Coherent control of an NV\(^{-}\) center with one adjacent \(^{13}\)C

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Received 10 April 2014, revised 24 July 2014
Accepted for publication 11 August 2014
Published 25 September 2014

Abstract
We investigate the theoretically achievable fidelities when coherently controlling an effective three-qubit system consisting of a negatively charged nitrogen vacancy (\(^{15}\)NV\(^{-}\)) center in diamond with an additional nearby carbon \(^{13}\)C spin \(I_C = 1/2\) via square and two frequency component radio and microwave frequency pulses in different magnetic field regimes. Such a system has potentially interesting applications in quantum information-related tasks such as distributed quantum computation or quantum repeater schemes. We find that the best fidelities can be achieved in an intermediate magnetic field regime. However, with only square pulses it will be challenging to reach the fidelity threshold(s) predicted by current models of fault-tolerant quantum computing.

Keywords: quantum control, nitrogen vacancy, defect centers, impurity spins, quantum repeaters

1. Introduction

Impurity spins in solids have long been known for their potential to be used in quantum information processing devices [1–3]. Among these, the negatively charged vacancy centers (NV\(^{-}\)) in diamond has stood out for its exceptional properties. It is a well-localized, stable and optically controllable spin in the ‘vacuum’ of a mostly spinless carbon lattice [4]. Given these
virtues, it had soon been recognized as a good solid-state qubit, showing long coherence times even at room temperature [5–8]. Moreover, NV centers have been employed in a host of applications beyond quantum information, ranging from use as a single-photon source [9–11], to high-resolution sensor in electrometry [12], magnetometry [8, 13–19], decoherence microscopy [20–22], nano-scale NMR sensor [23–25] and thermometer [26].

A nitrogen vacancy center consists of a vacancy site in a diamond lattice adjacent to a substitutional nitrogen atom resulting in a defect of \( \text{C}_{3\text{v}} \) symmetry [27, 28]. In the negative charge state NV\(^{-} \), the electron wave function is a spin \( S = 1 \) for both a ground state manifold (GSM) with orbital symmetry \( \text{A}_2 \) as well as an excited state manifold (ESM) of E-type orbital symmetry separated from the GSM by an optical 637 nm (ZPL) transition. The NV\(^{-} \) center exhibits the useful properties of optical polarizability and spin-dependent fluorescence, allowing initialization and readout of the electron spin even at room temperature. These are possible due to the presence of energetically intermediate levels between the GSM and ESM, which allow spin non-conserving, non-radiative transitions which preferentially (but not exclusively) populate the \( m_S = 0 \) sub-level (for a detailed review see [4]). Together with the electronic spin of the vacancy, hyperfine-coupled nitrogen and possibly carbon nuclear spins found in the vicinity can form a quantum register of several qubits. In such a register, the nuclear spins with their excellent coherence times [29, 30] would serve as quantum memories accessed via the more directly controllable electronic spin of the vacancy. This system was proposed as the node in a quantum repeater [31, 32] as well as for quantum information processing [33] and has been intensely studied by numerous experiments both at room and low temperature (\( \approx 4–8 \) K). The important milestones demonstrated are initialization and single-shot readout of electronic and nuclear spins in both temperature regimes [34–36], as well as, at low temperature, creation of entanglement between vacancy electron and nuclear spins [37], the polarization of single photons [38] and other (distant) NV centers [39]. Further important steps on the way to a scalable quantum computation architecture are a demonstration of room temperature quantum registers formed by long-range dipolar coupled NV\(^{-} \) centers [40] and entanglement swapping to nuclear spins [41]. Moreover, in quantum registers made up of a single NV\(^{-} \) and multiple proximate carbon nuclear spins, decoherence-protected operations were performed [42], and recently the first implementations of quantum error correction in diamond-based qubits were also demonstrated [43, 44].

While these experiments serve as beautiful proofs-of-principle, and fidelities achieved are remarkable given the practical technical difficulties, they are not yet at thresholds required for scaleable, fault-tolerant quantum computation [45] or even for quantum repeaters [46, 47], which are generally believed to be easier to implement. In particular, even with error correction a general computation will require many gate executions before the system is reset/corrected, and this quickly degrades fidelity. From the perspective of architecture selection and design, it would be highly desirable to have a better theoretical understanding of the ultimate limits to the achievable fidelities, given the inherent properties of the NV\(^{-} \) system. Previous studies looking at a bare NV\(^{-} \) center in a pure carbon lattice have shown that in principle such a system might indeed allow operations with high enough accuracy for large-scale quantum computation even when using only simple control pulses [48], as long as exciting the NV electron spin out of the GSM is avoided. The hyperfine interaction strength in the ESM (\( \approx 60 \) MHz) is relatively stronger than in the GSM (\( \approx 3 \) MHz) [49, 50], hence any excitation from the GSM could result in dephasing on the nitrogen nuclear spin. As quantum information requires not only gate operation but also readout and initialization, this difference in coupling strength adds significant
constraints to the operational regimes of physical parameters and setups. By contrast, nearby, strongly coupled $^{13}\text{C}$ nuclear spins do not show this difference in hyperfine coupling strength, and it might thus be used to design a device immune to this source of dephasing.

This leads to the question investigated in the present work: whether high-fidelity control by simple means is still possible in an effective three-qubit system ($^{15}\text{NV}^{−}+^{13}\text{C}$), where the carbon introduces interactions which potentially make high-fidelity control more difficult. In our study we focus on NV$^{−}$ centers containing the $^{15}\text{N}$ isotope of nitrogen, since this is a natural qubit, eliminating all worries about leaving the computational space during operations. As nitrogen $^{14}\text{N}$ is much more abundant in nature, so far the experimental study of NV centers has focused on the $^{14}\text{NV}^{−}$ variety. Still, using isotopically selective implantation techniques fabricating samples containing predominantly $^{15}\text{NV}^{−}$ centers should be possible.

This paper is structured as follows. In section II we introduce the effective spin model we used and discuss the magnetic field regimes we investigated it in, which were low and intermediate magnetic fields. In section III we investigate the primitive gates obtainable with single-pulse control in the two interesting magnetic field regimes, possible choices for computational basis, and we comment on the driving power requirements. Section IV contains an analysis of times and fidelities for derived gates based on the results from the previous section and an assessment of our system’s performance as a node of a quantum repeater. We present a concluding discussion in section V.

