}$ quasi-energy) gives the the dip position $\tau_{\text{dip}}$ for average Hamiltonian theory; the precise dip position is given by $E_{\ell}^F(2\tau_{\text{dip}}) = \frac{\pi}{4}$ and not the $E_{\ell}^F(\tau_{\text{dip}}) = \pi$ one might infer from geometrical models. (d) The the dip contrast increases with the degree of curvature of the crossing, i.e., $E_{\ell}^F(\tau_{\text{dip}}) - E_{\ell}^F(\frac{\pi}{2})$. 
The decoherence is given by:
\[ \mathcal{L}(\tau) \propto \langle B_d(N_p 4\tau) B_u(N_p 4\tau) \rangle = \sum_{l,v} e^{-iN_p(\xi_l(\tau)-\xi_v(\tau))} c_{dl}^*(\tau) c_{ul}(\tau) \langle \Phi_{dl}^F(\tau) | \Phi_{ul}^F(\tau) \rangle \]  
\hspace{1cm} (7)

In the Floquet picture, a minimum (or dip) in coherence occurs because \( \langle \Phi_{dl}^F(\tau) | \Phi_{ul}^F(\tau) \rangle = 0 \), thus whenever a pair of important Floquet states becomes orthogonal. This determines the dip positions regardless of the dimensionality of the system. The Floquet state overlaps exhibit similar temporal periodicity to the eigenphases. This can be shown analytically for the one spin and pseudospin models as seen below. At \( \tau = 0 \) the two \( u, d \) Floquet basis states are aligned; with increasing \( \tau \) they evolve away from each other and once per cycle attain orthogonality.

Defining a quasienergy \( \epsilon_l \equiv \xi_l/4\tau \), we propose that in general, such a periodic minimum in overlap means the fundamental pulse interval which yields a dip is:
\[ \tau_{\text{dip}} \simeq \frac{2\pi}{\epsilon_l - \epsilon_{l'}} \]  
\hspace{1cm} (8)
related to the difference between two Floquet quasienergies, with further harmonics at \( \tau \simeq 3\tau_{\text{dip}}, 5\tau_{\text{dip}} \ldots \) since the Floquet system periodicity is modulo \( 2\pi \). For the two-dimensional one-spin pair problem, where the eigenphases occur in \( \pm \epsilon \) pairs, \( \tau_{\text{dip}} \simeq \frac{\pi}{2} \).

Below we first investigate the behaviour for single-spin or single pseudospin models where we can present closed form analytical expressions and can compare directly with geometric approaches e.g.[13][22]. We exploit these approximations, but also present comparisons with numerical diagonalisations of the full joint central spin-cluster Hamiltonian (without assuming state-conditionial dynamics).

This investigation demonstrates that Floquet theory offers a completely new perspective regarding the origin of the dips: our key finding is that each dip is associated with an anti-crossing of a pair of Floquet eigenstates; and that the shape and depth of the dip is directly related to the curvature and splitting of the anti-crossing. In addition, the Floquet method is equally valid for larger interacting clusters, including the higher-dimensional 3-cluster problem depicted in Fig.1(c), so it is in many-body sensing that Floquet analysis may prove essential in future.

**Pseudospin models** Both pair flip-flop dynamics as well as single spin-dynamics (in systems like NV centres where a crystal field leads to non trivial one-spin dynamics) can be approximated by a two-state Hamiltonian. We term this the pseudospin model, on the understanding that for the single-spin detection by NV-centers case, this is a genuine spin. For the detection of the pair-dynamics of a flip-flopping cluster it is a pseudospin.

The evolution of the nuclear spin or pseudospin is conditional on the state \( i = u, d \) of the probe and reduces to precession about an effective magnetic field:
\[ H_i = \frac{1}{2} \mathbf{h} \cdot \mathbf{\Sigma} = \frac{1}{2}(X\sigma_x + Z_i\sigma_z) \]  
\hspace{1cm} (9)
where \( \sigma_x, \sigma_z \) are Pauli matrices in the usual spin basis; in the pseudospin case of course, we have \( |\uparrow\rangle \rightarrow |\uparrow\rangle \) and \( |\downarrow\rangle \rightarrow |\downarrow\rangle \).

The \( X, Z_i \) depend on the physical system (see Appendix for details); but for NV centers \( h^u \approx (\omega_x, 0, A u + \omega_z) \) while \( h^u \approx (\omega_x, 0, \omega_z) \) where \( \omega_x \) is the Larmor precession and \( A u \) the parallel component of the hyperfine interaction. For spin pair-sensing on the other hand, \( h = \frac{1}{2} (C_{12}, 0, \cos \beta i \Delta J) \).

\[ \sum \left| \mathcal{B}_l(\tau) \right|^2 \left| \mathcal{B}_u(\tau) \right|^2 \]  
\hspace{1cm} (10)

The eigenvalues of \( H_i \) (the pseudospin frequencies) are:
\[ \omega_i = \pm \frac{1}{2} \sqrt{X^2 + Z_i^2} \]
and the orientation of the effective field is \( \theta_i = \)
arctan(X/Z).  

**Floquet states** For a two-state problem, the time-evolution operator is given by:

\[
T_i^{(0)}(t) = R_y(\theta_i)R_z(2\omega_i t)R_y(\theta_i)
\]

where \(R_y, R_z\) are rotation matrices. The propagator \(T_i^{(2)}(t = 4\tau)\) given by Eq.\[A5\] can be diagonalised and the corresponding Floquet states can also be represented on the Bloch sphere. In Appendix B we show the Floquet states are real, thus:

\[
T_i^{(2)}(t = 4\tau) = R_y(\theta_F)R_z(2\mathcal{E}_F)R_y(\theta_F)
\]

which may be compared with Eq.\[11\]. We denote \(\theta_{u,d}\) the Floquet angles. Since \(T_i^{(2)}(t = 4\tau)\) is unitary, the eigenvalues \(\lambda^\pm\) are complex so we can write \(\lambda^\pm = \exp \pm i\mathcal{E}_F\). This defines the Floquet phases \(\mathcal{E}_u^F(\tau) = \mathcal{E}_d^F(\tau) \equiv \mathcal{E}_F(\tau)\) which are state independent.

