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Storage and retrieval of nonclassical photon pairs and conditional single photons generated by the parametric down-conversion process

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Abstract. Storage and retrieval of parametric down-conversion (PDC) photons are demonstrated with electromagnetically induced transparency (EIT). Extreme frequency filtering is performed for the THz order of broadband PDC light and the frequency bandwidth of the light is reduced to the MHz order. Storage and retrieval procedures are carried out for frequency-filtered PDC photons. Since the filtered bandwidth (full width at half-maximum (FWHM) = 9 MHz) is within the EIT window (FWHM = 12.6 MHz), the flux of the PDC light is successfully stored and retrieved. The nonclassicality of the retrieved light is confirmed by using the photon counting method, where the classical inequality that is only satisfied for classical light fields is introduced. Since PDC photons can be utilized for producing a single-photon state conditionally, storage and retrieval procedures are also performed for conditional single photons. The anticorrelation parameter used for checking the property of the single-photon state shows a value of less than 1, which means that the retrieved light is in a nonclassical region.

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1. Introduction

Coherently transferring a quantum state of light to an atomic ensemble enables us to implement quantum memory for photons [1, 2], generate a nonclassical state of an atomic ensemble, and manipulate a quantum state of light [3–5]. A coherent transfer can be made by using a phenomenon called electromagnetically induced transparency (EIT) [6]. Recently, storage and retrieval of nonclassical light and generation of entanglement between two distant atomic ensembles have been demonstrated [7–9]. In these experiments, Raman scattered photons generated from atomic ensembles were used as nonclassical light [10]. Another method of generating nonclassical light, the parametric down-conversion (PDC) process, has been widely used in fundamental experiments in quantum optics [11] and for demonstrating various quantum information protocols [12, 13]. Light fields produced by the PDC process have been used as rich nonclassical photonic states such as the conditional single-photon state, antibunched state [14, 15], hyperentangled state [16], GHZ state [17], \( W \) state [18] and cluster state [19]; that is, the PDC photon pair is a fundamental resource for creating various nonclassical light fields. Storing the PDC photons in atomic ensemble, therefore, enables us to store and manipulate various nonclassical lights and can be utilized in the present photonic quantum information processing.

Storing nonclassical light generated from the PDC process, however, has a problem in that the bandwidth of the light is not matched by the atomic interaction width [20]. The spectral bandwidth of the PDC photons is typically of the order of THz, whereas the spectral widths for the atomic dipolar transition are of the order of MHz. This problem becomes crucial for photon counting, since a photon interacting with atoms cannot be selectively counted. Frequency filtering of PDC photons is thus required to solve this problem [21]. It should be noted that frequency filtering does not impose fundamental limitations on the current protocols in photonic quantum information, while it does have a disadvantage in that the filtering decreases the photon counting rate. The storage of PDC photons was demonstrated by using time filtering in our previous study [22]. However, the nonclassicality of the incident and the retrieved lights was not confirmed in the study. Furthermore, there was a large difference in frequency bandwidth between the incident and the retrieved light.

In this paper, we have demonstrated storage and retrieval of PDC photons, where we have avoided the problem of the bandwidth mismatch between PDC photons and EIT spectral width by using the frequency filtering technique. Preservation of the nonclassicality of the retrieved

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PDC photons was confirmed by using the photon counting method, where we introduced an inequality which is only satisfied for classical light fields. The PDC photons exhibit pair correlation, since a single pump photon is converted to two photons. This pair correlation is essential for preparing single photons conditionally. Therefore, we also checked preservation of the single-photon property through the storage process by using triggered PDC photons.

