Photon Statistics in Higher Order Raman Processes

A. HENGEITRAUB, M.P. SHARMA, F.G. DOS REIS and L.M. BRESCANSIN
Instituto de Fíísica, Universidade Estadual de Campinas, Gixa Postal 6165, Campinas, 13081, SP, Brasil

Received em 20 de fevereiro de 1987

Abstract We present a new technique for deriving the short-time approximation of Agrawal and Mehta. The technique is applied to the nonlinear problem of generation of higher order Stokes radiation. We start by applying the time-translation operator to the boson operators that create the photons involved. This generates a power series in the evolution-time variable \( t \) which, when truncated at \( t^2 \), yields the short-time approximation. We show how one can obtain higher-order approximations. Furthermore we show how to calculate the joint normally-ordered quantum characteristic function and how to derive the covariance and variance functions.

I. INTRODUCTION

Raman scattering processes occur when incident light photons are inelastically scattered. A photon of frequency \( \omega_L \), incident on a Raman active medium, is annihilated giving rise to a photon of frequency \( \omega_P \) and a Stokes photon with frequency \( \omega_S = \omega_L - \omega_P \). Alternatively, an incident laser photon may be annihilated together with a phonon while an anti-Stokes photon at the sum frequency \( \omega_A = \omega_L + \omega_P \) is created. Under conditions of high Raman gain, the Stokes beam of frequency \( \omega_S_1 \) may reach a sufficiently high intensity to undergo Stokes scattering itself producing 2\(^{nd} \) Stokes radiation with frequency \( \omega_S_2 = \omega_S_1 - \omega_P \). The intensity of the 2\(^{nd} \) Stokes mode could also build up to serve as the pump of 3\(^{rd} \) Stokes lines and so forth. In single mode optical fibers\(^1 \) Stokes lines up to the 10\(^{th} \) order are observed whereas, due to phase mismatch, no anti-Stokes mode was observed.

In the present paper we discuss a mathematical approach that renders the study of correlations between the laser and the Stokes modes amenable. We show that it contains the short-time approximation, and we derive general equations for the two mode correlation functions which yield, as a particular case, the variance.

We acknowledge financial support from CNPq and FINEP of Brasil.
The choice of the Hamiltonian describing the dynamics of the processes is discussed in sec. 2, together with the equations of motion of the creation and annihilation operators of the photons involved. Sec. 3 contains the discussion of the way of obtaining the explicit time dependent of the operators starting with the time translation operator. The quantum characteristic function is obtained in general form for two modes in sec. 4. The variance and covariance functions are calculated in sec. 5. Results and conclusions are discussed in sec. 6.

2. THE MODEL HAMILTONIAN AND THE EQUATIONS OF MOTION

The non-linear problem we are considering has been treated, with classical fields, by Bloembergen and Shen\(^2\) and a modified classical theory has been put forward by Linde, Maier and Kaizer\(^3\). We consider a generalized form of the Hamiltonian given by Walls\(^4\) to take into account higher-order Stokes modes. The rotating wave approximation is taken with all fields quantized:

\[
H = H_0 + H_I
\]

\[
H_0 = \hbar \sum_j \omega_j a_j^+ a_j^\dagger ; j = L, S_1, S_2, \ldots, S_n, P
\]

\[
H_I = \hbar \sum_{k=1}^{S_n+1} K_k (a_k^+ a_{k+1}^+ + h.c.)
\]

H is the free field Hamiltonian and \(H_I\) takes into account the interactions. The \(a_j\) are boson annihilation operators for the laser, \(L\), first Stokes, etc., modes. \(K_k\) are the coupling constants for the laser and first Stokes, \(L\), and second Stokes modes, and so on.

3. THE TRANSFORMATION METHOD FOR THE TIME EVOLUTION OF THE OPERATORS

To determine the time dependence of the operators, we make use of the time-translation operator

\[
S(t) = \exp\{iHt/\hbar\} = \exp\{i(H_0 + H_I)t/\hbar\}
\]

where \(H_0\) and \(H_I\) are given in eqs. (2) and (3). Due to the energy conservation condition, \(\omega_k = \omega_{k+1} + \omega_P\), the Hamiltonians \(H_0\) and \(H_I\) com-
mute. This makes possible to write the operator $S(t)$ as a product of two factors. One of these is the unitary operator for transformation to the interaction representation

$$S_I(t) = \exp\{iH_0 t/\hbar\}.$$  \hspace{1cm} (5)

