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1998 J. Phys. A: Math. Gen. 31 565

(http://iopscience.iop.org/0305-4470/31/2/016)

J. Phys. A: Math. Gen. **31** (1998) 565–583. Printed in the UK PII: S0305-4470(98)81215-8

Characterizations of classical and nonclassical states of quantized radiation

Arvind†‖, N Mukunda‡¶ and R Simon§

† Department of Physics, Indian Institute of Science, Bangalore-560012, India
‡ Center for Theoretical Studies and Department of Physics, Indian Institute of Science, Bangalore-560012, India
§ Institute of Mathematical Sciences, CIT Campus, Madras-600113, India

Received 22 January 1997, in final form 30 July 1997

Abstract. A new operator-based condition for distinguishing classical from nonclassical states of quantized radiation is developed. It exploits the fact that the normal ordering rule of correspondence to go from classical to quantum-dynamical variables does not in general maintain positivity. It is shown that the approach naturally leads to distinguishing several layers of increasing nonclassicality, with more layers as the number of modes increases. A generalization of the notion of sub-Poissonian statistics for two-mode radiation fields is achieved by analysing completely all correlations and fluctuations in quadratic combinations of mode annihilation and creation operators conserving the total photon number. This generalization is nontrivial and intrinsically two-mode as it goes beyond all possible single mode projections of the two-mode field. The nonclassicality of pair-coherent states, squeezed vacuum and squeezed thermal states is analysed and contrasted to one another, comparing the generalized sub-Poissonian statistics with extant signatures of nonclassical behaviour.

1. Introduction

Electromagnetic radiation is intrinsically quantum mechanical in nature. Nevertheless it has been found extremely fruitful, at both conceptual and practical levels, to designate certain states of quantized radiation as being essentially ‘classical’ and others as being ‘nonclassical’ [1]. It is the latter that show the specific quantum features of radiation most sharply. Some of the well known signs of nonclassicality in this context are quadrature squeezing [2], antibunching [3] and sub-Poissonian photon statistics [4]; none of which can be accounted for by a classical statistical ensemble of solutions of the classical Maxwell equations. Phase-space distribution functions such as the diagonal coherent state distribution function, the Wigner function etc are central to such a classification and recently, there have been several experiments to directly reconstruct such functions and thus locate nonclassicality through them [5].

The purposes of this paper are to present a new physically equivalent way of distinguishing classical from nonclassical states of radiation, dual to the customary definition and based on operator properties; to point out the existence of several levels of classical behaviour, with a structure that gets progressively more elaborate as the number of modes

‖ E-mail address: arvind@physics.iisc.ernet.in
¶ Also at: Jawaharlal Nehru Centre for Advanced Scientific Research, Jakkur-560064, India.

0305-4470/98/020565+19$19.50

increases; and finally to give a complete discussion of signatures of nonclassical photon statistics for two-mode fields, working at the level of fluctuations in photon numbers.

For single-mode fields, the nonclassical states further divide into strongly nonclassical and weakly nonclassical. The nonclassicality of strongly nonclassical states is already revealed via the expectation values of phase-insensitive operators while it is necessary to consider phase-sensitive operators to unearth the nonclassicality of weakly nonclassical states. The consequences of strong nonclassicality can be measured in a rather interesting way by mixing the signal with a local oscillator having random phase as has been done recently [6]. Generalizations of similar experimental schemes will be useful to study levels of nonclassicality for two-mode radiation and to measure the intrinsically two-mode sub-Poissonian statistics introduced by us.

The contents of this paper are arranged as follows. In section 2 we develop a criterion based on operator expectation values, to distinguish between classical and nonclassical states of radiation. The basic idea is that the normal ordering rule of correspondence between classical dynamical variables and quantum operators, while being linear and translating reality into hermiticity, does not respect positivity. If this potential nonpositivity does not show up in the expectation values of operators in a certain state, then that state is classical; otherwise it is nonclassical. Section 3 explores this new approach further and shows that, as the number of independent modes increases, the classification of quantum states gets progressively finer; several levels of nonclassicality emerge. This is shown in detail for one and two mode fields and the trend becomes clear. Section 4 analyses in complete detail the properties of two-mode photon-number fluctuations, stressing the freedom to choose any normalized linear combination of the originally given modes as a variable single mode. The well known Mandel parameter criterion [7] for sub-Poissonian statistics for a single-mode field is extended in full generality to a matrix inequality in the two-mode case. It is shown that certain consequences of this inequality transcend the set of all single-mode projections of it and are thus intrinsically two-mode in character. Explicit physically interesting examples of this situation are provided, and the well known pair-coherent states are also examined from this point of view. Section 5 presents some concluding remarks.

2. The distinction between classical and nonclassical states—an operator criterion

We deal for simplicity with states of a single-mode radiation field, though our arguments generalize immediately to any number of modes. The photon creation and annihilation operators $\hat{a}^\dagger$ and $\hat{a}$ obey the customary commutation relation

$$[\hat{a}, \hat{a}^\dagger] \equiv \hat{a}\hat{a}^\dagger - \hat{a}^\dagger\hat{a} = 1. \tag{2.1}$$

Coherent states $|z\rangle$ are right eigenstates of $\hat{a}$ with a (generally complex) eigenvalue z; they are related to the states $|n\rangle$ of definite photon number (eigenstates of $\hat{a}^\dagger\hat{a}$) in a well known way. A general (pure or mixed) state of the one-mode field is described by a corresponding normalized density matrix $\hat{\rho}$:

$$\hat{\rho}^\dagger = \hat{\rho} \geqslant 0 \qquad \mathrm{Tr}\,\hat{\rho} = 1. \tag{2.2}$$

It can be expanded in the so-called diagonal coherent state representation [8]:

$$\begin{aligned} \hat{\rho} &= \int \frac{\mathrm{d}^2 z}{\pi} \phi(z) |z\rangle\langle z| \\ \int \frac{\mathrm{d}^2 z}{\pi} \phi(z) &= 1. \end{aligned} \tag{2.3}$$

While hermiticity of $\hat{\rho}$ corresponds to reality of the weight function $\phi(z)$, the latter is in general a singular mathematical quantity, namely a distribution of a well-defined class.

The conventional designation of $\hat{\rho}$ as being classical or nonclassical is based on the properties of $\phi(z)$. Namely, $\hat{\rho}$ is said to be classical if $\phi(z)$ is everywhere non-negative and not more singular than a delta function [1]:

$$\begin{aligned} &\hat{\rho} \text{ classical} \Leftrightarrow \phi(z) \geqslant 0 \qquad \text{no worse than delta function} \\ &\hat{\rho} \text{ nonclassical} \Leftrightarrow \phi(z) \not\geqslant 0. \end{aligned} \tag{2.4}$$

It is clear that the conditions to be classical involve an infinite number of independent inequalities, since $\phi(z) \geqslant 0$ has to be obeyed at each point z in the complex plane. This is true despite the fact that the condition $\hat{\rho} \geqslant 0$ means that the 'values' of $\phi(z)$ at different points z are not quite 'independent'. One realizes this by recalling that every classical probability distribution over the complex plane is certainly a possible choice for $\phi(z)$, with the corresponding $\hat{\rho}$ being classical. When $\hat{\rho}$ is classical in the above sense, quantum expectation values acquire a classical statistical interpretation (see below).

The above familiar definition of classical states deals directly with $\hat{\rho}$ and $\phi(z)$. Now we develop an equivalent definition based on operators and their expectation values. As is well known, the representation (2.3) for $\hat{\rho}$ is closely allied to the normal ordering rule for passing from classical c-number dynamical variables to quantum operators. Within quantum mechanics we know that an operator $\hat{F}$ is completely and uniquely determined by its diagonal coherent-state matrix elements (expectation values) $\langle z|\hat{F}|z\rangle$. Moreover, the hermiticity of $\hat{F}$ and reality of $\langle z|\hat{F}|z\rangle$ are precisely equivalent. Any (real) classical function $f(z^*, z)$ determines uniquely, by the normal ordering rule of placing $\hat{a}^\dagger$ always to the left of $\hat{a}$ after substituting $z \to \hat{a}$ and $z^* \to \hat{a}^\dagger$, a corresponding (Hermitian) operator $\hat{F}_N$ as follows.

Normal ordering rule:

$$\begin{aligned} &f(z^*, z) \to \hat{F}_N \\ &\langle z|\hat{F}_N|z\rangle = f(z^*, z) \\ &f \text{ real} \Leftrightarrow \hat{F}_N \text{ Hermitian.} \end{aligned} \tag{2.5}$$

The connection with the representation (2.3) for $\hat{\rho}$ is given by

$$\mathrm{Tr}(\hat{\rho}\hat{F}_N) = \int \frac{\mathrm{d}^2 z}{\pi} \phi(z) f(z^*, z). \tag{2.6}$$

It is an important property of the normal ordering rule that, while it translates classical reality to quantum hermiticity, *it does not preserve positive semidefiniteness*. More explicitly, while by equation (2.5) $\hat{F}_N \geqslant 0$ implies $f(z^*, z) \geqslant 0$, *the converse is not true*. Here are some simple examples of non-negative classical real $f(z^*, z)$ leading to indefinite Hermitian $\hat{F}_N$:

$$\begin{aligned} &f(z^*, z) = (z^* + z)^2 \longrightarrow \hat{F}_N = (\hat{a}^\dagger + \hat{a})^2 - 1 \\ &f(z^*, z) = (z^* + z)^4 \longrightarrow \hat{F}_N = ((\hat{a}^\dagger + \hat{a})^2 - 3)^2 - 6 \\ &f(z^*, z) = \mathrm{e}^{-z^* z} \sum_{n=0}^{\infty} \frac{C_n}{n!} z^{*n} z^n \longrightarrow \hat{F}_N = \sum_{n=0}^{\infty} C_n |n\rangle\langle n|. \end{aligned} \tag{2.7}$$

In the last example, the real constants C_n can certainly be chosen so that some of them are negative while maintaining $f(z^*, z) \geqslant 0$; this results in $\hat{F}_N$ being indefinite.