2. Method

In our experiment, we used single-mode PDC photons (degenerate parametric fluorescence) because of the experimental simplicity of the frequency filtering. There are several methods for estimating the nonclassicality of single-mode PDC photons [23]–[26]. Especially in [26], nonclassical photon statistics has been successfully observed by using photon counting. This method, however, cannot be applied to our experiment because of the low overall photon detection efficiency which is caused by frequency filtering, EIT and photon counters. We thus introduce a different type of inequality here. When the Glauber $P$ function [27] is non-negative, the following inequality is obtained:

$$\int \frac{(|\alpha|^4 t - |\alpha|^2)^2}{|\alpha|^2} P(\alpha) \, d^2 \alpha \geq 0,$$

where $t$ is any real number. The left part can be expanded in series on $t$. To satisfy the inequality for any $t$, the discriminant should be negative and the following inequality is obtained:

$$w \equiv \frac{\langle \hat{a}^\dagger \hat{\phi} \rangle \langle \hat{a}^\dagger \hat{\phi} \hat{a}^\dagger \hat{\phi} \rangle}{\langle \hat{a}^\dagger \hat{\phi} \hat{a}^\dagger \hat{\phi} \rangle^2} \geq 1,$$

where $\hat{a}$ and $\hat{a}^\dagger$ are the annihilation and creation operators of the single-mode light. The violation of this inequality (2) means that the non-negative $P$ function does not exist; that is, the light is nonclassical.

In our experiment, we also prepared the single-photon state by using the single-mode PDC photons. The single-photon property can be estimated from the anticorrelation parameter [28, 29],

$$\alpha = \frac{\langle \hat{a}_s^\dagger \hat{a}_s \rangle \langle \hat{a}_t^\dagger \hat{a}_t \rangle \langle \hat{a}_s^\dagger \hat{a}_s \hat{a}_t^\dagger \hat{a}_t \rangle}{\langle \hat{a}_s^\dagger \hat{a}_s \rangle^2 \langle \hat{a}_t^\dagger \hat{a}_t \rangle^2},$$

where $\hat{a}_s$,$(\hat{a}_t^\dagger)$ and $\hat{a}_t$,$(\hat{a}_s^\dagger)$ are the annihilation (creation) operators of the signal and trigger photons, respectively. The anticorrelation parameter $\alpha$ corresponds to the conditionally estimated intensity correlation function. In this sense, when $\alpha$ is less than 1, the resultant signal light is a nonclassical field. Especially, when $\alpha$ is close to 0, the resultant signal light can be regarded as the conditional single-photon state. When the single-mode parametric fluorescence with low excitation is split into two beams (trigger and signal beams) and one photon is detected at the trigger beam, the resultant signal light can also be regarded as the conditional single-photon state.

3. Experiment

3.1. Storage and retrieval of the frequency-filtered PDC photons

Figure 1 shows a schematic diagram of the experimental setup. The states \{\ket{a}, \ket{b}\} and \ket{c} of a $\Lambda$-type three-level system correspond to $5^2S_{1/2}$, $F = \{1, 2\}$ and $5^2P_{1/2}$, $F = 2$ of the $^{87}$Rb.

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Figure 1. Schematic diagram of the experimental setup. TiS: Ti:sapphire laser; AOM: acousto-optic modulator; LN: quasi-phase-matched MgO:LiNbO₃ waveguide; PBP: Pellin–Broca prism; DM: dichroic mirror; DP: dispersing prism; FC: filtering cavity; ET: etalon; HG: holographic grating; λ/₂, λ/₄: half and quarter wave plates; PBS: polarizing beam splitter; PMF: polarization maintaining single-mode fiber; SPCM: single photon counting module. The arrows indicate the polarization of light.

$D_1$ line, respectively. We generated PDC photons by using two independent type-0 quasi-phase-matched MgO:LiNbO₃ waveguides (LN1 and LN2). We employed ridge waveguides that were 5.0 µm wide, 3.0 µm thick and 8.5 mm long, which were fabricated so that quasi-phase matching was obtained at room temperature. Optical pulses (full width at half-maximum (FWHM) of the pulse temporal width $= 50$ ns) were generated by an acousto-optic modulator (AOM1) from the linearly polarized continuous output of a Ti:sapphire laser (TiS1) tuned to the $|a⟩ → |c⟩$ transition. The pulses were coupled to LN1 with 60% efficiency and generated second harmonic pulses, the polarizations of which were the same as the fundamental light. The second harmonic light was separated from the fundamental light by using a Pellin Broca prism (PBP) and a dichroic mirror (DM). Only the second harmonic light was injected into LN2 (coupling efficiency: 45%) and the PDC light was generated through the spontaneous PDC process, where the polarization of the PDC light was the same as the fundamental light. The second harmonic light was separated from the second harmonic light by using a dispersing prism (DP). We reduced the bandwidth of the PDC photons from 10 THz to 9 MHz by using a filtering cavity (FC) (FWHM of the bandwidth $= 9$ MHz, free spectral range (FSR) $= 1.5$ GHz), an etalon (ET) (FWHM of the bandwidth $= 300$ MHz, FSR $= 37$ GHz), and a combination of a holographic grating (HG).
and a polarization maintaining single-mode fiber (PMF) (FWHM of the bandwidth = 23 GHz).
The total transmittance of the frequency filtering at the center frequency of the PDC light
(resonant on the $|a\rangle \rightarrow |c\rangle$ transition) was about 20%.

