Fast phase manipulation of the single nuclear spin in solids by rotating fields

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We propose fast phase gates of single nuclear spins interacting with single electron spins. The gate operation utilizes geometric phase shifts of the electron spin induced by fast and slow rotating fields; the path difference depending on nuclear-spin states enables nuclear phase shifts. The gate time is inversely proportional to the frequency of the slow rotating field. As an example, we use nitrogen-vacancy centers in diamond, and show, in principle, the phase-gate time orders of magnitude to be shorter than previously reported. We also confirmed the robustness of the gate against decoherence and systematic errors.

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I. INTRODUCTION

The realization of long-lived qubits with sufficient operability is an essential issue in the field of quantum information. Nuclear spins are one of the most attractive platforms for their prominent spin coherence. On the other hand, nuclear Rabi oscillations are much slower than those of electron spins because of their small magnetic moment. As a solution to this problem, nuclear-spin phase controls via electron-spin transition have been theoretically proposed [1] and experimentally demonstrated [2]. The phase gate is enabled by the hyperfine coupling and off-resonant transitions of the electron spin. Using this method, the gate time can be improved up to the inverse of the hyperfine constant. These phase controls initially require the nuclear-spin preparations ($\pi/2$ pulse) taking several tens of microseconds [2]. On the other hand, they suggested that the nuclear $\pi/2$ pulse is only necessary upon initialization and measurement in many of the models for computation [1]; here, we only focus on the time for phase shifts.

Here, we propose a method for fast phase control of single nuclear spins. As the gate time is not limited by the hyperfine constant, in principle this gate operation is orders of magnitude faster than previously reported. The proposed gate operation utilizes geometric phase shifts of single electron spins [3–7]; here, the electron-spin dynamics is controlled by rotating fields. As the time evolution of the electron spin depends on nuclear-spin states, the rotating field enables nuclear-spin phase gates. We also found that our gates are robust against decoherence and systematic errors. Finally, we show how to implement conditional phase gates using two different nuclear spins.

In the proposed method, we consider single nuclear spins interacting with single electron spins of nitrogen-vacancy (NV) centers in diamond [8–11]. The NV electron spins are well known for their outstanding spin coherence [$T_2 > 1$ ms at room temperature (RT)] [12]. The electron spin couples to a wide range of forces—magnetic, optical, and electrical. Optical excitations allow for spin initialization and readout [13]. Coherent coupling with single photons [14] is used at low temperature in order to generate quantum networks among separate NV centers [15,16]. Stark shifts in the excited state make spin-sublevel shifts in the ground state via spin-orbit couplings at RT [17], and the electric-field effect enables electron-spin controls between $m_s = \pm 1$ states [18]. In the proposed methods, the electron-spin dynamics is controlled by oscillating electric and magnetic fields; for reasons that are discussed later, the electric- and magnetic-field control is superior to solely magnetic-field controls for fast operations.

II. SPIN HAMILTONIAN AND DYNAMICS

In the proposed method, an NV electron spin ($S = 1$) interacts with a single nitrogen-15 (^{15}N) nuclear spin ($I = 1/2$). The ground-state spin Hamiltonian is described [17,19,20] as ($\hbar = 1$ from now on)

$$H_0 = DS_z^2 + \gamma_e B_z S_z + d_\perp E_x\left(S_x^2 - S_y^2\right) + d_\perp E_y(S_x S_y + S_y S_x) + A_{||} S_z I_z + \frac{A_\perp}{2}(S_+ I_- + S_- I_+), \quad (1)$$

where γ_e is the electron gyromagnetic ratio; B_z is the magnetic field oriented along the NV axis. $d_\perp/2\pi (=17$ Hz cm/V) is the electric dipole moment; $E_{x,y}$ are electric fields with directions perpendicular to the NV axis. D is the zero-field splitting parameter, and $A_\parallel(A_\perp)$ is the hyperfine coupling constant. These parameters are set to their reported values of $D/2\pi = 2.87$ GHz, $A_\parallel/2\pi = 3.03$ MHz, and $A_\perp/2\pi = 3.65$ MHz. Here we omit the nuclear Zeeman terms for simplicity as these terms are much smaller than the others. In the following, we use $|\,\rangle_E (|\,\rangle_{\rm N})$ to represent electron (nitrogen nuclear) spin states. We also assume that the large energy splitting, $[A_\perp/(D - \gamma_e B_z)]^2 \sim 10^{-6}$, allows us to neglect the electron-nuclear flip-flop terms of Eq. (1), and the electron spin can be described as a two-level system. The Hamiltonian can be thus simplified in the interaction picture of the zero-field splitting:

$$H = \gamma_e B_z \sigma_z + d_\perp E_x \sigma_x + d_\perp E_y \sigma_y + A_{||}\sigma_z I_z, \quad (2)$$

where $\sigma_i (i = x,y,z)$ are the Pauli operators.

In the proposed method, spin input states are prepared under static electric and magnetic fields. During phase gating of the nuclear spin, we set the static field to zero and apply oscillating electric and magnetic fields simultaneously $\gamma_e B_z = \omega_1 \cos(\omega t), d_\perp E_x = \omega_1 \sin(\omega t)\cos(\varphi)$, and $d_\perp E_y = \omega_1 \sin(\omega t)\sin(\varphi)$. The oscillation of each field amplitude

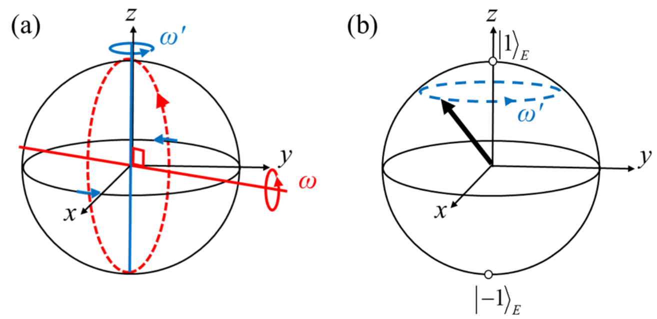

FIG. 1. (a) The inertial force is generated by the rotating field in the laboratory frame (red arrow). Gate operations are performed by adiabatic rotations of the inertial force (blue arrow). (b) The adiabatic rotation of the inertial force leads to the electron-spin cyclic evolution (blue dashed arrow) in the NV electron Bloch square.

