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THE QUANTUM THEORY OF DAMPING  
AND THE RAMAN EFFECT

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## ABSTRACT

The basis of this dissertation is a review of the quantum mechanical formulation of damping and stimulation, particularly in non-linear optical processes. The basic problems of the quantum theory of damping are discussed, and the formalism for the quantum theory is introduced. The study of a particular example, that of a damped simple harmonic oscillator, provides an introduction to the handling of the basic tool, the master equation.

The physical aspects of the non-linear processes are contained in the systems' photon statistics, and the master equation provides several approaches for obtaining these. This theory is then used to formulate a quantum mechanical model of Raman scattering by phonons, and to thus obtain the photon statistics of the scattered radiation.

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## INTRODUCTION

We usually try to account for damping in quantum systems phenomenologically, which is not a very successful approach for many systems. By allowing the damping to be described by interaction with a heat bath, we obtain a formalism by which damping can be treated successfully. This method is used widely in the theory of non-linear optical processes, particularly in laser processes. Haken (1970) gives a comprehensive discussion of the method and its applications.

Using a form of perturbation theory, we can derive an equation of motion (the "Master Equation") for the density matrix describing our damped system. By adopting a particular form for the bath, or reservoir, we obtain a working equation. We follow Louisell (1969), who takes the bath to be a large number of simple harmonic oscillators.

The master equation gives us an operator equation for the density matrix. It is convenient to reduce this to a c-number equation, and this may be done in several ways, depending on the information required and on the initial conditions of the system. Once the equation is solved, we have a complete statistical description of the system, since the average of any dynamical variable (e.g. photon number) may be found. This theory may then be applied to the non-linear process of Raman scattering, which is the inelastic scattering of light by a medium.

Until Walls' paper (Walls 1972), the only work on Raman scattering using a master equation type approach was that of Shen (1967) who dealt explicitly with electronic type interactions. A thorough classical description of the stimulated Raman effect has been given by Bloembergen and Shen (1964 and 1965) and this allows the photon-phonon damping to be easily introduced phenomenologically. This is not the case in a quantum approach, and earlier papers (Walls 1970, Mishkin and Walls 1969) accounted for the interaction by considering coupling to a single phonon mode only. This approach is particularly deficient when describing the stokes - antistokes coupling, since it allows coupling through a single mode only. The master equation approach allows interaction with a whole phonon reservoir, and this is much nearer reality. By solving the master equation, the stokes and antistokes fields' statistics, and hence a complete description of the effect, is obtained.

## 1. QUANTUM THEORY BACKGROUND

### 1.1 The Density Matrix:- Quantum Statistical Mechanics

(Dirac 1958)

In general, the processes we study are noisy (due mainly to spontaneous emission). Thus, even though initial conditions may be known exactly, we cannot obtain exact information about the system's state at any later time. For this reason a statistical approach is necessary. We consider an ensemble of a large number of systems similar to the one describing the process of interest, with each system being set up in an identical manner to the others (i.e. all have the same initial conditions). If  $|\psi(t)\rangle$  represent possible pure states of the system, let  $p_\psi$  be the distribution of these states throughout the ensemble. Let  $M$  be some dynamical quantity of interest. In a pure state  $|\psi(t)\rangle$ ,  $M$ 's average value is  $\langle M \rangle = \langle \psi(t) | M | \psi(t) \rangle$ . For the average value of  $M$  with the system in its mixed state, we make the standard hypothesis of statistical mechanics. That is:

$$\langle M \rangle = \text{the } \underline{\text{ensemble}} \text{ average of } \langle \psi(t) | M | \psi(t) \rangle$$

i.e.  $\langle M \rangle = \sum_{\psi} p_{\psi} \langle \psi(t) | M | \psi(t) \rangle$  represents the actual average value of  $M$  in our real system.



This may be written  $\langle M \rangle = \text{Tr} [M \sum_{\psi} \rho_{\psi} |\psi\rangle\langle\psi|]$ .

We then define our density operator by

$$\rho = \sum_{\psi} \rho_{\psi} |\psi\rangle\langle\psi|. \quad (1.1.1.)$$

Then  $\langle M \rangle = \text{Tr} [M \rho]. \quad (1.1.2.)$

## 1.2 Second Quantization (Dirac 1958)

In the light-scattering processes, we describe the light as an electric field, and carry out second quantization. We consider a free electromagnetic field, in a cavity, so that eigenvalue expansions may be used. The fields  $\underline{E}$  and  $\underline{B}$  may be described by the magnetic vector potential  $\underline{A}$  and the scalar potential  $\phi$ . In the absence of free charges, it is convenient to work in the coulomb gauge, in which  $\nabla \cdot \underline{A} = 0$ , and

$$\underline{E}(\underline{r}, t) = -\frac{1}{c} \frac{\partial \underline{A}(\underline{r}, t)}{\partial t}, \quad \underline{B}(\underline{r}, t) = \nabla \times \underline{A}(\underline{r}, t). \quad (1.2.1.)$$

The Maxwell's equations then yield

$$\square \underline{A}(\underline{r}, t) = 0 \quad (1.2.2.)$$

Sturm-Liouville eigenfunction theory shows that in the cavity, a general solution of (1.2.2.) is

$$\underline{A}(\underline{r}, t) = \sum_{\underline{k}} b_{\underline{k}} \underline{u}_{\underline{k}}(\underline{r}) e^{-i\omega_{\underline{k}} t} + \text{complex conjugate.}$$

where  $\omega_{\underline{k}} = \frac{ck}{\sqrt{\epsilon_{\underline{k}}}} = \frac{c}{\sqrt{\epsilon_{\underline{k}}}} \left( \frac{2\pi}{V^{1/3}} \right) (n_1^2 + n_2^2 + n_3^2)$ ,  $n_1, n_2, n_3 \in \mathbb{Z}^+$

The  $\underline{u}_{\underline{k}}(\underline{r})$  are orthogonal w.r.t. the weighting function

$$\frac{\epsilon_{\underline{k}} \omega_{\underline{k}}^2}{c^2}, \quad \text{or, since } \omega_{\underline{k}} \text{ and } c \text{ are constants, } (\epsilon_{\underline{k}} \text{ may vary})$$

spatially and/or temporally)

Possible mode functions  $u_{\underline{k}}(\underline{r})$  are  $u_{\underline{k}}(\underline{r}) = \frac{e_{\underline{k}}}{\sqrt{V\epsilon_{\underline{k}}}} e^{i\underline{k}\cdot\underline{r}}$ .

where  $e_{\underline{k}}$  is a unit polarization vector.

Redefining a few constants, we may write

$$A(\underline{r}, t) = \sum_{\underline{k}} \sqrt{\frac{\hbar c^2}{2\omega_{\underline{k}}}} \{ a_{\underline{k}} u_{\underline{k}}(\underline{r}) e^{-i\omega_{\underline{k}}t} + a_{\underline{k}}^* u_{\underline{k}}^*(\underline{r}) e^{i\omega_{\underline{k}}t} \} \quad (1.2.3.)$$

Second quantization involves the canonical quantization of  $A$ . This means  $a_{\underline{k}}$  and  $a_{\underline{k}}^*$  become operators  $a_{\underline{k}}, a_{\underline{k}}^\dagger$  and the canonical commutation rules for  $A$  give the following commutation rules:

$$[a_{\underline{k}}, a_{\underline{k}'}] = [a_{\underline{k}}^\dagger, a_{\underline{k}'}^\dagger] = 0, \quad [a_{\underline{k}}, a_{\underline{k}'}^\dagger] = \delta_{\underline{k}\underline{k}'} \quad (1.2.4.)$$

These (boson) commutation rules imply  $a_{\underline{k}}$  may be interpreted as a "destruction" operator, and  $a_{\underline{k}}^\dagger$  as a "creation" operator. If we let  $n_{\underline{k}}$  be the number of quanta in the mode of the field described by  $u_{\underline{k}}(\underline{r}) e^{-i\omega_{\underline{k}}t}$ , then a basis of pure states of the field is of the form  $\{ |n_1, n_2, \dots, n_{\underline{k}}, \dots\rangle \}$

$$a_{\underline{k}} |n_1, n_2, \dots, n_{\underline{k}}, \dots\rangle = \sqrt{n_{\underline{k}}} |n_1, n_2, \dots, n_{\underline{k}}-1, \dots\rangle. \quad (1.2.5.)$$

$$a_{\underline{k}}^\dagger |n_1, n_2, \dots, n_{\underline{k}}, \dots\rangle = \sqrt{n_{\underline{k}}+1} |n_1, n_2, \dots, n_{\underline{k}}+1, \dots\rangle. \quad (1.2.6.)$$

The vacuum state  $|0\rangle$  of a mode is defined by  $a_{\underline{k}}|0\rangle = 0$ .

