Polarization-Entangled Light Pulses of 10^5 Photons

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We experimentally demonstrate polarization entanglement for squeezed vacuum pulses containing more than 10^5 photons. We also study photon-number entanglement by calculating the Schmidt number and measuring its operational counterpart. Theoretically, our pulses are the more entangled the brighter they are. This promises important applications in quantum technologies, especially photonic quantum gates and quantum memories.

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Entanglement is the signature of the quantum world. One part of an entangled system has its properties fully undefined yet fully correlated with the properties of its counterpart [1]. Can this behavior be observed for large objects? Recently, entanglement was discovered for macroscopic material systems [2,3]. It is tempting to observe it for bright photonic states [4,5], because bright light is much more efficient in interactions than single photons. For bright squeezed vacuum (SV), very different from usual squeezed light, entanglement was discussed theoretically [6–10] but never tested experimentally. Coincidence measurements could only reveal entanglement for up to 12 photons [11].

The clue to the observation of entanglement for bright SV is in registering, instead of single photons and coincidences [9,11,12], fluctuations of macroscopic intensities and measuring the variances of intensity differences [13]. A great advantage of this measurement is that it is robust against the multimode detection. In our experiment, by applying this technique to entangled bright SV, the states of entangled SV, also known as macroscopic Bell states, can be obtained via parametric down-conversion in two type-I nonlinear crystals [13]. For example, the singlet state is generated by the Hamiltonian [8,10,11,13]

$$\hat{H} = i\hbar G(a_H^\dagger b_V^\dagger - a_V^\dagger b_H^\dagger) + \text{H.c.}, \tag{1}$$

where $a^\dagger$ and $b^\dagger$ are photon creation operators in beams A and B, respectively, which in our case have the same direction but different wavelengths λ_A and λ_B. The subscripts H and V stand for the horizontal and vertical polarization, respectively, and the parameter G depends on the crystal properties and the pump power. The state can be written as a Fock-state expansion [10]:

$$|\Psi_{\text{mac}}^{(-)}\rangle = \frac{1}{\cosh^2\Gamma}\sum_{N=0}^{\infty}\sqrt{N+1}\tanh^N\Gamma|\Psi_-^{(N)}\rangle, \tag{2}$$

where $|\Psi_-^{(N)}\rangle \equiv (1/\sqrt{N+1})\frac{1}{N!}(a_H^\dagger b_V^\dagger - a_V^\dagger b_H^\dagger)^N|0\rangle$ and Γ is the parametric gain.

Clearly, photon numbers in beams A and B are exactly the same. Moreover, if polarization beam splitters are placed in the beams, the number of transmitted photons in beam A will be equal to the number of reflected photons in beam B, and vice versa (Fig. 1). Correlations will be maintained at any orientations of the polarizers, or quarter-wave plates in front of them, as long as they are the same for both beams. This is because the operator expression in $|\Psi_-^{(N)}\rangle$ is invariant to polarization transformations. Note that such perfect correlations are manifested only by SV and not by displaced squeezed states, which contain a huge coherent component and only a small part of SV.

Such polarization correlations are similar to the ones manifested by two-photon Bell states but involve far larger photon numbers. Besides their fundamental interest, they are important for quantum information protocols based on light-light (quantum gates) and light-matter (quantum memory) interactions. One can mention quantum memory proposal [14] and up-conversion of such states [15]. The latter suggests a new field of research, nonlinear optics of entangled states.

Separability condition.—How can we prove in experiment that the state (2) is entangled? According to Ref. [10], if a bipartite system containing two macroscopic light beams A and B (Fig. 1) is separable, it satisfies a certain

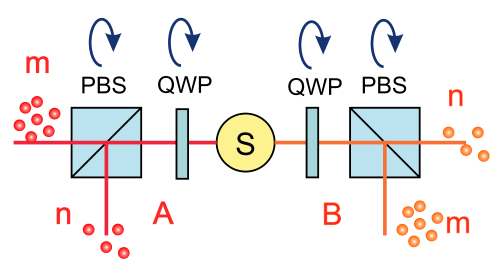

FIG. 1 (color online). Quantum correlations of the macroscopic singlet Bell state. The source S emits light pulses into beams A and B (spatially separated for clarity). In each pulse, photon numbers emitted into any orthogonal polarization modes in the two beams are random but exactly equal.

condition. Violation of this condition indicates that the state is nonseparable (entangled if it is pure).

