Higher-order squeezing in Raman scattering

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The occurrence of fourth and fifth-order squeezing in the fundamental mode of the electromagnetic field in spontaneous Raman process under the short-time approximation is investigated based on a fully quantum mechanical approach. The coupled Heisenberg equations of motion involving real and imaginary parts of the quadrature operators are established. The dependence of the higher-order squeezing on the number of photons is investigated. It is shown that higher-order squeezing allows a much larger fractional noise reduction.

[Keywords: Higher-order squeezing, Raman scattering, Quantum optics, Optical oscillator, Non-linear optical processes, Non-classical states of electromagnetic field, Quadrature operators]

1 Introduction

Squeezing is de-amplifying of noise in one quadrature at the expense of amplifying the noise in the other orthogonal quadrature. The concept of squeezing of electromagnetic field has gained great deal of importance in view of the possibility of reducing the noise of an optical signal below the vacuum limit. It is a special class of minimum uncertainty states of the quantized oscillator and hence, their study is expected to lead to new fundamental insights.

Squeezing has been either experimentally observed or theoretically predicted in a variety of non-linear optical processes, such as, harmonic generation, multi-wave mixing processes, Raman, hyper-Raman etc. Hong & Mandel and Hillery have introduced the notion of higher-order squeezing of quantized electromagnetic field as generalization of normal squeezing. More recently, higher-order squeezing has also been studied in various higher-order non-linear optical processes. In an earlier publication, squeezing up to third-order has been studied in Raman process.

The aim of the present paper is to study higher-order squeezing up to fifth-order in Raman process and also investigate the dependence of squeezing on the photon number.

2 Definition of Higher-Order Squeezing

In order to define the higher-order squeezing effect, the authors consider a single mode of the radiation field with frequency ω and creation & annihilation operators $a$ & $a^\dagger$. When examining squeezing effects, it is more useful to work with the slowly varying operators.

$$ A = a \exp (i \omega t) \text{ and } A^\dagger = a^\dagger \exp (-i \omega t) \ldots (1) $$

It is possible to characterize the fourth-order amplitude by its real and imaginary parts as:

$$ F_1 = \frac{1}{2} (A^4 + A^{A^4}) \text{ and } F_2 = \frac{1}{2} i (A^{\dagger} A^{-1} A^{\dagger}) \ldots (2) $$

These operators do not commute and obey the commutation relation:

$$ [ F_1, F_2 ] = \frac{i}{2} \left( 16 N^3 + 24 N^2 + 56 N + 24 \right) \ldots (3) $$

where $N = A^\dagger A$ is the photon number operator.

This leads to the uncertainty relation ($\hbar = 1$)

$$ \Delta F_1 \Delta F_2 \geq \frac{1}{4} \left( 16 N^3 + 24 N^2 + 56 N + 24 \right) \ldots (4) $$

The fourth-order squeezing state is said to exist in $\Delta F_j$, ($j = 1, 2$) if:

$$ (\Delta F_j)^2 < \frac{1}{4} \left( 16 N^3 + 24 N^2 + 56 N + 24 \right) \ldots (5) $$
Similarly, fifth-order squeezing is defined in terms of the operators:

\[ G_1 = \frac{1}{2} (A^2 + A^*^2) \quad \text{and} \quad G_2 = \frac{1}{2i} (A^2 - A^*^2) \ldots (6) \]

The operators obey the commutation relation:

\[ [G_1, G_2] = \frac{i}{2} (25 N^4 + 50 N^3 + 275 N^2 + 250N + 120) \ldots (7) \]

and, as a result, satisfy the uncertainty relation

\[ \Delta G_1 \Delta G_2 \geq \frac{1}{4} (25 N^4 + 50 N^3 + 275 N^2 + 250N + 120) \ldots (8) \]

Fifth-order squeezing is said to exist if:

\[ \left( \Delta G_1 \right)^2 < \frac{1}{4} (25 N^4 + 50 N^3 + 275 N^2 + 250N + 120) \ldots (9) \]

3 Higher-order Squeezing of the Fundamental Mode in Raman Scattering

Raman scattering is looked upon as the interaction between a pump field of frequency \( \omega_1 \) (mode A), a stokes field of frequency \( \omega_2 \) (mode B) and a vibrational phonon of frequency \( \omega_3 \) (mode C) and the corresponding Hamiltonian can be written as:

\[ H = \omega_a a^+ a + \omega_b b^+ b + \omega_c c^+ c + g (ab^+ c + a^+ bc) \ldots (10) \]

where \( a^+ (a) \), \( b^+ (b) \) and \( c^+ (c) \) are the creation (annihilation) operators of the A, B and C modes respectively and \( g \) is the coupling constant of the process.

Eq. (10) leads to coupled Heisenberg equations of motion:

\[ \dot{A} = -i g B C, \quad \dot{B} = -i g A C^* \quad \text{and} \quad \dot{C} = -i g A B^* \ldots (11) \]

where \( A, B \) and \( C \) are slowly varying operators, which are defined by \( A = a \exp (i\omega_1 t), B = b \exp (i\omega_2 t) \) and \( C = c \exp (i\omega_3 t) \), with the relation \( \omega_1 = \omega_2 + \omega_3 \).