Therefore, in Heisenberg's picture we have, due to the factorization of $S(t)$

$$a_j(t) = \exp\{iHt/\hbar\} a_j \exp\{-iHt/\hbar\}$$

$$= \exp\{iH_I t/\hbar\} a_j^I(t) \exp\{-iH_I t/\hbar\},$$  \hspace{1cm} (6)

where

$$a_j^I(t) = \exp\{iH_0 t/\hbar\} a_j \exp\{-iH_0 t/\hbar\}$$  \hspace{1cm} (7)

is the operator $a_j$ in the interaction representation, From eqs. (7) and (2) we obtain

$$a_j^I(t) = a_j \exp(i\omega_j t),$$  \hspace{1cm} (8)

which, with the help of eq. (6), leads to

$$a_j(t) = A_j(t) \exp(-i\omega_j t).$$  \hspace{1cm} (9)

Finally, through the Baker-Hausdorff identity, keeping terms up to the second power in $t$, we get

$$A_j(t) = \exp(-iH_I t)a_j \exp(iH_I t)$$

$$= a_j + \frac{i}{\hbar} [H_I, a_j] + \frac{1}{2} \left( \frac{i}{\hbar} \right)^2 [H_I, [H_I, a_j]].$$  \hspace{1cm} (10)

These equations are the same as those obtained with the short-time approximation. We shall refer to this approximation as the STL approximation. It renders possible the determination of the time dependence of the boson operators by just doing two commutators

$$[H_I, a_j] = -\hbar \sum_\ell k_\ell (\delta_{pj} a_j a_\ell^+ + \delta_{j+1,\ell} a_j a_\ell^+ + \delta_{j,\ell+1} a_\ell a_j + \delta_{j,\ell} a_\ell a_j)$$  \hspace{1cm} (11)
4. THE QUANNM CHARACTERISTIC FUNCTION

To describe the statistical properties of the non-linear interaction in higher order Raman scattering we calculate the joint normally ordered characteristic function. For two different modes $i$ and $j$ it is defined by the relation

$$C_N(\beta_i, \beta_j; t) = \text{Tr}\{[\hat{p}(0)\exp[\beta_i^{+}a_i(t)] + \beta_j^{+}a_j(t)]\exp[-\beta_i^{+}a_i(t) - \beta_j^{+}a_j(t)]\} \quad (13)$$

The density operator $\hat{p}(0)$, for coherent laser and Stokes modes and chaotic phonons is written as

$$\rho(0) = \frac{1}{n_p} \int \exp\left[-\frac{|\xi_p|^2}{n_p}\right] |\xi_L, \xi_{s_1}, \ldots, \xi_{s_n}, \xi_p\rangle \langle \xi_L, \xi_{s_1}, \ldots, \xi_{s_n}, \xi_p| d^2\xi_p, \quad (14)$$

where $n_p$ is the average number of phonons at $t=0$ and

$$a_j|\xi_L, \xi_{s_1}, \ldots, \xi_{s_n}, \xi_p\rangle = \xi_j|\xi_L, \xi_{s_1}, \ldots, \xi_{s_n}, \xi_p\rangle ; \quad j = L, s_1, \ldots, s_n, p \quad (15)$$
the $\xi_j$ being complex numbers.

The linear combinations of creation and annihilation operators in the exponentials in eq. (13) are obtained as explicit functions of time in the STL approximation. The result is

$$B_i a^+_{ij}(t) + B_j a^+_{ij}(t) = \gamma_{ij} a^+_{ij} + \gamma_{ji} a^+_{ij} + \delta_{i,j} t + \delta_{ij} t^2 \quad (16)$$

with

$$\delta_{i,j} = \frac{i}{\hbar} \{ \gamma_{ij} [H_I, a^+_{ij}] + \gamma_{ji} [H_I, a^+_{ij}] \} \quad (17)$$

$$\delta_{ij} = \frac{1}{\hbar} \{ \gamma_{ij} [H_I, [H_I, a^+_{ij}]] + \gamma_{ji} [H_I, [H_I, a^+_{ij}]] \} \quad (18)$$

$$\gamma_{ij} = \beta_{ij} \exp(i \omega_{ij} t) \quad (19)$$

The commutators can be calculated, for each specific pair of the indices $i,j$, from eqs. (11) and (12). It can be easily shown that