2. Effective spin model

The system we investigated effectively consists of three qubits: the electronic spins of the vacancy defect (V) and two nuclear spins, one belonging to the always present nitrogen (N) and the other to a nearby carbon $^{13}\text{C}$ (C). Throughout we assume the nitrogen to be a $^{15}\text{N}$ isotope, and thus both nuclei in our system have spin $I = 1/2$, while the electronic spin state is a triplet $S = 1$. Since we did not consider excitations out of the $^3\text{A}_2$ GSM, the free time evolution of the system is well described by the Hamiltonian [51]

\[
H_{\text{NVC}} = H_{\text{V}} + H_{\text{N}} + H_{\text{C}} + H_{\text{VN}} + H_{\text{VC}} \\
H_{\text{V}} = DS_\perp^2 + E\left(S_\perp^2 - S_\parallel^2\right) + \gamma_e BS_\parallel \\
H_{\text{C/N}} = \gamma_{\text{C/N}} BI_{\text{C/N, z}} \\
H_{\text{VN}} = \vec{S}\vec{A}_\text{N} \\
H_{\text{VC}} = \vec{S}\vec{C}_\text{C} ,
\]

where $\vec{S} = (S_\perp, S_\parallel, S_\perp)^T$ is the vacancy and $\vec{I} = (I_\perp, I_\parallel, I_\perp)^T$ the nuclear spin operator and we define the magnetic moments $\gamma_e = g_e\mu_B = 28 \text{ MHz/mT}$ for the electronic spin as well as the nuclear spins of carbon $\gamma_C = g_C\mu_B = +10.6 \text{ kHz/mT}$ and nitrogen $\gamma_N = g_N\mu_B = -4.3 \text{ kHz/mT}$. $D$ is a zero-field splitting of 2.88 GHz (at low temperature) coming from the spin-spin interaction, $B$ denotes the magnetic field we assume to be parallel to the NV-axis, and $E$ is the crystal strain which is very weak in the GSM ($0...10 \text{ MHz}$) and could be canceled entirely by applying an appropriate electric field. Finally, $\vec{A}$ and $\vec{C}$ are the hyperfine tensors of nitrogen and carbon respectively.

For symmetry reasons $\vec{A}$ is exactly axial, while $\vec{C}$ is approximately so, even for nearest-neighbor carbons where one might expect the contact term to give a significant non-axial
contribution. As we consider the nitrogen to be an $^{15}$N isotope ($I = 1/2$), we do not need to include a nuclear quadrupolar term in (1). Also, the direct dipolar interaction between the two nuclear spins is negligible, as even for nearest-neighbor $^{13}$C it does not exceed $\sim 0.5$ kHz.

The hyperfine interaction term for the nitrogen consists of parallel and exchange contribution and reads $\tilde{S}A_{NN} = A_0 S_z I_{N,z} + \frac{1}{2} A_\perp (S^+ I_C^- + S^- I_C^+)$, while the carbon hyperfine-term looks the same in its principal axis system, there are additional terms after transforming into NV-adapted coordinates (with $z$ along the NV’s symmetry axis):

$$\tilde{S}C_C = C_{||}(\theta) S_z I_{C,z} + \frac{1}{2} C_{\perp}(\theta) (S^+ I_C^- + S^- I_C^+)$$
$$+ \frac{1}{2} C_R(\theta) (S^+ I_C^+ + S^- I_C^-)$$
$$+ C_\Delta(\theta) (S_z I_{C,y} + S_y I_{C,x}) \ ,$$

where the $C_\Delta$-term contains $z$- and $y$-operators because we used an $x$-axis rotation in the coordinate transformation. The four coefficients depend on the angle $\theta$ between the NV axis and the vacancy-carbon axis and are given by

$$C_{||}(\theta) = C_{||} \cos^2 \theta + C_{\perp} \sin^2 \theta$$
$$C_{\perp}(\theta) = \frac{1}{2} \left( C_{\perp} (1 + \cos^2 \theta) + C_{||} \sin^2 \theta \right)$$
$$C_R(\theta) = \frac{1}{2} \left( C_{\perp} (1 - \cos^2 \theta) - C_{||} \sin^2 \theta \right)$$
$$C_\Delta(\theta) = (C_{\perp} - C_{||}) \sin \theta \cos \theta \ .$$

The effect of the two additional terms $C_R$ and $C_\Delta$ on energy levels and states in the magnetic field regime are minimal except that for the $m_S = 0$ states at low field, where $C_R$ causes a splitting between even-parity states ($|0, \uparrow \uparrow \rangle_{VC}$ and $|0, \downarrow \downarrow \rangle_{VC}$) while the odd-parity states ($|0, \uparrow \downarrow \rangle_{VC}$ and $|0, \downarrow \uparrow \rangle_{VC}$) are split by the exchange term.

The value for $C_{||}(\theta)$ can be observed directly in ODMR (optically detected magnetic resonance) experiments as the hyperfine splitting between different carbon spin orientations. The other parameters are, however, harder to confirm. A rough estimate can be gained by setting the magnetic field to $B = B_s = 103$ mT and observing the splitting at the avoided crossing between the $m_S = -1$ and 0 levels. Since the Hamiltonian is highly connected, this will not yield good results even for $C_{\perp}$. A better strategy is measuring the level splitting while sweeping the magnetic field and fitting the model parameters to the obtained data. As an analytic approximation to this, we can look at the curvature of the $m_S = 0$, $-1$ levels in a field region around $60$–$80$ mT. There, at least in 2nd-order perturbation theory, the curvatures are directly proportional to $C_{\perp}^2$ (mixing $| -1, \uparrow \rangle_{VC} \leftrightarrow |0, \downarrow \rangle_{VC}$) and $C_R^2 + C_\Delta^2$ respectively (mixing $| -1, \downarrow \rangle_{VC} \leftrightarrow |0, \uparrow \rangle_{VC}$).

We considered two different carbon positions, nearest-neighbor and third-neighbor, because these show the strongest hyperfine interaction and thus offer the potentially fastest gate times. For a nearest-neighbor carbon, hyperfine interaction strength in the principal basis is $C_{||,nn} = 199$ MHz and $C_{\perp,nn} = 123$ MHz, while in the NV-basis this corresponds to $C_{||}(\theta_{nn}) = 129$ MHz, $C_{\perp}(\theta_{nn}) = 155$ MHz, $C_R(\theta_{nn}) = -35$ MHz and $C_\Delta(\theta_{nn}) = 25$ MHz (‘nn’ stands for ‘nearest neighbor’). Numerical $ab\ initio$ calculations found two different classes.
of third-neighbor positions showing a strong hyperfine coupling [49]: planar (out of plane) third neighbors (see figure 1) with coupling constants of $C_{||,3rd} = 19$ (18)MHz and $C_{\perp,3rd} = 14$ (13) MHz. In ensemble measurements [52], hyperfine ESR lines associated with third neighbors have been identified showing interaction strengths of $C_{||} = 18.53$ and $C_{\perp} = 13.26$, which is right between the theoretically predicted values. We use these latter values as the best estimate of third-neighbor interaction strength.

In comparison to the bare NV center, the level structure of the Hamiltonian (1) shows a much larger splitting of the $m_S = \pm 1$ levels, due to the much stronger parallel hyperfine interaction for both carbon positions we considered. There are two avoided crossings, one strain-avoided at $B_{str, nn} = C_{||}/2\gamma_e \approx 2.6$ mT ($B_{str, 3rd} = 0.28$mT for third neighbors) and the other (mainly) exchange-avoided at $B_x = D/\gamma_e \approx 103$mT.

