Experimental Violation of Local Realism by Four-Photon Greenberger-Horne-Zeilinger Entanglement

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We report the first experimental violation of local realism by four-photon Greenberger-Horne-Zeilinger (GHZ) entanglement. In the experiment, the nonstatistical GHZ conflicts between quantum mechanics and local realism are confirmed, within the experimental accuracy, by four specific measurements of polarization correlations between four photons. In addition, our experimental results also demonstrate a strong violation of Mermin-Ardehali-Belinskii-Klyshko inequality by 76 standard deviations. Such a violation can only be attributed to genuine four-photon entanglement.

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Multiparticle entanglement not only plays a crucial role in fundamental tests of quantum mechanics (QM) versus local realism (LR), but is also at the basis of nearly all quantum information protocols such as quantum communication and quantum computation [1]. Since the seminal work of Greenberger, Horne, and Zeilinger (GHZ) [2], the research on multiparticle entanglement has received much attention. In contrast with the case of two-particle entanglement where only statistical correlation predicted by QM is inconsistent with LR, in the case of maximally entangled states of more than two particles (i.e., the so-called GHZ states) a conflict with LR arises even for nonstatistical predictions of QM [2]. Further, QM can violate the multiparticle Bell-type inequalities imposed by LR by an amount that grows exponentially with the number of entangled particles [3–6], that is, going to higher entangled systems the conflict between QM and LR becomes ever stronger.

In recent years, entanglement of three photons has been demonstrated experimentally [7] and used to obtain the GHZ contradiction between QM and LR [8]. Meanwhile, entanglement of three atoms [9] or four ions [10] has also been demonstrated. Though significant experimental progress has been achieved in the demonstration of GHZ theorem [8], the three-photon experiments did not reveal correlations which are strong enough to confirm genuine multiparticle entanglement [11]. This is due to the fact that the data measured in the above three-photon entanglement experiments can be explained by a hybrid model in which only less than three particles is entangled [11]. Using the highly pure four-photon entanglement achieved in a recent experiment [12], it is, in principle, possible to exclude such a hybrid model by showing a sufficient violation of Bell-type inequalities. However, due to the very low coincidence rate, the data for such a violation were not collected. Therefore, genuine multiphoton entanglement still remains to be demonstrated experimentally.

In this Letter, we present a high intensity source of four-photon GHZ entanglement [13], and we report the first four-observer test of GHZ contradiction and provide sufficient experimental evidence to confirm the existence of genuine four-photon entanglement, hence closing the possibility of a hybrid model.

To demonstrate the four-photon GHZ contradiction, we first generate four-photon entanglement using the technique developed in a previous experiment [12]. As shown in Fig. 1, a pulse of ultraviolet (UV) light passes through a beta-barium borate (BBO) crystal twice to produce two polarization-entangled photon pairs, where both pairs are in the state $(1/\sqrt{2})(|H\rangle|V\rangle - |V\rangle|H\rangle)$, and H (V) denotes horizontal (vertical) linear polarization. One photon out of each pair is then steered to a polarization beam splitter (PBS) where the path lengths of each photon have been adjusted such that they arrive simultaneously. The PBS transmits H and reflects V polarization. After the two photons pass through the PBS, and exit it by a different output port each, and there is no way whatsoever to distinguish from which emission which of the photons originated, then correlations due to four-photon GHZ entanglement,

$$|\Psi\rangle = \frac{1}{\sqrt{2}}(|H\rangle_1|V\rangle_2|V\rangle_3|H\rangle_4 + |V\rangle_1|H\rangle_2|H\rangle_3|V\rangle_4), \quad (1)$$

can be observed [15]. With emphasis we note that, as shown in Refs. [12,16], this conditional feature of observing four-photon GHZ entanglement does not prevent us from performing an experimental demonstration of LR or practical applications in quantum information.

