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

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July 30, 2001  

Abstract  

We review two conditions that distinguish between states of \( N \) particles in which all \( N \) particles are entangled to each other and states in which only \( M \) particles entangled (with \( M < N \)). These conditions are used to analyse 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)). It is shown that these experiments do not meet the conditions discussed here and can therefore not be considered as definitive confirmation of three-particle entanglement. We also discuss the modifications of the experiments which would make such confirmation possible.  
PACS: 03.65 Ud

1 Introduction

The experimental production and detection of multi-particle entanglement has 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 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].

$N$-particle entanglement differs from the more well-known two-particle entanglement, not only because the classification of different types of this form of the entanglement is still an open problem [6], 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 [7, 8].

For $N$-particle systems, generalised Bell inequalities have been reported by Mermin [9] and Ardehali [10]. These $N$-particle inequalities are likewise derived under the assumption of local realism. More explicitly, it is assumed that each particle can 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 $N$ [9, 10]. $N$-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 whether the data admit a model in which less than $N$ particles are entangled. The standard generalised Bell-inequalities mentioned above are not designed to deal with this issue. All they test for is whether the observed data exhibit some entanglement. In order to demonstrate true $N$-particle entanglement more stringent conditions are needed.

It is the purpose of this letter to review two experimentally accessible conditions, presented in section 2 as conditions $A$ and $B$. Further, in section 3, recent experiments to observe three-particle entanglement by Bouwmeester et al. [1] by Pan et al. [4]) and of Rauschenbeutel et al. [2] are analysed to see whether or not they meet these conditions. It is shown that this is not the case. We therefore conclude that these experiments cannot yet be considered as undisputable confirmation of three-particle entanglement. We also discuss modifications of the experimental procedure which would allow for a test of these conditions.
2 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 \( \mathcal{H} = \mathcal{H}_1 \otimes \cdots \otimes \mathcal{H}_N \). A general mixed state \( \rho \) of this system is called \( N \)-particles entangled iff no convex decomposition of the form

\[
\rho = \sum_i p_i \rho_i, \quad \text{with } p_i \geq 0, \sum_i p_i = 1,
\]

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

\[
\rho = \sum_i p_i \langle \psi_i | \psi_i \rangle.
\]

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

\[
\rho = \sum_i p_i \rho_{K_1}^{(i)} \otimes \cdots \otimes \rho_{K_{r_i}}^{(i)}
\]

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 which is \( N \)-particle entangled is the Greenberger-Horne-Zeilinger-state

\[
|\psi_{\text{GHZ}}\rangle = \frac{1}{\sqrt{2}} (|\uparrow \cdots \uparrow\rangle + |\downarrow \cdots \downarrow\rangle),
\]

where \(|\uparrow\rangle\) and \(|\downarrow\rangle\) denote the eigenstates of some dichotomic observable (e.g. spin or polarisation) which we will take, by convention, as oriented along the \( z \)-axis. On the other hand, the 3-particle state

\[
\rho = \frac{1}{2} (\hat{P}_1^{(1)} \otimes \hat{P}_S^{(23)} + \hat{P}_1^{(1)} \otimes \hat{P}_T^{(23)})
\]

is only two-particle entangled. Here, \( \hat{P}_T^{(23)} \) and \( \hat{P}_S^{(23)} \) denote projectors on the triplet state \( \frac{1}{\sqrt{2}} (|\uparrow \uparrow \rangle + |\downarrow \downarrow \rangle) \) and singlet state \( \frac{1}{\sqrt{2}} (|\uparrow \downarrow \rangle - |\downarrow \uparrow \rangle) \) respectively for particles 2 and 3, and \( \hat{P}_1^{(1)} = |\downarrow\rangle \langle \downarrow| \) and \( \hat{P}_1^{(1)} = |\uparrow\rangle \langle \uparrow| \) 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 [11] for a system of $N$ two-level particles (q-bits). As a start, consider the well-known Bell-CHSH inequality [12] for two particles. Let $A$ and $A'$ be dichotomous observables on the first particle, with possible outcomes $\pm 1$, and similarly for observables $B$ and $B'$ on the second particle. Consider the expression:

$$F_2 := AB + A'B + AB' - A'B' = (A + A')B + (A - A')B' \leq 2. \quad (6)$$

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

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

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

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

we obtain the standard two-particle Bell-CHSH inequality [12]:

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

In quantum mechanics the observable $A$ is represented by the spin operator $\hat{A} = \vec{a} \cdot \vec{\sigma}$ with unit three-dimensional vector $\vec{a}$, and similarly for the other three observables. The expected correlation in a state $\rho$ is given by $E_{\rho}(AB) = \text{Tr}(\rho \vec{a} \cdot \vec{\sigma} \otimes \vec{b} \cdot \vec{\sigma})$. In terms of these expectation values the Bell-CHSH inequality can be violated by entangled quantum states. The largest violation of this inequality by a quantum state is $2\sqrt{2}$ [13].

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

$$F_N := \frac{1}{2}(A_N + A'_N)F_{N-1} + \frac{1}{2}(A_N - A'_N)F'_{N-1} \leq 2, \quad (9)$$

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 [11],

$$|E_{lr}(F_N)| \leq 2. \quad (10)$$

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

$$\hat{F}_N := \frac{1}{2}(\hat{A}_N + \hat{A}'_N) \otimes \hat{F}_{N-1} + \frac{1}{2}(\hat{A}_N - \hat{A}'_N) \otimes \hat{F}'_{N-1} \leq 2 \quad (11)$$
may violate the bound (10) for entangled quantum states. As shown in reference [11], the maximal value is
\[ |E_\rho(\tilde{F}_N)| \leq 2^{(N+1)/2}, \]  
(12)
i.e. a violation by a factor \(2^{(N-1)/2}\).

The inequality (10) can now be extended into a test of \(N-1\)-entanglement. Consider a state in which one particle (say the \(N\)-th) is independent from the others, i.e.: \(\rho = \rho_N \otimes \rho_{1,...,N-1}\). One then obtains:
\[
|E_\rho(\tilde{F}_N)| = \left| \text{Tr } \rho \left( \frac{1}{2}(\hat{A}_N + \hat{A'}_N) \otimes \tilde{F}_{N-1} + \frac{1}{2}(\hat{A}_N - \hat{A'}_N) \otimes \tilde{F}'_{N-1} \right) \right| \\
= \frac{1}{2} \left| \left( \langle \hat{A}_N \rangle \rho + \langle \hat{A'}_N \rangle \rho \right) \text{Tr } \rho \tilde{F}_{N-1} + \left( \langle \hat{A}_N \rangle \rho - \langle \hat{A'}_N \rangle \rho \right) \text{Tr } \rho \tilde{F}'_{N-1} \right| \\
= \frac{1}{2} \left| \langle \hat{A}_N \rangle \rho \left( E_\rho(\tilde{F}_{N-1}) + E_\rho(\tilde{F}'_{N-1}) \right) + \langle \hat{A'}_N \rangle \rho \left( E_\rho(\tilde{F}_N) - E_\rho(\tilde{F}'_{N-1}) \right) \right| \\
\leq \frac{1}{2} \left| E_\rho(\tilde{F}_{N-1}) + E_\rho(\tilde{F}'_{N-1}) \right| + \frac{1}{2} \left| E_\rho(\tilde{F}_{N}) - E_\rho(\tilde{F}'_{N-1}) \right| \\
= \max(|E_\rho(\tilde{F}_{N-1})|, |E_\rho(\tilde{F}'_{N-1})|) \leq 2^{N/2} \tag{13}
\]
where we have used \(|\langle \hat{A}_N \rangle | \leq 1, |\langle \hat{A'}_N \rangle | \leq 1\) and the bound (12).

