Optical holonomic single quantum gates with a geometric spin under a zero field

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The realization of fast fault-tolerant quantum gates on a single spin is the core requirement for solid-state quantum-information processing. As polarized light shows geometric interference, spin coherence is also geometrically controlled with light via the spin–orbit interaction. Here, we show that a geometric spin in a degenerate subspace of a spin-1 electronic system under a zero field in a nitrogen vacancy centre in diamond allows implementation of optical non-adiabatic holonomic quantum gates. The geometric spin under quasi-resonant light exposure undergoes a cyclic evolution in the spin–orbit space, and acquires a geometric phase or holonomy that results in rotations about an arbitrary axis by any angle defined by the light polarization and detuning. This enables universal holonomic quantum gates with a single operation. We demonstrate a complete set of Pauli quantum gates using the geometric spin preparation and readout techniques. The new scheme opens a path to holonomic quantum computers and repeaters.

A quantum bit or qubit must be capable of being precisely and quickly manipulated, as well as being robust against noise. These criteria pose a dilemma in that the qubit must be open for a driving field but not for a noise field. It has been demonstrated that the degenerate subspace of a spin-1 electronic system under a zero field, which we call a geometric spin, can serve as a promising memory qubit that is robust against environmental noise. The challenge is to manipulate the degenerate qubit with the help of a geometric phase.

The concept of the geometric phase was first proposed by Pancharatnam in 1956 in reference to light polarization. Since then, two kinds of geometric phase have been discussed. Adiabatic geometric phases were first proposed by Berry in 1984 and non-adiabatic non-Abelian geometric phases were proposed by Anandan in 1988. Holonomic quantum computation (HQC) based on the adiabatic geometric phase was then proposed for fault-tolerant quantum gates by Zanardi and Rasetti in 1999 and generalized to non-adiabatic HQC by Wang and Matsumoto in 2001 and Zhu and Wang in 2002. The geometric phase has been experimentally demonstrated in molecular ensembles, in a superconducting qubit, in trapped ions, in a quantum dot and in a single nitrogen-vacancy (NV) centre in diamond.

These demonstrations, however, introduced fundamentally unnecessary energy splitting to the qubit to energetically select a well-defined eigenstate, or to use spin precession for arbitrary axis rotation. The splitting in turn prevented fast rotation, even in the non-adiabatic case, to avoid unwanted interference between the eigenstates. To overcome this problem, Sjöqvist proposed an optical scheme with a three-level Hamiltonian where the degenerate spin is prohibited because it requires a double transition. A classical light for rotation in the polarization state $|p\rangle = \cos(\theta/2)|+1\rangle_p + e^{i\phi}\sin(\theta/2)|-1\rangle_p$ based on circular polarizations $|\pm1\rangle_p$ couples the $|A_2\rangle$ with the corresponding state called the bright state $|B\rangle = \sin(\theta/2)|+1\rangle + e^{i\phi}\cos(\theta/2)|-1\rangle$, to induce orbital evolution in the spin–orbit space spanned by $|A_2\rangle$ and $|B\rangle$ states; in contrast, it uncouples the orthogonal state called the dark state $|D\rangle = \cos(\theta/2)|+1\rangle - e^{i\phi}\sin(\theta/2)|-1\rangle$, which remains unchanged, as described by the parameter-dependent interaction Hamiltonian $\hat{H} = (\Omega/2)(|A_2\rangle\langle B| + |B\rangle\langle A_2| - \Delta|A_2\rangle\langle A_2|)$, where $\Omega$ is the Rabi frequency on resonance and $\Delta$ denotes the detuning (see Methods). The unitary operator after a cyclic evolution in the spin–orbit space following the holonomy matrix $U(t_2) = e^{-i\Omega t_2/\hbar}(|B\rangle\langle B| + |D\rangle\langle D|) e^{-i\Omega t_2/\hbar}$, where the bright state acquires a geometric phase or holonomy of $\gamma = \pi(1 - (\Delta/\sqrt{D^2 + \Delta^2}))$, which corresponds to one-half of the solid angle enclosed by the orbital trajectory in the spin–orbit space (Fig. 1c). The cycle time (Rabi period) is shortened by the detuning $\Delta = (2\pi/\sqrt{D^2 + \Delta^2})$. The proper parameter set of $\{\theta, \phi, \Delta\}$ allows the geometric spin to be rotated by any angle $\gamma$ about an arbitrary axis defined by the unit gyration vector $\mathbf{B}$ pointing to $|B\rangle$, as shown in Fig. 1c. Also note that the unitary operators with different sets of $\{\theta, \phi, \Delta\}$ do not generally

