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Note	2光子状態は、2つの光子の波動関数が時空間的に完全に重なりあった状態で、単純に光子が2つある状態とは異なります。たとえば、その偏光状態は、単純に考えた場合の $2 \times 2 = 4$ ではなく、3つに限られます。今回の研究は、その2つの光子状態間のもつれ合いの生成を、単純な「2つのもつれ合い光子対」から区別して検証した世界初の成果です。今後の、多粒子状態間の量子相関の実験的な研究への先駆けになる物と期待されます。
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Distinguishing Genuine Entangled Two-Photon-Polarization States from Independently Generated Pairs of Entangled Photons

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A scheme to distinguish entangled two-photon-polarization (ETP) states from two independent entangled one-photon-polarization (EOP) states is proposed. Using this scheme, the experimental generation of ETP by parametric down-conversion is confirmed through the anticorrelations among three orthogonal two-photon-polarization states. The estimated fraction of ETP among the correlated photon pairs is 37% in the present experimental setup.

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Entanglement is one of the key features of quantum theory, and the generation of entangled one-photon-polarization (EOP) states by parametric down-conversion has been the focus of much experimental research [1–3]. Such entangled photon pairs have also been employed in several experiments on quantum information processing and quantum communication [4]. At present, there appear to be two ways to expand the concept of entangled photons: increase the number of local systems or increase the number of photons in the local system. In the former case of multiparty entanglement, single photons are distributed to multiple parties. This case has been realized through the generation of three-party entangled states such as Greenberger-Horne-Zeilinger states [5–7] and various four-party entangled states [8]. In the latter case, n photons conformed to the same spatiotemporal mode are distributed to each party. The first step in this direction is the generation of entangled two-photon-polarization (ETP) states.

The two-photon-polarization states can be expressed using a basis of three orthogonal states [9], exploiting the indistinguishability or bosonic nature of the two photons. Thus, two-photon-polarization states may be used as physical representations of three-level systems. Howell *et al.* [10] recently reported the violation of a Bell's inequality for spin-1 systems (three-level systems) proposed by Gisin and Peres [11] using ETP. The quantum mechanical prediction of the maximum value of the conclusion for the ETP is 2.55, whereas the classically predicted maximum is 2. Howell *et al.* experimentally confirmed the violation of this inequality, obtaining an experimental value of 2.27 ± 0.02 , attributing the result to ETP. However, the present authors have found that two independent EOPs, which may have been generated in the same experimental setup, can also violate the Bell's inequality used by Howell *et al.*, with a value of up to 2.41 for the correlations. This is possible because the correlations in Bell's inequalities are not a measure of indistinguishability, but rather of nonlocality. Thus, the violation of a Bell's inequality reported by Howell *et al.* in [10] does not require the generation of ETP, and an alternative

method for obtaining direct evidence of the generation of ETP is desirable.

In this Letter, we therefore propose a novel method to distinguish ETP from two independent EOPs. In the proposed method, the orthogonality of three two-photon-polarization basis states is checked via the correlations between the two local systems. The experimental generation of ETP using parametric down-conversion can then be evaluated using the proposed method. The results clearly reveal an anticorrelation between the corresponding basis states, indicating the successful generation of ETP.

EOP can be generated by pulsed type-II parametric down-conversion [2]. When one pair of photons is emitted into two modes A and B , the quantum state of the pair is ideally given by

$$|EOP\rangle = (|H\rangle_A|V\rangle_B - |V\rangle_A|H\rangle_B)/\sqrt{2}, \quad (1)$$

where H and V represent horizontal and vertical polarization. However, in general, multipair states are also generated in higher-order processes [12]. When two pairs are emitted simultaneously, the quantum state of the pairs is ideally given by

$$|ETP\rangle = \frac{1}{\sqrt{3}}(|HH\rangle_A|VV\rangle_B - |HV\rangle_A|HV\rangle_B + |VV\rangle_A|HH\rangle_B), \quad (2)$$

where $|HH\rangle$, $|HV\rangle$, and $|VV\rangle$ are the three orthogonal basis states of two-photon-polarization states, corresponding to two H -polarized photons, one photon H -polarized and V -polarized each, and two V -polarized photons, respectively. Note that these states differ from the simple direct products of two photons, $|H\rangle_1 \otimes |V\rangle_2$, where 1 and 2 denote spatially or temporally independent modes.

