Parallel selective nuclear spin addressing for fast high-fidelity quantum gates

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Due to their long coherence times, nuclear spins have gained considerable attention as physical qubits. Two-qubit gates between nuclear spins of distinct resonance frequencies can be mediated by electron spins, usually employing a sequence of electron-nuclear gates. Here we present a different approach inspired by, but not limited to, NV centers in diamond and discuss possible applications. To this end we generalize external electron spin control sequences for nuclear spin initialization and hyperpolarization to achieve the simultaneous control of distinct nuclear spins via an electron spin. This approach results in efficient entangling gates that, compared to standard techniques, reduce the gate time by more than 50% when the gate time is limited by off-resonant coupling to other spins, and by up to 22% when the gate time is limited by small electron-nuclear coupling.

INTRODUCTION

Spin carrying material impurities represent a promising platform for various near term applications of quantum technology. In particular, the nitrogen-vacancy (NV) center has proven itself to be a valid candidate for quantum computation, communication and sensing applications [1–4]. Recent progress manifests in a number of works, including NV-nuclear quantum gates [5], NV-NV gates [6], sensing of single nuclei [7] and polarization of spin ensembles [8, 9]. However, the coherence time of NV-centers limits achievable fidelities. Nuclear spins in diamond may be fully controlled by NV centers and offer far longer coherence times due to their smaller gyromagnetic ratio which renders them less susceptible to environmental noise and thus as ideal candidates for physical qubits, e.g. for quantum memories with long storage times.

Numerous approaches to realize gates between NV centers and nuclear spins have been presented, both with [10, 11] and without additional external radiofrequency (RF) control [12–14] on the nuclear spins themselves. Their common foundation are pulsed dynamical decoupling sequences which exhibit resonance frequencies that are multiples of $\pi/\tau$ where $\tau$ is the time between subsequent pulses. At present, these sequences are tuned to a single resonance which implies the major drawback that only one nuclear spin can be controlled at a time.

In this work, we overcome this limitation with an approach based on polarization sequences [9] that originally have been developed to initialize nuclear spins [8, 15]. Our modifications allow for simultaneously resonant addressing of two different frequencies at the same time without requiring additional RF control, using a sequence with two free parameters incorporated by a pulse spacing and pulse phases. This paves the way for a wide range of applications, such as the realization of gates for quantum computation applications or the protection from magnetic field fluctuations. These are not limited to diamond based material, for example carbon-13 and silicon-29 spins with silicon vacancies in silicon carbide [16] can be controlled jointly. The approach presented here can be used to initialize and manipulate different nuclear spin species simultaneously, extending approaches for quantum simulators made of NV-controlled nuclei [17].

We start by explaining the basic pulse sequences that create the desired Hamiltonian and continue with modifications that allow to manipulate the effective coupling strength in this Hamiltonian. These ideas are then applied to investigate the efficiency with which our protocol can create nuclear spin entanglement in two relevant systems, namely between a silicon-29 and a carbon-13 spin like inside silicon-carbide and between carbon-13 spins close to an NV-center in diamond that only differ by their hyperfine coupling to the electron spin. Furthermore we discuss the manipulation of states that are insensitive to magnetic field fluctuations, which therefore offer excellent coherence properties for quantum sensing and computation applications. Finally we compare our approach to standard techniques for different experimentally relevant situations.

TUNING PULSED POLARIZATION SEQUENCES RESONANT TO TWO ARBITRARY FREQUENCIES

We assume an NV center placed at $\vec{r} = 0$ in a magnetic field $B(\vec{r}) = B_z(\vec{r}) \hat{z}$, which is aligned with its symmetry axis $\hat{z}$. Under the application of an external microwave

