Deterministic and Storable Single-Photon Source Based on a Quantum Memory

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A single-photon source is realized with a cold atomic ensemble (^{87}Rb atoms). A single excitation, written in an atomic quantum memory by Raman scattering of a laser pulse, is retrieved deterministically as a single photon at a predetermined time. It is shown that the production rate of single photons can be enhanced considerably by a feedback circuit while the single-photon quality is conserved. Such a single-photon source is well suited for future large-scale realization of quantum communication and linear optical quantum computation.

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Although weak coherent beams can be used as a pseudo single-photon source, the advent of quantum information processing (QIP) has placed stringent requirements on single photons either on demand or heralded [1]. In particular, linear optical quantum computation [2] depends on the availability of such single-photon sources. The single-photon nature guarantees unconditional security and high efficiency in quantum cryptography [3]. Different approaches have been attempted in the last decade to develop an on-demand single-photon source, such as quantum dots [4,5], single atoms and ions [6,7], and color centers [8]. However, all of them are confronted with different challenges. For example, the single-atom implementation provides spectrally narrow single photons with a well-defined spatial mode, but the manipulation of single atoms requires sophisticated techniques and expensive setups [6]. Quantum dots are a potential source with high single-photon rate, but the requirement of spectral filtering entails inevitable losses. It is very difficult to prepare truly identical sources due to inhomogeneities in both the environment of the emitters and the emitters themselves [9]. Color centers are excellent sources, even at room temperature; however, the high peak intensities of a pulsed excitation can lead to complex and uncontrollable dark states [1]. So it has been taken as a formidable task to develop a promising deterministic single-photon source.

Moreover, an important challenge in distributed QIP is the controllable transfer of a quantum state between a flying qubit and macroscopic matter. Starting from a recent proposal for long-distance quantum communication with atomic ensembles [10], it is possible to implement both a single-photon source on demand and controllable transfer of a quantum state between a photonic qubit and macroscopic matter, provided that proper feedback is applied. A single spin excitation can be generated in an atomic ensemble by applying a series of subsequent clear (optical pumping) and write pulses stimulating spontaneous Raman scattering. The successful generation of a spin excitation is indicated by the detection of a corresponding Raman photon. This information is used as feedback to stop the sequence, and further on to start the next process, for example, to convert the spin excitation back into a single photon. Such a sequence can be taken as having a feedforward ability for the deterministically converted single photon.

Recently, significant experimental progresses have been achieved in demonstration of quantum storage and single-photon sources [11–14], and even entanglement between two atomic ensembles [15,16] has been generated. However, coincidence-based postselection was used in these experiments. No feedback was applied, and consequently the requirement of resources would increase exponentially with each new step of operation. This significantly limits the scalability of the schemes [2,10].

In this Letter, we present an experimental realization of a deterministic and storable single-photon source. Single spin excitations in an atomic ensemble are generated by detecting anti-Stokes photons from spontaneous Raman scattering. This detection allows one to implement feedforward and convert the spin excitations into single photons at a predetermined time. It is shown that the single-photon quality is conserved while the production rate of single photons can be enhanced significantly by the feedback circuit. In principle, the spatial mode, bandwidth, and frequency of single-photon pulses are determined by the spatial mode, intensity, and frequency of the retrieve laser [14]. It is feasible to integrate such a single-photon source with the storage medium, atomic ensembles. Together with the technology developed in previous experiments [11–16], our controllable single-photon source potentially paves the way for the construction of scalable quantum communication networks [10,17] and linear optical quantum computation [2].

The basic concept of our experiments is shown in Fig. 1. Cold atoms with Λ-type level configuration (two ground states $|a\rangle$, $|b\rangle$ and an excited state $|e\rangle$) collected by a

