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A master equation approach to the Raman effect

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Abstract. A quantum-mechanical model of Raman scattering from a phonon bath is formulated. A master equation is derived for the density operator of the light fields alone. This model correctly describes the stochastic coupling of Stokes and anti-Stokes radiation through the phonon bath. Since quantum fluctuations are included, this treatment contains the spontaneous and stimulated Raman effects in one self-consistent formalism. The photon statistics of the light fields are obtained by the solution of a Fokker–Planck equation.

1. Introduction

The temporal and spatial evolution of the light field amplitudes in the Raman effect has been successively described by the classical coupled wave theory of Bloembergen and Shen (1964) (Shen and Bloembergen 1965). The strong damping of the optical phonons relative to that of the light field may be readily introduced in classical theory. However, this feature is not so readily incorporated in quantum theory and has been a limitation of previous quantum-mechanical treatments (Mishkin and Walls 1969, Walls 1970), In these quantum-mechanical approaches the electromagnetic fields and the phonons have been treated as single monochromatic modes. While the assumption of monochromatic light field modes is a reasonable approximation that of a single phonon mode has considerably less validity. In particular the deterministic coupling of the Stokes and anti-Stokes modes through a single phonon mode is a serious shortcoming of these models.

In this paper we attempt to improve on previous quantum-mechanical models by considering Raman scattering from a large number of phonon modes, that is, a phonon bath. The Stokes and anti-Stokes modes are now coupled in a random or stochastic manner through the phonon bath.

The method of the master equation widely used in laser theory (for a full discussion and extensive bibliography see Haken (1970)), is directly applicable to the present problem. By tracing out over the operators of the phonon bath an equation of motion for the density operator of the field modes alone is obtained. Equations of motion for the expectation values of the operators representing the field amplitudes and photon numbers may be derived from the master equation. In addition the photon distributions of the field modes are obtained by solving the corresponding Fokker–Planck equation.

2. Master equation for the Stokes field

We first consider the production of Stokes radiation in a tuned cavity by an incident laser field on a Raman active medium, neglecting anti-Stokes and higher order Stokes production. The interaction of an incident laser beam with frequency ω_L, a Stokes mode with frequency ω_s and a phonon bath may be described phenomenologically by the Hamiltonian

$$H = H_0 + H_1 \tag{2.1}$$

$$H_0 = \hbar\omega_L b_L^\dagger b_L + \hbar\omega_s b_s^\dagger b_s + \sum \hbar\omega_j b_j^\dagger b_j \tag{2.2}$$

$$H_1 = \hbar b_L b_s^\dagger \sum \lambda_j^s b_j^\dagger + \hbar b_L^\dagger b_s \sum \lambda_j^{s*} b_j \tag{2.3}$$

where b_L, b_s and b_j are the annihilation operators for the laser, Stokes and phonon modes respectively, all obeying the boson commutation relations

$$[b_m, b_n^\dagger] = \delta_{mn}. \tag{2.4}$$

The λ_j^s, the coupling constants for the Stokes interaction, contain the phase integrals $\int_V \exp\{-i(\boldsymbol{k}_L - \boldsymbol{k}_s - \boldsymbol{k}_j) . \boldsymbol{r}\}\, d^3r$ where $\boldsymbol{k}_m$ are the wavevectors of the modes.

In deriving the above Hamiltonian it has been assumed that at the comparatively long wavelengths involved in optical processes the oscillations of molecules at neighbouring lattice sites are nearly in phase, that is intermolecular interactions have been neglected. In this long wavelength approximation the lattice may be regarded as a continuum and the optical-mode vibration assumes the form of a simple harmonic oscillator (Pantell and Puthof 1969 § 7.3.3). The sum over lattice sites is replaced by an integral over the volume of the crystal.

The finite size of the crystal destroys the translational invariance of the medium and momentum is no longer precisely conserved in the interaction. Hence a single Stokes mode generated in a finite medium is coupled to a finite band of optical phonons whose wavevectors $\boldsymbol{k}_j$ may differ from $\boldsymbol{k}_L - \boldsymbol{k}_s$ by amounts of the order of the reciprocal of the dimensions of the medium. For a full discussion of the derivation of the Hamiltonian for Raman scattering from optical phonons see von Foerster and Glauber (1971) and Pantell and Puthof (1969).