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drive $H_{mw}(t)$, the total Hamiltonian then reads
\begin{equation}
H(t) = DS_z^2 + \gamma_e B_z(0)S_z + \sum_n \gamma_n B_z(\vec{r}_n) I_n^z + S_z \sum_n \vec{A}_n \cdot \vec{I}_n + H_{mw}(t),
\end{equation}
where $S_z$ is the spin-1 $z$-operator of the NV and we have the nuclear spin-1/2 operators $I_n^z = \sigma_n^z/2$, $i = x, y, z \text{ with } \sigma_i$ the corresponding Pauli matrix. Furthermore, we have the electronic (nuclear) gyromagnetic ratio $\gamma_e (\gamma_n)$. The large zero field splitting $D = 2\pi \times 2,87 \text{ GHz}$ permits application of the secular approximation, hence the coupling of each nuclear spin to the electron spin is solely determined by the hyperfine vector $\vec{A}_n$ given by
\begin{equation}
\vec{A}_n = \frac{\mu_0 \gamma_n \gamma_e}{4\pi|\vec{r}_n|^3} \left[ \vec{z} - 3 \left( \frac{\vec{z} \cdot \vec{r}_n}{|\vec{r}_n|^2} \right) \right],
\end{equation}
where $\vec{r}_n$ describes the position of the corresponding nucleus. We now reduce the electronic Hilbert space to the space spanned by $|m_s = 0\rangle$ and $|m_s = +1\rangle$, effectively choosing our working qubit. Adapting the microwave control to drive transitions between those levels, it takes the form
\begin{equation}
H_{mw}(t) = 2\Omega(t) \cos(\omega_{mw} + \varphi)\sigma_x,
\end{equation}
where $\Omega(t)$ is a time dependent Rabi frequency and $\sigma_x = |0\rangle\langle+1| + |+1\rangle\langle0|$. In a frame rotating with respect to the zero field splitting and the microwave frequency $\omega_{mw}$, the total Hamiltonian can be written as $H = H_0 + H_{mw}$, where
\begin{equation}
H_0 = \sum_n \omega_n \vec{I}_n + \frac{1}{2} \sigma_z \sum_n \vec{A}_n \cdot \vec{I}_n,
\end{equation}
\begin{equation}
H_{mw} = \Delta \sigma_z/2 + \Omega(t) \left[ \sigma_x \cos(\varphi) + \sigma_y \sin(\varphi) \right] / 2
\end{equation}
and the strongly oscillating terms are neglected. Here we defined $\sigma_\pm = |+1\rangle\langle-1| - |\pm1\rangle\langle0|$ and $\sigma_\mp = i|\mp\rangle\langle\pm1| - i|\pm\rangle\langle\mp1|$. The detuning from the electronic transition frequency is denoted as $\Delta$ and the phase $\varphi$ of the drive selects the axis around which the qubit’s Bloch vector rotates. Note that the Larmor vector of the nuclear spins is now further modified by the hyperfine coupling as
\begin{equation}
\vec{\omega}_n = \gamma_n B_z(\vec{r}_n) \vec{z} + \frac{1}{2} \vec{A}_n.
\end{equation}
This shift originates from the reduction to the two-level system and can be understood as an additional magnetic field created by the electron spin, with its magnitude being the average of the vanishing field in $|0\rangle$ and the field $\vec{A}_n$ in $|1\rangle$. In particular, this is the reason why single nuclear spin addressing becomes possible, even for identical species and homogeneous magnetic fields.
Changing the basis such that the Larmor term $\vec{\omega}_n$ defines the new $z$-axis and the new $y$-axis is chosen such that the Hamiltonian is
\begin{equation}
H_0 = \sum_n \omega_n I_n^z + \frac{1}{2} \sigma_z \sum_n (a_+ I_n^x + a_- I_n^y),
\end{equation}
where $\omega_n = \omega_n \hat{\omega}_n$, $a_\pm = \hat{\omega}_n \hat{A}_n$ describes the coupling parallel to the Larmor vector and $a_\pm = \sqrt{|\hat{A}_n|^2 - a_\|^2}$ is the orthogonal component.

Instead of the natural occurring interaction in equation (6) that, without additional microwave control $\Omega(t) = 0$, conserves the energy of the electronic spin, we now aim to create a polarization exchange interaction of the form $\sigma_x I_n^x \pm \sigma_y I_n^y$. This can be achieved by applying a sequence of control pulses on the electronic spin that we will detail in the following.

Sensing sequences consisting of equally spaced population inverting pulses like the Carr-Purcell-Meiboom-Gill (CPMG) sequence [18] or the XY-family [19] create an effective $\sigma_x I_{x,y}$ interaction between the electron spin qubit and nuclei with Larmor frequency $\omega_j$ by choosing the delay between pulses $\tau$ according to $\tau = k\pi/\omega_j$, where $k$ is an odd integer. Note that delaying the start of the sequence with respect to $t = 0$ effectively chooses the nuclear spin interaction operator $I_{x,y}$, i.e. a convex combination of $I_x$ and $I_y$. Further, additional pulses can map the effective electronic operator, e.g. a $\pi/2$ pulse transforms the interaction into $\sigma_y I_{x,y}$.

Polarization sequences [9] extend this concept and create an effective $\sigma_x I_x \pm \sigma_y I_y$ interaction. Furthermore, they can be tuned to resonance with two arbitrary frequencies $\omega_j (j = 1, 2)$, which can be achieved with either of the two sequences illustrated in Figure 1 a) and b). In both cases, the control over the resonance frequencies is achieved by a parameter $\phi$ that affects the pulse phases (Figure 1 a)) or pulse duration (Figure 1 b)). Note that for $\phi = \pi/2$ we recover the PulsePol sequence [9] for the sequence in Figure 1 a), while for $\phi = \pi$ we obtain a standard XY-sequence for both sequences. In Appendix A we analyze the robustness of both sequences against control errors, that is detuning and amplitude deviations of the driving field which are corrected by the carefully chosen pulse phases. In particular we show the sequence in Figure 1 a) is robust to first order errors up to a term independent of the number of sequence repetitions $N$. Moreover, the equidistant $\pi$-pulses cancel slow dephasing noise originating from drifts of the externally applied magnetic field and magnetic impurities in the diamond sample. For the rest of this work we will focus on the sequence a) due to its better robustness against amplitude errors but the ideas presented here apply to the sequence b) in a similar way.