induces an inertial force, which controls NV spin states. In the laboratory frame, the Hamiltonian is then rewritten as

$$H(t) = \vec{\omega}_1(\omega t,\varphi)\cdot\vec{\sigma} + A_{||}\sigma_z I_z, \tag{3}$$

where $\vec{\omega}_1(\omega t,\varphi)$ represents the amplitude of each oscillating field on the polar coordinate system. The oscillation of the electric and magnetic fields plays a key role in the proposed gate operation; if the amplitude ω_1 is too small, the effect of the oscillating field is suppressed by the orthogonal static magnetic field (Appendix A). Therefore, we set the amplitude ω_1 about the same as the hyperfine constant $A_{||}$. On the other hand, if we use solely magnetic-field controls instead of electric- and magnetic-field controls, the large zero-field splitting makes it difficult to observe the effect of transverse magnetic fields.

The three oscillating-field components of Eq. (3) correspond to one rotating field, whose rotational axis is perpendicular to the z axis of the NV-defect coordinate system, (x, y, z) [Fig. 1(a)]. Thus it is useful to set another coordinate system, (X', Y', Z'), where Z' indicates the rotational axis. The coordinate transform is described as

$$\begin{pmatrix}\sigma_x\\ \sigma_y\\ \sigma_z\end{pmatrix} = \begin{pmatrix}\cos\varphi & -\sin\varphi & \\ \sin\varphi & \cos\varphi & \\ 1 & & \end{pmatrix}\begin{pmatrix}\sigma'_X(\varphi)\\ \sigma'_Y(\varphi)\\ \sigma'_Z(\varphi)\end{pmatrix}. \tag{4}$$

The Hamiltonian in the (X', Y', Z')-coordinate system is then represented as

$$H(t) = \omega_1[\cos(\omega t)\sigma'_X(\varphi) + \sin(\omega t)\sigma'_Y(\varphi)] + A_{||}\sigma'_X(\varphi)I_z. \tag{5}$$

In analyzing the effect of this rotating field, it is useful to employ a rotating frame. Using a unitary operator,

$$U_1(t) = \exp\left[-i\frac{\omega t}{2}\sigma'{}_Z(\varphi)\right], \tag{6}$$

the rotating-frame Hamiltonian is described as $U_1^\dagger(t)H(t)U_1(t) - iU_1^\dagger(t)d/dtU_1(t) = H_1 + V_1(t)$, where

$$H_1 = \omega_1\sigma'_X(\varphi) - \frac{\omega}{2}\sigma'_Z(\varphi), \tag{7a}$$

$$V_1(t) = A_{||}\exp\left[i\frac{\omega t}{2}\sigma'{}_Z(\varphi)\right]\sigma'_X(\varphi)\exp\left[-i\frac{\omega t}{2}\sigma'{}_Z(\varphi)\right]I_z. \tag{7b}$$

Here, H_1 are the nonperturbation terms, and $V_1(t)$ is the perturbation term. We next represent the operator $U_1(t)$ in the (x, y, z)-coordinate system. From Eq. (4), the matrix, $\sigma'_Z(\varphi)$, is written as $\sigma'_Z(\varphi) = \exp(-i\varphi\sigma_z/2)\sigma_y\exp(i\varphi\sigma_z/2)$. The operator $U_1(t)$ can then be described in the (x, y, z)-coordinate system as

$$U_1(t) = \exp\left(-i\frac{\varphi}{2}\sigma_z\right)\exp\left(-i\frac{\omega t}{2}\sigma_y\right)\exp\left(i\frac{\varphi}{2}\sigma_z\right). \tag{8}$$

The rotating-frame Hamiltonian of Eq. (7) denotes that the electron spin interacts with the inertial force of the rotational field, $-iU_1^\dagger(t)d/dtU_1(t)$. However, in rotating fields with too high frequency, $|A_{\|}/\omega|$, $|\omega_1/\omega| \ll 1$, dynamic and geometric phases induced by the inertial force are nearly canceled [21]. Thus it is difficult to perform fast phase gates by one rotating field with too high frequency. By contrast, we next show that using two rotating fields with different frequencies allows for accumulation of the geometric phase without cancellation, even in rotational fields with sufficiently high frequency.

We consider that when the rotating-frame Hamiltonian changes adiabatically, the spin follows it [21]. This inertial-force change can be implemented through phase shifts of the electric field, $\varphi \to \omega' t$, which satisfy the following condition, $\omega'/\omega \ll 1$. From Eq. (8), the inertial-force change causes z operation in the (x, y, z)-coordinate system. Thus in analyzing the inertial-force change, it is useful to use the (x, y, z)-coordinate system. The nonperturbation term, H_1, is described in another rotating frame by using a unitary operator,

$$U_2(t) = \exp\left(-i\frac{\omega' t}{2}\sigma_z\right), \tag{9}$$

as $H_2 \equiv U_2^\dagger(t)H_1U_2(t) - iU_2^\dagger(t)d/dt\,U_2(t) = (\omega_1 - \omega'/2)\sigma_z - \omega\sigma_y/2$. The eigenvectors can be described under the following conditions, $\omega \gg \omega' - 2\omega_1$ as

$$|+'\rangle_E \equiv \frac{1}{\sqrt{2(1+\delta)}}[i(1+\delta)|1\rangle_E + |-1\rangle_E], \tag{10a}$$

$$|-'\rangle_E \equiv \frac{1}{\sqrt{2(1+\delta)}}[|1\rangle_E + i(1+\delta)|-1\rangle_E], \tag{10b}$$

where $\delta \equiv 2(\omega_1 - \omega'/2)/\omega$. Under the approximation $\delta \sim 0$, the diagonal Hamiltonian, H_2, is simplified as

$$H_2 \sim -\frac{\omega}{2}\sigma_y. \tag{11}$$

We next consider the perturbation term [Eq. (7b)] in the (x, y, z)-coordinate system. According to Eqs. (8), (9), and (11), the perturbation term is described in the interaction picture of H_2 as

$$V_I = \exp(iH_2t)U_2^\dagger(t)V_1(t)U_2(t)\exp(-iH_2t) = A_{||}\sigma_z I_z. \tag{12}$$

