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Quantum correlations in optics

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Abstract. In many nonlinear optical problems, for example in down-conversion and four-wave mixing, the photons are generated in pairs. The strong correlation between the photons in a pair, characterized by either the correlations between operators corresponding to observables associated with individual photons, or the correlated state describing the two photons, may lead to various nonclassicalities. We discuss some of these nonclassical effects and their experimental demonstrations in nonlinear optical processes.

Keywords. Quantum correlations; squeezing; nonlinear mixing; Wigner distribution.

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1. Introduction

The discussion of quantum phenomena traditionally focussed on the case of microscopic systems with one or few particles or quanta. However, there are examples in optics where the quantum effects show up in processes involving a large number of photons. In nonlinear optical processes of down-conversion and four-wave mixing, the photons are generated in pairs. The strong correlation between the photons in a pair may lead to nonclassicalities such as sub-Poissonian statistics, quantum interference and squeezing. In the case of squeezing the number of photons involved in the process is large. In the following we discuss some of these nonclassical effects and their experimental demonstrations in nonlinear optical processes.

We begin by introducing the photon counting statistics and the intensity-intensity correlation function (which is proportional to the joint probability of two-photon detection with intensity expressed in units of photon-number density). The properties of the coherent states of the radiation field are reviewed. The conditions for demonstrations of the nonclasssical features of antibunching, sub-Poissonian statistics are then indicated and the squeezed states of the radiation field are defined. A brief description of the nonlinear processes generating nonclassical fields is given, with particular emphasis on parametric down-conversion and four-wave mixing interactions. Finally, various classical and local inequalities that are violated by quantum fields are expressed as inequalities relating to the parameters of the underlying Wigner distribution function.

2. Correlation functions

2.1 *Classical fields*

It is well known that when a fluctuating, quasimonochromatic classical light beam of instantaneous intensity $I(t)$ falls on a photoelectric detector, the differential probability $P_1(t)$ for the emission of a photoelectron and its detection within a short time interval δt is given by [1]

$$P_1(t) = \kappa\langle I(t)\rangle\delta t, \tag{1}$$

where κ is a constant characteristic of the detector, and $\langle\ \rangle$ denotes the average over the ensemble for repeated measurements. Here $I(t)$ is a classical field intensity. It is often convenient to express $I(t)$ in units of photons per second, in which case κ is the dimensionless quantum efficiency of the detector.

When two light beams of intensities $I_1(t)$ and $I_2(t)$ fall on two photodetectors of quantum efficiencies κ_1, κ_2, the joint probability of photoelectric detections at both detectors at times t_1 within δt_1 and t_2 within δt_2 is given by

$$P_2(t_1,t_2) = \kappa_1\kappa_2\langle I_1(t_1)I_2(t_2)\rangle\delta t_1\delta t_2. \tag{2}$$

Introducing the normalized intensity correlation function $\gamma(t_1,t_2)$ as

$$\gamma(t_1,t_2) + 1 \equiv \frac{\langle I_1(t_1)I_2(t_2)\rangle}{\langle I_1(t_1)\rangle\langle I_2(t_2)\rangle}, \tag{3}$$

we can rewrite the joint probability in the form

$$P_2(t_1,t_2) = P_1(t_1)P_2(t_2)[1+\gamma(t_1,t_2)]. \tag{4}$$

The first term represents the random contribution to the joint probability and the second describes correlated detections.

2.2 *Quantum fields*

Consider the mode expansion of a quantum mechanical field vector operator $\mathbf{F}(\mathbf{r},t)$ in the second-quantized notation :

$$\mathbf{F}(\mathbf{r},t) = L^{-3/2}\sum_{\mathbf{k}s} l(\omega)a_{\mathbf{k}s}\epsilon_{\mathbf{k}s}\exp[i(\mathbf{k}.\mathbf{r}-\omega t)] + \text{ h.c.} \tag{5}$$

$$= \mathbf{V}(\mathbf{r},t) + \mathbf{V}^\dagger(\mathbf{r},t), \tag{6}$$

where L^3 is the quantization volume, $l(\omega)$ is a slowly varying function of frequency $\omega \equiv \omega_{\mathbf{k}s}$, $\epsilon_{\mathbf{k}s}$ is the unit polarization vector, and $\mathbf{V}$ is the positive frequency part, going as $\exp(-i\omega t)$, which is analytic and regular in the lower half of the complex t-plane, and it contains the photon annihilation operator a.

