High-fidelity transfer and storage of photon states in a single nuclear spin

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Long-distance quantum communication requires photons and quantum nodes that comprise qubits for interaction with light and good memory capabilities, as well as processing qubits for the storage and manipulation of photons. Owing to the unavoidable photon losses, robust quantum communication over lossy transmission channels requires quantum repeater networks1,2. A necessary and highly demanding prerequisite for these networks is the existence of quantum memories with long coherence times to reliably store the incident photon states. Here we demonstrate the high-fidelity (∼98%) coherent transfer of a photon polarization state to a single solid-state nuclear spin that has a coherence time of over 10 s. The storage process is achieved by coherently transferring the polarization state of a photon to an entangled electron–nuclear spin state of a nitrogen–vacancy centre in diamond. The nuclear spin-based optical quantum memory demonstrated here paves the way towards an absorption-based quantum repeater network.

Quantum communication protocols rely on building quantum networks with stationary quantum nodes separated by large distances and photons (information carriers) to distribute entanglement between them1. Hence a quantum node should have the ability to coherently absorb and/or emit as well as store the information encoded in the photon state onto a memory element that has a very long coherence time2. Further, to maximize the entanglement among the nodes, each node should have additional processing qubits for entanglement purification or to perform quantum error corrections3–5. Finding a suitable system that meets all the requirements to be a functional quantum node is quite challenging. In this regard, ensembles of atomic gases, trapped ions and solid-state systems were intensively studied6–11. Although ensemble systems provide high interaction efficiency with photons, single qubits typically have longer storage times due to the absence of inhomogeneous broadening and weaker dephasing, as well as more important advantage for quantum computation networks due to their ability for in situ information processing14–16, for example entanglement purification15. Thus a hybrid quantum system composed of different physical components with complementary functionalities could be an ideal quantum node for any given quantum network.

The nitrogen–vacancy (NV) defect centre in diamond provides one such quantum hybrid (spin) system made of electron and nuclear spins. Although its electron spin is used for interaction with photons9, for fast18 and high-fidelity control19 and for readout of the spin state20–22, its surrounding nuclear spins are well-isolated from their environment and yield very long coherence times. Thus nuclear spins make natural candidates for information storage and electron spins for spin photon interface. In addition, electron and nuclear spins form a multiqubit quantum register allowing for quantum information processing, for example high-fidelity quantum error correction19 and quantum memory23.

Although there has been much progress in photon transfer and storage onto ensembles of atoms/spins that have strong coupling to photons, a direct transfer of the photon state to a single qubit has only been demonstrated in single atoms13,16. Although the non-linear effects induced by strong light–matter coupling facilitated the transfer process, the low coherence times allowed storage for only few hundred microseconds. However, achieving such strong light–matter coupling for single solid-state qubits still remains a large technical challenge. In this work, we propose a new scheme that does not require a strong coupling of solid-state qubits to the optical photons. We also store photons onto a qubit (nuclear spin) that has a long coherence time, a key requirement for practical quantum applications. Consequently, we succeed in the coherent transfer of an optical photon polarization state to a single nuclear spin in an NV defect centre in diamond that has a coherence time of 10 s.

The basic element of our system is a single NV centre consisting of an electronic spin (S = 1) and an intrinsic 14N nuclear spin (I = 1), coupled by hyperfine interaction. Owing to its C3v symmetry, the ground state of the NV centre is an orbital singlet (|Eg⟩) and the first optical excited state is an orbital doublet (|En⟩) (Ref. 24). At liquid helium temperature, one of the excited states, |En⟩, forms a lambda system with |±1⟩, in the ground states through σ– and σ+ polarized optical transitions as shown in Fig. 1b. Another excited state that is relevant in our protocol |En⟩ couples with |0⟩, in the ground state through a cycling spin–conserving optical transition (Fig. 1c).

