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NUMBER AND PHASE CORRELATIONS IN RAMAN SCATTERING

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ABSTRACT

Number and phase correlations of fields in a two-mode Raman scattering model are analyzed. The existence of nonclassical two-mode effects including violations of the Cauchy-Schwarz inequality and Muirhead inequality, negative values of the normally ordered number-difference variance and intermode photon anticorrelations are investigated applying four criteria. We distinguish 16 cases and show explicitly that 6 of them are forbidden whereas the remaining 10 occur. Some general relationships between various criteria for the intermode number correlations are derived. Optical phase correlations are studied within the Pegg-Barnett phase formalism. A general expression for the phase-difference variance is obtained.

1. Model of Raman scattering

Models of Raman scattering of few radiation modes have attracted much interest[1-4]. In the Raman effect, photons of the pump field at frequency ω_1 are annihilated emitting photons of the Stokes mode at frequency ω_1 and of the anti-Stokes mode at frequency ω_2. The few-mode assumption implies that the theory is the best suited for scattering in a tuned cavity. The model can describe scattering from a gas of two-level atoms[2] or from a large number of phonon modes, that is, a phonon bath[3,4]. We are interested in the nonlinear problem describing also depletion of the pump field. Thus we do not apply the parametric approximation[3], which would effectively linearize the problem. But for simplicity, we neglect anti-Stokes generation, which is physically justified for a scattering medium at low temperatures[1]. The master equation for the reduced density operator $\widehat{\rho}$, describing the Raman effect under Markov approximation, was given by McNeil and Walls in the form[1]

$$\frac{\partial\widehat{\rho}}{\partial\tau}=\frac{\beta_1}{2}\left([\widehat{a}_1\widehat{a}_2^\dagger\widehat{\rho},\widehat{a}_1^\dagger\widehat{a}_2]+[\widehat{a}_1\widehat{a}_2^\dagger,\widehat{\rho}\widehat{a}_1^\dagger\widehat{a}_2]\right)-\frac{\beta_2}{2}\left([\widehat{a}_1\widehat{a}_2^\dagger,\widehat{a}_1^\dagger\widehat{a}_2\widehat{\rho}]+[\widehat{\rho}\,\widehat{a}_1\widehat{a}_2^\dagger,\widehat{a}_1^\dagger\widehat{a}_2]\right), \quad (1)$$

where the subscript "1" denotes the pump operators, and "2" is for the Stokes variables; $\widehat{a}_{1,2}$ ($\widehat{a}_{1,2}^\dagger$) are the annihilation (creation) operators; τ is the rescaled time γt, where γ is the gain constant for the Stokes mode. By considering the process at low temperature, the second term in eq. (1) can be neglected ($\beta_2\approx 0$), whereas the first term remains with $\beta_1\approx 1$.

The complete solution of the master equation (1) was given by Miranowicz and Kielich[4]. Here, we rewrite their solution more compactly. We find the elements of $\widehat{\rho}(\tau)$ in Fock basis in the form

$$\begin{aligned}\langle n_1 n_2|\widehat{\rho}(\tau)|n_1' n_2'\rangle &= \sum_{l=0}^{n_2} F_{\lambda l}\,(l!)^2 \left[\binom{n_1+l}{l}\binom{n_1'+l}{l}\binom{n_2}{l}\binom{n_2'}{l}\right]^{1/2} \\ &\quad \times \langle n_1+l|\widehat{\rho}_1(0)|n_1'+l\rangle\langle n_2-l|\widehat{\rho}_2(0)|n_2'-l\rangle, \end{aligned} \tag{2}$$

where the function $F_{\lambda l}$, for $0 \le \lambda < l$, is

$$F_{\lambda l} = \sum_{q=0}^{\lambda} \sum_{q'=\lambda+1}^{l} \prod_{\substack{p=0\\ p\neq q}}^{\lambda} [f(p)-f(q)]^{-1} \prod_{\substack{p'=\lambda+1\\ p'\neq q'}}^{l} [f(p')-f(q')]^{-1} \tag{3}$$
$$\times \left(\tau\delta_{f(q)f(q')}\exp[-f(q)\tau] + (\delta_{f(q)f(q')}-1)\frac{\exp[-f(q)\tau]-\exp[-f(q')\tau]}{f(q)-f(q')}\right)$$

