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Sufficient conditions for three-particle entanglement and their tests in recent experiments

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We point out a loophole problem in some recent experimental claims to produce three-particle entanglement. The problem consists in the question whether mixtures of two-particle entangled states might suffice to explain the experimental data. In an attempt to close this loophole, we review two sufficient conditions that distinguish between N-particle states in which all N particles are entangled to each other and states in which only M particles are entangled (with $M<N$). It is shown that three recent experiments to obtain three-particle entangled states [Bouwmeester *et al.*, Phys. Rev. Lett. **82**, 1345 (1999); Pan *et al.*, Nature **403**, 515 (2000); and Rauschenbeutel *et al.*, Science **288**, 2024, (2000)] do not meet these conditions. We conclude that the question whether these experiments provide confirmation of three-particle entanglement remains unresolved. We also propose modifications of the experiments that would make such confirmation feasible.

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I. INTRODUCTION

The experimental production and detection of multiparticle entanglement have seen much progress during the last years. Manipulation of such highly entangled N-particle states is of great interest for implementing quantum information techniques, such as quantum computing and quantum cryptography, as well as for fundamental tests of quantum mechanics. Extended efforts have resulted in recent claims of experimental confirmation of both three- and four-particle entanglement using photons and atom-cavity techniques [1–5]. In this paper, we examine a possible loophole in such claims.

N-particle entanglement differs from the more well-known two-particle entanglement, not only because the classification of different types of this form of entanglement is still an open problem [6,7], but also because it requires different conditions for actual experimental confirmation. In the case of two-particle entangled states, it suffices to show that the observed data cannot be explained by a ‘‘local realist’’ model. That is, it is sufficient for the correlations between the observed data to violate a certain Bell inequality. In fact, for pure states, this condition is also necessary, because all pure two-particle entangled states can be made to violate such a Bell inequality by an appropriate choice of the observables [8,9].

For N-particle systems, generalized Bell inequalities have been reported by Mermin [10] and Ardehali [11]. These N-particle inequalities are likewise derived under the assumption of local realism. More explicitly, it is assumed that each particle may be assigned independent elements of reality corresponding to certain measurement outcomes. A bound on the expected correlations is then obtained and shown to be violated by the corresponding quantum mechanical expectation values by a maximal factor that grows exponentially with N [10,11]. N-particle experiments that violate these inequalities are then, again, disproofs of the assumptions of local realism.

However, the violation of local realism is not sufficient for confirmation of the entanglement of all N particles. For this purpose, one must also address the question of whether the data admit a model in which less than N particles are entangled. The standard generalized Bell inequalities mentioned above are not designed to deal with this issue, and thus, leave the loophole open that the data might be explained by mixtures of states in which less than N particles are entangled. In fact, as shown in more detail below, the data of some experiments aimed to produce three-particle entanglement may be approximated surprisingly closely by a mixture of two-particle entangled states. This forms the motivation for a closer investigation of conditions needed to close this particular loophole.

Some conditions of this kind have been formulated in the recent literature [7] in terms of partial transpositions of the N-particle density matrix. Unfortunately, it is not clear at present how these conditions may be tested experimentally. In this paper, we review two experimentally accessible conditions, presented in Sec. II as conditions A and B. In Sec. III, we analyze some recent experiments [1,2,4] to produce three-particle entanglement, in order to see whether these conditions are met. It is shown that this is not the case. This, of course, does not prove that there is no three-particle entanglement in these experiments. Rather, we conclude that on the basis of the conditions reviewed here, the above loophole problem remains unresolved. However, we propose modifications of the experimental procedure that would allow for a more definite confirmation of three-particle entanglement.

II. SUFFICIENT CONDITIONS FOR *N*-PARTICLE ENTANGLEMENT

We start with the definition of the basic concept. Consider an arbitrary N-particles system described by a Hilbert space

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$\mathcal{H}=\mathcal{H}_1\otimes\cdots\otimes\mathcal{H}_N$. A general mixed state ρ of this system is called N-particles entangled if no convex decomposition of the form

$$\rho=\sum_i p_i\rho_i \quad \text{with } p_i\geqslant 0, \ \sum_i p_i=1 \tag{1}$$

exists in which all the states ρ_i are factorizable into products of states of less than N particles. Of course, since each factorizable mixed state is a mixture of factorizable pure states, one may equivalently assume that factorizable states ρ_i are pure, so that the decomposition (1) takes the form

$$\rho=\sum_i p_i|\psi_i\rangle\langle\psi_i|. \tag{2}$$

In order to extend the above terminology, let K be any subset $K\subset\{1,\ldots,N\}$ and let ρ^K denote a state of the subsystem composed of the particles labeled by K. We will call an N-particle state M particle entangled ($M<N$) if a decomposition exists of the form

$$\rho=\sum_i p_i\rho_i^{K_1^{(i)}}\otimes\cdots\otimes\rho_i^{K_{r_i}^{(i)}}, \tag{3}$$

where, for each i, $K_1^{(i)},\ldots,K_{r_i}^{(i)}$ is some partition of $\{1,\ldots,N\}$ into r_i disjoint subsets, each subset $K_j^{(i)}$ containing at most M elements; but no such decomposition is possible when these subsets are required to contain less than M elements.

An example of an N-particle state that is N particle entangled is the Greenberger-Horne-Zeilinger (GHZ) state

$$|\psi_{\mathrm{GHZ}}\rangle=\frac{1}{\sqrt{2}}(|\uparrow\uparrow\cdots\uparrow\rangle+|\downarrow\downarrow\cdots\downarrow\rangle), \tag{4}$$

where $|\uparrow\rangle$ and $|\downarrow\rangle$ denote the eigenstates of some dichotomic observable (e.g., spin or polarization) which we will take, by convention, as oriented along the z axis. On the other hand, the three-particle state

$$\rho=\frac{1}{2}(\hat{P}_{\uparrow}^{(1)}\otimes\hat{P}_S^{(23)}+\hat{P}_{\downarrow}^{(1)}\otimes\hat{P}_T^{(23)}) \tag{5}$$

is only two-particle entangled. Here, $\hat{P}_T^{(23)}$ and $\hat{P}_S^{(23)}$ denote projectors on the triplet state $1/\sqrt{2}(|\uparrow\downarrow\rangle+|\downarrow\uparrow\rangle)$ and singlet state $1/\sqrt{2}(|\uparrow\downarrow\rangle-|\downarrow\uparrow\rangle)$, respectively, for particles 2 and 3, and $\hat{P}(1)_{\downarrow}=|\downarrow\rangle\langle\downarrow|$ and $\hat{P}_{\uparrow}^{(1)}=|\uparrow\rangle\langle\uparrow|$ are the "down" and "up" states for particle 1. Note that, as the state (5) exemplifies, an N-particle state can be M particle entangled even if it has no M particle subsystem whose (reduced) state is M-particles entangled. In the remainder of this section, we review two inequalities that allow for a test between N-particle and M-particle entangled states, focusing mainly on $N=3$ and $M=2$.