To simplify comparison with experiment, we derive a necessary condition of separability in terms of the Stokes parameters and their variances [16]. This approach enables us to prove a stronger condition than the one of Ref. [10]. It is important that our consideration is also valid for multimode beams.

As shown in Sec. A of the Supplemental Material [16], for a separable state, the sum of the three Stokes variances ΔS_i^2, $i = 1, 2, 3$, cannot be smaller than twice the total photon number $\langle \hat{S}_0 \rangle$:

$$\sum_{i=1}^{3} \Delta S_i^2 / \langle \hat{S}_0 \rangle \geq 2. \quad (3)$$

A similar inequality was proved for atomic ensembles [17].

Inequality (3) is often mentioned as one of the uncertainty relations in polarization quantum optics (see, for instance, [18,19]). Indeed, it follows directly from the well-known equality for the Stokes operators $\hat{S}_i$ [18], $\hat{S}_1^2 + \hat{S}_2^2 + \hat{S}_3^2 = \hat{S}_0(\hat{S}_0 + 2)$. It should be noted, however, that this operator equality holds true only in the case where, apart from the two polarization modes, the light beam contains only a single frequency and angular mode [20,21]. Thus, inequality (3) is not of general validity. In fact, it is a necessary condition of separability. Its violation indicates that a beam is nonseparable, i.e., is a sufficient condition of nonseparability. As we show below, Eq. (3) is violated in our experiment.

The experiment was performed with the macroscopic singlet Bell state $|\Psi_{\rm mac}^{(-)}\rangle$, similar to the one considered in [8–11]. The setup (Fig. 2) is described in detail in Refs. [13,16].

Theoretically, the singlet state $|\Psi_{\rm mac}^{(-)}\rangle$ has three Stokes parameters equal to zero, $\langle \hat{S}_{1,2,3} \rangle = 0$, as well as the corresponding variances, $\Delta S_{1,2,3}^2 = 0$, and higher-order moments [20]. Thus, in theory condition (3) is always violated, as its left-hand side is zero. In practice, achieving a zero variance of any Stokes observable is impossible. The noise is caused by the inevitable losses (including the nonideal quantum efficiency of the detectors) and the imperfect mode matching. To optimize the mode matching, beams A and B are filtered in the angle separately.

Testing condition (3) requires the measurement of variances for three Stokes observables and the total photon number $\langle \hat{S}_0 \rangle$, which is the shot-noise level. The variances of $S_{1,2,3}$ and the mean photon number $\langle \hat{S}_0 \rangle$ were measured by analyzing the statistics over 20 000 pulses [22]. Typical photon numbers per pulse were 10^5, due to a very large number of modes collected. It is known that collinear type-I phase matching is characterized by a large number of angular Schmidt modes [23]. By accepting, with our angular apertures, nearly whole angular spectra at wavelengths λ_A and λ_B, we collected about 10^4 angular modes

