Entanglement of the orbital angular momentum states of the photon pairs generated in a hot atomic ensemble

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In this paper, we experimentally demonstrate that the photon pairs generated via spontaneous four-wave mixing in a hot atomic ensemble are in entangled orbital angular momentum (OAM) states. The density matrix of the OAM states of the photon pair is reconstructed, from which the fidelity to the maximal entangled state and the concurrence are estimated to be about 0.89 and 0.81, respectively. The experimental result also suggests the existence of the entanglement concerned with spatial degrees of freedom between the hot atomic ensemble and the Stokes photon.

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Entanglement, as one of the most interesting phenomena of quantum mechanics, plays a central role in quantuminformation field [1]. Higher-dimensional entangled states enable us to achieve more efficient quantum-information processes [2,3], for example, a quantum key distribution scheme with higher alphabets allows one to increase the flux of information. Such higher-dimensional entangled states can be created by using orbital angular momentum (OAM) states of photons, because the OAM states can be used to define an infinite-dimensional Hilbert space [4]. Since the pioneering experiment of generating photon pairs with OAM entangled states using parametric down-conversion [5], many protocols based on higher-dimensional entangled states have been demonstrated by using such photonic states [6–8].

Photons are very promising carriers in the quantum communication. Because of the losses and decoherence of the photons in the channel, quantum repeaters are needed in long-distance quantum communication. Duan et al. have shown that an atomic ensemble is a good candidate of the quantum repeater [9]. To realize long-distance quantum communication with higher-dimensional quantum states, the transferring of the OAM states between a photon and an atomic ensemble is required. Such transferring between classical light and cold atoms [10-12] or hot atoms [13] has been reported in the past years. Very recently, Inoue et al. have clarified that a photon and a cold atomic ensemble can be in an entangled OAM state. However, so far there has been no similar discussion or experiment in a hot atomic ensemble. In this paper, we experimentally demonstrate that the photon pairs generated via spontaneous four-wave mixing (SFWM) in a hot atomic ensemble are in the entangled OAM states. The density matrix of the OAM states of the photon pairs is reconstructed, from which, the concurrence and the fidelity are estimated. This entanglement suggests that it is possible to entangle the OAM states of a photon and a hot atomic ensemble.

We consider an ensemble of atoms with a four-level structure of $|a\rangle$, $|b\rangle$, $|c\rangle$, and $|d\rangle$ as shown in Fig. 1(a). A strong coupling laser resonant with the $|b\rangle \rightarrow |c\rangle$ transition continuously drives the atoms into the level $|a\rangle$. A weak pump laser tuned to the $|a\rangle \rightarrow |d\rangle$ transition counter propagates with the coupling laser. The pump laser is weak and will induce the $|d\rangle \rightarrow |b\rangle$ transition occasionally. When a Stokes photon is emitted, the atomic ensemble collapses into the state $\frac{1}{\sqrt{N}} \sum_j |a_1, a_2, \dots, b_j, \dots, a_N\rangle$, which is the superposition of all possible final states with one of the atoms transferred to $|b\rangle$. The strong coupling laser restores the ensemble to the state $|a_1, a_2, \dots, a_N\rangle$, and at the same time an anti-Stokes photon is generated. This process is some kind of four-wave-mixing process, therefore the momentums and the OAMs of the photons are expected to be conserved [13,14], i.e.,



FIG. 1. (Color online) (a) Energy levels of ⁸⁷Rb, and the frequency arrangement of the lasers used in this experiment, where $|a\rangle$ and $|b\rangle$ are the hyperfine sublevels of the ground state, and $|c\rangle$ and $|d\rangle$ are excited states. (b) Schematic setup of our experiment. A strong coupling laser and a weak pump laser with orthogonal linear polarization counterpropagate through the rubidium cell. Paired Stokes photons and anti-Stokes photons are generated in phasematched directions. H1 and H2 are computer-generated holograms; SMF1 and SMF2 are single-mode fibers, which collect the ∓ 1 diffraction of H1 and H2, and connect to single photon-counting modules (SPCM); F1 and F2 are filters, each of which is combined with an optical pumped rubidium cell and a ruled diffraction grating.