The storage scheme resembles quantum teleportation with heralding. It consists of three steps (Fig. 1): Bell state preparation of the electron–nuclear spin system; optical transfer; and heralding. We start with the NV centre spin system prepared in an initial Bell state |Ψ⟩ = (1/√2)(|+⟩|0⟩ − |−⟩|1⟩) in the polarization state |ψ⟩p = (1/√2)(|↑⟩|σ+⟩ + e^iθ|↓⟩|σ−⟩) and in resonance with the Λ (A4) transition is absorbed by the NV centre. After the...
absorption of a photon, the collective photon–NV centre spin system evolves into the state $|\Psi\rangle \otimes |\rho_{\text{n}}\rangle$, where $|\Psi\rangle = (1/\sqrt{2}) (|+1\rangle_n - e^{i\phi}|−1\rangle_n)$. Conditional on the heralding result that is described in detail below, the information is transferred from the photon to the nuclear spin.

Figure 1 shows the circuit diagram and pulse sequence used in implementing the protocol. Initially, both the electron spin and $^{14}$N nuclear spin are thermally populated among three spin states. Before preparing the Bell state, we first initialize both of them to $|0\rangle_e|0\rangle_n$ with a fidelity higher than 98% (Fig. 2a, b and Methods).

To prepare the Bell state from this initialized spin state, we take advantage of the degeneracy of $|0\rangle_e|+1\rangle_n$ and $|0\rangle_e|-1\rangle_n$ in the absence of a magnetic field. Under resonant radiofrequency (RF) driving of the nuclear spin to state $|0\rangle_n|0\rangle_n$, this degeneracy leads to two new eigenstates, the bright state $|0\rangle_e|b\rangle_n = (1/\sqrt{2})(|+1\rangle_n + |−1\rangle_n)$, which couples with $|0\rangle_e|0\rangle_n$ and the dark state $|0\rangle_e|d\rangle_n = (1/\sqrt{2})(|+1\rangle_n - |−1\rangle_n)$, which remains uncoupled. The Bell state $|\Psi\rangle$ is then prepared from $|0\rangle_e|0\rangle_n$ by applying a nuclear spin $\pi$ pulse to $|0\rangle_e|b\rangle_n$ followed by an electron spin $\pi$ pulse (see Fig. 1a and Methods).

In the second step after the Bell state is created, the electron is resonantly excited by a photon in polarization state $|\psi\rangle$ to $|\Psi\rangle$ via the $\Lambda$ system (see Fig. 1b). We synchronize the excitation to be within 20 ns after the preparation of the Bell state to avoid any decoherence due to the surrounding spin bath. Owing to the low absorption rate (≈0.1%, see Methods) of the NV centre, a 12 ns laser pulse with a peak power of 200 nW (corresponding to an optical Rabi frequency of 27 MHz; ref. 25), containing a few thousand photons, is used to excite the NV centre. Although we use a classical light field to demonstrate state conversion, the defect can only absorb and emit single photons. The probability of absorbing more than one photon due to re-excitation is very small (~0.025, see Supplementary Information).

In the third and last step of the protocol, a heralding process is required to confirm the transfer. As the excited state $|\Psi\rangle$ decays back to the three ground states $|±1\rangle_s$, which by emitting a photon, the transfer could be heralded by detecting this photon. However, this approach suffers from a low photon collection efficiency of 2%. Instead, we herald the transfer process by detecting the electronic spin state in $|0\rangle_e$ through a high-fidelity single-shot readout based on the $|0\rangle_e \rightarrow |E_+\rangle$ spin-conserving transition (Fig. 1c)22. After absorption into $|\Psi\rangle$, subsequent relaxation takes place either to $|0\rangle_e$ (with 40% possibility) or $|±1\rangle_s$ (with 30% possibility respectively). The absorption to $|E_+\rangle$ is only possible if the electron has relaxed to $|0\rangle_e$, and hence the efficiency of the heralding reaches a value of $\approx$ 40%. Overall, the successful rate of transferring the photon to the nuclear spin is approximately 20% (for further detail see Methods).