We thus see that when the normal ordering rule is used, every $\hat{F}_N \geqslant 0$ arises from a unique $f(z^*, z) \geqslant 0$, but some (real) $f(z^*, z) \geqslant 0$ lead to (Hermitian) indefinite $\hat{F}_N$.

So in a given quantum state $\hat{\rho}$, the operator $\hat{F}_N$ corresponding to a non-negative real classical $f(z^*, z)$ could well have a negative expectation value. *If this never happens, then $\hat{\rho}$ is classical*. That is, as we see upon combining equations (2.4) and (2.6): if for every $f(z^*, z) \geqslant 0$ the corresponding $\hat{F}_N$ has a non-negative expectation value even though $\hat{F}_N$ may be indefinite, then $\hat{\rho}$ is classical. Conversely, $\hat{\rho}$ is nonclassical if there is at least one $f(z^*, z) \geqslant 0$ which leads to an indefinite $\hat{F}_N$ whose expectation value is negative.

We can also convey the content of this dual-operator way of defining classical states as follows: while the normal ordering rule allows for the appearance of 'negativity' in an operator $\hat{F}_N$ even when none is present in the corresponding classical $f(z^*, z)$, in a classical state such negativity never shows up in expectation values.

It is well to re-emphasize at this point that the above distinction between 'classical' and 'nonclassical' states is based on a convention within the quantum theory of radiation. The physical content of the convention is that in a 'classical state' all *quantum expectation values* of normal ordered operators can be equally well regarded as arising from a truly classical statistical ensemble with a corresponding bonafide probability distribution at the amplitude level.

Since it is positivity that may be lost when we use the normal ordering rule to pass from classical to quantum variables, it is of interest to ask what happens when other rules of correspondence are used. Two familiar alternative rules are antinormal ordering ($\hat{a}$ to the left and $\hat{a}^\dagger$ to the right) [9] and Weyl ordering ($\hat{q}$ and $\hat{p}$ treated symmetrically) [10, 11]; in fact in an algebraic sense we may say that the latter stands midway between the other two. With antinormal ordering it turns out that classical positivity certainly implies quantum positivity but not conversely. For example, by this rule we find:

$$f(z^*, z) = (z^* + z)^2 - 1 \longrightarrow \hat{F}_A = (\hat{a}^\dagger + \hat{a})^2. \tag{2.8}$$

With the Weyl rule, positivity can fail in both directions, as shown by these examples:

$$\begin{aligned} &f(q, p) = \delta(q)\delta(p) \longrightarrow \hat{F}_W = \text{parity operator} \\ &\hat{F}_W = |1\rangle\langle 1| \longrightarrow f(q, p) = \frac{2}{\pi}\left(q^2 + p^2 - \frac{1}{2}\right)\exp(-q^2 - p^2). \end{aligned} \tag{2.9}$$

While these remarks illuminate in terms of operator properties the relations among the three ordering rules, the classification of states $\hat{\rho}$ into classical and nonclassical ones is based most simply on the normal ordering rule. It is clear that all these considerations extend easily to any number of modes of radiation.

All the familiar criteria of nonclassicality can be cast in our operator-based approach in a rather straightforward way. We consider here two examples, namely, sub-Poissonian statistics and quadrature squeezing which involve fluctuations in photon number and quadrature components respectively. At a first glance, they seem to involve more than one operator in a nonlinear way and thus are not directly analysable by the operator-based dual method of defining nonclassicality. However, it turns out that we can incorporate these notions in our formalism quite easily. A single-mode state exhibits sub-Poissonian photon statistics if the Mandel parameter

$$Q = \frac{\langle \hat{a}^{\dagger 2}\hat{a}^2\rangle - \langle \hat{a}^\dagger \hat{a}\rangle^2}{\langle \hat{a}^\dagger \hat{a}\rangle} \tag{2.10}$$

(which is the same as $\langle \hat{a}^{\dagger 2}\hat{a}^2\rangle - \langle \hat{a}^\dagger \hat{a}\rangle^2$, except for a positive denominator) is negative for that state. In order to connect this notion with our approach, consider the non-negative classical function

$$f_Q(z, z^*) = (|z|^2 - c^2)^2 \tag{2.11}$$

where c is a real parameter. The operator corresponding to this function, by the normal ordering rule, is

$$\hat{F}_Q(a^\dagger, a) = \hat{a}^{\dagger 2}\hat{a}^2 - 2\hat{a}^\dagger\hat{a}c^2 + c^4 \tag{2.12}$$

which is indefinite for all values of c. Therefore, if the expectation value of this operator in a given state is negative for some value of c, the state is nonclassical. To further strengthen the potential of this function to explore nonclassicality, we minimize its expectation value with respect to c. The value of c for which the minimum occurs is state dependent and can be written in a universal form:

$$c \text{ (for which } \langle \hat{F}_Q(a^\dagger, a)\rangle \text{ is minimum)} = \langle \hat{a}^\dagger\hat{a}\rangle \tag{2.13}$$

(the expectation here is calculated for the state of interest). If we now set c to this value the expectation value of the operator in the above analysis becomes identical to Mandel's Q parameter for sub-Poissonian statistics.

The analysis for quadrature squeezing is very similar; the classical positive function is now

$$f_{\text{sq}}(z, z^*) = (z^*\mathrm{e}^{\mathrm{i}\varphi} + z\mathrm{e}^{-\mathrm{i}\varphi} - c)^2 \tag{2.14}$$

with the corresponding indefinite operator being

$$\hat{F}_{\text{sq}}(a^\dagger, a) = (\hat{a}^\dagger\mathrm{e}^{\mathrm{i}\varphi} + \hat{a}\mathrm{e}^{-\mathrm{i}\varphi})^2 - 2(\hat{a}^\dagger\mathrm{e}^{\mathrm{i}\varphi} + \hat{a}\mathrm{e}^{-\mathrm{i}\varphi})c + c^2 - 1. \tag{2.15}$$

The negativity of the expectation value for this operator becomes identical to the squeezing of the quadrature component $\frac{1}{\sqrt{2}}(\hat{a}^\dagger\mathrm{e}^{\mathrm{i}\varphi} + \hat{a}\mathrm{e}^{-\mathrm{i}\varphi})$ when we set $c = \langle \hat{a}^\dagger\mathrm{e}^{\mathrm{i}\varphi} + \hat{a}\mathrm{e}^{-\mathrm{i}\varphi}\rangle$ (with the expectation calculated for the state of interest). The appearance of φ indicates the phase sensitive nature of quadrature squeezing as opposed to that of sub-Poissonian statistics. By choosing different values of φ we can analyse different quadrature components. Similar analysis can also be carried out for other criteria of nonclassicality such as higher-order squeezing [12] and the one based on matrices constructed out of factorial moments of the photon number distribution [13].

3. Levels of classicality

3.1. The single-mode case

We begin again with the single-mode situation and hereafter deal exclusively with the normal ordering prescription. (Therefore the subscript N on $\hat{F}_N$ will be omitted). Suppose we limit ourselves to classical functions $f(z^*, z)$ which are real, non-negative and phase invariant, that is, invariant under $z \longrightarrow \mathrm{e}^{\mathrm{i}\alpha}z$. An independent and complete set of these can be taken to be

$$f_n(z^*, z) = \mathrm{e}^{-z^*z}z^{*n}z^n/n! \qquad n = 0, 1, 2, \ldots \tag{3.1}$$

since they map conveniently to the number state projection operators:

$$f_n(z^*, z) \longrightarrow \hat{F}^{(n)} = |n\rangle\langle n| \qquad n = 0, 1, 2, \ldots. \tag{3.2}$$

A general real linear combination $f(z^*, z) = \sum_n C_n f_n(z^*, z)$, even if non-negative, may lead to an indefinite $\hat{F}$, as seen at equation (2.7).

If we are interested only in the expectation values of such variables, we are concerned only with the probabilities $p(n)$ for finding various numbers of photons; for this purpose an angular average of $\phi(z)$ is all that is required:

$$\begin{aligned} p(n) &= \langle n|\hat{\rho}|n\rangle = \mathrm{Tr}(\hat{\rho}|n\rangle\langle n|) \\ &= \int \frac{\mathrm{d}^2 z}{\pi}\phi(z)\mathrm{e}^{-z^* z} z^{*n} z^n/n! \\ &= \int_0^\infty \mathrm{d}I\, P(I)\mathrm{e}^{-I} I^n/n! \\ P(I) &= \frac{1}{2\pi}\int_0^{2\pi} \mathrm{d}\theta\, \phi(I^{1/2}\mathrm{e}^{\mathrm{i}\theta}). \end{aligned} \tag{3.3}$$

Now while $\phi(z) \geqslant 0$ certainly implies $P(I) \geqslant 0$, the converse is not true. Thus one is led to a three-fold classification of quantum states $\hat{\rho}$ [14]:

$$\begin{aligned} &\hat{\rho}\ \text{classical} \Longleftrightarrow \phi(z) \geqslant 0 \qquad \text{hence } P(I) \geqslant 0 \\ &\hat{\rho}\ \text{weakly nonclassical} \Longleftrightarrow P(I) \geqslant 0 \qquad \text{but } \phi(z) \ngeqslant 0 \\ &\hat{\rho}\ \text{strongly nonclassical} \Longleftrightarrow P(I) \ngeqslant 0 \qquad \text{so necessarily } \phi(z) \ngeqslant 0. \end{aligned} \tag{3.4}$$

The previous definition (2.4) of nonclassical $\hat{\rho}$ based on $\phi(z)$ alone is now refined to yield two subsets of states, the weakly nonclassical and the strongly nonclassical. The former states do have the following property:

$$\hat{\rho}\ \text{weakly nonclassical} \Longrightarrow \mathrm{Tr}(\hat{\rho}\hat{F}) \geqslant 0 \text{ if } f(z^*, z) = \sum_{n=0}^{\infty} C_n f_n(z^*, z) \geqslant 0. \tag{3.5}$$

However, in addition, there would definitely be some phase noninvariant $f(z^*, z) \geqslant 0$ for which $\hat{F}$ is indefinite and $\mathrm{Tr}(\hat{\rho}\hat{F}) < 0$. It is just that this extent of nonclassicality in $\hat{\rho}$ is not revealed by the expectation values of phase invariant variables, or at the level of the probabilities $p(n)$†.