According to Eqs. (8), (9), (11), and (12), the time evolution in the laboratory frame is

$$U(t) = \exp\left(-i\frac{\omega' t}{2}\sigma_z\right)\exp(-iA_{||}t\sigma_z I_z). \tag{13}$$

We last consider the cyclic evolution of the electron spin. After one cyclic operation, the spin acquires a global phase,

TABLE I. Applied electric and magnetic fields. The amplitudes of the static fields satisfy $|\gamma_e B_0| \gg |A_{||}|, |d_\perp E_0|$. The frequencies of oscillating fields satisfy $\omega \gg \omega'$, and the amplitude ω_1 is about the same as the hyperfine constant $A_{||}$.

	Static fields	Oscillating fields
Magnetic fields (z axis)	$\gamma_e B_0$	$\omega_1 \cos(\omega t)$
Electric fields (x axis)	$d_\perp E_0$	$\omega_1 \sin(\omega t)\cos(\omega' t)$
Electric fields (y axis)	0	$\omega_1 \sin(\omega t)\sin(\omega' t)$

$U(\tau)|\psi(0)\rangle_E|\uparrow(\downarrow)\rangle_{\rm N} = \exp[-i\Omega_{\uparrow(\downarrow)}(\tau)]|\psi(0)\rangle_E|\uparrow(\downarrow)\rangle_{\rm N}$, whose phase factor is determined to be

$$- i\Omega_{\uparrow(\downarrow)}(\tau) = \int_0^\tau dt \langle\uparrow(\downarrow)|\langle\psi(0)|U^\dagger(t) \times \left[\frac{d}{dt}U(t)\right]|\psi(0)\rangle_E|\uparrow(\downarrow)\rangle_{\rm N}. \quad (14)$$

This global phase depends on the time evolution operator, $U(t)$, and on the input state of the electron spin. According to Eq. (7), the inertial force does not interact with nuclear spins; thus the nuclear gate operation requires that the input state of the electron spin depends on nuclear-spin states.

III. PHASE GATING OPERATION

We first summarize the outline of our phase controls and the physical assumptions (Table I). First, we prepare the total spin state by electron-spin π pulses and nuclear-spin $\pi/2$ pulses as $1/\sqrt{2}|1'\rangle_E(|\uparrow\rangle_{\rm N} + |\downarrow\rangle_{\rm N})$ under static electric and magnetic fields (E_0, B_0), where $|1'\rangle_E$ is one of the electron-spin eigenstates (Appendix B); for keeping long coherence time, the amplitudes of the fields should satisfy $|\gamma_e B_0| \gg |A_{||}|, |d_\perp E_0|$ (Appendix C). Before phase gating, we instantly turn off the static fields, and turn on oscillating electric and magnetic fields simultaneously [Eq. (3)] having an equal amplitude ω_1 and oscillating frequency ω. The fast gate operations are implemented by the phase shift of the oscillating electric fields as $\varphi = \omega' t$. Here, the field amplitude ω_1 is about the same as the hyperfine constant $A_{||}$ (Appendix A), and each frequency satisfies $\omega \gg \omega'$. In addition, during the switch processes between the static or oscillating fields, the spin state should not be disturbed; thus this process should be done suddenly. This time scale should be much shorter than the electron-spin precession. Therefore, the static (oscillating) fields should be turned on or off much faster than $2\pi/\gamma_e B_0$, $(2\pi/\omega')$. In these conditions, the spin state is not changed in the switching process.

We first discuss the input spin state under static fields (E_0, B_0). The input state of the electron spin, $|\psi(0)\rangle_E$, corresponds to the eigenstate of the static-field Hamiltonian (Appendix B),

$$|1'\rangle_E|\uparrow(\downarrow)\rangle_{\rm N} = \left[\cos\left(\frac{1}{2}\theta_{\uparrow(\downarrow)}\right)|1\rangle_E + \sin\left(\frac{1}{2}\theta_{\uparrow(\downarrow)}\right)|-1\rangle_E\right]|\uparrow(\downarrow)\rangle_{\rm N}, \quad (15a)$$

$$\cos\left(\frac{1}{2}\theta_{\uparrow(\downarrow)}\right) = \frac{1}{C_{\uparrow(\downarrow)}}\left\{\gamma_e B_0 + (-)\frac{A_{||}}{2} + \sqrt{\left[\gamma_e B_0 + (-)\frac{A_{||}}{2}\right]^2 + (d_\perp E_0)^2}\right\}, \quad (15b)$$

$$\sin\left(\frac{1}{2}\theta_{\uparrow(\downarrow)}\right) = \frac{d_\perp E_0}{C_{\uparrow(\downarrow)}}, \quad (15c)$$

where $C_{\uparrow(\downarrow)}$ is a normalized constant. The input state of the nuclear spin is prepared by magnetic-resonance controls as $1/\sqrt{2}|1'\rangle_E(|\uparrow\rangle_{\rm N} + |\downarrow\rangle_{\rm N})$. According to Eq. (15), we found that using the large magnetic field is necessary to suppress decoherence under the static field, because the electric-field effect induces decoherence of the nuclear spin via the hyperfine interaction. In large magnetic fields, the coherence time can exceed 1 ms (Appendix C), which is almost equal to the coherence time of the nuclear spin limited by the relaxation time of the NV electron spin at RT ($T_{1e} \sim 5$ ms) [22].