Traditional photodetectors operate by **absorption** of photons and hence the measured quantities correspond to the normally-ordered correlations [2]. One can define a quantum correlation function Γ_{ij} in the normal order as

$$\Gamma_{ij} \equiv \langle V_i^\dagger(\mathbf{r}_1,t_1)V_j(\mathbf{r}_2,t_2)\rangle. \tag{7}$$

For a quantized electromagnetic field, the light intensity becomes a Hilbert space operator $I(t)$, and the ensemble average $\langle I(t)\rangle$ has to be interpreted as a quantum mechanical expectation value as in (7). In the two-time correlation function, the operators have to be written in normal order and in time order [3], so that

$$P_2(t_1,t_2) = \kappa_1\kappa_2\langle T : I_1(t_1)I_2(t_2) :\rangle\delta t_1\delta t_2. \tag{8}$$

Here T stands for time-ordering and the colons :: for normal ordering of the operators, i.e., $\langle T : I(t_1)I(t_2) :\rangle = \langle a^\dagger(t_1)a^\dagger(t_2)a(t_2)a(t_1)\rangle$ when $t_1 < t_2$. For a stationary field, $\gamma(t_1,t_2)$ depends only on the time-difference $t_2 - t_1 = \tau$. For thermal fields, the joint probability of detecting two photoelectric pulses separated by time τ is maximum at $\tau = 0$, it decreases as τ increases. Thus, for thermal light, $\gamma(\tau = 0) = 1$, and in the limit $\tau \to \infty, \gamma(\tau) = 0$.

3. The coherent state

The coherent state $|\alpha\rangle$ [3] of the radiation field is the eigenstate of the photon annihilation operator a :

$$a|\alpha\rangle = \alpha|\alpha\rangle, \tag{9}$$

where α is the complex eigenvalue. The corresponding wave-packet 'coheres' (does not spread) in time. This can be explicitly seen from the following equivalent definition of a coherent state as a state ψ_{cs} in a harmonic oscillator potential which, at time $t = 0$, is a Gaussian that is displaced in phase-space from the origin by x_0, has a phase which is proportional to the position x, and the width of the Gaussian is that of the ground state, $\sigma_0 \equiv (\hbar/2mw)^{1/2}$, i.e.,

$$\psi_{\text{cs}}(t=0) = (2\pi\sigma_0^2)^{-1/4}\exp[-(x-x_0)^2 + ip_0x/\hbar]. \tag{10}$$

A coherent state may be generated by applying a unitary displacement operator $D(\alpha)$ to the ground state $|0\rangle$ of a simple harmonic oscillator, i.e.,

$$|\alpha\rangle = D(\alpha)|0\rangle, \tag{11}$$

where the displacement operator is given as

$$D(\alpha) = \exp(\alpha a^\dagger - \alpha^* a). \tag{12}$$

The correlation function (8) in the coherent state of a multimode field factorizes, and the degree of coherence which is defined as the magnitude of the normalized correlation function given in the RHS of (3), is thus unity in a coherent state.

When completely coherent light (in a coherent state $|\alpha\rangle$) falls on a photoelectric detector, the number of photoelectric counts n (proportional to the light intensity I) registered in some finite time interval obeys Poisson statistics for which the variance $\langle(\Delta n)^2\rangle$ of n equals the mean number $\langle n\rangle$. For such a coherent field, the above joint probability (8) is constant for all τ, and $\gamma(\tau) = 0$, for all $\tau \geq 0$.

