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

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In this paper the complete normal quantum characteristic function is calculated in the short-time approximation for degenerate hyper-Raman scattering from which fluctuations in separate modes and correlations among these are deduced. The results are discussed from the view-point of anticorrelation or antibunching assuming that (*i*) the phonon mode is initially chaotic whereas all photon modes are initially coherent, and (*ii*) the laser mode is initially coherent and all other modes are initially chaotic. A comparison with non-degenerate hyper--Raman scattering and Raman scattering is made.

1. *Introduction*

This paper is a continuation of previous papers [1, 2] concerning the quantum statistical properties of radiation scattered in the process of non-degenerate hyper-Raman scattering. In a similar way we calculate the complete normal quantum characteristic function using the short-time approximation up to t^2 (t being the interaction time), from which we are able to obtain easily fluctuations in separate modes as well as the correlations among the modes. Special attention is given again to states of anticorrelation. As before [1, 2], we consider two cases (*i*) the phonon mode is initially chaotic whereas all radiation modes are initially coherent, and (*ii*) the laser mode is initially coherent and all other modes (Stokes, anti-Stokes and phonon) are initially chaotic. The method of investigation is identical to that used for non-degenerate hyper-Raman scattering [1, 2] and Raman scattering [3].

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2. General description

The degenerate hyper-Raman scattering is described by the re-normalized "effective" Hamiltonian:

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

where a_L, a_S, a_A, a_V and $a_L^\dagger$, $a_S^\dagger$, $a_A^\dagger$, $a_V^\dagger$ are the annihilation and creation operators of the laser, Stokes, anti-Stokes and vibration (phonon) modes with the corresponding frequencies ω_L, ω_S, ω_A, ω_V, respectively, fulfilling the frequency resonance conditions $\omega_{,SA} = 2\omega_L \mp \omega_V$. The corresponding coupling constants are denoted as κ_S and κ_A. The Heisenberg equations describing this process have the following form:

$$\dot{a}_L = -i(\omega_L a_L + 2\kappa_S^* a_L^\dagger a_S a_V + 2\kappa_A a_L^\dagger a_A a_V^\dagger),$$

$$\dot{a}_S = -i(\omega_S a_S + \kappa_S a_L^2 a_V^\dagger), \quad \dot{a}_A = -i(\omega_A a_A + \kappa_A a_L^2 a_V),$$

$$\dot{a}_V = -i(\omega_V a_V + \kappa_S a_L^2 a_S^\dagger + \kappa_A^* a_L^{\dagger 2} a_A). \tag{2.2}$$

This can be solved approximately up to t^2 as follows:

$$a_L(t) = \exp(-i\omega_L t)\{a_L - 2it(\kappa_S^* a_L^\dagger a_S a_V + \kappa_A^* a_L^\dagger a_A a_V^\dagger) + t^2[|\kappa_S|^2(2a_L a_S^\dagger a_S a_V^\dagger a_V - a_L^\dagger a_L^2(a_V^\dagger a_V + a_S^\dagger a_S + 1)) + |\kappa_A|^2(2a_L a_A^\dagger a_A(a_V^\dagger a_V + 1) + a_L^\dagger a_L^2(a_A^\dagger a_A - a_V^\dagger a_V)) + 2a_L(\kappa_S^* \kappa_A a_S a_A^\dagger a_V^2 + \mathrm{h.c.})]\},$$

$$a_S(t) = \exp(-i\omega_S t)\{a_S - i\kappa_S t a_L^2 a_V^\dagger + t^2[|\kappa_S|^2 a_S(-2(a_L^\dagger a_L + 1)(a_V^\dagger a_V + 1) + \tfrac{1}{2} a_L^{\dagger 2} a_L^2 + 2a_L^\dagger a_L + 1) + \tfrac{1}{2}\kappa_S \kappa_A a_L^4 a_A^\dagger - 2\kappa_S \kappa_A^* (a_L^\dagger a_L + 1) a_A a_V^{\dagger 2}]\},$$