Under oscillating fields, the electron spin performs cyclic evolutions, following which the electron spin incurs phase shifts depending on the nuclear spin states, $\Omega_{\uparrow(\downarrow)}$. From Eqs. (14) and (15), the phase factor $\Omega_{\uparrow(\downarrow)}$ depends on the expected value $\langle\uparrow(\downarrow)|\langle 1'|\sigma_z|1'\rangle_E|\uparrow(\downarrow)\rangle_{\rm N} = \cos\theta_{\uparrow(\downarrow)}$. This is approximated under large magnetic fields, $|\gamma_e B_0| \gg |A_{||}|$, $|d_\perp E_0|$, as

$$\cos\theta_{\uparrow(\downarrow)} \sim 1 - \frac{1}{2}\left[\frac{d_\perp E_0}{\gamma_e B_0 + (-)A_{||}/2}\right]^2, \quad (16)$$

where we take lowest-order terms. The gate speed is defined as $\Delta\Omega \equiv d/dt(\Omega_\uparrow - \Omega_\downarrow)$, which is calculated as (Appendix D)

$$\Delta\Omega \sim \frac{\omega'}{2}\left(\frac{d_\perp E_0}{\gamma_e B_0}\right)^2 \frac{A_{||}}{\gamma_e B_0} + A_{||}. \quad (17)$$

Here, the nuclear-spin phase is detected in a rotating frame whose angular velocity corresponds to energy splitting of the nuclear spin under the static field. Thus the second term of Eq. (17) is canceled out. This result denotes that the gate speed is proportional to the frequency ω'. When, for example, the rotational frequency of the electric field is set to $\omega'/2\pi = 1.0$ GHz, the gate time is about 165 ns (Fig. 2); this is almost equal to the theoretical limit of nuclear phase gates using hyperfine interactions and electron-spin transitions ($\pi/A_{||} \sim 165$ ns) [1,2]. Moreover, we can apply the oscillating fields with much higher frequency. In experiments, the key point is how to apply electric fields with high frequency. We consider that the oscillating fields with THz frequency have little loss in intrinsic bulk diamonds [23,24]. Moreover, diamonds are not piezoelectric [25], and we need not consider the time delay by strain effects. Thus we estimate that the theoretical limit of our gate time is in principle much shorter than 100 ns.

IV. DECOHERENCE

Geometric phase shifts are, in general, very sensitive to random fluctuation about the path. According to Eq. (17),

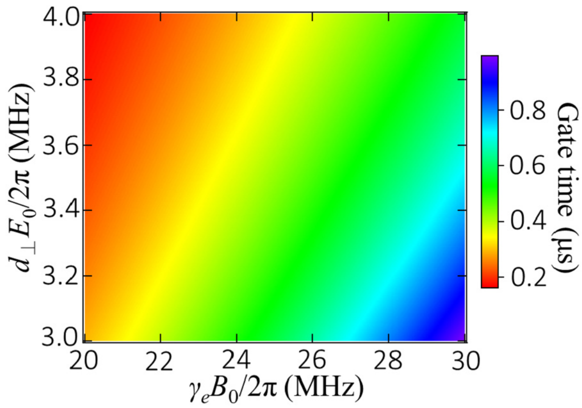

FIG. 2. Gate time of the proposed nuclear-spin phase gate. $\gamma_e B_0$ and $d_\perp E_0$ correspond to the amplitude of the static magnetic and electric fields, respectively. The frequency of the slow electric-field rotation is $\omega'/2\pi = 1.0$ GHz.

decoherence is mainly caused by magnetic-field fluctuations of nuclear-spin baths [26,27] and by the frequency fluctuations of the oscillating fields, ω'. Here, we assume a nuclear-spin bath composed of ^{13}C at a concentration of 0.03%, and the noise amplitude is $\gamma_e \delta B/2\pi = 0.02$ MHz [26]. The noise magnetic field, δB, can be described by adding it to the static magnetic field as $B_0 \to B_0 + \delta B$. Similarly, we assume that the unwanted shift of the field frequencies, $\delta\omega$, is less than several kHz. The unwanted shift, $\delta\omega$, can be also described as $\omega' \to \omega' + \delta\omega$.

The effective noise amplitude of the nuclear spin can be defined from the nuclear phase shifts, $\Delta\Omega(B_0,\omega')$, in Eq. (17) as $\gamma_n \delta B_N \equiv |\Delta\Omega(B_0 + \delta B, \omega' + \delta\omega) - \Delta\Omega(B_0,\omega')| \sim |(\Delta\Omega(B_0,\omega') - A_{||})\{\delta\omega/\omega' - 3\delta B/B_0\}|$, where the error parameters are sufficiently small, $|\delta\omega/\omega'|, |\delta B/B_0| \ll 1$, and we take lowest-order terms. Moreover, when the frequency, ω', is large enough, as $|\delta\omega/\omega'| \ll |\delta B/B_0|$, the frequency error, $\delta\omega$, is not significant. Thus the effective noise amplitude, $\gamma_n \delta B_{\rm N}$, can be simplified as $\gamma_n \delta B_{\rm N} \sim |3(\Delta\Omega(B_0,\omega') - A_{||})\delta B/B_0|$. In addition, the random fluctuation can be described by a correlation function of random classical fields, $f(t)$ [28]. The field average is zero, $\langle f(t)\rangle = 0$, and the autocorrelation is $\langle f(t) f(0)\rangle = \exp(-t/\tau_c)$, where τ_c is the correlation time of the nuclear-spin bath. The nuclear noise Hamiltonian is then represented as $H^{\rm N}_{\rm noise} = \gamma_n \delta B_{\rm N} f(t) I_z$.