The observed fourfold coincident rate of the desired component $HVVH$ or $VHHV$ was about 1.3 per second, which is almost 2 orders of magnitude brighter than the previous experiment [12]. The ratio between any of the desired fourfold events $HVVH$ and $VHHV$ to any of the 14 other nondesired ones, e.g., $HHHV$, is better than

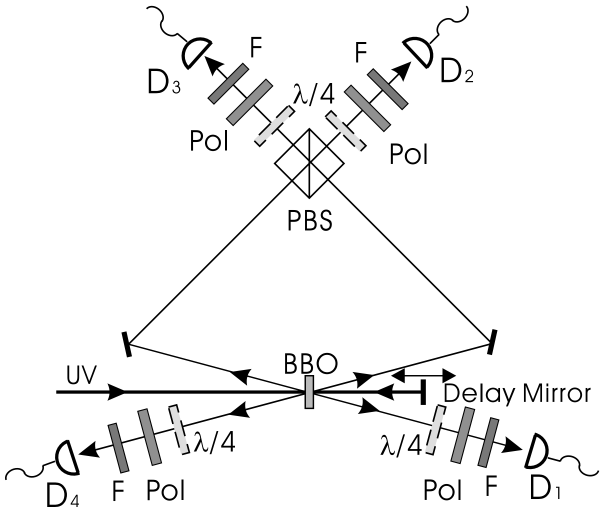

FIG. 1. Experimental setup for observing high intensity four-photon GHZ entanglement. Two pairs of entangled photons are produced by passing a UV laser pulse through a BBO crystal twice. The UV laser with a central wavelength of 394 nm has a pulse duration of 200 fs, a repetition rate of 76 MHz, and an average pump power of 450 mW. By optimizing the collection efficiency [14], we are able to observe about 2×10^4 entangled pairs per second for each pair behind 3.6 nm filters (F) of central wavelength 788 nm. Coincidences between detectors D_1, D_2, D_3, and D_4 exhibit four-photon GHZ entanglement. Polarizers (Pol) and quarter wave plates ($\lambda/4$) are used to perform the measurement of linear H'/V' or R/L polarization.

60:1. To confirm that the state (1) is indeed in a coherent superposition, we have performed polarization measurements on the four photons in the H'/V' basis, where $|H'\rangle = 1/\sqrt{2}(|H\rangle + |V\rangle)$ and $|V'\rangle = 1/\sqrt{2}(|H\rangle - |V\rangle)$. In Fig. 2, we compare the count rates of $H'H'H'H'$ and $H'H'H'V'$ components as we move the delay mirror. At zero delay, the latter component is suppressed with a visibility of 0.84 ± 0.03, hence confirming the coherent superposition of $HVVH$ and $VHHV$.

We now analyze the QM predictions for the state (1). Since the polarization states of a photon are a realization of a qubit, one can treat $|H\rangle$ and $|V\rangle$ as the two eigenvectors of Pauli operator σ_x of eigenvalues $+1$ and -1, respectively. Adopting the methods of Refs. [8,17], we consider measurements of linear polarization H'/V' or of circular polarization R/L, where $|R\rangle = (1/\sqrt{2})(|H\rangle + i|V\rangle)$ and $|L\rangle = (1/\sqrt{2})(|H\rangle - i|V\rangle)$ can be represented as the two eigenstates of Pauli operator σ_y with eigenvalues ± 1. We shall call a measurement of H'/V' as a σ_x measurement and one of R/L as a σ_y measurement.

To illustrate the GHZ conflict between QM and LR, we first consider three specific measurements of

$$\sigma_x\sigma_x\sigma_x\sigma_x, \quad \sigma_x\sigma_y\sigma_x\sigma_y, \quad \sigma_x\sigma_x\sigma_y\sigma_y, \tag{2}$$

where, for example, $\sigma_x\sigma_x\sigma_y\sigma_y$ denotes a joint measurement of linear polarization H'/V' on photons 1 and 2, and

