

Quantification of Quantum Correlations in Two-Beam Gaussian States Using Photon-Number Measurements

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Identification, and subsequent quantification of quantum correlations, is critical for understanding, controlling, and engineering quantum devices and processes. We derive and implement a general method to quantify various forms of quantum correlations using solely the experimental intensity moments up to the fourth order. This is possible as these moments allow for an exact determination of the global and marginal impurities of two-beam Gaussian fields. This leads to the determination of steering, tight lower and upper bounds for the negativity, and the Kullback-Leibler divergence used as a quantifier of state nonseparability. The principal squeezing variances are determined as well using the intensity moments. The approach is demonstrated on the experimental twin beams with increasing intensity and the squeezed super-Gaussian beams composed of photon pairs. Our method is readily applicable to multibeam Gaussian fields to characterize their quantum correlations.

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Quantum theory allows for correlations between spatially separated systems or degrees of freedom that are fundamentally different from their classical counterparts. For composite systems, quantum correlations (QCs) manifest themselves in many different (inequivalent) forms [1–4], including the Bell nonlocality, quantum steering, and entanglement. They can be exploited to achieve qualitatively better performance in information processing tasks compared to purely classical scenarios. Though the question of which kind of QCs is a necessary resource for a given quantum information protocol remains still open, particularly when multipart systems are considered, the analysis of QCs among different subsystems is extraordinarily important [5,6]. It belongs to fundamental problems in quantum information science and quantum many-body physics at present. Applying very general quantum theories and models, the structure of QCs is elucidated and nontrivial limitations on the strength of physically allowed QCs are revealed.

Over the years, several methods for the analysis of QCs have been proposed based on, e.g., the violation of various inequalities [7–10], geometrical considerations [11], or even interference among multiple copies of the investigated state [12–14]. However, such methods usually require certain initial knowledge about the analyzed state density matrix, which is never experimentally acquired without technical difficulties. Similarly, the homodyne tomography [15] in quantum optical experiments with continuous-variable states, which provides the complete characterization of the detected field, has to rely on a coherent local oscillator with the varying phase [16]. Contrary to this, we routinely measure the photocount distributions in numerous

experiments by photon-number-resolving detectors [17]. So it is very important to develop methods allowing to extract the maximum information about QCs in the analyzed field using just these photoncount distributions. While there are numerous nonclassicality witnesses at our disposal [18–20], revealing the structure of QCs represents a much more complicated task. Several schemes for solving this problem have already been suggested using, however, some form of homodyning of the analyzed field [21–23]. Interestingly, an exact copy of the analyzed state can be used in a suitable interferometric setup [13] instead of a local coherent oscillator of the homodyne scheme to reveal all four invariants of a two-beam Gaussian field, which is equivalent to the determination of all elements of its covariance matrix.

In this Letter, we address this problem for a wide group of in-practice important Gaussian fields. We propose a scheme based solely on the intensity moments of optical fields to estimate their global and marginal purities. Although the photocount measurements do not allow for the determination of all coefficients characterizing such Gaussian fields (information about the phases of complex coefficients is missing), the needed information can partly be inferred from the values of higher-order intensity moments [24]. In our scheme, both the global and marginal field purities are expressed in terms of higher-order intensity moments. This opens the door for direct determination of important QC quantifiers, involving the Rényi-2 entropy, Kullback-Leibler divergence, one- and two-way Gaussian steering, and even tight lower and upper bounds for the logarithmic negativity. Also, the squeezing of Gaussian states can be determined. In practice, direct measurements of the intensity

moments of optical fields are standardly performed using various types of photon-number-resolving detectors including intensified CCD cameras [25,26] or superconducting bolometers [27], to name a few. This makes our scheme qualitatively simpler compared to those based on various forms of homodyning. Using two-beam states originated in parametric down-conversion, we experimentally demonstrate the suggested approach.

