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QUANTUM STATISTICS OF RAMAN SCATTERING MODEL WITH STOKES MODE GENERATION

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Abstract

The model describing three coupled quantum oscillators with decay of Rayleigh mode into the Stokes and vibration (phonon) modes is examined. Due to the Manley-Rowe relations the problem of exact eigenvalues and eigenstates is reduced to the calculation of new orthogonal polynomials defined both by the difference and differential equations. The quantum statistical properties are examined in the case when initially: the Stokes mode is in the vacuum state; the Rayleigh mode is in the number state; and the vibration mode is in the number or squeezed states. The collapses and revivals are obtained for different initial conditions as well as the change in time the sub-Poisson distribution by the super-Poisson distribution and vice versa.

1 Introduction

Recently quantum statistical properties of scattered light in the Raman process have attracted considerable interest.[1, 2] In particular, the anticorrelation between the Stokes and Rayleigh lines in the resonance scattering have been examined[3, 4], and the generation of squeezed light have been considered.[5, 6, 7] At the same time, the strong quantum fluctuations of energy have been observed[8, 9] experimentally. It is known that the Raman scattering is an example of optical parametric process in which one of the interacting waves is a medium vibration mode of boson type[10] (phonons). In the case of condensed matter, such a mode is usually in thermal equilibrium with a given temperature. The state of that mode is determined by different mechanisms of microscopic interactions in the medium and in some cases can lead to a strong number fluctuation.[11] An example is provided by a polariton-type system in which the equilibrium state is a squeezed one.[12] Evidently, the statistical properties of vibration mode must have influence on the statistics of scattered light.

In this paper we consider the quantum properties of scattered light, and its dependence on the type of statistical distribution function of the vibration mode in the Raman scattering. For simplicity, we suppose the resonance steady state process with generation of inelastic Stokes component only. The initial state of Rayleigh mode is assumed to be a number state, while vibration mode can be initially in a number state or in a squeezed vacuum state. The Stokes field is initially in the vacuum state. The simplest model of three bounded oscillators is used for description of process under consideration.[13] Using the representation of Schrödinger equation in terms of new orthogonal polynomials[14, 15], we examine the dynamics of the Mandel's factor

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of scattered light, and show the qualitative difference between two choices of initial states of the vibration mode.

The rest of this paper is organized as follows. We first introduce the model Hamiltonian for which we calculate the dynamical properties. Evaluation of the eigenvalues and eigenfunctions, and a discussion of how to construct the time dependent Mandel's factor is given in the next section. We then present our results, and conclude with an emphasis on the experimental implications.

2 Raman scattering model

To discuss some of the interesting statistical properties of light we study the Raman scattering model described by the Hamiltonian

$$H = \omega a^\dagger a + \omega_s a_s^\dagger a_s + \omega_b b^\dagger b + \gamma(b^\dagger a_s^\dagger a + a^\dagger a_s b) \quad (1)$$

where a^\dagger and a_s^\dagger are the creation operators for the Rayleigh and Stokes modes, respectively, with the corresponding frequencies ω and ω_s , b^\dagger is the creation operator for the vibration mode with frequency ω_b , and γ is the coupling constant. We consider here only the Stokes process because we will examine the case of low intensity initial field for which anti-Stokes component is negligible.

As a consequence of the Manley-Rowe relations, an exact eigenstate of the above Hamiltonian can be chosen as

$$|n, m\rangle = \sum_{j=0}^n \lambda_j^{n,m} |n-j\rangle_r |j\rangle_s |m\rangle_v \quad (2)$$

where $|\dots\rangle_g$ is the number state of g -th mode. The coefficients $\lambda_j^{n,m}$ are determined by the recursion relation[15]

$$\lambda_{j+1}^{n,m} [(n-j)(j+1)(m+j+1)]^{1/2} = x^{n,m} \lambda_j^{n,m} - \lambda_{j-1}^{n,m} [(n-j+1)j(m+j)]^{1/2} \quad (3)$$

Here $x^{n,m} = (E^{n,m} - \omega_r n - \omega_v m)/\gamma$, and $E^{n,m}$ is an eigenvalue of the Hamiltonian corresponding to the eigenstate given above. The above relation [eq. (3)] can be represented in the form

$$P_{j+1}^{n,m}(x) = x P_j^{n,m}(x) - q_j^{n,m} P_{j-1}^{n,m}(x); \quad \text{with} \quad q_j^{n,m} = (n-j+1)j(m+j), \quad (4)$$

defining some orthogonal polynomials $P(x)$, which can be expressed in terms of the Bernoulli polynomials B as

$$P_j^{n,m}(x) = \sum_{i=0}^j \beta_{ij}^{n,m} B_{ij}^{n,m} x^i, \quad (5)$$

where the coefficients β satisfy

$$\beta_{ij}(j-i) = -jq_j \beta_{i,j-1}, \quad \text{and} \quad \beta_{i,j+1}(n-j)i = j\beta_{i-1,j}.$$

