Observation of stronger than binary correlations with entangled photonic qutrits

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Can the occurrence of a particular outcome of a quantum measurement with three possible outcomes always be explained as a two-step process in which, in the first step, some classical mechanism precludes one of the outcomes and, in the second step, a binary measurement generates the outcome? [1]. This way of viewing a measurement reduces the non-classical phenomena to a single binary measurement. A related question is: Are there correlations stronger than those produced when the correlation between the binary measurements are nonsignaling but arbitrarily strong? This includes Popescu and Rohrlich’s maximally nonlocal correlations [2]. Here we provide the first experimental confirmation of the existence of stronger-than-binary correlations, which implies a negative answer to the first question and a positive answer to the second. The experiment uses a high-visibility source of pairs of photons each of them encoding a qutrit. The two photons are distributed to two laboratories, where randomly chosen three-outcome measurements are performed. The entangled state and the measurements are chosen aiming to the maximum quantum violation of the unique optimal inequality for nonsignaling binary correlations [3]. A violation of this inequality with a significance corresponding to 9.3 standard deviations is observed. This provides a compelling evidence against two-step explanations of the measurement process and falsifies nonsignaling binary theories as possible descriptions of nature.

Quantum mechanics is so successful that it is difficult to imagine how to go beyond the present theory without contradicting existing results. However, going beyond our present understanding is needed to, e.g., extending quantum theory into gravity. The critical point in quantum mechanics is the measurement process [4]. One way to go beyond our present description of a measurement is by reconstructing quantum theory [5–9] and identify axioms related to measurements which could be modified in a way not contradicting existing experimental evidence, but making different predictions. This is the approach that we follow in this work. A related approach is investigating stochastic nonlinear modification of the Schrödinger equation [10].

Here we consider a class of general probabilistic theories motivated by the following observation. In quantum theory, two-outcome measurements \((E, \mathbb{I} \rightarrow E)\) are feasible whenever \(E\) is a self-adjoint operator satisfying \(0 \leq E \leq \mathbb{I}\). On the other hand, in any general probabilistic theory, if \((\mathcal{E}_1, \mathcal{E}_2, \ldots, \mathcal{E}_n)\) represents an \(n\)-outcome feasible measurement, then any postprocessing to a binary measurement \((\mathcal{E}_1', \mathcal{E}_2')\) is also a feasible measurement. However, in quantum theory, \((E_1, E_2, \ldots, E_n)\) is an \(n\)-outcome feasible measurement whenever all postprocessings \((E', \mathbb{I} \rightarrow E')\) are feasible. This suggests a natural alternative, namely, assuming that feasible \(n\)-outcome measurements are only those which can be implemented by selecting from binary measurements.

A binary measurement [1] is one that can be implemented as

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a two-step process in which, in the first step, some classical mechanism excludes all but two of the outcomes and, in a second step, the final output is produced by a binary measurement. The concept is illustrated in Fig. 1.

Correlations between the outcomes of measurements performed by two parties, called Alice and Bob, are described by joint probabilities $P(a, b|x, y)$, where $x$ and $y$ are Alice’s and Bob’s measurement settings, respectively, and $a$ and $b$ are Alice’s and Bob’s measurement outcomes, respectively. Binary nonsignaling correlations are those which are both nonsignaling, i.e., $\sum_a P(a, b|x, y) = P(a|x)$ and $\sum_a P(a, b|x, y) = P(b|y)$, and binary, i.e., $P(a|x) = 0$ except for two outcomes $a$ and $P(b|y) = 0$ except for two outcomes $b$, and the convex hull thereof [1]. For example, Popescu–Rohrlich boxes [2] maximally violating the CHSH-Bell inequality provide an example of binary nonsignaling correlations which are forbidden in quantum theory. Interestingly, according to quantum theory, there exist stronger than binary nonsignaling correlations [1]. A major problem however has been identifying how they can be actually observed.

The experiment presented here aims at the maximum violation predicted by quantum theory of the optimal and unique inequality satisfied by binary nonsignaling correlations. The experiment is a bipartite Bell-like inequality experiment in which Alice randomly chooses between two different measurements, $x = 0, 1$, each of them with three possible outcomes, $a = 0, 1, 2$, and Bob randomly chooses between two different measurements, $y = 0, 1$, each of them with three possible outcomes, $b = 0, 1, 2$. Any binary nonsignaling correlations satisfy the inequality

$$I_a \leq 1,$$  \hspace{1cm} (1)

where

$$I_a = \sum_{k,x,y=0,1} (-1)^{k+x+y} P(k, k|x, y),$$ \hspace{1cm} (2)

and the outcomes $a = 2$ and $b = 2$ do not occur explicitly (see Methods). In contrast, according to quantum theory, inequality (1) can be violated up to

$$I_a = 2(2/3)^{3/2} \approx 1.089.$$

This maximum quantum value can be achieved by preparing two qutrits in a particular state and making some particular three-outcome local measurements (see Methods).

