QUANTUM STATISTICAL PROPERTIES OF PHOTON AND PHONON FIELDS IN NON-DEGENERATE HYPER-RAMAN SCATTERING

By V. Peřinová, J. Peřina

Laboratory of Optics, Palacký University, Olomouc*

and P. Szlachetka, S. Kielich

Institute of Physics, A. Mickiewicz University, Poznań**

(Received November 7, 1978)

The complete normal quantum characteristic function is calculated in the short-time approximation from which fluctuations in separate modes and correlations among them were deduced. The results obtained are particularly discussed from the view-point of anticorrelation (antibunching) assuming that (*i*) the phonon mode is initially chaotic whereas the photon modes are initially coherent, and (*ii*) the laser mode is initially coherent whereas all other modes are initially chaotic.

1. *Introduction*

This paper is a continuation of paper [1] (cf. the references therein), devoted to the quantum statistical properties of radiation involved in Raman or Brillouin scattering. Here we derive the complete quantum characteristic function using the short-time approximation. From it we deduce fluctuations in separate modes and their correlation and discuss them from the view-point of anticorrelation (antibunching) of photons. We consider the following cases: (*i*) the phonon mode is initially chaotic whereas all other modes are initially coherent, (*ii*) the laser modes are initially coherent whereas all other (phonon, Stokes and anti-Stokes) modes are initially chaotic. Typical for this higher-order non-linear optical process, as compared to Raman scattering, is that in the quantum characteristic function the third and fourth-order terms occur in its parameters.

* Address: Laboratory of Optics, Palacký University, Leninova 26, 771 46 Olomouc, Czechoslovakia.

** Address: Instytut Fizyki UAM, Grunwaldzka 6, 60-780 Poznań, Poland.

2. *General description*

Non-degenerate hyper-Raman scattering is described by the renormalized "effective" Hamiltonian:

$$H = \sum_{j=1,2,S,A,V} \hbar\omega_j a_j^\dagger a_j + \hbar(\kappa_S a_1 a_2 a_S^\dagger a_V^\dagger + \kappa_A a_1 a_2 a_A^\dagger a_V + \text{h.c.}), \tag{2.1}$$

where a_1, a_2, a_S, a_A, a_V, and $a_1^\dagger, a_2^\dagger, a_S^\dagger, a_A^\dagger, a_V^\dagger$ are, respectively, the annihilation and creation operators for the first and second laser, Stokes, anti-Stokes and phonon modes with frequencies ω_1, ω_2, ω_S, ω_A, ω_V, κ_S and κ_A are coupling constants for the Stokes and anti-Stokes modes and $\omega_{S,A} = \omega_1 + \omega_2 \mp \omega_V$. The corresponding Heisenberg equations are of the form:

$$\begin{aligned}
\dot{a}_1 &= -i(\omega_1 a_1 + \kappa_S^* a_2^\dagger a_S a_V + \kappa_A^* a_2^\dagger a_A a_V^\dagger),\\
\dot{a}_2 &= -i(\omega_2 a_2 + \kappa_S^* a_1^\dagger a_S a_V + \kappa_A^* a_1^\dagger a_A a_V^\dagger),\\
\dot{a}_S &= -i(\omega_S a_S + \kappa_S a_1 a_2 a_V^\dagger),\\
\dot{a}_A &= -i(\omega_A a_A + \kappa_A a_1 a_2 a_V),\\
\dot{a}_V &= -i(\omega_V a_V + \kappa_S a_1 a_2 a_S^\dagger + \kappa_A^* a_1^\dagger a_2^\dagger a_A);
\end{aligned} \tag{2.2}$$

they can be solved approximately up to t^2 in the form:

$$\begin{aligned}
a_1(t) = \exp(-i\omega_1 t)\Big\{&a_1 - it(\kappa_S^* a_2^\dagger a_S a_V + \kappa_A^* a_2^\dagger a_A a_V^\dagger) + \frac{t^2}{2}[|\kappa_S|^2 a_1(a_S^\dagger a_S a_V^\dagger a_V\\
&- a_2^\dagger a_2(a_S^\dagger a_S + a_V^\dagger a_V + 1)) + |\kappa_A|^2 a_1(a_A^\dagger a_A(a_V^\dagger a_V + 1) + a_2^\dagger a_2(a_A^\dagger a_A - a_V^\dagger a_V))\\
&+ a_1(\kappa_S^* \kappa_A a_S a_A^\dagger a_V^2 + \text{h.c.})]\Big\},
\end{aligned}$$

