

Supplementary information

Geometric entanglement of a photon and spin qubits in diamond

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Supplementary Note 1. GRAPE pulse

To increase the fidelity of quantum operations, we use gradient ascent pulse engineering (GRAPE) algorithm¹ to search for the optimal pulses. This allows us to perform the operations with relatively high fidelity without requiring a sufficiently large Rabi frequency compared to the nitrogen-induced hyperfine splitting, which mainly degrades the operational fidelity. In this work, we optimize the amplitude modulation of the pulses for qubit state preparation and readout, which transfer the ancillary state $|0\rangle_S$ to the target qubit state $|\psi\rangle_S = \cos\theta_{MW}|+1\rangle_S + e^{-i\phi_{MW}}\sin\theta_{MW}|-1\rangle_S$ and vice versa. The optimized pulse is made with microwave polarization $|\psi\rangle_{MW} = \cos\theta_{MW}|+1\rangle_{MW} + e^{-i\phi_{MW}}\sin\theta_{MW}|-1\rangle_{MW}$ according to the target state, while the amplitude modulation depends on θ_{MW} . This is because the effect of hyperfine splitting is independent of ϕ_{MW} . Figure S1 shows the amplitude modulation of the GRAPE pulses for each θ_{MW} .

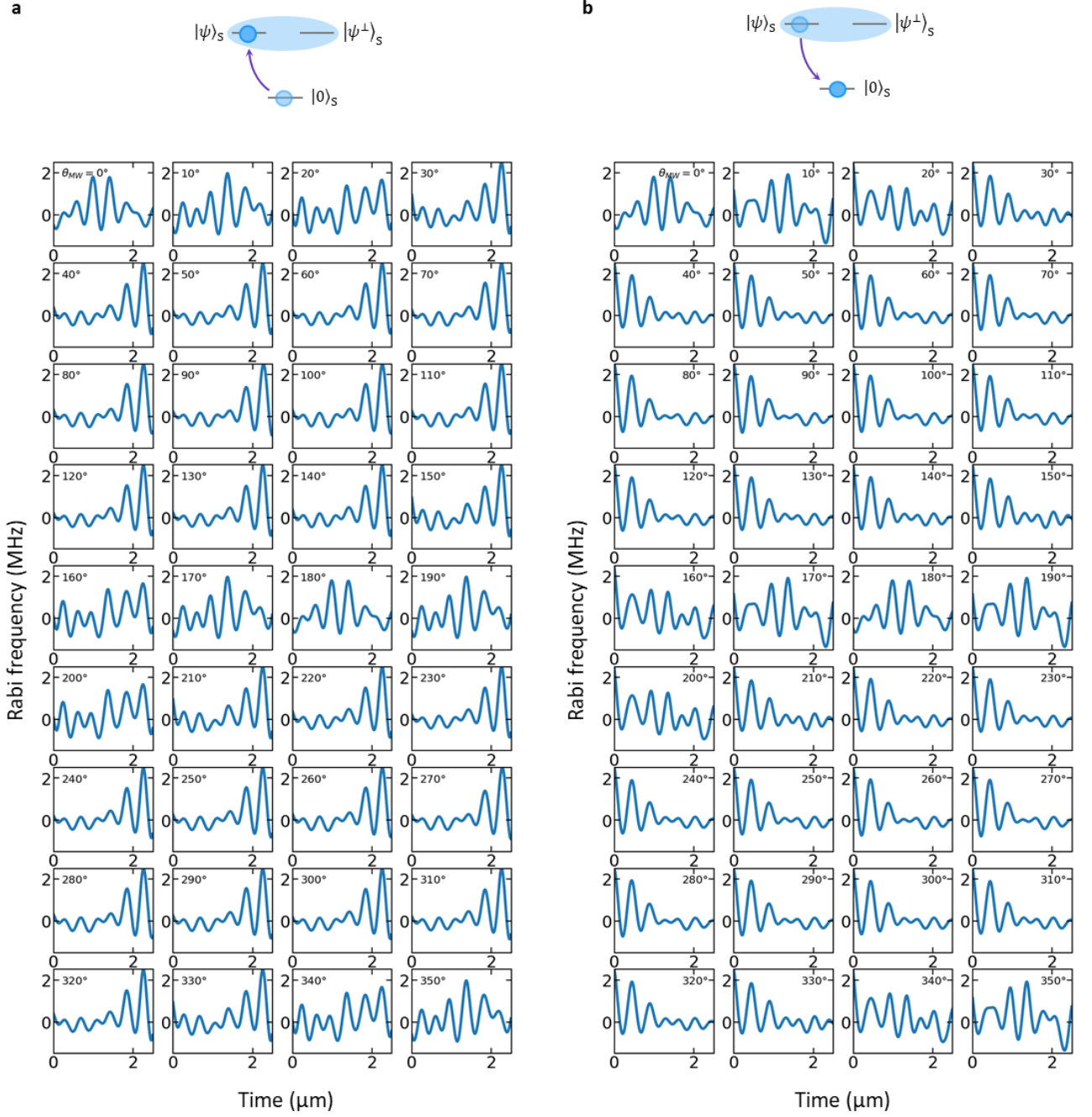


Fig. S1| Amplitude modulation of GRAPE pulses. **a**, Pulse envelope (Rabi frequency) of the state transfer from the ancillary state $|0\rangle_S$ to the target qubit state $|\psi\rangle_S$ depending on θ_{MW} , where $|\psi^\perp\rangle_S$ is the orthogonal state of $|\psi\rangle_S$. **b**, Pulse envelope of the state transfer from $|\psi\rangle_S$ to $|0\rangle_S$.

Supplementary Note 2. Qubit preparation and readout

The experimental sequence of spin preparation and measurement in Fig. 2 is described below (Fig. S2). First, the electron spin is initialized to the zero state by spin pumping with resonant excitation to the $E_{1,2}$ excited states. An arbitrary qubit state is then prepared with a GRAPE pulse, and immediately transferred to the zero state with another GRAPE pulse for qubit readout. The population transferred to the zero state reflect the population of the readout basis selected by the polarization of the GRAPE pulse. To read out the zero state, we perform a resonant excitation to the spin-conserving E_y excited state and detect the subsequent PSB emission with an APD.

