Multistep Two-Copy Distillation of Squeezed States via Two-Photon Subtraction

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(Received 15 June 2022; accepted 5 December 2022; published 29 December 2022)

Squeezed states are nonclassical resources of quantum cryptography and photonic quantum computing. The higher the squeeze factor, the greater the quantum advantage. Limitations are set by the effective nonlinearity of the pumped medium and energy loss on the squeezed states produced. Here, we experimentally analyze for the first time the multistep distillation of squeezed states that in the ideal case can approach an infinite squeeze factor. Heralded by the probabilistic subtraction of two photons, the first step increased our squeezing from 2.4 to 2.8 dB. The second step was a two-copy Gaussification, which we emulated. For this, we simultaneously measured orthogonal quadratures of the distilled state and found by probabilistic postprocessing an enhancement from 2.8 to 3.4 dB. Our new approach is able to increase the squeeze factor beyond the limit set by the effective nonlinearity of the pumped medium.

DOI: 10.1103/PhysRevLett.129.273604

Squeezed states of light exhibit Gaussian uncertainties of the electromagnetic field, some of whose variance is smaller than that of the ground state [1–4]. They enable continuous-variable quantum teleportation [5,6], the sensitivity enhancements of atomic spectroscopy [7], and gravitational wave detectors beyond photon shot noise [8–10], and they are the basic resource of one-sided device-independent quantum key distribution (QKD) [11] and optical quantum computing [12]. Squeezed states of light can be deterministically generated by a nonlinear process. The most efficient approach uses resonator-enhanced parametric down-conversion pumped by conventional laser light [13,14]. Squeezing the variance of the photon shot noise by factors larger than 10 (10 dB) at some near infrared wavelengths can be realized [15–17]. At other wavelengths or in other systems, the available nonlinearities are much smaller and limit the squeeze factors. Recently, squeeze factors of (just) 1.1 and 1.3 were realized ponderomotively by levitated nanoparticles in free-space optical tweezers [18,19].

Gaussian squeezing is an irreducible resource [20]; any combination of interference in passive linear interferometers, homodyne detection, and feed-forward cannot distill from an ensemble of arbitrary size another (smaller) one with an enhanced squeeze factor [21]. This no-go theorem is similar to the one about entanglement distillation of Gaussian-squeezed two-mode states with local Gaussian operations and classical communication [22–24]. So far, just the de-Gaussifying photon subtraction was realized to work around this no-go theorem, which resulted in non-Gaussian entanglement or two-mode squeezing [25–28]. Also certain types of non-Gaussian squeezed states were distilled solely by Gaussification [29,30]. It is now of fundamental interest to prove that the combination of these methods is efficient enough to outperform the Gaussian squeeze factor limitation imposed by the available nonlinearity.

Here, we analyze multistep distillation of Gaussian squeezed states and experimentally prove the principle of enhancing squeezing beyond the nonlinearity limit. The successful first distillation step is heralded by the probabilistic coincidence of two subtracted photons. Theoretically, we find that an infinite number of additional two-copy Gaussification and distillation steps produces an infinitely large Gaussian squeeze factor, if and only if the initial state is pure and squeezed by just 3 dB. This condition is not fulfilled in our experiment, nevertheless, we realize a second step to demonstrate further distillation and Gaussification. For this we split the heralded distilled states and simultaneously measure the squeezed and antisqueezed quadrature fields $\hat{X}^Q$ and $\hat{Y}^Q$. The data provide all the information of the first step distilled state and allow us to emulate two-copy Gaussification and distillation by probabilistic data postprocessing. The emulation saves us from additional hardware. The second step can be repeated arbitrary times depending only on the amount of sampled data. Our emulation approach is a rigorous, data-based quantum simulation of optical state processing.