$$a_A(t) = \exp(-i\omega_A t)\{a_A - i\kappa_A t a_L^2 a_V - t^2[|\kappa_A|^2 a_A(2(a_L^\dagger a_L + 1) a_V^\dagger a_V + \tfrac{1}{2} a_L^{\dagger 2} a_L^2 + 2a_L^\dagger a_L + 1) + \tfrac{1}{2}\kappa_S \kappa_A a_L^4 a_S^\dagger + 2\kappa_S^* \kappa_A (a_L^\dagger a_L + 1) a_S a_V^2]\},$$

$$a_V(t) = \exp(-i\omega_V t)\{a_V - it(\kappa_S a_L^2 a_S^\dagger + \kappa_A^* a_L^{\dagger 2} a_A) + t^2[|\kappa_S|^2 a_V(-2(a_L^\dagger a_L + 1)(a_S^\dagger a_S + 1) + \tfrac{1}{2} a_L^{\dagger 2} a_L^2 + 2a_L^\dagger a_L + 1) + |\kappa_A|^2 a_V(2a_L^\dagger a_L a_A^\dagger a_A - \tfrac{1}{2} a_L^{\dagger 2} a_L^2) - 2\kappa_S \kappa_A^* a_S^\dagger a_A a_V^\dagger]\}, \tag{2.3}$$

where $a_j = a_j(0)$. It can be verified that the following conservation laws are valid:

$$\frac{d}{dt}(a_L^\dagger(t) a_L(t) + 2a_S^\dagger(t) a_S(t) + 2a_A^\dagger(t) a_A(t)) = 0,$$

$$\frac{d}{dt}(a_V^\dagger(t) a_V(t) + a_A^\dagger(t) a_A(t) - a_S^\dagger(t) a_S(t)) = 0. \tag{2.4}$$

The time evolution of the system is fully described by the normal quantum characteristic function [4]

$$C_N(\beta_L, \beta_S, \beta_A, \beta_V, t) = \mathrm{Tr}\{\varrho(0) \prod_{j=\mathrm{L,S,A,V}} \exp(\beta_j a_j^\dagger(t)) \exp(-\beta_j^* a_j(t))\}, \tag{2.5}$$

with $\varrho(0)$ being the initial density matrix of the system. Substituting from (2.3) and performing the normal ordering of (2.5) in the initial operators using the Baker–Hausdorff identity (up to t^2) and if we further apply the Glauber–Sudarshan representation of $\varrho(0)$, we arrive at the complete normal quantum characteristic function valid up to t^2:

$$\begin{aligned} C_N(\{\beta_j\}, t) = \langle \exp \{ &\sum_{j=L,S,A,V} (\beta_j \xi_j^*(t) - \beta_j^* \xi_j(t)) - \sum_{j=L,S,V} |\beta_j|^2 B_j + \tfrac{1}{2} \sum_{j=L,V} (\beta_j^{*2} C_j + \text{c.c.}) \\ &+ \Big(\sum_{\substack{j<k \\ j,k=L,S,A,V}} \beta_j^* \beta_k^* D_{jk} + \beta_L \beta_S^* \bar{D}_{LS} + \beta_L \beta_V^* \bar{D}_{LV} + \text{c.c.} \Big) - (\beta_L^3 D_{LLL}^* + \beta_L \beta_A \beta_V D_{LAV}^* \\ &+ \beta_L \beta_S \beta_V D_{LSV}^* + \beta_L^2 \beta_A D_{LLA}^* + \beta_L^2 \beta_V D_{LLV}^* + \beta_L^2 \beta_S D_{LLS}^* + \beta_L \beta_V^2 D_{LVV}^* + \beta_L |\beta_L|^2 \bar{D}_{LLL} \\ &+ |\beta_L|^2 \beta_V \bar{D}_{LVL} + \beta_L |\beta_V|^2 \bar{D}_{LVV} + \beta_L^2 \beta_S^* \bar{D}_{LLS} - \text{c.c.}) + (\beta_L^4 D_{LLLL}^* \\ &+ \beta_L \beta_S \beta_V^2 D_{LSVV}^* + \text{c.c.}) + |\beta_L|^2 |\beta_V|^2 \bar{D}_{LVLV} + |\beta_L|^4 \bar{D}_{LLLL} \} \rangle, \end{aligned} \tag{2.6}$$

