Quantum Correlations beyond Entanglement and Discord

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Dissimilar notions of quantum correlations have been established, each being motivated through particular applications in quantum information science and each competing for being recognized as the most relevant measure of quantumness. In this contribution, we experimentally realize a form of quantum correlation that exists even in the absence of entanglement and discord. We certify the presence of such quantum correlations via negativities in the regularized two-mode Glauber-Sudarshan function. Our data show compatibility with an incoherent mixture of orthonormal photon-number states, ruling out quantum coherence and other kinds of quantum resources. By construction, the quantumness of our state is robust against dephasing, thus requiring fewer experimental resources to ensure stability. In addition, we theoretically show how multimode entanglement can be activated based on the generated, nonentangled state. Therefore, we implement a robust kind of nonclassical photon-photon correlated state with useful applications in quantum information processing.

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Introduction.—The certification of quantum correlations is essential for the ever-accelerating development of quantum technologies. Beyond this practical demand, a fundamental understanding of quantum correlations, including their characterization and quantification, plays a key role when exploring the boundary between classical theories and the unique features of quantum physics. Still, the question remains which kinds of correlation are genuinely quantum. That is, which of the many contenders—be it an established or recently proposed concept (e.g., quantum coherence and resource theory [1,2], entanglement [3], discord [4], etc.)—does describe the concept of a nonclassical correlation best? In this work, we show that the notion of nonclassicality in quantum optics [5,6] can supersede contemporary forms of quantumness in its ability to unveil quantum correlations.

In the context of quantum optics, any observations which cannot be fully described in terms of Maxwell's wave theory of light are called nonclassical [5,6]. This well-established concept of nonclassicality is based on the impossibility of describing field correlations as done in classical electrodynamics, and it is commonly defined in terms of the Glauber-Sudarshan P representation [7,8]. The latter describes nonclassical light through phase-space distributions that are, in the case of quantum light, incompatible with a classical concept of a non-negative probability distribution.

The more recently developed concept of quantum coherence adapts some ideas of the notion of nonclassicality to quantify resources required for quantum information processing [1,2]. In this framework, quantum superpositions equally serve as the origin of quantumness in a system. However, in most cases, the classical reference is defined through incoherent mixtures of orthonormal basis states, contrasting the notion of quantum-optical nonclassicality in terms of nonorthogonal eigenstates of the non-Hermitian annihilation operator.

Entanglement, which can be embedded into the concept of coherence [9,10], is by far the most frequently studied form of quantum correlation among the many contenders [3]. This is due to its fundamental role as well as its many applications, e.g., in quantum metrology, cryptography, computing, and teleportation. The phenomenon of entanglement was discovered in early seminal discussions about the implications of quantum physics [11,12], long before the conception of the relatively young field of quantum information processing.

Many other notions and measures of quantum correlations have been proposed, too [4]. For instance, discord is a feature which includes correlations caused by entangled but also by nonentangled states [13,14], and it can be connected to quantum coherence as well [15]. In this context, it is worth mentioning that the label quantum for this sort of correlation is a topic of ongoing debates [16]. Nonetheless, it has been demonstrated that discord is maximally inequivalent to the notion of quantum-optical nonclassicality [17]. To date, it remains an open problem to decide—not only in theory, but also experimentally—which of the candidates is best suited for characterizing quantum correlations.

In this Letter, we address this issue experimentally by realizing and analyzing a fully phase-randomized two-mode squeezed vacuum (TMSV) state [18]. This state of quantum light has the following properties: It is non-entangled; it has zero discord; it does not exhibit quantum coherence in the photon-number basis; its reduced single-mode states are classical; and it has a non-negative, two-mode Wigner function. Despite these strong signatures of classicality, we demonstrate the presence of quantum correlations as defined through the notion of nonclassicality in quantum optics with a statistical significance next to certainty. Because of the phase independence of the generated state, this nonclassical feature is robust against dephasing. Furthermore, the activation of entanglement using this kind of state is developed to show its resourcefulness for quantum information processing applications.

Quantum correlations.—For analyzing quantum correlations, we consider a class of two-mode states that are phase insensitive [17,18]. Still, intensity-intensity (likewise, photon-photon) correlations are present in such states. For instance, this can be achieved by a full phase randomization of a TMSV state, resulting in

$$\hat{\rho} = \sum_{n\in\mathbb{N}} (1-p)p^n |n\rangle\langle n| \otimes |n\rangle\langle n|, \tag{1}$$

where $p = \tanh|\xi|$ is a value between zero and one and ξ is the complex squeezing parameter. Such states are, for example, a relevant resource for boson sampling tasks [19].