For the sake of simplicity in both analysis and application, it made sense to investigate the model in magnetic field regimes in which the eigenstates have high ‘z-fidelity’, i.e., are close to the $S_z$-$I_z$-basis. In the NV”, in principle three such regimes exist. The z-fidelity can be achieved for very high magnetic fields of $B \gg B_x$, for which the $|m_S = -1\rangle$ levels are lowest in energy. However, such large magnetic fields are not very desirable from a practical point of view, as they are difficult to keep stable and the fast Larmor precession of the electronic spin makes accurate timing harder. We therefore chose to concentrate on the low and intermediate field strengths, which are around $B = 1 – 2$ mT and $B = 15 – 50$ mT respectively. For nearest-neighbor $^{13}$C in the low-B regime, a good compromise between z-fidelity and low field strength that still provided sufficient level separation was $B = 1.1$mT, which is the value we chose. However, in the case of third-nearest-neighbor carbon, $B = 2$mT is a better choice, since 1.1mT is still somewhat too close to the strain avoided crossing with correspondingly lower z-fidelity (see figure 2b). For both carbon positions, we used $B = 25$mT as the intermediate B-field strength, since this gives the best z-fidelity.

Figure 1. Carbon nuclear spin positions. NV center and the sites where the lattice positions for the (one) carbon $^{13}$C we considered in this study: on the left a free 3D view, and on the right along the [111] direction. The color coding of the spheres is as follows: (small) blue = vacancy, green = nitrogen, black = nearest neighbors (of V), gray = next-nearest neighbors, yellow and orange = third neighbors for which numerical ab initio calculations suggest strong hyperfine interaction with the NV electron spin due to finite spin density. These calculations find slightly different coupling strength for the two positions yellow and orange, but this has not yet been resolved experimentally. The dashed cage shows a diamond lattice unit-cell.
To simulate dissipative time evolution in our system, we solved a time-dependent master equation with Lindblad operators, describing relaxation and dephasing for each subsystem individually (the details can be found in appendix B). In order to model the experimentally well-

Figure 2. Levels and $z$-fidelity dependence on an axial magnetic field. (a) Energy levels as function of magnetic field strength for nearest-neighbor $^{13}$C. The insets zoom in on the avoided crossings at $B = 2.6\text{mT}$ and $103\text{mT}$ respectively. For this plot, an unrealistically high strain was assumed in order to clearly show the former. (b) Fidelity of eigenstates with $S_z, I_z$-basis (‘$z$-fidelity’) for low and intermediate magnetic field (left and right column respectively). The same colors represent the same states in both pictures.

2.1. Decoherence model

To simulate dissipative time evolution in our system, we solved a time-dependent master equation with Lindblad operators, describing relaxation and dephasing for each subsystem individually (the details can be found in appendix B). In order to model the experimentally well-
established Gaussian dephasing of the NV electronic spin \([53, 54]\), we assumed time-dependent rates 
\(\gamma_{\text{V},a/b}(t) = t/(T^2_{\text{V}})\) (i.e. the same for both dephasing channels ‘a’ and ‘b’). Since the hyperfine coupling is quite strong for close-by carbons, one should in general use Lindblad operators adapted to the eigenbasis of the total system. However, since we were only interested in magnetic field regimes in which the eigenbasis is very close to the computational \((S_z,I_z)\) basis, the error due to the simplified decoherence model was inconsequential. The decoherence times we assumed were 
\(T_{1,\text{V}} = 10\,\text{ms}, T_{2,\text{V}} = 100\,\mu\text{s}, T_{1,\text{C}} = T_{1,\text{N}} = 10\,\text{s}, T_{2,\text{C}} = T_{2,\text{N}} = 10\,\text{ms}\). These are conservative estimates, and each individually has already been demonstrated or even surpassed in experiment \([6–8]\).

2.2. Driving

We modeled microwave (MW) and radio-frequency (RF) driving with a Hamiltonian

\[
H_{\text{drive}} = u(t) \left( S_x + \frac{\gamma_c}{\gamma_e} I_{C,x} + \frac{\gamma_n}{\gamma_e} I_{N,x} \right),
\]

where the driving field is a sum of square pulses \(u(t) = \sum_{n=1}^{N_f} \Omega_{0,n} \cos(\nu_n t + \phi_0)\). The number of frequency components, \(N_f\), was in practice either one (pure square pulse) or two and \(\nu_n\) usually chosen in resonance with some transition. This left the \(\Omega_{0,n}\) as the main parameter(s) to be optimized. However, we limited our search to values still in the RWA regime, so that the relative phase \(\phi_0\) provided control of the driving axis and a direct handle (direct coupling to \(y\)-direction operators) was unnecessary.

In this work we have not considered pulse shaping (varying \(\Omega_0\) and \(\phi_0\) continuously in time), leaving this as a further optimization to achieve fully fault-tolerant quantum computation in the future.

3. Single pulse gates

In this section, we present an overview of the results of single-pulse driving for the two interesting magnetic field regimes identified previously. We find that the intermediate magnetic field regime allows much cleaner gates compared to the low magnetic field regime, as in the latter, insufficient level separation makes it difficult to exclusively address desired transitions. The natural choice of basis is \(\left\{|m_S = 0\rangle_V, |m_S = \pm 1\rangle_V\right\}\), where our results were obtained for \(|-1\rangle_V\) but the other spin orientation should yield very similar numbers.

3.1. Low and intermediate fields

Low \(B\). — In the low magnetic field regime, 1.1mT for nearest and 2mT for third-nearest-neighbor (see section 2), the separation between \(m_S = 0 \rightarrow \pm 1\) transitions is on the order of 50–100 MHz (see figure 3). This sets a lower limit for pulse duration of about 10–20 ns when driving the vacancy. While this kind of bound is somewhat less stringent for the nuclear spins, which have transition frequencies separated by 120 (10) MHz in the cases of (third) nearest-neighbor carbon, their weak coupling to the driving field necessitates comparatively long pulse times in any case. In fact, at the driving strengths we found to be optimal, pulse times of 300–1000 ns and 6–7 \(\mu\text{s}\) respectively revealed that both carbon and nitrogen nuclear spins are being driven mostly indirectly via the hyperfine interaction with the NV electronic spin.
Without hyperfine interaction, expected direct driving pulse times would be on the order of \( \gamma \mu \approx -25 \text{ ms} \) for the carbon and \( \mu \approx -50 \text{ ms} \) for the nitrogen respectively.

