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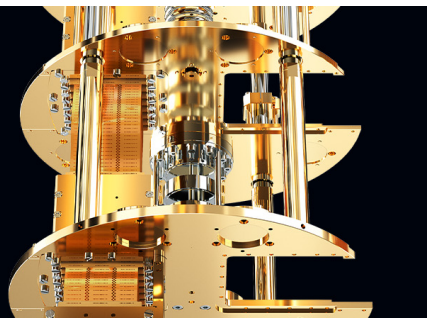

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ABSTRACT

We propose a scheme of heralded storages of single photons via spontaneous Raman scattering (SRS) enhanced by a chiral standing-wave cavity, which creates a non-classically correlated pair of a spin wave and a Stokes photon (heralded photon) when a single-photon "write" pulse is applied to an atomic ensemble. In this scheme, to decrease the power of the write pulse for SRS, the write laser and the Stokes fields are aligned to propagate near-collinearly through the atoms. The short storage lifetime remains a challenge in standing-wave cavity-enhanced SRS. To prolong the storage lifetime, we achieve a long-wavelength spin-wave memory by removing the backward-scattering Stokes field from the cavity, which is realized by using non-reciprocal couplings between the Stokes fields and the atoms. In the experiment, the memory lifetime is demonstrated up to ∼100 μs, which is beyond the limitation (∼350 ns) of the storage in the atomic ensembles coupled to standing-wave cavities. We achieve a spin-wave memory for the write pulse containing a mean photon number of ∼4. The quantum correlation of the Stokes-spin-wave pairs is beyond 2, which gives strong evidence of non-classical behavior.

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Quantum memories[1–4] are devices that can faithfully store and recall the quantum states of single photons and are critical elements in quantum networks based on quantum repeaters. The quantum networks hold promise for the distributed quantum computers and long-distance quantum communication.[5–7] Quantum storages of single photons in atomic ensembles have been experimentally demonstrated via various schemes, which include the Duan–Lukin–Cirac–Zoller (DLCZ) protocol,[8–25] electromagnetically induced transparency (EIT),[26–34] atomic frequency comb (AFC),[35–44] and Raman storage[45,46] as well as its variants.[47–49] However, these storages are done in a non-heralded way. Heralded storages of the quantum states of a single photon are required for the demonstration of measurement-device-independent quantum key distribution (QKD)[50] and the construction of quantum networks via quantum repeaters with satellite links.[51,52] Note that, in Refs. 51 and 52, the heralded storages of the single-photon quantum states are done by using the combination of quantum nondemolition (QND) measurement and quantum memory (QM), where QND heralds the coming of the quantum states and QM is used for the storage of the quantum states. Based on nonlinear cross-phase modulations in a solid-state atomic ensemble (Tm:LiNbO3 waveguide), Sinclair *et al.* reported a proposal of nondestructive detection of photonic time-bin qubits and made a proof-of-principle demonstration[53] for the proposal. For improving the cross-phase modulations, Goswami *et al.* proposed a cavity-enhanced scheme.[54] Another way of realizing the heralded storages of the single-photon quantum states is based on quantum teleportation of a photonic qubit into a quantum memory. Quantum teleportation of a photonic qubit to an atomic ensemble[55] or to a solid-state spin[56] has been demonstrated. By coupling the input photon into the mode of a high-finesse cavity loading a single atom, G. Rempe's group experimentally demonstrated the heralded storages of the single-photon quantum states with cavity-QED system.[57,58]

QMs based on atomic ensembles feature collective enhancement and promise higher retrieval efficiencies.[59] In 2009, V. Vuletic's group demonstrated a heralded quantum memory for the polarization states of a coherent light pulse containing ∼500 photons with atomic

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ensemble in a low-finesse cavity.[60] So far, heralded storages of the single-photon quantum states have not been experimentally realized with atomic ensemble-based memories.