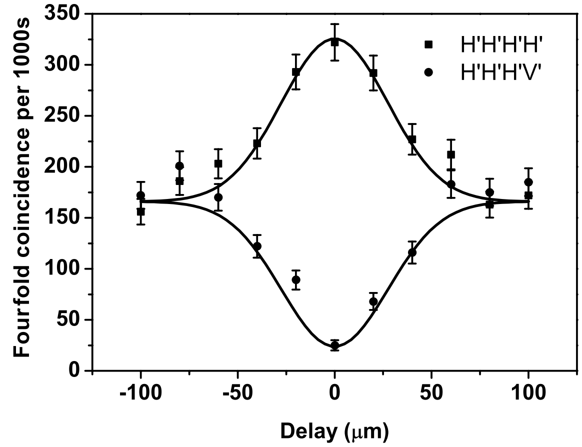

FIG. 2. Typical experimental results for polarization measurements on all four photons in the H'/V' basis. The coincidence rates of $H'H'H'H'$ and $H'H'H'V'$ components are shown as a function of the pump delay mirror position. The high visibility obtained at zero delay implies that four photons are indeed in a coherent superposition.

circular polarization R/L on photons 3 and 4. The three operators in Eq. (2) commute with each other and the state (1) is their common eigenstate with the eigenvalue $+1$. Thus, in any of the three measurements, the total number of photons that carry either V' or L polarization (i.e., with eigenvalue -1) must be even. For example, in a $\sigma_x\sigma_x\sigma_y\sigma_y$ measurement, only polarization combinations $H'H'RR$, $H'H'LL$, $H'V'RL$, $H'V'LR$, $V'H'RL$, $V'H'LR$, $V'V'RR$, and $V'V'LL$ arise.

What are the implications for LR? One can presume that, each photon carries Einstein-Podolsky_Rosen elements of reality [18] for both σ_x and σ_y measurements that determine the specific individual measurement result [19]. This is because in every one of the three measurements, any individual measurement result—both for L/R and for H'/V' bases—can be predicted with certainty for every photon given the corresponding measurement results of the other three [8,17].

For any photon i we call these elements of reality X_i with values $+1$ (-1) for $H'(V')$ polarizations and Y_i with values $+1$ (-1) for $R(L)$; we thus obtain the relations $X_1X_2X_3X_4 = X_1Y_2X_3Y_4 = X_1X_2Y_3Y_4 = +1$, in order to be able to reproduce the quantum predictions on all three measurements in Eq. (2). Furthermore, according to LR, any specific measurement for σ_x or σ_y must be independent of whether a σ_x or σ_y measurement is performed on the other photons. As $X_1Y_2Y_3X_4 = (X_1X_2X_3X_4) \times (X_1Y_2X_3Y_4)(X_1X_2Y_3Y_4)$, we obtain $X_1Y_2Y_3X_4 = +1$.

Therefore, the existence of the elements of reality implies that, performing a $\sigma_x\sigma_y\sigma_y\sigma_x$ measurement on the state (1), one should obtain the product of the eigenvalues with $+1$. Thus, from a LR point of view the only possible results for a $\sigma_x\sigma_y\sigma_y\sigma_x$ measurement are $H'RRH'$, $H'RLV'$, $H'LRV'$, $H'LLH'$, $V'RRV'$, $V'RLH'$, $V'LRH'$, and $V'LLV'$ [as shown in Fig. 3(e)].

