Fast Room Temperature Phase Gate on a Single Nuclear Spin in Diamond

S. Sangtawesin, T. O. Brundage, and J. R. Petta

Department of Physics, Princeton University, Princeton, NJ 08544, USA

Nuclear spins support long lived quantum coherence due to weak coupling to the environment, but are difficult to rapidly control using nuclear magnetic resonance (NMR) as a result of the small nuclear magnetic moment. We demonstrate a fast ~ 500 ns nuclear spin phase gate on a $^{14}$N nuclear spin qubit intrinsic to a nitrogen-vacancy (NV) center in diamond. The phase gate is enabled by the hyperfine interaction and off-resonance driving of electron spin transitions. Repeated applications of the phase gate bang-bang decouple the nuclear spin from the environment, locking the spin state for up to ~ 140 μs.

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The NV center in diamond is one of the most promising systems for quantum computation due to its convenient optical spin initialization and readout schemes, which can be performed at room temperature [1,3]. In addition to effective manipulation of the electronic spin, hyperfine coupling provides a means to detect and control proximal nuclear spins, enabling multiple qubit operations [4-10]. Recent works have demonstrated high-fidelity initialization and readout of nuclear spins in diamond using an NV center as an auxiliary qubit [9,11]. Robust control and preservation of the qubit state for as long as ~500 ns nuclear spin phase gate on a single NV center by utilizing off-resonant Rabi oscillations of the nuclear spin further splits each electronic state, generating on the timescale of the electron Rabi oscillations. Through this quantum control approach, we can achieve a π-phase gate in less than 500 ns, a speed far exceeding that of the nuclear Rabi oscillations $\tau_n \sim 40 \mu$s. The pulses can be applied repeatedly, providing rapid phase shifts to bang-bang decouple the qubit from the environment and preserve the qubit state for as long as ~140 μs [10,18].

Our sample is a high purity type IIA diamond (Element Six) with naturally occurring NV centers. We determine the locations of single NV centers relative to pre-patterned alignment marks using fluorescence confocal microscopy, as shown in Fig. 1(a). We verify single photon emission from a single NV center by measuring the photon correlation function $g^2(0) \geq 0.5$ [21,22]. Microwave (MW) and radio frequency (RF) pulses are applied to the NV center to drive electron spin and nuclear spin transitions, respectively. The DC electrodes allow Stark shifting of the NV center energy levels [21,22], but are not used in this experiment.

The ground state manifold of the NV center is described by the Hamiltonian:

$$H = D S_z^2 + g_e \mu_B \vec{B} \cdot \vec{S} + \frac{1}{2} A_\perp (S_+ I_- + S_- I_+) + A_\parallel S_z I_z + Q I_z^2 - g_N \mu_N \vec{B} \cdot \vec{I}. \quad (1)$$

Here $g_e$ is the electronic g-factor, $\mu_B$ is the Bohr magneton, $\vec{B}$ is the external magnetic field, and $\vec{S}$ ($\vec{I}$) are the electron (nuclear) spin operators. The energy level diagram is shown in Fig. 1(b). Electronic spin levels $m_s = -1, +1$ are separated from $m_s = 0$ by a zero-field splitting $D = 2.87$ GHz. The $m_s = -1$ and $m_s = +1$ energy level degeneracy is lifted by an external magnetic field that results in a Zeeman splitting $g_e \mu_B B_z$, where $g_e \mu_B = 2.802$ MHz/G and the $z$-axis is defined along the NV symmetry axis. The $m_s = +1$ state is far detuned and therefore not shown in the diagram. We use the $m_s = 0$ and $m_s = -1$ levels to encode the electron spin qubit, allowing optical initialization and readout of the NV center electron spin state [23,24]. Hyperfine coupling to the $^{14}$N nuclear spin further splits each electronic state into three sublevels, corresponding to the nuclear spin projections $m_I = -1, 0, +1$. The $m_I = -1, +1$ states are split from $m_I = 0$ by the nuclear quadrupole coupling $Q = -4.962$ MHz, and their degeneracy is lifted slightly by nuclear Zeeman splitting, with $g_N \mu_N = 0.308$ kHz/G. The degeneracy is further lifted by the hyperfine coupling to the electronic spin with $A_\perp = -2.70$ MHz and $A_\parallel = -2.16$ MHz in the $m_s = \pm 1$ subspaces [10,12].