The interaction time is taken to be short, of the order of \( 10^{-6} \) s and \( \chi = \frac{\omega}{c} \) and terms are kept up to second order in \( \chi \). Expanding \( A(t) \) in Taylor’s expansion, we get:

\[ A(t) = A - i g B C - \frac{1}{2} [g B^2 C (N_A A + N_A A A)] \ldots (12) \]

and \( A(t) = A + i g B C \cdot \frac{1}{2} [g B^2 C (N_A A + N_A A A)] \ldots (13) \)

where \( N_A = A^\dagger A, \quad N_B = B^\dagger B \) and \( N_C = C^\dagger C \).

In order to examine the fourth-order squeezing effects in the fundamental mode as a function of time, the quadrature components are defined:

\[ F_{A_1}(t) = 1/2 [A^4(t) + A'^4(t)] \ldots (14) \]

With the help of Eqs (12) and (13), we get the fourth-order equation for fundamental mode \( A\):

\[ A^4(t) = A^4[1 - \frac{1}{2}g B^2 C] \ldots (15) \]

and \( A'^4(t) = A'^4[1 - \frac{1}{2}g B^2 C] \ldots (16) \)

Using Eqs (15) and (16), the real quadrature component for the fundamental mode \( A \) in fourth-order squeezing may be written as:

\[ F_{A_1}(t) = 1/2 [(A^4 + A'^4) - \frac{1}{2}g B^2 C (A^4 + A'^4)] \ldots (17) \]

For the case of spontaneous interaction, it is assumed the initial quantum state as a product of the coherent state \( \alpha \) for the fundamental mode \( A \) and vacuum state for modes \( B \) and \( C \), i.e.

\[ 1\psi = \alpha^\dagger \alpha > | 0 > , \alpha > | 0 > , \alpha > | c > \ldots (18) \]

As given by Lo 28, the wave function of the system starts as a coherent state at \( t = 0 \), and evolves as a squeezed state at a later time, which has motivated the authors to use initial coherent state.

Using Eqs (13) and (14), we get:

\[ <\psi | F_{A_1}(t) | \psi > = \frac{1}{2}[(\alpha^2 + \alpha^*^2) - \frac{1}{2}g B^2 C (\alpha^2 + \alpha^*^2)] \ldots (19) \]

where \( \alpha = | \alpha \rangle \exp (i\theta) \) and \( \alpha^* = | \alpha \rangle \exp (-i\theta) \), \( \theta \) is the phase angle.

We obtain from Eq. (19):

\[ < F_{A_1}^2 (t) > = 1/4 \{1 - 4|g|^2 [\alpha^2 + \alpha^*^2 + 2\alpha^6 + 16|\alpha|^4 + 72|\alpha|^8 + 96|\alpha|^12 + 24] \ldots (20) \]

and \( < F_{A_1}^2 (t) >^2 = 1/4 \{1 - 4|g|^2 [\alpha^2 + \alpha^*^2 + 2\alpha^6] \ldots (21) \}

Hence, \[ \Delta F_{A_1}^2 (t) = < F_{A_1}^2 (t) > - < F_{A_1} (t) >^2 = 1/4 \{1 - 4|g|^2 [\alpha^2 + 72|\alpha|^4 + 96|\alpha|^8 + 24] \ldots (22) \]

The number of photons in mode \( A \) may be defined as:
\[ N_1(t) = A'(t) A(t) = A'A \left[ 1 - |g|^2 t^2 \right] \] ... (23)
and \[ N_2(t) = A'N_A A + N_A \left[ 1 - 2|g|^2 t^2 \right] \] ... (24)
similarly, \[ N_3(t) = \left[ A'A + 3A^3A^2 + A^3A \right] \left[ 1 - 3|g|^2 t^2 \right] \] ... (25)
So, \[ \frac{1}{4} \left( 16N^3 + 24N^2 + 56N + 24 \right) \]
\[ = \frac{1}{4} \left[ 16|\alpha|^6 + 12|\alpha|^4 + 72|\alpha|^2 + 48 |\alpha|^4 + 38 |\alpha|^2 \right] \] ... (26)
which gives:
\[ [\Delta F_{g_1}(t)]^2 = \frac{1}{4} \left( 16N^3 + 24N^2 + 56N + 24 \right) \]
\[ = -4|g|^2 t^2 \left[ |\alpha|^6 + 6|\alpha|^4 + 14.5 |\alpha|^2 + 6 \right] \] ... (27)

![Fig. 1 — Dependence of the fourth-order squeezing \( S_R \) with \( |\alpha|^2 \) in spontaneous Raman process (when \( |g|^2t^2=10^{-3} \))](image)

The right hand side of Eq. (27) is always negative, showing the existence of squeezing in the fourth-order amplitude of the fundamental mode.

