Nonclassical photon pairs generated from a room-temperature atomic ensemble

Wei Jiang$^1$, Chao Han$^1$, Peng Xue$^1$, L.-M. Duan$^{1,2}$, G.-C. Guo$^1$

$^1$Key Laboratory of Quantum Information, University of Science and Technology of China, Hefei 230026, P. R. China

$^2$Department of Physics, University of Michigan, Ann Arbor, MI 48109-1120

We report experimental generation of non-classically correlated photon pairs from collective emission in a room-temperature atomic vapor cell. The nonclassical feature of the emission is demonstrated by observing a violation of the Cauchy-Schwarz inequality. Each pair of correlated photons are separated by a controllable time delay up to 2 microseconds. This experiment demonstrates an important step towards the realization of the Duan-Lukin-Cirac-Zoller scheme for scalable long-distance quantum communication.

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Implementation of quantum communication and computation is of large current interest [1]. Recently, significant interests arise in using atomic ensembles for realization of spin squeezing [2–5], quantum memory [6–9], and quantum information processing [10–15]. A significant result in this direction is a scheme proposed by Duan, Lukin, Cirac and Zoller (hereafter “DLCZ”) which gives a promising approach to realization of scalable long-distance quantum communication [14]. This scheme overcomes the photon attenuation problem and can communicate quantum states over arbitrarily long distance with only polynomial costs based on a clever implementation of the quantum repeater architecture [16,17]. In the DLCZ scheme, one generates and purifies entanglement between distant atomic ensembles by interfering and counting single-photons emitted from them. The same setup can also be used to prepare maximally entangled states between many such ensembles [15]. The challenging and important enabling step for these schemes is to demonstrate quantum correlation between the emitted single-photons and the long-lived collective atomic excitations. In this experiment, we realize this important step by observing non-classical correlation between them in a room-temperature atomic vapor. The collective atomic excitation is transferred subsequently to a photon in experiments, so actually we observe nonclassical correlation between two successively emitted photons.

Very recently, two other experiments have been reported contributing to the realization of the DLCZ scheme [18,19]. In particular, Kuzmich et al. have reported striking results on generation of nonclassical photon pairs in a sample of cold atoms. Compared with those experiments, our experiment is distinctive by the following features: (i) We observe nonclassical correlation between photon pairs generated from a room-temperature atomic vapor cell. The observed correlation is comparable to that reported in Ref. [18], but our setup of room-temperature atoms is much cheaper compared with the magnetic-optical trap used for cold atoms [18], and the use of cheaper setups could have advantage in future full realization of the DLCZ scheme where many such basic setups are required. The experiment in Ref. [19] is also based on the use of an atomic vapor cell, however, it is not performed in the single-photon region as required by the DLCZ scheme. (ii) Compared with instantaneous photon pairs produced previously in atomic cascades [20] or in parametric down conversion [21], the atomic ensemble experiments are distinct in that the correlated photon pairs generated in this system can be separated with a controllable time delay, which is in principle only limited by the single-photon storage time in the atomic ensemble. In our experiment, we demonstrate a time delay of 2 microseconds between the pair of non-classically correlated photons, which is considerably longer than the 400-nanosecond time delay reported before [18]. It is important to experimentally improve this storage time to facilitate the full realization of the DLCZ scheme which requires quantum memory [14,15].

The basic idea behind this experiment can be understood by considering a sample of three-level atoms in a Λ type configuration as illustrated in Fig. 1b. Most of the atoms are initially prepared in the state $|a\rangle$ through optical pumping. A write laser pulse is then sent through the atomic ensemble which couples off-resonantly to the atomic transition $|a\rangle \rightarrow |c\rangle$. This pulse induces a Raman scattering, bringing a small fraction of the atoms into the level $|b\rangle$ by emitting Stokes photons from the transition $|c\rangle \rightarrow |b\rangle$. The write pulse is controlled to be weak so that the average Stokes photon number scattered into the specified forward propagating mode $\psi_w (\mathbf{r})$ is much smaller than 1 for each pulse [14,22]. We detect the photon in this mode, and upon a detector click, one atom will be excited to the collective atomic mode $s^\dagger$ defined as [14,22]

