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Improved light-matter interaction for storage of quantum states of light in a thulium-doped crystal cavity

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We design and implement an atomic frequency comb quantum memory for 793-nm wavelength photons using a monolithic cavity based on a thulium- (Tm-) doped $\text{Y}_3\text{Al}_5\text{O}_{12}$ crystal. Approximate impedance matching results in the absorption of 90% of input photons and a memory efficiency of $(27.5 \pm 2.7)\%$ over a 500-MHz bandwidth. The cavity enhancement leads to a significant improvement over the previous efficiency in Tm-doped crystals using a quantum memory protocol. In turn, this allows us to store and recall quantum states of light in such a memory. Our results demonstrate progress toward efficient and faithful storage of single-photon qubits with a large time-bandwidth product and multimode capacity for quantum networking.

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I. INTRODUCTION

Quantum memories that can map fast moving quantum states of light reversibly onto matter are an invaluable component of future quantum networks [1]. These light-matter interfaces increase the efficiency of complex quantum networking schemes and allow network tasks to be accomplished over long distances and via error correction and local processing [2]. For instance, quantum memories for light play a crucial role in the efficient distribution of entanglement over long distances thereby securing classical communication by means of quantum key distribution and allowing the lossless distribution of qubits through quantum teleportation. Common to all these applications is the need for efficient quantum memories that can store many qubits encoded into different modes of light, which is often referred to as the multiplexing capacity or time-bandwidth product [3].

For quantum networking, it has been shown that the rate of entanglement generation or complex multiphoton state generation scales with the product of memory efficiency and time-bandwidth product [4,5]. The intuition behind this follows from that of classical communications: Higher efficiency devices allow more equipment to be connected over greater distances before loss takes a toll. Similarly, communication rates scale with the number of temporal and spectral channels, e.g., a memory's multiplexing capacity or time-bandwidth product [6–8]. Finally, quantum memories for light should feature sufficiently large bandwidths to interface with a diverse range of single-photon sources, including spontaneous

parametric down-conversion (SPDC), quantum dots, and single molecule emitters [9–11].

Starting with the description of suitable quantum storage protocols around 15 yr ago [12,13], cryogenically cooled rare-earth crystals have rapidly demonstrated their potential as suitable storage materials. In particular, in conjunction with the so-called atomic frequency comb (AFC) protocol [14], they have allowed storing nonclassical states of light, such as single and entangled photons [15–19]. In order to increase efficiencies to values close to 100%, there has been a push towards cavity-enhanced light-matter interaction [20]. A lot of progress has been reported towards this end [21–23], but cavity-enhanced storage of nonclassical light remains to be demonstrated, in part, due to the limited available memory bandwidth. Here, we demonstrate a 500-MHz broad quantum memory with efficiency up to 27.5% as well as high-fidelity storage of heralded single photons over more than 1.5-GHz bandwidth using an impedance-matched cavity. Our findings further support the potential of rare-earth crystals to meet the stringent demands of future quantum networks for memories that allow quantum state storage.

II. EXPERIMENT

The memory cavity is made of a $l = 4$ -mm-long 0.1% thulium (Tm): $\text{Y}_3\text{Al}_5\text{O}_{12}$ (Tm:YAG) crystal and placed in a cryostat operating at a temperature of 600 mK. The end facets of the crystal are reflection coated with reflectivities $R_2 = 99\%$ on the rear and $R_1 = 40\%$ on the front sides. The reflectivity value for the front facet is chosen to allow for impedance matching at the 793-nm Tm:YAG absorption wavelength by meeting the condition $R_1 = R_2 e^{(-2\alpha l)}$ with α as the average absorption coefficient across the cavity resonance bandwidth [20]. These coatings create a planar optical cavity with a free spectral range of 20 GHz and a finesse of seven.

