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 in four-photon Greenberger-Horne-Zeilinger (GHZ) entanglement. In the experiment, the non-statistical 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 not only demonstrate a violation of Mermin-Ardehali-Belinskii-Klyshko inequality by 76 standard deviations, but also for the first time provide sufficient evidence to confirm the existence of genuine four-particle entanglement.

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Multi-particle 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, Zeilinger (GHZ) [2], the research on multi-particle entanglement has received much attention. In contrast to 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 multi-particle Bell-type inequalities imposed by LR by an amount that grows exponentially with the number of entangled particles [3, 4, 5, 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 realized experimentally [7] and used to demonstrate the extreme GHZ contradiction between QM and LR [8]. Meanwhile, entanglement of three atoms [9] or four ions [10] has also been demonstrated, yet in these two cases the quality of the entangled states still needs to be improved significantly in order to be useful for tests of LR and for quantum information processing. Though significant experimental progress has been achieved, all the above experiments suffer from a loophole in confirming genuine multi-particle entanglement [11]. This is due to the fact that the data measured in any of the above N-particle entanglement experiments can be explained by a hybrid model in which only less than N particles are entangled [11]. Using the highly pure four-photon entanglement achieved in a recent experiment [12], we are able to observe about $2 \times 10^4$ entangled pairs per second for each pair behind 3.6nm filters (F) of central wavelength 788nm. Coincidences between detectors $D_1$, $D_2$, $D_3$ and $D_4$ exhibit four-photon GHZ entanglement.

In this Letter, we develop a high intensity source of four-photon GHZ entanglement [13], by which we report the first four-observer test of GHZ contradiction, and for the first time provide sufficient experimental evidence to confirm the existence of genuine four-particle entanglement, hence closing the possible loophole of a hybrid model. To demonstrate the four-photon GHZ contradiction, we first generate four-photon entanglement using the
Let us analyze the QM predictions for the four-photon state (1). Since the polarization states of a photon are a realization of a qubit, one can represent $|H\rangle$ by column vector \[
\begin{pmatrix} 1 \\ 0 \end{pmatrix}\] and $|V\rangle$ by column vector \[
\begin{pmatrix} 0 \\ 1 \end{pmatrix}.
\]

That is, they can be treated as the two eigenvectors of Pauli operator $\sigma_x$ of eigenvalues $+1$ and $-1$ respectively. Adopting the methods of Refs. [8, 15], we consider measurements of linear polarization $H'/V'$, or of circular polarization $R/L$, where $|R\rangle = \frac{1}{\sqrt{2}}(|H\rangle + i|V\rangle)$ and $|L\rangle = \frac{1}{\sqrt{2}}(|H\rangle - i|V\rangle)$ can be represented as the two eigenstates of Pauli operator $\sigma_y$ with eigenvalues $\pm 1$. We shall call a measurement of $H'/V'$ linear polarization as a $\sigma_x$ measurement and one of $R/L$ circular polarization as a $\sigma_y$ measurement.

To illustrate the GHZ conflict between QM and LR, we first consider three specific measurements of polarization correlations between four photons:

\[
\sigma_x\sigma_x\sigma_x\sigma_x, \quad \sigma_x\sigma_y\sigma_x\sigma_y, \quad \sigma_x\sigma_y\sigma_y\sigma_y
\]

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 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 $H'HRR$, $H'HRLL$, $H'V'RL$, $H'V'LR$, $V'H'RL$, $V'H'LL$, $V'V'RR$, and $V'V'LL$ arise. Similar constraints can also be obtained for the other two measurements of (2).

We now analyze what are the implications for LR. Although any specific result obtained in any joint measurement on less than four photons is maximally random, one can still presume that, each photon carries Einstein-Podolsky-Rosen (EPR) elements of reality [17] for both $\sigma_x$ and $\sigma_y$ measurements that determine the specific individual measurement result [15]. This is because in every one of the three measurements, any individual measurement result - both for circular polarization and for linear polarization $H'/V'$ - can be predicted with certainty for every photon given the corresponding measurement results of the other three [8, 15].

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 $\sigma_x$ or $\sigma_y$ must be independent of whether a $\sigma_x$ or $\sigma_y$ measurement is performed on the other photons. As $X_1X_i = +1$ and $Y_iY_i = +1$, we can write $X_1Y_2Y_3X_4 = (X_1X_2X_3X_4)(X_1Y_2X_3Y_4)(X_1X_2Y_3Y_4)$ and obtain $X_1Y_2Y_3X_4 = +1$. 

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.
QM predicts that the only possible results for a state (1) is an eigenstate with eigenvalue -1 for operator $\sigma_x$. Thus, from a local realistic point of view one should obtain the product of the eigenvalues (a)-(c), and predictions of QM and of LR (normalized), $\langle \sigma_x \rangle$, $\langle \sigma_y \rangle$, and $\langle \sigma_z \rangle$. The observed results for the first three measurements are shown in Figs. 3a, 3b, 3c. 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. 3d, 3e, 3f, we compare the predictions of QM and LR with the results of the fourth $\sigma_x \sigma_y \sigma_z \sigma_x$ measurement. The results show that, within experimental error, the four-fold 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 [19], that is for rates much higher than observed in the experiment (≈ 11%).