In order to achieve the long-term stability of the frequency filtering, we utilized both the
locking and the alignment beams (not shown in figure 1). A part of the output beam from
TiS1 (locking beam) was phase modulated by an electro-optic modulator and its frequency was
adequately shifted by an acousto-optic modulator. The locking beam was incident on FC and the
resonant frequency of FC was actively stabilized to the $|a\rangle \rightarrow |c\rangle$ transition by using a piezo-
electric transducer attached to the cavity mirror, where the conventional FM-sideband locking
technique was utilized and the fluctuation of the resonant frequency was suppressed by less than
1 MHz. In order to prevent the locking beam from being mixed with the PDC light, the beams
were in opposite circulations of the cavity. A fraction of the laser beam diffracted by the AOM1
was tapped by a beam splitter and was used as the alignment beam. The alignment beam was
coupled to LN2 so that its spatial mode was made identical with the PDC light. The resonant
frequency of the ET was tuned to the $|a\rangle \rightarrow |c\rangle$ transition by monitoring the transmittance of
the alignment beam and adjusting the temperature of the ET. Once the proper resonance was
obtained, it was stable for one day and the alignment beam was thus cut off when the data were
acquired for the PDC light. When it was needed to take data for the coherent state of light, the
alignment beam was utilized instead of the PDC light.

Magneto-optically trapped $^{87}$Rb atoms were used as an optically thick medium, placed
in a vacuum cell magnetically shielded by a single permalloy. One cycle of the experiment
consisted of an atomic medium preparation period (1.5 ms) and a measurement period (1.0 ms).
After laser cooling, the magnetic field and the cooling ($-20$ MHz detuned from $^{5}\!\!\!S_{1/2}, F = 2 \rightarrow ^{5}\!\!\!P_{3/2}, F' = 3$) and repumping ($^{5}\!\!\!S_{1/2}, F = 1 \rightarrow ^{5}\!\!\!P_{3/2}, F' = 2$) lights were turned off,
and depumping lights ($^{5}\!\!\!S_{1/2}, F = 2 \rightarrow ^{5}\!\!\!P_{3/2}, F' = 2$) illuminated the atomic ensemble for
$100 \mu s$ so that all the atoms were prepared in state $|a\rangle$, where the optical depth of the $|a\rangle \rightarrow |c\rangle$
transition was $\sim 7$. In the measurement period, a probe light (beam diameter: 250 $\mu$m) and
a 250 $\mu$W control light (beam diameter: 500 $\mu$m) with the same circular polarizations were
injected into the unpolarized atomic ensemble. The probe and the control lights were made to
overlap at the cold atoms with a crossing angle of 3°. The control light was generated from the
TiS2 and its intensity was varied using an acousto-optic modulator (AOM2). The probe light
was detected by single-photon counting modules using silicon avalanche photodiodes (SPCM;
Perkin–Elmer model SPCM-AQR; detection efficiency: 62%).

Figure 2(a) shows intensity transmission spectra when a weak ($\sim$ pW) laser light (alignment
beam; coherent state of light) was used as the probe light. In the absence of the control light
(dotted line), the probe laser light was absorbed by the atomic medium. With the addition of
the control light (solid line), the atomic medium was rendered transparent around the resonant
frequency (peak transmission: about 100%, FWHM of the transparency window: 12.6 MHz).
Figure 2(b) shows typical experimental results of the storage and retrieval of frequency-filtered
PDC photons. The probe PDC pulses were injected into the cold atoms 1817 times during
the measurement period. Repeating the preparation and measurement periods, we accumulated
photon counts, shown as a function of time. Curve (A) corresponds to the probe PDC pulse in
the absence of cold atoms. The temporal shape of the probe PDC pulse was Gaussian with an
FWHM of 50 ns. Curve (B) corresponds to a probe PDC pulse with cold atoms and a constant
intensity of the control light. Here, the offset caused by the control light was subtracted. (The
unwanted photon flux was mixed with the retrieved light, which was the leakage of the control

