

CHCRUS

This is the accepted manuscript made available via CHORUS. The article has been published as:

Storage and Reemission of Heralded Telecommunication-Wavelength Photons Using a Crystal Waveguide

Mohsen Falamarzi Askarani, Marcel.li Grimau Puigibert, Thomas Lutz, Varun B. Verma, Matthew D. Shaw, Sae Woo Nam, Neil Sinclair, Daniel Oblak, and Wolfgang Tittel Phys. Rev. Applied **11**, 054056 — Published 21 May 2019 DOI: 10.1103/PhysRevApplied.11.054056

Storage and reemission of heralded telecommunication-wavelength photons using a crystal waveguide

Mohsen Falamarzi Askarani,^{1,2} Marcel.li Grimau Puigibert,^{1,3} Thomas Lutz,^{1,4} Varun B. Verma,⁵

Matthew D. Shaw,⁶ Sae Woo Nam,⁵ Neil Sinclair,^{1,7} Daniel Oblak,¹ and Wolfgang Tittel^{1,2}

¹Institute for Quantum Science and Technology, and Department of Physics & Astronomy,

University of Calgary, 2500 University Drive NW, Calgary, Alberta, T2N 1N4, Canada

QuTech, Delft University of Technology, 2600 GA Delft, The Netherlands

³University of Basel, Klingelbergstrasse 82, CH-4056 Basel, Switzerland

⁴ETH Zürich, Otto-Stern-Weg 1, 8093 Zrich, Switzerland

⁵National Institute of Standards and Technology, 325 Broadway, Boulder, Colorado 80305, USA

⁶ Jet Propulsion Laboratory, California Institute of Technology,

4800 Oak Grove Drive, Pasadena, California 91109, USA

⁷ California Institute of Technology, 1200 East California Boulevard, Pasadena, California 91125, USA

(Dated: May 5, 2019)

Large-scale fibre-based quantum networks will likely employ telecommunication-wavelength photons of around 1550 nm wavelength to exchange quantum information between remote nodes, and quantum memories, idemally operating at the same wavelength, that allow increasing the transmission distances as a key element of a quantum repeater. However, the development of a suitable memory remains an open challenge. Here, we demonstrate the storage and reemission of single, heralded, 1532 nm-wavelength photons using a crystal waveguide. The photons are emitted from a photon-pair source based on spontaneous parametric down-conversion, and the memory is based on an atomic frequency comb of 6 GHz bandwidth, prepared through persistent spectral hole burning of the inhomogeneously broadened absorption line of a cryogenically-cooled erbium-doped lithium niobate waveguide. Despite currently limited storage time and efficiency, this demonstration represents an important step towards quantum networks that operate in the telecommunication band, and the development of integrated (on-chip) quantum technology using industry-standard crystals.

I. INTRODUCTION.

Many efforts towards future quantum networks [1] have focused on employing telecommunication-wavelength photons in the C-band (1530-1565 nm) due to their suitability for connecting remote nodes via existing and lowloss fibre infrastructure. The synchronization of information in such nodes is enabled by quantum memories that allow storing and later retrieving quantum information [2]. As such, quantum memories are essential for the operation of quantum repeaters, which promise the transmission of quantum information over large distances [3].

The push to realize long-distance quantum communication has been enabled by significant progress in developing quantum-optical technology [4]. However, a lot of this work is not compatible with C-band photons without the use of complexity-adding and potentiallylossy frequency conversion [5]. To avoid this conversion step, supplementary efforts have focused on developing quantum technology that operates in the telecom C-band, which has resulted in the development of efficient single-photon detectors [6] and single-photon sources [7], for example. Furthermore, significant progress towards quantum memories using various platforms has been made in the last decade [8]. One promising approach is based on cryogenically-cooled rare-earth-ion-doped crystals as they often feature suitable properties such as long optical and spin coherence times [9]. Achievements include storage of entangled photons [10, 11], teleportation into a quantum memory [12], storage in nano-fabricated structures[13], and storage assisted by impedance-matched cavities [14, 15]. In addition, quantum storage and signal processing has been demonstrated using thulium doped into an industry-standard titaniumindiffused lithium niobate waveguide [10] – a promising approach to efficient on-chip information processing. However, developing a C-band quantum memory, especially one that can be integrated on-chip, has turned out to be a challenging task.

One class of materials that has the potential to fill this gap is that of erbium-doped crystals. They are unique in that they offer a ground-to-excited level transition at around 1532 nm wavelength and optical coherence times up to 4.4 ms – the longest in any optical transition in a solid [16]. This has prompted the demonstration of a photon-echo quantum memory protocol with attenuated laser pulses at the single-photon level in $Er: Y_2SiO_5$ [17], but not for non-classical (single photon) light. The difficulty with erbium-doped materials is the relatively long lifetime of the excited level—up to $\sim 11 \text{ ms}$ —compared to that of sub-levels within the ground-state manifold (around 100 ms). This makes optical pumping to the latter—a necessary step in photon-echo quantum memory [2]—challenging and poses a significant obstacle to achieving high storage efficiency. This problem is partially solved in Er-doped fibres, which, due to their amorphous (rather than crystalline) structure, feature reduced spin-spin interactions and, as a result, extended groundstate (Zeeman-level) lifetimes [18]. This has allowed recent demonstrations of storage and reemission of nonclassical and entangled states of light at telecommunication wavelengths [19]. However, despite the appeal of Erdoped fibres for all-fibre implementations, decoherence arising from their amorphous structure [20] currently restricts the storage time to about 50 nanoseconds, though improvements at ultra-low temperatures may be possible.

