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Noise-resilient architecture of a hybrid electron-nuclear quantum register in diamond

Michael A Perlin1,2, Zhen-Yu Wang1, Jorge Casanova1 and Martin B Plenio1

1 Institut für Theoretische Physik und IQST, Albert-Einstein-Allee 11, Universität Ulm, D-89081 Ulm, Germany
2 JILA, National Institute of Standards and Technology, and University of Colorado, 440 UCB, Boulder, Colorado 89081, USA

E-mail: zhenyu.wang@uni-ulm.de

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Abstract

A hybrid quantum register consisting of nuclear spins in a solid-state platform coupled to a central electron spin is expected to combine the advantages of its elements. However, the potential to exploit long nuclear spin coherence times is severely limited by magnetic noise from the central electron spin during external interrogation. We overcome this obstacle and present protocols for addressing a decoherence-free nuclear spin subspace which was not accessible by previously existing methods. We demonstrate the efficacy of our protocols using detailed numerical simulations of a nitrogen-vacancy centre with nearby 13C nuclei, and show that the resulting hybrid quantum register is immune to electron spin noise and external magnetic field drifts. Our work takes an important step toward realising robust quantum registers that can be easily manipulated, entangled, and, at the same time, well isolated from external noise, with applications from quantum information processing and communication to quantum sensing.

1. Introduction

A primary goal in the development of quantum technologies, such as quantum information processors, quantum simulators, and quantum sensors, is the design of quantum systems that can be manipulated in a reliable manner and, at the same time, well isolated from unwanted environmental noise. A promising route to this end is the design of hybrid quantum devices which combine different quantum resources with distinct characteristics and advantages. A remarkable platform containing such resources is the nitrogen-vacancy (NV) centre in diamond [1, 2]. The electron spins of NV centres have excellent controllability and can be polarised, detected, and coherently manipulated with high fidelity via optical fields and microwave radiation. The 13C nuclear spins surrounding NV centres, meanwhile, exhibit exceptionally long coherence times [3]. Crucially, the electron-nuclear hyperfine coupling provides a route to selectively manipulate and entangle nuclear spins through the NV centre. Using dynamical decoupling (DD) control sequences [4–9], the characteristic frequency of the NV electron spin can be tuned to the precession frequency of a particular nucleus, thereby coupling the NV electron and nuclear spins through the electron-nuclear hyperfine interaction. This individual addressing [9–11] allows for the realisation of decoherence-protected quantum gates [12], quantum error correcting codes [13–15], quantum computing [16, 17], and quantum simulation protocols [18] in the hybrid system of a NV centre and its surrounding nuclei. In addition, the NV electron spin provides an optical interface to entangle spins with photons [19], which together with the available nuclear memory establishes a key prerequisite for scalable quantum architectures [20]. To this end, recent remarkable experiments have established photon-mediated entanglement between distant NV quantum-network nodes [21, 22], as well as entanglement distillation on entangled spin qubits [23].

The same electron–nuclear hyperfine coupling which is central to this hybrid quantum system, however, also dephases nuclear spins [24] due to, for example, electron spin relaxation. More specifically, the electron undergoes stochastic spin flips ($T_1$ processes), thereby acting as a source of magnetic noise for the nuclear spins.
via the hyperfine coupling. This noise is particularly severe during optical initialisation and readout of NV electrons, as well as during the reset for establishing electron–photon entanglement [25]. Electron spin noise thus limits the use of the NV centre together with surrounding nuclear spins as quantum resources for performing information processing and sensing tasks. While nuclear spins with weaker hyperfine coupling to the NV centre are less sensitive to electron spin flip noise, weak coupling also unavoidably implies poor nuclear spin controllability, which again limits their utility for quantum memory registers. Moreover, current protocols [6–11] for selective addressing and control of nuclear spins rely on the distinct nuclear precession frequencies \( \omega_j \) with \( j \) indexing a particular nucleus. Therefore, the time to selectively address a nucleus with a frequency \( \omega_j \) can not be shorter than \( 1/|\omega_j - \omega| \) for all other nuclei \( j \neq 1 \) coupled to the NV electron. Because the nuclear precession frequency differences are produced by the hyperfine interactions at the locations of the nuclei, current selective addressing protocols for \(^{13}\text{C} \) spins thus prefer stronger hyperfine interactions, unavoidably imposing stronger electron spin noise on the \(^{13}\text{C} \) nuclei.

