Robust quantum-network memory based on spin qubits in isotopically engineered diamond

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Quantum networks can enable quantum communication and modular quantum computation. A powerful approach is to use multi-qubit nodes that provide quantum memory and computational power. Nuclear spins associated with defects in diamond are promising qubits for this role. However, dephasing during optical entanglement distribution hinders scaling to larger systems. Here, we show that a 13C-spin quantum memory in isotopically engineered diamond is robust to the optical link operation of a nitrogen-vacancy centre. The memory lifetime is improved by two orders-of-magnitude upon the state-of-the-art, surpassing reported times for entanglement distribution. Additionally, we demonstrate that the nuclear-spin state can survive ionisation and recapture of the nitrogen-vacancy electron. Finally, we use simulations to show that combining this memory with previously demonstrated entanglement links and gates can enable key network primitives, such as deterministic non-local two-qubit gates, paving the way for test-bed quantum networks capable of investigating complex algorithms and error correction.

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INTRODUCTION

Quantum networks have the potential to enable a wealth of applications that go beyond classical technologies, including secure communication, quantum sensor networks, and distributed quantum computation1–4. Such a network might consist of nodes with stationary ‘communication’ qubits that are connected together by entanglement through photonic channels (Fig. 1a). Each node ideally contains multiple additional ‘data’ qubits that can be used to store and process quantum states. Universal operations over the network can then be performed by repeatedly distributing entangled states and subsequently consuming them2,3.

Large-scale networks and universal quantum computation become possible if imperfections can be overcome through entanglement distillation and quantum error correction4,5. Besides high-fidelity operations, this requires the faithful storage of quantum states while new entangled states are repeatedly distributed over the network2–5. This capability is captured by the (active) ‘link efficiency’, \( \eta_{\text{link}} = r_{\text{ent}}/r_{\text{dec}} \), given by the ratio of the inter-node entanglement generation rate \( r_{\text{ent}} \) and the decoherence rate \( r_{\text{dec}} \) of the data qubits during network operation (see Supplementary Note 1 for further discussion). Without error correction, \( \eta_{\text{link}} \) sets the available number of cycles of entanglement distribution, and thereby the depth of protocols and computations that can be performed effectively. While quantum error correction can ultimately increase achievable circuit depths, this will require \( \eta_{\text{link}} \gg 1 \)2–4,6.

Various systems have demonstrated basic building blocks for optical quantum networks7–17. The nitrogen-vacancy (NV) centre in diamond is a promising platform because it combines a spin–photon interface for heralded remote entanglement18–20, with access to multiple 13C nuclear-spin data qubits that can store quantum states for long times21–27. Entanglement generation rates \( r_{\text{ent}} \) larger than the idle qubit decoherence rate have been shown, enabling a single cycle of entanglement delivery deterministically on a clock cycle7. As a first step towards exploiting additional computational power in the nodes, experiments with up to two qubits per node demonstrated two-cycle network protocols such as entanglement distillation28 and entanglement swapping in a three-node network29,30. However, in all these experiments \( \eta_{\text{link}} \ll 1 \). The resulting network operation is inherently probabilistic—if entanglement generation cycles do not succeed early enough, all previously established quantum states stored in the network memory are lost—hindering the scaling to larger systems operating over many cycles. Realising large \( \eta_{\text{link}} \) will require higher entanglement rates \( r_{\text{ent}} \) through efficient spin-photon interfaces31–34 and/or reduced dephasing of the nuclear spin qubits during the network operation \( r_{\text{dec}} \)32,33.

In this work, we focus on the latter challenge. We demonstrate that 13C spin qubits in isotopically engineered diamond with a reduced 13C concentration provide robust data qubits for quantum networks. We develop control and single-shot readout of an individual 13C spin that is weakly coupled to a single NV centre. We then show that an arbitrary quantum state can be stored in this data qubit for over \( 10^5 \) repetitions of a remote-entanglement sequence. For current entanglement link success rates, this would imply \( \eta_{\text{link}} \approx 10 \). We show through numerical network simulations that such a high \( \eta_{\text{link}} \) value can enable a next generation of network protocols, including deterministic remote two-qubit gates and the distillation of entangled states over a four-node network, which provides a primitive for surface-code quantum error correction (Fig. 1a)34,46. Finally, we identify ionisation of the NV centre to the neutral charge state as a limiting mechanism and show that the data qubit can be protected through fast controlled resetting of the NV charge state. When combined with recent progress with optical cavities towards enhanced entanglement rates32,34 and high-fidelity quantum gates32,34,36, these results indicate that nuclear spins in isotopically engineered samples provide a promising test-bed for quantum networks.

