Coherence time of over a second in a telecom-compatible quantum memory storage material

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Quantum memories for light will be essential elements in future long-range quantum communication networks. These memories operate by reversibly mapping the quantum state of light onto the quantum transitions of a material system. For networks, the quantum coherence times of these transitions must be long compared to the network transmission times, approximately 100 ms for a global communication network. Due to a lack of a suitable storage material, a quantum memory that operates in the 1,550 nm optical fibre communication band with a storage time greater than 1 μs has not been demonstrated. Here we describe the spin dynamics of 167Er3+:Y2SiO5 in a high magnetic field and demonstrate that this material has the characteristics for a practical quantum memory in the 1,550 nm communication band. We observe a hyperfine coherence time of 1.3 s. We also demonstrate efficient spin pumping of the entire ensemble into a single hyperfine state, a requirement for broadband spin-wave storage. With an absorption of 70 dB cm−1 at 1,538 nm and Δ transitions enabling spin-wave storage, this material is the first candidate identified for an efficient, broadband quantum memory at telecommunication wavelengths.

Any future globally deployed quantum communication network will require nodes connected by optical fibre. To minimize transmission loss and maintain high data throughput, all elements of such a network, particularly quantum repeater nodes, should transmit in one of the low-loss telecom bands for optical fibre at 1,310 and 1,550 nm. In its simplest implementation, a quantum repeater relies on an efficient, long-lived quantum memory.

Developing such a memory has proven very challenging. None of the proposed systems that operate directly in the low-loss telecom band have the potential for long-term storage. For this reason, more complex ways of interfacing these candidate quantum memories with telecom are being investigated, including frequency conversion or non-degenerate photon pairs. One of the most promising candidate memory systems is rare earth ions in solids. The potential for developing practical memories in these systems has been highlighted through a series of recent demonstrations using non-Kramers ions (ions with an even number of electrons). Crystals doped with either praseodymium or europium have demonstrated long storage on the optical transitions. However, the hyperfine lifetime is still very short (100 ms), preventing long-term storage. In comparison, the hyperfine lifetimes for Pr3+:Y2SiO5 and Eu3+:Y2SiO5 are 5 min and 23 days, respectively.

Nevertheless, the advantage of direct telecom compatibility means there has been considerable work towards developing quantum memories using erbium. Instead of storing on spin states, this work has largely focused on storing on the optical transitions. Atomic frequency comb (AFC) delay lines have delayed quantum states of light in Er-doped glass fibre; however, the efficiency was limited to ~1% and the storage time to 50 ns. Weak coherent states have also been stored in crystals using a two-level gradient echo memory (GEM) technique with an efficiency of 0.25%. The fidelity of the recalled state in this demonstration was, however, well below the non-classical limit. For both these demonstrations the inefficiency of the optical pumping of the ground-state electron spin levels was identified as limiting the memories’ efficiency. To avoid this limitation, new on-demand memory techniques, not requiring optical pumping to initialize the ensemble, have been proposed and demonstrated. Although the latter technique has shown

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The material used for this investigation was a 0.005\% doped
\(^{167}\text{Er}^{3+}:\text{Y}_2\text{SiO}_5\) crystal, enriched to 92\% isotopic purity. The Er ions
substitute for Y ions in two non-equivalent \(C_i\) symmetry sites, and
this work used the site labelled ‘site 2’ (ref. 33). To most easily access
the regime where the electron spin was frozen, the magnetic field
was applied along a direction with a large ground-state Zeeman
splitting—namely, the \(D_1\) optical extinction axis (214 GHz T\(^{-1}\)).

The ground-state hyperfine structure was studied optically by
exciting the \(^{1}S_{0} \rightarrow ^{1}P_{1/2}\) optical transition between the two spin-
down projections of the lowest crystal field levels, at a wavelength
of 1,538 nm (see Supplementary Methods for the energy level
structure). In a field of 7 T, the Zeeman splitting of the electronic
spin projections is more than sufficient to resolve this transition.

With a nuclear moment of
\(I = |7/2\rangle\), the \(^{167}\text{Er}^{3+}:\text{Y}_2\text{SiO}_5\) exhibits eight hyperfine
spin states \(m_i = |−7/2, \ldots, +7/2\rangle\) and the entire system studied
comprises sixteen energy levels, as shown Fig. 1a. The hyperfine
energy spacings were obtained by hole-burning measurements
(see Supplementary Methods).