In the following we want to derive the conditions under which an effective $\sigma_x I_x \pm \sigma_y I_y$ interaction is created. For this purpose we determine the modulation of the $\sigma_z$ operator that appears in equation (4) in the rotating frame of the control (i.e., the subsequently applied pulses). In this
derivation we assume instantaneous pulses, the calculation for non-instantaneous pulses is used in Appendix C to derive the effective coupling strengths.

The unitary evolution after the first three pulses can be rewritten in the rotating frame of the control as

$$U_1 = e^{-i\frac{\pi}{2}\sigma_y/2U_{\text{free}}(\sigma_z, \tau)e^{-i\frac{\pi}{2}\sigma_z/2U_{\text{free}}(\sigma_x, \tau)e^{-i\frac{\pi}{2}\sigma_x/2U_{\text{free}}(\sigma_y, \tau)} = \pi_X U_{\text{free}}(-\sigma_x, \tau)U_{\text{free}}(\sigma_x, \tau),}$$

where $\pi_X = e^{-i\pi\sigma_x/2}$ is a $\pi$-pulse and we use the interaction Hamiltonian of equation (6) in the free evolution

$$U_{\text{free}}(\sigma_j, \tau) \equiv \exp(-i\tau(\omega I_j + \frac{\sigma_j}{2}(a_\perp I_x + a_\parallel I_z))),$$

simplifying to only one nuclear spin for the moment.

This means on positions 1 and 2 of the sequence in Figure 1 a) we modulated $\sigma_x$ to $\sigma_x$ and $\sigma_x$, which is also illustrated in Figure 1 c). Continuing this scheme for the next two pulses, we make use of the phase $\phi + \pi$ that all pulses in the second part of the sequence have compared to first part. These phases can be interpreted as additional rotation around the $z$-axis, for example the $\pi_{\phi+180^\circ} = \pi_{\phi+\pi}$ pulse is then $R_+ e^{-i\pi\sigma_z/2}R_-$ where $R_+ = e^{i\phi+\pi/2\sigma_z/2}$ describes the rotation. As $R_+$ commutes with all free evolutions, we immediately see that the unitary containing the free evolutions 3 and 4 with the surrounding pulses is

$$U_2 = R_+U_1R_-.$$  

Those unitary evolutions are repeated $N$ times in the sequence, so we can use $R_+\pi_X = \pi_X R_+$ and the commutation of $\pi_X$ and $U_{\text{free}}(\pm \sigma_x, \tau)$ to express the total evolution as

$$U_{\text{tot}} = (U_2U_1)^N = (R_+\pi_X U_{\text{free}}(-\sigma_x, \tau)U_{\text{free}}(\sigma_x, \tau)R_-\pi_X U_{\text{free}}(-\sigma_x, \tau)U_{\text{free}}(\sigma_x, \tau)) \times (R_+U_{\text{free}}(-\sigma_x, \tau)U_{\text{free}}(\sigma_x, \tau)^N)$$

$$= (R_+U_{\text{free}}(-\sigma_x, \tau)U_{\text{free}}(\sigma_x, \tau)^N \sum_{k=0}^{2N-1} U_{\text{free}}(-R_+^k\sigma_x R_+^k, \tau)U_{\text{free}}(R_+^k\sigma_x R_+^k, \tau).$$

This shows that the rotation on all phases of the second part of the sequence $U_2$ was constructed such that the free evolution operator $U_{\text{free}}(\theta, \tau)$ is rotated every $2\tau$
by an additional phase $\phi + \pi$ in addition to the $\pi$ rotations between position 1 and 2, 3 and 4 etc. A graphical representation is presented in Figure 1 c), where the first effective free evolution $U_\text{free}(\sigma_x, \tau)$ is depicted as position 1. The second effective free evolution $U_\text{free}(-\sigma_x, \tau)$ at position 2 is followed by $U_\text{free}(R_{-\sigma_z}R_+, \tau) = U_\text{free}(\sigma_x \cos(\phi + \pi) + \sigma_y \sin(\phi + \pi), \tau)$ at position 3.

