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Photon correlations in multi-photon Raman processes

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Abstract. Non-linear interactions between incident and scattered photons (Stokes and anti-Stokes radiation) in Raman and hyper-Raman processes considered as elementary acts, are analysed. Fluctuations of phonons are shown to cause anticorrelation effects between incident and scattered photons. Antibunching of incident laser photons in spontaneous hyper-Raman processes occurs in a similar way to their antibunching in second-harmonic generation.

1. Introduction

The present paper is a continuation of earlier ones [1, 2], in which we dealt with the dynamics of anticorrelation effects in Raman and hyper-Raman scattering, assuming that (i) the phonon mode is initially chaotic, whereas the laser, Stokes and anti-Stokes modes are coherent ; and (ii) the laser mode is initially coherent but all other modes are chaotic.

Our aim is to study the intermodal statistical relationship between incident and scattered photons in Raman and hyper-Raman events when (i) the laser and Stokes (anti-Stokes) modes are initially coherent whereas the phonon mode is chaotic and the anti-Stokes (Stokes) mode is in vacuum state ; (ii) the laser and phonon modes are initially chaotic whereas the Stokes and anti-Stokes modes are coherent ; and (iii) all modes are initially chaotic. We show that in some cases fluctuations of photons may cause the appearance of photon anticorrelation in Raman as well as hyper-Raman events. Moreover, in spontaneous hyper-Raman a special case of anticorrelation effect can occur, namely photon antibunching, which does not take place in Raman scattering. Attention has been drawn to the occurrence of this effect by Simaan [3]. In addition, we consider some aspects of the existence of the Glauber–Sudarshan representation related to non-linear interaction between incident and scattered modes.

2. Equations of motion

In a previous paper [4], the foundations of the semiclassical theory of multi-photon Rayleigh and Raman scattering were formulated taking into account multipolar electric and magnetic quantum transitions. Here, we develop in full the quantum theory of multi-photon Raman scattering, albeit in the approximation of electric (in general non-linear) dipole transitions.

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The total hamiltonian describing multi-photon Raman scattering may be expressed as the sum of a free hamiltonian $\hat{H}_0$ and an interaction hamiltonian $\hat{H}_{\text{int}}$

$$\hat{H}=\hat{H}_0+\hat{H}_{\text{int}}. \tag{2.1}$$

The free hamiltonian has the form

$$\hat{H}_0=\hbar\omega_l\hat{a}_l{}^\dagger\hat{a}_l+\hbar\omega_s\hat{a}_s{}^\dagger\hat{a}_s+\hbar\omega_a\hat{a}_a{}^\dagger\hat{a}_a+\hbar\omega_p\hat{a}_p{}^\dagger\hat{a}_p, \tag{2.2}$$

where the frequencies and boson operators of the laser, Stokes, anti-Stokes and phonon modes are labelled by the subscripts l, s, a and p respectively. The effective interaction hamiltonian has the form

$$\hat{H}_{\text{int}}=\hbar(\kappa_s{}^{(k)}\hat{a}_l{}^k\hat{a}_p{}^\dagger\hat{a}_s{}^\dagger+\text{h.c.})+\hbar(\kappa_a{}^{(k)}\hat{a}_l{}^k\hat{a}_p\hat{a}_a{}^\dagger+\text{h.c.}), \tag{2.3}$$

where $\kappa_s{}^{(k)}$ and $\kappa_a{}^{(k)}$ are the coupling constants for the Stokes and anti-Stokes processes respectively. The energy conservation condition

$$\omega_{s,a}=k\omega_l\mp\omega_p \tag{2.4}$$

is assumed to hold. As seen from (2.3), a Stokes (anti-Stokes) photon is the result of annihilation of k laser photons incident from a single laser mode, and the creation (annihilation) of a phonon in a non-linear medium. If in (2.3) and (2.4) $k=1$, we have the well-known hamiltonian describing the single act of Raman scattering. If $k=2$, the hamiltonian (2.3) models the degenerate hyper-Raman effect† [5].

The Heisenberg equations of motion resulting from the model hamiltonian (2.1)–(2.3) are given by

$$\frac{d\hat{a}_l(t)}{dt}=-i\omega_l\hat{a}_l(t)-ik\kappa_s{}^{(k)*}\,\hat{a}_l{}^{\dagger(k-1)}(t)\hat{a}_s(t)\hat{a}_p(t)-ik\kappa_a{}^{(k)*}\,\hat{a}_l{}^{\dagger(k-1)}(t)\hat{a}_a(t)\hat{a}_p{}^\dagger(t), \tag{2.5}$$

$$\frac{d\hat{a}_s(t)}{dt}=-i\omega_s\hat{a}_s(t)-i\kappa_s{}^{(k)}\hat{a}_l{}^k(t)\hat{a}_p{}^\dagger(t), \tag{2.6}$$