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Figure 2. (a) Measured intensity transmission spectra of a weak coherent probe as a function of probe detuning to the $|a\rangle \rightarrow |c\rangle$ transition with (solid line) and without (dotted line) the control light. (b) Typical storage and retrieval of PDC photons: (A) incident, (B) slowed and (C) stored and retrieved pulses.

light and also the spontaneous emission from the background gas.) It should be noted that the spectrum of the frequency-filtered PDC photons is not Gaussian but close to Lorentzian, since the probe PDC pulse passed through the FC. A Lorentzian function has a long tail and therefore the probe PDC pulse included a frequency component that did not interact with atoms. This component passed through the cold atoms at the speed of light in vacuum. In contrast, the frequency component within the EIT window exhibited a slow propagation. The observed pulse delay of the component is about 35 ns, corresponding to more than three orders of magnitude reduction in group velocity. Due to the contribution of these two frequency components, the temporal shape of the probe PDC pulse was broadened. Curve (C) corresponds to the temporal variation of the probe PDC pulse with atoms and dynamically changing control light, where the control light was turned off at 140 ns and turned on at 390 ns. The falling/rising time was 30 ns. The total photon count for the retrieved light was 14% of that for the incident light, which clearly shows that the photon flux of the PDC photons was stored and retrieved.
3.2. Estimation of nonclassicality for the PDC photons

Firstly, we checked the nonclassical property of the retrieved PDC photons, where the rotating angle of the half-wave plate was adjusted so that all the probe light passed through the polarizing beam splitter (PBS). The transmitted probe was detected by SPCM-A, -B and -C (figure 1). The total transmissions for frequency-filtered PDC photons at SPCM-A, -B and -C were 36, 16 and 14%, respectively. Figure 3(a) shows the auto-intensity correlation of incident frequency-filtered PDC photons (curve (A) in figure 2(b)) obtained by a multi-channel scaler (MCS). The obtained normalized autocorrelation function was $g^{(2)}(0) = 7.9 \pm 0.1$, which indicates a superbunching effect, a significant property of PDC photons. The inset of figure 3(a) shows a magnification of the coincidence counts around 0 time delay. The FWHM of the correlation time is about 25 ns, which agrees with the value obtained from the bandwidth (9 MHz) of the FC [30]. The obtained single counts of SPCM-A was $N_A = 29,689,09$. The coincidence counts between SPCM-A
and -B and between SPCM-A and -C were $N_{AB} = 18,956$ and $N_{AC} = 15,274$, respectively. The triple counts between SPCM-A, -B and -C were $N_{ABC} = 75$. These were measured for 1700 s. The value of $w = N_A/N_{ABC}/N_{AB}N_{AC}$ was $0.77 \pm 0.09$. (The value of $w$ for the incident alignment beam was $1.07 \pm 0.06$.) The violation of the inequality (2) means that the incident PDC photons were in the nonclassical highly bunched photon pair state [22].

To estimate the retrieved component (curve (C) in figure 2(b)), the outputs of SPCM-A, -B and -C were gated from 400 ns to 450 ns. While the offset caused by the control beam was subtracted in figure 2(b), such a subtraction cannot be performed for the photon counts. The ratio of the retrieved component to the offset in gated time was 2.0. Figure 3(b) shows the auto-intensity correlation of the retrieved light. The obtained normalized autocorrelation function was $g^{(2)}(0) = 7.7 \pm 0.2$ and the superbunching effect was thus preserved [22]. The value of $w$ for the retrieved light was estimated to be $0.52 \pm 0.26$ from the obtained values of $N_A = 10,420,108$, $N_{AB} = 10,255$, $N_{AC} = 7782$ and $N_{ABC} = 4$, which were measured for 10 h. (The value of $w$ for the retrieved alignment beam was $1.11 \pm 0.13$.) The violation of the inequality (2) is direct evidence that the nonclassical highly bunched photon pair state was stored and retrieved (the probability that the retrieved light was in the classical region is 0.05). Despite the fact that the retrieved light included the offset component caused by the control light, the values of $g^{(2)}(0)$ and $w$ for the retrieved light were not degraded compared with those for the incident light. The long tail of the Lorentzian spectrum outside the EIT window was not stored, which would have improved the values of $g^{(2)}(0)$ and $w$. On the other hand, the retrieved light had an associated offset which should have degraded these values. We guess that these two effects balanced and hence the values of $g^{(2)}(0)$ and $w$ did not show significant difference between the incident and retrieved lights.

