POLARISATION DEPENDENCE OF PHOTON ANTIBUNCHING PHENOMENA INVOLVING LIGHT PROPAGATION IN ISOTROPIC MEDIA

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By the method of "short optical paths", the dynamics of photon antibunching is shown to depend on the polarisation state of the photons and their two-photon absorption in an isotropic medium. In the case of self-induced optical birefringence, a change in helicity of the elliptically polarized beam or a change in sign of the angle between the large semi-axis of the ellipse and the analyzer causes a reversal of antibunching into bunching. Linear and circular polarisation of the photons are discussed as well for antibunching related with two-photon absorption, defined by the imaginary part of the molecular hyperpolarizability tensor.

1. Introduction

The production of a radiation field exhibiting photon antibunching (anticorrelation) has recently been proved feasible by Kimble et al. [1] in experiments on the resonance fluorescence of sodium atoms, illuminated continuously with the light of a dye laser. As shown by us previously [2,3] applying the "short optical path" method, the effect of photon anticorrelation can also occur in phenomena of second and higher light harmonics generation. Similar results have been derived independently by Mišta and Peřine [4]. Lately, Drummond et al. [5] have considered the problem of antibunching and bistability with regard to second light harmonic generation inside a Fabry-Pérot cavity. Changes in the polarisation and statistical properties of the light beam due to nonlinear interaction with the system of molecules have been discussed earlier by Atkins and Wilson [6], Tanaś,[7], as well as Kielich and Tanaś [8]. Previously derived formulae contain in fact, implicitly, the antibunching effect in phenomena of nonlinear optical activity [6,7] and self-induced optical birefringence [6,8] of isotropic molecular media. Quite recently, Bandilla and Ritze [9,10] have discussed the role of interference and polarisational effects in the enhancement of photon antibunching.

In the present letter we propose explicit formulae, defining the occurrence of antibunching and its dependence on the polarisation state of the incident photons at light propagation in an isotropic medium composed of N mutually independent nonlinearly polarizable atoms or molecules.

2. Equations of motion, and field correlation tensors

We shall be considering the interaction of an intense light beam and an isotropic medium, consisting of N atoms (molecules). Nonlinear interaction between the light and an individual atom will be described in terms of the following phenomenological interaction hamiltonian:

$$H_{\mathrm{I}} = -\tfrac{1}{2}\gamma_{\sigma\tau\nu\rho}(\omega)E_\sigma^{(-)}E_\tau^{(-)}E_\nu^{(+)}E_\rho^{(+)}, \tag{1}$$

where $\gamma_{\sigma\tau\nu\rho}(\omega)$ is the hyperpolarizability tensor of the molecule [11], and $E_\sigma^{(+)}$, $E_\sigma^{(-)}$ are operators of the electromagnetic field of the light beam [12]. Since we shall be dealing with a single radiation mode, of elliptical polarisation and propagation in the z-direction, the free field operators are of the form:

$$E_\sigma^{(+)}(k) = [E_\sigma^{(-)}(k)]^\dagger = \mathrm{i}(\hbar\omega/2\epsilon_0)^{1/2} e_\sigma a \exp(\mathrm{i}kz), \tag{2}$$

with ω the radiation frequency, e_σ the polarisation vector component, and a the photon annihilation operator fulfilling the boson commutation rules

$$[a, a^\dagger] = 1. \tag{3}$$

The polarisation vector of light elliptically polarized and propagating along z has the following components [6]:

$$\mathrm{i}e_x = \cos\eta\cos\theta - \mathrm{i}\sin\eta\sin\theta, \qquad \mathrm{i}e_y = \cos\eta\sin\theta + \mathrm{i}\sin\eta\cos\theta, \tag{4}$$

θ denoting the azimuth and η the ellipticity of the incident beam.

Applying the hamiltonian [1] and the commutation rules [2], one obtains the quantum equations of motion (in the Heisenberg picture) for the field operators. Since the problem under consideration is that of propagation of the beam and not that of a field in a cavity, we perform the interchange $z = -ct$. As a result, the quantum equations of motion for the slowly varying parts of the field operators take the form:

$$\frac{\mathrm{d}}{\mathrm{d}z} E_\sigma^{(+)}(z) = \mathrm{i}\frac{1}{\hbar c}\frac{\hbar\omega}{2\epsilon_0} N\langle\gamma_{\sigma\tau\nu\rho}(\omega)\rangle_\Omega E_\tau^{(-)}(z)E_\nu^{(+)}(z)E_\rho^{(+)}(z), \tag{5}$$

where $\langle\ \rangle_\Omega$ stands for averaging over all possible orientations Ω of the molecule in the hyperpolarizability tensor:

$$\langle\gamma_{\sigma\tau\nu\rho}(\omega)\rangle_\Omega = \gamma_1(\omega)\delta_{\sigma\tau}\delta_{\nu\rho} + \gamma_2(\omega)\delta_{\sigma\nu}\delta_{\tau\rho} + \gamma_3(\omega)\delta_{\sigma\rho}\delta_{\tau\nu}. \tag{6}$$

