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Entangling Billions of Motional Atoms and an Optical Loop at Ambient Condition

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Quantum entanglement recognized by Einstein, Podolsky, Rosen and Schrödinger is the essence of quantum physics, and has become a real available resource in the laboratory over recent years. The observation of quantum entanglement in macroscopic matters has implications in the fundamental studies of quantum mechanics, as well as the implementations of quantum information technologies, such as quantum communication, enhanced sensing and distributed quantum computing. Here, we report the creation and storage of the heralded entanglement between two different types of broadband quantum memories located on separated platforms at room temperature: a single-photon entangled state delocalized between billions of motional atoms as a collective excitation and an all-optical loop as a flying qubit. The stored entangled state is subsequently retrieved after a programmable storage time, and is verified by measuring the nonclassical correlations, quantum interference, and concurrence of the mapped-out Raman-scattered photons. Our results show that quantum entanglement can be sustained in macroscopic matters at ambient condition, which pushes the fundamental researches of the transition boundary between quantum and classical worlds. Also, the two quantum memories constitute a hybrid memory-built-in quantum network, which highlights the potential of the cooperation between an stationary atomic ensemble and an all-optical loop as quantum nodes at ambient condition, and brings a significant step towards practical quantum networks.

Human perception of the real-life world is mainly derived from the phenomena of macroscopic matter movements, described by the laws of classical physics. Quantum mechanics, however, endows the nature of superposition and entanglement is able to express the universe in a clearer and more concise manner. Therefore, expanding quantum theory to more general ambient systems and exploring the quantum phenomena in macroscopic objects push the researches of the transition boundary between classical and quantum realm. On the other hand, the capability of establishing and sharing remote high-fidelity entanglement over long distances is necessary for scalable and high speed quantum networks, which outperforms classical world in the areas of communication, computing, sensing and metrology [1–10].

Over the past two decades, heralded quantum entanglement has been observed in some matter systems, such as atomic ensembles [3, 11–14], individual atoms [15], trapped ions [16], quantum dots [17] and NV centers [18–20]. Unfortunately, strong internal interactions and external coupling with the environment make it hard to observe quantum superposition and entanglement with very short decoherence time in macroscopic systems. Photons, as the most widely-used state carriers in quantum world today, still suffer from inevitable loss, either propagating in free space or optical fibers [21, 22]. Together with the probabilistic nature of quantum mechanics [23, 24], the creation and dissemination of the entangled states are hard to be scalable for longer distances or for more nodes in a quantum network.

Quantum-memory-assisted repeaters provide an elegant solution, with which the heralded remote entanglement can be stored and retrieved on demand [25–35], enabling the polynomial increase in time consumption in a network. Practical applications of quantum memories require some advantageous features, such as high efficiency, long lifetime, low noise level, operable at high bandwidth and room temperature. Such quantum-memory-enabled networks is pursued for generating, storing, processing and disseminating heralded entanglement, in real-life quantum information technologies, overcoming the most critical barriers including photon loss and probabilistic sources.

In this paper, we study the creation and retrieval of the collective excitation state in billions of motional atoms, and the broadband storage of the loop architecture. Such two memory-built-in quantum nodes constitute a hybrid quantum network at ambient condition. Based on the implementations, we observe the heralded quantum entanglement between billions of motional atoms and an optical loop located on separated platforms at room temperature.

As is shown in FIG. 1a, the entangled state is established once the first read pulse applied, with a fixed time delay \( \tau_0 \), and it can be hold on for coherence times of both quantum memories, which are all at microsecond level [36]. To reveal the quantum entanglement, the stored states are mapped back to optical modes, then

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FIG. 1. Schematic of establishing and verifying the hybrid entanglement between an optical loop and billions of motional atoms. a. Entanglement map depends on the storage times of two quantum memories. The top three pulses represent the control light addressing on the atoms. W: Write pulse, R: Read pulse. The entangled state at the marked point \((t_2, t_3)\) is verified as follows. b. Time sequences of the control pulses and the generated photons. c. Sequences for photons generation, entanglement creation and verification. Insets above describe write and read processes of the far off-resonance Duan-Lukin-Cirac-Zoller protocol, with the three-level A-type configuration of atoms. \(|g\rangle\) and \(|s\rangle\) represent hyperfine ground states. Insets below describe the polarization states of the optical modes, shown in Bloch spheres. The first polarization beam splitter (PBS1) combines the retrieved two optical modes, and the following half-wave plate (HWP) and two quarter-wave plates (QWP) rotate and mix the fields, resulting the single-photon interference on PBS2. \(D_{S/A}\): Detector of Stokes/anti-Stokes photons.