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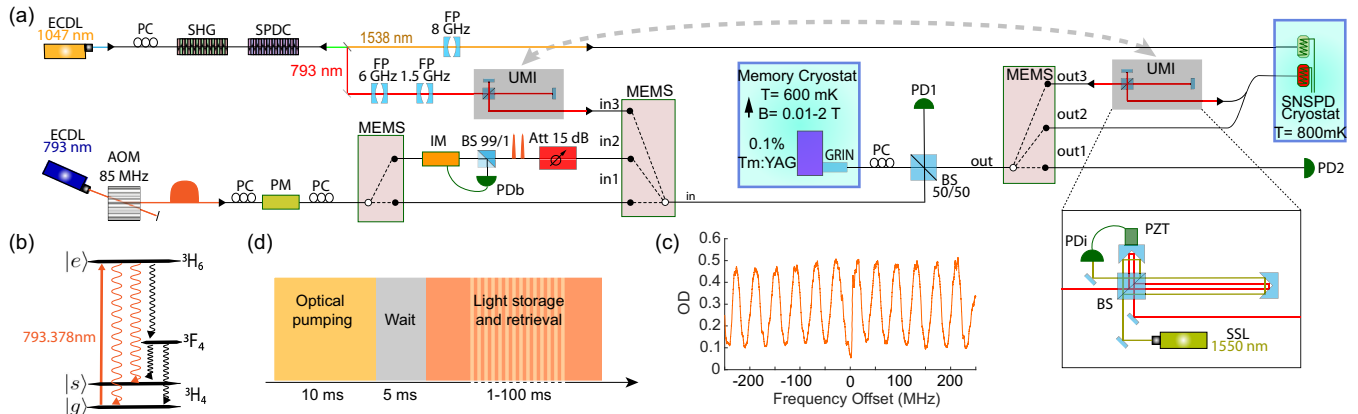


FIG. 1. (a) Schematic of the experimental setup; FP: Fabry-Perot filter cavity; ECDL: external cavity diode laser; SSL: solid-state laser; SHG: second-harmonic generation; SPDC: spontaneous parametric down-conversion; FP: cavity filter; BS: beam splitter; PC: polarization controller; GRIN: gradient index lens; UMI: unbalanced Michelson interferometer; PZT: Piezoelectric actuator; Att: optical attenuator; AOM: acousto-optic modulator; PD: photodiode; MEMS; SNSPD: superconducting nanowire single-photon detectors; IM: intensity modulator; PM: phase modulator. Light from three paths was directed to an input beam splitter for AFC creation and photon storage. After reemission from the memory, the light was switched between different analyzers. (b) Level structure of Tm:YAG. (c) Example 500-MHz AFC scan of a weak read pulse across the comb. (d) Experimental duty cycle, spectral hole burning time, period of spontaneous emission, and a period for memory use.

The Tm:YAG crystal was cut and mounted such that a magnetic field (aligned along the [001] crystal axis) splits the Zeeman degeneracy for four of the six crystallographic sites equally to create a λ system for optical pumping [24]. Optical access was provided by ferrule-tipped (single-mode) fibers and collimation lensing through the planar crystal cavity, and alignment to the fundamental cavity mode was achieved through nanopositioning stages that allow angular steering of the ferrules. All input signals were routed into the memory by a set of microelectromechanical systems (MEMS) switches and a 50:50 fiber beam splitter, which also allowed collection of the reflected signals [see Fig. 1(a)].

A single AFC memory consists of a spectrally periodic distribution of atomic absorption peaks with spacing δ (see Refs. [14,17,25] for more information). Overlapping an optical signal field with this shaped absorption feature causes the AFC to absorb the input pulse of light. The rare-earth dopants within the crystal are put into an entangled superposition state, primed for reemission after a preset interval $\tau = 1/\delta$. We addressed the rare-earth ion-doped crystal sample using three paths as seen in Fig. 1(a)—one for creation of the AFC, and two more for delivering various signals for storage. We sculpt this spectral comb feature through optical pumping on the Tm $^3H_6 \rightarrow ^3H_4$ transition, shown in Fig. 1(b), to drive population with transition frequencies matching the comb troughs into magnetically separated Zeeman level $|s\rangle$. When the Zeeman level splitting is matched to the desired comb tooth spacing, a spectral grating with finesse (the ratio between peak spacing δ and peak width) $F_{\text{AFC}} \approx 2$ is prepared without altering the average optical depth. The pumping process, pictured in Fig. 1(d), allowed the creation of memory features between 100-MHz–10-GHz bandwidth with tooth spacing from 4 to 100 MHz. These memories allow storage of 100-ps-10-ns long pulses for times between 10 and 250 ns, limited by a pump laser linewidth. An example of one such comb,

created in an uncoated region of the crystal, is pictured in Fig. 1(c).