From the Lagrangian formulation, we may obtain the free field Hamiltonian:

$$H = \frac{1}{2} \int_V (E^2 + B^2) d^3r = \sum_{\underline{k}} \hbar \omega_{\underline{k}} (a_{\underline{k}}^\dagger a_{\underline{k}} + \frac{1}{2}) \quad (1.2.7.)$$

In the second quantized form, we have the field as follows:

$$\underline{E}(\underline{r}, t) = i \sum_{\underline{k}} \sqrt{\frac{\hbar}{2\omega_{\underline{k}}}} \{ a_{\underline{k}} u_{\underline{k}}(\underline{r}) e^{-i\omega_{\underline{k}}t} - a_{\underline{k}}^\dagger u_{\underline{k}}^*(\underline{r}) e^{i\omega_{\underline{k}}t} \}. \quad (1.2.8.)$$

Note: If we define canonical co-ordinates and momenta

$q_k, p_k$  by

$$\begin{aligned} a_k &= \frac{1}{\sqrt{2\hbar\omega_k}} (\omega_k q_k + i p_k) \\ a_k^\dagger &= \frac{1}{\sqrt{2\hbar\omega_k}} (\omega_k q_k - i p_k). \end{aligned} \quad (1.2.9.)$$

then the Hamiltonian for a single mode,  $H = \hbar\omega_k (a_k^\dagger a_k + \frac{1}{2})$  becomes  $H = \frac{1}{2}(\omega_k^2 q_k^2 + p_k^2)$ .

which is a simple harmonic oscillator Hamilton. Thus in the second quantization, a field mode is formally equivalent to a simple harmonic oscillator.

### 1.3 Coherent States of the Field (Glauber 1963)

We may define  $n$ th order correlation functions for the electric field:

$$G^{(n)}(x_1, \dots, x_{2n}) = \text{Tr} [\rho E^-(x_1) \dots E^-(x_n) E^+(x_{n+1}) \dots E^+(x_{2n})] \quad (1.3.1.)$$

where:  $E^-$  and  $E^+$  are the -ve and +ve frequency components respectively of (1.2.8.), and  $x_i = (r_i, t_i)$ .

By a simple analogy with classical optics, we may say that the field exhibits  $n$ th order coherence if all correlation functions up to the  $n$ th factorize. i.e. if

$$G^{(n)}(x_1, \dots, x_{2n}) = \prod_{j=1}^n G^{(1)}(x_{2j-1}, x_{2j}) \quad i = 1, 2, \dots, n.$$

There exist certain pure states in which the field exhibits coherence to all orders. These states are known

as coherent states of the field. For a single mode, they are written  $|\alpha\rangle$  and  $a|\alpha\rangle = \alpha|\alpha\rangle$ . For a number of modes, they are written  $|\alpha_1, \alpha_2, \dots, \alpha_k, \dots\rangle$  and

$$a_k |\alpha_1, \alpha_2, \dots, \alpha_k, \dots\rangle = \alpha_k |\alpha_1, \alpha_2, \dots, \alpha_k, \dots\rangle. \quad (1.3.2.)$$

$$\alpha_i \in \mathbb{C}, \quad i=1,2,\dots$$

In terms of number states  $|n\rangle$

$$|\alpha\rangle = e^{-|\alpha|^2/2} \sum_{n=0}^{\infty} \frac{\alpha^n}{\sqrt{n!}} |n\rangle. \quad (1.3.3.)$$

Using  $\langle m|n\rangle = \delta_{mn}$  we have

$$|\langle \beta|\alpha\rangle|^2 = e^{-|\alpha-\beta|^2} \quad (1.3.4.)$$

so that the coherent states do not form an orthogonal set. However, using the completeness relation  $\sum_{n=0}^{\infty} |n\rangle\langle n| = 1$  for the number states, we can show

$$\int \frac{d^2\alpha}{\pi} |\alpha\rangle\langle\alpha| = 1 \quad (1.3.5.)$$

where  $d^2\alpha = d(\text{Re}\alpha)d(\text{Im}\alpha)$ . so that the coherent states do form a complete set. (In fact the set is over-complete.) This makes the coherent states extremely useful.

NOTE: As defined, the coherent states are in the Heisenberg picture. In the Schrödinger picture they are

$$|\alpha, t\rangle = |\alpha e^{-i\omega(t-t_0)}\rangle. \quad (1.3.6.)$$

where  $\omega$  is the  $\omega_k$  of the mode in question.

#### 1.4 Damping and Stimulation (Louisell 1964)

Consider the Heisenberg equations of motion for for a free field in a cavity:

$$\frac{da_k}{dt} = \frac{1}{i\hbar} [a_k, H] = -i\omega_k a_k$$

$$\frac{da_k^\dagger}{dt} = \frac{1}{i\hbar} [a_k^\dagger, H] = i\omega_k a_k^\dagger$$

Thus  $a_k(t) = a_k e^{-i\omega_k t}$  and  $a_k^\dagger(t) = a_k^\dagger e^{i\omega_k t}$  where  $a_k = a_k(0)$  is the Schrödinger picture operator.

Thus  $[a_k(t), a_k^\dagger(t)] = [a_k, a_k^\dagger] = 1$ , which does not violate the uncertainty principle for canonically conjugate operators.

In order to describe any damping of the mode, we might attempt to add phenomenological damping terms, guided by classical damping:

$$\frac{da_k}{dt} = -i\omega_k a_k - \frac{\gamma}{2} a_k$$

$$\frac{da_k^\dagger}{dt} = i\omega_k a_k^\dagger - \frac{\gamma}{2} a_k^\dagger$$

This gives  $a_k(t) = a_k e^{-i\omega_k t - \frac{\gamma}{2} t}$  and  $a_k^\dagger(t) = a_k^\dagger e^{i\omega_k t - \frac{\gamma}{2} t}$

Then  $[a_k(t), a_k^\dagger(t)] = e^{-\gamma t}$  This violates the uncertainty principle since for large  $t$ ,  $e^{-\gamma t} \rightarrow 0$

This is because the phenomenological terms account only for the action of the mode  $a_k$  on the loss, but not vice-versa. For  $t \leq \frac{1}{\gamma} = \text{mode "lifetime"}$ , only the action of the mode on the loss is important, and the above method is suitable, since  $e^{-\gamma t}$  is not close to zero. For  $t \gg \frac{1}{\gamma}$  the mode has certainly acted on the loss, which now acts back on the mode. For a self-consistent theory of damping we must look further than the phenomenological approach.

## 2. THE QUANTUM THEORY OF DAMPING

### 2.1. The Damping Mechanism (Louisell 1969)

The loss mechanism for a system (optical field modes in our case) may be represented by a heat bath, or reservoir, with which the system may interact. We enclose the system and reservoir in a cavity, so that the normal mode decomposition may be used.

