Quantum Phase Control by Electric Field in a Rare-Earth Material

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Manipulating quantum properties by electric fields promises individual spin addressability and energy efficiency compared to magnetic control. The electric field couples directly to the electron charge and can thereby control the spin dynamics via spin-orbit coupling. However, most spin carriers in electric field control research are light elements, for which the spin-orbit coupling is weak. To enhance electric-spin coupling, one option is to improve the spin-orbit coupling of the qubits. Here, we employ a rare-earth qubit to show that the electric field can work as a highly efficient quantum phase gate. The electric-spin interaction can be decoupled by microwave pulses to realize quantum bang-bang control and the quantum Zeno effect. This quantum phase gate is also utilized to demonstrate the refined Deutsch-Jozsa algorithm in the rare-earth qubit.

Rare-earth elements are increasingly important in technology, and they are being used in widespread applications¹, such as luminescent materials², magnetic materials³ and medical contrast agents⁴. The magnetic properties of rare-earth ions are largely dominated by the presence of strong spin-orbit coupling⁵,⁶. This advantage offers a pass to control the spin coherently with high efficiency and spatial selectivity, which is the central technique in quantum information processing. A general approach to prepare a cat state and manipulate its quantum phase is accomplished by magnetic resonance in which trains of resonant oscillating magnetic field pulses are applied⁷. Nevertheless, down to the single spin level⁸, this turns out to be inconvenient due to its bad localization and addressability, and moreover, the fabrication of cavities is also challenging. By contrast, individual spin resonance driven by an electric (E) field could be much more practical based on the present development of the electronic industry⁹.

In terms of magnetic exchange, the magnetolectric effect does not necessarily rely on spin-orbit coupling¹⁰. However, for individual spin carriers, the E field couples directly to the electron charge and therefore controls the spin dynamics via spin-orbit coupling¹¹. The unquenched orbital momentum and the large spin-orbit coupling can easily enhance the efficiency of controlling the spin by an E field. This pushes the rare-earth elements to come into the stage. The inner shell 4f orbital degeneracy is weakly removed by the crystal field resulting in a large residual orbital momentum. The relativistic
effect of rare-earth elements, which are heavy, is significant, affording strong spin-orbit couplings. The electric-spin coupling is responsible for the Stark effect, and the related magnetic resonance researches were first published in the 1960s\textsuperscript{12,13} and later reviewed by Mims\textsuperscript{14}. The emergence of the Stark effect in magnetic resonance requires a lack of inversion symmetry in the spin carrier. The electric control of spin has been proposed to realize electric-dipole-induced spin resonance in semiconductors\textsuperscript{9}, and numerous experiments have been performed to control magnetic quantum dots\textsuperscript{15}, single electron spins\textsuperscript{16}, diamond defects\textsuperscript{17} and the single nuclear spin in a molecular magnet\textsuperscript{18}. The coherent control of an electron spin ensemble has been demonstrated in pniitogen-doped silicane semiconductors\textsuperscript{19–21}, piezo-diluted magnetic semiconductors\textsuperscript{22} and molecular magnets\textsuperscript{23}.

Herein, we demonstrate the advantage of a rare-earth ion in the quantum manipulation by an $E$ field based on its strong electric-spin coupling. The $E$ field plays the role of a phase gate of high efficiency in a two-level system. Quantum bang-bang decoupling is thereby realized in the electric phase gate by the microwave (mw). Furthermore, the refined Deutsch-Jozsa (DJ) algorithm is also demonstrated using this electric phase gate.

![Figure 1](image.png)

**Figure 1** The Ce:YAG single crystal structure and the device used in our experiments. **a**, The location of the six magnetically inequival-ve sites of Ce\textsuperscript{3+} marked as Ce-1 to Ce-6. Each Ce\textsuperscript{3+} is coordinated by 8 oxygen nuclei. The six groups of their principle axes are shown as coloured arrows (red, green and blue for $x$, $y$ and $z$, respectively). Inversion centres located at (0.5, 0.5, 0.5) and (0.5±0.25, 0.5±0.25, 0.5±0.25). The inversion pairs of Ce-1 to Ce-6 generated by (0.5, 0.5, 0.5) are also shown. **b**, Ce:YAG single crystal of size 2×6×0.5 mm\textsuperscript{3} is mounted on the electrodes, and a 50-$\Omega$ impedance is added. The top surface of the Ce:YAG single crystal is connected to the electrode through 0.2 mm gold wires.