One may reduce the above trilinear Hamiltonian to a bilinear one by replacing the operator b_L by the constant laser amplitude E_L times the phase factor $\exp(-i\omega_L t)$. This approximation, known as the parametric approximation treats the laser as effectively undepleted by the Raman interaction and as such is valid only for weak interactions or short interaction times. This restriction was lifted in the paper by Walls (1970), however, the interaction considered there was limited to a single phonon mode and a one photon scattering process.

Employing the parametric approximation the interaction Hamiltonian H_1 becomes

$$H_1 = \hbar E_L \exp(-i\omega_L t) b_s^\dagger \sum \lambda_j^s b_j^\dagger + \hbar E_L^* \exp(i\omega_L t) b_s \sum \lambda_j^{s*} b_j. \tag{2.5}$$

This interaction may be considered as an oscillator coupled to a heat bath though the coupling displayed here differs from the usual oscillator (a) reservoir (Γ_B) coupling $a\Gamma_B^\dagger$. However, since energy is conserved due to the factor $\exp(-i\omega_L t)$ the master equation for the reduced density operator ρ of the Stokes field may be derived using standard techniques (see Haken 1970, Louisell 1969). The result for the markoffian master equation in the interaction picture is

$$\frac{\partial \rho}{\partial t} = \frac{\kappa}{2}([b_s^\dagger \rho, b_s] + [b_s^\dagger, \rho b_s]) + \kappa \bar{n}_{ph}([[b_s^\dagger, \rho], b_s]) \tag{2.6}$$

where

$$\kappa = 2\pi g(\omega_L - \omega_s)|\lambda(\omega_L - \omega_s)|^2 |E_L|^2, \tag{2.7}$$

$g(\omega_j)$ is the density of the phonon modes and $\bar{n}_{\rm ph}$ the mean number of phonons per mode in the bath is defined by

$$\bar{n}_{\rm ph} = \left\{\exp\left(\frac{\hbar(\omega_{\rm L}-\omega_{\rm s})}{kT}\right)-1\right\}^{-1} \tag{2.8}$$

where T is the temperature of the phonon bath and k is the Boltzmann constant. In the derivation of equation (2.6) the reservoir spectrum $g(\omega_j)$ and the Stokes coupling constant $\lambda^{\rm s}(\omega_j)$ have been assumed to be flat functions of frequency in the vicinity of $\omega_j = \omega_{\rm L}-\omega_{\rm s}$. This condition is expected to be fulfilled by optical phonons. Implicit in the derivation of the master equation is the assumption that the phonon bath is unaffected by its interaction with the light field. This is equivalent to the assumption used in the classical analysis that the phonons are so quickly damped that they are in their steady state.

From the master equation we may derive equations of motion for the following expectation values:

$$\frac{\rm d}{{\rm d}t}\langle b_{\rm s}^{\dagger}\rangle = \tfrac{1}{2}\kappa\langle b_{\rm s}^{\dagger}\rangle \tag{2.9}$$

$$\frac{\rm d}{{\rm d}t}\langle b_{\rm s}^{\dagger}b_{\rm s}\rangle = \kappa\langle b_{\rm s}^{\dagger}b_{\rm s}\rangle+\kappa(\bar{n}_{\rm ph}+1) \tag{2.10}$$

which are readily solved to yield for the expectation value of the field amplitude

$$\langle b_{\rm s}^{\dagger}(t)\rangle = \langle b_{\rm s}^{\dagger}(0)\rangle \exp(\tfrac{1}{2}\kappa t) \tag{2.11}$$

and for the mean number of Stokes photons

$$\begin{aligned}\bar{n}_{\rm s}(t) &= \langle b_{\rm s}^{\dagger}(t)b_{\rm s}(t)\rangle\\ &= \bar{n}_{\rm s}(0)\,{\rm e}^{\kappa t}+(\bar{n}_{\rm ph}+1)({\rm e}^{\kappa t}-1).\end{aligned} \tag{2.12}$$