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that the $^{13}$C-spin dynamics depend on the NV electron-spin state; however, induced additional dephasing of the nuclear-spin Larmor frequency, where $\gamma_i$ is the $^{13}$C gyromagnetic ratio and $B_0$ is the external magnetic field along the NV axis (here $B_0 = 47$ G), $I_{\parallel}$ is the spin-1/2 projection of the electron spin. In the NV$^-$ charge state that is used for network operation, $m_i \in \{-1, 0, 1\}$, from which we define a qubit in the $\{-1, 0\}$ basis ($\{\pm 1\}$). Additionally, however, stochastic ionisation events can convert NV$^-$ to the NV$^0$ state with $m_i \in \{-1/2, 1/2\}$. Note that fast orbital relaxation in the NV$^0$ ground state rapidly dephases the spin state, making it unsuitable as a qubit.

As seen from Eq. 1, the $^{13}$C spin undergoes different precession dependent on the NV charge- and spin-state. This conditional precession enables complete control over the $^{13}$C spins by controlled inversions of the NV electron spin. Uncontrolled electron-spin dynamics, however, induce additional dephasing of the $^{13}$C spin, which sets a limit on the achievable link efficiency $\eta_{\text{link}}$. Our approach to creating robust memory is to reduce the coupling between the NV electron spin and the $^{13}$C data qubits by reducing the $^{13}$C concentration. While this approach results in slower gate speeds, these are not the rate-limiting step in current network experiments. Note that, in the case of $^{13}$C-spin bath-limited decoherence, the coherence times increase proportionally with the reduction in coupling strength, such that no intrinsic reduction in gate fidelity is expected.

Our experiments are performed on a single NV centre in a type-IIa isotopically-purified diamond (targeted $^{13}$C concentration of 0.01%) at a temperature of 4 K. The hardware setup and NV centre properties are described in the Methods section.

Dynamical decoupling (DD) spectroscopy with the NV centre reveals coupling to an isolated $^{13}$C spin, along with the wider spin bath (Fig. 1c). We characterise the electron-nuclear hyperfine components parallel (perpendicular) to the NV axis to be $A_{\parallel} = 2\pi \cdot 80(1)$ Hz and $A_{\perp} = 2\pi \cdot 271(4)$ Hz. The resonance at 44.794 $\mu$s (red) corresponds to the $^{13}$C spin bath. Solid lines correspond to a theoretical model (Supplementary Note 2). Intrinsic decoherence timescales of the nuclear spin for different electron states. Solid lines are fits (see Methods). Dashed lines is a guide to the eye.

RESULTS

System overview

We consider quantum-network nodes consisting of a single NV centre coupled to multiple $^{13}$C nuclear spins (Fig. 1a). The optically active NV electron spin acts as a communication qubit and the $^{13}$C nuclear spins are additional data qubits that also function as quantum memory to store states while new entanglement links. A key feature of this system is that the $^{13}$C-spin dynamics depend on the NV electron-spin state (Fig. 1b). This is captured by the Hamiltonian for a single $^{13}$C spin:

$$H = \omega_i I_z + A_i m_i I_z + A_{\perp} m_z I_z. \tag{1}$$

Here, we have made the secular approximation, $\omega_i = \gamma_i B_0$ is the nuclear-spin Larmor frequency, where $\gamma_i$ is the $^{13}$C gyromagnetic ratio and $B_0$ is the external magnetic field along the NV axis (here $B_0 = 47$ G), $I_{\parallel}$ are the nuclear spin-1/2 operators, while $m_i$ is the spin-$z$ projection of the electron spin. In the NV$^-$ charge state that is used for network operation, $m_i \in \{-1, 0, 1\}, \{\pm 1\}$. Note that fast orbital relaxation in the NV$^0$ ground state rapidly dephases the spin state, making it unsuitable as a qubit.