where the set (L, S, A, V) is assumed to be ordered, $\xi_j(t)$ are time-dependent complex amplitudes obtained from (2.3) if the initial complex amplitudes ξ_j are substituted for the initial operators a_j (ξ_j are eigenvalues of a_j in the coherent state $|\{\xi_j\}\rangle$). The brackets in (2.6) represent the average over ξ_j and

$$B_L = 4t^2[|\kappa_S|^2 |\xi_S|^2 |\xi_V|^2 + |\kappa_A|^2(|\xi_L|^2 + |\xi_V|^2 + 1) + (\kappa_S^* \kappa_A \xi_S \xi_A^* \xi_V^2 + \text{c.c.})],$$

$$B_S = t^2 |\kappa_S|^2 |\xi_L|^4, \qquad B_V = t^2[|\kappa_S|^2 |\xi_L|^4 + 2|\kappa_A|^2 (2|\xi_L|^2 + 1)\, |\xi_A|^2],$$

$$\begin{aligned} C_L = {}&-\exp(-i2\omega_L t)\, \{2it(\kappa_S^* \xi_S \xi_V + \kappa_A^* \xi_A \xi_V^*) \\ &+ t^2[|\kappa_S|^2 \xi_L{}^2 (|\xi_S|^2 + |\xi_V|^2 + 1) - |\kappa_A|^2 \xi_L{}^2 (|\xi_A|^2 - |\xi_V|^2) + 4\kappa_S^* \kappa_A^* \xi_L^{*2} \xi_S \xi_A]\}, \end{aligned}$$

$$C_V = -\exp(-i2\omega_V t)\, [4t^2 \kappa_S \kappa_A^* (|\xi_L|^2 + 1) \xi_S^* \xi_A],$$

$$D_{LS} = -\exp(-i(\omega_L + \omega_S)t) t^2 [|\kappa_S|^2 \xi_L \xi_S (|\xi_L|^2 + 2|\xi_V|^2) + 2\kappa_S \kappa_A^* \xi_L \xi_A \xi_V^{*2}],$$

$$D_{LA} = -\exp(-i(\omega_L + \omega_A)t) t^2 [|\kappa_A|^2 \xi_L \xi_A (|\xi_L|^2 + 2|\xi_V|^2 + 2) + 2\kappa_S^* \kappa_A \xi_L \xi_S \xi_V^2],$$

$$\begin{aligned} D_{LV} = {}&-\exp(-i(\omega_L + \omega_V)t)\, \{2it\kappa_A^* \xi_L^* \xi_A + t^2 \xi_L \xi_V \\ &\times [|\kappa_S|^2 (|\xi_L|^2 + 2|\xi_S|^2) + |\kappa_A|^2 (|\xi_L|^2 - 2|\xi_A|^2)]\}, \end{aligned}$$

$$D_{SA} = -\tfrac{1}{2} \exp(-i(\omega_S + \omega_A)t) t^2 \kappa_S \kappa_A \xi_L^4,$$

$$D_{SV} = -\exp(-i(\omega_S + \omega_V)t)\, \{i\kappa_S t \xi_L^2 + 2t^2 (|\xi_L|^2 + 1)\, [|\kappa_S|^2 \xi_S \xi_V + 2\kappa_S \kappa_A^* \xi_A \xi_V^*]\},$$

$$D_{AV} = -\exp(-i(\omega_A + \omega_V)t) 2t^2 |\kappa_A|^2 (|\xi_L|^2 + 1) \xi_A \xi_V,$$

$$D_{LS} = -\exp(i(\omega_L - \omega_S)t) 2t^2 \kappa_S \kappa_A \xi_L^3 \xi_A^*,$$

$$D_{LV} = -\exp(i(\omega_L - \omega_V)t) 4t^2 \xi_L^* (|\kappa_A|^2 |\xi_A|^2 \xi_V + \kappa_S \kappa_A^* \xi_S^* \xi_A \xi_V^*),$$

$$D_{LLL} = \exp(-i3\omega_L t) 2t^2 \kappa_S^* \kappa_A^* \xi_L^* \xi_S \xi_A,$$