Purity estimation of Gaussian fields.—We begin with defining the normal characteristic function $C_{\mathcal{N}}(\beta_1, \beta_2)$ [24] for general single-mode two-beam fields,

$$C_{\mathcal{N}}(\beta_1, \beta_2) = \left\langle \exp \left[\sum_{j=1,2} \beta_j \hat{a}_j^\dagger \right] \exp \left[- \sum_{j=1,2} \beta_j^* \hat{a}_j \right] \right\rangle, \quad (1)$$

where \hat{a}_j (\hat{a}_j^\dagger) stands for the annihilation (creation) operator of beam j , $j = 1, 2$, and $\langle \cdots \rangle$ denotes quantum-mechanical averaging. For quantum Gaussian fields the characteristic function $C_{\mathcal{N}}(\beta_1, \beta_2)$ takes the form [24]

$$C_{\mathcal{N}}(\beta_1, \beta_2, \beta_1^*, \beta_2^*) = \exp \left[- \sum_{j=1,2} [B_j |\beta_j|^2 + (C_j \beta_j^{*2}/2 + \text{c.c.})] + (D_{12} \beta_1^* \beta_2^* + \bar{D}_{12} \beta_1 \beta_2 + \text{c.c.}) \right] \quad (2)$$

with real (B_j) and complex (C_j , D_{jk} , \bar{D}_{jk}) parameters characterizing the Gaussian state; c.c. replaces the complex-conjugated terms.

The measured intensity moments $\langle W_1^k W_2^l \rangle$, given as the normally ordered photon-number moments and obtained via the Stirling numbers from the measured photon-number moments [24], contain partial information about the state parameters:

$$\langle W_1^k W_2^l \rangle = (-1)^{k+l} \frac{\partial^{2(k+l)} C_{\mathcal{N}}(\beta_1, \beta_2, \beta_1^*, \beta_2^*)}{\partial^k \beta_1 \partial^k (\beta_1^*) \partial^l \beta_2 \partial^l (\beta_2^*)} \Big|_{\beta_1 = \dots = \beta_2^* = 0}. \quad (3)$$

Considering the intensity moments up to the second order, we reveal the following relations for the looked-for parameters:

$$\begin{aligned} B_j &= \langle W_j \rangle, \\ |C_j|^2 &= \langle W_j^2 \rangle - 2\langle W_j \rangle^2, \quad j = 1, 2, \\ |D_{12}|^2 + |\bar{D}_{12}|^2 &= \langle W_1 W_2 \rangle - \langle W_1 \rangle \langle W_2 \rangle. \end{aligned} \quad (4)$$

The third-order intensity moments give us additional information about the looked-for parameters:

$$\begin{aligned} -4\Re\{C_1 \bar{D}_{12} D_{12}^*\} &= -4\langle W_1 \rangle (\langle W_1 W_2 \rangle - \langle W_1 \rangle \langle W_2 \rangle) \\ &\quad + \langle W_1^2 W_2 \rangle - \langle W_1^2 \rangle \langle W_2 \rangle, \\ -4\Re\{C_2 \bar{D}_{12} D_{12}\} &= -4\langle W_2 \rangle (\langle W_1 W_2 \rangle - \langle W_1 \rangle \langle W_2 \rangle) \\ &\quad + \langle W_1 W_2^2 \rangle - \langle W_1 \rangle \langle W_2^2 \rangle. \end{aligned} \quad (5)$$

We note that, alternatively, the expressions in Eq. (5) can be obtained from the third-order moments $\langle W_j^3 \rangle$, $j = 1, 2$ or even fourth-order moments $\langle W_1^3 W_2 \rangle$ and $\langle W_1 W_2^3 \rangle$. Finally, also the fourth-order intensity moment $\langle W_1^2 W_2^2 \rangle$ reveals a useful relation among the looked-for parameters,