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We realise universal control over the electron-$^{13}$C two-qubit system by microwave (MW) and radio-frequency (RF) single-qubit gates, and a DD-based electron-nuclear two-qubit gate. Furthermore, we develop repetitive readout of the nuclear spin to improve the single-shot readout fidelity (see Supplementary Note 2) giving a maximum state preparation and measurement (SPAM) fidelity of 91(1)%. For the measurements reported here we focus on the system dynamics rather than maximising the fidelities, and thus use a faster initialisation procedure to optimise the signal-to-noise ratio, with lower SPAM fidelity of 79.4(9%). The fidelities are predominantly limited by electron-spin decoherence during two-qubit gates, likely arising from electron-nuclear hyperfine coupling.
from the high concentration of P1 centre impurities in this specific device (~75 ppb\textsuperscript{31}). Multi-qubit registers with comparable two-qubit gate fidelities to those achieved in natural-abundance \textsuperscript{13}C devices (~99\%\textsuperscript{23}) are expected to be feasible with reduced impurity concentration in future diamond growth.

**Memory robustness while idling**

We first investigate the intrinsic decoherence processes of the \textsuperscript{13}C nuclear spin, i.e. when the electron spin is idle. The electron spin-state affects spin-bath dynamics, as flip–flop interactions between nuclear spins are suppressed when the hyperfine interaction is present (the ‘frozen core’ effect)\textsuperscript{23,25,26}. Therefore, we separately characterise the decoherence timescales for the NV electron in the \{1\} or \{0\} states.

Figure 1d summarises the measured timescales. The observation of vastly longer \(T_{1,e-1}\) (\(\gg 5\) s) than \(T_{1,e-0}\) (\(= 2.8(2)\) s) indicates that the spin relaxes primarily through flip–flop interactions with other \textsuperscript{13}C spins. The measured \(T_{1,e-0}\) is \(\sim 30–100\) times longer than typical values for natural-abundance samples, consistent with the expectation of a linear dependence on isotopic concentration\textsuperscript{39,42}.

The measured dephasing times are \(T_{2,e-1}\) = 0.38(1) s and \(T_{2,e-0}\) = 0.42(2) s. We first isolate the timescale associated with couplings to the nuclear-spin bath by performing a spin-echo experiment on all \textsuperscript{13}C spins, decoupling the \textsuperscript{13}C bath from other processes but leaving their mutual interactions unperturbed. We find a characteristic timescale, \(T_{2,\text{bath}}\) = 0.66(3) s (see Supplementary Note 3). The contribution from P1 impurities can similarly be estimated using the NV electron dephasing time during double electron-electron resonance measurements with the P1 impurities, \(T_{2,\text{P1-bath}}\). From the respective gyromagnetic ratios, \(\gamma_{C}\) and \(\gamma_{E}\), we infer \(T_{2,\text{P1-bath}}\) = 0.61 s. Combining these processes gives an estimate for \(T_{2,\text{e-0}}\) = 0.45 s, close to the measured values.

We finally consider the application of spin echoes which mitigate quasi-static noise. The single-echo \(T_{2,1}\) = 1.11(8) s and eight-echo \(T_{2,8}\) = 1.62(9) s are likely limited by the \(m_{I} = 0\) nuclear \(T_{1}\). The observation of some underlying structure in these measurements is likely due to the dynamics of other proximal \textsuperscript{13}C spins\textsuperscript{20}. In contrast, for the electron \{1\} state, the nuclear-spin frozen core has previously been shown to significantly enhance \textsuperscript{13}C spin-echo times\textsuperscript{25}. While \(T_{2,1}\) = 1.82(6) s and \(T_{2,8}\) = 2.91(8) s do exceed \(T_{1}\), respectively, they are shorter than would be predicted from the \textsuperscript{13}C concentration alone (~1 min). We ascribe this to the presence of the P1 bath, which exhibits faster dynamics which are not well mitigated by echo pulses. This decoherence might be further mitigated by strong driving of the P1 bath\textsuperscript{51,54} or by using higher purity diamonds.

