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On Nonlinear Optical Activity and Photon Statistics

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Abstract

A method of determining n-th order correlation tensors for the emergent beam is proposed for arbitrary statistics and polarisation of the incident beam. By this method, we obtained results for the signal in a correlator with two as well as with three symmetrically disposed detectors.

Inhalt

Über nichtlineare optische Aktivität und Photonen-Statistik. Eine Methode für die Bestimmung von Korrelationstensoren n-ter Ordnung für den austretenden Strahl wird vorgeschlagen, bei beliebiger Statistik und Polarisation des einfallenden Strahles. Die Methode ergibt Resultate für das Signal in einem Korrelator mit zwei oder mit drei symmetrisch aufgestellten Detektoren.

Introduction

Recently, *Atkins* and *Wilson* [1] have discussed changes in the statistical properties of the field due to nonlinear interactions with a system of freely orienting molecules. They envisaged a generalized birefringence experiment, consisting in a modification of the *Hanbury-Brown* and *Twiss* [2] and *Rebka-Pound* [3] experiments. We propose a method, different from that of Ref. [1], for the calculation of changes in the statistical properties of a beam traversing an isotropic, optically active medium. Our method presents the following two advantages: (i) the calculations can be conducted in terms of field

[1] 60–780 Poznań, Grunwaldzka 6.

operators, without having recourse to a particularized representation for quantal states of the field, and (ii) the transition to classical quantities is performed easily by replacement of the field operators by classical field strengths. Also, we believe the method is more highly effective with regard to calculations of correlation tensors of higher orders. As an example, we adduce results for tensors of correlations of order 1, 2 and 3 (tensors of ranks 2, 4 and 6) permitting the calculation of the signal in a correlator with two as well as in a correlator with three symmetrically disposed detectors, for arbitrary statistics of the incident beam. In particular, for a two-detector correlator and certain specialized statistics of the incident beam, our results reduce to those of *Atkins* and *Wilson* [1].

Theoretical

We shall be considering the electric field of a light beam, traversing a medium as a plane wave propagating in the z-direction of laboratory coordinates. We assume the field as stationary.

$$E_\sigma^{(+)}(z, t) = E_\sigma^{(+)}(k; z)\, e^{i(kz-\omega_k t)}. \tag{1}$$

Above, $E_\sigma^{(+)}(k; z)$ is a slowly varying function of z.

$$E_\sigma^{(+)}(k; 0) = i\left(\frac{\hbar\omega_k}{2\varepsilon_0}\right)^{\frac{1}{2}} e_\sigma^{\lambda}(k)\, a_{k\lambda} \tag{2}$$

where $e_\sigma^{\lambda}(k)$ is the polarisation vector of the incident beam and $a_{k\lambda}$ the annihilation operator of a photon with the wave vector $\vec{k}$ and polarisation λ fulfilling the commutation rules:

$$[a_{k\lambda}, a_{k'\lambda'}^{+}] = \delta_{kk'}\,\delta_{\lambda\lambda'}. \tag{3}$$

Since in (2) we have omitted the quantisation volume, the operator $n_{k\lambda} = a_{k\lambda}^{+}\, a_{k\lambda}$ has to be interpreted as the photon density. For light elliptically polarized and propagating along z, the polarisation vector $\vec{e}^{\lambda}$ has the following components [4]:

$$\begin{aligned} i\, e_x^{\lambda}(k) &= \cos\eta\cos\Theta - i\sin\eta\sin\Theta \\ i\, e_y^{\lambda}(k) &= \cos\eta\sin\Theta + i\sin\eta\cos\Theta \end{aligned} \tag{4}$$

where Θ is the azimuth and η the ellipticity of the incident beam.

The quantum-mechanical equation of motion for the operator of the field interacting with a system of optically active molecules (N per unit volume) can be written as follows[2]

$$\begin{aligned} \frac{d\,E_x^{(+)}(k; z)}{dz} = \frac{N\omega_k}{2\varepsilon_0 c^2}\{ & \varrho(\omega_k)\, E_y^{(+)}(k; z) \\ & + \sigma(\omega_k)\,[E_x^{(-)}(k; z)\, E_x^{(+)}(k; z)\, E_y^{(+)}(k; z) \\ & + E_y^{(-)}(k; z)\, E_y^{(+)}(k; z)\, E_y^{(+)}(k; z)]\} \end{aligned} \tag{5}$$

[2] Eq. (5) accounts for rotation effects only. The complete equation describing nonlinear interaction between the field and the system of N freely orienting molecules will be given in Ref. [5]. Its derivation is similar to the procedure of Refs. [6] and [7].

where $\varrho(\omega_k)$ and $\sigma(\omega_k)$ are isotropic averages of the tensors of optical rotation and hyper-rotation (identical to those of [1]).

