Nuclear electric resonance

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Nuclear-spin qubits have long coherence time and are desirably applied into quantum information processing. However, the existing methods either fail to address single nucleus (such as nuclear magnetic resonance), or severely affect nuclear coherence time (such as electrical nuclear manipulation based on hyperfine stark effect, ENMHSE). Here we propose an electrical nuclear manipulation called nuclear electric resonance which can address the single nuclear qubit, and on the other keep the long coherence time. Applying this, we construct universal quantum gates with external electric field. These universal gates are practicable for arbitrary $S \geq 1$ spin nuclei. Given the much longer coherence time of nuclear electric resonance, we improve the number of single-qubit operations by three orders of magnitude compared with that of ENMHSE.

I. INTRODUCTION

Quantum computing [1–8] can solve some important complex problems which go beyond the power of classical information processing. Decoherence is the biggest barrier to realize a quantum computer in the real world. Nuclear spin is an important candidate for quantum computing because of its long coherence time [7–10]. Naturally, we may try to manipulate nuclear spins by magnetic field. However, this method such as nuclear magnetic resonance is technically difficult to modulate the magnetic field exactly in micro areas and therefore it only manipulates ensemble states rather than a single quantum state, though it can have a long coherence time. This makes it difficult to design a scalable quantum computer. Electric field (EF) can be tuned exactly in micro areas by existing matured technology, and some electrical nuclear manipulation proposals [11–13] based on hyperfine stark effect, ENMHSE are presented. But hyperfine stark effect affects coherence time since it is the mutual coupling between nuclear spin and electron angular momentum. Remarkably, it has been experimentally demonstrated [14] that EF can couple with single nucleus based on electric quadrupole effect (EQE) [15, 16]. EQE holds a long coherence time since nuclear spin is unilaterally driven by electric field gradient (EFG) produced by electrons. This EFG is an ‘external’ field for nucleus. However, the existing study [14, 16] of EQE is essentially based on the model of static electric field, while there is no study on manipulating nuclear states. Here we present a general theoretical model for nuclear EQE with time-dependent electromagnetic field (EMF). With the model we systematically shows how to realize nuclear electric resonance (NER) which can precisely manipulate a single nucleus by oscillating EF. Universal quantum gates are achieved by NER with nuclear interaction control. Our NER quantum computing proposal has huge advantages in application: it can be applied to arbitrary $S \geq 1$ spin nuclei and has a remarkably excellent performance on single-qubit operation since its coherence time is much longer than ENMHSE’s.

II. NUCLEAR ELECTRIC QUADRUPOLE EFFECT IN ELECTROMAGNETIC FIELD

Consider a nucleus at the origin forced by vacuum EMF. We take scalar potential $\phi(r, t) = 0$ [17], and vector potential $A_\alpha(r, t)(\alpha = x, y, z)$ to first order

$$
A_\alpha(r, t) = A_\alpha(0, t) + \partial_\beta A_\beta(0, t)r_\beta, \quad (1)
$$

with the Einstein summation convention, i.e., when an index variable appears twice in a single term it implies summation over $\{x, y, z\}$. Consider a scalar function

$$
G(r, t) \equiv A_\alpha(0, t)r_\alpha + \frac{1}{2}\partial_\alpha A_\beta(0, t)r_\alpha r_\beta. \quad (2)
$$

Taking the gauge transformation

$$
A'_\alpha(r, t) = A_\alpha(r, t) - \partial_\alpha G(r, t),
\phi'(r, t) = \phi(r, t) + \partial_\alpha G(r, t), \quad (3)
$$

and meanwhile noticing that

$$
\partial_\alpha A_\beta(0, t) - \partial_\beta A_\alpha(0, t) = \varepsilon_{\alpha\beta\gamma} B_\gamma(0, t),
-\partial_\alpha A_\alpha(0, t) = -\partial_\alpha A_\alpha(0, t) - \partial_\alpha \phi(0, t) = E_\alpha(0, t), \quad (4)
$$

where $\varepsilon_{\alpha\beta\gamma}$ is the Levi-Civita symbol, we obtain

$$
A'_\alpha(r, t) = -\frac{1}{2}\varepsilon_{\alpha\beta\gamma} r_\beta B_\gamma(0, t),
\phi'(r, t) = -E_\alpha(0, t)r_\alpha - \frac{1}{2}\partial_\alpha E_\beta(0, t)r_\alpha r_\beta. \quad (5)
$$
With this we can write down the Hamiltonian for our system, a nucleus under the time-dependent EMF