We may stress that this three-fold classification is again based on a physically motivated convention within quantum theory. In general, $\mathcal{P}(I)$ could be regarded as a candidate for the probability distribution for the intensity variable, with its moments (along with the exponential factor e^{-I}) yielding the true probabilities $p(n)$. If a quantum state $\hat{\rho}$ is weakly nonclassical, it means that as far as the photon number probabilities $p(n)$ are concerned, a truly classical statistical ensemble at the intensity level is available to reproduce these $p(n)$ but this is not true at the deeper amplitude level. If $\hat{\rho}$ is strongly nonclassical, then even for the limited information contained in the $p(n)$, a classical ensemble description is impossible since $\mathcal{P}(I)$ is not non-negative. We may also clarify that this discussion deals exclusively with the search for possible classical statistical ensemble descriptions at various levels and not at all with the semiclassical approach to quantum theory in the limit that Planck's constant 'tends to zero'.

It is clear that the classification (3.4) is $U(1)$ or phase invariant. That is, $\hat{\rho}$ retains its classical, weakly nonclassical or strongly nonclassical character under the transformation $\phi(z) \longrightarrow \phi'(z) = \phi(z\mathrm{e}^{\mathrm{i}\alpha})$.

As examples of interesting inequalities obeyed if $\hat{\rho}$ is either classical or weakly nonclassical, we may quote the following involving the factorial moments of the photon

† See Klauder and Sudarshan [8].

number probabilities $p(n)$:

$$\begin{aligned}\gamma_m &= \mathrm{Tr}(\hat{\rho}\hat{a}^{\dagger m}\hat{a}^m)\\ &= \int \frac{\mathrm{d}^2 z}{\pi}\phi(z)(z^* z)^m\\ &= \int_0^\infty \mathrm{d}I\ P(I)I^m \\ &= \sum_{n=m}^{\infty} p(n)n!/(n-m)! \geqslant 0 \qquad m = 0, 1, 2, \ldots\end{aligned} \tag{3.6}$$

$\hat{\rho}$ classical or weakly nonclassical $\Longleftrightarrow P(I) \geqslant 0 \Longrightarrow \gamma_m\gamma_n \leqslant \gamma_{m+n} \leqslant \sqrt{\gamma_{2m}\gamma_{2n}}$.

Violation of any one of these inequalities implies $\hat{\rho}$ is strongly nonclassical.

The inequalities quoted in equation (3.6) above clearly involve an infinite subset of the photon number probabilities $p(n)$. However one can easily construct far simpler inequalities involving a small number of the $p(n)$, violation of any of which also implies that $\hat{\rho}$ is strongly nonclassical. For example, from equations (3.1) and (3.2), for any non-negative integer n_0 and any real a, b we have the correspondence

$$\begin{aligned}&f(z^*, z) = \mathrm{e}^{-z^* z}\frac{(z^* z)^{n_0}}{n_0!}(a + bzz^*)^2 \rightarrow\\ &\hat{F} = a^2|n_0\rangle\langle n_0| + 2(n_0+1)ab|n_0+1\rangle\langle n_0+1| + (n_0+1)(n_0+2)b^2|n_0+2\rangle\langle n_0+2|.\end{aligned} \tag{3.7}$$

Here $f(z^*, z)$ is non-negative while $\hat{F}$ is indefinite if $ab < 0$. We then have the result:

$$\begin{aligned}&\hat{\rho} \text{ classical or weakly nonclassical} \Leftrightarrow P(I) \geqslant 0 \Rightarrow\\ &a^2 p(n_0) + 2(n_0+1)abp(n_0+1) + (n_0+1)(n_0+2)b^2 p(n_0+2)\\ &\qquad = \frac{1}{n_0!}\int_0^\infty \mathrm{d}I\ P(I)\mathrm{e}^{-I}I^{n_0}(a+bI)^2 \geqslant 0\\ &\text{i.e. } p(n_0+1)^2 \leqslant \left(\frac{n_0+2}{n_0+1}\right)p(n_0)p(n_0+2).\end{aligned} \tag{3.8}$$

So again, violation of any of these 'local' inequalities in $p(n)$ implies that $\hat{\rho}$ is strongly nonclassical.

A physically illuminating example of the distinction between classical and weakly nonclassical $\hat{\rho}$, and passage from one to the other, is provided by the case of the Kerr medium. The argument is intricate and rests on two well known results. The first is Hudson's theorem [15]: if a (pure-state) wavefunction $\psi_0(q)$ has a non-negative Wigner function $W_0(q, p)$, then $\psi_0(q)$ is Gaussian and conversely; in that case $W_0(q, p)$ is also Gaussian. The second result is the general connection between $\phi(z)$ and $W(q, p)$ for any $\hat{\rho}$:

$$W(q, p) = 2\int \frac{\mathrm{d}^2 z'}{\pi}\mathrm{e}^{-2|z-z'|^2}\phi(z') \qquad z = \frac{1}{\sqrt{2}}(q + \mathrm{i}p). \tag{3.9}$$

This means that for classical $\hat{\rho}$ with $\phi(z) \geqslant 0$, $W(q, p) \geqslant 0$ as well. Now imagine a single mode radiation field in an initial coherent state $|z_0\rangle$ with $\phi_0(z) = \pi\delta^{(2)}(z - z_0)$, incident upon a Kerr medium [16]. This initial state is pure, classical and has a Gaussian wavefunction $\psi_0(q)$. The Kerr-medium Hamiltonian is of the form

$$H_{\mathrm{Kerr}} = \alpha\hat{a}^\dagger\hat{a} + \beta(\hat{a}^\dagger\hat{a})^2. \tag{3.10}$$

Clearly the number states $|n\rangle$ are eigenstates of this Hamiltonian. Therefore the Poissonian photon number distribution

$$p(n) = e^{-I_0} I_0^n / n! \qquad I_0 = z_0^* z_0 \tag{3.11}$$

of the input state $|z_0\rangle$ is preserved under passage through the Kerr medium. Likewise the function $P(I) = \delta(I - I_0)$ is left unaltered. Therefore the output state $|\psi\rangle$, which of course is pure, is either classical or weakly nonclassical. However, the form of H_{Kerr} shows that the output wavefunction is non-Gaussian. Therefore by Hudson's theorem the corresponding $W(q, p)$ must become negative somewhere. Therefore by equation (3.9) the output $\phi(z)$ cannot be non-negative. Thus passage through the Kerr medium converts an incident coherent state, which is classical, into a final state which is weakly nonclassical.

3.2. *The two-mode case*

Now we sketch the extension of these ideas to the two-mode case. Here the operator commutation relations, number and coherent states and the diagonal representation for $\hat{\rho}$, are straightforward generalizations of the relations in the single-mode case, and of equation (2.3):

$$\begin{aligned} \hat{\rho} &= \int \mathrm{d}\mu\,(\underline{z})\phi(\underline{z})|\underline{z}\rangle\langle\underline{z}| \\ \mathrm{d}\mu\,(\underline{z}) &= \mathrm{d}^2 z_1\, \mathrm{d}^2 z_2/\pi^2. \end{aligned} \tag{3.12}$$

The symbol $\underline{z}$ denotes a pair of complex numbers (z_1, z_2).

It is convenient at this point to go when necessary beyond purely real classical functions $f(\underline{z})$ in applying the normal ordering rule to obtain corresponding operators. From the general number states matrix elements of $\hat{\rho}$ we read off some operator correspondences generalizing equations (3.1) and (3.2):

$$\begin{aligned} \langle n_3, n_4|\hat{\rho}|n_1, n_2\rangle &= \mathrm{Tr}(\hat{\rho}|n_1, n_2\rangle\langle n_3, n_4|) \\ &= \int \mathrm{d}\mu\,(\underline{z})\phi(\underline{z})\mathrm{e}^{-\underline{z}^\dagger\underline{z}} \frac{z_1^{*n_1} z_2^{*n_2} z_1^{n_3} z_2^{n_4}}{\sqrt{n_1! n_2! n_3! n_4!}} \Longrightarrow \\ \mathrm{e}^{-\underline{z}^\dagger\underline{z}} \frac{z_1^{*n_1} z_2^{*n_2} z_1^{n_3} z_2^{n_4}}{\sqrt{n_1! n_2! n_3! n_4!}} &\longrightarrow |n_1, n_2\rangle\langle n_3, n_4|. \end{aligned} \tag{3.13}$$

