

Cavity-Enhanced Spin-Wave Solid-State Quantum Memory

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We report on the realization of an efficient solid-state spin-wave quantum memory, with on-demand readout, using the full atomic frequency comb (AFC) scheme in a $\text{Pr}^{3+}:\text{Y}_2\text{SiO}_5$ crystal embedded in an impedance-matched cavity. We demonstrate operation at the single-photon level by storing weak coherent states with an efficiency up to $(40 \pm 2)\%$ and a signal-to-noise ratio of 14 for an input photon number of 0.42 photons per pulse. We also investigated the enhancement of the incoherent noise due to the impedance-matched cavity and characterized the quantum memory performance, showing a two-way transfer from excited to spin states and back of up to 83%. Finally, we confirmed the quantum nature of our memory by storing nonclassical states of light, i.e., a heralded single photon from a nondegenerate spontaneous parametric down-conversion source, and achieved nonclassical correlations between the heralding and the stored-and-retrieved photon. These results demonstrate that impedance-matched AFC spin-wave quantum memories with on-demand readout can be used for experiments involving the storage of photonic quantum states. They also open the door to solid-state on-demand quantum memories with very high efficiencies, serving as a key resource for quantum networks and quantum repeaters.

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One of the key challenges in building large quantum networks [1] is the implementation of quantum memories that can store quantum information in the form of photons and retrieve it at a later time in an on-demand fashion. This technology would enable the realization of quantum repeaters [2,3] and unlock long-distance distribution of entangled states. The requirements for quantum memories to fulfill this promise as part of a quantum repeater include high efficiency, high fidelity, long storage time, and large multiplexing capacity. Although so far no system has been demonstrated that fulfills all these requirements at the same time, many implementations across a variety of systems have been reported, including cold atomic clouds [4–10], warm atomic vapors [11–16], nanomechanical resonators [17], and rare-earth-doped solids. Single quantum systems such as single atoms [18,19], vacancy centers in diamonds [20–22], and single trapped ions [23–26] have also been shown to be viable quantum memories. Up to now, cold

atomic gases have reported the highest storage and retrieval efficiency, around 90% [6,7,10].

Rare-earth-ion-doped crystals are a highly promising platform for quantum memory applications, as they enable ensemble-based storage without requiring complex laser cooling and trapping techniques. These crystals inherently provide a large ensemble of atoms with optical and spin transitions embedded in a solid-state matrix. At cryogenic temperatures, the ions exhibit long coherence times for both optical and spin transitions, enabling demonstrations of extremely long storage durations with bright classical light [27–29] and up to 1 s for light at the single-photon level [30,31]. High efficiencies for storage in the excited state at the single-photon level have been demonstrated as well [32,33]. This system also offers a large multiplexing potential due to its static inhomogeneous broadening, spanning several GHz [34,35] and providing a resource for temporal and spectral multiplexing [36]. In particular, the atomic frequency comb (AFC) protocol [37,38], which uses a comb-shaped structure prepared in the inhomogeneous broadening, inherently supports temporal multimodality [34,35,39]. Furthermore, frequency [35,40,41] and spatial [42,43] multiplexing have also been demonstrated using the AFC protocol.

To date, the AFC protocol is the only one that has demonstrated storage of externally generated single and entangled photons in rare-earth-doped crystals. This capability has facilitated key demonstrations of quantum

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repeater components, including entanglement between quantum memories and telecom photons [44–48], light-matter quantum teleportation [49,50], and heralded entanglement between spatially separated quantum memories [39,51–53]. The intrinsic temporal multimodality of the AFC protocol significantly enhances entanglement distribution rates [34,39,54] in quantum networks and naturally supports the storage of states encoded in time bins. However, the AFC efficiency is typically constrained by the optical density of the material, which is difficult to increase in millimeter- or centimeter-long crystals. Theoretically, the forward retrieval efficiency is limited to (54%) [37] due to reabsorption. This limitation can be overcome using the cavity-assisted AFC protocol, where the crystal is placed within a low-finesse cavity operating in the impedance-matched regime [55].

