

Experimental observation of four-photon entanglement from down-conversion

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We observe polarization-entanglement between four photons produced from a single down-conversion source. The non-classical correlations between the measurement results violate a generalized Bell inequality for four qubits. The characteristic properties and its easy generation with high interferometric contrast make the observed four-photon state well-suited for implementing advanced quantum communication schemes such as multi-party quantum key distribution, secret sharing and telecloning.

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Entanglement between more than two particles is the key ingredient for advanced multi-party quantum communication. A number of proposals, e.g. telecloning [1], reduction of the communication complexity [2], or secret sharing [3] utilize multi-particle entanglement for quantum communication protocols.

Only few experiments have demonstrated entanglement between more than two qubits. Whereas the strong coupling between atoms enables engineered state preparation [4] as required for quantum computation, entangled multi-photon states are best suited for communication purposes. Interference of independently created photon pairs was used together with conditional detection for the first observations of three- and four-photon GHZ states [5]. Yet, those schemes required interferometric setups which limit their applicability and make detailed investigations difficult. Moreover, it is important to generate also different types of entangled multi-photon states required for quantum communication.

In this letter, we show that a polarization-entangled four-photon state can be directly observed behind a single pulsed spontaneous parametric down-conversion (SPDC) source. In contrast to previous techniques, the state forms without overlapping photons at beam splitters and the need of matching path length differences. The observed state can be used for telecloning and, as shown here, for multi-party key distribution and secret sharing.

In spontaneous parametric down-conversion, there is a reasonable probability of simultaneously producing four photons for strong pump power. However, if the type-II down-conversion [6] is adjusted to give polarization entanglement for a pair emitted into the two spatial modes a_0 and b_0 , the state of the four photons emitted into these modes is not simply the product of two entangled pairs [7]. Due to their bosonic nature, the emission of two otherwise indistinguishable photons with identical polarization into the same direc-

tion is twice as probable as the emission of two photons with orthogonal polarization [8]. Splitting each of the two modes at a non-polarizing beam splitter enables the observation of correlations due to the entanglement between four photons.

In our experiment we select events such that one photon is detected in each of the four outputs (a , a' , b , and b') of the beam splitters (Fig. 1). These four-photon coincidences can be explained with the four-photon state

$$\begin{aligned} |\Psi^{(4)}\rangle = & \sqrt{\frac{1}{3}} (|HHVV\rangle + |VVHH\rangle \\ & - \frac{1}{2} (|HVHV\rangle - |HVVH\rangle - |VHHV\rangle + |VH VH\rangle)), \end{aligned} \quad (1)$$

where the four entries in the state vectors indicate horizontal (H) or vertical (V) polarizations of the photons in arm a , a' , b and b' . Generally, with such a setup one obtains a superposition of a four-photon Greenberger-Horne-Zeilinger (GHZ) state and a product of two Einstein-Podolsky-Rosen (EPR) pairs [7],

$$|\Psi^{(4)}\rangle = \sqrt{\frac{2}{3}} |GHZ\rangle_{aa'bb'} - \sqrt{\frac{1}{3}} |EPR\rangle_{aa'} |EPR\rangle_{bb'} \quad (2)$$

For the particular state in eq. 1, the GHZ state is equal to $\frac{1}{\sqrt{2}}(|HHVV\rangle_{aa'bb'} + |VVHH\rangle_{aa'bb'})$ and the EPR state is the so-called Bell state $|\Psi^-\rangle = \frac{1}{\sqrt{2}}(|HV\rangle_{xx'} - |VH\rangle_{xx'})$ with $x = a, b$.

The generic form of state $|\Psi^{(4)}\rangle$ (eq. 2) is invariant under *identical* basis changes by the four observers, i.e., $|\Psi^{(4)}\rangle$ remains a superposition of a four-photon GHZ state and a product of two EPR pairs when all four observers use identically, but otherwise arbitrarily oriented polarization analyzers [10]. This contrasts with the GHZ states which lose their characteristic two-component form under such a basis change. This feature is related to the fact that the four-photon state of equation (1) can be viewed as the result of cloning an EPR pair. It is known that quantum cloning cannot be perfect. Thus, a four-photon GHZ contribution is created in addition to the product of two EPR pairs [12]. Nevertheless, the property of maintaining the characteristic form under basis transformation is carried over from the EPR singlet state to its quantum clone $|\Psi^{(4)}\rangle$.

To observe the four-photon entangled state, it is necessary to select single spatial modes and to erase the possible frequency correlations of the original photon pairs. This can be achieved by using *pulsed* parametric down conversion and by

detecting the photons behind narrowband filters, resulting in a coherence time longer than the pump pulse duration [13].