and for other values of λ

$$F_{\lambda l} = \sum_{q=0}^{l} \exp[-f(q)\tau] \prod_{\substack{p=0\\ p\neq q}}^{l} [f(p)-f(q)]^{-1}, \tag{4}$$

where $f(x) = \frac{1}{2}[(n_1+x)(n_2-x+1) + (n_1'+x)(n_2'-x+1)]$. The coefficient $\lambda = [\![(n_2+n_2')/4 - (n_1+n_1')/4]\!]$ is given in terms of the integer-value function $[\![x]\!]$ (the maximum integer $\le x$). We assume that the Stokes and pump beams are initially independent. Eq. (2) holds for n_2' greater than n_2. Nonetheless, the time-dependence of the complete density matrix $\widehat{\rho}(\tau)$ is determined since the elements $\langle n_1 n_2|\widehat{\rho}(\tau)|n_1' n_2'\rangle$ for $n_2' < n_2$ are given by the complex conjugate of (2) with interchanged $n_1 \leftrightarrow n_1'$ and $n_2 \leftrightarrow n_2'$. The solution of eq. (1) for the diagonal matrix elements $\langle n_1 n_2|\widehat{\rho}(\tau)|n_1 n_2\rangle$, being a special case of the solution (2) for $n_1 = n_1'$ and $n_2 = n_2'$, was obtained by McNeil and Walls[1] and, for arbitrary initial fields, by Simaan[2] . The solution (2) provides a complete specification of all measurable properties of the light field. Here, we analyze number and phase photon correlations only.

2. Photon-number correlations

Various criteria for the existence of nonclassical intermode phenomena in the two-mode radiation fields have been proposed[5-7]. However, the complete set of their interrelations has, as yet, not been established. We demonstrate some general properties of the four criteria by way of their comparison in the two-mode model of Raman scattering.

To examine the intermode photon anticorrelation (antibunching)[6] we calculate the second-order cross-correlation function Q_{12},

$$Q_{12} = \frac{\langle\Delta\widehat{n}_1\Delta\widehat{n}_2\rangle}{\langle\widehat{n}_1\rangle\langle\widehat{n}_2\rangle} = \frac{\langle\widehat{n}_1\widehat{n}_2\rangle}{\langle\widehat{n}_1\rangle\langle\widehat{n}_2\rangle} - 1. \tag{5}$$

The function Q_{12} vanishes for uncorrelated states; it is positive for correlated and negative for anticorrelated states. To check whether the intermode statistics are nonclassical we study violations of the classical inequalities. Violation of the Muirhead inequality can be measured by Lee's D_{12} parameter[6]

$$D_{12} = \langle : \hat{n}_1^2 : \rangle + \langle : \hat{n}_2^2 : \rangle - 2\langle \hat{n}_1 \hat{n}_2 \rangle. \tag{6}$$

Negative values of the normally ordered number-difference variance

$$V_{12} \equiv \langle : [\Delta(\hat{n}_1 - \hat{n}_2)]^2 : \rangle = \langle : \Delta\hat{n}_1^2 : \rangle + \langle : \Delta\hat{n}_2^2 : \rangle - 2\langle \Delta\hat{n}_1 \Delta\hat{n}_2 \rangle \tag{7}$$

can be interpreted as number-difference sub-Poissonian behavior in the two-mode radiation, whereas positive values of (7) will be referred to as number-difference super-Poissonian statistics. We omit the analysis of the number-sum variance[3] $\langle : [\Delta(\hat{n}_1 + \hat{n}_2)]^2 : \rangle$, since this quantity is time independent in the Raman model. Violation of the Cauchy-Schwarz inequality can be determined by Agarwal's I_{12} parameter[7]

$$I_{12} = \frac{\sqrt{\langle : \hat{n}_1^2 : \rangle \langle : \hat{n}_2^2 : \rangle}}{\langle \hat{n}_1 \hat{n}_2 \rangle} - 1. \tag{8}$$

