Adiabatic Dynamical-Decoupling Based Control of Nuclear Spin Registers

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The use of the nuclear spins surrounding electron spin qubits as quantum registers and long-lived memories opens the way to new applications in quantum information and biological sensing. Hence, there is a need for generic and robust forms of control of the nuclear registers. Although adiabatic gates are widely used in quantum information, they can become too slow to outpace decoherence. Here, we introduce a technique whereby adiabatic gates arise from the dynamical decoupling protocols that simultaneously extend coherence.

We illustrate this pulse-based adiabatic control for nuclear spins around NV centers in diamond. We obtain a closed-form expression from Landau-Zener theory and show that it reliably describes the dynamics. By identifying robust Floquet states, we show that the technique enables polarisation, one-shot flips and state storage for nuclear spins. These results introduce a new control paradigm that combines dynamical decoupling with adiabatic evolution.

There is enormous interest in the development of quantum technologies based on spins in the solid state. Optically active defects, such as the Nitrogen vacancy (NV) center in diamond, offer well-isolated individual electronic spins with long coherence times, optical addressability and operation from cryogenic to room-temperatures [1–7]. In combination with nearby coupled nuclear spins, these systems provide a multi-qubit platform with a wide range of applications including detection, imaging and atomic-scale characterisation of the spin samples [8–12], quantum computation and quantum networks [13–15].

Both sensing and quantum information applications typically rely, for control of the central electronic spin, on sequences of periodically repeated microwave pulses, known as dynamical decoupling (DD). Although DD was initially employed to decouple the central electron spin qubit from the decohering effect of the surrounding spin bath [16], it was soon identified that the same sequences could also be used to sense individual nuclear spins [17–18]. For the case of the NV center in diamond, the entanglement generated between the electron spin and the $I = 1/2$ $^{13}$C nuclear spins is being explored as a tool for control, with nuclear spins as multi-qubit registers and quantum memories [19,20]. Furthermore, state storage in nuclear spins has been used to augment protocols for quantum sensing and nanoscale NMR [21].

Adiabatic quantum gates are well established in quantum information but, as adiabaticity entails slow parameter sweeps, outpacing decoherence represents a problem [22]. Proposals employing dynamical decoupling to extend coherence alongside the adiabatic gates have been investigated [23,24]. However, to our knowledge, the adiabatic gates that are intrinsic to the commonly used pulse-DD protocols have not previously been considered: this may be surprising as they offer the important advantage that the coherence-protecting repeated pulse-DD protocols, in some sense, “come for free”.

Here we introduce and investigate a new technique that combines adiabatic passage with coherence protection through dynamical decoupling. We name this method Ad-Pulse. This involves adiabatically sweeping over the Floquet eigenstates of the DD pulse protocol. We show that this type of control is quite generic and, for various applications, does not require knowledge of the individual resonances. Ad-Pulse can also be applied to a many-spin bath and we show some applications are insensitive to the number of spins. We illustrate Ad-Pulse using the example of an NV centre and surrounding nuclear registers. We show also it may polarise and initialise small nuclear clusters more effectively than the recently pro-

![Image](https://example.com/image.png)
For $\tau$ period the is no period averaging and in fact one sweeps coherently over of an effective static Hamiltonian. Bath [30] using an adiabatic sweep of the energy eigenvalues been proposed theoretically for polarisation of a nuclear spin may tune over the effective Hamiltonian. This approach has driving. Varying the amplitude renormalised parameters, by averaging over the period of the typically strong or high frequency (non-resonant) field can be shown to correspond to an effective, static Hamiltonian with characteristic resonances of the nuclear spins of interest. Previously it has been shown that the loss of coherence identified in DD studies at resonances correspond to avoided crossings of the Floquet eigenphases [31, 32]. Recognising this, Ad-Pulse sweeps over these eigenphases: if sufficiently slow, the sweep follows each eigenstate adiabatically and coherently.

A system with a temporally periodic Hamiltonian, $\hat{H}(t+T) = \hat{H}(t)$, Floquet’s theorem allows one to write solutions of the Schrödinger equation in terms of quasi-energy states $|\psi_l(t)\rangle = \exp(-i\epsilon_l t)|\Phi_l\rangle$ where $\epsilon_l$ is the quasi energy, $|\Phi_l(t)\rangle = |\Phi_l(t+T)\rangle$, $T$ is the period and $l = 1,..,D$ ($D$ is the dimension of the state space).