Other promising approaches to overcoming the limitations of erbium are the use of magnetic fields of several Tesla, where ground state lifetimes can exceed one minute [21], and crystals inside cavities with high quality factors and small mode volumes, which reduce the excited state lifetime by means of the Purcell effect [22]. But despite a lot of research aimed at developing quantum memory at telecommunication wavelength, the storage and reemission of non-classical light using an erbiumdoped crystal, in particular a crystalline waveguide that can be integrated with telecommunication-industry devices, remains a yet-to-be-achieved goal.

Here we demonstrate quantum memory for heralded single photons at 1532.05 nm wavelength using an Er-doped lithium niobate waveguide created through titanium indiffusion. The heralded photons are generated using a photon-pair source based on spontaneous parametric down-conversion (SPDC), and storage and reemission relies on the atomic frequency comb (AFC) protocol [23] and optical pumping into superhyperfine levels, which have previously not been exploited towards this end. The non-classical nature of the re-emitted (heralded) photons is verified by measuring the crosscorrelation function $g_{12}^{(2)}$ with the heralding photons.

II. THE AFC QUANTUM MEMORY PROTOCOL

The storage and reemission process of single photons is based on the AFC protocol, which has underpinned a lot of progress towards efficient and broadband quantum memory during the past decade. For example, storage times as long as milliseconds [24], efficiencies as high as 56% [14], storage bandwidths of several GHz [10] and fidelities up to 99.9% [25] have been achieved.

An atomic frequency comb is comprised of a series of narrow spectral absorption lines that are equidistantly detuned by Δ (a 'comb' of absorption features) [23]. It can be prepared from an inhomogeneously broadened absorption line of rare-earth ions by frequency-selective optical pumping of population into long-lived auxiliary levels. The absorption of a photon by an AFC leads to a collective atomic excitation described by

$$|\Psi\rangle_{A} = \frac{1}{\sqrt{N}} \sum_{j=1}^{N} c_{j} e^{-i2\pi\delta_{j}t} e^{ikz_{j}} |g_{1}, ... e_{j}, ... g_{N}\rangle.$$
(1)

Here N is the number of ions and δ_j the detuning of the *j*th ion's transition frequency with respect to the carrier

frequency of the incoming photon. The longitudinal position of the ions is denoted by z_j , and the coefficient c_j is related to the ion's excitation probability (i.e. it's position and transition frequency). After absorption, each term in Eq. (1) accumulates a phase that depend on the detuning $\delta_j = m_j \Delta$, where m_j is an integer. Consequently, all terms coherently rephase at a time $\tau = 1/\Delta$, leading to collective reemission of the photon in its original quantum state. Under certain conditions the reemission process can reach unit efficiency [23, 26, 27].

III. SPECTROSCOPIC CHARACTERIZATION

For AFC-based quantum state storage in an ensemble of absorbers, in our case rare-earth ions doped into a crystal, the storage medium has to meet several criteria. In addition to suitable level structure (e.g. absorption at the desired wavelength), they include an inhomogeneously broadened absorption profile with a bandwidth that exceeds the inverse duration of the photons to be stored; the possibility for optical pumping into long-lived auxiliary shelving levels (so-called persistent spectral hole burning); and a coherence time in excess of the desired storage time.

To date, only a few spectroscopic studies of the lowtemperature properties of the 1532 nm ${}^4I_{15/2} \leftrightarrow {}^4I_{13/2}$ transition of erbium-doped lithium niobate-both in bulk crystals as well as waveguides created by means of titanium indiffusion (see Appendix A for details about the waveguide fabrication)—have been reported [9, 28– 31]. For light polarized orthogonal to the crystal's c-axis $(\mathbf{E} \perp \mathbf{c})$, the 1532 nm transition of Er:LiNbO₃ features a 180 GHz-wide inhomogeneously-broadened line, a 2 ms population lifetime of the ${}^{4}I_{13/2}$ level, and, at a temperature of 1.6 K and a magnetic field of 50 kG oriented parallel to the c-axis $(\mathbf{B} \| \mathbf{c})$, an optical coherence time of 117 μ s [9, 30]. The same transition of Er³⁺ was studied in a Ti⁴⁺:LiNbO₃ waveguide at 3 K, resulting in a 250 GHzwide inhomogeneously broadened line, a population lifetime matching that of the bulk material, and a coherence lifetime of 18.2 μ s in a 4 kG field oriented **B**||**c** [28]. We expect that the coherence lifetime of the waveguide increases and eventually equals that of the bulk crystal at lower temperatures and under the application of an optimized magnetic field, similarly to what we previously observed for a Tm³⁺:Ti⁴⁺:LiNbO₃ waveguide compared to bulk Tm^{3+} :LiNbO₃ [32].