The decoherence rate can be estimated by second-order calculations of the von Neumann equation. We set the input state of the nuclear spin as $\rho_{\rm N}(0) = |+\rangle\langle+|_{\rm N}$, where $|+\rangle_{\rm N}$ is a superposition state, $|+\rangle_N = 1/\sqrt{2}(|\uparrow\rangle_{\rm N} + |\downarrow\rangle_{\rm N})$. The ensemble-averaged density matrix $\bar\rho_{\rm N}(t)$ shows nuclear-spin coherence, which is approximated as $\langle+|\bar\rho_{\rm N}(t)|+\rangle_{\rm N} \sim \exp[-\xi(t)]$, where $\xi(t) \sim (\gamma_n \delta B_{\rm N} t/2)^2$ under slow fluctuations of the nuclear-spin bath, $\tau_c \gg t$ [28]. The coherence time without echo is

$$\frac{1}{T^*_{2\rm N}} = \frac{3\omega'}{4}\left(\frac{d_\perp E_0}{\gamma_e B_0}\right)^2 \frac{A_{||}}{\gamma_e B_0}\frac{\delta B}{B_0}. \tag{18}$$

Here, gate errors by decoherence can be estimated as $\varepsilon_{\rm dec} = 1$–fidelity; fidelity is defined by ${\rm tr}[\rho_{\rm N}(t)\rho'_{\rm N}(t)]$ [22], where $\rho_{\rm N}(t)$ is the ideal density matrix, and $\rho'_{\rm N}(t)$ is the density matrix with errors. The fidelity is calculated as $\frac{1}{2}\{1 + \exp[-(t/T^*_{2\rm N})^2]\}$. According to Eq. (17), the gate error of the nuclear phase gate can be approximated as

$$\varepsilon_{\rm dec}(B_0) \sim \left(\frac{3\pi}{2\sqrt{2}}\frac{\delta B}{B_0}\right)^2, \tag{19}$$

where we take lowest-order terms. In the simulation shown in Fig. 3(a), it is seen that large static magnetic fields allow for robust spin control.

V. SYSTEMATIC ERRORS

Robustness against systematic errors is of practical importance in quantum information processing [29,30]. Here, we consider systematic errors by unwanted shifts of the static field (E_0, B_0) and by the amplitude mismatch of the oscillating field, ω_1.

Unwanted shifts of the static fields can be represented as $E_0 \to E_0 + \Delta E, B_0 \to B_0 + \Delta B$. In the proposed method, we focus on nuclear spins, and the gate error can be also estimated as $\varepsilon_{\rm sys} = 1$–fidelity. The systematic error

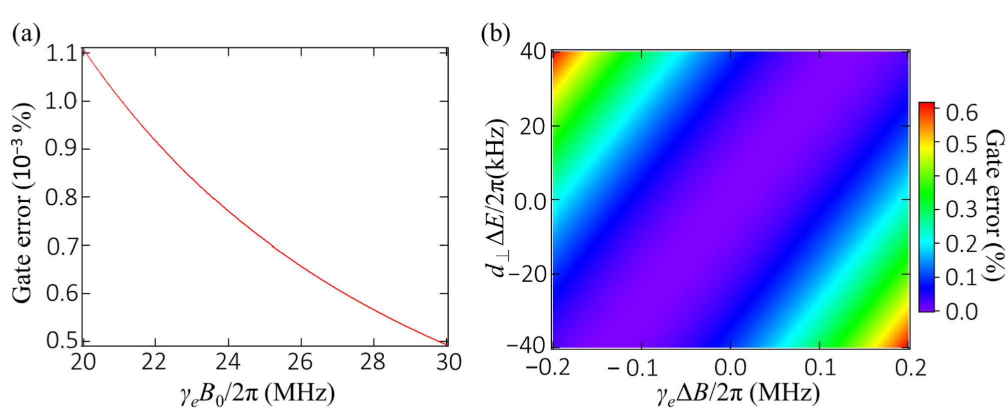

FIG. 3. Robustness against decoherence and systematic errors. (a) Stochastic errors by decoherence: $\gamma_e B_0$ correspond to the amplitude of static magnetic fields. The noise amplitude is $\gamma_e \delta B/2\pi = 0.02$ MHz. (b) Systematic errors: ΔB, ΔE denote unwanted shifts of the static magnetic and electric fields, respectively. The amplitudes of the static electric and magnetic fields are $d_\perp E_0/2\pi = 4.0$ MHz and $\gamma_e B_0/2\pi = 20$ MHz, respectively.

causes unwanted unitary transformations, but does not cause decoherence; thus ρ_N and ρ'_N are calculated as pure states. The systematic error of the nuclear phase gate can be described as

$$\varepsilon_{sys}(\Delta B,\Delta E) \sim 4\pi^2\left(-\frac{3\Delta B}{4B_0}+\frac{\Delta E}{2E_0}\right)^2, \tag{20}$$

where we assume that the unwanted shifts are sufficiently small, $|\Delta E/E_0|, |\Delta B/B_0| \ll 1$, and take lowest-order terms. Based on this, it is seen that large static electric and magnetic fields allow for robust spin control [Fig. 3(b)].