Where we require only “stroboscopic” knowledge of our system at times $t = t + kT$, one may obtain eigenstates of the one-period unitary evolution operator $\hat{U}(T) \equiv \hat{U}(T,0)$ for the joint electron-nuclear spin bath system under pulse-DD. The Floquet states $|\Phi_l\rangle$, obey the eigenvalue equation:

$$\hat{U}(T)|\Phi_l\rangle = \lambda_l|\Phi_l\rangle \equiv \exp(-i\epsilon_l)|\Phi_l\rangle$$

**FIG. 2.** Comparison of standard DD with Ad-Pulse DD, using the PolCPMG polarisation protocol, applied to an NV centre electron coupled to one or more nuclear spins. In previously demonstrated PolCPMG experiments [27], a slight under/overrotation of the $\pi$ pulses was shown to split the normal CPMG resonance at $\tau_{\pi}$ into two $\tau_{\pi} \equiv \tau^\pm$ resonances that are nuclear state selective. The initial state is $|X^\pm\rangle_{NV} \otimes \rho_n$, so the nuclei are initially in a mixed state and $|X^\pm\rangle \equiv |0\rangle \pm |1\rangle$. (a) Single spin case. DD resonances correspond to avoided crossings of the Floquet eigenphases (upper panel). Applying PolCPMG at $\tau^\pm$ or $\tau^+$ polars the mixed state as shown (lower panel). The corresponding adiabatic protocol (Ad-PolCPMG) sweeps slowly over a $\tau$ range of $\equiv \tau^\pm \pm \Delta\tau/2$ without reinitialisation of the NV for different $\tau$. As the sweep step size, $\delta\tau$, is reduced the sweep becomes adiabatic and full polarisation is achieved: the behavior for all parameters is well described by Eq.4, based on Landau Zener theory (red curves). (b) Multi-spin cluster. The Floquet level structure is illustrated for the case of $N_{nuc} = 7$ nuclei. Although $M_z$ is not a good quantum number during the Ad-Pulse, for clarity, states are coloured according to the asymptotic states $|\Phi_l\rangle$ is not a good quantum number during the Ad-Pulse, for clarity, states are coloured according to the asymptotic $|\Phi_l\rangle$ is not a good quantum number during the Ad-Pulse, for clarity, states are coloured according to the asymptotic. As the $\tau^+$, the sweep saturates, possibly because the end points are not asymptotic. Similar results with $\delta\tau = 1$ ns are obtained for any subset of the $N_{nuc} = 7$ spin cluster investigated here. Coupling strengths are listed in [34].

Posed DD-based polarisation protocols PulsePol [25, 26] or PolCPMG [27] and we investigate possibilities for quantum state storage and read-out.

Floquet theory is well-established in many fields in physics, mostly relating to continuous driving, including applications in NMR [28]. However, the term covers a wide range of scenarios: the Floquet theorem is applicable to any temporally periodic system. One important application is so-called Floquet engineering (FE), [29] where a system driven by a typically strong or high frequency (non-resonant) field can be shown to correspond to an effective, static Hamiltonian with renormalised parameters, by averaging over the period of the driving. Varying the amplitude of the non-resonant drive, one may tune over the effective Hamiltonian. This approach has been proposed theoretically for polarisation of a nuclear spin bath [30] using an adiabatic sweep of the energy eigenvalues of an effective static Hamiltonian.

The Ad-Pulse proposed here is quite distinct from FE: there is no period averaging and in fact one sweeps coherently over the period $T$ of the pulse protocol, over a range containing the
where $E_i \equiv \tan^{-1} \text{Im} \lambda_i / \text{Re} \lambda_i$ is the eigenphase (the Floquet phase).

An NV electron spin system surrounded by $N_{\text{nuc}}$ nuclear spins is described by the Hamiltonian:

$$\hat{H}(t) = \hat{H}_p(t) + \omega_L \sum_n \hat{I}^{(n)}_z + \hat{S}_z \sum_n A^{(n)} \cdot \mathbf{I}^{(n)}$$  \hspace{1cm} (2)

$$\hat{H}_p(t) = \Omega(t) \hat{S}_x$$

is the pulse control Hamiltonian. $\Omega(t)$ is the microwave drive strength which is non-zero during the pulses. For the CPMG sequence, the microwave pulses are applied along the $x$-axis at regular intervals, $\tau$, as shown in Fig. 1.