via a single-photon interference, we obtain the concurrence and density matrix of the retrieved light modes. We also observe a violation of Cauchy-Schwarz inequality \([37]\) up to 209 SDs, with a cross-correlation value up to 15.39 ± 0.26 well exceeds 6, which enables the violation of Bell’s inequality \([38]\) and promises further quantum applications. The experimental time sequences are shown in FIG. 1b.

The experimental scheme used for generating photons is based on the far off-resonance Duan-Lukin-Cirac-Zoller protocol in a memory-built-in fashion, and its implementations have been demonstrated as intrinsically broadband and low-noise at room temperature \([36, 39, 40]\). As is shown in FIG. 1c, initially, billions of motional atoms are prepared into the ground state \(|g\rangle\), waiting for the write pulse to produce an excitation among atoms, and meanwhile, the process is accompanied by a flying Stokes photon via spontaneous Raman scattering to herald a success excitation. After programmable storage times, the collective excitation state can be mapped out as an anti-Stokes photon, and into various temporal modes by a series of read pulse. The probability distribution can
be tuned by applying different amplitudes of each read pulse.

An all-optical loop serves as another quantum memory node, for mapping flying anti-Stokes modes in and out with programmable storage times individually. Once an Stokes photon occurs, the horizontally-polarized anti-Stokes mode entering the loop will be converted to vertically-polarized, so that the photon will be trapped in the optical loop until its polarization is converted back.

The entanglement is established when the first read pulse is applied on atoms, to convert the collective excited state back to anti-Stokes photons with a tunable retrieval efficiency. By sending this retrieved anti-Stokes mode A to the all-optical loop, the detection of a Stokes photon at $D_S$ heralds the creation of a single excitation delocalized between the two quantum memories, and the entangled state can be sustained until the second read pulse is applied to atoms for retrieving the state. The established entangled state can be written as

$$|\psi_{\text{Joint}}\rangle = (\alpha |1\rangle_{\text{Loop}} |0\rangle_{\text{Atoms}} + \beta |0\rangle_{\text{Loop}} |1\rangle_{\text{Atoms}})/\sqrt{2}$$  \hspace{1cm} (1)$$

where $|1\rangle_{\text{Atoms}}$ represents a collective excitation state $[41]$ in the atomic ensemble, and $|1\rangle_{\text{Loop}}$ represents an anti-Stokes photon trapped in the optical loop. The value of $\alpha$ and $\beta$ are configurable by applying various pulse energy of the retrieval light.

In our experiment, two read pulses are applied for retrieving the stored excitation state, and the energy of each read pulse is finely tuned to obtain the approximately same retrieval efficiency (see Methods for details), so that a maximally entangled state can be created. Once a Stokes photon is detected, the excitation will be mapped out into two temporal modes (denoted by mode A at $t_1$ and mode B at $t_2$) with the same probability, as a single-photon entangled state $(|1\rangle_{t_1}|0\rangle_{t_2} + |0\rangle_{t_1}|1\rangle_{t_2})/\sqrt{2}$. We will be able to read the entangled state by programing the storage time of both quantum memories in a coordinated fashion. Experimental setup and time sequences of the control and pump pulses are illustrated in FIG. 2.

Observing a strike at detector $D_S$ therefore heralds the presence of the entanglement between two quantum memories with a predictable time delay. To reveal the
quantum entanglement, the second incident read pulse is applied to convert the excitation state in motional atoms to optical mode, meanwhile, a read signal is applied to map out the flying photon in the optical loop by converting its polarization state back to horizontal. Therefore, the entanglement between two quantum memories is mapped to the entanglement between two anti-Stokes modes, which are then combined on a polarizing beam splitter.