$$\frac{d\hat{a}_a(t)}{dt}=-i\omega_a\hat{a}_a(t)-i\kappa_a{}^{(k)}\hat{a}_l{}^k(t)\hat{a}_p(t), \tag{2.7}$$

$$\frac{d\hat{a}_p(t)}{dt}=-i\omega_p\hat{a}_p(t)-i\kappa_s{}^{(k)}\hat{a}_l{}^k(t)\hat{a}_s{}^\dagger(t)-i\kappa_a{}^{(k)*}\,\hat{a}_l{}^{\dagger k}(t)\hat{a}_a(t). \tag{2.8}$$

Denoting the number operators $\hat{a}_l{}^\dagger(t)\hat{a}_l(t)$, $\hat{a}_s{}^\dagger(t)\hat{a}_s(t)$, $\hat{a}_a{}^\dagger(t)\hat{a}_a(t)$, and $\hat{a}_p{}^\dagger(t)\hat{a}_p(t)$ by $\hat{n}_l(t)$, $\hat{n}_s(t)$, $\hat{n}_a(t)$ and $\hat{n}_p(t)$ respectively, we may verify that

$$\left.\begin{aligned}\frac{d}{dt}(\hat{n}_l(t)+k\hat{n}_s(t)+k\hat{n}_a(t))&=0,\\ \frac{d}{dt}(\hat{n}_p(t)+\hat{n}_a(t)-\hat{n}_s(t))&=0.\end{aligned}\right\} \tag{2.9}$$

Therefore $\hat{n}_l(t)+k\hat{n}_s(t)+k\hat{n}_a(t)$ and $\hat{n}_p(t)+\hat{n}_a(t)-\hat{n}_s(t)$ are constants of motion and are simultaneously satisfied for any time.

† Non-degenerate hyper-Raman processes have been considered by the present authors [6].

The solutions of the Heisenberg equations of motion (2.5)–(2.8) in the short time approximation for $k=1$ and $k=2$ have been obtained by Szlachetka *et al.* [1] and Peřinová *et al.* [2] respectively.

3. Correlation functions

In this paper we consider two kinds of statistical moments—variances of the intensities and covariance functions for the number of photons, i.e.

$$\langle(\Delta W_i)^2\rangle = \mathrm{Tr}\,\{\hat{a}_i^{\dagger 2}(t)\hat{a}_i^2(t)\hat{\rho}(0)\} - (\mathrm{Tr}\,\{\hat{a}_i^\dagger(t)\hat{a}_i(t)\hat{\rho}(0)\})^2, \tag{3.1}$$

$$\begin{aligned}\langle\Delta W_i \Delta W_j\rangle = {} & \mathrm{Tr}\,\{\hat{a}_i^\dagger(t)\hat{a}_i(t)\hat{a}_j^\dagger(t)\hat{a}_j(t)\hat{\rho}(0)\} \\ & - \mathrm{Tr}\,\{\hat{a}_i^\dagger(t)\hat{a}_i(t)\hat{\rho}(0)\}\,\mathrm{Tr}\,\{\hat{a}_j^\dagger(t)\hat{a}_j(t)\hat{\rho}(0)\},\ i, j = \mathrm{l, s, a},\end{aligned} \tag{3.2}$$

where $\hat{\rho}(0)$ is the total density operator of the system at the time $t=0$ in the Glauber–Sudarshan representation.

We evaluate the functions (3.1) and (3.2) for the following cases:

(i) The laser and Stokes modes are initially coherent while the phonon mode is chaotic and the anti-Stokes mode is in vacuum state, i.e.

$$\hat{\rho}(0) = (\pi n_\mathrm{p})^{-1} \int \exp\left\{-\frac{|\xi_\mathrm{p}|^2}{n_\mathrm{p}}\right\} |\xi_\mathrm{l}, \xi_\mathrm{s}, 0, \xi_\mathrm{p}\rangle\langle\xi_\mathrm{l}, \xi_\mathrm{s}, 0, \xi_\mathrm{p}|\, d^2\xi_\mathrm{p}. \tag{3.3}$$

(ii) The laser and anti-Stokes modes are initially coherent while the phonon mode is chaotic and the Stokes mode is in vacuum state, i.e.

$$\hat{\rho}(0) = (\pi n_\mathrm{p})^{-1} \int \exp\left\{-\frac{|\xi_\mathrm{p}|^2}{n_\mathrm{p}}\right\} |\xi_\mathrm{l}, 0, \xi_\mathrm{a}, \xi_\mathrm{p}\rangle\langle\xi_\mathrm{l}, 0, \xi_\mathrm{a}, \xi_\mathrm{p}|\, d^2\xi_\mathrm{p}. \tag{3.4}$$