The trade-off between noise strength and gate times is generally unavoidable for control methods based on spectroscopic discrimination [6–11], even when using encoded nuclear spin subspaces. By pairing nuclear spins, for example, one can reduce the magnitude of NV electron spin noise on a logical qubit in a decoherence-protected subspace (DPS) [25] (see figure 1(b)). As existing methods [25–27] to access this DPS still rely on distinct nuclear precession frequencies \( \omega_1, \omega_2 \) for selective addressing and control, however, the strength of noise acting on the DPS is proportional to \( \delta_{1,2} = |\omega_1 - \omega_2| \), which cannot be made too small because the time to distinguish the two nuclei has to be \( \gtrsim 1/\delta_{1,2} \). Very recent works [28, 29] propose dissipative preparation of nuclear singlet states by frequent reset on the NV electron spin. These procedures, however, would also destroy the electron spin state, and lack the ability to manipulate nuclear spins for storage of quantum information.

In this work, we address the problem of selectively manipulating nuclear spins with identical precession frequencies, i.e. \( \omega_1 = \omega_j \), by combining DD techniques with radio-frequency (RF) nuclear spin control. As a result, we are able to construct an accessible decoherence-free subspace (DFS) [30] in a solid-state platform, and thereby realise a robust hybrid quantum register which is resilient to dephasing processes such as NV initialisation and readout, in addition to external magnetic field noise (see figure 1). We provide a protocol for storage and retrieval of information from this DFS, and test its efficacy under realistic conditions via detailed numerical simulations.

### 2. Noise from electron-nuclear coupling

In the typical situation with the electron–nuclear flip-flop terms suppressed by a large energy mismatch, on the order of GHz in the case of an NV centre and nearby \(^{13}\text{C} \) nuclei, the hyperfine interactions between the electron and nuclear spins reads [1] \( H_{\text{hf}} = S_z \sum_j \vec{A}_j \cdot \vec{I}_j \), where \( S_z \) and \( \vec{I} \) are respectively electron and nuclear spin operators. Denoting by \( \hat{z} \) a unit vector along the NV symmetry axis, the hyperfine fields \( \vec{A}_j \) can be decomposed into parallel and perpendicular components \( A_j^\parallel = \vec{A}_j \cdot \hat{z} \) and \( A_j^\perp = \vec{A}_j - A_j^\parallel \hat{z} \). In the presence of a static magnetic field \( B_z \hat{z} \) with strength \( B_z \gg A_j^\parallel /\gamma_n \) where \( A_j^\parallel = |\vec{A}_j| \) and \( \gamma_n \) is the nuclear \(^{13}\text{C} \) gyromagnetic ratio,
the perpendicular components $\vec{A}_j^\perp \cdot \vec{I}_j = A_j^\perp I_j^z$ in $H_{\text{hf}}$ are suppressed, resulting in the coupling $H_{\text{hf}} = \sum_j A_j^\perp I_j^z$, where $I_j^z = \hat{I}_j \cdot \hat{z}$. While nuclear spins are excellent candidates for storing quantum information, uncontrolled electron spin flips generate random noise with the strengths $A_j^\parallel$ on the nuclei through the coupling $H_{\text{hf}}$.

In the following, we demonstrate how to reliably control a DFS with $\delta_{1,2} = 0$ under realistic experimental conditions, i.e., in the presence of noisy nuclear spins and imperfect control.

3. Larmor pairs and their identification

It is possible for multiple $^{13}$C nuclear spins to share the same hyperfine components $A_j^\parallel = A_i^\parallel$ and $A_j^\perp = A_i^\perp$, e.g., due to the symmetries of the diamond lattice. Such nuclei manifestly satisfy the necessary conditions for containing a DFS. We refer to exactly two $^{13}$C nuclei in such a symmetric configuration (see figure 1) as a Larmor pair, and observe their relatively large probability of occurrence in an NV system with natural $^{13}$C abundance (see figure 2). Because the parallel component $A_i^\parallel$ for the Larmor pair is different from $A_j^\parallel$ of other spins, we can use existing methods based on spectroscopic discrimination [6–11] to address and to identify the Larmor pair via the NV electron.