When considering single driving pulses, achievable fidelities for certain initial-target state pairs can be more than 99% assuming perfect timing, but, even so, gate fidelities (the minimum over the entire Bloch sphere, see appendix A) are much lower. This still means that by applying sequences of pulses some interesting states, e.g. entangled states involving either nuclear spin, or both, can be prepared with reasonably high fidelity of >97%. For instance, hyperfine enabled double-spin flip processes allow preparation of odd-parity entangled states between NV electronic and carbon nuclear spin by only a single pulse in the case of the \( m_S = 0, +1 \) or \( m_S = 0, -1 \) bases and two pulses in the case of the \( m_S = +1, -1 \) basis (see appendix D).

**Intermediate B.** — At a field strength of 25mT, the improved separation between transitions (see figure 4) made it possible to achieve good gate fidelities in the range of 94% to 97% for both single- and multi-qubit gates. These figures hold for an assumed timing error of \( \Delta t = 250 \text{ ps} \), which we think is achievable. Moreover, these fidelities are for all three subsystems combined (as opposed to looking at only the subsystems directly involved in a certain gate).
A more detailed account of the results in both magnetic field regimes can be found in appendix D.

3.2. Choice of computational basis

Depending on the chosen computational basis, different single- and multi-qubit gates require more than one pulse: as depicted in figure 4, in the \(|m_S = 0\rangle_V, |m_S = -1\rangle_V\) basis, several two- and three-qubit operations can be performed with only a single driving pulse, but at the cost of having to polarize the NV electronic spin (to \(|-1\rangle_V\)) in order to be able to do single-qubit gates on the nuclear spins. A basis more suited for the latter is \(|m_S = +1\rangle_V, |m_S = -1\rangle_V\), since the level splittings for both carbon and nitrogen do not depend on the sign of \(m_S\) (this remains approximately true even in the intermediate field regime up to \(B \approx 30\) mT). However, in that basis, single-qubit operations on the vacancy require two pulses (\(|+1\rangle_V \rightarrow |0\rangle_V \rightarrow |-1\rangle_V\), this involves using two frequency components), and multi-qubit gates even more (e.g. a \(C_{CNOT}\) requires three: pulse 1 and 3, \(\pi\)-pulses on \(|+1, +\rangle_V \leftrightarrow |0, +\rangle_V\); and pulse 2, a \(\pi\)-pulses on \(|0, +\rangle_V \leftrightarrow |-1, +\rangle_V\)). Since the fidelity of a pulse sequence is likely to be close to the product of the individual fidelities, one will probably prefer the \(|m_S = 0\rangle_V, |m_S = -1\rangle_V\) basis, unless a certain application demands otherwise.

3.3. Driving power

Our optimization of the driving power \(\Omega_0\) yielded complementary results for nearest- and third-nearest neighbors. While in the former case, a medium driving power for V and C gates and a quite strong power for N yielded the highest maximum gate fidelities, the situation was reversed for third-nearest neighbors. That third-nearest neighbor V driving should be done fast is understandable because we need line-widths to be larger than the C-spin level splitting of \(C_{||} \approx 13.5\) MHz. This is well satisfied for \(\Omega_0 \geq 150\) MHz with peaks in the achievable fidelity occurring whenever the phases can be best lined up. A resulting \(\Omega_0\)-dependence of the gate fidelity is figure 5 for the example of a (third neighbor) NV electronic spin. While the highest peak in absolute terms occurs at \(\Omega_0 \approx 240\) MHz in the raw fidelity, taking into account a finite timing accuracy of, assumed, 250ps shows that values around 160 MHz are in fact the most preferable choice.

However, in an experimental setup, all optimal driving powers identified in this study are rather technically challenging. Since we consider our system to operate at cryogenic temperatures (\(\sim 4–8\) K), sample heating due to the MW and RF-radiation is a serious issue: a rough estimate for the maximum permissible ‘true’ driving power is \(O(1\)W), for which \(\pi\)-pulse times are roughly 50ns for the NV electron spin. In our model, this gate time occurs for \(\Omega_0 = 15\) MHz, which is significantly lower than any of the optimal values we identified (see table 2). This could provide the motivation for an extended search in the low-\(\Omega_0\) regime. However, the difficulty in such a search would be that computation time is proportional to gate time and thus roughly inversely proportional to \(\Omega_0\). Thus, for all but the NV electronic spin, this would make an extensive search very difficult.
4. Derived gates and sequences

In the previous section we looked at gates and operations implementable with a single pulse. Now we want to extend this to sequences of pulses in order to realize a set of useful gate operations on the three-qubit NVC system at intermediate magnetic field. In table 1 we give an overview of relevant gates in the NV system, both primitive and derived ones, showing for both carbon positions gate times, fidelities and equivalent circuits used to derive them.

The primitives presented in the previous section include all single-qubit rotations around an axis in the x-y plane (from which one can construct z-rotations), as well as the four entangling operations CROT$_V$C, CROT$_C$, CROT$_V$, and CROT$_{CN,V}$, where CROT$_{1[2],3}$ denotes a conditional rotation applied to qubit three controlled by the state of qubit(s) one (and two). For instance, if we chose to perform an X($\pi/2$) rotation, i.e. a $\pi$-pulse around the x-axis, the resulting operation would be a CiNOT, which is equivalent to a CNOT up to a $\pi/2$ z-rotation on the control qubit. In addition, there is the non-unitary initialization of the NV electron spin into the $|0\rangle_V$ state.

The standard technique at room temperature, to employ off-resonant excitation with green laser light, has been used in virtually all NV experiments to date. However, at low temperature, resonant driving to a state with preferential decay to the $m_S = 0$ state, e.g. $|A_2\rangle$, is much faster and we should be able to reach high fidelities after only a few cycles.

These primitives clearly form a universal set which has in fact some redundancy. For instance, we need only one out of CROT$_V$C and CROT$_C$, as well as CROT$_V$ and CROT$_{CN,V}$. Having them all at our disposal potentially improves both gate time and fidelity.

In table 1 there are some gates where this is indeed the case. For instance, a CPHASE$_{CN}$ can be derived from CNOT$_{V,N}$ and CNOT$_{V,C}$, but it is much faster and cleaner to simply apply the Toffoli gate CNOT$_{CN,V}$ twice. Another good example is the BELL gates: logically the two options shown in the righthand column of table 1 are equivalent (both map the computational basis onto a Bell basis), but in practice there is difference in both gate time and fidelity.

