Nonclassicality and entanglement for wave packets

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Mode-entanglement-based criteria and measures become insufficient for broadband emission, e.g., from spasers (plasmonic nanolasers). We introduce criteria and measures for the (i) total entanglement of two wave packets, (ii) entanglement of a wave packet with an ensemble, and (iii) total nonclassicality of a wave packet. We discuss these criteria in the context of (i) entanglement of two wave packets emitted from two initially entangled atoms) and (ii) entanglement of an emitted wave packet with an ensemble or atom for spontaneous emission and single-photon superradiance. We also show that, (iii) when two constituent modes of a wave packet are entangled, this creates nonclassicality in the wave packet as a noise reduction below the standard quantum limit. The criteria we introduce are all compatible with near-field detectors.

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I. INTRODUCTION

Quantum entanglement, a purely nonclassical effect, has been observed both at the macroscopic [1] and microscopic scales [2]. Observation of novel effects such as quantum teleportation [3] with satellites [4], between two matter waves [5], and detection of stealth jets [6] with entangled microwave photons [7] (quantum radars) point to the impact of entangled light on the current technology. This makes the generation, detection, and quantification of nonclassical states—such as quadrature or number-squeezed states [8], two-mode entangled states, and many-particle entangled states [9,10]—an important area of research.

Rapid and dramatic progresses in (quantum) plasmonics not only enabled plasmonic nanolasers (i.e., spasers [11,12]), nanometer-size optical (scanning near-field optical microscopy (SNOM) [13,14]) and Raman (surface enhanced Raman scattering (SERS) [15,16]) imaging, but also allowed quantum optics effects to appear in nanostructures. Analogs of electromagnetically induced transparency (EIT)-like [17] path interference effects appear also in linear [18-21] and nonlinear [22–24] plasmonic response. Surprisingly, squeezed or entangled photons, converted into and back from nanowire plasmon oscillations, are experimentally shown to keep quantum features for times (i.e., 10^{-10} s [25–28]) much longer than plasmons' decay intervals. Considering also miniaturization and high data transfer capacity, plasmons offer in microchips [29], entanglement of two plasmon wave packets or entanglement of a plasmon wave packet with ensembles, e.g., for quantum data storage, appear to likely dominate the quantum optics field in the following decades [30,31].

Entanglement of matter waves is also an interesting phenomenon [32-36], which can be utilized, e.g., for matter-

Witnesses and measures of quantum entanglement usually rely on the inseparability of the two modes which are commonly represented by a single wave vector **k**, i.e., $\hat{a}_{\mathbf{k}_1}$ and $\hat{a}_{\mathbf{k}_2}$. Here, $\omega_{1,2} = ck_{1,2}$ are the carrier frequencies of the two nonclassical beams. Such a treatment is acceptable for pulses of narrow frequency width, especially when the detector is placed (measurement is performed) in the far field, where the choice of a single component k is justified also with the directional (small solid angle) arguments. When a broadband, e.g., a plasmon, emission is measured in the far field, a single (carrier) k-mode is detected, still, due to the small angle arguments. Hence, in a broadband source, too, detection of entanglement via carrier frequencies does still work.

However, quantification of the entanglement or nonclassicality via detecting the inseparability of only the two modes, e.g., carrier frequencies of the two beams, is highly insufficient in the detection and "use" of the whole entanglement potential of the two broadband pulses. Maximum entanglement or nonclassicality harvesting, e.g., in quantum teleportation [3,5] and quantum thermodynamics (heat engines) [37,38], is important in the efficiency of such devices. The situation (insufficiency of mode-entanglement-based criteria) becomes even more adverse if the quantification is performed via two near-field detectors [39], where separation of two modes becomes impossible. Therefore, the entanglement of two wave packets, which once could be questioned due to curiosity, now becomes a necessity [40] with the development of fast-response nanocontrol [12] and nanoimaging [39] techniques.

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wave quantum teleportation [5]. While entanglement between the two components of a Bose-Einstein condensate, achieved via hopping and collision interactions, refers to two sharp, e.g., momentum, modes [32–36], a stronger entanglement, obtained, e.g., via molecular dissociation [5] and collisions possesses a wide (e.g., momentum) spectrum.

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In this paper, we aim to extend the notion of two-mode entanglement to the entanglement of two wave packets, each containing broadband frequency components. We also introduce the notion for the nonclassicality of a wave packet, which is referred to as single-mode nonclassicality. Furthermore, we extend the definition of entanglement between an ensemble of quantum emitters and the emitted mode [41] to the ensemblewave-packet entanglement [42].

After a survey among the possible extensions or generalizations of the entanglement of wave packets, we demonstrate that the most meaningful definition could be achieved via making a replacement, $\hat{a} \rightarrow \sum_{\mathbf{r}} \hat{a}_{\mathbf{r}}$, from a single mode to a wave packet. The summation $\sum_{\mathbf{r}}$ stands for the volume or area of the detector, for the measurement via a near-field detector, and $\sum_{\mathbf{r}}$ stands for the whole space for the calculation of the total entanglement existing between the two wave packets. Here, $\hat{a}_{\mathbf{r}}$ is the annihilation operator of a photon at position \mathbf{r} . In particular, we study the entanglement of wave packets emitted from either two initially entangled cavities or two initially-entangled atoms. Similar to electromagnetic wave packets, continuous-variable entanglement between two matter waves can be witnessed by referring to their center-ofmass position or momentum.

The paper is organized as follows. In Sec. II, we introduce the entanglement of two wave packets using the electric fields of the two wave packets, i.e., $\hat{a}_i \rightarrow \hat{E}_i^{(+)} = \sum_{\mathbf{k}} \varepsilon_k e^{i\mathbf{k}\cdot\mathbf{r}} \hat{a}_{\mathbf{k}}$. We show that the generation (onset) of entanglement between the two pulses, at positions \mathbf{r}_1 and \mathbf{r}_2 , propagates with the speed of light, *c*. This definition is demonstrated to be not useful for two purposes:

(1) Entanglement does not quantify the inseparability of the two wave packets, instead it witnesses on the inseparability (correlations) of the electric field measurements at the positions \mathbf{r}_1 and \mathbf{r}_2 .

(2) Using such a definition, we are faced with a divergence problem, in $\sum_{\mathbf{k}} \varepsilon_k$, when we desire to use the analogs of the stronger Simon-Peres-Horodecki (SPH) criterion [43] or the Duan-Giedke-Cirac-Zoller (DGCZ) criterion [44] for the wave packets. We face the same divergence problem when we introduce $\hat{a}_i \rightarrow \sum_{\mathbf{k}_i} \hat{a}_{i,\mathbf{k}_i}$, although this definition has the potential to detect the inseparability of any two modes selected from each wave packet.

In Sec. III, we introduce $\hat{a}_i \rightarrow \sum_{\mathbf{r}_i} \hat{a}_{i,\mathbf{r}_i}$. This one can circumvent both the divergence problem in item 2 and calculate the total entanglement which two near-field detectors measure. We can therefore calculate the whole entanglement (potential) between the two wave packets. Here, i = 1, 2 refers to the two wave packets.

In Sec. IV, we define the total entanglement between two wave packets by introducing the annihilation operator $\hat{A}_i = \sum_{\mathbf{r}_i} \hat{a}_{i,\mathbf{r}_i}$. We introduce the criteria for wave-packet entanglement that are analogs to the SPH criterion [43] and the Hillery-Zubairy (HZ) criterion [45]. We study the time development of the total entanglement of two wave packets, emitted from two initially entangled cavities or atoms, using both HZ and SPH criteria. Use of criteria for detecting the entanglement between two matter waves is also discussed. In Sec. V, we introduce ensemble-wave-packet entanglement criteria by replacing $\hat{a}_i \rightarrow \hat{A}_i$. We study the spontaneous emission of a single atom and superradiant single-photon emission from a many-particle entangled ensemble. In Sec. VI, we define the nonclassicality of a wave packet both via a noise matrix of \hat{X} , \hat{P} operators, defined over \hat{A} , and via a beam splitter (BS): by measuring the wave-packet–wave-packet entanglement generated at the beam-splitter output when a nonclassical wave packet is incident on the beam splitter. We show that, (a) when some of the constituent modes belonging to the wave packet are squeezed or (b) when two modes of the wave packet are entangled, the wave packet becomes nonclassical, i.e., with reduced noise in an \hat{X}_{ϕ} operator, with $\hat{A}_{\phi} = e^{i\phi}\hat{A}$. In Sec. VII, we present a summary of our results.