To selectively couple the NV electron to only the Larmor pair, we use DD sequences on two electron spin levels $|0\rangle$ and $|m_i\rangle (m_i = +1$ or $-1)$ to realise the effective interaction Hamiltonian (see appendix)

$$H_{\text{int}} = \frac{1}{4} f_{\text{int}} A_i^\parallel \sigma_z (I_i^x + I_i^z),$$  \hspace{1cm} (1)

where $\sigma_z$ is the two-level electron spin Pauli operator and $f_{\text{int}}$ is a tunable parameter provided by the adaptive-XY (AXY) sequence [9]. Note that the spin operators $I_i^x$ are quantised along the azimuthal directions of their

![Figure 2. Probability to find at least one Larmor pair near an NV electron spin.](image-url)
respective local hyperfine fields $A_j^\pm$. The Hamiltonian in equation (1) leads to the entangling gate

$$U_{\text{int}}(\theta) = \exp \left[ -i\theta \sigma_z (I_1^x + I_2^x) \right]$$

(2)

with an interaction time $4\theta / (f_{\text{RF}} A^\pm)$. Note that the gate in equation (2) is realised in a decoherence-protected manner; that is, the DD sequence suppresses dephasing noise from e.g. unwanted nuclear spins and, at the same time, realises non-trivial addressing of the target nuclear spins [6–10, 12, 13].

To find a Larmor pair, we can use the delayed entanglement echo (DEE) recently developed in [11] (see figure 3(a) for a DEE signal spectrum with a single spin versus with a Larmor pair), or the resonance fingerprints in [31] to find spectrally resolvable resonances and identify the number of nuclear spins within them. Having found a Larmor pair, we can measure the corresponding hyperfine fields $A_j^\pm$ via our recently developed nuclear positioning method in [10]. Therein, we turn on an external RF field on resonance with the nuclear spin precession frequency. In the rotating frame of the nuclear spin precession, the phase of this RF field corresponds to an effective static magnetic field direction $\phi_{\text{RF}}$ in the $x$–$y$ plane (see figure 3(b)). When the effective RF field direction is parallel to $A_j^\perp$, the electron–nuclear spin coupling for spin $j$ is not affected. In general, the effective RF field will suppress coupling along directions orthogonal to the effective field axis. By varying the phase of the RF field, one can thus measure the positions of both nuclei in the Larmor pair. This measurement can be simplified by taking symmetries of the diamond lattice into account (see figures 3(c), (d) for the NV transition signal in a strong magnetic field). The directions of $A_j^\perp$ can be firstly identified by the signal reduction in figure 3(c) where the signal is not very strong, and can be more accurately measured by using longer interaction times or a stronger coupling as in figure 3(d). The ambiguity of $180^\circ$ in figures 3(c), (d) can be eliminated by checking the NV transition signal in the presence of a weak magnetic field ($B_z \sim A_j^\perp /\hbar$) [10]. As we will see, however, resolving this ambiguity is not necessary for our protocol.

### 4. Controlling spectrally-indistinguishable nuclear spins

The symmetry between nuclear spins in equation (1) can be broken by a RF control field, allowing for individual control of each nucleus in a Larmor pair. Using the technique outlined in [10], we can decouple all electron–nuclear interactions, and introduce RF control fields targeting the desired Larmor pair in a coherence-protected manner to realise the Hamiltonian

$$H_{\text{RF}} = \Omega (\phi_{\text{RF}}) \cdot (I_1^x + I_2^x) = \Omega (I_1^x \cos \phi_1 + I_2^x \sin \phi_1),$$

(3)

where $\phi_1$ are the azimuthal angles of the perpendicular components of the local hyperfine fields (see figure 3(b)); $I_j^\phi = I_j^x \cos \phi + I_j^y \sin \phi$ and $\Omega, \phi_{\text{RF}}$ are respectively the Rabi frequency and phase of the RF drive. Note that the spin operators $I_j^\phi$ are quantised along the azimuthal directions of their respective local hyperfine fields $A_j^\perp$. By combining the non-selective controls of $H_{\text{RF}}$ and $H_{\text{int}}$, we can selectively address only one of the two nuclei in the Larmor pair. Without loss of the generality, we let the index $j = 1$ for the target nucleus.
The control in equation (3) allows us to apply \( \pi \) pulses on both nuclei simultaneously by applying the RF drive for a time \( \pi / \Omega \). Using \( \phi_{\text{RF}} = \phi_2 + \pi / 2 \) and defining \( \alpha = \phi_2 - \phi_1 + \frac{\pi}{2} \), we can thus generate the gate

\[
U = e^{-i \phi_2 I_2^Z} e^{i \phi_1 I_1^Z}.
\]