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Figure 1 | Optical geometric spin rotation. a, Molecular structure of a nitrogen vacancy in diamond. b, Degenerate three-level Λ system and conceptual explanation for the geometric spin rotation. c, Correspondence between the polarization vector for the rotation light represented in the Poincaré sphere (left) and the rotation vector for the geometric spin represented in the Bloch sphere (right). Rotation angle γ is determined by the solid angle 2γ of the cyclic evolution in the spin-orbit space based on the relevant excited state |A2⟩ and the bright state |B⟩ for the rotation light (middle).

Figure 2 | Experimental procedure and characterization of the NV centre. a, Experimental set-up. b, Experimental pulse sequence and related transition in the three-level Λ system. The degenerate |±1⟩ states are reconfigured into bright |B⟩ and dark |D⟩ states defined by the polarizations for the preparation, rotation and readout lights. c, Photoluminescence excitation (PLE) spectrum with respect to the optical transition from the |±1⟩ states to the A2 state, which has a lifetime of 12 ns. d, Optically driven Rabi oscillation for the vertically polarized light |−⟩p = (|+⟩p − |−⟩p)/√2 in spin-orbit space. The vertical axis indicates the bright-state population. e, Geometric rotation angle (red) and 2π pulse length in the spin-orbit space (blue) as a function of detuning frequency from the A2 resonance, where the Rabi frequency on resonance is 250 MHz.
The reconstructed states, however, largely deviate from the expected states, which are expected to trace the great circles in the Bloch sphere. 

The geometric spin states rotated about the X, Y and Z axes, beginning at the |+, +⟩ and |−⟩ states, respectively (yellow dots), are plotted in the Bloch sphere. The red (blue) dots show the states rotating around the positive (negative) bright-state vector (indicated with arrows). Solid lines show the fitting curves, assuming the X and Y components of the off-alignment of the NV centre and the crystal strain based on the Hamiltonian analysis. The same plots after polarization compensation for the state preparation, rotation and readout. Arrows indicate the rotation axes. The pulse length is thus calibrated depending on light polarization and detuning to provide the expected rotation angle, as shown in Fig. 2e.

We evaluated the quantum process of the optical geometric spin rotation by reconstructing the geometric spin-state vectors rotated around the X, Y and Z axes based on quantum state tomography, which are expected to trace the great circles in the Bloch sphere. The reconstructed states, however, largely deviate from the expected trajectories, as shown in Fig. 3a. These state vectors are reproduced well by the Hamiltonian analysis, including the off-alignment of the NV centre and the crystal strain (see Methods). The unwanted effects are well compensated by adapting the light polarization to correct the dark states to prepare ideal states and bright states that faithfully rotate and read out the states, as shown in Fig. 3b.

The values of the optical holonomic quantum gates were evaluated by quantum process tomography (see Methods). The process fidelities for the holonomic Pauli-X (bit flip or NOT), Y (bit and phase flip) and Z (phase flip) gates are 92(11)%, 89(3)% and 90(10)%, respectively (Fig. 3c). The fidelities are mainly limited by dephasing induced by the real excitation to the A₂ state followed by the spontaneous relaxation back to the ±1 states, seen as amplitude damping of the optical Rabi oscillation in Fig. 2d. Other elemental operations required to build universal quantum gates such as the Hadamard gate

\[ H = \frac{1}{\sqrt{2}} \begin{pmatrix} 1 & 1 \\ 1 & -1 \end{pmatrix} \]
phase gate

\[ S = \begin{bmatrix} 1 & 0 \\ 0 & i \end{bmatrix} \]

and \( \pi/8 \)-rotation gate

\[ T = \begin{bmatrix} 1 & 0 \\ 0 & e^{\pi i / 8} \end{bmatrix} \]

can also be implemented with a single operation by choosing the proper parameter sets of \( [\theta, \phi, \Delta] = [\pi/4, 0, 0] \) for \( \text{H} \), \( [0, 0, (\Delta / \sqrt{3})] \) for \( S \) and \( [0, 0, (3\Delta / \sqrt{7})] \) for \( T \) to achieve the shortest pulse length of the rotation light. The fidelities could be further increased by increasing the power to decrease the pulse length up to the limit given by the splitting between the \( A_2 \) and \( A_1 \) states (typically around 3 GHz). Although we showed only one-qubit gates, strong dependence on detuning in the nearly resonant scheme, shown in Fig. 2c, in contrast to the conventional far-detuned or off-resonant scheme, allows implementation of two-qubit gates, such as the C-phase gate. For example, if we use a proximal \(^{13}\text{C}\) isotope with hyperfine coupling stronger than the natural linewidth of the \( A_2 \) state, we can selectively gate the electron spin under the conditions of the nuclear spin, such as \( \hat{Z}_S |+1\rangle \langle +1| + |−1\rangle \langle −1| \), which is simply the C-phase gate. With those single-qubit and two-qubit gates, we could construct a universal non-Abelian non-adiabatic HQC (refs 5,19).