$$\begin{aligned} &4(2|D_{12}|^2 |\bar{D}_{12}|^2 + \Re\{C_1 C_2 \bar{D}_{12}^2\} + \Re\{C_1 C_2 (D_{12}^*)^2\}) \\ &= \langle W_1^2 W_2^2 \rangle - 4\langle W_1 W_2 \rangle^2 - \langle W_1 \rangle \langle W_2 \rangle \langle W_1 W_2 \rangle + \langle W_1 \rangle^2 \langle W_2 \rangle^2 \\ &\quad - 2[\langle W_1 \rangle^2 (\langle W_2^2 \rangle - 2\langle W_2 \rangle^2) + \langle W_2 \rangle^2 (\langle W_1^2 \rangle - 2\langle W_1 \rangle^2)] \\ &\quad - (\langle W_2^2 \rangle - 2\langle W_2 \rangle^2) (\langle W_1^2 \rangle - 2\langle W_1 \rangle^2) \\ &\quad + 16\langle W_2 \rangle \Re\{C_1 \bar{D}_{12} D_{12}^*\} + 16\langle W_1 \rangle \Re\{C_2 \bar{D}_{12} D_{12}\}. \end{aligned} \quad (6)$$

Surprisingly, Eqs. (4)–(6) are sufficient to obtain the global and marginal purities of the two-beam Gaussian fields. In order to show that, we need to know the corresponding covariance matrix $\sigma \equiv \langle \hat{\xi} \hat{\xi}^T \rangle - \langle \hat{\xi} \rangle \langle \hat{\xi}^T \rangle$ containing the second-order moments of the position $\hat{x}_j = (\hat{a}_j + \hat{a}_j^\dagger)/2$ and momentum $\hat{p}_j = (\hat{a}_j - \hat{a}_j^\dagger)/(2i)$ operators embedded in vector $\hat{\xi}^T = (\hat{x}_1, \hat{p}_1, \hat{x}_2, \hat{p}_2)$. Using the parameters of the normal characteristic function $C_{\mathcal{N}}$ in Eq. (2), the covariance matrix σ is expressed as

$$\sigma = \begin{bmatrix} \sigma_1 & \gamma \\ \gamma^T & \sigma_2 \end{bmatrix}, \quad (7)$$

where

$$\begin{aligned} \sigma_j &= \begin{bmatrix} 1 + 2B_j + 2\Re\{C_j\} & 2\Im\{C_j\} \\ 2\Im\{C_j\} & 1 + 2B_j - 2\Re\{C_j\} \end{bmatrix}, \\ \gamma &= \begin{bmatrix} 2\Re\{D_{12} - \bar{D}_{12}\} & 2\Im\{D_{12} - \bar{D}_{12}\} \\ 2\Im\{D_{12} + \bar{D}_{12}\} & -2\Re\{D_{12} + \bar{D}_{12}\} \end{bmatrix}. \end{aligned} \quad (8)$$

Now, one can easily verify that the determinants of the global σ and local σ_j , $j = 1, 2$, covariance matrices are given in terms of the intensity moments as

$$\begin{aligned} \det \sigma &= 1 + 4(\langle W_1 \rangle + \langle W_2 \rangle) + 12(\langle W_1 \rangle + \langle W_2 \rangle)^2 - 4\langle W_1^2 \rangle (1 + 6\langle W_2 \rangle + 24\langle W_2 \rangle^2) - 4\langle W_2^2 \rangle (1 + 6\langle W_1 \rangle + 24\langle W_1 \rangle^2) \\ &\quad + 8\langle W_1^2 W_2 \rangle (1 + 6\langle W_2 \rangle) + 8\langle W_1 W_2^2 \rangle (1 + 6\langle W_1 \rangle) - 8\langle W_1 W_2 \rangle (1 + 6\langle W_1 \rangle + 6\langle W_2 \rangle) + 48\langle W_1 \rangle \langle W_2 \rangle \\ &\quad + 96\langle W_1 \rangle \langle W_2 \rangle (\langle W_1 \rangle + \langle W_2 \rangle) + 5\langle W_1 \rangle \langle W_2 \rangle + 24\langle W_1^2 \rangle \langle W_2^2 \rangle - 8\langle W_1^2 W_2^2 \rangle + 48\langle W_1 W_2 \rangle^2, \end{aligned} \quad (9)$$

$$\det \sigma_j = 1 + 4\langle W_j \rangle + 12\langle W_j \rangle^2 - 4\langle W_j^2 \rangle, \quad j = 1, 2. \quad (10)$$

Knowing these determinants, the corresponding purities $\mu = 1/(\det \sigma)^{1/2}$ and $\mu_j = 1/(\det \sigma_j)^{1/2}$ are established [1]. Contrary to this, seralian Δ , the last of four global invariants of two-beam Gaussian states, requires γ to be determined, $\Delta = \det \sigma_1 + \det \sigma_2 + 2 \det \gamma$ [3,28].