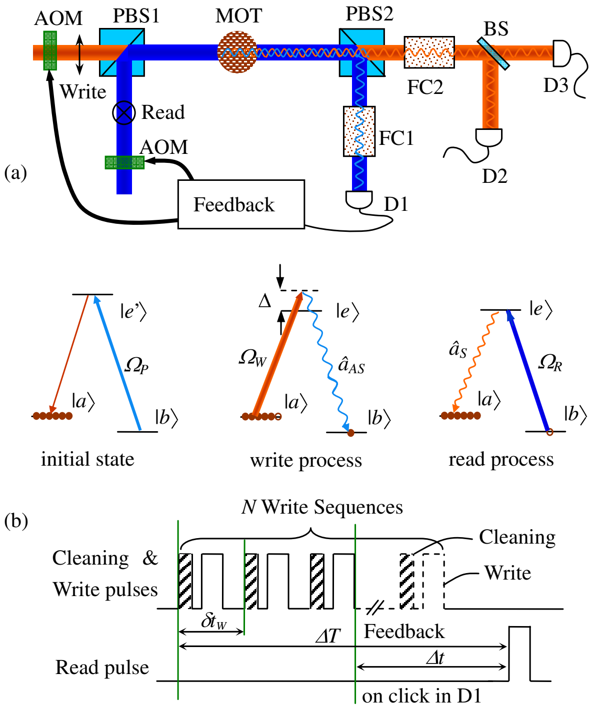

FIG. 1 (color online). (a) Illustration of the experimental setup and (b) the time sequence with the feedback circuit for the write and read process. The atomic ensemble is first prepared in the initial state $|a\rangle$ by applying a pump beam resonant with the transition $|b\rangle$ to $|e'\rangle$. A write pulse with the Rabi frequency Ω_W is applied to generate the spin excitation and an accompanying photon of the mode $\hat{a}_{\mathrm{AS}}$. Waiting for a duration Δt, a read pulse is applied with orthogonal polarization and spatially overlap with the write beam in PBS1. The photons, whose polarization is orthogonal to that of the write beam, in the mode $\hat{a}_{\mathrm{AS}}$ are spatially extracted from the write beam by PBS2 and detected by detector D1. Similarly, the field $\hat{a}_{\mathrm{S}}$ is spatially extracted from the read beam and detected by detector D2 (or D3). Here, FC1 and FC2 are two filter cells, BS is a 50/50 beam splitter, and AOM1 and AOM2 are two acousto-optic modulators.

magneto-optical trap (MOT) are used as the media for quantum memory. The atoms are initially optically pumped to state $|a\rangle$ by a pump laser. Then a weak classical *write* pulse, with the Rabi frequency Ω_W, close to the resonance of transition $|a\rangle$ to $|e\rangle$ is introduced in the atomic cloud. Because of the spontaneous Raman process, a photon of anti-Stokes field $\hat{a}_{\mathrm{AS}}$ is emitted into the forward scattering mode. Simultaneously, a collective spin excitation corresponding to the mode of the anti-Stokes field $\hat{a}_{\mathrm{AS}}$ is generated in the atomic ensemble [10,18]. The state of the field $\hat{a}_{\mathrm{AS}}$ and the collective spin state of the atoms can be expressed by the superposed state

$$|\Psi\rangle \sim |0_{\mathrm{AS}}0_b\rangle + \sqrt{\chi}|1_{\mathrm{AS}}1_b\rangle + \chi|2_{\mathrm{AS}}2_b\rangle + O(\chi^{3/2}), \quad (1)$$

where χ is the excitation probability of one spin flip and $|i_{\mathrm{AS}}i_b\rangle$ denotes the i-fold excitation of the anti-Stokes field and the collective spin. Ideally, conditioned on detecting one and only one anti-Stokes photon in detector D1, a single spin excitation is generated in the atomic ensemble with certainty. After a controllable time delay δt (in the order of the lifetime τ_c of the spin excitation), another classical *read* pulse with the Rabi frequency Ω_{R}, which is on resonance with the transition from $|b\rangle$ to $|e\rangle$, is applied to retrieve the spin excitation and to generate a photon of Stokes field $\hat{a}_{\mathrm{S}}$.