II. CORRELATIONS OF ELECTRIC-FIELD MEASUREMENTS

Arriving at a convenient definition, or a notion, of the entanglement of two wave packets (WPs) necessitates the exploration of correlations between electric fields of the two wave packets at different positions \mathbf{r}_1 and \mathbf{r}_2 . It is straightforward to see that one can obtain the same forms with the two criteria, DGCZ [43] and HZ [45,46], for $\hat{a}_1 \rightarrow \hat{E}_1^{(+)}(\mathbf{r}_1)$ and $\hat{a}_2 \rightarrow \hat{E}_2^{(+)}(\mathbf{r}_2)$, where

$$\hat{E}_{i}^{(+)}(\mathbf{r}_{i}) = \sum_{\mathbf{k}_{i}} \varepsilon_{k_{i}} e^{i\mathbf{k}_{i}\cdot\mathbf{r}_{i}} \hat{a}_{i,\mathbf{k}_{i}}$$
(2.1)

are the positive parts of the electric field operators associated with the two wave packets, i = 1, 2. $\varepsilon_{k_i} = \sqrt{\hbar c k_i / \epsilon_0 V_i}$ is the electric field of a single photon, depending on the quantization volume V_i of the *i*th wave packet. Following the same steps, given in Ref. [45], a criterion analogous to the HZ criterion can be obtained as

$$\lambda_{\rm HZ} = \langle \hat{E}_2^{(+)}(\mathbf{r}_2) \hat{E}_2^{(-)}(\mathbf{r}_2) \hat{E}_1^{(+)}(\mathbf{r}_1) \hat{E}_1^{(-)}(\mathbf{r}_1) \rangle - |\langle \hat{E}_2^{(+)}(\mathbf{r}_2) \hat{E}_1^{(-)}(\mathbf{r}_1) \rangle|^2, \qquad (2.2)$$

where $\lambda_{\text{HZ}} < 0$ witnesses the inseparability of the two wave packets, or the presence of nonlocal correlations between electric field measurements of the two wave packets at positions \mathbf{r}_1 and \mathbf{r}_2 . $\hat{E}_i^{(-)}(\mathbf{r}_i)$ is the Hermitian conjugate of $\hat{E}_i^{(+)}(\mathbf{r}_i)$. The Hillery-Zubairy criterion does not lead to any divergence problem, in contrast to SPH or DGCZ criteria, since it does not necessitate the evaluation of a term like $\langle \hat{E}_i^{(+)}(\mathbf{r}_i) \hat{E}_i^{(-)}(\mathbf{r}_i) \rangle$.

One can also derive the analog of the DGCZ criterion for the entanglement of two wave packets with the replacement $\hat{x}_1 \rightarrow \hat{E}_1(\mathbf{r}_1)$ and $\hat{x}_2 \rightarrow \hat{E}_2(\mathbf{r}_2)$ using the same arguments in Ref. [44], i.e., the Cauchy-Schwarz inequality for separable states. Here, $\hat{E}_i(\mathbf{r}_i) = \hat{E}_i^{(+)}(\mathbf{r}_i) + \hat{E}_i^{(-)}(\mathbf{r}_i)$ is the electric field operator. This criterion, however, is not a useful one since it contains terms like $\langle \hat{E}_i^{(+)}(\mathbf{r}_i) \hat{E}_i^{(-)}(\mathbf{r}_i) \rangle$ which do diverge. The SPH criterion also includes similar divergent terms and does not have any practical use here.

Our experience shows us that the DGCZ criterion works well for quadrature-squeezed-like states, while the HZ criterion works well mainly for number-squeezed-like states and superpositions of Fock states [47]. Here, in this section, we consider the entanglement of two wave packets, emitted from two initially entangled cavities, $|\psi(0)\rangle = a_1(0)|1\rangle_{c_1}|0\rangle_{c_2} + a_2(0)|0\rangle_{c_1}|1\rangle_{c_2}$, into two different reservoirs, or from two initially entangled atoms $|\psi(0)\rangle = a_1(0)|e\rangle|g\rangle + a_2(0)|g\rangle_1|e\rangle_2$. (We study the extended version of the system considered



FIG. 1. Two cavities are initially in an entangled state and they decay into two different reservoirs. We examine the time evolution of the onset of the entanglement between the two reservoirs or, in other words, correlations in the electric field measurements of the wave packets (WPs) emitted into the two reservoirs. We also calculate the total entanglement of the two wave packets in Sec. IV.

in Ref. [48], where the reservoirs are treated as two single modes.) Fortunately, we can study the correlations in such a system. Because the system emits the superpositions of Fock states, the HZ criterion, which does not diverge, can be used.

In Fig. 1, the two cavities are initially in an entangled state, $|\psi(0)\rangle = (a_1(0)|1\rangle_{c_1}|0\rangle_{c_2} + a_2(0)|0\rangle_{c_1}|1\rangle_{c_2})|0\rangle_{R_1}|0\rangle_{R_2}$, where $|\rangle_{c_{1,2}}$ and $|\rangle_{R_{1,2}}$ are the Fock states for the two entangled cavities and the two reservoirs the cavities decay, respectively. The solution of the interaction picture Hamiltonian [48]

$$\hat{V} = \sum_{i=1}^{2} \sum_{\mathbf{k}_{i}} \hbar g_{\mathbf{k}_{i}} \hat{a}_{i,\mathbf{k}_{i}}^{\dagger} \hat{c}_{i} e^{-i(\Omega_{i}-\omega_{\mathbf{k}_{i}})t} + \text{H.c.}$$
(2.3)

in the subspace of possible states

$$\begin{aligned} |\psi(t)\rangle &= (b_{1}(t)|0\rangle_{c_{1}}|1\rangle_{c_{2}} + b_{2}(t)|1\rangle_{c_{1}}|0\rangle_{c_{2}}|0\rangle_{R_{1}}|0\rangle_{R_{2}} \\ &+ |0\rangle_{c_{1}}|0\rangle_{c_{2}} \left(\sum_{\mathbf{k}_{1}} d_{1,\mathbf{k}_{1}}(t)|1_{\mathbf{k}_{1}}\rangle_{R_{1}}|0\rangle_{R_{2}} \\ &+ |0\rangle_{R_{1}}\sum_{\mathbf{k}_{2}} d_{2,\mathbf{k}_{2}}(t)|1_{\mathbf{k}_{2}}\rangle_{R_{2}} \right) \end{aligned}$$
(2.4)

is determined by the coefficients

$$b_i(t) = e^{-\gamma_i t/2} a_i(0),$$
 (2.5)

$$d_{i,\mathbf{k}_{i}}(t) = g_{k_{i}}a_{i}(0)\frac{1 - e^{-i(\Omega_{i} - \omega_{k_{i}})t - \gamma_{i}t/2}}{(\omega_{k_{i}} - \Omega_{i}) + i\gamma_{i}/2},$$
(2.6)

where Ω_i and γ_i are the cavity resonance and damping rate, respectively. g_{k_i} is the coupling strength between the *i*th cavity and the *i*th reservoir. When we consider sufficiently long cavities, and thin mirrors which couple the cavities to the reservoirs, the HZ criterion for the entanglement of the two wave packets can be calculated as

$$\lambda_{\rm HZ}(t) \simeq -(2\pi)^2 g_1^2(\Omega_1) D_1(\Omega_1) g_2^2(\Omega_1) D_2(\Omega_2) \varepsilon_{K_1} \varepsilon_{K_2} \times e^{-\gamma_1 |z_1 - ct|/2c} e^{-\gamma_2 |z_2 - ct|/2c} \times \Theta(t - z_1/c) \Theta(t - z_2/c),$$
(2.7)

where we assume that dispersion of the cavity emission is negligible in the transverse directions, \hat{x}_i and \hat{y}_i . $D(\Omega_i)$ is the density of states at the cavity resonance Ω_i and can be related to the damping rate as $\gamma_i = \pi D_i(\Omega_i)g^2(\Omega_i)$. $\varepsilon_{K_1} = \sqrt{\hbar\Omega_i/\epsilon_0 V_i}$ with $K_i = \Omega_i/c$. The step functions in Eq. (2.7), $\Theta(t - z_i/c)$, reveal the luminal "onset" of correlations (entanglement) between the two wave packets, at z_1 and z_2 . We note that this approximate result for entanglement is realistic in the following aspect. For two collimated wave packets of narrow frequency band, the entanglement does not decay (or decays negligibly) with *z* propagation. We also evaluate the $\lambda_{HZ}(t)$ for an uncollimated emission, where we find that the absolute value of its negativity decreases with spatial spreading.