Since \(\tilde{F}_N\) is invariant under a permutation of the \(N\) particles, this bound holds also for a state in which another particle than the \(N\)-th factorises, and, since \(E_\rho(F_N)\) is convex as a function of \(\rho\), it holds also for mixtures of such states. Hence, for every \((N-1)\)-particle entangled state we have
\[
|E_\rho(\tilde{F}_N)| \leq 2^{N/2}. \tag{14}
\]

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

Specialising now to the case where \(N = 3\), inequality (14) can be written more conveniently as
\[
|E(ABC') + E(AB'C) + E(A'B'BC) - E(A'B'C')| \leq 2^{3/2}, \tag{15}
\]
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 summarise here
the derivation presented by Sackett et al. [3]. Consider the so-called state preparation
fidelity \( F \) of a \( N \)-particle state \( \rho \) defined as
\[
F(\rho) := \langle \psi_{\text{GHZ}} | \rho | \psi_{\text{GHZ}} \rangle = \frac{1}{2}(P_1 + P_4) + \text{Re} \, \rho_{\uparrow\downarrow},
\]
where \( | \psi_{\text{GHZ}} \rangle \) is given by (4), \( P_1 := \langle \uparrow \cdots \uparrow | \rho | \uparrow \cdots \uparrow \rangle \) and \( \rho_{\uparrow\downarrow} := \langle \uparrow \cdots \uparrow | \rho | \downarrow \cdots \downarrow \rangle \) 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
\[
| \phi \rangle = \left( a | \uparrow \cdots \uparrow \rangle^K + \cdots + b | \downarrow \cdots \downarrow \rangle^K \right) \otimes \left( c | \uparrow \cdots \uparrow \rangle^{K'} + \cdots + d | \downarrow \cdots \downarrow \rangle^{K'} \right).
\]
where \( | \uparrow \cdots \uparrow \rangle^K \) is the state with all particles in subset \( K \) in the ‘up’-state and similarly for the other terms. Normalisation of \( | \phi \rangle \) leads to \( |a|^2 + |b|^2 \leq 1 \) and \( |c|^2 + |d|^2 \leq 1 \). It then follows that
\[
2F(| \phi \rangle \langle \phi |) = |ac|^2 + |bd|^2 + 2\text{Re} \left( ab^* cd^* \right) \leq \left( |a|^2 + |b|^2 \right) \left( |c|^2 + |d|^2 \right) \leq 1.
\]
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 \( \rho \) as defined in Eqn. (2).

We have thus found a second sufficient condition for \( N \)-particle entanglement, namely
\[
F(\rho) > 1/2.
\]
Of course, analogous conditions can be obtained by replacing the special state \( | \psi_{\text{GHZ}} \rangle \) in
definition (16) by another maximally entangled state, such as \( \frac{1}{\sqrt{2}}(| \uparrow \cdots \uparrow \rangle \pm | \downarrow \cdots \downarrow \rangle) \)
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, \( \text{Re} \, \rho_{\uparrow\downarrow} \) is not the expectation
value of a product observable, and information about this quantity can only be obtained
indirectly. In the next section we discuss several experimental procedures by which this
information can 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.

3 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
\[
| \psi_B \rangle = \frac{1}{\sqrt{2}}(| HHV \rangle + | VHV \rangle)
\]
is claimed to be experimentally observed. Here \( | H \rangle \)
and $|V\rangle$ are the horizontal and vertical polarisation states of the photons. We represent this state in the $z$-basis using $|H\rangle = |\uparrow\rangle$ and $|V\rangle = |\downarrow\rangle$ as

$$|\psi_B\rangle = \frac{1}{\sqrt{2}} (|\uparrow\uparrow\downarrow\rangle + |\downarrow\downarrow\uparrow\rangle).$$

(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 $|\psi_B\rangle$. 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 can 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 salient results of this experiment can in fact be reproduced by a simple two-particle entangled state. Thus, the question remains whether or not the observed data can 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 $|\psi_{GHZ}\rangle = \frac{1}{\sqrt{2}} (|\uparrow\uparrow\uparrow\rangle + |\downarrow\downarrow\downarrow\rangle)$ 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 [9]. This inequality is presented in [14] and reads:

$$|\langle xyy \rangle + \langle yxy \rangle + \langle yyx \rangle - \langle xxx \rangle| \leq 2,$$