To identify long-lived auxiliary levels with lifetimes that significantly exceed that of the ${}^{4}I_{13/2}$ excited level, we perform spectral hole burning (SHB) to study the level structure and dynamics of $\mathrm{Er}^{3+}:\mathrm{Ti}^{4+}:\mathrm{LiNbO}_{3}$ at various magnetic fields. Towards this end we excite the inhomogeneously broadened 1532 nm transition using a narrowband laser, and then read the frequencydependent absorption profile by scanning the laser over a spectral interval centered at the excitation frequency.



FIG. 1. a) Experimental timing sequence. The sequence is continuously repeated during the experimental run. Switch 2 is closed (and connected to the photodetector, PD) during the AFC preparation for monitoring the AFC structure and for recording the frequency-depending optical depth shown in Figs. 2a and 3b. The experimental set-up consists of two main parts. The part depicted in the top half allows for optical pumping (spectral hole burning) and AFC generation – it consists of a pump laser, a phase modulator (PM), an acousto-optic modulator (AOM), a polarization controller (PC), optical switches, a circulator, and a photo-detector (PD). The part in the bottom half depicts the heralded single photon generation and the detection setup – it consists of a pulsed pump laser, second harmonic generation (SHG), spontaneous parametric down conversion (SPDC), longpass filters (LF), Fabry-Perot cavities (FP), superconducting nanowire single-photon detectors (SNSPD), and a time-to-digital converter (TDC). See Appendix C to E for details of all components.

An absorption spectrum obtained at a magnetic field of 19 kG and taken after 30 s—a time that exceeds by far the lifetime of the ${}^{4}I_{13/2}$ level—is shown in Fig. 2a. Regions of increased (decreased) transparency that are detuned from the central hole (at the excitation frequency) are referred to as side- (anti-) holes – additional studies with varying waiting times between excitation and read-out reveal that they persist for several minutes.

To determine the cause of the level splitting and hence the sub-structure, we perform SHB at different magnetic fields with $\mathbf{B} \| \mathbf{c}$. Fig. 2b depicts the field-dependent detunings for three pairs of distinguishable side-holes. Fits yield values of 1.15 ± 0.01 , 1.721 ± 0.006 , and 0.572 ± 0.002 kHz/G, respectively. This agrees with the superhyperfine coupling between the Er^{3+} electronic spin (S = 1/2)and the nuclear spins of ⁹³Nb (I = 9/2), ⁷Li (I = 3/2), and ⁶Li (I = 1) in the host crystal that was previously reported for an Er^{3+} :LiNbO₃ bulk crystal [30]. The sidehole positions are determined by the excited-level splitting, whereas the position of the anti-holes are given by the ground state splitting as well as by differences between ground and excited state splittings. These observations suggest the possibility for persistent spectral hole burning using, for the first time, superhyperfine levels.

In addition, we also found evidence of population transfer into long-lived electronic Zeeman levels of erbium at small magnetic fields. However, a large absorption background in AFCs exceeding 1 GHz bandwidth prevented us from using these levels for shelving. For more information see [33].

IV. HERALDED SINGLE PHOTON STORAGE

To demonstrate storage of single photons in the Erdoped crystal, we apply a magnetic field of 16.5 kG and $\mathbf{B} \| \mathbf{c}$ across the crystal, and create an AFC with approximately 21 MHz tooth spacing, i.e. a storage time of $\tau = 48$ ns. The tooth spacing was chosen such that the comb periodicity coincides with the excited-level splitting caused by the super-hyperfine interactions between the erbium electronic spin and the nuclear spin of ⁷Li and ⁹³Nb. The finesse of the comb – the ratio of tooth width γ to tooth spacing Δ – is two. Combined with the optical depths of the comb and the background obtained from the AFC trace in Fig. 3, it limits the AFC storage efficiency to around 1%. However, insufficient laser in-



FIG. 2. a) Optical depth over a 100 MHz-wide spectral range centered at 1532.05 nm after applying a 19 kG magnetic field. Side holes due to the interaction between the Er^{3+} electronic spin and 93 Nb, 6 Li, and 7 Li nuclear spins are visible. The apparent side-holes at ± 50 MHz are due to an imperfect frequency sweep. b) Detuning of 7 Li and 93 Nb side holes as a function of the magnetic field. Side holes for 6 Li are resolved only at the highest field. Uncertainty bars indicate the difference between the detunings of the positive and negative side hole compared to the central hole.



FIG. 3. a) A 200 MHz wide section of our 6 GHz-bandwidth AFC. b) Simplified energy level diagram of erbium in lithium niobate. An AFC can be created by redistributing—optically pumping through the ${}^{4}I_{13/2}$ excited level—ions between superhyperfine levels of the ${}^{4}I_{15/2}$ ground state manifold.

tensity at large detunings and hence non-optimized hole burning leads to a non-uniform AFC and an averaged AFC efficiency of around 0.1%.

Next, we generate 6 GHz wide heralded single photons at 1532 nm wavelength using spontaneous parametric down conversion (SPDC) of short, 523 nm laser pulses in a periodically-poled lithium-niobate crystal. This nonlinear process results in the probabilistic generation of photon pairs at 795 nm and 1532 nm wavelength, and detecting a 795 nm photon hence indicates (heralds) the presence of another photon at 1532 nm. (The experimental setup is outlined in Fig. 1 and its components are described in detail in Appendix C.) These photons are then sent into, absorbed (stored) in, and re-emitted from the erbium memory after the pre-programmed time delay of 48 ns, see Fig. 4.