Similarly, for fifth-order squeezing in the fundamental mode as a function of time is defined as:
\[ G_{g_1}(t) = 1/2 \left[ A'(t) + A''(t) \right] \] ... (28)
Hence, the real quadrature component for the fundamental mode \( A \) in fifth-order squeezing may be written as:
\[ G_{g_1}(t) = 1/2 \left[ \left( A^5 + A^3 \right) - 1/2|g|^2 t^2 \left( A^5 + A^3 \right) \right] \] ... (29)

Using Eqs (18) and (29), we get:
\[ <\psi | G_{g_1}(t) | \psi > = 1/2 \left[ (|\alpha|^2 + |\alpha'|^2) - 1/2|g|^2 t^2 (|\alpha|^2 + |\alpha'|^2) \right] \] ... (30)

It is obtained in the fifth-order squeezing, keeping terms up to second order in \( g \):
\[ <G_{g_1}^2(t) > = 1/4 \left[ 1 - 5|g|^2 t^2 \right] \]
\[ [|\alpha|^2 + |\alpha'|^2 + 2|\alpha''|^2 + 25|\alpha|^4 + 50|\alpha'|^4 + 275|\alpha|^6 + 250|\alpha'|^6 + 120] \] ... (31)
and \[ <G_{g_1}^2(t) > = 1/4 \left[ 1 - 5|g|^2 t^2 \right] [2|\alpha|^2 + 275|\alpha|^4 + 250|\alpha'|^4 + 120] \] ... (32)

Hence, \[ [\Delta G_{g_1}(t)]^2 < G_{g_1}^2(t) > - < G_{g_1}(t) >^2 \]
\[ = 1/4 \left[ 1 - 5|g|^2 t^2 \right] \]
\[ [2|\alpha|^2 + 275|\alpha|^4 + 250|\alpha'|^4 + 120] \] ... (33)

and \[ N_4(t) = \left[ N_A^4 A^4 + 6A^3A^2 + 7A^2A^2 + N_A \right] \left[ 1 - 4|g|^2 t^2 \right] \] ... (34)
So, \[ \frac{1}{4} \left( 25N^4 + 50N^3 + 275N^2 + 250N + 120 \right) \]
\[ = \frac{1}{4} \left[ 25|\alpha|^4 + 200|\alpha|^2 + 600|\alpha|^4 + 600|\alpha|^2 + 120 \right. \]
\[ - 25|g|^2 t^2 \left[ 4|\alpha|^2 + 30|\alpha|^4 + 68|\alpha'|^4 + 42|\alpha'|^2 \right] \] ... (35)

which give \[ [\Delta G_{g_1}(t)]^2 \]
\[ = \frac{1}{4} \left( 25N^4 + 50N^3 + 275N^2 + 250N + 120 \right) \]
\[ = - 25/4|g|^2 t^2 \left[ |\alpha|^2 + 10|\alpha'|^2 + 52|\alpha|^4 + 78|\alpha'|^4 + 24 \right] \] ... (36)

The right hand side of Eq. (36) is always negative, showing the existence of squeezing in the fifth-order amplitude of the fundamental mode in spontaneous interaction under short-time approximation.

In the Hamiltonian interaction, the coupling constant \( g \) is taken as real. However, to take care of complex \( g \), \( g^2 \) is replaced by \( |g|^2 \) as the authors are not considering the phase terms.

4 Results and Discussion

The results show the presence of squeezing in fundamental mode in Raman scattering in fourth- and fifth-order. In order to study higher-order variations of the squeezing in spontaneous Raman interaction, the right hand side of Eqs (27) and (36) is denoted as \( S_R \) and \( S_{g_1} \) respectively and taking \( |g|^2 \)
$= 10^4$, the variation of squeezing with $|\alpha|^2$ are shown in Figs 1 and 2.

\begin{figure}[h]
\centering
\includegraphics[width=0.5\textwidth]{squeezing.pdf}
\caption{Dependence of the fifth-order squeezing $S_5$ with $|\alpha|^2$ in spontaneous Raman process (when $|\alpha|^2=10^4$)}
\end{figure}

Figs 1 and 2 show that, the squeezing increases with $|\alpha|^2$, which is directly dependent upon the increase of number of photons. This confirms that, the squeezed states are associated with large number of photons.

5 Conclusion

The important findings of the paper can be listed as follows:

A comparison of the fourth-order squeezing with fifth-order squeezing in amplitude having same number of photons shows more squeezing in fifth-order. Hence, this confirms that the squeezed states are associated with large number of photons.

Hence, it is concluded that, higher-order squeezing makes it possible to achieve significantly larger noise reduction than ordinary squeezing. This also establishes the fact that, processes with higher-order non-linearity are more suitable for generation of squeezed light. This study is being extended to $n$th-order squeezing in a future communication. These results may pave way to obtain desired degree of squeezing through different higher-order non-linear optical processes.

References