(iii) The laser and phonon modes are initially chaotic while the Stokes and anti-Stokes modes are coherent, i.e.

$$\begin{aligned}\hat{\rho}(0) = (\pi^2 n_\mathrm{l} n_\mathrm{p})^{-1} \int \exp & \left\{-\frac{|\xi_\mathrm{l}|^2}{n_\mathrm{l}} - \frac{|\xi_\mathrm{p}|^2}{n_\mathrm{p}}\right\} \\ & \times |\xi_\mathrm{l}, \xi_\mathrm{s}, \xi_\mathrm{a}, \xi_\mathrm{p}\rangle\langle\xi_\mathrm{l}, \xi_\mathrm{s}, \xi_\mathrm{a}, \xi_\mathrm{p}|\, d^2\xi_\mathrm{l} d^2\xi_\mathrm{p}.\end{aligned} \tag{3.5}$$

(iv) All modes are initially chaotic, i.e.

$$\hat{\rho}(0) = (\pi^4 n_\mathrm{l} n_\mathrm{s} n_\mathrm{a} n_\mathrm{p})^{-1} \int \prod_{\mathrm{l, s, a, p}} \exp\left\{-\frac{|\xi_i|^2}{n_i}\right\} |\{\xi_i\}\rangle\langle\{\xi_i\}|\, d^2\xi_i, \tag{3.6}$$

where $|\xi_i\rangle$ is the normalized eigenstate of the annihilation operator $\hat{a}_i$ with complex eigenvalue ξ_i, i.e.

$$\hat{a}_i|\xi_i\rangle = \xi_i|\xi_i\rangle.$$

The average numbers of bosons being initially chaotic are denoted by n_i.

4. Interaction between laser and Stokes photons

4.1. *The laser and Stokes modes are initially coherent while the phonon mode is chaotic and the anti-Stokes mode is in vacuum state*

In order to evaluate $\langle\Delta W_\mathrm{l} \Delta W_\mathrm{s}\rangle$ for the Raman effect we use the solutions (2.11) and (2.12) of the Heisenberg equations of motion considered in our previous paper [1]. Inserting those solutions and (3.3) into (3.2) and taking the trace over initial states we obtain with accuracy up to t^2

$$\langle \Delta W_l \Delta W_s \rangle_R = -|\kappa_s^{(1)}|^2 t^2 |\xi_l|^2 |\xi_s|^2 (2n_p + 1). \tag{4.1}$$

The relative covariance function $\langle \Delta W_l \Delta W_s \rangle_R / \langle W_l \rangle_R \langle W_s \rangle_R$ is given by

$$\frac{\langle \Delta W_l \, \Delta W_s \rangle_R}{\langle W_l \rangle_R \langle W_s \rangle_R} = -|\kappa_s^{(1)}|^2 t^2 (2n_p + 1), \tag{4.1 a}$$

where $\langle W_l \rangle_R$ and $\langle W_s \rangle_R$ is the average number of laser and Stokes photons respectively, defined as

$$\langle W_i \rangle = \text{Tr}\,\{\hat{\rho}(0)\hat{a}_i^\dagger(t)\hat{a}_i(t)\}, \quad i = l, \text{s}.$$

From the experimental point of view it is of interest how the short-time approximation restricts the power of the incident laser beam and what is the order of magnitude of the anticorrelation effect (4.1 *a*). Let us consider this problem. The following condition [7]

$$|\kappa_s^{(1)} \xi_l t| < 1$$

is a consequence of the short-time approximation, where $t = z/c$ is the interaction time, z the path traversed in the Raman medium, and c the velocity of light. For a medium of $z = 1$ cm, the interaction time is of order 10^{-10} s. Taking the coupling constant $\kappa_s^{(1)} = 100\ \text{s}^{-1}$ [8] we have

$$|\xi_l|^2 < |\kappa_s^{(1)} t|^{-2} = 10^{16},$$

which is equivalent to a radiation intensity of $I < 10^{26}$ photons/s cm^2 < 10 MW/cm^2. From (4.1 *a*), we see that relative covariance of photons depends strongly on the initial average number of phonons. For instance, for $n_p = 10^{12}$ (hydrogen gas at $p = 10$ atm and 300 K [9]) and $\kappa_s^{(1)} = 100\ \text{s}^{-1}$, the order of magnitude of the function (4.1 *a*) is 10^{-4}.