The arbitrary angle rotation obtained here is extremely important for achieving a fast quantum Fourier transform, which requires controlled-\( R_k \) gates, where

\[ R_k = \begin{bmatrix} 1 & 0 \\ 0 & e^{2\pi i k / 2^N} \end{bmatrix} \]

and \( k \) is an integer less than the system size \( N \). Although it is possible to construct \( R_2 \) gates with a combination of \([X, H, S, T]\) gates, this is not efficient for handling large systems. On the other hand, the holonomic quantum gate directly performs the \( R_k \) gates.

The geometric rotation near resonance is essentially different from the conventional spin rotation based on the three-level \( A \)
system with far detuning or off resonance27–29. Figure 4a shows the simulated gate fidelity for X gates as a function of detuning and pulse length. When the detuning is smaller than the Rabi frequency of 250 MHz, one turn (or a few turns) of cyclic evolution in the spin–orbit space induces the geometric phase in the spin space. In contrast, when the detuning is much larger than the Rabi frequency, the dynamic phase, instead of the geometric phase, accumulates continuously to result in the spin rotation, which is known as the stimulated Raman transition27–29 or the optical Stark effect27–28. Although the conventional off-resonant scheme seems better for reducing the probability of transition into the excited state and the subsequent spontaneous relaxation, it requires a longer operation time than in the present near-resonant stimulated-Raman scheme (Fig. 4a). The operation time is further reduced with a smaller rotation angle, leading to further high fidelity. Note that the gate fidelities for the 2π rotation shown in Fig. 3c are the worst case in the X, Y and Z axes. Note also that the geometric rotation is extremely robust against pulse-length error in comparison to the straightforward dynamic rotation in the two-level system, especially for larger angles rotations, as shown in Fig. 4b,c. The detailed tolerance analyses against various types of energy shifts are shown in Supplementary Fig. 1. Although some tendencies are seen to depend on rotation angle, the tolerances are generally large enough to withstand unexpected field fluctuations; more importantly, optical dynamic rotation can never be used for the ground state spin in practice.

The geometric rotation relies only on light polarization, not on the spin precession conventionally used to compose an arbitrary axis rotation. It was performed under a completely zero field, which is desirable for the quantum coherence of the geometric spin, because it means the surrounding 13C nuclear spins are completely frozen3. Moreover, the unwanted timing problem caused by interference from those spins is avoided. Although detuning was used in the demonstration to change the rotation angle, the same operation could be performed by a discrete phase change, instead of the continuous phase change, called detuning, used here. The typical approach to a discrete phase change is to change the phase of the rotation light at the north pole (|A2⟩) in the spin–orbit space. The general composite pulse scheme ideally relaxes the requirements for specific pulse lengths, and pulse length error will not depend on the rotation angle. However, this scheme lowers the fidelity somewhat, especially for small angle rotations, due to the longer total operation time. We therefore chose the continuous phase change scheme rather than the discrete phase change scheme for its faster operation and higher fidelity.

The demonstration by Yale et al.17 used the dark state in the (±1) space to acquire the geometric phase known as the Berry phase8 by adiabatically changing the dark state to make a cyclic evolution. In contrast, our demonstration used the bright state to acquire the geometric phase known as the Aharonov–Anandan phase1 by non-adiabatically applying a short optical pulse to make a cyclic evolution in the spin–orbit space, thus enabling faster quantum gate operations within a few nanoseconds instead of a few hundred nanoseconds, as in Yale’s demonstration. We also achieved complete rotation about an arbitrary axis by any angle corresponding to the light polarization and the phase, with fidelities as high as 90%, in strong contrast to Yale’s experiments.