In recent years, several experiments have employed this technique, achieving efficiencies up to 56% with classical pulses [56,57] and up to 62% [33] at the single-photon level [58]. These experiments used only excited-state storage, resulting in preprogrammed storage times. To gain access to longer storage times and on-demand readout, optical control pulses can be used to transfer the excitations to spin states. While progress is ongoing at the quantum level in free space, both for AFC [46] and other protocols [59,60], the only experiment to date that has employed spin-wave storage in a cavity-enhanced solid-state system [57] used classical input pulses at an efficiency of 12%. Performing spin-wave storage at the single-photon level is considerably more challenging, requiring low noise levels to enter the quantum regime. In particular, it has remained so far unclear whether the noise collection enhancement resulting from the presence of the cavity would prevent the storage of single photons with high signal-to-noise ratio in an impedance-matched cavity solid-state memory.

In this Letter, we demonstrate a spin-wave cavity-enhanced quantum memory with on-demand readout. We report the highest spin-wave storage (SWS) efficiency to date in a solid-state system, reaching up to $(40 \pm 2)\%$ for weak coherent pulses at the single-photon level, together with a signal-to-noise ratio of 33 for input pulses containing one photon on average. We also thoroughly characterized the storage process and noise generation, which are both enhanced by the presence of the cavity, but still allow for nonclassical correlations. Finally, we confirmed the quantum nature of the memory by storing heralded single photons from a cavity-enhanced spontaneous parametric down-conversion (SPDC) source with a storage efficiency of $(18.2 \pm 1.4)\%$ and a cross-correlation value of (6.3 ± 0.8) .

We used a $\text{Pr}^{3+}:\text{Y}_2\text{SiO}_5$ crystal as quantum memory, specifically a 3-mm-long crystal placed inside a Montana Cryostation cryostat at 3.4 K. The relevant level scheme is shown in Fig. 1(a). An AFC consisting of a series of narrow periodic absorption peaks was prepared on the $\pm 1/2g - \pm 3/2e$ transition of the $^3H_4 - ^1D_2$ manifold. Input pulses

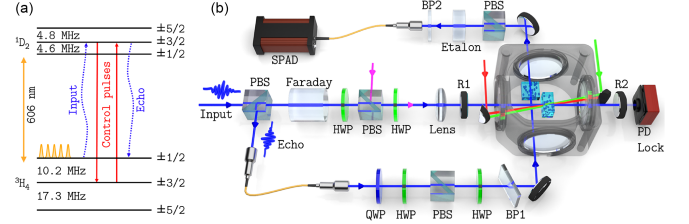


FIG. 1. (a) Energy level scheme. The $^3H_4 - ^1D_2$ transition of $\text{Pr}^{3+}:\text{Y}_2\text{SiO}_5$. The optical transition, where the AFC is prepared, is $\pm 1/2g - \pm 3/2e$. (b) Schematic of the experimental setup for spin-wave storage. The pulses come from the left side and are spatially separated from the photons that were reflected or stored in the memory using a Faraday rotator and polarizing beam splitter (PBS). The retrieved photons are then fiber coupled and sent through a 2-MHz-wide spectral pit in the filter crystal before detection (the filter crystal preparation beam is not shown). Two auxiliary beams off axis with respect to the cavity were used for the memory preparation (green beam) and operation via the application of control pulses (red beam). Definitions: R1/R2, cavity mirrors; Faraday, Faraday rotator; HWP, half-wave plate; PD, photodiode; QWP, quarter-wave plate; SPAD, single photon avalanche detector; BP1, bandpass filter (center 610 nm, FWHM 10 nm); BP2, bandpass filter (center 600 nm, FWHM 20 nm).

can then be stored in the AFC with a preprogrammed storage time τ_{AFC} or converted to a spin wave on the $\pm 3/2g$ state using strong optical control pulses and later retrieved on demand. To maximize transfer efficiency, the control pulses were chirped over 1.06 MHz following a hyperbolic tangent profile. A sketch of the setup is shown in Fig. 1(b). The photons retrieved from the quantum memory were fiber coupled and sent through a 2-MHz-wide spectral pit in a second $\text{Pr}^{3+}:\text{Y}_2\text{SiO}_5$ crystal (filter crystal) before detection by single-photon detectors.