The quantum character of a field can be demonstrated by violating these classical results either in measurements of time intervals between detected photons (demonstrating

antibunching [4]), or in photon counting measurements (yielding **sub-Poissonian statistics** [5]). The condition for sub-Poissonian photon statistics is given by $\langle(\Delta n)^2\rangle - \langle n\rangle < 0$, which gives $\gamma(0) < 0$. Antibunching occurs when $\gamma(0) < \gamma(\tau)$. These conditions violate the Schwarz inequality for classical fields, and hence both sub-Poissonian statistics and antibunching are quantum phenomena.

4. Squeezed states of the electromagnetic field

For a single-mode field, squeezed states are a unique set of quantum states in which fluctuations in the amplitude of one of the quadrature phases, $(a^\dagger + a)$ or $i(a^\dagger - a)$, is less than that in a coherent state.

A single-mode squeezed state $|\alpha, s\rangle$ may be generated by first acting with the squeeze operator $S(s)$ on the vacuum $|0\rangle$ followed by the displacement operator $D(\alpha)$:

$$|\alpha, s\rangle = D(\alpha)S(s)|0\rangle, \tag{13}$$

where

$$S(s) = \exp(sa^{\dagger 2} - s^* a^2), \tag{14}$$

s being a complex parameter, and $D(\alpha)$ is given by eq. (12). The squeeze operator creates and destroys photons in pairs. If the order of the two operators $D(\alpha)$ and $S(s)$ in eq. (13) is reversed, a squeezed coherent state $|s, \alpha\rangle$ is obtained.

Two-mode squeezed states can be generated by applying the following generalized squeeze operator to the two-mode vacuum $|0, 0\rangle$:

$$S_{12}(s) = \exp(sa_1^\dagger a_2^\dagger - s^* a_1 a_2), \tag{15}$$

where the subscripts 1,2 refer to the two modes. This may be followed by displacements in each mode represented by the operators

$$D_1(\alpha_1) = \exp(\alpha_1 a_1^\dagger - \alpha_1^* a_1), \tag{16}$$

$$D_2(\alpha_2) = \exp(\alpha_2 a_2^\dagger - \alpha_2^* a_2). \tag{17}$$

Two photons of the same frequency ω from the same radiation mode can be absorbed in a single atomic transition between two levels (of the same parity) via an intermediate state. Such a transition is second-order in the interaction **p.A**. The atom-radiation interaction responsible for the two-photon transitions can in general be expressed in the form :

$$H = h(G\Sigma a^{\dagger 2} + G^* \Sigma^\dagger a^2), \tag{18}$$

where G is the coupling coefficient which depends on the nonlinear susceptibility of the medium for the process under consideration, and Σ is the atomic polarization operator which flips the state of the atom. In the Markov case, when the atomic memory effects can be ignored, all interactions generating two output beams from an intense (classical) input pump field, can be written as an effective bilinear interaction of the form

$$H_{\text{eff}} = ga_1^\dagger a_2^\dagger + \text{h.c.}, \tag{19}$$

where a_1, a_2 ($a_1^\dagger$, $a_2^\dagger$) are the annihilation (creation) operators corresponding to the two output modes. The output fields are taken to be initially in the vacuum state $|0, 0\rangle$, and the state at time t will then be a state $|TP\rangle$ given by

$$|TP\rangle = \exp(-iH_{\text{eff}}t/\hbar)|0,0\rangle \tag{20}$$

$$= \text{sech}(|g|t/\hbar)\sum_{n=0}^{\infty}[(-ig/|g|)\tanh(|g|t/\hbar)]^n|n,n\rangle \tag{21}$$

If we restrict ourselves only to the first two terms of the exponential series corresponding to the lowest-order excitations, we get the two-photon state $|1,1\rangle$.

In some nonlinear optical interactions, for example in the three-wave mixing interaction of frequency down-conversion and in four-wave mixing, the photons are generated and absorbed in pairs as described above. Such two-photon processes have been shown [6–9] to exhibit rich quantum features such as sub-Poissonian statistics, squeezing, and violation of Cauchy–Schwarz inequalities. In the following we first briefly describe the nonlinear processes of interest.