One can then show that the sequence \( U_{\text{DEE}} = U_{\text{int}}(\phi)U_rU_{\text{int}}(\theta) \) leads to the evolution \( U_{\text{DEE}} = e^{-i \phi_2 I_2^Z} \), where \( U_1 = e^{-i \phi_1 I_1^Z} e^{i \phi_1 I_1^Z} e^{-i \phi_1 I_1^Z} \) entangles only the target nucleus with the NV electron. We can cancel the single-qubit operation on the second spin by repeating the sequence \( U_{\text{DEE}} \) twice to get

\[
U_{\text{DEE}}^2 = -2i(r_a I_a^Z + r_b I_b^Z)\sigma_z + \cos^2 \theta - \cos(2\alpha)\sin^2 \theta,
\]

where we define \( r_a = 2 \cos \alpha \sin \theta [\cos^2 \alpha \cos \theta + \sin^2 \alpha] ; r_b = 4 \cos^2 \alpha \sin \theta \sin \theta [\theta^2] ; \) and \( \alpha = \alpha + \pi / 2 \). The last line of equation (4) vanishes when

\[
\cos^2 \alpha \sin \theta = \frac{1}{2},
\]

which can be satisfied by choosing a suitable value of \( \theta \) if

\[
|\sin(\phi_2 - \phi_1)| \geq 1 / \sqrt{2}.
\]

While we will work within the constraint of equation (6), we note that it is possible to relax this constraint by applying additional repetitions of \( U_{\text{DEE}} \) (see supplementary materials is available online at stacks.iop.org/QST/4/015007/mmedia). By using a value of \( \theta \) which satisfies equation (5), we can implement a selective \( \pi \) rotation \( R_z \) on the target spin along a direction perpendicular to \( \hat{z} \) by applying \( U_{\text{DEE}}^7 \) followed by a \( \sigma_z \) gate on the electron spin, i.e.

\[
R_z(\beta) = \sigma_z U_{\text{DEE}}^7 = e^{-i \beta I_1^Z},
\]

where the azimuthal angle \( \beta \) can be determined from equations (4) and (5). By adding a delay window in which DD is used to negate electron-nuclear coupling, the nuclear spins are rotated around their quantisation axis and hence the rotation equation (7) can be shifted to \( R_z(\phi) \) for any angle \( \phi \). From equation (4) and the definition of \( \alpha \), we can see that the ambiguity of \( \pi \) in \( \phi_1 \) and \( \phi_2 \) (see figures (b)–(d)) is inconsequential as it merely introduces a possible global phase in the nuclear spin gate equation (7).

The selective rotation \( R_z(\beta) \) in equation (7) is essential for our following protocols to manipulate a nuclear DFS, so it is important to verify that this operation can indeed be performed with high fidelity. The elementary gates which are used to construct \( R_z(\beta) \), namely \( U_{\text{int}}(\theta) \) and \( U_r \), can be implemented by existing DD methods, which suppress perturbations from environmental spins [6–10, 12, 13]. The use of robust DD sequences can further compensate for control errors [5, 9]. We perform numerical simulations to demonstrate the control \( R_z(\beta) \) on a Larmor pair via AXY-8 sequences [9]. Supplementary materials shows our simulated fidelities of \( R_z(\beta) \), which are above 0.99 for a wide range of detuning and amplitude errors in the DD pulses. As detailed in Supplementary Materials, these gate fidelities are achieved in the presence of additional nuclei which introduce noise on the NV electron spin coherence and the selective nuclear spin addressing protocol. The entire protocol in the simulation uses 880 DD pulses and a total time \( \approx 1 \) ms, which are of orders of magnitude smaller than the number (e.g., 10240) of pulses and coherence times \( T_2 > 1 \) s reported in recent experiments [32]. Because the NV electron spin relaxation time \( T_1 > 1 \) h at low temperatures [32], this relaxation process is neglected in our gate simulations.