In terms of quantum correlations, it can be directly observed that the state in Eq. (1) is an incoherent mixture of photon-number states, thus exhibiting no quantum coherence in the form of quantum superpositions of photon-number states; it is a classical mixture of tensor-product states, thus exhibiting no entanglement; and it has zero discord, because $\hat{\rho} = \sum_{n\in\mathbb{N}} \hat{\rho}_{A|n} \otimes |n\rangle\langle n|$ holds true, where the photon-number states form the eigenbasis to $\hat{\rho}_{A|n} = (\hat{1} \otimes \langle n|)\hat{\rho}(\hat{1} \otimes |n\rangle)$ and $\mathrm{tr}_A\hat{\rho}$ [20]. For those criteria of quantum correlations, it suffices in our scenario to consider the contribution of off-diagonal elements,

$$\mathcal{C}(\hat{\rho}) \stackrel{\mathrm{def}}{=} \sum_{\substack{m,n,k,l\in\mathbb{N}:\\ m\neq n, k\neq l}} |(\langle m| \otimes \langle k|)\hat{\rho}(|n\rangle \otimes |l\rangle)|, \tag{2}$$

which quantifies the coherent contributions [21] and is $\mathcal{C}(\hat{\rho}) = 0$ for the state in Eq. (1). It is worth emphasizing that the nullity of coherence in the two-mode photon-number basis implies the nullity of discord, which further implies no entanglement. In addition, the incoherent mixture of photon-number states under study further implies a classical interpretation in a particle picture [22]. Furthermore, the marginal states $\mathrm{tr}_A\hat{\rho}$ and $\mathrm{tr}_B\hat{\rho}$ are thermal states and, thus, classical, too. Also, the two-mode state under study is a mixture of Gaussian TMSV states, implying a non-negative Wigner function.

At this point, one sees no indication of quantum correlations. Yet, we have not considered the notion of nonclassicality in quantum optics so far. This concept is defined through the Glauber-Sudarshan P representation [7,8]

$$\hat{\rho} = \int d^2\alpha \int d^2\beta P(\alpha,\beta) |\alpha\rangle\langle\alpha| \otimes |\beta\rangle\langle\beta|, \tag{3}$$

where $|\alpha\rangle$ and $|\beta\rangle$ denote classically coherent states of the harmonic oscillator [23]. Whenever P cannot be interpreted as a classical probability density, the state of light $\hat{\rho}$ refers to a nonclassical one [5,6]. Since the P distribution is in many cases highly singular [24], thus experimentally inaccessible, regularization and direct sampling procedures have been proposed and implemented to reconstruct a function P_Ω which is always regular and non-negative for any classical states of light [25,26]. This is achieved by a convolution of the Glauber-Sudarshan P function with a suitable, non-Gaussian kernel Ω, resulting in P_Ω. Our previous theoretical studies suggest that the state in Eq. (1) indeed demonstrates nonclassical correlations [18], i.e.,

$$P_\Omega(\alpha,\beta) \stackrel{\mathrm{ncl}}{<} 0 \tag{4}$$

for some complex phase-space amplitudes α and β.

Experimental implementation.—Figure 1 shows our experimental setup for the preparation and detection of the quantum state in Eq. (1). The challenge here is that a full two-mode state tomography with long-term stability, phase control, and phase readout is paramount for our coherence analysis. See Supplemental Material [27] for technical details.

Two amplitude-squeezed fields at 1064 nm are produced by optical parametric amplifiers (OPAs). One OPA consists of a type-I hemilithic, standing wave, nonlinear cavity with a 7% $MgO:LiNbO_3$ crystal; the other OPA uses a periodically poled potassium titanyl phosphate crystal instead. To combine distinct sources of squeezed light in one setup, the seed and pump powers are adjusted such that the squeezed output fields of the two crystals are of equal intensity and squeezing. For this purpose, the two pump powers of the second harmonic are chosen as 242 and 50 mW, respectively. Moreover, we have to minimize power fluctuations of the pump light, since the optical parametric gain of each OPA responds differently [27].