In our present experiment, more than 10^8 ^{87}Rb atoms are collected by the MOT with an optical depth of about 5 and the temperature of about 100 μK. The earth magnetic field is compensated by three pairs of Helmholtz coils. The two ground states $|a\rangle$ and $|b\rangle$ and the excited state $|e\rangle$ in the Λ-type system are $|5S_{1/2}, F = 2\rangle$, $|5S_{1/2}, F = 1\rangle$, and $|5P_{1/2}, F = 2\rangle$, respectively. The write laser is tuned to the transition from $|5S_{1/2}, F = 2\rangle$ to $|5P_{1/2}, F = 2\rangle$ with detuning of 10 MHz, and the read laser is locked on resonance to the transition from $|5S_{1/2}, F = 1\rangle$ to $|5P_{1/2}, F = 2\rangle$. By using orthogonal polarizations, write and read beams are spatially overlapped on a polarized beam splitter (PBS1), and then focused into the cold atoms with the beam waist of 35 μm. After passing the atomic cloud, the two beams are split by PBS2, which serves as the first stage of filtering the write (read) beam out from the anti-Stokes (Stokes) field. The leakage of the write (read) field from PBS2 propagating with the anti-Stokes (Stokes) field will be further filtered by a thermal cell filled with ^{87}Rb atoms, in which the rubidium atoms are prepared in state $|5S_{1/2}, F = 2\rangle$ ($|5S_{1/2}, F = 1\rangle$) initially. Coincident measurements among D1, D2, and D3 are performed with a time resolution of 2 ns.

After switching off the MOT, the atoms are optically pumped to the initial state $|a\rangle$. The write pulse containing about 10^4 photons with a duration of 100 ns is applied onto the atomic ensemble, to induce the spontaneous Raman scattering via $|a\rangle \rightarrow |e\rangle \rightarrow |b\rangle$. The state of the induced anti-Stokes field and the collective spin in Eq. (1) is generated with a probability $\chi \ll 1$. After a controllable delay of δt, the read pulse with the duration of 75 ns is applied for converting the collective excitation into the Stokes field. In comparison, the intensity of the read pulse is about 100 times stronger than that of the write one.

Assume that the probability of having an anti-Stokes (Stokes) photon is p_{AS} (p_{S}), and that the coincident probability between the Stokes and anti-Stokes channels is $p_{\mathrm{AS,S}}$, then the intensity correlation function $g^{(2)}_{\mathrm{AS,S}} = p_{\mathrm{AS,S}}/(p_{\mathrm{AS}}p_{\mathrm{S}})$. We measured the variation of $g^{(2)}_{\mathrm{AS,S}}$ as a function of p_{AS} shown in Fig. 2(a) with a time delay of $\delta t = 500$ ns. Considering the background in each channel, we obtain

$$p_{\mathrm{AS}} = \chi\eta_{\mathrm{AS}} + B\eta_{\mathrm{AS}}, \quad (2a)$$

$$p_{\mathrm{S}} = \chi\gamma\eta_{\mathrm{S}} + C\eta_{\mathrm{S}}, \quad (2b)$$

$$p_{\mathrm{AS,S}} = \chi\gamma\eta_{\mathrm{AS}}\eta_{\mathrm{S}} + p_{\mathrm{AS}}p_{\mathrm{S}}. \quad (2c)$$

Here, η_{AS} and η_{S} are the overall detection efficiencies in

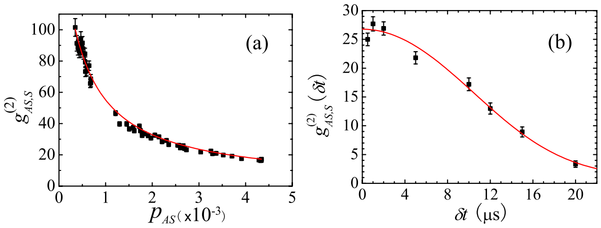

FIG. 2 (color online). Intensity correlation function $g^{(2)}_{\mathrm{AS,S}}$ along the excitation probability p_{AS} with $\delta t = 500$ ns (a) and along the time delay δt between read and write pulses with $p_{\mathrm{AS}} = 3 \times 10^{-3}$ (b). The black symbols are obtained from the current experiment, and the curves correspond to a least-square fit procedure according to Eqs. (2) and (3). The observed lifetime is $\tau_c = 12.5 \pm 2.6$ μs.

the anti-Stokes and Stokes channels, respectively, which include the transmission efficiency η_t of filters and optical components, the coupling efficiency η_c of the fiber couplers, and the quantum efficiency η_q of single-photon detectors (η_{AS} includes an additional spatial mode-match efficiency η_m [13]), γ is the retrieve efficiency which is a time-dependent factor, and B (C) is determined by the background in the anti-Stokes (Stokes) channel. The solid curve in Fig. 2(a) is the least-square fit result according to Eq. (2), assuming $B = 0$ for simplicity. The efficiency in the anti-Stokes channel is observed as $\eta_{\mathrm{AS}} \sim 0.07$ and the retrieve efficiency $\gamma \sim 0.3$. The largest correlation $g^{(2)}_{\mathrm{AS,S}}$ (101 ± 6) appears at the lowest excitation probability p_{AS} (3.5×10^{-4}).