The prediction of exponential growth for the Stokes photons is a result of the parametric approximation which as previously mentioned breaks down for sizeable laser depletion. The first term in equation (2.12) corresponding to the amplification of the initial Stokes field is known as the stimulated Raman effect. The second term due to the spontaneous Raman effect occurs even for no Stokes photons initially present ($\bar{n}_{\rm s}(0) = 0$) and corresponds to an amplification of the vacuum fluctuations. Emission of photons at the Stokes frequency may even occur from a phonon bath at zero temperature ($\bar{n}_{\rm ph} = 0$) by the spontaneous Raman effect. This is an intrinsically quantum-mechanical effect which is not contained in classical theory.

3. Photon statistics of the Stokes light

Besides the mean values of the field amplitude and photon number the master equation also contains information on the fluctuations present in the process.

The photon statistics may either be obtained directly by solving for the diagonal matrix elements ρ_{nn} of the density operator (see Scully and Lamb 1967, Pike 1969) or by transforming the master equation into a Fokker–Planck equation dependent on classical variables only. In this paper we shall adopt the latter approach.

The transformation to the Fokker–Planck equation may be accomplished by assuming that a P representation exists for the Stokes field at time t

$$\rho = \int P(\beta, t)|\beta\rangle\langle\beta|\, d^2\beta. \tag{3.1}$$

Here P is a quasi-probability function and $|\beta\rangle$ is the eigenstate of the operator b_s. This diagonal representation for the radiation field in terms of coherent states was introduced by Glauber (1963a, b).

Substitution of equation (3.1) into the master equation (2.6) and using standard techniques (see Haken 1970, Louisell 1969) yields the following Fokker–Planck equation for P:

$$\frac{\partial P}{\partial t} = \left\{-\frac{\kappa}{2}\left(\frac{\partial}{\partial \beta}\beta + \frac{\partial}{\partial \beta^*}\beta^*\right) + \kappa(\bar{n}_{ph}+1)\frac{\partial^2}{\partial\beta\partial\beta^*}\right\}P. \tag{3.2}$$

If we assume that the Stokes field is initially in a coherent state, that is

$$P(\beta, 0) = \delta^2(\beta - \beta_0) \tag{3.3}$$

the solution of equation (3.2) is readily shown to be

$$P(\beta, t) = \frac{1}{\pi(\bar{n}_{ph}+1)(e^{\kappa t}-1)}\exp\left(\frac{-|\beta-\beta_0\, e^{\kappa t/2}|^2}{(\bar{n}_{ph}+1)(e^{\kappa t}-1)}\right). \tag{3.4}$$

This is a gaussian distribution centred at $\beta_0\, e^{\kappa t/2}$ with variance $\frac{1}{2}(\bar{n}_{ph}+1)(e^{\kappa t}-1)$. This describes the superposition of noise quanta from the spontaneous Raman process on the coherent amplification of the stimulated Raman effect.

We note here the close similarity with the results of Mollow and Glauber (1967) on the parametric amplifier. The Hamiltonian they chose for parametric amplification may be derived from equation (2.5) by considering only one phonon mode as the idler mode. Their solution for the P representation at time t (Mollow and Glauber 1967 equation (7.14)) for an initially coherent signal mode and initially chaotic idler mode exhibits the same characteristics as our result (equation (3.4)).

4. Master equation for the coupled Stokes and anti-Stokes fields

We now consider the coupling of Stokes and anti-Stokes radiation through the phonon bath. Higher order Stokes and anti-Stokes production is neglected. Again we assume monochromatic Stokes and anti-Stokes modes appropriate to cavity modes. The interaction may be described by the following phenomenological Hamiltonian:

$$H = H_0 + H_1 \tag{4.1}$$

$$H_0 = \hbar\omega_L b_L^\dagger b_L + \hbar\omega_s b_s^\dagger b_s + \hbar\omega_a b_a^\dagger b_a + \sum \hbar\omega_j b_j^\dagger b_j \tag{4.2}$$

$$H_1 = \hbar b_L b_s^\dagger \sum \lambda_j^s b_j^\dagger + \hbar b_L b_a^\dagger \sum \lambda_j^a b_j + \text{adjoint} \tag{4.3}$$

where the symbols are as defined in § 2; b_a is the annihilation operator for the anti-Stokes mode with frequency ω_a and obeys the boson commutation relations (equation (2.4)). The coupling constant for the anti-Stokes process λ_j^a contains the phase integral $\int_V \exp\{-i(\boldsymbol{k}_L - \boldsymbol{k}_a + \boldsymbol{k}_j) \cdot \boldsymbol{r}\}\, d^3r$.