The absorption spectrum of the optical transition is shown in
Fig. 1b. Three bands are clearly resolved in this spectrum, which
we associate with the \(\Delta m_l = −1, 0 \) and \(1\) transitions. The
splitting between bands is 1 GHz, and they can still be resolved
at fields as low as 1 T. The absorption spectrum was fitted with
a population model, shown in the figure, based on the measured
energy level structure and including the contribution of \(I = 0\)
impurity isotopes. The fitted lineshape was a Voigt profile with
equal Gaussian and Lorentzian contributions and a linewidth
of 150 MHz. The fit was used to determine the oscillator strengths
of each hyperfine transition. Whilst the strongest optical transitions
have \(\Delta m_l = 0\), there is appreciable oscillator strength in the fourteen
\(\Delta m_l = ±1\) transitions. Relative to the \(\Delta m_l = 0\) transitions, the

**Figure 1** 1,538 nm optical transition of \(^{167}\text{Er}^{3+}:\text{Y}_2\text{SiO}_5\). a, Energy level
diagram for a field of 7 T. The red-detuned (\(\Delta m_l = −1\)) transitions are
represented by red arrows, and the blue-detuned (\(\Delta m_l = +1\)) transitions
are represented by blue arrows. The energy spacing between the hyperfine
states was determined by hole-burning. b, Orange: absorption spectrum at
1.4 K with a field of 7 T along the \(D_1\) axis. Black: model of absorption based
on hole-burning measurements. Vertical black dashes: the centroids of the
optical transitions used for the spectrum model shown in black.

significantly higher efficiencies—up to 40%—the storage times
are still limited by the optical coherence time and quantum storage has
yet to be demonstrated. Currently, there is no proposed strategy
to achieve the fidelities and storage times required for quantum
repeater applications without using long-lived hyperfine states.

Here, we investigate the hyperfine spin dynamics of
\(^{167}\text{Er}^{3+}:\text{Y}_2\text{SiO}_5\) in the presence of a large magnetic field, with
the aim of increasing both the hyperfine lifetime and coherence
time to be comparable to non-Kramers systems. As mentioned
above, the hyperfine lifetime is short because it is coupled to
the electron spin, which itself flips rapidly due to coupling to
the lattice and to other electron spins. To increase the hyperfine
lifetime, therefore, it is necessary to slow the electron spin flips.
Böttger et al. previously demonstrated that a large magnetic field
can suppress electronic relaxation in \(^{167}\text{Er}^{3+}:\text{Y}_2\text{SiO}_5\) (ref. 31). The field
decreases both the electronic spin–lattice coupling and electronic
cross-relaxation. The spin–lattice is turned off because, with the
electron spin, which itself flips rapidly due to coupling to
the lattice, the hyperfine level is more than sufficient to resolve this transition.

The absorption spectrum of the optical transition is shown in
Fig. 1b. Three bands are clearly resolved in this spectrum, which
we associate with the \(\Delta m_l = −1, 0 \) and \(1\) transitions. The
splitting between bands is 1 GHz, and they can still be resolved
at fields as low as 1 T. The absorption spectrum was fitted with
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of each hyperfine transition. Whilst the strongest optical transitions
have \(\Delta m_l = 0\), there is appreciable oscillator strength in the fourteen
\(\Delta m_l = ±1\) transitions. Relative to the \(\Delta m_l = 0\) transitions, the
We studied hyperfine cross-relaxation by measuring the lifetime of spectral holes burnt into the $\Delta m = +1$ state, the population slowly relaxes to thermal equilibrium via spin–lattice coupling. Three main processes can contribute to this relaxation: the one-phonon direct process, and the two-phonon Raman and Orbach processes. A general expression for the relaxation is:

$$\gamma(T) = \gamma_0 T + \gamma_1 T^3 + \gamma_2 T^5 \exp\left(-\frac{hf}{kT}\right)$$  \hspace{2cm} (1)

where $T$ is the temperature, $f$ is the ground state electronic splitting, $h$ is the Planck constant and $k$ is the Boltzmann constant. $\gamma_0$, $\gamma_1$, and $\gamma_2$ represent the coupling strengths of the direct, Raman, and Orbach processes, respectively.