Squeezed-state distillation.—Let us consider an ensemble of a pure single-mode squeezed vacuum state $|\psi(r)\rangle$ with a squeeze factor $\beta = e^{2r} > 1$ as a canonical example, where r is the squeeze parameter [1]. The state

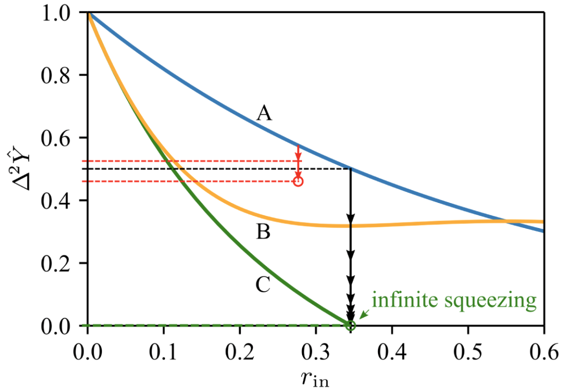

FIG. 1. Potential of our distillation protocol. Variances of squeezed quadrature uncertainties $\Delta^2\hat{Y}$ as a function of the squeeze parameter $r_{\rm in}$ before distillation. A: Initial, *pure* Gaussian squeezed vacuum state. B: Two-photon subtracted non-Gaussian squeezed vacuum state. C: Gaussian squeezed vacuum state in the asymptotic limit of multistep distillation Gaussification. For an initially pure 3 dB squeezed state ($r_{\rm in} = r_{\rm th} \approx 0.3466$), $\Delta^2\hat{Y} \to 0$. The Gaussification converges only for $r < r_{\rm th}$, where curve C ends. Note, our experiment (red arrows) started from a slightly *mixed* Gaussian state.

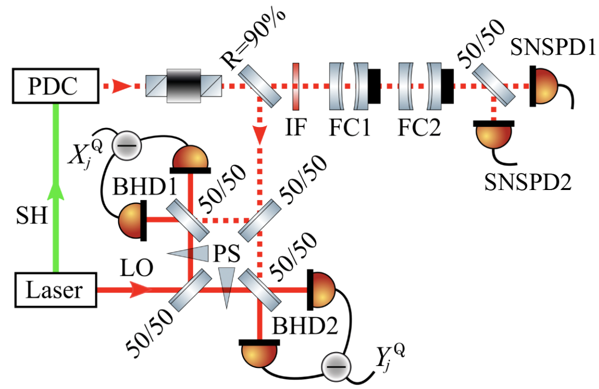

FIG. 2. Optical setup. Resonator-enhanced parametric down-conversion (PDC) produced a beam of subsequent modes in identical squeezed vacuum states. 10% of the states' energy was tapped and distributed onto two superconducting nanowire single-photon detectors (SNSPD1,2). 90% of the optical energy was also split and absorbed by two balanced homodyne detectors (BHD1,2) that simultaneously measured the quadratures $\hat{X}^Q$ and $\hat{Y}^Q$. The subscript "Q" indicates data taken on halves of the beam. An interference filter (IF) and two optical filter cavities of different lengths (FC1,2) rejected the optical spectrum outside the BHD bandwidth. LO: continuous-wave local oscillator (1064 nm); PS: phase shifter; SH: second-harmonic pump field (532 nm).

$|\psi(r)\rangle$ is a minimum uncertainty state with Gaussian Wigner function and quadrature variances $\Delta^2\hat{X} = e^{2r}$ and $\Delta^2\hat{Y} = e^{-2r}$, and it can be written as a superposition of the ground state $|0\rangle$ and even number states $|2n\rangle$ (Fock states) with monotonically decreasing probability amplitudes as n increases [2]. "Distillation" describes the process of creating a smaller ensemble of states with a larger squeeze factor $\beta' > \beta$ by selecting only some of the initial states, conditioned on the successful subtraction of two photons. The subtraction of two photons preserves the structure of the state in the Fock basis and it enhances the amplitude of the two-photon state with respect to the amplitude of the vacuum state. The (nonnormalized) state after subtraction of two photons $|\psi_{2S}(r)\rangle = \hat{a}^2|\psi(r)\rangle$ reads

$$|\psi_{2S}(r)\rangle = \frac{\tanh r}{\sqrt{\cosh(r)}}\sum_{n=0}^{\infty}(2n+1)(\tanh r)^n\frac{\sqrt{(2n)!}}{2^n n!}|2n\rangle.$$