We again introduce the parametric approximation replacing b_L by $E_L \exp(-i\omega_L t)$. The basic problem here is that of two modes coupled to the same heat bath, one mode coupled in the usual $a\Gamma_B^\dagger$ manner the other coupled in a $a^\dagger\Gamma_B^\dagger$ fashion. The case of two modes coupled to the same heat bath in the usual $a\Gamma_B^\dagger$ manner has been considered by Hubner (1970). The master equation in our case may be derived in an analogous fashion to that of Hubner's since energy is conserved apart from a frequency mismatch

$$-2\Delta = 2\omega_L - \omega_s - \omega_a. \tag{4.4}$$

The result for the master equation in the interaction picture is

$$\begin{aligned}\frac{\partial \rho}{\partial t} = {} & \tfrac{1}{2}\kappa_{ss}([b_s^\dagger\rho, b_s] + [b_s^\dagger, \rho b_s]) + \tfrac{1}{2}\kappa_{aa}([b_a\rho, b_a^\dagger] + [b_a, \rho b_a^\dagger]) + \tfrac{1}{2}\kappa_{sa}\exp(2i\Delta t)([b_s^\dagger\rho, b_a^\dagger] \\ & + [b_s^\dagger, \rho b_a^\dagger]) + \tfrac{1}{2}\kappa_{as}\exp(-2i\Delta t)([b_a\rho, b_s] + [b_a, \rho b_s]) + \bar{n}_{ph}\kappa_{ss}[[b_s^\dagger, \rho], b_s] \\ & + \bar{n}_{ph}\kappa_{aa}[[b_a, \rho], b_a^\dagger] + \bar{n}_{ph}\kappa_{as}\exp(-2i\Delta t)[[b_s^\dagger, \rho], b_a^\dagger] \\ & + \bar{n}_{ph}\kappa_{sa}\exp(2i\Delta t)[[b_a, \rho], b_s]\end{aligned} \tag{4.5}$$

where

$$\begin{aligned}\kappa_{ss} &= 2\pi g(\omega_c)|\lambda^s(\omega_c)|^2|E_L|^2 \\ \kappa_{aa} &= 2\pi g(\omega_c)|\lambda^a(\omega_c)|^2|E_L|^2 \\ \kappa_{as} &= 2\pi g(\omega_c)\lambda^a(\omega_c)\lambda^{s*}(\omega_c)|E_L|^2 \\ \kappa_{sa} &= \kappa_{as}^*\end{aligned} \tag{4.6}$$

and $\omega_L - \omega_s \simeq \omega_c \simeq \omega_a - \omega_L$.

In the derivation of the above equation the reservoir spectrum $g(\omega)$ and the Raman coupling constants $\lambda^s(\omega)$, $\lambda^a(\omega)$ have been assumed to be flat functions of frequency in the vicinity of $\omega = \omega_c$.

The equations of motion for the expectation values of the operators $\langle b_s\rangle$ and $\langle b_a^\dagger\rangle$ which follow from the above master equation are

$$\begin{aligned}\frac{d\langle b_s\rangle}{dt} &= \tfrac{1}{2}\kappa_{ss}\langle b_s\rangle + \tfrac{1}{2}\kappa_{sa}\langle b_a^\dagger\rangle\exp(2i\Delta t) \\ \frac{d\langle b_a^\dagger\rangle}{dt} &= -\tfrac{1}{2}\kappa_{aa}\langle b_a^\dagger\rangle - \tfrac{1}{2}\kappa_{as}\langle b_s\rangle\exp(-2i\Delta t).\end{aligned} \tag{4.7}$$

