Preparing the bound instance of quantum entanglement

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Among the possibly most intriguing aspects of quantum entanglement is that it comes in "free" and "bound" instances. Bound entangled states require entangled states in preparation but, once realized, no free entanglement and therefore no pure maximally entangled pairs can be regained. Their existence hence certifies an intrinsic irreversibility of entanglement in nature and suggests a connection with thermodynamics. In this work, we present a first experimental unconditional preparation and detection of a bound entangled state of light. We consider continuous-variable entanglement, use convex optimization to identify regimes rendering its bound character well certifiable, and realize an experiment that continuously produced a distributed bound entangled state with an extraordinary and unprecedented significance of more than ten 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.

The preparation of complex multi-mode entangled states of light distributed to two or more parties is a necessary starting point for applications in quantum information processing [1–5], quantum metrology [6–8] 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 multi-mode quantum states [9]. 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 [9] by any local device allowed by the rules of quantum mechanics. This irreversible character has triggered entire theoretical research programmes [10], in particular by linking entanglement theory to a thermodynamical picture, with this irreversibility reminiscent of—but being inequivalent with—the second law of thermodynamics [11, 12]. In order to investigate such connections both new theoretical as well as experimental means of constructing multi-mode 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 [1–5]. 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 (post-selection) 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 momentum like variables having a continuous spectrum and a 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 (EPR) paradox [13, 14] and unconditional quantum teleportation [15, 16]. Recently, the CV platform has been extended to investigate multimode entangled states [17–22]; however, the significance of their nonclassical properties have typically been smaller compared to their postselected counterparts.

In this work, we demonstrate the continuous unconditional preparation of one of the rarest types of multi-mode 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 [23]. 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. [24]. In ref. [25] 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 "pseudo-bound 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. [26] bipartite bound entangled states of trapped ions have been verified by the unconditional detection of resonance fluorescence. In ref. [27] the first bound entangled states of light have been generated, albeit of multipartite and not of bipartite nature. Similar to ref. [23], 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 (non-pure) 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 multi-dimensional 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. [28], will have both free entangled and separable states very nearby. Optimal entanglement witnesses can be efficiently constructed for Gaussian states [29], yet to maximize the distance of an optimal hyperplane separating separable states to the boundary of non-distillable states—hence maximizing robustness of a preparation—is a non-convex 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 $\hat{\rho}$ [30–32]. 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 annihiliation and creation operators, respectively. Collecting these $2n$ coordinates in a vector $\hat{O} = (\hat{x}_1, \hat{p}_1, \ldots, \hat{x}_n, \hat{p}_n)$, 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 *symplectic matrix*. The second moments are embodied in the $2n \times 2n$ covariance matrix

$$\gamma_{j,k} = 2\mathrm{Re}\,\mathrm{tr}\left(\hat{\rho}(\hat{O}_j - d_j)(\hat{O}_k - d_k)\right), \tag{1}$$

with $d_j = \mathrm{tr}(\rho\hat{O}_j)$, giving rise to a real-valued symmetric matrix γ, see supplementary material.

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 [9, 28] proving that the state is not distillable.

The state is said to be *entangled* if physical covariance matrices γ_A and γ_B exist of states in modes A and B, respectively, so real matrices satisfying $\gamma_A, \gamma_B \geq -\mathrm{i}\sigma$, such that [31, 32]

$$\gamma \geq \gamma_A \oplus \gamma_B. \tag{2}$$

This idea suggests a natural entanglement measure [33] for Gaussian states, defined as the solution of

$$E(\gamma) = 1 - \max_{\gamma_A, \gamma_B} x \tag{3}$$
$$\gamma \geq \gamma_A \oplus \gamma_B, \quad \gamma_A, \gamma_B \geq -\mathrm{i}x\sigma.$$

$E(\gamma) > 0$ indeed implies that the state is entangled. The above problem is known as a semi-definite program, a convex optimization problem that can efficiently be solved.