The solution for arbitrary time propagation (for arbitrary number of pairs of pulses \(N_p\)) where \(N = 2N_p\) is obtained by scaling the eigenphases, thus

\[
T_i^{(2N_p)}(t = 4N_p\tau) = R_y(\theta_F)R_z(2N_p\mathcal{E}_F)R_y(\theta_F).
\]

The decoherence is then obtained as:

\[
\mathcal{L}(t = 4N_p\tau) = 1 - 2 \left[ \cos^2 \left( \frac{\mathcal{E}_F(\tau)/2}{2} - \cos^2 \left( \frac{\mathcal{E}_F(\tau/2)}{2} \right) \right) \right] \sin^2 \left[ N_p\mathcal{E}_F(\tau) \right]
\]

In order to obtain both the Floquet eigenphase \(\mathcal{E}_F(\tau)\), as well as its half-period value \(\mathcal{E}_F(\tau/2)\) in Eq.\[15\] one may use \(\cos \mathcal{E}_F(t) = \cos(2\omega_dt)\cos(2\omega_dt) - \sin(2\omega_u t)\sin(2\omega_d t)\cos(\theta_u - \theta_d)\) with \(t = \tau\) or \(t = \tau/2\).

Equ.\[13\] is a key result of this work. It is instructive to compare this form with equations derived previously, using geometrical arguments and the precession about effective pseudofield.\[B19\]

\[
\mathcal{L}(t = 4N_p\tau) = 1 - 2\hat{h}_u \times \hat{h}_d \sin^2 \omega_u \tau \sin^2 \omega_d \tau \frac{\sin^2(N_p\alpha)}{\cos^2(\alpha/2)}
\]

with

\[
\cos \alpha = \cos(2\omega_u t) \cos(2\omega_d t) - \hat{h}_u \cdot \hat{h}_d \sin(2\omega_u t) \sin(2\omega_d t).
\]

By comparison with Eq.\[15\] we identify \(\alpha\) above with the Floquet phase \(\mathcal{E}_F(\tau)\); with appropriate manipulation, we can also show that the expressions are equivalent and both yield \(\mathcal{L} = 1 - 2F(\tau)\sin^2(N_p\mathcal{E}_F)\). However, they encompass quite different physical perspectives, as summarised by Fig.\[2\] in terms of precessions on the Bloch sphere in one case, or in terms of avoided crossings in the other; and they suggest somewhat different position for the dips. From Eq.\[14\] one could be led to infer that dips occur where \(\alpha = \pi\), since

\[
\lim_{\alpha \to \pi} \frac{\sin^2(N_p\alpha)}{\cos^2(\alpha/2)} \to N_p^2.
\]

Thus there are two possibilities for the dip positions:

\[
\cos^{-1}(\alpha/2) \text{ divergence- dips at: } \mathcal{E}_F(\tau/2) = \pi/2
\]

Floquet expression-dips at: \(\mathcal{E}_F(\tau/2) = \pi/2\)

which in general yield different positions for \(\tau_{\text{dip}}\). In the Flo-
The width and shape of the dip is also determined by the Floquet anti-crossing. For low enough $N$, the eigenvalue splitting at the anti-crossing determines the depth of the dip. If $\alpha \equiv \mathcal{E}F = \pi$ the dip depth is zero.

Instead, it is the condition $\mathcal{E}F(\tau_{\text{dip}})/2 = \pi/2$ which rigorously select the dip positions. We show in the Appendix, that this is the precise condition for orthogonality of the Floquet states.

Fig. 2 clarifies the difference. The $F(\tau)$ prefactor represents the curvature of an avoided crossing where the two eigenphases approach (but do not reach if a dip exists) $|\mathcal{E}F(\tau_{\text{dip}})| = \pi$. It is the difference $\pi - \mathcal{E}F(\tau_{\text{dip}}) = \delta$ which is significant; since $F(\tau_{\text{dip}}) = 1$ and $\mathcal{E}^+ - \mathcal{E}^- = 2\delta$, the eigenvalue splitting at the anti-crossing determines the depth of the dip:

$$\mathcal{L}(t = 4Np\tau_{\text{dip}}) = 1 - 2 \sin^2(Np\delta). \quad (16)$$

The width and shape of the dip is also determined by the crossing. For low enough $Np$, the central height increases as $(Np\delta)^2$; however, for larger $Np\delta \gtrsim \pi/2$, the $\sin^2(Np\delta)$ prefactor simply supersedes an oscillatory background of “sidebands” on the $F(\tau)$ envelope and the dip shape is no longer strongly dependent on the number of pulses $Np$. A narrow avoided-crossing (low splitting, $\delta$ small) gives a single, sharp (but weaker) coherence dip. A broad (large $\delta$) crossing has a very broad envelope, modulated by sidebands dependent on $Np$.