With the transformations

$$\begin{aligned}\langle b_s\rangle &= \langle \tilde{b}_s\rangle e^{i\Delta t} \\ \langle b_a\rangle &= \langle \tilde{b}_a\rangle e^{i\Delta t}\end{aligned} \tag{4.8}$$

these equations become

$$\frac{d}{dt}\begin{pmatrix}\langle \tilde{b}_s\rangle \\ \langle \tilde{b}_a^\dagger\rangle\end{pmatrix} = \begin{pmatrix}\tfrac{1}{2}\kappa_{ss} - i\Delta & \tfrac{1}{2}\kappa_{sa} \\ -\tfrac{1}{2}\kappa_{as} & -\tfrac{1}{2}\kappa_{aa} + i\Delta\end{pmatrix}\begin{pmatrix}\langle \tilde{b}_s\rangle \\ \langle \tilde{b}_a^\dagger\rangle\end{pmatrix}. \tag{4.9}$$

To simplify the results we shall henceforth ignore the small Raman dispersion effects in the coupling constants and set $\kappa_{ss} = \kappa_{aa} = \kappa_{sa} = \kappa_{as} = \kappa$. The solutions for $\langle b_s\rangle$ and $\langle b_a\rangle$ have a time dependence e^{Gt} where the gain coefficient

$$G = i\Delta \pm i(\Delta^2 + i\kappa\Delta)^{1/2}. \tag{4.10}$$

We note that for perfect frequency matching ($\Delta = 0$) the gain coefficient is zero. This, however, is not the case if laser depletion is included in the analysis (Walls 1971). Amplification of the vacuum fluctuations by the laser pump occurs even for $\Delta = 0$

(Walls 1970). This, however, is a second order effect and the principal Raman gain occurs for large Δ.

For large Δ we obtain two waves, one with almost entirely Stokes character which is amplified and one with almost entirely anti-Stokes character which is attenuated. These waves have the following gain coefficients:

$$G_s = \tfrac{1}{2}\kappa \qquad G_a = 2i\Delta - \tfrac{1}{2}\kappa. \tag{4.11}$$

Thus as a justification of the quantum-mechanical model of the Raman process we have formulated we recover the classical results of Shen and Bloembergen (1965). Note that their classical model considered travelling waves and a spatial variation of the fields with momentum mismatch Δk. Our quantum-mechanical analysis deals with standing waves, and thus considers a temporal variation of the fields with frequency mismatch Δ. For a quantum-mechanical treatment of propagation problems see Tucker and Walls (1969) and von Foerster and Glauber (1971), also Haus (1970).

Further we may derive equations of motion for the expectation values of the following operator products:

$$\begin{aligned}
\frac{d}{dt}\langle b_a^\dagger b_a\rangle &= -\kappa\langle b_a^\dagger b_a\rangle - \tfrac{1}{2}\kappa\langle \tilde{b_a^\dagger b_s^\dagger}\rangle - \tfrac{1}{2}\kappa\langle \tilde{b_a b_s}\rangle + \kappa\bar{n}_{ph} \\
\frac{d}{dt}\langle b_s^\dagger b_s\rangle &= \kappa\langle b_s^\dagger b_s\rangle + \tfrac{1}{2}\kappa\langle \tilde{b_a^\dagger b_s^\dagger}\rangle + \tfrac{1}{2}\kappa\langle \tilde{b_a b_s}\rangle + \kappa(\bar{n}_{ph}+1) \\
\frac{d}{dt}\langle \tilde{b_a b_s}\rangle &= -2i\Delta\langle \tilde{b_a b_s}\rangle + \tfrac{1}{2}\kappa\langle b_a^\dagger b_a\rangle - \tfrac{1}{2}\kappa\langle b_s^\dagger b_s\rangle - \kappa(\bar{n}_{ph}+\tfrac{1}{2}) \\
\frac{d}{dt}\langle \tilde{b_a^\dagger b_s^\dagger}\rangle &= 2i\Delta\langle \tilde{b_a^\dagger b_s^\dagger}\rangle + \tfrac{1}{2}\kappa\langle b_a^\dagger b_a\rangle - \tfrac{1}{2}\kappa\langle b_s^\dagger b_s\rangle - \kappa(\bar{n}_{ph}+\tfrac{1}{2})
\end{aligned} \tag{4.12}$$