Such a definition of entanglement (correlations) between two wave packets is instructive especially for exploring the onset of the entanglement in spatial dimensions. However, such a definition fails to work for most useful nonclassical states, the Gaussian states, which are the ones convenient to generate and use in the experiments.

Moreover, it has a potential only to quantify the wavepacket-wave-packet entanglement on a position-to-position basis. That is, it does not quantify the "total" entanglement between the two wave packets. A candidate for quantifying the total entanglement, i.e., between all of the modes, could be

$$\hat{a}_i \to \sum_{\mathbf{k}_i} \hat{a}_{i,\mathbf{k}_i} \quad \text{or} \quad \hat{a}_i \to \sum_{\mathbf{k}_i} \varepsilon_{k_i} \hat{a}_{i,\mathbf{k}_i},$$
(2.8)

which have the potential to address the entanglement of any two modes, \hat{a}_{1,\mathbf{k}_1} and \hat{a}_{2,\mathbf{k}_2} , between the two wave packets.¹ Such definitions, however, are again not useful for Gaussian states since they lead to divergence in SPH and DGCZ criteria.

III. CONVENIENCE OF WORKING IN THE SPATIAL DOMAIN: CONVERGENCE

Next, we realize that we cannot avoid the divergence of $\sum_{\mathbf{k}}$ summation, since we cannot adopt a bound for the **k**-space. In contrast to momentum space, fortunately, a $\sum_{\mathbf{r}}$ summation is bound by the volume *V* which can be handled theoretically or can be limited in the experiments. Thus, we choose to work in the spatial domain by introducing the mode expansion [49,50]

$$\hat{a}(\mathbf{r}) = \sum_{\mathbf{k}} e^{i\mathbf{k}\cdot\mathbf{r}}\hat{a}_{\mathbf{k}},\tag{3.1}$$

which can be Fourier transformed as

$$\sum_{\mathbf{r}} \hat{a}(\mathbf{r}) e^{-i\mathbf{k}\cdot\mathbf{r}} = \sum_{\mathbf{k}'} \left(\sum_{\mathbf{r}} e^{i(\mathbf{k}-\mathbf{k}')\cdot\mathbf{r}} \right) \hat{a}_{\mathbf{k}'} = \hat{a}_{\mathbf{k}}$$
(3.2)

by defining the normalized summation $\sum_{\mathbf{r}} \rightarrow \int d^3 \mathbf{r} / V$ and using $\sum_{\mathbf{k}} \rightarrow \frac{V}{(2\pi)^3} \int d^3 \mathbf{k}$ as usual [17]. Hermitian conjugates of Eqs. (3.1) and (3.2) can be employed, applied in vacuum, to relate the spatial and momentum Fock spaces, e.g., as

$$|1_{\mathbf{r}}\rangle = \sum_{\mathbf{k}} e^{-i\mathbf{k}\cdot\mathbf{r}} |1_{\mathbf{k}}\rangle$$
 and $|1_{\mathbf{k}}\rangle = \sum_{\mathbf{k}} e^{i\mathbf{k}\cdot\mathbf{r}} |1_{\mathbf{r}}\rangle.$ (3.3)

The advantage of working in the spatial domain, by defining the annihilation operator

$$\hat{a}_i \to \hat{A}_i = \sum_{\mathbf{r}_i} \hat{a}_i(\mathbf{r}_i),$$
 (3.4)

¹We use the phrase "has the potential to detect entanglement" on purpose, because noise reduction due to $\hat{a}_{1,\mathbf{k}_1} \leftrightarrow \hat{a}_{2,\mathbf{k}_2}$ entanglement can be screened by a noise increase due to two other modes $\hat{a}_{1,\mathbf{k}'_1} \leftrightarrow \hat{a}_{2,\mathbf{k}'_2}$.

is, now, the quantity $\langle \hat{A}_i \hat{A}_i^{\dagger} \rangle$ does not diverge. Here, i = 1, 2 enumerates the two wave packets. Moreover, Eq. (3.4), when used in an entanglement criterion, has the potential to detect correlations between any two spatial modes, $\hat{a}_{1,\mathbf{r}_1} \leftrightarrow \hat{a}_{2,\mathbf{r}_2}$, of the two wave packets. One can obtain the commutation

$$[\hat{A}, \hat{A}^{\dagger}] = 1$$
 (3.5)

from the relation $[\hat{a}(\mathbf{r}), \hat{a}(\mathbf{r}')] = V\delta(\mathbf{r} - \mathbf{r}')$, which deduces from Eq. (3.1) and $[\hat{a}_{\mathbf{k}}, \hat{a}_{\mathbf{k}'}] = \delta_{\mathbf{k},\mathbf{k}'}$. Commutation (3.5) remains convergent and dimensionless via normalized definition of the spatial integration $\sum_{\mathbf{r}} \rightarrow \frac{1}{V} \int d^3\mathbf{r}$.

In the next section, we use the annihilation operator \hat{A} , defined in Eq. (3.4), to obtain wave packet analogs of DGCZ [44], HZ [45], and SPH [43] criteria. We also use the same form, \hat{A} , for introducing the ensemble-wave-packet entanglement (Sec. V) and nonclassicality of a wave packet (Sec. VI).

IV. WAVE-PACKET-WAVE-PACKET ENTANGLEMENT

In order to obtain a "convergent" entanglement criterion which has the potential to address a kind of "total" entanglement, e.g., taking all spatial or *k*-mode correlations into account, we introduce $\hat{A}_i = \sum_{\mathbf{r}_i} \hat{a}_i(\mathbf{r}_i)$, for instance, for the DGCZ criterion [44]

$$\lambda_{\text{DGCZ}} = \langle (\Delta \hat{u})^2 \rangle + \langle (\Delta \hat{v})^2 \rangle - (\alpha^2 + \beta^2), \qquad (4.1)$$

where $\lambda_{DGCZ} < 0$ witnesses the inseparability of the two wave packets. Here, the operators are

$$\hat{u} = \alpha \hat{X}_1 + \beta \hat{X}_2, \tag{4.2}$$

$$\hat{v} = \alpha \hat{P}_1 - \beta \hat{P}_2, \tag{4.3}$$

where

$$\hat{X}_i = (\hat{A}_i^{\dagger} + \hat{A}_i)/\sqrt{2} = \sum_{\mathbf{r}} \hat{x}_i(\mathbf{r}_i),$$
 (4.4)

$$\hat{P}_{i} = i(\hat{A}_{i}^{\dagger} - \hat{A}_{i})/\sqrt{2} = \sum_{\mathbf{r}_{i}}^{r_{i}} \hat{p}_{i}(\mathbf{r}_{i}).$$
(4.5)

 \hat{X}_i and \hat{P}_i satisfy the usual commutation relation

$$[\hat{X}_i, \hat{P}_i] = i. \tag{4.6}$$

Equation (4.6) is a central result of the paper. Most useful criteria are shown to be derived, even in stronger forms, using the Heisenberg uncertainty and Schrödinger-Roberson inequalities, via a partial transpose method [51,52]. These inequalities, and the derivation of the criteria, are based on the uncertainty of observables, e.g., \hat{X}_i and \hat{P}_i here, and their commutations, e.g., $[\hat{X}_i, \hat{P}_i]$. Thus, any two-mode entanglement criterion derived for $\hat{a}_1 \leftrightarrow \hat{a}_2$, see also Ref. [53], is valid also for the inseparability of the two wave packets, when \hat{X}_i and \hat{P}_i are defined as in Eqs. (4.4) and (4.5).

More explicitly, if one defines the operators

$$\hat{\xi} = [\hat{X}_1 \ \hat{P}_1 \ \hat{X}_2 \ \hat{P}_1] \tag{4.7}$$

and calculates the noise matrix

$$V_{ij} = \frac{1}{2} \langle \hat{\xi}_i \hat{\xi}_j + \hat{\xi}_j \hat{\xi}_i \rangle - \langle \hat{\xi}_i \rangle \langle \hat{\xi}_j \rangle, \qquad (4.8)$$

the SPH criterion [43]

$$\lambda_{\text{SPH}} = \det A \det B + \left(\frac{1}{4} - |\det C|\right)^2 - \operatorname{tr}(AJCJBJC^T J) - \frac{1}{4}(\det A + \det B)$$
(4.9)

is also valid for the entanglement of two wave packets. Here, A, B, and C are 2 × 2 matrices defining the 4 × 4 noise matrix $V = [A, C; C^T, B]$. The SPH criterion [43] is a particularly important one, since it accounts for any intramode rotations, i.e., $\hat{A}_{\phi} = e^{i\phi}\hat{A}$, in the X_i - P_i plane [53].

