Quantum memories for light play a vital role in optical quantum networks (1) for long-distance secure communications and interconnecting future quantum computers. Although optical quantum memories—devices that faithfully store input photonic qubits—have already been implemented in macroscopic systems such as atomic ensembles (2), solid-state crystals (3–5), and waveguides (6–8), realizing scalable networks requires integration of micro- or nanoscale memories with other guided-wave components for on-chip quantum information routing and processing. Crystals doped with rare-earth ions (REIs) are appealing solid-state materials for quantum storage because of their highly coherent optical transitions between few levels, which are further split into Zeeman and hyperfine levels with coherent transitions in the microwave regime (4, 9, 10). So far, memories based on macroscopic rare-earth crystals have been realized with electromagnetically induced transparency (11), gradient echo, controlled reversible inhomogeneous broadening (CRIB) (12), and atomic frequency comb (AFC) protocols (3, 13). REI-doped waveguide quantum memories have also been developed (6–8), but the macroscopic lengths of these devices are not ideal for on-chip integration. Coherent coupling of a mesoscopic REI ensemble to a nanophotonic cavity has been achieved recently (14, 15). To show the versatile functionality of this nanoscale platform, we demonstrate storage of photonic quantum bits with high fidelity, fast memory initialization, and a method for dynamically controlling the storage time.

Our optical quantum memory is based on a triangular nanobeam photonic crystal resonator (16, 10) fabricated in a Nd:YVO crystal nominally doped at 100 parts per million [materials and methods (17)] (Fig. 1). The device is a one-sided cavity; the input mirror (left side in Fig. 1, B and C) was fabricated using a smaller number of photonic crystal lattice periods and thus has a lower reflectivity. The optical coupling in and out of the device was implemented by means of a 45°-angled coupler at one end of the nanobeam that totally internally reflects light into the device. An aspheric doublet lens matched the mode of the single-mode fiber to the mode of the nanobeam waveguide (Fig. 1A). The coupling efficiency was optimized to 27% (from fiber to waveguide) by positioning the device using a three-axis nano-positioner. The loss of the cavity fundamental mode volume is $V_{\text{mode}} = 0.52(\lambda/\ln)^3 = 0.0564 \text{ mm}^3$ (simulated, where $\lambda$ is the resonance wavelength and $n$ is the effective modal refractive index), with a measured, near-critically coupled quality factor $Q = 3700$ (energy decay rate $\kappa = x_{\text{in}}$ from the input mirror was $\sim 50\%$ of $x_{\text{in}}$ is the in-trapping loss, and $x_{\text{leak}}$ is the leakage through the long mirror. The device was cooled to $-0.5 \text{ K}$ in a helium-3 refrigerator. The laser used to prepare and probe the memory was modulated by two double-pass acousto-optic modulators (AOMs) and two electro-optic modulators (EOMs) connected in series, and it was delivered to the sample by means of a single-mode fiber. The reflected signal from the device was sent via a circulator and an optical switch to either a spectrometer for characterization or an 82%-efficient superconducting nanowire single-photon detector (18) [supplementary text (17)] mounted in the same refrigerator.