4.1. Repeaters

An early application for NV centers could be quantum repeaters. In this context, adding a third qubit in the form of the carbon has the benefit of allowing limited error correction. A three-qubit
**Table 1.** List of the derived gates. Values for primitive gates were taken from the results in section 3 (see also appendix D). These were then used to estimate those of derived gates. When there was an ambiguity between gate time and fidelity for gates with multiple equivalent circuits, we gave precedence to fidelity. Note that the efficient implementation of SWAP and BELL via CROts shown in the right column works only for the carbon nuclear spin. (*): assumed values.

| gate   | time (μs) | fid (%) | circuit                      |
|--------|-----------|---------|------------------------------|
| INITᵣ | 100 ns    | 99.9(*) | INITᵣ =                     |
| MEASᵥ  | 100 ns    | "      | MEASᵥ =                     |
| X, Yᵥ/C/N | 0.016/0.004/0.33/6.2 | 95.9/96.8/94.1 | X, Yᵥ/C/N = |
| CROTᵥ/C,N | 16 ns/60 ns | 96.8/93.0 | CROTᵥ/C,N = |
| CROTᵥ,N | 6.55/6.55 | 97.7/98.0 | CROTᵥ,N = |
| Zᵥ/C/N | 0.032/0.66/12.5 | 92/97/89 | Zᵥ/C/N = |
| Hᵥ/C/N | 0.04/0.83/15.6 | 91/94/92 | Hᵥ/C/N = |
| CNOTᵥ/C,N | 0.35/1.06 | 91/94 | CNOTᵥ/C,N = |
| CNOTᵥ,N | 0.34/1.08 | 90/94 | CNOTᵥ,N = |
| CNOTᵥ,ŋ | 12.8/12.9 | 91/94 | CNOTᵥ,ŋ = |
| CPHASEᵥ,N | 1.86/1.26 | 90/95 | CPHASEᵥ,N = |
| CNOTᵥ,N | 44.1/44.6 | 55/70 | CNOTᵥ,N = |
| INITᵥ | 0.79/57.0 | 85/82 | INITᵥ = |
| INIT甯 | 57.0/75.6 | ~50/66 | INIT甯 = |
| SWAPᵥ/C | 0.71/2.23 | 85/85 | SWAPᵥ/C = |
| SWAPᵥ甯 | 69.7/70.4 | <50/60 | SWAPᵥ甯 = |
| BELLᵥ/C | 0.67/2.16 | 94/82/95 | BELLᵥ/C = |
| BELLᵥ甯 | 25.6/25.8 | 73/82 | BELLᵥ甯 = |
| BELLᵥ/C | 1.4/4.4 | 80/81 | BELLᵥ/C = |
| BELLᵥ甯 | 95.5/96.4 | <50/50 | BELLᵥ甯 = |
system is, however, incapable of correcting a general error (bit and phase flip). This would require introducing another qubit, for instance another $^{13}\text{C}$ into the system, an approach that is worth investigating in future work. We can, however, also contemplate going in the other direction and working with only two qubits, at the cost of having no error correction: as we can see, all gates involving the nitrogen nuclear spin are much slower than those with the carbon, and offer no advantage in fidelity. Thus, in order to improve the rate of the device we could restrict ourselves to using only the latter approach. This would rely on the initial entangling link being high-fidelity in the first place. Ways to establish such links probabilistically have been proposed [31, 55] using state-dependent reflectivity of cavities together with path-erasure techniques.

We want to briefly explore how far the NVC system with the simple driving paradigm investigated here could be used as a node in a basic nested repeater scheme (see e.g. [31] and references therein to learn about quantum repeaters). If we assume that entanglement links between two NVC systems are established with fidelity exceeding 99.9% using this method, a Bell measurement could be performed with fidelity $F_{\text{BELLM}} = 80\%$ (81%) for third-nearest-neighbor carbon, resulting in a final link fidelity of 68% (as we also have to perform a SWAP operation to move one of the links to the nuclear spin, before we can apply the Bell measurement), which is below the classical threshold. However, performing one or more rounds of purification could extend the repeater link further. Note that while purification schemes normally employ CNOT gates, they can also be adapted to the CROT gates available here as primitive gates. Given that initial link and measurement fidelities are high, the limit fidelity for a purified link that is being stored in the carbon nuclear spins would be roughly $\sim 75\%$, achieved after $k \geq 5$ rounds of purification. However, this does not take into account decoherence by a factor of $\eta \sim .99$ reduction during the $T_{\text{roundtrip}} + T_{\text{gates}} \sim 10\mu s$ one round of entanglement swapping and purification take with our gates and an assumed node separation of a few tens of meters. Doing so leads to the appearance of maxima at higher nesting levels in the fidelity-vs-rounds of purification plot shown in figure 6, while at the first level there is just a reduction of overall fidelity by $\eta$. As can be seen, at the first nesting level, three rounds of purification suffice to get a link fidelity above the classical threshold. But already at the second level gate infidelity and decoherence are too strong to reach the quantum regime.

This shows that for NV$^-$ with strongly coupling carbon $^{13}\text{C}$ to be truly useful as a component in a scalable architecture it is necessary to go beyond the simple pulse paradigm investigated here and consider further optimization strategies. In technical applications, this would mean a complication that is avoidable in bare NV centers, where square pulses are already good enough. However, we think it is still interesting to pursue this course, as the carbon offers a single-qubit gate speed-up by a factor of more than ten for nearest- and still about five for third-nearest neighbors.

5. Conclusion

We numerically investigated a system consisting of an $^{15}\text{NV}^-$ center and a nearby, strongly hyperfine-coupled carbon $^{13}\text{C}$ nuclear spin in two different magnetic field regimes. Within a conservative yet realistic model, we determined the achievable fidelities for specific states as well as gates using only simulated square pulses of microwave and radio-frequency radiation. We found that in the low magnetic field regime only some special starting and target state
combinations allowed high fidelity operations. This suggests that careful selection of states gives us sufficient fidelity to perform some quantum information tasks. Gate fidelity suffers from the limited state $z$-fidelity and level separation. The situation is much better at intermediate fields. There, we found fidelities of up to 98% for single-qubit gates on the carbon nuclear spin and 97% for the NV electronic spin. The nitrogen single-qubit as well as multi-qubit gate fidelities were somewhat lower than that. If we analyze the expected gate times of gates derived from these primitives via straightforward concatenation, we find that using a strongly bound carbon does indeed offer potential speed-up of operations. However, the fidelities of these derived gates quickly deteriorates with the nesting level. Thus this study indicates that gates implemented via square pulses can be used only in limited applications such as a proof-of-principle repeater experiment involving one NV$^{-}$$+$$C$-center. Also of experimentation interest might be using hyperfine-enabled double-spin flip processes to have faster and higher fidelity state preparation of entangled states. For general applications, going beyond the square-pulse paradigm and using pulse-shaping techniques like optimal control is required.

Acknowledgments

We thank M S Everitt, S J Devitt and H Kosaka for valuable comment and discussion. This research was partially supported under the Commissioned Research of the National Institute of Information and Communications Technology (A & B) project.

Appendix A. Measuring fidelity

The fidelity between two quantum states described by density matrices $\rho$ and $\sigma$ can be determined by [56]
Table 2. $\pi$-pulse fidelities and times for resonant, square-pulse driving of the NV+C system in the low ($B \approx 2 \text{ mT}$) and intermediate magnetic field ($B = 25 \text{ mT}$) regimes for selected states, single- and multi-qubit gates. Gate fidelities were computed from a set of 25, 16 and 8 initial-final state pairs for vacancy, carbon and nitrogen respectively. Uncertainties were computed assuming a timing accuracy of $\Delta t = 250 \text{ ps}$.