In Sec. III.3 of Ref. [53], we show that such a strong criterion is possible to be derived also for number-phase-squeezedlike states [8]. Similar to the SPH criterion [43], it accounts for intramode rotations in the n- Φ , number-phase, plane. This new criterion is also valid for detecting the entanglement of two wave packets.

Similarly, the HZ criterion [45]

$$\lambda_{\rm HZ} = \langle \hat{A}_2^{\dagger} \hat{A}_2 \hat{A}_1^{\dagger} \hat{A}_1 \rangle - |\langle \hat{A}_2^{\dagger} \hat{A}_1 \rangle|^2 \tag{4.10}$$

can be derived, using the same arguments in Ref. [45], for the two wave packets.

A. Two entangled cavities

In the following, we calculate the total entanglement between two WPs emitted from two initially entangled cavities into two different reservoirs. This is depicted in Fig. 1. First, we calculate the $\lambda_{HZ}(t)$ given in Eq. (4.10), since the emitted pulses are superpositions of Fock states. Second, we perform the same calculation for λ_{SPH} given in Eq. (4.9). Similar results can be obtained also for the emission of two initially entangled atoms.

1. HZ criterion

The solution of Eq. (2.4) for the emission of two entangled cavities can be transformed to the spatial domain of the two reservoirs as

$$\begin{aligned} |\psi(t)\rangle &= (b_1(t)|0\rangle_{c_1}|1\rangle_{c_2} + b_2(t)|1\rangle_{c_1}|0\rangle_{c_2})|0\rangle_{R_1}|0\rangle_{R_2} \\ &+ |0\rangle_{c_1}|0\rangle_{c_2} \bigg[|0\rangle_{R_1} \bigg(\sum_{\mathbf{r}_2} I_2(\mathbf{r}_2, t)|1_{\mathbf{r}_2}\rangle_{R_2}\bigg) \ (4.11) \\ &+ \bigg(\sum_{\mathbf{r}_2} I_1(\mathbf{r}_1, t)|1_{\mathbf{r}_2}\rangle_{R_2}\bigg)|0\rangle_{R_2}\bigg] \ (4.12) \end{aligned}$$

$$+ \left(\sum_{\mathbf{r}_{1}} I_{1}(\mathbf{r}_{1},t) |\mathbf{1}_{\mathbf{r}_{1}}\rangle_{\mathbf{R}_{1}}\right) |0\rangle_{\mathbf{R}_{2}} \right], \qquad (4.12)$$

where $I_i(\mathbf{r}_i, t) = \sum_{\mathbf{k}_i} d_{i,\mathbf{k}_i}(t)e^{i\mathbf{k}_i \cdot \mathbf{r}_i}$ with $d_{i,\mathbf{k}_i}(t)$ is given in Eq. (2.6). Using the contour-integration method, the momentum integral can be calculated as

$$I_i(\mathbf{r}_i, t) = \frac{Vb_i(0)}{2\pi c r_i} K_i g_i(\Omega_i) e^{-(i\Omega_i + \gamma_i/2)r_i/c} \Theta(ct - r_i), \quad (4.13)$$

where $K_i = \Omega_i/c$ and $g_i(\Omega_i)$ is the cavity-reservoir coupling evaluated at the cavity resonance $\omega = \Omega_i$. We remark that in the evaluation of I_i we did not make a collimated-beam approximation, i.e., $\mathbf{k} \simeq k_z$, which we performed in Eq. (2.7). In Eq. (2.7), we perform a collimated-beam approximation to provide an easier understanding of the experiments. The notion of entanglement would not change if we did or did not perform such an approximation.



FIG. 2. Hillery-Zubairy and Simon-Peres-Horodecki criteria, which become $\lambda_{\text{HZ}}(t) = \lambda_{\text{SPH}}(t)$ for the two wave packets emitted from two initially entangled cavities depicted in Fig. 1. In contrast to pointwise, $E_1(\mathbf{r}_1, t) \leftrightarrow E_2(\mathbf{r}_2, t)$, electric-field correlations studied in Sec. II, $\lambda_{\text{HZ,SPH}}(t) < 0$ witnesses a kind of total entanglement between the two wave packets emitted into two different reservoirs.

When the \hat{A}_1 operator acts on $|\psi(t)\rangle$, we obtain

$$\hat{A}_{1}|\psi(t)\rangle = \left(\sum_{\mathbf{r}_{1}}\sum_{\mathbf{r}_{1}'}I_{1}(\mathbf{r}_{1},t)\hat{a}_{1}(\mathbf{r}_{1}')|1_{\mathbf{r}_{1}}\rangle_{R_{1}}\right)|0\rangle_{R_{2}}|0\rangle_{c_{1}}|0\rangle_{c_{2}}$$
$$= \left(\sum_{\mathbf{r}_{1}}I_{1}(\mathbf{r}_{1},t)\right)|0\rangle_{R_{1}}|0\rangle_{R_{2}}|0\rangle_{c_{1}}|0\rangle_{c_{2}}.$$
(4.14)

The same form appears for $(\hat{A}_2|\psi(t)\rangle)^{\dagger} = \langle \psi(t)|\hat{A}_2^{\dagger}$. If we define the spatial integral in Eq. (4.14) as $J_i(t) = \sum_{\mathbf{r}_i} I_i(\mathbf{r}_i, t)$, the second term of the λ_{HZ} , in Eq. (4.10), can be identified as $-|J_1(t)|^2 |J_2(t)|^2$. It is evident from Eq. (4.14) that $\hat{A}_2\hat{A}_1|\psi(t)\rangle = 0$. Hence, the first term in Eq. (4.10) is zero. Then, the HZ criterion for two wave packets reduces to

$$\lambda_{\rm HZ}(t) = -|J_1(t)|^2 |J_2(t)|^2, \qquad (4.15)$$

where spatial integrals can be evaluated as

$$J_i(t) = \frac{2b_i(0)}{c} K_i g_i(\Omega_i) \frac{1 - e^{\alpha_i ct} + e^{\alpha_i ct} \alpha_i ct}{\alpha_i^2}, \quad (4.16)$$

with $\alpha_i ct = -(i\Omega_i + \gamma_i/2)t$, which do not depend on the reservoir volume. In Fig. 2, we plot $\lambda_{\text{HZ}}(t)$. The total entanglement increases until the two wave packets leave the two cavities (or the two atoms) completely. Then, it drops but approaches a constant value as $\gamma t \gg 1$. We scale the y axis of Fig. 2 with $4a(0)b(0)K_1K_2g_1(\Omega_1)g_2(\Omega_2)/c^2\alpha_1^2\alpha_2^2$. We consider emission from a plasmonic cavity, and thus choose $\gamma = 10^{-2}\Omega$ where Ω is in the optical regime.

2. SPH criterion

We can also calculate the total entanglement between the two wave packets, using the SPH criterion defined in Eq. (4.9). The terms like $\langle \hat{A}_i^2 \rangle$ and $\langle \hat{A}_2 \hat{A}_1 \rangle$ do vanish. So, the 2 × 2 matrices become

$$A = \begin{bmatrix} \ell_1 & 0\\ 0 & \ell_1 \end{bmatrix}, \ B = \begin{bmatrix} \ell_2 & 0\\ 0 & \ell_2 \end{bmatrix}, \text{ and } C = \begin{bmatrix} a & b\\ -b & a \end{bmatrix}, (4.17)$$

where $\ell_{1,2} = \frac{1}{2} + |J_{1,2}|^2$, $a = (J_2^*J_1 + J_1^*J_2)/2$, and $b = i(J_2J_1^* - J_1J_2^*)/2$. The SPH criterion is evaluated as

$$\lambda_{\text{SPH}} = \ell_1^2 \ell_2^2 + \left(\frac{1}{4} - (a^2 + b^2)^2\right)^2 - 2\ell_1 \ell_2 (a^2 + b^2) - \frac{1}{4} \left(\ell_1^2 + \ell_2^2\right), \quad (4.18)$$

which reduces to

$$\lambda_{\rm SPH}(t) = -|J_1(t)|^2 |J_2(t)|^2 = \lambda_{\rm HZ}(t)$$
(4.19)

for the particular system we consider here.