To verify the transfer process, conditioned on the heralding result, we perform tomography on the transferred state $(1/\sqrt{2}) (|+1\rangle_n - e^{i\phi}|−1\rangle_n)$. This state can be rewritten on the basis of the nuclear dark and bright states as $\cos(\phi/2)|b\rangle_n - i \sin(\phi/2)|d\rangle_n$ for convenience. We map the phase information of the nuclear
spin state onto its population by a RF pulse (see Fig. 1c), that is \( \cos \left( \frac{\phi}{2} \right) |b\rangle - i \sin \left( \frac{\phi}{2} \right) |d\rangle \rightarrow \cos \left( \frac{\phi}{2} \right) |0\rangle - i \sin \left( \frac{\phi}{2} \right) |1\rangle \). By doing so, the phase information is preserved even if there is nuclear spin dephasing during the subsequent illumination of the \( E_{1} \) heralding laser pulse.

By measuring the amplitude of the electron spin Rabi oscillation conditioned on the nuclear spin states \( |0\rangle \) and \( |\pm 1\rangle \), we obtain the populations of nuclear spin state, from which we infer the phase \( \phi \). Fig. 2c,d shows the Rabi oscillations of the electronic spin for \( \phi = 0 \) and the density matrix of the corresponding nuclear spin state. Repeating the experiment with varying \( \phi \) we readout the coherent phase between the photon polarization states via the nuclear spin state with an average fidelity of 98% (see Fig. 3).

So far, we have demonstrated the transfer of the polarization states of a photon to a single nuclear spin. To be functional as a node in quantum networks, the nuclear spin is required to have a long coherence time. The free precession of the nuclear spin is measured via a Ramsey sequence (see Supplementary Information). The coherence dephases on a timescale of \( T_{2m} = 0.31 \pm 0.05 \) s for the electron spin in \( m_s = 0 \) and on a timescale of \( T_{2m} = 12.0 \pm 1.9 \) ms for the electron spin in \( m_s = \pm 1 \). The dephasing arises from the noise generated by the surrounding \( ^{13}\text{C} \) nuclear spin bath. The dependence on the electron spin state can be understood by noting the different spin bath dynamics in these two cases: for \( m_s = 0 \) the bath’s dynamics is dominated by nuclear magnetic dipole-dipole interactions, whereas the electron spin-induced gradient magnetic field dominates in the \( m_s = \pm 1 \) case (see Supplementary Information).

To suppress the dephasing noise we apply a Hahn echo sequence. Fig. 4a,b shows the corresponding signal decay. For the electron spin in the manifold \( m_s = \pm 1 \) the gradient magnetic field-induced energy mismatch strongly suppresses flip-flop processes for the \( ^{13}\text{C} \) nuclear spins that are close to the electron spin. Calculations based on a cluster-correlation expansion model yield a coherence time that exceeds 5 s (see Supplementary Information), which disagrees with the experimentally observed coherence time of \( T_{2m} = 0.53 \pm 0.09 \) s. This large discrepancy.