Condition A. The following condition has been derived by Gisin and Bechmann-Pasquinucci [12] for a system of N two-level particles (q bits). As a start, consider the well-known Bell inequality of Clauser, Horne, Shimony, and Holt (CHSH) [13] for two particles. Let A and A' be dichotomous observables on the first particle, with possible outcomes ± 1, and similarly for observables B and B' on the second particle. Consider the expression

$$F_2:=AB+A'B+AB'-A'B'=(A+A')B+(A-A')B'\leqslant 2. \tag{6}$$

Assuming local realism, the pair A and B are conditionally independent

$$p_{AB}^{\mathrm{lr}}(a,b)=\int_{\Lambda}p_A(a|\lambda)p_B(b|\lambda)\rho(\lambda)d\lambda \tag{7}$$

and similarly for the pairs A',B, A,B', and A',B', where p_A and p_B are probabilities conditional on the hidden variable $\lambda\in\Lambda$. If we denote the expected correlations as

$$E_{\mathrm{lr}}(AB)=\sum_{ab} ab\, p_{AB}^{\mathrm{lr}}(a,b),$$

we obtain the standard two-particle Bell-CHSH inequality [13]

$$|E_{\mathrm{lr}}(F_2)|=|(E_{\mathrm{lr}}(AB)+E_{\mathrm{lr}}(A'B)+E_{\mathrm{lr}}(AB')-E_{\mathrm{lr}}(A'B')| \leqslant 2. \tag{8}$$

In quantum mechanics, the observable A is represented by the spin operator $\hat{A}=\vec{\mathbf{a}}\cdot\vec{\sigma}$ with unit three-dimensional vector $\vec{\mathbf{a}}$, and similarly for the other three observables. The expected correlation in a state ρ is given by $E_\rho(AB)=\mathrm{Tr}\,(\rho\vec{\mathbf{a}}\cdot\vec{\boldsymbol{\sigma}}\otimes\vec{\mathbf{b}}\cdot\vec{\sigma})$. In terms of these expectation values, the Bell-CHSH inequality may be violated by entangled quantum states. The largest violation of this inequality by a quantum state is $2\sqrt{2}$ [14].

The Bell-CHSH inequality is generalized by Gisin and Bechmann-Pasquinucci to N particles through a recursive definition. Let A_j and A_j' denote dichotomous observables on the jth particle, $(j=1,2,\ldots,N)$, and define

$$F_N:=\frac{1}{2}(A_N+A_N')F_{N-1}+\frac{1}{2}(A_N-A_N')F_{N-1}'\leqslant 2, \tag{9}$$

where F_{N-1}' is the same expression as F_{N-1} but with all A_j and A_j' interchanged. Here, the upper bound on F_N follows by natural induction from the bound (6) on F_2. One now obtains the so-called Bell-Klyshko inequality [12],

$$|E_{\mathrm{lr}}(F_N)|\leqslant 2. \tag{10}$$

This Bell-Klyshko inequality is also violated in quantum mechanics. That is to say, the expectation value of the corresponding operator

$$\hat{F}_N:=\frac{1}{2}(\hat{A}_N+\hat{A}_N')\otimes\hat{F}_{N-1}+\frac{1}{2}(\hat{A}_N-\hat{A}_N')\otimes\hat{F}_{N-1}'\leqslant 2 \tag{11}$$

may violate the bound (10) for entangled quantum states. As shown in reference [12], the maximal value is

$$|E_\rho(\hat{F}_N)| \leqslant 2^{(N+1)/2}, \tag{12}$$

i.e., a violation by a factor of $2^{(N-1)/2}$.

The inequality (10) may now be extended into a test of $N-1$ entanglement. Consider a state in which one particle (say the Nth) is independent from the others; i.e., $\rho = \rho_{\{N\}} \otimes \rho_{\{1,\ldots,N-1\}}$. One then obtains

$$\begin{aligned} |E_\rho(\hat{F}_N)| &= \left| \mathrm{Tr} \ \ \rho\left(\frac{1}{2}(\hat{A}_N + \hat{A}'_N) \otimes \hat{F}_{N-1} + \frac{1}{2}(\hat{A}_N - \hat{A}'_N) \otimes \hat{F}'_{N-1}\right)\right| \\ &= \frac{1}{2}\left|(\langle \hat{A}_N\rangle_\rho + \langle \hat{A}'_N\rangle_\rho)\mathrm{Tr} \ \ \rho\hat{F}_{N-1} + (\langle \hat{A}_N\rangle_\rho - \langle \hat{A}'_N\rangle_\rho)\mathrm{Tr} \ \ \rho\hat{F}'_{N-1}\right| \\ &= \frac{1}{2}\left|\langle \hat{A}_N\rangle_\rho (E_\rho(\hat{F}_{N-1}) + E_\rho(\hat{F}'_{N-1})) + \langle \hat{A}'_N\rangle_\rho (E_\rho(\hat{F}_{N-1}) - E_\rho(\hat{F}'_{N-1}))\right| \\ &\leqslant \frac{1}{2}\left|E_\rho(\hat{F}_{N-1}) + E_\rho(\hat{F}'_{N-1})\right| + \frac{1}{2}\left|E_\rho(\hat{F}_{N-1}) - E_\rho(\hat{F}'_{N-1})\right| \\ &= \max(|E_\rho(\hat{F}_{N-1})|, |E_\rho(\hat{F}'_{N-1})|) \leqslant 2^{N/2}, \end{aligned} \tag{13}$$

where we have used $|\langle \hat{A}_N\rangle| \leqslant 1, |\langle \hat{A}'_N\rangle| \leqslant 1$ and the bound (12).