Above, $\gamma_1(\omega)$, $\gamma_2(\omega)$ and $\gamma_3(\omega)$ are the respective rotational invariants of the molecular hyperpolarizability tensor [11]. For the configuration applied by us, eq. (5) with regard to [6] becomes:

$$\frac{\mathrm{d}}{\mathrm{d}z} E_x^{(+)}(z) = \frac{\mathrm{i}N}{\hbar c}\left(\frac{\hbar\omega}{2\epsilon_0}\right)\{[\gamma_2(\omega)+\gamma_3(\omega)]\,[E_x^{(-)}(z)E_x^{(+)}(z)E_x^{(+)}(z) + E_y^{(-)}(z)E_y^{(+)}E_x^{(+)}(z)]$$
$$+\gamma_1(\omega)[E_x^{(-)}(z)E_x^{(+)}(z)E_x^{(+)}(z) + E_x^{(-)}(z)E_y^{(+)}(z)E_y^{(+)}(z)]\}. \tag{7}$$

We obtain the equation for the y-component by way of the interchange $x \leftrightarrow y$ and that for the creation operators by taking the hermitean conjugate of (7). Since eqs. (7) cannot be solved strictly, we have recourse to a procedure used by us earlier [7] for the calculation of the variations undergone by the field correlation tensors of the beam along a sufficiently short path z in the isotropic medium.

The correlation tensor of nth order (which is a tensor of rank $2n$ with respect to the components of the fields) can be defined as follows [12]:

$$G^{(n)}_{\mu_1\ldots\mu_n\mu_{n+1}\ldots\mu_{2n}}(z) = \langle E_{\mu_1}^{(-)}(z)\ldots E_{\mu_n}^{(-)}(z)E_{\mu_{n+1}}^{(+)}(z)\ldots E_{\mu_{2n}}^{(+)}(z)\rangle, \tag{8}$$

where $\langle\ \rangle$ stands for the quantum-mechanical mean value over the states of the field. If the field variations along the propagation path z are small, the correlation tensor (8) can be expanded in a Taylor series in which only the term linear in z is of relevance:

$$G^{(n)}_{\cdots}(z) = G^{(n)}_{\cdots}(0) + \mathrm{d}G^{(n)}_{\cdots}(z)/\mathrm{d}z|_{z=0}z + \ldots \tag{9}$$

Applying (9), (7) and (3), one can calculate within an approximation linear in z the variations in coherence tensors of the beam traversing the medium.

The results for the correlation tensors of the 1st and 2nd orders are, in an approximation linear in z (we give but one of the components):

$$\Delta G_{xx}^{(1)}(z) = G_{xx}^{(1)}(z) - G_{xx}^{(1)}(0) = -(Nz/\hbar c)(\hbar\omega/2\epsilon_0)^3 \{[\gamma_2''(\omega) + \gamma_3''(\omega)](1 + \cos 2\eta \cos 2\theta)$$
$$+ \tfrac{1}{2}\gamma_1''(\omega)(1 + 2\cos 2\eta \cos 2\theta + \cos 4\eta) + \tfrac{1}{2}\gamma_1'(\omega) \sin 4\eta \sin 2\theta\}\langle a^{\dagger 2} a^2\rangle, \tag{10}$$

$$\Delta G_{xxxx}^{(2)}(z) = -(Nz/\hbar c)(\hbar\omega/2\epsilon_0)^4 \{\tfrac{1}{2}[\gamma_2''(\omega) + \gamma_3''(\omega)](1 + \cos 2\eta \cos 2\theta)^2$$
$$+ \tfrac{1}{4}\gamma_1''(\omega)(1 + \cos 2\eta \cos 2\theta)(1 + 2\cos 2\eta \cos 2\theta + \cos 4\eta)$$
$$+ \tfrac{1}{4}\gamma_1'(\omega)(1 + \cos 2\eta \cos 2\theta) \sin 4\eta \sin 2\theta\}[2\langle a^{\dagger 3} a^3\rangle + \langle a^{\dagger 2} a^2\rangle]. \tag{11}$$

In (10) and (11), the hyperpolarizability tensor has been split into real (prime) and imaginary (bis) parts. Averages of the type $\langle a^{\dagger n} a^n\rangle$ are nth order correlation functions of the incident beam. It is worth noting that the term proportional to $\langle a^{\dagger 2} a^2\rangle$ in (11) appears owing to our application of the commutation rules (3) and reflects the quantum properties of the field. This term is decisive for the dynamics of the photon antibunching effect.

