Multiplexed storage and real-time manipulation based on a multiple degree-of-freedom quantum memory

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The faithful storage and coherent manipulation of quantum states with matter-systems would enable the realization of large-scale quantum networks based on quantum repeaters. To achieve useful communication rates, highly multimode quantum memories are required to construct a multiplexed quantum repeater. Here, we present a demonstration of on-demand storage of orbital-angular-momentum states with weak coherent pulses at the single-photon-level in a rare-earth-ion-doped crystal. Through the combination of this spatial degree-of-freedom (DOF) with temporal and spectral degrees of freedom, we create a multiple-DOF memory with high multimode capacity. This device can serve as a quantum mode converter with high fidelity, which is a fundamental requirement for the construction of a multiplexed quantum repeater. This device further enables essentially arbitrary spectral and temporal manipulations of spatial-qutrit-encoded photonic pulses in real time. Therefore, the developed quantum memory can serve as a building block for scalable photonic quantum information processing architectures.

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large-scale quantum networks would enable long-distance quantum communication and optical quantum computing. Due to the exponential photon loss in optical fibers, quantum communication via ground-based optical fibers is currently limited to distances of hundred of kilometers. To overcome this problem, the idea of quantum repeater has been proposed to establish quantum entanglement over long distances based on quantum memories and entanglement swapping. It has been shown that to reach practical data rates using this approach, the most significant improvements can be achieved through the use of multiplexed quantum memories.

The multiplexing of quantum memories can be implemented using any degree-of-freedom (DOF) of the photons, such as those in the temporal, spectral, and spatial domains. Rare-earth-ion-doped crystals (REIC) offer interesting possibilities as multiple-DOF quantum memories for photons by virtue of their large inhomogeneous bandwidths and long coherence time at cryogenic temperatures. Recently, there have been several important demonstrations using REIC, such as the simultaneous storage of 100 temporal modes by atomic frequency comb (AFC) featuring preprogrammed delays, the storage of tens of temporal modes by spin-wave AFC with on-demand readout and the storage of 26 frequency modes with feed-forward controlled readout. The orbital-angular-momentum (OAM) of a photon receives much attention because of the high capacity of OAM states for information transmission and spatial multimode operation. Tremendous developments have recently been achieved in quantum memories for OAM states, paving the way to quantum networks and scalable communication architectures based on this DOF.

To date, most experiments with quantum memories have been confined to the storage of multiple modes using only one DOF, e.g., temporal, spectral, or spatial. To significantly improve the communication capacity of quantum memories and quantum channels, we consider a quantum memory using more than one DOF simultaneously.

Here, we report on the experimental realization of an on-demand quantum memory storing single photons encoded with three-dimensional OAM states in a REIC. We present the results of a multiplexed spin-wave memory operating simultaneously in temporal, spectral and spatial DOF. In addition to expanding the number of modes in the memory through parallel multiplexing, a quantum mode converter (QMC) can also be realized that can perform mode conversion in the temporal and spectral domains simultaneously and independently. Indeed, our quantum memory enables arbitrary temporal and spectral manipulations of spatial-qutrit-encoded photonic pulses, and thus can serve as a real-time sequencer, a real-time multiplexer/demultiplexer, a real-time beam splitter, a real-time frequency shifter, a real-time temporal/spatial filter, among other functionalities.

**Results**

**Experimental setup.** A scheme of our experimental setup and relevant atomic level structure of Pr\(^{3+}\) ions is presented in Fig. 1. The memory crystal (MC) and filter crystal (FC) used in this setup are 3 × 6 × 3 mm crystals of 0.05% doped Pr\(^{3+}\) -Y\(_2\)SiO\(_5\), which are cooled to 3.2 K using a cryogen-free cryostat (Montana Instruments Cryostation). In order to maximize absorption, the polarization of input light is close to the D\(_2\)-axis of Y\(_2\)SiO\(_5\) crystal. To realize reliable quantum storage with high multimode capacity, we created a high-contrast AFC in MC (the AFC structure is shown in Supplementary Note 1). Spin-wave storage is employed to enable on-demand retrieval and extend storage time. The control and input light are steered towards the MC in opposite directions with an angular offset ~4° to reject the strong control field and avoid the detection of free induction decay noise. To achieve a low noise floor, we increase the absorption depth of the FC by employing a double-pass configuration. Figure 2a presents the experimental setup.*