In our experiment we used the UV-pulses of a frequency-doubled mode-locked Ti:Sapphire laser (pulse length 180 fs) to pump SPDC in a 2 mm thick properly oriented BBO crystal at a center wavelength of 390 nm. The pump beam was focused to a waist of 100 μm inside the crystal, and the repetition rate was 76 MHz with an average power of 450 mW. The degenerate down-conversion emission into the two characteristic type-II crossing directions was coupled into single mode optical fibers to define the spatial emission modes. Behind the fibers the down-conversion light passed interference filters ($\Delta\lambda = 3$ nm), and was split at dielectric 50%-50% beam splitters into four distinct spatial modes. Polarization analysis in each of the four outputs was performed by a combination of quarter- and half-wave plates together with polarizing beam splitters. The four photons were detected by single photon Si-avalanche diodes, and registered with an eight-channel multi-coincidence unit. This unit recorded every possible coincidence between the eight detectors, and thus allowed efficient registration of the 16 relevant fourfold coincidences. The detectors exhibit different efficiencies between 40% and 50% due to production tolerances. If not stated otherwise, the rates presented here are therefore corrected for the separately calibrated efficiencies, and the errors given are deduced from propagated Poissonian counting statistics.

Fig. 2a shows the 16 possible fourfold coincidence probabilities for detecting one photon in each of the four outputs of the beam splitters, with all four polarization analyzers oriented along H/V . The rates of the $HHVV$ and the $VVHH$ events are in very good agreement with the state in equation (1) and, within errors, equal to the sum of all events where the two photons detected in arms a and a' , or in arms b and b' , have orthogonal polarization. The four-photon state $|\Psi^{(4)}\rangle$ exhibits the mentioned invariance under identical change of the four detection bases. Fig. 2b shows the four-photon coincidence probability when analyzed along $+45^\circ / -45^\circ$ linear polarization. Again, one observes two types of coincidences, the GHZ part, and the fourfold coincidences due to the EPR pairs with average rates lower by a factor of four. Integration times were 5 h, and 17.5 h, respectively, with four-fold coincidence rates between 300 and 100 per hour, varying mainly due to drifts of the fiber coupling.

As a first step in the characterization of entangled states it is customary to analyze the correlations between measurement results. For this purpose, polarization measurements corresponding to dichotomic observables with eigenvectors $|l_x, \phi_x\rangle = \sqrt{1/2}(|V\rangle_x + l_x e^{-i\phi_x} |H\rangle_x)$ and eigenvalues $l_x = \pm 1$ are performed by the observation stations in the four modes ($x = a, a', b, b'$). The theoretical prediction for the correlation function defined as the expectation value of the product of the four local results is given by [7]

$$E_{QM}(\phi_a, \phi_{a'}, \phi_b, \phi_{b'}) = \frac{2}{3} \cos(\phi_a + \phi_{a'} - \phi_b - \phi_{b'}) + \frac{1}{3} \cos(\phi_a - \phi_{a'}) \cos(\phi_b - \phi_{b'}). \quad (3)$$

The experimental value of the correlation function can be

obtained from the 16 four-photon coincidence rates, as shown in Fig. 2a and b, via

$$E(\phi_a, \phi_{a'}, \phi_b, \phi_{b'}) = \sum_{\substack{l_a, l_{a'}, \\ l_b, l_{b'} = \pm 1}} l_a l_{a'} l_b l_{b'} p_{l_a, l_{a'}, l_b, l_{b'}}(\phi_a, \phi_{a'}, \phi_b, \phi_{b'}). \quad (4)$$

Therein, the four-photon probabilities $p_{l_a, l_{a'}, l_b, l_{b'}}$ are given by

$$p_{l_a, l_{a'}, l_b, l_{b'}}(\phi_a, \phi_{a'}, \phi_b, \phi_{b'}) = c_{l_a, l_{a'}, l_b, l_{b'}} / \sum c,$$

where $c_{l_a, l_{a'}, l_b, l_{b'}}$ is the number of recorded four-fold events at the detectors specified by the indices (for the specific settings), and the sum is the total number of relevant four-fold events. Fig. 2c shows the dependence of E on the angle ϕ_a , for the other analyzers fixed at angles $\phi_{a'} = \phi_b = \phi_{b'} = 0$, corresponding to 45° linear polarization. For its visibility, which here is equal to the maximal absolute value of the correlation function (eq. 4), we obtain $V = 79.3\% \pm 1.4\%$, compared to $V = 100\%$ expected from (eq. 3) for a pure state. It serves as a measure for the quality of our state preparation, and largely depends on the ratio between the spectral bandwidths of the pump light and the detected photons [13].