This state is non-Gaussian and well approximates an "even" Schrödinger-cat-like state formed by the superposition of two displaced coherent (squeezed) states [31,32]. The variance of the squeezed quadrature of the non-Gaussian state $|\psi_{2S}(r)\rangle$ can be analytically expressed,

$$\Delta^2\hat{Y}^B = e^{-2r}\left[1 - 4\frac{\sinh r\cosh r - 2\sinh^2 r}{2\sinh^2 r + \cosh^2 r}\right],$$

where "B" refers to curve B in Fig. 1 with $r = r_{\rm in}$. We find that the subtraction of two photons enhances the squeezing if and only if $\tanh(r) < 1/2$ ($\beta < 3$), i.e., only for moderate squeeze factors, see Fig. 1 for $r_{\rm in} \approx 0.55$. Since $|\psi_{2S}(r)\rangle$ is non-Gaussian, its squeezing can be further enhanced by an iterative Gaussification procedure [33,34], where two copies of the state are overlapped at a balanced beam splitter, and one output is accepted as distilled if the other output mode is projected onto the vacuum state. The distilled output then forms the input for another iteration of the Gaussification [33,34]. The squeeze parameter r_{2SG} of an asymptotic Gaussian state obtained by iterative Gaussification of the state $|\psi_{2S}(r)\rangle$ is given by $\tanh(r_{2SG}) = 3\tanh(r)$. The Gaussification of $|\psi_{2S}(r)\rangle$ is beneficial if the initial squeezing is not too small, because for $r \ll 1$ the two-photon subtracted state $|\psi_{2S}(r)\rangle$ remains close to a Gaussian state. The Gaussification converges only for $\tanh(r) < 1/3$ ($\beta < 2$), and when $\tanh(r) \to 1/3$, arbitrarily strong squeezing can be distilled in principle from the initially close to 3 dB-squeezed state, where the decibel scale is given by $10\,{\rm dB}\log_{10}\beta$. This is illustrated in Fig. 1. Not illustrated is the fact that all other input squeeze strengths can also be distilled to arbitrarily strong squeezing, if the two-photon subtraction is combined with appropriate coherent displacements [35].

Experimental.—Figure 2 shows the schematic of the optical setup. The master laser was a continuous-wave Nd:YAG laser that provided an ultrastable light beam of up to 2 W at 1064 nm in the TEM00 mode. Most of this light was frequency doubled and used to pump resonator-enhanced, type I degenerate parametric down-conversion (PDC) process below the oscillation threshold. The nonlinear material inside the resonator was periodically poled $KTiOPO_4$. The PDC resonator had a HWHM linewidth of about 0.05 GHz and produced a continuous stream of squeezed vacuum states in a TEM 00 beam with a squeeze factor of up to $\beta = 10$ (10 dB) with a free spectral range of 4.6 GHz. For the experiments here, we reduced the pump power, limiting the effective nonlinearity and producing a 2.4 dB squeezed state with barely measurable mixedness [17]. The squeezed vacuum beam was split with a power ratio of 90/10. The "signal" beam (higher fraction) was measured with a pair of balanced homodyne detectors (BHD) with quantum efficiencies above 98%. One balanced homodyne detector continuously measured the squeezed quadrature $\hat{Y}^Q$ and the other one *simultaneously* the anti-squeezed quadrature $\hat{X}^Q$ on halves of the beam. Each pair $(X_j^Q; Y_j^Q)$ corresponded to a point in two-dimensional (2D) phase space, where the quasiprobability density function is the Husimi Q function [36]. This type of detection has been called "8-port homodyne detection" or "heterodyne detection." We introduce here the term "simultaneous 2D BHD" because we consider it more descriptive and less ambiguous than previous terms.

The "trigger" beam (10% fraction) was spectrally filtered by an interference filter with a transmission peak at 1064 nm and a FWHM of 0.64 nm and subsequently by two length-controlled Fabry-Perot resonators [37]. They had large free spectral ranges of 56.8 and 47.8 GHz and FWHM linewidths of 181 and 153 MHz to filter out all squeezed field components that were outside the ≈200 MHz HWHM detection bandwidth of the BHDs. The filtered beam was distributed to two superconducting nanowire single-photon detectors with quantum efficiencies greater than 93% (SNSPD1,2). The data of the BHDs was only analyzed, when both of the SNSPDs detected a photon within a window of 6.4 ns. In this case, the signal beam contained a mode in a squeezed vacuum state of which two photons were subtracted. We determined the FWHM width of the temporal mode $f(t)$ to about 12 ns, see Fig. 1 of Supplemental Material [38].

