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A master equation approach to the hyper-Raman effect

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Abstract. The hyper-Raman effect is treated theoretically from the quantum statistical point of view. For this purpose a master equation, based on a microscopically correct Hamiltonian, is derived and then solved analytically. The solution obtained is quite general and enables computation of the complete joint probability distribution for arbitrary time and any initial conditions.

It is well known that when sufficiently intense monochromatic radiation of frequency ω_1 is scattered by a system, the scattered radiation will contain not only the frequencies ω_1 (Rayleigh scattering) and $\omega_1 \pm |\omega|$ (Raman scattering) but also frequencies of the type $2\omega_1$ and $2\omega_1 \pm |\omega|$ where ω is the transition frequency (see Long 1972). The new frequencies are in fact respectively associated with what is called hyper-Rayleigh and hyper-Raman scattering. A theoretical treatment of the hyper-Raman effect has been considered by Long and Stanton (1970). On using the density matrix method they derived general formulae which give information on the frequencies of the scattered radiation, the scattering mechanism and resonance processes. From these results it has been shown that the anti-Stokes hyper-Raman emission at $2\omega_1 + |\omega|$ only takes place in the case where a downward transition is involved and the transition frequency ω has a positive value. In a similar manner, the Stokes hyper-Raman emission at $2\omega_1 - |\omega|$ only takes place in the case where an upward transition is involved and the frequency ω has a negative value.

The purpose of this paper is to consider the problem of the Stokes hyper-Raman effect (SHRE) from the quantum statistical point of view. No such treatment has so far appeared, and therefore it should be of interest to know how the statistical natures of the light fields involved are disturbed. On using a microscopically correct Hamiltonian, an equation which describes the rate of change of the joint probability distribution $P_{n,m}$, caused by the SHRE, is derived and then solved analytically by using a Laplace transform method similar to that used by Simman (1975) for the analogous process of the stimulated Raman effect.

Consider that the SHRE occurs, simply due to the interaction of the incident and scattered light beams with a gas of N identical two-level atoms. In each elementary scattering event of the process this kind of interaction simultaneously destroys two photons, each of frequency ω_1, and creates a single scattered photon with frequency $\omega_2 = 2\omega_1 - |\omega|$ where ω is the frequency separation between the atomic ground state $|1\rangle$ and the excited state $|2\rangle$. It is worth mentioning here (see Long and Stanton 1970) that the SHRE proceeds by three transitions; photons ω_1 are absorbed at two of the transitions, whereas a photon ω_2 is emitted at the third. This implies that there are three electric-dipole interactions altogether, and the atom which is in its ground state $|1\rangle$ before the process gets excited to two virtual intermediate states during the SHRE and finally settles down into its excited state $|2\rangle$ after the process. It is assumed that the atoms only have transitions of the required frequency for the Stokes component of the hyper-Raman effect, where its anti-Stokes component is ignored, and the conditions needed for any other process to occur are also taken to be badly satisfied. The total Hamiltonian H, which describes our system, then has the form

$$H=\hbar\omega_1 a_1^\dagger a_1+\hbar\omega_2 a_2^\dagger a_2+\tfrac{1}{2}\hbar\omega\sum_j (c_{2j}^\dagger c_{2j}-c_{1j}^\dagger c_{1j})+\hbar a_1 a_1 a_2^\dagger \sum_j X_j c_{2j}^\dagger c_{1j}$$
$$+\hbar a_1^\dagger a_1^\dagger a_2 \sum_j X_j c_{2j} c_{1j}^\dagger, \tag{1}$$

where $\hbar=h/2\pi$, h is Planck's constant, $a_k^\dagger$ and a_k are identified with the photon creation and destruction operators for the kth mode, X_j is the dipole matrix element for the SHRE, and finally $c_{1j}^\dagger$, $c_{2j}^\dagger$, c_{1j} and c_{2j} are the creation and destruction operators for the jth atom in the states $|1\rangle$ and $|2\rangle$, respectively. In the above description no account of losses has been included.