The finite lifetime of the spin excitation results from the dephasing of the collective state due to the Larmor precession of the spins in the residual magnetic field. It can be characterized by the decay of the retrieve efficiency $\gamma(\delta t) = \gamma_0 \exp(-\delta t^2/\tau_c^2)$ [13], where τ_c is the lifetime of the collective state. It can be determined from the decay of the measured intensity correlation function $g^{(2)}_{\mathrm{AS,S}}(\delta t)$ as shown in Fig. 2(b), taken at $p_{\mathrm{AS}} = 0.003$. Using Eq. (2), the intensity correlation function reads

$$g^{(2)}_{\mathrm{AS,S}}(\delta t) = 1 + \frac{\gamma(\delta t)}{(B + \chi)\gamma(\delta t) + D}, \tag{3}$$

where C is absorbed by the new constant D. Our data give a lifetime of $\tau_c = 12.5 \pm 2.6$ μs. The cross correlation of the first point is slightly lower, which might be caused by noise arising from the elastic scattering of the write beam.

In order to increase the efficiency of the single-photon source we apply a feedback protocol. As shown in Fig. 1(b), in the time interval ΔT, N independent write sequences with a period of δt_W are applied to the atomic ensemble. Each write sequence contains a cleaning pulse (the optical pumping to the initial state) and a write pulse. Once an anti-Stokes photon is detected by D1, the feedback circuit stops the further write sequence and enables the read pulse to retrieve the single Stokes photon after a time delay Δt. The maximum number of trials (N) is given by the lifetime of the excitation. The feedback protocol enhances the production rate of Stokes photons according to the new excitation probability $P_{\mathrm{AS}} = \sum_{i=0}^{N-1} p_{\mathrm{AS}}(1 - p_{\mathrm{AS}})^i$ while the single-photon quality is conserved.

Our protocol can be executed in different modes. In a first mode, one can fix the retrieve time ΔT. Therefore, the delay Δt varies because the spin excitation is created randomly by one of the write sequences. Single photons are produced at a given time with a high probability, ideally approaching unity if $N \gg 1$. Furthermore, the retrieve efficiency could be improved significantly by an increased optical depth of the atomic ensemble and an optimal retrieve protocol [19]. This mode serves as a deterministic single-photon source. In a second mode, we retrieve the single photon with a fixed delay Δt after a successful write. More generally, the imprinted single excitation can be converted into a single photon at any given time within the lifetime τ_c. This is well suited for a quantum repeater [10,17] where one needs to synchronize the nodes.

In the first experiment, we fixed $\Delta T = 12.5$ μs and $\delta t_W = 1$ μs, and $N = 12$ subsequent write sequences were applied. The quality of the single-photon source can be characterized by the anticorrelation parameter α [20], which is equivalent to the second-order autocorrelation function $g^{(2)}_{\mathrm{S,S}}$ of the Stokes photon on the condition of an anti-Stokes photon is detected. When we use N write pulses and the feedback protocol, the detection probabilities in D2, D3, and the coincidence detection probability D23 conditioned on a registration of an anti-Stokes photon in D1 are

$$P_{m|\mathrm{AS}} = \frac{\sum_{i=0}^{N-1} p_{\mathrm{AS}}(1 - p_{\mathrm{AS}})^i p_{m|\mathrm{AS}}(\Delta T - i\delta t_W)}{\sum_{i=0}^{N-1} p_{\mathrm{AS}}(1 - p_{\mathrm{AS}})^i}, \tag{4}$$

where $m = 2, 3, 23$ and $p_{m|\mathrm{AS}}(\Delta T - i\delta t_W)$ is a time-dependent probability conditioned on a click in the anti-Stokes channel. The anticorrelation parameter α is given by $P_{23|\mathrm{AS}}/(P_{2|\mathrm{AS}}P_{3|\mathrm{AS}})$.