The impedance matching scheme relies on interference between an electric field leaking through the front cavity facet after each cavity round trip and the incoming field initially reflected from the front facet. At the impedance matched condition, these fields create perfectly destructive interference resulting in perfect intake of light by the lossy cavity mode. In our case, the engineered loss (in the form of an AFC) guarantees heightened interaction between light and rare-earth ions. In the following, we describe initial characterizations of the memory. First, we examined how cavity resonances interact with the Tm:YAG absorption profile. Using an IM to carve Gaussian pulses of 4-ns duration at 1000-Hz repetition rate, we slowly swept the laser frequency across the inhomogeneous Tm absorption line centered at 793 nm. Shown in Fig. 2(a), the reflected part of the input pulse was detected and normalized to the input pulse intensity. No signal was detected transmitting through the cavity. On resonance with the cavity mode and ~ 4 GHz from the thulium line center, more than 90% of the input energy was absorbed within the rare-earth cavity system.

Next, we created a 500-MHz-wide AFC and moved its center frequency in 500-MHz increments from the Tm:YAG absorption peak past the cavity resonance. For each detuning, we measured the storage efficiency using laser pulses containing many photons. As expected, the optimum central AFC frequency matched the minimum of the cavity’s reflection spectrum. As seen in Fig. 2(b), the absorption and reemission of light then becomes more likely than the reflection of the signal light from the cavity, peaking at a system efficiency of $12 \pm 1\%$. Taking into account 50% loss of the input-output splitter as well as 6% coupling and lensing loss, we find a memory efficiency of $27.5 \pm 2.7\%$. It decreases to 7% for a 1.6-GHz-wide AFC due to the limited bandwidth of the impedance-matched cavity (see the Supplemental Material

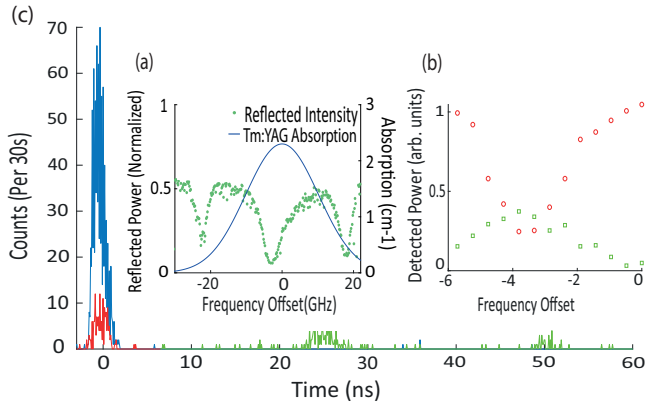


FIG. 2. (a) The line (blue) shows the absorption of the Tm:YAG crystal without a cavity [28], whereas the points (green) show the normalized reflected power obtained by sweeping 4-ns-long pulses of light in frequency across the cavity features. Close-to-perfect impedance matching is obtained at a detuning of -4 GHz. (b) Detected intensity of strong 2-ns-long light pulses after reflection (red circles) and reemission after 50-ns storage from the cavity (green squares). The maximum reemission is visible at (approximate) impedance matching. All AFCs were of 500-MHz width. (c) Detection histograms for 25-ns storage of weak coherent pulses. The upper (blue) peak occurring at 0-ns offset is the memory input pulse; the lower (red) peak occurring at 0 ns is the portion of the input pulse that is reflected from the cavity (not stored), and the (green) peaks at 25 and 50 ns show recalled pulses after a multiples of the storage time.

for details [26]). These values, which are insensitive to the type of input signal (strong or weak laser pulses or heralded single photons), correspond to a 20- to 30-fold increase in the single-pass efficiency in the same crystal (no cavity), which we estimate to be below 1%. Note as well that, due to the combination of our sample's small single-pass absorption and the broad bandwidth of our spectral features, we measured no evidence of strong dispersion, which has previously limited bandwidths of similar cavity-based memories [21–23].

We used several light sources to test the memory. First, to create time-bin qubits encoded into temporal modes of attenuated laser pulses, we employed an AOM combined with an IM to tailor a continuous-wave laser beam at a 793-nm wavelength. Early and late temporal modes were of 800-ps lengths and separated by 1.4 ns with spectral and phase control achieved via laser diode grating adjustment and a serrodyne-driven phase modulator. Second, we prepared quantum-correlated pairs of photons at 1538- and 793-nm wavelength by means of SPDC in a periodically poled lithium niobate waveguide. After filtering, their spectra were narrowed to 8 and 1.5 GHz, respectively. Third, passing the 793-nm photons before storage through an unbalanced Michelson interferometer, we could, furthermore, generate heralded time-bin qubits with the same mode separation. All single-photon-level signals were detected using low jitter WSi SNSPDs [27] followed by suitable coincidence electronics. In addition, to analyze photons in qubit states, we employed an actively phase-locked Michelson interferometer with 42-cm path-length difference.