**Determination of electric-spin coupling parameters**

An yttrium aluminium garnet (YAG) single crystal doped with Ce\textsuperscript{3+} is used to probe the Stark effect. YAG crystalizes in a cubic unit cell of $Ia\overline{3}d$. Various metallic cations including rare-earth ions can be doped into the YAG crystal by occupying the $Y^{3+}$ position\textsuperscript{24}. In Ce:YAG, there are 6 magnetically inequivalent sites\textsuperscript{25}. The local environment of all the Ce\textsuperscript{3+} is identically coordinated by 8 oxygens with a local symmetry of $D_2$. The three $C_2$ axes form the principal axes. The spin carriers appear as inversion pairs due to the centre symmetry of the crystal (Fig. 1a).

In the present research, only the ground doublets transition is observed; the hyperfine interaction of Ce\textsuperscript{3+} with adjacent nuclear spins is not observed. We simplify the Ce\textsuperscript{3+} ion as an effective $S = 1/2$ with a highly anisotropic $g$-tensor. Therefore, the Hamiltonian of the Ce\textsuperscript{3+} ion in the external static magnetic field is $\hat{H} = \mu_B \vec{B} \cdot \vec{g} \hat{S}$. Based on previous studies and our measurements, the principal values of the $g$ tensor are $g_{xx}=1.85$, $g_{yy}=0.90$ and $g_{zz}=2.74$\textsuperscript{25}.
Figure 2 Spin echo oscillation upon the application of an $E$ field. a, The pulse sequence employed in the experiment. The rectangular $E$ field pulse is applied between the mw $\pi/2$ and $\pi$ pulses of the standard Hahn echo pulse sequence. b, The transient spin echo oscillates with increasing frequencies upon enhancing the applied $E$ field strength from 0 to $10^6$ V/m, indicating that the relative quantum phase evolution is coherently controlled by the $E$ field. c, The quantum phase evolution frequency is obtained by the fast Fourier transformation (FFT) of the integrated echo intensity. It demonstrates a linear response to the $E$ field strength. d, The quantum phase evolution frequency is simulated by varying the direction of the $E$ and $B_0$ fields. The most efficient frequency for quantum phase control is optimized to be 0.96 MHz when $E \parallel z$ and $B_0 \parallel [0.44, 0.90, 0]$ with respected to the local Ce$^{3+}$ coordinate.

The Stark effect can be observed from the spin echoes as a function of the time for which the $E$ field is applied (Fig. 2a). The spin echo signal oscillates during this time (Fig. 2b). The oscillation frequency is proportional to the strength of the $E$ field from 0 to $10^6$ V/m, demonstrating a linear behaviour (Fig. 2c). In the $E$ field effect experiment, the inversion pair
cancels out the signal in the imaginary channel. Moreover, the spin echo oscillation decays with respect to the time of the applied $E$ field. We attribute this damping to the $E$ field inhomogeneity, which could be well simulated (SI).

The $E$ field Hamiltonian is generally expressed as $\hat{H}_E = \sum_{ijk} E_i T_{ijk} \mu_B B_i \hat{S}_k$, where $T_{ijk} = \frac{g_{jk}}{\partial E_i}$ and $g_{jk}$ is a symmetric matrix with $g_{jk} = g_{kj}$. The local symmetry $D_7$ of Ce$^{3+}$ largely reduces the non-vanishing elements of $T_{ijk}$ from 18 to 3. Therefore, the electric Hamiltonian is written as

$$\hat{H}_E = \mu_B \left( E_x T_{xyz} (B_x \hat{S}_x + B_y \hat{S}_y) + E_y T_{yxz} (B_y \hat{S}_y + B_z \hat{S}_z) + E_z T_{xyz} (B_z \hat{S}_z + B_x \hat{S}_x) \right),$$