Figure 2 | Nuclear spin readout. a,b, Deterministic initialization of the \( ^{14}\text{N} \) nuclear spin. a, Energy-level diagram and pulse sequence describing the initialization of the \( ^{14}\text{N} \) nuclear spin. The colours used for the various transitions involved are same as those in Fig. 1. b, Nuclear spin population measurements before (green) and after (red) nuclear initialization. The two different scales shown correspond to the different methods involved in the measurement procedure. To measure the nuclear spin population in its thermal state (green) we use the optically detected magnetic resonance of the electron spin, and to measure the nuclear spin population after initialization (red) we perform a single-shot readout of the electron spin state \( |0\rangle \). Here the MW frequency is 2.879 GHz and RF frequency is 2.76 MHz. Data in the red/green curves are accumulated for 600 and 2,000 rounds, respectively. Error bars indicate the shot-noise errors. c, Nuclear spin state measurement after storing the photonic state \( (1/\sqrt{2})(|+\rangle_{e} + |\rangle_{n}) \). We measure the electron spin Rabi oscillation conditioned on the nuclear spin states \( |b\rangle = (1/\sqrt{2})(|+\rangle_{e} - |\rangle_{n}) \) (green) and \( |d\rangle = (1/\sqrt{2})(|+\rangle_{e} + |\rangle_{n}) \) (red), to obtain the nuclear spin state populations \( P_{b} \) and \( P_{d} \). The transferred phase is then found from the simple equation, \( \phi = \cos^{-1}(P_{d} - P_{b})/(P_{d} + P_{b}) \), which in the present case is zero. Data are accumulated for 350 rounds. d, Reconstructed density matrix of the nuclear spin from data in c. The shaded areas in the plots mark 95% confidence intervals.
indicates that noise sources other than the nuclear spin bath are limiting the spin coherence time in this case. In the \( m_s = 0 \) manifold, however, such a limitation is not observed. A spin coherence time beyond 10 s is measured, which can be explained by treating the nuclear spin bath effects as an effective random classical field (see Supplementary Information). By comparing these two cases, the most probable factors causing the deviation in the \( m_s = \pm 1 \) case would be the fluctuation of the hyperfine interaction between the NV centre electron spin and the \(^{14}\text{N}\) nuclear spin, or the fluctuation of the axial quadruple interaction of the \(^{14}\text{N}\) nuclear spin.

In conclusion, we demonstrate the heralded storage of a photon polarization state in a solid nuclear spin with high fidelity and a long storage time. This protocol can be applied in register arrays made of a single NV centre with several individual controllable nuclear spins nearby\(^\text{19}\) to implement entanglement purification algorithms\(^\text{26}\) making the protocol fault-tolerant. The photon storage at each node can also be further improved by employing dynamical decoupling noise spectroscopy or a \(^{13}\text{C}\)-enriched diamond\(^\text{27}\) to achieve a longer electron spin coherence times as well as using weakly coupled \(^{13}\text{C}\) nuclear spins to yield increased stability of the memory qubits under repetitive excitation.

**Methods**

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

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**Author contributions**
H.K. conceived the original idea, S.Y., Y.W. and P.N. designed the experiment, S.Y. and T.H.T. performed the experiment, S.Y., Y.W. and D.D.B. analysed data and wrote the paper, J.W. supervised the project and all authors commented on the manuscript.

**Additional information**
Supplementary information is available in the online version of the paper. Reprints and permissions information is available online at www.nature.com/reprints. Correspondence and requests for materials should be addressed to S.Y. and J.W.

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

Sample and experimental conditions. We use a [111] oriented NV centre with a low strain (\( \approx 1.2 \) GHz) to suppress strain-induced effects that lower the symmetry of the NV and alter the configuration of the excited states. In the experiment the net magnetic field is set to zero to ensure degeneracy of the ground states. By working at the liquid helium temperature (\( T < 8 \) K) we can resolve optical transitions and enable resonant excitation to perform efficient initialization and projective high-fidelity single-shot readout on the electron spin. The RF \( \pi \) pulse duration in the experiment is on the order of 100 \( \mu s \) whereas those of MW are all below 1,200 ns. A 250 ns waiting time is applied after the A1 laser pulse for relaxation of the electron spin through the singlet state. Otherwise, the pulse delays between subsequent pulses are below 20 ns. The single-shot readout pulse length ranges from 750 ns (Figs 2c and 4c) to 6,000 ns (Figs 2b and 4a).