$$[a^+_{ij}, \delta_{i,j}] = 0 \quad (20)$$

$$[a^+_{ij}, \delta_{ij}] = 0 \quad (21)$$

This then allows us to calculate, within the STL approximation,

$$\exp[B_i a^+(t) + B_j a^+(t)] = \exp(\gamma_{ij} a^+_{ij} + \gamma_{ji} a^+_{ij}) \exp(\delta_{i,j} t + \delta_{ij} t^2)$$

$$\approx \exp(\gamma_{ij} a^+_{ij} + \gamma_{ji} a^+_{ij}) \exp(\delta_{i,j} t + (\delta_{ij} t + \frac{1}{2} \delta_{ij}^2) t^2) \quad (22)$$

It follows that the second exponential factor in eq. (13) can be written as a product of $\exp(-\gamma_{ij} a^+_{ij} - \gamma_{ji} a^+_{ij})$ times a polynomial of the second degree in $t$.

Finally, the characteristic function for two modes is written in general form as

$$C_H(B_i, B_j) = \text{Tr}(\rho(0) A_{i,j}(t)) \quad (23)$$

where
\[ a(t) = \exp(\gamma_{ij} a_i^+ a_j^+ \rho_{ij}) \exp(-\gamma_{ij} a_i a_j) \]  

(24)

with

\[ \rho_{ij} = 1 + (\sigma^i_{ij} - \sigma_{ij}^0) t + (\sigma^i_{ij} - \sigma_{ij}^0 + \frac{1}{2} \sigma^i_{ij}^2 + \frac{1}{2} \sigma^j_{ij}^2 - \sigma^i_{ij} \sigma^j_{ij}) t^2 \]  

(25)

and \( \rho(0) \) given in eq. (14).

We can therefore obtain the two-mode characteristic function, as given in eq. (23), with the help of eqs. (17) through (19), from the commutators of eqs. (11) and (12), for each specific case. In particular we have performed the calculations for \((i,j) = (L,s_1), (L,s_2), (s_1,s_2)\)

\[ \sigma_{Ls_1}^i = i(\gamma L s_1 a_i^+ a_i + \gamma s_1 K a_s a_i + \gamma s_1 K a_s^+ a_i^+) \]  

(26)

\[ \sigma_{Ls_1}^{ii} = -\frac{1}{2} \left[ \gamma L s_1^2 L (a_i^+ a_i + a_i^+ a_i) + \gamma s_1 K s_2 (a_s^+ a_s + a_s^+ a_s) + \gamma s_1 K s_1 (a_s^+ a_s + a_i^+ a_i) \right] \]  

(27)

\[ \sigma_{Ls_2}^i = i(\gamma L s_1 a_i^+ a_i + \gamma s_2 K a_s a_i) \]  

(28)

\[ \sigma_{Ls_2}^{ii} = -\frac{1}{2} \left[ \gamma L s_1^2 L (a_i^+ a_i + a_i^+ a_i) + \gamma s_1 K s_2 (a_s^+ a_s + a_s^+ a_s) + \gamma s_1 K s_1 (a_s^+ a_s + a_i^+ a_i) \right] \]  

(29)

\[ \sigma_{s_1 s_2}^i = i(\gamma s_1 K a_i a_i^+ + \gamma s_1 K a_i a_i^+ a_i + \gamma s_1 K a_i a_i a_i) \]  

(30)

\[ \sigma_{s_1 s_2}^{ii} = -\frac{1}{2} \left[ \gamma s_1 K s_2 L (a_i^+ a_i + a_i^+ a_i) + \gamma s_1 K s_1 (a_s^+ a_s + a_s^+ a_s) + \gamma s_1 K s_1 (a_i^+ a_i + a_i^+ a_i) \right] \]  

(31)

Eqs. (23) through (31) determine

\[ \mathcal{C}_N(\beta_i, \beta_j) = (1 + \psi_{ij} t^2) \exp(\gamma_{ij} E_i^+ E_j^+ - \gamma_{ij} E_i E_j - \gamma_{ij} E_i^+ E_j^+) \]  