$$s^\dagger = \left(1/\sqrt{N_a}\right) \sum_{i=1}^{N_a} |b\rangle_i \langle a|,$$  \hspace{1cm} (1)

where $N_a$ is the total involved atom number. It has been predicted that there will be a definite correlation between the photon number in the forward propagating mode $\psi_f (\mathbf{r})$ and the atom number in the collective mode $s$ [14], and this correlation is critical for all the applications based on this setup, including the implementation of quantum repeaters. To experimentally confirm this correlation, we transfer the collective atomic excitation...
to a photonic excitation after a time delay $\delta t$ by shining a read laser pulse on the ensemble which couples to the transition $|b\rangle \rightarrow |c\rangle$. This read pulse will bring the atom back to the state $|a\rangle$ by emitting an anti-Stokes photon in a specified forward-propagating mode $\psi_r(\mathbf{r})$. We can then detect the photon coincidences in the write and the read modes $\psi_w(\mathbf{r})$ and $\psi_r(\mathbf{r})$. The mode function $\psi_r(\mathbf{r})$ is determined by the spatial shapes of the write and the read pulses, the geometry of the atomic ensemble, and the mode structure of $\psi_w(\mathbf{r})$ [22]. In experiments, one can choose the waists of the write and the read laser beams so that $\psi_w(\mathbf{r})$ and $\psi_r(\mathbf{r})$ largely overlap with the spatial shapes of these pumping beams. The Stokes and anti-Stokes photons in these modes are coupled into single-mode fibers, which direct them to single-photon detectors for coincidence measurements.

In our experiment, we use an optically thick ensemble of $^{87}\text{Rb}$ atomic vapor contained in a silica cell. There is also some buffer gas (Ne, 30 Torr) inside the cell which is used to increase the spin relaxation time of the $^{87}\text{Rb}$ atoms. The number density of $^{87}\text{Rb}$ atoms is estimated to be around $1.3 \times 10^{10}$ cm$^{-3}$ under room temperature. The silica cell is placed in a three-layer permalloy box for shielding magnetic fields. The residue magnetic field inside the box is estimated to be below 1 mG. The desired ground states $|a\rangle$ and $|b\rangle$ are chosen respectively as the hyperfine states $|5S_{1/2}, F=1\rangle$ and $|5S_{1/2}, F=2\rangle$ of the $^{87}\text{Rb}$ atoms, and the excited state $|c\rangle$ is provided by the hyperfine states in the $|5P_{1/2}\rangle$ manifold.

The schematic set up for this experiment is shown by Fig. 1a. The write and the read pulses are from two different semiconductor lasers working at a wavelength about 795 nm, with the frequency difference of 6.8 GHz matching the hyperfine splitting of the $^{87}\text{Rb}$ atoms. These pumping laser beams are shined from different sides of the first polarization beam splitter PBS$_1$ so that they have orthogonal polarizations when going through the silica atomic vapor cell. After the cell, we need to separate the weak signal of Stokes or anti-Stokes photons from the strong write and read laser pulses, and this is done through both polarization and frequency selection. Right after the cell, the second polarized beam splitter PBS$_2$ will separate the signal from the pumping laser beams with an extinction ratio of about 7 $\times$ 10$^3$. Further frequency filtering is achieved by the glass cells F1 and F2, each containing Rubidium atoms initially optically pumped to the hyperfine levels $|5S_{1/2}, F=1\rangle$ and $|5S_{1/2}, F=2\rangle$, respectively. As in Ref. [18], the residual write (read) laser pulses after the PBS$_2$ will be strongly attenuated ($>10^6$) by the atomic cells F1 (F2) through resonant absorption while the signal Stokes (anti-Stokes) photons transmit with a high efficiency due to the large hyperfine detuning. After the filters, both of the Stokes and anti-Stokes photons are split by a 50% – 50% beam splitter, and then coupled into single-mode fibers, which direct them to the four single photon detectors (PerkinElmer Model SPCM-AQR). With this setup, we can measure both the auto-correlations and cross-correlations between the Stokes and the anti-Stokes photons.