To first order, these processes cause decay only through the $\Delta m = \pm 1$ hyperfine transitions. This allowed us to model the population dynamics as a series of coupled rate equations, with the same coupling rate $\gamma$ between all adjacent hyperfine states (see Supplementary Methods for more information). This population-dynamics model accurately fits the recorded time series of the spectra, hence supporting the assumptions of the dynamics.

The resulting temperature dependence of the spin–lattice relaxation rate is shown in Fig. 3. The fit shows an exponential increase in $T$, from 2.2 to 1.8 K. This agrees with the exponential reduction in high-energy phonons associated with the Orbach process. Meanwhile, the plateau in transition lifetimes below 1.8 K can be attributed to the direct phonon process. Fitting equation (1) to the low-temperature data gave direct and Orbach coefficients of $\gamma_0 = 9 \times 10^{-4} \text{s}^{-1} \text{K}^{-1}$ and $\gamma_2 = 8 \times 10^{-30} \text{s}^{-1} \text{Hz}^{-3}$, respectively. Above 2.6 K, the plateau in lifetimes indicates the presence of a phonon bottleneck, limiting the rate of hyperfine relaxation, which was not included in the model.

The other important decay mechanism in this system is hyperfine spin–spin cross-relaxation. This process is masked in the previous measurement because it does not redistribute the hyperfine levels across the series. With the $\Delta m = -1$ and $+1$ bands clearly resolved, pumping of the nuclear spin into a single state is possible through frequency selection. By exciting only the seven transitions (the peak at 3 GHz in Fig. 1b), we were able to pump 95 ± 3% of the $^{167}$Er population into the $|+7/2\rangle$ hyperfine ground state. This results in a nearly eightfold increase in the already high optical depth of this transition. Such a high optical depth is difficult to measure accurately with traditional absorption spectroscopy, and instead amplitude modulation (AM) spectroscopy was used. This method is sensitive to both the absorption and the phase shift of the transmitted light, allowing a more accurate determination of the optical depth (for more detail, refer to the Methods).

The resulting spectrum is shown in Fig. 2. Also shown in this figure is the model fitted to the spectrum to obtain the optical depth, and the resulting absorption lineshape. This absorption line has two components—a shoulder indicated at arrow B due to the impurity isotopes, and a central peak at A due to ions in the $m_I = |+7/2\rangle$ state. The AM spectrum for this peak has a central dip due to the changing phase of the transmitted light as the laser was swept through the highly absorbing peak. At this frequency the absorption was determined to be 70 ± 4 dB cm$^{-1}$.

When hole-burning at the frequency indicated by arrow A, the blue spectrum in the inset of Fig. 2 is observed. The hole and anti-holes demonstrate that spin-pumped population is shifted into $|+5/2\rangle$, $|+3/2\rangle$ and $|+1/2\rangle$ hyperfine levels via the $\Delta m = -(1, 2$ and 3) optical decay paths.

The efficiency of the spin pumping (95 ± 3%) was measured by fitting the model to only the $\Delta m = -1$ transitions, where the decreasing trend in oscillator strengths and low absorption improve the accuracy of the population estimate. The ability to polarize the $^{167}$Er ensemble into a single hyperfine state opens the path towards several high-bandwidth memory techniques, which are discussed later.

Once the $^{167}$Er ensemble is pumped into the $|+7/2\rangle$ hyperfine state, the population slowly relaxes to thermal equilibrium via spin–lattice coupling. Three main processes can contribute to this...
states is established and manipulated via a common optical excited state, rather than by direct magnetic resonance. The decay of the echo amplitude with time is shown in Fig. 5. Below 1 s, the decay is sub-exponential, and the $e^{-1}$ decay time is $T_{1\text{echo}} = 1.30 \pm 10$ ms.

With the electron spin frozen, this coherence time will be dominated by the dynamics of the nuclear spins in the crystal—principally the spin-1/2 Y ions. These dynamics are substantially altered from those in undoped $Y_2SiO_5$. The large magnetic field of the Er ion creates a large frozen core of Y spins whose frequencies are detuned from the bulk. This means that the spin flips of those Y ions having the biggest impact on the Er ion are slowed substantially, resulting in a much longer coherence time than expected if all spins were flipping at the bulk rate (3.6 ms, see Supplementary Methods for this calculation). The spectral diffusion caused by the slow spin flips of the frozen core also leads to the non-exponential shape of the echo observed in Fig. 5.