Comparison with Average Hamiltonian models A frequently used approximation in spin sensing is the average Hamiltonian model wherein the eigenvalues $\omega_{\text{av}}$ of $H_d$ of the time-averaged Hamiltonian provide an estimate of the dip positions and that $\tau_{\text{dip}} = \frac{Np\pi}{\omega_{\text{av}}}$ is significant; since $|\mathcal{E}F(\tau_{\text{dip}})| = \pi$, we can show that this is equivalent to $\tau_{\text{dip}} = \frac{Np\pi}{\omega_{\text{av}}}$. The reason why the short time, linear behavior is equivalent to average Hamiltonian behavior is clear. The quantum time propagator for CPMG-2N may be formally written:

$$T(0,4Np\tau) = T \left[ e^{ijH_u\tau} e^{-iH_d\tau} e^{-iH_u\tau} e^{iH_d\tau} \right]^{Np} \quad (19)$$

where the time-ordering operator $T$ reflects the non-commutative nature of the time propagation under $H_u$ and $H_d$. For sufficiently short time, each cycle may be expressed in 2nd-order Suzuki-Trotter form $T(0,4\tau) \approx e^{-iH_u\tau} e^{-iH_d\tau} e^{-iH_u\tau} e^{-iH_d\tau} \approx e^{-2i(H_u+H_d)\tau} + \delta E...$ where the first term represents propagation under the average Hamiltonian and the error $\delta E \sim O(\tau^3)$. In fact, for CPMG-1 (Hahn echo) the error is $O(\tau^2)...$ thus average Hamiltonian propagation is more accurate under CPMG-2N. For sufficiently short times, CPMG-2 behaviour is exactly equal to propagation under $\frac{1}{2}(H_u + H_d)$.

In sum, the validity of average Hamiltonian models can in our model, once again be related to the curvature of the Floquet anti-crossing. A low-curvature crossing, which remains linear for most times so $\mathcal{E}F(\tau) \approx \epsilon_0\tau$ up to $\tau \approx \tau_{\text{dip}}$ gives good agreement with Eq.[18] while for high-curvature avoided crossings, average Hamiltonian models fail; the short-time quasi-energy is only a good approximation to the true quasienergy if $\epsilon_{\text{nor}} = \frac{\mathcal{E}F(\tau_{\text{nor}})}{\pi} \approx \epsilon_0$. In other cases, one must obtain exact solutions of the orthogonality condition $\mathcal{E}F(\tau_{\text{dip}})/2 = \pi/2$.

In order to estimate the error in the average Hamiltonian, we consider the cubic error term (using the Baker- Campbells-Hausdorff relation) in terms of the pseudo-field components:

$$\frac{1}{12} \left( [H_u, [H_u, H_d]] - [H_d, [H_u, H_d]] \right) \tau^3 = \frac{1}{12} X (Z_u - Z_d)^2 \tau^3 \quad (20)$$

at the dip, $\tau \sim h^{-1}$ ie some representative mean Hamiltonian; for small interaction term (small $X$) then, $h \sim Z = \frac{1}{2} (Z_u + Z_d)$ thus it is not unreasonable to expect the error at

FIG. 5: Comparisons between the dip positions obtained with Eq.[13] (blue line) and average Hamiltonian theory Eq.[18] (red line) for the full coherence function (left panels) as well as its envelope (right). (a) and (d) R=100 (b) and (e) R=20 (c) and (f) R=10.
small times to scale with $\delta E \sim \frac{X}{Z} (\frac{Z_u - Z_d}{Z})^2$. We test this numerically, by examining the discrepancy between the exact and average Hamiltonian results as a function of the ratio $r = (Z_u - Z_d)^2/(\frac{Z}{Z})$ and also as a function of interaction $\eta = \frac{X}{Z}$.

In Fig. [5], we plot the effect of varying magnetic field on the ratio $r$ for NV centres and also for an Si:Bi spin. Under a field sweep, the Si:Bi system spans a wide range of regimes. The figure shows a rich pattern of avoided crossings which widen and narrow successively. Agreement with average models is excellent for smaller $X/Z$ at all fields, but not so for larger $X/Z$, as shown in Fig. [5]. For small $Z_u - Z_d$, the discrepancy grows quadratically, as expected from Eq. [20]. Fig. [4] shows the comparable behaviour for an NV center. NV centers show a strikingly different pattern for the $F(\tau)$ envelope: they produce a distinctive pattern of coherence “diamonds” with the width of avoided crossings growing linearly, rather than quadratically (consistent with what one expects from BCH: $\frac{(Z_u - Z_d)^2}{Z}$) is held fixed while $X$ increases from the origin.

We have presented here the Floquet theory analysis of sensing under periodic dynamical decoupling sequences. This formalism is equivalent to previous geometrical approaches but offers new insights. The theory is fully general for the one-spin problem and can be applied to a range of sensing systems such as NV centres, silicon vacancy centres and electron donors in silicon.

The key change in understanding is that dips in the coherence envelope appear at avoided crossings of the Floquet donor systems such as NV centres, silicon vacancy centres and electron donors in silicon.

The analysis with Floquet theory presented here is ideally complemented by field sweeps- i.e. not to fit a single set of dips but rather to the locus of dips whose position varies sensitively with experimental parameters. Unlike pseudospin models, Floquet theory is valid for larger dimensions and is a promising tool for studying higher order cluster effects.

I. SENSING WITH DONORS IN SILICON

Central spin systems in silicon are also of technological interest, for example because of their very long coherence times [21], which attain the seconds timescale without any dynamical decoupling control. Although electron donor atoms in silicon offer a promising source of spin qubits, they do not yet offer the level of optical control possible with single defects in diamond which makes it possible to efficiently polarise the central spin and to accurately read it out optically; however, rapid progress is being made in control and read-out at the single spin level [22–24] and thus it is useful to already examine, at least in principle, their potential as sensors. Certain donor electron spins (for atoms such as bismuth or arsenic) have very strong mixing with the host nuclear spin, making their coherence properties sensitively dependent on magnetic field strength [25–28]. The coherence times can be very simply characterised by considering the electronic spin projection on the $z$ axis as a function of the external field $B_0$ as was illustrated in Fig. [1] and exhibit orders of magnitude changes within a span of 100 Gauss [29]. The sensitivity of the behavior to external applied field may potentially be exploited for sensing and is investigated here.

