Optik 127 (2016) 4447–4453

Contents lists available at ScienceDirect

Optik

journal homepage: www.elsevier.de/ijleo

Nonclassical effects in the second harmonic generation

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ARTICLE INFO

Article history:
Received 14 December 2015
Accepted 18 January 2016

Keywords:
Second harmonic
Variance squeezing
Quantum entanglement
Coherent state
Information entropy

ABSTRACT

The higher-order nonclassical squeezing and quantum entanglement effects emerging from the second harmonic generation of the associated two-mode and two-photon Hamiltonian are investigated in the dispersive limit. The squeezed states of the field, including the normal and amplitude squared (higher-order) squeezing factors are generated in two ways, i.e., from the bosonic operators via amplitude powered quadrature variables, and through the $SU(2)$ characterization of a passive and lossless device with two input and two output ports, which then allows one to visualize the operations of beam splitters and phase shifters as rotations of angular momentum operators in 3-space. Two criteria for intermodal higher-order quantum entanglement and different coherent states for the two modes in the initial state are used to compute these nonclassical effects. The unitary time evolution of the linear entropy, computed from the partial trace of the density matrix over the secondary mode, is also used as a criterion of quantum entanglement. These approaches show, in fact, that the present model exhibits a considerable amount of this nonclassical effect. The unitary time evolution of the linear entropy shows that the present nonlinear optical model does not preserve the modulus of the Bloch vector.

1. Introduction

Second harmonic generation (SHG) is a nonlinear optical process, in which photons with the same frequency interacting with a nonlinear material are effectively "combined" to generate new photons with twice the energy, and therefore twice the frequency and half the wavelength of the initial photons. In biological and medical science, the effect of SHG is used for high-resolution optical microscopy. Because of the non-zero second harmonic coefficient, only non-centrosymmetric structures are capable of emitting SHG light. One such structure is collagen, which is found in most load-bearing tissues. SHG microscopy has been used for extensive studies of the cornea [1] and lamina cribrosa sclerae [2], both of which consist primarily of collagen. SHG is also used by the laser industry to make green 532 nm lasers from a 1064 nm source and for measuring ultra short pulse width by means of intensity auto-correlation. Generating the second harmonic, often called frequency doubling, is also a process in radio communication; it was developed in the 20th century, and has been used with frequencies in the megahertz range.

Generally speaking, these *two-photon states* can be useful for solving various fundamental physical and technological problems. For example, one can use the two-photon states in order to improve optical communications by reducing the quantum fluctuations in one (signal) quadrature component of the field at the expense of the amplified fluctuations in another (unobservable) component. These interesting properties and applications in various fields of applied and basic theoretical research of the SHG, motivated us to explore the so far uninvestigated potential existence of nonclassical squeezing and intermodal higher-order quantum entanglement effects in the associated electromagnetic field. In this context it is worth noting that several new applications of these nonclassical states have been reported in recent past [3–7]. As a consequence of these recently reported applications, generation of nonclassical states in various quantum systems emerged as one of the most important areas of interest in quantum information theory and quantum optics [8,9]. Several systems are already investigated and have been shown to produce entanglement and other nonclassical states [10,11].

A state having negative or highly singular (more singular than δ function) Glauber–Sudarshan P function is referred to as a nonclassical state as it cannot be expressed as a classical mixture of coherent states [3]. P function provides us an essential as well as sufficient criterion for detection of nonclassicality. However, P function is not directly experimentally measurable. Consequently, several operational criteria for nonclassicality have

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http://dx.doi.org/10.1016/j.ijleo.2016.01.152
0030-4026/

been proposed in the past. A large number of these criteria are expressed as inequalities involving expectation values of functions of annihilation and creation operators. As mentioned above we are interested in the higher-order nonclassical properties of radiation fields. In quantum optics and quantum information higher-order nonclassical properties of bosons (e.g., higher-order Hong-Mandel squeezing, higher-order antibunching, higher-order sub-Poissonian statistics, higher-order entanglement, etc.) are often studied [12]. Until recently, past studies on higher-order nonclassicalities were predominantly restricted to theoretical investigations. However, a bunch of exciting experimental demonstrations of higher-order nonclassicalities have been recently reported [13–15].

Higher-order squeezing is usually studied using two different approaches. In the first approach introduced by Hillery [16] reduction of variance of an amplitude powered quadrature variable for a quantum state with respect to its coherent state counterpart reflects nonclassicality. In contrast, in the second type of higher-order squeezing introduced by Hong and Mandel [17,18], higher-order squeezing is reflected through the reduction of higher-order moments of usual quadrature operators with respect to their coherent state counterparts.

The present contribution aims to study higher-order nonclassical properties emerging from the SHG Hamiltonian with specific attention to higher-order squeezing and higher-order quantum entanglement.