(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

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

(22)

in clear violation of (21). However, as mentioned in the Introduction, it is not sufficient to violate a generalised Bell-inequality of this type to provide confirmation of three-particle entanglement. Thus, again, the question remains whether the reported data can be regarded as a 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 can 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 when the desired state is actually produced. This is because only measurements were performed in various directions in the $xz$ plane. However, for any observable $\vec{a} \cdot \vec{\sigma} \otimes \vec{b} \cdot \vec{\sigma} \otimes \vec{c} \cdot \vec{\tau}$ with $\vec{a}, \vec{b}, \vec{c}$ unit vectors in the $xz$ plane, we obtain

$$\langle \psi_B | \vec{a} \cdot \vec{\sigma} \otimes \vec{b} \cdot \vec{\sigma} \otimes \vec{c} \cdot \vec{\tau} | \psi_B \rangle = \cos \alpha \cos \beta \cos \gamma,$$

with $\alpha$, $\beta$, and $\gamma$ the angles these vectors span from the $x$-axis. These expectation values are factorisable, 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., $\langle \psi_B | \rho | \psi_B \rangle$ of the experimentally produced state $\rho$. But the reported data do not allow for an estimate of the relevant off-diagonal element $\text{Re} \langle \uparrow \uparrow \downarrow | \rho | \downarrow \downarrow \uparrow \rangle$. Indeed, the only measurements which 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) which 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 can be simply adjusted to test both conditions. If, in the experiment of Bouwmeester et al., one measures spin observables in directions $\vec{a}, \vec{b}$ and $\vec{c}$ in the $xy$-plane, rather than the $xz$-plane, one obtains $E_{\psi_B}(ABC) = \langle \psi_B | \vec{a} \cdot \vec{\sigma} \otimes \vec{b} \cdot \vec{\sigma} \otimes \vec{c} \cdot \vec{\tau} | \psi_B \rangle = \cos(\alpha + \beta - \gamma)$ where $\alpha, \beta,$ and $\gamma$ 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 4.

For the state $|\psi_{\text{GHZ}}\rangle = \frac{1}{\sqrt{2}}(|\uparrow \uparrow \uparrow\rangle + |\downarrow \downarrow \downarrow\rangle)$, used in the experiment of Pan et al., it follows likewise that $E_{\psi_{\text{GHZ}}}(ABC) = \langle \psi_{\text{GHZ}} | \vec{a} \cdot \vec{\sigma} \otimes \vec{b} \cdot \vec{\sigma} \otimes \vec{c} \cdot \vec{\tau} | \psi_{\text{GHZ}} \rangle = \cos(\alpha + \beta + \gamma)$ when the vectors are chosen in the $xy$-plane. Then, inequality (15) will be violated maximally by the value 4 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.

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

$$\sigma_x \otimes \sigma_y \otimes \sigma_y + \sigma_y \otimes \sigma_x \otimes \sigma_y + \sigma_y \otimes \sigma_y \otimes \sigma_x - \sigma_x \otimes \sigma_x \otimes \sigma_x = -4 (|\down\down\down\rangle \langle \uparrow\up\up\up\up| + |\up\up\up\down\down\rangle \langle \down\down\down\down\down|),$$

so that for all states $\rho$:

$$\langle xyy + yxy + yyx - xxx \rangle_{\rho} = -8\text{Re} \langle \up\up\up\down | \rho | \down\down\down \rangle.$$  \hfill (23)

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

$$|\text{Re} (\rho_{\uparrow\downarrow})| = \frac{2.83 \pm 0.09}{8} = 0.35 \pm 0.01.$$

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 set-up of Pan et al. too, one should expect to obtain an experimental value of $F(\rho) \approx 0.75$, 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-$\frac{1}{2}$ 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 3-atom state is $|\Psi_{\text{triplet}}\rangle = \frac{1}{\sqrt{2}}(|e_1, i2, g_3\rangle + |g_1, g_2, e_3\rangle)$ is represented here as $|\psi_B\rangle = \frac{1}{\sqrt{2}}(|\up\up\down\rangle + |\down\down\up\rangle)$.