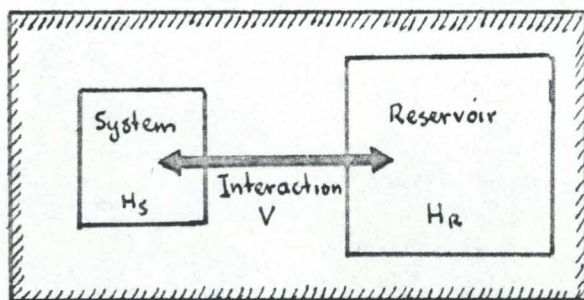


Fig.1

The complete Hamiltonian may be written as

$$H = H_S + H_R + V \quad (2.1.1.)$$

where  $H_S$  = free Hamiltonian for system

$H_R$  = free Hamiltonian for reservoir

$V$  = system-reservoir interaction Hamiltonian.

### 2.2 Density Matrix Equation of Motion (Dirac 1958)

Let  $|\psi(t)\rangle$  be pure states of the complete system plus reservoir.

Then the density matrix is  $\rho_{S,R} = \sum_{\psi} p_{\psi} |\psi\rangle\langle\psi|$ .

In the ensemble, the  $\rho_\psi$  are time independent, hence

$$i\hbar \frac{\partial \rho_{s,r}}{\partial t} = \sum_{\psi} \rho_{\psi} [i\hbar \frac{\partial |\psi\rangle}{\partial t} \langle \psi| + |\psi\rangle i\hbar \frac{\partial \langle \psi|}{\partial t}] .$$

Now the  $|\psi\rangle$  are in the Schrödinger picture, so we have

$$i\hbar \frac{\partial |\psi\rangle}{\partial t} = H|\psi\rangle \quad \text{with the adjoint relation:-}$$

$$-i\hbar \frac{\partial \langle \psi|}{\partial t} = \langle \psi| H \quad \text{using} \quad H^\dagger = H$$

Hence we obtain the equation of motion for  $\rho_{s,r}$ .

$$i\hbar \frac{\partial \rho_{s,r}}{\partial t} = [H, \rho_{s,r}] . \quad (2.2.1.)$$

### 2.3 The Reduced Density Operator. The Master Equation

(Louisell 1969)

$\rho_{s,r}$  contains information about the system and the reservoir. Clearly, the reservoir information is unwanted information as far as we are concerned, since:

In general we are interested in system operators  $M_s(t)$  only:

$$\begin{aligned} \langle M_s(t) \rangle &= \text{Tr}_{s,r} [M_s(t) \rho_{s,r}(t)] . \\ &= \text{Tr}_s [M_s(t) \text{Tr}_r \rho_{s,r}(t)] . \end{aligned}$$

Define the reduced density operator for the system by

$$\rho_s(t) = \text{Tr}_r [\rho_{s,r}(t)] . \quad (2.3.1.)$$

$$\text{Then} \quad \langle M_s(t) \rangle = \text{Tr}_s [M_s(t) \rho_s(t)] . \quad (2.3.2.)$$

We proceed to use (2.2.1.) to obtain an equation for  $\rho_s(t)$ . As a first step, we transform to the interaction picture, (see e.g. Merzbacher, 1961) which removes high frequency motion due to the free Hamiltonians:

Define the unitary operator  $U(t) = \exp\left[\frac{1}{i\hbar}(H_S + H_R)t\right]$

Let  $\chi(t) = U^{-1}(t) \rho_{S,R}(t) U(t)$ .

Then  $\rho_{S,R}(t) = U(t) \chi(t) U^{-1}(t)$ . (2.3.3.)

Substituting (2.3.3.) into (2.2.1.) gives the following equation of motion for  $\chi(t)$ .

$$i\hbar \frac{\partial \chi(t)}{\partial t} = [V(t), \chi(t)]. \quad (2.3.4.)$$

where  $V(t) = U^{-1}(t) V U(t)$  is the interaction Hamiltonian in the interaction picture.

We transform (2.3.4.) to an integral equation:

$$\chi(t) = \frac{1}{i\hbar} \int_0^t [V(t'), \chi(t')] dt' + \chi(0).$$

and proceed as in normal perturbation theory by iteration.

Two iterations yield:

$$\begin{aligned} \chi(t) = \chi(0) &+ \frac{1}{i\hbar} \int_0^t [V(t'), \chi(0)] dt' \\ &+ \frac{1}{(i\hbar)^2} \int_0^t \int_0^{t''} [V(t'), [V(t''), \chi(t'')]] dt'' dt' \end{aligned} \quad (2.3.5.)$$

We could keep on iterating, but this standard technique does not converge sufficiently quickly to yield exponential behaviour. Instead we use a different technique.

Differentiating (2.3.5.) gives:

$$\frac{\partial \chi(t)}{\partial t} = \frac{1}{i\hbar} [V(t), \chi(0)] + \frac{1}{(i\hbar)^2} \int_0^t [V(t), [V(t'), \chi(t')]] dt' \quad (2.3.6.)$$

To remove the reservoir information, we now trace (2.3.6.) over the reservoir:

$$\begin{aligned} \frac{\partial S(t)}{\partial t} &= \frac{1}{i\hbar} \text{Tr}_R [V(t), \chi(0)] \\ &+ \frac{1}{(i\hbar)^2} \int_0^t \text{Tr}_R [V(t), [V(t'), \chi(t')]] dt' \end{aligned}$$

where  $S(t) = \text{Tr}_R \chi(t) = \text{Tr}_R [U^{-1}(t) \rho_{S,R}(t) U(t)]$ . (2.3.7.)  
 $= U^{-1}(t) \rho_S(t) U(t)$ .



We assume the system-reservoir interaction is turned on at  $t = 0$ . Then  $\chi(0) = \rho_{S,R}(0) = \rho_S(0) f_0(H_R) = S(0) f_0(H_R)$ . i.e. because there is no interaction at  $t = 0$ , the operator factorizes. We have taken the reservoir to be at thermal equilibrium at  $t = 0$ , so the reservoir density operator at  $t = 0$  is

$$f_0(H_R) = \exp(-\frac{H_R}{kT}) / \text{Tr}_R [\exp(-\frac{H_R}{kT})] \quad (2.3.8.)$$

which is the normalized Boltzmann distribution.

We choose a representation in which  $H_R$  is diagonal, and put any diagonal elements of  $V(t)$  into  $H_S + H_R$ , so that  $V(t)$  has only zeros on the diagonals. Then

$$\text{Tr}_R [V(t), \chi(0)] = \text{Tr}_R [V(t), S(0) f_0(H_R)] = 0.$$

We now assume that  $V$  is so small, and the reservoir so large, that the reservoir remains in thermal equilibrium. Then the reservoir is always described by  $f_0(H_R)$ .

If  $V = 0$ , we would have  $\chi(t) = S(t) f_0(H_R)$ . However,  $V \neq 0$ .

But  $V \ll H_S$  and  $H_R$ , hence we may write

$$\chi(t) = S(t) f_0(H_R) + \Delta\chi(t). \quad (2.3.9.)$$

where  $\Delta\chi(t) = O(V)$ .

$$\text{Now } S(t) = \text{Tr}_R \chi(t), \therefore (2.3.9) \Rightarrow \text{Tr}_R \Delta\chi(t) = 0. \quad (2.3.10)$$

Using the above, equation (2.3.6.) becomes:

$$\frac{\partial S(t)}{\partial t} \approx \frac{1}{(i\hbar)^2} \int_0^t \text{Tr}_R [V(t'), [V(t'), S(t') f_0(H_R) + \Delta\chi(t')]] dt'$$

Retaining only those terms up to  $O(V^2)$  we obtain:

$$\frac{\partial S(t)}{\partial t} \approx \frac{1}{(i\hbar)^2} \int_0^t \text{Tr}_R [V(t'), [V(t'), S(t') f_0(H_R)]] dt' \quad (2.3.11)$$

(2.3.11) is an integro-differential equation for  $S(t)$ ,

showing that  $\frac{\partial S}{\partial t}$  at present (and hence  $S$  in the future)

depends on the whole of the past history of  $S$ . ( $S$  appears

integrated from 0 to t on the R.H.S. of (2.3.11)).