$$a_2(t) = a_1(t) \text{ with } 1 \leftrightarrow 2,$$

$$\begin{aligned}
a_S(t) = \exp(-i\omega_S t)\Big\{&a_S - it\kappa_S a_1 a_2 a_V^\dagger + \frac{t^2}{2}[|\kappa_S|^2 a_S(a_1^\dagger a_1 a_2^\dagger a_2 - (a_1^\dagger a_1 + a_2^\dagger a_2 + 1)a_V^\dagger a_V)\\
&- \kappa_S \kappa_A^*(a_1^\dagger a_1 + a_2^\dagger a_2 + 1)a_A a_V^{\dagger 2} + \kappa_S \kappa_A a_1^2 a_2^2 a_A^\dagger]\Big\},
\end{aligned}$$

$$\begin{aligned}
a_A(t) = \exp(-i\omega_A t)\Big\{&a_A - it\kappa_A a_1 a_2 a_V - \frac{t^2}{2}[|\kappa_A|^2 a_A((a_1^\dagger a_1 + 1)(a_2^\dagger a_2 + 1)\\
&+ (a_1^\dagger a_1 + a_2^\dagger a_2 + 1)a_V^\dagger a_V) + \kappa_S^* \kappa_A(a_1^\dagger a_1 + a_2^\dagger a_2 + 1)a_S a_V^2 + \kappa_S \kappa_A a_1^2 a_2^2 a_S^\dagger]\Big\},
\end{aligned}$$

$$\begin{aligned}
a_V(t) = \exp(-i\omega_V t)\Big\{&a_V - it(\kappa_S a_1 a_2 a_S^\dagger + \kappa_A^* a_1^\dagger a_2^\dagger a_A) + \frac{t^2}{2}[|\kappa_S|^2 a_V(a_1^\dagger a_1 a_2^\dagger a_2\\
&- (a_1^\dagger a_1 + a_2^\dagger a_2 + 1)a_S^\dagger a_S) + |\kappa_A|^2 a_V(-a_1^\dagger a_1 a_2^\dagger a_2 + (a_1^\dagger a_1 + a_2^\dagger a_2 + 1)a_A^\dagger a_A)]\Big\},
\end{aligned} \tag{2.3}$$

where $a_j = a_j\ (0)$.

The time evolution of the statistical properties of the system is described by the normal quantum characteristic function (cf. e. g. [2]):

$$C_N(\beta_1, \beta_2, \beta_S, \beta_A, \beta_V, t) = \mathrm{Tr}\,\{\varrho(0) \prod_{j=1,2,S,A,V} \exp(\beta_j a_j^\dagger(t)) \exp(-\beta_j^* a_j(t))\}, \tag{2.4}$$

where $a_j(t)$ are solutions (2.3), $\varrho(0)$ is the density matrix specifying the statistical properties of the initial fields. The normal ordering of (2.4) in the initial operators can be performed using the Baker–Hausdorff identity for which the assumptions are fulfilled up to t^2. Moreover, use is made of the fact that the first terms a_j in (2.3) commute with the other terms. With further application of the Glauber–Sudarshan representation of $\varrho(0)$, we arrive at the complete normal quantum characteristic function valid up to t^2:

$$\begin{aligned} C_N(\{\beta_j\}, t) = \langle \exp\{ & \sum_{j=1,2,S,A,V} (\beta_j \xi_j^*(t) - \beta_j^* \xi_j(t)) - \sum_{j=1,2,S,V} |\beta_j|^2 B_j \\ & + \tfrac{1}{2} \sum_{j=1,2,V} (\beta_j^{*2} C_j + \text{c.c.}) + \sum_{\substack{j<k \\ j,k=1,2,S,A,V}} (\beta_j^* \beta_k^* D_{jk} + \beta_j \beta_k^* \bar{D}_{jk} + \text{c.c.}) \\ & + (\beta_1^* \beta_2^{*2} D_{122} + \beta_1^{*2} \beta_2^* D_{112} + \beta_1^* \beta_S^* \beta_V^* D_{1SV} + \beta_2^* \beta_S^* \beta_V^* D_{2SV} + \beta_1^* \beta_2^* \beta_A^* D_{12A} + \beta_1^* \beta_A^* \beta_V^* D_{1AV} \\ & + \beta_2^* \beta_A^* \beta_V^* D_{2AV} + \beta_1^* \beta_V^{*2} D_{1VV} + \beta_2^* \beta_V^{*2} D_{2VV} + \beta_1^* \beta_2^* \beta_V^* D_{12V} + \beta_1^* \beta_2^* \beta_S^* D_{12S} - |\beta_1|^2 \beta_2 \bar{D}_{121} \\ & - \beta_1 \beta_2 \beta_S^* \bar{D}_{12S} - \beta_1 |\beta_V|^2 \bar{D}_{1VV} - |\beta_2|^2 \beta_V \bar{D}_{2V2} - \beta_2 |\beta_V|^2 \bar{D}_{2VV} - \beta_1 |\beta_2|^2 \bar{D}_{122} - |\beta_1|^2 \beta_V \bar{D}_{1V1} \\ & - \text{c.c.}) + (\beta_1^{*2} \beta_2^{*2} D_{1122} + \beta_1^* \beta_S^* \beta_V^{*2} D_{1SVV} + \beta_2^* \beta_S^* \beta_V^{*2} D_{2SVV} + \text{c.c.}) + |\beta_1|^2 |\beta_2|^2 \bar{D}_{1212} \\ & + |\beta_1|^2 |\beta_V|^2 \bar{D}_{1V1V} + |\beta_2|^2 |\beta_V|^2 \bar{D}_{2V2V}\} \rangle, \end{aligned} \tag{2.5}$$