Negative values of either D_{12}, V_{12}, or I_{12} occur only for a not well-behaved Glauber-Sudarshan P function of the field, and hence these three parameters can be treated as criteria for the existence of nonclassical correlations[3,5-7]. We find that

$$D_{12} - 2\langle \hat{n}_1 \hat{n}_2 \rangle I_{12} = \left(\sqrt{\langle : \hat{n}_1^2 : \rangle} - \sqrt{\langle : \hat{n}_2^2 : \rangle} \right)^2 \geq 0, \tag{9}$$

$$D_{12} - V_{12} = (\langle \hat{n}_1 \rangle - \langle \hat{n}_2 \rangle)^2 \geq 0. \tag{10}$$

We conclude from eq. (9) that violation of the Muirhead inequality, in the form $D_{12} < 0$, implies the violation of the Cauchy-Schwarz inequality, $I_{12} < 0$, and by eq. (10) implies number-difference sub-Poissonian statistics of the field, $V_{12} < 0$. Moreover, we find that intermode photon anticorrelation ($Q_{12} < 0$) can occur for two-mode states both satisfying and violating the Cauchy-Schwarz inequality and Muirhead inequality, and for both number-difference sub- and super-Poissonian behavior. We briefly analyze photon-number correlations in the two-mode model of Raman scattering. The correlation parameters (5)–(8) for the pump (subscript "1") and Stokes ("2") modes can be positive or negative for different initial conditions and evolution times τ. Analyzing the signs of any 4 parameters, one can distinguish 16 cases. However for the parameters (5)–(8), 6 cases cannot occur due to our constraints (9) and (10). All the remaining ten cases are observed in Fig.1. Using the notation (sign(Q_{12}), sign(D_{12}), sign(V_{12}), sign(I_{12})), we observe the following cases: **1.** all four parameters positive, i.e. $(+,+,+,+)$, for $0 < \tau < 0.71$ in Fig.1b and for $0.14 < \tau < 0.47$ in Fig.1f for initially chaotic pump field of intensity initially dominant over the intensity of the Stokes mode. Four cases where only one parameter is negative: **2.**