A. Sensor spins with variable $z$ projection

In the absence of the periodic dynamical decoupling, the sensor-nuclear spin dynamics takes a well known form. The Hamiltonian for the joint central spin system and environment cluster takes the form:

$$\hat{H}_{\text{tot}} = \hat{H}_{\text{CS}} + \hat{H}_{\text{int}} + \hat{H}_{\text{nucl}}.$$  \hspace{1cm} (21)

$\hat{H}_{\text{CS}}$ is our central spin system Hamiltonian:

$$\hat{H}_{\text{CS}} = B_0 \gamma_c \hat{S}_z + \hat{H}_\beta$$  \hspace{1cm} (22)

where the first term is the Zeeman interaction with the external magnetic field and the second is a (system-dependent) interaction which means that the eigenstates of $\hat{H}_{\text{CS}}$ are not $|Sm_S\rangle$, but have a different projection $\langle i | \hat{S}_z | i \rangle = S \cos \beta_i$ for the $i$-th eigenstate. In general, the tilt $\beta_i(B_0)$ depends on the magnitude of $B_0$. For example, for donors in silicon, the additional term arises from the strong coupling to the host nuclear spin thus $\hat{H}_\beta \simeq A \hat{S} \cdot \mathbf{S}$; for NV− centers, $\hat{H}_\beta \simeq D \hat{S}_z^2$ the interaction is a crystal field which only causes a tilt if there is an angle $\theta \neq 0$ to the magnetic field axis; for silicon vacancies in diamond the tilting, attributed to spin-orbit effects, is described in [9].

Donors have a rich mixing structure and $\beta_i(B_0)$ is easily evaluated analytically [23–26]. The parameter, $\cos \beta_i$, is obtained from the central spin Hamiltonian. For donors in silicon such as arsenic or bismuth this is

$$\hat{H}_{\text{CS}} = \omega_0 \hat{S}_z - \omega_0 \delta \hat{I}_z + A \hat{S} \cdot \hat{I}.$$  \hspace{1cm} (23)

where $\omega_0 = B g \beta$ is the electron Zeeman frequency ($g$ is the electron g-factor and $\beta$ is the Bohr magneton). $\delta = \omega_I/\omega_0 =$
2.488 \times 10^{-4} \text{ is the ratio of nuclear and electron Zeeman frequencies and } A \text{ is the hyperfine coupling between nuclear and electron spin. }

For the Si:Bi system \( S = 1/2 \) and \( I = 9/2 \) which produces 20 different energy levels. At intermediate magnetic fields the energy levels are mixed and \( m_s \) and \( m_I \) are not good quantum numbers. Instead a good quantum number is \( m = m_s + m_I \). The Hamiltonian can be decoupled into a direct sum of two-dimensional sub Hamiltonians, \( H_m \), with constant \( m \) and basis states \( |m_s = \pm 1/2, m_I = m \mp 1/2 \).

The sub Hamiltonians, \( H_m = \frac{1}{2} J m \), are given by

\[
\hat{h}_m = Z_m \sigma_z + X_m \sigma_x - (\frac{1}{2} + 2m \delta \omega_0)I,
\]

where

\[
Z_m = [m + \omega_0(1 + \delta)]
\]

\[
X_m = (I(I + 1) + \frac{1}{4} - m^2)^{1/2}
\]

If we denote \( R_m = \sqrt{Z_m^2 + X_m^2} \), then

\[
\cos \beta_{u,l} = \pm Z_m / R_m.
\]

The form of \( \beta_i(B_0) \) is illustrated in Fig.6 for a key ESR spectral line of bismuth. At high fields \( B_0 \), the angles correspond to the untitled “bare spin” regime where \( \beta_u = 0, \beta_d = \pi \), but at lower \( B_0 \), we highlight three regimes:

(i) and (ii) where the tilt angles pass through two separate field values, where one angle \( \beta_{u,d} = \pi/2 \) (at \( B_0 = 0.16 \) and 0.21T); this regime is in fact analogous to the behaviour of typical NV\(^-\) experiments for which \( m_s = 0 \) (also corresponding to \( \beta = \pi/2 \)). The height and width of the decoherence “dips” is maximal around these points. We refer to them as LZ points since the bismuth electronic states with \( \beta_{u,d} = \pi/2 \) are at a Landau-Zener crossing (not to be confused with the Floquet states anticrossing).

(ii) In between there is a field value (in this particular example at \( B_0 = 0.188 \)) where \( \beta_u = \beta_d \), corresponding to an “Optimal Working Point” for decoherence suppression here, with much lower visibility of the coherence “dips”. However, we show below that the coherence function \( C(B_0, \tau) \) then becomes considerably simpler, with less peak-splitting and fewer sidebands, which can also be advantageous.