To demonstrate the non-classical nature of the reemitted photons, and hence the memory, we measure the second-order cross-correlation function $g_{12}^{(2)} = \frac{P_{12}}{P_1 P_2}$. Here, the probability of detecting a coincidence between



FIG. 4. Storage and reemission of heralded single photons at telecommunication wavelength. The histogram depicts the time-resolved rate of coincidence detections of heralding 795 nm and heralded 1532 nm photons. The overall acquisition time is 2 min. The time axis is defined relative to the detection of 1532 nm photons that are directly transmitted (not absorbed) by the memory. Stored photons are re-emitted after 48 ns and cause the coincidence peak highlighted in red in the zoom in. The number of coincidences in the 500 pslong window is 18 ± 4.24 , clearly exceeding the background of 1.9 ± 0.45 per 500 ps window, obtained by averaging coincidences between 44.5 ns and 49.5 ns. The coincidence peaks at $t \approx 2.5, 5, 7.5, 10$ ns are due to spurious temporal modes of the pump laser - a consequence of imperfect alignment of the laser cavity - causing additional photon-pair generation by the SPDC crystal. The sequence of coincidence detection peaks is repeated every 12.5 ns, matching the 80 MHz repetition rate of the pulsed SPDC pump laser. These repeated patterns, often referred-to as accidental coincidences, are due to the detection of photons belonging to pairs that have been generated during different laser cycles.

the two photons is P_{12} , and P_1 (P_2) are the probabilities of detecting the two photons individually (see Ref. [34] for more details). To verify the presence of non-classical correlations, $g_{12}^{(2)}$ must be greater than 2 [34, 35]. First, bypassing the waveguide, we determine $g_{12}^{(2)}$ of our photon pair source (without additional storage) to be around 20. It is limited by multi-pair emissions that are caused by the high pump intensity [35]. Repeating the measurement with re-emitted photons, we then find $g_{12}^{(2)}=7.1\pm4$ (see Appendix E for details). This demonstrates that non-classical correlations between the members of photon pairs remain throughout the storage process, i.e. that the memory indeed operates in the quantum domain.

V. DISCUSSION

While this result is promising, several improvements are necessary to create a quantum memory that can be used in a quantum repeater. First, the total system efficiency—evaluated by comparing the heralded photon detection rates measured with the full setup with that after circumventing the cryostat (including the memory) is currently only of around 0.01%. This number is limited by a coupling efficiency of around 10% and, as mentioned above, the efficiency of the storage and reemission process (i.e. that of the AFC itself) of 0.1%. The latter is determined by several factors, including the lack of phase-matching that allows triggering backwards emission [23], low finesse (F=2) of the AFC due to the chosen pumping scheme, insufficient optical depth of the comb teeth, and a remaining absorption background, especially at large detuning.

The coupling efficiency can be improved by optimized matching of the modes guided by the fibre and the waveguide. Furthermore, the limited optical depth and the lack of phase matching can be countered with an impedancematched cavity [26, 27]. However, this solution is not beneficial without reducing the remaining background loss, which we attribute to the complexity of the superhyperfine level structure and the possibility of laser-induced enhancement of spin relaxation [31, 33] – an effect that we have previously observed, and mitigated, in the creation of AFCs in $\text{Tm}^{3+}:\text{Ti}^{4+}:\text{LiNbO}_3$ [32].

We believe that further characterization of the atomic level structure and population dynamics, in particular through more detailed SHB measurements (e.g. with varying magnetic field strength and orientation), as well as optimization of the AFC preparation steps (e.g. using back-pumping [36] or spin-mixing methods [37]), will allow a significant reduction of the background absorption. Another possibility may be exploring persistent spectral hole burning into Zeeman levels [33]. Furthermore, wavelength-dependent measurements may allow the identification of ions belonging to different magnetic sub-classes and featuring favorable (superhyperfine- or Zeeman-) levels. Ultimately, a lower sample temperature and erbium doping concentration could allow for more efficient SHB in addition to suppressing decoherence caused, e.g., by spin flip-flops.

Another limitation of the current demonstration is the storage time, which, as described above, is indirectly determined by the strength of the external magnetic field. More precisely, the field sets the splitting of the superhyperfine levels, which in turn determines the frequency spacing between peaks and troughs of the AFC and hence the comb periodicity Δ . If this were the only factor, then using small magnetic fields would allow for arbitrarily small teeth spacing and hence arbitrarily long storage times. However, the minimum teeth spacing (and hence the maximum storage time) is given by the material's homogeneous linewidth (the inverse of the coherence time, possibly rendered worse by spectral diffusion), and - being the limiting factor in our case – instability of the frequency of the (free running) laser used for spectral hole burning. We estimate that locking to a stable reference cavity would allow increasing the storage time to a few μ s, which could be further increased to hundreds of μ sec by further reducing the temperature.