(1)

where the $T_{xyz}$, $T_{yxz}$ and $T_{xyz}$ are the coupling parameters. The determination of these parameters is similar to the approach of Mims$^{14}$ (SI). Four of the six magnetic inequivalent Ce$^{3+}$ positions are selected to perform the experiments of the angular dependent Stark effect. The parameters are determined to be $T_{xyz} = 3.30 \times 10^{-8} \text{ m/V}$, $T_{yxz} = 8.76 \times 10^{-8} \text{ m/V}$ and $T_{xyz} = 12.13 \times 10^{-8} \text{ m/V}$.

**Electric phase gate**

When the $E$ field is applied between the two mw pulses, the spin echo gains a phase $\phi_E = \Delta \omega t$, where $\Delta \omega$ is the resonance frequency shift, and $t$ is the acting time of the $E$ field. This $\phi_E$ is also the relative quantum phase acquired by the superposition state. Therefore, the electric pulse is a controllable electric phase gate,

$$\hat{R}(\phi) = \begin{pmatrix} 1 & 0 \\ e^{i\phi} & 1 \end{pmatrix},$$

(2)

which offers the superposition state $|0\rangle + |1\rangle$ a relative phase $e^{i\phi}$, yielding $e^{i\phi} |0\rangle + e^{i\phi} |1\rangle$.

Next, we discuss the effect of $\hat{R}_E$ on $\Delta \omega$ term by term. The quantized $z$ axis is defined by the $B_0$ field direction, and therefore, in the absence of the $E$ field, the Hamiltonian can be written as $\hat{H}_0 = g_{\text{eff}} \mu_B B_0 \hat{S}_z = \omega_0 \hat{S}_z$. When the $E$ field acts on $\hat{S}_z$, the $E$ field Hamiltonian is $\hat{H}_{E1} = \sum E_i T_{ijk} \mu_B B_i \hat{S}_k = \omega_1 \hat{S}_z$, and the frequency shift $\Delta \omega$ is simply $\omega_1$.

However, if $\hat{R}_E$ does not commute with $\hat{R}_0$, i.e., $\hat{R}_{E2} = \sum E_i T_{ijk} \mu_B B_i \hat{S}_k = \omega_2 \hat{S}_{xy}$, the frequency shift is $\Delta \omega = \sqrt{\omega_1^2 + \omega_2^2} - \omega_0 \approx \omega_2^2/2\omega_0$ to the second order.

Based on the above discussion, it is easy to explain the observed linear Stark effect in the experiments. When $\hat{R}_0$ commutes with $\hat{R}_E$, the acquired phase is a linear function of the $E$ field. Conversely, when $\hat{R}_0$ and $\hat{R}_E$ are noncommutative, this phase is a quadratic term of the $E$ field. Because $\hat{R}_E$ is normally much smaller than $\hat{R}_0$, this quadratic term is too small to probe, and the linear effect dominates when $\hat{R}_{E1}$ and $\hat{R}_{E2}$ coexist. The bipolar $E$ field pulse between the mw pulses can eliminate the first order and accumulate the quadratic effect. This technique has been employed to investigate the Stark effect of pnictogen-doped-silicane semiconductors for which the first order of the $E$-field-induced hyperfine coupling shift cancels out.$^{19}$

The electric-spin coupling originates from the spin-orbit interaction. For the reported pnictogen-doped-silicane semiconductors$^{21}$ or piezo materials$^{22}$, the coupling strength of spin-obit interaction of the spin carriers is small. However, the applied $E$ field dramatically redistributes the electron wave function with respect to its original environment, and thus significantly changes the orbital momenta of the electrons. This results in a sizable effective electric-spin coupling. In the present research, we employ rare-earth ion which possesses a very large coupling strength of spin-obit interaction. Although the applied $E$ field slightly affects the orbital momenta of the electrons, the resulting effective electric-spin coupling is comparable with those of the reported materials. Therefore, rare-earth materials can equivalently work as an efficient phase gate.

