



Experimental Unconditional Preparation and Detection of a Continuous Bound Entangled State of Light

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Among the possibly most intriguing aspects of quantum entanglement is that it comes in free and bound instances. The existence of bound entangled states certifies an intrinsic irreversibility of entanglement in nature and suggests a connection with thermodynamics. In this Letter, we present a first unconditional, continuous-variable preparation and detection of a bound entangled state of light. We use convex optimization to identify regimes rendering its bound character well certifiable, and continuously produce a distributed bound entangled state with an extraordinary and unprecedented significance of more than 10 standard deviations away from both separability and distillability. Our results show that the approach chosen allows for the efficient and precise preparation of multimode entangled states of light with various applications in quantum information, quantum state engineering, and high precision metrology.

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The preparation of complex multimode entangled states of light distributed to two or more parties is a necessary starting point for applications in quantum information processing [1,2] as well as for fundamental physics research. An aggressively pursued example of the latter is the preparation of the bound instance of entanglement, a type of entanglement that can only exist in higher-dimensional or multimode quantum states [3]. Bound entanglement is fundamentally interesting since, in contrast to free entanglement, it can not be distilled to form fewer copies of more strongly entangled pure states [3] by any local device allowed by the rules of quantum mechanics. This irreversible character has triggered entire theoretical research programs [4], in particular, by linking entanglement theory to a thermodynamical picture, with this irreversibility reminiscent of—but being inequivalent with—the second law of thermodynamics [5]. In order to investigate such connections both new theoretical as well as experimental means of constructing multimode states must be innovated.

In recent years, great progress in information processing, metrology and fundamental research has actually been achieved in the photon counting (discrete variable, DV) regime using postselection [1]. States of light are the optimal systems for entanglement distribution because they propagate fast and can preserve their coherence over long distances. *Postselection* means that the measurement outcome of the detectors which characterizes the quantum state is also used to select the state, conditioned on certain measurement outcomes. In such an approach, conditional applications are possible; however, an unconditional application of the states in downstream experiments is

conceptually not possible. Another limitation that any postselected architecture will eventually face is that without challenging prescriptions of measurement, quantum memories, and conditional feedforward, the preparation (postselection) efficiency will exponentially decay with an increasing number of modes. In parallel to postselected architectures of light, unconditional platforms for research in quantum information have been developed which build on the detection of position and momentumlike variables having a continuous spectrum and Gaussian statistics. In such platforms the preparation efficiency of one mode is identical to the preparation efficiency of N modes. In the past, this continuous-variable (CV) platform has been used to demonstrate the Einstein-Podolsky-Rosen paradox [6] and unconditional quantum teleportation [7]. Recently, the CV platform has been extended to investigate multimode entangled states [8]; however, the significance of their nonclassical properties have typically been smaller compared to their postselected counterparts.

In this Letter, we demonstrate the continuous unconditional preparation of one of the rarest types of multimode entangled states—bipartite bound entangled states—using the CV platform. The property of *bound entanglement* is verified by four downstream balanced homodyne detectors with a detection efficiency of almost unity. Alternatively, our setup can make available bound entangled states for any downstream application. The bound entanglement is generated with unprecedented significance, i.e., with state preparation error bars small with respect to the distance to the free entanglement regime and with respect to the distance to the separability regime. Our result is achieved by the convex optimization of state preparation parameters,

and by introducing the experimental techniques of single-sideband quantum state control and classical generation of hot squeezed states.

The first ever generation of bound entangled states was claimed in 2009 [9]. This work used photon counting and postselection; however, the data presented did not support this claim, an issue which has been addressed in a comment, see Ref. [10]. In Ref. [11], a DV nuclear magnetic resonance state whose density matrix has a small contribution of bound entanglement has been observed. Such a state has been called a “pseudobound entangled state.” Very recently, the actual first bound entangled states have been generated in two experiments, both on the basis of discrete variables. In Ref. [12] bipartite bound entangled states of trapped ions have been verified by the unconditional detection of resonance fluorescence. In Ref. [13] the first bound entangled states of light have been generated, albeit of multipartite and not of bipartite nature. Similar to Ref. [9], photon counting and postselection have been used. An unconditional application of the distributed entanglement in a downstream experiment is hence not possible. This is now made possible in our work, with a significance of bound entanglement that has not been achieved using postselection.