Since $\hat{F}_N$ is invariant under a permutation of the N particles, this bound holds also for a state in which another particle than the Nth factorizes, and, since $E_\rho(F_N)$ is convex as a function of ρ, it holds also for mixtures of such states. Hence, for every $(N-1)$-particle entangled state we have

$$|E_\rho(\hat{F}_N)| \leqslant 2^{N/2}. \tag{14}$$

Thus, a sufficient condition for N-particle entanglement is a violation of Eq. (14), i.e., inequality (10) should be violated by a factor larger than $2^{(N/2-1)}$.

Specializing now to the case where $N=3$, inequality (14) may be written more conveniently as

$$|E(ABC') + E(AB'C) + E(A'BC) - E(A'B'C')| \leqslant 2^{3/2}, \tag{15}$$

where we have put $A_1 = A, \ A_2 = B$, and $A_3 = C$.

For example, for a choice of spin directions $\vec{a} = \vec{a}'$ along the z axis, and $\vec{b}, \ \vec{b}', \ \vec{c}, \ \vec{c}'$ in the xy plane with angles $\beta = 0, \ \beta' = \pi/2, \ \gamma = \pi/4$, and $\gamma' = -\pi/4$ from the x axis, the mixed state (5) gives $E_\rho(F_3) = 2\sqrt{2}$. This violates inequality (10), thus indicating two-particle entanglement, but does not violate inequality (15), and thus shows no three-particle entanglement.

Condition B. Another condition for N-particle entanglement follows from the fact that the internal correlations of a quantum state are encoded in the off-diagonal elements of the density matrix that represents the state in a product basis. We summarize here the derivation presented by Sackett *et al.* [3]. Consider the so-called state preparation fidelity F of a N-particle state ρ defined as

$$F(\rho) := \langle \psi_{\mathrm{GHZ}}|\rho|\psi_{\mathrm{GHZ}}\rangle = \frac{1}{2}(P_\uparrow + P_\downarrow) + \mathrm{Re}\,\rho_{\uparrow\downarrow}, \tag{16}$$

where $|\psi_{\mathrm{GHZ}}\rangle$ is given by (4), $P_\uparrow := \langle \uparrow \cdots \uparrow|\rho|\uparrow \cdots \uparrow\rangle, \ P_\downarrow := \langle \downarrow\downarrow \cdots \downarrow|\rho|\downarrow\downarrow \cdots \downarrow\rangle$ and $\rho_{\uparrow\downarrow} := \langle \uparrow\uparrow \cdots \uparrow|\rho|\downarrow\downarrow \cdots \downarrow\rangle$ is the far off-diagonal matrix element in the z basis. Now, partition the set of N particles into two disjoint subsets K and K' and consider a pure state of the form

$$\begin{aligned} |\phi\rangle = (a|\uparrow\uparrow \cdots \uparrow\rangle^K + \cdots + b|\downarrow\downarrow \cdots \downarrow\rangle^K) \otimes (c|\uparrow\uparrow \cdots \uparrow\rangle^{K'} \\ + \cdots + d|\downarrow\downarrow \cdots \downarrow\rangle^{K'}), \end{aligned} \tag{17}$$

where $|\uparrow\uparrow \cdots \uparrow\rangle^K$ is the state with all particles in subset K in the ''up'' state and similarly for the other terms. Normalization of $|\phi\rangle$ leads to $|a|^2 + |b|^2 \leqslant 1$ and $|c|^2 + |d|^2 \leqslant 1$. It then follows that

$$\begin{aligned} 2F(|\phi\rangle\langle\phi|) &= |ac|^2 + |bd|^2 + 2\,\mathrm{Re}(ab^*cd^*) \\ &\leqslant (|a|^2 + |b|^2)(|c|^2 + |d|^2) \leqslant 1. \end{aligned} \tag{18}$$

Thus, the state preparation fidelity is at most 1/2 for any state of the form (17). From the convexity of $F(\rho)$ it follows that this inequality also holds for any mixture of such product states, i.e., for any state ρ as defined in Eq. (2).

We have thus found a second sufficient condition for N-particle entanglement, namely,

$$F(\rho) > 1/2. \tag{19}$$

Of course, analogous conditions may be obtained by replacing the special state $|\psi_{\mathrm{GHZ}}\rangle$ in definition (16) by another maximally entangled state, such as $1/\sqrt{2}(|\uparrow \ldots \uparrow\downarrow\rangle \pm |\downarrow \ldots \downarrow\uparrow\rangle)$, etc. An experimental test of condition B requires the determination of the real part of the far off-diagonal matrix element $\rho_{\uparrow\downarrow}$. Now, obviously, $\mathrm{Re}\,\rho_{\uparrow\downarrow}$ is not the expectation value of a product observable, and information about this quantity may only be obtained indirectly. In the next section, we discuss several experimental procedures by which this information may be obtained. As we shall see, it is important that such procedures make sure that no unwanted matrix elements contribute to the determination of this quantity.

III. ANALYSIS OF EXPERIMENTS

Using the conditions *A* and *B* discussed above, we now turn to the analysis of three recent experimental tests for three-particle entangled states.