The asymmetric cavity is formed by two mirrors with reflectivities $R_1 = 0.4$ and $R_2 = 0.99$ and curvatures of 125 and 100 mm, respectively, resulting in an empty cavity finesse $F_{\text{cav}} = 6.5$. The 208-mm-long cavity is built around the vacuum chamber, while the preparation and spin-wave transfer (control) beams are overlapped via d-shaped mirrors and enter the crystal at an angle of about 5° . R_1 and the sample thickness were chosen to satisfy the impedance-matching condition $R_1 = R_2 e^{-2\tilde{d}}$. Here, R_2 is the reflectivity of the second mirror, and $\tilde{d}$ is the average optical depth of ions in the comb region given by Eq. (1) as $\tilde{d} = d/\mathcal{F}_{\text{AFC}}$ [37]. In this case, d is the optical depth of the crystal and $\mathcal{F}_{\text{AFC}} = \Delta/\gamma$ is the comb finesse, the ratio between the comb tooth spacing $\Delta = 1/\tau_{\text{AFC}}$ and their width γ . When impedance matching is achieved and spatial mode matching is perfect, the input pulse can be completely absorbed by the sample and, assuming square-shaped peaks, the efficiency can be modeled as [55]

$$\eta_{\text{cav}} = \frac{4\tilde{d}^2 R_1 \text{sinc}^2(\pi/F_{\text{AFC}})}{(1 - R_1)^2} \eta_i e^{-\frac{(\tau_s \gamma_{\text{inhom}} \pi)^2}{2 \log(2)}}. \quad (1)$$

The first fraction refers to the AFC efficiency in the cavity, while η_t^2 denotes the transfer efficiency to the spin state $3/2_g$ and back. The Gaussian decay models spin-state dephasing due to the spin-inhomogeneous broadening γ_{inhom} during spin-wave storage for time τ_s [46]. Intracavity round trip loss ϵ can be considered by replacing R_2 with $R_2 - \epsilon$ to model a realistic experiment [57]. In our case, an intracavity round trip loss of $\epsilon = 7\%$ was inferred.

Each experimental cycle comprised two phases: (i) A 600 ms preparation phase, during which an AFC with $F_{\text{AFC}} = 3.3$ was prepared using a coherent technique [61] and the cavity length was stabilized using a 960-MHz-detuned beam, orthogonally polarized to the input beam to avoid interaction with Pr^{3+} ions; (ii) the measurement phase, during which 150–500 weak coherent pulses were sent with 300 μs spacing to allow fluorescent noise to decay. Detector clicks and trigger signals heralding the pulses were time tagged. Typically, the first 75 pulses of the pulse train were used for data analysis because the memory changed its characteristics due to the effect of the control pulses on the residual population in the $3/2_g$ state (Supplemental Material [62]). The experimental sequence was repeated at about 1 Hz and synchronized with the cryostat cycle to prevent vibrations from disturbing the spectral structure and impairing the performance of the memory [63]. To mitigate slow experimental drifts, we collected all the required data to extract the various efficiencies and noise in the same experimental run by sequentially using six different measurements, with or without inputs and with or without control pulses. The AFC efficiency was optimized using a machine learning algorithm [62].

We now turn to the storage experiments. We started by storing weak coherent pulses with a duration of 1 μs FWHM and a mean value of 0.42 photons per pulse measured before the cavity. Using the AFC protocol with $\tau_{\text{AFC}} = 8 \mu\text{s}$, we obtained a storage and retrieval efficiency of $(51.5 \pm 0.8)\%$ in the excited state. We then applied two control pulses separated by 4.5 μs (peak to peak) to transfer the excitations to the $3/2_g$ state and then back to the excited state. Figure 2 (top) shows a detection time histogram of this measurement, yielding a spin-wave efficiency of $(40 \pm 2)\%$, which represents, to our knowledge, the highest spin-wave storage efficiency to date for any solid-state quantum memory. The efficiency can be interpreted as the device efficiency, where the quantum memory is formed by the cavity surrounding the vacuum chamber of the cryostat, cooling the $\text{Pr}^{3+}:\text{Y}_2\text{SiO}_5$ crystal.