Similarly, using the solutions of the Heisenberg equations for hyper-Raman processes obtained by Peřinová *et al.* [2], we have

$$\langle \Delta W_l \Delta W_s \rangle_{h-R} = -2|\kappa_s^{(2)}|^2 t^2 |\xi_l|^2 |\xi_s|^2 (2|\xi_l|^2 n_p + 2n_p + |\xi_l|^2). \tag{4.2}$$

The variances of the intensities are given by

$$\langle (\Delta W_l)^2 \rangle_R = 2|\kappa_s^{(1)}|^2 t^2 |\xi_l|^2 |\xi_s|^2 n_p, \tag{4.3}$$

$$\langle (\Delta W_l)^2 \rangle_{h-R} = |\kappa_s^{(2)}|^2 t^2 \{4|\xi_s|^2 n_p (2|\xi_l|^4 + 6|\xi_l|^2 + 1) - 2|\xi_l|^4 (|\xi_s|^2 + n_p + 1)\} - 2|\kappa_a^{(2)}|^2 t^2 |\xi_l|^4 n_p, \tag{4.4}$$

$$\langle (\Delta W_s)^2 \rangle_R = 2|\kappa_s^{(1)}|^2 t^2 |\xi_l|^2 |\xi_s|^2 (n_p + 1), \tag{4.5}$$

$$\langle (\Delta W_s)^2 \rangle_{h-R} = 2|\kappa_s^{(2)}|^2 t^2 |\xi_l|^4 |\xi_s|^2 (n_p + 1). \tag{4.6}$$

From (4.1)–(4.6) we see that in the presence of stimulated Stokes emission ($\xi_s \neq 0$) : (1) the functions (4.1) and (4.2) exhibit anticorrelation between incident laser photons and Stokes photons and (2) the functions (4.3)–(4.6) exhibit bunching of photons in laser and Stokes modes. Anticorrelation (4.1) and (4.2) occurs even at $n_p = 0$, i.e. when the phonon mode is initially in vacuum state, as a result of spontaneous phonon emission.

In the presence of spontaneous Stokes emission ($\xi_s=0$): (1) the functions (4.1) and (4.2) equal zero meaning that no correlation occurs between laser and Stokes photons; (2) the variances of the intensities (4.3), (4.5) and (4.6) show that in the laser and Stokes modes in Raman scattering and in the Stokes mode in hyper-Raman scattering the photons are uncorrelated; and (3) antibunching of laser photons in the hyper-Raman effect takes place. This is a reflection of the fact that two laser photons are simultaneously annihilated from the laser mode in this non-linear process. The above antibunching is similar to antibunching of laser photons in second-harmonic generation [10, 11], where two photons are simultaneously annihilated from the fundamental laser beam.

One may note that in the presence of stimulated Stokes emission, there occurs antibunching of laser photons in hyper-Raman processes if $n_p=0$, namely

$$\langle(\Delta W_l)^2\rangle_{\text{h-R}} = -2|\kappa_s^{(2)}|^2 t^2 |\xi_l|^4(|\xi_s|^2+1). \tag{4.7}$$

This effect is a result of the intrinsically quantum mechanical phenomenon, consisting in phonon vacuum fluctuation.

4.2. *The laser and phonon modes are initially chaotic while the Stokes and anti-Stokes modes are coherent*

If, at the time $t=0$, the systems are described by the density operator (3.4), we obtain the following results:

$$\langle\Delta W_l \Delta W_s\rangle_{\text{R}} = |\kappa_s^{(1)}|^2 t^2\{n_l^2(n_p+|\xi_s|^2+1) - n_l|\xi_s|^2(2n_p+1)\}, \tag{4.8}$$

$$\langle\Delta W_l \Delta W_s\rangle_{\text{h-R}} = |\kappa_s^{(2)}|^2 t^2\{4n_l^3(n_p+|\xi_s|^2+1) - 4n_l|\xi_s|^2(3n_l n_p + n_l + n_p)\}, \tag{4.9}$$

$$\langle(\Delta W_l)^2\rangle_{\text{R}} = n_l^2 + 2|\kappa_s^{(1)}|^2 t^2 n_l|\xi_s|^2 n_p - 2|\kappa_s^{(1)}|^2 t^2 n_l^2(|\xi_s|^2+n_p+1) + 2|\kappa_a^{(1)}|^2 t^2 n_l|\xi_a|^2(n_p+1) - 2|\kappa_a^{(1)}|^2 t^2 n_l^2(n_p-|\xi_a|^2), \tag{4.10}$$

$$\langle(\Delta W_l)^2\rangle_{\text{h-R}} = n_l^2 + 2|\kappa_s^{(2)}|^2 t^2\{4|\xi_s|^2 n_p(4n_l^2+6n_l+1) - 4n_l^2(|\xi_s|^2 + n_p+1)\} + 2|\kappa_a^{(2)}|^2 t^2\{4|\xi_a|^2(n_p+1) \times (4n_l^2+6n_l+1) + 4n_l^2(|\xi_a|^2 - n_p)\}, \tag{4.11}$$

$$\langle(\Delta W_s)^2\rangle_{\text{R}} = 2|\kappa_s^{(1)}|^2 t^2 n_l|\xi_s|^2(n_p+1), \tag{4.12}$$

$$\langle(\Delta W_s)^2\rangle_{\text{h-R}} = 4|\kappa_s^{(2)}|^2 t^2 n_l^2|\xi_s|^2(n_p+1). \tag{4.13}$$

