Experimental Violation of a Spin-1 Bell Inequality Using Maximally Entangled Four-Photon States

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We demonstrate the experimental violation of a spin-1 Bell inequality. The spin-1 inequality is based on the Clauser, Horne, Shimony, and Holt formalism. For entangled spin-1 particles, the maximum quantum-mechanical prediction is 2.55 as opposed to a maximum of 2, predicted using local hidden variables. We obtained an experimental value of 2.27 ± 0.02 using the four-photon state generated by pulsed, type-II, stimulated parametric down-conversion. This is a violation of the spin-1 Bell inequality by more than 13 standard deviations.

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The assumption of local realism led Einstein, Podolsky, and Rosen (EPR) to argue that quantum mechanics cannot be a complete theory [1]. In 1951 Bohm discussed the system of two spatially separated and entangled spin-$1/2$ particles in order to illustrate the essential features of the EPR paradox [2]. The famous Bell inequalities [3], based on entangled spin-$1/2$ particles, expresses the remarkable fact that whatever additional variables are supplemented to the quantum theory, the conflict between quantum theory and local realism remains. Since the formulation of the Bell inequalities and later of the Clauser, Horne, Shimony, and Holt inequalities [4], several spin-$1/2$ experiments based on polarization-entangled photons [5–9], time-energy entangled photons [10,11], and trapped ions [12] have been performed that verified the quantum-mechanical predictions. The nonlocal features of these entangled states have been employed in several applications in the field of quantum information such as dense coding [13], quantum cryptography [14,15], and quantum teleportation [16].

A natural extension of the research on entangled particles is the study of entangled states of spin-s objects ($s > 1/2$). Gisin and Peres showed that entangled particles with arbitrarily large spins still violated a Bell inequality [17]. This result implies that large quantum numbers are no guarantee of classical behavior. The inequalities that can be derived for entangled spin-s particles are not unique. There are choices one can make in the type of measurements performed on the particles and in the values assigned to the measurement outcomes, leading to different degrees of discrepancy between quantum-mechanical and local-realistic predictions. The question of which choices lead to a maximum discrepancy is hard to answer and only recently progress has been made for certain classes of measurements [18–20]. In this Letter we will consider a spin-1 Bell inequality based on Stern-Gerlach type of measurements and on a most simple value assignment to the measurement outcomes. Under these restrictions there remains a significant discrepancy between the quantum-mechanical predictions, $S = 2.55$, and the local-realistic predictions $S \leq 2$, where S is a function of the correlations between measurement results on the two entangled and spatially separated spin-1 particles.

Apart from its fundamental interest [17,18,21], entangled states of spin-s objects are of clear interest for applications in quantum information due to the higher dimensional Hilbert space associated with these states (e.g., quantum cryptography, dense coding, and bound entanglement [22]). Recently, various alternatives for addressing higher dimensional Hilbert spaces in quantum information science have been explored, for example, by using angular momentum of light [23,24] and multistate BB84 protocols [25,26].

We present the first experimental demonstration of a violation of a Bell inequality for entangled spin-1 objects. We use the fact that the polarization-entangled four-photon fields (two photons in each of two spatial modes) of pulsed parametric down-conversion are formally equivalent to two maximally entangled spin-1 particles [27]. This is related to theoretical work by Drummond [28] in which he describes cooperative emission of wave packets containing n bosons and proves that multiparticle states can violate the Bell inequalities. The connection between states produced in parametric down-conversion and the n-boson multiparticle states has recently been discussed by Reid *et al.* [29].

A spin-1 particle has three distinct basis states ($|-1\rangle$, $|0\rangle$, and $|1\rangle$) The spin-1 analog of Bohm's entangled spin-$1/2$ particles is given by

$$|\Psi_1\rangle = \frac{1}{\sqrt{3}}(|1,-1\rangle - |0,0\rangle + |-1,1\rangle). \qquad (1)$$

Consider the case that one spin-1 particle is sent to Alice who performs a Stern-Gerlach type of measurement (a projection measurement onto the basis states $|-1\rangle$, $|0\rangle$, and $|1\rangle$) along the direction α. The other particle is sent to Bob who performs the same type of measurement along the direction β.