For one mode the phase transformations form the group $U(1)$. For two modes this generalizes to the group $U(2)$ of (passive) transformations mixing the two orthonormal single photon modes. A general element $u = (u_{rs}) \in U(2)$ can be obtained as a phase factor $\mathrm{e}^{\mathrm{i}\alpha} \in U(1)$ times an element $a \in SU(2), u = \mathrm{e}^{\mathrm{i}\alpha} a$. At the operator level the unitary $U(2)$ action on the two-mode Hilbert space is generated by the well known Schwinger construction of angular momentum,

$$\underline{J} = \tfrac{1}{2}\underline{\hat{a}}^\dagger \underline{\sigma}\,\underline{\hat{a}} \tag{3.14}$$

where $\underline{\sigma}$ are the Pauli matrices and $\underline{\hat{a}} = (\hat{a}_1, \hat{a}_2)^T$ are the two annihilation operators, and the total number operator $\underline{\hat{a}}^\dagger\underline{\hat{a}}$. Namely, the unitary operator $\mathcal{U}(a)$ on the Hilbert space, for $a \in SU(2)$, is obtained as a suitable exponential of the $\underline{J}$'s; while the $U(1)$ generator is the total number operator. Actually, $\mathcal{U}(u)$ for $u \in U(2)$ can also be easily defined by its action on the coherent states [17],

$$\mathcal{U}(u)|\underline{z}\rangle = |u^*\underline{z}\rangle. \tag{3.15}$$

For our later purposes it is also useful to have the effect of these unitary operators on $\underline{\hat{a}}, \underline{\hat{a}}^\dagger$ and monomials in them:

$$\begin{aligned}
&\mathcal{U}(u)\hat{a}_r\mathcal{U}(u)^{-1} = \sum_{s=1}^{2} u_{sr}\hat{a}_s \\
&\mathcal{U}(u)\hat{a}_r^\dagger\mathcal{U}(u)^{-1} = \sum_{s=1}^{2} u_{sr}^*\hat{a}_s^\dagger \\
&\mathcal{U}(u)\frac{\hat{a}_1^{\dagger j+m}\hat{a}_2^{\dagger j-m}}{\sqrt{(j+m)!(j-m)!}}\mathcal{U}(u)^{-1} = \mathrm{e}^{-2\mathrm{i}\alpha j}\sum_{m'} D^{(j)}_{m'm}(a)\frac{\hat{a}_1^{\dagger j+m'}\hat{a}_2^{\dagger j-m'}}{\sqrt{(j+m')!(j-m')!}} \\
&\mathcal{U}(u)\frac{\hat{a}_1^{j+m}\hat{a}_2^{j-m}}{\sqrt{(j+m)!(j-m)!}}\mathcal{U}(u)^{-1} = \mathrm{e}^{2\mathrm{i}\alpha j}\sum_{m'} D^{(j)}_{m'm}(a)^*\frac{\hat{a}_1^{j+m'}\hat{a}_2^{j-m'}}{\sqrt{(j+m')!(j-m')!}} \\
&j = 0, \tfrac{1}{2}, 1, \ldots \\
&m, m' = j, j-1, \ldots, -j.
\end{aligned} \tag{3.16}$$

We have chosen the exponents of $\hat{a}$'s and $\hat{a}^\dagger$'s and numerical factors in such a way that the results can be expressed via the $SU(2)$ irreducible representation matrices, namely the $\mathcal{D}$-functions of quantum angular-momentum theory [18]. As is also well known, the states with a fixed total number of photons, say $2j$, transform under $\mathcal{U}(u)$ via the matrices $\mathcal{D}^{(j)}(a)$.

To motivate the existence of several layers of classicality, we now generalize the single mode $U(1)$-invariant real factorial moments γ_n of equation (3.6) to two-mode quantities which conserve total photon number and also transform in a closed and covariant manner under $SU(2)$. For this purpose, keeping in mind equations (3.16), it is convenient to start with the (in general complex) classical monomials

$$\begin{aligned}
&f^j_{m_1m_2}(\underline{z}^\dagger, \underline{z}) = N_{jm_1m_2} z_1^{*j+m_1} z_2^{*j-m_1} z_1^{j+m_2} z_2^{j-m_2} \\
&N_{jm_1m_2} = [(j+m_1)!(j-m_1)!(j+m_2)!(j-m_2)!]^{-1/2} \\
&j = 0, \tfrac{1}{2}, 1, \ldots \\
&m_1, m_2 = j, j-1, \ldots, -j.
\end{aligned} \tag{3.17}$$

The total power of $\underline{z}$ is equal to that of $\underline{z}^\dagger$, hence these are $U(1)$-invariant. The corresponding operators and their $SU(2)$ transformation laws are, using equation (3.16):

$$\begin{aligned}
&f^j_{m_1m_2}(\underline{z}^\dagger, \underline{z}) \to \hat{F}^j_{m_1m_2} = N_{jm_1m_2}\hat{a}_1^{\dagger j+m_1}\hat{a}_2^{\dagger j-m_1}\hat{a}_1^{j+m_2}\hat{a}_2^{j-m_2} \\
&a \in SU(2) : \mathcal{U}(a)\hat{F}^j_{m_1m_2}\mathcal{U}(a)^{-1} = \sum_{m_1', m_2'} D^{(j)}_{m_1'm_1}(a) D^{(j)}_{m_2'm_2}(a)^* \hat{F}^j_{m_1'm_2'}.
\end{aligned} \tag{3.18}$$

For a given two-mode state $\hat{\rho}$ we now generalize the factorial moments γ_n of equation (3.6) to the following three-index quantities:

$$\begin{aligned}
\gamma^{(j)}_{m_2m_1} &= \mathrm{Tr}(\hat{\rho}\hat{F}^j_{m_1m_2}) \\
&= N_{jm_1m_2}\,\mathrm{Tr}(\hat{\rho}\hat{a}_1^{\dagger j+m_1}\hat{a}_2^{\dagger j-m_1}\hat{a}_1^{j+m_2}\hat{a}_2^{j-m_2}).
\end{aligned} \tag{3.19}$$

Their $SU(2)$ transformation law is clearly

$$\begin{aligned}
&\hat{\rho}' = \mathcal{U}(a)\hat{\rho}\mathcal{U}(a)^{-1} : \\
&\gamma'^{(j)}_{m_2m_1} = \sum_{m_1', m_2'} D^{(j)}_{m_1'm_1}(a^{-1}) D^{(j)}_{m_2'm_2}(a^{-1})^* \gamma^{(j)}_{m_2'm_1'} \\
&\text{i.e. } \gamma'^{(j)} = D^{(j)}(a)\gamma^{(j)}D^{(j)}(a)^\dagger.
\end{aligned} \tag{3.20}$$

In the last line for each fixed j the generalized moments $\gamma^{(j)}_{m_1m_2}$ have been regarded as a (Hermitian) matrix of dimension $(2j+1)$.

On account of the fact that the total photon number is conserved in the definition of these moments, calculation of $\gamma^{(j)}_{m_1m_2}$ does not require complete knowledge of $\phi(\underline{z})$ but only of a partly angle-averaged quantity $\mathcal{P}(I_1, I_2, \theta)$:

$$\begin{aligned}\gamma^{(j)}_{m_1m_2} &= N_{jm_1m_2}\int_0^\infty \mathrm{d}I_1\int_0^\infty \mathrm{d}I_2\int_0^{2\pi}\frac{\mathrm{d}\theta}{2\pi}\mathcal{P}(I_1, I_2, \theta)(I_1I_2)^j \\ &\quad\times(I_1/I_2)^{1/2(m_2+m_1)}\mathrm{e}^{\mathrm{i}(m_1-m_2)\theta} \\ \mathcal{P}(I_1, I_2, \theta) &= \int_0^{2\pi}\frac{\mathrm{d}\theta_1}{2\pi}\phi(I_1^{1/2}\mathrm{e}^{\mathrm{i}\theta_1}, I_2^{1/2}\mathrm{e}^{\mathrm{i}(\theta_1+\theta)}).\end{aligned}\tag{3.21}$$

It is clear that these moments $\gamma^{(j)}_{m_1m_2}$ involve more than just the photon number probabilities $p(n_1, n_2)$ which are just the 'diagonal' case of the general matrix element in equation (3.13):

$$\begin{aligned}p(n_1, n_2) &= \langle n_1, n_2|\hat{\rho}|n_1, n_2\rangle \\ &= \int_0^\infty \mathrm{d}I_1\int_0^\infty \mathrm{d}I_2\, P(I_1, I_2)\mathrm{e}^{-I_1-I_2}I_1^{n_1}I_2^{n_2}/n_1!n_2! \\ P(I_1, I_2) &= \int_0^{2\pi}\frac{\mathrm{d}\theta}{2\pi}\mathcal{P}(I_1, I_2, \theta).\end{aligned}\tag{3.22}$$

This is the two-mode version of equation (3.3). The subset of 'diagonal' moments $\gamma^{(j)}_{mm}$ are calculable in terms of $p(n_1, n_2)$ or $P(I_1, I_2)$:

$$\begin{aligned}\gamma^{(j)}_{mm} &= \int_0^\infty \mathrm{d}I_1\int_0^\infty \mathrm{d}I_2\, P(I_1, I_2)I_1^{j+m}I_2^{j-m}/(j+m)!(j-m)! \\ &= \sum_{n_1,n_2} p(n_1, n_2)n_1!n_2!/(n_1-j-m)!(n_2-j+m)!(j+m)!(j-m)!.\end{aligned}\tag{3.23}$$

However, under a general $SU(2)$ mixing of the modes, the expressions $\gamma^{(j)}_{mm}$, $p(n_1, n_2)$, $P(I_1, I_2)$ do not transform in any neat way among themselves and one is obliged to enlarge the set to include the more general $\gamma^{(j)}_{m_1m_2}$ and $\mathcal{P}(I_1, I_2, \theta)$. (In particular, for these, the probabilities $p(n_1, n_2)$ are inadequate.) When this is done we see the need to deal with both the quantities $\mathcal{P}(I_1, I_2, \theta)$, $P(I_1, I_2)$ derived from $\phi(\underline{z})$ by a single or a double angular average. One can therefore distinguish four levels of classicality for two-mode states:

$$\begin{aligned}&\hat{\rho}\text{ classical} \Leftrightarrow \phi(\underline{z}) \geqslant 0 \qquad (\text{hence } \mathcal{P}(I_1, I_2, \theta), P(I_1, I_2) \geqslant 0) \\ &\hat{\rho}\text{ weakly nonclassical } I \Leftrightarrow \mathcal{P}(I_1, I_2, \theta) \geqslant 0 \qquad (\text{hence } P(I_1, I_2) \geqslant 0) \\ &\qquad\qquad \text{but } \phi(\underline{z}) \ngeqslant 0 \\ &\hat{\rho}\text{ weakly nonclassical } II \Leftrightarrow P(I_1, I_2) \geqslant 0 \qquad \text{but } \mathcal{P}(I_1, I_2, \theta) \ngeqslant 0 \\ &\qquad\qquad (\text{hence } \phi(z) \ngeqslant 0) \\ &\hat{\rho}\text{ strongly nonclassical } \Leftrightarrow P(I_1, I_2) \ngeqslant 0 \qquad (\text{hence } \phi(\underline{z}),\ \mathcal{P}(I_1, I_2, \theta) \ngeqslant 0).\end{aligned}\tag{3.24}$$

These definitions can be cast in dual operator forms. For example, for weakly nonclassical-I states, we can say that for any classical real non-negative overall $U(1)$ phase invariant $f(\underline{z}^\dagger, \underline{z})$ the corresponding operator $\hat{F}$ has a non-negative expectation value, while this fails for some $f(\underline{z}^\dagger, \underline{z})$ outside this class. In the weakly nonclassical-II case, we have to further limit $f(\underline{z}^\dagger, \underline{z})$ to be real non-negative and invariant under independent $U(1)\times U(1)$ phase transformations in the two modes, to be sure that the expectation value of $\hat{F}$ is non-negative.