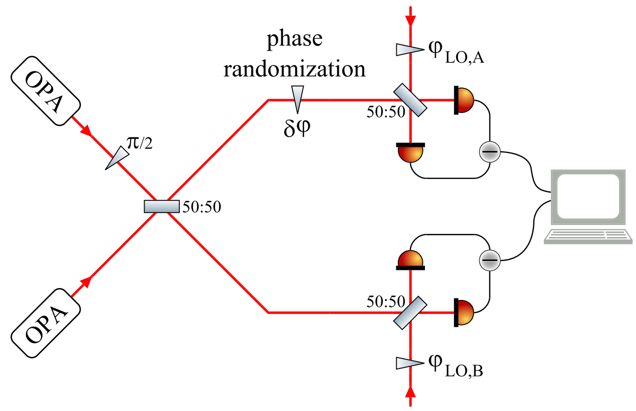

FIG. 1. Setup outline. Two single-mode squeezed states are prepared by two OPAs. Combining these beams on a 50∶50 beam splitter with a $\pi/2$ phase shift yields a TMSV state. Introducing a phase randomization (indicated by the phase fluctuation $\delta\varphi$) in one of the output arms approximates the desired target state in Eq. (1) [18]. Both final beams are probed by balanced homodyne detectors with controllable phases $\varphi_{\mathrm{LO},A}$ and $\varphi_{\mathrm{LO},B}$.

The two squeezed fields are superposed with a visibility of 96.5% and a relative phase of $\pi/2$ on a 50∶50 beam splitter. The high visibility demonstrates a successful combination of the two sources that results in a TMSV state:

$$|\mathrm{TMSV}\rangle = \frac{1}{\cosh|\xi|}\sum_{n=0}^{\infty}(e^{i\arg\xi}\tanh|\xi|)^{n}|n\rangle\otimes|n\rangle. \quad (5)$$

Both output modes A and B of the state are probed by balanced homodyne detectors. We observed $(96.7\pm0.7)\%$ visibility between the fields and their corresponding local oscillators. For one OPA, we measured a single-mode squeezing variance of -1.3 dB and $+3.7$ dB antisqueezing. This yields an initial squeezing of -7.3 dB and an overall efficiency of $(63\pm2)\%$. The latter figure was multiplied by two to compensate for the vacuum input, because blocking the second OPA effectively introduces additional 50% loss at the first (i.e., leftmost) beam splitter in Fig. 1.

We use piezoelectric transducers to control the optical phases and realize a random phase shift $\delta\varphi$ in one of the arms. To achieve a uniform dephasing over the full 2π interval, white noise is applied with sufficiently high amplitude. Because of the bandwidth limitations of the transducers, a uniformly distributed phase—as required to exactly obtain the state in Eq. (1)—can be approximated only via long measurement times, requiring us to maintain a high stability of our setup. Further technical details are provided in Supplemental Material [27].

Results.—Figure 2 depicts the first 625 density-matrix elements of the reconstructed state, without (top panel) and with (bottom panel) phase randomization, which requires the full two-mode state tomography. The initially generated