Finally, we note that our current approach does not allow photons to be re-emitted on-demand, which requires an additional step, i.e. the reversible, coherent, mapping to a third long-lived level [38]. Nonetheless, applications such as quantum repeaters may utilize memories of fixed storage times provided that spectral or spatial multiplexing is performed [39].

VI. CONCLUSION

In conclusion, we have demonstrated the storage and reemission of heralded single photons at telecommunication-wavelength using a cryogenicallycooled erbium-doped lithium niobate waveguide. The non-classical nature of the storage process is demonstrated by a measurement of the cross-correlation coefficient between the heralding and the heralded photons after reemission. We employed an AFC that is based on optical pumping of atomic population into superhyperfine ground levels, and we have detailed the limitations as well as possible improvements of our memory, in particular its limited storage efficiency. Our work is a step towards on-chip quantum network technology operating in the important telecommunication C-band using standard telecommunication industry materials.

APPENDIX A: ER-DOPED LITHIUM NIOBATE WAVEGUIDE

To fabricate the Er³⁺:Ti⁴⁺:LiNbO₃ waveguide, z-cut congruent $LiNbO_3$ is Er-doped over a length of 10 mm by indiffusion of a vacuum-deposited (electron-beam evaporated) 8 nm-thick Er layer at 1130 °C for 150 h in an Aratmosphere. This step is followed by a post-treatment in Oxygen (1 h) to get a full re-oxidization of the crystal. Er substitutes for Li when incorporated into the LiNbO₃ lattice. The indiffusion results in a $3.6 \times 10^{19} \text{ cm}^{-3}$ near-surface concentration and a Gaussian concentration profile that features a 1/e penetration depth of 8.2 μm [40]. Next, the waveguide is created by indiffusion of Ti. To do this, a 98 nm-thick titanium layer is deposited on the Er-doped surface of the LiNbO₃ substrate using electron beam evaporation. From this layer, 7 μ m-wide Ti strips are defined by photo-lithography and chemical etching, and subsequently in-diffused at 1060 °C for 8.55 h. This process leads to a single-mode waveguide with a $4.5 \times 3 \ \mu m$ full-width-at-half-maximum intensity distribution for transverse magnetic-polarization. Note that an analogous procedure, which is detailed in Ref. [10], was used to fabricate the similar $Tm^{3+}:Ti^{4+}:LiNbO_3$ waveguide.

For our experiments, the Er^{3+} :Ti⁴⁺:LiNbO₃ waveguide is mounted in an adiabatic demagnetization refrigerator that can be operated at temperatures of around 0.6 K (Fig. 1). A magnetic field oriented $\mathbf{B} \| \mathbf{c}$ of up to 20 kG is applied using a solenoid. Light is coupled into, and out of, the waveguide by fibre butt-coupling. The overall in-out coupling transmission of our cryogenic setup is around 10% (measured at a wavelength of 1532.05 nm).

APPENDIX B: SHB AND AFC PREPARATION

Time-dependent SHB is performed by exciting a subset of ions within a few-MHz-broad spectral region of the inhomogeneously-broadened absorption line using continuous-wave laser light at 1532.05 nm wavelength. The experimental setup is depicted in the top half of Fig. 1b. Optical pumping then leads to a modification of the absorption profile, which is characterized by sweeping the frequency of the light across the spectral hole using a phase modulator and serrodyne modulation. This allows characterization of level spacings and, if repeated after various delays, population dynamics.

The AFC is generated by optical pumping during 300 ms, as illustrated by the timing sequence in Fig. 1a. During this stage, the laser light is frequency-swept over a 6 GHz bandwidth by serrodyne phase-modulation while its intensity is modulated using an AOM. This procedure creates features of high (low pump power intensity) and low (high pump power intensity) absorption, as shown in Fig. 3 for a magnetic field of 16.5 kG and $\mathbf{B} \| \mathbf{c}$. A 30 ms time delay following the optical pumping step (see Fig. 1a) ensures that no noise-inducing photons that are spontaneously emitted from the ${}^4I_{13/2}$ excited-level mask the stored and re-emitted photons. Finally, photons are repetitively stored and re-emitted after 48 ns during a 200 ms time interval (see Fig. 1a). The closing of the first optical switch (which is open during the AFC preparation) in conjunction with the circulator allows suppressing stray pump light during the storage and reemission of the heralded single photons. Note that the light polarization and the magnetic field orientation are, respectively, set perpendicular and parallel to the c-axis of the crystal, i.e. $\mathbf{E} \perp \mathbf{c}$ and $\mathbf{B} \parallel \mathbf{c}$.

APPENDIX C: SPDC-BASED HERALDED SINGLE PHOTON SOURCE

As shown in Fig. 1, the creation of photon pairs begins with a mode-locked laser that emits pulses of 6 ps-duration at a rate of 80 MHz and a wavelength of 1047 nm. These pulses are frequency-doubled to 523.5 nm by means of second harmonic generation using a PPLN crystal and then used to pump a second PPLN crystal phase-matched for SPDC and creating frequencycorrelated photon pairs at 795 nm and 1532 nm wavelength. After passing through an interference filter that removes the remaining 523 nm light, a dichroic mirror separates the photons from each pair. Finally, the bandwidth of each photon is reduced by filtering: we employ a 12 (6) GHz bandwidth fibre-Bragg grating (Fabry-Perot filter) for filtering the 1532 (795) nm photons. The 1532 nm photons are coupled into the waveguide (the second optical switch is open at this stage) and then stored. After reemission, they are detected by a superconducting nanowire single-photon detector (SNSPD) [41]. The heralding 795 nm photon is directly detected by another SNSPD.