where the set (1, 2, S, A, V) is assumed to be ordered, $\xi_j(t)$ are time-dependent complex amplitudes which are obtained from (2.3) by substituting the initial complex amplitudes ξ_j for the initial operators a_j. The brackets in (2.5) represent the average over ξ_j and

$$B_1 = t^2[|\kappa_S|^2 |\xi_S|^2 |\xi_V|^2 + |\kappa_A|^2 (|\xi_2|^2 + |\xi_V|^2 + 1)\, |\xi_A|^2 + (\kappa_S \kappa_A^* \xi_S^* \xi_A \xi_V^{*2} + \text{c.c.})],$$

$$B_2 = B_1(2 \to 1), \qquad B_S = |\kappa_S|^2 t^2 |\xi_1|^2 |\xi_2|^2,$$

$$B_V = t^2[|\kappa_S|^2 |\xi_1|^2 |\xi_2|^2 + |\kappa_A|^2 (|\xi_1|^2 + |\xi_2|^2 + 1)\, |\xi_A|^2],$$

$$C_1 = -\exp(-i2\omega_1 t) \kappa_S^* \kappa_A^* t^2 \xi_2^{*2} \xi_S \xi_A, \qquad C_2 = C_1(1 \leftrightarrow 2),$$

$$C_V = -\exp(-i2\omega_V t) \kappa_S \kappa_A^* t^2 (|\xi_1|^2 + |\xi_2|^2 + 1) \xi_S^* \xi_A,$$

$$\begin{aligned} D_{12} = -\exp[-i(\omega_1 + \omega_2)t] \Big\{ & it(\kappa_S^* \xi_S \xi_V + \kappa_A^* \xi_A \xi_V^*) + \frac{t^2}{2} [|\kappa_S|^2 \xi_1 \xi_2 (|\xi_S|^2 + |\xi_V|^2 + 1) \\ & + |\kappa_A|^2 \xi_1 \xi_2 (|\xi_V|^2 - |\xi_A|^2) + 2\kappa_S^* \kappa_A^* \xi_1^* \xi_2^* \xi_S \xi_A] \Big\}, \end{aligned}$$

$$D_{1S} = -\exp[-i(\omega_1 + \omega_S)t] \frac{t^2}{2} [|\kappa_S|^2 \xi_1 \xi_S (|\xi_2|^2 + |\xi_V|^2) + \kappa_S \kappa_A^* \xi_1 \xi_A \xi_V^{*2}],$$

$$D_{1A} = D_{1S}(S \leftrightarrow A),$$

$$D_{1\mathrm{V}} = -\exp[-i(\omega_1+\omega_\mathrm{V})t]\left\{it\kappa_\mathrm{A}^*\xi_2^*\xi_\mathrm{A}+\frac{t^2}{2}\,\xi_1\xi_\mathrm{V}[|\kappa_\mathrm{S}|^2(|\xi_2|^2+|\xi_\mathrm{S}|^2)\right.$$

$$\left.+|\kappa_\mathrm{A}|^2(|\xi_2|^2-|\xi_\mathrm{A}|^2)]\right\},$$

$$D_{2\mathrm{S}} = D_{1\mathrm{S}}(1\leftrightarrow 2),\quad D_{2\mathrm{A}} = D_{1\mathrm{A}}(1\leftrightarrow 2),\quad D_{2\mathrm{V}} = D_{1\mathrm{V}}(1\leftrightarrow 2),$$

$$D_{\mathrm{SA}} = -\exp[-i(\omega_\mathrm{S}+\omega_\mathrm{A})t]\,\frac{t^2}{2}\,\kappa_\mathrm{S}\kappa_\mathrm{A}\xi_1^2\xi_2^2,$$