$$D_{LAV} = \exp(-i(\omega_L + \omega_A + \omega_V)t) 2t^2 |\kappa_A|^2 \xi_L \xi_A \xi_V,$$

$$D_{\mathrm{LSV}} = \exp(-i(\omega_{\mathrm{L}}+\omega_{\mathrm{S}}+\omega_{\mathrm{V}})t)2t^2(|\kappa_{\mathrm{S}}|^2\xi_{\mathrm{L}}\xi_{\mathrm{S}}\xi_{\mathrm{V}}+2\kappa_{\mathrm{S}}\kappa_{\mathrm{A}}^*\xi_{\mathrm{L}}\xi_{\mathrm{A}}\xi_{\mathrm{V}}^*),$$

$$D_{\mathrm{LLA}} = \exp(-i(2\omega_{\mathrm{L}}+\omega_{\mathrm{A}})t)\frac{t^2}{2}|\kappa_{\mathrm{A}}|^2\xi_{\mathrm{L}}^2\xi_{\mathrm{A}},$$

$$D_{\mathrm{LLV}} = \exp(-i(2\omega_{\mathrm{L}}+\omega_{\mathrm{V}})t)\left[i\kappa_{\mathrm{A}}^*t\xi_{\mathrm{A}}+\frac{t^2}{2}(|\kappa_{\mathrm{S}}|^2+|\kappa_{\mathrm{A}}|^2)\xi_{\mathrm{L}}^2\xi_{\mathrm{V}}\right],$$

$$D_{\mathrm{LLS}} = \exp(-i(2\omega_{\mathrm{L}}+\omega_{\mathrm{S}})t)\frac{t^2}{2}|\kappa_{\mathrm{S}}|^2|\xi_{\mathrm{L}}|^2\xi_{\mathrm{S}},$$

$$D_{\mathrm{LVV}} = \exp(-i(\omega_{\mathrm{L}}+2\omega_{\mathrm{V}})t)4t^2\kappa_{\mathrm{S}}\kappa_{\mathrm{A}}^*\xi_{\mathrm{L}}\xi_{\mathrm{S}}^*\xi_{\mathrm{A}},$$

$$\bar{D}_{\mathrm{LLL}} = \exp(i\omega_{\mathrm{L}}t)2t^2|\kappa_{\mathrm{A}}|^2\xi_{\mathrm{L}}^*|\xi_{\mathrm{A}}|^2,$$

$$\bar{D}_{\mathrm{LVL}} = \exp(i\omega_{\mathrm{V}}t)4t^2(|\kappa_{\mathrm{A}}|^2|\xi_{\mathrm{A}}|^2\xi_{\mathrm{V}}^*+\kappa_{\mathrm{S}}^*\kappa_{\mathrm{A}}\xi_{\mathrm{S}}\xi_{\mathrm{A}}^*\xi_{\mathrm{V}}),$$

$$\bar{D}_{\mathrm{LVV}} = \exp(i\omega_{\mathrm{L}}t)4t^2|\kappa_{\mathrm{A}}|^2\xi_{\mathrm{L}}^*|\xi_{\mathrm{A}}|^2,$$

$$\bar{D}_{\mathrm{LLS}} = \exp(i(2\omega_{\mathrm{L}}-\omega_{\mathrm{S}})t)t^2\kappa_{\mathrm{S}}\kappa_{\mathrm{A}}\xi_{\mathrm{L}}^2\xi_{\mathrm{A}}^*,$$

$$D_{\mathrm{LLLL}} = -\exp(-i4\omega_{\mathrm{L}}t)\frac{t^2}{2}\kappa_{\mathrm{S}}^*\kappa_{\mathrm{A}}^*\xi_{\mathrm{S}}\xi_{\mathrm{A}},$$

$$D_{\mathrm{LSVV}} = -\exp(-i(\omega_{\mathrm{L}}+\omega_{\mathrm{S}}+2\omega_{\mathrm{V}})t)4t^2\kappa_{\mathrm{S}}\kappa_{\mathrm{A}}^*\xi_{\mathrm{L}}\xi_{\mathrm{A}},$$

$$\bar{D}_{\mathrm{LVLV}} = 4t^2|\kappa_{\mathrm{A}}|^2|\xi_{\mathrm{A}}|^2, \quad \bar{D}_{\mathrm{LLLL}} = t^2|\kappa_{\mathrm{A}}|^2|\xi_{\mathrm{A}}|^2. \tag{2.7}$$