The pulse duration for a $\pi$-flip of the electron spin is denoted $T_\pi$. $\omega_L$ is the nuclear Larmor frequency; the hyperfine field $A^{(n)}$ felt by the nuclear spin has components $A^{(n)}_x, A^{(n)}_y$ relative to the $z$-axis. Without loss of generality, we take $A^{(n)}_y \equiv A^{(n)}_z$. We omit the $n$ superscript for single-nucleus spin calculations below and denote as $\tau_0$, the resonant pulse spacing for which the electron and nuclear spins entangle.

For CPMG, $\tau_0 = j\pi/(\omega_L + A_z/2)$, where $j$ is an odd integer.

In Fig 1, we illustrate an application of Ad-Pulse using the well-known CPMG-2$N_p$ dynamical decoupling protocol. We consider a bath of 5 nuclear spins, C1,C2...C5 (for list of couplings see table in [34]) from a cluster employed as registers in recent experiments in [20], with $A_z/(2\pi)$ within the range [20 : 60] kHz. The Floquet eigenphases $E_i \equiv E_i(\tau)$ are plotted over a range centered around $\tau = \pi/\omega_L$, for $B_0 = 0.0403$ T. We plot $E_i \in [-\pi : \pi]$ noting the spectra are multiply degenerate under the shift $E_i \rightarrow E_i + 2\pi n$, for integer $n$ [32].

In a standard DD-based sensing protocol, $N_p$ cycles would be applied at each value of $\tau$. The NV electronic spin is reinitialised optically after each value of $\tau$. Ad-Pulse instead sweeps over a range $\Delta \tau$ of pulse spacings without reinitialisation, moving adiabatically on a single Floquet eigenstate in $k = 1, \ldots N_s$ steps. Thus $\tau_{k+1} = \tau_k + \delta \tau$ where $\delta \tau$ is the step in $\tau_k$ and the total sequence time up to step $k$ is

$$t_k = \sum_{l=1}^{l=k} 2\pi N_p \text{ where } \tau_k \equiv \tau(t_k).$$  \hspace{1cm} (3)

For a coherent sweep, the total sweep time is $t_{\text{Ttot}} = \sum_{l=1}^{l=N_s} 2\pi N_p$ and is required to be less than the NV centre’s coherence time under a DD protocol, $T_2$.

A generic feature of the Floquet spectra, for arbitrary number of of nuclei $N_{\text{nuc}}$, is the gap seen between two extremal states which asymptotically tend to maximal/minimal polarised states $M_z = \pm N_{\text{nuc}}/2$; and the other $M_z$ manifolds. Each and every one of the states in the adjoining $M_z = \pm (N_{\text{nuc}}/2 - 1)$ manifolds experiences a coupling $A^{(n)}_z \hat{I}^{(n)}_z \hat{S}_z$ associated with an anti-crossing [32] - and level repulsion- with the extremal states. Hence there is always a gap with the adjoining manifolds.

The lower panel of Fig 1 shows the NV spin coherence $C(\tau) = \langle \hat{S}_z \rangle$ and nuclear bath polarisation $P(\tau) = \frac{1}{N_{\text{nuc}}} \sum_n \frac{2}{\pi} \langle I_z^{(n)} \rangle$ as an adiabatic sweep over $\tau$ is carried out, using the common CPMG protocol. The sweep over a single eigenstate is shown to invert the entire nuclear bath: an initial “down” polarised bath state $|X_{\text{nuc}} \rangle$ follows the Floquet state trajectory to a fully ‘up’ polarised bath state $|X_{\text{nuc}} \rangle$. Provided $t_{\text{Ttot}} \lesssim T_2$, a coherent NV state evolves into another coherent NV state (from $|X_+ \rangle$ to $|X_- \rangle$). Unlike the FE polarisation method [30], the total bath polarisation $M_z h = \sum_n (I_z^{(n)})$ is not a good quantum number, allowing the whole-bath flip illustrated in Fig I