Using equation (7), we can implement a selective controlled gate on only one spin in a Larmor pair, such as

\[
U_{\text{ent}} = [i \sigma_z R_z(\pi/2) e^{-i \theta t} ]^2 = \sigma_z R_z(\pi/2) e^{-i \theta t},
\]

by using two blocks of the control-free Hamiltonian (see methods)

\[
H_{\text{free}} = \frac{1}{2} \sigma_z \sum_j A_j^Z I_j^Z + \Delta \sum_j I_j^Z,
\]

where \( \Delta \) corresponds to a shift from the use of a rotating frame during the application of \( H_{\text{free}} \). The gate \( U_{\text{ent}}^\prime \) cancels the electron-nuclear coupling with all nuclear spins except that addressed by \( R_z \). Neglecting the evolutions of other nuclear spins outside the Larmor pair, \( U_{\text{ent}}^\prime \) is thus the controlled phase gate

\[
U_z = \exp[-i \phi A_1^Z \sigma_z I_1^Z] \exp[-i 2 \pi \Delta I_1^Z],
\]

with a tunable single nuclear spin operation. A single nuclear spin operation \( e^{i \theta I_1^Z} \) can also be realized by the sequence \( R_z(\phi_1) R_z(\phi_2) \) with \( \phi_2 - \phi_1 + 2 \pi = \phi / 2 \).

5. Nuclear spin DFS in solids

Using selective control on each of the spins in the Larmor pair, we can construct a DFS in the span of \( \{ | +1 1 \rangle, | +1 0 \rangle \} \) within their joint Hilbert space. Collective dephasing of nuclear spin states which results from
coupling to the NV centre and external fields yields an identical phase factor on all states in this DFS. The coherence of states in the DFS is insensitive to optical and microwave control on the NV electron spin, yielding much longer coherence times than the case of a DPS, as shown in figure 4.

One can initialise the Larmor pair into a state within the DFS in a variety of ways, e.g. by first polarising the nuclear spins to $|\downarrow_1 \downarrow_2\rangle$ via swapping with the polarised NV electron spin (see [11] for a protocol), and then using the nuclear-selective $\pi$ rotation in equation (7) to flip the first spin, thereby preparing the state $|\uparrow_1 \downarrow_2\rangle$.

Below, we describe protocols for storage and retrieval of quantum information from the DFS, using similar ideas in [33]. These protocols use our selective control in equation (7), in addition to established methods of electron spin initialisation, readout, and the electron-nuclear spin gate in equation (2). The protocols have the advantage that the dominant electron spin noise from NV initialisation and readout does not dephase the quantum information.

5.1. Storing the electron spin state in a DFS

A quantum state of the NV electron spin takes the general form $c_0 |0\rangle + c_1 |1\rangle$, where $c_{0(1)}$ can be complex numbers or $c_{0(1)} = \tilde{c}_{0(1)} |\tilde{\varphi}_{0(1)}\rangle$ can include the states $|\tilde{\varphi}_{0(1)}\rangle$ of other quantum systems, e.g., other remote electron or nuclear spins in a quantum network. When $|\tilde{\varphi}_0\rangle = |\tilde{\varphi}_1\rangle$ and the complex numbers $\tilde{c}_{0(1)} = 0$, the NV electron spin is initially entangled with other quantum systems.