(**I**) In the experiment of Bouwmeester *et al.* [1], the three-photon entangled state $|\psi_{\mathrm{B}}\rangle = 1/\sqrt{2}(|HHV\rangle + |VVH\rangle)$ is claimed to be experimentally observed. Here, $|H\rangle$ and $|V\rangle$ are the horizontal and vertical polarization states of the photons. We represent this state in the z basis using $|H\rangle = |\uparrow\rangle$ and $|V\rangle = |\downarrow\rangle$ as

$$|\psi_{\mathrm{B}}\rangle = \frac{1}{\sqrt{2}}(|\uparrow\uparrow\downarrow\rangle + |\downarrow\downarrow\uparrow\rangle). \tag{20}$$

The experiment consisted, first, of a set of threefold coincidence measurements in the zzz directions, in which the fraction of the desired outcomes, i.e., the components $|\uparrow\uparrow\downarrow\rangle$ and $|\downarrow\downarrow\uparrow\rangle$ out of the 2^3 possible outcomes was determined and found to be in a ratio of 12:1. Furthermore, to show coherent superposition of these components, a second set of measurements was performed in the xxx directions. For a large fraction of the observed data, this second set of measurements shows correlations as expected from the desired state $|\psi_B\rangle$. A third series of measurements performed in the zxx directions showed no such correlations, again, as expected from the state $|\psi_{\mathrm{B}}\rangle$. Bouwmeester *et al.* concluded that: "The data clearly indicate the absence of two-photon correlations and thereby confirm our claim of the observation of GHZ entanglement between three spatially separated photons [1]." However, no quantitative analysis was made to determine whether two-particle entangled states may account for or contribute to the observed data. In order to show that such an analysis is not superfluous, it is shown in Appendix A how most of the salient results of this experiment may in fact be reproduced by a simple two-particle entangled state. Thus, we are presented with the loophole problem whether or not the observed data may be regarded as hard evidence for true three-particle entanglement.

The experiment of Pan *et al.* [4], performed by the same group, aimed to produce the GHZ state $|\psi_{\mathrm{GHZ}}\rangle = 1/\sqrt{2}(|\uparrow\uparrow\uparrow\rangle + |\downarrow\downarrow\downarrow\rangle)$ by a procedure similar to the previous experiment. Although their main goal was to show a conflict with local realism, Pan *et al.* also claim to have provided evidence for three-particle entanglement. For this purpose, they performed four series of measurements, in the xxx, xyy, yxy, and yyx directions, and tested a three-particle Bell inequality of the form derived by Mermin [10]. This inequality is presented in [15] and reads

$$|\langle xyy\rangle + \langle yxy\rangle + \langle yyx\rangle - \langle xxx\rangle| \leqslant 2, \tag{21}$$

where $\langle xyy\rangle$ is the expectation value of $\sigma_x^{(1)}\otimes\sigma_y^{(2)}\otimes\sigma_y^{(3)}$, etc. The reported experimental data are

$$|\langle xyy\rangle + \langle yxy\rangle + \langle yyx\rangle - \langle xxx\rangle| = 2.83 \pm 0.09, \tag{22}$$

in clear violation of Eq. (21). However, as mentioned in the Introduction, violating a generalized Bell inequality of this type is not sufficient to confirm three-particle entanglement. Thus, again, the question remains whether the reported data may be regarded as confirmation of three-particle entanglement. In particular, one might ask, do these experiments meet either of the conditions *A* or *B*?

Upon further analysis, we may answer this question. First, we note that the procedure followed by Bouwmeester *et al.* does not allow for a test of condition *A* even in the ideal case where the desired state is actually produced. This is because measurements were performed only in various directions in the xz plane. However, for any observable $\vec{\mathbf{a}}\cdot\vec{\sigma}\otimes\vec{\mathbf{b}}\cdot\vec{\sigma}\otimes\vec{\mathbf{c}}\cdot\vec{\sigma}$ with $\vec{\mathbf{a}},\vec{\mathbf{b}},\vec{\mathbf{c}}$ unit vectors in the xz plane, we obtain $\langle\psi_{\mathrm{B}}|\vec{\mathbf{a}}\cdot\vec{\sigma}\otimes\vec{\mathbf{b}}\cdot\vec{\sigma}\otimes\vec{\mathbf{c}}\cdot\vec{\sigma}|\psi_{\mathrm{B}}\rangle = \cos\alpha\cos\beta\cos\gamma$, with α, β, and γ the angles these vectors span from the x axis. These expectation values are factorizable, and measurements of spin observables in the xz plane cannot lead to a violation of condition *A*, i.e., the inequality (15). Neither does the choice of measurements in this experiment allow for a test of condition *B*. For such a test, one would have to determine the relevant state preparation fidelity, i.e., $\langle\psi_{\mathrm{B}}|\rho|\psi_{\mathrm{B}}\rangle$ of the experimentally produced state ρ. But the reported data do not allow for an estimate of the relevant off-diagonal element $\mathrm{Re}\langle\uparrow\uparrow\downarrow|\rho|\downarrow\downarrow\uparrow\rangle$. Indeed, the only measurements that are sensitive to the value of this matrix element, namely, those in the xxx directions, are also sensitive to all other matrix elements on the cross diagonal in the zzz eigenbasis.

The experiment by Pan *et al.* is more rewarding in this respect. The inequality (21) tested in this experiment is identical to a Bell-Klyshko inequality (10) for $N=3$. Since the inequality is violated, the experiment is indeed a violation of local realism. However, within experimental errors, the measured value $E(F_3) = 2.83 \approx 2^{3/2}$ does not violate inequality (15) that would be sufficient for evidence of three-particle entanglement. Thus, although the experimental procedure allowed for a test of condition *A*, it did not violate it. Further, the experiment of Pan *et al.* did not attempt to test condition *B* either.

However, both experiments may be simply adjusted to test both conditions. If, in the experiment of Bouwmeester *et al.*, one measures spin observables in directions $\vec{\mathbf{a}}$, $\vec{\mathbf{b}}$, and $\vec{\mathbf{c}}$ in the xy plane, rather than the xz plane, one obtains $E_{|\psi_{\mathrm{B}}\rangle}(ABC) = \langle\psi_{\mathrm{B}}|\vec{\mathbf{a}}\cdot\vec{\sigma}\otimes\vec{\mathbf{b}}\cdot\vec{\sigma}\otimes\vec{\mathbf{c}}\cdot\vec{\sigma}|\psi_{\mathrm{B}}\rangle = \cos(\alpha+\beta-\gamma)$ where α, β, and γ again denote the angles from the x axis. For the choice: $\alpha=\pi/2$, $\alpha'=0$, $\beta=\pi/4$, $\beta'=-\pi/4$, $\gamma=\pi/4$, and $\gamma'=3\pi/4$, the inequality (15) will be violated maximally by the value four.