We select the two sublevels $|m_s, m_I\rangle = | -1, +1\rangle, \quad | -1, 0\rangle$ for our nuclear qubit as the transition is well isolated from the others, allowing selective excitation using
FIG. 1: (a) Confocal image of the NV center used in the experiment. Two DC electrodes (not used) and a RF & MW stripe line are fabricated near the NV center. Inset: Measurements of the second order correlation function \( g^{(2)}(\tau) \), with \( g^{(2)}(0) < 0.5 \) indicating emission from a single NV center. (b) Energy level diagram, with energy levels indexed by the electron spin quantum number \( m_1 \) and nuclear spin quantum number \( m_2 \). The \( m_S = +1 \) level is not shown in the level diagram due to the relatively large electron Zeeman energy. Microwave excitations (MW1 and MW2) drive electronic transitions, while RF excitation is used to drive nuclear spin transitions. (c) Pulse sequence used to implement the nuclear spin phase gate during nuclear Rabi oscillations.

the RF field. Moreover, it allows us to perform readout of the nuclear spin state using a microwave \( \pi \)-pulse (MW1, frequency \( \nu_{\text{MW1}} \)), tuned to resonance with \( m_I = +1 \) transition \( \nu_{+1} \), mapping the nuclear spin state to the electronic spin state. The electronic spin state is subsequently measured using optical readout [5, 12]. Unlike \(^{13}\text{C}\) nuclear spins, which exhibit Larmor precession in the presence of an external magnetic field, \(^{14}\text{N}\) nuclear spin precession is prohibited in NVs due to the large quadrupole coupling [4]. Therefore, we induce well-defined nuclear spin precession using RF pulses, adjusting the RF power to control the nuclear Rabi frequency \( \Omega_n \) [12].

In detail, when the system is in an arbitrary state \( |\psi\rangle = \alpha| -1, 0 \rangle + \beta| -1, +1 \rangle \), with \( |\alpha|^2 + |\beta|^2 = 1 \), we apply a MW pulse (MW2) with frequency \( \nu_{\text{MW2}} \) for a short duration \( t \), driving the electronic transitions \( | -1, 0 \rangle \leftrightarrow |0, 0 \rangle \) and \( | -1, +1 \rangle \leftrightarrow |0, +1 \rangle \) (with transition frequencies \( \nu_0 \) and \( \nu_{+1} \), respectively). In the strong driving limit, where the electron Rabi frequency greatly exceeds the detunings \( \Omega_E > \Delta m_I = \nu_{\text{MW2}} - \nu_{m_I} \), electrons undergo fast Rabi oscillations between \( m_S = 0 \) and \( m_S = -1 \) regardless of the nuclear spin state. We ensure that the electron Rabi frequency is the same on both transitions by setting \( \nu_{\text{MW2}} \) midway between \( \nu_0 \) and \( \nu_{+1} \). If \( t \) is chosen such that \( \Omega_E t = 2n\pi \), the electron spin state will return to the original \( m_S = -1 \) subspace, with phase accumulation generated by the off-resonant electron spin Rabi oscillations:

\[
U(t)|\psi\rangle = \alpha e^{i\delta_0 t/2} | -1, 0 \rangle + \beta e^{i\delta_{+1} t/2} | -1, +1 \rangle.
\]

This process implements a phase gate on the nuclear spin, with the phase difference \( \Delta \phi = (\delta_{+1} - \delta_0)t/2 = (\nu_0 - \nu_{+1})t/2 \equiv A_m t/2 \) set by the hyperfine coupling \( A_m \) and the duration of the MW2 pulse \( t \).

The full experimental sequence, including optical initialization and readout, is illustrated in Fig. (1c). A DC magnetic field \( B_z \approx 500 \text{ G} \) is applied along the NV-axis, bringing the system close to the excited state level anti-crossing (ESLAC) [22]. At the ESLAC, the NV center electronic spin is first polarized to \( m_S = 0 \) via optical pumping with a 532 nm laser. During this process, the \(^{14}\text{N}\) nuclear spin is dynamically polarized to \( m_I = +1 \) [20, 26]. The choice of working at the ESLAC provides high fidelity initialization and readout of the nuclear spin without requiring a Ramsey-type pulse sequence, where the fidelity is limited by a weak selective MW pulse and electron dephasing during the Larmor precession period in the initialization protocol [5]. After the system is polarized optically, a selective \( \pi \)-pulse (MW1) is applied to transfer the population to \( | -1, +1 \rangle \), completing the initialization process. Spin manipulation is performed by applying a RF pulse with duration \( \tau_{\text{RF}} \), where phase gate “kicks” from MW2 are simultaneously applied. Finally, optical readout is performed after applying another selective MW1 \( \pi \)-pulse that converts the population from \( | -1, +1 \rangle \) to the bright state \( |0, +1 \rangle \). This yields a photoluminescence (PL) output that is proportional to the \( | -1, +1 \rangle \) population at the end of the pulse sequence.