The measured signal-to-noise ratio (SNR) was 14 ± 3 , corresponding to $\mu_1 = \mu_{\text{in}}/\text{SNR} = 3\%$. Thus, a heralded single photon source with just 3% heralding efficiency would yield $\text{SNR} = 1$ using this memory. A plot of the spin-wave storage efficiency as a function of time between the control pulses is shown in Fig. 2 (bottom). We observed a decay of the efficiency as a function of storage time with a

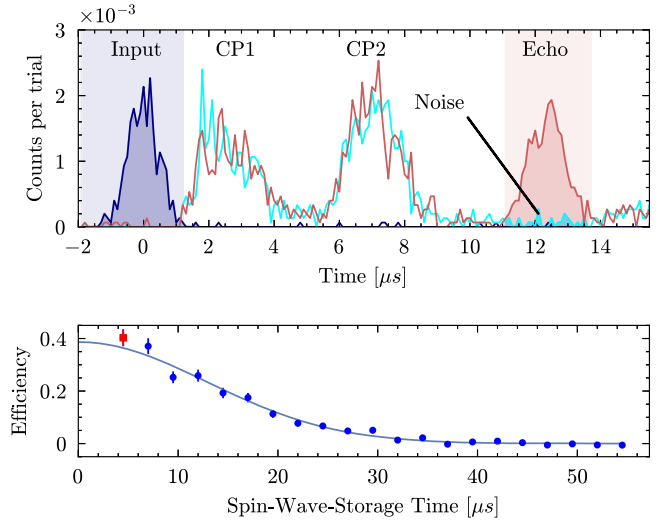


FIG. 2. Top: input pulse (blue) and its echo (red pulse) after 12.5 μs . The recorded input pulse with a mean photon number of 0.42 photons per pulse and length of 1 μs FWHM was reflected from the first cavity mirror with 40% reflectivity. Taking into account the reflectivity, the ratio between the echo and the input pulse yields $(40 \pm 2)\%$ storage efficiency. The noise measurement (cyan) was taken by sending the control pulses without an input. The efficiency was estimated by comparing the signal from the reflection of the cavity mirror with the AFC or SWS echo. The control pulse energy was 53 nJ per pulse for a pulse duration of 1.5 μs FWHM. The detection window size for the echo and the noise is 2.5 μs ; the bin size of the histogram is 100 ns. Bottom: scan of the memory efficiency as a function of the storage time in the spin state. The decay is fitted with a Gaussian function, allowing us to determine the spin-inhomogeneous broadening to (20.5 ± 0.5) kHz, which is close to the previously observed value of 16.1 kHz [46]. The AFC efficiency during this scan was $(51.5 \pm 0.8)\%$ and the first point of the series (red square) corresponds to the measurement displayed in the top graph.

$1/e^2$ decay time of 26 μs , due to the inhomogeneous broadening of the spin transition.

By measuring the depletion of the AFC echo with the first control pulse, we inferred a transfer efficiency from the excited state to the $3/2_g$ state of $\eta_{t1} = (94.5 \pm 2.2)\%$ for the measurement with the shortest spin-wave storage time reported in Fig. 2. Taking into account the decay due to the spin-inhomogeneous broadening, the combined transfer efficiency to the spin state and back to the excited state was $\eta_t^2 = (83 \pm 9)\%$, among the highest demonstrated in this system [64]. Further improvement is possible by optimizing the pulse shape or increasing the pulse length [64].

These results show that the spin-wave quantum memory can operate with a high signal-to-noise ratio at the single-photon level. However, the noise remained significant, especially compared to the essentially noiseless excited-state storage of the AFC. We attribute part of this noise to scattering of the control pulses, which are deflected into the cavity mode, and to free induction decay of off-resonantly excited ions. This coherent noise, at the control pulse

frequency, could, in principle, be blocked by the filter crystal, prepared with a 2-MHz-wide spectral pit. Additional incoherent fluorescence arose from imperfect spectral preparation, e.g., residual population in the $3/2_g$ spin state, leading to an incoherent fluorescence emission which decayed over the next ~ 160 μ s, partially emitting into the cavity mode at the frequency of the input photons.