\[ \mathcal{H} = \sum_z \left[ \frac{1}{2m(z)} (p^{(z)}_\alpha - eA'_\alpha(r^{(z)}, t))(p^{(z)}_\alpha - eA'_\alpha(r^{(z)}, t)) + e\phi'(r^{(z)}, t) + V(r^{(z)}) + \gamma^{(z)} s^{(z)} B_\alpha(0, t) \right] + \sum_n \left[ \frac{1}{2m(n)} p^{(n)}_\alpha p^{(n)}_\alpha + V(r^{(n)}) + \gamma^{(n)} s^{(n)} B_\alpha(0, t) \right], \]  

(6)

Consider the terms in right hand side of Eq. (7) one by one. The first summation is simply the interaction between nuclear magnetic moment and magnetic field, hence equivalent to \( \gamma_n s_\beta B_\beta(0, t) \) where \( \gamma_n \) is the nuclear gyromagnetic ratio and \( S \) is the nuclear spin. Compared with this, the second summation can be ignored since it is a second order small quantity. By the Wigner-Eckart theorem the third summation is omitted since it is a zero-order spherical harmonic tensor operator and equivalent to a constant. The forth summation is also negligible because nucleus is so stable that we only concern about the protons move around the origin. Applying the Wigner-Eckart theorem and the Gauss’s law \( \partial_\alpha E_\alpha(0, t) = 0 \) we can rewrite the fifth summation into the following equivalent form after some tedious calculations:

\[ \mathcal{H}_Q \equiv -\frac{1}{2} \bar{Q} \partial_\alpha E_\beta(0, t)(S_\alpha S_\beta + S_\beta S_\alpha), \]  

(8)

where \( \bar{Q} = \frac{eQ}{2n(2S^2 - 1)n^2} \) and \( Q \equiv Z(SS|3z^2 - r^2|SS) \) is the nuclear electric quadrupole momentum. Eq. (8) is the general form of EQE in EMF. Given the discussion above, Eq. (7) can be written more neatly as

\[ \mathcal{H} = \gamma_n S_\alpha B_\alpha(0, t) + \mathcal{H}_Q. \]  

(9)

This is our major equation for Hamiltonian. To confirm the correctness of Eq. (9), we suppose magnetic field at the origin is independent with time and all components of electric field have the same frequency and phase, it turns to the Hamiltonian in Ref. [14]. If we only consider the time-independent electric field, Eq. (9) will be the same as EQE in a static electric field presented in Ref. [10].

where \( V(r) = -\frac{V_0}{1 + \exp(\frac{r}{a})} \) is the Woods-Saxon potential that describes the nuclear forces applied on each nucleon, index \((z)\) and \((n)\) represent protons and neutrons respectively, \( Z \) and \( N \) are the number of protons and neutrons respectively, \( m \) is the mass, \( e \) is the elementary charge, \( \gamma \) is the gyromagnetic ratio, \( s \) is the spin, \( R_0 \) is the nuclear radius, \( V_0 \) and \( a \) are constants. Applying Eq. (5) into Eq. (6) we obtain

\[ \mathcal{H} = \sum_{z,n} \left[ -\frac{e}{2m(z)} z_\alpha \gamma_\gamma p_\gamma + \gamma^{(z)} s^{(z)} + \gamma^{(n)} s^{(n)} \right] B_\alpha(0, t) + \sum_{z} \frac{e^2}{8m(z)} \left[ s^{(z)} r^{(z)} B_\beta(0, t) B_\beta(0, t) - r^{(z)} r^{(z)} B_\alpha(0, t) B_\beta(0, t) \right] + \sum_{z} \left[ -eE_\alpha(0, t) r^{(z)} \right] - \sum_{z} \left[ \frac{e}{2} \partial_\alpha E_\beta(0, t) r^{(z)} r^{(z)} \right]. \]  

(7)

III. NUCLEAR ELECTRIC RESONANCE

Nuclear electric quadrupole momentum \( Q \) is extremely small, about \( 10^{-26} \sim 10^{-28} \text{m}^2 \). Straightly using Eq. (9), we need a huge EFG which is impossible for the existing technology. To solve this problem, we use external oscillating EF to manipulate atomic electron states and they produce a sufficiently strong EFG on the nucleus. We can precisely manipulate nuclear state by controlling oscillating EF, called nuclear electric resonance.