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*Fig. 1.** Experimental setup and atomic levels. **a** Schematic illustrations of the experiment. The AFC is prepared in a memory crystal (MC) and a narrow spectral filter has been prepared in a filter crystal (FC). The beam waist of the input light is 65 μm at the middle of the MC. The pump/control light has a beam waist of 300 μm inside the MC to ensure good overlap with the high-dimensional input light. The input pulses are attenuated to single-photon level by neutral density filters (NDF). The spatial modes of these photons are converted into OAM superposition states by a spatial light modulator (SLM1). After storage in the MC, the retrieved signal passes through two consecutive acousto-optical modulators (AOM), which act as a temporal gate and a frequency shifter. The AOM are used in double-pass configuration to ensure the photons’ spatial mode unchanged when the frequency of photons is swept over tens of MHz. The SLM2 and a single-mode fiber (SMF) are employed to analyze the OAM states of the retrieved photons. The FC is double-passed with help of a polarization beam splitter (PBS), a half-wave plate (HWP) and a Faraday rotator (FR). Two bandpass filters (BPF) centered at 606 nm are employed to further suppress noises before the final detection of signal photons using single-photon detector (SPD). QWP quarter-wave plate. **b** Hyperfine states of the first sublevels of the ground and the excited states of Pr\(^{3+}\) in Y\(_2\)SiO\(_5\). The input field is resonant with 1/2g-3/2e, and the control field is resonant with 3/2g-3/2e (see Methods section for details).
modulators and single-mode fibers. We then characterized the memory operation using quantum process tomography. Figure 2b presents the real part of the experimentally reconstructed process matrix $\chi$. It is found to have a fidelity of $0.909 \pm 0.010$ with respect to the identity operation. This fidelity exceeds the classical bound of $0.831$ (see Supplementary Note 4 for details), thereby confirming the quantum nature of the memory operation. The nonunity value of the memory fidelity may be caused by the limited beam waist of the pump/control light, which may result in imperfect overlap with different OAM modes. Moreover, we noted that the memory performance for superposition states of $|L\rangle$ and $|R\rangle$ is much better than that achieved here (as detailed in Supplementary Note 2). The visibility of such two-dimensional superposition states is higher than the fidelity of the memory process in all three dimensions. This result indicates that the storage efficiency is balanced for the symmetrical LG modes but is not balanced for all the three considered spatial modes.

**Multiplexing storage in multiple DOF.** Carrying information in multiple DOF on photons can expand the channel capacity of quantum communication protocols. Here, we show that our solid-state memory can be simultaneously multiplexed in temporal, spectral, and spatial DOF. As shown in Fig. 3a, two AFC are created in the MC with an interval of 80 MHz between them to achieve spectral multiplexing. The two AFC have the same peak spacing of $\Delta = 200$ kHz and the same bandwidth of $\Gamma_{\text{AFC}} = 2$ MHz. The spin-wave storage efficiencies are 5.05% and 5.13%, for the first AFC and the second AFC, respectively. The temporal multimode capacity of an AFC is limited by $\Gamma_{\text{AFC}}/\Delta$. However, increasing the number of modes, the time interval between the last control pulse and the first output signal pulse will be reduced. Therefore, we employed only two temporal modes to reduce the noise caused by the last control pulse. The spatial multiplexing is realized by using three independent paths as input as shown in Fig. 3b. These paths, $s_1, s_2,$ and $s_3$, correspond to the OAM states as $|L\rangle$, $|R\rangle$, and $|G\rangle$ defined above. By combining all three DOF together, we obtain $2 \times 2 \times 3 = 12$ modes in total. The FC is employed to select out the desired spectral modes. Figure 3c illustrates the results of multimode storage over these three DOF for $\mu = 1.04$. The minimum crosstalk as obtained from mode crosstalk for each mode is $19.7 \pm 3.41$, which is calculated as one takes the counts in the diagonal term as the signal and then locates the large peaks over the range of output modes as the noise.