$$\vec{k}_S + \vec{k}_{AS} = \vec{k}_P + \vec{k}_C,$$

$$L_S + L_{AS} = L_P + L_C,$$
(1)

where \vec{k}_i and L_i represent the wave vectors and the OAMs of the corresponding photons, respectively. According to Eq. (1), when the pump laser and the coupling laser carry zero OAM, the Stokes photon and the anti-Stokes photon are in the entangled state

$$|\Psi\rangle = C \sum_{i=-\infty}^{+\infty} \alpha_i |i\rangle_S |-i\rangle_{AS}, \qquad (2)$$

where *C* is the normalization coefficient, and α_i is the relative amplitude of the OAM state. In this work we only investigate the entanglement concerned with *i*=0 and 1, thus the expected entangled state can be written as

$$|\Psi\rangle = C(|0\rangle_S |0\rangle_{AS} + \alpha_1 |1\rangle_S |-1\rangle_{AS}). \tag{3}$$

Although we only discuss the two-dimensional case, it is natural to presume that our discussion can be extended into high-dimensional cases over a wide range of OAM [15].

A Gaussian mode beam, which can be discriminated by using a single-mode fiber, is in the Laguerre-Gaussian (LG) mode when it carries a well-defined OAM [16], and can be described by the LG_{pl} mode, where p+1 is the number of the radial nodes, and l is the number of the 2π -phase variations along a closed path around the beam center. Here we consider the case of p=0, in which the LG_{0l} mode carries the corresponding OAM of $l\hbar$ per photon. The LG_{0l} mode has a doughnut-shape intensity distribution

$$E_{0l}(r,\varphi) = E_{00}(r) \frac{1}{\sqrt{|l|!}} \left(\frac{r\sqrt{2}}{w}\right)^{|l|} e^{-il\varphi},$$
(4)

where w is the beam radius, and

$$E_{00}(r) = \sqrt{\frac{2}{\pi} \frac{1}{w}} \exp\left(-\frac{r^2}{w^2}\right)$$

is the radial amplitude distribution of a Gaussian mode beam which carries zero OAM (LG_{00}) . A computer-generated hologram (CGH), with a fork structure at the center, is used to change the order of the OAM of a beam [17]. The ± 1 order diffraction of the CGH increases the OAM of the input beam by $\pm 1\hbar$ per photon, when the dislocation of the hologram overlaps with the beam center. The superposition of the LG_{00} mode and the LG₀₁ mode can be achieved by shifting the dislocation of the hologram out of the beam center a certain amount [5,18]. Therefore, the combination of the CGH and a single-mode fiber can be used to collect the various superpositions of the $LG_{0\mp 1}$ mode and the LG_{00} mode. It should be noted that there are also higher-order LG modes in the firstorder diffraction, but their intensities are small compared with the $LG_{0\pm 1}$ mode and the LG_{00} mode [18], so the influence of them is ignored in this paper.

The schematic setup of the experiment is shown in Fig. 1(b). A natural rubidium cell with a length of 5 cm is used as the working medium. The temperature of the cell is kept at about 50 °C, corresponding to an atomic intensity of about



FIG. 2. (Color online) Time-resolved coincidence counts between the Stokes photons and the anti-Stokes photons. The data is accumulated for about 1000 seconds and then normalized in time. τ is the relative delay between the Stokes photons and the anti-Stokes photons.

 1×10^{11} /cm³. The coupling laser vertically linear polarized has an intensity of about 7 mW. The pump laser horizontally polarized has an intensity of about 60 μ W. The $1/e^2$ diameters of these two lasers are about 2 mm. The vertically polarized Stokes photons emitted at an angle of about 4° to the lasers are diffracted by a CGH (H1), and the -1 order diffraction is coupled into a single-mode fiber (SMF1). The horizontally polarized anti-Stokes photons in the phasematched direction are diffracted by H2, and the +1 order diffraction is coupled into SMF2. The diffraction efficiencies of these CGHs are about 40%. The SMF1 (SMF2) collects the desired superposition of $|0\rangle_{S}$ ($|0\rangle_{AS}$) and $|1\rangle_{S}$ ($|-1\rangle_{AS}$) according to the displacement of the H1 (H2), where the displacement of the CGH is defined as the distance between the dislocation of the CGH and the beam center. The collected photons are detected by photon-counting modules (Perkin-Elmer SPCM-AQR-15), and the time-resolved coincidence counts between the Stokes photons and the anti-Stokes photons are recorded by using a time digitizer (FAST ComTec P7888-1E) with a 2 ns bin width and total of 160 bins. The Stokes photons are used as the "start" signals of the time digitizer, and the anti-Stokes photons with certain delay are used as the "stop" signals.