5. Optical nonlinearities

The response of a medium to an applied electric field is usually taken to be linear given by an induced electric polarization **P** proportional to the electric field **E**. For a nonlinear material subject to an intense electric field **E**, the proportionality constant or the susceptibility χ is no longer a constant independent of the strength of the field. In the regime of weak nonlinearities, one may adapt a perturbative approach to the problem in which the polarization P is written as a power series expansion in E:

$$P_i = \epsilon_0[\chi_{ij}^{(1)}E_j + \chi_{ijk}^{(2)}E_jE_k + \chi_{ijkl}^{(3)}E_jE_kE_l + \cdots], \tag{22}$$

where ϵ_0 is the dielectric permittivity of vacuum and $\chi^{(n)}$ is the nth-order susceptibility tensor characteristic of the medium. The second- and all even-order susceptibilities are identically zero in the bulk of a centrosymmetric medium. The number of beams taking part in a nonlinear mixing interaction in a medium is the number of beams entering the medium plus the number of beams generated by the medium itself. The $\chi^{(2)}$-processes are three-wave mixing interactions of the type

$$\omega_1 + \omega_2 \rightleftharpoons \omega_0, \tag{23}$$

where ω's are the frequencies.

5.1 *Down-conversion*

For the degenerate case of $\omega_1 = \omega_2 = \omega$, (23) leads to the second-harmonic generation, $\omega + \omega \to 2\omega$, the reverse of which is called the degenerate parametric down-conversion. In the process of spontaneous frequency down-conversion, photons in the monochromatic pump laser beam of frequency ω spontaneously 'split' into pairs of lower-frequency signal and idler photons that emerge from the nonlinear medium within a cone around the pump beam axis. If the frequencies of the monochromatic signal and idler light beams are ω_1 and ω_2, respectively, then as given by (23), energy must be conserved for an interaction to take place with appreciable probability. Efficient parametric down-conversion also requires conservation of momentum, i.e.,

$$\mathbf{k}_0 = \mathbf{k}_1 + \mathbf{k}_2, \tag{24}$$

where $\mathbf{k}_0$, $\mathbf{k}_1$, $\mathbf{k}_2$ are the momenta of the pump, signal and idler photons respectively. Equations (23) and (24) are together known as the phase-matching conditions. These conditions can be met in a uniaxial non-centrosymmetric crystal exhibiting birefringence, such as KDP (KH_2PO_4), LI ($LiIO_3$). For a negative birefringent crystal with the index of refraction for the ordinary wave (n_0) larger than that for the extra-ordinary ray (n_e) at a particular frequency, phase-matching for degenerate down-conversion may be achieved by choosing a direction of propagation θ of the pump with the optic axis of the crystal such that $n_e(\theta)$ (at 2ω) $= n_o$ (at ω).

5.2 *Four-wave mixing*

The $\chi^{(3)}$-processes of degenerate four-wave mixing, optical phase conjugation, two-beam coupling have enormous potential for application in dynamic holography, wave-front reversal, optical computing and squeezed state generation. Recently a lot of attention has been paid to the study of nonlinearity due to saturated absorption in resonant samples of dye-doped glasses. The radiative lifetime of the lowest triplet state in dyes, particularly in the rigid environment of a solid host, is quite long. This results in a low value of the saturation intensity and thus allows the observation of nonlinear effects in dye-doped solids at very low power levels [10].

The nonlinear process of self-diffraction refers to the diffraction of the pump beams from the laser-induced grating created by the pumps themselves. The saturation characteristics of dyes are usually described by a three-level model, and the rate equation formalism is applied for the population dynamics. The generation of four-wave mixing signals with chaotic (thermal) and phase-diffusing pump fields has been extensively studied in recent years using a microscopic approach, based on numerical solution of the Bloch vectors for the atomic density matrix elements, but only for the case when one of the pump beams is weak [11]. For the basic experimental studies, the samples of dye-doped solids are popular candidates, as they are easy to prepare and have a high value of nonlinear susceptibility arising from the long life-time of the excited state. Because of this long life-time, on the other hand, these samples were thought to be not useful for applications based on the correlation of the signals generated in nonlinear mixing. However, our recent calculations show that squeezing effects may persist in a four-wave mixing system with long-lived states [7,8], and it has an advantage in requiring less pump powers.