Here, the temporal, spectral and spatial DOF are employed as classical DOF for multiplexing. One can choose any DOF to carry quantum information. As a typical example, now we use the temporal and spectral DOF for multiplexing and each channel is encoded with spatial-qutrit state for each channel $\eta_{\text{SW}} = 5.51\%$. For an input with a mean photon number $\mu = 1.12$ per pulse, we have measured a signal-to-noise ratio (SNR) $\sim 39.7 \pm 6.7$ with the input photons in the Gaussian mode.

**Quantum process tomography.** The ability to realize the on-demand storage of photonic OAM superposition states in solid-state systems is crucial for the construction of OAM-based high-dimensional quantum networks. Quantum process tomography for qutrit operations benchmarks the storage performance for OAM qutrit in our solid-state quantum memory. The qutrit states are prepared in the following basis of OAM states: $|L_{G_{p=0}}^{l=0}\rangle$, $|L_{G_{p=0}}^{l=1}\rangle$, $|L_{G_{p=0}}^{l=2}\rangle$. Here, $|L_{G_{p=0}}^{l}\rangle$ corresponds to OAM states defined as Laguerre-Gaussian (LG) modes, where $l$ and $p$ are the azimuthal and radial indices, respectively. In the following, we use the kets $|L\rangle$, $|G\rangle$, and $|R\rangle$ to denote the OAM states $|L_{G_{p=0}}^{l=0}\rangle$, $|L_{G_{p=0}}^{l=1}\rangle$, and $|L_{G_{p=0}}^{l=2}\rangle$, respectively. For $\mu = 1.12$, we first characterized the input states before the quantum memory using quantum state tomography (see Methods section for details). The reconstructed density matrices of input are not ideal because of imperfect preparation and measurements based on spatial light
which is used as |G⟩ modes may need to be converted30. This device can ensure the preserving their quantum properties. Our device is expected to convert arbitrary temporal and spectral modes in real-time while for single-photon level inputs.

Arbitrary manipulations in real time. The precise and arbitrary manipulation of photonic pulses while preserving photonic coherence is an important requirement for many proposed photonic technologies31. In addition to the QMC functionality demonstrated above, the developed quantum memory can enable arbitrary manipulations of photonic pulses in the temporal and spectral domains in real time. As an example, we prepared the OAM qutrit state |ψ1⟩ in the f1t1 and f2t2 modes (Fig. 5a) as the input. Four typical operations were demonstrated, i.e., exchange of the readout times for the f1 and f2 photons, the simultaneous retrieval of the f1 and f2 photons at t1, shifting the frequency of f1 photons to f2 but keeping the frequency of f2 photons unchanged and temporal beam splitting the f2 photons but filtering out the f2 photons. These operations correspond to output of |ψ1⟩f1t2f2t1, |ψ1⟩f1t1f2t1, |ψ1⟩f1t1f2t2∗, and |ψ1⟩f1t2f2t2∗, respectively. Another example was implemented with the OAM qutrit state |ψ2⟩ = (|L⟩ + |G⟩ − i|R⟩)/√3 encoded in the f1t2 and f2t2 modes as the input, as shown in Fig. 5b with same output. The retrieved states were then characterized via quantum state tomography as usual (see Methods). Table 1 shows the fidelities between output states and input states.

Discussion
In conclusion, we have experimentally demonstrated a multiplexed solid-state quantum memory that operates simultaneously
in three DOF. The currently achieved multimode capacity is certainly not the fundamental limit for the physical system. Pr$^{3+}$: Y$_2$SiO$_5$ has an inhomogeneous linewidth of 5 GHz, which can support more than 60 independent spectral modes. The number of temporal modes that can be achieved using the AFC protocol is proportional to the number of absorption in the comb, which has already been improved to 50 in Eu$^{3+}$:Y$_2$SiO$_5$.$^{22}$ There is no fundamental limit on the multimode capacity in the OAM DOF since it is independent on the AFC bandwidth. The capacity in this DOF is simply determined by the useful size of the memory in practice. We have recently demonstrated the faithful storage of 51 OAM spatial modes in a Nd$^{3+}$:YVO$_4$ crystal.$^{26}$ The combination of these state-of-the-art technologies could result in a multimode capacity of 60 × 50 × 51 = 1,53,000 modes. This large capacity could greatly enhance the data rate in memory-based quantum networks and in portable quantum hard drives with extremely long lifetimes.$^{15}$