It should be clear that the underlying motivations for these definitions are similar in spirit to the single mode case. As before, the main idea is to see to what extent quantum expectation values can be mimicked by truly classical statistical ensembles. In this sense, we see clearly that in the weakly nonclassical-I case there are more observables whose expectation values are reproducible on a classical statistical basis, than in the weakly nonclassical-II case. The general idea then is to seek, for a given quantum state, all those observables whose expectation values are reproducible on a classical basis, and separate them from those for which this is not possible.

At this point we can see that these levels of classicality possess different covariance groups. Since under a general $U(2)$ transformation $\mathcal{U}(u), u \in U(2)$, the function $\phi(\underset{\sim}{z})$ undergoes a point transformation, $\phi(\underset{\sim}{z}) \rightarrow \phi'(\underset{\sim}{z}) = \phi(u^T \underset{\sim}{z})$, we see that the property of being classical is preserved by all $U(2)$ transformations. On the other hand, a general $U(2)$ transformation can cause transitions among the other three levels. The point transformation property is obtained for $\mathcal{P}(I_1, I_2, \theta)$ and $P(I_1, I_2)$ only under the diagonal $U(1) \times U(1)$ subgroup of $U(2)$; in fact $P(I_1, I_2)$ is invariant under $U(1) \times U(1)$, while $\mathcal{P}(I_1, I_2, \theta)$ suffers a shift in the angle argument θ. Thus one can see that each of the three properties of being weakly nonclassical-I, weakly nonclassical-II or strongly nonclassical is only $U(1) \times U(1)$ invariant.

As the number of modes increases further, clearly the hierarchy of levels of classicality also increases.

Generalizing inequalities of the form (3.6) for the diagonal quantities $\gamma_{mm}^{(j)}$, for $\hat{\rho}$ classical or weakly nonclassical-I or weakly nonclassical-II, is quite straightforward, since then we deal with the two modes separately. The more interesting, and quite nontrivial, problem is to look for matrix generalizations of equation (3.6), bringing in the entire matrices $\gamma^{(j)} = (\gamma_{m_1 m_2}^{(j)})$, and looking for inequalities valid for states of the classical or weakly nonclassical-I types. (Of course for any quantum state $\hat{\rho}$ we have the obvious property that $\gamma^{(j)}$, for each j, is Hermitian positive semidefinite. This is the two-mode generalization of $\gamma_n \geqslant 0$ in the one-mode case.) However, this is expected to involve use of the Racah–Wigner calculus for coupling of tensor operators, familiar from quantum angular-momentum theory, inequalities for reduced matrix elements, etc [19].

In the next section we undertake a study of the particular case $j = 1$ which involves at most quartic expressions in $\hat{a}$'s and $\hat{a}^\dagger$'s. This is just what is involved in giving a complete account of the two-mode generalization of the Mandel Q-parameter familiar in the single-mode case.

4. Generalized photon-number fluctuation matrix for two-mode fields

For the one-mode case, with the single photon number operator $\hat{N} = \hat{a}^\dagger \hat{a}$, we have some obvious inequalities valid in all quantum states and others valid in weakly nonclassical and classical states as defined in equation (3.4):

any state:

$$\langle \hat{N} \rangle \equiv \mathrm{Tr}(\hat{\rho}\hat{N}) \equiv \gamma_1 \geqslant 0 \qquad (4.1a)$$

$$\langle : \hat{N}^2 : \rangle \equiv \mathrm{Tr}(\hat{\rho}\hat{a}^{\dagger 2}\hat{a}^2) \equiv \gamma_2 \geqslant 0 \qquad (4.1b)$$

$$\langle \hat{N}^2 \rangle \equiv \mathrm{Tr}(\hat{\rho}\hat{a}^\dagger\hat{a}\hat{a}^\dagger\hat{a}) \equiv \gamma_2 + \gamma_1 \geqslant 0 \qquad (4.1c)$$

$$(\Delta N)^2 \equiv \langle \hat{N}^2 \rangle - \langle \hat{N} \rangle^2 \equiv \langle (\hat{N} - \langle \hat{N} \rangle)^2 \rangle \equiv \gamma_2 + \gamma_1 - \gamma_1^2 \geqslant 0. \qquad (4.1d)$$

Weakly nonclassical or classical state:

$$\langle : \hat{N}^2 : \rangle - \langle \hat{N} \rangle^2 \equiv (\Delta N)^2 - \langle \hat{N} \rangle \equiv \gamma_2 - \gamma_1^2 \geqslant 0. \qquad (4.1e)$$

(Here the dots : : denote normal ordering.) The Mandel Q-parameter is defined as [7]

$$Q \equiv \frac{(\Delta N)^2 - \langle \hat{N} \rangle}{\langle \hat{N} \rangle} \equiv \frac{\gamma_2 - \gamma_1^2}{\gamma_1} \tag{4.2}$$

and it has the property of being non-negative in classical and weakly nonclassical states. Conversely if Q is negative, the state is definitely strongly nonclassical. The two cases $Q > 0$ and $Q < 0$ correspond respectively to super- and sub-Poissonian photon number distributions.

The inequalities (4.1) are not all independent, as some imply others. We now give the generalization of these in matrix form, to two-mode states.

We have to deal with four independent number-like operators $\hat{N}_\mu$, $\mu = 0, 1, 2, 3$ which we define thus:

$$\begin{aligned} \hat{N}_\mu &= \underline{\hat{a}}^\dagger \sigma_\mu \underline{\hat{a}} = (\sigma_\mu)_{rs} \hat{a}_r^\dagger \hat{a}_s \\ \hat{a}_r^\dagger \hat{a}_s &= \tfrac{1}{2} (\sigma_\mu)_{rs} \hat{N}_\mu. \end{aligned} \tag{4.3}$$

(Here σ_0 and σ_j are the unit and the Pauli matrices and the sum on μ goes from 0 to 3.) The expectation values of $\hat{N}_\mu$ in a general state $\hat{\rho}$ are written as n_μ:

$$\begin{aligned} \langle \hat{N}_\mu \rangle &\equiv \mathrm{Tr}(\hat{\rho} \hat{N}_\mu) = \int \mathrm{d}\mu\, (\underline{z}) \phi(\underline{z}) \underline{z}^\dagger \sigma_\mu \underline{z} = n_\mu \\ \langle \hat{a}_r^\dagger \hat{a}_s \rangle &\equiv \mathrm{Tr}(\hat{\rho} \hat{a}_r^\dagger \hat{a}_s) = \tfrac{1}{2} (\sigma_\mu)_{rs} n_\mu. \end{aligned} \tag{4.4}$$

Thus n_μ and the matrix $\gamma^{(1/2)} = (\gamma^{(1/2)}_{m_1 m_2})$ are essentially the same. Since the 2×2 matrix $(\langle \hat{a}_r^\dagger \hat{a}_s \rangle)$ is always Hermitian positive semidefinite, we see that the generalization of inequality (4.1*a*) to the two-mode case is

$$n_0 - |\underline{n}| \geqslant 0. \tag{4.5}$$

(All components of n_μ are real.) It may be helpful to remark that the matrix $\gamma^{(1/2)}$ is analogous to the coherency matrix, and the quantities n_μ to the Stokes parameters, in polarization optics [20].

We now consider quadratic expressions in $\hat{N}_\mu$ which are up to quartic in $\hat{a}_r^\dagger$ and $\hat{a}_r$ combined. To handle their normal ordering compactly, we first define certain quadratic expressions in $\hat{a}_r$ and their Hermitian conjugates:

$$\begin{aligned} &\hat{A}_j = \mathrm{i} \underline{\hat{a}}^T \sigma_2 \sigma_j \underline{\hat{a}} \qquad \hat{A}_j^\dagger = -\mathrm{i} \underline{\hat{a}}^\dagger \sigma_j \sigma_2 \underline{\hat{a}}^* \qquad j = 1, 2, 3 \\ &\hat{a}_r \hat{a}_s = -\frac{\mathrm{i}}{2} (\sigma_j \sigma_2)_{rs} \hat{A}_j \qquad \hat{a}_r^\dagger \hat{a}_s^\dagger = \frac{i}{2} (\sigma_2 \sigma_j)_{rs} \hat{A}_j^\dagger. \end{aligned} \tag{4.6}$$