This allows to rewrite the Hamiltonian in a rotating frame with respect to a nuclear Larmor frequency as

$$H = \frac{\sigma_x \cos(\phi(t)) + \sigma_y \sin(\phi(t))}{2} \left[ a^\dagger (I_x \cos(\omega t) - I_y \sin(\omega t)) + a^\dagger I^z \right],$$

with piecewise constant functions $\phi(t)$. The interaction does only contain nonrotating terms that will not vanish during a long evolution when the function $\phi(t)$ has a Fourier component with frequency $\omega$, and the $a^\dagger$ term always vanishes after the application of the rotating wave approximation yielding the effective Hamiltonian

$$H_\text{eff} = \frac{a^\dagger}{2} (\sigma_x I_x + \sigma_y I_y).$$

In the visualization of Figure 1 c), the nuclear spin operators of equation (11) proportional to $a^\dagger$ evolve with a periodicity of $2\pi/\omega$. As calculated in equation (10), the effective $\sigma_x$ operator gains a phase $\pi \pm \phi$ every $2\tau$, depending on if we view the rotation as being clockwise or anticlockwise. Resonance is achieved if two free evolution periods of a time $2\omega \tau$ result in the same phase for the electron and nuclear spin operators up to $2\pi n$, where $n$ is an integer.

We conclude from this picture that all nuclear spins whose Larmor frequencies $\omega$ fulfill

$$2\tau = \left(n + \frac{1}{2} \pm \phi \frac{2\pi}{2\omega}\right) \frac{2\pi}{\omega}$$

interact resonantly with the electronic spin, while the polarization direction depends on the $\pm$ sign ($\sigma_x$ rotating in same/opposite direction as the $I_x$ operator). Here, $n > 0$ is the the order of the resonance corresponding to the number of full cycles per step of $2\tau$, which motivates to denote a resonance $\tau$ by $(n, \pm)$ in the following. For the specific case $\phi = \pi/2$ we recover the PulsePol resonances $\tau \omega/\pi = 1/4, 3/4, 5/4, \ldots$ [9]. Figure 1 d) shows the first three resonances for $\phi = 3\pi/4$.

In order to simultaneously put two chosen frequencies into resonance, the free parameter $\phi$ may be chosen as follows. For example, for frequencies $\omega_1 < \omega_2 < 3\omega_1$ we can use the $(n = 0, +)$ and $(n = 1, -)$ conditions. Hence we need to solve

$$\frac{1}{2} + \phi \frac{2\pi}{\omega_1} = \frac{3}{2} - \phi \frac{2\pi}{\omega_2},$$

for $\phi$, which yields

$$\phi = \frac{\frac{3}{2} - \frac{2\pi}{\omega_1} \omega_2}{1 + \frac{2\pi}{\omega_2}}.$$  \hspace{1cm} (15)

Here, the nuclear spins precessing at two frequencies $\omega_1$ and $\omega_2$ are polarized in opposite directions. Polarization in the same direction, i.e. applying a flipflop-interaction for both nuclei instead of applying a flipflop-interaction to one nucleus and a flipflip-interaction to the other, can be achieved by employing the $(n, \pm)$ and $(n + 1, \pm)$ conditions, where we obtain

$$\pm \phi = \frac{2\pi}{\omega_2/\omega_1 - 1} - n - \frac{1}{2}.$$  \hspace{1cm} (16)

We remark that the effective electron-nuclear coupling strength varies depending on the chosen resonance $(n, \pm)$ and the parallel coupling $a_{\perp}$. In the case of instantaneous pulses we obtain the effective coupling

$$a_{\text{eff}} = \frac{a_{\perp}}{2n\pi \pm \phi} \left(1 - \cos \left(n\pi \pm \frac{\phi}{2}\right)\right)$$

for the $(n, \pm)$ resonance.

This value is constant once the resonance $(n, \pm)$ is fixed. However, for applying these sequences in quantum information processing or simulation, it is required to tune the effective coupling strength for different resonances independently, otherwise only one specific interaction that depends on the system configuration can be created. In the next section we solve this limitation by extending the sequences to allow for an arbitrary tuning of the coupling strength.

**Using Polarization Sequences for High-Fidelity Nuclear-Nuclear Gates**

We will now show how we can tune the coupling strengths $a_1$ and $a_2$ to two nuclear spins which we aim to couple simultaneously to the NV center. This allows to realize arbitrary entangling gates between nuclei with different coupling strength and Larmor frequency. There are two general gates to distinguish, that is, whether the nuclei interact with the electron spin with one flipflop and one flipflop interaction or two flipflop (flipflip) interactions.