Comparing the above results with those obtained in § 4.1 we see essential differences between them. The functions (4.8) and (4.9) are positive for $n_l > n_p$ and $n_l > n_s$, which in practice takes place. Therefore, in the presence of stimulated Stokes emission, we have correlation effects between the laser and Stokes photons. Correlation effects between these modes occur in spontaneous Stokes emission as well. Moreover, from (4.10) and (4.11) we see that the initially chaotic laser beam remains chaotic for $t>0$ meaning that bunching of laser photons takes place throughout.

Generally, we may conclude that an initially chaotic laser beam damages anticorrelation effects between laser and Stokes photons in Raman and hyper-Raman scattering as well as antibunching of laser photons in hyper-Raman scattering.

4.3. *The laser, Stokes, anti-Stokes and phonon modes are initially chaotic*

In this case we have

$$\langle \Delta W_l \Delta W_s \rangle_R = |\kappa_s^{(1)}|^2 t^2 \{ n_l^2 (n_p + n_s + 1) - n_s^2 (n_l - n_p) - n_l n_s (2n_p + 1) \}, \quad (4.14)$$

$$\langle \Delta W_l \Delta W_s \rangle_{h-R} = |\kappa_s^{(2)}|^2 t^2 \{ 4n_l^3 (n_p + n_s + 1) - 4n_s^2 (n_l^2 - n_p) + 8 n_l n_s n_p - 4 n_l n_s (3 n_l n_p + n_l + n_p) \}. \quad (4.15)$$

As is seen from (4.14) and (4.15), the laser and Stokes photons are mutually correlated in spontaneous as well as stimulated scattering, similarly as in § 4.2. Moreover, one can prove that, in the laser and Stokes modes, bunching of photons takes place always, i.e.

$$\langle (\Delta W_i)^2 \rangle_R > 0, \quad \langle (\Delta W_i)^2 \rangle_{h-R} > 0, \quad i = l, s.$$

5. Interaction between laser and anti-Stokes modes

5.1. *The laser and anti-Stokes modes are initially coherent while the phonon mode is chaotic and the Stokes mode is in vacuum state*

In this case we have

$$\langle \Delta W_l \Delta W_a \rangle_R = - |\kappa_a^{(1)}|^2 t^2 |\xi_l|^2 |\xi_a|^2 (2n_p + 1), \quad (5.1)$$

$$\langle \Delta W_l \Delta W_a \rangle_{h-R} = -2 |\kappa_a^{(2)}|^2 t^2 |\xi_l|^2 |\xi_a|^2 \{ |\xi_l|^2 (2n_p + 1) + 2(n_p + 1) \}, \quad (5.2)$$

$$\langle (\Delta W_l)^2 \rangle_R = 2 |\kappa_a^{(1)}|^2 t^2 |\xi_l|^2 |\xi_a|^2 (n_p + 1), \quad (5.3)$$

$$\langle (\Delta W_l)^2 \rangle_{h-R} = |\kappa_a^{(2)}|^2 t^2 \{ 4 |\xi_a|^2 (n_p + 1)(2 |\xi_l|^4 + 6 |\xi_l|^2 + 1) + 2 |\xi_l|^4 (|\xi_a|^2 - n_p) \} - |\kappa_s^{(2)}|^2 t^2 \, 2 |\xi_l|^4 (n_p + 1), \quad (5.4)$$

$$\langle (\Delta W_a)^2 \rangle_R = 2 |\kappa_a^{(1)}|^2 t^2 |\xi_l|^2 |\xi_a|^2 n_p, \quad (5.5)$$

$$\langle (\Delta W_a)^2 \rangle_{h-R} = 2 |\kappa_a^{(2)}|^2 t^2 |\xi_l|^4 |\xi_a|^2 n_p. \quad (5.6)$$

In the presence of stimulated anti-Stokes emission the above expressions lead to the following results.: (1) between the laser and anti-Stokes modes, photon anticorrelation effects take place ; (2) in individual modes, bunching of photons occurs.

In the absence of stimulated anti-Stokes emission : (1) the anticorrelation effects (5.1) and (5.2) vanish, whereas with the hyper-Raman effect, in the laser mode antibunching appears ; and (2) with the Raman effect, the photons in laser and anti-Stokes modes, and in hyper-Raman in anti-Stokes modes, are uncorrelated.