This central result allows us to determine various quantities that characterize the structure of two-beam QCs [29–32]. Using the purities μ and μ_j , $j = 1, 2$, we immediately obtain the Rényi-2 entropies along the formula $S_R = -\ln(\mu)$ [31]. We note that S_R represents the continuous analog of the Shannon entropy. The Rényi-2 entropy S_R can then be used to quantify the total quadrature correlations via the Kullback-Leibler divergence (distance) H between the analyzed two-beam state $\hat{\rho}$ and its factorized counterpart $\text{Tr}_2\{\hat{\rho}\}\text{Tr}_1\{\hat{\rho}\}$ [31]:

$$H = S_{R,1} + S_{R,2} - S_R = \ln\left(\frac{\mu}{\mu_1\mu_2}\right). \quad (11)$$

Also the degree of (one-way) Gaussian steering of beam 2 by beam 1 [30] is expressed in terms of purities [32]:

$$G_{1 \rightarrow 2} = \max\{0, \ln(\mu/\mu_1)\}. \quad (12)$$

We note that two-way steering occurs provided that both $G_{1 \rightarrow 2}$ and $G_{2 \rightarrow 1}$ are nonzero.

Using purities, even the logarithmic negativity E_N [29], giving the degree of entanglement revealed, e.g., by the Simon criterion [33], is established through its tight lower and upper bounds derived by Adesso *et al.* [34]:

$$\begin{aligned} E_{\max}(\rho) &= -\frac{1}{2} \ln \left[-\frac{1}{\mu} + \left(\frac{\mu_1 + \mu_2}{2\mu_1^2\mu_2^2} \right) \right. \\ &\quad \left. \times \left(\mu_1 + \mu_2 - \sqrt{(\mu_1 + \mu_2)^2 - \frac{4\mu_1^2\mu_2^2}{\mu}} \right) \right], \\ E_{\min}(\rho) &= -\frac{1}{2} \ln \left[\frac{1}{\mu_1^2} + \frac{1}{\mu_2^2} - \frac{1}{2\mu^2} - \frac{1}{2} \right. \\ &\quad \left. - \sqrt{\left(\frac{1}{\mu_1^2} + \frac{1}{\mu_2^2} - \frac{1}{2\mu^2} - \frac{1}{2} \right)^2 - \frac{1}{\mu^2}} \right]. \quad (13) \end{aligned}$$

Application to experimental data.—We tested the derived formulas on a set of the experimental spatio-spectrally multimode twin beams (TWBs) [35] with increasing intensity [for mean photon number $\langle n_1 \rangle$ of beam 1, see Fig. 1(a)] that were obtained by adding the photocounts registered by two single-photon counting modules positioned in the signal (1) and idler (2) beams in subsequent detection windows (for details, see Supplemental Material [36] and [37]). The TWBs at 710 nm originated in type-I parametric down-conversion in a LiIO₃ nonlinear crystal pumped by the third harmonic of an Nd-YAG laser at 355 nm. We arrived this way at the compound multimode