The samples of thin films of Rhodamine 6G dye-doped boric acid are prepared by the following method [10]. 10^{-3} M concentration of the dye in orthoboric acid powder is mixed and heated in a test-tube to around 270 deg C. The resultant homogeneous melt is poured and pressed between two glass plates preheated to 130 deg C. While pressing, care is taken to apply pressure uniformly, so that the thickness variations in the sample are minimal.

Using a narrow-band (bandwidth $\approx$ 20 MHz) argon-ion laser, we have been able to generate a phase-conjugate signal in a simple configuration with very few optical elements [12]. We have measured the transient build-up and decay of the first-order diffracted signal generated by four-wave mixing in our samples of dye-doped glasses. By applying the rate equation formalism to a three-level saturable absorber, it can be easily shown that the effective time τ in which a saturable absorber recovers from saturation, after the pump field is removed, is given by the product of the triplet state life-time T_1 and the quantum yield of the triplet state [10]. The saturation intensity I_s is inversely proportional to the recovery time τ. The build-up time τ_r of the saturation depends on the incident intensity I, and for homogeneous broadening, $\tau_r \cong \tau/(1 + I/I_s)$. By studying the decay and rise of the self-diffracted signal from the saturable absorber, when the pump beams are modulated, one experimentally measures τ and τ_r [12]. The validity of the rate equation treatment is then checked, and as indicated earlier, the predictions of the detailed numerical calculations involving atomic density matrices can also be verified.

6. Quantum interference in down-conversion

The nonclassical features in interference with independent sources have been the subject of many recent investigations [13]. These effects show up readily in intensity correlations that are fourth-order in field amplitudes. Fourth-order effects are predicted even classically, but there are significant differences between the predictions of the classical and the quantum mechanical theories regarding the relative depth of modulation or 'visibility' of the fourth-order interference pattern. For classical fields there are theoretical upper bounds for the 'visibility' and the quantum mechanical states can be made to violate these new kind of inequalities. For a two-photon source, the joint probability of detecting two photons at two points in the interference plane vanishes when the separation between the detectors is an odd integral multiple of half fringe-spacing, and the relative modulation amplitude or 'visibility' σ of the fringe pattern is 100%, unlike the classical situation where it can be shown that for two sources of monochromatic fixed-intensity waves, $\sigma \leq 50\%$ [13]. The quantum mechanical result calls for the Dirac interpretation of interference – viz. the fact that one of the detected photons must have come from one source and one from the other, but we cannot tell which came from which. We therefore have to add the two probability amplitudes corresponding to the two indistinguishable paths, and the probability amplitudes are in anti-phase when the two detectors are separated by an odd number of half-fringes. The first observation of nonclassical effects in fourth-order interference was reported in an experiment [14] involving the down-converted photons.

7. Squeezing by four-wave mixing

The first experiment [15] to successfully generate squeezed states (two-mode) employed the process of degenerate four-wave mixing. In this technique, the nonlinear medium takes some photons from two strong pump waves and feeds them into two weaker conjugate beams. When these two correlated signal beams are combined at a beam-splitter, the resulting light exhibits the amplified and reduced quadrature fluctuations. A few years back, we proposed a theoretical model for a two-photon squeezed laser [7], in which the gain medium is an active nonlinear medium. An intense pump laser beam causes two-photon excitations in the medium and generates two radiation fields due to four-wave mixing. The generated photons can get reabsorbed by a two-photon absorption process. A strong competition among four-wave mixing, two-photon absorption and linear cavity losses leads to lasing action above a certain threshold. This is an example of a laser where amplification is obtained without population inversion. The phase correlations of the two photons generated inside the cavity in this process yields a narrower than the usual Schawlow–Townes linewidth of the two-photon laser far above threshold. The spectrum of fluctuations in the intensity difference between the two output modes shows squeezing, as the photon-number fluctuations of the two modes try to balance each other. As mentioned earlier, our recent calculations show that such effects persist in systems such as the dye-doped solids with long-lived excited states, and these systems have an additional advantage in requiring less pump powers [8].