It is assumed here that almost all the N atoms are maintained in their ground states by some external influence. Standard techniques (Shen 1967, McNeil and Walls 1974) then lead to the following master equation for the reduced density operator ρ of the light field:

$$\mathrm{d}\rho/\mathrm{d}t=NJ(a_1a_1a_2^\dagger\rho a_1^\dagger a_1^\dagger a_2-2a_1^\dagger a_1^\dagger a_2 a_1 a_1 a_2^\dagger\rho-2\rho a_1^\dagger a_1^\dagger a_2 a_1 a_1 a_2^\dagger), \tag{2}$$

where J is proportional to the modulus squared of the dipole matrix element X_j. The master equation for the diagonal matrix elements $P_{n,m}$ of the density operator ρ in the Fock representation which follows immediately from (2), can then be written as

$$\mathrm{d}P_{n,m}(\tau)/\mathrm{d}\tau=(n+1)(n+2)mP_{n+2,m-1}-n(n-1)(m+1)P_{n,m}, \tag{3}$$

where $\tau=NJt$ is a new time variable. Equation (3) is very often called the photon rate equation for the probability distribution $P_{n,m}(\tau)$ due to the fact that obviously it represents an equation for the probability that at time τ, there are n and m photon numbers present respectively in the incident and scattered light beams. The probability distribution is assumed to be normalised, and it is seen by summation of (3) that a normalised distribution remains normalised as the SHRE proceeds.

In a previous paper (Simaan 1975), a Laplace transform method has been used to solve a photon rate equation which describes the stimulated Raman effect. Equation (3) in fact also proves tractable to solution by the method of Laplace transform, and therefore by following exactly the same general procedure as that of Simaan (1975), the solution of equation (3) can be written as follows:

$$P_{n,m}(\tau)=\sum_{\alpha=0}^{\lambda}\sum_{\gamma=0}^{\alpha}\frac{\prod_{\beta=0}^{\alpha-1}(F_{\beta+1})}{\prod_{\beta=0,\beta\neq\gamma}^{\alpha}(F_\beta-F_\gamma)}\exp(-F_\gamma\tau)Q_{n+2\alpha}(0)R_{m-\alpha}(0)$$
$$+\theta(m)\sum_{\alpha=\lambda+1}^{m}\sum_{\gamma=0}^{\lambda}\sum_{\gamma'=\lambda+1}^{\alpha}\frac{\prod_{\beta=0}^{\alpha-1}(F_{\beta+1})}{\prod_{\beta=0,\beta\neq\gamma}^{\lambda}(F_\beta-F_\gamma)\prod_{\beta=\lambda+1,\beta'\neq\gamma'}^{\alpha}(F_{\beta'}-F_{\gamma'})}$$
$$\times\left(\delta_{F_{\gamma'}F_\gamma}\tau\exp(-F_\gamma\tau)+(1-\delta_{F_{\gamma'}F_\gamma})\frac{\exp(-F_\gamma\tau)-\exp(-F_{\gamma'}\tau)}{(F_{\gamma'}-F_\gamma)}\right)$$
$$\times Q_{n+2\alpha}(0)R_{m-\alpha}(0), \tag{4}$$

where

$$F_\beta = (n+2\beta)(n+2\beta-1)(m-\beta+1),$$

$$\theta(m) = \begin{cases} 0 & \text{for } m=0 \\ 1 & \text{for } m>0, \end{cases}$$

$$\delta_{F_{\gamma'}F_\gamma} = \begin{cases} 1 & \text{for } F_{\gamma'} = F_\gamma \\ 0 & \text{otherwise,} \end{cases}$$

$$\lambda = \begin{cases} \frac{1}{2}m & \text{for } m \text{ even or zero} \\ \frac{1}{2}(m-1) & \text{for } m \text{ odd} \end{cases} \qquad n \geqslant m,$$

$$\lambda = \begin{cases} \frac{2}{3}(m-n) & \text{for } \frac{1}{3}(m-n) \text{ integer} \\ \frac{2}{3}(m-n+1) & \text{for } \frac{1}{3}(m-n+1) \text{ integer} \\ \frac{2}{3}(m-n-1) & \text{for } \frac{1}{3}(m-n-1) \text{ integer} \end{cases} \qquad n<m,$$

and the initial joint distribution $P_{n+2\alpha,m-\alpha}(0)$ has been replaced by a product of the single initial distributions $Q_{n+2\alpha}(0)$ and $R_{m-\alpha}(0)$.