**A: Low-field regime**

| State | Nearest Neighbor | Third Neighbor | Fidelity (%) | Time $T_\pi$ (ns) | $\Omega_0^{\text{opt}}$ (MHz) | Fidelity (%) | Time $T_\pi$ (ns) |
|-------|------------------|----------------|--------------|-------------------|-----------------------------|--------------|-------------------|
| $|1\rangle_C \rightarrow |1\rangle_C$ | 1↑ , 1↑ | 24 (22.5) | 93.2 (94.2) | 31.0 (31.1) | 74 (76) | 89.3 (90.1) | 9.0 (9.9) |
| $|1\rangle_C \rightarrow |1\rangle_C$ | 1↑ , x+ | " | 95.3 (95.7) | 31.8 (31.5) | " | 94.1 (94.4) | 9.3 (10.5) |
| $|1\rangle_C \rightarrow |1\rangle_C$ | " | 22.5 (19.0) | 4 (91.3)% | 48.4 (38.8) | 70 (70) | 13.4% (89.2%) | 3.9 (10.4) |
| $|1\rangle_C \rightarrow |1\rangle_C$ | Avg | 90.6 (94.3)% | 32 (38.7) | " | 87.1% (91.3%) | 9.8 (" ) |

**B: Intermediate-field regime**

| State | Nearest Neighbor | Third Neighbor | Fidelity (%) | Time $T_\pi$ (ns) | $\Omega_0^{\text{opt}}$ (MHz) | Fidelity (%) | Time $T_\pi$ (ns) |
|-------|------------------|----------------|--------------|-------------------|-----------------------------|--------------|-------------------|
| $|1\rangle_C \rightarrow |1\rangle_C$ | 1↑ , 1↑ | 31 | 98.5 ± 0.2 | 23.4 | 72 | 99.7 ± 0.2 | 9.6 |
| $|1\rangle_C \rightarrow |1\rangle_C$ | 1↑ , x+ | 44 | 98.5 ± 0.3 | 16.0 | 90 | 99.4 ± 0.2 | 7.9 |
| Operation | | | | | |
| l + , ↑ )_{CN} | " | 98.2 ± 1.3 | 16.4 | 129 | 97.2 ± 1.5 | 5.3 |
| \( \frac{1}{2}\sigma_0)_C \otimes (\frac{1}{2}\sigma_0)_N \) | " | 95.3 ± 0.3 | 16.4 | 230 | 97.5 ± 1.6 | 3.0 |
| gate | 44 | 95.9 ± 1.3 | 16.0 | 165 | 97.5 ± 0.8 | 4.4 |
| CROT_{C,V} | 43 | 96.8 ± 0.3 | 15.8 | 12 | 93.0 ± 0.1 | 60.0 |
| CROT_{CN,V} | 0.8 | 95.2 ± 0.0(1) | 932 | 1.1 | 97.3 ± 0.0(1) | 634 |
| l ↑ )_C → l ↓ )_C | l - 1, ↑ )_{VN} | 31 | 99.6 ± 0.1 | 486 | 110 | 99.6 ± 0.2 | 1336 |
| l - 1, x + )_{VN} | " | 99.6 ± 0.1 | 486 | 51 | 99.4 ± 0.0(1) | 2973 |
| \( \frac{i}{\sqrt{2}}(|0 \rangle + |1 \rangle)\sqrt{v} \uparrow )_N \) | " | 32.5 | 98.4 ± 0.4 | 453 | 72 | 99.3 ± 0.6 | 1981 |
| \( \frac{i}{\sqrt{2}}(|1 \rangle - |0 \rangle)\sqrt{v} \uparrow )_N \) | gate | 52.5 | 96.0 ± 0.4 | 331 | 72.5 | 99.2 ± 0.1 | 1980 |
| CROT_{V,C} | gate | 52.5 | 96.8 ± 0.1 | 332 | 130.5 | 96.9 ± 0.2 | 1001 |
| l ↑ )_N → l ↓ )_N | l - 1, ↑ )_{VC} | 110 | 99.2 ± 0.0(0) | 6719 | 83 | 99.4 ± 0.0(0) | 8847 |
| l - 1, x + )_{VC} | " | 96.2 ± 1.0 | 7146 | 101 | 98.4 ± 0.1(1) | 7649 |
| \( \frac{i}{\sqrt{2}}(|0 \rangle + |1 \rangle)\sqrt{v} \uparrow )_C \) | gate | 121 | 99.0 ± 0.0(0) | 6143 | 100.5 | 98.7 ± 0.0(03) | 7563 |
| CROT_{V,N} | | 109.5 | 94.1 ± 0.0(0) | 6232 | 109 | 96.6 ± 0.0(0) | 6922 |
| | | 109 | 97.7 ± 0.0(0) | 6552 | 110 | 98.0 ± 0.0(0) | 6553 |
\[ F(\rho, \sigma) = \left( \text{Tr} \sqrt{\rho \sigma \sqrt{\rho}} \right)^2. \]  

(A.1)

The square-root is defined for hermitian operators and can be computed from the eigenspectrum via

\[ \sqrt{\rho} = \text{diag}\{ \sqrt{\rho_1}, \ldots, \sqrt{\rho_D} \} \]

where \( D = \dim H \) and \( \rho_n \) are the eigenvalues of \( \rho \). If one of the states, say \( \sigma \), is a pure state, this simplifies to

\[ F(\rho, \sigma = |\phi\rangle \langle \phi|) = \text{Tr} (\rho |\phi\rangle \langle \phi|) = \langle \phi | \rho | \phi \rangle. \]

(A.2)

For purely unitary time evolutions, the exact gate fidelity can be computed by just considering (the time evolution of) an ONB of the Hilbert space \( H \). In practice, this would require computing the time evolution for \( D \) different starting states. However, since \( \mathcal{S}_\rho(\rho) \), i.e. the actual time evolution of the system, is dissipative in our case, the exhaustive description necessary for calculating the exact gate fidelity requires computing the time evolution for all \( D^2 \) generators of the space of hermitian operators on \( H \). As a standard way of assessing gate-fidelity, this was computationally too costly even for our modest Hilbert-space size of \( D = 12 \).

Therefore, we settled on the practical solution of computing the state fidelities for a suitably large subset of states from \( H \) and taking as our gate fidelity the minimum among all the obtained values. The size of these sets were 25, 16 and 8 states when assessing vacancy, carbon and nitrogen driving pulses, respectively. In detail, the state sets were

Vacancy (25 states): \[ \{ |0, k, l\rangle, |0\rangle \otimes (\rho_{\text{mixed}})_C \otimes (\rho_{\text{mixed}})_N \} \]

Carbon (16 states): \[ \{ |m, \downarrow, l\rangle \} \]

Nitrogen (8 states): \[ \{ |n, l, \downarrow\rangle \} \]

where \( k \in \{ \uparrow, \downarrow, x\pm, y\pm \} \), \( l \in \{ \uparrow, \downarrow, x\pm \} \), \( m \in \{ 0, -1, x\pm \} \) and \( n \in \{ 0, -1 \} \).