(32)
where

\[
\psi_{s_1} = iK^2 \left( n_p + |\xi_{s_1}|^2 + 1 \right) \text{Im}(\gamma^{*} \xi_{s_1}) + iK \left( n_{s_2} \text{Im}(\gamma^{*} \xi_{s_2}) - n_{s_2} \text{Im}(\gamma^{*} \xi_{s_2}) \right) + iK^2 \left( n_{s_2} \text{Im}(\gamma^{*} \xi_{s_2}) \right) - K^2 \left( (2n + 1) \text{Re}(\gamma L_{s_2} \xi_{s_2}) \right) - K^2 \left( (2n + 1) \text{Re}(\gamma L_{s_2} \xi_{s_2}) \right) - K^2 \left( (2n + 1) \text{Re}(\gamma L_{s_2} \xi_{s_2}) \right) - K^2 \left( (2n + 1) \text{Re}(\gamma L_{s_2} \xi_{s_2}) \right) - K^2 \left( (2n + 1) \text{Re}(\gamma L_{s_2} \xi_{s_2}) \right)
\]

(33)

\[
\psi_{s_2} = iK^2 \left( n_p + |\xi_{s_1}|^2 + 1 \right) \text{Im}(\gamma^{*} \xi_{s_1}) + iK \left( n_{s_2} \text{Im}(\gamma^{*} \xi_{s_2}) - n_{s_2} \text{Im}(\gamma^{*} \xi_{s_2}) \right) + iK^2 \left( n_{s_2} \text{Im}(\gamma^{*} \xi_{s_2}) \right) - K^2 \left( (2n + 1) \text{Re}(\gamma L_{s_2} \xi_{s_2}) \right) - K^2 \left( (2n + 1) \text{Re}(\gamma L_{s_2} \xi_{s_2}) \right)
\]

(34)

and

\[
\psi_{s_1 s_2} = iK^2 \left( n_p + |\xi_{s_1}|^2 \right) \text{Im}(\gamma^{*} \xi_{s_1}) + iK^2 \left( n_{s_2} + |\xi_{s_2}|^2 + 1 \right) \text{Im}(\gamma^{*} \xi_{s_2}) + iK \left( n_{s_2} + |\xi_{s_2}|^2 \right) \text{Im}(\gamma^{*} \xi_{s_2}) + iK^2 \left( n_{s_2} + |\xi_{s_2}|^2 \right) \text{Im}(\gamma^{*} \xi_{s_2}) - K^2 \left( (2n + 1) \text{Re}(\gamma L_{s_2} \xi_{s_2}) \right) - K^2 \left( (2n + 1) \text{Re}(\gamma L_{s_2} \xi_{s_2}) \right) - K^2 \left( (2n + 1) \text{Re}(\gamma L_{s_2} \xi_{s_2}) \right)
\]

(35)
5. PHOTONPHOTON COVARIANCE AND VARIANCE FUNCTIONS

The photon statistical properties we are interested in can be obtained from the joint normally ordered characteristic function.

The correlation between the number of photons of species $i$ and $j$ is studied through the photon-photon covariance functions

$$
\langle \Delta n_i \Delta n_j \rangle = \langle a_i^+(t) a_i(t) a_j^+(t) a_j(t) \rangle - \langle a_i^+(t) a_i(t) \rangle \langle a_j^+(t) a_j(t) \rangle
$$

$$
\langle \Delta n_i \Delta n_j \rangle \equiv \frac{\partial^2 C_N(\beta_i, \beta_j, t)}{\partial \beta_i \partial (-\beta_i^*) \partial \beta_j \partial (-\beta_j^*)} \bigg|_{\beta_i = 0, \beta_j^* = 0} - \frac{\partial^2 C_N(\beta_i, \beta_j, t)}{\partial \beta_i \partial (-\beta_i^*) \partial \beta_j \partial (-\beta_j^*)} \bigg|_{\beta_i = 0, \beta_j^* = 0}
$$

The fluctuation in intensity of mode $i$ is studied by means of the variance, defined as

$$
\langle (\Delta n_i)^2 \rangle = \langle a_i^+(t) a_i(t) \rangle - \langle a_i^+(t) a_i(t) \rangle^2
$$

$$
\equiv \frac{\partial^2 C_N(\beta_i, \beta_j, t)}{\partial \beta_i \partial (-\beta_i^*) \partial \beta_j \partial (-\beta_j^*)} \bigg|_{\beta_i = 0, \beta_j^* = 0}^2 - \frac{\partial^2 C_N(\beta_i, \beta_j, t)}{\partial \beta_i \partial (-\beta_i^*) \partial \beta_j \partial (-\beta_j^*)} \bigg|_{\beta_i = 0, \beta_j^* = 0}
$$