We probe the \( | -1, +1 \rangle \leftrightarrow | -1, 0 \rangle \) nuclear spin transition by sweeping the RF frequency \( \nu_{\text{RF}} \) for a fixed \( \tau_{\text{RF}} \) that is set to achieve a \( \pi \)-pulse when on resonance. When \( \nu_{\text{RF}} \) is on resonance with the nuclear spin transition, population will be transferred from \( | -1, +1 \rangle \) to \( | -1, 0 \rangle \). Since \( | -1, 0 \rangle \) is off resonance with MW1, the population transfer leads to a “dark” readout and reduces the PL intensity [Fig. 2(a)]. \( \nu_{\text{RF}} \) is then tuned to resonance with this transition. By varying \( \nu_{\text{RF}} \), nuclear spin oscillations can be observed with a Rabi frequency \( \Omega_n \sim 25 \text{ kHz} \) [Fig. 2(b)]. This nuclear Rabi frequency is much greater than the Rabi frequency calculated using only the nuclear gyromagnetic ratio, due to the additional \( \sim 10 \times \) enhancement from the electron-nuclear flip-flop, \( \Omega_n \approx g_{\text{N}} \mu_{\text{N}} B_{\text{RF}} + A_{1g} g_{\text{E}} \mu_{\text{E}} B_{\text{RF}}/D \).

To measure the electronic transition frequency, we perform electron-nuclear double resonance spectroscopy by preparing the system in \( | -1, 0 \rangle \) or \( | -1, +1 \rangle \) using the calibrated MW1 and RF pulses. A selective MW2 \( \pi \)-pulse is applied with varying frequency \( \nu_{\text{MW2}} \) and the population remaining in \( | -1, 0 \rangle \) or \( | -1, +1 \rangle \) is measured. A decrease in PL occurs when \( \nu_{\text{MW2}} \) is on resonance with the electron spin transitions [Fig. 2(c)], allowing us to extract the hyperfine coupling \( \sim 2.16 \text{ MHz} \), consistent
Here the system is prepared in a superposition state \( \psi = \frac{1}{\sqrt{2}}(-1, +1) + | -1, 0 \rangle \) with a nuclear spin \( \frac{\pi}{2} \)-pulse and allowed to freely precess for a duration \( \tau_{\text{free}} \). Before a \( \pi \)-pulse is applied to rotate the spin back to the original basis \( |20\rangle \). We do not observe any damping in the Ramsey fringes during this measurement interval, indicating a long nuclear spin phase coherence time \( T_{20} > 100 \mu s \).

Nuclear spin phase gates are calibrated by tuning the MW2 frequency \( \nu_{\text{MW2}} \) to the midpoint between the \( m_1 = 0 \) and \( m_1 = +1 \) transitions. We apply one phase gate of varying duration \( t \) during the Ramsey sequence \([\text{Fig. 2(d)}]\) and extract the phase of the fringes afterwards.

The inset of Fig. 3(a) shows that the nuclear spin phase accumulation \( \Delta \phi \) is linearly proportional to the gate duration \( t \). For this measurement the MW2 power is tuned such that six full cycles of electron Rabi oscillations correspond to a \( \pi \)-phase gate \( \Omega_{e} t / 2\pi = 6 \). We note that the \( \pi \)-phase gate duration \( t = 2\pi / A_{||} = 462 \) ns is fixed by the hyperfine coupling strength \( A_{||} = 2.16 \) MHz \([\text{Fig. 2(c)}]\). This corresponds to an electron Rabi frequency \( \Omega_{e} = 12.96 \) MHz.

We now show that the phase gate can be applied during nuclear Rabi oscillations. Results with one and two \( \pi \)-phase gates are shown in Fig. 3(b) and (c), respectively.

The red curves show PL intensity as a function of \( \nu_{\text{RF}} \) with phase gates applied. For direct comparison, the black curves show nuclear Rabi oscillations that are not interrupted by phase gates. Phase gates are applied at the times indicated by the red arrows in the figures. The phase shifts \( \Delta \phi = \pi \), evident in the two plots, indicate that we successfully applied \( \pi \)-phase gates on the nuclear spin qubit with each \( t = 462 \) ns gate operation time, far exceeding the speed of nuclear spin Rabi oscillations \( \tau_{n} \sim 40 \mu s \) shown in Fig. 2(b).