Considering the second case, the effective evolution operator under the developed sequences for polarization in the same direction might be written as

$$U(t) = \exp \left\{ -it/2 \left[ a_1 (\sigma_x I^z_x + \sigma_y I^z_y) + a_2 (\sigma_x I^z_x + \sigma_y I^z_y) \right] \right\}. \hspace{1cm} (18)$$

For an equal coupling strength $a_1 = a_2$ and an evolution time $t$ such that $\sin(\sqrt{a_1^2 + a_2^2} t/4) = 1$, the operator
simplifies to (see Appendix C)
\[
V = \mathbb{1} (\langle -|0\rangle \langle 0| - |0\rangle \langle 1|)
+ \sigma_z (|1\rangle \langle 1| - |0\rangle \langle 0|).
\] (19)

Within the nuclear spin subspace, this gate acts like the composition of a CPHASE and a SWAP gate. Together with local rotations on the nuclear spins, this would be sufficient for universal quantum computation [20]. For example, application of this gate in combination with a \(\pi/2\) pulse around the \(y\) axis on the second nuclei can create the minimal GHZ state
\[
e^{-i\pi/2t_y(2)}V|0\rangle_{y-} = \frac{|0\rangle + |1\rangle}{\sqrt{2}}.
\]
These states are maximally entangled and advantageous in quantum metrology [21].

Similar results are obtained using the gate implemented with one flipflop and one flipflop interaction, that is
\[
U(t) = \exp \left\{ -it/2 \left[ a_1 (\sigma_x I_x^f + \sigma_y I_y^f) + a_2 (\sigma_x I_x^f - \sigma_y I_y^f) \right] \right\}.
\] (20)

In particular, for equal coupling and the gate time fulfilling \(\sin(\sqrt{a_1^2 + a_2^2}/4) = 1\) we arrive at
\[
V = \mathbb{1} (\langle -|1\rangle \langle 0| - |0\rangle \langle 1|)
+ \sigma_z (|0\rangle \langle 1| - |1\rangle \langle 0|).
\] (21)

However, the condition \(a_1 = a_2\) only holds for the special case where the ratio between the Larmor frequencies is the ratio between the perpendicular coupling strengths and a miss-match heavily decreases the fidelity of the gate. We thus extend the PulsePol sequence in Figure 1 a) by replacing each \(\pi\)-pulse by a composite pulse as proposed in [22–25]. As shown in Figure 2 a), this introduces two new parameters \(\tau_1\) and \(\tau_2\) that can be employed to tune the effective coupling to the nuclei. In particular, for instantaneous pulses it reads
\[
\begin{align*}
a_{\text{eff}} &= \frac{a_{1/2}}{\pi + \phi_{n,\pm}} \left[ 1 + 2 \cos \frac{(\beta_1 + \beta_2)(\phi_{n,\pm} + \pi)}{2} 
- 2 \cos \frac{\beta_2(\phi_{n,\pm} + \pi)}{2} + \sin \frac{\phi_{n,\pm}}{2} \right].
\end{align*}
\] (22)

where we defined \(\beta_{1,2} = \tau_{1,2} \frac{2\pi}{T_2}\) and \(\phi_{n,\pm} = (2n - 1)\pi \pm \phi\). Using this expression the right values of the \(\tau_j\) for a given pair of nuclear spins and the desired effective coupling strengths \(a_{1/2}^2\) can be deduced numerically. The expression for non-instantaneous pulses is given in Appendix C.

We apply this concept and evaluate the performance of the gate under amplitude and detuning errors of the drive which induces the pulses. To do so, we employ the process fidelity of the gate, which is given by the overlap of the Choi states corresponding to the implemented evolution via the pulse sequences and the desired target evolution [26]. Figure 2 b) shows >99.99% process-fidelity for a gate with a silicon-29 (\(\omega_1 = (2\pi)2\mathrm{MHz}\)) and a carbon-13 spin (\(\omega_2 = 1.265\omega_1\)) coupled to an NV center (\(a_1^2 = (2\pi)20\mathrm{kHz}\), \(a_2^2 = (2\pi)25\mathrm{kHz}\)), using a Rabi frequency \(\Omega = (2\pi)50\mathrm{MHz}\) and a total time \(T \approx 418\mu\mathrm{s}\). Pulse control errors of < 2% amplitude and < (2\(\pi\)) 2MHz detuning errors do not affect the fidelity. While the shape of the optimal region looks different for different gate times, this robustness is always sufficient for common experimental systems that operate with a single NV center.

Adding a third spin with \(a_1^2 = (2\pi)20\mathrm{kHz}\) and a frequency close to that of one of the other nuclei as sketched in Figure 2 c) impairs the fidelity in a frequency range \(\sim 1/T\), where \(T\) is the total gate duration. This is seen in Figure 2 d), where, for fixed total gate time, an infidelity larger than 1% appears in a larger range around the first two nuclear spin frequencies. This is a result of the frequency filter scaling \(\sim 1/T\), that stems from the effective Fourier transformation we perform in equation (12). We investigate this in more detail after the following section. Note that the values for larger total gate times (decreasing fidelity for \(T > 700\mu\mathrm{s}\)) can be improved by choosing a higher order resonance.

While this section used a gate with one flipflop and one flipflop Hamiltonian, we will demonstrate the application of a gate with two flipflop Hamiltonians in the example presented in the next section.