Compared to Raman scattering [3], there are third and fourth-order terms in β_j in the characteristic function similar to the case of non-degenerate hyper-Raman scattering [1, 2] even if all modes are initially coherent. Moreover, in the present case the third and fourth-order terms in β_{L} also occur. Fluctuations of the intensity, W_j, in separate modes are given by:

$$\langle(\Delta W_{\mathrm{L}})^2\rangle = \left.\frac{\partial^4 C_{\mathrm{N}}}{\partial\beta_{\mathrm{L}}^2\partial(-\beta_{\mathrm{L}}^*)^2}\right|_{\{\beta_j\}=0} - \left.\left(\frac{\partial^2 C_{\mathrm{N}}}{\partial\beta_{\mathrm{L}}\partial(-\beta_{\mathrm{L}}^*)}\right)^2\right|_{\{\beta_j\}=0}$$

$$= \langle B_{\mathrm{L}}^2+|C_{\mathrm{L}}|^2+2B_{\mathrm{L}}|\xi_{\mathrm{L}}(t)|^2+(C_{\mathrm{L}}^*\xi_{\mathrm{L}}^2(t)+\mathrm{c.c.})$$

$$+4(\bar{D}_{\mathrm{LLL}}\xi_{\mathrm{L}}(t)+\mathrm{c.c.})+4\bar{D}_{\mathrm{LLLL}}\rangle+\langle(B_{\mathrm{L}}+|\xi_{\mathrm{L}}(t)|^2)^2\rangle-\langle B_{\mathrm{L}}+|\xi_{\mathrm{L}}(t)|^2\rangle^2,$$

$$\langle(\Delta W_j)^2\rangle = \left.\frac{\partial^4 C_{\mathrm{N}}}{\partial\beta_j^2\partial(-\beta_j^*)^2}\right|_{\{\beta_j\}=0} - \left.\left(\frac{\partial^2 C_{\mathrm{N}}}{\partial\beta_j\partial(-\beta_j^*)}\right)^2\right|_{\{\beta_j\}=0}$$

$$= \langle B_j^2+|C_j|^2+2B_j|\xi_j(t)|^2+(C_j^*\xi_j^2(t)+\mathrm{c.c.})\rangle+\langle(B_j+|\xi_j(t)|^2)^2\rangle$$

$$-\langle B_j+|\xi_j(t)|^2\rangle^2, \quad j = \mathrm{S, A, V} \tag{2.8}$$

and the correlations among modes are given by:

$$\langle \Delta W_j \Delta W_k \rangle = \frac{\partial^4 C_N}{\partial \beta_j \partial(-\beta_j^*) \partial \beta_k \partial(-\beta_k^*)}\bigg|_{\{\beta_j\}=0} - \frac{\partial^2 C_N}{\partial \beta_j \partial(-\beta_j^*)} \frac{\partial^2 C_N}{\partial \beta_k \partial(-\beta_k^*)}\bigg|_{\{\beta_j\}=0}$$

$$= \langle |D_{jk}|^2 + |\bar{D}_{jk}|^2 + (D_{jk}\xi_j^*(t)\xi_k^*(t) - \bar{D}_{jk}\xi_j(t)\xi_k^*(t)$$

$$+ \bar{D}_{jkk}\xi_j(t) + \bar{D}_{jkj}\xi_k(t) + \text{c.c.}) + \bar{D}_{jkjk}\rangle + \langle (B_j + |\xi_j(t)|^2)(B_k + |\xi_k(t)|^2)\rangle$$

$$- \langle B_j + |\xi_j(t)|^2 \rangle \langle B_k + |\xi_k(t)|^2 \rangle, \quad j < k. \tag{2.9}$$