$$D_{\mathrm{SV}} = -\exp[-i(\omega_\mathrm{S}+\omega_\mathrm{V})t]\,[it\kappa_\mathrm{S}\xi_1\xi_2+t^2(\tfrac{1}{2}\,|\kappa_\mathrm{S}|^2\xi_\mathrm{S}\xi_\mathrm{V}+\kappa_\mathrm{S}\kappa_\mathrm{A}^*\xi_\mathrm{A}\xi_\mathrm{V}^*)\,(|\xi_1|^2+|\xi_2|^2+1)],$$

$$D_{\mathrm{AV}} = -\exp[-i(\omega_\mathrm{A}+\omega_\mathrm{V})t]\,\frac{t^2}{2}\,|\kappa_\mathrm{A}|^2\xi_\mathrm{A}\xi_\mathrm{V}(|\xi_1|^2+|\xi_2|^2+1),$$

$$\bar{D}_{12} = -\exp[i(\omega_1-\omega_2)t]t^2|\kappa_\mathrm{A}|^2\xi_1^*\xi_2|\xi_\mathrm{A}|^2,$$

$$\bar{D}_{1\mathrm{S}} = -\exp[i(\omega_1-\omega_\mathrm{S})t]t^2\kappa_\mathrm{S}\kappa_\mathrm{A}\xi_1\xi_2^2\xi_\mathrm{A}^*,$$

$$\bar{D}_{1\mathrm{V}} = -\exp[i(\omega_1-\omega_\mathrm{V})t]t^2[|\kappa_\mathrm{A}|^2\xi_1^*|\xi_\mathrm{A}|^2\xi_\mathrm{V}+\kappa_\mathrm{S}\kappa_\mathrm{A}^*\xi_1^*\xi_\mathrm{S}^*\xi_\mathrm{A}\xi_\mathrm{V}^*],$$

$$\bar{D}_{2\mathrm{S}} = \bar{D}_{1\mathrm{S}}(1\leftrightarrow 2),\quad \bar{D}_{2\mathrm{V}} = \bar{D}_{1\mathrm{V}}(1\leftrightarrow 2),$$

all other $\bar{D}_{jk} = 0$,

$$D_{122} = \exp[-i(\omega_1+2\omega_2)t]t^2\kappa_\mathrm{S}^*\kappa_\mathrm{A}^*\xi_1^*\xi_\mathrm{S}\xi_\mathrm{A},\quad D_{112} = D_{122}(1\leftrightarrow 2),$$

$$D_{1\mathrm{SV}} = \exp[-i(\omega_1+\omega_\mathrm{S}+\omega_\mathrm{V})t]t^2[\tfrac{1}{2}\,|\kappa_\mathrm{S}|^2\xi_1\xi_\mathrm{S}\xi_\mathrm{V}+\kappa_\mathrm{S}\kappa_\mathrm{A}^*\xi_1\xi_\mathrm{A}\xi_\mathrm{V}^*],$$

$$D_{2\mathrm{SV}} = D_{1\mathrm{SV}}(1\leftrightarrow 2),\quad D_{12\mathrm{A}} = \exp[-i(\omega_1+\omega_2+\omega_\mathrm{A})t]\,\frac{t^2}{2}\,|\kappa_\mathrm{A}|^2\xi_1\xi_2\xi_\mathrm{A},$$

$$D_{1\mathrm{AV}} = D_{12\mathrm{A}}(2\to \mathrm{V}),\quad D_{2\mathrm{AV}} = D_{1\mathrm{AV}}(1\to 2),$$

$$D_{1\mathrm{VV}} = \exp[-i(\omega_1+2\omega_\mathrm{V})t]t^2\kappa_\mathrm{S}\kappa_\mathrm{A}^*\xi_1\xi_\mathrm{S}^*\xi_\mathrm{A},\quad D_{2\mathrm{VV}} = D_{1\mathrm{VV}}(1\to 2),$$

$$D_{12\mathrm{V}} = \exp[-i(\omega_1+\omega_2+\omega_\mathrm{V})t]\left[it\kappa_\mathrm{A}^*\xi_\mathrm{A}+\frac{t^2}{2}\,(|\kappa_\mathrm{S}|^2+|\kappa_\mathrm{A}|^2)\xi_1\xi_2\xi_\mathrm{V}\right],$$

$$D_{12\mathrm{S}} = D_{12\mathrm{A}}(\mathrm{A}\to \mathrm{S}),\quad \bar{D}_{121} = \exp(i\omega_2 t)t^2|\kappa_\mathrm{A}|^2|\xi_\mathrm{A}|^2\xi_2^*,$$