The developed multiple-DOF quantum memory can serve as a QMC, which is a fundamental requirement for the construction of scalable networks based on multiplexed quantum repeaters. Although it is not demonstrated in the current work, mode conversion in the spatial domain should also be feasible using a high-speed digital micromirror device.$^{38}$ QMC can also find applications in linear optical quantum computations. One typical example is to solve the mode mismatch caused by fiber-loop length effects and the time jitter of the photon sources in a boson sampling protocol.$^{19,40}$

Quantum communication and quantum computation in a large-scale quantum network rely on the ability to faithfully store and manipulate photonic pulses carrying quantum information. The presented quantum memory can apply arbitrary temporal and spectral manipulations to photonic pulses in real time, which indicates that this single device can serve as a variable temporal beam splitter$^{32,41}$ and a relative phase shifter$^{42}$ that enables arbitrary control of splitting ratio and phase for each output. Therefore, this device can perform arbitrary single-qubit operations.$^{43}$ Combining with the recent achievements on generation of heralded single photons$^{44,45}$, this device should provide the sufficient set of operations to allow for universal quantum computing in the Knill–Laflamme–Milburn scheme$^{46}$. Our results are expected to find applications in large-scale memory-based quantum networks and advanced photonic information processing architectures.

**Methods**

**AFC preparation.** We tailored the absorption spectrum of Pr$^{3+}$ ions to prepare the AFC using spectral hole burning.$^{35}$ The frequency of the pump light was first scanned over 16 MHz to create a wide transparent window in the Pr$^{3+}$ absorption line. Then, a 1.6 MHz sweep was performed outside the pit to prepare the atoms into the 1/2g state. The broad-band procedure created an absorbing feature of 2 MHz in width resonant with the 1/2g–3/2e transition, but simultaneously populated the 3/2g state, which, in principle, must be empty for spin-wave storage. Thus, a clean pulse was applied at the 3/2g–3/2e transition to empty this ground state. After the successful preparation of absorbing band in the 1/2g state, a stream of hole-burning pulses was applied on the 1/2g–3/2e transition. An AFC structure with a periodicity of Δ = 200 kHz is prepared in this step. These pulses burned the desired spectral comb of ions on the 1/2g–3/2e transition and antiholes at the 3/2g–3/2e transition; thus, a short burst of clean pulses was applied to maintain the emptiness of the 3/2g state. For AFC preparation, the remaining 3/2g ground state is used as an auxiliary state, which stores those atoms which do not contribute to the AFC components. To reduce the noise generated by the control pulses during spin-wave storage, we applied 100 control pulses separated by 25 μs and another 50 control pulses with a separation of 100 μs after the preparation of the comb.$^{35}$ An example of the AFC with a periodicity Δ = 200 kHz is illustrated in Fig. 3a. A detailed estimation of the structure and storage efficiency of the AFC memory is presented in Supplementary Note 1. The signal photons are mapped onto the AFC, leading to an AFC echo after a time $1/\Delta$. Spin-wave storage is achieved by applying two on-resonance control pulses to induce reversible transfer between the 3/2e state and 3/2g state before the AFC echo emission. The complete storage time is 12.68 μs in our experiment which includes an AFC storage time of 5 μs and a spin-wave storage time of 7.68 μs.
Filtering the noise. In order to achieve a low noise floor, temporal, spectral, and spatial filter methods are employed. The input and control beams are sent to the MC in opposite directions with a small angular offset for spatial filtering. Temporal filtering is achieved by means of a temporal gate implemented with two AOM. This AOM gate temporally blocked the strong control pulses. This is important to avoid burning a spectral hole in the FC and to avoid blinding the single-photon detector. We used two 2-nm bandpass filters at 606 nm to filter out incoherent fluorescence noise. The spectral of the filter mode was achieved by narrow-band spectral filter in the FC (shown by the dashed black line in Fig. 3a), which is created by 0.8 MHz sweep around the input light frequency, leading to a transparent window of approximately 1.84 MHz due to the power broadening effect. Furthermore, the FC is implemented in a double-pass configuration to achieve high absorption.