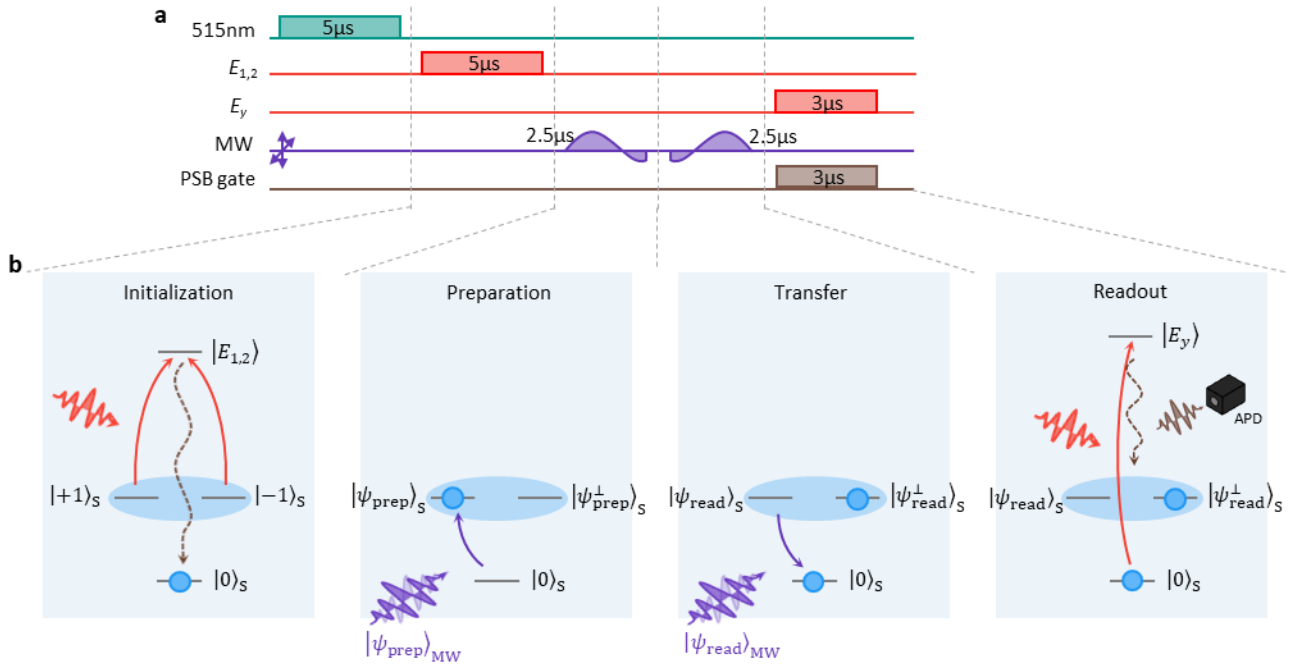


Fig. S2| Experimental sequence of qubit preparation and readout. a, Timing sequence of the experiment, which include the charge initialization, the spin initialization, preparation, transfer and readout. **b**. Schematic dynamics illustrated in the energy level structure.

Supplementary Note 3. Fidelity of entangled state

The ideal entangled state in this study $|\Psi_+\rangle = \frac{|+1\rangle_p|-1\rangle_s + |-1\rangle_p|+1\rangle_s}{\sqrt{2}}$ can be expressed in the form of a density matrix as follows:

$$\rho_{\Psi^+} = |\Psi^+\rangle\langle\Psi^+| = \frac{\sigma_0\sigma_0 + \sigma_x\sigma_x + \sigma_y\sigma_y - \sigma_z\sigma_z}{4} \quad (\text{S1})$$

where σ_0 , σ_x , σ_y and σ_z are identity operator and the Pauli operators. Therefore, the fidelity of the experimentally generated two-quantum state ρ_{ex} can be decomposed into the following elements:

$$F = \text{Tr}(\rho_{\Psi^+}\rho_{\text{ex}}) = \frac{1 + \text{Tr}(\sigma_x\sigma_x\rho_{\text{ex}}) + \text{Tr}(\sigma_y\sigma_y\rho_{\text{ex}}) - \text{Tr}(\sigma_z\sigma_z\rho_{\text{ex}})}{4}, \quad (\text{S2})$$

where

$$\text{Tr}(\sigma_x\sigma_x\rho_{\text{ex}}) = \langle+,+|\rho_{\text{ex}}|+,+\rangle - \langle+,-|\rho_{\text{ex}}|+,-\rangle \quad (\text{S3})$$

$$- \langle- ,+|\rho_{\text{ex}}|- ,+\rangle + \langle- , -|\rho_{\text{ex}}|- , -\rangle ,$$

$$\text{Tr}(\sigma_y\sigma_y\rho_{\text{ex}}) = \langle+i,+i|\rho_{\text{ex}}|+i,+i\rangle - \langle+i,-i|\rho_{\text{ex}}|+i,-i\rangle \quad (\text{S4})$$

$$- \langle-i,+i|\rho_{\text{ex}}|-i,+i\rangle + \langle-i,-i|\rho_{\text{ex}}|-i,-i\rangle ,$$

$$\text{Tr}(\sigma_z\sigma_z\rho_{\text{ex}}) = \langle+1,+1|\rho_{\text{ex}}|+1,+1\rangle - \langle+1,-1|\rho_{\text{ex}}|+1,-1\rangle \quad (\text{S5})$$

$$- \langle-1,+1|\rho_{\text{ex}}|-1,+1\rangle + \langle-1,-1|\rho_{\text{ex}}|-1,-1\rangle ,$$