APPENDIX D: DATA ACQUISITION AND $g^{(2)}$ MEASUREMENT

To collect data we employ a start-stop triggering method using a time-to-digital converter (TDC). The TDC starts with the detection of the heralding 795 nm photons, and it stops with the detection of heralded 1532 nm photons after being transmitted through, or re-emitted from, the waveguide. The TDC generates a histogram (shown in Fig. 4) that indicates the number of coincidence detections as a function of time delay between the start and the stop.

The measured coincidence rate before adding the memory was around 2300 s^{-1} . It is determined by the repetition rate of the pump laser (80 MHz), the mean photonpair generation probability per pump pulse (1.6%), and several factors that limit the probability for detecting the created photons. They are the coupling efficiencies into fibre of around 70% for photons of both wavelength; transmission of the photons through longpass filters (85% for both wavelength) and Fabry Perot cavities (40% and 60%), bandwidth mismatch between the pump and down-converted photons (17% and 35%) [42]; and detection efficiencies of the single photon detectors of around 70%. All brackets denote first the respective value for 795 nm, and then for 1532 nm photons. The multiplication of these factors leads to a predicted coincidence rate of 3170 s^{-1} – not far from the measured result. We attribute the difference to loss in fibre connectors and underestimation of the effect of bandwidth mismatch. Including the quantum memory system efficiency of 0.01%, and 33% extra loss due to an additional optical switch and a circulator then leads to a calculated coincidence rate for re-emitted photons of around 0.21 s^{-1} . This approximately matches the experimental value of 0.16 s^{-1} that can be extracted from the coincidence peak centered around 48 ns in Fig. 4.

As explained in more detail in [43], the cross correlation function $g_{12}^{(2)}$ can be evaluated directly from the histogram by comparing true and accidental coincidence rates:

$$g_{12}^{(2)} = \frac{P_{12}}{P_1 P_2} = \frac{R_{true}}{R_{acc}}.$$

Here, R_{true} , which is proportional to P₁₂, denotes the coincidence detection rate of photons emitted simultaneously. These coincidences are represented by the peak at t=48 ns. Furthermore R_{acc} denotes the rate of accidental

coincidences – it is proportional to P_1P_2 and the coincidences are represented by the peaks highlighted in gray in Fig. 4. For increased accuracy, we evaluated R_{acc} by averaging over five accidental coincidence peaks.

We note that an adsorber that is part of the cryogenic system broke shortly after we started measuring. This made it impossible to improve the data depicted in Fig. 4 and hence the cross-correlation function.

APPENDIX E: SUPERCONDUCTING NANOWIRE SINGLE-PHOTON DETECTORS (SNSPDS)

The SNSPDs used in our experiment feature a detection efficiency of around 70%, a time resolution of around 100 ps, a dead time of approximately 100 ns, and a dark count rate of 10-20 Hz. The signal created by each SNSPD is first amplified, shaped to a square pulse by a comparator, and then sent to a digital delay generator. The generator's two outputs are connected to the TDC and a counter.

ACKNOWLEDGMENTS

The authors thank Wolfgang Sohler, Mathew George, and Raimund Ricken for providing the Er³⁺:Ti⁴⁺:LiNbO₃ waveguide, Francesco Marsili for assistance with development of the SNSPDs, Jakob H. Davidson for help with aligning the waveguide, and Gustavo Amaral and Erhan Saglamyurek for useful discussions. This work is funded through Alberta Innovates Technology Futures (AITF), the National Science and Engineering Research Council of Canada (NSERC), and the Defense Advanced Research Projects Agency (DARPA) Quiness program. W.T. furthermore acknowledges funding by the Netherlands Organization for Scientific Research (NWO) and as a Senior Fellow of the Canadian Institute for Advanced Research (CIFAR), and V.B.V. and S.W.N. partial funding for detector development from the Defense Advanced Research Projects Agency (DARPA) Information in a Photon (InPho) program. Part of the detector research was carried out at the Jet Propulsion Laboratory, California Institute of Technology, under a contract with the National Aeronautics and Space Administration (NASA).