The experiment goes as follows: we first optically pump the $^{87}\text{Rb}$ atoms in the silica cell to the ground state $|5S_{1/2}, F=1\rangle$. Then the write pulse, with about 5 $\times$ 10$^4$ photons, is sent through the atomic ensemble. The duration of the write pulse is about 1 $\mu$s. With this pulse, the probability to generate a Stokes photon in the spatial mode collected in this experiment is estimated to be about $p_w \approx 0.14$. The Stokes photons are detected by the single photon detectors $D_A$ and $D_B$. After a controllable time delay $\delta t$, which is typically 2 $\mu$s for our experiment, we send the read pulse through the atomic ensemble. This pulse has the same duration as the write pulse, but is stronger in intensity than the latter by a factor of 10. This read pulse transfers the collective atomic excitations back to anti-Stokes photons, with the retrieving probability of about 0.32 estimated from our experimental data (including the half polarization loss due to the unpolarized ensemble). The anti-Stokes photons are registered through the single-photon detectors $D_C$ and $D_D$. To reduce noise, before the detectors ($D_A$, $D_B$) and ($D_C$, $D_D$), we apply two gates with the time window of about 1 $\mu$s, synchronized respectively with the write and the read pulses. The time sequences of the write and the read pulses together with the gating windows are shown by Fig. 1c. The above steps form a full duty cycle (one trial), and this cycle is repeated at a rate of 5 kHz.

In experiments, the detectors for the Stokes and anti-Stokes photons typically have count rates of 220 s$^{-1}$ and 70 s$^{-1}$, respectively. The outputs of these four single photon detectors are sent to a time interval analyzer (TIA) for measuring the coincidence between any chosen pair of detectors. This coincidence can be measured by using the output from one detector as the start signal of the TIA and recording the arrival time of the output from the other detector. The coincidence between ($D_A$, $D_B$), ($D_C$, $D_D$), and ($D_A$, $D_C$) (or ($D_B$, $D_D$)) are denoted in the following by $n_{1,1}(t)$, $n_{2,2}(t)$, and $n_{1,2}(t)$, respectively.

The experimental results for these coincidences are shown by Fig. 2a (the left column). The first peak represents the coincidence within the same duty cycle (say $i$), and the following peaks are coincidences between the $i$th trial and the following trials. In Fig. 2b (the right column), by expanding the time axis, we show the detailed time shape $n_{\alpha,\beta}(\tau)$ of the first peak as well as the average time shape $m_{\alpha,\beta}(\tau)$ of the seven following peaks, where $\alpha, \beta = 1, 2$, and $\tau$ denotes the shifted arrival time whose width is smaller than 1 $\mu$s as set by the gating window. The average shape $m_{\alpha,\beta}(\tau)$ of the coincidence rates from different trials are defined as $m_{\alpha,\beta}(\tau) = (1/7) \sum_{j=1}^7 n_{\alpha,\beta}(\tau+j\Delta t)$, where $\Delta t = 200 \mu$s is the time interval between subsequent duty cycles.