**Memory robustness during the remote-entanglement sequence**

Armed with a characterisation of the intrinsic decoherence processes, we now turn to the remote-entanglement sequence shown in Fig. 2a, b. This entangling primitive is compatible with single-photon schemes used in recent NV network experiments\textsuperscript{29,55}. In this work, we implement the protocol on a single network node to investigate its effect on the \textsuperscript{13}C data qubit.

As discussed in the previous sections, imperfect knowledge of the electron-spin state leads to spurious phases acquired by the nuclear spin. Within each entangling attempt, there are a number of processes which can lead to such dephasing: the stochastic nature of the electronic spin-reset time, infidelities in the initialised electron-spin state, excited state spin-flips after the optical \(n\)-pulse, and MW pulse errors. Reducing the electron-nuclear coupling strength enhances the robustness to such events.

We implement the entangling protocol experimentally. For simplicity, we omit the optical \(n\)-pulses, but note that the small associated electron spin-flip probability is expected to play a negligible effect on the \textsuperscript{13}C decoherence\textsuperscript{22,23}. As a trade-off between the optical reset rate and stochastic ionisation to NV\textsuperscript{56,57}, spin reset is performed using 30 nW of optical power.
for 5 μs, resulting in an initialisation fidelity $\geq 98\%$ (Supplementary Note 4). For the MW pulse used to create the electron superposition state, we implement a weak multi-tone driving pulse ($\Omega_{\text{drive}} \sim 93$ KHz), see Supplementary Note 5. This comes at the expense of a time overhead, but mitigates heating. We set the MW rotation angle to n/2, for which dephasing due to the entangling primitive is expected to be maximal for the given sequence (Supplementary Note 7). Finally, we focus on networks for distributed quantum computation, rather than long-distance communication, and assume that the distance between network nodes is small ($< 100$ m). Thus, any decision logic can be completed within 1 μs. With these choices, the total duration of each primitive is 9 μs.

We prepare the nuclear spin in each of the six cardinal states ($\Psi_i \in \{X, i=1\cdots 6\}$) by applying N repetitions of the primitive, and measure the expectation value in the associated eigenbasis, $(\sigma_j) \equiv \text{Tr}(\rho \sigma_j)$, where $\sigma_j$ are nuclear-spin Pauli operators, $j \in \{x, y, z\}$ (Fig. 2c). We interleave one cycle of XY8 decoupling pulses on the nuclear spin to mitigate dephasing. Without any post-selection of the data we find that the data qubit can preserve an arbitrary quantum state for $N_{1/e} = 1.33(4) \times 10^5$ entangling primitives (1.20(4) s of continuous entanglement attempts).

We now consider the effects of ionisation and spectral diffusion on the data. After each experimental run, we perform two-laser probe measurements ($E'$, $E_c$ transitions, denoted charge-resonance (CR) checks (see Supplementary Note 8))13. The number of photons detected in this check is used to verify that the NV remained in NV$^-$ and on resonance throughout the experiment. By varying the post-selection threshold for the CR check, we can reject measurements in which the NV ionised or underwent spectral diffusion with increasing confidence. We find a further improvement in the data qubit lifetime when omitting these cases, showing that they are currently a significant limitation (Fig. 2c). After accounting for ionisation and spectral diffusion, we fit a decay time $N_{1/e} = 2.07(8) \times 10^5$. Considering spin superposition-and eigen-states separately, we find $N_{1/e,x} = 1.90(8) \times 10^5 [1.71(7) s]$ and $N_{1/e,y} = 2.6(2) \times 10^5$ attempts [2.4(2) s].

In Supplementary Note 6, we additionally present results for the decay of a superposition state when using strong microwave pulses ($\Omega_{\text{drive}} \sim 27$ KHz), so that the sequence is shorter (primary duration 6.3 μs). We find a further improved corrected decay constant of $N_{1/e,x} = 4(1) \times 10^5$ attempts [2.6(6) s]. However, in this case, we cannot rule out that heating due to the strong pulses changes the spin-reset dynamics, although the observation of similar ionisation statistics suggests that this is not the case. Such heating can be addressed by improved device engineering in future work.

For both primitive durations (6.3 and 9 μs), the measured decoherence timescales are comparable to those arising from intrinsic spin-bath dynamics. This suggests that the entanglement sequence only weakly increases the dephasing of the $^{13}$C spin.