**MANIPULATION OF NOISE-PROTECTED STATES**

The states in the subspace \(|10\rangle,|01\rangle\) for nuclei with the same gyromagnetic ratio are protected from fluctuations of the global magnetic field. Therefore, it is an excellent candidate to store quantum information since its coherence time is substantially prolonged [27]. Efficient manipulation in this submanifold can further be used to store and manipulate quantum information in the protected manifold or to obtain information on the frequency difference and therefore the magnetic field gradient on a small lengthscale.

In this section we show fast high-fidelity manipulation in this submanifold using the protocol described in the previous subsection, but generalizing the operator \(V\) from the previous section. A major drawback of using the subspace \(|10\rangle,|01\rangle\) is that resonance conditions that polarize in the same direction, e.g. two flip-flop Hamiltonians, have to be used. This demands higher resonance conditions and therefore longer gate times, because we now need to solve...
for \( \phi \) and \( n \). We are particularly interested in similar frequencies where

\[
1 \ll \frac{\omega_2}{\omega_1} - 1 = \frac{1}{\frac{2n+1}{2} + \frac{\phi}{2\pi}} \tag{24}
\]

and therefore large \( n \) is required as \( \phi \in [0, 2\pi] \).

In the previous section we effectively used the \( \{11\}, \{00\} \) manifold using one flip-flip and one flip-flop Hamiltonian, which is beneficial when the gates are not required to affect the \( \{10\}, \{01\} \) subspace.

To create the desired gate we again fix the gate time to \( \sin(\sqrt{a_1^2 + a_2^2/4}) = 1 \). By defining \( \cos \alpha \equiv (a_1^2 - a_2^2)/(a_1^2 + a_2^2) \) we obtain an evolution operator (see Appendix B for frame linestyle explanation)

\[
U = \begin{pmatrix}
1 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0
\end{pmatrix},
\]

where the space spanned by \( |10\rangle \) and \( |01\rangle \) is mapped onto itself, which means arbitrary rotations controlled by the NV center are performed using the unitary

\[
U = -\sin(\alpha)|1\rangle_{NV} \otimes \sigma_x^{(1)} + \cos(\alpha)\sigma_z^{NV} \otimes \sigma_z^{(1)},
\]

where \( \sigma_x^{(1)} = |10\rangle \langle 01| + |01\rangle \langle 10| \) and \( \sigma_z^{(1)} = |10\rangle \langle 10| - |01\rangle \langle 01| \). After initializing the nuclei into \( |10\rangle \), for example by using a polarization sequence repeatedly, information can be swapped from the electron spin onto the nuclear spins or to perform local gates in the subspace \( \{10\}, \{01\} \). As an example, we consider a Hadamard gate on the encoded subspace in Figure 3 that transforms an initially prepared state \( |10\rangle \) into \( (|10\rangle + |01\rangle)/\sqrt{2} \). From the condition \( \cos \alpha = 1/\sqrt{2} \) we obtain \( \alpha = (\sqrt{2} \pm 1)b \), which we can again control by choosing the waiting times between the 5 \( \pi \)-pulses correctly.

Figure 3 uses the resonances (22,-) and (23,-) for two nuclei \( \omega_1 = (2\pi)2MHz, \omega_2 = 1.045\omega_1, \alpha_1 = (2\pi)20kHz, \alpha_2 = (2\pi)25kHz \) and a Rabi frequency \( \Omega = (2\pi)30MHz \). An additional third spin \( \alpha_2 = (2\pi)20kHz \) decreases the fidelity significantly if it is close in frequency to the first or second spin or to other resonances (e.g. (21,+) at \( \sim 0.98\omega_1 \) and (22,+) at \( \sim 1.025\omega_1 \)). For higher effective couplings the fidelity is higher as less perturbations can accumulate during the smaller gate time, but also the
The phase from an external magnetic field compared to this section. Those states accumulate the two times gates that achieve \( \sigma \) be equivalently realized by a synthesis of electron-nuclear gates that achieve \( \sigma \) be equivalently realized by a synthesis of electron-nuclear gates.

The action of the gates constructed in this work can be equivalently realized by a synthesis of electron-nuclear gates that achieve \( \sigma _x I_x \) interactions using standard sensing sequences [13]. Importantly, these gates are mainly limited by the decoherence time \( T_2 \) of the electron spin, during which at least two electron-nuclear gate operations have to be performed. While our work is motivated by NV centers in diamond controlling nearby C-13 spins, the same considerations also apply for shallow NV centers near the diamond surface that suffer from smaller decoherence times due to various defects on the diamond surface. However they can be used to control nuclear spins of various materials near the surface, including hexagonal boron nitride (hBN), silicon carbide and fluorine attached to the diamond surface. While still being prone to electronic dephasing during the gate time, the smaller total gate time achieved by the polarization sequence approach presented in this work can increase fidelities compared to gates synthesized by electron-nuclear gates.