The Hamiltonian for an electron in an atom with static magnetic field \((0, 0, B_0)\) is

\[ \mathcal{H}_0 = \frac{\mathbf{p}_e^2}{2m_e} - \frac{kZe^2}{r} + \gamma_e B_0 I_z, \]  

(10)

where \( \mathbf{p}_e \) is the momentum, \( m_e \) is the mass, \( Z \) is the atomic number, \( k \) is the Coulomb’s constant, \( e \) is the elementary charge, \( \gamma_e \) is the gyromagnetic ratio, and \( I_z \) is the orbital angular momentum. We do not consider electron spin since it does not couple with EF. The eigenvalue of this Hamiltonian is \( E_{nm} = -\frac{Z^2 m_e e^4}{2kZe^2} + \gamma_e B_0 n \hbar, \) and related eigenstate is \(|nlm\rangle\). Here \( n, l, m \) are principal, azimuthal and magnetic quantum number. Applying the external oscillating EF \((E_e(t), E_n(t), E_z(t))\) with angular frequency \( \omega \), we get the perturbation Hamiltonian

\[ \mathcal{H}_1 = \varepsilon E_\alpha(t) r_\alpha. \]  

(11)

The time evolution by the Hamiltonion \( \mathcal{H}_0 + \mathcal{H}_1 \) is

\[ \langle \psi(t) \rangle = \exp \left\{ -\frac{i}{\hbar} \int_0^t (\mathcal{H}_0 + \mathcal{H}_1) \, dt \right\} \langle \psi(0) \rangle. \]  

The electron on the \( E_{nm} \) energy level has \( n - |m| \) degrees of degeneracy. Given initial state \(|\psi(0)\rangle = \sum_{l=-|m|}^{|m|} \xi_l |nlm\rangle\), we
have:

\[ |ψ(t)⟩ = \exp \left\{ -\frac{i}{\hbar}E_{nm}t \right\} \exp \left\{ \frac{i}{\hbarω}\tilde{E}_α(t)r_α \right\} \sum_l c_l |nlm⟩ \]

\[ \approx \exp\left\{ -\frac{i}{\hbar E_{nm}}t \right\} \sum_l c_l (1 + \frac{i}{\hbarω}\tilde{E}_α(t)r_α) |nlm⟩ \] \tag{12}

where \( \tilde{E}_α(t) = -ω \int_0^t E_α(t)dt \). After some simple calculations, we get EFG at the origin (the nuclear position) generated by the electron of state \( |ψ(t)⟩ \):

\[ \partial_α E_α = ke ⟨\psi(t)⟩ \frac{3r_α r_β}{r^5} - \frac{δ_α δ_β}{r^3} |ψ(t)⟩ \] \tag{13}

Using \( r \cos θ, r \sin θ \cos φ, r \sin θ \sin φ \) coordinate and the recurrence formulas of associated Legendre polynomials we get each component of EFG to the first order after some tedious calculations:

\[ \partial_x E_x = \partial_y E_y = C(n, m) - B(n, m) \tilde{E}_z(t), \]

\[ \partial_z E_z = -2C(n, m) + 2B(n, m) \tilde{E}_z(t), \]

\[ \partial_x E_y = \partial_y E_x = 0, \]

\[ \partial_x E_z = \partial_y E_z = -3A(n, m) \tilde{E}_x(t), \]

\[ \partial_y E_z = \partial_x E_y = -3A(n, m) \tilde{E}_y(t), \] \tag{14}

where

\[ A(n, m) = \frac{ke^2}{\hbar ω} \sum_{l't} \langle nl'm | \cos θ - \cos^3 θ/r^2 |nlm⟩ \Im(\epsilon_l^∗ \epsilon_l), \]

\[ B(n, m) = \frac{ke^2}{\hbar ω} \sum_{l't} \langle nl'm | \cos θ - 3 \cos^3 θ/r^2 |nlm⟩ \Im(\epsilon_l^∗ \epsilon_l), \]

\[ C(n, m) = \frac{3ke^2}{2} \sum_{l't} \langle nl'm | 1 - 3 \cos^2 θ/r^3 |nlm⟩ \epsilon_l^∗ \epsilon_l. \] \tag{15}