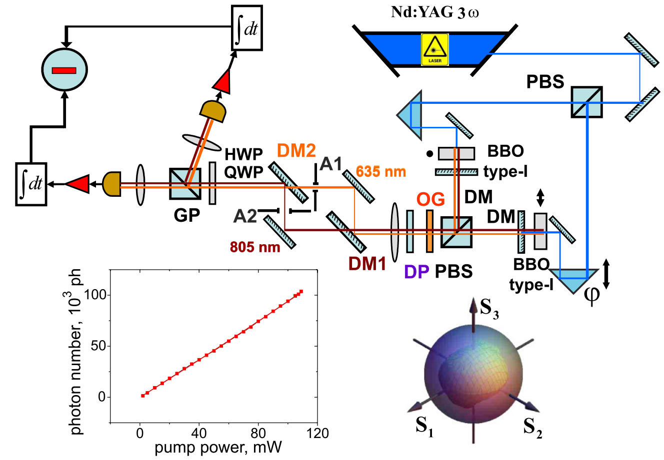

FIG. 2 (color online). Top: The experimental setup. Orthogonally polarized SVs are generated in two β-barium borate (BBO) crystals and overlapped at a polarizing beam splitter (PBS); the pump is eliminated by dichroic mirrors (DM) and a long-pass filter (OG). The interferometer is balanced using trombone prisms. A dichroic plate (DP) is inserted for producing the macroscopic singlet state. Two apertures ($A1$ and $A2$) placed in the focal plane of a lens select the angular spectra of the beams at two wavelengths, separated by dichroic mirror DM1 and joined together by dichroic mirror DM2. The measurement part also includes a Glan prism (GP), a half-wave or quarter-wave plate (HWP and QWP, respectively), and two detectors. Bottom left: Number of photons per pulse versus the pump power. Bottom right: Variance of the Stokes observable versus the direction in 3D (the object inside the sphere, which shows the shot-noise level).

and 10^2 frequency modes [24]. At the same time, the number of photons per mode was mesoscopic. The bottom-left part of Fig. 2 shows the output versus input characteristic of the down-converter. It is almost linear, and the fit yields the maximum gain 0.33 ± 0.06 corresponding to the number of photons per mode 0.12 ± 0.04. However, condition (3) is invariant to the number of modes [16]. This is why it is suitable for testing multimode states; on the contrary, traditional Wigner-function measurement requires single-mode states and is therefore not applicable here. Besides, measurement of the photon-number variance for SV has been shown to be invariant to the gain, at least up to values $\Gamma \sim 2$ [24].

Figure 3 shows the left-hand side of inequality (3) plotted against the diameter of the $A1$ aperture, D_1. As expected, the best noise suppression is observed for D_1 satisfying the mode matching condition $D_1/D_2 = \lambda_A/\lambda_B$ [24]. For all points below the dashed line, the necessary condition of separability is violated. We see that with the transverse modes properly matched, it is violated by more than 5 standard deviations. Similar behavior is observed if D_2 is scanned at fixed D_1.

Hence, we have experimentally demonstrated that the macroscopic singlet Bell state is polarization nonseparable. Note that it is prepared pure, and only imperfections

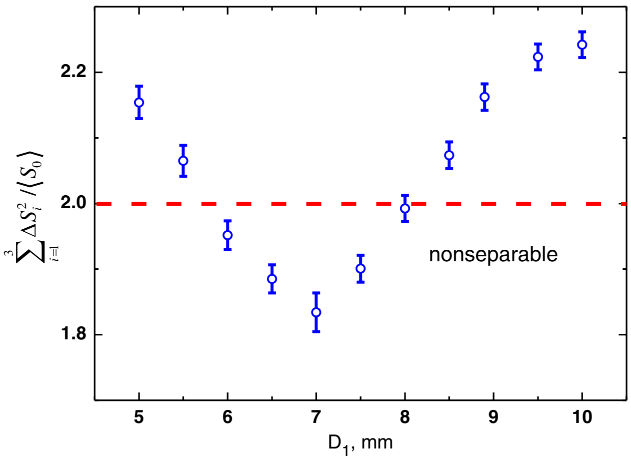

FIG. 3 (color online). The left-hand part of the separability condition (3) versus the diameter of aperture $A1$, the other aperture diameter being 8.9 mm. The dashed line is the boundary set by the separability condition.

of the detection setup make it mix with the vacuum. A pure nonseparable state should be able to violate Bell's inequalities [25], but apparently a new form of Bell's inequality should be derived for this state.

Photon-number entanglement of the singlet Bell state (2) can be characterized by noticing that the state can be rewritten as a product of two Schmidt decompositions in the Fock basis:

$$|\Psi_{\text{mac}}^{(-)}\rangle = |\Psi_1\rangle \bigotimes |\Psi_2\rangle,$$

$$|\Psi_1\rangle \equiv \sum_{n=0}^{\infty} \sqrt{\lambda_n}\, |n\rangle_{AH} |n\rangle_{BV},$$

$$|\Psi_2\rangle \equiv \sum_{m=0}^{\infty} (-1)^m \sqrt{\lambda_m} |m\rangle_{AV} |m\rangle_{BH}.$$

Here, $\lambda_n \equiv \tanh^{2n}\Gamma/\cosh^2\Gamma$, and the notation $|n\rangle_{AH}$ means a Fock state in beam A with n photons in the horizontally polarized mode. The notation for beam B and mode V is similar. Clearly, the state can be represented as a product of two entangled states, one of them involving modes AH and BV and the other one modes AV and BH.