Under the action of the unitary operators $\mathcal{U}(a)$ representing $SU(2)$, both $\hat{A}_j$ and $\hat{A}_j^\dagger$ transform as real three-dimensional Cartesian vectors. We can now easily express the result of writing the product $\hat{N}_\mu \hat{N}_\nu$ as a leading normally ordered quartic term plus a remainder:

$$\begin{aligned} \hat{N}_\mu \hat{N}_\nu &=: \hat{N}_\mu \hat{N}_\nu : + (\ell_{\mu\nu\lambda} + \mathrm{i}\epsilon_{0\mu\nu\lambda}) \hat{N}_\lambda \\ : \hat{N}_\mu \hat{N}_\nu :&= t_{\mu\nu jk} \hat{A}_j^\dagger \hat{A}_k \\ t_{\mu\nu jk} &= \tfrac{1}{2} (\delta_{\mu\nu} \delta_{jk} - \delta_{\mu j} \delta_{\nu k} - \delta_{\nu j} \delta_{\mu k} - \mathrm{i} \delta_{\mu 0} \epsilon_{0\nu jk} - \mathrm{i} \delta_{\nu 0} \epsilon_{0\mu jk}) \\ \ell_{\mu\nu\lambda} &= \delta_{\mu\nu} \delta_{\lambda 0} + \delta_{\mu 0} \delta_{\nu\lambda} + \delta_{\nu 0} \delta_{\mu\lambda} - 2 \delta_{\mu 0} \delta_{\nu 0} \delta_{\lambda 0}. \end{aligned} \tag{4.7}$$

Here $\epsilon_{\sigma\mu\nu\lambda}$ is the four-index Levi–Civita symbol with $\epsilon_{0123} = 1$. So the anticommutators and commutators among $\hat{N}_\mu$ and $\hat{N}_\nu$ are:

$$\tfrac{1}{2} \{\hat{N}_\mu, \hat{N}_\nu\} = t_{\mu\nu jk} \hat{A}_j^\dagger \hat{A}_k + \ell_{\mu\nu\lambda} \hat{N}_\lambda \tag{4.8a}$$

$$[\hat{N}_\mu, \hat{N}_\nu] = 2\mathrm{i} \epsilon_{0\mu\nu\lambda} \hat{N}_\lambda. \tag{4.8b}$$

(These latter are just the $U(2)$ Lie algebra relations.) To accompany n_μ, in a general state we denote the expectation values of $\hat{A}_j^\dagger \hat{A}_k$ by q_{jk}:

$$\langle \hat{A}_j^\dagger \hat{A}_k \rangle \equiv \mathrm{Tr}(\hat{\rho} \hat{A}_j^\dagger \hat{A}_k) = q_{jk}. \tag{4.9}$$

Clearly, (q_{jk}) is basically the matrix $\gamma^{(1)} = (\gamma^{(1)}_{m_1 m_2})$ and is always a 3×3 Hermitian positive semidefinite matrix. This statement is the generalization of inequality (4.1*b*). We can also generalize the inequalities (4.1*c*) and (4.1*d*) by saying that for any quantum state the two matrices with elements given by

$$\langle \tfrac{1}{2}\{\hat{N}_\mu, \hat{N}_\nu\}\rangle = t_{\mu\nu jk} q_{jk} + \ell_{\mu\nu\lambda} n_\lambda \tag{4.10a}$$

$$\begin{aligned} \Delta(\hat{N}_\mu, \hat{N}_\nu) &\equiv \tfrac{1}{2}\langle\{\hat{N}_\mu - \langle\hat{N}_\mu\rangle, \hat{N}_\nu - \langle\hat{N}_\nu\rangle\}\rangle \\ &= t_{\mu\nu jk} q_{jk} + \ell_{\mu\nu\lambda} n_\lambda - n_\mu n_\nu \end{aligned} \tag{4.10b}$$

are both 4×4 real symmetric positive semidefinite. As in the one-mode case the inequality obeyed by $(\Delta(\hat{N}_\mu, \hat{N}_\nu))$ implies the one obeyed by the anticommutator matrix $(\langle \frac{1}{2}\{\hat{N}_\mu, \hat{N}_\nu\}\rangle)$.

Now we search for matrix inequalities which are valid in two-mode classical or weakly nonclassical-I states, but not necessarily in weakly nonclassical-II or strongly nonclassical states. The key ingredient is the formula

$$\begin{aligned} \langle : \tfrac{1}{2}\{\hat{N}_\mu, \hat{N}_\nu\} : \rangle - \langle\hat{N}_\mu\rangle\langle\hat{N}_\nu\rangle &= \int \mathrm{d}\mu\,(\underline{z})\phi(\underline{z})(\underline{z}^\dagger\sigma_\mu\underline{z} - n_\mu)(\underline{z}^\dagger\sigma_\nu\underline{z} - n_\nu) \\ &= \int_0^\infty\int_0^\infty \mathrm{d}I_1\,\mathrm{d}I_2 \int_0^{2\pi} \frac{\mathrm{d}\theta}{2\pi}\mathcal{P}(I_1, I_2, \theta)(\underline{\zeta}^\dagger\sigma_\mu\underline{\zeta} - n_\mu)(\underline{\zeta}^\dagger\sigma_\nu\underline{\zeta} - n_\nu) \end{aligned} \tag{4.11}$$

$$\underline{\zeta} = \begin{pmatrix} I_1^{1/2} \\ I_2^{1/2}\mathrm{e}^{\mathrm{i}\theta} \end{pmatrix}.$$

We can draw the following conclusion:

classical or weakly nonclassical-I state:

$$(\langle : \tfrac{1}{2}\{\hat{N}_\mu, \hat{N}_\nu\} : \rangle - \langle\hat{N}_\mu\rangle\langle\hat{N}_\nu\rangle) \equiv (\Delta(\hat{N}_\mu, \hat{N}_\nu) - l_{\mu\nu\lambda}\langle\hat{N}_\lambda\rangle) \geqslant 0. \tag{4.12}$$

This is the intrinsic two-mode expression of super-Poissonian statistics and its violation (possible only in weakly nonclassical-II or strongly nonclassical states) is an intrinsic signature of two-mode sub-Poissonian photon statistics. What makes this criterion nontrivial is the fact that for any n_μ obeying equation (4.5) the 4×4 matrix $(l_{\mu\nu\lambda} n_\lambda)$ is real symmetric positive semidefinite.

It is interesting to pin down the way in which this matrix inequality (4.12) can go beyond a single-mode condition [21]. The most general normalized linear combination of the two mode-operators $\hat{a}_r$ is determined by a complex two-component unit vector $\underline{\alpha}$:

$$\begin{aligned} &\hat{a}(\underline{\alpha}) = \underline{\alpha}^\dagger\underline{\hat{a}} = \alpha_r^*\hat{a}_r \\ &\underline{\alpha}^\dagger\underline{\alpha} = 1 \\ &[\hat{a}(\underline{\alpha}), \hat{a}(\underline{\alpha})^\dagger] = 1. \end{aligned} \tag{4.13}$$

For every such choice of a single mode, the inequality (4.12) does imply the single-mode inequality (4.1*e*). We can see this quite simply as follows. Given $\underline{\alpha}$, we define the real four-component quantity $\xi_\mu(\underline{\alpha})$ by

$$\begin{aligned} &\xi_\mu(\underline{\alpha}) = \tfrac{1}{2}\underline{\alpha}^\dagger\sigma_\mu\underline{\alpha} : \\ &\xi_0(\underline{\alpha}) = |\underline{\xi}(\underline{\alpha})| = \tfrac{1}{2}. \end{aligned} \tag{4.14}$$

Then, using the completeness of σ_μ expressed by

$$(\sigma_\mu)_{rs}(\sigma_\mu)_{tu} = 2\delta_{ru}\delta_{st} \tag{4.15}$$

we have the consequences:

$$\begin{aligned} &\xi_\mu(\underline{\alpha})\hat{N}_\mu = \hat{a}(\underline{\alpha})^\dagger \hat{a}(\underline{\alpha}) \equiv \hat{N}(\underline{\alpha}) \\ &\ell_{\mu\nu\lambda}\xi_\mu(\underline{\alpha})\xi_\nu(\underline{\alpha}) = \xi_\lambda(\underline{\alpha}). \end{aligned} \tag{4.16}$$

Indeed we easily verify that (leaving aside $\xi_\mu = 0$ identically)

$$\begin{aligned} l_{\mu\nu\lambda}\xi_\mu\xi_\nu = \xi_\lambda \Rightarrow \; &\text{either } \xi_0 = |\underline{\xi}| = \tfrac{1}{2} \\ &\Leftrightarrow \xi_\mu = \tfrac{1}{2}\underline{\alpha}^\dagger \sigma_\mu \underline{\alpha} \\ &\text{some } \underline{\alpha} \text{ obeying } \underline{\alpha}^\dagger \underline{\alpha} = 1 \\ &\text{or } \xi_0 = 1,\; \underline{\xi} = 0. \end{aligned} \tag{4.17}$$

Saturating the left-hand side of (4.12) with the latter possibility, $\xi_\mu = \delta_{\mu 0}$, leads to the super-Poissonian condition for the total photon number distribution. Saturating it with $\xi_\mu(\underline{\alpha})\xi_\nu(\underline{\alpha})$ we obtain as a consequence,

$$(\Delta \hat{N}(\underline{\alpha}))^2 - \langle \hat{N}(\underline{\alpha}) \rangle \geqslant 0 \qquad \text{any } \underline{\alpha}. \tag{4.18}$$

In this way the two-mode matrix 'super-Poissonian' condition (4.12) implies the scalar single-mode super-Poissonian condition (4.1*e*) for every choice for normalized single mode with annihilation operator $\hat{a}(\underline{\alpha})$, as well as for the total photon number.