B. Matter-wave entanglement

The criteria developed for electromagnetic radiation above can also be used for matter waves when $\hat{x}_i(\mathbf{r}_i)$ and $\hat{p}_i(\mathbf{r}_i)$, in Eqs. (4.4) and (4.5), are replaced by the individual positions and momenta of the particles. That is, $\hat{X}_{1,2} =$ $\frac{1}{N_{1,2}} \sum_{i_{1,2}=1}^{N_{1,2}} \hat{x}_{i_{1,2}}(\mathbf{r}_{i_{1,2}}). \text{ Now, } \hat{X}_1, \hat{P}_1 \text{ and } \hat{X}_2, \hat{P}_2 \text{ in Eq. (4.7) refer}$ to the center-of-mass coordinates on which measurements are conducted to witness the entanglement. We use a particle number normalization instead of volume normalization. Depending on the type of the interaction between the two matter waves used in the generation of the entanglement, both SPH and HZ criteria can be used in detecting the entanglement. Unlike the collective operators $\hat{S}_{+}^{(i)}$, introduced in the following section, these criteria witness the continuous-variable entanglement between the two matter waves.² When one of the two observable sets in Eq. (4.7) belongs to the electromagnetic wave packet, e.g., in a superradiant emission, one can witness the continuous-variable entanglement between a matter wave [55] and an electromagnetic wave packet.

When the entanglement between two matter waves is generated via interactions like hopping or weak interspecies collisions [32–35], one can obtain a large portion of the total entanglement by calculating the entanglement between the two recoil (momentum) modes. When techniques like molecular dissociation [5,36] are used, where strong interactions in diatomic molecules come into play, however, momentum distribution in each matter-wave ensemble can be broadened. In such a case, continuous-variable entanglement detection based on the criteria studied above becomes more feasible regarding quantum optics applications.

V. ENSEMBLE-WAVE-PACKET ENTANGLEMENT

Similarly, we can introduce an entanglement criterion between an ensemble and a (e.g., emitted) wave packet. When we change $\hat{a} \rightarrow \hat{A}$ in Eq. (4) of Ref. [10], it is straightforward to obtain the criterion

$$\mu_{\rm HZ} = \langle \hat{S}_+ \hat{S}_- \hat{A}^\dagger \hat{A} \rangle - |\langle \hat{S}_+ \hat{A} \rangle|^2, \qquad (5.1)$$

which works better for the entanglement of numberlike (Focklike) states with an ensemble. This is the case for the spontaneous emission of a single atom [17] or superradiant singlephoton emission from an ensemble of many-particle entangled

²For a network of matter waves or wave packets, a multimode generalization can be performed [54].

atoms [10,56,57]. Here, $\hat{S}_{+} = \sum_{i=1}^{N} \sigma_{i}^{(+)}$ is the collective raising operator for the ensemble containing N two-level atoms where $\sigma_i^{(+)}$ is the Pauli matrix of the *j*th atom, and $\hat{S}_{-} = \hat{S}_{+}^{\dagger}$.

One can also obtain the analog of the DGCZ criterion for ensemble-wave-packet entanglement, $\hat{a} \rightarrow \hat{A}$ in Ref. [41], by examining the uncertainty bound for $\langle (\Delta \hat{u})^2 \rangle + \langle (\Delta \hat{v})^2 \rangle$ using 2)

$$\hat{v} = \hat{S}_x + \hat{X} \quad \text{and} \quad \hat{v} = \hat{S}_y - \hat{P},$$
 (5.2)

where $\hat{X} = (\hat{A}^{\dagger} + \hat{A})/\sqrt{2}, \ \hat{P} = i(\hat{A}^{\dagger} - \hat{A})/\sqrt{2}, \ \hat{S}_x = (\hat{S}_+ + \hat{A})/\sqrt{2}$ $\hat{S}_{-})/2$, and $\hat{S}_{v} = i(\hat{S}_{-} - \hat{S}_{+})/2$. Such a criterion has already been studied for the entanglement between an ensemble and a single mode of light [41], in the context of squeezing transfer from a nonclassical light to an ensemble resulting in spin squeezing. Here, we only make the replacement $\hat{a} \rightarrow \hat{A}$ and introduce ensemble-wave-packet entanglement. The DGCZ criterion works fine for Gaussian or quadrature-squeezed-like states.

Below, first, we calculate $\mu_{\rm HZ}(t)$ for the spontaneous emission of a single atom. Next, we evaluate $\mu_{\rm HZ}(t)$ for singlephoton superradiant emission [56,57] from an initially entangled ensemble of atoms [10].

A. Spontaneous emission of a single atom

The wave function of a two-level atom, initially in the excited state, is given by [17]

$$|\psi(t)\rangle = \beta(t)|e\rangle|0\rangle + |g\rangle \sum_{\mathbf{k}} \gamma_{\mathbf{k}}(t)|\mathbf{1}_{\mathbf{k}}\rangle, \qquad (5.3)$$

where spontaneous emission is possible into many \mathbf{k} modes with probability amplitudes

$$\gamma_{\mathbf{k}}(t) = e^{-i\mathbf{k}\cdot\mathbf{r}_0} g_k \frac{1 - e^{i(\omega_k - \omega_{eg})t - \Gamma t/2}}{(\omega_k - \omega_{eg}) + i\Gamma/2},$$
(5.4)

where \mathbf{r}_0 is the position of the atom and $\beta(t) = e^{-\Gamma t/2}$. Spontaneous emission takes place into a vacuum. ω_{eg} and Γ are the level spacing and damping rate of the atom, respectively. g_k is the coupling strength of the **k** vacuum mode with the atomic dipole. When \hat{A} acts on this state, it results in

$$\hat{A}|\psi(t)\rangle = \left[\sum_{\mathbf{r}} \left(\sum_{\mathbf{k}} e^{i\mathbf{k}\cdot\mathbf{r}} \gamma_{\mathbf{k}}(t)\right)\right] |g\rangle|0\rangle, \qquad (5.5)$$

where $\sum_{\mathbf{k}}$ integration in the inner parentheses, I_A , yields

$$I_A(\mathbf{r},t) = \frac{V}{2\pi \ cr_s} g(\omega_{eg}) K_{eg} e^{-(i\omega_{eg} + \Gamma/2)r_s/c} \Theta(ct - r_s), \quad (5.6)$$

with $r_s = |\mathbf{r} - \mathbf{r}_0|$, $K_{eg} = \omega_{eg}/c$, and $\Theta(x)$ is the step function. Then, the $\sum_{\mathbf{r}}$ spatial integration results in

$$J_A(t) = \frac{2g(\omega_{eg})K_{eg}}{c} \frac{1 - e^{\alpha ct} + e^{\alpha ct}\alpha ct}{\alpha^2},$$
 (5.7)

similar to Eq. (4.16) of the previous section. Here, $\alpha ct =$ $-(i\omega_{eg}+\Gamma/2)t$. It is easy to see from Eq. (5.5) that $\hat{S}_{-}\hat{A}|\psi(t)\rangle = 0$, which turns the first term in $\mu_{\rm HZ}$, Eq. (5.1), equal to zero. The $(\langle \psi(t) | \hat{S}_+)^{\dagger} = \hat{S}_- | \psi(t) \rangle$ is

$$\hat{S}_{-}|\psi(t)\rangle = \beta(t)|g\rangle|0\rangle.$$
(5.8)

So, the HZ criterion becomes

$$\mu_{\rm HZ}(t) = -|\beta(t)|^2 |J_A(t)|^2 = -e^{-\Gamma t} |J_A(t)|^2.$$
 (5.9)

In Fig. 3, we plot $\mu_{\rm HZ}(t)$.



FIG. 3. Spontaneous emission from a single atom into the vacuum. Evolution of the entanglement, $\mu_{HZ}(t) < 0$, between the atom and the emitted wave packet. Superradiant single-photon emission from an ensemble shows a similar behavior except for the emission time determined by collective decay γ_N in place of single-atom decay γ .

B. Superradiant emission from an ensemble

We also study the entanglement of the superradiantly emitted single photon from an initially entangled ensemble of atoms $|\phi(0)\rangle_{\text{ens}} = \sum_{j=1}^{N} e^{i\mathbf{k}_0 \cdot \mathbf{r}_j} |e_j\rangle$, where $|e_j\rangle$ indicates that the *j*th atom is in the excited state and the remaining N - 1ones are in the ground state. The method for the generation of such a state is described in Ref. [57]. \mathbf{r}_i are the positions of the atoms in the ensemble which can be much larger than the emission wavelength $\lambda_0 = 2\pi/k_0$ [56]. In Fig. 4 of Ref. [10], we demonstrated the entanglement between the central mode (carrier frequency) of the emitted light and the ensemble. Here, in contrast, we examine the entanglement of the ensemble with the whole emitted light, the WP.