8. Wigner function description of quantum features

For fields generated in a large class of nonlinear optical processes, including those with losses, the quantum state of the generated radiation corresponds to a Gaussian Wigner function which is centred around the mean value of the field [16]:

$$W(z,z^*) = [\pi(t^2-4|\mu|^2)^{1/2}]^{-1}\exp[-\{\mu(z-z_0)^2+\mu^*(z^*-z_0^*)^2 + t|z-z_0|^2\}/(t^2-4|\mu|^2)], \qquad (25)$$

where

$$\langle a\rangle = z_0, \qquad (26)$$
$$\langle a^2\rangle = -2\mu^* + z_0^2, \qquad (27)$$
$$\langle a^{\dagger 2}\rangle = -2\mu + z_0^{*2}, \qquad (28)$$
$$\langle n\rangle \equiv \langle a^\dagger a\rangle = t - \frac{1}{2} + |z_0|^2, \qquad (29)$$
$$t > \mu + \mu^*. \qquad (30)$$

This distribution can be explicitly shown to emerge as the solution around steady-state for the processes under consideration [17]. The fluctuations in the photon-number $n \equiv a^\dagger a$ can be written in terms of the Wigner parameters as

$$\langle(\Delta n)^2\rangle \equiv \langle n^2\rangle - \langle n\rangle^2 = t^2 + 2|z_0|^2 t - \frac{1}{4} - 2z_0^2\mu - 2z_0^{*2}\mu^* + 4|\mu|^2. \quad (31)$$

The condition for sub-Poissonian photon statistics, viz. $\langle(\Delta n)^2\rangle - \langle n\rangle < 0$, can now be written in terms of the Wigner parameters (μ, μ^*, t) as

$$t^2 + 2|z_0|^2 t + \frac{1}{4} - 2z_0^2\mu - 2z_0^{*2}\mu^* + 4|\mu|^2 - |z_0|^2 - t < 0. \qquad (32)$$

The condition for squeezing the quadrature component $(a + a^\dagger)$, viz. $\langle\Delta(a+a^\dagger)^2\rangle < 1$, leads to the following condition on the Wigner parameters :

$$0 < t - \mu - \mu^* < \frac{1}{2}. \qquad (33)$$

Similarly, conditions for the violation of classical inequalities pertaining to the 'visibility' or relative depth of modulation in higher order interference [13,14] can be written down in terms of the Wigner parameters. In a typical polarization correlation set-up demonstrating nonlocality of the Einstein–Podolsky–Rosen type [18], the appropriate Bell inequality [19,20] is shown to be violated. Such violation has been observed experimentally in correlation measurements of mixed signal and idler photons produced in the process of parametric down-conversion [21]. In such a set-up, the two correlated modes coming out of a nonlinear material are made to fall from opposite sides on a beam-splitter. The mixed beams arrive at the detectors via two polarizers set at variable angles, and the condition for the violation of nonlocality for optimum choice of polarization angles of the two detected beams in this case is the following [17]:

$$\frac{\langle n_1(n_1-1)\rangle + \langle n_2(n_2-1)\rangle}{\langle n_1 n_2\rangle} < 0.5, \tag{34}$$

where 1,2 refer to the signal and idler beams incident on the beam-splitter. For the down-conversion process with nearly-Poissonian photon statistics, this condition can be expressed in terms of the Wigner parameters as [17]

$$t + |z_0|^2 < 0.83. \tag{35}$$

Thus instead of having different inequalities to describe different quantum features of the electromagnetic field, one can identify a generalized description in terms of the nonclassical distribution function of the field, and classify the degree of nonclassicality in such cases [22].

Summary

Two-photon processes have the potential of generating nonclassical states of light. Many quantum effects have been observed experimentally in dissipative nonlinear systems producing correlated two-photon states. A description of the various nonclassical and nonlocal features is given in terms of the parameters of the Wigner distribution function of the fields generated in a large class of nonlinear processes. The field acquires quantum features only for certain ranges of the Wigner parameters. This may provide a unifying description of the known quantum features of the radiation field.

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