The crux of Bell inequalities is that from a local-realistic point of view the probability P for joint measurement outcomes can be decoupled as

$$P(A, B|\alpha, \beta, \lambda) = P(A|\alpha, \lambda)P(B|\beta, \lambda), \quad (2)$$

where λ accounts for all possible local hidden variables. A and B refer to the measurement results ($|1\rangle$, $|0\rangle$, or $|-1\rangle$) obtained by Alice and Bob using detection orientations α and β, respectively. We define a local-realistic spin-1 measurement combination

$$E^{HV}(\alpha, \beta) = \int d\lambda\, f(\lambda)\overline{A}(\alpha, \lambda)\overline{B}(\beta, \lambda), \quad (3)$$

where

$$\overline{A}(\alpha, \lambda) = P(1|\alpha, \lambda) - P(0|\alpha, \lambda) + P(-1|\alpha, \lambda), \quad (4)$$

$$\overline{B}(\beta, \lambda) = P(1|\beta, \lambda) - P(0|\beta, \lambda) + P(-1|\beta, \lambda), \quad (5)$$

which implies $|\overline{A}(\alpha, \lambda)| \le 1$ and $|\overline{B}(\beta, \lambda)| \le 1$. The derivation of the spin-1 Bell inequality proceeds exactly as the spin- $1/2$ formalism [4,30], leading to

$$S = |E(\alpha, \beta) - E(\alpha, \beta') + E(\alpha', \beta) + E(\alpha', \beta')| \le 2. \quad (6)$$

On the other hand, quantum mechanics predicts that the measurements cannot be decoupled yielding

$$E^{QM}(\alpha, \beta) = P(1,1|\alpha, \beta) - P(1,0|\alpha, \beta) + P(1,-1|\alpha, \beta) - P(0,1|\alpha, \beta) + P(0,0|\alpha, \beta) - P(0,-1|\alpha, \beta) + P(-1,1|\alpha, \beta) - P(-1,0|\alpha, \beta) + P(-1,-1|\alpha, \beta). \quad (7)$$

Using the Bell inequality in Eq. (6), a theoretical maximum violation of 2.55 is achieved, which is in agreement with Gisin and Peres [17]. This prediction was obtained using analyzer rotations of $\alpha = 0°$, $\alpha' = 22.5°$, $\beta = 11.25°$, and $\beta' = 33.75°$. As mentioned in the introduction, in arriving at the spin-1 Bell inequality we restricted ourselves in the following way. First we consider only Stern-Gerlach type of measurements. And second we assigned the value $+1$ to both measurement results $|1\rangle$ and $|-1\rangle$ and the value -1 to measurement result $|0\rangle$. These choices do not maximally profit from the three-dimensionality of spin-1 states, and for recent theoretical progress on optimizing Bell inequalities we refer to Refs. [18–20].

The entangled quanta we use are the multiphoton modes of a polarization-entangled light field [27] produced by pulsed type-II parametric down-conversion. The first order term of parametric down-conversion is $1/\sqrt{2}(|H, V\rangle - |V, H\rangle)$, which is used in spin-1/2 Bell inequality experiments. However, we are interested in the second order term of the down-converted field. By postselection we can measure this term, which is given by

$$\frac{1}{\sqrt{3}}(|2H, 2V\rangle - |HV, VH\rangle + |2V, 2H\rangle), \quad (8)$$

where, for example, the $|2H, 2V\rangle$ means that if Alice measures two horizontal photons, then Bob will measure two vertical photons. As we have shown in [27] this four-photon state is rotationally invariant. The photons sent to Alice (and Bob) have three possible polarization measurement outcomes with equal probabilities, namely $|2H\rangle$, $|HV\rangle$, and $|2V\rangle$, which we will define as the $|1\rangle$, $|0\rangle$, and $|-1\rangle$ state, respectively. This polarization measurement is the analog of a Stern-Gerlach measurement for spin-1 particles. Thus, it is *not* the photons that are the spin-1 particles, but the two-photon polarization-entangled modes.

A schematic of our experimental setup is shown in Fig. 1. The pump laser is a 120 fs pulsed, frequency doubled, Ti:sapphire laser operating at 390 nm with an 80 MHz repetition rate. The pump enters a nonlinear beta-barium borate (BBO) crystal cut for type-II phase matching [7]. The down-converted field is then fed back into the crystal along with the retroreflected pump beam. The difference in the round-trip path length of the pump beam and down-converted field is much smaller than the coherence length of the 5 nm bandwidth frequency filtered down-converted photons. The feedback loop for the entangled fields contains a 2 mm BBO crystal rotated 90° with respect to the optical axis of the down-conversion crystal, which compensates for the temporal walk-off. Such alignment yields very good spatial and temporal overlap with which-pass interference visibilities of 98%.