3.3. Estimation of the single-photon property for the PDC photons

Secondly, we estimated the property of conditional single photons prepared from the PDC photons. The detection scheme shown in the inset of figure 1 was used. A part of the frequency-filtered PDC light was reflected by the PBS, coupled to a single-mode fiber and detected by SPCM-t, where the coupling efficiency to the fiber was 67%. Total transmissions after the PBS were 23 and 22% at SPCM-1 and -2, respectively. The splitting ratio of the PBS was adjusted so that the counting rate of each SPCM became comparable.

Figure 4 shows the anticorrelation parameter $\alpha$ for the (a) incident and (b) retrieved states as a function of $g_{11}^{(2)}$ of the incident states, which is the cross intensity correlation function between SPCM-t and SPCM-1. The error bars indicate the standard deviations and the solid line shows a theoretical curve based on the degenerate parametric process. As the value of $g_{11}^{(2)}$ increased, both the anticorrelation parameters for incident and retrieved states decreased. This property is essential for the PDC light to be the conditional single-photon state. The triggered incident light showed a single-photon property with a minimum value of $\alpha = 0.19 \pm 0.06$. It is noted that the value of $\alpha$ was $1.00 \pm 0.03$ when the coherent state of the alignment beam was utilized as the incident light pulse. While the measurement error increased due to a decrease of the photon flux caused by an imperfect storage process, the triggered retrieved light showed the nonclassical feature associated with a single-photon state, where a minimum value of $\alpha = 0.52 \pm 0.30$ was obtained. The probability that the $\alpha \geq 1$ is 0.08 (the probability for the point at $g_{11}^{(2)} = 13.5$ is 0.05). While the anticorrelation parameter for the retrieved light does not show a sufficiently small value to be considered as the single photon, the conditional retrieved
state is in the nonclassical region. The value of $\alpha$ was also measured for the retrieved alignment beam pulse, which was $1.00 \pm 0.07$. The incident light included noncorrelated components due to the Lorentzian shape of the filtered spectrum. The retrieved light also included the offset component. Nevertheless, as is shown in figure 4, the anticorrelation parameters for both incident and retrieved light were in agreement with the simple theoretical curve without a fitting parameter. This is because the anticorrelation parameter was plotted as a function of $g_{t1}^{(2)}$, which is sensitive to the strength of the photon pair correlation in the light.

4. Conclusion

In conclusion, we have demonstrated storage and retrieval of PDC photons with EIT, where the bandwidth of the light was reduced from the THz to MHz order. Storage and retrieval of the flux of the PDC photons were clearly observed due to the frequency filtering. We introduced an inequality which is only satisfied for the classical light field and we verified the
nonclassicality of the retrieved light from the violation of this inequality. This result suggests that the atomic ensemble storing the photonic information was in a nonclassical state. We also prepared conditional single photons by using the PDC photons and performed the storage and retrieval of the single photons. While the anticorrelation parameter $\alpha$ for the retrieved light did not show a value close to 0, $\alpha$ decreased as $g^{(2)}_1$ increased and showed a value of less than 1 (nonclassicality). Since PDC photons have been widely utilized as a source of the nonclassical state of light, the results obtained here should have a wide range of applications which are not limited to quantum information processing. For example, manipulation of nonclassical light will be possible by controlling the state of atoms storing the photonic information. A variety of nonclassical atomic states could also be generated, which might be utilized for testing the bizarre nature of quantum physics or measuring fundamental physical constants with ultrahigh precision.

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