To show the effect of the cavity on the observed noise, we measured the detected noise and memory efficiency as a function of cavity detuning, which can be seen in Fig. 3. A large noise detection window of 260 μ s was chosen to accumulate good statistics. We fitted an exponential decay to the noise histograms and extracted the noise amplitude from the fit (Supplemental Material [62]). The cavity enhanced the incoherent noise resonant with the $1/2_g \leftrightarrow 3/2_e$ transition by a factor of 1.4 compared to the case without cavity, which is less than the expected enhancement of $4F/\pi \sim 4.3$ [65], given that the cavity has a finesse F of about 3.4 when the impedance-matching condition is fulfilled. One possible cause for the lower noise enhancement could be a constant background fluorescence emitted by the filter crystal caused by control pulse leakage, which is independent of the cavity detuning. The spin-wave and

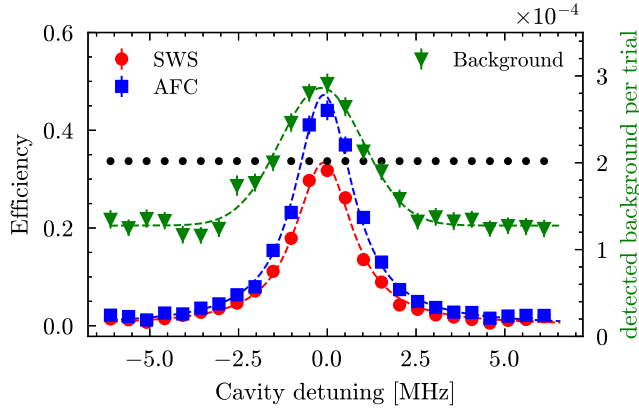


FIG. 3. Scan of the AFC and SWS efficiency as well as the background noise as a function of the cavity detuning. Black dotted line indicates noise level measured in single pass without a cavity. The background represents the probability of a noise click per trial in a 2 μ s bin at the echo time. It is extrapolated from an exponential decay fit to the dark-count corrected histogram (bin size 2 μ s) in a 260- μ s-long window beginning 22 μ s after the SWS echo. For a better statistical significance, all 500 pulses per experimental cycle were summed for the noise curve, while the efficiencies were extracted from the first 75 pulses of each cycle. For each cavity detuning step, 2625 one-second cycles were conducted to assess the background noise and SWS efficiency, and 875 cycles were conducted to evaluate the AFC efficiency. The data for both efficiencies were fitted with Voigt curves with a fixed standard deviation of the normal distribution given by the spectral width of the input pulse of 440 kHz. The noise was fitted with a Gaussian with a FWHM of 2.8 MHz, yielding a noise level increase by about 1.4 times when the cavity is on resonance with the stored photon.

AFC efficiency follow a Voigt profile, resulting from the convolution of the Gaussian pulse of 440 kHz FWHM and the Lorentzian cavity resonance. The fit with a Voigt profile yields a total FWHM of 1.96 MHz with a Lorentzian cavity linewidth of 1.86 MHz.

Finally, to confirm the quantum nature of our memory, we stored heralded single photons generated by a cavity-enhanced spontaneous parametric down-conversion source [66]. The source emitted narrow band nondegenerate photon pairs with one photon resonant with the memory and the other photon at a wavelength of 1552 nm. The detection of a telecom photon heralded the presence of a photon at 606 nm in front of the memory, with a heralding efficiency of 25%. We inferred a photon linewidth of about 2.1 MHz and a heralded autocorrelation of $g_{i;s,s}^{(2)} = 0.06 \pm 0.01$ for the input field, confirming that the source indeed emits heralded single photons. Because of the unknown pair-creation time in a continuous-wave pumped SPDC process, the control pulses were sent conditioned on the detection of an idler photon. With heralded single photons, an AFC efficiency of $(30.6 \pm 1.8)\%$ was reached with a detection window size of 1.1 μ s. Spin-wave storage trials yielded an efficiency of $(18.2 \pm 1.4)\%$, with a coincidence histogram shown in Fig. 4. These values are lower than those for weak coherent pulses, mostly because of the bandwidth mismatch between the single photons and the quantum memory; however, they are compatible with the expected efficiency given the bandwidth of the photons [62]. The bandwidth mismatch is also visible in the red