$$\bar{D}_{12\mathrm{S}} = \exp[i(\omega_1+\omega_2-\omega_\mathrm{S})t]t^2\kappa_\mathrm{S}\kappa_\mathrm{A}\xi_1\xi_2\xi_\mathrm{A}^*,$$

$$\bar{D}_{122} = \bar{D}_{1\mathrm{VV}} = \bar{D}_{121}(2\to 1),$$

$$\bar{D}_{1\mathrm{V}1} = \bar{D}_{2\mathrm{V}2} = \exp(i\omega_\mathrm{V}t)t^2[|\kappa_\mathrm{A}|^2|\xi_\mathrm{A}|^2\xi_\mathrm{V}^*+\kappa_\mathrm{S}^*\kappa_\mathrm{A}\xi_\mathrm{S}\xi_\mathrm{V}^*\xi_\mathrm{A}],$$

$$\bar{D}_{2\mathrm{VV}} = \bar{D}_{1\mathrm{VV}}(1\to 2),\quad D_{1122} = -\exp[-2i(\omega_1+\omega_2)t]\,\frac{t^2}{2}\,\kappa_\mathrm{S}^*\kappa_\mathrm{A}^*\xi_\mathrm{S}\xi_\mathrm{A},$$

$$D_{1\mathrm{SVV}} = -\exp[-i(\omega_1+\omega_\mathrm{S}+2\omega_\mathrm{V})t]t^2\kappa_\mathrm{S}\kappa_\mathrm{A}^*\xi_1\xi_\mathrm{A},$$

$$D_{2\mathrm{SVV}} = D_{1\mathrm{SVV}}(1\to 2),\quad \bar{D}_{1212} = \bar{D}_{1\mathrm{V}1\mathrm{V}} = \bar{D}_{2\mathrm{V}2\mathrm{V}} = |\kappa_\mathrm{A}|^2t^2|\xi_\mathrm{A}|^2. \tag{2.6}$$

It should be noted that, compared to the Raman scattering case [1], in (2.5) the third and fourth-order terms in β_j occur even if the field is initially coherent. Fluctuations of the intensity, W_j, in separate modes are:

$$\langle(\Delta W_j)^2\rangle = \frac{\partial^4 C_N}{\partial\beta_j^2\partial(-\beta_j^*)^2}\bigg|_{\{\beta_j\}=0} - \left(\frac{\partial^2 C_N}{\partial\beta_j\partial(-\beta_j^*)}\right)^2\bigg|_{\{\beta_j\}=0}$$
$$= \langle B_j^2+|C_j|^2+2B_j|\xi_j(t)|^2+C_j^*\xi_j^2(t)+C_j\xi_j^{*2}(t)\rangle$$
$$+\langle(B_j+|\xi_j(t)|^2)^2\rangle-\langle B_j+|\xi_j(t)|^2\rangle^2 \tag{2.7a}$$

and correlations of fluctuations in various modes are given by:

$$\langle\Delta W_j\Delta W_k\rangle = \frac{\partial^4 C_N}{\partial\beta_j\partial(-\beta_j^*)\partial\beta_k\partial(-\beta_k^*)}\bigg|_{\{\beta_j\}=0} - \frac{\partial^2 C_N}{\partial\beta_j\partial(-\beta_j^*)}\frac{\partial^2 C_N}{\partial\beta_k\partial(-\beta_k^*)}\bigg|_{\{\beta_j\}=0}$$
$$= \langle|D_{jk}|^2+|\bar{D}_{jk}|^2+(D_{jk}\xi_j^*(t)\xi_k^*(t)-\bar{D}_{jk}\xi_j(t)\xi_k^*(t)+\bar{D}_{jkk}\xi_j(t)+\bar{D}_{jkj}\xi_k(t)$$
$$+\text{c.c.})+\bar{D}_{jkjk}\rangle+\langle(B_j+|\xi_j(t)|^2)(B_k+|\xi_k(t)|^2)\rangle-\langle B_j+|\xi_j(t)|^2\rangle\langle B_k+|\xi_k(t)|^2\rangle,$$
$$j<k. \tag{2.7b}$$

The brackets in (2.7a, b) are omitted and the last two terms cancel out if all modes are initially coherent.