The proposed method for distinguishing ETP from two independent EOPs is as follows. The basis states of Eq. (2) can be transformed into three orthogonal unpolarized photon states, i.e.,

$$|ETP\rangle = \frac{-1}{\sqrt{3}}(|HV\rangle_A|HV\rangle_B + |PM\rangle_A|PM\rangle_B + |RL\rangle_A|RL\rangle_B), \quad (3)$$

where

$$|PM\rangle = \frac{1}{\sqrt{2}}(|HH\rangle - |VV\rangle), \quad (4)$$

$$|RL\rangle = \frac{i}{\sqrt{2}}(|HH\rangle + |VV\rangle). \quad (5)$$

Here, P , M , R , and L represent plus-diagonal ($|P\rangle = [|H\rangle + |V\rangle]/\sqrt{2}$), minus-diagonal ($|M\rangle = [|H\rangle - |V\rangle]/\sqrt{2}$), right-circular ($|R\rangle = [|H\rangle + i|V\rangle]/\sqrt{2}$), and left-circular ($|L\rangle = [|H\rangle - i|V\rangle]/\sqrt{2}$) polarizations. Thus, $|PM\rangle$ ($|RL\rangle$) represents a P - (R -) polarized photon and an M - (L -) polarized photon generated in the same mode. Note that these three states also form a set of orthogonal basis states [9].

Consider the case that one of the two photons of Eq. (3), say, that in path A , is detected in the H -polarized state, and the other photon also in path A is detected in the V -polarized state. In this situation, the two-photon-polarization states in mode A would be projected into the $|HV\rangle$ state, and the two-photon-polarization state in mode B would also be projected automatically into the $|HV\rangle$ state due to the entanglement between paths A and B . Consequently, the probability of detecting two photons in mode B in $|RL\rangle$ or $|PM\rangle$ states will be zero. Therefore, in actual experiments, a fourfold coincidence event in which the two photons in either mode are detected with different polarization [H and V in mode A , P (R) and M (L) in mode B] will never occur for pure state ETP. In the following, the number of coincidence events given the same measurement bases is defined as $C_{\parallel}$, and that for different measurement bases is defined as $C_{\perp}$. In the case of ideal pure state ETP, the ratio $r = C_{\perp}/C_{\parallel}$ is obviously 0.

Next, consider the case in which two independent EOPs described by Eq. (1) are emitted into spatially or temporally separable modes. The state of these two pairs is given by

$$|EOP \otimes EOP\rangle = \frac{1}{\sqrt{2}}(|H\rangle_{A1}|V\rangle_{B1} - |V\rangle_{A1}|H\rangle_{B1}) \otimes \frac{1}{\sqrt{2}}(|H\rangle_{A2}|V\rangle_{B2} - |V\rangle_{A2}|H\rangle_{B2}), \quad (6)$$

where modes $A1$ and $A2$ ($B1$ and $B2$) are in the same optical path A (B) but are spatially or temporally distinguishable. For example, when two photons of Eq. (6) in path A are measured in the H/V basis, $|V\rangle_{A1} \otimes |H\rangle_{A2}$, the state in path B will be $|H\rangle_{B1} \otimes |V\rangle_{B2}$ due to the polarization entanglement. Suppose the two photons in modes $B1$ and $B2$ are detected in the R/L basis, i.e., $|R\rangle_{B1} \otimes |L\rangle_{B2}$ or $|L\rangle_{B1} \otimes |R\rangle_{B2}$. Since the two photons are independent, the conditional probability for detecting two photons with

polarization $|R\rangle$ and $|L\rangle$ when the other two photons in path A are detected with polarization $|H\rangle$ and $|V\rangle$ is $|_{B1}\langle R|H\rangle_{B1B2}\langle L|V\rangle_{B2}|^2 + |_{B1}\langle L|H\rangle_{B1B2}\langle R|V\rangle_{B2}|^2 = 1/2$. Therefore, the ratio r for two independent EOPs should be $1/2$ even in the ideal case of pure state emission.

Since the correlations between the polarizations in A and B should be maximal for the ideal pure state, it is reasonable to assume that r will be greater than or equal to $1/2$ in the more general case of mixed state EOPs. Specifically, depolarization effects due to experimental imperfections in the alignment of the optical setup will typically increase the value of r . For realistic mixed state EOPs, we can therefore assume that $r \geq 1/2$ [13].