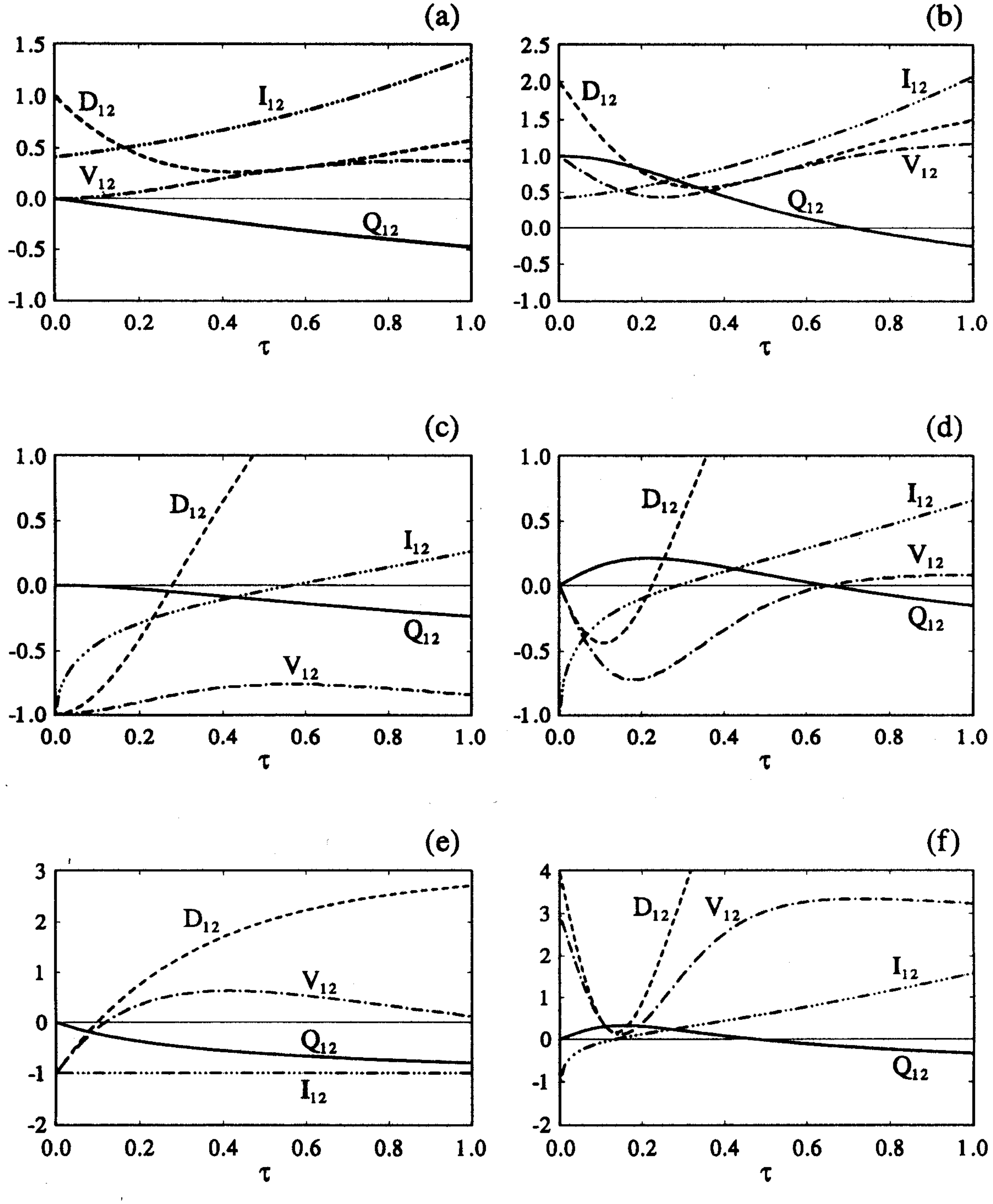

Fig.1. Evolution of Q_{12} (thick solid lines), D_{12} (dashed lines), V_{12} (dot-dashed lines) and I_{12} (dot-dot-dashed lines) for various initial fields with mean number of photons in the pump, $\langle\widehat{n}_1\rangle$, and Stokes, $\langle\widehat{n}_2\rangle$, modes: **(a)** $\langle\widehat{n}_1\rangle = 1$ in coherent state and $n_2 = 0$ in vacuum; **(b)** $\langle\widehat{n}_1\rangle = 1$ in chaotic state and $n_2 = 0$ vacuum; **(c)** $\langle\widehat{n}_1\rangle = 1$ in coherent and $n_2 = 1$ in Fock state; **(d)** $\langle\widehat{n}_1\rangle = 1$ in chaotic and $n_2 = 1$ in Fock state; **(e)** $n_1 = 1$ in Fock and $\langle\widehat{n}_2\rangle = 1$ in coherent state; **(f)** $\langle\widehat{n}_1\rangle = 2$ chaotic and $n_2 = 1$ in Fock state.