Condition $B$ was used to test for 3-particle entanglement. The measured fidelity is claimed to be $F = 0.54 \pm 0.03$ 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 can no longer be claimed to hold, since one cannot exclude that other off-diagonal density matrix elements contribute to their determination of $\text{Re} \rho_{\uparrow\downarrow}$.
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)

\[
\begin{array}{cccccccc}
P_{\uparrow\uparrow\uparrow} & P_{\uparrow\uparrow\downarrow} & P_{\uparrow\downarrow\uparrow} & P_{\uparrow\downarrow\downarrow} & P_{\downarrow\uparrow\uparrow} & P_{\downarrow\uparrow\downarrow} & P_{\downarrow\downarrow\uparrow} & P_{\downarrow\down\downarrow} \\
0.1 & 0.22 & 0.06 & 0.04 & 0.1 & 0.09 & 0.36 & 0.03
\end{array}
\]  

(25)

This gives \(\frac{1}{2}(P_{\uparrow\uparrow\uparrow} + P_{\downarrow\down\downarrow}) = 0.29\). Next, the off-diagonal matrix element \(\text{Re} \langle \uparrow\uparrow\downarrow | \rho | \uparrow\uparrow\downarrow \rangle\) is determined by first projecting particle 2 onto either |+\rangle_2 or |−\rangle_2, and measuring the so-called ‘Bell-signals’ \(\hat{B}_\pm(\phi) := \sigma_x^{(1)} \otimes \mathbf{n}_\phi \cdot \sigma^{(3)}\) on the remaining pair. Here, again, \(\mathbf{n}_\phi = (\cos \phi, \sin \phi, 0)\).

Thus, the expectation of these Bell signals is given by \(\langle \hat{B}_\pm(\phi) \rangle = \text{Tr}(\rho \sigma_x^{(1)} \otimes \hat{P}_\pm(2) \otimes \mathbf{n}_\phi \cdot \sigma^{(3)})\). 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 \(\pi\) 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 \(A = 2|\langle \uparrow\uparrow\downarrow | \rho | \downarrow\down\downarrow \rangle|\). The experimental data give a value of \(A = 0.28 \pm 0.04\), leading to the result \(F = \frac{1}{2}(P_{\uparrow\uparrow\uparrow} + P_{\downarrow\down\downarrow} + A) = 0.54 \pm 0.03\).

However, if one assumes a general unknown state, it turns out that not only the matrix element \(\langle \uparrow\uparrow\downarrow | \rho | \uparrow\uparrow\downarrow \rangle\) (and its complex conjugate), but also the elements \(\langle \uparrow\uparrow\uparrow | \rho | \downarrow\down\downarrow \rangle\), \(\langle \uparrow\down\down | \rho | \down\uparrow\uparrow \rangle\) and \(\langle \up\down\down | \rho | \down\up\up \rangle\) 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 can conclude that \(2 \text{Re} \rho_{\uparrow\down\down}\) has the ‘worst-case’ value of \(A - w\).

Using the data from [2] such an analysis has been performed from which we obtain \(w = 0.26 \pm 0.04\) (see Appendix B for details). \(2 \text{Re} \rho_{\uparrow\down\down}\) then has the approximate value of \(0.02 \pm 0.05\) instead of the value \(0.28 \pm 0.04\) reported by Rauschenbeutel et al. This value gives an approximate fidelity \(F = 0.31 \pm 0.05\) which no longer meets the inequality \(F \geq 1/2\) 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 can contaminate the data from this experiment even for two-particle entangled states. For example consider the incoherent mixture of two pure Bell-signal states:

$$\rho_{\text{mix}} = \frac{1}{2} \left( \hat{P}_+^{(2)} \otimes \hat{P}_S^{(13)} + \hat{P}_-^{(2)} \otimes \hat{P}_T^{(13)} \right),$$