The NV electron spin state can be stored in the DFS in the following steps. With the Larmor pair initialised to $|\uparrow_1 \downarrow_2\rangle$, we apply the conditional evolution $U_{\text{DFS}}(z)$ (equation (2)) to achieve the entangled electron-nuclear state $c_0 |0\rangle |\tilde{\varphi}_0\rangle + c_1 |1\rangle \tilde{\varphi}_1\rangle$, where $|\tilde{\varphi}_i\rangle$ denotes $\exp(\pm i z T) |\tilde{\varphi}_i\rangle$. Then we apply a Hadamard gate $H_y$ (i.e., a $\pi/2$ pulse applied along the $y$ direction) on the NV electron qubit followed by another $U_{\text{DFS}}(z)$ to get $\frac{1}{\sqrt{2}}(|0\rangle \ket{\psi_1} - |0\rangle \ket{\psi_0})$, where $|\psi_1\rangle = c_0 |\uparrow_1 \downarrow_2\rangle - c_1 |\downarrow_1 \uparrow_2\rangle$ and $|\psi_0\rangle = c_0 |\downarrow_1 \uparrow_2\rangle + c_1 |\uparrow_1 \downarrow_2\rangle$ are the states stored in the DFS. A projective measurement on the NV electron spin then stores the quantum information in the DFS qubit as $|\psi_{\text{DFS}}\rangle$ for the outcome $m = 0(1)$. The storage in $|\psi_{\text{DFS}}\rangle$ is equivalent to the case of $|\psi_{\text{DFS}}\rangle$ up to a single-qubit operation.

5.2. Retrieving a quantum state from the DFS

To retrieve the quantum information stored in the DFS, say, $c_0 |\uparrow_1 \downarrow_2\rangle + c_1 |\downarrow_1 \uparrow_2\rangle$ (which can be an entangled state as in the storage protocol), we can use the following steps: (i) We initialise the electron spin state to the superposition $|x^+\rangle = |0\rangle + |1\rangle \sqrt{2}$. (ii) We apply a selective $\pi$ rotation as equation (7) to flip the second spin and get $c_0 |\uparrow_1 \downarrow_2\rangle + c_1 |\downarrow_1 \uparrow_2\rangle$. (iii) We apply a standard DEE protocol as in [11] to achieve the interaction $A^\dagger \sigma_z (I_x^+ + I_y^+)$, which implements a conditional phase gate on the electron spin and yields $c_0 |\chi^+_0\rangle |\uparrow_1 \downarrow_2\rangle + c_1 |\chi^+_1\rangle |\downarrow_1 \uparrow_2\rangle$, where $|\chi^+_i\rangle = \exp(\mp i \pi \sigma_2) |x^+_i\rangle$. (iv) We apply a $\pi/2$ pulse on the electron spin to get $c_0 |\uparrow_1 \downarrow_2\rangle - c_1 |\downarrow_1 \uparrow_2\rangle$. (v) Finally, we use the conditional evolution $U_{\text{DFS}}(z)$ (Equation (2)) to retrieve the state as $(c_0 |0\rangle + c_1 |1\rangle) |\chi^+_0\rangle |\chi^+_0\rangle$, where the nuclear spin states can be reset to the state $|\uparrow_1 \downarrow_2\rangle$ in the DFS by a $\pi$ pulse as in equation (7).
effects of experimental errors

The overall fidelities of the storage and retrieval protocols provided above are determined by the fidelities of state initialization, gate operations, and of state readout. A probability of not getting the right initial state will reduce the success rate and hence the overall fidelity. For NV centres, the fidelity of electron spin preparation can be larger than 0.99 [36]. Gate operations can have a high fidelity by using robust pulse sequences developed in the fields of magnetic resonance [37] and DD [5, 9]. With the use of AXY sequences, the combined fidelity after the gate operations for storage or retrieval can be higher than 0.99, even with relatively large control errors (see supplementary materials). Because the protocols are implemented in a coherence protected manner and the times for storage and retrieval are orders of magnitude smaller than reported coherence times (e.g. over one
second in a diamond with the natural abundance 1.1% of $^{13}$C [32], the errors due to electron spin flip noise and the nuclear spin bath are negligible. For NV centres, the main factor limiting the overall fidelity could be the single-shot readout, with a fidelity of $\sim 0.93$ reported in [36]. Single-shot readout fidelities above 0.99 might be achievable by coupling the NV centre to an optical cavity [20].