However, it is easy to see that *the information contained in the matrix inequality (4.12) is not exhausted by the collection of single-mode inequalities (4.18) for all possible choices of (normalized)* $\underline{\alpha}$. Denoting the real symmetric matrix appearing on the left-hand side of (4.12) by $(A_{\mu\nu})$,

$$A_{\mu\nu} = \Delta(\hat{N}_\mu, \hat{N}_\nu) - \ell_{\mu\nu\lambda}\langle \hat{N}_\lambda \rangle \tag{4.19}$$

it is clear that

$$\begin{aligned} \xi_\mu A_{\mu\nu}\xi_\nu \geqslant 0 \qquad &\text{for all } \xi_\mu \text{ obeying } \xi_0 = |\underline{\xi}| = \tfrac{1}{2} \\ &\not\Rightarrow (A_{\mu\nu}) \geqslant 0. \end{aligned} \tag{4.20}$$

Indeed, the left-hand side here reads in detail:

$$\xi_\mu A_{\mu\nu}\xi_\nu = \tfrac{1}{4}A_{00} + A_{0j}\xi_j + \xi_j\xi_k A_{jk} \tag{4.21}$$

and the non-negativity of this expression for all 3-vectors ξ_j with $|\underline{\xi}| = \frac{1}{2}$ cannot exclude the possibility of the 3×3 matrix (A_{jk}) having some negative eigenvalues. Part of the information contained in the matrix condition (4.12) is thus irreducibly two-mode in character, a sample of this being,

$$(A_{\mu\nu}) \geqslant 0 \Rightarrow (A_{jk}) \geqslant 0. \tag{4.22}$$

Admittedly to a limited extent, this situation is analogous to some well known properties of Wigner distributions. Thus the marginal distributions in a single variable obtained by integrating $W(q,p)$ with respect to p or with respect to q (or any real linear combination of q and p) are always non-negative probability distributions, even though $W(q,p)$ is in general indefinite. So also here, it can well happen that for a certain state both A_{00} and $\xi_\mu(\underline{\alpha})A_{\mu\nu}\xi_\nu(\underline{\alpha})$ are non-negative for all $\underline{\alpha}$, yet $(A_{\mu\nu})$ is indefinite.

There exists in the literature a well known inequality for two-mode fields, which when violated is a sign of nonclassicality [22]. It reads:

$$\begin{aligned}&\langle \hat{n}_1(\hat{n}_1 - 1) + \hat{n}_2(\hat{n}_2 - 1) - 2\hat{n}_1\hat{n}_2\rangle \geqslant 0\\ &\hat{n}_1 = \hat{a}_1^\dagger \hat{a}_1 \qquad \hat{n}_2 = \hat{a}_2^\dagger \hat{a}_2\end{aligned} \tag{4.23}$$

and evidently involves only diagonal elements of the matrix $(A_{\mu\nu})$. After rearranging the operators in normal ordered form one can see that

$$\begin{aligned}\langle \hat{n}_1(\hat{n}_1 - 1) + \hat{n}_2(\hat{n}_2 - 1) - 2\hat{n}_1\hat{n}_2\rangle &= \langle \hat{a}_1^{\dagger 2}\hat{a}_1^2 + \hat{a}_2^{\dagger 2}\hat{a}_2^2 - 2\hat{a}_1^\dagger\hat{a}_2^\dagger\hat{a}_1\hat{a}_2\rangle\\ &= \tfrac{1}{2}(q_{11} + q_{22} - q_{33})\\ &= A_{33} + n_3^2 \qquad n_3 = \langle \hat{a}_1^\dagger\hat{a}_1 - \hat{a}_2^\dagger\hat{a}_2\rangle.\end{aligned} \tag{4.24}$$

By our analysis, in any classical or weakly nonclassical-I state the matrix $(A_{\mu\nu})$ is positive semidefinite, so in particular A_{33} and even more so the expression $A_{33} + n_3^2$, are both non-negative. Thus the inequality (4.23) is certainly a necessary condition for classical and weakly nonclassical-I states. Conversely, if (4.23) is violated and $A_{33} + n_3^2$ is negative, then certainly A_{33} is negative as well and the state is either weakly nonclassical-II or strongly nonclassical. However, this condition is unnecessarily strong since it asks for A_{33} to be less than $-n_3^2$; as we have shown, even the weaker condition $A_{33} < 0$ is sufficient to imply that the state is weakly nonclassical-II or strongly nonclassical. Vice versa, our necessary condition $A_{33} \geqslant 0$ for a classical state or weakly nonclassical-I state is stronger than the condition (4.23). In both directions, then, our conditions are sharper than the ones existing in the literature.

We conclude this section by presenting a few examples bringing out the content of the matrix condition (4.12), in particular the possibility of its containing more information than all single-mode projections of it.

(a) *Pair-coherent states*. These are simultaneous eigenstates of $\hat{a}_1\hat{a}_2$ and $\hat{a}_1^\dagger\hat{a}_1 - \hat{a}_2^\dagger\hat{a}_2$ [23]:

$$\begin{aligned}&\hat{a}_1\hat{a}_2|\zeta, q\rangle = \zeta|\zeta, q\rangle \qquad \zeta \in \mathcal{C}\\ &(\hat{a}_1^\dagger\hat{a}_1 - \hat{a}_2^\dagger\hat{a}_2)|\zeta, q\rangle = q|\zeta, q\rangle \qquad q = 0, \pm 1, \pm 2 \ldots.\end{aligned} \tag{4.25}$$

For $q \geqslant 0$ these states are given by

$$|\zeta, q\rangle = N_q \sum_{n=0}^{\infty} \frac{\zeta^n}{[n!(n+q)!]^{1/2}} |n + q, n\rangle \tag{4.26}$$

where N_q is a normalization constant. It is known that in these states the second mode already shows sub-Poissonian statistics†. Thus if we write the matrix (4.19) for these states as $(A_{\mu\nu}(\zeta, q))$, then even without having to nontrivially mix the modes we find:

$$\begin{aligned}&\underline{\alpha} = (0, 1)^T \qquad \xi_\mu(\underline{\alpha}) = \tfrac{1}{2}\underline{\alpha}^T\sigma_\mu\underline{\alpha} = (\tfrac{1}{2}, 0, 0, -\tfrac{1}{2}) :\\ &\xi_\mu(\underline{\alpha})A_{\mu\nu}(\zeta, q)\xi_\nu(\underline{\alpha}) < 0.\end{aligned} \tag{4.27}$$

The matrix $A(\zeta, q)$ is indefinite and the pair coherent states are therefore neither classical nor even weakly nonclassical-I. Consistent with this, a direct numerical study of the least eigenvalue $l(A(\zeta, q))$ of $(A_{\mu\nu}(\zeta, q))$ for sample values of ζ and q, does show it to be negative.

† See Agarwal [2].

(b) *Two-mode squeezed vacuum*. It has been shown elsewhere [25] that a two-mode squeezing transformation is characterized by two independent intrinsic squeeze parameters a and b obeying $a \geqslant b \geqslant 0$. A representative of such a transformation is

$$\mathcal{U}^{(0)}(a,b) = \exp\left[\frac{(a-b)}{4}(\hat{a}_1^{\dagger 2} - \hat{a}_1^2)\right]\exp\left[\frac{(a+b)}{4}(\hat{a}_2^{\dagger 2} - \hat{a}_2^2)\right]. \tag{4.28}$$

The case $a = b$ essentially corresponds to the second mode alone being squeezed. For general $a \neq b$ we have genuine two-mode squeezing; while the (Caves–Schumaker) limit $b = 0$ involves maximal entanglement of the two modes. We restrict our analysis to this limit in the sequel. Then the two-mode squeezed vacuum is characterized by the single parameter a and is

$$\mathcal{U}^{(0)}(a,0)|0,0\rangle = \exp\left[\frac{a}{4}(\hat{a}_1^{\dagger 2} - \hat{a}_1^2 + \hat{a}_2^{\dagger 2} - \hat{a}_2^2)\right]|0,0\rangle. \tag{4.29}$$

The matrix $(A_{\mu\nu}(a))$ can be explicitly computed and happens to be diagonal:

$$\begin{aligned}(A_{\mu\nu}(a)) = \mathrm{diag}(&\tfrac{1}{2}(-3+7\cosh(2a))\sinh(a)^2, 2\cosh(2a)\sinh(a)^2,\\ &-2\sinh(a)^2, 2\cosh(2a)\sinh(a)^2).\end{aligned} \tag{4.30}$$

We see that for all $a > 0$ this is indefinite, since the third eigenvalue $A_{22}(a)$ is strictly negative. This is displayed in figure 1(a), for a in the range $0 < a < 1$. Thus for all $a > 0$ the state (4.29) is definitely neither classical nor weakly nonclassical-I. On the other hand the leading diagonal element (eigenvalue) $A_{00}(a)$ dominates the others in the sense that for all choices of single mode the 'expectation value' of $A(a)$ is non-negative:

$$\xi_\mu(\underline{\alpha})A_{\mu\nu}(a)\xi_\nu(\underline{\alpha}) \geqslant 0 \qquad \text{all } \underline{\alpha}. \tag{4.31}$$

Thus the squeezed vacuum (4.29) displays nonclassicality via sub-Poissonian statistics in an intrinsic or irreducible two-mode sense which never shows up at the one-mode level for any choice of that mode. This is to be contrasted to the case of pair-coherent states discussed previously. At the same time the state (4.29) is also quadrature squeezed for all $a > 0$. Thus both these nonclassical features are present simultaneously.