The bath Hamiltonian consists of nuclear Zeeman terms and dipolar coupling within each cluster of bath spins:

\[
\hat{H}_{bath} = \hat{H}_{D} + \hat{H}_{NZ},
\]

\[
\hat{H}_{NZ} = \sum_a \gamma_N B I_a^z,
\]

\[
\hat{H}_{D} = \sum_{a < b} I_a C(r_{ab}) I_b,
\]

where \( \gamma_N \) is the nuclear (bath) gyromagnetic ratio and \( r_{ab} \) denotes the relative position of bath spins at lattice sites \( a \) and \( b \). The components of the dipolar tensor \( D(r) \) are given by

\[
C_{ij}(r) = \frac{\mu_B h Z_N}{4 \pi r^3} \left( \delta_{ij} - \frac{3 r_i r_j}{r^2} \right),
\]

where \( \delta_{ij} \) denotes the Kronecker delta, \( \mu_B = 4 \pi \times 10^{-7} \text{ NA}^{-2} \)

and \( i, j = x, y, z \). For the \( k \)-th pair of bath spins, the Zeeman interaction and pairwise dipolar interaction are given by

\[
\hat{H}^{(n)}_{bath} = \gamma_N B_0 (I_1^z + I_2^z) + 2 C_{12} (I_1^+ I_2^- - C_{12}^{(n)} / 2) (I_1^+ I_2^- + I_2^- I_1^-),
\]

where \( C_{12}^{(n)} \) is the dipolar coupling strength between the two bath spins.

A common and extremely convenient approximation is to neglect non-secular terms in \( \hat{H}_{int} \), valid for typical values of \( B_0 \), due to the disparity in the energy cost of an electronic spin-flip, relative to a nuclear, spin-flip:

\[
\hat{H}_{int} \approx \sum_k J_k \hat{S}_k \hat{I}_k
\]

where \( \hat{S} \) represents the central electron spin, \( J_k \) is the hyperfine coupling tensor and \( k \) labels the bath spins \( \hat{I}_k \).

In that case, the total Hamiltonian \( \langle u | \hat{H}_{tot} | d \rangle \approx 0 \) does not couple electronic states and evolution becomes conditional on the state of the central spin system. \( i = u, d \). The problem is then diagonalised using a basis of bath states \( | \uparrow \rangle \) and \( | \downarrow \rangle \), for each bath spin. Given Eq.31 one can also assume that the total \( I^z \) is conserved in each cluster. Thus, for a pair, the basis of \( | \uparrow \downarrow \rangle \) and \( | \downarrow \uparrow \rangle \) states are coupled by the dynamics, resulting in a familiar “flip-flop” dynamics; in contrast, states \( | \uparrow \uparrow \rangle \) and \( | \downarrow \downarrow \rangle \) are largely decoupled from the flip-flop states and each other. As outlined in the next section, the coherence dynamics due to the \( k \)-th pair then reduces to the competition between
the detuning due to the central spin $\Delta J = J_1 - J_2$ and the intra-pair dipolar coupling $C_{12}$.

For a three cluster, similarly, the 8 basis states couple into (i) two sets of three coupled states which can flip flop and contribute to decoherence $eg \{ |\uparrow\uparrow\uparrow\rangle, |\uparrow\downarrow\uparrow\rangle \}$ and $|\downarrow\uparrow\uparrow\rangle$) the states with total quantum number $I^z = +1/2$; the equivalent with total $I^z = -1/2$; and (ii) the non-mixing states $|\uparrow\uparrow\uparrow\rangle$ and $|\downarrow\downarrow\downarrow\rangle$, which contribute only weakly to the decoherence.

The $T_i^{(0)}(\tau)$ matrices ($i = u, d$) can be constructed for a given cluster of two, three or more spins using the eigenvalues and eigenvectors of the total Hamiltonian and the corresponding $T_i^{(2)}(\tau)$ are in turn diagonalised to obtain the complex eigenvalues $e^{-i\xi F}$ and hence the Floquet eigenphases $\xi F$. For the pairs, we can use the analysis of the previous section but use full numerics as a check.

We note that at the LZ points, the form of the bismuth pseudospin Hamiltonian is quite analogous to the form of the NV centre pseudospin and is described by effective fields of the form $h_u = (X,0,0)$ and $h_d = (X,0,0)$ (for the low field LZ point) thus one field has zero $z$ projection while the other does not, with $X = C_{12}/4$ and $2Z_i = \cos \beta_i \Delta J$. In particular, the experiments were interpreted using the average Hamiltonian model and Eq. [17] to obtain $T_{uv} \approx 2 N_p \pi/(X^2 + (Z/2)^2)1/2$ for a cluster with $\tau \approx 20$. Fig. 7 illustrates the fact that the Si:Bi LZ points and Optimal Working Points mimic some of the characteristics of NV center dimer sensing and NV center single-spin sensing respectively.

II. MANY-BODY INTERACTIONS

A. 3-cluster model

In this section we apply the Floquet approach to the system depicted in Fig. 1(c): we compare the decoherence “fingerprint” of three distinct spin pairs (analogous, formally, to the detection of three independent spins by NMR) with a “fingerprint” of three distinct spin pairs (analogous, formally, to the detection of three independent spins by NMR) with a unique signature.

For the 3-cluster, we take three spins, with hyperfine couplings $J_k \equiv J_1, J_2, J_3$ to the sensor spin and with mutual dipolar interactions $C_{ij} \equiv C_{12}, C_{23}, C_{31}$. Disregarding interactions, the energy cost of the spin flips is $\Delta_{ij} = J_i - J_j$.