7. Effects of inter-nuclear interactions

Inter-nuclear interactions, which are the remaining source of noise acting on the DFS qubits, can be suppressed by DD on nuclear spins. These interactions are weak, e.g. with a strength $g_{n}$, of at most $2\pi \times 50$ Hz for the samples counted in figure 2. In the protocol to realise large-scale graph states, the times required to implement the electron and nuclear spin gates can be short enough to neglect inter-nuclear interactions. However, if this noise is not properly suppressed, it can dephase the DFS qubit after sufficiently long storage times. Because the Larmor pair has a precession frequency that is different from the frequencies of bath spins by a minimum amount $\delta_{\text{min}}$ of a few to tens of kHz (see figure 2), the noise from bath spins does not flip the nuclear spins of the Larmor pair, and only acts on the individual spin operators $I^{e}$ and $I^{n}$ in the Larmor pair. Furthermore, the large separation of scales between Larmor precession frequency differences $\delta_{\text{min}}$ and nuclear-spin coupling strengths $g_{n}$ leaves room for bath-selective nuclear-spin $\pi$ pulses using external RF fields with Rabi frequencies $\Omega$ satisfying $g_{n} \ll \Omega \ll \delta_{\text{min}}$ (e.g., $\Omega \sim 2\pi \times 0.5$ kHz). As a consequence, one can apply DD sequences on the nuclear spin bath [38] to suppress this noise on DFS qubits during long-time information storage.

The dipolar coupling between the two spins of the Larmor pair splits the energies of the states $|\pm\rangle = \frac{1}{\sqrt{2}} (|\uparrow\rangle \pm |\downarrow\rangle)$ of the DFS qubit. This constant splitting can be determined by measuring the positions of the Larmor pair, which can be performed to high accuracy due to the lattice structure of diamond [10, 39]. With a known value of the splitting, its effect on the DFS qubit can be removed in an appropriate rotating frame. Alternatively, one can eliminate the effect of this coupling by a Hahn echo (i.e. DD with one $\pi$ pulse) on the levels $|\pm\rangle$, e.g. via the use of a single nuclear spin gate $e^{i\pi I_{z}}$ to exchange $|\pm\rangle \leftrightarrow |\mp\rangle$.

8. Discussion

We have shown that electron-nuclear hybrid quantum registers can be made free from external magnetic noise and internal electron spin noise by using our proposed control protocols. Combining recently developed DD techniques with RF control fields, we overcome fundamental limitations of previous methods for nuclear spin control based on spectroscopic discrimination, which allows us to selectively address nuclear spins with identical Larmor precession frequencies in the vicinity of an NV centre. As a consequence, our scheme enables the construction of an addressable DFS and makes a major step towards the realisation of robust quantum registers. In this regard, we have provided a protocol to store quantum states of the NV electron in a DFS, a complementary protocol to retrieve a quantum state from the DFS back into the NV electron spin, and a recipe for constructing long-lived, large-scale graph states. Our schemes can be realised via existing experimental techniques. For example, by using DD, which is also incorporated in our scheme, a recent experiment [32] has demonstrated a coherence time exceeding one second for a single NV electron spin and the control of 19 nuclear spins in a diamond with a natural 1.1% abundance of $^{13}$C.

The work we have developed has important applications. For example, our work allows to create large-scale many-body quantum states distributed over multiple quantum network nodes, e.g., for quantum communication and quantum computing tasks [34, 35], because the entangled-state decoherence rate in the DFS is negligible compared with the entanglement-generation rate 39 Hz reported in an NV quantum network [22]. In quantum sensing, our work could significantly enhance existing proposals to use the NV electron spin together with surrounding $^{13}$C nuclear spins as a memory-assisted nano-scale sensor, as such sensors generally limited by the coherence time of the available quantum memory [11, 40]. For the purposes of computing, it is also possible to incorporate additional $^{13}$C nuclei or a $^{15}$N nuclear spin into the electron-nuclear hybrid module so as to include auxiliary processing qubits for performing fast few-qubit gate operations through the NV centre. This capability is enhanced by the insensitivity of our DFS qubit to magnetic field noise from the NV centre, which allows the previously limited parallel use of the NV electron and its surrounding nuclear spins as quantum resources. Moreover, our methods are general and may be applied to other colour centres such as silicon carbide [41] and other scalable architectures [26].
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Appendix. Electron-nuclear Hamiltonian

Under a magnetic field $B_z \hat{z}$ along the NV symmetry axis, the electron-nuclear coupling is described by the Hamiltonian ($\hbar = 1$)

$$H = S_z \sum_j A_j \cdot \hat{I}_j - \sum_j \gamma_n B_z I^z_j. \quad (11)$$

where $\hat{I}_j = I^x_j \hat{x} + I^y_j \hat{y} + I^z_j \hat{z}$ are nuclear spin operators, $S_z = \sum_{m_z=\pm1,0} |m_z\rangle \langle m_z|$ is the electron spin operator, $\gamma_n$ is the nuclear $^{13}$C gyromagnetic ratio, and the hyperfine fields $A_j$ have components $A^j_1 = A_j \cdot \hat{z}$ and $A^j_2 = A^j_1 \times \hat{x}$. We consider a strong magnetic field $B_z \gg \gamma_n$.