where $|\psi_p, \psi_s\rangle$ denotes $|\psi_p\rangle_p |\psi_s\rangle_s$ for the simplicity. All terms in equations (S3-5) can be directly measured experimentally. In our experiment, since the spin measurement is conditioned on the detection of the photon polarization in one basis, the polarization balance cannot be accurately determined from the measuremental results due to the fluctuation of the excitation power in time for each photon measurement basis and the variation of the measurement efficiency. Hence, we calculate the fidelity assuming that the polarization imbalance can be corrected by a polarization-dependent attenuator in practice. Table S1 lists the 12 different event counts obtained in our experiments. The number of error events is estimated from Fig. 3a. corresponding to the gate delay time selected for each measurement. Since it is assumed that the spin is in a maximally mixed state when an error event is detected, the events are subtracted from the spin state probability.

In the following, we will discuss the main errors of the entangled state mentioned in the main text. The ideal entangled state $|\Psi^+\rangle$ is expressed in terms of the basis of attenuated polarization $|e^{2i\phi_{\text{NV}}}\rangle_{\text{p}}$ and the orthogonal polarization $|-e^{2i\phi_{\text{NV}}}\rangle_{\text{p}}$ estimated from Fig. 4d as follows:

$$|\Psi^+\rangle = e^{-2i\phi_{\text{NV}}} \frac{|e^{2i\phi_{\text{NV}}}\rangle_{\text{p}}|e^{2i\phi_{\text{NV}}}\rangle_{\text{s}} - |-e^{2i\phi_{\text{NV}}}\rangle_{\text{p}}|-e^{2i\phi_{\text{NV}}}\rangle_{\text{s}}}{\sqrt{2}}, \quad (\text{S6})$$

where $|\pm e^{2i\phi_{\text{NV}}}\rangle = \frac{|+1\rangle \pm e^{2i\phi_{\text{NV}}}|-1\rangle}{\sqrt{2}}$ and ϕ_{NV} denotes the angle of the NV axis (see Supplementary Note 4). When the attenuation coefficient α is applied to $|-e^{2i\phi_{\text{NV}}}\rangle_{\text{p}}$ basis, $|\Psi^+\rangle$ turns into

$$|\Psi^{+\prime}\rangle = \frac{|e^{2i\phi_{\text{NV}}}\rangle_{\text{p}}|e^{2i\phi_{\text{NV}}}\rangle_{\text{s}} + \alpha |-e^{2i\phi_{\text{NV}}}\rangle_{\text{p}}|-e^{2i\phi_{\text{NV}}}\rangle_{\text{s}}}{\sqrt{1+\alpha^2}}, \quad (\text{S7})$$

where the global phase is ignored. The state fidelity in the presence of polarization distortion is calculated by

$$F = |\langle\Psi^+|\Psi^{+\prime}\rangle|^2 = \frac{(1+\alpha)^2}{2(1+\alpha^2)}. \quad (\text{S8})$$

Therefore, the error due to NV-axis misalignment is estimated to be 6%. The spin measurement error is estimated to be 2% from the state preparation and readout experiments shown in Fig. 2, assuming that the preparation and readout operations contain similar errors. For the hybridization of excited states, the error from the ideal maximally entangled state is estimated to be 1% by calculating the eigenstate of A_2 state from the Hamiltonian considering the magnitude of the crystal distortion of NV used in this experiment.

Table S1. The number of measured events.

Photon state	$ +1\rangle_{\text{p}}$		$ -1\rangle_{\text{p}}$		$ +\rangle_{\text{p}}$		$ -\rangle_{\text{p}}$		$ +i\rangle_{\text{p}}$		$ -i\rangle_{\text{p}}$	
Spin state	$ +1\rangle_{\text{s}}$	$ -1\rangle_{\text{s}}$	$ +1\rangle_{\text{s}}$	$ -1\rangle_{\text{s}}$	$ +\rangle_{\text{s}}$	$ -\rangle_{\text{s}}$	$ +\rangle_{\text{s}}$	$ -\rangle_{\text{s}}$	$ +i\rangle_{\text{s}}$	$ -i\rangle_{\text{s}}$	$ +i\rangle_{\text{s}}$	$ -i\rangle_{\text{s}}$
Total events	17	100	107	23	140	7	17	184	205	32	16	151
Estimated error events	11.3		12.6		0		0		23.0		16.2	