Here, we have shown that applying a large magnetic field of 7 T can extend the hyperfine coherence time of Er to 1.3 s. In fact, Fig. 4 indicates that a similar result could be achieved at fields as low as 3 T. 1.3 s is the longest coherence time obtained in any Kramers system, and is comparable to the longest coherence times obtained in non-Kramers systems using the well-known ZEFOZ technique. From a practical standpoint, the technique shown here has an advantage over ZEFOZ, since it does not require precise alignment of the magnetic field. This technique could be applied to achieve long coherence times in other Kramers ions considered for quantum memories, such as Nd.

In the context of quantum repeater applications, the coherence time seen here is already sufficient for a large-scale network. Ravazi et al. show that a coherence time of 1 s is more than sufficient for a 1,000 km repeater network, even without error correction.

In addition to its long coherence time, $^{167}$Er:$Y_2SiO_5$ in the regime studied here has similar optical depth and optical pumping efficiency to Pr:$Y_2SiO_5$, the non-Kramers material most widely used for quantum memory demonstrations. It also has 100 times larger hyperfine splittings than Pr, which means that it should have larger memory bandwidths and reduced noise from off-resonant excitation during the memory protocol. Collectively, the parameters presented here suggest $^{167}$Er will rival, or exceed, the performance demonstrated by non-Kramers memories.

This system could also be considered for applications requiring long-term storage (that is, a quantum ‘hard drive’). For this application, it should be possible to achieve a coherence time approaching the population $T_1$ limit (10 min) at 1.6 K and 7 T using ZEFOZ. To achieve this longer coherence time it is necessary to turn off the hyperfine cross-relaxation. Since cross-relaxation couples only strongly states with $\Delta m_s = \pm 1$, it can be eliminated by preparing storage qubits using non-adjacent hyperfine levels, for instance $|\pm 7/2\rangle \leftrightarrow |\pm 3/2\rangle$.

Finally, the ability to efficiently pump the $^{167}$Er ensemble into a single hyperfine state is crucial for high-bandwidth quantum communication. It paves the way for broadband Raman memory techniques which, until now, have been limited to atomic vapour systems and potentially nitrogen vacancies in diamond. Alternatively, spin pumping allows for potentially GHz-bandwidth spin-wave storage using techniques already demonstrated in rare earth systems, such as GEM, and drastically improves the efficiency of AFC protocols.

**Methods**

Methods, including statements of data availability and any associated accession codes and references, are available in the online version of this paper.

Received 1 December 2016; accepted 11 August 2017; published online 11 September 2017
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Acknowledgements
M.J.S. would like to thank C. Thiel for insightful discussions. This work was supported by the Australian Research Council Centre of Excellence for Quantum Computation and Communication Technology (Grant No. CE110001027). M.J.S. was supported by an Australian Research Council Future Fellowship (Grant No. FT110100919).

Author contributions
M.J.S. and M.P.H. conceived the initial project. M.J.S., M.P.H. and M.R. designed the experimental set-up. M.R. carried out the experiment. M.R. and R.L.A. analysed the results. All authors contributed to writing the manuscript.

Additional information
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Competing financial interests
The authors declare no competing financial interests.
Methods

Experimental setup. A 0.005% doped 152Er3+:Y2SiO5 3 × 4 × 5 mm (D1, D2, b) crystal provided by Scientific Materials Corp (Bozeman, Montana) was maintained at 1.4 K in an Oxford helium bath cryostat with a 1.5 T super-conducting magnet (see Figure 1 of the Supplementary Methods). Both the magnetic field and light propagating direction were parallel with the D1 optical extinction axis. As there was only one cryostat window, the crystal was placed against a mirror, onto which the beam was focused with a 70 µm waist. The reflection down the D1 axis gave a total absorption length of 6 mm. The optical transitions were excited with a Thorlabs TLK-1550R laser, stabilized to a home-made fibre reference cavity with a 1 s linewidth less than 100 kHz. Intensity and frequency modulation of the light was achieved with a 10 GHz JDSU AM-EOM and detected with a Lab Buddy 10 GHz detector.

AM spectroscopy. This method was based on detecting the optical beat between the sidebands and carrier of an EOM as a function of the modulation frequency. The radiofrequency (RF) modulation sweep was generated by a spectrum analyser, which was also used for detection. This formed a closed modulation loop, as shown in Fig. 1 of the Supplementary Methods. The optical sidebands were weak (~1%) compared to the carrier power, and the laser was kept far detuned (0.5–1 GHz) from the absorption.