This is the EFG generated by an electron of state \( |ψ(t)⟩ \). The total EFG is simply the summation for all electrons in the atom:

\[ \partial_x E_x = \partial_y E_y = C - B \tilde{E}_z(t), \]

\[ \partial_z E_z = -2C + 2B \tilde{E}_z(t), \]

\[ \partial_x E_y = \partial_y E_x = 0, \]

\[ \partial_x E_z = \partial_y E_z = -3A \tilde{E}_x(t), \]

\[ \partial_y E_z = \partial_x E_y = -3A \tilde{E}_y(t), \] \tag{16}

where

\[ (A, B, C) = \sum_{\text{all electrons}} (A(n, m), B(n, m), C(n, m)). \] \tag{17}

With this total EFG, Eq. (9) leads to the Hamiltonian for nucleus:

\[ H_n = \gamma_n B_0 S_z + \tilde{Q}(C - B \tilde{E}_z(t))(-S_x^2 - S_y^2 + 2S_z^2) + 3QA[(S_x S_z + S_z S_x) \tilde{E}_x(t) + (S_y S_z + S_z S_y) \tilde{E}_y(t)] \]

\[ = \gamma_n B_0 S_z + 3\tilde{Q}(C - B \tilde{E}_z(t))S_z^2 + 3\tilde{Q}A \times \]

\[ [(S_x S_z + S_z S_x) \tilde{E}_x(t) + (S_y S_z + S_z S_y) \tilde{E}_y(t)]. \] \tag{18}

In order to manipulate nuclear state, we take the external EF to be the following form:

\[ (E_x(t), E_y(t), E_z(t)) = E(\sin(ωt + φ), -\cos(ωt + φ), 0), \] \tag{19}

Then Eq. (13) becomes to

\[ H_{n, \text{ER}} = \gamma_n B_0 S_z + 3\tilde{Q}CS_z^2 + 3\tilde{Q}AE \times \]

\[ [(S_x S_z + S_z S_x) \cos(ωt + φ) + (S_y S_z + S_z S_y) \sin(ωt + φ)], \] \tag{20}

where the time-independent small quantities have been ignored. This is our major Hamiltonian for NER. Using the rotating frame \( Ψ_{n, \text{ER}}(t) = e^{-iS_z(ωt + φ)}Ψ_{n, \text{ER}}(t) \) in the Schrödinger equation \( i\hbar \partial_\tau Ψ_{n, \text{ER}}(t) = H_{n, \text{ER}}Ψ_{n, \text{ER}}(t) \) we obtain

\[ Ψ_{n, \text{ER}}(t) = \exp \left\{ -\frac{i}{\hbar}3\tilde{Q}CS_z^2 + AE[(S_x S_z + S_z S_x) \cos φ + (S_y S_z + S_z S_y) \sin φ] \right\} Ψ_{n, \text{ER}}(0), \] \tag{21}

where resonance condition \( \gamma_n B_0 = \hbar ω \) and the identity \( I_x \cos(ωt + φ) + I_y \sin(ωt + φ) = e^{-iS_z(ωt + φ)}I_x e^{iS_z(ωt + φ)} \) have been used. With this we can precisely manipulate nuclear state.

Some notes about Eqs. (20) and (21):

(i) The term \( 3\tilde{Q}CS_z^2 \) is independent of our oscillating EF so it gives inhomogeneous energy level splittings all the time. This effect has been observed in experiment [14].

(ii) The factor \( 3AE \) is the amplitude of EFG in Eq. (19), which is much larger than the amplitude \( E \) of oscillating EF in Eq. (19). We give a rough evaluation \( A ≃ \frac{ke^2}{h \omega} \approx 8 \times 10^{10} \text{m}^{-1} \) where \( h_0 \) is the Bohr radius. This provides a sufficiently large Rabi frequency.

(iii) NER will divide the Hilbert space into two parts if \( S \) is a half-integer. Explicitly, the operator \( (S_x S_z + S_z S_x) \cos φ + (S_y S_z + S_z S_y) \sin φ \) appearing in Eq. (21) shows that the transitions can only appear between neighbor energy levels. But the matrix element of this operator will be zero if \( ms = -m' \) for \( ms \gg \frac{1}{2} \) so the subspaces of \( ms \leq -\frac{1}{2} \) and of \( ms \geq \frac{1}{2} \) are independent. This agrees with the experiment [14]. One more thing is that there does not exist NER for spin \( S = \frac{1}{2} \).