Spin system initialization. The electron spin can be initialized to \( |0\rangle \), near 100% by pumping with A1 laser\(^{22}\). Then by applying a polarization transfer from the electron spin to the \( ^{14}N \) nuclear spin, we initialize the \( ^{14}N \) nuclear spin to \( |0\rangle \) with a fidelity higher than 98% (Fig. 2a). We achieve such a high fidelity because the initialization via the excited state \( |4\rangle \) is barely affected by the hyperfine interaction. The procedure is as follows: we start from state \( |0\rangle \) of the electron spin and a thermal state of the \( ^{14}N \) nuclear spin that is characterized by identical populations in states \( |0\rangle \) and \( |1\rangle \). Populations in states \( |0\rangle \) and \( |1\rangle \) are then transferred to state \( |+1\rangle \) and \( |-1\rangle \) through a nuclear spin-controlled NOT gate followed by an electron spin-controlled NOT gate on the nuclear spin. After reinitializing the electron spin into state \( |0\rangle \), the final state is \( |0\rangle |0\rangle \). Unlike existing schemes, our approach here neither relies on level anticrossings\(^{28}\) nor requires post-selection measurements\(^{22}\).

Bell state preparation. We start from state \( |0\rangle |0\rangle \). A nuclear \( \pi \) pulse (nuclear spin-controlled NOT gate) drives this state to \( (1/\sqrt{2})|0\rangle |+\rangle + |+\rangle |0\rangle \). In the absence of a magnetic field, transitions from \( |\pm 1\rangle |\pm 1\rangle \) to \( |\pm 1\rangle |\pm 1\rangle \) are degenerate. An electron spin \( \pi \) pulse (electron spin-controlled NOT gate), which flips state \( |0\rangle |+\rangle \) to \( |+\rangle |0\rangle \) and state \( |0\rangle |-\rangle \) to \( |-\rangle |0\rangle \) simultaneously, creates the final state \( (1/\sqrt{2})|+\rangle |0\rangle + |0\rangle |+\rangle \).

Photon absorption rate. The photon absorption rate is proportional to \( (\lambda_0^2/\pi)(\Gamma_0/\Gamma) \), where \( \lambda_0 \) is the photon wavelength in diamond, \( \pi \) is Euler’s number and \( \Gamma_0/\Gamma \) is the ratio of the zero-phonon-line transition rate to the total transition rate (the Debye–Waller factor of the NV centre in diamond), which is around 4%. So the photon absorption rate is proportional to \( (4\%/\pi)(\text{NA}^2/\text{NA}^2) \), which is on the order of 0.1%. \( \text{NA} \) is numerical aperture of objective and \( \text{NA} \) is the refraction index of diamond. In principle, an optical cavity can boost this absorption rate by increasing the number of photons passing through the NV centre, and also boost the \( \Gamma_0/\Gamma \) ratio by Purcell enhancement.

Optical writing of nuclear spins. The electronic spin prepared in the superposition state \( (1/\sqrt{2})|+\rangle + e^{i\phi}|-\rangle \) will deterministically absorb the incoming photon only if it is in the state \( |\psi\rangle = (1/\sqrt{2})|\sigma^+\rangle + e^{i\phi}|\sigma^-\rangle \) and vice-versa. Owing to the dark state feature in a lambda system, an incoming photon in the state orthogonal to the electron spin state does not get absorbed. Thus the excitation becomes probabilistic due to this intrinsic dark state feature. By rewriting the electron–nuclear Bell state in the photon basis as

\[
|\psi^\prime\rangle = (1/\sqrt{2})(|\psi\rangle |\psi\rangle + |\psi^\prime\rangle |\psi^\prime\rangle),
\]

where \( |\psi\rangle = (1/\sqrt{2})|+\rangle + e^{i\phi}|-\rangle \) and \( |\psi^\prime\rangle = (1/\sqrt{2})|+\rangle + e^{i\phi}|-\rangle \) it can be seen that absorption takes place with only a 50% probability. When the photon is absorbed one can see from the above equation that the nuclear spin will be projected onto \( |\psi\rangle \). As there is only a 50% probability of this happening, one needs to herald either on the emitted photons or on the electronic spin state to confirm the storage process.

Spin coherence time. For the single-spin system studied here there is no inhomogeneous broadening due to averaging over a spatial ensemble. Instead, temporal averaging is needed to collect sufficient statistics to characterize the spin dynamics. This averaging can also lead to fast apparent dephasing that can be reversed using a Hahn echo technique\(^{29}\).

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