For the state $|\psi_{\mathrm{GHZ}}\rangle = 1/\sqrt{2}(|\uparrow\uparrow\uparrow\rangle + |\downarrow\downarrow\downarrow\rangle)$, used in the experiment of Pan *et al.*, it follows likewise that $E_{\mathrm{GHZ}}(ABC) = \langle\psi_{\mathrm{GHZ}}|\vec{\mathbf{a}}\cdot\vec{\sigma}\otimes\vec{\mathbf{b}}\cdot\vec{\sigma}\otimes\vec{\mathbf{c}}\cdot\vec{\sigma}|\psi_{\mathrm{GHZ}}\rangle = \cos(\alpha$

$+\beta+\gamma$) when the vectors are chosen in the xy plane. Then, inequality (15) will be violated maximally by the value four for the choice: $\alpha=\pi/2$, $\alpha'=0$, $\beta=\pi/2$, $\beta'=0$, $\gamma=\pi/2$, and $\gamma'=0$. Using these angles in future experiments will thus allow for tests of three-particle entanglement.

Finally, we discuss how the experiments can be adjusted in order to test condition *B*. Determining the populations $P_{\uparrow}$ and $P_{\downarrow}$ in Eq. (16) is rather trivial and will not be discussed. Here, we mention two possible procedures to determine $\mathrm{Re}\,\rho_{\uparrow\downarrow}$. The first is to use a three-particle analogue of the method used by Sackett *et al.* [3]. Consider the observable $\hat{S}_{\pm}(\phi) \coloneqq \vec{\mathbf{n}}_{\phi}\cdot\vec{\sigma}\otimes\vec{n}_{\phi}\cdot\vec{\sigma}\otimes\vec{n}_{\pm\phi}\cdot\vec{\sigma}$ where $\vec{\mathbf{n}}_{\phi}=(\cos\phi,\sin\phi,0)$. The expectation values $\langle\psi_{\mathrm{GHZ}}|\hat{S}_{+}(\phi)|\psi_{\mathrm{GHZ}}\rangle$ and $\langle\psi_{\mathrm{B}}|\hat{S}_{-}(\phi)|\psi_{\mathrm{B}}\rangle$, considered as functions of ϕ, oscillate as $A\cos(3\phi+\alpha_0)+B\cos(\phi+\beta)+\mathrm{const.}$, where $A=2\,\mathrm{Re}\,\rho_{\uparrow\downarrow}$. (That is, $A=2\,\mathrm{Re}\langle\uparrow\uparrow\uparrow|\rho|\downarrow\downarrow\downarrow\rangle$ in the first, and $A=2\,\mathrm{Re}\langle\uparrow\uparrow\downarrow|\rho|\downarrow\downarrow\uparrow\rangle$ in the second case.) Hence, by measuring $S_{+}(\phi)$ for the GHZ state (4), or $S_{-}(\phi)$ for the state (20), for a variety of angles ϕ, and by filtering out the amplitude that oscillates as $\cos 3\phi$, one obtains an estimate of the relevant off-diagonal element $|\mathrm{Re}\,\rho_{\uparrow\downarrow}|$ needed to test condition *B*.

However, a simpler way to determine this off-diagonal matrix element is to take advantage of the simple operator identity:

$$\sigma_x\otimes\sigma_y\otimes\sigma_y+\sigma_y\otimes\sigma_x\otimes\sigma_y+\sigma_y\otimes\sigma_y\otimes\sigma_x-\sigma_x\otimes\sigma_x\otimes\sigma_x$$
$$=-4(|\downarrow\downarrow\downarrow\rangle\langle\uparrow\uparrow\uparrow|+|\uparrow\uparrow\uparrow\rangle\langle\downarrow\downarrow\downarrow|), \tag{23}$$

so that for all states ρ

$$\langle xyy+yxy+yyx-xxx\rangle_{\rho}=-8\,\mathrm{Re}\langle\uparrow\uparrow\uparrow|\rho|\downarrow\downarrow\downarrow\rangle. \tag{24}$$

Since the expectation value in the left-hand side of Eq. (24) has already been measured in the experiment of Pan *et al.*, one may infer from their reported result (22) that

$$|\mathrm{Re}(\rho_{\uparrow\downarrow})|=\frac{2.83\pm0.09}{8}=0.35\pm0.01.$$

Thus, only one additional measurement in the zzz directions would have been sufficient for a full test of condition *B*. If the ratio reported in the experiment of Bouwmeester *et al.* of 12:1 (corresponding to populations of 0.40) is a feasible result for the setup of Pan *et al.* too, one should expect to obtain an experimental value of $F(\rho)\approx0.75$, well above the threshold value of 1/2.

(II). The experiment of Rauschenbeutel *et al.* [2] was set up to measure three-particle entanglement for three spin-1/2 systems (two atoms and a single-photon cavity field mode). The state of the cavity field is not directly observable, and was therefore copied onto a third atom, so that the actual measurement was carried out on a three-atom system. Let us first adapt the notation of [2] to the notation of this paper: Their target three-atom state $|\Psi_{\mathrm{triplet}}\rangle=1/\sqrt{2}(|e_1,i_2,g_3\rangle+|g_1,g_2,e_3\rangle)$ is represented here as $|\psi_B\rangle=1/\sqrt{2}(|\uparrow\uparrow\downarrow\rangle+|\downarrow\downarrow\uparrow\rangle)$.

Condition *B* was used to test for three-particle entanglement. The measured fidelity is claimed to be $F=0.54\pm0.03$ and this is, within experimental accuracy, only just greater than the sufficient value of 1/2. However, we will argue that upon a ‘‘worst-case’’ analysis of the data, this result may no longer be claimed to hold, since one cannot exclude that other off-diagonal density-matrix elements contribute to their determination of $\mathrm{Re}\,\rho_{\uparrow\downarrow}$.