Note that the analysis angles giving perfect correlations of $|\Psi^{(4)}\rangle$ are different from those for a four-photon GHZ state. Due to the EPR contributions, this state cannot be used in a GHZ-type argument refuting local hidden variable models of quantum mechanics. However, the invariance mentioned earlier enables perfect correlations for all possible sets of common analysis directions, a feature which does not hold for GHZ states, but which is of importance for the multiparty key distribution schemes presented below and other quantum communication schemes [1,11].

The contribution of the product of the EPR states also leads to a different nature of the four-photon entanglement. The seemingly innocent question of how much entanglement is in the state $|\Psi^{(4)}\rangle$ cannot be answered for the moment, because clear measures of multi-particle entanglement are still missing [14]. Keeping in mind possible applications for multiparty quantum cryptography and secret sharing, we analyze the entanglement of the state in terms of violation of a (non-conventional) Bell inequality.

One can write down a *single* Bell inequality which summarizes all possible local realistic constraints on the correlation function for the case of each local observer measuring the polarizations along *two* alternative directions [7,15,16]. Let us introduce a shorthand notation, $E(\phi_a^k, \phi_{a'}^l, \phi_b^m, \phi_{b'}^n)$, for the correlation functions deduced from the observed count rates for the full set of 16 local directions, with $k, l, m, n = 1, 2$ denoting which of the two alternative phase settings was chosen at the local observation station measuring in arm $x = a, a', b, b'$. The generalized Bell inequality reads [16]

$$S^{(4)} = \frac{1}{2^4} \sum_{\substack{s_x = \pm 1 \\ x=a, a', b, b'}} \left| \sum_{\substack{k, l, m, n \\ = 1, 2}} s_a^k s_{a'}^l s_b^m s_{b'}^n E(\phi_a^k, \phi_{a'}^l, \phi_b^m, \phi_{b'}^n) \right| \leq 1. \quad (5)$$

The maximal violation of this inequality for $|\Psi^{(4)}\rangle$ is obtained when three observers (one in each mode $x = a', b, b'$) perform polarization analysis along $\phi_x^{1,2} = \pm\pi/4$, and the observer in mode a chooses between $\phi_a^1 = 0$ or $\phi_a^2 = \pi/2$. Then the quantum prediction is as high as $S_{QM}^{(4)} = 1.886$, and results in a violation of the above inequality whenever the correlation function implied by the studied state $|\Psi^{(4)}\rangle$ has visibility greater 53% [7]. In comparison, for a four-photon GHZ state one obtains $S_{QM}^{(4)} = \sqrt{8}$ and a critical visibility of $1/\sqrt{8} \approx 35\%$. The visibility requirement for an experimental violation of the inequality (5) is therefore more demanding for $|\Psi^{(4)}\rangle$. But since it is much simpler to generate $|\Psi^{(4)}\rangle$, an experimental violation of the local realistic condition (5) becomes feasible.

Fig. 3 shows all 256 fourfold coincidence probabilities necessary for such an analysis. They were recorded in blocks of 16 coincidence rates, each corresponding to one of the 16 phase settings appearing in the inequality (5). Integration times were ranging from 2.75 to 4.75 hours per frame. For evaluating the generalized Bell-inequality we used the *raw* data without any correction for efficiencies. The resulting value $S^{(4)} = 1.301 \pm 0.040$ strongly violates the boundary for local realistic theories and confirms the entanglement of $|\Psi^{(4)}\rangle$.

Perfect correlations and the violation of a Bell-inequality are the key ingredients of entanglement based quantum cryptography [17], which with the aid of $|\Psi^{(4)}\rangle$ can be now expanded for multi-party quantum communication. Similar to a four-observer Bell experiment, parties A', B, and B' (observing photons in mode a', b and b') switch between analysis angles $\phi_x = \pm\pi/4$, while party A observing photons in mode a switches ϕ_a between 0 or $\pi/2$, and with a certain probability to $\pi/4$. After a number of quartets are registered, party A announces when having analyzed along 0 or $\pi/2$. For these detection quartets the other parties publicly announce their settings and results, which now can be used to evaluate $S^{(4)}$. The degree of violation of the Bell inequality (5) is a measure of the security of the key exchange. Any eavesdropper attack on any of the four quantum channels would reduce the entanglement and thus the violation of inequality (5). Thus, the four parties can assume that the remaining instances have been securely transmitted. Since in these cases party A measured along $\phi_a = \pi/4$, perfect correlations exist between the four measurement results according to equation (3) and enable the four parties to obtain a random, secure key [18].