For the disjoint pairs, we take three spin pairs, with the same dipolar interactions $C_{ij}$ as the 3-cluster, but which are independent of each other. To have similar frequencies as the 3-cluster, we must have similar energy cost of a all three spin flips; and they must obey the cyclic condition of the 3-cluster $\Delta_{12} + \Delta_{23} + \Delta_{31} = 0$. Pair 1 has two spins with interaction $C_{12}$ and a pair of hyperfine couplings $(J_1, J_2)$; pair 2 has interaction $C_{23}$ and hyperfine couplings $(J_2, J_3)$; pair 3 has $C_{31}$ and hyperfine couplings $(J_3, J_1)$. We take $C_{12} = C_{23} = 1.05 \times 10^5$ kHz and $C_{31} = 2.2 \times 10^5$ kHz, realistic values for nuclear impurities in the silicon lattice. We take $J_1 = 180$ kHz, $J_3 = 100$ kHz and $J_2 = 0$, thus our pairs correspond to $r \approx 100 - 40$ so the interactions are sufficiently weak to make their detection challenging but sufficiently strong to, below, illustrate important features. The choice of $J_2 = 0$ does not involve much loss of generality. If a state-dependent Hamiltonian is chosen, the $J_1, J_2, J_3$ values can be shifted by an arbitrary constant without perturbing the dynamics. If the full Hamiltonian is considered, there can be higher order effects such as hyperfine mediated corrections to $C_{ij}$. This correction is very small for our parameters but is tested by full numerics below.

Although our model corresponds to disjoint pairs versus interacting spins, it is straightforward to generalise to the molecular NMR sensing situation where we detect (say) three independent independent nuclear spins with intrinsic frequencies $\omega_k$, determined by the molecular environment as well as mag-
netic Zeeman energies the scenario investigated in [11]. In that situation too, in future it will be useful to detect also mutual interactions between the spins. In our case, the intrinsic frequency of each spin pair or pseudo-spin is largely set by \( \Delta_{ij} \).

### B. Solution of total Hamiltonian

First we set aside all pseudospin approximations and do full diagonalisations using the complete 8-state basis of the 3-cluster as well as the complete basis of the bismuth sensor including the host nuclear spin. Thus, unlike Fig.3 we do not use the analytical form for the \( z \) projection angle \( \beta \); it emerges from the numerics. We evaluate the decoherence numerically rather than using Eq.13. A similar calculation was carried out with the three disjoint pairs, then the decoherence was averaged over the corresponding bath states (eight and four respectively). Figure 8 shows a map of the coherence in the \((\tau, B_0)\)-space in both cases.

One conclusion to be drawn from comparisons between full numerics and the analytical (one-pair) Eq.13 is that the complex sideband structure in Fig.3 is surprisingly robust; without bath state averaging, full numerics give very similar structure to Fig.3 (obtained from Eq.13 for one bath state). Current realisations are heavily damped by incoherent contributions, but future realisations using sensors with long coherence times \((T_2\) times on the order of seconds have been measured for Si:Bi\(^{28}\)) could in principle reveal this structure.

### C. Floquet Spectroscopy

One striking feature of the 3-cluster decoherence map in Fig.8(b) is that some lines are split into “doublets” with very similar structure. The origin of these is in the average over the bath states; examining maps for the individual 8 bath states, we see that while the \( I_z = \pm 3/2 \) cluster states \(|\uparrow\uparrow\uparrow\rangle\) and \(|\downarrow\downarrow\downarrow\rangle\) make no appreciable contribution, the doublets arise from the separate \( I^2 \) \(= \pm 1/2 \) subspaces, which do not mix. In other words, the \(|\uparrow\downarrow\rangle\), \(|\downarrow\uparrow\rangle\) and \(|\downarrow\downarrow\rangle\) states with total quantum number \( I^2 = -1/2 \) interact only weakly with the equivalent with total \( I^2 = -1/2 \) subspace, but each provides a locus of dips with a slightly different shift. In contrast to the spin pairs, in the case of the 3-cluster, the secular Ising \((C_{ijk})\) components yield a non-trivial dynamical effect.

In order to capture the most significant secular corrections from the dipolar interactions, we can estimate a Floquet quasienergy by considering only the diagonals. We obtain:

\[
\epsilon_l = \frac{1}{2}(J_{ij} - J_{jk}(\cos \beta_u + \cos \beta_d) + C_{ij} + C_{ik} - C_{jk} \quad (32)
\]

where \( i, j, k = 1, 2, 3 \) or cyclic permutations give \( \epsilon_{l=1,2,3} \) quasienergies.

Thus we estimate the dip positions:

\[
\tau_{\text{dip}}^{(l,m)} \simeq \frac{2\pi}{\epsilon_l - \epsilon_m} \quad (33)
\]

for the dip arising from the difference between the \( l \) and \( m \)-th quasienergies. This secular contribution greatly amplifies the dipolar coupling as it is a linear contribution. This is in contrast to disjoint pairs; if the dipolar coupling is weak, since \( \omega_l = \pm \sqrt{C^{2}_{12} + (\cos \beta_1 \Delta_j)^2} \), for \( C_{12} \ll \cos \beta_1 \Delta_j \) the non-secular contributions in the disjoint pairs represent a

---

**Fig. 8:** Fingerprinting multiple environmental spin cluster-pairs via their decoherence “bar-code”. Illustrates the effect of 3-body correlations. The figure shows the coherence as a function of magnetic field \( B_0 \) and pulse interval \( \tau \), calculated with a full numerical diagonalisation of the total Hamiltonian for \( N = 100 \). The top panel denotes three independent pairs while the lower panel shows three interacting spins, with otherwise equivalent dipolar couplings and intrabath interactions as illustrated in Fig.8(b). One evident difference (and signature of a cluster of three spins) are the doublets due to the two separate subspaces of the three interacting spins. The splittings are directly related to the interactions. For the 3-cluster, in fact there is a secular contribution from interactions between spins, greatly amplifying their contribution.
quadratic shift. This is shown in Fig. 9 where Eq. 33 accurately follows the numerical dips.

FIG. 9: Decoherence for the same 3-cluster as in Fig. 8. The coloured lines show comparisons with Eq. 32 showing excellent agreement with numerics. The horizontal lines (i) and (ii) corresponding to $B_0 = 0.185$T and 0.2T respectively correspond to the traces in Fig 1(b) and are also the two traces analysed below.