The remaining part of the paper is organized as follows. Section 2 introduces the theoretical background associated with the SHG Hamiltonian together with the two approaches of squeezing based on the variances of the quadrature modes. We also introduce a scattering matrix, proper of the $SU(2)$ group, which will in general transform the angular momentum operators among themselves. Since $SU(2)$ is equivalent to the rotation group in three dimensions, introduction of the Schwinger representation of the angular momentum operators through an homomorphism with the unitary group, will allow one to visualize the operations of beam splitter and phase shifters as rotations in 3-space. This section ends with the description of the conditions for the intermodal higher-order quantum entanglement. In Section 3 we use the criteria described in the previous section to numerically illustrate the nonclassical character of the radiation field of this nonlinear optical process using different initial coherent states. This is a novel feature of the present approach, since coherent states can be considered as squeezed states. It will be shown that the variances of different canonically conjugated variables can even assume (small) values which are less than the ground state variances. It will be also shown that there exists the possibility of a weak intermodal higher-order quantum entanglement of this specific two-mode optical process. This is reflected, in particular, in the unitary evolution of the linear entropy which shows that the present nonlinear optical process does not preserve the modulus of the Bloch vector. The paper ends up with conclusions in Section 4.

2. Theoretical background

2.1. Second harmonic generation Hamiltonian and initial conditions

With the widespread use of large-amplitude beams from powerful lasers, it is necessary to assume that the relationship between the polarization and the electric field is nonlinear. In fact, the second-order term of the expansion of the polarization in powers of the electric field shows that the resultant second-order nonlinear response generates an oscillating polarization at the sum and difference frequencies of the input fields. If the two frequencies are equal, the sum frequency is at twice the input frequency, and the effect is called frequency doubling or second harmonic generation. This nonlinear process is governed by the Hamiltonian

$$\mathcal{H} = \mathcal{H}_0 + \mathcal{H}_{\text{int}} = \omega_a a^\dagger a + \omega_b b^\dagger b + g(a^2 b^\dagger + a^{\dagger 2} b), \tag{1}$$

where $a(a^\dagger)$ and $b(b^\dagger)$ are the annihilation (creation) operators of the fundamental mode of frequency ω_a and of the second hermonic mode of frequency ω_b, respectively, satisfying standard commutation relations for the Lie algebra of $SU(2)$. When perfect matching conditions are satisfied, we have the relation $\omega_a = 2\omega_b$. The constant g describes phenomenologically the coupling between the modes. It can always be chosen as real.

The nonlinear Hamiltonian (1) can be diagonalized through the method of small rotations pioneered by Klimov et al. [19,20] and we briefly describe it. After noting that it admits the constant of motion [19]

$$\mathcal{R} = a^\dagger a + 2b^\dagger b, \tag{2}$$

then the Hamiltonian can be rewritten in the following form

$$\begin{aligned} \mathcal{H}_0 &= \frac{\omega_a + \omega_b}{3}\mathcal{R} \\ \mathcal{H}_{\text{int}} &= \frac{\Delta}{3}(b^\dagger b - a^\dagger a) + g(a^2 b^\dagger + a^{\dagger 2} b), \end{aligned} \tag{3}$$

where $\Delta = \omega_b - 2\omega_a$ is the detuning.

Since $\mathcal{H}_0$ determines the total energy stored in both modes, which is conserved, $[\mathcal{H}_0, \mathcal{H}_{\text{int}}] = 0$, we can factor out $\exp(-i\mathcal{H}_0 t)$ from the evolution operator and drop it altogether.

In the present work we are interested in the dispersive limit of this model, when

$$|\Delta| \gg g(\bar{n}_1 + 1)(\bar{n}_2 + 1), \tag{4}$$

where $\bar{n}_1$ and $\bar{n}_2$ denote the average photon numbers in the first and second harmonic modes a and b, respectively. Then, using the Lie transformation method [19–21], and applying a unitary transformation to the interaction Hamiltonian (1), an effective Hamiltonian is obtained as

$$\mathcal{H}_{eff} = \mathcal{T}\mathcal{H}_{\text{int}}\mathcal{T}^\dagger, \tag{5}$$

where

$$\mathcal{T} = \exp[\lambda(a^2 b^\dagger - a^{\dagger 2} b)], \tag{6}$$

with $\lambda = g/\Delta \ll 1$. By expanding Eq. (5) in a power series and keeping terms up to the order $(g/\Delta)^2$, yields

$$\mathcal{H}_{eff} = \frac{\Delta}{3}(b^\dagger b - a^\dagger a) + \frac{g^2}{\Delta}[4b^\dagger b a^\dagger a - (a^\dagger a)^2]. \tag{7}$$

The effective Hamiltonian Eq. (7) describes the dispersive evolution of the fields, but the essential point is that it is diagonal, which implies that there is no population transfer between the modes (as expected in the far resonant limit). The first term in Eq. (7) does not affect the dynamics and just leads to rapid oscillations of the wave function. The dynamics of the present model is not stationary and depends on the initial conditions of the model. Thus, it is assumed that, initially, the field modes are in generalized Glauber boson coherent states $|z_1 z_2>$ [22] and in a disentangled state with density operator

$$\rho(0) = \rho_a(0) \otimes \rho_b(0) = |\psi(0)><\psi(0)|, \tag{8}$$

where $\rho_a(0)$ and $\rho_b(0)$ are density operators at $t = 0$ of the field modes, and

$$|\psi(0)> = \sum_{m_1=0}^{\infty}\sum_{m_2=0}^{\infty} C_{m_1 m_2}(0)|m_1> \otimes |m_2>, \tag{9}$$