Our theoretical search for CV Gaussian bound entangled states of light begins with three (nonpure) squeezed input modes and a vacuum mode overlapped on four beam splitters acting as phase-gates. This yields several independent parameters to be chosen that includes three pairs of quadrature variances and the splitting ratios and the relative phases of the phase gates. Additional vacuum contributions due to optical losses at different locations in the experiment have to be considered as well. As it turns out, bound entanglement is extremely rare in this multidimensional parameter space. Hence, to theoretically identify suitable regimes for experimental certification is a challenging task: Known examples of CV bound entangled states, including those of Ref. [14], will have both free entangled and separable states very nearby. Optimal entanglement witnesses can be efficiently constructed for Gaussian states [15], yet to maximize the distance of an optimal hyperplane separating separable states to the boundary of nondistillable states—hence maximizing robustness of a preparation—is a nonconvex difficult problem. What is more, a reasonable compromise with the preparation complexity has to be found, with a surprisingly simple feasible scheme being shown in Fig. 1.

We now present the measures required for verifying the presence of bound entanglement. Since the studied states are Gaussian they are fully described by their first—which will not play a role here—and second moments, specified by the covariance matrix of a state $\hat{\rho}$ [16–18]. We define a set of quadratures for each optical mode given by $\hat{x}_j = (\hat{a}_j + \hat{a}_j^\dagger)/2^{1/2}$ and $\hat{p}_j = -i(\hat{a}_j - \hat{a}_j^\dagger)/2^{1/2}$ where $\hat{a}_j, \hat{a}_j^\dagger$ are the annihilation and creation operators, respectively.

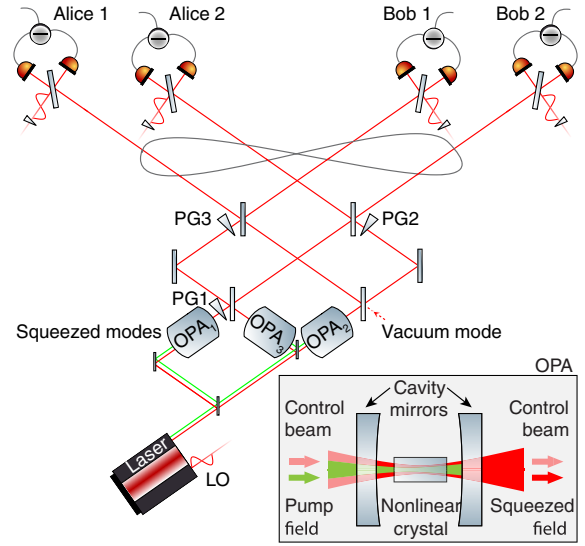


FIG. 1 (color online). Experimental setup: The experiment is composed of three optical parametric amplifiers (OPA_{1-3}), three actively controlled piezo mounted mirrors forming phase gates (PG1 – 3) and four homodyne detectors which are independent of the preparation. The inset shows the construction of an OPA as a nonlinear crystal inside a resonator producing a spatial TEM_{00} (transverse electromagnetic) mode. The bound entangled state is obtained through the bipartite splitting such that Alice and Bob each possess two of the four modes.

Collecting these $2n$ coordinates in a vector $\hat{O} = (\hat{x}_1, \hat{p}_1, \dots, \hat{x}_n, \hat{p}_n)$, we can write the commutation relations as $[\hat{O}_j, \hat{O}_k] = i\sigma_{j,k}$, where $\hbar = 1$ and is a matrix σ often known as a *symplectic matrix*. The second moments are embodied in the $2n \times 2n$ covariance matrix

$$\gamma_{j,k} = 2 \operatorname{Re} \operatorname{tr}[\hat{\rho}(\hat{O}_j - d_j)(\hat{O}_k - d_k)], \quad (1)$$

with $d_j = \operatorname{tr}(\hat{\rho}\hat{O}_j)$, giving rise to a real-valued symmetric matrix γ , see Supplemental Material [20].

Verification of bipartite bound entanglement requires showing that the state is entangled (inseparable) with respect to a bipartition of the modes and that the state remains positive under partial transposition [3,14,19] proving that the state is not distillable. The state is said to be *entangled* if no physical covariance matrices γ_A and γ_B exist of states in modes A and B , respectively, so real matrices satisfying $\gamma_A, \gamma_B \geq -i\sigma$, such that [14] $\gamma \geq \gamma_A \oplus \gamma_B$. This idea suggests a natural entanglement measure [21] for Gaussian states, defined as the solution of

$$E(\gamma) = 1 - \max_{\gamma_A, \gamma_B} x, \quad \gamma \geq \gamma_A \oplus \gamma_B, \quad \gamma_A, \gamma_B \geq -ix\sigma. \quad (2)$$

$E(\gamma) > 0$ indeed implies that the state is entangled related to the entanglement measure negativity [22]. The above problem is known as a semidefinite program, a convex optimization problem that can efficiently be solved.