The primary purpose for using the two-pass scheme is to increase the count rates. For pulsed four-photon down-conversion the count rates increased by a factor of 16 for two passes as opposed to one pass, provided that both down-conversion fields are exactly in phase and completely indistinguishable. This leads to approximately 5 four-photon coincidence detections per second. To perform active stabilization of the phase we use the fact that under the same conditions there is maximum constructive interference for the much more intense two-photon state (singlet spin-$\frac{1}{2}$). Thus the two-photon coincidences can then act as a precision, low-noise four-photon intensity reference.

The analysis setup is shown in the dashed box in Fig. 1. Each analyzer contains a $\lambda/2$ wave plate, a polarizer, a $\lambda/4$ wave plate, a polarizing beam splitter (PBS), narrow bandwidth filters (5 nm), and two single photon detectors. The half-wave plates are used to set α and β on Alice's and Bob's sides. The $|HV\rangle$ state is detected by having the quarter wave plate oriented 0° with respect to the horizontal polarization axis. The photons then pass through the quarter wave plate unaltered and are split up at the PBS. The $|2H\rangle$ state is measured by inserting a linear polarizer oriented such that only horizontally polarized

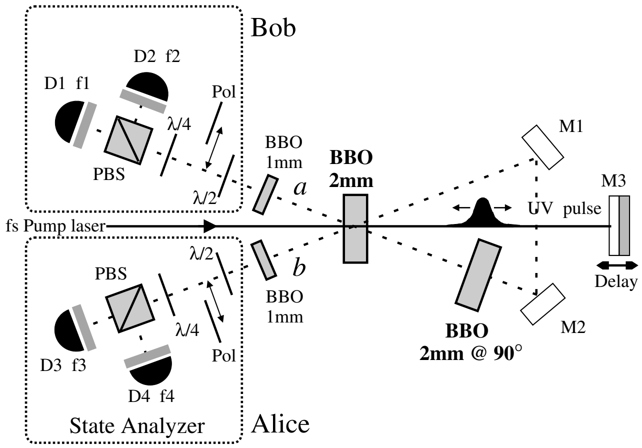

FIG. 1. Experimental setup for generation and detection of entangled spin-1 singlets. A type-II noncollinear parametric down-conversion process creates four-photon states which are amplified by the double pass configuration. The detection is done at Alice's and Bob's sides by postselection as described in the main text.

photons are transmitted. The quarter wave plate rotated by 45° followed by the PBS is an effective 50/50 beam splitter. Thus, the probability for measuring two photons (one in each detector) on Alice's or Bob's side is reduced by a factor of 2 due to the binomial measurement statistics. In addition, inserting a polarizer introduces unavoidable losses in the mode and further reduces the probability of measurement compared to that of the $|HV\rangle$ state. It was experimentally determined that the two-photon measurement probability of the $|2H\rangle$ state was 43.1% on Alice's side and 43.4% on Bob's side compared to 50% for an ideal 50/50 beam splitter and lossless polarizer. Measuring the $|2V\rangle$ is the same as the $|2H\rangle$ except that the polarizer is rotated by 90°.

With the configuration just described, it is necessary to measure 36 probabilities, nine from Eq. (7), for each of the four analyzer settings in Eq. (6). The experimental results for one analyzer setting (namely, $\alpha = -16°$, $\beta' = 14°$) are listed in Table I. The measurements were taken by observing the raw fourfold coincidence counts of all nine measurement possibilities. Each data point is the average over twelve 60 sec intervals. The data obtained using two polarizers were then multiplied by a factor $1/(0.431)(0.434)$. The data obtained using a polarizer on Alice's (Bob's) side were multiplied by a factor of $1/0.431$ ($1/0.434$). These modified data are shown under the "Mod." column and the corresponding probability under "Prob." Similar tables have been measured for the other three analyzer orientations (using $\alpha' = 4°$, $\beta = 6°$). Combining all these data we arrive at a single value, $S = 2.27 \pm 0.02$. This is the primary result of the paper and is more than 13 standard deviations away from the maximum value explainable by local realistic theories, $S = 2$. In the following we take a look at the specific noise in the system which will explain the difference between the ideal prediction ($S = 2.55$) and the measured result ($S = 2.27$).

TABLE I. Experimental results for one setting of the analyzers.