Here, we propose a scheme of heralded storage of single photons via the DLCZ process. A write pulse induces spontaneous Raman scattering (SRS) of a Stokes photon, which can simultaneously create a spin wave non-classically correlated with the Stokes photon. In the case of the write pulse at the single-photon level, a detection event of the Stokes photon heralds that the write photon is stored as one spin wave, which can be retrieved after a storage time. Toward the heralded storage of an input write photon, we demonstrate the storage of a write laser pulse containing $\langle N_w \rangle \approx 4$ photons in an atomic ensemble coupled into a standing-wave cavity. With standing-wave cavities, the previous works have reported the enhanced DLCZ storage process.[10,60] However, in those works, the spin waves include a short-wavelength component, which severely limits the storage lifetime (~350 ns[10]). In the present work, we eliminate the short-wavelength spin-wave component by using non-reciprocal light-atom couplings based on the polarization-direction correlation.[61] Thus, the achieved heralded storage lifetime is up to ~100 μs with the standing-wave cavity.

Figure 1(a) shows our experimental setup, where a cold ensemble of ^{87}Rb atoms is coupled into a standing-wave optical cavity. The standing-wave cavity consists of two mirrors M1 (output coupler) and M2 separated by 1200 mm, and the reflectivity of M1 and M2 is R1 = 90% and R2 = 99.99%, respectively. The cavity is inserted by two lenses (L1 and L2), two quarter-wave plates [$(\lambda/4)_1$ and $(\lambda/4)_2$], and two polarization beam-splitting mirrors (PBSM1 and PBSM2), where each of the PBSMs has a transmissivity of 99.3% for horizontal (H)-polarization, and a reflectivity of 95% for vertical (V)-polarization. The cavity round trip loss is L = 6.5% for the H-polarized Stokes field. The waist radius of the cavity mode (TEM00) is $\omega_c \approx 30\ \mu$m and cavity fineness is F = 34.

The relative levels of the atoms include $|a\rangle = |5S_{1/2}, F = 1\rangle$, $|b\rangle = |5S_{1/2}, F = 2\rangle$, $|e_1\rangle = |5P_{1/2}, F' = 1\rangle$, and $|e_2\rangle = |5P_{1/2}, F' = 2\rangle$, which are shown in Fig. 1(b). We apply a bias magnetic field $B = 4G$ along the z axis to define quantum axis and split the Zeeman degeneracy of the atomic levels. Releasing from the MOT, the cold atoms are initially prepared in the Zeeman state $|a, m_F = -1\rangle$ via optical pumping, where m_F denotes the magnetic quantum number. A write laser beam (purple solid arrow) with a frequency red detuned from the $|a\rangle \rightarrow |e_1\rangle$ transition by ~15 MHz is applied on the atoms. In the process of spontaneous Raman scattering, the emitted Stokes photons are basically collinear with the write laser, with a distribution only over a very small solid angle.[62] So, toward the heralded storage of single photons for write laser pulses, we combine the write and Stokes fields collinearly to reduce the write pulse power.

As shown in Fig. 1(a), the write laser beam is V-polarized and is introduced to the atoms via the PBSM1. The write beam is changed from V-polarized into left-circularly polarized by the wave-plate $(\lambda/4)_1$, and then interacts with the atoms on the transition $|a, m_F = -1\rangle \rightarrow |e_1, m_F = 0\rangle$. The "write" pulses induce Stokes emission on the $|e_1, m_F = 0\rangle \rightarrow |b, m_F = 1\rangle$ ($\Delta m_F = -1$) Zeeman transition, which simultaneously create spin waves stored in the coherence $|a, m_F = -1\rangle \leftrightarrow |b, m_F = 1\rangle$. The emitted Stokes field on the transition is resonant with the standing-wave cavity, whose length is stabilized by introducing a locking laser beam into the cavity. The Stokes emissions on other transitions, for example, $|e_1, m_F = 0\rangle \rightarrow |b, m_F = -1\rangle$, etc., will not be resonant with the cavity since their frequencies are different with that from the transition $|a, m_F = -1\rangle \leftrightarrow |b, m_F = 1\rangle$ due to Zeeman splitting. In the following, the Stokes field only refers to the emission from the $|e_1, m_F = 0\rangle \rightarrow |b, m_F = 1\rangle$ ($\Delta m_F = -1$) transition.