The conflict between the quantum predictions for the GHZ states and local realism can also be shown via violation of a suitable Bell inequality. In this case taking account of the errors is straightforward. A number of inequalities for $N$-particle GHZ states have been derived [3, 4, 5, 6]. According to the optimal MABK inequality for GHZ states [4, 6], LR imposes a constraint on statistical correlations of polarization measurements on the four-photon system as the following:

$$|\langle A \rangle| \leq 2,$$  (3)

where

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

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

and $\sigma_a = \frac{1}{\sqrt{2}}(\sigma_x + \sigma_y)$, $\sigma_b = \frac{1}{\sqrt{2}}(\sigma_x - \sigma_y)$, and they correspond to measurements of two (orthogonal) pairs of elliptic polarizations. In Eq. 4, for example, $\langle \sigma_x \sigma_x \sigma_x \sigma_x \rangle$ denotes the expectation value of a $\sigma_x \sigma_x \sigma_x \sigma_x$ measurement on the four photons. QM predicts a maximal violation of the constraint by a factor of $2\sqrt{2}$. For the prefect quantum prediction the visibility of the correlations can be reduced to as little as 35.4%. Interestingly, a different set of measurements, than those for the GHZ contradiction, are optimal in the case of this inequality. Further, one could note that the inequalities derived in [6] require only a visibility of 32.9%. All this should be contrasted with the visibility consistent with the result of ref. [19], concerning the GHZ contradiction, which is 50%. Therefore in order to get maximal possible disagreement with LR, we performed another set of measurements.

To measure the expectation value of $A$, we need to perform sixteen specific measurements such as

![Fig. 3: Experimental results observed in the first three experiments (a)-(c), and predictions of QM and of LR (normalized), and observed results for the $\sigma_x \sigma_y \sigma_z \sigma_x$ measurement (d)-(f). The visibilities in (a)-(c) are $0.820 \pm 0.011$, $0.807 \pm 0.011$ and $0.781 \pm 0.012$, respectively. The experimental results in (f) are in agreement with the QM predictions (d) while in conflict with LR (e), with a visibility of $0.789 \pm 0.012$. The integration time of each four-fold coincidence is 1000s.](image)
A \sigma_{n} \text{ measurement on photon 4 is obtained if we insert in its path a quarter wave plate (QWP), whose optical axis is set at 45° with respect to the horizontal direction. Then, the two eigenstates of operator \sigma_{n} \text{ are converted into linear polarizations which are polarized along the directions of } -22.5° \text{ and } 67.5°. In the same way, the two eigenstates of operator \sigma_{b} \text{ can be converted into } -67.5° \text{ and } 22.5° \text{ linear polarizations.}

The average visibility observed in the experiment for the state (1) is 78.4% and thus greatly exceed the minimum of 35.4%. Substituting the experimental results into the left-hand side of inequality (3) gives

\[ |\langle A \rangle| = 4.433 \pm 0.032, \tag{5} \]

which violate the inequality (3) by over 76 standard deviations, hence demonstrating the conflict between QM and LR in four-photon GHZ entanglement.

Furthermore, the high visibilities also confirm the existence of genuine four-photon entanglement in our experiment. To demonstrate a full four-photon entanglement, two sufficient conditions, i.e. the inequality \[ |\langle A \rangle| > 4 \]

and the so-called state preparation fidelity \( F(\rho) > 1/2 \), must be satisfied [11, 20]. Here the state preparation fidelity is defined as

\[ F(\rho) = \langle \Psi | \rho | \Psi \rangle = \frac{1}{2} \left( \langle HVVH | \rho | HVVH \rangle + \langle VHVV | \rho | VHVV \rangle \right) + \text{Re} \langle HVVH | \rho | VHVV \rangle \tag{6} \]

and for any state \( \rho \), there is a simple identity:

\[ |\langle A \rangle| = 8\sqrt{2} \text{Re} \langle HVVH | \rho | VHVV \rangle. \tag{7} \]

Not only does the experimental result in Eq. (5) significantly violate the inequality \[ |\langle A \rangle| > 4 \], together with the observed fractions of the desired components and the undesired ones in the \( H/V \) basis it also gives \( F(\rho) = 0.840 \pm 0.007 \), which is well above the threshold of 1/2. Thus, our experiment for the first time provides unambiguous evidence for a full test of four-particle entanglement, which excludes any hybrid hidden-variable model to explain our experimental data.

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 space-like separated randomly switched measurements [21]. Our work, besides its significance in quantum foundations, could also be applied to investigate the basic elements of quantum computation with linear optics [22] and implement multi-photon quantum secret sharing [23].

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