Compared with macroscopic bulk memories that rely on long crystals (for large optical depth) to achieve high storage efficiency, memories based on nanocavities exploit the enhanced light-matter interaction at the single-photon level and thus can achieve high efficiency using a very small volume of material containing a mesoscopic ensemble of atoms (19–21). Moreover, to efficiently map a photon in and out of the ensemble, the device needs to be impedance-matched so that the atoms are coupled to the cavity with a collective cooperativity $C = 4N\gamma^2/\kappa_\text{in}$ close to unity (20, 21). Here $g$ is the atom-cavity coupling rate, $\Gamma_c$ is the atomic homogeneous linewidth, and $N$ is the number of atoms per $\Gamma_c$ bandwidth. This impedance-matching condition applies to a pulsed operation to maximize the efficiency of absorbing a pulse in the atomic ensemble. The reflection spectrum of the cavity on resonance with the Nd ensemble shows a center peak (Fig. 2A, red dashed box) resulting from the coupling between the cavity field and the Nd${}^{3+}\,^4I_{15/2}\rightarrow^4I_{17/2}$ transition at 880 nm. From the contrast of the peak, we extracted an effective $C = 0.75$ at the center frequency of the ensemble absorption [materials and methods (17)]. Figure 2B plots the photoluminescence lifetime (optical $T_2$) in the cavity (blue), showing a $1/\epsilon$ decay time of 4.5 $\mu$s that is 20 times as fast as that of the bulk (red). This Purcell enhancement increases the branching ratio of the $Y_{17/2}$-to-$Z_{17/2}$ transition from initially 27% (22) to $\sim 97\%$, which facilitates an efficient A system for optical pumping experiments. The optical coherence time $T_2$ from two-pulse photon echo measurements was $3.1 \pm 0.03 \mu$s [$\Gamma_\text{in} = 1/(\pi T_2) = 100 \text{ kHz}$] for cavity-coupled ions when the cavity was detuned from the ensemble resonance by $\sim 50$ GHz (blue curve in Fig. 2C), which is close to the $3.2 \pm 0.2 \mu$s found for the bulk crystal. When the cavity was on resonance, the strong Purcell enhancement modified the $T_2$ to $T_2^{\text{cav}(\text{on})} = 2.3 \pm 0.3 \mu$s. With an inhomogeneous broadening of $\sim 2$ GHz, our nanocavity contained a total of $4 \times 10^5$ ions, with a peak spectral density of $N = 20$ ions per $\Gamma_c$ estimated from the cooperativity and a maximum single-photon Rabi frequency $\hbar \gamma_{\text{max}} = 2\pi \times 30 \text{ MHz}$ (22).

Ensemble-based optical quantum memory protocols such as AFC and CRIB require preparing high-contrast spectral features within the inhomogeneous profile by means of persistent hole-burning. In REI-doped crystals, this preparation step is typically slow because of the long excited-state optical lifetimes $T_1$. Additionally, the resultant spin polarization, measured by the ratio of two spin-ground-state populations, can be poor for Kramers ions that, compared with non-Kramers ions, have shorter Zeeman lifetimes $T_1$ relative to optical $T_2$ (e.g., erbium) (23). Here we show that the nanocavity increases the optical pumping efficiency and spin polarization by enhancing the spontaneous decay rate from excited states to the Zeeman ground state.

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**Quantum Optics**

**Nanophotonic rare-earth quantum memory with optically controlled retrieval**


Optical quantum memories are essential elements in quantum networks for long-distance distribution of quantum entanglement. Scalable development of quantum network nodes requires on-chip qubit storage functionality with control of the readout time. We demonstrate a high-fidelity nanophotonic quantum memory based on a mesoscopic neodymium ensemble coupled to a photonic crystal cavity. The nanocavity enables $>95\%$ spin polarization for a high-fidelity nanophotonic quantum memory based on a mesoscopic neodymium ensemble and time-bin-selective readout through an enhanced optical Stark shift of the comb frequencies. Our solid-state memory is integrable with other chip-scale photon source and detector devices for multiplexed quantum and classical information processing at the network nodes.
Fig. 1. Schematics of the experiment. (A) The Nd:YVO device was cooled to 480 mK in a helium-3 refrigerator. Laser light was modulated by a combination of AOMs and EOMs and was delivered to the device with a single-mode fiber. An aspheric doublet lens focused the fiber output to the nanobeam coupler (close-up view shown in the red dashed box). The reflected light was sent either to a room-temperature apparatus for characterization or to a WSi superconducting nanowire detector in the same refrigerator. Scale bar, 1 μm. a, b, and c are crystallographic axes. SNSPD, superconducting nanowire single-photon detector; MEMS, micro-electromechanical system; SMF, single-mode fiber; pol., polarization; PM, phase modulation; AM, amplitude modulation. (B) A scanning electron microscope image of the one-sided nanobeam optical cavity. (C) Simulated fundamental transverse-magnetic (electric field polarization along c) mode profiles. The black dashed line defines the input-output boundary for characterizing the memory device efficiency.