3. *Special cases of initial fields*

3.1. The phonon mode chaotic and all photon modes coherent

In this case we obtain from (2.7a) and (2.6) for fluctuations of the intensity in separate modes

$$\langle(\Delta W_1)^2\rangle = t^2\{2|\xi_1|^2(1+|\xi_2|^2)[|\kappa_S|^2|\xi_S|^2\langle n_V\rangle+|\kappa_A|^2|\xi_A|^2(\langle n_V\rangle+1)]-(2\langle n_V\rangle+1)R\},$$
$$\langle(\Delta W_2)^2\rangle = \langle(\Delta W_1)^2\rangle \quad \text{with} \quad 1\leftrightarrow 2,$$

$$\langle(\Delta W_S)^2\rangle = 2|\kappa_S|^2t^2|\xi_1|^2|\xi_2|^2|\xi_S|^2(\langle n_V\rangle+1),$$
$$\langle(\Delta W_A)^2\rangle = 2|\kappa_A|^2t^2|\xi_1|^2|\xi_2|^2|\xi_A|^2\langle n_V\rangle,$$
$$\langle(\Delta W_V)^2\rangle = \langle n_V\rangle^2+2t^2\{|\kappa_S|^2[-(|\xi_1|^2+|\xi_2|^2+1)|\xi_S|^2\langle n_V\rangle^2+|\xi_1|^2|\xi_2|^2\langle n_V\rangle(|\xi_S|^2$$
$$+\langle n_V\rangle+1)]+|\kappa_A|^2[(|\xi_1|^2+|\xi_2|^2+1)|\xi_A|^2\langle n_V\rangle(\langle n_V\rangle+1)$$
$$+|\xi_1|^2|\xi_2|^2\langle n_V\rangle(|\xi_A|^2-\langle n_V\rangle)+R\langle n_V\rangle\}, \tag{3.1}$$

which closely correspond to the Raman scattering case [1]. Here $\langle n_V\rangle$ represents the mean number of phonons and $R = \kappa_S\kappa_A\xi_1{}^2\xi_2{}^2\xi_S^*\xi_A^*+\text{c. c.}$ The antibunching cannot be expected to occur in this case.

Using (2.7b) we obtain for the correlations of fluctuations:

$$\langle \Delta W_1 \Delta W_2\rangle = t^2\{|\kappa_S|^2[(1+2(|\xi_1|^2+|\xi_2|^2))\,|\xi_S|^2\langle n_V\rangle$$
$$-|\xi_1|^2|\xi_2|^2(|\xi_S|^2(1-2\langle n_V\rangle)+\langle n_V\rangle+1]+|\kappa_A|^2[(1+2(|\xi_1|^2+|\xi_2|^2))\,|\xi_A|^2(\langle n_V\rangle+1)$$
$$+|\xi_1|^2|\xi_2|^2((3+2\langle n_V\rangle)\,|\xi_A|^2-\langle n_V\rangle)]-(2\langle n_V\rangle+1)R\},$$
$$\langle \Delta W_1 \Delta W_S\rangle = t^2\{-|\kappa_S|^2|\xi_1|^2|\xi_S|^2[(2\langle n_V\rangle+1)\,|\xi_2|^2+\langle n_V\rangle]+R(\langle n_V\rangle+1)\},$$
$$\langle \Delta W_1 \Delta W_A\rangle = t^2\{-|\kappa_A|^2|\xi_1|^2|\xi_A|^2[(2\langle n_V\rangle+1)\,|\xi_2|^2+\langle n_V\rangle+1]+R\langle n_V\rangle\},$$
$$\langle \Delta W_1 \Delta W_V\rangle = t^2\{|\kappa_S|^2[(1+|\xi_1|^2+|\xi_2|^2)\,|\xi_S|^2\langle n_V\rangle^2-|\xi_1|^2|\xi_2|^2\langle n_V\rangle\,(2|\xi_S|^2+\langle n_V\rangle+1)$$
$$-|\xi_1|^2|\xi_S|^2\langle n_V\rangle]+|\kappa_A|^2[(1+|\xi_1|^2+|\xi_2|^2)\,((\langle n_V\rangle+1)^2|\xi_A|^2$$
$$+|\xi_1|^2|\xi_2|^2(\langle n_V\rangle+1)\,(2|\xi_A|^2-\langle n_V\rangle)+|\xi_1|^2|\xi_A|^2(\langle n_V\rangle+1)]+R\},$$
$$\langle \Delta W_2 \Delta W_S\rangle = \langle \Delta W_1 \Delta W_S\rangle \text{ with } 1\leftrightarrow 2, \quad \langle \Delta W_2 \Delta W_A\rangle = \langle \Delta W_1 \Delta W_A\rangle \text{ with } 1\leftrightarrow 2,$$
$$\langle \Delta W_2 \Delta W_V\rangle = \langle \Delta W_1 \Delta W_V\rangle \text{ with } 1\leftrightarrow 2, \quad \langle \Delta W_S \Delta W_A\rangle = -\frac{t^2}{2}(2\langle n_V\rangle+1)R,$$
$$\langle \Delta W_S \Delta W_V\rangle = t^2\{|\kappa_S|^2[-(|\xi_1|^2+|\xi_2|^2+1)\,|\xi_S|^2\langle n_V\rangle\,(\langle n_V\rangle+1)+|\xi_1|^2|\xi_2|^2(\langle n_V\rangle+1)^2$$
$$+2|\xi_1|^2|\xi_2|^2|\xi_S|^2(\langle n_V\rangle+1)]+(\langle n_V\rangle+1)R\},$$
$$\langle \Delta W_A \Delta W_V\rangle = -t^2\{|\kappa_A|^2[(|\xi_1|^2+|\xi_2|^2+1)\,|\xi_A|^2\langle n_V\rangle\,(\langle n_V\rangle+1)$$
$$+|\xi_1|^2|\xi_2|^2\langle n_V\rangle\,(2|\xi_A|^2-\langle n_V\rangle)]+R\langle n_V\rangle\}. \qquad (3.2)$$