Supplementary Note 4. Distortion of optical polarization

Since the electric dipole of the electron in a NV center is directed perpendicular to the NV axis, the absorption efficiency is maximum when the polarization of the incident light is also in the same direction. However, since the diamond sample used in this experiment has a crystal orientation of 100, the NV axis is expected to be tilted by about 54.7° relative to the optical axis. The polarization of light propagating perpendicular to the diamond surface can be decomposed into a component perpendicular to the NV axis and a component orthogonal to it, whose amplitude is projected onto a plane perpendicular to the NV axis, reducing its effective amplitude. As a result, the effective polarization is distorted (Fig. S3). In the main text, we analyze the polarization distortion by fitting the experimental results with the tilt of the NV axis as free parameters. Although crystal strain can also modulate the effective polarization by perturbing $|A_2\rangle$ state, we here incorporate it into the fidelity degradation of $|A_2\rangle$ state as the origin of the entanglement.

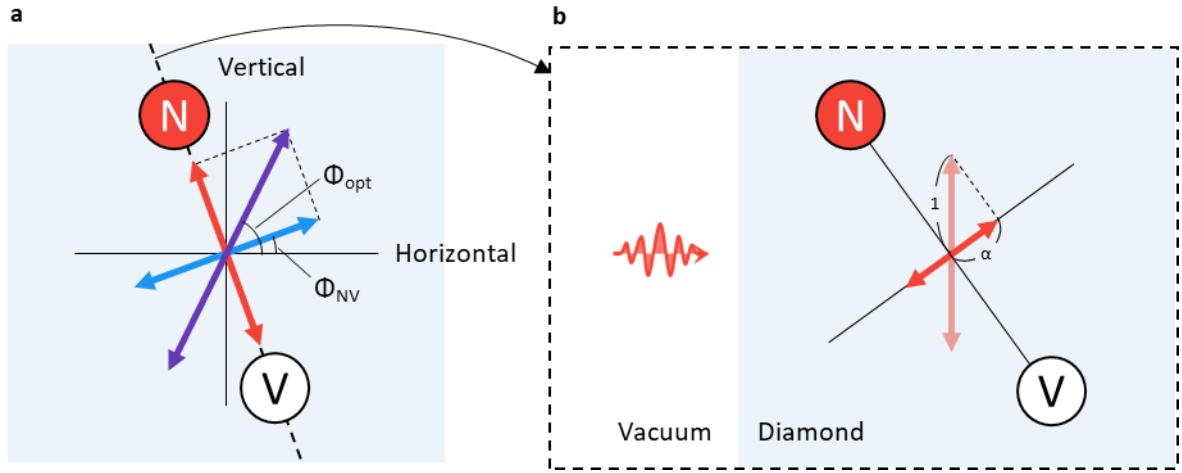


Fig. S3| Tilt of the NV axis. **a**, Front view of a diamond, showing the arbitrary polarization of light field defined by Φ_{opt} (purple arrows) decomposed into a component perpendicular to the NV axis (blue arrows) and a component orthogonal to it (red arrows). **b**, View of a plane perpendicular to the surface and parallel to the NV axis. The light field sensed by the NV electron is the component projected onto the plane perpendicular to the NV axis, which reduces the amplitude of the light field by a factor of α , thus distorting the polarization.