The emitted Stokes fields propagating along the z and -z directions are both coupled into the cavity modes. Constrained to the

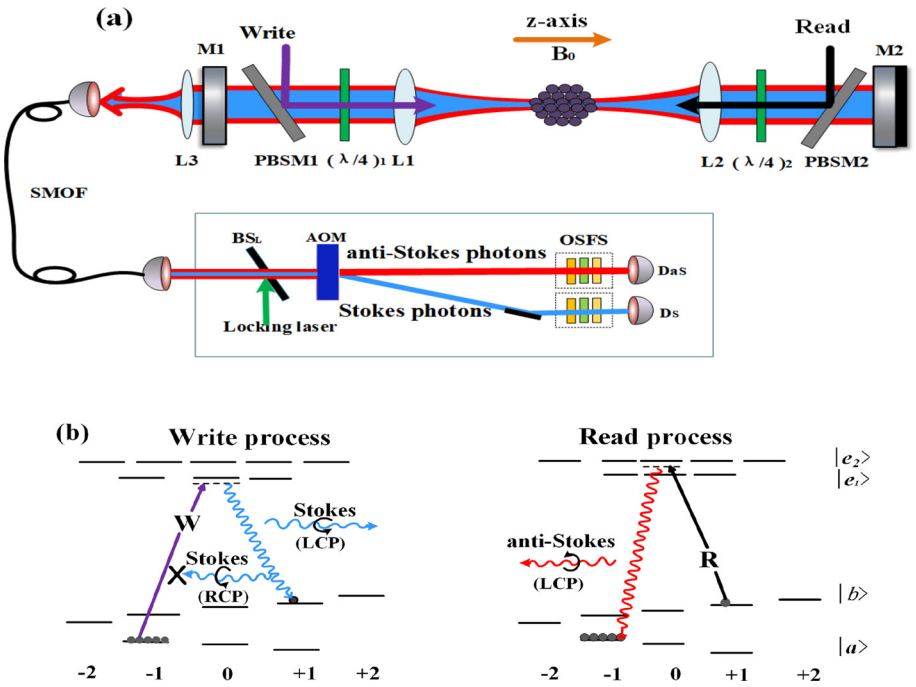

FIG. 1. (a) Experimental setup: the cold atomic ensemble is coupled with a standing-wave cavity. The Stokes and anti-Stokes photons are resonant with the cavity. W: write laser pulse; R: read laser pulse, OSFS: optical spectrum filters for Stokes (anti-Stokes) channels; SMF: single-mode fiber; PBSM, polarization beam splitting mirror; AOM: acousto-optic modulator. (b) Relevant atomic levels coupled with the light fields. ×: the backward (−z axis) emitted Stokes field is removed off the cavity. RCP: right circular polarization; LCP: left circular polarization.

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spin-momentum correlation,[61] the Stokes fields initially emitting from the atoms on $|e_1, m_F = 0\rangle \rightarrow |b, m_F = 1\rangle$ transition propagating along the z and -z directions are left-circularly polarized (LCP) and right-circularly polarized (RCP), respectively, which are labeled as fields E_{+k} and E_{-k}. The RCP Stokes field E_{-k} propagating along the -z direction is changed into V-polarized after the quarter wave-plate $(\lambda/4)_1$, which then is reflected off the cavity by PBSM1. So, the Stokes field along the backward direction (-z-axis) is removed out the cavity.