5.2. *The laser and phonon modes are initially chaotic, while the Stokes and anti-Stokes modes are coherent*

In this case we have

$$\langle\Delta W_l \Delta W_a\rangle_R = |\kappa_a^{(1)}|^2 t^2\{n_l^2(n_p - |\xi_a|^2) - n_l|\xi_a|^2(2n_p+1)\}, \quad (5.7)$$

$$\langle\Delta W_l \Delta W_a\rangle_{h-R} = |\kappa_a^{(2)}|^2 t^2\{-4n_l^3(|\xi_a|^2 - n_p) - 4n_l|\xi_a|^2(3n_l n_p + 2n_l + n_p + 1) - 4n_l^2 n_a\}, \quad (5.8)$$

$$\langle(\Delta W_a)^2\rangle_R = 2|\kappa_a^{(1)}|^2 t^2 n_l|\xi_a|^2 n_p, \quad (5.9)$$

$$\langle(\Delta W_a)^2\rangle_{h-R} = 4|\kappa_a^{(2)}|^2 t^2 n_l^2|\xi_a|^2 n_p. \quad (5.10)$$

The functions $\langle(\Delta W_l)^2\rangle_R$ and $\langle(\Delta W_l)^2\rangle_{h-R}$ are identical to (4.10) and (4.11) respectively.

Comparing the above results with those derived in § 5.1, we see that anti-correlations between the laser and anti-Stokes photons may occur as a result of phonon vacuum fluctuations. Therefore, if $n_p = 0$, (1) the functions (5.7) and (5.8) exhibit photon anticorrelation, and (2) the photons in anti-Stokes mode are uncorrelated.

5.3. *The laser, Stokes, anti-Stokes and phonon modes are initially chaotic*

In this case we have

$$\langle\Delta W_l \Delta W_a\rangle_R = |\kappa_a^{(1)}|^2 t^2\{n_l^2(n_p - n_a) + n_a^2(n_l + n_p + 1) - n_l n_a(2n_p+1)\}, \quad (5.11)$$

$$\langle\Delta W_l \Delta W_a\rangle_{h-R} = |\kappa_a^{(2)}|^2 t^2\{-4n_l^3(n_a - n_p) - 4n_l^2 n_a - 4n_l n_a(3n_l n_p + 2n_l + n_p + 1) + 2n_a^2 n_l(n_l + 4n_p + 1)\}. \quad (5.12)$$

The variances of the intensities

$$\langle(\Delta W_i)^2\rangle_R, \quad \langle(\Delta W_i)^2\rangle_{h-R}, \quad i = l, a,$$

are always positive.

The functions (5.11) and (5.12) are negative provided that $n_p = 0$ and $n_l > n_a$.

Generally, we may emphasize that fluctuations of photons can provoke anticorrelation between the laser and anti-Stokes photons but are unable to create anticorrelation between the laser and Stokes photons.

6. Characteristic functions and quasidistributions

All the results obtained in §§ 4 and 5 can be derived easily from quantum characteristic functions; however, these functions are mathematically cumbersome. In this section, we draw attention to certain characteristic functions of a mathematically interesting form, which have not been considered in our earlier articles [1, 2].

If all modes are initially chaotic, the joint normally ordered characteristic functions for the laser and Stokes (anti-Stokes) modes have the form

$$C_N(\beta_l, \beta_i, t) = \exp[-|\beta_l|^2 B_l(t) - |\beta_i|^2 B_i(t) + |\beta_l|^2|\beta_i|^2 B_{li}(t)], \quad i = s, a, \quad (6.1)$$

where

$$B_l(t)=\langle\hat{a}_l^\dagger(t)\hat{a}_l(t)\rangle=n_l-|\kappa_s^{(1)}|^2t^2n_l(n_s+n_p+1)+|\kappa_s^{(1)}|^2t^2n_sn_p$$
$$+|\kappa_a^{(1)}|^2t^2n_l(n_a-n_p)+|\kappa_a^{(1)}|^2t^2n_a(n_p+1), \qquad (6.2)$$

$$B_s(t)=\langle\hat{a}_s^\dagger(t)\hat{a}_s(t)\rangle=n_s+|\kappa_s^{(1)}|^2t^2n_l(n_s+n_p+1)-|\kappa_s^{(1)}|^2t^2n_sn_p, \qquad (6.3)$$

$$B_a(t)=\langle\hat{a}_a^\dagger(t)\hat{a}_a(t)\rangle=n_a+|\kappa_a^{(1)}|^2t^2n_l(n_p-n_a)$$
$$-|\kappa_a^{(1)}|^2t^2n_a(n_p+1), \qquad (6.4)$$

$$B_{la}(t)=\langle\Delta W_l\Delta W_a\rangle=|\kappa_a^{(1)}|^2t^2\{n_l^2(n_p-n_a)+n_a^2(n_p+n_l+1)$$
$$-n_ln_a(2n_p+1)\}, \qquad (6.5)$$

$$B_{ls}(t)=\langle\Delta W_l\Delta W_s\rangle=|\kappa_s^{(1)}|^2t^2\{n_l^2(n_p+n_s+1)$$
$$-n_s^2(n_l-n_p)-n_ln_s(2n_p+1)\}. \qquad (6.6)$$