Quantum tomography. To characterize the memory performance for three-dimensional OAM states, quantum process tomography for the quantum memory operation is performed. Reconstructing the process matrix \( \chi \) of any three-dimensional state requires nine independent measurements. We chose three OAM eigenstates and six OAM superposition states as our nine input states, which are listed as follows: \( |L\rangle, |G\rangle, |R\rangle, (|L\rangle + |G\rangle)/\sqrt{2}, (|R\rangle + |G\rangle)/\sqrt{2}, (|L\rangle + |G\rangle)/\sqrt{2}, (-i|R\rangle + |G\rangle)/\sqrt{2}, (|L\rangle + |R\rangle)/\sqrt{2}, \) and \( (|L\rangle - i|R\rangle)/\sqrt{2} \). The complete operators for the reconstruction of the matrix \( \chi \) are as follows: 

\[
\lambda_1 = \begin{bmatrix} 1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & 1 \end{bmatrix}, \quad \lambda_2 = \begin{bmatrix} 0 & 1 & 0 \\ 1 & 0 & 0 \\ 0 & 0 & 0 \end{bmatrix}, \\
\lambda_3 = \begin{bmatrix} 0 & -i & 0 \\ i & 0 & 0 \\ 0 & 0 & 0 \end{bmatrix}, \quad \lambda_4 = \begin{bmatrix} 1 & 0 & 0 \\ 0 & -1 & 0 \\ 0 & 0 & 0 \end{bmatrix}, \\
\lambda_5 = \begin{bmatrix} 0 & 0 & 1 \\ 0 & 0 & 0 \\ 1 & 0 & 0 \end{bmatrix}, \quad \lambda_6 = \begin{bmatrix} 0 & 0 & -i \\ 0 & 0 & 0 \\ i & 0 & 0 \end{bmatrix}, \\
\lambda_7 = \begin{bmatrix} 0 & 0 & 0 \\ 0 & 0 & 1 \\ 0 & 1 & 0 \end{bmatrix}, \quad \lambda_8 = \begin{bmatrix} 0 & 0 & 0 \\ 0 & 0 & -i \\ 0 & i & 0 \end{bmatrix}, \\
\lambda_9 = \frac{1}{\sqrt{3}} \begin{bmatrix} 1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & -2 \end{bmatrix}.
\]

Table 1 Fidelities of qutrit states after temporal and spectral manipulations

| Input mode | Output mode | Fidelity |
|------------|-------------|----------|
| \( |\psi_1\rangle/C_11 \) | \( |\psi_1\rangle/C_11 \) | 0.881 ± 0.022 |
| \( |\psi_1\rangle/C_11 \) | \( |\psi_2\rangle/C_12 \) | 0.876 ± 0.022 |
| \( |\psi_1\rangle/C_11 \) | \( |\psi_1\rangle/C_11 \) | 0.897 ± 0.020 |
| \( |\psi_2\rangle/C_12 \) | \( |\psi_2\rangle/C_11 \) | 0.828 ± 0.019 |
| \( |\psi_2\rangle/C_12 \) | \( |\psi_2\rangle/C_12 \) | 0.896 ± 0.017 |
| \( |\psi_2\rangle/C_12 \) | \( |\psi_2\rangle/C_11 \) | 0.898 ± 0.013 |
| \( |\psi_2\rangle/C_12 \) | \( |\psi_2\rangle/C_11 \) | 0.898 ± 0.016 |
| \( |\psi_2\rangle/C_12 \) | \( |\psi_2\rangle/C_11 \) | 0.829 ± 0.025 |

The fidelity for the output mode of \( |\psi_1\rangle/C_11 \) is a little lower than the others because of the less photon counts in each output caused by the temporal splitting operation. The error bars for the fidelities correspond to one standard deviation caused by the statistical uncertainty of photon counts.
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