We use Monte Carlo simulations to model the nuclear-spin dephasing induced by the entanglement attempts, taking into account all known control errors (see Supplementary Note 7) but neglecting the intrinsic decoherence rates$^{22}$. We find that even using pessimistic parameters, an $A_j = 2n \cdot 80$ Hz coupled nuclear spin is predicted to retain a fidelity of 77.6(4)% with respect to an initial superposition state after $10^6$ entangling attempts. This finding is consistent with the interpretation of spin-bath limited decoherence in the present experiment.

Our simulations also show that the dephasing infidelity after $10^6$ entangling attempts can be reduced below 1% by using optimised primitives incorporating an additional electron-spin echo. Further improvement of the performance can be realised by reducing the entangling primitive duration, such that more attempts can be performed within the intrinsic decoherence timescales. These intrinsic timescales may themselves be extended by optimised decoupling schemes$^{20}$ as well as using samples with lower nitrogen defect concentrations. It is clear, however, that ionisation currently is a limiting factor. We next probe the nuclear-spin dynamics after such ionisation events to understand if they can be mitigated.

### Mitigating ionisation

The previous section indicates that ionisation of the NV centre limits achievable memory performance. In the above experiments, and in previous work$^{22,29,35}$, an ionisation event (NV$^- \rightarrow$ NV$^0$) at some point in the remote-entanglement sequence causes complete dephasing of the $^{13}$C spins. This is because such events occur stochastically, the subsequent electron-spin dynamics are unknown, and, as discussed above (see Eq. 1 and Fig. 1b), each electron-spin state causes different $^{13}$C-spin evolution.

While ionisation rates can be reduced by decreasing optical powers for the spin-reset process, this results in longer sequence times. Additionally, the optical $n$-pulses cannot be removed, and therefore it is challenging to completely prevent ionisation. An alternative approach is to instead develop techniques which make the data qubit robust to this process.

In order to study the effect of charge-state switching, we implement resonant (zero-phonon line, ZPL) optical pulses that efficiently induce ionisation (NV$^- \rightarrow$ NV$^0$, 1 ms, 500 nW) and recharging (NV$^0 \rightarrow$ NV$^-$, 1 ms, 500 nW) with minimal disturbance to the local charge environment. Combining these pulses with CR checks, we can use heralding and post-selection to perform verified charge-state switching from NV$^- \rightarrow$ NV$^0 \rightarrow$ NV$^-$ (see Supplementary Note 8).

We first combine the verified charge-state switching protocol with nuclear-spin control to investigate the properties of the $^{13}$C spin for the NV$^0$ state. After preparing NV$^-$ and initialising the nuclear spin in the state $|1\rangle$, we apply the ionisation pulse, and proceed with the experiment if a subsequent CR check heralds preparation in NV$^0$. We then apply a nuclear-spin RF $n$-pulse with a Rabi frequency of 5.4(2) Hz, for which we sweep the carrier frequency f. After applying the recharge pulse, we read-out the nuclear spin in the Z-basis and post-select the data on finding the NV in NV$^-$. As shown in Fig. 3a, we observe a single nuclear-spin resonance at a frequency of 50229.8(1) Hz, which matches the $^{13}$C Larmor frequency. The observation of a single transition, rather than two transitions at $f = \omega_c \pm A_j/2$, suggest a fast averaging over the two NV$^0$ electron-spin states, akin to motional narrowing$^{21,41}$.

To characterise the nuclear-spin dephasing, we prepare the nuclear spin in a superposition state, and let it evolve freely while switching to NV$^0$ for a variable time. The inset of Fig. 3b reveals coherent oscillations, corresponding to precession of the nuclear spin at a frequency of 50231(1) Hz. The nuclear-spin coherence $(\langle X \rangle^2 + \langle Y \rangle^2)^{1/2}$ decays as $T_2^*/NV^0 = 57(3)$ ms. This dephasing time is significantly shorter than for NV$^-$ ($T_2^*/NV^- = 0.4$ s, Fig. 1d), indicating that the NV$^0$ state introduces an additional dephasing mechanism. The fitted decay exponent of $n = 1.2(1)$ (see Methods) matches the value of $n = 1$ expected for the motional-narrowing-like regime$^{21}$.