Non-distillability can be tested by evaluating the partial transposition of a state [34] 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 PPT if its partial transpose is positive, i.e. is again a legitimate covariance matrix, or equivalently, $\gamma^\Gamma + \mathrm{i}\sigma \geq 0$. A measure as to the quantitative extent a state is PPT can be taken to be the minimum eigenvalue of this matrix,

$$P(\gamma) = \min \mathrm{eig}(\gamma^\Gamma + \mathrm{i}\sigma). \tag{4}$$

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.

Finally, we test whether the reconstructed covariance matrix satisfies the Heisenberg uncertainty relation as this is a test if the matrix corresponds to a physical state. (Unphysical states might occur if the error bars of the quantum state preparation or the tomographic characterization are too large.) This is performed by checking that the inequality

$$\gamma + i\sigma \geq 0, \tag{5}$$

is satisfied for the reconstructed state.

RESULTS

Based on our theoretical parameter search our final experimental setup is realized as shown in fig. 1. In total three optical parameter amplifiers (OPAs), 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 OPA_1, (0.54, 5.16) from OPA_2 and finally from OPA_3 (0.63, 2.54). The phase-gates were set to $\phi_1 = 90°$, $\phi_2 = 41°$ and $\phi_3 = 140°$, respectively. The first OPA produces a classically squeezed (thermal) state we refer to as *hot squeezing*. It manifests a non-uniform stationary noise distribution amongst its two quadratures without having the smallest quadrature fall below the vacuum noise level.

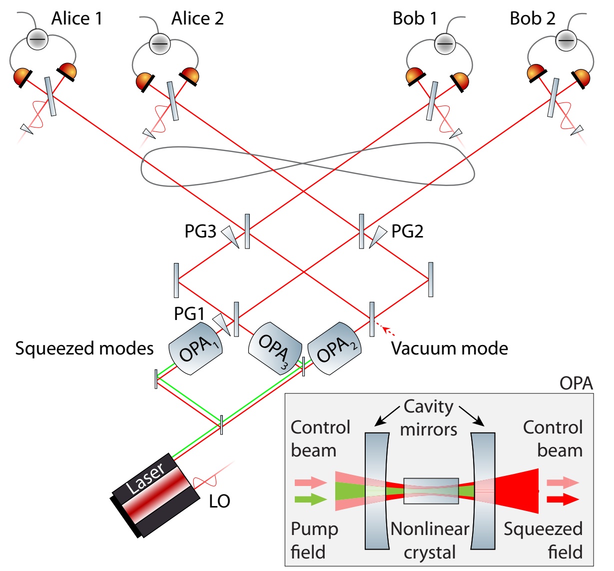

FIG. 1. Experimental setup: The experiment is composed of three optical parametric amplifiers (OPA_{1-3}), three actively controlled piezo mounted mirrors forming phase-gates ($\mathrm{PG}1-3$) and four homodyne detectors which are independent of the preparation. The inset shows the construction of an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

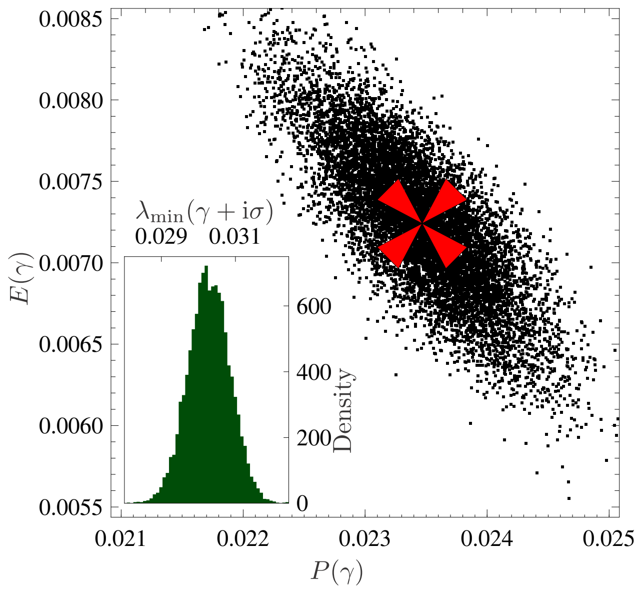

FIG. 2. Experimental results: The state measured after 4 million sets of raw quadrature data points yields the entanglement E and non-distillability P indicated by the red cross. Other 10^4 points are obtained by bootstrapping the original 4 million data points and show that we are 16σ away from separability 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. The fact that the involved states are Gaussian up to the experimental accuracy reached, as can be assessed by estimating higher cumulants.