The brackets in (2.8) and (2.9) 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 simply obtain from (2.8):

$$\langle (\Delta W_L)^2 \rangle = t^2\{|\kappa_S|^2[4|\xi_S|^2\langle n_V\rangle(2|\xi_L|^4 + 6|\xi_L|^2 + 1)$$

$$-2|\xi_L|^4(|\xi_S|^2 + \langle n_V\rangle + 1)] + |\kappa_A|^2[4|\xi_A|^2(\langle n_V\rangle + 1)$$

$$\times(2|\xi_L|^4 + 6|\xi_L|^2 + 1) + 2|\xi_L|^4(|\xi_A|^2 - \langle n_V\rangle)] - 4(2\langle n_V\rangle + 1)R\},$$

$$\langle (\Delta W_S)^2 \rangle = 2|\kappa_S|^2 t^2 |\xi_L|^4 |\xi_S|^2(\langle n_V\rangle + 1), \qquad \langle (\Delta W_A)^2 \rangle = 2|\kappa_A|^2 t^2 |\xi_L|^4 |\xi_A|^2 \langle n_V\rangle,$$

$$\langle (\Delta W_V)^2 \rangle = \langle n_V\rangle^2 + 2t^2\langle n_V\rangle\{|\kappa_S|^2[|\xi_L|^4(|\xi_S|^2 + \langle n_V\rangle + 1) - 4(|\xi_L|^2 + 1)|\xi_S|^2\langle n_V\rangle$$

$$-2\langle n_V\rangle] + |\kappa_A|^2[|\xi_L|^4(|\xi_A|^2 - \langle n_V\rangle) + 4|\xi_L|^2|\xi_A|^2(\langle n_V\rangle + 1) + 2|\xi_A|^2] + R\}, \tag{3.1}$$

where $R = \kappa_S\kappa_A\xi_L^4\xi_S^*\xi_A^* + \text{c. c.}$, $\langle n_V\rangle = \langle|\xi_V|^2\rangle$. Thus we find that the antibunching can be observed in the laser mode in spontaneous scattering ($\xi_S = \xi_A = 0$) to the amount $-2|\kappa_S|^2t^2|\xi_L|^4(\langle n_V\rangle + 1) - 2|\kappa_A|^2t^2|\xi_L|^4\langle n_V\rangle$. This property is closely analogous to the possibility of measuring the anticorrelation of fluctuations between the first and the second laser modes, $\langle \Delta W_1 \Delta W_2\rangle$, in non-degenerate hyper-Raman scattering and in the subfrequency mode in the course of second harmonic generation from vacuum. It should be noted that $\langle(\Delta W_{1,2})^2\rangle = 0$, that is $\langle(\Delta W_1 + \Delta W_2)^2\rangle = 2\langle\Delta W_1\Delta W_2\rangle = -2t^2|\xi_1|^2|\xi_2|^2(|\kappa_S|^2(\langle n_V\rangle + 1) + |\kappa_A|^2\langle n_V\rangle)$ (ξ_1 and ξ_2 are the initial complex amplitudes in laser modes 1 and 2 respectively) in non-degenerate hyper-Raman scattering and $\langle(\Delta W_L)^2\rangle = 0$ in Raman scattering if $\xi_S = \xi_A = 0$, so that this effect is quite typical for the degenerate hyper-Raman scattering. In all other cases $\langle(\Delta W_j)^2\rangle = 0$, $j =$ S, A, V.

Similarly from (2.9):

$$\langle \Delta W_L \Delta W_S\rangle = t^2\{-2|\kappa_S|^2|\xi_L|^2|\xi_S|^2[|\xi_L|^2(2\langle n_V\rangle + 1) + 2\langle n_V\rangle] + 2(\langle n_V\rangle + 1)R\},$$

$$\langle \Delta W_L \Delta W_A\rangle = t^2\{-2|\kappa_A|^2|\xi_L|^2|\xi_A|^2[|\xi_L|^2(2\langle n_V\rangle + 1) + 2(\langle n_V\rangle + 1)] + 2\langle n_V\rangle R\},$$

$$\langle \Delta W_L \Delta W_V\rangle = t^2\{|\kappa_S|^2[4(|\xi_L|^2 + 1)|\xi_S|^2\langle n_V\rangle^2 + 4|\xi_L|^2|\xi_S|^2\langle n_V\rangle(\langle n_V\rangle - 1) - 2|\xi_L|^4\langle n_V\rangle$$