This technique is phase sensitive, allowing for accurate measurement of large optical depths and, as the laser remains locked to the reference cavity, spectra can be recorded in conjunction with precise optical pulses and sweeps.

Energy structure and spin pumping. The hyperfine structure of the I1/2, and I3/2 states was determined at 7 T and 1.4 K through a series of hole-burning measurements. A 0 to 4 GHz tunable RF source was used to generate a series of 20 spectral holes across three sets of measurements (see Supplementary Section 3 for an example spectrum). Each spectrum was made by applying fixed frequency RF for 100 ms. The absorption spectrum was recorded by a 0.01 to 2.9 GHz RF scan with the tracking generator and RF input on the spectrum analyser (50 ms scan, 1 MHz RBW).

To spin pump the ensemble into either the mI = ± 7/2 or ± 7/2 hyperfine ground state, a voltage-controlled oscillator (VCO) was used to drive the EOM. A 3 s saw-tooth scan (100 ms rep rate) was performed over the ΔmI = ± 1 or ± 1 absorption bands, respectively, with ~2 mW of optical power in each sideband. The spin-pumped absorption spectrum was recorded once again using AM spectroscopy. Repeating this with the laser (carrier) frequency on either side of the absorption feature, it was possible to stitch two scans together to span more than 2.9 GHz.

Magnetic field dependence of spectral holes. Spectral holes were burnt into the centre of the ΔmI = + 1 absorption band, as it afforded higher optical depth than the ΔmI = −1 band and deeper spectral holes than the ΔmI = 0 band, which had isotopic impurities and excessive optical depth. Between 0 and 1.5 T, spectral holes were generated using a tunable RF source. Above 1.5 T, 10-MHz-wide spectral trenches were burnt instead, as there was negligible spectral diffusion and long hole-burning lifetimes. This was achieved by sweeping the spectrum analyser tracking generator output over 10 MHz for 10 s. The tracking generator output was centered at approximately 1.5 GHz to keep the laser carrier and second sideband off-resonant. The spectral features were measured using phase modulation (PM) spectroscopy.

Temperature dependence of spin pumping. In a 7 T field, spin pumping into the mI = ± 7/2 state was achieved using the same technique as described above. At each temperature, four to five spectra were recorded at time intervals spaced long enough to observe appreciable relaxation back to thermal equilibrium. For example, at 1.6 K, spectra were recorded after 10 s and (10, 20, 40, 80) min. See Section 5 of the Supplementary Methods for example spectra.

Optical Raman echoes. Optical pulses (π and π/2) were generated using the EOM sidebands and two RF generators tuned to 1,155.3 MHz and 2,150 MHz. The pulse lengths were optimized using two pulse optical echoes at either frequency, with 1.5 mW of optical power in each sideband. For the |I1/2, −5/2⟩ ↔ |I1/2, −7/2⟩ transition (1,155.3 MHz), the optimal π-pulse length is 4 ± 0.5 μs, while for the |I1/2, −7/2⟩ ↔ |I1/2, −7/2⟩, a π/2-pulse was 1.5 μs (2,150 MHz). This pair of transitions were chosen because they have the largest oscillator strengths, so shorter pulses could be used. The hyperfine spin ensemble was prepared by first pumping into the mI = ± 7/2 state with a VCO for 10 s, as described above. This was followed by applying a 100 μW pulse at 2,150 MHz for 100 ms, to generate a 100-kHz-wide absorption feature (anti-hole) in the mI = −5/2 subgroup. After the pulse sequence, the echo that re-formed at the wavelength of the 1,155.3 MHz sideband was detected as a 1,155.3 MHz optical beat between the sideband and the carrier. This RF signal was then mixed with a 1,145.3 MHz RF source, and the 10 MHz beat signal was recorded. Note: the detected echo amplitude in this technique depends on the proportion of the initial ensemble transferred into the optical |− 7/2⟩ ↔ |− 7/2⟩ transition, which is determined by the probing power of sideband 2.

One advantage of this technique is that the bandwidth of the ~1 μs optical pulses used is much larger than the inhomogeneous broadening of the hyperfine transition, and so the width of the echo envelope gives a direct measure of this inhomogeneous broadening. As shown in the inset to Fig. 5, the echoes had a full-width at half-maximum of 7 ± 1 μs, corresponding to an inhomogeneous linewidth of 130 ± 20 kHz.

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.