where the transformation

$$\langle b_a b_s\rangle = \exp(2i\Delta t)\langle \tilde{b_a b_s}\rangle \tag{4.13}$$

has been introduced. These equations may be simplified by defining

$$\begin{aligned}
\bar{n}'_a(t) &= \bar{n}_a(t) - \bar{n}_{ph} \\
\bar{n}'_s(t) &= \bar{n}_s(t) - (\bar{n}_{ph}+1)
\end{aligned} \tag{4.14}$$

where $\bar{n}_j(t) = \langle b_j^\dagger b_j\rangle$ and with the following change of variables:

$$\begin{aligned}
A &= \tfrac{1}{2}(\bar{n}'_a(t)+\bar{n}'_s(t)), \qquad B = \tfrac{1}{2}(\bar{n}'_a(t)-\bar{n}'_s(t)), \\
X &= \tfrac{1}{2}(\langle \tilde{b_a b_s}\rangle + \langle \tilde{b_a^\dagger b_s^\dagger}\rangle), \qquad Y = \tfrac{1}{2}(\langle \tilde{b_a b_s}\rangle - \langle \tilde{b_a^\dagger b_s^\dagger}\rangle)
\end{aligned} \tag{4.15}$$

we obtain

$$\begin{aligned}
\frac{dA}{dt} &= -\kappa B, \qquad \frac{dB}{dt} = -\kappa A - \kappa X, \\
\frac{dX}{dt} &= -2i\Delta Y + \kappa B, \qquad \frac{dY}{dt} = -2i\Delta X.
\end{aligned} \tag{4.16}$$

The solutions to these equations vary as e^{Kt} where K assumes the values

$$K = \pm i\sqrt{(2)}\Delta\left\{1 \pm \left(1+\frac{\kappa^2}{\Delta^2}\right)^{1/2}\right\}^{1/2}. \tag{4.17}$$

We shall consider the explicit solutions for the case of large Δ for which

$$K = \pm\kappa, \ \pm i(4\Delta^2+\kappa^2)^{1/2}. \tag{4.18}$$

Here as with the field amplitudes we find one amplified solution with predominantly Stokes character and one attenuated solution with mainly anti-Stokes character. However, even in this large Δ limit correlations between the two field modes are still present.

5. Photon statistics for the coupled Stokes and anti-Stokes fields

We assume that the joint density operator for the Stokes and anti-Stokes modes has a P representation at time t

$$\rho = \int P(\alpha, \beta, t)|\alpha, \beta\rangle\langle\alpha, \beta| \, d^2\alpha \, d^2\beta \tag{5.1}$$

where $|\alpha\rangle$, $|\beta\rangle$ are the eigenstates of b_a and b_s respectively. Substitution of equation (5.1) into the master equation (4.5) and using standard techniques yields the following Fokker–Planck equation:

$$\frac{\partial P}{\partial t} = \left\{\left(-\frac{\kappa}{2}+i\Delta\right)\frac{\partial}{\partial\tilde{\beta}}\tilde{\beta}+cc+\left(\frac{\kappa}{2}+i\Delta\right)\frac{\partial}{\partial\tilde{\alpha}}\tilde{\alpha}+cc+\frac{\kappa}{2}\left(\tilde{\beta}\frac{\partial}{\partial\tilde{\alpha}^*}-\tilde{\alpha}\frac{\partial}{\partial\tilde{\beta}^*}\right)+cc\right.$$
$$\left.+\kappa(\bar{n}_{ph}+1)\frac{\partial^2}{\partial\tilde{\beta}\partial\tilde{\beta}^*}+\kappa\bar{n}_{ph}\frac{\partial^2}{\partial\tilde{\alpha}\partial\tilde{\alpha}^*}-\kappa(\bar{n}_{ph}+\tfrac{1}{2})\frac{\partial^2}{\partial\tilde{\alpha}\partial\tilde{\beta}}+cc\right\}P \tag{5.2}$$

where we have made the transformation of variables

$$\alpha = \tilde{\alpha}\, e^{i\Delta t}, \qquad \beta = \tilde{\beta}\, e^{i\Delta t}. \tag{5.3}$$