Let us consider two limiting factors. The first one applies when operating in samples where nuclear spins are abundant. In this case we are faced with a complicated and dense environmental frequency spectrum which requires a narrow frequency window to ensure selective addressing of nuclei. Figure 4 shows a comparison of our direct gate approach to a gate composed of electron-nuclear gates as proposed in [13]. For this example polarization based sequences are not only faster (see second case), but also provide better robustness to disturbance from a third spin. For equal total gate times, the frequency range where a third spin disturbs the nuclear gate (red solid curve) is only about one third of the range for the electron sequence envelope counterpart (green dotted curve). Our approach employing polarization sequences then only requires 50% of the electron-nuclear composed gate time since at all times two nuclei interact with the electron spin. This is reflected in the blue curve FWHM and clear from the fact that the nuclear-nuclear gate approach manipulates both frequencies simultaneously, while the electron-nuclear gates address the two frequencies during two different parts of the sequence. Note that for electron-nuclear gates only half of the total sequence is used to decouple the respective frequency from the environment, since two gates have to be applied. The property of decoupling both nuclei simultaneously while entangling them at the same time is fundamentally related to the fact that for polarization sequences the effective interaction operators for the respective nuclei, i.e. \( \sigma _x I_x + \sigma _y I_y \) and \( \sigma _x I_x^2 \pm \sigma _y I_y^2 \), do not commute. Contrary, the effective operators \( \sigma _x I_x^1 \) and \( \sigma _x I_x^2 \) created via the sensing sequences commute and therefore cannot be used to entangle simultaneously. Another advantage of polarization sequence based gates in Figure 4 is that there are no sidepeaks of infidelity, unlike for the electron-nuclear gate approach. In the latter case different sinus cardinalis (sinc) expressions appear in the frequency domain filter [28, 29], resulting in a broader envelope. In typical experimental settings, where a given fidelity needs to be achieved by decoupling from an impairing spin bath with a range of frequencies, it is required to compare the envelopes of the curves. The envelope for the electron-nuclear gate approach (green dotted) shows an almost three times larger FWHM than the polarization sequence based approach for the same total gate time.

In cases where frequency selective addressing is not the limiting factor, the strength of the available effective dipole-dipole coupling \( A_\text{pp} \) between the electron and nuclear spins becomes a key factor as it determines the gate time for a given gate angle. In the ideal case where the coupling strengths already have the preferred ratio, e.g.
We show that the resulting effective flipflop and flipflop electron-nuclear interactions can be used to realize entangling nuclear-nuclear gates and describe how to manipulate the effective Hamiltonian to perform desired gates by introducing additional pulses. Finally we show that these gates are faster than gates composed of single electron-nuclear interactions, in particular when simultaneous decoupling from other spins is required, and discuss relevant experimental settings.

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APPENDIX

Appendix A: Sequence robustness to errors

We can describe every pulse with a unitary evolution

\[ U = \exp(-i\Omega,t (\delta\sigma_z/2 + (1 + \epsilon)(\cos \varphi\sigma_x/2 + \cos \varphi\sigma_y/2))) \]  

(26)

where \( \Omega,t \) is the intended rotation angle (e.g. \( \pi \) for a \( \pi \)-pulse) with the pulse duration \( t \) and the intended Rabi frequency \( \Omega_r \), \( \delta = \Delta/\Omega_r \) describes detuning errors and \( \epsilon = \Omega/\Omega_r - 1 \) describes driving amplitude errors. Analyzing the pulse sequence in Figure 1 a), we see that Rabi frequency errors are cancelled to first order and the detuning errors lead in first order to a correction term after \( 2N \) repetitions

\[ U_{\text{sequence}}, 2N = U_{\text{perfect-pulses}} \times (1 + \delta \left( \frac{0}{(1 + e^{i\phi})(1 + e^{-2i\phi})...(1 + e^{2N\epsilon\phi})} \right) \). \]  

(27)

The magnitude of the correction term is bounded (unless \( \phi = 0 + 2k\pi \) which is not useful for our sequence even without errors) as we can calculate the offdiagonal terms using a geometric series

\[ \left| (1 + e^{i\phi})(1 + e^{-2i\phi})...(1 + e^{2N\epsilon\phi}) \right| = \left| \frac{1 - e^{4N\epsilon\phi}}{1 - e^{i\phi}} \right| \leq 1/\cos(\phi/2) \]  

(28)

In particular, for \( \phi = \pi/2 \) (PulsePol [9]) first order errors cancel already for \( N = 1 \), and the XY4-sequence for \( \phi = \pi \) cancels even for \( 2N = 1 \).