In terms of the interaction strengths, the two dips of the first doublet correspond to:

$$
\tau_{dip}^{(\pm 12)} \approx \frac{2\pi}{|\Delta_{12}(\cos \beta_u + \cos \beta_d) \pm (C_{31} - C_{23})|} \quad (34)
$$

and similarly for other doublets. Thus the mean position exposes the value of $\Delta_{12}$ while the splittings expose the dipolar coefficients.

The dipolar interactions $C_{ij}$ also give a weaker off-diagonal contribution which may be analysed with Floquet spectroscopy. In this case, interactions appear as avoided crossings of the Floquet eigenphases. Some key effects are illustrated in Figs. 10, 11 and 12 which compare the positions of decoherence dips with the underlying Floquet eigenspectrum of our model problem (parameters as defined above) comparing 3 disjoint pairs with a single equivalent 3-cluster. The Floquet spectroscopy in different regimes exhibits a rich structure but here for brevity we highlight only a subset of the key features.

**III. CONCLUSION**

We have shown here that Floquet theory provides a versatile tool for the analysis of data for sensing by dynamical decoupling control applied to arbitrary sensor spins—provided the applied pulse sequence is temporally periodic. It predicts the decoherence peaks with greater accuracy and reliability than current models. For complex clusters, Floquet spectroscopy offers a new handle on potentially complex decoherence spectra. Even with the additional theoretical tool, experimental sensing of potentially weak interactions remains a considerable challenge. Thus the analysis with Floquet theory presented here is ideally complemented by field sweeps—ie not to fit a single set of dips but rather to the locus of dips whose position varies sensitively with experimental parameters. The variation of pulse number $N$ is a well-tried alternative but only scales and does not vary the underlying frequency. For NV centres in diamond, the tilt angle of the field can be varied. But we showed here that the extreme sensitivity of the decoherence of donors in silicon to external field offers an exceptionally promising sensor, at least in principle: actual realisations must await technical developments which permit bismuth or arsenic to achieve the same capability in initialisation and read-out as the NV defects in diamond. But other defects such as
silicon vacancies in diamond also show rapid variation of $z$ projection with $B_0$ and could also be investigated.

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**Appendix A: Spin sensing via coherence decays**

The detection of nearby spins via their effect on the coherence of a central spin is now well understood, theoretically and experimentally\[15\] and is best understood as arising from their mutual entanglement. The central state is initially prepared in a superposition of upper and lower states at $t = 0$ so the central-spin bath state is:

$$|\Psi(0)\rangle = \frac{1}{\sqrt{2}}(|u\rangle + |d\rangle) \otimes |B(0)\rangle. \quad (A1)$$

The full state evolves under the joint Hamiltonian $\hat{H}_{tot}$ and the two components entangle, so at time $t$, $|\psi(t)\rangle = \frac{1}{\sqrt{2}}(|u\rangle|B_u(t)\rangle + |d\rangle|B_d(t)\rangle)$. While a full numerical quantum solution solves for the effect of $\hat{H}_{tot}$, analysis is considerably simplified by the usual assumption (valid for reasonable magnetic fields, see below) that $\langle u|\hat{H}_{tot}|d \rangle \simeq 0$ and thus:

$$|\psi(t)\rangle = \frac{1}{\sqrt{2}}(|u\rangle T_u(t)|B(0)\rangle + |d\rangle T_d(t)|B(0)\rangle) \quad (A2)$$

The bath state evolution is given by unitaries given by effective Hamiltonians i.e. $T_u(t) = \exp (-iH_u t)$ or $T_d(t) = \exp (-iH_d t)$, which are conditional on the state of the central spin. The bath dynamics (such as the flip-flopping of environment spin pairs) is then given by the eigenvalues of $H_{u,d}$ which are $\omega_u$ and $\omega_d$, where $l = 1, ..., D$ and $D$ is the dimension of the effective Hamiltonian. Phases arising from the evolution of $|u, d\rangle$ are removed experimentally by spin echo measurements.

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The application of dynamical decoupling sequences (as a train of $\pi$ pulses) does not significantly modify the above. However each $\pi$ pulse switches the populations in the $u \rightarrow d$ and vice-versa. Thus the global time evolution operators are constructed by concatenating the unitaries for each time step. For instance the time evolution unitaries for a Hahn echo (or CPMG-1), given by the sequence $\tau - \pi - \tau$ are denoted below by:

$$T_u^{(1)}(t = 2\tau) = T_u^{(0)}(\tau)T_d^{(0)}(\tau) \quad (A4)$$

$$T_d^{(1)}(t = 2\tau) = T_d^{(0)}(\tau)T_u^{(0)}(\tau) \quad (A4)$$
while for CPMG-2 dynamical decoupling, given by the sequence $\tau - \pi - \tau - \pi - \tau$ is given as:

$$
T_u^{(2)}(t = 4\tau) = T_u^{(1)}(2\tau)T_d^{(1)}(2\tau)
$$

$$
T_d^{(2)}(t = 4\tau) = T_d^{(1)}(2\tau)T_u^{(1)}(2\tau)
$$

(A5)

and so forth, for longer sequences, such as CPMG-N, denoted by $T_u^{(N)}(t = 2N\tau)$ and $T_d^{(N)}(t = 2N\tau)$. Even numbers of pulses are simply periodic repetitions of $T_u^{(2)}$.