$$(2|\xi_S|^2+\langle n_V\rangle+1)]+|\kappa_A|^2[4(|\xi_L|^2+1)\,|\xi_A|^2(\langle n_V\rangle+1)^2+2|\xi_L|^4(\langle n_V\rangle+1)\,(2|\xi_A|^2-\langle n_V\rangle)$$
$$+4|\xi_L|^2|\xi_A|^2(\langle n_V\rangle^2+\langle n_V\rangle+2)]+2R\},$$
$$\langle \Delta W_S \Delta W_A\rangle = -t^2(\langle n_V\rangle+1/2)R,$$
$$\langle \Delta W_S \Delta W_V\rangle = t^2\{|\kappa_S|^2[2|\xi_L|^4|\xi_S|^2(\langle n_V\rangle+1)$$
$$+|\xi_L|^4(\langle n_V\rangle+1)^2-4(|\xi_L|^2+1)\,|\xi_S|^2\langle n_V\rangle\,(\langle n_V\rangle+1)]+(\langle n_V\rangle+1)R\},$$
$$\langle \Delta W_A \Delta W_V\rangle = t^2\{|\kappa_A|^2[|\xi_L|^4\langle n_V\rangle\,(\langle n_V\rangle-2|\xi_A|^2)-4(|\xi_L|^2+1)$$
$$\times|\xi_A|^2\langle n_V\rangle\,(\langle n_V\rangle+1)]-\langle n_V\rangle R\}. \tag{3.2}$$

Thus, the anticorrelation in $\langle \Delta W_L \Delta W_{S,A}\rangle$ can occur in a manner similar to that in Raman scattering or in non-degenerate hyper-Raman scattering. It is maximal if $\Delta\varphi = 4\varphi_L+\psi_S+\psi_A-\varphi_S-\varphi_A = 0$, φ_j being phases of ξ_j and $\psi_{S,A}$ those of $\kappa_{S,A}$; for $\Delta\varphi = \pi/2$ the phase-dependent terms vanish, $R = 0$. Between modes L and V there is anticorrelation in the spontaneous scattering ($\xi_S = \xi_A = 0$), $\langle \Delta W_L \Delta W_V\rangle = -2(|\kappa_S|^2+|\kappa_A|^2)t^2|\xi_L|^4 \langle n_V\rangle(\langle n_V +1)$, but it also occurs in practice if

$$|\xi_L|^2 \gg \langle n_V\rangle \gg |\xi_S|^2, |\xi_A|^2. \tag{3.3}$$

Also between modes S and A anticorrelation occurs if $\Delta\varphi = 0$. However, $\langle \Delta W_{S,A} \Delta W_V\rangle \geqslant 0$ provided that (3.3) holds.

3.2. The laser mode coherent and all other modes chaotic

In this case we additionally average (3.1) and (3.2) over ξ_S and ξ_A which are now chaotic, i. e., we substitute $\langle|\xi_S|^2\rangle = \langle n_S\rangle, \langle|\xi_A|^2\rangle = \langle n_A\rangle$, $R = 0$ and we add the following terms to the shown quantities arising from the last two terms in (2.8) and (2.9)

$$\langle(\Delta W_S)^2\rangle : \langle n_S\rangle^2+2|\kappa_S|^2t^2[|\xi_L|^4-2(1+2(|\xi_L|^2+1)\,\langle n_V\rangle)]\,\langle n_S\rangle^2,$$
$$\langle(\Delta W_A)^2\rangle : \langle n_A\rangle^2-2|\kappa_A|^2t^2[|\xi_L|^4+2(1+2\langle n_V\rangle+2|\xi_L|^2(\langle n_V\rangle+1))]\,\langle n_A\rangle^2,$$
$$\langle \Delta W_L \Delta W_S\rangle : 2|\kappa_S|^2t^2[-|\xi_L|^4+2\langle n_V\rangle\,(2|\xi_L|^2+1)]\,\langle n_S\rangle^2,$$
$$\langle \Delta W_L \Delta W_A\rangle : 2|\kappa_A|^2t^2[|\xi_L|^4+2(\langle n_V\rangle+1)\,(2|\xi_L|^2+1)]\,\langle n_A\rangle^2,$$
$$\langle \Delta W_S \Delta W_V\rangle : |\kappa_S|^2t^2[|\xi_L|^4-4(|\xi_L|^2+1)\,\langle n_V\rangle]\,\langle n_S\rangle^2,$$
$$\langle \Delta W_A \Delta W_V\rangle : |\kappa_A|^2t^2[|\xi_L|^4+4|\xi_L|^2(\langle n_V\rangle+1)+2]\,\langle n_A\rangle^2, \tag{3.4}$$