In terms of the polynomials $P(x)$, the equation for eigenvalues E has now the form

$$P_{n+1}^{n,m}(x) = 0, \quad (6)$$

while the coefficients of the eigenstate given by eq. (2) are determined by the expression

$$\lambda_j^{n,m}(x_i) = \lambda_0^{n,m} \frac{F_j^{n,m}(x_i)}{F_j^{n,m}}, \quad \text{with} \quad F_j^{n,m} = \sqrt{[j!]^3 \binom{n}{j} \binom{m+j}{j}}. \quad (7)$$

The coefficient λ_0 is determined from the normalization condition.

Having constructed the solution to the eigenvalue and eigenstate problem, it is not hard to examine the dynamics of the system. In order to discuss our results, we shall use the time-dependent Mandel's Q -factor defined as

$$Q_r(t) = \frac{V_t(a_r^\dagger a_r) - \langle a_r^\dagger a_r \rangle_t}{\langle a_r^\dagger a_r \rangle_t}. \quad (8)$$

Here a_r^\dagger and a_r are the Bose operators for the Rayleigh mode, $V_t(a_r^\dagger a_r)$ is the time-dependent number variance, and $\langle \dots \rangle_t$ denotes a time-dependent expectation value. $Q(t)$ is positive in the case of super-Poisson statistics and negative for sub-Poisson number distribution. Zero value corresponds to the coherent state (Poisson distribution). Q -factors for the other modes may be defined similarly. The time dependent terms in the Mandel's factor must be calculated as corresponding expectation values with time dependent wave function.

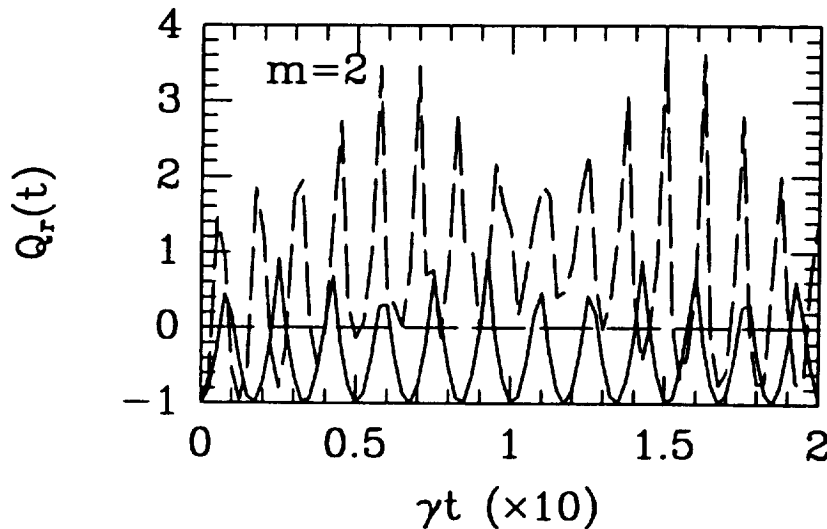


Fig. 1. Q -factor for Rayleigh mode initially in a number state with $n = 2$ (solid) and $n = 10$ (dashed), while the vibration mode is in a number state with $m = 2$.

3 Results and discussion

We have obtained qualitative differences in the quantum statistical properties of the Rayleigh mode depending on the initial state of the vibration mode. Using a number state for the Rayleigh mode, we consider uncorrelated (number state) and correlated (squeezed state) phonons in the vibration mode, and calculate the time evolution of the Mandel's Q -factor for the Rayleigh mode.

In Fig. 1 we show $Q_r(t)$ when the Rayleigh mode is initially in a number state with $n = 2$ and $n = 10$, indicated by solid and dashed lines, respectively. The vibration mode is also in the number state with $m = 2$. We observe that for a small value of n $Q_r(t)$ periodically fluctuates between sub-Poisson and super-Poisson statistics. As n is increased, quantum statistical distribution of the Rayleigh mode becomes more of super-Poissonian. The situation is qualitatively different, when the vibration mode is a correlated one described by a squeezed state as shown in Fig. 2. Here the squeezed state of the vibration mode is prepared such that the mean number of phonons is 2, viz., $|\nu|^2 = 2$. Solid, and dashed lines refer to the Rayleigh mode in the number state with $n = 2$ and $n = 10$, respectively. Here, we note that the distribution of photons remain mostly super-Poissonian, for large enough n . Similar results are obtained when the Rayleigh mode is initially in a coherent state.