The function (6.1) can be obtained with ease from the function (4.29) considered in our earlier publication [1] on additionally averaging that function over the initially chaotic laser mode.

With $C_N(\beta_l, \beta_i, t)$ available, let us consider the existence problem of the Glauber–Sudarshan quasidistribution function $\Phi_N(\xi_l, \xi_i, t)$ which is the inverse Fourier transform of $C_N(\beta_l, \beta_i, t)$, i.e.

$$\Phi_N(\xi_l, \xi_i, t)=\pi^{-4}\int C_N(\beta_l, \beta_i, t)$$
$$\times\exp(-\beta_l\xi_l^*-\beta_i\xi_i^*+\beta_l^*\xi_l+\beta_i^*\xi_i)d^2\beta_l d^2\beta_i, \quad i=\text{s, a}. \qquad (6.7)$$

If $B_{li}(t)>0$, the function $C_N(\beta_l, \beta_i, t)$ does not belong to the class of tempered distributions. Thus $C_N(\beta_l, \beta_i, t)$ does not possess a Fourier transform. On the other hand, if $B_{li}(t)<0$ anticorrelation takes place which excludes the existence of Φ_N as an ordinary function. Concluding, interaction between initially chaotic laser and Stokes (anti-Stokes) modes excludes the existence of the function $\Phi_N(\xi_l, \xi_i, t)$. Nevertheless, it may be of some interest to note that $\Phi_N(\xi_l, t)$ and $\Phi_N(\xi_i, t)$ exist separately and have the form

$$\Phi_N(\xi_j, t)=(\pi B_j(t))^{-1}\exp\left\{-\frac{|\xi'_j|^2}{B_j(t)}\right\}, \quad j=\text{l, s, a}. \qquad (6.8)$$

In order to illustrate antibunching of laser photons in the spontaneous hyper-Raman effect let us consider the characteristic function $C_N(\beta_l, t)$ for the laser mode. Using earlier results [2] we obtain, after some calculation,

$$C_N(\beta_l, t)=\exp(-\beta_l^*\xi_l(t)+\beta_l\xi_l^*(t)+\tfrac{1}{2}\beta_l^{*2}C_l(t)+\tfrac{1}{2}\beta_l^2C_l^*(t)), \qquad (6.9)$$

where

$$\xi_l(t)=(\xi_l-|\kappa_s^{(2)}|^2t^2\xi_l|\xi_l|^2(n_p+1)-|\kappa_a^{(2)}|^2t^2\xi_l|\xi_l|^2n_p)\exp(-i\omega_lt),$$
$$C_l(t)=(-|\kappa_s^{(2)}|^2t^2\xi_l^2(n_p+1)-|\kappa_a^{(2)}|^2t^2\xi_l^2n_p)\exp(-2i\omega_lt).$$

From (6.9) we easily obtain

$$\langle(\Delta W_l)^2\rangle_{\text{h-R}}=C_l(t)\xi_l^{*2}(t)+C_l^*(t)\xi_l^2(t)$$
$$=-2|\kappa_s^{(2)}|^2t^2|\xi_l|^4(n_p+1)-2|\kappa_a^{(2)}|^2t^2|\xi_l|^4n_p. \qquad (6.10)$$

Initial state of the system / Correlation function	L, s-coherent, p-chaotic, a-vacuum	L, a-coherent, p-chaotic, s-vacuum	L-coherent, p-chaotic, s, a-vacuum	L, p-chaotic, s, a-coherent	L, p-chaotic, s, a-vacuum	L-chaotic, s, a-coherent, p-vacuum	L, s, a, p-chaotic	L, s, a-chaotic, p-vacuum
$\langle(\Delta W_L)^2\rangle$	bunching	bunching	uncorrelation	bunching	bunching	bunching	bunching	bunching
$\langle(\Delta W_s)^2\rangle$	bunching	uncorrelation	uncorrelation	bunching	uncorrelation	bunching	bunching	bunching
$\langle(\Delta W_a)^2\rangle$	uncorrelation	bunching	uncorrelation	bunching	uncorrelation	uncorrelation	bunching	bunching
$\langle\Delta W_L \Delta W_s\rangle$	anticorrelation	uncorrelation	uncorrelation	correlation	correlation	correlation	correlation	correlation
$\langle\Delta W_L \Delta W_a\rangle$	uncorrelation	anticorrelation	uncorrelation	correlation	correlation	anticorrelation	correlation	anticorrelation

Figure 1. Raman scattering. Correlation, anticorrelation and uncorrelation among photons as a function of the initial statistical properties of the system.