IV. NUCLEAR ENERGY SPLITTINGS WITH EXTERNAL STATIC ELECTRIC FIELD

Consider the following external static EF

\[ (E_x(t), E_y(t), E_z(t)) = (0, 0, E_0). \] \tag{22}

Now the perturbation Hamiltonian for an atomic electron changes to

\[ H_1 = ezE_0. \] \tag{23}

The main Hamiltonian is still given by Eq. (10). The degenerate subspace \( D \) of the \( E_{nm} \) energy level has \( n - |m| \) degrees of degeneracy. The \( k \)th energy and state in the
space $\mathcal{D}$ are
\[
E_{nmk} = E^{(0)} + E_k^{(1)},
\]
\[
|\psi_{nmk}^{(0)}\rangle = |\psi_k^{(0)}\rangle + |\psi_k^{(1)}\rangle,
\]
where $E^{(0)} = E_{nm}$, $E_k^{(1)}$ and $|\psi_k^{(0)}\rangle$ are the $k$th eigenvalue and eigenstate for the projection matrix of $H^0$ into space $\mathcal{D}$, i.e., $H_{nm}^{(0)}|\psi_k^{(0)}\rangle = E_k^{(0)}|\psi_k^{(0)}\rangle$. $|\psi_k^{(1)}\rangle$ can be written as $|\psi_k^{(1)}\rangle = \sum c_l|nlm\rangle$ and then
\[
|\psi_k^{(1)}\rangle = \sum_{n'k' \neq k} \frac{|\psi_k^{(0)}\rangle \langle H_k | \psi_k^{(0)}\rangle}{E_{nm} - E_{n'm'}} c_{n'k'}\langle n'l'm|z|nlm\rangle.
\]
(24)

Taking the same procedure as Eqs. (13)–(17), we obtain the total EFG on the nucleus:
\[
\partial_x E_x = \partial_y E_y = C + B'E_0, \quad \partial_z E_z = -2(C + B'E_0),
\]
\[
\partial_x E_y = \partial_y E_x = \partial_z E_x = \partial_x E_y = \partial_y E_z = \partial_z E_y = 0
\]
(26)

where $C$ takes the same form as in Eqs. (15) and (17), and
\[
B' \equiv ke^2 \sum_{\text{all electrons}} \sum_{n',n,l,t',t''} \frac{(1 - 3 \cos^2 \theta)}{r^3} |n'l'm\rangle \langle n'l'm|z|nlm\rangle \frac{\langle l|QCS(2)}{E_{nm} - E_{n'm'}} \mathrm{Re}(c_{n'k}c_{l})
\]
(27)

Then the Hamiltonian for the nucleus in this atom is:
\[
H_{\mathcal{O}} = \gamma_n B_0 S_z + 3\tilde{Q}C S_x^2 + 3\tilde{Q}B'E_0 S_x^2.
\]
(28)

Remark:
(i) The term $3\tilde{Q}C S_x^2$ is independent of our static EF. It is a natural effect for a nucleus in an atom. This agrees with Eq. (20).

(ii) The phenomenon that static EFG induces inhomogeneous nuclear energy level splittings is called linear quadrupole Stark effect (LQSE) [10]. However, with the term $3\tilde{Q}B'E_0 S_x^2$ appearing in the above Hamiltonian we can control these nuclear energy level splittings by a simple EF instead of an unachievable EFG. A rough evaluation shows that $B' \approx ke^2 \sum_{(2m-1)n} \approx 10^{20} m^{-1}$ makes a remarkable energy gap.

These two terms are useful in quantum computing as shown in section V and VI.

V. SINGLE-QUBIT OPERATION FOR $S \geq 1$

Combining Eqs. (19) and (22) we obtain the external electric field:
\[
E_x(t) = E \sin(\omega t + \varphi), \quad E_y(t) = E_0, \quad E_y(t) = -E \cos(\omega t + \varphi).
\]
(29)

Given Eqs. (20) and (28) we have the following Hamiltonian for nucleus:
\[
H_{\mathcal{O}} = \gamma_n B_0 S_z + 3Q(C + B'E_0) S_x^2 + 3QAE \times \left[(S_z S_x + S_x S_z) \cos(\omega t + \varphi) + (S_y S_z + S_z S_y) \sin(\omega t + \varphi)\right].
\]
(30)