In the experiment (see Methods) we have obtained

$$I_a = 1.066 \pm 0.007$$ \hspace{1cm} (4)

which implies a violation of inequality (1) with a statistical significance corresponding to 9.3 standard deviations (see Methods). A careful analysis of the data (see Methods) shows that residual systematic errors are not enough to explain with nonsignaling binary theories the observed violation of inequality (1).

Consequently, the experiment falsifies binary general probabilistic theories as possible descriptions of nature by showing that there are correlations which are not binary nonsignaling. It also shows that in nature there are genuinely ternary measurements, thus demonstrating that the measurement process in quantum theory cannot be explained as a two-step process as in Fig. 1 a). In fact, the result of the experiment demonstrates [3] that none of the four measurements (Alice’s or Bob’s) can be binary.

The experiment also illustrates how the maturity and refinement achieved by the experimental techniques developed for quantum communication and information processing can be used to test very subtle predictions of quantum theory and obtain new insights about how nature works.

**METHODS**

**Bound on binary nonsignaling correlations.** Here we prove that the bound $I_a \leq 1$ in Eq. (1) is valid for all binary nonsignaling theories. First note that due to standard analysis the bound is $I_a \leq 1$ in the case of Bell local correlations, which is a subset of binary nonsignaling ones. Let us also note that in the derivation we can focus on extremal correlations. Hence it suffices to consider extremal binary nonlocal resources in the following.

Let us rewrite $I_a \leq 1$ in an equivalent form. To this end, we first subtract the quantity $\sum_{a,b=0,1,2} P(a, b|1, 1) = 1$ from both sides of Eq. (1). After relabeling certain measurement outcomes, we obtain the inequality

$$I_a' = P(00|00) - P(22|00) - \sum_{a+b\leq 2} P(a, b|1, 1)$$

$$- \sum_{a+b<2} P(a, b|1, 0) - \sum_{a+b<2} P(a, b|0, 1) \leq 0,$$

which is equivalent to $I_a \leq 1$. Therefore, if we can show that $I_a' \leq 0$ cannot be violated with extremal binary nonsignaling resources, it entails that $I_a \leq 1$ holds true as well using these resources. These binary correlations have the following properties. There exist certain $a_x \in \{0, 1, 2\}$ for $x = 0, 1$ and $b_y \in \{0, 1, 2\}$ for $y = 0, 1$ such that the respective marginal distributions $P(a_x|x) = 0$ and $P(b_y|y) = 0$. This implies $P(a_x, b_y|x, y) = 0$ for all $a_x, b_y, x, y$ and $P(a_x, b_y|x, y) = 0$ for all $a_x, b_y, x, y$. Due to the condition for extremal nonsignaling correlations, the rest of the entries of the distribution $P(a, b|x, y)$ form a Popescu–Rohrlich box [2], which can take up the entries 0 and 1/2. Notice that in order to violate $I_a'$ in Eq. (5) with such a distribution $P(a, b|x, y)$, one has to demand (i) that $P(00|00) = 1/2$ (the one appearing with positive coefficient) and (ii) all other probabilities (i.e., the ones entering the inequality with negative coefficients) are zero. Constraints (i) and (ii) along with the nonsignaling constraints of the distribution $P(a, b|x, y)$ imply the following chain of equalities:

**TABLE I. Angles of the half wave plates used in the measurement setups.**

<table>
<thead>
<tr>
<th>Measurement</th>
<th>HWP4</th>
<th>HWP5</th>
<th>HWP6</th>
<th>HWP7</th>
<th>HWP8</th>
</tr>
</thead>
<tbody>
<tr>
<td>Setting 0</td>
<td>$-22.5^\circ$</td>
<td>$0^\circ$</td>
<td>$45^\circ$</td>
<td>$17.63^\circ$</td>
<td>$37.50^\circ$</td>
</tr>
<tr>
<td>Setting 1</td>
<td>$22.5^\circ$</td>
<td>$0^\circ$</td>
<td>$45^\circ$</td>
<td>$17.63^\circ$</td>
<td>$-37.50^\circ$</td>
</tr>
</tbody>
</table>
The source of pairs of photons and the first party, Alice, are in lab 1, while the second party, Bob, is in lab 2. The distance between Alice and Bob is approximately 8 m. The source is a continuous wave laser of 404 nm wavelength and 100 mW power. To generate qutrit-qutrit entanglement, the pump laser is separated into two paths by three Half wave plates (HWPs) and one beam displacer (BD), where HWP1 is set at 15°, HWP2 is set at 27.37°, and HWP3 is set at 0°. Then the pump laser is directed into two 0.5 mm thick type-I cut barium borate (BBO) crystals. After the BD and three HWPs, the pump state is

$$|\psi\rangle = \frac{1}{2} \sqrt{2} (|00\rangle + |11\rangle - |22\rangle).$$

Then, each photon is distributed to a different laboratory. The laboratories are separated 8 m. In each laboratory, a local measurement is randomly decided between two possibilities by means of a random number generator. The whole process is repeated 4500 times. The data collection time for each run is 0.5 s.