In practice, the reservoir correlation times are such that the system very rapidly loses all knowledge of its past; i.e. the damping of the system is Markovian. Hence we may use  $S(t)$  in the R.H.S., there being no contribution for  $t' < t$ .

$$\frac{\delta S(t)}{\delta t} = \frac{1}{(i\hbar)^2} \int_0^t \text{Tr}_R [V(t), [V(t'), S(t)] f_0(H_R)] dt' \quad (2.3.12)$$

This is the "Master equation" for  $\rho_s(t)$  in the interaction picture, and is valid to  $O(V^2)$ .

#### 2.4 Example: A Damped Simple Harmonic Oscillator

We take the bath to be a large assembly of simple harmonic oscillators with which a system comprising a single harmonic oscillator interacts. (This form of interaction is well known classically.)

Using the standard boson operators for simple harmonic oscillators, we may write the Hamiltonians:

$$\begin{aligned} H_s &= \hbar\omega_0 (a^\dagger a + \frac{1}{2}). \\ H_R &= \sum_j \hbar\omega_j (b_j^\dagger b_j + \frac{1}{2}). \\ V &= \sum_j \hbar K_j a^\dagger b_j + \text{h.c.} \end{aligned} \quad (2.4.1.)$$

$\omega_0$  is the resonant frequency of the system oscillator  
 $\omega_j$  is the frequency of the jth bath oscillator.  $V$  is a phenomenological interaction Hamiltonian. Its physical applicability can be seen as follows:

The term  $a^\dagger b_j$  represents the destruction of a bath quantum with a system quantum being created simultaneously. The terms in the hermitian conjugate (which must be

included in order that  $V$  be hermitian) represent reverse processes.  $K_j$  is a coupling constant ( $K_j \ll \omega_j$ ) which measures the coupling between the system oscillator and the  $j$ th bath oscillator. We expect that  $K_j$  is appreciably different from zero only for  $j$  s.t.  $\omega_j \approx \omega_0$ .

In the interaction picture, we have:

$$V(t) = \sum_j \hbar K_j U^{-1}(t) a^\dagger b_j U(t) + \sum_j \hbar K_j^* U^{-1}(t) a b_j^\dagger U(t)$$

where  $U(t) = \exp[-it(\omega_0 a^\dagger a + \sum_j \omega_j b_j^\dagger b_j)]$ .

Rearrangement yields terms such as  $e^{i\omega_0 a^\dagger t} a^\dagger e^{-i\omega_0 a^\dagger t}$ .

Having earlier solved the Heisenberg equations of motion for a free field mode we can readily evaluate such terms. Recalling section (1.4), we see that for  $H_0 =$

$\hbar\omega_k (a_k^\dagger a_k + \frac{1}{2})$ ,  $a_k^\dagger(t) = a_k^\dagger e^{i\omega_k t}$  in the Heisenberg picture.  $a_k^\dagger$  is the Schrödinger picture operator  $a_k^\dagger(0)$ .

But we know also that any Heisenberg operator  $A_H(t)$  is connected with its Schrödinger picture counterpart  $A_S$  by

$$A_H(t) = \exp[iH_0 t/\hbar] A_S \exp[-iH_0 t/\hbar] = e^{i\omega_k a^\dagger a t} A_S e^{-i\omega_k a^\dagger a t}$$

Hence  $a_k^\dagger(t) = e^{i\omega_k a^\dagger a t} a_k^\dagger e^{-i\omega_k a^\dagger a t} = a_k^\dagger e^{i\omega_k t}$

We may thus write

$$V(t) = \hbar g(t) a^\dagger + \hbar g^\dagger(t) a \quad (2.4.2.)$$

where  $g(t) = \sum_j K_j e^{-i(\omega_j - \omega_0)t} b_j$

Upon inserting  $V(t)$  into the master equation (2.3.12) we find we have to evaluate terms such as

$$\text{Tr}_R [b_e b_m f_0(H_R)] = \langle b_e b_m \rangle_R,$$

$$\text{Tr}_R [b_e^\dagger b_m f_0(H_R)] = \langle b_e^\dagger b_m \rangle_R,$$

etc, on the R.H.S.

These may be easily evaluated, using the following:

In the number state representation, if A is a reservoir operator, 
$$\text{Tr}_R [A] = \sum_{n_1=0}^{\infty} \sum_{n_2=0}^{\infty} \dots \langle n_1, n_2, \dots | A | n_1, n_2, \dots \rangle .$$

$b_e b_m, b_e^+ b_m$  etc. are reservoir operators, hence this is applicable. We find:

$$(i) \quad \langle b_e b_m \rangle_R = 0 \quad (ii) \quad \langle b_e^+ b_m^+ \rangle_R = 0 \quad (2.4.3.)$$

$$(iii) \quad \langle b_e^+ b_m \rangle_R = \delta_{em} \bar{n}_e \quad (iv) \quad \langle b_e b_m^+ \rangle_R = \delta_{em} (1 + \bar{n}_e)$$

$$\text{where } \bar{n}_e = (e^{\hbar \omega_e / kT} - 1)^{-1} .$$

As an example, we evaluate the 4th term of the R.H.S. of the Master equation:

$$\begin{aligned} \text{This is } & \frac{1}{i\hbar} \int_0^t \text{Tr}_R \{ S(t) f_0(H_R) V(t') V(t) \} dt' \\ & = -S(t) \int_0^t \text{Tr}_R \{ f_0(H_R) [g(t') a^+ + g^+(t') a] [g(t) a^+ + g^+(t) a] \} dt' \\ & = -S(t) a^+ a^+ \int_0^t \text{Tr}_R \{ f_0(H_R) g(t') g(t) \} dt' - S(t) a^+ a \int_0^t \text{Tr}_R \{ f_0(H_R) g(t') g^+(t) \} dt' \\ & \quad - S(t) a a^+ \int_0^t \text{Tr}_R \{ f_0(H_R) g^+(t') g(t) \} dt' - S(t) a a \int_0^t \text{Tr}_R \{ f_0(H_R) g^+(t') g^+(t) \} dt' \end{aligned}$$

Using the results (2.4.3.) and definitions (2.4.2.) we

see

$$\begin{aligned} \text{Tr}_R \{ f_0(H_R) g(t') g(t) \} & = \langle g(t') g(t) \rangle_R = 0 \\ \text{Tr}_R \{ f_0(H_R) g^+(t') g^+(t) \} & = \langle g^+(t') g^+(t) \rangle_R = 0 \\ \text{Tr}_R \{ f_0(H_R) g(t') g^+(t) \} & = \langle g(t') g^+(t) \rangle_R = \sum_j |K_j|^2 (1 + \bar{n}_j) e^{-i(\omega_j - \omega_0)(t' - t)} \\ \text{Tr}_R \{ f_0(H_R) g^+(t') g(t) \} & = \langle g^+(t') g(t) \rangle_R = \sum_j |K_j|^2 \bar{n}_j e^{i(\omega_j - \omega_0)(t' - t)} \end{aligned}$$

To evaluate the sums, we make two assumptions. The first

is that the reservoir modes are sufficiently close

together in  $\omega$  space that the sum over  $\omega_j$  may be replaced

by an integral over  $\omega$ : i.e.  $\sum_j f(\omega_j) \rightarrow \int_{-\infty}^{\infty} f(\omega) g(\omega) d\omega$

where  $f$  is some function of the  $\omega_j$ 's and  $g(\omega)$  is the  $\omega$ -space

density of reservoir modes.