With the determined $\hat{R}_E$ parameters, it is possible to optimize the most efficient direction for $E$ field control (Fig. 2d). The simulation provides that, with an $E$ field strength of $10^6 \text{ V/m}$, the spin precession frequency originating from the $E$ field is 0.96 MHz when the $B_0$ field is along (0.44, 0.90, 0) and the $E$ field is along (0, 0, 1) with respect to the local Ce$^{3+}$ ion coordinate, yielding an electric-spin coupling of 1.6 Hz/(T·V·m$^{-1}$). Unlike crystal-field and hyperfine-coupling terms, the energy shift originating from the Zeeman term shows a response to the external magnetic field because the magnetic
field is also proportional to the energy shift induced by the $E$ field, as seen in Eq.1. Therefore, the operation speed of electric phase gate can be enhanced by a $B_0$ field. Moreover, the strong magnetic field provides plenty of advantages, e.g., resolution enhancement, pure state for initialization, and long phase memory time. Conclusively, in the present research, we can safely work with a relatively weak $E$ field.

Quantum manipulation
The optimized manipulation frequency for the present experimental conditions reaches nearly 1 MHz and can finish a $\pi/2$ rotation within 260 ns, which is much shorter than the phase memory time of 15 us at 10 K. This offers us the possibility to realize several quantum manipulation demonstrations by the $E$ field. When the superposition state evolves in the $E$ field, the spin rotates in the $xy$ plane of the Bloch sphere. This rotation is a Rabi cycle between a pair of orthogonal bases in the $xy$ plane, e.g., $\frac{1}{\sqrt{2}}(|0\rangle \pm |1\rangle)$, with a pseudo driven field along the $z$ axis.

The Rabi cycle evolution can be decoupled from the $E$ field by the fast mw pulses. To ensure the final state after the mw interruption remains in the $xy$ plane, the mw $\pi$ pulses are chosen to kick the spin. The cat state $\frac{1}{\sqrt{2}}(|0\rangle + |1\rangle)$ evolves to be $\frac{1}{\sqrt{2}}(|0\rangle + e^{i\phi}|1\rangle)$ by a controllable phase gate $\hat{R}(\phi)$. The basis $|1\rangle$ picks up a phase $e^{i(\pi-2\phi)}$ with respect to $|0\rangle$ after the mw $\pi$ pulse in the $+y$ direction, whereas the other inversion pair acquires phases $e^{-i\phi}$ and $e^{i(\pi+2\phi)}$. Fig. 3a shows the mw $\pi$ pulse effect in this Rabi cycle. The $\pi$ pulses can be applied successively at arbitrary moments, and the polarization evolution is reversed after each $\pi$ pulse. As illustrated in Fig. 3b, the Rabi cycle evolves in the $xy$ plane, while a mw $\pi$ pulse drives the spin to process rapidly around the $B_1$ field direction via a small circle. This makes the spin evolve via a shortcut and continue its previous evolution in the $xy$ plane.

In this demonstration of bang-bang control, the mw pulses are much faster than the electric phase gate. In the original proposal of bang-bang control, the rapid controllable spin flip is employed to prevent the unwanted phase evolution. Herein, similar to the ultrafast phase gate in a fullerene qubit, we can realize this by the rapid mw pulses as well. As seen in Fig. 3c, when the system evolves in the $xy$ plane driven by the $E$ field and a train of mw $\pi$ pulses are continuously applied, one can see that the spin evolution is locked, and the spin is decoupled from the $E$ field. This is actually the quantum Zeno effect which suppresses the quantum evolution by repeated measurements or controllable interactions with the environment. Apparently, ceasing the mw pulses releases the spin evolution, which can be locked again. It is necessary to mention that, unlike the projective Zeno experiment, which locks the system in the eigenstate, this experiment can freeze and release any cat state, as shown in Fig. 3c.
**Figure 3** The $E$ field phase gate demonstration of quantum bang-bang control and the quantum Zeno effect realized by mw pulses. 