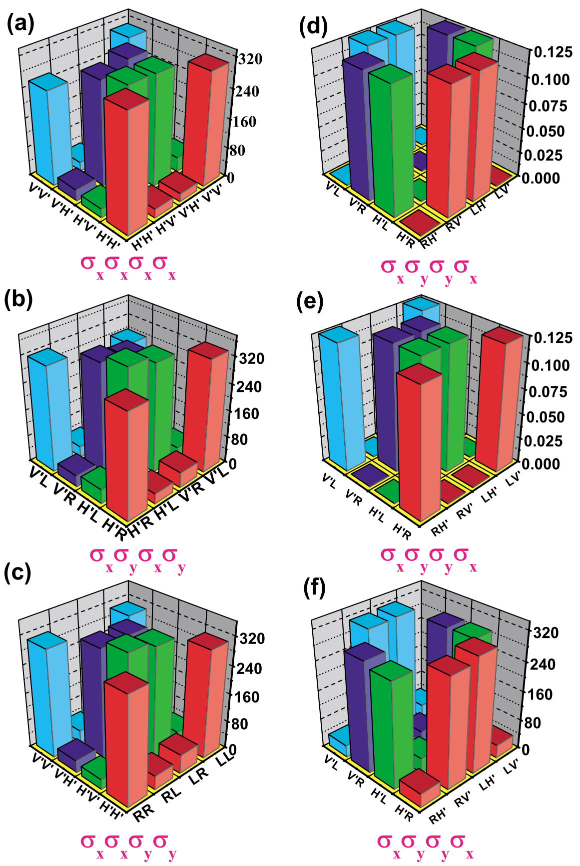

FIG. 3 (color online). Experimental results observed in the first three experiments (a)–(c), and predictions of QM and of LR (normalized), and observed results for the $\sigma_x\sigma_y\sigma_y\sigma_x$ measurement (d)–(f). The visibilities in (a)–(c) are 0.820 ± 0.011, 0.807 ± 0.011, and 0.781 ± 0.012, respectively. The experimental results in (f) are in agreement with the QM predictions (d) while in conflict with LR (e), with a visibility of 0.789 ± 0.012. The integration time of each fourfold coincidence is 1000 s.

However, according to QM, the state (1) is an eigenstate with eigenvalue -1 for operator $\sigma_x\sigma_y\sigma_y\sigma_x$. Thus, QM predicts that the only possible results for a $\sigma_x\sigma_y\sigma_y\sigma_x$ measurement are $H'RRV'$, $H'RLH'$, $H'LRH'$, $H'LLV'$, $V'RRH'$, $V'RLV'$, $V'LRV'$, and $V'LLH'$ [as shown in Fig. 3(d)]. Thus we conclude that the predictions by LR are completely opposite to the predictions by QM. It is the GHZ contradiction between LR and QM.

Experimentally, the observed results for the first three measurements are shown in Figs. 3(a)–3(c). Each measurement consists of 16 possible outcomes and ideally only eight of them should occur. However, since in reality no experiment can ever be perfect, even the outcomes which should not occur will occur with some small probabilities. Thus, if we are allowed to assume that the spurious events are attributable to the unavoidable experimental errors, then within the experimental accuracy we can conclude that the desired correlations in the three measurements confirm the quantum predictions for our GHZ entanglement.

In Figs. 3(d)–3(f), we compare the predictions of QM and LR with the results of the fourth $\sigma_x\sigma_y\sigma_y\sigma_x$ measurement. The results show that, within the experimental error, the fourfold coincidences predicted by QM occur, and not those predicted by LR. In this sense, we claim that we have experimentally realized the first four-particle test of local realism following the GHZ argument. For the purists, we may note that there is a derivation of the GHZ paradox for situations involving up to 25% (data flipping) error rate [20], that is for rates much higher than observed in the experiment ($\sim$11%).

The conflict between the quantum predictions for the GHZ states and LR can also be shown via violation of a suitable Bell inequality. In this case taking account of the errors is straightforward. According to the optimal Bell inequality for four-particle GHZ state [4], LR imposes the following constraint on correlations of polarization measurements on the four-photon system:

$$|\langle A\rangle| \leq 2, \tag{3}$$

where

$$A = \frac{1}{2}(\sigma_x\sigma_x\sigma_x - \sigma_x\sigma_y\sigma_y + \sigma_y\sigma_x\sigma_y + \sigma_y\sigma_y\sigma_x)(\sigma_a + \sigma_b) + \frac{1}{2}(\sigma_y\sigma_y\sigma_y + \sigma_x\sigma_y\sigma_x + \sigma_x\sigma_x\sigma_y - \sigma_y\sigma_x\sigma_x)(\sigma_a - \sigma_b) \tag{4}$$

and $\sigma_a = (1/\sqrt{2})(\sigma_x + \sigma_y)$, $\sigma_b = (1/\sqrt{2})(\sigma_x - \sigma_y)$, they correspond to measurements of two (orthogonal) pairs of elliptic polarizations. In Eq. (3), e.g., $\langle\sigma_x\sigma_x\sigma_x\sigma_a\rangle$ denotes the expectation value of a $\sigma_x\sigma_x\sigma_x\sigma_a$ measurement on the four photons. QM predicts a maximal violation of the inequality (3) by a factor of $2\sqrt{2}$. Hence, the threshold visibility to violate the constraint of Eq. (3) is given by $1/2\sqrt{2} \simeq 35.4\%$. This should be contrasted with the visibility consistent with the result of Ref. [20], concerning the GHZ contradiction, which is 50%. Interestingly, this implies that in contrast with three-particle entanglement, in the case of four-particle entanglement a different set of measurements than those for the GHZ contradiction is optimal in the inequality-based violation of LR.