INITIAL STATE OF THE SYSTEM / CORRELATION FUNCTION	L,s-coherent p-chaotic a-vacuum	L,a-coherent p-chaotic s-vacuum	L-coherent p-chaotic s,a-vacuum	L,p-chaotic s,a-coherent	L,p-chaotic s,a-vacuum	L-chaotic s,a-coherent p-vacuum	L,s,p,a-chaotic	L,s,a-chaotic p-vacuum
$\langle(\Delta W_L)^2\rangle$	bunching	bunching	antibunching	bunching	bunching	bunching	bunching	bunching
$\langle(\Delta W_s)^2\rangle$	bunching	uncorrelation	uncorrelation	bunching	uncorrelation	bunching	bunching	bunching
$\langle(\Delta W_a)^2\rangle$	uncorrelation	bunching	uncorrelation	bunching	uncorrelation	uncorrelation	bunching	bunching
$\langle\Delta W_L \Delta W_s\rangle$	anticorrelation	uncorrelation	uncorrelation	correlation	correlation	correlation	correlation	correlation
$\langle\Delta W_L \Delta W_a\rangle$	uncorrelation	anticorrelation	uncorrelation	correlation	correlation	anticorrelation	correlation	anticorrelation

Figure 2. Hyper-Raman scattering. Correlation, anticorrelation and uncorrelation among photons as a function of the initial statistical properties of the system.

The existence of antibunching of photons (6.10) excludes the existence of a quasidistribution function $\Phi_N(\xi_l, t)$ for the laser mode in spontaneous hyper-Raman scattering.

7. Conclusions

The quantum statistical properties of the Raman and hyper-Raman effects have been considered in self-consistent formalism. Correlation, anticorrelation or uncorrelation among the photons depend strongly on the initial statistical properties of the photons and phonons as displayed in figures 1 and 2.

The validity of the short-time approximation as well as of the above results is defined by the condition $|\kappa_{s,a}^{(1,2)} t\xi_l| < 1$ (cf., e.g., [12]). Even if some of these effects are rather small, others are measurable, for instance

$$\langle \Delta W_l \Delta W_s \rangle_R / \langle W_l \rangle_R \langle W_s \rangle_R \approx -2 \times 10^{-4}$$

and

$$\langle \Delta W_l \Delta W_a \rangle_R / \langle W_l \rangle_R \langle W_a \rangle_R \approx -2 \times 10^{-4}$$

assuming $\kappa_s^{(1)} \approx \kappa_a^{(1)} \approx 10^2\ s^{-1}$, $t \approx 10^{-10}$ s (for a sample of linear dimensions of the order of centimetres) so that $|\xi_l|^2 \approx 10^{16}$ (corresponding to a photon flux of 10^{26} photons/s, i.e. to a power of 10 MW) and $n_p \approx 10^{12}$ phonons/cm^3. Further, it should be noted that $\langle \Delta W_l \Delta W_{s,a} \rangle_R / [\langle (\Delta W_l)^2 \rangle_R \langle (\Delta W_{s,a})^2 \rangle_R]^{1/2} \approx -1$.

Our considerations did not include explicitly the polarization states of the photons (the polarization density matrix discussed by Atkins and Wilson [13]) which affect the dynamics of the photon correlation and anticorrelation effects [14]. Obviously, this would require the discussion of correlation tensors (in place of correlation functions) involving the photon polarization states [15]. These matters will be the subject of a separate paper.

On analyse les interactions non-linéaires entre photons incidents et diffusés (rayonnement Stokes et anti-Stokes) en Raman et hyper-Raman, processus considérés comme des événements élémentaires. On montre que des fluctuations de phonons produisent des effets d'anticorrélation entre les photons incidents et diffusés. L'antibunching des photons laser incident en hyper-Raman spontané se produit de la même manière que leur antibunching en génération de second harmonique.

Es werden nichtlineare Wechselwirkungen zwischen einfallenden und gestreuten Photonen (Stokes und Anti-Stokes Strahlung) in elementaren Raman und Hyper-Raman Prozessen analysiert. Fluktuationen von Phononen verursachen Antikorrelationen zwischen einfallenden und gestreuten Photonen. Antibunching einfallender Laserphotonen in spontanen Hyper-Raman Prozessen erfolgt ähnlich dem Antibunching bei der Erzeugung zweiter Harmonischer.

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