$$\text{Thus } \sum_j |K_j|^2 (1 + \bar{n}_j) e^{-i(\omega_j - \omega_0)(t' - t)} \approx \int_{-\infty}^{\infty} |K(\omega)|^2 [1 + \bar{n}(\omega)] g(\omega) e^{-i(\omega - \omega_0)u} d\omega$$

$$\text{and } \sum_j |K_j|^2 \bar{n}_j e^{i(\omega_j - \omega_0)(t' - t)} \approx \int_{-\infty}^{\infty} |K(\omega)|^2 \bar{n}(\omega) g(\omega) e^{i(\omega - \omega_0)u} d\omega$$

where  $u = t' - t$

We note that  $e^{i(\omega-\omega_0)u}$  oscillates rapidly except near  $\omega = \omega_0$ , so only those values of  $\omega$  near  $\omega_0$  contribute to the integrals. We thus make a second assumption, which is that  $|K(\omega)|^2 \bar{n}(\omega) g(\omega)$  and  $|K(\omega)|^2 (1+\bar{n}(\omega)) g(\omega)$  are slowly varying functions of  $\omega$  near  $\omega = \omega_0$ .

$$\text{Then } \int_{-\infty}^{\infty} |K(\omega)|^2 (1+\bar{n}(\omega)) g(\omega) e^{-i(\omega-\omega_0)u} d\omega \approx |K(\omega_0)|^2 (1+\bar{n}(\omega_0)) g(\omega_0) \int_{-\infty}^{\infty} e^{-i(\omega-\omega_0)u} d\omega$$

$$\text{and } \int_{-\infty}^{\infty} |K(\omega)|^2 \bar{n}(\omega) g(\omega) e^{i(\omega-\omega_0)u} d\omega \approx |K(\omega_0)|^2 \bar{n}(\omega_0) g(\omega_0) \int_{-\infty}^{\infty} e^{i(\omega-\omega_0)u} d\omega$$

From the theory of distribution, it is well known that

$$\frac{1}{2\pi} \int_{-\infty}^{\infty} e^{\pm i\omega' u} d\omega' \quad \text{is a representation of } \delta(u), \text{ the}$$

Dirac delta function.

$$\text{Since } \omega_0 \text{ is a constant, } \int_{-\infty}^{\infty} e^{\pm i(\omega-\omega_0)u} d\omega = \int_{-\infty}^{\infty} e^{\pm i\omega' u} d\omega' = 2\pi \delta(u).$$

$$\text{If we write } \gamma = 2\pi |K(\omega_0)|^2 g(\omega_0)$$

$$(2.4.4.)$$

$$\text{and } \bar{n} = \bar{n}(\omega_0)$$

the 4th term in the master equation becomes

$$-S(t) a^\dagger a \gamma (1+\bar{n}) \int_0^t \delta(u) dt' - S(t) a a^\dagger \gamma \bar{n} \int_0^t \delta(u) dt'$$

$$= -\frac{\gamma}{2} (1+\bar{n}) S(t) a^\dagger a - \frac{\gamma}{2} \bar{n} S(t) a a^\dagger \quad (2.4.5.)$$

$$\text{since } \int_0^t \delta(u) dt' = \frac{1}{2}.$$

The other terms in the master equation can be obtained in an analogous manner, yielding the following interaction - picture master equation for  $\rho_s$ , the oscillator's reduced density matrix:

$$\frac{\partial S(t)}{\partial t} = \frac{\gamma}{2} (2a s a^\dagger - a^\dagger a s - s a^\dagger a) + \gamma \bar{n} (a^\dagger s a - a s a^\dagger - a^\dagger a s - s a a^\dagger).$$

or, in more compact notation,

$$\frac{\partial S(t)}{\partial t} = \frac{\gamma}{2} ([a s, a^\dagger] + [a, s a^\dagger]) + \gamma \bar{n} [[a, s], a^\dagger]. \quad (2.4.6.)$$

We can convert this result to the Schrödinger picture by

$$\text{using } S(t) = U^{-1}(t) \rho_s(t) U(t), \quad a = U^{-1}(t) a(t) U(t) \quad \text{etc., along}$$

$$\text{with } U^{-1} U = U U^{-1} = 1.$$

$$\begin{aligned}
 S(t) &= \exp\left[\frac{i}{\hbar}(H_S + H_R)t\right] \rho_S(t) \exp\left[-\frac{i}{\hbar}(H_S + H_R)t\right] \\
 &= \exp\{i\omega_0 a^\dagger a t\} \rho_S(t) \exp(-i\omega_0 a^\dagger a t)
 \end{aligned}$$

$$\therefore \frac{\partial \rho_S}{\partial t} = U^{-1} \left\{ i\omega_0 [a^\dagger a, \rho_S(t)] + \frac{\partial \rho_S(t)}{\partial t} \right\} U.$$

Using all this in (2.4.6.) gives the following Schrödinger picture master equation:

$$\frac{\partial \rho_S(t)}{\partial t} = -i\omega_0 [a^\dagger a, \rho_S] + \frac{\gamma}{2} ([a, \rho_S a] + [a, \rho_S a^\dagger]) + \gamma \bar{n} [[a, \rho_S], a^\dagger]. \quad (2.4.7.)$$

This example may be used to describe the damping of an electric field mode in a cavity due to interaction with all other field modes, since a quantized field mode is analogous to a simple harmonic oscillator. The above equations may be obtained from the familiar interaction Hamiltonian:

$$H = \int_V \underline{E} \cdot \underline{P} \, d^3r = \int_V \epsilon \underline{E}^2 \, d^3r.$$

$\underline{P}$  the polarization is  $\underline{P} = \epsilon \underline{E}$ .

This S.H.O. example extends quite generally to other cases. We often find that the interaction mechanism can be decomposed into a large number of normal modes. These are then quantized, giving a reservoir of oscillators as in the above example. This is the case for the Raman scattering, which is discussed in Chapter 4.

### 3. PHOTON STATISTICS

As mentioned earlier, the electric field may be described by either of two complete sets, the number states  $\{|n\rangle\}$ , or the coherent states  $\{|\alpha\rangle\}$ . In evaluating the photon statistics, that is, the field's reduced density matrix, of our light field system, we may make use of either description. The initial conditions dictate which description is more convenient.

#### 3.1 The P-Representation (Glauber 1963)

Even though the coherent states are not orthogonal they are over-complete, so they form a possible representation for states and operators of the field; e.g. we may write any field operator  $A$  as  $A = \frac{1}{\pi^2} \iint \langle \alpha | A | \beta \rangle |\alpha\rangle \langle \beta| d^2\alpha d^2\beta$ . If the operator is diagonal in this representation,

$$A = \frac{1}{\pi} \int a(\alpha) |\alpha\rangle \langle \alpha| d^2\alpha, \quad a(\alpha) = \langle \alpha | A | \alpha \rangle.$$

We assume the system's density matrix may be written in this form. i.e.

$$\rho(t) = \int P(\alpha, t) |\alpha\rangle \langle \alpha| d^2\alpha. \quad (3.1.1.)$$

say (for a single field mode).

This is the "P-representation" for  $\rho$ , and  $P(\alpha, t)$  is the P-function which may be time dependent. Not all systems will possess a P-representation. The P-function is useful because it has some of the properties of a classical probability distribution:-

Consider the (ensemble) average of a normally ordered operator  $M(t)$  :-

(a normally ordered operator is one in which the commutation relations have been used to place all creation operators to the left of annihilation operators in each term. This eliminates vacuum fluctuations)

$$\begin{aligned} \langle M(t) \rangle &= \text{Tr}(\rho M) = \text{Tr} \left( \int P(\alpha, t) |\alpha\rangle \langle \alpha| M(t) d^2\alpha \right) \\ &= \int m(\alpha) P(\alpha, t) d^2\alpha. \end{aligned} \quad (3.1.2.)$$

where  $m(\alpha) = \langle \alpha | M(a^\dagger, a) | \alpha \rangle = M(\alpha^*, \alpha)$  - that is, the operators  $a, a^\dagger$  in  $M$  are replaced by the numbers  $\alpha, \alpha^*$ . (3.1.2.) is rather like a classical averaging procedure. (Note that the Hermiticity of  $\rho$  implies  $P$  must be real.) However, since  $P$  may take negative values, and the  $\{|\alpha\rangle\}$  are not an orthogonal set,  $P$  is not a true probability function.