Fig. 2. Cavity-enhanced impedance-matched light-matter interface. (A) Cavity reflection spectra for an empty cavity (dotted blue line), frequency shifted to overlap with the on-resonance spectrum and a cavity on resonance with the Nd transition (solid blue line). The peak (red dashed box) results from collective coupling of the Nd ensemble with the cavity. The flat spectrum (brown line) is the reflection from the right coupler, indicating that the cavity is dominantly one-sided. λ, wavelength. (B) Photoluminescence (PL) decays (optical $T_2^c$) for ions coupled to the nanocavity (blue) and in the bulk crystal (red). The decay in the nanocavity is not a single exponential (inset), because the ions randomly distributed in the cavity experience different Purcell enhancement. $t$, time; a.u., arbitrary units. (C) Two-pulse photon echo decays in the nanocavity that is on resonance with the ensemble (gray), detuned by ~50 GHz (blue), and in the bulk (red). The inset shows a typical photon echo signal from the nanocavity. The vertical axis is in linear scale and arbitrary units. The horizontal axis is in microseconds. $\tau$, delay between the first and second pulse.

The experimental magnetic field ($B = 340$ mT) configuration is illustrated in Fig. 3A. The misalignment of the magnet with respect to the sample caused a slight deviation $\theta$ of the field orientation from the c axis (crystal symmetry axis). From the bulk optical pumping measurement [supplementary text (I7)], we estimated this deviation to be $\sim 8.2^\circ$. A representative spectrum for the four transitions is plotted next to the Zeeman-level splitting (24) in Fig. 3A. The spin-preserving transitions a and c overlapped, which favored higher optical depth and better impedance-matching when using their overlapped spectral region as a photon-atom interface. The b and d transitions had smaller branching ratios, but both were enhanced, similar to the a and c transitions in the cavity. Using the pulse modulation sequence shown in the inset of Fig. 3C, we verified that the Zeeman spin lifetime in the cavity, $T_2^c = 12.5 \pm 1.0$ ms, was not degraded relative to the bulk, $T_2^{\text{bulk}} = 12.7 \pm 1.5$ ms (Fig. 3B). Figure 3C plots the spin population as a function of optical pumping time $t_{\text{pump}}$. In the nanocavity, a maximum spin polarization of $p_2/p_1 = 20$ ($p_1 < 5\%$) was achieved with $t_{\text{pump}}$ less than 1 ms, whereas in the bulk, it took more than 10 ms to reach $p_2/p_1 = 3$. This enhancement can be understood from a rate-equation model taking into account the field angle–dependent branching ratios given by the spin Hamiltonian (23, 24) [supplementary text (I7)].

To demonstrate storage of photonic qubits by means of the AFC protocol, we sent a sequence of 10-ns pulse pairs separated by a time interval $t = 1/\Delta$, where $\Delta$ is the frequency spacing of the
cavity as the probability that an AFC echo photon (ods (measured to characterize the AFC efficiency and to the comb and the background, respectively. a time-bin state

Fig. 3. Efficient optical pumping and quantum storage in a Nd:YVO nanocavity. (A) Magnetic field configuration with Zeeman spin levels and transitions (a to d) of Nd3+ (only isotopes with 0 nuclear spins). (B) Spectral hole decays from which spin lifetimes were extracted. $t_{\text{pump}}$, wait time between optical pumping and the probe pulse. (C) Optical pumping dynamics showing an enhanced spin polarization in the nanocavity [$p_t = 5\%$ (blue), versus $-20\%$ (red) in the bulk]. The inset shows the AOM modulation sequence. The bulk spin population was measured by the transmission of the probe pulse (red). The cavity was probed using photoluminescence counts after the probe excitation (blue). (D) An AFC with $F = 3.3$ and $\sim400$ ions in each tooth. Gray, 100-kHz resolution; black, 500-kHz resolution. (E) Input (black line), reflected (blue area), and AFC echo signal of a coherent state (mean photon number $\alpha = 0.58$) time-bin mode $\{|+/\rangle\}$ from the nanocavity. The inset shows the lower bounds on the qubit storage fidelity for a set of inputs, with an arithmetic mean fidelity of 96.8%. Error bars, standard deviation.