In the experiment, first the individual populations of eigenstates in the zzz directions was determined. These populations are the so-called longitudinal correlations in Fig. 3 of [2] and give the following results: (all numbers ±0.01)

P↑↑↑	P↑↑↓	P↑↓↑	P↑↓↓	P↓↑↑	P↓↑↓	P↓↓↑	P↓↓↓
0.1	0.22	0.06	0.04	0.1	0.09	0.36	0.03

(25)

This gives $1/2(P_{\uparrow\uparrow\downarrow}+P_{\downarrow\downarrow\uparrow})=0.29$. Next, the off-diagonal matrix element $\mathrm{Re}\langle\uparrow\uparrow\downarrow|\rho|\uparrow\uparrow\downarrow\rangle$ is determined by first projecting particle 2 onto either $|+\rangle_x$ or $|-\rangle_x$, and measuring the so-called ‘‘Bell signals’’ $\hat{B}_{\pm}(\phi)\coloneqq\sigma_x^{(1)}\otimes\vec{\mathbf{n}}_{\phi}\cdot\sigma^{(3)}$ on the remaining pair. Here, again, $\vec{\mathbf{n}}_{\phi}=(\cos\phi,\sin\phi,0)$.

Thus, the expectation of these Bell signals is given by $\langle\hat{B}_{\pm}(\phi)\rangle=\mathrm{Tr}\,(\rho\sigma_x^{(1)}\otimes\hat{P}_{\pm}^{(2)}\otimes\vec{\mathbf{n}}_{\phi}\cdot\vec{\sigma}^{(3)})$. The Bell signal $\langle\hat{B}_{+}(\phi)\rangle$ is predicted to oscillate as $A\cos\phi$. The other Bell signal $\langle\hat{B}_{-}(\phi)\rangle$ has a phase shift of π and thus oscillates as $-A\cos\phi$. In the case of the desired three-particle state (20), the amplitude A of the oscillatory Bell signals is equal to $A=2|\langle\uparrow\uparrow\downarrow|\rho|\downarrow\downarrow\uparrow\rangle|$. The experimental data give a value of $A=0.28\pm0.04$, leading to the result $F=1/2(P_{\uparrow\uparrow\downarrow}+P_{\downarrow\downarrow\uparrow}+A)=0.54\pm0.03$.

However, if one assumes a general unknown state, it turns out that not only the matrix element $\langle\uparrow\uparrow\downarrow|\rho|\downarrow\downarrow\uparrow\rangle$ (and its complex conjugate), but also the elements $\langle\uparrow\uparrow\uparrow|\rho|\downarrow\downarrow\downarrow\rangle$, $\langle\uparrow\downarrow\downarrow|\rho|\downarrow\uparrow\uparrow\rangle$ and $\langle\uparrow\downarrow\uparrow|\rho|\downarrow\uparrow\downarrow\rangle$ and their respective complex conjugates contribute to the measured amplitude *A*. In a ‘‘worst-case’’ analysis, these unwanted density-matrix elements should be assigned the highest possible value compatible with the values of the measured populations in table (25). Suppose these contributions sum up to the maximal value w in the amplitude A, then we may conclude that $2\,\mathrm{Re}\,\rho_{\uparrow\downarrow}$ has the ‘‘worst-case’’ value of $A-w$.

Using the data from [2], such an analysis has been performed from which we obtain $w=0.26\pm0.04$ (see Appendix B for details). $2\,\mathrm{Re}\,\rho_{\uparrow\downarrow}$ then has the approximate value of 0.02 ± 0.05 instead of the value 0.28 ± 0.04 reported by Rauschenbeutel *et al.* This value gives an approximate fidelity $F=0.31\pm0.05$, which no longer meets the inequality $F\geqslant1/2$ of Condition *B*.

One might object to our worst case analysis because it assumes a maximal contribution from other three-particle entangled states. This is not only physically implausible, but would also give rise to the hope that at least *some* three-particle entangled state has been observed. The prospects of this hope are difficult to assess. Of course, one has to take into account that a mixture of different three-particle entangled states is not necessarily a three-particle entangled state. But it is difficult to say whether or not this holds for the worst-case mixture discussed in Appendix B.

However this may be, it is straightforward to show that the unwanted matrix elements may contaminate the data from this experiment, even for two-particle entangled states. For example, consider the incoherent mixture of two pure Bell signal states

$$\rho_{\mathrm{mix}}=\frac{1}{2}(\hat{P}_{+}^{(2)}\otimes\hat{P}_{S}^{(13)}+\hat{P}_{-}^{(2)}\otimes\hat{P}_{T}^{(13)}), \tag{26}$$

where $\hat{P}_{T}^{(13)}$ and $\hat{P}_{S}^{(13)}$ denote projectors on the triplet state $1/\sqrt{2}(|\uparrow\downarrow\rangle+|\downarrow\uparrow\rangle)$ and singlet state $1/\sqrt{2}(|\uparrow\downarrow\rangle-|\downarrow\uparrow\rangle)$, respectively, for the particles 1 and 3, and $\hat{P}_{\pm}^{(2)}$ are the eigenprojectors in the x direction for particle 2. For this state, the expected values of $P_{\uparrow\uparrow\downarrow}$ and $P_{\downarrow\downarrow\uparrow}$ are 0.25, and $A :=\max_{\phi}|\mathrm{Tr}\;\;\rho_{\mathrm{mix}}\hat{B}_{\pm}(\phi)|=1$, while $\langle\uparrow\uparrow\downarrow|\rho_{\mathrm{mix}}|\downarrow\downarrow\uparrow\rangle=1/4$. In the experimental procedure of Rauschenbeutel *et al.*, this would lead one to conclude that the state preparation fidelity is $F=1/2(P_{\uparrow}+P_{\downarrow}+A)=0.75$, even though its actual value is only 0.5. This shows clearly how the contribution by unwanted matrix elements may corrupt the data for two-particle entangled states.

We conclude that this experiment does not provide evidence of three-particle entanglement. In order to exclude the contribution by undesired matrix density elements in the experimental determination of $\mathrm{Re}\,\rho_{\uparrow\downarrow}$, another experimental procedure is needed, e.g., an analog of the methods discussed above, or a test of conditions A and/or B is needed to warrant such a claim.