We can use Landau-Zener theory to analyse the adiabatic sweep from an initial $\tau = \tau_{\text{ini}}$ to a final $\tau_{\text{fin}}$. The range $\tau_{\text{ini}} : \tau_{\text{fin}}$ contains the anticrossing region near $\tau \simeq \tau_0$, but $\tau_{\text{ini}}, \tau_{\text{fin}}$ both lie outside it. For a single nuclear spin, for any value of $\tau \in [\tau_{\text{ini}} : \tau_{\text{fin}}]$, we find the probability of losses from the eigenstate trajectory are given by $e^{-\Gamma_{LZ}(\tau)}$, where

$$\Gamma_{LZ}(\tau(t)) = \frac{2A_z^2 \tau_0^2 T_r}{\beta^2 \delta \tau} F(\tau_{\text{ini}}, \tau) \equiv \Gamma_0 F(\tau_{\text{ini}}, \tau)$$

and $\Gamma_0$ is the well-known Landau Zener exponent (see [34] for further details). $F(\tau_{\text{ini}}, \tau)$ is required is another important consideration: i.e., although $\Gamma_0 \propto A_z^2$, since $\Delta \tau \propto A_z$, we find the total time required increases linearly $t_{\text{Ttot}} \propto A_z^{-1}$ [34]. For optimised Ad-Pulse, from Eq 3 we can take $\tau_1 \equiv \tau_{\text{ini}}$ and $\tau_{\text{fin}} \equiv \tau_{N_s}$.

$\beta$ is a protocol dependent parameter. We investigated adiabatic control in CPMG as well as the DD polarisation protocols PulsePol and PolCPMG. PulsePol is a recently proposed robust nuclear polarisation technique with a multi-pulse protocol [20, 25]. For pulse spacings $\tau_0 \simeq j\pi/(2w_1)$, where $j = 1, 3, 5...$ it yields an effective flip-flop Hamiltonian $\hat{H}_{PP} = gI_z S^z$. PolCPMG on the other hand, is a variant of CPMG, obtained by applying an over/under-rotation to the pulses i.e. $\theta = (1+\delta \theta)\pi$ where $\delta \theta = 0$ corresponds to CPMG. It was also shown experimentally to hyperpolarise a nuclear bath [27] for $\delta \theta \simeq 0.05 - 0.25\pi$. $\beta = (\pi + \delta \theta)$ for Ad-PolCPMG/CPMG while $\beta = 6\pi/(2+\sqrt{2})$ for Ad-PulsePol. $T_r = 4\tau_r$ for PulsePol, $T_r = 2\tau_r$ for CPMG/PolCPMG.

Fig I assumed nuclei start from a pure, polarised initial state. Generally the nuclear bath is in a mixture; initialising the NV spin can be achieved optically and is comparitively straightforward. In Fig 2, we show how Ad-Pulse adiabatic sweeps with PolCPMG or PulsePol may also be used to prepare a pure nuclear state from a mixture. We show results for Ad-PolCPMG, but Ad-PulsePol yields similar behaviours.
Fig. 2a (left panels) shows the Floquet spectrum for Pol-CPMG for a single spin for \( \theta_0 = 0.25\pi \). The degeneracy of the level anticrossings in CPMG is lifted into two distinct crossings at \( \tau_{\pm} \), and hence there are two coherence dips in the NV’s trace. Applying PolCPMG at \( \tau_{-} \) (\( \tau_{+} \)) will target the \( |\uparrow\rangle \) (\( |\downarrow\rangle \)) nuclear state and evolve it into the \( |\downarrow\rangle \) (\( |\uparrow\rangle \)) state, polarising the spin. Fig. 2a (right panels) shows the application of Ad-PolCPMG to initialise a single spin and shows the polarisation is well-described by Eq. 4 full polarisation is achieved when the sweep is adiabatic. In fact, if we allow repeated sweeps/reinitialisation of the NV electron spin, we find the spin bath polarisation is robust to a degree of non-adiabaticity.

Fig. 2b examines the case of multiple nuclear spins. The right panel shows the multi-spin Floquet spectra. The levels group into manifolds of different \( M_z \) (for the 5 spins case, \( M_z = -5/2, -3/2, \ldots +5/2 \) and there are 6 manifolds). Hyperfine coupling only allows couplings (or avoided crossings) between neighbouring \( M_z \) and \( M_z \pm 1 \) manifolds.

For the multi-spin Ad-PolCPMG, and the \( \tau = \tau^- \) resonance, we find that about \( r = 10 - 15 \) repetitions is sufficient to fully polarise (> 99.5%), whereas standard PolCPMG requires 100s of repeated sequences and NV re-initialisation. For \( \tau^+ \), a limit corresponding to about 95% polarisation is reached, possibly because the initial and end states do not correspond to the asymptotic \( |X^\pm\rangle|M_z\rangle \) basis. A sweep from \( \tau = 0 \) avoids these issues. Similar behaviour was observed for clusters with \( N_{\text{nuc}} \) up to 7.