The equation for the y-component is obtained by performing the interchange $x \leftrightarrow y$ and a change in sign of Eq. (5).

On traversing the path z in the medium, with the field (1), the n-th order correlation tensor (which is mathematically a tensor of rank 2n) can be defined as follows [8]:

$$G^{(n)}{}_{\mu_1 \ldots \mu_n \mu_{n+1} \ldots \mu_{2n}}(z) = \langle E_{\mu_1}{}^{(-)}(k; z) \ldots E_{\mu_n}{}^{(-)}(k; z)\, E_{\mu_{n+1}}{}^{(+)}(k; z) \ldots E_{\mu_{2n}}{}^{(+)}(k; z) \rangle_f \tag{6}$$

with $\langle\ \rangle_f$ denoting the quantum-mechanical mean value in the unknown final state of the field. If the changes in correlation tensors along the path z are small, one can write:

$$G^{(n)}{}_{\mu_1 \ldots \mu_n \mu_{n+1} \ldots \mu_{2n}}(z) = G^{(n)}{}_{\mu_1 \ldots \mu_n \mu_{n+1} \ldots \mu_{2n}}(0) + \left. \frac{d\, G^{(n)}{}_{\mu_1 \ldots \mu_n \mu_{n+1} \ldots \mu_{2n}}(z)}{dz} \right|_{z=0} z + \ldots \tag{7}$$

In the Taylor series expansion (7) of the correlation tensor, the expansion parameter is in fact not z but $\varrho(\omega_k)\, z$ or $\sigma(\omega_k)\, z$. With regard to the smallness of the tensors $\varrho(\omega_k)$ and $\sigma(\omega_k)$, we can neglect the terms of higher order. On having recourse to Eqs. (1) – (7), one can calculate the changes in coherence tensors in an approximation linear in z.

Results

For correlation tensors of order 1 (i.e. strictly of rank 2), we get:

$$\Delta\, G_{xx}{}^{(1)}(z) = G_{xx}{}^{(1)}(z) - G_{xx}{}^{(1)}(0) = -\Delta\, G_{yy}{}^{(1)}(z)$$
$$= \frac{\hbar\, \omega_k{}^2\, N\, z}{4\, \varepsilon_0{}^2\, c^2} \cos 2\eta \sin 2\Theta \left[\varrho(\omega_k) \langle a_{k\lambda}{}^+ a_{k\lambda} \rangle + \frac{\hbar \omega_k}{2\, \varepsilon_0} \sigma(\omega_k) \langle a_{k\lambda}{}^{+2} a_{k\lambda}{}^2 \rangle \right] \tag{8}$$

$$\Delta\, G_{xy}{}^{(1)}(z) = \Delta\, G_{yx}{}^{(1)}(z) = -\frac{\hbar\, \omega_k{}^2\, N\, z}{4\, \varepsilon_0{}^2\, c^2} \cos 2\,\eta \cos 2\,\Theta \left[\varrho(\omega_k) \langle a_{k\lambda}{}^+ a_{k\lambda} \rangle + \frac{\hbar \omega_k}{2\, \varepsilon_0} \sigma(\omega_k) \langle a_{k\lambda}{}^{+2} a_{k\lambda}{}^2 \rangle \right]. \tag{9}$$

The symbol $\langle\ \rangle$ denotes everywhere the quantum-mechanical mean values in the initial state of the field. Eqs. (8) and (9) permit the calculation of the rotation of the polarisation plane:

$$\varphi(z) \approx \frac{1}{2}\, \omega_k\, \mu_0\, N z \left[\varrho(\omega_k) + \frac{\hbar \omega_k}{2\, \varepsilon_0} \sigma(\omega_k) \frac{\langle a_{k\lambda}{}^{+2}\, \alpha_{k\lambda}{}^2 \rangle}{\langle a_{k\lambda}{}^+ a_{k\lambda} \rangle} \right]. \tag{10}$$

The first term of Eq. (10) accounts for natural optical activity. The second term describes nonlinear optical activity (cf. [9], [4] and the references there-

in). From Eq. (10), nonlinear optical activity is seen to depend not only on the intensity but moreover on the statistics of the incident beam. The result expressed by the nonlinear terms of Eq. (10) is twice larger for a chaotic beam than for a coherent beam of the same intensity.