These results suggest that the ETP can be distinguished from two independent EOPs by measuring the correlation of the three orthogonal polarization basis states. According to the results given above, the following condition indicates the presence of ETP among emitted photons,

$$r < \frac{1}{2}. \quad (7)$$

The proposed method was used to evaluate the generation of ETP using the experimental setup shown in Fig. 1. The pump laser was a 100 fs-pulsed, frequency-doubled Ti:sapphire laser (82 MHz repetition rate, $\lambda = 390$ nm). The pump laser was focused onto a type-II beta-barium borate (BBO, 1.5 mm) crystal using a convex lens ($f = 45$ cm) in order to collect emitted photons efficiently [14]. The Kwiat '95 source condition [2] was adopted for the generation of EOP. A half-wave plate (HWP_0) and 0.75 mm BBO crystal were inserted in each path for compensation of spatiotemporal walk-off. In each of the paths, the generated photons pass through an interference

FIG. 1. Experimental setup for generation of entangled photon pairs and measurement of polarization correlation between paths A and B . Entangled pairs are generated in a beta-barium borate (BBO) crystal by pulsed parametric down-conversion. $|HV\rangle$, $|RL\rangle$, and $|PM\rangle$ states are measured using combinations of a polarizing beam splitter (PBS), a quarter-wave plate (QWP) at 45° , and a half-wave plate (HWP) at 22.5° in each path. The events are counted when the photon detectors D_{A+} , D_{A-} , D_{B+} , and D_{B-} are fired simultaneously.

filter IF (bandwidth 3.6 nm, centered at 780 nm) and a 5 mm iris set 1 m from the crystal.

The three basis states of two-photon polarization were measured in each path using a half-wave plate (HWP), a quarter-wave plate (QWP), a polarizing beam splitter (PBS), and a pair of photon detectors (SPCM AQ/AQR series, PerkinElmer). Continuous transfer of the measurement basis from one to the next among the three orthogonal basis states of Eq. (3) was achieved by rotating the wave plate continuously. In order to measure correlation between paths *A* and *B* in terms of polarization, fourfold coincidence events were counted using an electronic circuit and photon counter (SR400, Stanford Research Systems).

Figure 2 shows an example of the experimental data. Figure 2(a) shows the results for fourfold coincidence events while rotating the QWP in path *B*, with the polarization in path *A* fixed at the *H/V* basis. The fourfold coincidence counts were maximal at QWP angles of 0°, 90°, and 180°, corresponding to the *H/V* basis measurement in path *B*, whereas minimal counts were recorded at

FIG. 2. Fourfold coincidence counts for (a) fixed *H/V* polarization measurement basis in path *A* while rotating QWP in path *B*, (b) fixed *R/L* basis in path *A* while rotating HWP in path *B*, and (c) fixed *P/M* basis in path *A* while rotating HWP in path *B*. Note that in Fig. 2(b) a QWP at 45° was inserted in path *B*. The error bars represent the statistical errors estimated for a Poisson distribution of the measurement results. Pumping power was (a) 380 mW, (b) 410 mW, and (c) 420 mW. Solid lines represent the fitted theoretical prediction.

45° and 135° (*R/L* basis). Taking the averages of these values for $C_{\parallel}$ and $C_{\perp}$ over five experiments, the ratio r was obtained as 0.36 ± 0.06 . As this result satisfies the condition given in Eq. (7), we find that this correlation indicates the successful generation of ETP.

Similarly, Fig. 2(b) [Fig. 2(c)] shows the results of fourfold coincidence counts with the HWP rotated in path *B* and *R/L* (*P/M*) basis measurement setup in path *A*. In this case, the polarization basis was transformed from the *R/L* (*H/V*) basis to the *P/M* (*P/M*) basis, allowing different and similar polarization settings between paths *A* and *B* to be measured. Note that in order to transform from the *R/L* basis to the *P/M* basis, we set a QWP rotated by 45° before the HWP in path *B*. The obtained values of the ratio r are 0.38 ± 0.05 for *R/L* and 0.36 ± 0.02 for *P/M*, which also satisfy condition (7) and are consistent with the result in Fig. 2(a). Figures 2(a)–2(c) show a correlation between the same measurement outcomes ($|HV\rangle$, $|RL\rangle$, and $|PM\rangle$) and anticorrelation between different measurement outcomes ($|HV\rangle$ – $|RL\rangle$, $|RL\rangle$ – $|PM\rangle$, and $|PM\rangle$ – $|HV\rangle$). Based on these results, specifically the correlation/anticorrelation ratios r satisfying Eq. (7), it appears that the present system successfully generated ETP.