After an extra-added controllable Pancharatnam-Berry’s phase [42] outside the all optical loop, two modes interfere on a half-wave plate and the following polarizing beam splitter. The interference can be observed by tuning the Pancharatnam-Berry’s phase, and the projected results of the retrieved single photon delocalized in two memories are recorded by the detectors after two ports N⁺ and N⁻, shown in FIG. 3. We observe the single-photon interference curve, and the calculated visibility value of 0.86 indicates that the coherence between two stored modes is preserved well during the storage time of 60 ns. Note that the phase difference induced by environment fluctuation between two optical modes in our experiment is rigorously stabilized by a beam of continuous wave light (phase-locking light) and a feedback circuit equipped with piezoelectric ceramics. In addition, the extra-added controllable Pancharatnam-Berry’s phase we introduce here is free of the phase-locking process, making it possible to control the phase arbitrarily. Details can be found in Methods.

The second-order correlation function $g_{AS}^{(2)}$ evaluates the performance of the systems to preserve quantum correlations. We therefore measure the second-order correlation functions between the heralding Stokes photons and the retrieved anti-Stokes modes after transmitting through the whole system, coming up to $15.39 \pm 0.26$. A violation of Cauchy-Schwarz inequality $\left(g_{AS}^{(2)}\right)^2 \leq g_{S-S}^{(2)} \cdot g_{AS-AS}^{(2)}$ [37] after the whole hybrid system is also observed as 209 SDs ($g_{S-S}^{(2)} = 1.72 \pm 0.12$ and $g_{AS-AS}^{(2)} = 1.56 \pm 0.26$), which indicates a high-fidelity generation and preservation of non-classical correlation at ambient condition.

The joint state of the retrieved anti-Stokes modes is described by a density matrix $\rho$ in the Fork state basis, in which the entanglement can be revealed by a tomographic approach [11, 43, 44]. According to the measurement results of the correlated photons, the density matrix can be deduced in the form in FIG. 4. The heralded probabilities of $p_{mn}$ represent the registration of $m$ photons in mode A, and $n$ photons in mode B, conditioned on a detected Stokes photons, $\{m, n\} = \{0, 1\}$. The off-diagonal terms represent coherence $d = V(p_{01} + p_{10})/2$, and $V$ is the visibility of interfering two heralded anti-Stokes modes obtained previously. Higher-order photon numbers are ignored. The concurrence of the density matrix $\rho$, therefore, is given by [11, 45]

$$C = \max(0, V(p_{01} + p_{10}) - 2\sqrt{p_{00}p_{11}}) \quad (2)$$

The positive value of concurrence indicates the entangled state, while the value of zero indicates separable state. To maximize the concurrence, from equation (2), one may increase the probabilities of detecting heralded states $(p_{01}, p_{10})$, and decrease the probabilities of detecting the separable states $(0|0)$ $(p_{00})$ and $(1|1) (p_{11})$, as well as improve the interference visibility ($V$). We calculate the concurrence in our system to be $1.63 \times 10^{-4}$, which is on the same order of magnitude as the maximum value $(C_{max} = 4.00 \times 10^{-4}$, when $V = 1$ and $p_{11} = 0$), here the $p_{11}$ is calculated according to the cross-correlation value with the relationship of $p_{11} = 4p_{10}p_{01}/(g_{S-AS}^{(2)} - 1)$ [46] based on the assumption
that the Duan-Lukin-Cirac-Zoller light source generates a two-mode-squeezed state [41]. Since local process cannot not increase the entanglement, our measurement gives a lower bound for the entanglement between two quantum memories. Note that the maximum concurrence is limited by the efficiency of the whole system, including heralding, retrieval, coupling and detecting efficiencies. The heralding and retrieval efficiency can be further improved in our experiment by introducing individual control of the write and read lasers, and employing higher transmittance optical elements, which is quite promising since the unit efficiency has been proven to be accessible [25, 34].

Since the two retrieved anti-Stokes modes are all vertically polarized when entering the optical loop, to map the first-arrived mode into the loop, a half-wave plate set at 22.5 degrees is placed just before the optical loop, as is shown in FIG. 2. Then we map the horizontally polarized component of the first mode into the loop, and after a programmable storage, it is retrieved and combined with the vertically polarized component of the second mode on PBS1 in both time and space domain. Alternatively, the half-wave plate can be replaced by a high-speed electrooptical modulator, so that the certain anti-Stokes modes can be selected with a doubled efficiency.