in all other cases the corrections are zero. Thus, antibunching occurs in $\langle(\Delta W_L)^2\rangle$ as before, which corresponds to anticorrelation between laser modes 1 and 2 in the non--degenerate case. In the other modes no antibunching is possible. The correlation of modes L and V has the same form as in Section 3.1 with $R = 0$. The anticorrelation between modes L and S, and L and A is practically unaffected by the corrections in (3.4) if (3.3) holds ($|\xi_{S,A}|^2 = \langle n_{S,A}\rangle$). Modes S and A are uncorrelated whereas the correlations between modes S and V, and A and V are always non-negative in the present case.

Note that when measuring fluctuations in pairs of modes, i. e., $\langle(\Delta W_j+\Delta W_k)^2\rangle$, $j \neq k$, $j, k = $ L, S, A, V, these quantities behave similarly to those in the Raman scattering case [3] and antibunching can generally occur under special conditions if all modes are initially coherent. For instance, in $\langle(\Delta W_L)^2\rangle$, phase-dependent antibunching of the first order in $\kappa_S t$ and $\kappa_A t$ occurs if $\varphi_S+\varphi_V-2\varphi_L-\psi_S = -\pi/2$ and $\varphi_A-\varphi_V-2\varphi_L-\psi_A = -\pi/2$. In the cases considered here these quantities are non-negative except $\langle(\Delta W_L+\Delta W_{S,A})^2\rangle = \langle(\Delta W_L)^2\rangle$ provided that $\xi_S = \xi_A = 0$ ($\langle n_S\rangle = \langle n_A\rangle = 0$) giving antibunching in spontaneous scattering, including the case when all modes are initially coherent.

4. *Conclusion*

In this paper we have shown that the laser mode exhibits antibunching in the course of spontaneous scattering. In the other modes the variances of the intensity are non-negative. Between the laser and Stokes or anti-Stokes modes anticorrelation possibly depends on the values of the phase-dependent terms. It always occurs if the phase-dependent terms are absent or if $|\xi_L|^2 \gg \langle n_V\rangle \gg \langle n_{S,A}\rangle$ provided that phonon, Stokes and anti-Stokes modes are chaotic at $t = 0$. Between the laser and phonon modes there is anticorrelation in the course of spontaneous scattering or if the above number condition is fulfilled. Stokes and anti-Stokes modes may be phase-dependently anticorrelated if the phonon mode is initially chaotic and all photon modes are initially coherent, whereas they are uncorrelated if the laser mode is initially coherent and the other modes are chaotic. The correlations between the Stokes or anti-Stokes and phonon modes are always non-negative. While in Raman scattering the antibunching can be observed in $\langle(\Delta W_j+\Delta W_k)^2\rangle$, $j \neq k$ only if all modes are initially coherent; in the present process $\langle(\Delta W_L+\Delta W_{S,A})^2\rangle < 0$ in the course of spontaneous scattering.

The statistical properties of degenerate hyper-Raman scattering are a natural degeneracy of those for non-degenerate hyper-Raman scattering; this is reflected by the similar behaviour of $\langle(\Delta W_L)^2\rangle$ in the degenerate case and $\langle(\Delta W_1\Delta W_2\rangle$ in the non--degenerate case. The other corresponding quantities behave, of course, similarly.

We may conclude that degenerate hyper-Raman scattering also represents a higher-order non-linear optical process providing various possibilities for the observation of antibunching or anticorrelation.

We note that Simaan in his recent paper [5] considered the quantum statistical properties of the Stokes hyper-Raman effect using the master equation and Fock states.

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