3. Photon antibunching effect and its polarisation dependence

Eqs. (10) and (11) permit the calculation, in an approximation linear in z, of the magnitude of the Hanbury–Brown and Twiss effect. We obtain:

$$G_{xxxx}^{(2)}(z) - [G_{xx}^{(1)}(z)]^2 = G_{xxxx}^{(2)}(0) - [G_{xx}^{(1)}(0)]^2 - (Nz/\hbar c)(\hbar\omega/2\epsilon_0)^4$$
$$\times \{[\gamma_2''(\omega) + \gamma_3''(\omega)](1 + \cos 2\eta \cos 2\theta)^2 + \tfrac{1}{2}\gamma_1''(\omega)(1 + \cos 2\eta \cos 2\theta)(1 + 2\cos 2\eta \cos 2\theta + \cos 4\eta)$$
$$+ \tfrac{1}{2}\gamma_1'(\omega)(1 + \cos 2\eta \cos 2\theta) \sin 4\eta \sin 2\theta\}[\tfrac{1}{2}\langle a^{\dagger 2} a^2\rangle + \langle a^{\dagger 3} a^3\rangle - \langle a^{\dagger 2} a^2\rangle\langle a^\dagger a\rangle], \tag{12}$$

The expression (12) holds for arbitrary parameters θ and η of the polarisation ellipse and arbitrary photon statistics of the incident beam. A particularly interesting result is obtained if the incident beam is coherent i.e. if $\langle a^{\dagger n} a^n\rangle = \langle a^\dagger a\rangle^n$ and $G_{xxxx}^{(2)}(0) - [G_{xx}^{(1)}(0)]^2 = 0$. In this case, with the incident beam linearly polarized in the x-direction $\eta = 0$ and $\theta = 0$, we obtain from (12):

$$G_{xxxx}^{(2)}(z) - [G_{xx}^{(1)}(z)]^2 = -(2Nz/\hbar c)(\hbar\omega/2\epsilon_0)^4 [\gamma_1''(\omega) + \gamma_2''(\omega) + \gamma_3''(\omega)]\langle a^\dagger a\rangle^2. \tag{13}$$

We thus obtain a negative Hanbury–Brown and Twiss effect i.e. antibunching, of a magnitude proportional to the imaginary parts of the rotational invariants of the hyperpolarizability tensor. Since the imaginary part of the tensor $\gamma(\omega)$ describes two-photon absorption, the preceding result is in agreement with results derived earlier for two-photon absorption and proving the possibility of antibunching [13,14].

At circular polarisation of the incident beam $\eta = \pi/4$ we obtain, irrespective of θ:

$$G_{xxxx}^{(2)}(z) - [G_{xx}^{(1)}(z)]^2 = -\tfrac{1}{2}(Nz/\hbar c)(\hbar\omega/2\epsilon_0)^4 [\gamma_2''(\omega) + \gamma_3''(\omega)]\langle a^\dagger a\rangle^2. \tag{14}$$

Here, again, we obtain antibunching, related with two-photon absorption but dependent only on two parts of the hyperpolarizability tensor, $\gamma_2''(\omega)$ and $\gamma_3''(\omega)$.

For an incident beam elliptically polarized with the azimuth $\theta = \pi/4$, eq. (12) leads to:

$$G_{xxxx}^{(2)}(z) - [G_{xx}^{(1)}(z)]^2 = -\tfrac{1}{2}(Nz/\hbar c)(\hbar\omega/2\epsilon_0)^4 [\gamma_2''(\omega) + \gamma_3''(\omega) + \tfrac{1}{2}\gamma_1''(\omega)(1 + \cos 4\eta)$$
$$+ \tfrac{1}{2}\gamma_1'(\omega) \sin 4\eta]\langle a^\dagger a\rangle^2. \tag{15}$$

In eq. (15), besides antibunching related with two-photon absorption (one part of which is dependent on the ellipticity of the beam), we have a term dependent on the real part of the hyperpolarizability tensor differing from zero even for molecules not exhibiting two-photon absorption and thus non-zero for all molecules. The last mentioned term, in fact, is related with self-induced birefringence, and has already been discussed by us [8]. It has yet another, interesting property, consisting in a change in sign for reversed helicity of the incident beam $\eta \to -\eta$, when antibunching goes over into bunching. The optimal ellipticity for the observation of antibunching in self-induced birefringence amounts to $\eta = \pi/8$. The term of relevance, as is obvious from (12), is moreover sensitive to a change in sign of θ which causes a change in sign of the term. Hence, with regard to self-induced birefringence antibunching we have at our disposal two possibilities leading from antibunching to bunching and vice versa: (i) by reversing the helicity of the incident beam $\eta \to -\eta$ at azimuth maintained constant, and (ii) by changing the angle θ to $-\theta$, maintaining the helicity constant. This is of particular interest since the terms related with two-photon absorption, being even, fail to exhibit such properties.

Our calculations are approximate and cease to hold in the case when the variations in correlation tensor along the propagation path are considerable, as they may be at saturation of two-photon absorption. The range of applicability of the "short optical path" approximation applied here has been discussed previously with regard to harmonics generation [15].

Direct measurements or the antibunching effect in phenomena of light propagation appear to be beset with difficulties because the magnitude of the effect is inversely proportional to the mean number of photons of the incident beam. Nonetheless, in this respect, the method proposed quite recently by Bandilla and Ritze appears to be promising [9,10]. More recently, Wagner et al. [16] have made a highly interesting attempt at measuring indirectly the antibunching effect in second-harmonic generation by simulation of the process using an appropriately constructed "filter". Their measurements are in agreement with our theoretical predictions [2].

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