The proposal method, according to Eq. (3), requires the three oscillating fields to have the exact same frequency (ω, ω'), and this can be implemented by using the same high-frequency signal generators corresponding to each frequency. On the other hand, the proposal method also requires the three oscillating fields to have the same amplitude; in practice, each field may have a small mismatch. According to the Hamiltonian $H(t)$ of Eq. (3), this error Hamiltonian, $H_{error}(t)$, can be described as

$$H_{error}(t) = \delta\omega_{||}\cos(\omega t)\sigma_z + \delta\omega_{\perp}\sin(\omega t) \times [\cos(\omega' t)\sigma_x - \sin(\omega' t)\sigma_y]. \tag{21}$$

The effective Hamiltonian is $H(t)+H_{error}(t)$, where the error parameters are small enough, $|\delta\omega_{||}|, |\delta\omega_{\perp}| \ll |\omega_1|, |A_{||}|$. This error Hamiltonian also includes three oscillating fields having the same frequency $(\omega, -\omega')$, but the error terms do not generate inertial forces. This is because, from the similar discussion of Appendix A, the field rotation of the error terms hardly affects the eigenvectors of the snapshot effective Hamiltonian. This denotes that the field rotation of the errors is almost neglected in the effective Hamiltonian. Here, these terms can be calculated as perturbations. From a similar discussion of Eq. (12), the perturbation terms of Eq. (12) are rewritten as

$$V'_I = A_{||}\sigma_z I_z + \delta\omega_{||}\cos(\omega t)\sigma_z + \delta\omega_{\perp}\sin(\omega t)[\cos(-2\omega' t)\sigma_x + \sin(-2\omega' t)\sigma_y]. \tag{22}$$

Under the following conditions, $|\delta\omega_{||}/\omega|, |\delta\omega_{\perp}/\omega| \ll 1$, $\omega' \ll \omega$, the fast oscillating terms can be neglected. Therefore, we estimate that the intensity mismatch of the oscillating fields, ω_1, is not significant when the error terms are small enough.

VI. CONDITIONAL GATE

Conditional phase gates are necessary for quantum computation. Here, we show ^{15}N nuclear phase gates controlled by nuclear spins of a third-nearest-neighbor carbon-13 (^{13}C). The hyperfine constant of the ^{13}C (^{15}N) nucleus is described as $A_{||}{}^{C}(A_{||}{}^{N})$, where $A_{||}{}^{C}/2\pi = 14$ MHz [26]. This operation requires phase shifts that differ depending on the ^{13}C nuclear-spin state, $\Delta\Omega_{C=\pm}$. To calculate these shifts, we add hyperfine coupling of the ^{13}C nucleus to the effective magnetic field as follows: $\gamma_e B_0 \to \gamma_e B_0 \pm A_{||}{}^{C}/2$. Here, we assume that the static magnetic field satisfies $|A_{||}{}^{N}/(2\gamma_e B_0 \pm A_{||}{}^{C})| \ll 1$, and from Eq. (17), the relative phase shift is

$$\Delta\Omega_{C=+} - \Delta\Omega_{C=-} \sim -\frac{\omega'}{2}\frac{A_{||}{}^{N}(d_{\perp}E_0)^2}{(\gamma_e B_0)^3}\left[\sum_{k=1} 2k(2k+1)\left(\frac{A_{||}{}^{C}}{2\gamma_e B_0}\right)^{2k-1}\right]. \tag{23}$$

The conditional gate corresponds to $(\Delta\Omega_{C=+} - \Delta\Omega_{C=-})t = \pi$. When we set the respective parameters to $\omega'/2\pi = 1.0$ GHz, $d_{\perp}E_0/2\pi = 4.0$ MHz, and $\gamma_e B_0/2\pi = 40$ MHz, the conditional gate time is about 1 μs.

VII. CONCLUSION

We have proposed a phase gate of single nuclear spins controlled by fast and slow rotating fields. The nuclear gate time is, in principle, much shorter than previously reported, which is limited by the hyperfine constant [1]. We showed the robustness of the proposed method against decoherence and systematic errors. Multinuclear operation was also confirmed. It should be noted that our methods are not limited only to NV-spin systems, and are applicable to many quantum systems such as ion trap, spins, and superconducting qubits. Our result is a significant step for outstanding operability of long-lived quantum memories.

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APPENDIX A: THRESHOLD OF THE ROTATING-FIELD AMPLITUDE

In the proposed gate operation, the oscillation of electric and magnetic fields plays a key role. If the amplitude ω_1 is too small, the oscillating-field effect is suppressed by the orthogonal static magnetic field. This is confirmed from the snapshot Hamiltonian of the electron spin,

$$H_{snap} = \omega_1\sigma_x + \frac{\omega_0}{2}\sigma_z, \tag{A1}$$

where the parameters $\varphi, \omega t$ of Eq. (3) are set to $\varphi = 0$ and $\omega t = \pi/2$, and ω_0 corresponds to the hyperfine constant $A_{||}$. Under small electric and magnetic-field conditions, $|\omega_1/\omega_0| \ll 1$, the eigenstates are

$$|\psi_1\rangle_E = \frac{1}{\sqrt{\omega_1{}^2+\omega_0{}^2}}(\omega_0|1\rangle_E + \omega_1|-1\rangle_E), \tag{A2a}$$

$$|\psi_{-1}\rangle_E = \frac{1}{\sqrt{\omega_1{}^2+\omega_0{}^2}}(\omega_1|1\rangle_E - \omega_0|-1\rangle_E). \tag{A2b}$$

If the oscillating-field effect is completely suppressed, these eigenstates are not changed from that of the nonoscillating field, $\omega_1 = 0$. The approximation of Eq. (2) denotes that, under the following conditions, the oscillating-field effect should be calculated without neglecting hyperfine off-diagonal terms:

$$|\langle -1 \mid \psi_1\rangle_E|^2 \sim |\langle 1 \mid \psi_{-1}\rangle_E|^2 \sim \left(\frac{\omega_1}{\omega_0}\right)^2 \leqslant 10^{-7}. \quad \text{(A3)}$$

Thus the amplitude of oscillating fields at least satisfies the condition $(\omega_1/\omega_0)^2 \gg 10^{-7}$.