To render the description of the vibration mode more realistic, we also use the Bose distribution at a given temperature. Here the mean number of phonons \bar{m} may be regarded as a parameter. In Fig. 3 we show $Q_r(t)$ when the Rayleigh mode is initially in a number state with $n = 2$. Solid, dashed, and dotted lines refer to vibration mode parameter $\bar{m} = 0.1, 1,$ and 10 , respectively. We observe that for small values of \bar{m} (low temperature) $Q_r(t)$ periodically fluctuates between sub-Poisson and super-Poisson statistics. As \bar{m} is increased, quantum statistical distribution of the Rayleigh mode becomes entirely super-Poissonian.

We have also carried out calculations with large numbers of n , and observed the collapse-revival phenomenon occurs in the system for large enough n . Since an increase in n implies an increase in the number of terms in the various sums, it is not surprising to observe the collapse-revival patterns as in the case of the Jaynes-Cummings model.[17, 18] It should be noted that similar behavior was obtained in the numerical calculations of Drobny and Jex[19], for the case of initial coherent state of the Rayleigh mode. In this connection, we emphasize that the collapse-revival phenomenon is a general property of the model described by the Hamiltonian irrespective of the initial state of the Rayleigh mode.

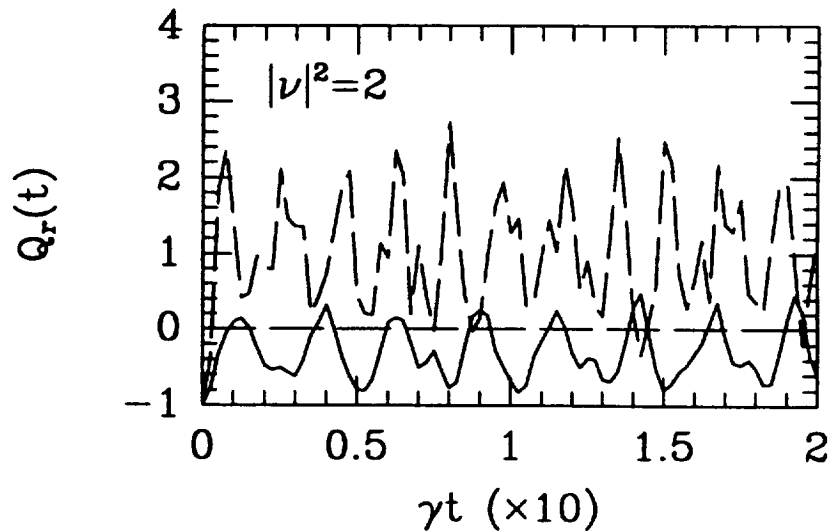


Fig. 2. Same as Fig. 1 when the vibration mode is initially in a squeezed state with parameter $|\nu|^2 = 2$.

We have obtained a qualitative difference in quantum statistical properties of scattered light depending on the statistics of the vibration mode. Our choice of initial state of the vibration mode can be considered as simulating correlated and uncorrelated phonons, and also phonons at finite temperature. We conclude stating that the experimental investigation of quantum statistical properties of scattered light in the Raman correlation spectroscopy with different types of incident light (e.g., in the number state or strongly sub-Poissonian) may yield important information about the correlations in the medium as well as in the molecules.

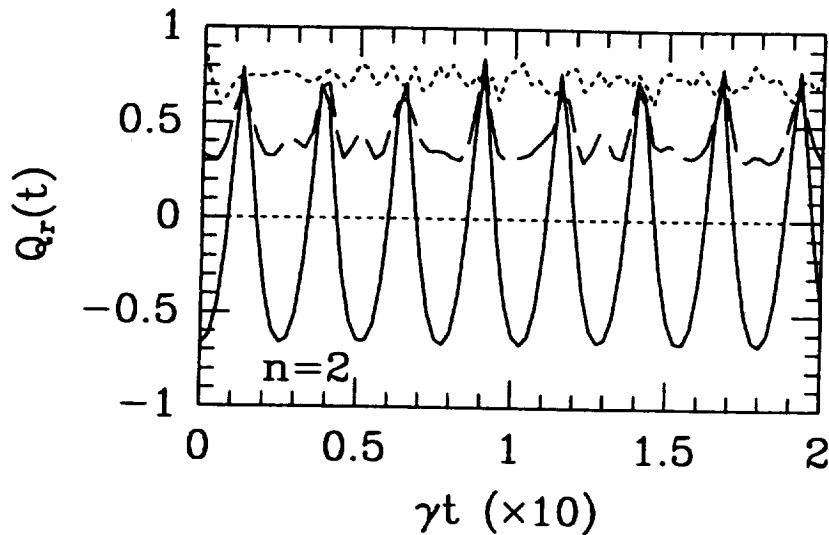


Fig. 3. Q -factor for Rayleigh mode initially in a number state ($n = 2$). Solid, dashed, and dotted lines indicate vibration mode parameter $\bar{m} = 0.1, 1,$ and $10,$ respectively.

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