The sequence in Figure 1 b) has a first order amplitude error that is only cancelled for \( \phi = \pi \) (XY-sequence), and is therefore only robust against Rabi frequency errors for \( \phi \equiv \pi \). The detuning errors affect the evolution as

\[ U_{\text{sequence}}, 2N = U_{\text{perfect-pulses}} \times (1 + \delta \left( \frac{i \cos(2N\phi)}{\sin(N\phi)} \sin(N\phi) - i \cos(2N\phi) \right) ) \]. \]  

(29)

that is always bounded and does not increase with the number of pulses.

Appendix B: Entangling by using polarization transfer Hamiltonians

A simultaneous flipflop between an NV and 2 nuclei can be written as

\[ \exp \left( -it/2 \left[ a_1(\sigma_z^{(NV)} \otimes I_x^{(1)} + \sigma_y^{(NV)} \otimes I_y^{(1)}) + a_2(\sigma_z^{(NV)} \otimes I_x^{(2)} + \sigma_y^{(NV)} \otimes I_y^{(2)}) \right] \right) \]

where the solid-framed matrix elements are NV-nuclear interactions, dotted matrix elements are to preserve unitarity and dashed matrix elements are nuclear-nuclear interactions. For \( \sin(\sqrt{a_1^2 + a_2^2}t/4) = 1 \) and \( a_1 = a_2 \), we obtain
\[
\exp \left( -it \left[ a_1 (\sigma_x^{(NV)} \otimes I_x^{(1)}) + \sigma_y^{(NV)} \otimes I_x^{(1)} \right] + a_2 (\sigma_x^{(NV)} \otimes I_x^{(2)}) \right) \right] \\
\left( \begin{array}{cccc}
1 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 \\
\end{array} \right)
\]

This means if the NV is prepared in |1\rangle, the nuclear states will swap and a CPHASE gate is applied.

**Appendix C: Calculation of the effective coupling**

We analytically calculate the effective coupling constant for the sequence in Figure 2 a) with finite pulses. It is sufficient to calculate the overlap of the modulation function with the intended cosine from the first \(\pi/2\) pulse until the center of the first \(\pi\)-pulse as shown in Figure 5. Mathematically this corresponds to calculating the Fourier coefficient for the desired resonance. We denote the fraction that the times \(\tau_{1,2}\) between the additional pulses by \(\beta_{1,2}\) such that \(\tau_{1,2} = \beta_{1,2} \pi + \phi\), see Figure 5. For the resonance condition \((n, \pm)\) we denote \(\phi_{n,\pm} = (2n - 1)\pi \pm \phi\) to obtain

\[
a_{\text{eff}} = \frac{a_1}{\pi + \phi} \int_{-\phi_{n,\pm}/2}^{\pi/2} dx \cos(x)f_1(x)
\]

\[
= \frac{a_1}{\pi + \phi_{n,\pm}} \frac{1}{1 - \epsilon^2} \left[ \epsilon \cos \frac{\phi_{n,\pm}}{2} + \cos \frac{\epsilon \pi}{2} + \cos \left( \frac{\beta_1 + \beta_2}{2} \right) \frac{\phi_{n,\pm} + \pi}{2} + \cos \left( \frac{\beta_1 + \beta_2}{2} \right) \frac{\phi_{n,\pm} + \pi}{2} + 5\epsilon \pi \right] + \cos \left( \frac{\beta_1 + \beta_2}{2} \right) \frac{\phi_{n,\pm} + \pi}{2} + \sin \frac{\phi_{n,\pm} - \epsilon \pi}{2}
\]

where we defined \(\epsilon = \omega_L/\Omega\) as the ratio between nuclear Larmor and electron Rabi frequency gives the expected \(a_1 (2 + \sqrt{2})/(6\pi)\) for PulsePol \((n=1, -)\) with instantaneous Pulses \((\epsilon = \beta_1 = \beta_2 = 0)\).

This equation can be used to achieve the same coupling for two different resonances. We use this equation to calculate equal coupling for the \(n = 0, +\) and \(n = 1, -\) resonances as shown in Figure 6.
Figure 5: Modulation function $f_1$ for the adapted sequence until the center of the first $\pi$-pulse that is replaced by a composite pulse: It rises to 1 during first $\pi/2$ pulse and remains there until the beginning of the composite pulse. For symmetry reasons it is sufficient to consider only the first $\pi$-pulse ($1 \rightarrow -1$), the evolution afterwards that takes a fraction $\beta_1$, the second $\pi$-pulse ($-1 \rightarrow 1$), the evolution afterwards that takes a fraction $\beta_2$ and half of the third $\pi$-pulse ($1 \rightarrow 0$).

Figure 6: Comparison of effective coupling (left) for $A_{\text{eff}}(\phi_1, \beta_1, \beta_2)$ (top) and $A_{\text{eff}}(2\pi - \phi_1, \beta_1, \beta_2)$ (bottom) for the parameters in Figure 2 (main text) and difference between those functions (right). The chosen $\beta_{1,2}$ is marked with a white cross where bot functions were equal and close to a desired value.