To describe systems of more than one mode, the definition (3.1.1.) is easily extended:

$$\rho(t) = \int P(\alpha_1, \alpha_2, \dots, t) |\alpha_1, \alpha_2, \dots\rangle \langle \alpha_1, \alpha_2, \dots| d^2\alpha_1 d^2\alpha_2 \dots$$

This gives the joint density operator for the system, and

$P(\alpha_1, \alpha_2, \dots, t)$  is the joint P function. To obtain the P function for a smaller number of modes we sum out  $P$  over the modes not required. e.g. for a two mode system,

$$P(\alpha_1, t) = \int P(\alpha_1, \alpha_2, t) d^2\alpha_2 \quad \text{is the P function for mode one.}$$

A Fokker-Planck Equation for P:

If we substitute  $\rho(t) = \int P(\{\alpha\}, t) |\{\alpha\}\rangle \langle \{\alpha\}| \prod d^2\alpha_i$  into our master equation, we obtain a Fokker-Planck type equation for  $P(\{\alpha\})$ , analogous to Fokker-Planck equations for classical damped and driven systems.

e.g. In the damped simple harmonic oscillator case, we have a single mode, hence  $\rho(t) = \int P(\alpha, t) |\alpha\rangle \langle \alpha| d^2\alpha$  is used.

We use the following results (see Appendix)



$$\begin{aligned}
\int P(\alpha) a^\dagger a |\alpha\rangle\langle\alpha| d^2\alpha &= \int |\alpha\rangle\langle\alpha| (\alpha^* \alpha - \frac{\partial}{\partial \alpha} \alpha) P(\alpha) d^2\alpha. \\
\int P(\alpha) |\alpha\rangle\langle\alpha| a^\dagger a d^2\alpha &= \int |\alpha\rangle\langle\alpha| (\alpha^* \alpha - \frac{\partial}{\partial \alpha^*} \alpha^*) P(\alpha) d^2\alpha \\
\int P(\alpha) a |\alpha\rangle\langle\alpha| a^\dagger d^2\alpha &= \int |\alpha\rangle\langle\alpha| (\alpha^* \alpha) P(\alpha) d^2\alpha \quad (3.1.3.) \\
\int P(\alpha) a^\dagger |\alpha\rangle\langle\alpha| a d^2\alpha &= \int |\alpha\rangle\langle\alpha| (1 + \alpha^* \alpha - \frac{\partial}{\partial \alpha} \alpha - \frac{\partial}{\partial \alpha^*} \alpha^* + \frac{\partial^2}{\partial \alpha \partial \alpha^*}) P(\alpha) d^2\alpha.
\end{aligned}$$

Any others follow from the  $a, a^\dagger$  commutations.

Inserting the P-representation for  $\rho$  into the Schrödinger picture master equation (2.4.7.) and using (3.1.3.), we

obtain:

$$\int |\alpha\rangle\langle\alpha| \left\{ \frac{\partial}{\partial t} - \left(\frac{\gamma}{2} + i\omega_0\right) \frac{\partial}{\partial \alpha} \alpha - \left(\frac{\gamma}{2} - i\omega_0\right) \frac{\partial}{\partial \alpha^*} \alpha^* - \gamma \bar{n} \frac{\partial^2}{\partial \alpha \partial \alpha^*} \right\} P(\alpha, t) d^2\alpha = 0$$

This is true for all times only if the term  $\{ \dots \} P(\alpha, t)$  vanishes. i.e.

$$\frac{\partial P}{\partial t} = \left(\frac{\gamma}{2} + i\omega_0\right) \frac{\partial}{\partial \alpha} (\alpha P) + \left(\frac{\gamma}{2} - i\omega_0\right) \frac{\partial}{\partial \alpha^*} (\alpha^* P) + \gamma \bar{n} \frac{\partial^2 P}{\partial \alpha \partial \alpha^*} \quad (3.1.4.)$$

This is the Fokker-Planck equation for the damped oscillator. The Fokker-Planck equation may be solved to give a completely general solution, with constants evaluated from the initial conditions. This has been done by Louisell. (Louisell 1969)

Another useful method is the "Green's function" method, (Wang & Uhlenbeck 1945) for which we assume the system is initially in a coherent state,  $|\alpha_0\rangle$ , say. Then  $P(\alpha, t=0) = \delta^2(\alpha - \alpha_0) = \delta(\alpha - \alpha_0) \delta(\alpha^* - \alpha_0^*)$ .

(Glauber 1963)

The solution of (3.1.4.) subject to this initial condition

$$\text{is } P(\alpha, t) = \frac{1}{\pi \bar{n} (1 - e^{-\gamma t})} \exp \left[ \frac{-|\alpha - \alpha_0 e^{-(i\omega_0 + \gamma/2)t}|^2}{\bar{n} - (1 - e^{-\gamma t})} \right]. \quad (3.1.5.)$$

An outline of the method for solution is given in the appendix.

### 3.2 Equations for $\rho$ in the Number State Representation

In the interaction picture, the number states are

$$\begin{aligned} |n\rangle_{\text{int}} &= \exp\left[-\frac{it}{\hbar}(H_S + H_R)\right] |n\rangle \\ &= |n\rangle \exp[-it(n\omega_0)]. \end{aligned}$$

Hence, taking the  $m, n$  th matrix element  $\langle m |_{\text{int}} \{ \dots \} | n \rangle_{\text{int}}$  of the master equation (in the interaction picture) for the simple harmonic oscillator gives the following differential - difference equation for the matrix elements  $\rho_{mn} = \langle m | \rho | n \rangle$ .

$$\begin{aligned} \frac{\partial \rho_{mn}}{\partial t} &= A(mn)^{1/2} \rho_{m-1, n-1} - \frac{1}{2} A(m+n+2) \rho_{mn} \\ &\quad - \frac{1}{2} C(m+n) \rho_{mn} + C[(m+1)(n+1)]^{1/2} \rho_{m+1, n+1}. \end{aligned} \quad (3.2.1.)$$

where  $A = \gamma \bar{n}$ ,  $C = \gamma(\bar{n}+1)$ .

Similar equations are obtained from master equations for other systems.

In many cases we need only know the diagonal elements of  $\rho$  because the system operator we are studying is diagonal in the number state representation. An example is the number of photons in a single mode system:

$$\begin{aligned} \langle n(t) \rangle &= \text{Tr}(n\rho) = \text{Tr}(a^\dagger a \rho) \\ &= \sum_n \langle n | a^\dagger a \rho | n \rangle \\ &= \sum_m \sum_n \langle n | a^\dagger a | m \rangle \langle m | \rho | n \rangle \end{aligned}$$

(since the completeness of the number states means  $\sum_n |n\rangle \langle n| = 1$ )

$$\langle n | a^\dagger a | m \rangle = \hbar \delta_{nm} \quad \text{ie: } n(t) = a^\dagger a \quad \text{is diagonal}$$

$$\therefore \langle n(t) \rangle = \sum_{n=0}^{\infty} n \rho_{nn}(t) \quad (3.2.2.)$$

To obtain  $\rho_{nn}(t)$  we set  $m = n$  in the equation of motion for the elements  $\rho_{mn}(t)$ . In the case of the oscillator,

(3.2.1.) we obtain:

$$\frac{\partial \rho_{nn}}{\partial t} = A n \rho_{n-1, n-1} - A(n+1) \rho_{nn} - C n \rho_{nn} - C(n+1) \rho_{n+1, n+1} \quad (3.2.3.)$$

A solution for this equation, subject to the initial condition  $\rho_{nn}(0) = \delta_{n0}$  (i.e: system initially in a vacuum state) is:

$$\rho_{nn}(t) = [\bar{n}(1-e^{-\gamma t})]^n / [1+\bar{n}(1-e^{-\gamma t})]^{n+1} \quad (3.2.4.)$$

(Pike 1970).

A general method of solution of such equations can be found in most books on probability (e.g. Bailey 1964).

NOTE:

(1) (a) The P-representation and Fokker-Planck equation approach is best suited to systems which are initially in a coherent state, because the Green's function solution is readily obtained.