**Appendix B. Numerical simulation**

**Decoherence model.**— We implemented a master equation of the form

\[ \dot{\rho} = -\frac{i}{\hbar} \left[ H_{VC}, \rho \right] + \sum_k \gamma_k \left( L_k \rho L_k^\dagger - \frac{1}{2} \left\{ L_k^\dagger L_k, \rho \right\} \right). \]  

(B.1)

Here \( k \in \{ (V, 0 \rightarrow +1), (V, 0 \rightarrow -1), (V, +1 \rightarrow 0), (V, -1 \rightarrow 0), (V, +1 \rightarrow -1), (V, z_{+10}), (V, z_{-10}), (C, \uparrow \rightarrow \downarrow), (C, \downarrow \rightarrow \uparrow), (C, z), (N, \uparrow \rightarrow \downarrow), (N, \downarrow \rightarrow \uparrow), (N, z) \} \) where \( z_{\pm 10} \) denotes a Pauli-z operator in the \( m_S = \pm 10 \) subspace. Thus, the Lindblad operators \( L_k \) describe relaxation and dephasing of each system \( (V, C \text{ and } N) \) individually and the \( \gamma_k \) are the inverses of the experimentally observed relaxation and decoherence times \( T_1 \) and \( T_2 \) of the individual subsystems except for vacancy dephasing rates \( \gamma_{2,Valb} \). This we chose to be time-dependent, to reproduce the experimentally observed Gaussian (and thus non-Markovian) dephasing of the NV electron spin. This kind of dephasing has been observed for the time evolution of a spin that evolves under the influence of a weak and randomly varying magnetic field, which in case of the NV stems from other spins in the vicinity (the electronic spins of nitrogen P1 centers as well as carbon \( ^{13}\text{C} \) nuclear spins). Gaussian dephasing is obtained for a linear time dependence of \( \gamma_{2,Valb} = 1/T_2^2 \).

**Simulation.**— Numerical simulations were performed in Mathematica (version 7.0) using the built-in NDSOLVE function to integrate the master equation (B.1) up to the desired final time starting in some state \( \rho(0) = \rho_0 \) of the entire system. Single-qubit gate fidelities were computed
as described in the previous section, while multi-qubit gates were extracted in a similar fashion, although comparing each final state to all other target states in addition to the desired one.

Appendix C. Derived gates

In the intermediate field regime ($B = 25\text{mT}$), we computed the fidelities of some interesting derived gates based on the simulation results obtained for primitive gates. Derived gates are constructed from sequences of primitive ones according to some gate identity. Following the prescription of these identities, we obtained derived gate parameters by multiplying the fidelities and summing the times of the constituent primitives. This is consistent with the limitation of the NV$-\text{C}$ system where in gates cannot be performed in parallel on different subsystems for physical reasons, even though this might be possible logically (e.g. single-qubit gates on different qubits commute). For dependency between primitive and derived gates, see figure C1; for the complete list of gates and the (highest fidelity) identities, see table 1.

Often there are several different ways to obtain a given gate, in particular since our set of primitives is redundant. For instance, it is possible to obtain a CNOT$_{C,N}$ either by applying the square of a CNOT$_{CN,V}$ (=$\text{TOFFOLI}_{CN,V}$) sandwiched between two Hadamard gates on the nitrogen, or, alternatively, via a CNOT$_{V,N}$ sandwiched between two SWAP$_{VC}$. In this case the former is clearly the faster and higher-fidelity alternative. However, there are also cases that require choosing between fidelity or speed. For example, a BELL$_{VC}$ gate can be achieved either via CNOT$_{V,C}$-$H_V$-CNOT$_{V,U}$ or with the same but with V and C switching roles.

Appendix D. Detailed results

D.1. Low field

As mentioned in section II, low magnetic fields offer the advantage of less stringent pulse-timing requirements. The magnetic field strength values we settled for were a trade-off between the z-fidelity of the eigenstates on one side and limiting level separation on the other. They were 1.1mT for nearest- and 2mT for third-neighbor carbon.

Single-qubit gates.— In the following, unless otherwise stated, fidelities and times given applied to single $\pi$-pulses for which the true gate fidelity was approximated by sampling over representative points in the Hilbert space of the combined system (see appendix appendix A for the details of how we measure fidelity).
Driving V. — The transition frequency between $|1\rangle_V$ and $|−1\rangle_V$ strongly depends on the state of the carbon nuclear spin for both nearest- and third-nearest-neighbor $^{13}$C, which clearly posed a problem for single-qubit operations in the $m_S = ±1$ basis. We had to solve this in two different ways for the two carbon positions: in the case of nearest-neighbor, using dual frequency driving ($N_f = 2$ in (4)) worked well, while it did not give good results for third neighbors. We attribute this to the much stronger parallel hyperfine interaction in the former case, resulting in a splitting of $\approx = 129$ MHz between carbon $|\uparrow\rangle_C$ and $|\downarrow\rangle_C$. This is resolvable within the $\pi$-pulse times giving the best fidelities, which are on the order of O(10 ns). In contrast, the splitting is only $\approx 13.5$ MHz for third-nearest-neighbor and therefore not big enough to allow resolution of the two-component pulse within a time of about 10 ns. This would instead require one order of magnitude longer pulses i.e., weaker driving power. We found, however, that for such slow pulses maximum fidelity invariably suffers. The best solution in this case is therefore to apply a fast pulse tuned to the average transition frequency. In principle it holds: the faster the better, but, for very short pulse times, timing error will start to seriously reduce the fidelity.

Driving C. — The level splitting for the carbon is independent of the state of either V or N, thus it is possible to manipulate the carbon-spin state independently. This probably explains why it shows the highest state fidelities of the three subsystems, reaching 96.8% and 96.9% for nearest- and third-nearest-neighbor carbon respectively (figure D1).

Driving N. — Similar to the NV electron spin, transition frequencies for the nitrogen nuclear spin depend on the state of the carbon, with a difference between level splittings of $\omega_{N,11} = 800$ kHz between $|\uparrow\rangle_C$ and $|\downarrow\rangle_C$. This means that while the electronic spin can in principle be in an arbitrary state (in the $m_S = ±1$ subspace), C must be polarized to either $\uparrow$ or $\downarrow$. The maximum fidelity is 97.9% for the starting state $|1, \uparrow\rangle_{VC}$ while gate fidelity is much
lower, mostly due to the energy-splitting difference mentioned as well as drift of the carbon spin phase.