In Fig 4(a) the CPMG-2 and CPMG-N sequence with even numbers of pairs (i.e. where $N = 2N_p$) is illustrated. These dynamical decoupling sequences suppress decoherence so $L(\tau) \simeq 1$; nevertheless, it has been observed in a number of experimental studies that proximate spins and spin-pairs (which interact strongly with the central spin) lead to “dips” in the background $L(\tau) = 1$ coherence function. Two typical traces are shown in Fig 4(d), for constant $B_0$; the positions and depths of the dips are related to the spin dynamical parameters. For single spins or pairs, pseudospin models have proved very useful. However, extracting information from a single experimental trace is non-trivial except for certain simple situations, such as detection of single C$^{13}$ (cluster of one) nuclear spins by NV$^-$ centres as the dynamics is characterized approximately by a single frequency, see Fig. 7(b).

**Appendix B: Pair Floquet states**

Here we give the Floquet states in terms of pseudospin parameters. For a donor spin, the effective spin field is $h = (X, 0, Z_i) = \frac{1}{2}(C_{12}, 0, \cos \beta_i \Delta_j)$. The pseudospin Hamiltonian is:

$$
H_i = \frac{1}{4}(C_{12}\sigma_x + P_i \Delta_j \sigma_z)
$$

(B1)

with $P_i = \cos \beta_i$, $i = u, d$, and with eigenvalues

$$
\omega_i = \pm \frac{1}{2}\sqrt{C_{12}^2 + (P_i \Delta_j)^2}
$$

(B2)

The Hamiltonian can be written as $H_i = R_y(\theta_i)\Omega_i R_y(\theta_i)$ where

$$
R_y(\theta) = \left( \begin{array}{cc} \cos(\theta/2) & -\sin(\theta/2) \\ \sin(\theta/2) & \cos(\theta/2) \end{array} \right), \Omega_i = \left( \begin{array}{cc} \omega_i & 0 \\ 0 & -\omega_i \end{array} \right)
$$

(B3)

and $\theta_i = \arctan(C_{12}/P_i \Delta_j) = \arctan(X/Z_i)$.

The time-evolution operator is given by $T^{(0)}_i(t) = R_y(\theta_i) \exp [-i\Omega_i t R_y(\theta_i)]$ where

$$
\exp [-i\Omega_i t] = \left( \begin{array}{cc} e^{-i\omega_i t} & 0 \\ 0 & e^{+i\omega_i t} \end{array} \right)
$$

(B4)

After a time $\tau$ a $\pi$-pulse is applied that swaps the states $|u\rangle$ and $|d\rangle$. The Hahn echo occurs after another time $\tau$ has elapsed. The Hahn echo bath propagator is

$$
T_u^{(1)}(\tau) = T_u^{(0)}(\tau)T_d^{(0)}(\tau) = A_0I - i(A_x\sigma_x + A_y\sigma_y + A_z\sigma_z)
$$

(B5)

where

$$
A_0 = \cos(\omega_u\tau) \cos(\omega_d\tau) - \sin(\omega_u\tau) \cos(\omega_d\tau) \cos(\theta_u - \theta_d)
$$

(B6)

$$
A_x = \sin(\omega_u\tau) \cos(\omega_d\tau) \sin(\theta_d) + \cos(\omega_d\tau) \sin(\omega_u\tau) \sin(\theta_u)
$$

(B7)

$$
A_y = -\sin(\omega_u\tau) \sin(\omega_d\tau) \sin(\theta_u - \theta_d)
$$

(B8)

$$
A_z = \sin(\omega_u\tau) \cos(\omega_d\tau) \cos(\theta_d) + \cos(\omega_d\tau) \sin(\omega_u\tau) \cos(\theta_u)
$$

(B9)

The other Hahn echo bath propagator is the same except $A_y$ changes sign, $T_u^{(1)}(\tau) = T_d^{(0)}(\tau)T_u^{(0)}(\tau) = A_0I + i(A_x\sigma_x - A_y\sigma_y + A_z\sigma_z)$.

To obtain the CPMG-2 propagator we concatenate the two Hahn echo propagators

$$
T_u^{(2)}(\tau) = T_u^{(1)}(\tau)T_d^{(1)}(\tau) = a_0I - i(a_x\sigma_x + a_z\sigma_z)
$$

(B10)

where

$$
a_0 = A_0^2 + A_y^2 - A_x^2 - A_z^2
$$

(B11)

$$
a_x = 2[A_x A_0 + A_y A_z]
$$

(B12)

$$
a_z = 2[A_x A_0 - A_y A_z]
$$

(B13)

and $T_d^{(2)}(\tau) = T_d^{(1)}(\tau)T_u^{(1)}(\tau) = a_0I - i(a'_x\sigma_x + a'_z\sigma_z)$ with

$$
a'_x = 2[A_x A_0 - A_y A_z]
$$

(B14)

$$
a'_z = 2[A_x A_0 + A_y A_z]
$$

(B15)

The CPMG-2 propagator can be written as $T_i^{(2)}(\tau) = R(\theta_i^F)ER_i^T(\theta_i^F)$ where $E$ is the diagonal matrix of $T_i^{(2)}$ eigenvalues (the same for $i = u$ and $i = d$). The phase of the eigenvalues is the Floquet phase $\mathcal{E}^F$

$$
E = \left( \begin{array}{cc} e^{-i\mathcal{E}^F} & 0 \\ 0 & e^{+i\mathcal{E}^F} \end{array} \right)
$$

(B16)

The Floquet angles and phase are functions of the pulse spacing, $\tau$:

$$
\mathcal{E}^F(\tau) = \arccos(a_0)
$$

(B17)

$$
\theta^F_u(\tau) = \arctan(a_x/a_z)
$$

(B18)

$$
\theta^F_d(\tau) = \arctan(a'_x/a'_z)
$$

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