The first term in equation (5.2) describes the amplification of the Stokes field, the second the attenuation of the anti-Stokes field and the third term describes the coupling between them. The final three terms describe the diffusion of noise into the system. This noise arises from two sources, one being the intrinsic chaotic nature of the phonon bath. This effect is proportional to the mean number of phonons $\bar{n}_{ph}$ present in the bath. The second source is quantum noise arising from spontaneous emission into the Stokes mode. Though there is no spontaneous emission into the anti-Stokes field, quantum noise filters into the anti-Stokes radiation through the coupling to the Stokes mode.

The Green function solution to equation (5.2) may be derived as follows.

Introducing the real and imaginary parts of α and β

$$\alpha = x_1 + ix_2 \qquad \beta = x_3 + ix_4 \tag{5.4}$$

we obtain the linear real Fokker–Planck equation of the form

$$\frac{\partial P}{\partial t} = -\sum_{ij=1}^{4} m_{ij}\frac{\partial}{\partial x_i}(x_j P)+\frac{1}{2}\sum_{ij=1}^{4} n_{ij}\frac{\partial^2 P}{\partial x_i \partial x_j} \tag{5.5}$$

where

$$[m_{ij}] = \begin{pmatrix} -\frac{1}{2}\kappa & -\Delta & \frac{1}{2}\kappa & 0 \\ \Delta & -\frac{1}{2}\kappa & 0 & \frac{1}{2}\kappa \\ -\frac{1}{2}\kappa & 0 & \frac{1}{2}\kappa & -\Delta \\ 0 & -\frac{1}{2}\kappa & \Delta & \frac{1}{2}\kappa \end{pmatrix} \tag{5.6}$$

and

$$[n_{ij}] = \frac{1}{2}\begin{pmatrix} \kappa\bar{n}_{\rm ph} & 0 & -\kappa(\bar{n}_{\rm ph}+\frac{1}{2}) & 0 \\ 0 & \kappa\bar{n}_{\rm ph} & 0 & \kappa(\bar{n}_{\rm ph}+\frac{1}{2}) \\ -\kappa(\bar{n}_{\rm ph}+\frac{1}{2}) & 0 & \kappa(\bar{n}_{\rm ph}+1) & 0 \\ 0 & \kappa(\bar{n}_{\rm ph}+\frac{1}{2}) & 0 & \kappa(\bar{n}_{\rm ph}+1) \end{pmatrix}. \tag{5.7}$$

Introducing the transformation

$$\tilde{x} = S\tilde{v} \tag{5.8}$$

such that

$$S^{-1}[m_{ij}]S = \mathrm{diag}(\lambda_1, \lambda_2, \lambda_3, \lambda_4) \tag{5.9}$$

where

$$\lambda = \pm \mathrm{i}\Delta\left(1 \pm \frac{\mathrm{i}\kappa}{\Delta}\right)^{1/2} \tag{5.10}$$

equation (5.5) becomes

$$\frac{\partial P}{\partial t} = -\sum_{i=1}^{4} \lambda_i \frac{\partial}{\partial x_i}(x_i P) + \frac{1}{2}\sum_{ij=1}^{4} \gamma_{ij}\frac{\partial^2 P}{\partial x_i \partial x_j} \tag{5.11}$$

where

$$[\gamma_{ij}] = S^{-1}[n_{ij}](S^{-1})^{\mathrm{T}}. \tag{5.12}$$

The solution to equation (5.11) corresponding to the Stokes and anti-Stokes fields initially in coherent states, that is,

$$P(x_i, 0) = \delta(x_i - x_i^0), \tag{5.13}$$

is given by Wang and Uhlenbeck (1945)

$$P(x_i, x_i^0, t) = \frac{1}{\pi^2(\det[\sigma_{ij}(t)])^{1/2}} \exp\left(-\sum_{ij=1}^{4} \sigma_{ij}(t)^{-1}\{x_i - x_i^0 \exp(\lambda_i t)\}\{x_j - x_j^0 \exp(\lambda_j t)\}\right) \tag{5.14}$$

where

$$\sigma_{ij}(t) = \frac{2\gamma_{ij}}{\lambda_i + \lambda_j}[1 - \exp\{-(\lambda_i + \lambda_j)t\}]. \tag{5.15}$$