Perfect anticorrelation, which should have been obtained for ideal pure state ETP, was not achieved in these experiments. One of the reasons for this may have been the presence of EOP due to a lack of coherence between the two emitted pairs. In the present experimental setup, temporal and spatial coherence was implemented using irises and interference filters. This may not have been sufficient to obtain single mode.

The ratio of ETP among the generated states can be estimated by calculating the fourfold coincidence rates for a mixture of pure state ETP [Eq. (3)] and two independent pure state EOPs [Eq. (6)]. If we define α as the probability of the generation of pure state ETP given by Eq. (3) and β as the probability of generating two independent pure state EOPs given by Eq. (6), the fourfold coincidence rates are given by

$$C_0 \left[\frac{\alpha}{3} \cos^4(2\theta_{\lambda/4}) + \frac{\beta}{2} \left(\frac{\cos^4(2\theta_{\lambda/4}) + 1}{2} \right) \right], \quad (8)$$

$$C_0 \left[\frac{\alpha}{3} \cos^2(4\theta_{\lambda/2}) + \frac{\beta}{2} \left(\frac{\cos^2(4\theta_{\lambda/2}) + 1}{2} \right) \right], \quad (9)$$

$$C_0 \left[\frac{\alpha}{3} \sin^2(4\theta_{\lambda/2}) + \frac{\beta}{2} \left(\frac{\sin^2(4\theta_{\lambda/2}) + 1}{2} \right) \right], \quad (10)$$

where Eqs. (8)–(10) correspond to Figs. 2(a)–2(c), respectively. C_0 is the total rate of fourfold coincidence counts, and $\theta_{\lambda/4}$ and $\theta_{\lambda/2}$ are the angles of the QWP and HWP in path *B*, respectively [solid lines in Figs. 2(a)–2(c)]. Defining $r = C_{\perp}/C_{\parallel}$ as the ratio of the minima and maxima in Eqs. (8)–(10), we obtain the relation between α and r as follows:

$$\alpha = \frac{1 - 2r}{1 - 2r/3}. \quad (11)$$

Therefore, for the ratio of $r = 0.36$ obtained in our experiments, we obtain an ETP fraction of $\alpha = 0.37$.

The analysis given above is based on the assumption that the polarization states are the maximally coherent pure states given by Eqs. (3) and (6) and that the only source of coincidences between H/V and R/L is the multimode component given by the EOP state in Eq. (6). However, decoherence resulting in mixed state outputs may also contribute to such coincidences.

In the present setup, the visibility of the coincidence fringes observed between all possible combinations of detector pairs in paths A and B were about 0.9 each, at a coincidence count rate of 3000 s^{-1} . In principle, complete quantum tomography would be needed to identify the effects of this decoherence on the two-photon-polarization states. In order to obtain a rough estimate of the effects of decoherence, it may be useful to consider the coincidence counts caused by white noise in the mixed state EOP emission. In this case, the rate of coincidence counts is equal to $1/4$ of the total rate of two pair coincidences, regardless of the polarizations measured. A white noise fraction of γ added to the total density matrix therefore adds a constant background coincidence rate of $C_0\gamma/4$ to the polarization dependence given by Eqs. (8)–(10).

Since the visibility of 0.9 observed in our experiment suggests a noise background of about 0.1 for one pair, we estimate that the two pair noise may be about $\gamma = 0.2$. With this assumption, the single mode component α is raised to 0.46, while the coherent multimode component β drops to 0.34. Thus, the single mode contribution is increased because the value of $r > 0$ observed in the experiment is now partially attributed to the effects of decoherence, reducing the multimode contribution necessary to explain the experimentally observed correlations.

A more precise investigation of decoherence effects will be presented elsewhere. Here, it should be noted that only decoherence effects generally increase the ratio r , since decoherence reduces the polarization correlations that are responsible for the differences between $C_{\parallel}$ and $C_{\perp}$. The minimal fraction of ETP emission required to explain the observed value of $r < 1/2$ is therefore obtained by assuming the emission of pure state ETP and EOP, as given in Eqs. (8)–(11). Our method has thus successfully verified the presence of a significant single mode component in the emission [15].

In conclusion, a scheme to distinguish ETP from two independent EOPs based on fourfold coincidence has been proposed and demonstrated experimentally. The experimental results indicate the generation of two photons in the same spatiotemporal mode in each path,

strongly suggesting the formation of ETP. Specifically, we have been able to confirm a minimum of 37% single mode emission among the entangled four photon states generated in our experimental setup.

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