Combined with our previous demonstrations of optical spin state tomography23 and optical spin state preparation23, our present observations established a complete set of elemental quantum operations for a solid-state spin composed of initialization, manipulation and readout. The developed method could also be applied to the manipulation of a nanospin to provide an optical pickup for a magnetoresistive random access memory (MRAM). It is also suitable for an ultrasensitive quantum sensor beyond the classical limit, because it works perfectly under zero magnetic field.

Geometric rotation was used to implement non-adiabatic holonomic quantum gates to manipulate the states of geometric spin qubits, which are known to be robust against control errors and environmental noise18–29 and are thus expected to be building blocks of HQC. Our approach is applicable to any other three-level system based on defect centres, ion traps, quantum dots and superconducting circuits. The optical control is especially useful to individually address integrated spins, which require nanoscale local access. It thus opens a path to building holonomic quantum processors for quantum computers. The holonomic quantum gates could also be used for basis transformation between four Bell states and the optimization or calibration of teleportation-based quantum state transfer23 to build holonomic quantum repeaters for long-distance quantum communication networks31.

Methods

Methods and any associated references are available in the online version of the paper.

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**Author contributions**
N.N. carried out the experiment. Y.S., R.K. and H.Ka. supported the experiment. Y.S. and H.Ko. analysed the data. Y.S. and H.Ko. wrote the manuscript. H.Ko. supervised the project. All authors discussed the results and commented on the manuscript.

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**Competing financial interests**
The authors declare no competing financial interests.
Methods

Experimental set-up. We used a native NV centre in a high-purity type-IIa chemical-vapour-deposition-grown bulk diamond with a (001) crystal orientation (electronic grade from Element Six) without ion implantation dose or annealing. A negatively charged NV centre located ~30 μm below the surface was found using a confocal laser microscope. A 25 μm copper wire mechanically attached to the surface of the diamond was used to apply a microwave. An external magnetic field was applied to carefully compensate for the geomagnetic field of ~0.045 mT using a permanent magnet with monitoring of the optically detected magnetic resonance (ODMR) spectrum within 0.1 MHz. The Rabi oscillation and Ramsey interference were also used to fine-tune the field. The NV centre used in the experiment showed hyperfine splittings caused by the 14N nuclear spin at 2.2 MHz and by 13C nuclear spins within 0.4 MHz. All experiments were performed at 5 K to reduce the optical line width of the A2 transition to as narrow as 54 MHz (Fig. 2c). The splitting between the E1 and E2 transitions indicated the crystal strain was 2.2 GHz (absolute value).

The experimental set-up was the same as used in ref. 2, except for an additional red laser for the spin projection (Fig. 2a). Figure 2b presents schematics of the pulse sequence used in the experiments. The arbitrary geometric spin state was prepared by the dark-state preparation method used in ref. 2. A green laser (532 nm, 100 µW) of 3 µs was first used to initialize the electron spin states to the ground mS = 0 state |0⟩, and a microwave (2.878 GHz) was then used to excite the electron spin to the geometric spin state. Finally, a red light resonant to the A2 state (-30 µW) in any arbitrary axis corresponding to the bright state of the light polarization is defined by the detuning. In this context, the optical geometric spin rotation is regarded as a bright-axis rotation. The rotated geometric spin state was then projected into any arbitrary state by the bright-state projection method used in ref. 2. The red light (0.1 µW, 10 ns) resonantly excited the bright state to the A2 state, which then relaxed to the ground state while emitting a photon with a different wavelength (photon sideband emission) for detection. All the light beams were focused onto the sample using a 0.7 NA ×100 objective inside the vacuum.

Hamiltonian used for the analysis. The fine structures of the orbital excited states in an NV centre are well defined at temperatures below ~10 K, and the eigenstates are individually accessible with resonant light. The Hamiltonian for the excited state is described as

$$H_{\text{exc}} = l_\lambda l_x S_x + D_{\text{A}} (l_x^2 S_x^2 + l_y^2 S_y^2) + e_x (l_x^2 S_x^2 + l_y^2 S_y^2) + i e_y (l_x^2 S_x^2 - l_y^2 S_y^2) \quad (1)$$

where $L_x$ and $S_x$ are the axial components of the orbital and spin angular momentum operators, $l_\lambda$ and $S_x$ are the raising and lowering operators defined as $L_x = (L_x \pm iL_y)/\sqrt{2}$ and $S_x = (S_x \pm iS_y)/\sqrt{2}$ on the SU(3) system in the bases of $|\{1,0,0\}, \{0,1,0\}\rangle$, $L_\lambda$ is the spin–orbit interaction, $D_{\text{A}}$ and $D_{\text{B}}$ are zero-field splittings arising from the axial and perpendicular spin–spin interaction, respectively, in the excited states and $e_x$ and $e_y$ are the x and y components of the crystal strain. The following six eigenstates are energetically well separated except for the $E_1$ and $E_2$ states under a low-strain regime:

- $|A_2\rangle = (|+1\rangle l_x|+1\rangle + |-1\rangle l_\lambda|+1\rangle)\sqrt{2}$
- $|A_1\rangle = (|+1\rangle l_x|-1\rangle - |-1\rangle l_\lambda|+1\rangle)\sqrt{2}$
- $|E_1\rangle = (|+1\rangle l_x|+1\rangle |+1\rangle + |-1\rangle l_\lambda|+1\rangle)\sqrt{2}$
- $|E_2\rangle = (|+1\rangle l_x|-1\rangle |+1\rangle + |-1\rangle l_\lambda|+1\rangle)\sqrt{2}$
- $|E_3\rangle = (|+1\rangle l_x|-1\rangle |+1\rangle)\sqrt{2}$
- $|E_4\rangle = (|+1\rangle l_x|+1\rangle |+1\rangle)\sqrt{2}$

On the other hand, the Hamiltonian for the orbital ground state is described as

$$H_{\text{gs}} = D_{\text{B}} S_x^2 \quad (8)$$

where $D_{\text{B}}$ is the zero-field splitting arising from the spin–spin interaction in the ground state. The optical excitation induces the orbital transition, depending on the light polarization. The driving Hamiltonian is described as

$$H_{\text{drive}} = \frac{\Omega}{\sqrt{2}} \left( \cos \left( \frac{\theta}{2} \right) |0\rangle \langle 1| + e^{i\phi} \sin \left( \frac{\theta}{2} \right) |1\rangle \langle 0| \right) + \text{H.c.} \quad (9)$$

where $\theta (\phi)$ denotes a polar (azimuth) angle in the Poincaré sphere, which represents a light polarization state, and H.c. indicates the Hermit conjugate. The driving Hamiltonian for the resonant optical transition between $|0\rangle_1|\pm1\rangle_2$ (hereafter indicated as $|\pm1\rangle$) and $|A_2\rangle$ is written as follows to conserve the spin angular momentum:

$$H_{\text{A}} = \frac{\Omega}{2} \cos \left( \frac{\theta}{2} \right) |A_2\rangle \langle -1| + e^{i\phi} \sin \left( \frac{\theta}{2} \right) |A_2\rangle \langle +1| \right) + \text{H.c.} \quad (10)$$

where $\Omega$ is the Rabi frequency. The Hamiltonian describes the dynamics in the degenerate three-level A system in the computational bases.

Bright-state driving. The $|\pm1\rangle$ basis states can be transformed into the bright state $|B\rangle$ and dark state $|D\rangle$, where the bright state is coupled with the excited state to create a new eigenstate, while the dark state is kept in its eigenstate. With this transformation, the driving Hamiltonian in the degenerate three-level A system is transformed as follows:

$$H_{\text{A}} = \frac{\Omega}{2} (|A_2\rangle \langle B| + |B\rangle \langle A_2|) \quad (11)$$

where

$$|B\rangle = \sin \left( \frac{\theta}{2} \right) |+1\rangle + e^{i\phi} \cos \left( \frac{\theta}{2} \right) |+1\rangle \quad (12)$$

and

$$|D\rangle = \cos \left( \frac{\theta}{2} \right) |+1\rangle - e^{i\phi} \sin \left( \frac{\theta}{2} \right) |+1\rangle \quad (13)$$

The bright state represented in the Bloch sphere shows one-to-one correspondence with the light polarization $|\psi_p\rangle$ represented in the Poincaré sphere as

$$|\psi_p\rangle = \cos \left( \frac{\theta}{2} \right) |+1\rangle + e^{i\phi} \sin \left( \frac{\theta}{2} \right) |+1\rangle \quad (14)$$

where $|\psi_p\rangle$ indicates right and left circular polarizations.

In general, the $A$ system Hamiltonian under driving light needs to add the detuning $\Delta$ as follows:

$$H_{\Delta} = \frac{\Omega}{2} (|A_2\rangle \langle B| + |B\rangle \langle A_2|) - \Delta |A_2\rangle \langle A_2| \quad (15)$$

where $\Delta = \sqrt{\gamma^2 + \Delta'}^2$ is the effective Rabi frequency, $\gamma = (\Omega/\Delta') (1 - \Delta/\Delta')$ is a unit vector indicating the rotation vector, and $\gamma_\Delta = \Omega_\Delta = (\gamma_x, \gamma_y, \gamma_z)$ are the Pauli operators and identity operator based on $|A_2\rangle$ and $|B\rangle$.