$\alpha = -16°$, $\beta' = 14°$	$\langle$Counts(60 s)$\rangle$	Mod.	Prob.
$P(1,1)$	2.20	11.71	2.25%
$P(1,-1)$	18.04	96.05	18.46%
$P(-1,1)$	17.37	92.48	17.77%
$P(-1,-1)$	1.78	9.48	1.82%
$P(1,0)$	21.92	50.47	9.70%
$P(0,1)$	33.67	77.86	14.96%
$P(-1,0)$	21.43	49.34	9.48%
$P(0,-1)$	28.74	66.46	12.77%
$P(0,0)$	66.50	66.50	12.78%
Total	$\cdots$	520.35	100%

In an ideal experiment, the only relevant experimental setting to obtain a maximum violation is the difference in angles between the analyzer settings ($\Delta\phi = \beta - \alpha = \alpha' - \beta = \beta' - \alpha'$). In our experiment, we observed two primary forms of noise. First, and most important, due to limitations imposed by the broad bandwidth of the 120 fs pump together with imperfections in compensating spatial and temporal walk-off, the stringent indistinguishability conditions for entanglement are not perfectly fulfilled. This leads to two-photon and four-photon contributions that are strongly correlated in the H/V basis, corresponding to $\alpha = \beta = 0$ (the preferred axes of the down-conversion crystal), but uncorrelated when $\alpha = \beta = 45°$. The second, and much smaller contribution which we can neglect in a first approximation, is due to six-photon noise giving rise to four-photon detection events such as $|2H, HV\rangle$. These considerations lead to a simple one-parameter model for the noise of our source

$$\rho = p(|\psi_{\text{pure}}\rangle\langle\psi_{\text{pure}}|) + \frac{(1-p)}{3}(2H, 2V\rangle\langle 2H, 2V| + |HV, VH\rangle\langle HV, VH| + |2V, 2H\rangle\langle 2V, 2H|), \quad (9)$$

where p is the probability of having the pure entangled state. The equal weighting of all three terms of the noise is expected from stimulated emission and has been experimentally verified. The presence of the specific noise in our setup will break the rotational symmetry. Hence, it is advantageous to set our measurement axes $(\alpha, \beta, \alpha', \beta')$ such that they are symmetric around the 0° axis (the orientation of minimum correlation noise). The maximum violation for a given level of noise occurs at a reduced angle difference $\Delta\phi$ compared with the ideal noiseless case. The curves in Fig. 2 are calculated values of S as a function of the angle difference $\Delta\phi$ for various levels of noise. We determined $p = 0.69$ for our experiment by fixing $\alpha = 22.5°$ while varying β from 0° to 45° and looking at the coincidence counts of the $|1,1;1,1\rangle$ term. For this level of noise the maximum value of S equals 2.28 for

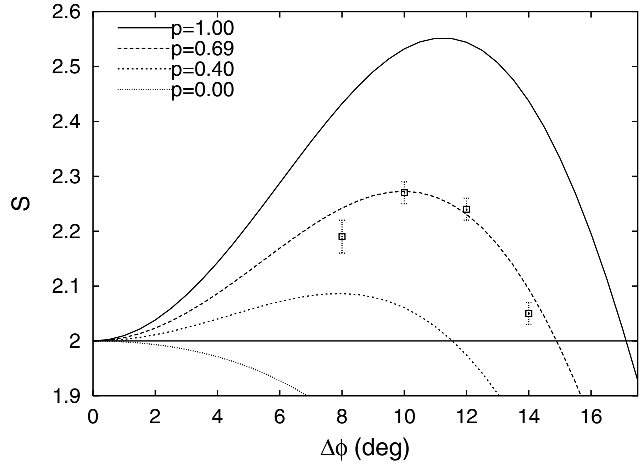

FIG. 2. The value of S is plotted as a function of the angle difference between analyzer axes. The curves correspond to different levels of noise. It was determined experimentally that $p = 0.69$, where p is the probability of having the pure entangled state. The experimental points are shown along with the corresponding theoretical prediction.

$\Delta\phi = 10°$. This is in good agreement with our measured value of $S = 2.27 \pm 0.02$ at $\Delta\phi = 10°$. In order to rule out systematic errors we measured three additional points $\Delta\phi$ along the curve of $p = 0.69$. Each of these also violates the Bell inequality as expected.

In summary, we have reported the experimental violation of a spin-1 Bell inequality. The experimentally determined value was 2.27 ± 0.02 which is in agreement with the value of 2.28 predicted for our system. In principle, the method can be extended to higher spin numbers. These results open up the exploration of spin-1 (and higher) states for optical quantum information.

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