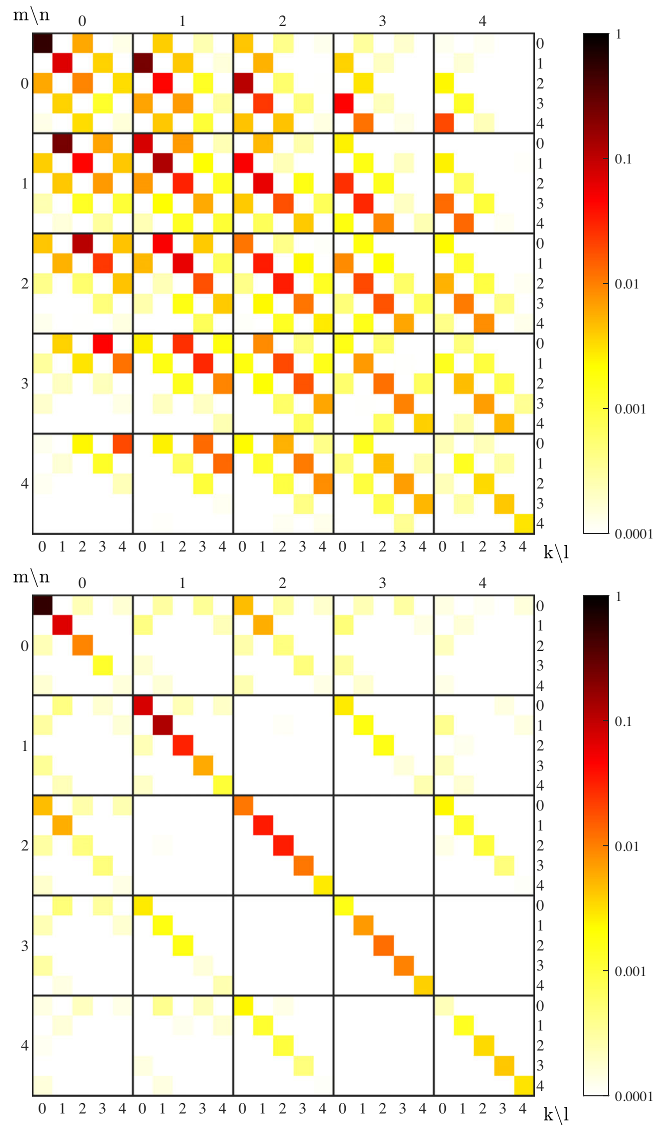

FIG. 2. Reconstructed density matrix elements of the TMSV state in photon-number bases, before (top) and after (bottom) phase randomization. Each entry provides the absolute values of the density matrix elements $|\rho_{(k,m),(l,n)}|$, where $\hat{\rho} = \sum_{k,l,m,n}\rho_{(k,m),(l,n)}|k\rangle\langle l|\otimes|m\rangle\langle n|$. Please note the logarithmic scale. In both plots, k (bottom axis) and l (right axis) denote photon numbers for A, and m (left axis) and n (top axis) indicate photon numbers for B. Large off-diagonal contributions certify the presence of quantum coherence (top). Strongly diminished off-diagonal elements indicate the absence of quantum coherence (bottom).

state shows strong contributions of off-diagonal elements, relating to the dominance of quantum coherence [Eq. (2)], $\mathcal{C}(|\mathrm{TMSV}\rangle\langle\mathrm{TMSV}|) = 1.789\pm0.021$. The implemented phase randomization then leads to a 45-fold suppression of the initial coherence, resulting in almost no subsisting coherence, $\mathcal{C}(\hat{\rho}) = 0.041\pm0.005$. (Errors have been obtained through a Monte Carlo approach; see Supplemental Material [27] for details on data processing and a discussion of the residual amount of quantum coherence, caused by experimental imperfections.) This loss of coherence implies that entanglement and discord do not contribute to the phase-randomized state that is almost completely characterized by its diagonal elements in the photon-number basis; see Eq. (1) and the bottom plot in Fig. 2.

We showed that quantum correlations in terms of quantum coherence are negligible for the produced state. That is, the phase-averaged state is mostly consistent with a classical statistical mixture of orthonormal two-mode, tensor-product, photon-number states, thus also ruling out entanglement and discord as a source of quantum correlations as discussed earlier. However, from our balanced homodyne detection data, we can further directly sample the regularized phase-space function P_Ω [26]. Here, the reconstruction is based on phase-insensitive pattern functions [27]. Thus, no amount of residual coherence can contribute to nonclassicality in the form of Eq. (4). The resulting distribution is shown in Fig. 3. We found a maximal statistical significance of more than 150 standard deviations for the negativity of the reconstructed quasiprobability distribution, $P_\Omega(\alpha=0,\beta=1.5)=(-1.570\pm0.010)\times 10^{-3}$. Because of these highly significant negativities, quantum correlations in the generated state are confirmed beyond the previously considered notions.

Again, we emphasize that quantum coherence, entanglement, and discord cannot contribute to the negativity, as our approach is, by construction, insensitive to such phase-sensitive phenomena. Furthermore, our method is, to our knowledge, the only existing approach to uncover this form of quantumness. As mentioned before, the Wigner function, for instance, is completely positive for our state. Thus, we experimentally generated quantum correlations which are inaccessible via other means. Moreover, phase stability is not required for the kind of quantum effect. In fact, we artificially introduced phase noise—which is often omnipresent in realistic quantum channels—to produce the sought-after state.

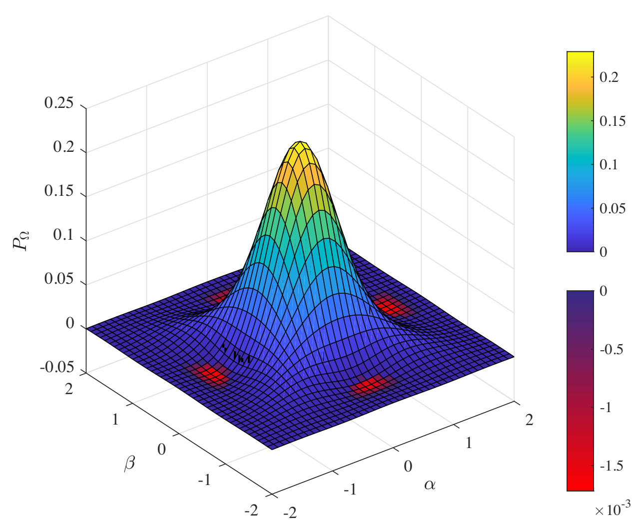

FIG. 3. Regularized P function sampled from experimental data for the phase-randomized TMSV state, including negativities.