IV. CONCLUSION

Experimental evidence for N-particle entanglement for N-particle states requires stronger conditions than a mere violation of local realism. M-particle entangled states, with $M<N$, have to be excluded as well. This leaves a loophole in recent experimental claims of evidence for multiparticle entangled states. We have reviewed two experimentally testable conditions that are sufficient to close this loophole, and analyzed three recent experiments to see whether they meet these conditions. Unfortunately, this is not the case. Hence, we conclude that the question remains unresolved whether these experiments provide confirmation of three-particle entanglement. However, we have proposed modifications of the experimental procedure that would make such confirmation possible. We hope that further experimental tests of N-particle entanglement (e.g., the recently published [5]), will take account of the specific requirements needed to test conditions such as A and B discussed above.

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APPENDIX A

The data obtained in the experiment of Bouwmeester *et al.* may be summarized as follows: (i) The measurements in the zzz basis give a value of 12:1 for the ratio between the desired outcomes and the remainder. This means that

$$\langle\uparrow\uparrow\downarrow|\rho|\uparrow\uparrow\downarrow\rangle=\langle\downarrow\uparrow\uparrow|\rho|\downarrow\uparrow\uparrow\rangle=0.4 \tag{A1}$$

and

$$\langle\uparrow\uparrow\uparrow|\rho|\uparrow\uparrow\uparrow\rangle=\cdots=\langle\downarrow\downarrow\downarrow|\rho|\downarrow\downarrow\downarrow\rangle=0.033 \tag{A2}$$

for the remaining six outcomes.

(ii) The measurements performed in the xxx directions determined the probability of $\hat{P}_{+}^{(1)}\otimes\hat{P}^{(2)}\otimes\hat{P}_{\pm}^{(3)}$. The experimental results are depicted in Fig. 2 of Ref. [1], and show a difference between the ± settings which is about 75% of the expected difference in the desired state $|\psi_{\mathrm{B}}\rangle$. Hence,

$$\begin{aligned}\mathrm{Tr}\;\;\rho\hat{P}_{+}^{(1)}\otimes\hat{P}_{-}^{(2)}\otimes\sigma_{x}^{(3)}&=\mathrm{Tr}\;\;\rho\hat{P}_{+}^{(1)}\otimes\hat{P}_{-}^{(2)}\otimes(\hat{P}_{+}^{(3)}-\hat{P}_{-}^{(3)})\\&=\frac{3}{4}\langle\psi_{\mathrm{B}}|\hat{P}_{+}^{(1)}\otimes\hat{P}_{-}^{(2)}\otimes\sigma_{x}^{(3)}|\psi_{\mathrm{B}}\rangle\\&=-\frac{3}{16}.\end{aligned}\tag{A3}$$

(iii) In a control measurement, the setting of the polarizer for the first particle was rotated to the $+z$ direction. This measurement thus determines the value of $\hat{P}_{\uparrow}^{(1)}\otimes\hat{P}^{(2)}\otimes\hat{P}_{\pm}^{(3)}$. In this case, no interference (i.e., no difference between the ± setting for particle three) was observed. This gives the constraint

$$\mathrm{Tr}\;\;\rho\hat{P}_{\uparrow}^{(1)}\otimes\hat{P}^{(2)}\otimes\sigma_{x}^{(3)}=0. \tag{A4}$$

We now show how most of these results may be reproduced by a simple two-particle entangled state. Consider the state

$$W=\alpha\hat{P}_{-}^{(2)}\otimes\hat{P}_{S}^{(13)}+\frac{1-\alpha}{2}(\hat{P}_{|\uparrow\uparrow\downarrow\rangle}+\hat{P}_{|\downarrow\downarrow\uparrow\rangle}), \tag{A5}$$

where $\hat{P}_{S}^{(13)}$ is the projector on the singlet state $1/\sqrt{2}(|\uparrow\downarrow\rangle-|\downarrow\uparrow\rangle)=1/\sqrt{2}(|+-\rangle-|-+\rangle)$.

Using this state (A5), one finds

$$\mathrm{Tr}\;\;W\hat{P}_{\uparrow}^{(1)}\otimes\hat{P}_{-}^{(2)}\otimes\sigma_{x}^{(3)}=0 \tag{A6}$$

in agreement with Eq. (A4). Moreover,

$$\mathrm{Tr}\;\;W\hat{P}_{+}^{(1)}\otimes\hat{P}^{(2)}\otimes\sigma_{x}^{(3)}=-\frac{\alpha}{2}, \tag{A7}$$

which gives agreement with Eq. (A3) for $\alpha=3/8$. Finally, using this choice for α we find

$$\langle\uparrow\uparrow\downarrow|W|\uparrow\uparrow\downarrow\rangle=\langle\downarrow\uparrow\uparrow|W|\downarrow\uparrow\uparrow\rangle=\frac{13}{32}\approx0.41, \tag{A8}$$

which is sufficiently close to Eq. (A1).

The only aspect in which the state (A5) fails to reproduce the experimental data is in the constraint (A2). Instead, the state W gives

$$\langle \uparrow\downarrow\downarrow | W | \uparrow\downarrow\downarrow \rangle = \langle \downarrow\uparrow\uparrow | W | \downarrow\uparrow\uparrow \rangle = \frac{3}{32} \approx 0.09, \qquad \text{(A9)}$$

$$\begin{aligned} \langle \uparrow\uparrow\uparrow | W | \uparrow\uparrow\uparrow \rangle &= \langle \uparrow\downarrow\uparrow | W | \uparrow\downarrow\uparrow \rangle \\ &= \langle \downarrow\uparrow\uparrow | W | \downarrow\uparrow\uparrow \rangle \\ &= \langle \downarrow\downarrow\downarrow | W | \downarrow\downarrow\downarrow \rangle = 0. \qquad \text{(A10)} \end{aligned}$$

Of course, the fit of the experimental data might be improved by varying some parameters of the state (A5) or utilizing the margins offered by the finite measurement accuracies. However, the purpose of this calculation is not to claim that all these data may consistently be reproduced by two-particle entangled state. Rather, we wish to point out that one may approximate the data unexpectedly closely, so that a serious quantitative test is needed before one may claim that these data confirm three-particle entanglement.