APPENDIX B: STATIC-FIELD HAMILTONIAN

The static-field Hamiltonian is, from Eq. (2), described as

$$H_{\rm sta} = \gamma_e B_0 \sigma_z + d_\perp E_0 \sigma_x + A_{||}\sigma_z I_z, \quad \text{(B1)}$$

where B_0 and E_0 are static magnetic and electric fields, respectively. The diagonal Hamiltonian is

$$\begin{aligned} H_{\rm sta} = {} & \sqrt{\left(\gamma_e B_0 + \frac{A_{||}}{2}\right)^2 + (d_\perp E_0)^2} |1'\rangle\langle 1'|_E |\uparrow\rangle\langle\uparrow|_N \\ & + \sqrt{\left(\gamma_e B_0 - \frac{A_{||}}{2}\right)^2 + (d_\perp E_0)^2} |1'\rangle\langle 1'|_E |\downarrow\rangle\langle\downarrow|_{\rm N} \\ & - \sqrt{\left(\gamma_e B_0 + \frac{A_{||}}{2}\right)^2 + (d_\perp E_0)^2} |-1'\rangle\langle -1'|_E |\uparrow\rangle\langle\uparrow|_N \\ & - \sqrt{\left(\gamma_e B_0 - \frac{A_{||}}{2}\right)^2 + (d_\perp E_0)^2} |-1'\rangle\langle -1'|_E |\downarrow\rangle\langle\downarrow|_{\rm N}, \end{aligned} \quad \text{(B2)}$$

and each electron eigenstate is

$$|1'\rangle_E|\uparrow(\downarrow)\rangle_{\rm N} = \frac{1}{C_{\uparrow(\downarrow)}}\left(\left\{\gamma_e B_0 + (-)\frac{A_{||}}{2} + \sqrt{\left[\gamma_e B_0 + (-)\frac{A_{||}}{2}\right]^2 + (d_\perp E_0)^2}\right\}|1\rangle_E + d_\perp E_0 |-1\rangle_E\right)|\uparrow(\downarrow)\rangle_{\rm N}, \quad \text{(B3a)}$$

$$|-1'\rangle_E|\uparrow(\downarrow)\rangle_{\rm N} = \frac{1}{C_{\uparrow(\downarrow)}}\left(d_\perp E_0|1\rangle_E - \left\{\gamma_e B_0 + (-)\frac{A_{||}}{2} + \sqrt{\left[\gamma_e B_0 + (-)\frac{A_{||}}{2}\right]^2 + (d_\perp E_0)^2}\right\}|-1\rangle_E\right)|\uparrow(\downarrow)\rangle_{\rm N}, \quad \text{(B3b)}$$

where $C_{\uparrow(\downarrow)}$ is a normalized constant. The electron eigenstates depend on the nuclear-spin states. Here, each factor can be approximated under large magnetic fields, $|\gamma_e B_0| \gg |d_\perp E_0|$, $|A_{||}|$, as

$$\begin{aligned} \sqrt{\left(\gamma_e B_0 \pm \frac{A_{||}}{2}\right)^2 + (d_\perp E_0)^2} \sim {} & \gamma_e B_0\left[1 + \frac{1}{2}\left(\frac{d_\perp E_0}{\gamma_e B_0}\right)^2\right] \\ & \pm \frac{A_{||}}{2}\left[1 - \frac{1}{2}\left(\frac{d_\perp E_0}{\gamma_e B_0}\right)^2\right], \end{aligned} \quad \text{(B4)}$$

where we take lowest-order terms. Thus the static-field Hamiltonian is rewritten as

$$\begin{aligned} H_{\rm sta} \sim {} & \gamma_e B_0\left[1 + \frac{1}{2}\left(\frac{d_\perp E_0}{\gamma_e B_0}\right)^2\right] S'_Z \\ & + A_{||}\left[1 - \frac{1}{2}\left(\frac{d_\perp E_0}{\gamma_e B_0}\right)^2\right] S'_Z I_Z, \end{aligned} \quad \text{(B5)}$$

where S'_z is defined as $S'_z \equiv |1'\rangle\langle 1'|_E - |-1'\rangle\langle -1'|_E$.

APPENDIX C: DECOHERENCE UNDER STATIC FIELDS

Here, we assume that the noisy magnetic fields are sufficiently small, $|\delta B/B_0| \ll 1$. By adding a noisy magnetic-field term as $B_0 + \delta B$, Eq. (B4) becomes

$$\sqrt{\left(\gamma_e B_0 \pm \frac{A_{||}}{2} + \gamma_e \delta B\right)^2 + (d_\perp E_0)^2}. \quad \text{(C1)}$$

Under large static magnetic fields, this is approximated as

$$\begin{aligned} & \sim \sqrt{\left(\gamma_e B_0 \pm \frac{A_{||}}{2}\right)^2 + (d_\perp E_0)^2} \\ & \quad \times \left[1 + \gamma_e \delta B \frac{\gamma_e B_0 \pm A_{||}/2}{(\gamma_e B_0 \pm A_{||}/2)^2 + (d_\perp E_0)^2}\right]. \end{aligned} \quad \text{(C2)}$$

Thus the noise terms are separated from the non-noise terms. Using the following approximations,

$$\begin{aligned} & \sqrt{\left(\gamma_e B_0 \pm \frac{A_{||}}{2}\right)^2 + (d_\perp E_0)^2} \\ & \quad \sim \left(\gamma_e B_0 \pm \frac{A_{||}}{2}\right)\left[1 + \frac{1}{2}\left(\frac{d_\perp E_0}{\gamma_e B_0 \pm A_{||}/2}\right)^2\right], \end{aligned} \quad \text{(C3a)}$$

$$\begin{aligned} & \frac{\gamma_e B_0 \pm A_{||}/2}{(\gamma_e B_0 \pm A_{||}/2)^2 + (d_\perp E_0)^2} \\ & \quad \sim \frac{1}{\gamma_e B_0 \pm A_{||}/2}\left[1 - \left(\frac{d_\perp E_0}{\gamma_e B_0 \pm A_{||}/2}\right)^2\right], \end{aligned} \quad \text{(C3b)}$$

the noise terms of Eq. (C2) can be rewritten as

$$\gamma_e \delta B\left[1 - \frac{1}{2}\left(\frac{d_\perp E_0}{\gamma_e B_0}\right)^2\left(1 \mp \frac{A_{||}}{\gamma_e B_0}\right)\right], \quad \text{(C4)}$$