The time evolution operator is now written as

$$U_{\Delta}(t) = \exp \left( -i \frac{\Delta}{\gamma} \left( |A_2\rangle \langle A_2| - |B\rangle \langle B| \right) \right) \exp \left( -i \frac{\Delta'}{\gamma} \left( |A_2\rangle \langle A_2| + |B\rangle \langle B| \right) \right) \quad (16)$$

The evolution operator for a round trip $t_{\text{round}} = (\pi/2) \Delta$ in the $(|A_2\rangle - |B\rangle)$ Bloch sphere becomes

$$U_{\Delta}(t_{\text{round}}) = \exp \left( -i \frac{\Delta}{\gamma} \right) \left( |A_2\rangle \langle A_2| - |B\rangle \langle B| \right) \exp \left( -i \frac{\Delta'}{\gamma} \right) \quad (17)$$

The operation is interpreted as the rotation around the bright state $|B\rangle$ by an angle $\pi (1 - \Delta/\Delta')$ in the Bloch sphere spanned with the $(|\pm1\rangle$) basis states. An arbitrary angle rotation around an arbitrary axis thus becomes possible by choosing the proper light polarization, a detuning $\Delta$ and the corresponding pulse length.

Hamiltonian compensation on the light polarization. As NV centres in diamond oriented along [100] are off-aligned from the optical axis by arccos $1/\sqrt{3}$, the electric-field amplitude of the light polarized along the inclined NV axis is decreased by a factor of $1/\sqrt{3}$ as the electric field is projected onto the plane normal to the NV axis, resulting in a reduction of the Rabi frequency. As a result, the polarization observed by the NV electron is different from the incident polarization due to the off-alignment. In addition, the crystal strain mixes the $|A_2\rangle$ state with the $|E_2\rangle$ and $|E_3\rangle$ states to rearrange the straightforward correspondence between the bright state, defined as equation (12), and the light polarization, defined as equation (14).

In this experiment, we estimated the strain parameters for $E_2 = -1.2$ GHz, $E_3 = -1.8$ GHz from the fitting to the quantum state tomography of the spin states rotated around the x, y, and z axes. As in equation (13), the ideal Hamiltonian is then created to calibrate both the off-alignment and strain effects by finding the adopted light polarization. The bright and dark states are obtained by projecting the eigenstates of the total Hamiltonian as the sum of equations (1), (8) and (9) onto the qubit space $|\pm1\rangle$. The light polarization adapting to the Hamiltonian can be found to realize the expected rotation. The light polarization parameters with and without
the compensation for the ±X, ±Y and ±Z rotations are summarized in Supplementary Table 1.

**Master equation.** We analysed the experimental data shown in Figs 2d and 4a based on the Lindblad master equation to simulate the relaxation process. The relaxation is caused by two processes: energy relaxation with a decay time of $T_1 = 12$ ns of the excited state $|A_2\rangle$ to the $|±1\rangle$ state, and phase relaxation with a dephasing time of $T_2^*$ between the $|±1\rangle$ states. The $T_2^*$ was estimated to be 4.6 ns from fitting to the Rabi oscillation shown in Fig. 2d. We neglected the relatively long energy relaxation from $|A_2\rangle$ to $|0\rangle$ for simplicity. The phase relaxation between the $|±1\rangle$ states was also negligible, because the dephasing time was on the order of a few microseconds.

**Quantum process tomography.** The $\chi$ matrices shown in Fig. 3c, representing the rotation gate operations in Fig. 3b, were reconstructed via quantum process tomography, which compares the final state after the gate operation with the initial state prepared in $|+\rangle, |+i\rangle, |+1\rangle, |−1\rangle$. As the obtained raw $\chi$ matrices are likely to be unphysical, the most likely $\chi$ matrices were deduced by assuming the trace conservation and the normal matrix based on the maximum likelihood estimation method. Optimization was performed using the differential_evolution function, which enables global optimization, in the scipy.optimize package of the python program.

**Data availability.** The data that support the plots within this paper and other findings of this study are available from the corresponding author upon reasonable request.

**References**
32. Howard, M. et al. Quantum process tomography and Lindblad estimation of a solid-state qubit. *New J. Phys.*, 8, 33–33 (2006).