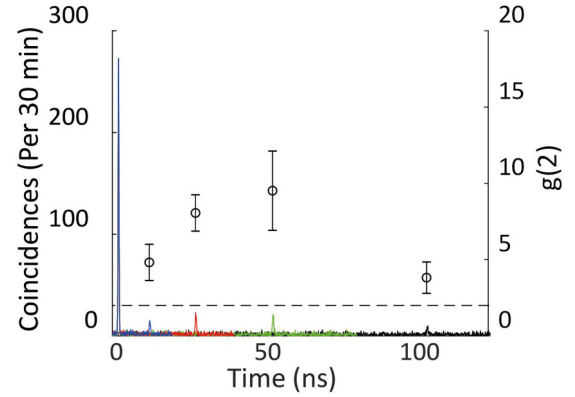


FIG. 3. Time-resolved coincidence detections of a 1538-nm heralding photon and a 793-nm photon stored in the cavity memory for various different storage times. The peaks visible at each of the set storage times (10, 25, 50, and 100 ns) verify that the nonclassical correlations created by SPDC persist after storage. The right-hand axis depicts the $g^{(2)}$ value for each peak with error bars multiple standard deviations above the classical limit (dashed line).

III. RESULTS

To study how well our memory stores nonclassical light, we first used photon pairs and measured cross correlations between heralding 1538-nm photons and 793-nm photons. At maximum pump laser power with no memory in place, we found a cross-correlation coefficient of $g^{(2)} = 61.8 \pm 3.8$. For values of $g^{(2)} > 2$, the correlations violate the Cauchy-Schwartz inequality, implying that they are nonclassical in nature [29]. Adding the memory for the 793-nm photons, we measured $g^{(2)} = 9.1 \pm 1.2$ after 25-ns storage, and we found Cauchy-Schwartz violations after as much as 100 ns (see Fig. 3). This shows the preservation of quantum correlations during storage in the cavity memory and, hence, establishes that the cavity-enhanced memory as a quantum memory for light.

Next, we created and stored various time-bin qubits encoded into attenuated laser pulses with a mean photon number of $\mu = 0.7$. For these measurements, we set the storage time to 25 ns. For Z -basis states, $|Z+\rangle \equiv |e\rangle$ and $|Z-\rangle \equiv |l\rangle$, we found a mean fidelity of $\mathcal{F}_z = 97.6 \pm 0.02\%$. Furthermore, characterization of $|X_{\pm}\rangle \equiv \frac{|e\rangle \pm |l\rangle}{\sqrt{2}}$ and $|Y_{\pm}\rangle \equiv \frac{|e\rangle \pm i|l\rangle}{\sqrt{2}}$ yielded $\mathcal{F}_{x,y} = 93.7 \pm 0.1\%$, resulting in a memory fidelity averaged over all six states of $\mathcal{F} = \frac{1}{3}\mathcal{F}_z + \frac{2}{3}\mathcal{F}_{x,y} = 95 \pm 0.1\%$ (see Fig. 4, the Supplemental Material [26], and Ref. [10] for more details). Taking into account the recall efficiency of 7% and the mean photon number of $\mu = 0.7$, all fidelities significantly exceed the upper bound of $\mathcal{F}(\mu, \eta) = 80.3\%$, established conservatively for classical memories under the assumption of intercept-resend attacks [30–32] as well as that imposed by the optimal universal quantum cloning machine [33].

Finally, we repeated these measurements after replacing the source of attenuated laser pulses by heralded single photons. But instead of using interferometers to analyze the qubits after storage in the X and Y bases, we configured the memory for double-comb storage with storage-time separation matching the qubit time-bin separation. This method