$(+,+,+,-)$ for $0 < \tau < 0.14$ in Fig.1f; **3.** $(-,+,+,+)$ for $\tau > 0$ in Fig.1a, $\tau > 0.71$ in Fig.1b, $\tau > 0.65$ in Fig.1d and $\tau > 0.47$ in Fig.1f; **4.** $(+,+,-,+)$ for $0.29 < \tau < 0.65$ in Fig.1d. The last case of that kind, i.e. **5.** $(+,-,+,+)$, does not occur since it is related to states which violate the Muirhead inequality and simultaneously fulfil the Cauchy-Schwarz inequality. Such a case is excluded by our constraint (9). Also in this case, violation of the Muirhead inequality is accompanied by number-difference super-Poissonian statistics, which in turn is forbidden by relation (10). One can distinguish six cases where exactly two parameters are negative: **6.** $(-,+,-,+)$ for $0.57 < \tau$ in Fig.1c; **7.** $(+,+,-,-)$ for $0.22 < \tau < 0.29$ in Fig.1d and **8.** $(-,+,+,-)$ for $0.11 < \tau < 1.12$ in Fig.1e. However, the remaining three cases cannot occur, namely **9.** $(-,-,+,+)$ is forbidden by both relations (9) and (10); **10.** $(+,-,-,+)$ is excluded by condition (9) and **11.** $(+,-,+,-)$ is excluded by (10). There are still the following four cases where exactly 3 parameters are negative, thus **12.** $(-,+,-,-)$ for $0.28 < \tau < 0.57$ in Fig.1c and for $0.09 < \tau < 0.11$ in Fig.1e **13.** $(+,-,-,-)$ for $0 < \tau < 0.22$ in Fig.1d, whereas the cases **14.** $(-,-,-,+)$ and **15.** $(-,-,+,-)$ are forbidden by conditions (9) and (10), respectively. In the Raman model, we can also observe the situation, for short-time evolution of fields initially in a single-photon Fock state ($n_{1,2} = 1$) and coherent state with $|\alpha_{2,1}| = 1$, that **16.** all four parameters are negative, i.e. $(-,-,-,-)$, for $0 < \tau < 0.28$ in Fig.1c and for $0 < \tau < 0.09$ in Fig.1e.

3. Phase correlations

We investigate the quantum phase properties of two-mode radiation Raman fields within the Pegg-Barnett formalism for the optical phase correlations[8] as a generalization of their theory of the single-mode Hermitian optical phase operator [9,10]. The phase distributions representing the difference or sum of the two single-mode phases can be defined in a 2π range with the help of the so-called casting procedure [8,10]. Here, we discuss only the phase-difference properties since, contrary to the phase-sum properties, they can be measured in experiment. The general expression for the mod(2π) Pegg-Barnett phase-difference distribution was obtained by Luis et al.[11,10]

$$P_{2\pi}(\theta_-) \;=\; \frac{1}{2\pi}\sum_{n=0}^{\infty}\sum_{k=0}^{n}\sum_{l=0}^{n}\exp[i(k-l)\theta_-]\langle l,n-l|\widehat{\rho}|k,n-k\rangle \tag{11}$$

by integration of the joint phase distribution $P(\theta_1,\theta_2)$ over the phase-sum $\theta_+ = \theta_1 + \theta_2$ and by application of the casting procedure to the resulting mod(4π) phase distribution $P_{4\pi}(\theta_-)$. Eq. (11) can also be derived from the phase-difference operator[12].

We calculate the second-order moment of the phase-difference operator[8,10] in order to make our presentation of the number and phase correlations more self-consistent. By application of eq. (11), we find the following general expression