The ability to store photons in multiple time bins and to retrieve one at an arbitrary bin is a key functionality in proposed multiplexed quantum repeater networks (25). Upon successful Bell-state measurement, a feedforward signal will be sent to retrieve the stored photon at a desired time bin for subsequent entanglement swap. Here we demonstrate time bin–selective readout of a coherent pulse, using the cavity-enhanced ac Stark shift. Two Stark pulses detuned at $\pm \Delta_{\text{st}} = 1$ GHz ($\Delta_{\text{st}} > \Delta$) from the center of the AFC [supplementary methods (17)] uniformly compressed the comb spacing by $\delta \Delta = 4\Delta_{\text{st}}(\Delta_{\text{st}}^2 + r^2)\Delta (27)$ (Fig. 4A) over the Stark pulse duration of $t_{\text{st}} = 16$ ns (where $n_{\text{st}}$ is the average number of photons in the cavity while the Stark pulse passes through it, and $g_{\text{st}}$ is the Stark Rabi frequency per single photon in the cavity). This comb compression resulted in an additional delay of the echo by $\delta t_{\text{echo}}/\Delta$. Figure 4C plots the measured echo retrieval time with increasing Stark pulse intensities from $n_{\text{st}} = 0$ to $\sim 170$ photons per pulse. The relative timing of the echo peaks is plotted against the Stark pulse photon number in Fig. 4D, with a linear fit of $\sim 50$ ps per photon (red dashed line). As the Stark pulse intensity increased, the echo efficiency gradually dropped (Fig. 4E), owing to broadening and distortion of the AFC teeth as a result of inhomogeneous ac Stark frequency shifts produced by random locations of ions in the cavity. Nevertheless, the echo signal (Fig. 4C) was still $12.3$ dB above the background at the highest Stark pulse intensity. The maximum delay of the measured echo

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was $10 \pm 1$ ns, which is comparable to the FWHM of the echo pulse—that is, a time bin. In the current device, inhomogeneous Rabi frequencies cause a smearing of the comb teeth while the Stark pulses are applied. This results in a weaker echo, thus limiting the maximum time shift without severely attenuating the echo intensity. Such limitations can be lifted if ions are selectively doped at the cavity antinodes by site-controlled implantation (28). Alternatively, isolating subensembles with more homogeneous Rabi frequencies could be possible by optical pumping with repeated $2\pi$ pulses, as demonstrated in (29).

The nanocavity scheme demonstrated here enables versatile engineering of the quantum light-matter interface and offers the distinctive advantage of faster and more efficient memory preparation. An on-demand quantum memory will require the incorporation of the spin-wave storage in long-lived hyperfine levels of REIs (30) or the use of controlled reversible inhomogeneous broadening. Both of these can benefit from the nanoscale platform—for instance, by adding micro-wave striplines or microelectrodes in proximity to the nanocavity device for control of the rare-earth spins. With advances in nanofabrication of the REI host crystals, medium- to large-scale quantum memory arrays can be envisioned, which would enable highly multiplexed repeater schemes. Furthermore, our platform also allows direct integration with single rare-earth qubits (31) and interfacing to superconducting quantum devices (32).

REFERENCES AND NOTES
5. W. Tittel et al., Laser Photonics Rev. 4, 244–267 (2010).
17. See the supplementary materials.

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SUPPLEMENTARY MATERIALS
www.sciencemag.org/content/357/6358/1392/suppl/DC1
Materials and Methods
Supplementary Text
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Fig. 4. Temporal-mode selective pulse retrieval using ac Stark pulses. (A) Schematic spectral configuration of two Stark pulses with respect to the AFC comb. The symmetric, far-detuned Stark pulses induce uniform compression of the AFC. (B) Stark pulses cause an additional delay in echo retrieval times. (C) Measured AFC echoes with increasing photon number in 16-ns Stark pulses. Here $A_{\text{det}} = 1$ GHz and $\Delta = 13.3$ MHz. The blue shaded area overlays the echo envelope with no Stark pulses. (D) AFC echo delay against Stark pulse intensity. A linear fit (red dashed line) corresponds to 50 ps per photon. (E) Decrease of AFC echo efficiency with Stark pulse intensity, caused by AFC distortion resulting from inhomogeneous Stark shifts in the nanocavity.
A rare-earth quantum memory

The development of global quantum networks will require chip-scale optically addressable quantum memories for quantum state storage, manipulation, and state swapping. Zhong et al. fabricated a nanostructured photonic crystal cavity in a rare-earth-doped material to form a high-fidelity quantum memory (see the Perspective by Waks and Goldschmidt). The cavity enhanced the light-matter interaction, allowing quantum states to be stored and retrieved from the memory on demand. The high fidelity and small footprint of the device offer a powerful building block for a quantum information platform.

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