(a) The electric field phase gate driven the Rabi cycle within the $xy$ plane of the Bloch sphere. (II) to (V), Four trains of mw pulses to kick the spin echo. The arrows indicate the moment that the mw pulses are applied. The quantum phase evolution reverses when applying the microwave pulses. b, The Bloch sphere demonstration of bang-bang control. The bold purple curve represents the $E$-field-phase-gate-induced spin phase evolution, and the thin green curves indicate the mw “kicking” operation. The straight red arrow is the $B_\parallel$ field direction. c, Successive mw pulses can lock and release the phase evolution at arbitrary positions.

**DJ algorithm demonstration**

The DJ problem is to identify whether a given function, $f: \{0,1\}^n \rightarrow \{0,1\}$, which takes $n$-digit binary inputs and produces 0 or 1 as the output, is balanced (returns 0 for half of the inputs and 1 for the other half) or constant (returns 0 for all inputs or 1 for all inputs)\(^{31}\). A classic algorithm requires $2^{n-1} + 1$ evaluations for $n$-bit inputs in the worst case, while the DJ algorithm can determine the function types in a single try. To ensure the intrinsic reversibility, the conventional DJ algorithm requires an additional ancilla bit to store the output\(^{32}\), while Collins proposed a refined version of the DJ algorithm allowing the $n$-bit function to be evaluated in $n$ qubits\(^{33}\). Therefore, the refined DJ algorithm with $n = 1$ can be implemented in this two-level system via an electric phase gate.
The refined DJ algorithm requires two types of quantum gates. The Hadamard gate is to generate a uniform superposition of the two initial inputs and also to convert the output to a readable eigenstate in the end. Here, the Hadamard gate is achieved by a mw $\pi/2$ pulse (Fig. 4a). The $f$-controlled gates are designed to encode the oracle functions to be determined as balanced or constant. As to the $n = 1$ situation, there are only 4 possibilities, $f_1(x) = 0$, $f_2(x) = 1$, $f_3(x) = 1 - x$, $f_4(x) = x$, where the only input variable $x$ can be either 0 or 1, and the output is also 0 or 1. These 4 functions are encoded in the 4 $f$-controlled gates: $U_{f_1} = \begin{pmatrix} 1 & 0 \\ 0 & 1 \end{pmatrix}$, $U_{f_2} = \begin{pmatrix} -1 & 0 \\ 0 & -1 \end{pmatrix}$, $U_{f_3} = \begin{pmatrix} -1 & 0 \\ 0 & 1 \end{pmatrix}$ and $U_{f_4} = \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix}$, which are realized by the electric phase gates $R(\phi)$ as $U_{f_1} = R(0)$, $U_{f_2} = R(2\pi)$, $U_{f_3} = R(\pi)$ and $U_{f_4} = R(-\pi)$. It is worth noting that $R(\pi)$ and $R(-\pi)$ yield a unitary transformation of $U_{f_3}$ and $U_{f_4}$ with a global phase $i$, which do not influence the measurement in this 1-qubit system. The cat state evolves to be $\frac{1}{\sqrt{2}}[(-1)^{f_1(0)}|0\rangle + (-1)^{f_1(1)}|1\rangle]$ by $U_{f_1}$, is then be transformed by a second Hadamard gate to $|0\rangle$ (for constant functions, $f_1$ and $f_2$, Fig. 4b and 4c) or $|1\rangle$ (for balanced functions, $f_3$ and $f_4$, Fig. 4d and 4e) and is finally read out by the mw Hahn echo with a positive or negative phase.
Unlike the previously reported implementation of the DJ algorithm in diamond, for which the phase shift was achieved by an auxiliary state via a $2\pi$ rotation, in the present research, the phase shift gate is realized by the electric phase gate, and only a two-level system is necessary.