Results and discussion.—We performed the entire distillation protocol twice on two different days to show reproducibility. In both runs, we recorded 1.65×10^6 two-photon subtraction events. For each event j we have simultaneously sampled time-resolved quadrature values $\hat{X}_j^Q(t)$ and $\hat{Y}_j^Q(t)$ within a 64 ns long time window centered on the subtraction event. Figure 3 shows the two pairs of time-resolved variances $\Delta^2\hat{X}^Q(t) = (\Delta^2\hat{X}(t) + 1)/2$ and $\Delta^2\hat{Y}^Q(t) = (\Delta^2\hat{Y}(t) + 1)/2$, where $\Delta^2\hat{X}(t)$ and $\Delta^2\hat{Y}(t)$ denote the corresponding variances of the signal beam before it was split for simultaneous 2D BHD. (The factor of 1/2 is due to the vacuum uncertainty entering the open

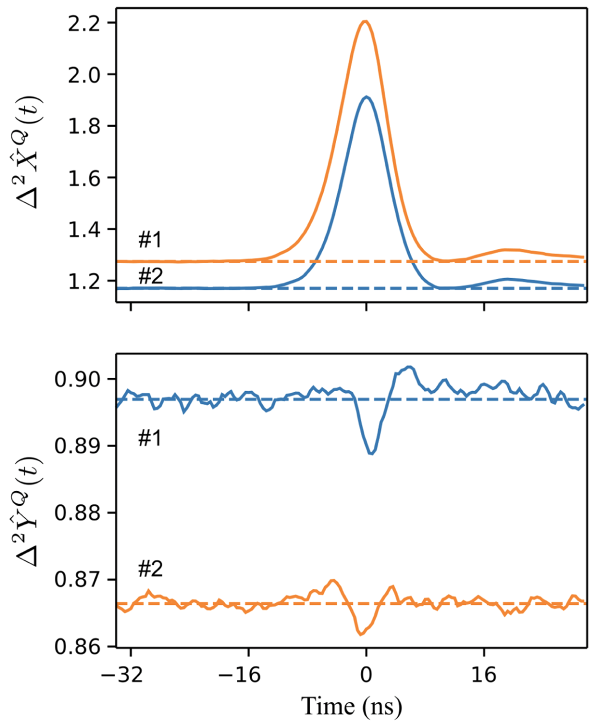

FIG. 3. Variances after two subtracted photons. Shown are the results of two independent measurement runs with slightly different initial squeeze factors. The quadrature variances $\Delta^2\hat{X}^Q(t)$ (top, from BHD1) and $\Delta^2\hat{Y}^Q(t)$ (bottom, from BHD2) include the time when both SNSPDs clicked, to which both x axes are referenced to $(t = 0)$. The traces are calculated from 10^6 individual measurements on halves of the beam and represent data from which the Husimi Q function [36] can be calculated. Around $t = 0$, the antisqueezing as well the squeezing are enhanced, which represents the distillation success of the first step of our protocol. Note that the data include frequencies outside the bandwidth of the squeezing resonator. This dilutes the actual squeeze factor from $\Delta^2\hat{Y}^Q(t) \approx 0.79$ (2.4 dB) to about 0.866 in #2.

port.) At the times around successful two-photon subtraction, which we deliberately set to zero in Fig. 3, the antisqueezed variance increased (upper plot) and the squeezed variance got reduced (lower plot). The observed dip represents the direct experimental manifestation of squeezing enhancement via two-photon subtraction. Variances sufficiently far away from the time of photon subtraction can be approximated by horizontal lines, which represent the levels of antisqueezing and squeezing without photon subtraction, respectively.