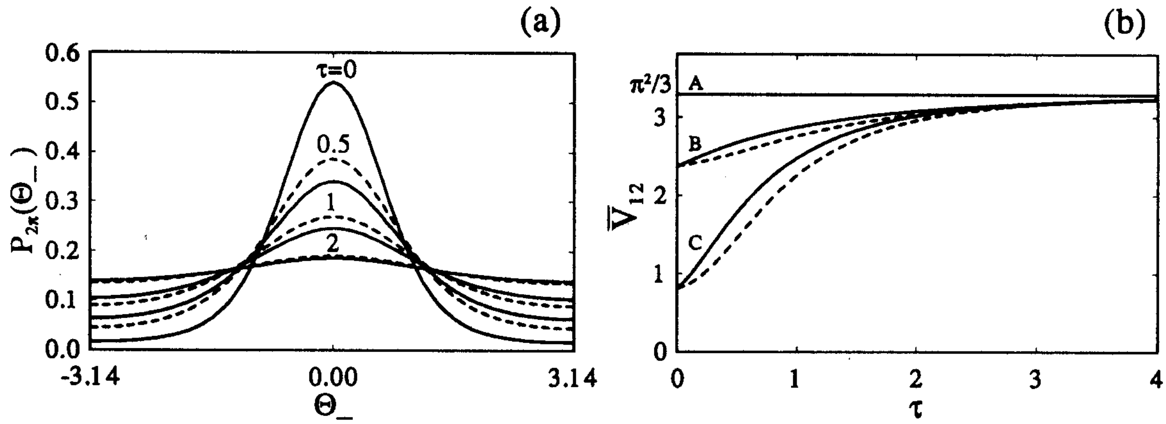

Fig.2. Evolution of (a) the phase-difference distributions $P_{2\pi}(\theta_-)$ and (b) phase-difference variances $\overline{V}_{12}$ for the fields initially coherent: $|\alpha_1|^2 = 1$, $|\alpha_2|^2 = 0$ and $|\alpha_1|^2 = 0$, $|\alpha_2|^2 = 1$ (line A), $|\alpha_1|^2 = 0.1$, $|\alpha_2|^2 = 1$ (solid line B) and $|\alpha_1|^2 = 1$, $|\alpha_2|^2 = 0.1$ (dashed line B), $|\alpha_1|^2 = 1$, $|\alpha_2|^2 = 2$ (solid line C and those in fig. 2(a)) and $|\alpha_1|^2 = 2$, $|\alpha_2|^2 = 1$ (dashed line B and those in fig. 2(a)).

for the mod(2π) phase-difference variance

$$\begin{aligned}\overline{V}_{12} &= \langle[\Delta(\widehat{\Phi}_{\theta_1} - \widehat{\Phi}_{\theta_2})]^2\rangle_{2\pi} = \int_{-\pi}^{\pi} \theta_-^2 P_{2\pi}(\theta_-)\mathrm{d}\theta_+ \\ &= \frac{\pi^2}{3} + 2\sum_{n=0}^{\infty}\sum_{\substack{k,l\\k\neq l}}^{n} \frac{(-1)^{k-l}}{(k-l)^2}\langle l, n-l|\widehat{\rho}|k, n-k\rangle. \qquad (12)\end{aligned}$$

The evolution of the mod(2π) phase-difference distributions (11) and variances (12) for the difference between the pump and Stokes phases are presented in Fig. 2a and Fig. 2b, respectively. We observe randomization of the phase d-ifference in the evolution of the Raman fields. Indeed, the distributions (11) for different initial fields, as chosen in Fig. 2a, are almost flat even for $\tau = 2$. We obtain a completely flat distribution $P_{2\pi}(\theta_-) = 1/(2\pi)$ for long evolution times. Hence in the time limit, the mod(2π) phase-difference variances go over into $\pi^2/3$, as is clearly seen in Fig. 2b. This phase-difference randomization process is typical for various two-mode optical phenomena[10]. Moreover, we find that the mod(2π) phase-difference distribution is flat whenever at least one of the single-mode phase distributions for the pump or Stokes fields is flat. Vacuum, Fock and chaotic states have random phase according to the Pegg-Barnett phase formalism[9]. So, the phase-difference distributions (11) are also uniform, and the variances (12) are equal to $\pi^2/3$ for arbitrary evolution times in all situations presented in Fig.1a–1f. We can generalize this interesting property of the mod(2π) phase-difference functions for arbitrary two-mode radiation fields.

4. Conclusions

We have presented a quantum-statistical approach to Raman scattering in a tuned cavity. We have derived a compact complete solution of the master equation for the model. We have calculated various number and phase correlation moments and obtained some new general relationships.

On a simple example of the two-mode Raman model, we show that the criteria of the intermode correlations measured by the parameters (5)–(8) are not equivalent and exemplify different aspects of photon correlations of the radiation fields.

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