Two ways to obtain a secure key can be formulated. Firstly, and similarly to recently proposed schemes of secret sharing [3], the parties could cooperate such that two of the four reveal their settings and results to the other two. Relying on the perfect correlations of the state, each of the two remaining parties can infer the result of the other and thus can evaluate a secure key only known to these two parties. Secondly, the measurement results can be used to *distribute* a key to three of the four parties. This can be achieved if, e.g., parties A and A' cooperate (forming now a single party A*) and compare their measurement results. The instances where they obtain the same results, are only due to the GHZ contribution

of $|\Psi^{(4)}\rangle$. For these cases the correlations of the GHZ state allow now the three parties A*, B, and B' to create a common secret key [19].

In summary, bosonic-type interference can be utilized to produce multi-photon entanglement directly from spontaneous parametric down-conversion. Without interferometric setups we could demonstrate the correlations between measurement results of four observers and the violation of a generalized four-photon Bell inequality. The high visibility of the quantum correlations and the ease of operation of our source show its potential for multi-party quantum communication applications like quantum secret sharing, three-party key distribution or for quantum telecloning.

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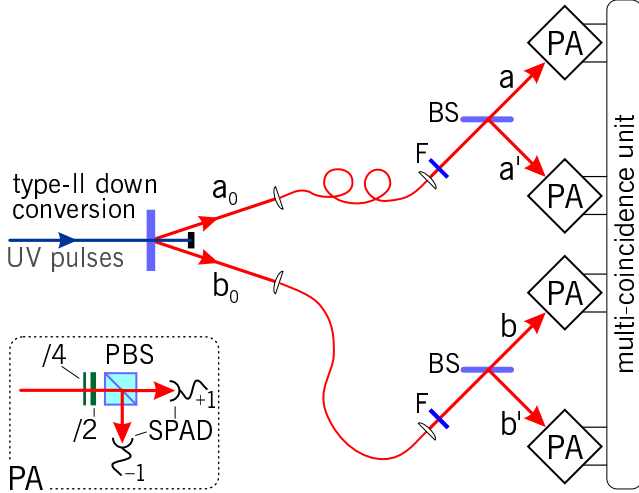


FIG. 1. Experimental setup. The four photons are emitted from the BBO crystal (type-II phase matching) into two spatial modes a_0 and b_0 , and distributed into the four modes a, a', b, b' by 50-50 beam splitters (BS) behind interference filters (F). To characterize the resulting four-photon state $|\Psi^{(4)}\rangle$, polarization analysis (PA) in various bases is performed for each mode using $\lambda/4$ and $\lambda/2$ wave plates in front of polarizing beam splitters (PBS) and single photon avalanche detectors (SPAD).

FIG. 2. Fourfold coincidence probabilities corresponding to a detection of one photon in each of the four polarization analyzers, oriented in (a) the H/V basis, and (b) the $\pm 45^\circ$ basis. (c) Four-photon polarization correlation with the detection basis of an observer in mode a varying from 45° linear at $\phi_a = 0$ to left circular, -45° linear and right circular polarization, while observers in a', b, b' analyze in the $\pm 45^\circ$ basis. The solid line shows a sinusoidal fit to the experimental results with a visibility of $79.3 \pm 1.4\%$.

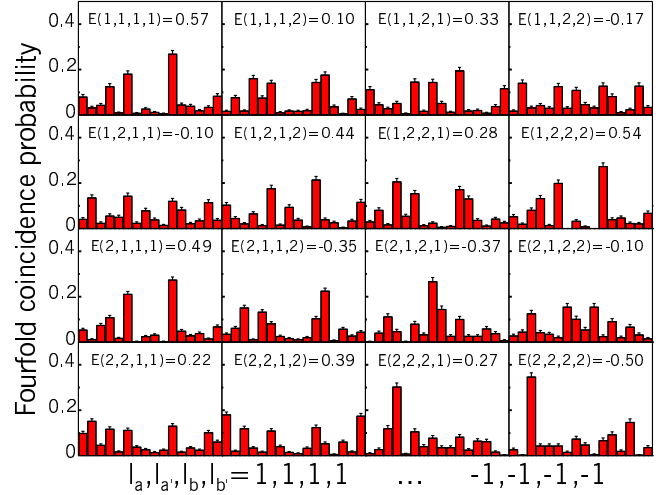


FIG. 3. Fourfold coincidence probabilities (raw data) for a four-particle test of local realistic theories. For the sixteen settings of the analyzer phases $\phi_a, \phi_{a'}, \phi_b, \phi_{b'}$, count rates *not* corrected for detection efficiencies are used to evaluate a generalized Bell inequality (5), leading to $S^{(4)} = 1.301 \pm 0.040$. This clearly exceeds the bound of 1 demanded by local realistic theories. Acquisition rates were around 150 fourfold coincidence events per hour.