**Conclusion**

We have illustrated that electric field pulses applied to the Ce:YAG can be a highly efficient quantum phase gate. As an example, the spin carrier Ce$^{3+}$ processes only one 4f/electron, illustrating that the rare-earth elements can indeed enhance the electric-spin coupling via their strong spin-orbit interaction. The optimized manipulation condition at the X-band and $10^8$ V/m allows over one hundred $\pi/2$ phase gates within a phase memory time of more than 15 $\mu$s at 10 K.

Prior to prospecting the future of rare-earth based qubits, we would like to highlight the importance of symmetry in the research of electric-spin coupling. Clearly, the inversion symmetry on the spin carrier shuts down the $E$-field-induced energy level shifts. Nevertheless, a non-centrosymmetric paramagnetic centre is very common for rare-earth complexes. A low local symmetry can cause the complexity in the determination of the $E$-field-involved Hamiltonian. In the present case, the $D_2$ local symmetry requires only 3 terms to describe the $E$ field behaviour within the ground doublet; however, in the lowest symmetry case, to fully address the electric-spin coupling anisotropy, one would need the full 18 terms. Moreover, in the present Ce:YAG lattice, the inversion pairs also cancel out the orthogonal channel and partially shade the full picture of this phase gate. Therefore, for potential applications, it is necessary to dope the rare-earth spin carriers in high symmetric lattices without any inversion symmetry.

The quantum coherence time is, of course, one of the factors that will determine if rare-earth ions can be applied in quantum information processing devices. Rare-earth ions can possess ultralong optical coherence times in crystal, which are applied to memorize the quantum states of photons. The hyperfine interactions between rare-earth electrons and their nuclei generate atomic clock transitions, which can enhance the phase memory time up to one minute and thus fulfil the application requirements. Another advantage of rare-earth ions is directly related to their optical properties. The individual rare-earth ion qubit can be initialized and read out optically. This was first realized with Ce:YAG, the same material reported in this research. This capability of easy initialization and individual spin readout is of crucial importance for error correction and scalability. One could argue that the optimized 1 MHz frequency shift reported here is not strong enough. This is majorly due to the weak strength of the applied $E$ field. In the single spin break junction devices, the $E$ field can easily reach $10^8$ V/m with a few volts. Such a high $E$ field can individually control spins. The aforementioned advantages and the high efficiency of electric-spin coupling reported in this research, could lead to a growth of rare-earth-based quantum information processing research in the near future.

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**Author contributions**
S.D.J. and S.G. conceived the research. Z.L. and S.D.J. performed the experiments and theoretical analysis. Y.H.F. helped in experiments of electric-spin coupling parameters determination. S.X.Q. provided the methods to simulate the spin evolutions. Z.M.W. performed the single crystal face index. S.D.J. and Z.L. wrote the manuscript with contributions from all authors.

**Competing interests**

The authors declare no competing interests.

**Methods**

Ce\(^{3+}\) ions doped in yttrium aluminium garnet (YAG) single crystal is employed to probe the Stark effect. To eliminate the electron-spin dipolar interactions, the concentration of Ce\(^{3+}\) ions was reduced to be less than 0.1\%, where the average distance between Ce\(^{3+}\) ions is more than 3 nm. In order to enhance the \(E\) field strength and maintain reasonable signal, the thickness of the Ce:YAG single crystal was cut to 0.5 mm with a 2 \(\times\) 6 mm\(^2\) size to fit the sample into the cavity. The single crystal sample was mounted on a support with its (111) face parallel to the electrodes (Fig. 1b), which connected to a pulsed voltage generator (AVR-5B from AVTECH) with 20-ns pulse edges. The 50-\(\Omega\) impedance was added to ensure the high speed and eliminate the spurious signals. The pulsed electron paramagnetic resonance (EPR) experiments were carried out on a Bruker E580 spectrometer equipped with Oxford CF935 cryostat. The single crystal was rotated around the \([1, 2, 0]\) axis with a goniometer of 0.1-degree resolution.