We now turn to discuss the propagation of the Stokes field E_{+k}. The LCP Stokes field E_{+k} propagating along the z direction is changed into H-polarization after the quarter wave-plate $(\lambda/4)_2$. The H-polarized Stokes field E_{+k} goes through the PBSM2 and is reflected by the cavity mirror M2. Then the reflected Stokes field E_{+k} propagates along the -z direction and is changed into LCP after $(\lambda/4)_2$. Passing through the atoms, the LCP E_{+k} propagating along the -z direction does not couple to them since the atoms emit only RCP Stokes field propagating along the -z direction.[61] After the quarter wave-plate $(\lambda/4)_1$, the E_{+k} is changed from LCP into H-polarization. The H-polarized E_{+k} passes through the PBSM1 and is reflected by cavity mirror M1. The reflected Stokes field E_{+k} propagates along the z axis, and its polarization is changed to LCP passing through $(\lambda/4)_1$. When it goes through the atoms, it couples with the atoms. With the cavity, the coupling between the LCP Stokes field with the atoms is enhanced by a factor of $\sim 2F/\pi$.[63]

According to the above-mentioned discussions, the coupling of the circularly polarized Stokes fields with the atoms on the chiral transition ($\Delta m_F = -1$) is non-reciprocal. Such non-reciprocal coupling enables us to remove the backward-scattering Stokes signal from the cavity by PBSM1. Thus, the successful detection of Stokes photons heralds the generation of a long-wavelength spin waves. Specifically, the wave-vector of the spin wave $\vec{k}_M^+$ is written as $\vec{k}_M^+ = \vec{k}_w - \vec{k}_S^+$, where $\vec{k}_w$ and $\vec{k}_S^+$ are the wave-vectors of the write beam and Stokes field, respectively. When the write laser and Stokes fields propagate in a nearly collinear configuration, the wave-vector $\vec{k}_M^+$ approaches to zero and the wavelength of the spin wave $\lambda_M = 2\pi/\vec{k}_M^+$ is very long. The relation between the lifetime τ_0 and the wavelength λ_M of the spin wave can be expressed as $\tau_0 = (\lambda_M/2\pi\nu_S)$, where ν_S is the one-dimensional average speed.[13] This long-wavelength spin wave promises us to achieve a long storage lifetime.[12,28,64] For the backward-scatting Stokes field E_{-k}, the stored spin wave $\vec{k}_M^- = \vec{k}_w - \vec{k}_S^- \approx 2\vec{k}_w$ due to $\vec{k}_w \approx -\vec{k}_S^-$. For this case, the wavelength of the stored spin wave is $\lambda_M = 2\pi/\vec{k}_M^- \approx 0.4\ \mu$m and corresponds to a 240 ns storage lifetime.[10] When both spin waves may be stored in a standing-wave cavity, the lifetime is ~350 ns in Ref. 10.

After a storage time τ, the spin wave is retrieved by a read laser beam with its frequency being tuned on the transition of $|b\rangle \rightarrow |e_2\rangle$ by ~ -90 MHz. As shown in Fig. 1(a). The read laser beam is V-polarized and combined into the cavity mode via PBSM2. It counter-propagates relative to the write beam. Before entering the atoms, the read beam is changed from V-polarization into RCP by the wave-plate $(\lambda/4)_2$. The RCP read laser interacts with the Zeeman transition $|b, m_F = +1\rangle \rightarrow |e_2, m_F = 0\rangle$ and converts the long-wavelength spin wave into an anti-Stokes photon that propagates along the -z axis. The anti-Stokes photon retrieved from the atoms on the transition $|e_2, m_F = 0\rangle \rightarrow |a, m_F = -1\rangle$ ($\Delta m_F = +1$) is LCP and is changed into H-polarized photon after the wave-plate $(\lambda/4)_1$. The H-polarized anti-Stokes photon goes through the PBSM1 and L1 and then is reflected by the cavity mirror M1. The anti-Stokes field propagates back and forth in the cavity. Similar to the coupling between the Stokes field and the atoms, the coupling between the anti-Stokes field and the atoms is also non-reciprocal. The LCP anti-Stokes field propagating along the -z axis is enhanced when passing the atoms. The anti-Stokes field is also resonant with cavity, which enhances the retrieval efficiency of the spin wave.[61]