Time evolution, i.e., superradiant emission, of this initial state into vacuum³ is given [56] by

$$|\psi(t)\rangle = \sum_{j=1}^{N} \beta_j(t) |e_j\rangle |0\rangle + \left(\sum_{\mathbf{k}} \gamma_{\mathbf{k}}(t) |\mathbf{1}_{\mathbf{k}}\rangle\right) |g\rangle, \quad (5.10)$$

where

$$\beta_j(t) = \frac{1}{\sqrt{N}} e^{-\gamma_N t} e^{i\mathbf{k}_0 \cdot \mathbf{r}_j}, \qquad (5.11)$$

$$\gamma_{\mathbf{k}}(t) = \frac{g_k}{\sqrt{N}} \frac{1 - e^{-\gamma_N t + i(\omega_k - \omega_{eg})t}}{(\omega_k - \omega_{eg} + i\gamma_N)} \sum_{j=1}^N e^{i(\mathbf{k}_0 - \mathbf{k}) \cdot \mathbf{r}_j}.$$
 (5.12)

This emission, from an extended $(L > \lambda_0)$ entangled ensemble, is referred to as timed superradiance and the initial state is called a timed Dicke state. Here, γ_N is the collective

³The results of Ref. [54] rely on Markovian approximation. Although we confine ourselves to a superradiant emission into vacuum modes, i.e., without the presence of a cavity, a stronger light-matter coupling can be achieved for an ensemble in a cavity. In such a case, i.e., when an ensemble of atoms collectively radiates into a cavity, one needs to take care of the validity of the Markovian approximation because, in a high-finesse cavity, evolution of the collective emission becomes non-Markovian [58].

(superradiant) decay rate, which can be much larger than the decay rate of a single atom [56].

 $\hat{A}|\psi(t)\rangle$ can be calculated similar to the spontaneous emission case, where now I_A in Eq. (5.6) becomes

$$I_A^{(\text{SR})}(\mathbf{r},t) = \sum_{j=1}^N \frac{e^{i\mathbf{k}_0 \cdot \mathbf{r}_j}}{\sqrt{N}} \frac{V}{2\pi \ cr_j} g(\omega_{eg}) K_{eg}$$
$$\times e^{-(i\omega eg + \gamma_N/2)r_j/c} \Theta(ct - r_j). \quad (5.13)$$

 $J_A^{(SR)}(t) = \sum_{\mathbf{r}} I_A^{(SR)}$ can also be calculated similarly, which results in

$$J_{A}^{(\text{SR})}(t) = J_{A}(t, \gamma_{N}) \sum_{j=1}^{N} \frac{e^{i\mathbf{k}_{0}\cdot\mathbf{r}_{j}}}{\sqrt{N}},$$
 (5.14)

where $J_A(t, \gamma_N)$ is the integral calculated for a singleatom emission in Eq. (5.7), with $\Gamma/2 \rightarrow \gamma_N$. We define the last term of Eq. (5.14), a phase-coherence term, as $\zeta = \sum_{j=1}^{N} e^{i\mathbf{k}_0 \cdot \mathbf{r}_j} / \sqrt{N}$.

Similar to the spontaneous emission of a single atom, $\hat{S}_{-}\hat{A}|\psi(t)\rangle = 0$ and $(\langle \psi(t)|\hat{S}_{+})^{\dagger} = \hat{S}_{-}|\psi(t)\rangle$ yields

$$\hat{S}_{-}|\psi(t)\rangle = \left(\sum_{j=1}^{N} \beta_{j}(t)\right)|g\rangle|0\rangle = e^{-\gamma_{N}t} \zeta |g\rangle|0\rangle.$$
(5.15)

Therefore, the ensemble–wave-packet entanglement criterion $\mu_{\rm HZ}$ becomes

$$\mu_{\rm HZ}^{\rm (SR)}(t) = e^{-2\gamma_N t} |\zeta|^2 |J_A(t, \gamma_N)|^2, \qquad (5.16)$$

where $J_A(t, \gamma_N)$ is given in Eq. (5.7) with $\Gamma/2 \rightarrow \gamma_N$. Thus, the time evolution of $\mu_{\rm HZ}^{\rm (SR)}(t)$ is the same as in Fig. 3, the single-atom spontaneous emission case, with replacement $\gamma \rightarrow \gamma_N$ for the scaling of the time, implying a much more rapid decay. We note that one cannot tell if a larger $\mu_{\rm HZ}$ implies a stronger entanglement or not, either in the wave-packet– wave-packet entanglement or in the ensemble–wave-packet entanglement. This is because, unlike logarithmic negativity [59], such entanglement criteria are not demonstrated to be employed as an entanglement measure.

VI. NONCLASSICALITY OF A WAVE PACKET

In this section, we introduce the nonclassicality of a WP. We show that a wave packet possesses nonclassicality either (a) when some of the constituent (**k**) modes are squeezed or (b) when, e.g., two constituent modes $\mathbf{k}_1 \leftrightarrow \mathbf{k}_2$ are entangled. Below, we first express the two methods used for the quantification or observation of the single-mode nonclassicality of a detected mode. Then, we apply these two methods for introducing the nonclassicality of a wave packet.

We remind that the single-mode nonclassicality of a light mode can be defined in two different ways:

(i) One may, e.g., for Gaussian states, examine the noise matrix, i.e., $V_{ij} = \langle \hat{\xi}_i \hat{\xi}_j + \hat{\xi}_j \hat{\xi}_i \rangle / 2 - \langle \hat{\xi}_i \rangle \langle \hat{\xi}_j \rangle$ for the real variables $\xi^{(r)} = [x_1, p_1]$ or using the complex representation $\xi^{(c)} = [\alpha_1, \alpha_1^*]$ [60,61]. One can show that quadrature squeezing, a single-mode nonclassicality, exists if $|\langle \hat{a}^2 \rangle| > \langle \hat{a}^{\dagger} \hat{a} \rangle$ [53], which derives from the eigenvalues of the noise matrix.

(ii) Alternatively, one can also observe or quantify the

nonclassicality of a single-mode \hat{a} via checking if it creates two-mode entanglement at a BS output [62–64]. For instance, the SPH criterion [43]—not only a necessary and sufficient condition for Gaussian states, but also a criterion that works well for superpositions of number states—can be used to determine the two-mode entanglement at the BS output. This approach may work better in witnessing the single-mode nonclassicality for a wider range of nonclassical states (see Fig. 2(c) in Ref. [65]).

Both approaches can be used in defining the nonclassicality of a wave packet. We first use method (i) to examine states (a) and (b), expressed in the first paragraph of the present section. In the second part of the section, we also mention briefly about the use of method (ii).

A. Method (i): Examining the noise matrix

Analogous to a single-mode (SM) state, we can define the noise matrix of a wave packet as

$$\begin{bmatrix} \frac{1}{2} + \langle \hat{A}^{\dagger} \hat{A} \rangle & \langle \hat{A}^{2} \rangle \\ \langle \hat{A}^{2} \rangle^{*} & \frac{1}{2} + \langle \hat{A}^{\dagger} \hat{A} \rangle \end{bmatrix}$$
(6.1)

in the complex representation, and as

$$\begin{bmatrix} \langle \hat{X}^2 \rangle - \langle \hat{X} \rangle^2 & \langle \hat{X}\hat{P} + \hat{P}\hat{X} \rangle/2 - \langle \hat{X} \rangle \langle \hat{P} \rangle \\ \langle \hat{X}\hat{P} + \hat{P}\hat{X} \rangle/2 - \langle \hat{X} \rangle \langle \hat{P} \rangle & \langle \hat{P}^2 \rangle - \langle \hat{P} \rangle^2 \end{bmatrix}$$
(6.2)

in real variables. Similar to the SM case [53], $\lambda_{\rm sm} = 1/2 + \langle \hat{A}^{\dagger} \hat{A} \rangle - |\langle \hat{A}^2 \rangle|$ determines the minimum noise (maximum squeezing) in the quadratures $\hat{X}_{\phi} = (\hat{A}_{\phi}^{\dagger} + \hat{A}_{\phi})/\sqrt{2}$ with $\hat{A}_{\phi} = e^{i\phi}\hat{A}$. Here, ϕ is chosen along the min noise direction.