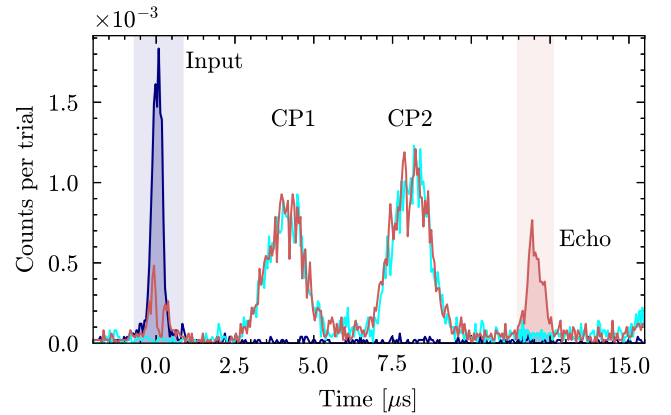


FIG. 4. Coincidence histogram showing the storage of heralded single photons per trial. A trial corresponds to the detection of an idler within the measurement window. Control pulses were sent conditionally upon idler detection. The efficiency observed was $(18.2 \pm 1.4)\%$, yielding a cross-correlation $g^{(2)} = (6.3 \pm 0.8)$ for a detection window of size 1100 ns. The bin size of the histogram is 50 ns. The 40% reflection from the first cavity mirror (blue) is labeled as “Input” and the spin-wave echo (red) as “Echo.” The leakage from the control pulses is labeled as “CP1” and “CP2.” A measurement with no input photon is shown in cyan. The heralding efficiency of the source was 25% and the pump power was 150 μ W. The data were collected in 30 min.

trace shown in Fig. 4—the input photon is broader than the cavity and is therefore partially reflected, in contrast to the measurement shown in Fig. 2, where this reflection is nearly fully suppressed. The observed cross-correlation was $g_{i,s}^{(2)} = (6.3 \pm 0.8)$. Selecting a smaller detection window increases the signal-to-noise ratio; for example, with a 480 ns detection window, we obtain $g_{i,s}^{(2)} = (10 \pm 2)$ at an efficiency of $(13.3 \pm 1.1)\%$. A value of $g_{i,s}^{(2)} > 5.8$ enables the violation of a Clauser-Holt-Shimony-Horne inequality when entangled qubits are stored, showing that our memory could, in principle, be used to store and preserve entanglement. Using the model from [67], given the signal-to-noise ratio of our system observed during the storage of single photons, we extrapolated that the photons at the output of the memory will have a heralded autocorrelation of 0.28 ± 0.03 , significantly below the bound of 0.5. This confirms that our memory preserves the single-photon character and therefore performs as a genuine quantum memory.

In summary, we have demonstrated the first cavity-enhanced spin-wave quantum memory in a rare-earth-doped solid, leading to the highest spin-wave storage efficiency at the quantum level demonstrated so far in this platform. Further improvements can be expected by reducing the losses inside the cavity and by increasing the finesse of the comb and cavity. A reduction from the current round trip losses down to 1% (using, e.g., low-loss random antireflection coatings or shorter crystals) together with a small increase in AFC finesse up to 3.8 would lead to a spin-wave storage efficiency of 58%, according to Eq. (1). Increasing further both the comb and cavity finesse to 5.8 and 16, respectively (Supplemental Material [62]), would lead to an efficiency close to 80%. A shorter crystal would mitigate the slow-light effect caused by steep absorption changes, increasing the memory bandwidth, thereby improving the storage efficiency for heralded single photons. With these improvements in place, this system is a promising building block for the realization of quantum repeaters, combining the unprecedented multiplexing capabilities of rare-earth-doped crystals with high-efficiency storage in a solid-state platform.

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Data availability—The data that support the findings of this Letter are openly available [75].

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