Notice that from eq. (37),

$$
\langle \Delta n_i \Delta n_i \rangle = \langle a_i^+(t) a_i(t) a_i^+(t) a_i(t) \rangle - \langle a_i^+(t) a_i(t) \rangle^2.
$$

Therefore, with the help of the commutation relation for boson creation and annihilation operators and eq. (37), eq. (38) becomes

$$
\langle \Delta n_i \Delta n_i \rangle = \langle (\Delta n_i)^2 \rangle + \langle a_i^+ a_i^+ \rangle,
$$

so that the notation can be somewhat misleading.
The correlation functions (either covariance or variance) can have: a) positive values, in which case we say that the photons show correlations, b) negative values, when the photons are anticorrelated, and c) zero values when the photons involved are statistically independent (uncorrelated).

From eqs. (32) and (36) we have obtained the following covariance functions

$$<\Delta n_L \Delta n_{s_1}> = t^2 K_{s_1} n_p |\xi_{s_1}|^2 |\xi_{L}| \{2K_{s_2} n_p |\xi_{s_2}| \cos(\psi_{L}+\psi_{s_2} - 2\psi_{s_1}) - K_{s_1} (2n_p + 1) |\xi_{s_1}| \}$$

(40)

$$<\Delta n_L \Delta n_{s_2}> = -t^2 K_{s_1} K_{s_2} (2n_p + 1) |\xi_{L}| |\xi_{s_1}|^2 |\xi_{s_2}| \cos(\psi_{L}+\psi_{s_2} - 2\psi_{s_1})$$

(41)

and

$$<\Delta n_{s_1} \Delta n_{s_2}> = t^2 K_{s_2} |\xi_{s_2}|^2 |\xi_{s_1}| \{2K_{s_1} n_p |\xi_{s_1}| \cos(\psi_{L}+\psi_{s_2} - 2\psi_{s_1}) - K_{s_1} K_{s_2} (2n_p + 1) |\xi_{s_1}| \}$$

(42)

For the variances, from eqs. (32) and (37), we calculate

$$\langle (\Delta n_L)^2 \rangle = 2t^2 K_{s_1} n_p |\xi_{L}|^2 |\xi_{s_1}|^2 ,$$

(43)

$$\langle (\Delta n_{s_1})^2 \rangle = 2t^2 \{K_{s_1} (n_p + 1) |\xi_{s_1}|^2 + K_{s_1} n_p |\xi_{s_1}|^2 |\xi_{s_2}| \}$$

(44)

and

$$\langle (\Delta n_{s_2})^2 \rangle = 2t^2 K_{s_2} (n_p + 1) |\xi_{s_1}|^2 |\xi_{s_2}|^2 .$$

(45)

In eqs. (40) to (45), the phase-angles \(\psi_k\), \(k = L, s_1, s_2\), are defined by

$$\xi_k = |\xi_k| \exp(i\psi_k) .$$

(46)

6. CONCLUSIONS

The equations for the covariance and variance functions that were obtained here, eqs. (40) through (45), are exactly the same as those obtained by two of the authors in a previous work9, employing the
short-time approximation and a direct calculation of the covariance and variance functions, that is through eqs. (36) and (37), without computing the characteristic function. We present here a new way of deriving the short-time approximation that can, in principle, be extended to include powers higher than $t^2$, as in eq. (10). We have further shown how to obtain in a consistent way the characteristic function for two modes, from which covariance and variance functions of any order are easily obtained. It can be seen without much difficulty that the commutation relations of eqs. (20) and (21) can be written for three or more different modes, allowing the calculation of multimode characteristic functions.

REFERENCES

Resumo
Apresentamos uma nova técnica para obter a aproximação de tempos curtos de Agrawal e Mehta. A técnica é aplicada ao problema não linear de geração de radiação Stokes de ordem superior. O operador de translação temporal é aplicado aos operadores de bosons que criam os vários fotoncs envolvidos. Um desenvolvimento em série de potenciais de $t$, a variável tempo de evolução, truncado em $t^2$, dá a aproximação de tempos curtos. É indicada a maneira para se obter aproximações de ordem superior. Ainda mais, mostra-se como calcular a função característica quântica com ordenação normal e como deduzir as funções variância e covariância.