(b) The number state representation approach is best suited to systems whose initial conditions are specified in terms of number states, since the solution of equations of type (3.2.3.) follows more readily from such conditions.

(2) The example of the simple harmonic oscillator is that of a damped system. The above formalism also covers stimulated systems if we make the substitutions

$$\gamma \rightarrow -\gamma \quad (3.2.5a)$$

$$\bar{n} \rightarrow -(\bar{n}+1) \quad (3.2.5b)$$

(3.2.5a) is equivalent to the damping constant becoming a gain constant. Since  $\bar{n} = (e^{\hbar\omega_0/kT} - 1)^{-1}$ , (3.2.5b) is equivalent to replacing the bath by a reservoir at negative temperature.

This will be seen more clearly in the work on Raman scattering. (see section 4.5)

### 3.3 Connection Between $P(\alpha, t)$ and the $\rho_{mn}(t)$

Using  $\rho = \int P(\alpha, t) |\alpha\rangle\langle\alpha| d^2\alpha$  and  $\rho_{mn}(t) = \langle m|\rho|n\rangle$   
 we have  $\rho_{mn}(t) = \int P(\alpha, t) \langle m|\alpha\rangle\langle\alpha|n\rangle d^2\alpha$ .

Using (1.3.3.) this gives:

$$\rho_{mn}(t) = \frac{1}{\sqrt{m!n!}} \int P(\alpha, t) \alpha^m \alpha^{*n} e^{-|\alpha|^2} d^2\alpha \quad (3.3.1.)$$

### 3.4 Further C-Number Equations of Motion

We may obtain equations of motion for expectation values directly from the master equation. These equations can then be solved, giving expectation values without having to calculate  $\rho$ .

We may write the master equation (2.3.12) as

$$\frac{\partial \rho}{\partial t} = M|\rho) \quad \text{say in the Schrödinger picture,}$$

where  $\rho$  is the system's reduced density operator.

Let  $A$  be a system operator, not explicitly time dependent.

$$\text{Then } \frac{\partial \langle A \rangle}{\partial t} = \frac{\partial}{\partial t} [\text{Tr}(A\rho)] = \text{Tr}[A \frac{\partial \rho}{\partial t}].$$

$$\text{i.e. } \frac{\partial \langle A \rangle}{\partial t} = \text{Tr}[AM|\rho)]. \quad (3.4.1.)$$

(3.4.1.) gives a differential equation for  $\langle A \rangle$ , first order in time.

### 3.5 The Diagonal Elements of $\rho$ in the $|\alpha\rangle$ Representation

If  $\rho$  has a P-representation, we have

$$\rho(t) = \int P(\alpha, t) |\alpha\rangle\langle\alpha| d^2\alpha$$

The diagonal elements of  $\rho$  are  $\langle \beta|\rho|\beta\rangle$  in the coherent state representation.

$$\begin{aligned}\langle \beta | \rho | \beta \rangle &= \int P(\alpha, t) \langle \beta | \alpha \rangle \langle \alpha | \beta \rangle d^2\alpha \\ &= \int P(\alpha, t) |\langle \beta | \alpha \rangle|^2 d^2\alpha.\end{aligned}$$

From section (1.3),  $|\langle \beta | \alpha \rangle|^2 = e^{-|\alpha - \beta|^2}$

so we have

$$\langle \beta | \rho | \beta \rangle = \int P(\alpha, t) e^{-|\alpha - \beta|^2} d^2\alpha. \quad (3.5.1.)$$

### 3.6 A Note on Averages

Since averages of observables are physical quantities, we expect them to be independent of the picture in which they are calculated. (Pictures are corrected by unitary transformations.)

This is in fact true, as the following shows:

Consider a variable  $M$  in one picture, with the corresponding variable in some new picture being  $M'$ , say. Now if  $U$  is the unitary transformation connecting the two pictures,

$$M' = U M U^{-1}. \quad \text{Hence:}$$

$$\begin{aligned}\langle M \rangle &= \text{Tr}(\rho M) = \text{Tr}(U^{-1} \rho U^{-1} U M) && \text{since } U^{-1} U = 1. \\ &= \text{Tr}(\rho U U^{-1} U M U^{-1}) && \text{by cyclic props.} \\ &= \text{Tr}(\rho M') \\ &= \langle M' \rangle.\end{aligned}$$

This fact is useful in work such as in (3.2), where the interaction picture is used.

## 4. RAMAN SCATTERING

### 4.1 Introduction

Raman scattering is the inelastic scattering of light by optical vibrations in a medium. The mechanism which couples the light field to the medium's vibrations is the polarizability of the medium, which is modulated by the vibrations. A simple study of a gaseous diatomic molecular medium gives the qualitative aspects:-

If the molecular polarizability tensor is  $\alpha_{ij}$ , then the dipole moment  $\underline{p} = (p_1, p_2, p_3)$  induced in a molecule by the light field  $\underline{E} = (E_1, E_2, E_3)$  is  $p_i = \alpha_{ij} E_j$  or  $\underline{p} = \underline{\alpha} \cdot \underline{E}$ .

In a gas, the molecules are free, so  $\alpha_{ij}$  is a function of the nuclear separation only. If  $Q$  is the instantaneous departure from equilibrium separation of the two nuclei, we may expand  $\alpha_{ij}$  thus:

$$\alpha_{ij} = \alpha_{ij}^0 + \alpha_{ij}^1 Q + \alpha_{ij}^2 Q^2 + \dots$$

where  $\alpha_{ij}^0 = \alpha_{ij}(Q=0)$ ,  $\alpha_{ij}^1 = \frac{1}{2} \frac{\partial \alpha_{ij}}{\partial Q}(Q=0)$ , ....

$$\therefore p_i = \alpha_{ij}^0 E_j + \alpha_{ij}^1 Q E_j + \dots \quad (4.1.1.)$$

which shows how the vibration (represented by the  $Q$  co-ordinate) and the optical field are coupled.

The term  $\alpha_{ij}^1 Q E_j$  gives first order Raman scattering, the next term gives second order scattering, and so on. Which orders occur, and their strength, is determined by the sizes of the  $\alpha_{ij}^1, \alpha_{ij}^2, \dots$  coefficients, which are determined by molecular symmetries. In general, in Raman

active media,  $\alpha_j^1$ , that is first order scattering, dominates.

We may write

$$\mu_i \approx \alpha_j^0 E_j + \alpha_j^1 Q E_j \quad (4.1.2.)$$

In terms of energy level transitions, light quanta are absorbed by transitions between molecular vibrational levels in the medium, and new light quanta are emitted.

Two first order processes occur:

(a) Stokes Emission

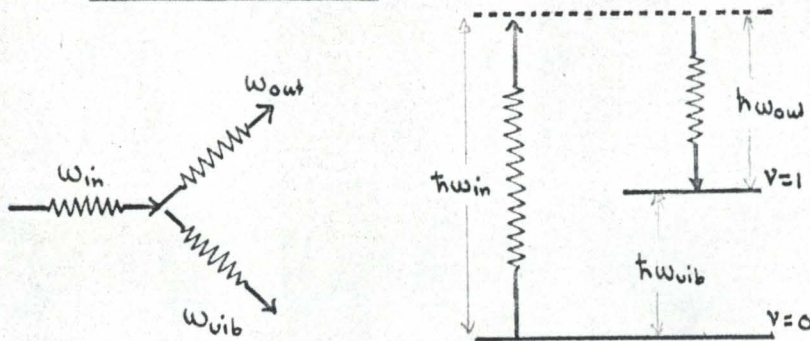


Fig. 2

The levels  $v$  are the vibrational quantum levels.