APPENDIX B

The two "Bell signals" measured in the experiment of Rauschenbeutel *et al.* correspond to $\langle \hat{B}_+(\phi) \rangle = \mathrm{Tr}\ \rho\sigma_x^{(1)} \otimes \hat{P}_+^{(2)} \otimes \sigma_\phi^{(3)}$ and $\langle \hat{B}_-(\phi) \rangle = \mathrm{Tr}\ \rho\sigma_x^{(1)} \otimes \hat{P}_-^{(2)} \otimes \sigma_\phi^{(3)}$ where $\hat{P}_\pm^{(2)}$ are projectors on the "up" and "down" states for spin in the x direction for particle 2. It is, however, more convenient to deal with their difference, i.e., $\langle \hat{B}_+(\phi) \rangle - \langle \hat{B}_-(\phi) \rangle = \mathrm{Tr}\ \rho\sigma_x^{(1)} \otimes \sigma_x^{(2)} \otimes \sigma_\phi^{(3)}$. Let us label the eight basis vectors $|\uparrow\uparrow\uparrow\rangle$, $|\uparrow\uparrow\downarrow\rangle$, $|\uparrow\downarrow\uparrow\rangle$, $|\uparrow\downarrow\downarrow\rangle$, $|\downarrow\uparrow\uparrow\rangle$, $|\downarrow\uparrow\downarrow\rangle$, $|\downarrow\downarrow\uparrow\rangle$, $|\downarrow\downarrow\downarrow\rangle$, consecutively by $1, \ldots, 8$. A straightforward calculation yields

$$\begin{aligned} \langle \hat{B}_+(\phi) - \hat{B}_-(\phi) \rangle = 2|\rho_{72}|\cos(\phi + \varphi_{72}) \\ + 2|\rho_{54}|\cos(\phi + \varphi_{54}) \\ + 2|\rho_{36}|\cos(\phi + \varphi_{36}) \\ + 2|\rho_{18}|\cos(\phi + \varphi_{18}), \end{aligned}$$

where $\rho_{72} = \rho_{27}^\star = |\rho_{72}|\exp(i\varphi_{72})$ and similarly for the other matrix elements.

In a worst-case analysis, all the phase factors such as φ_{72} are chosen equal to 0 and $|\rho_{54}|$, $|\rho_{36}|$ and $|\rho_{18}|$ should be given their maximal values compatible with the measured populations given in Eq. (25). These maximal values are obtained from the following worst-case decomposition of the unknown density matrix: $\rho = \alpha\sigma + \beta\tau + \gamma\upsilon + \delta\omega$ with σ, τ, and υ the density matrices of the entangled states $1/\sqrt{2}(|\uparrow\uparrow\uparrow\rangle + |\downarrow\downarrow\downarrow\rangle)$, $1/\sqrt{2}(|\uparrow\downarrow\downarrow\rangle + |\downarrow\uparrow\uparrow\rangle)$, and $1/\sqrt{2}(|\downarrow\uparrow\downarrow\rangle + |\uparrow\downarrow\uparrow\rangle)$, respectively. ω is an arbitrary density matrix, whose off-diagonal matrix elements, however, are assumed to have zero entries where any of the three other states σ, τ, and υ has nonzero entries. Using this decomposition, it follows that $|\rho_{18}| = \alpha/2$, $|\rho_{54}| = \beta/2$ and $|\rho_{36}| = \gamma/2$.

However, since $\sigma_{18} = \sigma_{11} = \sigma_{88}$, and similar relations for τ and υ, the fractions α, β, and γ also contribute to the populations ρ_{ii} of the total state, whose measured values are collected above in table (25). The maximal values compatible with these measured populations ρ_{ii} are: $\alpha/2 = 0.03 \pm 0.01, \beta/2 = 0.04 \pm 0.01, \gamma/2 = 0.06 \pm 0.01$ and the maximal value of w is thus $w = \alpha + \beta + \gamma = 0.26 \pm 0.04$, and $2\rho_{72} = A - w = 0.28 \pm 0.04 - 0.26 \pm 0.03 = 0.02 \pm 0.05$.

[1] D. Bouwmeester, J.-W. Pan, M. Daniell, H. Weinfurter, and A. Zeilinger, Phys. Rev. Lett. **82**, 1345 (1999).
[2] A. Rauschenbeutel, G. Nogues, S. Osnaghi, P. Bertet, M. Brune, J. Raimond, and S. Haroche, Science **288**, 2024 (2000).
[3] C. A. Sackett, D. Kielpinski, B. E. King, C. Langer, V. Meyer, C. J. Myatt, M. Rowe, Q. A. Turchette, W. M. Itano, D. J. Wineland, and C. Monroe, Nature (London) **404**, 256 (2000).
[4] J.-W. Pan, D. Bouwmeester, M. Daniell, H. Weinfurter, and A. Zeilinger, Nature (London) **403**, 515 (2000).
[5] J.-W. Pan, M. Daniell, S. Gasparoni, G. Weihs, and A. Zeilinger, Phys. Rev. Lett. **86**, 4435 (2001).
[6] S. Popescu, Phys. Rev. Lett. **74**, 2619 (1995).
[7] M. Lewenstein, B. Kraus, P. Horedecki, and J. I. Cirac, Phys. Rev. A **63**, 044304 (2001).
[8] N. Gisin, Phys. Lett. A **154**, 201 (1991).
[9] S. Popescu and D. Rohrlich, Phys. Lett. A **166**, 293 (1992).
[10] N. D. Mermin, Phys. Rev. Lett. **65**, 1838 (1990).
[11] M. Ardehali, Phys. Rev. A **46**, 5375 (1992).
[12] N. Gisin and H. Bechmann-Pasquinucci, Phys. Lett. A **246**, 1 (1998).
[13] J. F. Clauser, M. A. Horne, A. Shimony, and R. A. Holt, Phys. Rev. Lett. **23**, 880 (1969).
[14] B. S. Cirel'son, Lett. Math. Phys. **4**, 93 (1980).
[15] D. Bouwmeester, J.-W. Pan, M. Daniell, H. Weinfurter, and A. Zeilinger, in *The Physics of Quantum Information*, edited by D. Bouwmeester, A. Ekert, and A. Zeilinger (Springer, Berlin, 2000), pp. 197–209.