Figure 2 shows the time-resolved coincidence counts between the Stokes photons and the anti-Stokes photons in the LG₀₀ mode. This coincidence is obtained when the displacements of both CGHs are far larger than the radii of the beams, and the effects of the CGHs on the mode of the photons can be ignored. The figure shows that the maximum coincidence count is obtained at a relative delay of 12 ns between the Stokes photon and the anti-Stokes photon. This delay is caused by the time used for restoring the ensemble, which is mainly determined by the Rabi frequency of the coupling field [19]. The correlation function obtained at this relative delay is $g_{S,AS}(12 \text{ ns})=n(12 \text{ ns})/c_b=1.57\pm0.04$, where $n(\tau)$ is the coincidence count of each bin, and c_b is the background count obtained by averaging the coincidence counts at $\tau > 50$ ns. The counting rates of the Stokes photons



FIG. 3. (Color online) Coincident counts versus the displacements of H2 with different displacement of H1. (a) shows the results that the Stokes photons are in stationary states $|0\rangle$ (red squares) and $|1\rangle$ (green dots); (b) shows the results that the Stokes photons are in the superposition states $(|0\rangle \pm |1\rangle)/\sqrt{2}$. The data are fitted by using the square of Eq. (5) with w=0.8 mm.

and the anti-Stokes photons are $1.4 \times 10^4/\text{s}$ and $4.0 \times 10^4/\text{s}$, respectively. The larger counting rate of the anti-Stokes photons is caused by the quick moving of the atoms. The atoms in state $|b\rangle$ moving into the coupling laser contribute to uncorrelated anti-Stokes photons. Even when the pump beam is absent, the counting rate of the anti-Stokes photons is larger than 20 000/s. These uncorrelated photons cause the large background in the coincidence. The correlated time shown in Fig. 2 is about 30 ns.

In order to evaluate the quantum correlation of the OAM states, we measure the coincidence counts with certain displacements of the holograms. Figure 3 shows the results when H1 is fixed at various displacement while the displacement of H2 is swept. Every point is obtained by $N = \sum_{\tau=2}^{32} \frac{ns}{ns} [n(\tau) - c_b] / c_b$, which guarantees that most of the correlated anti-Stokes photons are taken into account. Every point is accumulated for more than 500 seconds. The data are fitted with the square of the projection function [17]

$$a(x0) = \int \int e^{-i \arg(r \cos \varphi - x0, r \sin \varphi)} \times u_{AS}(r) u_S(r, \varphi)^* r dr d\varphi,$$
(5)

where $\arg(x, y)$ is the argument of the complex number x + iy, $e^{-i \arg(r \cos \phi - x0, r \sin \phi)}$ represents the transmitting function of H2 with a displacement of x0, $u_{AS}(r) = E_{00}(r)$ is the field amplitude of the anti-Stokes photon collected by the singlemode fiber after being diffracted by the hologram (Gaussian mode), and $u_{S}(r, \phi) = \cos \theta E_{00}(r) + \sin \theta E_{01}(r, \phi)$ is the field amplitude of the Stokes photons collected by the singlemode fiber. The value of $\cos \theta$ (sin θ) is controlled by the displacement of H1. Equation (5) gives the projection between the different OAM modes. In this paper the u_i 's are the amplitudes of the Stokes photons and the anti-Stokes photons, respectively. This equation is tenable only when the collapse of the Stokes photon leads the anti-Stokes photon collapses into the corresponding state. Therefore, the Stokes photon and anti-Stokes photon should be in a quantum correlated state, if Eq. (5) always holds, no matter if the Stokes photon collapses to a stationary state or to a superposition state. In Fig. 3(a), the red squares show the results of the coincidence counts versus the displacements of H2 when the displacement of H1 is far larger than the radius of the Stokes