The temporal shape $f(t)$ of the mode that contained the two-photon subtracted state was extracted from the temporal covariance matrix of the antisqueezed quadrature [39]. We took into account the nontrivial structure of the covariance matrix for vacuum input which is related to the response function of our detector, see Supplemental Material [38] for details. The recorded

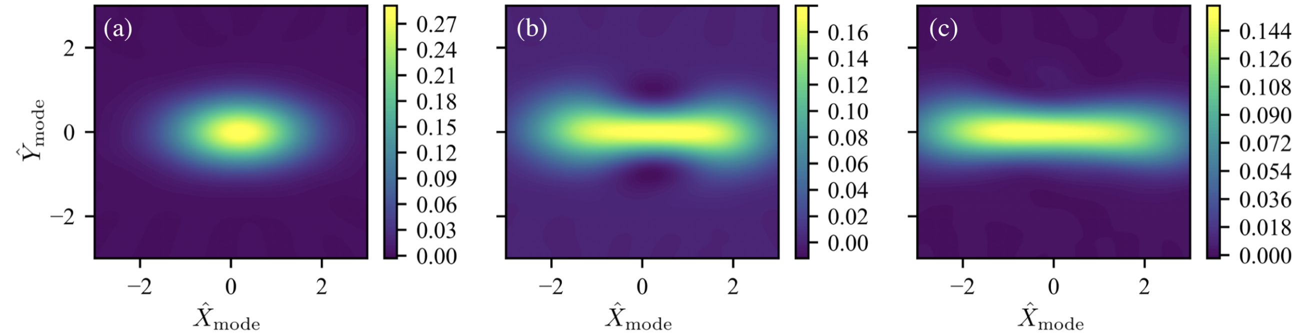

FIG. 4. Reconstructed Wigner functions. (a) The initial 2.4 dB-squeezed vacuum state. (b) The initial state distilled by the subtraction of two photons yielding 2.8 dB squeezing. (c) Example of the subsequently two-copy-two-step-distilled and Gaussified squeezed vacuum state for $\bar{n} = 1.3$ having 3.14 dB squeezing, see Fig. 5. The Gaussification step reduced the ensemble size to the fraction $P_{\rm svv} = 0.246$. Additional two-copy distillation steps are possible in principle if the amount of samples is sufficiently high. The "mode" is defined by the temporal shape $f(t)$ and its Fourier transform limited spectrum.

windows were weighted by $f(t)$ and integrated over time. The results were 1.65×10^6 quadrature pairs $\hat{X}^Q_{\rm mode}$ and $\hat{Y}^Q_{\rm mode}$ for each of the two runs, representing results of simultaneous 2D BHD on identical modes with the temporal profile $f(t)$. The same procedure was applied to characterize the same mode in a vacuum state to yield the quadrature variances for shot-noise normalization. The two-dimensional histogram of the measurement outcomes $\alpha = X^Q_{\rm mode} + iY^Q_{\rm mode}$ corresponded to the Husimi Q function, which completely characterizes the measured state. From this, we reconstructed the density matrix in Fock basis using the statistically motivated and robust maximum-likelihood reconstruction algorithm [40,41], see Supplemental Material [38] for details.

Figure 4 shows Wigner functions [42] of states of the mode with temporal profile $f(t)$ calculated from the reconstructed density matrices. It represents the result of our work. Panel *A* shows the initial Gaussian squeezed vacuum state before photon subtraction with a squeeze factor of 2.4 dB. Panel *B* shows the two-photon subtracted state of the same mode. It is clearly non-Gaussian. The squeeze factor increased to 2.8 dB. We also determined the quadrature variances directly from the measured quadratures and found excellent agreement. Since we used simultaneous 2D BHD, the full phase space data were recorded for each individual copy. This enabled us to emulate the second distillation step. We calculated the interference of two measurement outcomes α_{2j} and α_{2j+1}, where $1 \leq j \leq 8.25 \times 10^5$, at a balanced beam splitter by $\alpha_{j+} = (\alpha_{2j} + \alpha_{2j+1})/\sqrt{2}$ and $\alpha_{j-} = (\alpha_{2j} - \alpha_{2j+1})/\sqrt{2}$. If the amplitude in the constructively interfering output by chance obeys $|\alpha_+|^2 < \bar{n}$, where $\bar{n}$ is a freely choosable hard boundary, a state with improved squeezing emerges in the destructively interfering output port. The complex amplitude α_{j-} represents further distilled, partly Gaussified states. Figure 4(c) shows the result after one such additional Gaussification step with $\bar{n} = 1.3$. The squeeze factor is increased to 3.14 dB. Our emulated two-copy distillation is as efficient as the hardware-based version with *perfect* quantum memories at hand [43].