Entanglement activation.—Because of the ever-growing importance for quantum information processing [3,31], the question arises if entanglement can be activated, like for coherence and discord [9,10,15], from the nonclassically correlated state under study. Achieving such an activation renders this state a useful resource for quantum technologies.

For this purpose, let us recall that single-mode nonclassicality can be converted into entanglement via simple beam splitters [32–34]. Similarly, we consider combining each of our two modes separately on a 50:50 beam splitter with vacuum, where annihilation operators for the nonvacuum input map as $\hat{a}\mapsto(\hat{a}+\hat{a}')/\sqrt{2}$ and $\hat{b}\mapsto(\hat{b}+\hat{b}')/\sqrt{2}$; additional modes obtained from the splitting are indicated by prime superscripts. Such operations are free (i.e., classical) ones with respect to the reference $|\alpha\rangle\otimes|\alpha'\rangle$ of the Glauber-Sudarshan representation [Eq. (3)] since the beam-splitter output for mode A remains in this family of states, $|(\alpha+\alpha')/\sqrt{2}\rangle\otimes|(\alpha-\alpha')/\sqrt{2}\rangle$, and likewise for B. Furthermore, applied to a photon-number input state $|n\rangle\otimes|0\rangle$, the map yields the output state $|\Psi_n\rangle = 2^{-n/2}\sum_{j=0}^{n}\binom{n}{j}^{1/2}(-1)^{n-j}|j\rangle\otimes|n-j\rangle'$. Therefore, the state in Eq. (1) results in the final four-mode state

$$\hat{\rho}_{AA'BB'}=\sum_{n\in\mathbb{N}}(1-p)p^n|\Psi_n\rangle\langle\Psi_n|\otimes|\Psi_n\rangle\langle\Psi_n|. \quad (6)$$

Clearly, this state is still nonentangled when separating the joint subsystems AA' and BB' from each other. However, multimode entanglement is much richer, since entanglement in various mode decompositions can be considered; see, e.g., Refs. [35,36]. Here, we address the question whether there is entanglement between the primed and unprimed modes, i.e., with respect to the separation of AB and $A'B'$.

To answer this question, we consider the partial transposition criterion [37,38]. In one form [38], this criterion states that a state is entangled if the expectation value of a so-called entanglement witness $\hat{W}=(|\Phi\rangle\langle\Phi|)^{\mathrm{PT}'}$ is negative, where PT' denotes the partial transposition of the primed modes. For example, we can choose $|\Phi\rangle=|0\rangle\otimes|0\rangle'\otimes|1\rangle\otimes|1\rangle'-|1\rangle\otimes|1\rangle'\otimes|0\rangle\otimes|0\rangle'$, which results in

$$\mathrm{tr}(\hat{W}\hat{\rho}_{AA'BB'})=-\frac{(1-p)p}{2}<0, \quad (7)$$

for all nontrivial parameters $0<p<1$ that define our states in Eq. (1). Therefore, the two-mode nonclassical correlations of the generated state can be successfully activated to produce four-mode entanglement, which is a useful mixed-state quantum resource, such as for teleportation [39–42], and which can be further enhanced via entanglement distillation [43–47].

Conclusion.—We experimentally realized a quantum state with quantum correlations that are inaccessible by means of two-mode coherence, thus entanglement and discord, but can be intuitively visualized by negative quasiprobabilities. Quantum coherence—a recently explored resource for quantum information processing—exists in terms of superpositions of orthogonal photon-number states of the initial TMSV state. But a phase averaging destroys this and related kinds of quantum correlations. Thus, contemporary quantum-information-based concepts of correlation fail to certify the quantumness of the state under the challenging, but common, scenario of dephasing. However, the Glauber-Sudarshan-based concept of nonclassicality of light—frequently considered to be a dated notion, or not considered at all—is still capable of uncovering the quantum nature of the generated state.

Since the Glauber-Sudarshan distribution often displays a highly singular behavior, we employ a technique which enables us to directly sample a regularized version of such a two-mode phase-space function, allowing us to certify nonclassical negativities with a statistical significance of more than 150 standard deviations. Thus, the generated phase-independent state exhibits nonclassical correlations beyond entanglement and discord. Furthermore, we theoretically devised a method to activate multimode entanglement, utilizing only the produced state and simple beam splitter operations.

In conclusion, we realized a type of quantum correlation that can be accessed via phase-space approaches but not through more recent notions of quantumness. This kind of correlation is intrinsically robust against dephasing and can be easily converted into multimode entanglement. Therefore, this finding offers useful quantum correlations that can be realized without experimentally costly phase stabilization and is resourceful for modern quantum information protocols.

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