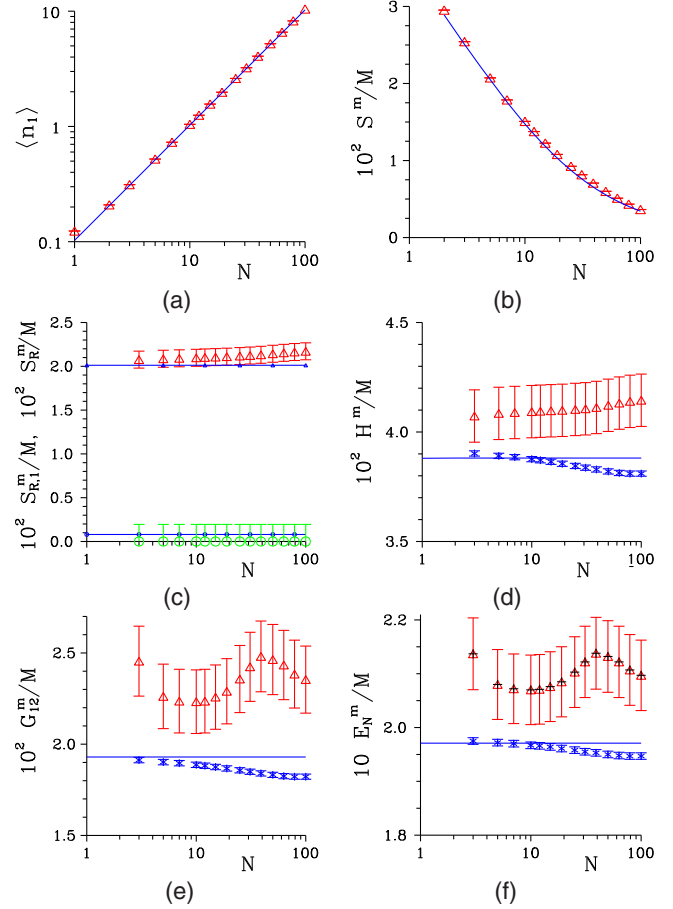


FIG. 1. (a) Mean photon number $\langle n_1 \rangle$ of beam 1 (red Δ). (b) Entropy S^m/M per mode (red Δ). (c) Rényi-2 entropies per mode $S_{R,1}^m/M$ and S_R^m/M for beam 1 (red Δ) and beams 12 (green \circ), respectively. (d) Kullback-Leibler divergence per mode H^m/M and its values for noisy TWBs (blue $*$). (e) Steering parameter per mode G_{12}^m/M (red Δ) and its values for noisy TWBs (blue $*$). (f) Lower (red Δ) and upper (black $+$) bounds for negativity per mode E_N^m/M and its values for noisy TWBs (blue $*$) as they depend on the number N of grouped detection windows; $M = 10N$ (for details, see Refs. [36,37]). In (a) and (b) error bars are smaller than the plotted symbols. In (f), red Δ and black $+$ nearly coincide. The solid blue curves originate in a model of M identical independent single-mode two-beam Gaussian fields with suitable parameters.

TWBs with mean photon numbers extending over 2 orders in magnitude (from 0.1 to 10 mean photons per beam). The experimental photocount histograms were reconstructed by the maximum-likelihood approach to obtain the joint photon-number distribution $p(n_1, n_2)$ and its photon number moments $\langle n_1^k n_2^l \rangle_m = \sum_{n_1, n_2=0}^{\infty} n_1^k n_2^l p(n_1, n_2)$. Also entropy S of the fields was determined along the formula $S = -\sum_{n_1, n_2=0}^{\infty} p(n_1, n_2) \ln[p(n_1, n_2)]$ and plotted in Fig. 1(b). The intensity moments, that are the normally ordered photon-number moments, were then derived as linear combinations of photon-number moments using the Stirling numbers of the first kind [18].

We note that, when correcting the experimental data for nonunit detection efficiencies η_1 and η_2 , the use of any reconstruction method is not required. We may simply determine the intensity moments $\langle W_1^k W_2^l \rangle_E$ directly from the detected photocount histogram and derive the needed intensity moments as $\langle W_1^k W_2^l \rangle = \langle W_1^k W_2^l \rangle_E / (\eta_1^k \eta_2^l)$. On the other hand, reconstruction methods allow us to correct also for other detector parameters like dark-count rates, cross talk, etc. In real experiments, sufficiently large detection efficiencies are needed to arrive at acceptably low errors in the determination of the reconstructed intensity moments.