The observation of a single nuclear-spin frequency and a decreased $T_2^*$ are consistent with a rapidly fluctuating NV$^0$ electron-spin state. In this regime, the dephasing time scales as $T_2^* = 8/(A_j^2 \cdot T_{11}^*/NV^0)^{21,41}$. Thus, we can extract $T_{11}^*/NV^0 = 570(30)$ μs. This value deviates from another recent cryogenic measurement ($T_{11}^*/NV^0 = 1.51(1.5)$)37. It is likely that this discrepancy is either due to the much higher magnetic field ($B_z = 1850$ G) or absence of a P1 bath in that work.

The observed nuclear-spin dephasing time in NV$^0$ is much longer than the time needed to recharge to NV$^-$. We now show that this enables the protection of the data qubit from ionisation events. We prepare the $^{13}$C in each of the six cardinal states, and compare the expectation values obtained when applying the
The nuclear-spin coherence in NV0. Inset: Ramsey fringes measured during the first 15 ms of free evolution (artificial detuning of 80 Hz with respect to 50230 Hz). Solid lines are fits (see Methods).

**Fig. 3** 13C dynamics during ionisation and recharging. a Nuclear magnetic resonance spectroscopy after heralded preparation of NV0. Solid line is a numerical evaluation of the optical Bloch equations for this system (see Methods). b Free induction decay of the nuclear-spin coherence in NV0. Inset: Ramsey fringes measured during the first 15 ms of free evolution (artificial detuning of 80 Hz with respect to 50230 Hz). Solid lines are fits (see Methods). c Protection of an arbitrary data qubit state under NV charge cycling. Orange (red) bars correspond to measurements with a charge-state cycle (idling in NV0) between nuclear-spin initialisation and readout. The data is post-selected on finding the NV in the negative charge state at the end of the measurement (see text).

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Numerical analysis of quantum-network protocols

Finally, we investigate the projected performance of a number of quantum-network protocols when combining the improved memory lifetime demonstrated in this work with previously demonstrated entanglement links and gate operations (in other devices). We focus on non-local two-qubit operations and the creation of distributed four-qubit GHZ states, a key building block for the distributed surface code3,4 (Fig. 1a).

These investigations are based upon density-matrix simulations of noisy quantum circuits. We use the following set of parameters to model the dominant error sources, which are either the measured state-of-the-art (on a variety of devices) or near-term predictions (denoted by *): two-qubit gates (1% infidelity23), NV optical measurement (1% infidelity20), and networked (NV–NV) Bell-pair creation (10% infidelity20). We account for the finite duration associated with each operation, including probabilistic NV–NV remote-entanglement (duration 6 μs, success probability
Second, we have the simplest known approach which fuses three distributed Bell pairs which approaches the limit of a single step of entanglement distillation. Finally, we have the protocol introduced by Nickerson et al. which extends the Plain protocol by one additional Bell-pair distillation, and, in principle, can lead to high-quality GHZ states during entangling sequences or idling.

The differences in performance between the schemes with $N_{1/e} = 2 \times 10^5$ give insights into the usefulness of entanglement distillation for the considered parameter regime. First, between the Plain and Modicum protocols, we see that the additional distillation step does improve the fidelity, from $F = 0.62$ to $F = 0.75$. However, this fidelity improvement is limited due to the need for an additional SWAP operation (three two-qubit gates). Furthermore, there is a slight increase in the average duration of the protocol, predominantly due to the probabilistic nature of the distillation step. For the Expedient protocol, we see that it takes significantly longer to successfully generate one GHZ state, and achieves lower fidelities. Additional rounds of entanglement distillation are only beneficial if the fidelity improvements (from distilling imperfect states) exceed the fidelity losses from decoherence of idle qubits. A further increase in $\eta_{\text{link}}$ is required to make use of such protocols.