- H. J. Kimble, The quantum internet, Nature 453, 1023 (2008).
- [2] A. I. Lvovsky, B. C. Sanders, and W. Tittel, Optical quantum memory, Nature Phot. 3, 706 (2009).
- [3] N. Sangouard, C. Simon, H. De Riedmatten, and N. Gisin, Quantum repeaters based on atomic ensembles and linar optics, Rev. Mod. Phys. 83, 33 (2011).
- [4] J. L. O'Brien, A. Furusawa, and J. Vučković, Photonic quantum technologies, Nature Phot. 3, 687 (2009).
- [5] S. Tanzilli, W. Tittel, M. Halder, O. Alibart, P. Baldi, N. Gisin, and H. Zbinden, A photonic quantum information interface, Nature 437, 116 (2005).
- [6] C. M. Natarajan, M. Tanner, and R. H. Hadfield, Superconducting nanowire single-photon detectors: physics and applications, Superconductor science and technology 25, 063001 (2012).
- [7] T. Müller, J. Skiba-Szymanska, A. B. Krysa, J. M. Felle, M. Anderson, R. M. Stevenson, J. Heffernan, D. A. Ritchie, and A. J. Shields, A quantum light-emitting diode for the standard telecom window around 1550 nm, Nature Comms. 9, 862 (2018).
- [8] F. Bussières, N. Sangouard, M. Afzelius, H. De Riedmatten, C. Simon, Christoph and W. Tittel, Prospective applications of optical quantum memories, J. Mod. Opt. 60, 1519 (2013).
- [9] C. W. Thiel, T. Böttger, and R. L. Cone, Rare-earthdoped materials for applications in quantum information storage and signal processing, J. Luminescence 131, 353 (2011).
- [10] E. Saglamyurek, N. Sinclair, J. Jin, J. A. Slater, D. Oblak, F. Bussieres, M. George, R. Ricken, W. Sohler, and W. Tittel, Broadband waveguide quantum memory for entangled photons, Nature 469, 512 (2011).

- [11] C. Clausen, I. Usmani, F. Bussières, N. Sangouard, M. Afzelius, H. De Riedmatten, and N. Gisin, Quantum storage of photonic entanglement in a crystal, Nature 469, 508 (2011).
- [12] F. Bussières, C. Clausen, A. Tiranov, B. Korzh, V. B. Verma, S. W. Nam, F. Marsili, A. Ferrier, P. Goldner, H. Herrmann, and C. Silberhorn, Quantum teleportation from a telecom-wavelength photon to a solid-state quantum memory, Nature Phot. 8, 775 (2014).
- [13] T. Zhong, J. M. Kindem, J. G. Bartholomew, J. Rochman, I. Craiciu, E. Miyazono, M. Bettinelli, E. Cavalli, V. Verma, S. W. Nam, F. Marsili, M. D. Shaw, A. D. Beyer, and A. Faraon, Nanophotonic rare-earth quantum memory with optically controlled retrieval, Science **357**, 1392 (2017).
- [14] M. Sabooni, S. Tornibue Kometa, A. Thuresson, S. Kröll, and L. Rippe, Cavity-enhanced storage - preparing for high-efficiency quantum memories, N. J. Phys. 15, 035025 (2013).
- [15] P. Jobez, I. Usmani, N. Timoney, C. Laplane, N. Gisin, and M. Afzelius, Cavity-enhanced storage in an optical spin-wave memory, N. J. Phys. 16, 083005 (2014).
- [16] T. Böttger, C. W. Thiel, R. L. Cone, and Y. Sun, Effects of magnetic field orientation on optical decoherence in Er³⁺:Y₂ SiO₅, Phys. Rev. B **79**, 115104 (2009).
- [17] B. Lauritzen, J. Minář, H. De Riedmatten, M. Afzelius, N. Sangouard, C. Simon, and N. Gisin, Telecommunication-wavelength solid-state memory at the single photon level, Phys. Rev. Lett. **104**, 080502 (2010).
- [18] E. Saglamyurek, T. Lutz, L. Veissier, M. P. Hedges, C. W. Thiel, R. L. Cone, and W. Tittel, Efficient and longlived Zeeman-sublevel atomic population storage in an

erbium-doped glass fiber, Phys. Rev. B $92,\ 241111(\mathrm{R})$ (2015).