These experimental beams are multimode as they are generated in parametric down-conversion in the running-wave configuration and detected in multiple detection windows. They also suffer from imperfections that occur both during the generation process and transmission to the detectors. As a consequence, they decline from the theoretically expected form of a multimode noisy TWB whose coefficients obey $C_1 = C_2 = \bar{D}_{12} = 0$ in Eq. (2). These declinations can be quantified using the derived formulas for the tested quantum information quantities. However, these quantities are derived for two single-mode Gaussian beams and so their application is conditioned by the reduction of the experimental multimode photon-number moments $\langle n_1^k n_2^l \rangle_m$ to one typical mode in each beam. Estimating the number M of effective modes in each beam [20,38], we may follow the procedure outlined in the Supplemental Material [36].

We compare the values of the obtained quantities per mode with those characterizing a single-mode Gaussian noisy TWB [39], that is fully characterized by three first- and second-order photon-number moments ($B_j = \langle w_j \rangle = \langle n_j \rangle$, $|D_{12}|^2 = \langle w_1 w_2 \rangle - \langle w_1 \rangle \langle w_2 \rangle = \langle n_1 n_2 \rangle - \langle n_1 \rangle \langle n_2 \rangle$). For the formulas, see Supplemental Material [36].

According to the experimental results reduced to one mode and plotted in Figs. 1(c)–1(f), the Rényi-2 entropies S_R^m/M , the Kullback-Leibler divergence H^m/M , the negativity E_N^m/M as well as the steering parameter $G_{1 \rightarrow 2}^m/M$ do not considerably change with the increasing field intensity, i.e., the increasing number N of grouped detection windows ($M = 10N$).

As the values of Rényi-2 entropy S_R^m of the two-beam fields are smaller than the entropies $S_{R,1}^m$ and $S_{R,2}^m$ of the constituting signal and idler beams [see Fig. 1(c)], the purities of the two-beam fields are greater than those of the constituting beams. This implies, according to the general classification of two-beam Gaussian states (see Table I in [40]), that the analyzed two-beam fields are entangled.

The experimental values of H_R^m/M , $G_{1 \rightarrow 2}^m/M$, and E_N^m/M reduced per one mode and determined by the derived formulas (11)–(13) are systematically greater (by approximately 10%–20%) than those characterizing the Gaussian noisy TWBs (determined by the formulas in the Supplemental Material [36]). This means that the states

of the detected two-beam fields are more general than those of the Gaussian noisy TWBs with the vanishing coefficients C_1 , C_2 , and \bar{D}_{12} . The consideration of the experimental third- and fourth-order intensity moments reveals that also the complex parameters (C_1 , C_2 , and \bar{D}_{12}) of the detected two-beam Gaussian fields are nonzero, which leads, according to our results, to stronger QCs described by the above quantities. We note here, that our results do not allow us to judge the declination (non-Gaussianity) of the analyzed state from the general form of Gaussian states as described by the characteristic function in Eq. (2).

The Kullback-Leibler divergence H_R^m , the steering parameter $G_{1 \rightarrow 2}^m$, and the negativity E_N^m of the two-beam fields increase practically linearly with the increasing TWB intensity. This contrasts with the behavior of the entropy S^m of the two-beam fields whose increase is smaller: The entropy S^m/M per mode plotted in Fig. 1(b) decreases with the increasing TWB intensity. This means that the capacity of available QCs increases linearly with the dimensionality (number of modes M) of the analyzed fields. The capacity of QCs thus grows faster than disorder in the analyzed fields quantified by the entropy S^m .

We note that nonzero negativity E_N^m and the Kullback-Leibler divergence H_R^m are obtained also directly for the experimental photocount histograms, i.e., without reconstructing the experimental data. This contrasts with the steering parameters $G_{1 \rightarrow 2}^m$ and $G_{2 \rightarrow 1}^m$ being zero in this case.