Together, these results show that the lifetime of the robust quantum-network memory demonstrated in this work is sufficient to demonstrate key distributed quantum computation protocols, such as deterministic two-qubit gates and basic distillation schemes, across optical quantum networks.
DISCUSSION
We have demonstrated a robust quantum-network memory based upon a single 13C-spin qubit in isotopically engineered diamond. Compared with previous work22,29, the data qubit lifetime during network operation is improved by two orders-of-magnitude. Critically, the data qubit now decoheres more slowly than state-of-the-art entanglement rates between NV centre network nodes7,29. Using numerical simulations, we show that the corresponding parameter regime—with a projected $\eta_{\text{link}} \sim 10$—enables a range of protocols for optical quantum networks, including deterministic two-qubit logic and creation of genuinely-entangled four-qubit GHZ states. Additionally, such robust memories would greatly speed up recently demonstrated protocols such as entanglement distillation and entanglement swapping18,29.

On the path towards reaching the fault-tolerance threshold for large-scale distributed quantum information processing, our simulations show that further improvements in $\eta_{\text{link}}$ are needed. The 13C decoherence rate $T_2^{\text{dec}}$ is currently limited by spurious ionisation of the NV centre to NV$^0$. Our results show that arbitrary states can be protected while cycling the charge-state back and forth with minimal loss of fidelity, suggesting that such events can be mitigated. Further improvements in $T_2^{\text{dec}}$ are possible by further improving the intrinsic coherence times, and reducing the time needed for the entanglement sequence. Additionally, an improvement of the optical entanglement success probability by a factor $\sim 100$ is feasible using Fabry-Perot micro-cavities32,34, which would also greatly improve the overall operation speed, as is required for advancing beyond prototype networks and towards technological applications. Together such improvements would yield $\eta_{\text{link}} > 1000$, which is anticipated to be sufficient to realise large-scale error corrected quantum networks5.

METHODS
Sample and hardware setup
Our experiments are performed on a type-IIa (100) diamond substrate, grown via chemical vapour deposition using isotopically-purified methane gas to reduce the 13C nuclear-spin concentration (targeted 0.01% 13C, Element Six). We address a single NV centre using a cryogenic (4 K) confocal microscope. A solid immersion lens and anti-reflection coating are fabricated to increase optical collection efficiency18,62,63. An external magnetic field is applied along the NV symmetry axis using three orthogonal permanent neodymium magnets mounted on linear actuators (Newport UT5100PP). We measure the field vector to be $(B_x, B_y, B_z) = (0.3(1), 0.06(8), 46.801(1))$ G by spectroscopy of the P1 bath transition frequencies51.

Resonant optical excitation of NV$^-$ at 637 nm (red, Toptica DLPro and New Focus TLP-6704-P) realises high-fidelity spin initialisation $(E_{1,2}$ transitions, $= E')$ and single-shot readout $(E_z$ transition)18. We measure readout fidelities of 68.5(2)% for the bright state $(m_z = 0, = |0\rangle)$ and 99.3(3)% for the dark state $(m_z = -1, = |1\rangle)$, giving $F_{\text{SSRO}} = 0.839(3)$. We employ 515 nm (green, Cobolt ML2) excitation to prepare the NV in NV$^-$ and on resonance with the 637 nm lasers (see Supplementary Note 8)19. Finally, for experiments involving the neutral charge state, NV$^0$, we use resonant 575 nm (yellow, Toptica DL-SHG Pro) light to realise fast recharging to NV$^-$. Through direct current modulation or cascaded acousto-optical modulators, we achieve on/off ratios $>100$ dB for all lasers.

Microwave (MW) driving via a lithographically defined gold stripline enables coherent control between $|0\rangle \leftrightarrow |1\rangle$. Pulses are applied using Hermite envelopes with maximum $\Omega_{\text{Rabi}} \sim 27$ MHz, aside from within the entangling primitive, where a multi-tone driving scheme is employed to mitigate heating (Supplementary Note 5). Using a fast microwave switch (TriQuint TG52355-SM, 40 dB) to suppress electronic noise, we measure long electron-spin relaxation $(T_1 > 30$ s), dephasing $(T_2^\ast = 94(2) \mu$s) and coherence $(T_2 < 0.992(4)$ ms) times, the latter of which is limited by the electron-spin bath formed by the P1 centres (75 ppb) which can be reduced in future diamond growth.