From these measured time resolved coincidences, we can confirm the nonclassical (quantum) correlation be-
between the Stokes and the anti-Stokes fields. As the anti-Stokes field is transferred from the collective atomic mode, this also confirms the nonclassical correlation between the collective atomic mode and the forward propagating Stokes mode. To confirm the nonclassical feature of the correlation between the Stokes and anti-Stokes fields, as in Ref. [18] we make use of the Cauchy-Schwarz inequality. As has been pointed out by Clauser [20] and discussed in detail in ref. [18], the normalized auto and cross correlations functions $g_{1,1}$, $g_{2,2}$, and $g_{1,2}$ between two arbitrary classical fields 1 and 2 need to satisfy the following Cauchy-Schwarz inequality (here “classical” means that there exists P-representation for these fields with a positive distribution [23])

$$[g_{1,2}]^2 \leq g_{1,1}g_{2,2}. \quad (2)$$

The auto and cross correlation functions between the Stokes filed 1 and the anti-Stokes filed 2 can be directly obtained from the measured coincidence rates $n_{\alpha,\beta}(\tau)$ and $m_{\alpha,\beta}(\tau)$ ($\alpha, \beta = 1, 2$). Let $N_{\alpha,\beta}$ and $M_{\alpha,\beta}$ represent the total number of coincidences within the time gating window for the same trial and different trails, respectively, so they actually represent respectively the time integrals of $n_{\alpha,\beta}(\tau)$ and $m_{\alpha,\beta}(\tau)$ over the whole peak, i.e., they are determined by the areas of the corresponding peaks. By definition, the normalized auto and cross correlation functions $g_{1,1}$, $g_{2,2}$, and $g_{1,2}$ between the Stokes and anti-Stokes fields are given respectively by $g_{1,1} = N_{1,1}/M_{1,1}$, $g_{2,2} = N_{2,2}/M_{2,2}$, and $g_{1,2}(\delta t) = N_{1,2}/M_{1,2}$, where $\delta t = 2 \mu s$ denotes explicitly the time delay between the Stokes and anti-Stokes fields.

The integrated coincidence rates $N_{\alpha,\beta}$ and $M_{\alpha,\beta}$ in our experiment are shown by Fig. 2c, from which we calculate the correlations $g_{1,1} = 1.764 \pm 0.026$, $g_{2,2} = 1.771 \pm 0.028$, $g_{1,2}(\delta t) = 2.043 \pm 0.031$. One can see that $[g_{1,2}^2(\delta t) = 4.17 \pm 0.09] > [g_{1,1}g_{2,2} = 3.12 \pm 0.08]$, so the Cauchy-Schwarz inequality (2) is manifestly violated by our experiment. This clearly demonstrates that up to a delay time of 2 $\mu s$, we still get non-classically (quantum) correlated photon pairs from our experiment.

In the ideal case, if there are no noise and imperfections, the Cauchy-Schwarz inequality could be violated to a much larger extent. If the excitation probability of the collective atomic mode is $p$ for each write pumping pulse, we could get a violation of the inequality (2) with $[g_{1,2}^2]/(g_{1,1}g_{2,2}) \approx [(1 + p)/(2p)]^2$ in the ideal case. In our experiment, the excitation probability $p$ is estimated to be between 0.1 and 0.2, which in principle could allow a significantly larger violation. In practice, however, several sources of noise and imperfection degrade the extent of violation of the Cauchy-Schwarz inequality. Firstly, in a room-temperature atomic vapor, due to the atomic motion and the Doppler broadening, the atomic excitation can be diffused from the collective atomic mode to some other modes, and vice versa. Such a diffusion will significantly reduce the cross-correlation between the Stokes and anti-Stokes fields. Secondly, there are significant background fields due to imperfect filtering. The background fields are uncorrelated and they will also reduce the cross-correlation between the Stokes and anti-Stokes fields. Unlike the cold atom ensemble [18], the optical thickness of the atomic vapor cell is pretty large, so we estimate that uncorrelated spontaneous emission is not a dominant source of noise in our case [22]. The signal-to-noise ratio of this experiment probably could be improved by increasing the detuning of the pumping laser, by improving the filtering ratios, by reducing the power of the write beam, or by adjusting the configuration of the atomic cell to prolong the spin relaxation time.