The negativity E_N is the most commonly used parameter to quantify QCs. However, we have only the lower and upper bounds in Eq. (13) at disposal for single-mode two-beam Gaussian fields. Nevertheless, the experimental data plotted in Fig. 1(f) show that these bounds are very close to each other: The uncertainty in determining E_N is practically given only by the experimental errors. This observation is valid in general. Indeed, from the point of view of the entanglement, two-beam Gaussian states are divided into groups of states with identical amount of E_N . These groups are parameterized by four parameters: purities μ , μ_1 , μ_2 , and serialian Δ . The minimal and maximal values of the negativity E_N given in Eq. (13) in fact represent the extremal values with respect to serialian Δ for fixed values of the purities. The general behavior of these extremal values can conveniently be quantified taking into account that the negativity E_N increases (decreases) with the increasing global purity μ (marginal purities μ_1 and μ_2). This suggests the ratios μ/μ_j , $j = 1, 2$, as suitable parameters for quantifying the uncertainty in the determination of E_N . The greater the ratios are, the greater the negativity E_N is. This is documented in the graph of Fig. 2(a) where the negativity E_N^{av} averaged over the states with fixed ratios μ/μ_1 and μ/μ_2 is plotted. The maximum of the relative error δE_N^{max} [34],

$$\delta E_N = \frac{E_N^{\text{max}} - E_N^{\text{min}}}{E_N^{\text{max}} + E_N^{\text{min}}}, \quad (14)$$

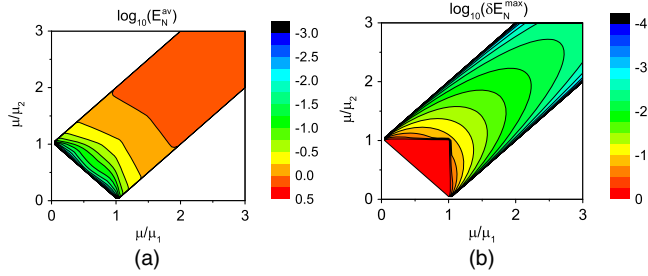


FIG. 2. (a) Average negativity E_N^{av} and (b) maximum δE_N^{max} of relative error as they depend on ratios μ/μ_1 and μ/μ_2 . In the white areas no entangled states exist. For $\mu_1 = \mu_2$, see also in [32].

taken over the states with the fixed ratios μ/μ_1 and μ/μ_2 is then shown in Fig. 2(b). According to Fig. 2(b), the relative error δE_N is smaller than 10% (1%) when the ratios μ/μ_j are greater than 1.5 (2.5), i.e., when the states exhibit considerable entanglement. This makes the use of the bounds for negativity E_N very efficient.

Single beam properties.—The approach presented above for two-beam Gaussian fields is applicable also to single-beam Gaussian fields. The intensity moments allow us to determine the principal squeezing variance [41,42] in this case. Merging the intensities of the signal and idler beams of the above discussed two-beam fields, we arrive at single-beam super-Gaussian fields with phase fluctuations reduced below the shot-noise limit, as discussed in detail in the Supplemental Material [36].

Further extension and application.—Our results also allow for the analysis of QCs of general n -beam Gaussian states. This is so as the appropriate covariance matrix is composed of blocks of 2×2 matrices similar to those written in Eq. (7) [43,44]. This allows us to analyze its properties by considering all possible two-beam subsystems of the whole n -beam Gaussian field. The formulas in Eqs. (9) and (10) hold for such subsystems and allow us to quantify QCs in these two-beam reductions. Relying on various monogamy relations, we may establish the lower bound for the genuine multipartite QCs in the whole n -beam field [31,45,46].

In *conclusion*, we have shown how various forms of quantum correlations of two-beam Gaussian fields (with spatio-spectral multimode structure), that naturally depend on the fields phase properties, can be quantified solely from the measured intensity moments up to the fourth order. The determination of the global and marginal purities of the involved beams in terms of the intensity moments represents the key step. The principal squeezing variances can solely be derived from the intensity moments as well. We have demonstrated usefulness and practicality of this approach by considering suitable experimental fields. Our method is readily applicable to multipartite systems for the detection and characterization of their quantum correlations. As the Gaussian states are exploited in numerous metrology applications and quantum-information protocols with continuous

variables, we foresee numerous applications of the suggested and demonstrated method in the near future.

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