For correlation tensors of order 2 (rank 4), we obtain $\left(\Theta = \frac{\pi}{4}\right)$

$$\Delta G^{(2)}{}_{xxxx}(z) = -\Delta G^{(2)}{}_{yyyy}(z) = \frac{\hbar^2 \omega_k{}^3 Nz}{8 \varepsilon_0{}^3 c^2} \cos 2\eta \left[\varrho(\omega_k) \langle a_{k\lambda}{}^{+2} a_{k\lambda}{}^2 \rangle + \frac{\hbar \omega_k}{2 \varepsilon_0} \sigma(\omega_k) \left(\langle a_{k\lambda}{}^{+3} a_{k\lambda}{}^3 \rangle + \frac{1}{2} \langle a_{k\lambda}{}^{+2} a_{k\lambda}{}^2 \rangle \right) \right] \tag{11}$$

$$\Delta G^{(2)}{}_{xyyx}(z) = -\Delta G^{(2)}{}_{yxxy}(z) = \frac{\hbar^3 \omega_k{}^4 Nz}{32 \varepsilon_0{}^4 c^2} \sigma(\omega_k) \cos 2\eta \langle a_{k\lambda}{}^{+2} a_{k\lambda}{}^2 \rangle \tag{12}$$

Eqs. (11) and (12) in conjunction with (8) and (9) permit calculations of the signal in the Rebka-Pound correlator [3]:

$$S_{xy}(z) \sim G^{(2)}{}_{xyyx}(z) - G^{(1)}{}_{xx}(z)\, G^{(1)}{}_{yy}(z)$$

for arbitrary statistics of the incident beam. In particular, for a coherent beam, the results obtained coincide with those of *Atkins* and *Wilson* [1].

For correlation tensors of order 3 (rank 6), the results are as follows $\left(\Theta = \frac{\pi}{4}\right)$:

$$\Delta G^{(3)}{}_{xxxxxx}(z) = -\Delta G^{(3)}{}_{yyyyyy}(z) = \frac{3 \hbar^3 \omega_k{}^4 Nz}{64 \varepsilon_0{}^4 c^2} \cos 2\eta \left[\varrho(\omega_k) \langle a_{k\lambda}{}^{+3} a_{k\lambda}{}^3 \rangle + \frac{\hbar \omega_k}{2 \varepsilon_0} \sigma(\omega_k) \left(\langle a_{k\lambda}{}^{+4} a_{k\lambda}{}^4 \rangle + \langle a_{k\lambda}{}^{+3} a_{k\lambda}{}^3 \rangle \right) \right] \tag{13}$$

$$\Delta G_{xyyyyx}(z) = -\frac{\hbar^3 \omega_k{}^4 Nz}{64 \varepsilon_0{}^4 c^2} \cos 2\eta \left[\varrho(\omega_k) \langle a_{k\lambda}{}^{+3} a_{k\lambda}{}^3 \rangle + \frac{\hbar \omega_k}{2 \varepsilon_0} \sigma(\omega_k) \left(\langle a_{k\lambda}{}^{+4} a_{k\lambda}{}^4 \rangle - \langle a_{k\lambda}{}^{+3} a_{k\lambda}{}^3 \rangle \right) \right] \tag{14}$$

$$\Delta G^{(3)}{}_{xxyyxx}(z) = \frac{\hbar^3 \omega_k{}^4 Nz}{64 \varepsilon_0{}^4 c^2} \cos 2\eta \left[\varrho(\omega_k) \langle a_{k\lambda}{}^{+3} a_{k\lambda}{}^3 \rangle + \frac{\hbar \omega_k}{2 \varepsilon_0} \sigma(\omega_k) \left(\langle a_{k\lambda}{}^{+4} a_{k\lambda}{}^4 \rangle + 3 \langle a_{k\lambda}{}^{+3} a_{k\lambda}{}^3 \rangle \right) \right]. \tag{15}$$

Eqs. (13)–(15) permit calculations of the signal in a correlator with three symmetrically disposed photomultipliers (see e.g. [10] and the references

therein). For certain particular states of the field, the mean values $\langle\ \rangle$ are readily calculated [8], [10]:

$$\begin{aligned} \langle a_{k\lambda}{}^{+n}\, a_{k\lambda}{}^{n}\rangle \text{ coherent} &= \langle a_{k\lambda}{}^{+}\, a_{k\lambda}\rangle^{n} \\ \langle a_{k\lambda}{}^{+n}\, a_{k\lambda}{}^{n}\rangle \text{ chaotic} &= n!\,\langle a_{k\lambda}{}^{+}\, a_{k\lambda}\rangle^{n} \\ \langle a_{k\lambda}{}^{+n}\, a_{k\lambda}{}^{n}\rangle\, n_{k\lambda}\text{-foton} &= n_{k\lambda}(n_{k\lambda}-1)\ldots(n_{k\lambda}-n+1). \end{aligned} \tag{16}$$

In the above cases, Eqs. (8)–(15) simplify considerably. A fuller discussion covering optical molecular reorientation [9] and other nonlinear effects will be given elsewhere [5].

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