(26)

where \(\hat{P}_T^{(13)}\) and \(\hat{P}_S^{(13)}\) denote projectors on the triplet state \(\frac{1}{\sqrt{2}}(|\uparrow\downarrow\rangle + |\downarrow\uparrow\rangle)\) and singlet state \(\frac{1}{\sqrt{2}}(|\uparrow\uparrow\rangle - |\downarrow\downarrow\rangle)\) 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 \(A := \max_\phi |\text{Tr} \rho_{\text{mix}} \hat{B}_\pm (\phi)| = 1\), while \(\langle \uparrow\uparrow\downarrow | \rho_{\text{mix}} | \downarrow\downarrow\uparrow \rangle = \frac{1}{4}\). In the experimental procedure of Rauschenbeutel et al., this would lead one to conclude that the state preparation fidelity is \(F = \frac{1}{2}(P_{\uparrow} + P_{\downarrow} + A) = 0.75\), even though its actual value is only 0.5. This shows clearly how the contribution by unwanted matrix elements can 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 matrix elements in the experimental determination of \(\text{Re} \rho_{\uparrow\uparrow\downarrow}\) another experimental procedure is needed, e.g. an analog of the methods discussed above, or else a test of Conditions \(A\) and/or \(B\) is needed to warrant such a claim.

4 Conclusion

Experimental evidence for \(N\)-particle entanglement for \(N\)-particle states requires stronger conditions than merely violating standard Bell-inequalities that test local realism. \(M\)-particle entangled states, with \(M < N\), have to be excluded as well. After reviewing two experimentally testable conditions which are sufficient for this purpose, we have shown that some recently performed experiments to detect three-particle entanglement do not meet these conditions. Hence, on the basis of these conditions, we conclude that these experiments have not provided hard evidence for three-particle entanglement. We have presented suggestions for improvement of these experiments. We hope that further experimental tests of \(N\)-particle entanglement (see e.g. [5]) will take account of the specific requirements needed to test conditions such as \(A\) and \(B\) discussed above.

Acknowledgements

The subject of this work was originally suggested by Sandu Popescu to one of the authors (M.S). We are very grateful to him for helpful comments, and also to Harvey Brown.
Appendix A

The data obtained in the experiment of Bouwmeester et al. can be summarised 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:

$$
\langle \uparrow \uparrow \downarrow | \rho | \uparrow \uparrow \downarrow \rangle = \langle \downarrow \uparrow \uparrow | \rho | \downarrow \uparrow \uparrow \rangle = 0.4,
$$

and

$$
\langle \uparrow \uparrow \uparrow | \rho | \uparrow \uparrow \uparrow \rangle = \ldots = \langle \downarrow \downarrow \downarrow | \rho | \downarrow \downarrow \downarrow \rangle = 0.033
$$

for the remaining six outcomes.

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

$$
\text{Tr} \, \rho \, \hat{P}_+^{(1)} \otimes \hat{P}_-^{(2)} \otimes \sigma_x^{(3)} = \frac{3}{4} \langle \psi_B | \hat{P}_+^{(1)} \otimes \hat{P}_-^{(2)} \otimes \sigma_x^{(3)} | \psi_B \rangle = -\frac{3}{16}.
$$

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

$$
\text{Tr} \, \rho \hat{P}_+^{(1)} \otimes \hat{P}_-^{(2)} \otimes \sigma_x^{(3)} = 0.
$$

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

$$
W = \alpha \hat{P}_+^{(2)} \otimes \hat{P}_S^{(13)} + \frac{1-\alpha}{2} \left( \hat{P}_+^{(11)} + \hat{P}_+^{(1)} \right),
$$

where $\hat{P}_S^{(13)}$ is the projector on the singlet state $\frac{1}{\sqrt{2}}(|\uparrow \downarrow \rangle - |\downarrow \uparrow \rangle) = \frac{1}{\sqrt{2}}(|+\downarrow \rangle - |\downarrow + \rangle)$. Using this state (31) one finds:

$$
\text{Tr} \, W \hat{P}_+^{(1)} \otimes \hat{P}_-^{(2)} \otimes \sigma_x^{(3)} = 0
$$
in agreement with (30). Moreover,
\[ \text{Tr } \hat{W} \hat{P}_+^{(1)} \otimes \hat{P}_+^{(2)} \otimes \sigma_x^{(3)} = -\frac{\alpha}{2}, \]  
(33)
which gives agreement with (29) for \( \alpha = 3/8 \). Finally, using this choice for \( \alpha \) we find
\[ \langle \uparrow \uparrow \downarrow | \hat{W} | \uparrow \uparrow \downarrow \rangle = \langle \downarrow \uparrow \uparrow | \hat{W} | \downarrow \uparrow \uparrow \rangle = \frac{13}{32} \approx 0.41, \]
(34)
which is sufficiently close to (27).