The term $\gamma_n B_0 S_z + 3\tilde{Q}(C + B'E_0) S_x^2$ makes the energy level splitting between $m_S$ and $m_S + 1$ to be $\gamma_n B_0 h + 3\tilde{Q}(C + B'E_0)(2m_S + 1)h^2$. So we select two specific energy levels by applying specific frequency $\omega$. We choose the subspace $\mathcal{O}$ of $m_S = \{S, S - 1\}$ for our qubit. The projection matrices of $S_x, S_y, S_z$ into this subspace are:
\[
S_x^{(\mathcal{O})} = \sqrt{2} S_x^{(z)}, \quad S_y^{(\mathcal{O})} = \sqrt{2} S_y^{(z)},
\]
\[
S_z^{(\mathcal{O})} = S_z^{(z)} + \frac{h}{2}(2S - 1),
\]
(31)

where $S_x^{(z)}, S_y^{(z)}, S_z^{(z)}$ are the spin $\frac{1}{2}$ matrices. In the $\mathcal{O}$ space Eq. (33) becomes to
\[
H_{\mathcal{O}}^{(\mathcal{O})} = \gamma_n B_0 S_z^{(\mathcal{O})} + 3\tilde{Q}(C + B'E_0) S_x^{(\mathcal{O})} + 3QAE \times \left[(S_z^{(\mathcal{O})} S_x^{(\mathcal{O})} + S_x^{(\mathcal{O})} S_z^{(\mathcal{O})}) \cos(\omega t + \varphi) + (S_y^{(\mathcal{O})} S_z^{(\mathcal{O})} + S_z^{(\mathcal{O})} S_y^{(\mathcal{O})}) \sin(\omega t + \varphi)\right]
\]
(32)

Using the rotating frame $\Psi_{\mathcal{O}}^{(\mathcal{O})}(t) = e^{-iS^{(\mathcal{O})}S_x^{(\mathcal{O})}t} \Psi_{\mathcal{O}}^{(\mathcal{O})}(t)$ in the Schrödinger equation $\hbar \partial_t \Psi_{\mathcal{O}}^{(\mathcal{O})}(t) = H_{\mathcal{O}}^{(\mathcal{O})} \Psi_{\mathcal{O}}^{(\mathcal{O})}(t)$ we obtain
\[
\Psi_{\mathcal{O}}^{(\mathcal{O})}(t) = \exp\left\{ -\frac{i}{\hbar} 3\sqrt{2}\sin(2S - 1) \hbar QAE \times \left[S_x^{(z)} \cos \varphi + S_y^{(z)} \sin \varphi\right]\right\} \Psi_{\mathcal{O}}^{(\mathcal{O})}(0)
\]
(33)
in the subspace $\mathcal{O}$. Here resonance condition $\gamma_n B_0 + 3(2S - 1)\hbar Q(C + B'E_0) = \hbar \omega$ has been used. With Eq. (33), any single-qubit gate can be achieved for nuclear spin $S \geq 1$.

There is a more remarkable result for $S = \frac{3}{2}$. We apply appropriate frequency EF to select the subspace $\mathcal{O} : \{m_S = \frac{3}{2}, \frac{1}{2}\}$ and neglect the other subspace $\mathcal{O}' : \{m_S = -\frac{3}{2}, -\frac{1}{2}\}$. But as we mentioned earlier in section III, NER naturally divides the Hilbert space of $S = \frac{3}{2}$ into two unconnected subspaces. So subspace $\mathcal{O}$ will be protected most robustly if $S = \frac{3}{2}$.

VI. TWO-QUBIT OPERATION FOR $S \geq 1$

Now we show two-qubit operations in $\mathcal{O}_1 \otimes \mathcal{O}_2$ of nucleus 1 and nucleus 2, which is not disturbed by other subspaces. This proposal can be realized in solid-state
We apply the EF \((0, 0, E_1)\) and \((0, 0, E_2)\) to two nuclei separately. Using Eq. (32) we obtain the Hamiltonian for two nuclei with interaction:

\[
\mathcal{H}_{\text{two}} = \gamma_{n_1} B_0 S_{1z} + \gamma_{n_2} B_0 S_{2z} + 3\hat{Q}_1 C_1 S_{1z}^2 + 3\hat{Q}_2 C_2 S_{2z}^2 + 3\hat{Q}_1 B_1 E_1 S_{1z}^2 + 3\hat{Q}_2 B_2 E_2 S_{2z}^2 + \mathcal{H}_1, \quad (34)
\]