To measure the expectation value of A, we need to perform 16 specific measurements as indicated in Eq. (4). A σ_a measurement on photon 4 is obtained if we insert in its path a quarter wave plate ($\lambda/4$), whose optical axis is set at 45° with respect to the horizontal direction. Then, the two eigenstates of operator σ_a are converted into linear polarizations which are polarized along the directions of

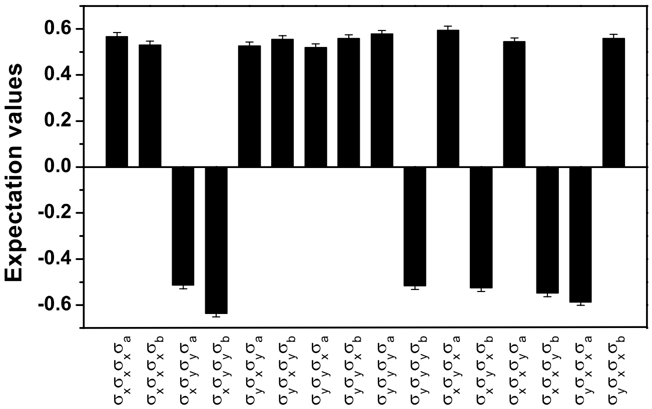

FIG. 4. Experimental results observed in 16 specific measurements to get the expectation value of A.

$-22.5°$ and $67.5°$. In the same way, the two eigenstates of operator σ_b can be converted into $-67.5°$ and $22.5°$ linear polarizations. The average visibility observed in the experiment for the state (1) is 78.4% and thus greatly exceeds the minimum of 35.4%. Substituting the experimental results (shown in Fig. 4) into the left-hand side of inequality (3) gives

$$|\langle A\rangle| = 4.433 \pm 0.032. \tag{5}$$

The violation of the inequality (3) is over by 76 standard deviations.

To demonstrate that the entanglement leading to the violation of Eq. (3) is a genuine four-photon entanglement (inexplicable by 3 or less photon entanglement), the sufficient condition is that $|\langle A\rangle| > 4$ [11] and it is satisfied by our data. Thus, our experiment provides unambiguous evidence for four-particle entanglement, which excludes any hybrid model.

Still one more question may be asked: with what fidelity the observed state reproduces the state $|\Psi\rangle$ of (1). Because of the specific form of $|\Psi\rangle$, the fidelity of a quantum state ρ with respect to $|\Psi\rangle$ is

$$\begin{aligned} F(\rho) &= \langle\Psi|\rho|\Psi\rangle \\ &= \frac{1}{2}(\langle HVVH|\rho|HVVH\rangle + \langle VHHV|\rho|VHHV\rangle) \\ &\quad + \mathrm{Re}\langle HVVH|\rho|VHHV\rangle. \end{aligned} \tag{6}$$

Using the identity $|\langle A\rangle| = 8\sqrt{2}\mathrm{Re}\langle HVVH|\rho|VHHV\rangle$ and the observed fractions of the desired components and the nondesired ones in the H/V basis, we obtain $F(\rho) = 0.840 \pm 0.007$, which is also well above the required threshold of 1/2 [11,21].

In conclusion, we have demonstrated the statistical and nonstatistical conflicts between QM and LR in four-photon GHZ entanglement. However, it is worth noting that, as for all existing photonic tests of LR, we also had to invoke the fair sampling hypothesis due to the very low detection efficiency in our experiment. Possible future experiments could include further study of GHZ correlations over large distances with spacelike separated randomly switched measurements [22]. Our work, besides its importance for foundations of QM, could also be applied to investigate the basic elements of quantum computation with linear optics [23] and implement multiphoton quantum secrete sharing [24].

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