Working in the electron spin levels $|0\rangle$ and $|m_z\rangle$ with $m_z = +1$ or $-1$, equation (11) becomes

$$H' = \frac{1}{2} \sigma_z \sum_j A_j \cdot \hat{I}_j - \sum_j \omega_j I^z_j. \quad (12)$$

Here we define the electron spin Pauli operator $\sigma_z = m_z (|m_z\rangle \langle m_z| - |0\rangle \langle 0|)$. The vectors $\hat{\omega}_j \equiv \gamma_n B_z \hat{z} - \frac{1}{2} m_z \hat{A}_j \approx \omega_j \hat{z}$, because of the strong magnetic field $B_z$.

Without control on the NV electron spin, the Hamiltonian in a rotating frame with respect to the nuclear spin Hamiltonian $-\sum_j \omega_j I^z_j$ for $\omega_j = \omega + \Delta_{\text{rot}}$ becomes

$$H'_{\text{free}} = \frac{1}{2} \sigma_z \sum_j A^1_j I^{1z}_j \cos(\omega_j t) + I^{1z}_j \sin(\omega_j t)$$

$$+ \frac{1}{2} \sigma_z \sum_j A^2_j I^{2z}_j + \Delta_{\text{rot}} \sum_j I^z_j. \quad (13)$$

Applying the rotating wave approximation to remove the oscillating terms in equation (13), we have the control-free Hamiltonian $H'_{\text{free}} = \frac{1}{2} \sigma_z \sum_j A^1_j I^{1z}_j + \Delta_{\text{rot}} \sum_j I^z_j$, i.e., equation (9) in the main text. Implementing the Hamiltonian $H'_{\text{free}}$ for a time $\tau$, a phase shift of $\Delta_{\text{rot}} \tau$ will be added to the phases of subsequent RF controls.

Selective coupling to only the nuclear spins with a frequency $\omega$ can be realised by DD control on the NV electron spin. Applying DD pulses on the electron spin transforms the spin operator $\sigma_z$ as $\sigma_z \rightarrow F(t) \sigma_z$, where the modulation function $F(t) = (-1)^{n(t)}$ when a number $n(t)$ of $\tau$ pulses have been applied in the time $t$. In the rotating frame with respect to the nuclear spin Hamiltonian $-\sum_j \omega_j I^z_j$ (i.e., $\Delta_{\text{rot}} = 0$), we have

$$H'_{\text{DD}} = \frac{1}{2} F(t) \sigma_z \sum_j A^1_j I^{1z}_j \cos(\omega_j t) + I^{1z}_j \sin(\omega_j t)$$

$$+ \frac{1}{2} F(t) \sigma_z \sum_j A^2_j I^{2z}_j. \quad (14)$$

We choose $F(t)$ to be of the form $F(t) = \sum_k f_k \cos(k \omega_{\text{DD}} t)$ in which $f_k = 0$ for even $k$. To selectively address the Larmor pair by the DD, we tune the $k_{\text{DD}}$-th harmonic on resonance with the nuclear frequency $\omega_j$ of the Larmor pair, i.e., $k_{\text{DD}} \omega_{\text{DD}} = \omega_j$. For CPMG or the XY family of sequences, $\omega_{\text{DD}}$ is fixed by the application of $\pi$ pulses at times $t_p = \pi (p - 1/2)/\omega_{\text{DD}}$, $p = 1, 2, \ldots$. The AXY sequences [9] allow for tuning $f_k$. As detailed in [9, 10], applying the rotating wave approximation to remove the oscillating terms in equation (14) will give equation (1) in the main text.

ORCID iDs

Michael A Perlin @ https://orcid.org/0000-0002-9316-1596
Zhen-Yu Wang @ https://orcid.org/0000-0002-8995-9669

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