where index 1 and 2 represent different nuclei, \(\mathcal{H}_1\) is the J-coupling interaction. We use two different kinds of nuclei or apply much different \(E_1\) and \(E_2\) to same kinds of nuclei. This means that the difference between two nuclear energy level splittings is much larger than J-coupling interaction and hence we can take the approximation \(\mathcal{H}_1 \approx 2\pi J(t)S_1S_2\). Notably, \(\mathcal{H}_1\) decouples the subspace \(O_1 \otimes O_2\) from other subspaces since it is a diagonal matrix. Similar to Eq. (32), we consider Eq. (34) in \(O_1 \otimes O_2\):

\[
\mathcal{H}_{\text{two}}^{(O_1 \otimes O_2)} = \gamma_{n_1} B_0 S_{1z} + 3(2S - 1)h\hat{Q}_1 C_1 \left(S_{1z}^{(1)} + S_{2z}^{(2)}\right) + \left[\gamma_{n_2} - \gamma_{n_1}\right] B_0 + 3(2S - 1)h(\hat{Q}_2 C_2 - \hat{Q}_1 C_1)S_{2z}^{(2)} + (2S - 1)h(3\hat{Q}_1 B_1 E_1 + \pi J(t))S_{1z}^{(1)} + (2S - 1)h(3\hat{Q}_2 B_2 E_2 + \pi J(t))S_{2z}^{(2)} + 2\pi J(t)S_{1z}^{(1)}S_{2z}^{(2)}, \quad (35)
\]

where Eq. (34) has been used. Using the rotating frame \(\Psi_{\text{two}}^{(O_1 \otimes O_2)}(t) = e^{-\frac{i}{\hbar} \left(S_{1z}^{(1)} + S_{2z}^{(2)}\right)(\omega)\left(t\right)} \Psi_{\text{two}}^{(O_1 \otimes O_2)}(t)\) in the Schrödinger equation \(i\hbar \partial_t \Psi_{\text{two}}^{(O_1 \otimes O_2)}(t) = \mathcal{H}_{\text{two}}^{(O_1 \otimes O_2)}(t)\) we obtain

\[
\Psi_{\text{two}}^{(O_1 \otimes O_2)}(t) = U_1(t)U_2(t)U_{1,2}(t)\Psi_{\text{two}}^{(O_1 \otimes O_2)}(0), \quad (36)
\]

where

\[
U_1(t) = \exp \left\{ -\frac{i}{\hbar} S_{1z}^{(1)} \int_0^t (2S - 1)h(3\hat{Q}_1 B_1 E_1 + \pi J(t))dt \right\}
\]

is the operation for the first qubit,

\[
U_2(t) = \exp \left\{ -\frac{i}{\hbar} S_{1z}^{(1)} \int_0^t (\gamma_{n_2} - \gamma_{n_1}) B_0 + (2S - 1)h \times \right. \\
\left. \left[3\hat{Q}_2 B_2 E_2 + \pi J(t) + 3(\hat{Q}_2 C_2 - \hat{Q}_1 C_1)\right]dt \right\}
\]

is the operation for the second qubit, and

\[
U_{1,2}(t) = \exp \left\{ -\frac{i}{\hbar} S_{1z}^{(1)} S_{2z}^{(2)} \int_0^t 2\pi J(t)dt \right\}
\]

is the two-qubit operation. Here resonance condition \(\gamma_{n_1} B_0 + (2S - 1)h\hat{Q}_1 E_0 = h\omega\) has been used. By tuning \(E_1, E_2, J(t)\) we can get a controlled-NOT gate or a controlled-Z gate \([7, 18]\).

### VII. PERFORMANCE EVALUATION

In quantum computing, all operations should be completed within the coherence time \(T_2^*\). We can calculate the number of flips \(N_t = T_2^* \times f_R\) of our NER proposal, where \(f_R\) is the Rabi frequency. It is the number of Rabi oscillations during a coherence time \(T_2^*\). After this calculation, we compare NER with ENMHSE as shown below.

From the study on Sb nucleus in Ref. [14], we get the following data: \(f_R = 684.2\text{Hz}, T_2^* = 92\text{ms}\) under the condition \(V_{\text{RF}} = 20\text{mV}\) which is the voltage of radio-frequency (RF) oscillating EF. According to Eq. (33) we obtain \(f_R = k_R E\) where \(k_R = \frac{3eAQ}{2\pi\sqrt{27h}}\). To our knowledge the voltage of oscillating EF can easily reach \(V_{\text{RF}} = 4\text{V}\) \([13]\), and the amplitude of EF is changed to \(E' = \frac{V_{\text{RF}}}{V_{\text{RF}}} E = 200\text{E}\). The Rabi frequency is improved to \(f'_R = k_R E' = 200k_R E = 200f_R = 136840\text{Hz}\). Finally we calculate \(N_t = T_2^* \times f'_R = 12589.28\) for our NER proposal. Comparing this result with ENMHSE as shown in Table II we find an improvement by three orders of magnitude on \(N_t\).