Figure 5 shows the improvement of the squeezed variance $\Delta^2\hat{Y}$ versus the success probability $P_{\rm svv}$ (the survival rate) in our second step Gaussification protocol. Since two input copies produce only one output copy, we have $P_{\rm svv} \leq 0.5$, and the lower the threshold $\bar{n}$, the more the probability of success is further reduced. At 25%, squeezing exceeds 3.1 dB. Convergence of the iterative Gaussification protocol can be analyzed for Gaussian acceptance probability $P(\alpha) = \exp(-|\alpha|^2/\bar{n})$ [44], see Supplemental Material [38]. For the condition in Fig. 4(b) we find convergence for $\bar{n} > 0.3$, with the maximum distillable squeeze factor of 4.6 dB. However, a single step can only be analyzed numerically, and our emulation, the results of which are shown in Fig. 5, provides an accurate characterization of the practical performance of the protocol.

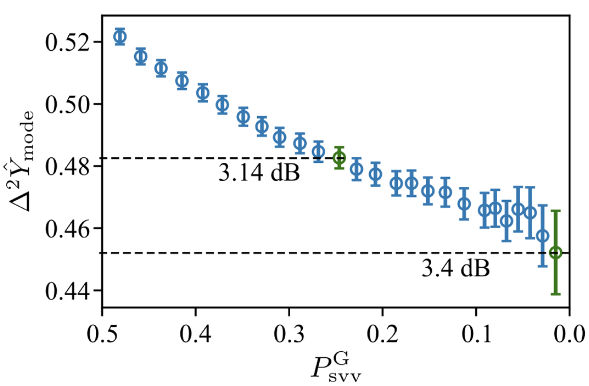

FIG. 5. Distilled squeezed variance as in Fig. 4(c) versus success probability of the Gaussification step $P^{\rm G}_{\rm svv}$. Our best result of about 0.453 (≈ -3.4 dB) occurs when the ensemble size decreases most.

Conclusion.—Distillation of the pure 3 dB squeezed Gaussian state can theoretically approximate the ideal, pure, and infinitely squeezed Gaussian state by following a two-photon subtraction with an infinite number of two-copy Gaussification and distillation steps. In our experiment, the distillation of an initially nearly pure Gaussian state at 2.4 dB was realized by two-photon subtraction followed by a single two-copy Gaussification and distillation step. We exceed the relevant threshold of 3 dB, which would allow us, for example, to overcome the no-cloning limit in quantum teleportation [6].

We achieved the two-copy Gaussification and distillation step by postprocessing subsequently measured Q-function data. The result is indistinguishable from the hardware approach that even uses *perfect* quantum memories [43]. Our "emulated" approach can in principle be applied to any quantum protocol where the intended distillation by Gaussification is directly followed by heterodyne detection. Then it has only advantages, such as efficient scalability to further two-copy Gaussification and distillation steps.

Our work addressed the situation where the maximum squeeze factor was limited by too low an effective nonlinearity of the source. The initial squeeze factor in our experiment was artificially reduced by an intentionally decreased pump power. In practice, there are situations where high pump powers must actually be avoided because otherwise the nonlinear medium is destroyed, for example, at shorter wavelengths [45]. Avoiding enhancement cavities [18,19] also leads to low effective nonlinearities, but in return to a wider squeeze bandwidth; it also reduces optical losses, which in combination with our approach can lead to higher squeeze factors. Another useful scenario is when suboptimal parameters of the source [46] cannot be easily improved, for instance, if the source is used for satellite-based QKD and thus located on a satellite.

It can be shown that photon subtraction followed by Gaussification can enhance the squeeze factor but it cannot counteract losses in quantum communication [35,47], and more advanced techniques are required to distill purified entangled states [47–49]. But we note that a pure single-mode squeezed state can be distilled from a large class of mixed states if the state is de-Gaussified by the operation $\hat{n} - 1$, which removes the single-photon term, followed by Gaussification. A detailed analysis of this procedure will be provided in future work [35].

This work was funded by the Deutsche Forschungsgemeinschaft (DFG, German Research Foundation)—SCHN 757/7-1. J. F. acknowledges support by the Czech Science Foundation under Grant No. 21-23120S.

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