In the present experiment, the waist radii of the write and read beams in the cavity are both $r_{w/r} = 95\ \mu$m. The peak power of the read pulse is ~5 mW. The pulse widths of the write and read pulses are 100 and 210 ns, respectively. The Stokes and anti-Stokes photons escape from the M1 mirror with the probability of $\eta_{esc} = \frac{T1}{T1+L} \approx 61\%$, where $T1 = 10\%$ is the transmittivity of M1, and L = 6.5% is the cavity round trip loss. The escaped photons are coupled into a single-mode fiber (SMF). The output of the SMF goes through a beam splitter BS_L, and then is directed to an acousto-optic modulator (AOM). The BS_L is used for combining the locking laser beam into the cavity. The Stokes and anti-Stokes photons have different times arriving to the AOM and they are split by it. The Stokes photons propagate along the -1-order diffraction channel and the anti-Stokes photons along the 0-order channel. The Stokes (anti-Stokes) field goes through a set of optical spectrum filter (OSFS) and is directed into the single-photon detector $D_S(D_{aS})$.

First, we measure the intrinsic retrieval efficiency γ of the spin wave as $\gamma = P_{S,aS}/\eta_S P_S$, where $P_{S,aS}$ denotes the coincident probability between the Stokes and anti-Stokes photons, which corresponds to the coincidence counter between the detectors D_S and D_{aS}. P_S denotes the detection probability of Stokes photons at the detector D_S, which can be written as $P_S = \chi\eta_S + B\eta_S$, χ denotes the excitation probability, i.e., the probability of generating a Stokes-spin-wave pair per write pulse, B the background noise, η_S detection efficiency for the Stokes channel. In the present experiment, the Stokes detection efficiency is $\eta_S \approx 7\%$ (supplementary material). Figure 2 is the measured intrinsic retrieval efficiency γ as a function of the storage time τ for $\chi = 1\%$. The power of the write pulse for $\chi = 1\%$ is 20 nW. The solid red curve is the fit to the intrinsic retrieval efficiencies according to the function $\gamma(\tau) = \gamma_0 e^{-\tau/\tau_0}$, which yields initial retrieval efficiency $\gamma_0 = 20\%$ and

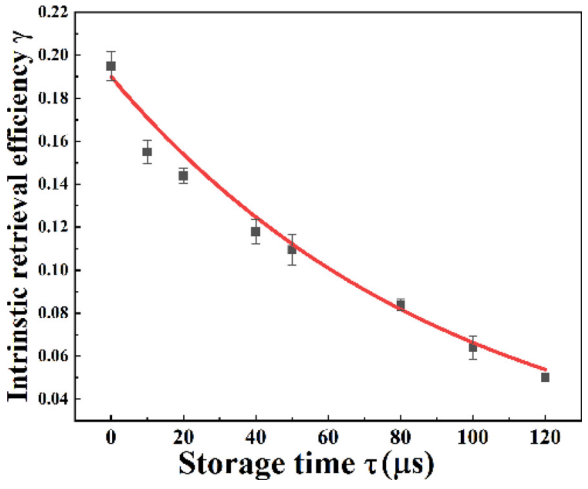

FIG. 2. The measured intrinsic retrieval efficiency as a function of the storage time τ for $\chi \approx 1\%$.

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the storage lifetime $\tau_0 \approx 100\,\mu s$. The achieved storage lifetime is close to that in free space under identical conditions ($\tau_0' \approx 110\,\mu s$), demonstrating that our approach breaks through the lifetime limitations by standing-wave cavity.