Equation (4) enables computation of the complete joint probability distribution $P_{n,m}(\tau)$ for an arbitrary time τ and any initial distributions $Q_{n+2\alpha}(0)$ and $R_{m-\alpha}(0)$. Figure 1 shows the distribution $P_{n,m}$ as a function of n and m with the fixed time $\tau = 0{\cdot}05$, the incident beam being initially coherent with a mean photon number equal to 10 and the scattered beam initially being a number state with no photons at the commencement of the SHRE. The time dependence of $P_{n,m}$ for the case of an initially chaotic incident beam is presented in figure 2 for the same conditions as in figure 1. The time evolution of the moments, degrees of second-order coherence and the correlation function of the two light beams involved in the SHRE can also be obtained directly from (4) by following a method similar to that of Simaan (1975). In fact we have carried out the calculations for these functions, and the results in general show that the statistical changes in the incident beam are in a way similar to those due to the

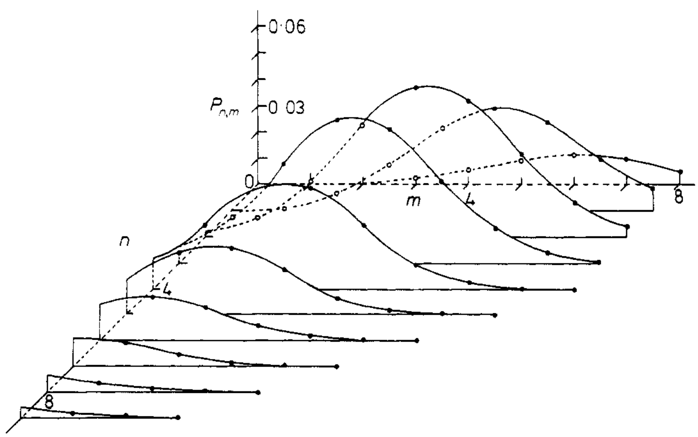

Figure 1. Joint probability distributions $P_{n,m}$ as functions of n and m for the time $\tau = 0{\cdot}05$, and initially coherent pump beam with a mean photon number $\bar{n}_0 = 10$, and an initially number-state scattered beam which has no photons in it.

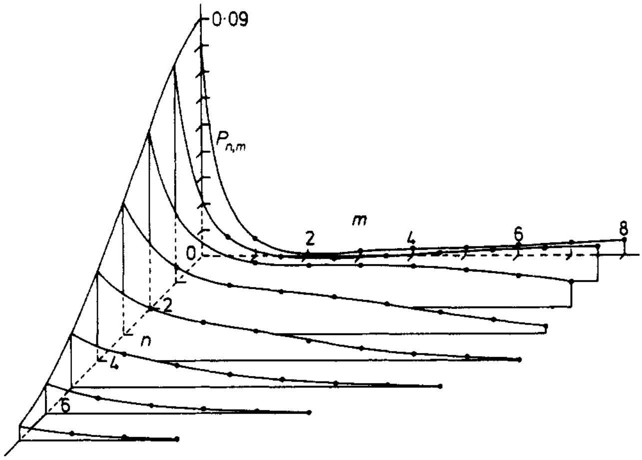

Figure 2. Joint probability distributions $P_{n,m}$ as functions of n and m for the time $\tau = 0{\cdot}05$, an initially chaotic pump beam with a mean photon number $\bar{n}_0 = 10$, and an initially scattered beam which has no photons in it.

process of single-beam two photon absorption (Simaan and Loudon 1975, 1978). On the other hand, the statistical behaviours of the scattered beam are found to be quite similar to those of the corresponding beam due to the stimulated Raman effect (Simaan 1975). Having clearly established these similarities, I was advised not to present the numberical calculations in detail as it was unnecessary. Therefore at this stage I shall conclude the discussion, but not before pointing out that numerical results on the SHRE also showed that the degree of second-order coherence for the incident beams falls below the unit value characteristic of coherent light. This is due to the effect of photon antibunching which has been the subject of much interest recently (Loudon 1976).

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