1. Method (i-a): Constituent modes of a wave packet are squeezed

As an example, we first examine the nonclassicality of a wave packet, for which some of the modes are squeezed, but the modes are all separable.

Only two modes are squeezed. For simplicity, as a warmup, first we assume that only two modes of the wave packet are in a squeezed vacuum state, i.e., $|\psi\rangle = |\xi_1\rangle_{\mathbf{k}_1} |\xi_2\rangle_{\mathbf{k}_2} |0\rangle_{\mathbf{k}_2} |0\rangle_{\mathbf{k}_4} \cdots$, and other modes are in a vacuum state.⁴ Here, ξ_i are squeezed vacuum states. In such a case, only four terms do not vanish in $\langle \hat{A}^2 \rangle$:

$$\langle \psi | \hat{A}^2 | \psi \rangle = \langle \xi_1 | \langle \xi_2 | \sum_{\mathbf{r}} \sum_{\mathbf{r}'} \left[e^{i\mathbf{k}_1 \cdot (\mathbf{r} + \mathbf{r}')} \hat{a}_{\mathbf{k}_1}^2 + e^{i\mathbf{k}_2 \cdot (\mathbf{r} + \mathbf{r}')} \hat{a}_{\mathbf{k}_2}^2 \right.$$

$$+ 2e^{i(\mathbf{k}_1 \cdot \mathbf{r} + \mathbf{k}_2 \cdot \mathbf{r}')} \hat{a}_{\mathbf{k}_1} \hat{a}_{\mathbf{k}_2} \left] |\xi_1\rangle |\xi_2\rangle.$$

$$(6.3)$$

We remark that, here, \mathbf{k}_1 and \mathbf{k}_2 are not variables, but they refer to two modes which are squeezed. The expectation values can be calculated by transforming the annihilation operators as $\hat{a}_i(\xi_i) = C_i \hat{a}_i - S_i \hat{a}_i^{\dagger}$, where $C_i \equiv \cosh r_i$ and $S_i \equiv \sinh r_i$, with r_i the squeezing parameters [17] $\xi_i = r_i e^{i\theta_i}$. We set the squeezing angles $\theta_i = 0$ for simplicity.

⁴Actually, this is equivalent to assuming that all other modes are in a coherent state, because only the noise operators $\delta \hat{a}_i$ determine the nonclassicality features. $\hat{D}(\alpha_i)$ displacement of each state does not alter the nonclassicality features [60] for Gaussian states.

In Eq. (6.3), only $\hat{a}_{\mathbf{k}}^2$ terms survive and we obtain

$$\langle \psi | \hat{A}^2 | \psi \rangle = -\sum_{\mathbf{r}} \sum_{\mathbf{r}'} (e^{i\mathbf{k}_1 \cdot (\mathbf{r} + \mathbf{r}')} S_1 C_1 + e^{i\mathbf{k}_2 \cdot (\mathbf{r} + \mathbf{r}')} S_2 C_2).$$
(6.4)

Similarly, $\langle \psi | \hat{A}^{\dagger} \hat{A} | \psi \rangle$ yields

$$\langle \psi | \hat{A}^{\dagger} \hat{A} | \psi \rangle = \sum_{\mathbf{r}} \sum_{\mathbf{r}'} \left(e^{i\mathbf{k}_1 \cdot (\mathbf{r} + \mathbf{r}')} S_1^2 + e^{i\mathbf{k}_2 \cdot (\mathbf{r} + \mathbf{r}')} S_2^2 \right).$$
(6.5)

One can note that

$$\sum_{\mathbf{r},\mathbf{r}'} e^{i\mathbf{k}_{i}\cdot(\mathbf{r}+\mathbf{r}')} = \sum_{\mathbf{r},\mathbf{r}'} e^{i\mathbf{k}_{i}\cdot(\mathbf{r}-\mathbf{r}')} = \left|\sum_{\mathbf{r}} e^{i\mathbf{k}_{i}\cdot\mathbf{r}}\right|^{2}$$
$$= \left(\sum_{\mathbf{r}} \sin(\mathbf{k}_{i}\cdot\mathbf{r})\right)^{2} + \left(\sum_{\mathbf{r}} \cos(\mathbf{k}_{i}\cdot\mathbf{r})\right)^{2}.$$
(6.6)

We remark that in the evaluation of $\langle \hat{A}^2 \rangle$, in Eq. (6.3), we consider only the two modes \mathbf{k}_1 , \mathbf{k}_2 among the summation, or ω integral, over an infinite number of modes. As could be anticipated, the contribution of the two modes remains only infinitesimal. Hence, a $|\sum_{\mathbf{r}} e^{i\mathbf{k}_i \cdot \mathbf{r}}|^2$ summation, when converted to integration $|\int d^3\mathbf{r} e^{i\mathbf{k}_i \cdot \mathbf{r}}/V|^2$, vanishes. Still, we can account for the infinitesimal contributions (squeezing) of the two modes to the nonclassicality of the wave packet as follows. The $\sin(\mathbf{k}_i \cdot \mathbf{r})$ summation in Eq. (6.6) gives exactly zero, since it is zero at $\mathbf{r} = 0$ and symmetric or periodic terms cancel each other. In the $\cos(\mathbf{k}_i \cdot \mathbf{r})$ summation, however, the central term at $\mathbf{r} = 0$, $\cos(0) = 1$, does not vanish. Hence, following our $\sum_{\mathbf{r}}$ definition in Sec. III, Eq. (6.6) becomes

$$\left|\sum_{\mathbf{r}} e^{\mathbf{k}_i \cdot \mathbf{r}}\right|^2 = \frac{(\Delta r)^3}{V},\tag{6.7}$$

which is dimensionless and becomes zero in a standard continuous integration, i.e., $(\Delta r)^3/V \rightarrow 0$.

When we include this infinitesimal contribution to the noise of our wave packet, we obtain

$$\lambda_{\rm sm} = \frac{1}{2} + \langle \hat{A}^{\dagger} \hat{A} \rangle - |\langle \hat{A}^2 \rangle| = \frac{1}{2} + \frac{(\Delta r)^3}{V} [(S_1^2 - S_1 C_1) + (S_2^2 - S_2 C_2)], \quad (6.8)$$

which is always less than 1/2 since $S_i^2 - S_iC_i < 0$ and becomes more negative as r_i increases.

Many modes are squeezed. We are aware that introducing the contribution from a single nonzero point, $(\Delta r)^3$ around $\mathbf{r} = 0$, leaves an ambiguity. However, we conduct this treatment because we do need it unavoidably in case (i-b), below. In order to leave the ambiguity, now, we also present the same treatment for a continuous distribution of the squeezing to many modes. We use the experience we obtained in our treatment with two modes.

When $|\xi_{\mathbf{k}}\rangle$ is a continuous function of **k** modes, we obtain

$$\langle \psi | \hat{A}^2 | \psi \rangle = \langle 0 | \sum_{\mathbf{r}, \mathbf{r}'} \sum_{\mathbf{k}, \mathbf{k}'} e^{i\mathbf{k} \cdot (\mathbf{r} + \mathbf{r}')} \delta_{\mathbf{k}, \mathbf{k}'} \hat{a}_{\mathbf{k}}^2(\xi_{\mathbf{k}}) | 0 \rangle.$$
(6.9)

We know from Eq. (6.3) that $\hat{a}_{\mathbf{k}}\hat{a}_{\mathbf{k}'}$ does not contribute. So, $\langle \psi | \hat{A}^2 | \psi \rangle$ becomes

$$\langle \psi | \hat{A}^2 | \psi \rangle = \sum_{\mathbf{r}, \mathbf{r}'} \sum_{\mathbf{k}} e^{i\mathbf{k} \cdot (\mathbf{r} \pm \mathbf{r}')} (-S_{\mathbf{k}} C_{\mathbf{k}}), \qquad (6.10)$$

where $S_{\mathbf{k}} \equiv \sinh r_{\mathbf{k}}$ and $C_{\mathbf{k}} \equiv \cosh r_{\mathbf{k}}$, and $r_{\mathbf{k}}$, the squeezing parameter for the **k** mode, is a continuous function of **k**.