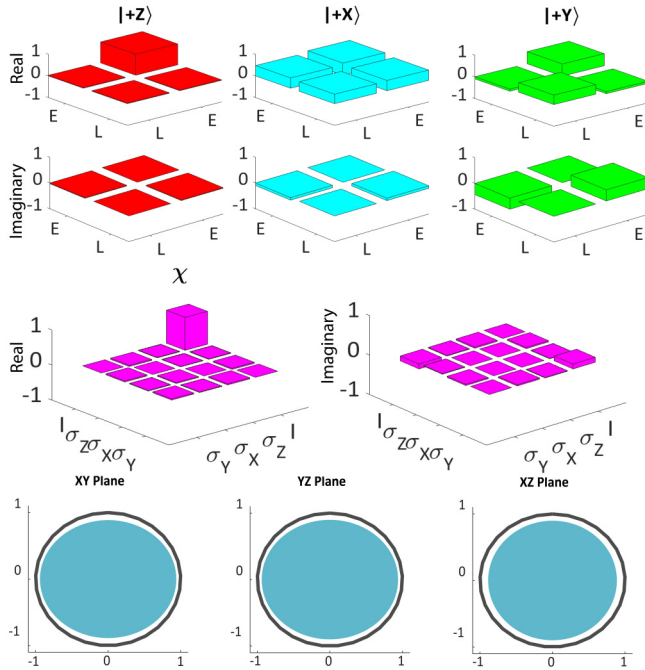


FIG. 4. (a) Density matrices of three nonorthogonal qubit states after reemission from the memory. (b) Quantum process matrix χ . (c) Cross sections through the Bloch sphere depicting the average over all possible output states. Black circular lines denote the output expected from a perfect memory, and the filled ovals (blue) depict qubits after the nonideal storage process [31].

allows for a convenient analysis of time-bin qubit states in the superposition bases [34]. The series of measurements resulted in a heralded single-photon qubit fidelity of $\mathcal{F} = 85 \pm 0.02\%$ (see the Supplemental Material [26]), again exceeding the thresholds imposed by classical storage and quantum cloning. It also allowed us to establish the density matrices of recalled qubits for various inputs by means of quantum state tomography [see Fig. 4(a)] and, in turn, to perform quantum process tomography [35,36]. The resulting storage process matrix χ , depicted in Fig. 4(b), completely describes the mapping between arbitrary input and output qubit states [31]. As expected, the dominant term describes the identity operation, but some small imperfections with magnitudes ≤ 0.1 are also visible.

Another way of presenting these imperfections is to look at the ensemble of output states averaged over all possible input states. This results in a deformed Bloch sphere as shown in Fig. 4(c). But note that this deformation as well as the unexpected elements in the process matrix in Fig. 4(b) provide the upper bound to imperfections in the storage process. Indeed, they are mostly caused by imperfect state preparation and measurement, rather than by an imperfect memory. For example, nonideal PM drive-pulse duration and timing cause small frequency shifts between temporal input modes, making them distinguishable and, therefore, reducing the quality of the interference required for analysis. Furthermore, imbal-

ances of the beam splitters used to split and couple temporal modes in our analysis interferometer also lower the measured fidelities.

IV. CONCLUSION

To summarize, we have demonstrated that solid-state quantum memory based on ensembles of rare-earth ions and cavity-enhanced light-matter interaction in a monolithic and fiber-coupled cavity allows storing quantum states of light. Our memory operates in the domain of preset storage times and allows feed-forward-based mode mapping using external frequency shifters [6]. It is capable of storing broadband quantum light with a fidelity of, at least, 95%, an efficiency η of up to 27.5%, and a time-bandwidth product (TB) of up to $100 \text{ ns} * 1.5 \text{ GHz} = 150$ where 100 ns is the longest time after which we observed a violation of the Cauchy-Schwartz inequality. We note that the factor of $\eta * \text{TB} = 1050$ (with $\eta = 7\%$), which is comparable to that obtained in previous demonstrations of rare-earth-ensemble-based storage of attenuated laser pulses [37].

To further improve key properties, several modifications are required. First, to reach an efficiency close to 100%, the finesse of the AFC has to be increased beyond its current value of two. As this is only possible if the total AFC width is smaller than the ground-state splitting of the rare-earth ion, in this case, 200 MHz/T, this limits the available bandwidth per spectral channel, implying the need for suitably adapted sources of quantum light. However, the time-bandwidth product can remain high as many spectral channels can be created in parallel [6]. Furthermore, fiber coupling loss must be reduced through better mode matching. Second, to increase the storage time to a few hundred microseconds, enough for an elementary link in a quantum repeater architecture of around 100 km [6], materials with improved coherence have to be employed. Possibilities include Tm:Y₃Ga₅O₁₂ for which optical coherence times of 490 μs have been reported [38] or materials featuring narrow ground-state transitions [24].

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