Gates via driving. — As we mentioned before, the hyperfine interaction causes transitions for the vacancy and nitrogen to be dependent on the state of the other qubits. While this is a problem when implementing single-qubit gates, it can be used to implement two-qubit gates via driving. Using a qubit basis consisting of $|m_\Sigma = 0\rangle$ and either of $|m_\Sigma = \pm 1\rangle$, such gates can be implemented with a single pulse. For the basis $\{|+\rangle, |-\rangle\}$ there is the difficulty that direct transition between these levels is not dipole-allowed and therefore exceedingly slow when driven directly. Thus, between the $|m_\Sigma = \pm 1\rangle$ states, all two-qubit gates involving V must be realized via sequences of at least three entangling pulses plus single-qubit rotations to tidy up factors of $i$.

For example a CNOT$_{C,V}$ would consist of the sequence $\pi(|+\rangle, |-\rangle)_B \leftrightarrow |0\rangle, |1\rangle \rangle$, $\pi(|0\rangle, |1\rangle \leftrightarrow |+\rangle, |-\rangle \rangle$, $\pi(|0\rangle, |1\rangle \leftrightarrow |+\rangle, |-\rangle \rangle)$. To be independent of the nitrogen, the pulse times must be fast compared to the nitrogen hyperfine level splitting of 3 MHz ($= 330$ ns), but slow enough to minimize off-resonant driving of the wrong transition (to $|m_\Sigma = -1\rangle$). For CNOT$_{C,V}$ the two transitions are separated by about 180 MHz at $B = 1$ mT corresponding to roughly 6 ns. Thus, both criteria can only be satisfied to limited degree, with the ideal pulse length being about 45 ns per pulse or 135 ns in total. A SWAP gate between vacancy and carbon state requires five $\pi$-pulses (see figure 3b) and has thus a lower fidelity still.

Entangled states.— As we have seen, implementing multi-qubit gates with high fidelities is difficult in the low magnetic field regime. However, if we aim for something less ambitious, such as preparing some useful state from a known starting state, high fidelities are achievable even when including the nitrogen. As examples, let us look at two entangles states, $(|+\rangle, |\downarrow\rangle + |+\rangle, |-\rangle \rangle)_B/\sqrt{2}$ and $(|+\rangle, |\downarrow\rangle + |+\rangle, |-\rangle \rangle)_{VN}/\sqrt{2}$. The standard way to reach these states are the three-pulse sequences depicted in figure D2, for which we find maximum fidelities of 97.4% (97.3%).

However, the presence of the strong hyperfine interaction makes it possible to directly drive ordinarily ‘forbidden’ transitions involving two simultaneous spin flips. This lets us reach the target states with the two-pulse sequences MW $- \pi (|0\rangle, |\downarrow\rangle \leftrightarrow |+\rangle, |\downarrow\rangle)$, RF $- \pi/2 (|+\rangle, |\downarrow\rangle \leftrightarrow |+\rangle, |\downarrow\rangle)$ and an equivalent sequence for the nitrogen (substitute ‘N’ for ‘C’). For these fidelities are 98.5% and 98.9% at optimum gate times of 286 ns (15 $\mu$s).
significantly better than for the ordinary three pulses. This two-pulse scheme works well only for setting up odd-parity Bell states, because the two-spin flip processes are mainly caused by the hyperfine exchange term. Even-parity Bell states would need a counter-rotating term, which is only present for the carbon nuclear spin and there too is much weaker than the exchange term.

D.2. Intermediate field

For magnetic field strengths between \( B \approx 15 \) mT and 50mT the eigenstates are much closer to the \( S_z^c I_z^c \)-basis than for low \( B \) (see section II, figure 2(b). If we choose the \(|m_S = 0\rangle\) and \(|-1\rangle\) levels as our vacancy-qubit basis, we see that while the \( z \)-fidelity of some states reaches a maximum only much later, the average peaks in the region around 25mT, which is the value we picked. It has the added benefit of large detuning with, and thus low leakage into the \(|+1\rangle\) subspace, which has to be avoided as it would constitute a qubit loss error.

Driving \( V \).— From the energy level structure at intermediate \( B \) (section III, figure 4) one can see that, like in the low-\( B \) regime, controlling the NV electronic independent of the carbon spin state is again not straightforward. As before, our solution was the dual-frequency driving technique in the case of nearest-neighbor carbon, and driving the average transition frequency in the case of third-neighbor carbon. With this, we were able to achieve maximum fidelities of 96.1% and 97.7% respectively. Plots of the gate fidelity for a \( \pi \)-pulse are shown in figure D3 for both carbon positions.

Driving \( C \).— Unlike at low magnetic field, fidelities of the carbon nuclear spin nearly match those for the NV electronic spin. Our choice of computational basis means, however, that we can only effect a \( \pi \)-rotation, if the NV electron spin is polarized into logical \(|1\rangle_{V}\) (the \(|m_S = -1\rangle\) state). In our numerical gate fidelity computations we nonetheless included starting states with \(|0\rangle_{V}\), in which case we checked how well the pulse preserves this state, i.e., we set a

**Figure D3.** Gate fidelities of NV electronic spin \( \pi \)-pulse driving at intermediate \( B \)-field for nearest-neighbor (upper row) and third-nearest-neighbor carbon (lower row). The horizontal axis shows time in \( \mu s \), and the vertical axis shows fidelity. Thick, purple traces show the gate, and the thin blue traces show average state-driving fidelity. The insets show a zoom-in on each maximum.
target state equal the starting state for the gate fidelity estimation. For nearest-neighbor carbon, starting states with the NV electron spin polarized show fidelities up to 99.6\%, while falling off somewhat if the vacancy starts in the state \(|x + \rangle = (|0\rangle_V + |−1\rangle_V)/\sqrt{2}\). There is no significant difference between these starting states in case of third-neighbor carbon.

**Driving N.**— Compared to the low magnetic field regime, nitrogen transition frequencies depend far less on the state of the carbon, which allows relatively good gate fidelities of 96(6)\% for third-nearest neighbor and 94(1)\% for nearest-neighbor $^{13}$C. With gate times on the order of 6 $\mu$s, non-polarized states of the NV electron spin would have d-ephased strongly due to the low assumed electron $T_2$ time of 100 $\mu$s (still relatively long for a solid state qubit), which is why we excluded them from our gate fidelity computation. Since relaxation is much slower, polarized electron spin states do not suffer appreciably during the gate time and thus state fidelity for such starting states is as high as for the other subsystems, 99.2\% and 99.4\% respectively for the two different carbon positions.

**Entangling gates.**— In the intermediate magnetic field regime, our choice of computational basis allowed several multi-qubit gates to be implemented by a single pulse (figure D4). The transitions involved are indicated schematically in figure 4. As can be seen, we can obtain multi-qubit gates between all qubits. This set of operations is redundant in that two CNOTs would already be universal, but this redundancy is very welcome since direct, single-pulse gates are faster and have a higher fidelity than those obtained from potentially lengthy gate sequences.

All results discussed here are summarized in table 2.

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