If we consider a simple function, e.g., were S_kC_k does not have any poles anywhere in the complex **k** plane, then the **k** integration in Eq. (6.10) vanishes unless $\mathbf{r}_1 = \mathbf{r}_2$, which leads to a single **r** summation

$$\langle \psi | \hat{A}^2 | \psi \rangle = \sum_{\mathbf{r}} \sum_{\mathbf{k}} (-S_{\mathbf{k}} C_{\mathbf{k}}) = -\frac{V}{(2\pi)^3} \int d^3 \mathbf{k} S_{\mathbf{k}} C_{\mathbf{k}},$$
(6.11)

where $\sum_{\mathbf{r}} = 1$ (see Sec. III) and $\sum_{\mathbf{k}} \rightarrow \frac{V}{(2\pi)^3} \int d^3 \mathbf{k}$ as usual [17]. $\langle \hat{A}^{\dagger} \hat{A} \rangle$ can be calculated similarly as

$$\langle \psi | \hat{A}^2 | \psi \rangle = \frac{V}{(2\pi)^3} \int d^3 \mathbf{k} \, S_{\mathbf{k}}^2, \qquad (6.12)$$

which gives a finite squeezing (reduction in noise)

$$\lambda_{\rm sm} = \frac{1}{2} + \langle \psi | \hat{A}^{\dagger} \hat{A} | \psi \rangle - |\langle \psi | \hat{A}^2 | \psi \rangle|$$
$$= \frac{1}{2} + \frac{V}{(2\pi)^3} \int d^3 \mathbf{k} \left(S_{\mathbf{k}}^2 - S_{\mathbf{k}} C_{\mathbf{k}} \right) \qquad (6.13)$$

for the wave packet. We note that $(S_k^2 - S_k C_k) < 0$ and we remind that $S_k \equiv \sinh r_k$ and $C_k \equiv \cosh r_k$.

2. Method (i-b): Entanglement of two constituent modes

We raise the following question. Does the entanglement between two constituent modes, letting them again be \mathbf{k}_1 and \mathbf{k}_2 , contribute to the nonclassicality of the wave packet?

We consider a state where there is no squeezing in the modes, but only the two modes \mathbf{k}_1 and \mathbf{k}_2 are entangled via a two-mode squeezing operator $\hat{\mathbf{E}} = e^{\beta \hat{a}_1^{\dagger} \hat{a}_2^{\dagger} - \beta^* \hat{a}_1 \hat{a}_2}$,

$$|\psi_{\text{ent}}\rangle = |\beta\rangle_{\mathbf{k}_1,\mathbf{k}_2} |0\rangle_{\mathbf{k}_3} |0\rangle_{\mathbf{k}_4} \cdots . \tag{6.14}$$

The reason we consider the entanglement due to the \hat{E} operator is that it creates "pure entanglement" between the k_1 and k_2 modes. That is, it does create single-mode nonclassicality in the modes (see Sec. II.5.(iii) in Ref. [53] and also Ref. [64]).

We can transform the $\hat{a}_i(\beta)$ operators as

$$\hat{a}_1(\beta) = C\hat{a}_1 + S\hat{a}_2^{\dagger}, \tag{6.15}$$

$$\hat{a}_2(\beta) = C\hat{a}_2 + S\hat{a}_1^{\dagger}, \qquad (6.16)$$

instead of working with the entangled state $|\beta\rangle_{\mathbf{k}_1,\mathbf{k}_2}$. Here, $C \equiv \cosh r$ and $S \equiv \sinh r$, where r determines the degree of the entanglement.

In this case, only the $\hat{a}_{\mathbf{k}_1}(\beta)\hat{a}_{\mathbf{k}_2}(\beta)$ and $\hat{a}_{\mathbf{k}_2}(\beta)\hat{a}_{\mathbf{k}_1}(\beta)$ terms contribute with *CS* in the calculation of $\langle \hat{A}^2 \rangle$ and only $\hat{a}^{\dagger}_{\mathbf{k}_{1,2}}(\beta)\hat{a}_{\mathbf{k}_{1,2}}(\beta)$ terms contribute with S^2 in the calculation of $\langle \hat{A}^{\dagger} \hat{A} \rangle$. Thus, we find

$$\langle \hat{A}^2 \rangle_\beta = 2 \frac{(\Delta r)^3}{V} CS, \qquad (6.17)$$

$$\langle \hat{A}^{\dagger} \hat{A} \rangle_{\beta} = 2 \frac{(\Delta r)^3}{V} S^2, \qquad (6.18)$$

which creates an infinitesimal squeezing in the wave packet as

$$\begin{split} \langle (\Delta \hat{X}_{\phi})^2 \rangle &= \lambda_{\rm sm} = \frac{1}{2} + \langle \hat{A}^{\dagger} \hat{A} \rangle - |\langle \hat{A}^2 \rangle| \\ &= \frac{1}{2} + 2 \frac{(\Delta r)^3}{V} (S^2 - SC), \end{split}$$
(6.19)

which is always less than the standard quantum limit 1/2. So, it creates a squeezed uncertainty wave packet.

B. Method (ii): Wave-packet nonclassicality via entanglement at a beam-splitter output

It is a known fact that the single-mode nonclassicality criterion $\langle \hat{a}^{\dagger} \hat{a} \rangle < |\langle \hat{a}^{2} \rangle|$, so $\langle \hat{A}^{\dagger} \hat{A} \rangle < |\langle \hat{A}^{2} \rangle|$, works well for quadrature-squeezed-like (and Gaussian-like) states. For more general states, such a nonclassicality criterion fails. In these cases, a BS can help us very much. When a nonclassical state is input to a BS, mixed with vacuum or a coherent state, it generates two-mode entanglement at the BS output. Hence, we can also decide that a wave packet is nonclassical if it produces wave-packet–wave-packet entanglement at the BS output. The BS transformation for a wave packet is given in Ref. [66].

It is well experienced that the SPH, two-mode entanglement, criterion [43] is able to reveal the two-mode entanglement in some states other than the Gaussian ones, e.g., some superpositions of two-mode Fock states. Hence, determining the wave-packet nonclassicality via BS provides us the advantage of being able to detect some of the non-Gaussian states, e.g., superposed number states, using the strength (enhanced generality) of the SPH criterion.⁵

For instance, use of a BS can resolve the singlemode nonclassicality of a superradiant-phase single-mode state (see Fig. 2(c) in Ref. [65]), whose nature is extremely different from the Gaussian-like states. It is a straightforward process to develop the same method (see Sec. II.b in Ref. [65]), with $\hat{a} \rightarrow \hat{A}$, also for wave-packet nonclassicality. Even though the SPH criterion [43] is a strong one which is able to determine also some of the other states, in Sec. III.3 of Ref. [53] we developed an SPH-like (strong, invariant) criterion for number-phase-squeezed-like states. This criterion is invariant under the rotations in the number-phase (n- Φ) plane. Although SPH is a strong criterion, it is defined with quadrature variables, while the other criterion is defined with \hat{n} and $\hat{\Phi}$ operators.

VII. SUMMARY

Developments in the current technology necessitate entanglement or nonclassicality criteria for broadband emitting sources, e.g., spasers [11,12,40]. Current mode-based criteria can still be used for the broadband states. However, they detect or measure the entanglement only associated with the two carrier frequencies. We introduce criteria and measures for the "total" entanglement of two wave packets. That is, the newly introduced criteria can measure the entanglement among all of the modes of the two wave packets. We also develop a "total" nonclassicality for a wave packet, which accounts for the nonclassicality of a wave packet both due to squeezing of the constituent modes and entanglement present among the constituent modes. In analogy with wave-packet-wavepacket entanglement and wave-packet nonclassicality, we also introduce criteria for ensemble-wave-packet entanglement. All the criteria and measures we introduce can also be used for measurements with near-field detectors [39].

The criteria we develop here can find applications in various media, for more productive utilization of broadband sources. Quantification of the continuous-variable entanglement between two counterpropagating plasmons, e.g., in nanosized interconnects for quantum computers utilizing our current infrastructures [29], or a collection of quantum emitters radiating into a plasmon mode [67], or the nonclassicality of a nanolaser emission [11,12], can employ the criteria developed here. Moreover, quantification or detection of continuous-variable entanglement between the center-ofmass coordinates of two matter waves, e.g., obtained via molecular dissociation methods [5], or the entanglement of a wave packet with a recoiled matter wave (motional degree of freedom), and nonclassicality of a matter wave itself, can employ the presented criteria.

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⁵The SPH criterion is a strong one since it is invariant under intramode rotations [53], i.e., $\hat{a}_{1,2} = e^{i\phi_{1,2}}\hat{a}_{1,2}$.

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