For each of the states $|\Psi_{1,2}\rangle$, the Schmidt number is $K_1 = K_2 = 1 + 2N_0$, where $N_0 \equiv \sinh^2\Gamma$ is the mean photon number. The total Schmidt number is their product. Note that neither of the states $|\Psi_{1,2}\rangle$, taken separately, violates condition (3), although they both manifest photon-number entanglement. This shows that there is a difference between polarization and photon-number entanglement of macroscopic Bell states.

If there are many (M) independent frequency-wave vector mode pairs, each containing a state of the form (2), then the total Schmidt number is given by the product $K = (1 + 2N_0)^{2M}$, which is extremely large. Under certain experimental conditions, this huge amount of entanglement could be used. However, in our experiment we treat the whole ensemble of modes jointly; moreover, the detection scheme also combines the two wavelengths. With the only partition being the polarization one, the Schmidt decomposition for our state can be written as for a two-mode SV, but with λ_n given by the Poissonian distribution [26] with the mean value $N \equiv 2MN_0$. Then the Schmidt number can be calculated as $K = e^{2N}/I_0(2N)$, where I_0 is the zero-order modified Bessel function of the first kind. At large N, $K \approx 2\sqrt{\pi N}$. In our experiment, this yields $K \approx 10^3$. This is much less than in the single-mode case, because we do not address each mode separately and deal only with their ensemble. We stress that the Schmidt number is not an operational measure as it cannot be directly measured in experiment. However, there exists its operational counterpart.

Operational measure.—The Schmidt number can be understood as the effective number of the Schmidt modes. For continuous variables like wave vectors or frequencies of single photons, its operational counterpart was proposed [27] as the ratio of the unconditional width of one subsystem with respect to some parameter to the width of the corresponding conditional distribution.

By analogy, consider the following procedure. In experiment, we obtain the photon-number probability distribution in beam B (unconditional distribution). Next, we measure the photon-number conditional distribution for beam B by postselecting only those pulses for which the photon number in beam A is fixed or within certain narrow bounds. The ratio R of the widths for the two distributions can be considered as a measure of entanglement. Note that the narrowing of the photon-number conditional distribution is typical for twin beams [28] but was never applied to quantify entanglement.

For each pair of correlated modes (for instance, $AH - BV$), the joint probability distribution contains a factor δ_{n_A,n_B} [Fig. 4(a)]. Therefore, while the unconditional distribution for beam B has a negative exponential shape, the conditional probability is only nonzero for a single value of n_B. In the case of many modes with small mean photon numbers, the unconditional distribution is Poissonian [Fig. 4(b)], while the conditional distribution is again of unity width. This gives $R = 2\sqrt{2N\ln 2}$, almost coinciding with the Schmidt number. For a more detailed analysis of R and other measures of entanglement for the state $|\Psi_{\text{mac}}^{(-)}\rangle$, see Ref. [29]. Unfortunately, the conditional distribution gets broadened due to losses, and the resulting R value becomes $R_\eta = 1/\sqrt{1-\eta}$, where η is the overall detection efficiency. Thus, the accessible degree of entanglement is given by only the detection efficiency and turns out to be much less than the Schmidt number. This is an agreement with the fact that the ratio R is applicable only to pure states, while the losses make the state not pure. Another disadvantage of R is that it is not sensitive to the phases contained in the wave function and thus can