Magnetic field stabilisation
The presence of a number of 6–9 T magnetic field systems in adjacent laboratories leads to slow magnetic field drifts (measured to be $\sim 200$ mG peak-to-peak across all presented measurements). These drifts are first mitigated using a feedback-loop. We measure the electron-spin resonance frequency approximately every 10 min, and compensate any deviation to within 3 kHz (~1 mG) of the set-point by moving the magnets. This comes at the cost of slight magnetic field misalignment. If this misalignment becomes too large, or nearby magnetic field systems are being swept too quickly, we observe degradation of the nuclear-spin operations. To remove such effects from our results, during 13C-related data-taking we interleave separate reference measurements of the nuclear-spin expectation value after simple state-preparation and measurement. These reference measurements do not contribute to the presented data-sets, but are used to discard measurement runs in the case that the reference measurement falls below 75% of the calibrated value.

Data analysis
Throughout this text, we do not correct the nuclear-spin data for SPAM errors.

The intrinsic decoherence timescales of the 13C data qubit, as presented in Fig. 1d, are fitted as described in Table 1.

| Metric          | $\tau_0$ (ms) | $\tau_1$ (ms) | $\tau_2$ (ms) | $\tau_3$ (ms) | $\tau_4$ (ms) | $\tau_5$ (ms) | $\tau_6$ (ms) | $\tau_7$ (ms) |
|-----------------|--------------|--------------|--------------|--------------|--------------|--------------|--------------|--------------|
| $T_2^{\text{dec}}$ | 0.55 (2)     | 0.46 (1)     | 0.51 (1)     | 0.52 (2)     | 0.58 (1)     | 0.60 (2)     | 0.43 (2)     | 0.49 (2)     |
| $T_2^{\text{rel}}$ | 0.38 (1)     | 0.42 (2)     | 1.82 (6)     | 1.11 (8)     | 2.91 (8)     | 1.62 (9)     | 2.8 (2)      | 2.8 (2)      |

The corresponding fraction of rejected data (lower panel) is fit as described in Table 2.

The 13C nuclear magnetic resonance spectrum for the NV$^0$ state (Fig. 3a) is modelled by numerically evaluating the optical Bloch equations for the system, using the measured Rabi frequency, $\Omega_{\text{Rabi}}$.

Table 1. Fit parameters for Fig. 1d.

| Metric          | $A$     | $T$ (s) | $n$   |
|-----------------|---------|--------|------|
| $T_2^{\text{dec}}$ | 0.55 (2) | 0.38 (1) | 1.6 (1) |
| $T_2^{\text{rel}}$ | 0.46 (1) | 0.42 (2) | 2.0 (2) |

Table 2. Fit parameters for Fig. 2c, upper panel.

| CR threshold | $A$     | $N_{1/2} \times 10^5$ | $n$   |
|--------------|---------|----------------------|------|
| 0            | 0.49 (1) | 1.33 (4)              | 1.00 (5) |
| 1            | 0.49 (1) | 1.80 (6)              | 1.09 (8) |
| 5            | 0.49 (1) | 2.07 (8)              | 1.09 (8) |
DATA AVAILABILITY
The data that support the findings of this study can be found on the 4TU repository: https://doi.org/10.4121/16887658.v3.

CODE AVAILABILITY
The code implementation for the noisy quantum-network simulations (Fig. 4c, d) can be found on the 4TU repository: https://doi.org/10.4121/16887658.v3.

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AUTHOR CONTRIBUTIONS

C.E.B., S.B., R.H., and T.H.T. designed the experiments. C.E.B. performed the experiments. C.E.B. and T.H.T. analysed the data. C.E.B., M.J.D., S.J.H.L., and H.P.B. prepared the experimental setup. S.W.d.B., P.F.W.M. and D.E. performed the network experiments. C.E.B. and T.H.T. wrote the paper with input from all authors. T.H.T. supervised the project.

COMPETING INTERESTS

C.E.B., S.B., R.H., and T.H.T. are co-inventors for patent N2029318, “Method and system for operating a quantum-network node”, submitted by the Delft University of Technology, 4th October 2021 at the Netherlands Patent Office. The patent focuses on techniques required to operate a quantum-network node which incorporates memory qubits which are robust to network operation and unwanted charge-state conversion. The authors declare that there are no other competing interests.

ADDITIONAL INFORMATION

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