In summary, we herald a hybrid quantum entanglement between two different types of broadband quantum memories located on separated platforms at ambient condition, and reveal the entangled state between an all-optical loop and billions of motional atoms. The measured high cross-correlation values between Stokes and anti-Stokes photons identify the capability of quantum memories to preserve quantum correlations, and the single-photon interference visibility and concurrence between two heralded anti-Stokes modes exhibit that the entangled state is protected well during the storage time.

Quantum entanglement built and observed in such macroscopic hybrid matters is profound for the fundamental researches of exploring both quantum and classical worlds, or more specifically, it pushes the transition boundary from quantum to classical. For real-life applications, arbitrary qubit could be teleported to the all-optical loop or billions of motional atoms. Also, quantum repeaters based on memories could be built for quantum communications, which promises for scalable and high-speed quantum networks. More importantly, the large time bandwidth product and the ability of operating at ambient condition of both quantum memories make the network promptly applicable.

**METHODS**

**Programmable and high-intensity light pulse generation system.** The width of the control pulses in our far off-resonance Duan-Lukin-Cirac-Zoller protocol is around 4 ns, and the corresponding storage bandwidth is measured to be 300 MHz. In our experiment, we develop a programmable and high-intensity light pulse generation system to create control pulses. The output frequency of the laser diode is locked to the transition $6S_{1/2}, F = 4$ to $6S_{3/2}, F' = 4$ co 5 line of cesium, with 10 MHz resolution. Then a high-speed (10 GHz) electrooptical modulator is applied to chop the continuous laser into short pulses. Combined with an arbitrary waveform generator (2.4 GHz bandwidth), we can finely tune both the shape and energy of the control light sequences. The working temperature of the electrooptical modulator is locked by a temperature controller, and a feedback circuit on its bias voltage is also added to minimize the real-time background noise by monitoring the modulated light signals. After amplifying and filtering, the signal to noise ratio of the pulse light reaches 200:1. In our experiment, the pulse energy of the write pulse, the first read pulse and the second read pulse are set at 370.46 pJ, 521.19 pJ and 372.98 pJ respectively, so that the amplitudes of the two retrieved anti-Stokes modes after the whole system are approximately the same. The beam waist is measured as 360 μm.

**Locking an unbalanced interferometer.** The interference in our implementation is realized with an unbalanced interferometer of 9-meter-length difference. To observe a high visibility interference, the impact of environmental noise should be eliminated rigorously. Thus as is indicated in FIG. 2, another beam of auxiliary phase-locking light is injected into the optical loop to stabilize the phase jitter induced by temperature drift and other mechanical vibrations in the ambient. Note that such phase-locking light have to satisfy that its coherence time is much longer than the unbalanced length of interferometer, as well as that its frequency jitter is negligible. Therefore, we utilize the same laser source with the signal photons (part of the control light), to minimized the phase difference caused by frequency jitter of the laser sources. The frequency of the control light (also the phase-locking light) is locked to the transition $6S_{1/2}, F = 4$ to $6S_{3/2}, F' = 4$ co 5 line of cesium, with 4 GHz detuning. The phase-locking light and the signal photons are set to be parallel but not collinear, with opposite directions to avoid introducing noise photons. A piezoelectric ceramics is mounted on one of the reflector in the optical loop, cooperating with the photodetector and voltage source, to form a feedback circuit. The interference of classical phase-locking light detected by the photodetector is then modulated, demodulated, and after passing through a low-pass electrical filter, the signal is handled in the Proportion Integration Differentiation controller. Finally, the electrical signal derived from the PID controller is applied to the PZT to finely tuning the phase difference of the unbalanced interferometer. Remaining phase jitter comes from the slight misalignment between phase-locking light and signal photons, as well as the finite precision of frequency locking process (around 5 MHz), and note that the phase-locking light propagates around the loop once (for 3-meter-long), while the signal photons propagate around three times (for 9-meter-long),
calling for more strict phase-locking robustness.

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