If the sweeps are far from adiabatic we may combine the contributions from Eq. 4 additively \( P_{\text{pol}}(t) = \sum_n P_{\text{pol}}^{(n)}(t) \) to accurately describe full simulations. The converse is not true: the highly adiabatic \( \delta \tau = 1 \) ns sweeps in Fig. 2b are not described by Eq. 4 each sweep achieves at most the equivalent of 1 spin-flip (as may be seen from the equispaced \( r = 1, 2, 3 \) sweeps). This is in contrast to the pure state trajectory in Fig. 1 where for the extremal state, the \( X^+ \rightarrow X^- \) corresponds to a whole bath flip.

In Fig. 3 we investigate Ad-PulsePol for quantum state storage. The corresponding Floquet spectrum is shown in Fig. 3a for the three nuclei C1-C3, either as independent spins (top panel) or for the 3-spin cluster (lower panel), for \( B_0 = 0.0403 \) T. For Ad-PulsePol, plotting the eigenphases \( \xi_i \in [-\pi/2 : \pi/2] \) is most insightful, as it shows that the important crossings come in pairs. They are labelled for spin C2 (blue) for the third harmonic (\( j = 3 \) for \( \tau_{\pi} \)) corresponding to \( \hat{H}_{\text{PP}} = gI^+ S^- \). They show that an initial state \( |\uparrow\downarrow\rangle \leftrightarrow |0\uparrow\rangle \), while \( |\downarrow\uparrow\rangle \rightarrow |1\downarrow\rangle \) and \( |0\downarrow\rangle \rightarrow |0\downarrow\rangle \).

Finally, we show that an adiabatic sweep passing through such a pair of crossings would map an arbitrary initial NV state onto the nuclear spin state:

\[
\begin{align*}
[a|0\rangle + b|1\rangle]_{\text{nuc}} \otimes |\downarrow\rangle_{\text{nuc}} &\rightarrow [0]_{\text{NV}} \otimes [a|\downarrow\rangle + b|\uparrow\rangle]_{\text{nuc}} \\
[a|0\rangle + b|1\rangle]_{\text{nuc}} \otimes |\uparrow\rangle_{\text{nuc}} &\rightarrow [0]_{\text{NV}} \otimes [a|\downarrow\rangle + b|\uparrow\rangle]_{\text{nuc}}
\end{align*}
\]

(5)

noting that for the second line, the adiabatic sweep leaves the NV in the \( |1\rangle \) state, so optical reinitialisation to \( |0\rangle \) is assumed. A \( Z \) gate on the nuclei is also assumed: the relative nuclear phases rotate as \( e^{\pm i\omega \mu t} \) thus may be chosen by timing the Larmor precession.

We test this for an adiabatic sweep over \( \Delta \tau \) centered on \( \tau_{PP} \) for spin C1 (\( \tau_{PP}/(2\pi) \approx 20 \) kHz), starting with the test state \( |\Psi_0\rangle = \frac{1}{\sqrt{3}} [0]_\text{NV} \otimes |\downarrow\rangle_{\text{nuc}} \) and testing the fidelity of the overlap \( \mathcal{F}(\tau) = \langle |\Psi_0\rangle |\Psi(\tau)\rangle \) where \( \langle |\Psi_0\rangle |\Psi(\tau)\rangle = |0\rangle_{\text{NV}} \otimes [\frac{1}{\sqrt{3}} |\downarrow\rangle + \frac{2}{\sqrt{3}} |\uparrow\rangle]_{\text{nuc}} \), but disregarding phases between the nuclear states for convenience (by taking modulus of the coefficients of \( \Psi(\tau) \)). The partial overlap between avoided crossings reduces the fidelity appreciably, indicating that although this technique may be used, it is restricted to very well isolated (well-resolved in \( \tau_{PP} \)) nuclear spins. Ad-PolCPMG can equally achieve an equivalent state storage gate.

Conclusions We introduce and discuss the new technique of adiabatic dynamical decoupling and have shown it has a broad range of potential applications that opens several avenues for experimental studies, ranging from polarisation and initialisation of nuclear mixtures to control of single multi-spin pure states for state storage and preparation. There are many different DD protocols and further studies will be re-
quired to identify new types of robust states, including engineered many body Hamiltonian approaches, that will allow adiabatic coherent control on multiple bath spins simultaneously.

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