**Evaluation of the data.** In total, 1060 complete data sets have been measured with 67.1 coincidences on average. We evaluate three conditions on the data: (i) Normalization, i.e., whether $$\sum_i N_s(i, j|x, y)$$ is independent of $$x$$ and $$y$$. (ii) Nonsignaling, i.e., whether $$\sum_j N_s(i, j|x, y)$$ is independent of $$x$$ and $$\sum_i N_s(i, j|x, y)$$ is independent of $$y$$. (iii) The inequality $$\sum_{k, x, y, k \neq 2} (-1)^{k+x+y} N_s(k, k|x, y) - 1\frac{1}{4} \sum_{i, j, x, y} N_s(i, j|x, y) \leq 0$$. Hereby $$N_s(i, j|x, y)$$ is the number of coincidences for each of the runs $$s = 1, \ldots, 1060$$. We compute the mean $$m$$ and the variance $$\nu$$ over the 1060 runs for each condition, so that $$t = m \sqrt{1060/\nu}$$ is distributed according to the Student-$$t$$ distribution with parameter $$\nu = 1059$$ degrees of freedom. Since in this regime the Student-$$t$$ distribution is very close to a normal distribution, we obtain the
Table II. Joint $p$-values for (i) normalization conditions, (ii) nonsignaling conditions, and (iii) inequality (1). “Coin tosses” is $k$ if the condition to hold is as plausible as obtaining $k$ times heads in a row when throwing a fair coin. “Standard deviations” is $\sigma$ if the condition to hold is as plausible as obtaining a value $|x| > \sigma$ from a normal distributed random variable. 1) Full data set using all 1060 repetitions. 2) Reduced data set using every 5th repetition. For the median of the $\chi^2$-distribution and the $p$-value the significance of more than 2 is unreasonably high. Therefore, the $p$-value given in the table is increased by a factor of $\sqrt{5} \approx 2.2$, a 4 $\sigma$ violation of inequality (1) remains while the violation of the nonsignaling conditions vanishes.

Finally, we compute $P_s(i,j|x,y) = N_s(i,j|x,y)/\sum_k \sum_\ell N_s(k,\ell|x,y)$ for each $s$. This allows us to compute for each repetitions the value of $I_a$. In equation (4) we report the mean value and standard deviation of these 1060 values.

Data repository. The complete data set is publicly available from http://personal.us.es/adan/binary.htm. We encourage readers who want to expand our work with further data analysis to do so.

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Author contribution. The experiment was performed by X-M.H., B-H.L., Y.G., G-Y.X., Y-F.H., C-F.L., and G-C.G. The theory was developed by M.K., T.V., and A.C. Data analysis was performed by M.K., with input from all authors. The paper was written by X-M.H., B-H.L., C-F.L., A.C., M.K., and T.V. with input from all authors. Experiment conceived by M.K., T.V., A.C., X-M.H., B-H.L., and C-F.L. The authors declare that they have no competing financial interests.

<table>
<thead>
<tr>
<th>Condition</th>
<th>Coin tosses</th>
<th>Standard deviations</th>
</tr>
</thead>
<tbody>
<tr>
<td>1) Full data set: $I_a = 1.066 \pm 0.006$</td>
<td></td>
<td></td>
</tr>
<tr>
<td>(i) 0.213</td>
<td>2.23</td>
<td>1.25 $\sigma$</td>
</tr>
<tr>
<td>(ii) $3.66 \times 10^{-4}$</td>
<td>11.4</td>
<td>3.56 $\sigma$</td>
</tr>
<tr>
<td>(iii) $1.19 \times 10^{-20}$</td>
<td>66.2</td>
<td>9.32 $\sigma$</td>
</tr>
<tr>
<td>2) Reduced data set: $I_a = 1.08 \pm 0.02$</td>
<td></td>
<td></td>
</tr>
<tr>
<td>(i) 0.340</td>
<td>1.56</td>
<td>0.954 $\sigma$</td>
</tr>
<tr>
<td>(ii) 0.0296$\times 2^*$</td>
<td>4.08</td>
<td>1.89 $\sigma$</td>
</tr>
<tr>
<td>(iii) $9.44 \times 10^{-6}$</td>
<td>16.7</td>
<td>4.43 $\sigma$</td>
</tr>
</tbody>
</table>