The fast \( \pi \)-phase gate can be applied repeatedly to decouple the nuclear spin from the RF pulse, effectively locking the nuclear spin state. We demonstrate nuclear spin locking by applying multiple \( \pi \)-phase gates in rapid succession for up to several nuclear Rabi oscillation periods \([\text{Fig. 3(a-b)}]\). While the state preservation is evident, there is \( \gtrsim 30\% \) decrease in the amplitude of nuclear Rabi oscillations after several phase gates are applied. The amplitude decrease is asymmetric; it reduces the bright state PL level while leaving the dark state PL level unchanged. This suggests that it is not caused by dephasing of the nuclear spin, in which case the amplitude would be dampened symmetrically. To understand this effect, we first show from simulations that the decrease in contrast is due to the population being driven out of the \( |m_S, m_I\rangle = | -1, 0 \rangle, | -1, +1 \rangle \) two-level subspace. Then, we argue that this missing population contributes to a “dark” readout, resulting in the asymmetric decrease in the oscillations.

To simulate the experiment we use Eq. 1 and add driving terms:

\[
H_{N}^{\text{ac}} = \Omega_{n} \sin(2\pi \nu_{\text{RF}} t) I_x
\]

\[
H_{e}^{\text{ac}} = \Omega_{e} \sin(2\pi \nu_{\text{MW2}} t) S_x + r \Omega_{n} \sin(2\pi \nu_{\text{RF}} t) S_z.
\]

The experimentally determined Rabi frequencies
(\Omega_e = 12.96 \text{ MHz} \text{ and } \Omega_n = 25 \text{ kHz}) \text{ are used in the simulation, with the DC magnetic field fixed at } B_z = 500 \text{ G. Here } r \text{ is a phenomenological parameter to account for an off-axis nuclear drive field that couples to the Zeeman splitting of the electron spin state } [21]. \text{ Dephasing of the electronic spin is modeled using the Lindblad master equation with the electron spin coherence time } T_2 = 300 \mu s \text{ extracted from spin echo experiments. We start the simulation with the system in a pure state } |0, 1 \rangle \text{ before the MW1 initialization pulse is applied. Electron spin relaxation and nuclear spin dephasing are neglected as there is no observable decay on the timescale of our experiments.}

The simulation results shown in Fig. 4(c) indicate that there is a population buildup in the \(|m_S,m_I\rangle = |0,0\rangle\) state after the application of multiple phase gates. This population buildup results in a decrease in the maximum population of \(|-1, +1\rangle\) Rabi oscillations after the phase gate, which agrees with the experimental result. This is due to a RF drive being applied simultaneously with the MW2 pulses, causing the electronic levels to oscillate at a frequency comparable to the electron Rabi frequency \([28]\). A non-ideal rotation on the \(|-1,0\rangle \leftrightarrow |0,0\rangle\) transition during the fast phase gates leaves some residual population in \(|0,0\rangle\). We account for this effect using the second term in Eq. [4] which couples the RF drive to the Zeeman splitting of the electron spin state \([20]\).

Next, we argue that this additional population in \(|0,0\rangle\) does indeed contribute to a “dark” readout. Typically the \(m_S = 0\) population would contribute to a “bright” readout, as the cycling transition associated with \(m_S = 0\) does not involve the intersystem crossing through the singlet state \(1A_1\). However, near the ESLAC, \(|0,0\rangle\) is strongly coupled to \(|-1, +1\rangle\) in the excited state manifold. In contrast to the experiment performed by Morton et al. \([16, 17]\) where the nuclear spin is initialized using thermal polarization, it is precisely this coupling that makes the nuclear spin polarization possible for our experiment as it allows for Larmor precession between the two states near the ESLAC during optical excitation. Thus, \(|0,0\rangle\) can be converted to \(|-1, +1\rangle\) and contribute to a dark state during optical readout due to the non-radiative decay of \(|-1, +1\rangle\) through the singlet state. This sequence of events, which is part of the nuclear spin polarization process, occurs on the same \(\sim 300 \text{ ns}\) timescale as the optical readout \([27]\).

We now show that the population loss is indeed due to energy level shifts associated with the RF pulse. We perform a Ramsey experiment [see Fig. 4(d)] and interrupt the free evolution by applying multiple \(\pi\)-phase gates. Within our measurement error, there is no visible decay of the Ramsey fringe amplitude after the phase gates are applied. These measurements indicate that there is no population loss after many phase gates have been applied during the time when RF is turned off \([20]\).

In conclusion, we have demonstrated fast phase gate operations on a nuclear spin qubit in diamond by driving electronic spin transitions of an NV center. \(\pi\)-phase gates are achieved in 462 ns, approximately 100 times faster than the bare nuclear Rabi frequency. These fast phase gates can be applied repeatedly to preserve the nuclear spin state, providing an alternative method for nuclear spin dynamical decoupling.

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