Nondistillability can be tested by evaluating the partial transposition of a state [3] which physically reflects time reversal. For covariance matrices, partial transposition amounts to changing the sign of momentum coordinates or by applying the operation $\gamma^\Gamma = M\gamma M$, where $M = (1, 1, 1, 1, 1, -1, 1, -1)$, with a -1 in all momentum coordinates belonging to B . A covariance matrix γ is said to be positive partial transpose (PPT) if its partial transpose is again a legitimate covariance matrix, or equivalently, $\gamma^\Gamma + i\sigma \geq 0$. A measure as to the quantitative extent a state is PPT can be taken to be the minimum eigenvalue of this matrix,

$$P(\gamma) = \min \text{eig}(\gamma^\Gamma + i\sigma). \quad (3)$$

The continuity of the eigenvalues with respect to variations in the matrix are enough to guarantee that the measure is meaningful. A strictly positive value of $P(\gamma)$ unambiguously certifies that the state is not distillable.

Based on our theoretical parameter search our final experimental setup is realized as shown in Fig. 1. In total, three optical parameter amplifiers (OPAs), similar to those described in Ref. [23] three phase-gates, consisting of a beam splitter and a piezo mounted mirror, and a vacuum mode are utilized as the base setup. The four homodyne detectors are only necessary for the verification of bound entanglement but not for its preparation. We set our OPAs to produce the minimum and maximum vacuum noise normalized variances to be: (2.0, 3.46) from OPA₁, (0.54, 5.16) from OPA₂ and finally (0.63, 2.54) from OPA₃. The phase gates were set to $\phi_1 = 90^\circ$, $\phi_2 = 41^\circ$, and $\phi_3 = 140^\circ$, respectively. For further details, see Ref. [20].

The first OPA produces a classically squeezed (thermal) state we refer to as *hot squeezing*. It manifests a nonuniform stationary noise distribution amongst its two quadratures without having the smallest quadrature fall below the vacuum noise level. Hot squeezing is generated when, for example, two amplitude squeezed modes of different squeezing factors are overlapped on a 50/50 beam splitter with a relative phase of 90° , thereby producing a two-mode squeezed state, but then one of the output modes is discarded. Without the presence of hot squeezing, we could not find any possibility to produce continuous-variable bound entanglement (between four modes), using the methods described in Ref. [20]. Indeed, the passive optics following the sources can no longer alter the eigenvalues of $\sigma\gamma$, which also define the degrees of squeezing and mixedness of the state. Hot squeezing therefore appears to give rise to a necessary ingredient of quantum and classical correlations in order to create robust bound entangled states. We demonstrate that the same state can also be prepared in a purely classical way by applying a local random displacement on the phase quadrature of a vacuum mode while parametrically amplifying the state's amplitude quadrature. The stationary random phase modulation is produced by using an electro-optical modulator driven

with the output from a homodyne detector measuring shot noise. The amplitude modulation is generated by operating OPA₁ in Fig. 1 in amplification mode, effectively anti-squeezing the amplitude quadrature and deamplifying the thermal noise phase quadrature of the input state. In principle, the random amplitude noise of the first input mode can also be provided by a second homodyne detector and an amplitude modulator, thereby replacing the parametric OPA₁ device. We note that pseudorandom numbers could be insufficient in this scheme since they could introduce artificial correlations and a nonstationary noise into the final state.

In order to hit the tiny regions in parameter space where bound entanglement does exist we introduce to our setup a new technique for precisely controlling phase gates at arbitrary angles. This method relies on an optical single-sideband scheme (see Supplemental Material [20]) that can be used to arbitrarily and independently set the working point of both a phase-gate network and multiple homodyne detectors. This scheme reduces setting the relative phase between interfering modes to selecting the electronic demodulation phase used in the control loop. A portion of the light leaving the phase gates, PG1-3 in Fig. 1, is redirected to control photodetectors. We are able to derive a strong error signal by tapping only $1 \mu\text{W}$ of power corresponding to no more than 1% of the signal mode's optical power. For applications where delicate quantum states must remain free from losses our method provides a means by which they can still be used for controlled interference without significant vacuum contribution due to loss.