- [19] E. Saglamyurek, J. Jin, V. B. Verma, M. D. Shaw, F. Marsili, S. W. Nam, D. Oblak, and W. Tittel, Quantum storage of entangled telecom-wavelength photons in an erbium-doped optical fibre, Nature Phot. 9, 83 (2015).
- [20] L. Veissier, M. Falamarzi, T. Lutz, E. Saglamyurek, C. W. Thiel, R. L. Cone, and W. Tittel, Optical decoherence and spectral diffusion in an erbium-doped silica glass fiber featuring long- lived spin sublevels, Phys. Rev. B 94, 195138 (2016).
- [21] M. Ranĉić, M. P. Hedges, R. L. Ahlefeldt, and M. J. Sellars, Coherence time of over a second in a telecomcompatible quantum memory storage material, Nature Physics 14, 50 (2018).
- [22] E. Miyazono, T. Zhong, I. Craiciu, J. M. Kindem, and A. Faraon, Coupling of erbium dopants to yttrium orthosilicate photonic crystal cavities for on-chip optical quantum memories, Appl. Phys. Lett. 108, 011111 (2016).
- [23] M. Afzelius, C. Simon, H. De Riedmatten, and N. Gisin, Multimode quantum memory based on atomic frequency combs, Phys. Rev. A 79, 052329 (2009).
- [24] P. Jobez, C. Laplane, N. Timoney, N. Gisin, A. Ferrier, P. Goldner, and M. Afzelius, Coherent spin control at the quantum level in an ensemble-based optical memory, Phys. Rev. Lett. **114**, 230502 (2015).
- [25] Z. Q. Zhou, W. B. Lin, M. Yang, C. F. Li, and G. C. Guo, Realization of reliable solid-state quantum memory for photonic polarization qubit, Phys. Rev. Lett. 108, 190505 (2012).
- [26] M. Afzelius, and C. Simon, Impedance-matched cavity quantum memory, Phys. Rev. A 82, 022310 (2010).
- [27] S. A. Moiseev, S. N. Andrianov, and F. Gubaidullin, Efficient multimode quantum memory based on photon echo in an optimal QED cavity, Phys. Rev. A 82, 022311 (2010).
- [28] A. Delfan Abazari, Photon echo quantum memory and state transformation (M.Sc. Thesis - University of Calgary, 2009).
- [29] C. W. Thiel, R. M. Macfarlane, R. L. Cone, Y. Sun, T. Böttger, K. D. Merkel, and W. R. Babbitt, Spectroscopy and dynamics of Er⁺³:LiNbO₃ at 1.5 microns for quantum information and signal processing applications, (10th Intl Meeting on Hole Burning, Single Molecule, and Related Spectroscopies: Science and Applications -HBSM 2009).
- [30] C. W. Thiel, R. M. Macfarlane, T. Böttger, Y. Sun, R. L. Cone, and W. R. Babbitt, Optical decoherence and persistent spectral hole burning in Er³⁺:LiNbO₃, J. Luminescence **130**, 1603 (2010).
- [31] C. W. Thiel, Y. Sun, R. M. Macfarlane, T. Böttger, and R. L. Cone, Rare-earth-doped LiNbO₃ and KTiOPO₄ (KTP) for waveguide quantum memories, J. Phys. B: Atomic, Molecular and Optical Physics 45, 124013 (2012).

- [32] N. Sinclair, D. Oblak, C. W. Thiel, R. L. Cone, and W. Tittel, Properties of a rare-earth-ion-doped waveguide at sub-Kelvin temperatures for quantum signal processing, Phys. Rev. Lett. **118**, 100504 (2017).
- [33] M. F. Askarani, T. Lutz, M. G. Puigibert, N. Sinclair, D. Oblak, and W. Tittel, Persistent atomic frequency comb based on Zeeman sub-levels of an erbium-doped crystalline waveguide, in preparation.
- [34] S. Fasel, O. Alibart, S. Tanzilli, P. Baldi, A. Beveratos, N. Gisin, and H. Zbinden, High-quality asynchronous heralded single- photon source at telecom wavelength, New J. Phys. 6, 163 (2004).
- [35] P. R. Tapster and J. G. Rarity, Photon statistics of pulsed parametric light, J. Mod. Opt. 45, 595 (1998).
- [36] M. Nilsson, L. Rippe, S. Kröll, R. Klieber, and D. Suter, Hole-burning techniques for isolation and study of individual hyperfine transitions in inhomogeneously broadened solids demonstrated in Pr³⁺:Y₂SiO₅, Phys. Rev. B **70**, 214116 (2004).
- [37] B. Lauritzen, S. R. Hastings-Simon, H. De Riedmatten, M. Afzelius, and N. Gisin, State preparation by optical pumping in erbium-doped solids using stimulated emission and spin mixing, Phys. Rev. A 78, 043402 (2008).
- [38] M. Afzelius, I. Usmani, A. Amari, B. Lauritzen, A. Walther, C. Simon, N. Sangouard, J. Minář, H. De Riedmatten, N. Gisin, and S. Kröll, Demonstration of atomic frequency comb memory for light with spin-wave storage, Phys. Rev. Lett. **104**, 040503 (2010).
- [39] N. Sinclair, E. Saglamyurek, H. Mallahzadeh, J. A. Slater, M. George, R. Ricken, M. P. Hedges, D. Oblak, C. Simon, W. Sohler, and W. Tittel, Spectral multiplexing for scalable quantum photonics using an atomic frequency comb quantum memory and feed-forward control, Phys. Rev. Lett. **113**, 053603 (2014).
- [40] I. Baumann, R. Brinkmann, M. Dinand, W. Sohler, L. Beckers, C. Buchal, M. Fleuster, H. Holzbrecher, H. Paulus, K. H. Müller, and T. Gog, Erbium incorporation in LiNbO3 by diffusion-doping, Applied Physics A 64, 33 (1996).
- [41] F. Marsili, V. B. Verma, J. A. Stern, S. Harrington, A. E. Lita, T. Gerrits, I. Vayshenker, B. Baek, M. D. Shaw, R. P. Mirin, and S. W. Nam, Detecting single infrared photons with 93% system efficiency, Nature Phot. 7, 210 (2013).
- [42] J. Jin, M. G. Puigibert, L. Giner, J. A. Slater, M. R. Lamont, V. B. Verma, M. D. Shaw, F. Marsili, S. W. Nam, D. Oblak, and W. Tittel, Entanglement swapping with quantum-memory-compatible photons, Phys. Rev. A 92, 012329 (2015).
- [43] A. Kuzmich, W. P. Bowen, A. D. Boozer, A. Boca, C. W. Chou, L.M. Duan, and H. J. Kimble, Generation of nonclassical photon pairs for scalable quantum communication with atomic ensembles, Nature 423, 731 (2003).