Figure 3(a) shows the measured α as a function of the excitation probability p_{AS}. For $N = 1$ (black) the variation of α is nearly linear in the region of $p_{\mathrm{AS}} = 0$–0.006. The black curve is the fit according to Eq. (4). When using 12 successive write sequences, we plot α vs $12p_{\mathrm{AS}}$ as solid circles. The red (gray) line is a no free parameter calculation from Eq. (4), taking the fitted parameters from $N = 1$, setting $N = 12$. We note that, for $p_{\mathrm{AS}} \to 0$, the value of α is 0.057 ± 0.028, which in principle should be 0. This offset comes from noise including residual leakage of the write and read beams, stray light, and dark counts of the detectors. However, the advantage of the feedback protocol is not degraded by such noise. It is verified that α is conserved even with enhanced excitation probability. If the lifetime of the spin excitation is sufficiently long to allow many write sequences, the excitation probability can reach unity while the single-photon nature is still con-

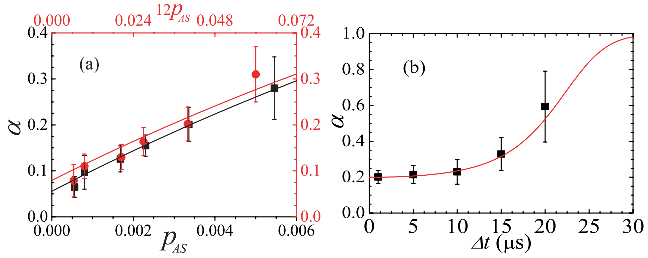

FIG. 3 (color online). The anticorrelation parameter as a function of (a) p_{AS} and (b) Δt. In panel (a), the data in black correspond to the experiment without feedback circuit, in which each write sequence is followed by one read pulse. The data in red (gray) correspond to the experiment with feedback circuit, in which 12 successive write sequences are followed by one read pulse. The red (gray) curve is the theoretical evaluation taking into account the fitted background of the black symbols. In panel (b), 12 write sequences were applied in each trial while measuring.

served. Then the generation efficiency depends on only the retrieve efficiency itself.

In the second experiment, we use $\delta t_W = 1\ \mu$s and $N = 12$. Figure 3(b) shows the measured α as a function of Δt. For every Δt, ΔT varies due to the random creation of the spin excitation by the N write sequences. The behavior of $\alpha(\Delta t)$ is related to a reversed profile of $g^{(2)}_{\mathrm{AS,S}}(\delta)$ in Fig. 2(b). For the delay $\Delta t < \tau_c$, the value of α stays at a low level and varies slowly. For $\Delta t > \tau_c$, $\alpha(\Delta t)$ increases towards 1. But even for a delay of 20 μs ($\sim 2\tau_c$) we find $\alpha \sim 0.6$. A satisfying agreement is observed between the theoretical curve and the experimental data.

Typically, the single spin excitation can be produced at a rate of 600 per second, while with a detected success probability per trial of 2.5%, the overall detection rate of single-photon production is $\sim 15\ \mathrm{s}^{-1}$. As demonstrated in the present work, the lifetime of collective states is important for the quality and production rate of single photons. In the atomic ensemble, the coherence time of the collective state suffers from the residual magnetic field around the MOT and the thermal motion of the atoms. The latter effect is negligible because of the very low temperature of the atomic cloud. Using a better compensation of residual magnetic field or using field insensitive clock states we can significantly increase the lifetime of the collective state. Moreover, by further improving the control circuit, i.e., reducing the period of write pulses, we can apply more write pulses within the lifetime. In particular, in the case with $p_{\mathrm{AS}} = 0.003$ and a write period of 300 ns, we can obtain a single-photon source with a probability as high as 95% within a lifetime of 300 μs.

In conclusion, we have demonstrated an experimental realization of an controllable single-photon source with atomic storage. The lifetime of the collective spin excitation reaches 12.5 μs. A feedback circuit was constructed to control the generation of the spin excitation and the storage time δt. Being a key device in the scalable quantum communication network, this circuit also shows a promising performance in the enhancement of the excitation probability while the single-photon quality is conserved. This single-photon source is able to work at either a deterministic mode or a time controllable mode heralded by the feedback circuit. The single-photon source based on an atomic ensemble has the advantages of narrow band, high quality, and controllable character, which is helpful for the construction of scalable quantum information processing system in the future.

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Note added.—During the final phases of our experiment we became aware of a related experiment by Matsukevich *et al.* [21].

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