We see that there occurs anticorrelation between laser modes 1 and 2 in the course of spontaneous scattering ($\xi_S = \xi_A = 0$) amounting to $\langle \Delta W_1 \Delta W_2\rangle = -|\kappa_S|^2 t^2|\xi_1|^2|\xi_2|^2 (\langle n_V\rangle+1)-|\kappa_A|^2t^2|\xi_1|^2|\xi_2|^2\langle n_V\rangle$. Also $\langle(\Delta W_1+\Delta W_1)^2\rangle = 2\langle \Delta W_1\Delta W_2\rangle$ is negative here. This behaviour is analogous to the case of double-beam two-photon absorption [3] and it cannot have any analogy in Raman scattering. The other results are similar to those obtained for Raman scattering [1]. Between modes 1 (2) and S, 1 (2) and A, and S and A we have enhanced anticorrelation compared to the case when all modes are initially coherent provided that $\Delta\varphi = 2\,\varphi_1+2\varphi_2+\psi_S+\psi_A-\varphi_S-\varphi_A = \pi$ in the first and second cases and $\Delta\varphi = 0$ in the third case (anticorrelation is maximal). Here φ_j are phases of $\xi_{S,A}$ and $\psi_{S,A}$ are phases of $\kappa_{S,A}$. The anticorrelation also occurs in the first (second) case if ξ_A (ξ_S) $= 0$ or if $\Delta\varphi = \pi/2$ when $R = 0$. Between modes 1 (2) and V the anticorrelation can be observed in spontaneous scattering ($\xi_S = \xi_A = 0$) or if

$$|\xi_1|^2,\,|\xi_2|^2 \gg \langle n_V\rangle \gg |\xi_S|^2,\,|\xi_A|^2, \qquad (3.3)$$

as occurs in practice. Under this condition, $\langle \Delta W_{S,A}, \Delta W_V\rangle \geq 0$ always holds.

3.2. The laser mode coherent and all other modes chaotic

In this case we additionally average (3.1) and (3.2) over chaotic initial complex amplitudes and put $\langle|\xi_S|^2\rangle = \langle n_S\rangle$ and $\langle|\xi_A|^2\rangle = \langle n_A\rangle$ and $R = 0$ and we add the following terms to $\langle(\Delta W_{S,A})^2\rangle$, $\langle\Delta W_{1,2}\Delta W_{S,A}\rangle$, $\langle\Delta W_{S,A}\Delta W_V\rangle$:

$$\langle(\Delta W_S)^2\rangle : \langle n_S\rangle^2+2|\kappa_S|^2t^2[|\xi_1|^2|\xi_2|^2\langle n_S\rangle^2-(|\xi_1|^2+|\xi_2|^2+1)\langle n_S\rangle^2\langle n_V\rangle],$$

$$\langle(\Delta W_A)^2\rangle : \langle n_A\rangle^2+2|\kappa_A|^2t^2[|\xi_1|^2|\xi_2|^2\langle n_A\rangle^2-(|\xi_1|^2+|\xi_2|^2+1)\langle n_A\rangle^2(\langle n_V\rangle+1)],$$

$$\langle(\Delta W_{1,2}\Delta W_S\rangle : |\kappa_S|^2t^2[|\xi_{1,2}|^2(\langle n_V\rangle-|\xi_{2,1}|^2)+\langle n_V\rangle(|\xi_{2,1}|^2+1)]\langle n_S\rangle^2,$$