FIG. 4. (Color online) Graphical representation of the reconstructed density matrix. (a) is the real part and (b) is the imaginary part.

photons, and the green dots show the results when the displacement of H1 is 0. The red line in Fig. 3(a) is fitted with $\theta = 0$ and the green dashed line is fitted with $\theta = \pi/2$, which means the Stokes photons are in LG₀₀ and LG₀₁ modes, respectively. This figure clearly indicates the correlation of OAM between the Stokes photons and the anti-Stokes photons. However, such a correlation can be obtained even when the photon pairs are in a mixture of the $|0\rangle_{S}|0\rangle_{AS}$ states and the $|1\rangle_{S}|-1\rangle_{AS}$ states. To further demonstrate the quantum correlation of the Stokes photons and the anti-Stokes photons, we measure the correlation in a different basis. We displace H1 by a certain amount, which makes the collected Stokes photons be in the superposition states $1/\sqrt{2}(|0\rangle \pm |1\rangle)$, and then sweep H2. The results are shown in Fig. 3(b). The data fit well with the theoretical prediction. Therefore, Fig. 3 shows that the Stokes photons and the anti-Stokes photons are in strongly quantum correlated OAM states.

Furthermore, we perform a two-qubit state tomography [20] to get the full state of the Stokes photons and the anti-Stokes photons. The density matrix is reconstructed from the coincidences with various combinations of the measurement basis. A graphical representation of the reconstructed density matrix is shown in Fig. 4. From the density matrix, the fidelity [21] to the maximally entangled state $|\Psi\rangle = (|0\rangle_S |0\rangle_{AS}$ $+|1\rangle_S |-1\rangle_{AS})/\sqrt{2}$, the concurrence, and the entanglement of formation are estimated to be about 0.89, 0.81, and 0.74, respectively. All of these results clearly demonstrate that the Stokes photon and the anti-Stokes photon are in an entangled OAM state [22].

In the SFWM process, the Stokes photon and the anti-Stokes photon are not generated simultaneously. When a Stokes photon is generated, the hot atomic ensemble collapses into the $\frac{1}{\sqrt{N}}\Sigma_j|a_1,a_2,\ldots,b_j,\ldots,a_N\rangle$ state, which may be entangled with the Stokes photon. Lately the information of the atomic ensemble is retrieved by using the coupling laser, and an anti-Stokes photon carrying the information of the atomic ensemble is emitted. Therefore, the entanglement of OAM states between the Stokes photon and the anti-Stokes photon may suggest the existence of the entanglement of OAM states between the Stokes photon and the hot atomic ensemble.

Here we discuss the influence of the CGH used for mode detection. It is mentioned above that there are also higherorder LG modes in the diffraction of the CGH. The influence of the higher-order modes to the experiment is estimated as follows. Using the projection function, the theoretical detected density matrix is reconstructed. Supposing that the photon pairs are in the maximal entangled state with full LG modes, the expected fidelity of the two-qubit density matrix is estimated to be about 0.93 when Eq. (5) is used directly, and will be about 0.98 if the Stokes photons are treated in the same way as the anti-Stokes photons in the projection function. These results show that the existence of higher modes in the first-order diffraction of the CGH will make the reconstructed density matrix imperfect, but the influence is small. The superposition of the OAM states is decided by the displacement of the CGH, which depends on the beam radius. Therefore, the fluctuation of the beam position or radius may bring error. A theoretical simulation shows that the variation of the fidelity is less than 0.01 when the change of the beam position or radius is 0.01w. These theoretical simulations suggest that the error imported by the CGH is acceptable. Therefore, we conjecture that the main cause of the low fidelity is the fluctuation of the coincidence counts, which might be caused by the Poisson fluctuation, by the large decay between the ground states of the atom or by the instability of laser.

In summary, we demonstrate that the Stokes photons and the anti-Stokes photons generated using SFWM in the hot atomic ensemble are in the OAM entangled states. The density matrix of the OAM state is reconstructed via a tow-qubit tomography, from which the fidelity to the maximal entangled state and the concurrence are estimated to be about 0.89 and 0.81, respectively. The experimental result also suggests the existence of the entanglement concerned with the spatial degree of freedom between the hot atomic ensemble and the Stokes photon.

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