The cross correlation function between the Stokes and anti-Stokes photons can be measured by

$$g_{S,aS} = \frac{P_{S,aS}}{P_S P_{aS}}, \quad (1)$$

where P_{aS} denotes the detection probability of anti-Stokes photons at the detector D_{aS}. In the supplementary material, we derive the expression of the cross correlation function $g_{S,aS}$, which is written as

$$g_{S,aS} = 1 + \frac{\gamma(\tau)}{(B+\chi)\gamma(\tau) + (B+\chi)(1-\gamma(\tau))\xi_{se}A + \frac{BC}{\chi} + C}, \quad (2)$$

where $\gamma(\tau)$ is the intrinsic retrieval efficiency after storage time τ, C is the background noises in anti-Stokes channels, ξ_{se} is the branching ratio of the anti-Stokes photon on the atomic transition, and A denotes the enhance of the cavity to the anti-Stokes field. Figure 3 plots the measured $g_{S,aS}$ data (black square dots) as a function of storage time τ for $\chi \approx 1\%$. The solid line is the fittings to the measured data according to Eq. (2), where $\gamma(\tau) = \gamma_0 e^{-\tau/\tau_0}$. The fitting is in good agreement with the measured $g_{S,aS}$.

Before demonstrating a heralded storage of a write pulse at single-photon level, we measure the dependence of the write pulse power p_w on the excitation probability χ, and find the relation between them can be described by $\chi = kp_w$, where $k \approx 5.7 \times 10^{-4}/nW$ (see the supplementary material). The photon number N_w of the write pulse can be calculated by $N_w = p_w \Delta t/h\nu \approx p_w \times (4 \times 10^2/\text{nw})$, where $h\nu$ is the energy of single write photon, and $\Delta t = 100$ ns is the write pulse width.

Subsequently, we measure the dependence of $g_{S,aS}$ on the write pulse power p_w. The measured results are plotted as Fig. 4, which

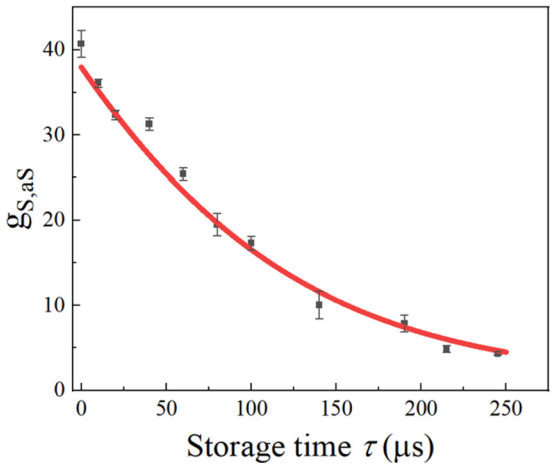

FIG. 3. Measured cross correlation function $g_{S,aS}$ as a function of storage time τ. The red line is the fittings to the measured data according to Eq. (2), where $B = 4.3 \times 10^{-4}$ and $C = 6 \times 10^{-4}$ are taken from the measured values, the cavity enhancement factor and the branching ratio are taken as $A = 1.3$ and $\xi_{se} = 0.27$, respectively.