Thus the joint P function for the Stokes and anti-Stokes fields is given by a four-dimensional gaussian distribution representing a mixture of coherent and chaotic states (Mollow and Glauber 1967). We shall discuss the physical significance of this solution for large Δ corresponding to a large Raman gain with gain coefficients

$$\mathrm{Re}\,\lambda_1 = \mathrm{Re}\,\lambda_2 = -\tfrac{1}{2}\kappa \qquad \mathrm{Re}\,\lambda_3 = \mathrm{Re}\,\lambda_4 = \tfrac{1}{2}\kappa. \tag{5.16}$$

In this case one sees that equation (5.14) describes the coherent amplification of the Stokes field amplitude with mean increasing as $e^{\kappa t/2}$ and corresponding attenuation of the anti-Stokes field amplitude as $e^{-\kappa t/2}$. The anti-Stokes amplitude, however, does not decrease to zero due to its coupling to the Stokes field which drags it along. The variance of the gaussian distribution describes the degree of chaos or noise present in the coupled fields. This noise component is seen to increase with time due to the amplification of the noise diffusing from the phonon bath and the spontaneous emission into the Stokes mode. The noise in an isolated anti-Stokes mode would reach a limit proportional to $\bar{n}_{\mathrm{ph}}$, however, due to the coupling to the Stokes field the chaotic component in the anti-Stokes field also continues to increase with time. The distribution function for the individual Stokes or anti-Stokes modes may be obtained by integrating the joint P distribution over the opposite variable.

6. Conclusions

A fully quantum-mechanical treatment of Raman scattering from a phonon heat bath has been described using the master equation technique. The Hamiltonian formulation successfully describes the stochastic coupling of the Stokes and anti-Stokes fields through the phonon bath. This model reproduces the classical predictions for the coupled field amplitudes. In addition the photon distributions for the field modes are obtained from the solution of a Fokker–Planck equation. Whereas a classical analysis was only applicable to the stimulated Raman effect this treatment contains the spontaneous and stimulated Raman effect in one self-consistent formalism.

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References

Bloembergen N and Shen Y R 1964 *Phys. Rev. Lett.* **12** 504–7
Glauber R J 1963a *Phys. Rev.* **130** 2529–39
—— 1963b *Phys. Rev.* **131** 2766–88
Haken H 1970 *Handb. Phys.* **25** 2c (Berlin: Springer-Verlag)
Haus H A 1970 *Proc IEEE* **58** 1599–611
Hubner H 1970 *Z. Phys.* **239** 103–19
Louisell W H 1969 *Quantum Optics* ed R J Glauber (New York: Academic Press) pp 680–742
Mishkin E A and Walls D F 1969 *Phys. Rev.* **185** 1618–28
Mollow B R and Glauber R J 1967 *Phys. Rev.* **160** 1076–96
Pantell R H and Pathoff H E 1969 *Fundamentals of Quantum Electronics* (New York: Wiley)
Pike E R 1969 *Riv. Nuovo Cim.* **1** 277–314
Risken H, Schmid C H and Weidlich W 1966 *Z. Phys.* **194** 337–59

Scully M O and Lamb W E Jr 1967 *Phys. Rev.* **159** 209–26
Shen Y R and Bloembergen N 1965 *Phys. Rev.* **137** A1787–805
Tucker J and Walls D F 1969 *Phys. Rev.* **178** 2036–43
von Foerster T and Glauber R J 1971 *Phys. Rev.* A **3** 1484–511
Walls D F 1970 *Z. Phys.* **237** 224–33
—— 1971 *Opto Electronics* **3** 57–60
Wang M C and Uhlenbeck G E 1945 *Rev. mod. Phys.* **17** 323–42