$$\langle\Delta W_{1,2}\Delta W_A\rangle : |\kappa_A|^2t^2[(|\xi_{2,1}|^2+\langle n_V\rangle+1)(|\xi_{1,2}|^2+1)+|\xi_{2,1}|^2\langle n_V\rangle]\langle n_A\rangle^2,$$

$$\langle\Delta W_S\Delta W_V\rangle : |\kappa_S|^2t^2[|\xi_1|^2|\xi_2|^2-(|\xi_1|^2+|\xi_2|^2+1)\langle n_V\rangle]\langle n_S\rangle^2,$$

$$\langle\Delta W_A\Delta W_V\rangle : |\kappa_A|^2t^2[|\xi_1|^2|\xi_2|^2+(|\xi_1|^2+|\xi_2|^2+1)(\langle n_V\rangle+1)]\langle n_A\rangle^2. \quad (3.4)$$

In the other cases the corrections are zero.

While no antibunching can occur in separate modes, anticorrelation is possible among modes in analogy to Raman scattering. The above is valid between laser modes 1, 2 and between a laser mode and the phonon mode, 1 (2), V; between modes 1 (2) and S, and 1 (2) and A there is anticorrelation provided that (3.3) holds ($[\xi_{S,A}|^2 = \langle n_{S,A}\rangle$). Modes S and A are not correlated at all and the correlations between modes S and V, and A and V are always non-negative in this case.

It should be noted that when detecting pairs of modes, i. e., when measuring fluctuations $\langle(\Delta W_j+\Delta W_k)^2$, $j \neq k$, $j, k = 1, 2, S, A, V$, these quantities behave similarly to those in Raman scattering [1] and antibunching can be observed under special conditions if all modes are initially coherent whereas they are non-negative in the cases considered here except $\langle(\Delta W_1+\Delta W_2)^2\rangle$, leading to antibunching $-2|\xi_1|^2|\xi_2|^2t^2(|\kappa_S|^2(\langle n_V\rangle+1) + |\kappa_A|^2\langle n_V\rangle)$ in spontaneous scattering. Moreover, if all modes are initially coherent, phase-dependent antibunching of the order $\kappa_S t$, $\kappa_A t$ is possible if $\varphi_S+\varphi_V-\varphi_1-\varphi_2-\psi_S = -\pi/2$ and $\varphi_A-\varphi_V-\varphi_1-\varphi_2-\psi_A = -\pi/2$.

4. *Conclusions*

We have found that in the cases under discussion, i. e. when the phonon mode is chaotic or phonon, Stokes and anti-Stokes modes are chaotic while the other modes are coherent at $t = 0$, no antibunching in separate modes can be observed. The anticorrelation is typical between the first (second) laser mode and Stokes or anti-Stokes modes, in analogy to Raman scattering, provided that the phase-dependent terms are negative (special initial phase condition) or absent (special initial phase condition or partly spontaneous scattering). Anticorrelation also occurs if phonon, Stokes and anti-Stokes modes are initially chaotic and $|\xi_{1,2}|^2 \gg \langle n_V\rangle \gg \langle n_{S,A}\rangle$ as occurs in practice. Between any laser and phonon modes anticorrelation is possible in the course of spontaneous scattering or if the above condition for mean numbers holds. The Stokes and anti-Stokes modes can be phase--dependently anticorrelated provided that the phonon mode is initially chaotic and the other modes are initially coherent but they are uncorrelated if the phonon and Stokes (or anti-Stokes) modes are initially chaotic. Similarly to Raman scattering, there is non-negative correlation between Stokes or anti-Stokes and phonon modes. A special feature of this higher-order non-linear process is that there is the anticorrelation between laser modes in spontaneous scattering.

In Raman scattering antibunching can be observed measuring fluctuations $\langle(\Delta W_j+\Delta W_k)^2\rangle, j \neq k$ only if all modes are initially coherent. In the cases under discussion here these quantities are non-negative except for $\langle(\Delta W_1+\Delta W_2)^2\rangle$ in spontaneous scattering.

Thus, also the non-degenerate hyper-Raman scattering process provides a number of possibilities to observe anticorrelation. Some other possibilities for the generation of radiation exhibiting anticorrelation in the process of non-degenerate hyper-Raman scattering have been discussed elsewhere [4]. In Raman scattering the quasi-distribution function related to antinormal ordering of field operators has been obtained as a shifted multidimensional Gaussian distribution and the photocounting distribution and its factorial moments can be expressed in terms of Laguerre polynomials. This is not feasible in the hyper-Raman scattering process.

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