In summary, we have observed generation of nonclassical photon pairs from a room-temperature atomic vapor cell. Each pair of correlated photons can be separated by a controllable time delay which is limit only by the coherence time in the atomic ground state manifold. Up to a delay time of 2 $\mu s$, we still clearly demonstrate non-classical correlation between the Stokes and anti-Stokes fields from the room-temperature atomic vapor. Compared with the cold atom experiment, the setup involved here are much cheaper. This experiment shows the prospect that such a cheap system could be used as a basic element for realization of quantum repeaters and long-distance quantum communication.

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[1] For a review, see D. P. DiVincenzo, Fortschr. Phys. 48, 771 (2000).
[2] A. Kuzmich, K. Molmer, and E. S. Polzik, Phys. Rev. Lett. 79, 4782 (1997).
[3] A. Kuzmich, N. P. Bigelow, and L. Mandel, Europhys. Lett. A 42, 481 (1998).
[4] J. Hald, J. L. Sørensen, C. Schori, and E. S. Polzik, Phys. Rev. Lett. 83, 1319 (1999).
[5] A. Kuzmich, L. Mandel, and N. P. Bigelow, Phys. Rev. Lett. 85, 1594 (2000).
[6] M. Fleischhauer, and M. D. Lukin, Phys. Rev. Lett. 84, 5904 (2000).
[7] C. Schori, B. Julsgaard, J. L. Sørensen, and E. S. Polzik, Phys. Rev. Lett. 89, 057903 (2002).
[8] C. Liu, Z. Dutton, C. H. Behroozi, L. V. Hau, Nature 409, 490 (2001).
[9] D. F. Phillips et al., Phys. Rev. Lett. 86, 783-786 (2001).
[10] A. S. Zibrov et al., Phys. Rev. Lett. 88, 103601 (2002).
[11] A. Kuzmich and E. S. Polzik, Phys. Rev. Lett. 85, 5639.
FIG. 1. The experimental configuration. (1a) The schematic setup of the experiment. The write and the read laser beams are sent from different sides of the PBS1, and go through the atomic cell C with orthogonal polarizations. The silica cell C has a length of 3 cm, containing isotopically pure 87Rb atomic vapor together with Ne buffer gas. The waist of the laser beams at the cell is about 4 mm. After the cell, the signals are separated from the strong pumping laser beams first through the polarization selection at the PBS2 and then through the frequency-selective absorption at the filter cells F1 and F2. The signals are split by a beam splitter, and coupled into single-mode fibers with an effective collection solid angle of about $2 \times 10^{-5}$. Then they are detected by four single-photon detectors with a detection efficiency of 64%. The transmission efficiency of the signal photons from the atomic cell to the detectors is estimated to be about 50%. The outputs of the detectors are sent to the time interval analyzer for measuring the coincidences of any pair of detectors, from which we can infer both auto-correlation and cross-correlation between the Stokes and anti-Stokes photons. (1b) The relevant level structure of 87Rb for this experiment. The desired Λ configuration is formed by the three hyperfine levels $|5S_{1/2}, F = 1\rangle$, $|5S_{1/2}, F = 2\rangle$ and $|5P_{3/2}, F = 1\rangle$. The frequencies of the write and the read laser beams are detuned from the frequencies of the corresponding atomic transitions both by a detuning of about 100 MHz. (1c) Time sequences for the optical pump, the write, and the read pulses, and for the synchronized gating windows. Gate1 and Gate 2 for the write and the read steps both have a time width of about 1 $\mu$s.

FIG. 2. The experimental data. (2a) (the left column) The time resolved coincidence rates $n_{\alpha,\beta}(t)$ with $(\alpha, \beta) = (1, 1)$, (2, 2) and (1, 2) over 8 successive trials of the experiment. (2b) (the right column) The shape of the coincidence peaks with the time axis expanded. The higher peak corresponds to the coincidence within the same i-th trial, and the lower peak represents the average of the following 7 coincidence peaks for different trials. (2c) The time integrated (total) coincidence events from different pairs of detectors.
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