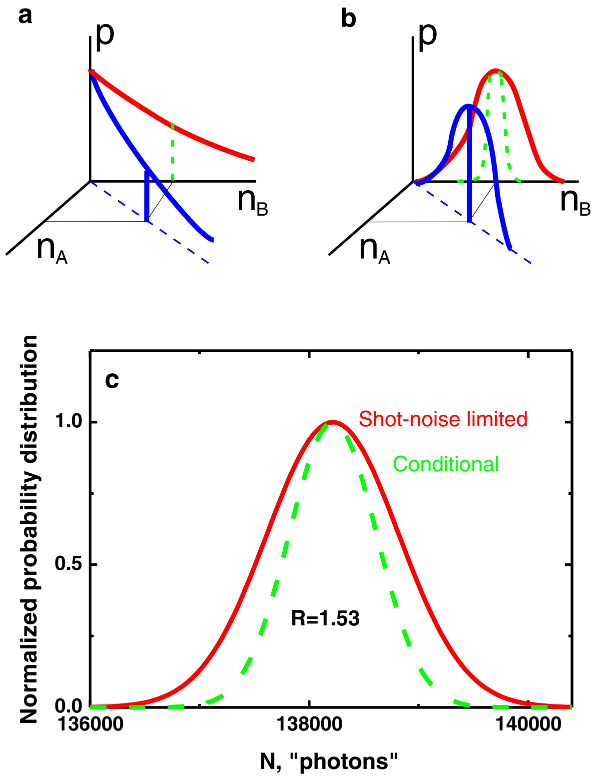

FIG. 4 (color online). Photon-number distributions. (a) Single-mode case: the joint probability distribution (blue solid line in the plane $n_A = n_B$), the unconditional distribution (exponentially decaying red line), and the conditional distribution (green dashed line). (b) The same distributions in the case of a realistic experiment (many modes, nonideal detection). (c) Experimentally obtained distributions: the unconditional one, with the excess noise eliminated (red solid line) and the conditional one (green dashed line). The electronic noise is subtracted.

underestimate the entanglement [30]. However, the state $|\Psi^{(-)}_{\rm mac}\rangle$, in theory, contains no nontrivial phases.

In our experiment, the unconditional distribution was obtained by plotting the photon-number histogram for an ensemble of 20 000 pulses. Because the distribution was broadened due to the excess noise of the pump, we found the numerator of the R ratio by measuring the width of photon-number histogram for a shot-noise limited source with the same mean photon number [Fig. 4(c), red solid line]. The conditional distribution for the $|\Psi^{(-)}_{\rm mac}\rangle$ state was measured by postselecting only those pulses for which the photon number in the idler channel differed from the mean value by not more than 50 and plotting the photon-number histogram in the signal channel [Fig. 4(c), green dashed line]. The degree of entanglement is then measured to be 1.53 ± 0.05. This corresponds to the detection efficiency $\eta = 0.57$.

The triplet states.—We have proved the violation of the separability condition (3) for the macroscopic singlet Bell state. Since the triplet states can be obtained from the singlet one via local unitary transformations, they are entangled as well. The separability conditions for these states can be derived from condition (3) via the corresponding unitary transformations [29].

The Schmidt decomposition for the triplet states is similar to the one for the singlet state, and the Schmidt number is the same. The only difference is that, for the singlet state, the polarization modes can be chosen in any way. For each of the triplet states, there is a unique choice of polarization modes to see entanglement: It should be horizontally and vertically polarized modes for $|\Psi^{(+)}_{\rm mac}\rangle$, diagonally polarized modes for $|\Phi^{(-)}_{\rm mac}\rangle$, and right- and left-circularly polarized modes for $|\Phi^{(+)}_{\rm mac}\rangle$ [13].

In conclusion, we have tested the macroscopic singlet Bell state, containing two beams of different wavelengths, for separability. The results convincingly prove that the state is nonseparable with respect to polarization observables. As the photon numbers per pulse are as high as 10^5, and the state is prepared pure, this can be considered as a proof of macroscopic entanglement. As a measure of photon-number entanglement, we have calculated the Schmidt number, which turned out to be given by the total number of photons in a pulse. Theoretically, our multiphoton state is highly entangled. The entanglement can be confirmed by the measurement of the photon-number distribution for one beam (unconditional distribution) and the distribution conditioned on registering a certain number of photons in the other beam (conditional distribution). As the former is broader than the latter, the state is photon-number entangled. The measured degree of entanglement and separability condition violation are both reduced due to the inefficient detection. However, there is apparently a difference between photon-number and polarization entanglement.

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[16] See Supplemental Material at http://link.aps.org/supplemental/10.1103/PhysRevLett.109.150502. The derivation of the separability condition (3) is given, and the experimental details on the preparation of the singlet macroscopic Bell state are described, including the operation of the dichroic plate.
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