The four balanced homodyne detectors are used for the full tomographic reconstruction of the covariance matrix. The results of the reconstruction are used to evaluate two characteristics of the state: namely, its entanglement E (2) and its PPTness P Eq. (3). In order to build the statistics of these characteristics we first continuously recorded 4×10^6 data points from the amplitude and phase quadratures of each mode. Using the bootstrapping method, we then randomly sampled from the total 4×10^6 points, with uniform distribution, points that were different, and produced a series of covariance matrices from which the entanglement, PPT and physical properties were calculated. Our results are represented in Fig. 2 by the black points. The cross corresponds to the average state inferred from the total data set. The abscissa of Fig. 2 is the PPTness and the ordinate the entanglement. By projecting the scatter plot onto the respective axes we calculate a significance of 46σ away from being distillable, i.e., $P(\gamma) < 0$ and 16σ away from being separable, i.e., $E(\gamma) \leq 0$. To demonstrate that the generated state is not close to the boundary of state space (and to confirm its physicality), the smallest eigenvalue of $\gamma + i\sigma$ is also depicted: This is shown in the inset as a histogram. The fact that it is more than 50σ away from being unphysical can be seen as an indication of the fact that our setup was stable over the entire measurement time

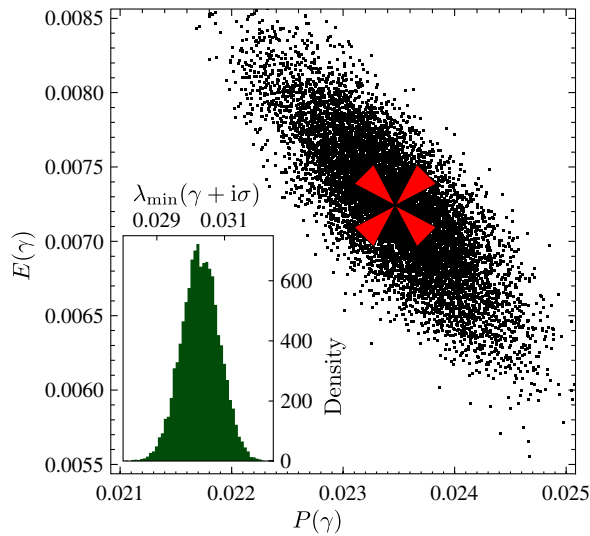


FIG. 2 (color online). Experimental results: The state measured after 4×10^6 sets of raw quadrature data points yields the entanglement E and nondistillability P indicated by the red cross. Other 10^4 points are obtained by bootstrapping the original 4×10^6 data points and show that we are 16σ away from separability and 46σ away from distillability. In the inset we depict the minimum eigenvalue of $\gamma + i\sigma$ of each of the 10^4 bootstrapped correlation matrices, showing that they are significantly far away from the boundary of covariance matrices allowed by the uncertainty principle.

and that our measured data exhibited little statistical uncertainty.

The fact that the involved states are Gaussian to a very high statistical significance is carefully checked by statistical methods (see Supplemental Material [20]): Indeed, we have not only estimated the second moments from the time series data from homodyning measurements, but in fact all moments. Based on these data, we have computed Q - Q -plots for measured distributions against perfectly Gaussian ones, showing a remarkable coincidence and confirming the precisely Gaussian character of the state. What is more, from a bound of the mean energy of the state and of its Gaussian character, one can derive rigorous bounds to the distillable entanglement of the state, confirming an at most negligible amount of distillable entanglement. Again, for details, see the Supplemental Material [20].

Our results present the first unconditional preparation of bound entangled states of a physical system characterized by (continuous) position- or momentum-like variables. With respect to systems composed of light, we demonstrate the first unconditional preparation of bound entanglement, and achieve an unprecedented significance of its features. Independent of any postselection, our platforms allow for the distribution of the entangled states. As with other states of light, our bound entangled states can be distributed to remote parties, which might be kilometers apart using optical fibers [24]. The decoherence on bound entangled

states due to photon loss and phase noise [25] and the ineffectiveness of distillation schemes [26] can be tested, as the applicability of thermodynamical pictures of entanglement can be studied, experimentally.

Our results clearly exemplify the potential of the continuous-variable platform for the precise engineering of complex multimode states of light. We underline that using this platform the state preparation efficiency does not depend on the number of entangled modes. That is to say, detecting, for example, one squeezed mode with one homodyne detector has exactly the same efficiency as detecting N squeezed states with N homodyne detectors simultaneously. Furthermore, we estimate our total quantum detection efficient to be between 90% and 95% being already considered in the preparation of bound entanglement. Alternatively, this loss could be mapped directly onto the measured state by inclusion of neutral density filters, and verification with perfect detectors would reveal the same statistics as depicted in Fig. 2.

We believe that the precise and unconditional preparation of (bipartite) bound entangled states of light demonstrated uplifts the theoretical and experimental research on the link between entanglement theory and statistical physics. From a more general and also technological perspective, the high efficiency and the high degree of control in multimode quantum state preparation achieved certainly promotes the application of the unconditional continuous-variable platform for the preparation of quantum states of light for fundamental research as well as quantum metrology.

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