### VIII. CONCLUSION

In summary, we present a new nuclear manipulation proposal called NER, and apply it in constructing universal quantum gates on arbitrary \(S \geq 1\) spin nuclei. Since NER can manipulate single nucleus and meanwhile hold a long coherence time, it is quiet useful for quantum information processing in the future.

We acknowledge the financial support in part by Ministry of Science and Technology of China through The National Key Research and Development Program of China grant No. 2017YFA0303901; National Natural Science Foundation of China grant No.11774198 and No.11974204.

| Method | Coherence time \(T_2^*/\text{ms}\) | Rabi frequency \(f_R/\text{kHz}\) | Number of flips \(N_t\) |
|--------|-----------------|-----------------|-----------------|
| ENMHSE | \(\sim 0.064\)  | \(\sim 180.8\)  | \(\sim 11.57\)  |
| ENMHSE | \(\sim 0.97\)   | \(\sim 5\)      | \(\sim 4.85\)   |
| NER    | \(\sim 92\)    | \(\sim 136.8\)  | \(\sim 12589.28\) |

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[1] D. P. DiVincenzo, Science 270, 255 (1995).
[2] A. Ekert and R. Jozsa, Reviews of Modern Physics 68, 733 (1996).
[3] J. Preskill, Quantum 2, 79 (2018).
[4] F. Arute, K. Arya, R. Babbush, D. Bacon, J. C. Bardin, R. Barends, R. Biswas, S. Boixo, F. G. Brandao, D. A. Buell, et al., Nature 574, 505 (2019).
[5] Y. Wu, W.-S. Bao, S. Cao, F. Chen, M.-C. Chen, X. Chen, T.-H. Chung, H. Deng, Y. Du, D. Fan, et al., Physical Review Letters 127, 180501 (2021).
[6] H.-S. Zhong, Y.-H. Deng, J. Qin, H. Wang, M.-C. Chen, L.-C. Peng, Y.-H. Luo, D. Wu, S.-Q. Gong, H. Su, et al., Physical Review Letters 127, 180502 (2021).
[7] M. A. Nielsen and I. Chuang, Quantum computation and quantum information (American Association of Physics Teachers, 2002).
[8] J. W. Park, Z. Z. Yan, H. Loh, S. A. Will, and M. W. Zwierlein, Science 357, 372 (2017).
[9] G. Sharma, T. Gaebel, E. Rej, D. J. Reilly, S. E. Economou, and E. Barnes, Physical Review B 99, 205423 (2019).
[10] A. Serafin, M. Fadel, P. Treutlein, and A. Sinatra, Physical Review Letters 127, 013601 (2021).
[11] S. Thiele, F. Balestro, R. Ballou, S. Klyatskaya, M. Ruben, and W. Wernsdorfer, Science 344, 1135 (2014).
[12] A. Laught, J. T. Muhonen, F. A. Mohiyaddin, R. Kalra, J. P. Dehollain, S. Freer, F. E. Hudson, M. Veldhorst, R. Rahman, G. Klimeck, et al., Science Advances 1, e1500022 (2015).
[13] A. J. Sigillito, A. M. Tyryshkin, T. Schenkel, A. A. Houck, and S. A. Lyon, Nature Nanotechnology 12, 958 (2017).
[14] S. Asaad, V. Mourik, B. Joecker, M. A. Johnson, A. D. Baczevski, H. R. Firgau, M. T. Mądzik, V. Schmitt, J. J. Pla, F. E. Hudson, et al., Nature 579, 205 (2020).
[15] N. Bloembergen, Science 133, 1363 (1961).
[16] C. P. Slichter, Principles of magnetic resonance, Vol. 1 (Springer Science & Business Media, 2013).
[17] M. O. Scully and M. S. Zubairy, Quantum optics (American Association of Physics Teachers, 1999).
[18] D. Loss and D. P. DiVincenzo, Physical Review A 57, 120 (1998).