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 discarded one of the output modes to complete the preparation. Without the presence of hot squeezing, bound entanglement cannot be prepared; it introduces quantum noise giving rise to the subtle interplay of quantum and classical correlations close to the boundary of bound entangled and separable quantum 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 EOM driven with the output from a homodyne detector measuring shot noise. The amplitude modulation is generated by operating OPA_1 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 OPA_1 device. It is important to note that pseudo-random numbers could be insufficient in this scheme since they could introduce artificial correlations and a non-stationary 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 supplementary material) 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 μW 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 eq. (4) and its PPTness P eq. (4). In order to build the statistics of these characteristics we first continuously recorded 4 million data points from the amplitude and phase quadratures of each mode. Using the bootstrapping method, we then randomly sampled from the total 4 million 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 red 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) eq. (5) 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 and that our measured data exhibited little statistical uncertainty.

DISCUSSION

Our results present the first unconditional preparation of bound entangled states of a physical system characterized by (continuous) position/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 allows for the distribution of the entangled states. As other states of light our bound entangled states can be distributed to remote parties, which might be kilometers apart using optical fibers [35]. The decoherence on bound entangled states due to photon loss and phase noise [36] and the ineffectiveness of distillation schemes [37] can be tested, as well as the applicability of thermodynamical pictures of entanglement be studied experimentally.

Our results clearly exemplify the potential of the continuous variable platform for the precise engineering of complex multi-mode 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 90-95% 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 (bi-partite) 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.

METHODS

Details of entanglement criteria

Explicitly, for n modes the *symplectic matrix* σ reads as

$$\sigma = \bigoplus_{j=1}^{n} \begin{pmatrix} 0 & 1 \\ -1 & 0 \end{pmatrix}. \tag{6}$$

The *Heisenberg uncertainty relation*, expressed in terms of the covariance matrix [30], is given by

$$\gamma + \mathrm{i}\sigma \geq 0. \tag{7}$$

Such operator valued inequalities $A \geq B$ for Hermitian A and B always refer to operator ordering, meaning that the real eigenvalues of $A - B$ are non-negative. The above measure E for covariance matrices, eq. (4), indeed indicates entanglement in states [33], and for two modes this is essentially nothing but the familiar *negativity* [38–40].

In the above discussion we show that the spectrum of $\gamma + \mathrm{i}\sigma$ is bounded from below by $\varepsilon > 0$, hence manifesting the Heisenberg uncertainty principle. It is worth mentioning that this also means that the smallest *symplectic eigenvalue* $s_1(\gamma)$ of γ is bounded away from 1.

Identifying robust bound entangled states

The relative volume of bound entangled states compared to all states is very small under every reasonable measure, and any verification as pursued here necessarily requires a careful analysis as to what parameter regime is most suitable. In this subsection, we report techniques that have been used to identify regimes of robust bound entangled states. We explore the space of correlation matrices considering the most general correlation matrix, modulo unitary local operations that do not change the entanglement properties of the system:

In order to find more robust states, we look at all physical covariance matrices, once the irrelevant parameters are taken away. The most general such covariance matrix of 4 modes, up to local unitaries that will not alter any entanglement properties, is of the form

$$\gamma = \begin{pmatrix} \lambda_1 & 0 & 0 & 0 & \lambda_5 & 0 & \lambda_9 & \lambda_{10} \\ 0 & \lambda_1 & 0 & 0 & 0 & \lambda_6 & \lambda_{11} & \lambda_{12} \\ 0 & 0 & \lambda_2 & 0 & \lambda_{13} & \lambda_{14} & \lambda_7 & 0 \\ 0 & 0 & 0 & \lambda_2 & \lambda_{15} & \lambda_{16} & 0 & \lambda_8 \\ \lambda_5 & 0 & \lambda_{13} & \lambda_{15} & \lambda_3 & 0 & 0 & 0 \\ 0 & \lambda_6 & \lambda_{14} & \lambda_{16} & 0 & \lambda_3 & 0 & 0 \\ \lambda_9 & \lambda_{11} & \lambda_7 & 0 & 0 & 0 & \lambda_4 & 0 \\ \lambda_{10} & \lambda_{12} & 0 & \lambda_8 & 0 & 0 & 0 & \lambda_4 \end{pmatrix}. \tag{8}$$

Once a state is obtained we now search for variations in which both $P(\gamma)$ and $E(\gamma)$ increase. Thus, the state after a successful variation will be "significantly bound entangled". This search was performed in a combination of using witnesses [29], accompanied with Monte Carlo sampling and running a semi-definite problem in each step. The most suitable state, as quantified by the biggest value of $\min\{E(\gamma), P(\gamma)\}$, are characterized by an entanglement value of $E(\gamma) = 0.054$ and $P(\gamma) = 0.132$, giving an idea of the limiting values that one can achieve.

However, experimentally it is too expensive to engineer a state with an arbitrary correlation matrix. We thus construct a circuit which, starting from a product of noisy Gaussian single mode states, can produce bound entangled states, but is simple enough to be producible in the lab with available technology. A (non unique) example of such a circuit is plotted in fig. 1. The resulting scheme is a result of a variation within the above parameterized family of circuits, maximizing the statistical significance of being bound entangled by running semi-definite problems in each step. Afterwards we filter the results allowing only those which require achievable values of squeezing at the input and which only require a single mode with hot squeezing, as this is also a precious resource that, at the moment, can only be input in a single mode. Within the resulting states we choose the most robust according to the aforementioned criteria.

Details of the experiment

The three OPAs used to produce the underlying quadrature squeezing at sideband frequency of 6.4 MHz were constructed from a type I non-critically phase-matched $\mathrm{MgO{:}LiNbO_3}$ crystal inside a standing wave resonator, similar to the design that previously has been used in ref. [41]. They were pumped with approximately 100 mW of green light at 532 nm each resulting in a classical gain of about 5. The length of the OPA cavity as well as the phase of the second harmonic pump beam were controlled using radio-frequency modulation/demodulation techniques.

Balanced homodyne detection was performed on each of the four modes in order to reconstruct the 8×8 covariance matrix. The optical local oscillator was filtered through a three mirror ring cavity operated in high finesse mode resulting in a linewidth of 55 kHz. The detector difference currents were electronically mixed with a 6.4 MHz local oscillator and low-pass filtered with a 400 kHz bandwidth. The dark noise separation from shot noise was measured to be more than 10 dB for each detector. The raw data was acquired using a 14 bit National Instruments DAQ-card and in total eight measurement settings including the shot noise measurement were required in order to reconstruct the covariance matrix.

The hot squeezed states were generated by randomly phase modulating the control beam used to set the length of the OPA cavity at the squeezing sideband frequency, 6.4 MHz, and locking the OPA cavity in amplification. This produces phase squeezed states whose smallest quadrature can be controlled by varying the strength of the random noise modulated on the control field and whose amplitude quadrature is controlled by the degree of classical gain.

The single-sideband was generated by overlapping the output of a second laser operating at around 1064 nm with the bright output of OPA1. The beams were phase-locked at a beat frequency of 15 MHz resulting in a field that corresponds to both a phase and amplitude modulation. The beat was detected by directing approximately 1% of the phase-gate outputs to photodetectors placed behind the phase-gates as well as in each homodyne detector. The relative phase between the carriers at both the phase-gates and the homodyne detectors could then be set to an arbitrary phase simply by changing the demodulation phase of the electronic local oscillator. We estimate a phase sensitivity at each phase-gate to be approximately 2 deg.

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