Supplementary Note 5. Interface of microwave and optical photons

In this section, we discuss the function of the interface between microwave and optical photons with a NV center. The four-level system of the electron under a zero magnetic field shown in Fig. 1b constitutes a hybrid system of V and Λ -type structure interacting with microwave and light fields respectively. Therefore, coherent operation across microwave and optical domains is feasible in principle. There are two possible schemes at least. The first scheme is quantum states transfer from microwave to optical photon. This is an implementation of the teleportation-like quantum state transfer technique at the V-type system instead of Λ -type system previously demonstrated by Yang et al.² and Tsurumoto et al.³. As shown in Fig. S4, after the generation of spin-photon entanglement, the microwave photon induces spin transition from the polarization-dependent state to the ancillary state. If the ancilla state is then measured by spin-dependent optical excitation, the polarization state of the optical photon is projected to the microwave-polarization-dependent state. Since the measurement of the ancilla state heralds the successful event of quantum state transfer, high fidelity transfer can be achieved even in the situation where the transfer probability is quite low. The second scheme is a probabilistic Bell measurement between microwave and optical photons. After preparation to $|0\rangle_S$, microwave photon probabilistically excites the spin to map the polarization state to the qubit state. Subsequently, if the absorption of optical photon is detected, it becomes a conditional Bell measurement of the qubit state and the photon polarization state, as previously demonstrated by Kosaka et al.⁴. As a result, a Bell measurement of microwave and photon polarization can be realized.

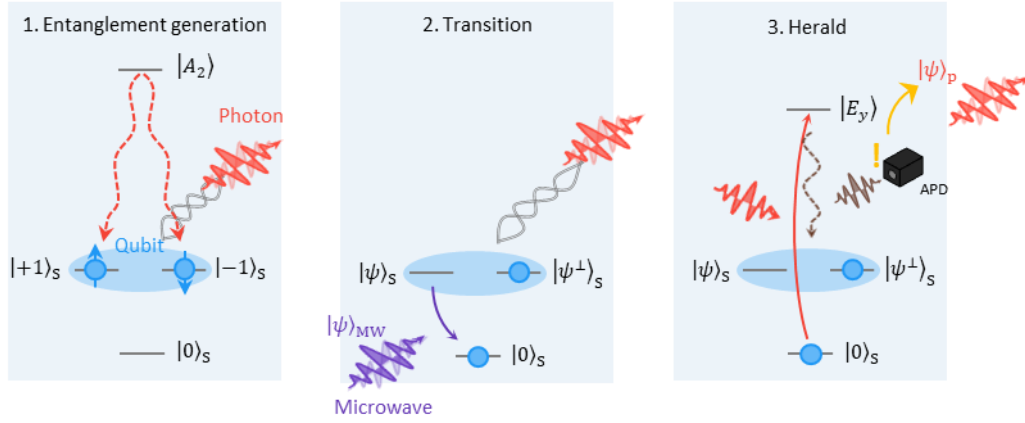


Fig S4| Protocol of quantum state transfer from a microwave photon to an optical photon. After the generation of spin-photon entanglement, the incoming microwave with arbitrary polarization $|\psi\rangle_{MW}$ probabilistically transfer the polarization-dependent spin state $|\psi\rangle_s$ to $|0\rangle_s$. When the transition to $|0\rangle_s$ is heralded by the resonant excitation to $|E_y\rangle$, a teleportation-like quantum state transfer from the microwave to optical photon is achieved.

Supplementary References

1. Khaneja, N., Reiss, T., Kehlet, C., Schulte-Herbrüggen, T. & Glaser, S. J. Optimal control of coupled spin dynamics: Design of NMR pulse sequences by gradient ascent algorithms. *J. Magn. Reson.* **172**, 296–305 (2005).
2. Yang, S. *et al.* High-fidelity transfer and storage of photon states in a single nuclear spin. *Nat. Photonics* **10**, 507–511 (2016).
3. Tsurumoto, K., Kuroiwa, R., Kano, H., Sekiguchi, Y. & Kosaka, H. Quantum teleportation-based state transfer of photon polarization into a carbon spin in diamond. *Commun. Phys.* **2**, 74 (2019).
4. Kosaka, H. & Niikura, N. Entangled Absorption of a Single Photon with a Single Spin in Diamond. *Phys. Rev. Lett.* **114**, 053603 (2015).