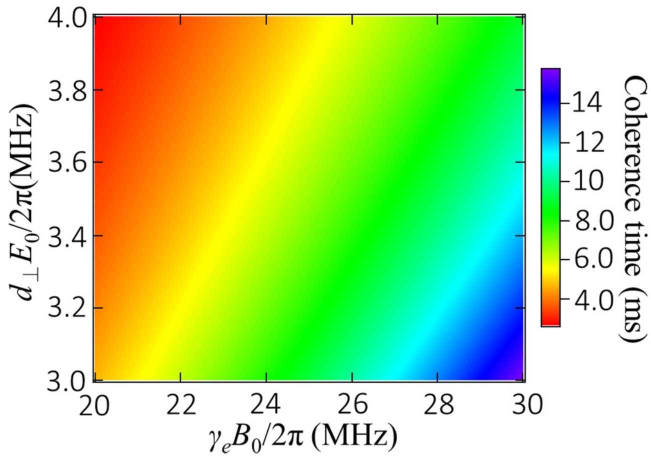

FIG. 4. Coherence time under static electric and magnetic fields: $\gamma_e B_0, d_\perp E_0$ correspond to the amplitude of the static magnetic and electric fields, respectively. The noise amplitude is $\gamma_e \delta B/2\pi = 0.02$ MHz.

where we take lowest-order terms. The noise static-field Hamiltonian is shown as

$$H_{\rm sta}^{\rm noise} \sim \gamma_e \delta B \left[1 - \frac{1}{2}\left(\frac{d_\perp E_0}{\gamma_e B_0}\right)^2\right] S_z' + \gamma_e \delta B \left(\frac{d_\perp E}{\gamma_e B_0}\right)^2 \frac{A_{||}}{\gamma_e B_0} S_z' I_z. \tag{C5}$$

In the proposed method, the electron spin is in its eigenstate under static fields, and we focus on nuclear-spin decoherence. From Eq. (C5), the effective noise Hamiltonian of the nuclear spin can be described as

$$V_{\rm noise}^{\rm N} = b f(t) I_z, \tag{C6}$$

where the noise amplitude b is

$$b = \gamma_e \delta B \left(\frac{d_\perp E_0}{\gamma_e B_0}\right)^2 \frac{A_{||}}{\gamma_e B_0}, \tag{C7}$$

where $f(t)$ is the correlation function described in Sec. IV.

The initial state of the nuclear spin is set as $\rho_{\rm N} = |+\rangle\langle+|_{\rm N}$, where $|+\rangle_{\rm N} = 1/\sqrt{2}(|\uparrow\rangle_{\rm N} + |\downarrow\rangle_{\rm N})$. The time evolution can be estimated by second-order calculations of the von Neumann equation. From the similar calculation of Eq. (18), the coherence time of the nuclear spin is

$$\frac{1}{T_{2\rm N}^*} \sim \frac{b}{2} = \frac{\gamma_e \delta B}{2}\left(\frac{d_\perp E_0}{\gamma_e B_0}\right)^2 \frac{A_{||}}{\gamma_e B_0}. \tag{C8}$$

In the simulation shown in Fig. 4, we assume the nuclear-spin bath composed of ^{13}C at a concentration of 0.03% and the noise amplitude of $\gamma_e \delta B/2\pi = 0.02$ MHz. The coherence time is longer than 1 ms, which is almost equal to the nuclear coherence time limited by the relaxation time of the NV electron spin at RT ($T_{1e} \sim 5$ ms).

APPENDIX D: PHASE SHIFT OF THE NUCLEAR SPIN

The gate speed, $\Delta\Omega \equiv d/dt(\Omega_\uparrow - \Omega_\downarrow)$, is, from Eqs. (13)–(16), calculated as

$$\Delta\Omega = A_{||} - \frac{d_\perp E_0}{2}\left(\frac{d_\perp E_0}{\gamma_e B_0 + A_{||}/2} - \frac{d_\perp E_0}{\gamma_e B_0 - A_{||}/2}\right) - \frac{\omega'}{4}\left[\left(\frac{d_\perp E_0}{\gamma_e B_0 + A_{||}/2}\right)^2 - \left(\frac{d_\perp E_0}{\gamma_e B_0 - A_{||}/2}\right)^2\right]. \tag{D1}$$

Under large magnetic fields, $|\gamma_e B_0| \gg |d_\perp E_0|, |A_{||}|$, the second terms are approximated as

$$\frac{d_\perp E_0}{2}\left(\frac{d_\perp E_0}{\gamma_e B_0 + A_{||}/2} - \frac{d_\perp E_0}{\gamma_e B_0 - A_{||}/2}\right) \sim -\frac{A_{||}}{2}\left(\frac{d_\perp E_0}{\gamma_e B_0}\right)^2. \tag{D2}$$

This term is much smaller than the first term, and we thus neglect it. The third terms are also approximated as

$$\frac{\omega'}{4}\left[\left(\frac{d_\perp E_0}{\gamma_e B_0 + A_{||}/2}\right)^2 - \left(\frac{d_\perp E_0}{\gamma_e B_0 - A_{||}/2}\right)^2\right] \sim -\frac{\omega'}{2}\left(\frac{d_\perp E_0}{\gamma_e B_0}\right)^2 \frac{A_{||}}{\gamma_e B_0}. \tag{D3}$$

Thus the gate speed can be described as

$$\Delta\Omega \sim \frac{\omega'}{2}\left(\frac{d_\perp E_0}{\gamma_e B_0}\right)^2 \frac{A_{||}}{\gamma_e B_0} + A_{||}. \tag{D4}$$

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