(c) *Two-mode squeezed thermal state*. This is defined as follows (we again limit ourselves to the case $b = 0$):

$$\begin{aligned}\hat{\rho}(a,\beta) &= \mathcal{U}^{(0)}(a,0)\hat{\rho}_0(\beta)\mathcal{U}^{(0)}(a,0)^{-1}\\ \hat{\rho}_0(\beta) &= (1-\mathrm{e}^{-\beta})^2\exp[-\beta(\hat{a}_1^\dagger\hat{a}_1 + \hat{a}_2^\dagger\hat{a}_2)].\end{aligned} \tag{4.32}$$

At zero temperature $\beta \to \infty$ this goes over to the previous example (b). Once again the matrix $(A_{\mu\nu}(a,\beta))$ can be computed analytically and it turns out to be diagonal:

$$(A_{\mu\nu}(a,\beta)) = (-1+\mathrm{e}^\beta)^2 \times \mathrm{Diag}\begin{pmatrix} \frac{1}{8}(13-14\mathrm{e}^\beta+13\mathrm{e}^{2\beta}+20(1-\mathrm{e}^{2\beta})\cosh(2a)+7(1+\mathrm{e}^\beta)^2\cosh(4a)) \\ \frac{1}{2}((1-\mathrm{e}^\beta)^2+2(1-\mathrm{e}^{2\beta})\cosh(2a)+(1+\mathrm{e}^\beta)^2\cosh(4a)) \\ 1+\mathrm{e}^{2\beta}+(1-\mathrm{e}^{2\beta})\cosh(2a) \\ \frac{1}{2}((1-\mathrm{e}^\beta)^2+2(1-\mathrm{e}^{2\beta})\cosh(2a)+(1+\mathrm{e}^\beta)^2\cosh(4a)) \end{pmatrix}. \tag{4.33}$$

Now the third element $A_{22}(a,\beta)$ can become negative for low enough temperature $T = \beta^{-1}$ or high enough squeeze parameter a. The variation of the least eigenvalue $l(A(a,\beta))$ of $A(a,\beta)$ with respect to a in the range $0 \leqslant a \leqslant 1$, for various choices of β, is shown in figures 1(b)–(d). One can see that if the temperature is not too high, for sufficiently large a the element $A_{22}(a,\beta)$ becomes negative, indicating that the state has then become weakly nonclassical-II or strongly nonclassical. (In comparison we recall that for quadrature

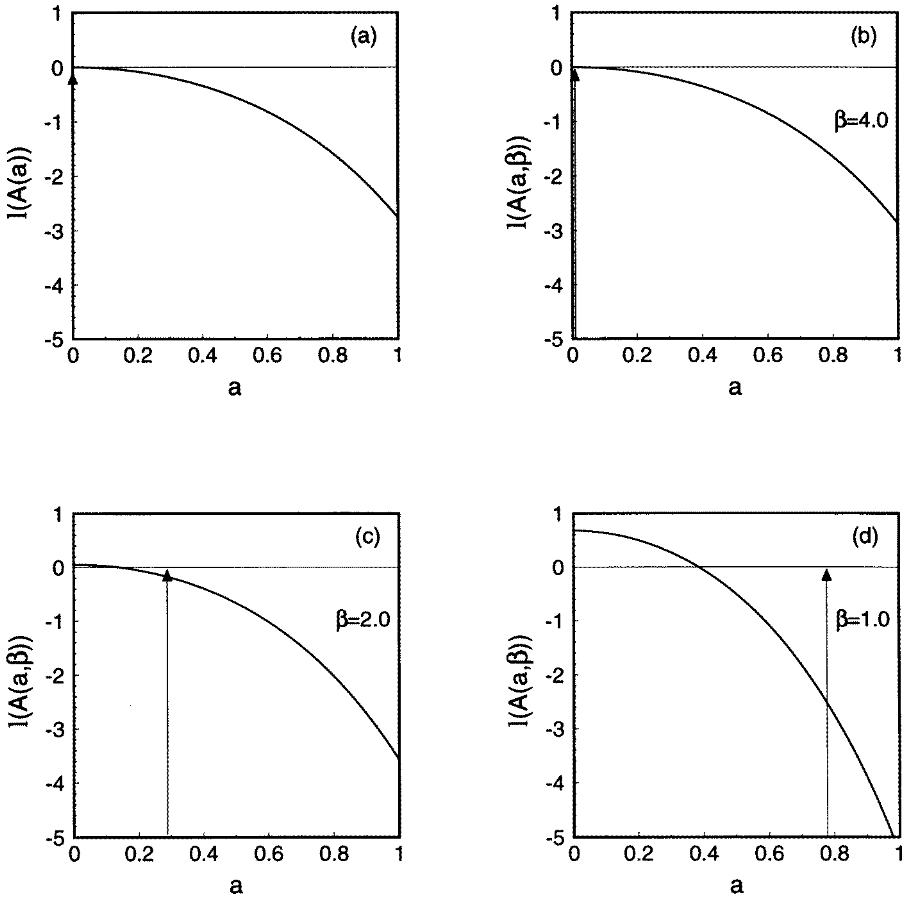

Figure 1. Plots of least eigenvalue of the matrix $(A_{\mu\nu})$ as a function of squeezing parameter a. (a) The least eigenvalue of $(A_{\mu\nu})$ for squeezed vacuum whereas (b)–(d) display the same for squeezed thermal states with inverse temperature β taking the values 4.0, 2.0 and 1.0 respectively. In (b)–(d) the arrows show the setting in of quadrature squeezing.

squeezing to set in the parameter a must obey the inequality $a > \ln\coth(\frac{\beta}{2})$ [24].) On the other hand as in example (b), the leading element $A_{00}(a, \beta)$ again dominates the others in the sense that

$$\xi_\mu(\underline{\alpha}) A_{\mu\nu}(a, \beta) \xi_\nu(\underline{\alpha}) \geqslant 0 \qquad \text{all } \underline{\alpha}. \tag{4.34}$$

So once again, when $A_{22}(a, \beta) < 0$, the sub-Poissonian statistics is irreducibly two-mode in character. In figures 1(b)–(d) we have also indicated the value of the squeeze parameter a at which quadrature squeezing sets in. It is interesting to see that, for the states described here, at each temperature, the irreducible two-mode sub-Poissonian statistics occurs before squeezing. Therefore (limiting ourselves to low-order moments of $\phi(\underline{z})$) there exists a range of squeeze parameter where the only visible nonclassicality is through such sub-Poissonian statistics.

The more general squeezed thermal state

$$\hat{\rho}(\beta, a, b) = \mathcal{U}^{(0)}(a, b) \hat{\rho}_0(\beta) \mathcal{U}^{(0)}(a, b)^{-1} \tag{4.35}$$

has qualitatively similar properties. Detailed numerical studies presented elsewhere [21] have shown that these states also do not show sub-Poissonian statistics at the one-mode level. On the other hand, a direct search for the least eigenvalue of $(A_{\mu\nu}(a, b, \beta))$ reveals that, for suitable values of β, a, b, this is negative.

We thus have several instructive examples of the situation indicated by equation (4.20).

5. Concluding remarks

We have presented a dual operator and expectation value based approach to the problem of distinguishing classical from nonclassical states of quantized radiation and thus brought out the significance of this classification in a new physically interesting manner. As the number of independent modes increases, this approach leads to finer and yet finer levels of nonclassical behaviour, in a steady progression. This has been followed up by a complete analysis of photon number fluctuations for two-mode fields and a comprehensive concept of sub-Poissonian statistics for such fields going beyond what can be handled by techniques developed at the one-mode level.

In a previous paper we have set up the formalism needed to examine the possibility of two-mode fields showing sub-Poissonian statistics at the one-mode level in an invariant manner, by following the variation of the Mandel Q-parameter as one continuously varies the combination of the two independent modes into a single mode. One can see through the work of the present paper that that preparatory analysis is a necessary prerequisite to be able to pinpoint the aspects of sub-Poissonian statistics which are irreducibly two-mode in character. Examples (b) and (c) at the end of section 4 bring out this aspect vividly.

The inequality (4.23) has been strengthened by our approach to a sharper criterion to distinguish various situations:

$$\begin{aligned}&\text{classical or weakly nonclassical-I} \Rightarrow A_{33} \geqslant 0\\ &A_{33} < 0 \Rightarrow \text{weakly nonclassical-II or strongly nonclassical.}\end{aligned} \tag{5.1}$$

From equation (4.19) and (4.24) we see that A_{33} has the following neat expression:

$$\begin{aligned}A_{33} &= (\Delta\hat{n}_1)^2 - \langle\hat{n}_1\rangle + (\Delta\hat{n}_2)^2 - \langle\hat{n}_2\rangle - 2\Delta(\hat{n}_1, \hat{n}_2)\\ &= \langle(\hat{n}_1 - \hat{n}_2)^2\rangle - (\langle\hat{n}_1 - \hat{n}_2\rangle)^2 - \langle\hat{n}_1 + \hat{n}_2\rangle.\end{aligned} \tag{5.2}$$

It is thus expressible solely in terms of expectations and fluctuations of the original (unmixed) mode number operators $\hat{n}_1, \hat{n}_2$ and their functions. One can now see easily, again from equation (4.19), that the statements (5.1) are part of a wider set of statements involving only expectations of functions of $\hat{n}_1, \hat{n}_2$:

$$\begin{aligned}&A_{00} = (\Delta\hat{N}_0)^2 - \langle\hat{N}_0\rangle\\ &A_{03} = A_{30} = \Delta(\hat{N}_0, \hat{N}_3) - \langle\hat{N}_3\rangle\\ &A_{33} = (\Delta\hat{N}_3)^2 - \langle\hat{N}_0\rangle\\ &\hat{N}_0 = \hat{n}_1 + \hat{n}_2 \qquad \hat{N}_3 = \hat{n}_1 - \hat{n}_2\end{aligned} \tag{5.3a}$$

$$\text{classical or weakly nonclassical-I} \Rightarrow \begin{pmatrix} A_{00} & A_{03}\\ A_{30} & A_{33}\end{pmatrix} \geqslant 0$$

$$\begin{pmatrix} A_{00} & A_{03}\\ A_{30} & A_{33}\end{pmatrix} < 0 \Rightarrow \text{weakly nonclassical-II or strongly nonclassical.} \tag{5.3b}$$

All other inequalities involving matrix elements such as $A_{01}, A_{02}, A_{13}\ldots$ involve 'phase sensitive' quantities going beyond $\hat{n}_1$ and $\hat{n}_2$.

Going back to the matrix $A = (A_{\mu\nu})$, we see that from its properties we cannot immediately distinguish between the classical and weakly nonclassical-I situations, or between the weakly nonclassical-II and strongly nonclassical situations. In both the former, A is positive semidefinite; while if A is indefinite, one of the latter two must occur. It would be interesting, for pair coherent states or squeezed thermal states for instance, to be able to see, when A is indefinite, whether we have a weakly nonclassical-II or a strongly nonclassical state, and whether this depends on and varies with the parameters in the state.

Acknowledgments

Arvind thanks University Grants Commission, India for financial support.

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