The only aspect in which the state (31) fails to reproduce the experimental data is in the constraint (28). Instead, the state \( W \) gives
\[ \langle \uparrow \downarrow \downarrow | \hat{W} | \uparrow \downarrow \downarrow \rangle = \langle \downarrow \uparrow \uparrow | \hat{W} | \downarrow \uparrow \uparrow \rangle = \frac{3}{32} \approx 0.09, \]
(35)
\[ \langle \uparrow \uparrow \uparrow | \hat{W} | \uparrow \uparrow \uparrow \rangle = \langle \uparrow \downarrow \uparrow | \hat{W} | \uparrow \downarrow \uparrow \rangle = \langle \downarrow \uparrow \uparrow | \hat{W} | \downarrow \uparrow \uparrow \rangle = \langle \downarrow \downarrow \downarrow | \hat{W} | \downarrow \downarrow \downarrow \rangle = 0. \]
(36)

Of course, the fit of the experimental data might be improved by varying some parameters of the state (31) or utilising the margins offered by the finite measurement accuracies. However, the purpose of this calculation is not to claim that all these data can consistently be reproduced by two-particle entangled state. Rather, we wish to point out that one can approximate the data unexpectedly closely, so that a serious quantitative test is needed before one can claim that these data confirm three-particle entanglement.

Appendix B

The two “Bell-signals” measured in the experiment of Rauschenbeutel et al. correspond to \( \langle \hat{B}_+ (\phi) \rangle = \text{Tr } \rho \sigma_x^{(1)} \otimes \hat{P}_+^{(2)} \otimes \sigma_\phi^{(3)} \) and \( \langle \hat{B}_- (\phi) \rangle = \text{Tr } \rho \sigma_x^{(1)} \otimes \hat{P}_-^{(2)} \otimes \sigma_\phi^{(3)} \) where \( \hat{P}_\pm \) 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. \( \langle \hat{B}_+ (\phi) \rangle - \langle \hat{B}_- (\phi) \rangle = \text{Tr } \rho \sigma_x^{(1)} \otimes \sigma_x^{(2)} \otimes \sigma_\phi^{(3)} \). 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,...,8. A straightforward calculation yields
\[ \langle \hat{B}_+ (\phi) - \hat{B}_- (\phi) \rangle = 2|\rho_{72}| \cos(\phi + \varphi_{72}) + 2|\rho_{54}| \cos(\phi + \varphi_{54}) + 2|\rho_{36}| \cos(\phi + \varphi_{36}) + 2|\rho_{18}| \cos(\phi + \varphi_{18}) \]
where \( \rho_{72} = \rho_{27} = |\rho_{72}| \exp(i\varphi_{72}) \) and similarly for the other matrix elements.

In a worst case analysis, all the phase factors such as \( \varphi_{72} \) 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 (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 \( \sigma, \tau, \) and
$\nu$ 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. $\omega$ is an arbitrary density matrix, whose off-diagonal matrix elements, however, are assumed to have zero entries where any of the three other states $\sigma, \tau$, and $\nu$ has non-zero 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 $\tau$ and $\nu$, the fractions $\alpha, \beta$ and $\gamma$ also contribute to the populations $\rho_{ii}$ of the total state, whose measured values are collected above in table (25). The maximal values compatible with these measured populations $\rho_{ii}$ 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 $2\rho_{72} = A - w = 0.28 \pm 0.04 - 0.26 \pm 0.03 = 0.02 \pm 0.05$.

References