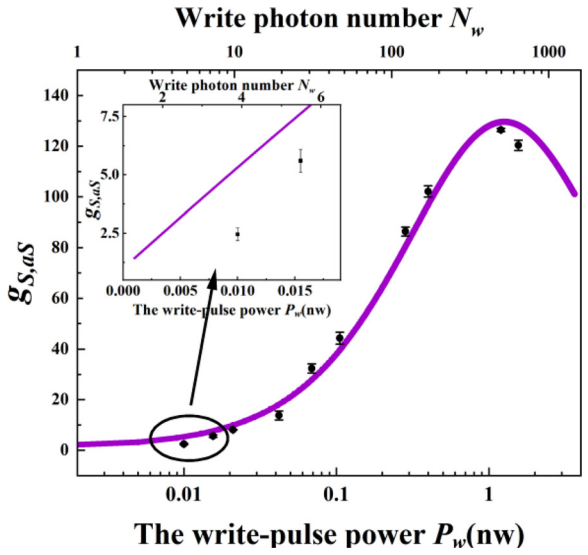

FIG. 4. Measured $g_{S,aS}$ as a function of p_w. The purple solid line is the fitting of $g_{S,aS}$ according to Eq. (2) and $\chi = kp_w$. In the fitting, the parameters $\gamma_0 = 20\%$, $B = 4.3 \times 10^{-4}$, $C = 6 \times 10^{-4}$, $k = 5.7 \times 10^{-4}$/nW, and $\xi_{se} = 0.27$.

show that the maximum value of $g_{S,aS} \approx 126$ at the write power $p_w = 1.23$ nW (corresponding to $N_w \approx 500$, $\chi = 7 \times 10^{-4}$). When the write pulse is attenuated to the single-photon level, i.e., the mean photon number $\langle N_w \rangle \approx 4$ in the present experiment corresponding to $p_w \approx 0.01$ nW, the cross correlation function $g_{S,aS} \approx 2.46$, which is still beyond 2 and gives a strong evidence of non-classical behavior. The purple solid line is the fit to the measured $g_{S,aS}$ according to Eq. (2) and $\chi = kp_w$, both are in good agreement.

As mentioned in the supplementary material, the excitation probability induced by a write pulse containing $\langle N_w \rangle \approx 4$ photons is $\chi \approx 5.7 \times 10^{-6}$, which is below the background-noise level $B \approx 4.3 \times 10^{-4}$. However, we still measure the non-classical correlation $g_{s,aS} \approx 2.46$, which indicates that our present experiment promises an enhanced quantum measurement.[65]

In summary, we experimentally demonstrate the storage of the input write pulse at the single-photon level via spontaneous Raman scattering enhanced by a standing-wave cavity. Based on the polarization-direction correlation, we design the cavity with the polarization optical elements, which enables the non-reciprocal coupling of the Stokes field with the atoms. This chiral cavity removes the backward-scattered Stokes photons from the cavity, thereby extending the storage lifetime of the atomic memory to 100 μs. Toward the heralded storages of single photons, we reduce the power of the write pulse by aligning the write laser and the Stokes fields to propagate near-collinearly through the atoms.

The detection probability of the Stokes photons induced by the write pulse at the single-photon level ($\langle N_w \rangle \approx 4$) is $\sim 10^{-6}$ per pulse, which is very low for realizing the heralded storage of single photons. By improving the cavity finesse, the heralding probability of the write single-photon storage can be increased accordingly. However, the increase in cavity fineness will reduce the retrieval efficiency of atomic spin waves due to the large intracavity loss. To enhance the heralding probability of single-photon storage ($\sim 10^{-5}$) while maintaining high

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spin-wave retrieval efficiency (>70%), this requires increasing cavity finesse ($F = 250$) and suppressing intracavity loss ($L = 0.5\%$). Through these technical advancements, we anticipate that the heralded storage of a single photon may be effectively demonstrated with atomic ensembles.

See the supplementary material for more information about the dependence of the cross correlation function $g_{S,aS}$ on the storage time τ and the relationship $\chi = kp_w$.

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AUTHOR DECLARATIONS

Conflict of Interest

The authors have no conflicts to disclose.

Author Contributions

Can Sun: Data curation (equal); Methodology (equal); Writing – original draft (equal). **Yibo Hou:** Data curation (equal). **Xiang Chen:** Data curation (equal); Methodology (equal). **Tianyu Wang:** Methodology (equal). **Chunyao He:** Methodology (equal). **Yulei Geng:** Methodology (equal). **Yafei Wen:** Methodology (equal). **Shujing Li:** Methodology (equal). **Hai Wang:** Funding acquisition (equal); Methodology (equal); Writing – review & editing (equal).

DATA AVAILABILITY

The data that support the findings of this study are available from the corresponding authors upon reasonable request.

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