(b) Antistokes Emission

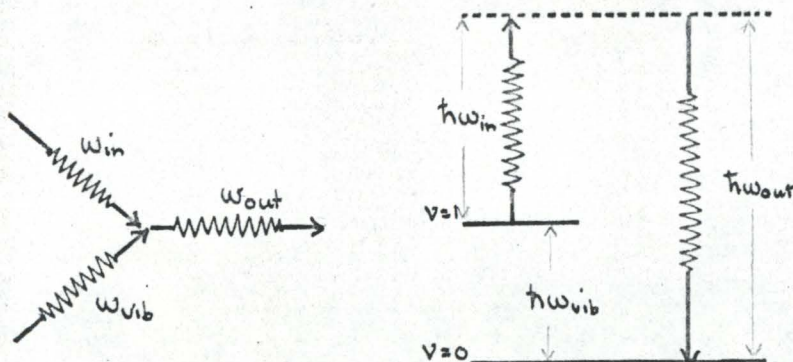


Fig. 3

In case (a), the molecule is excited vibrationally by the ingoing quantum.  $\omega_{out} < \omega_{in}$ .

In case (b), the molecule is initially excited, and is de-excited by the ingoing quantum.  $\omega_{out} > \omega_{in}$ .

In the case of scattering in a solid medium, or in more complex molecular gases, we use a normal co-ordinate

decomposition for the vibration co-ordinate  $Q$ . (In a diatomic molecular gas,  $Q$  is already its own normal co-ordinate under the simple harmonic potential assumption.)

#### 4.2 Raman Scattering and the Master Equation

In order to use the master equation to describe the process, we must carry out the scattering in a cavity.

Raman Hamiltonian:

We take the Hamiltonian 
$$H = \int_V \underline{E} \cdot \underline{P} \, d^3r. \quad (4.2.1.)$$

For a single molecule, 
$$p_i = \alpha_{ij}^0 E_j + \alpha_{ij}^1 Q E_j$$
 or 
$$\underline{P} = \underline{\alpha}^0 \cdot \underline{E} + \underline{\alpha}^1 Q \underline{E} \quad (4.2.2.)$$

(4.2.2.) is a microscopic-variable equation, with  $\underline{E}$  being the local field at a molecule. The  $\underline{E}$  of (4.2.1.) is the macroscopic field.  $\underline{E}_{\text{local}}$  and  $\underline{E}_{\text{mac}}$  are related by the Lorentz factor  $\frac{(n^2+2)}{3}$ , where  $n$  = average refractive index in optical range.

Macroscopic polarization is then  $\underline{P} = N \underline{p}$  where  $N$  is the number of molecules in the sample.

i.e. 
$$\underline{P} = \underline{\alpha}^0 \cdot N \frac{(n^2+2)}{3} \underline{E} + \underline{\alpha}^1 N Q \frac{(n^2+2)}{3} \underline{E}.$$

or: 
$$\underline{P} = \underline{\alpha} \cdot \underline{E} + \underline{\alpha} Q \underline{E} \quad \text{say} \quad (4.2.3.)$$

where  $\underline{\alpha}$  is redefined to include  $N \frac{(n^2+2)}{3}$ .

Hence the Hamiltonian  $H$  of (4.2.1.) becomes

$$H = \int_V \underline{E} \cdot \underline{\alpha} \cdot \underline{E} \, d^3r + \int_V \underline{E} \cdot \underline{\alpha} Q \underline{E} \, d^3r. \quad (4.2.4.)$$

Quantization of Molecular Vibrations (see e.g: Ziman 1969)

We make a standard decomposition of  $Q$  into normal

modes: 
$$Q(\underline{r}) = \sum_{\underline{k}} q(\underline{k}) e^{i \underline{k} \cdot \underline{r}}. \quad (4.2.6.)$$

We now quantize the vibration, by setting

$$q(\underline{k}) = \left( \frac{\hbar}{2M_{\underline{k}} \Omega_{\underline{k}}} \right)^{\frac{1}{2}} (b_{\underline{k}} + b_{\underline{k}}^{\dagger}) \quad (4.2.7.)$$



The  $b_{\underline{k}}, b_{\underline{k}}^\dagger$  are ordinary boson operators. The quantized normal modes are phonons:-  $b_{\underline{k}}$  destroys a phonon of wave-vector  $\underline{k}$ ,  $b_{\underline{k}}^\dagger$  creates a phonon of wave-vector  $\underline{k}$ ,  $\Omega_{\underline{k}}$  is the frequency of the phonon  $\underline{k}$ .  $M_{\underline{k}}$  is an effective mass. We shall see that the set of phonons behaves as a reservoir for the interaction.

In the Hamiltonian (4.2.4.) we insert the expression (4.2.6.) using (4.2.7.), for  $Q$ , and the expression (1.2.8.) for  $\underline{E}$ .

The term  $\int \underline{E} \cdot \underline{\epsilon} \cdot \underline{E} d^3r$  yields the following:

$$H_S = \sum_j \hbar \omega_j (a_j^\dagger a_j + \frac{1}{2}). \quad (4.2.8.)$$

which is the free electric field Hamiltonian.

The term  $\int \underline{E} \cdot \underline{\alpha} \cdot Q \underline{E} d^3r$  gives the interaction Hamiltonian  $V$  between the field and the phonons.

To the Hamiltonian (4.2.4.) we must add the free-phonon Hamiltonian, which is

$$H_R = \sum_{\underline{k}} \hbar \Omega_{\underline{k}} (b_{\underline{k}}^\dagger b_{\underline{k}} + \frac{1}{2}). \quad (4.2.9.)$$

We thus see that the interaction is describable in the formalism of Chapter 2.

### 4.3 Stokes Emission

Since anti-Stokes emission depends on the Stokes emission (see 4.1), if there is initially no anti-Stokes present, we may ignore the presence of anti-Stokes radiation for a short time. We can thus consider the electric field to consist of Stokes and laser photons only. We will deal with standing waves, so we may take the Stokes and laser radiation to be in monochromatic modes. Thus, if the laser mode has frequency  $\omega_L$ , and the Stokes mode has

Hamiltonian must be hermitian, in order to be an observable.

(ii) From (4.2.4.) we find that the coupling constant  $K_j$  is given by

$$\begin{aligned} K_j &= \left( \frac{\hbar \omega_L \omega_S}{2^3 \Omega_j M_j} \right)^{\frac{1}{2}} \int_V e^{i \mathbf{k}_j \cdot \mathbf{r}} \underline{u}_L^*(\mathbf{r}) \cdot \underline{\alpha}_j \underline{u}_S(\mathbf{r}) d^3 r. \\ &= \left( \frac{\hbar \omega_L \omega_S}{V^2 2^3 \Omega_j M_j \epsilon_L \epsilon_S} \right)^{\frac{1}{2}} \underline{e}_L \cdot \underline{\alpha}_j \underline{e}_S \int_V e^{i(\mathbf{k}_S + \mathbf{k}_j - \mathbf{k}_L) \cdot \mathbf{r}} d^3 r. \end{aligned} \quad (4.3.3.)$$

We see that  $K_j$  is only significant when  $\mathbf{k}_S + \mathbf{k}_j = \mathbf{k}_L$ .

To (4.3.1.) and (4.3.2.) we add the free phonons' Hamiltonian

$$H_R = \sum_j \hbar \Omega_j (b_j^\dagger b_j + \frac{1}{2}). \quad (4.3.4.)$$

The number of phonons is the number of molecules times the number of degrees of freedom per molecule, which is of the order of  $10^{23}$ . Hence we are justified in taking the aggregate of phonons to be a reservoir in thermal equilibrium, unperturbed by the interaction.

Thus our laser-stokes-phonon interaction is describable by the approach of chapter 2. In fact, if we compare equations (4.3.1.) - (4.3.4.) with equations (2.4.1.), we see that the laser-stokes system is exactly analogous to the damped oscillator system if we make the identification

$$a \leftrightarrow a_L a_S^\dagger$$

We can proceed as in section (2.4) to derive the analogous master equation

$$\begin{aligned} \frac{\partial S}{\partial t} &= \frac{\gamma}{2} ([a_L a_S^\dagger S, a_L^\dagger a_S] + [a_L a_S^\dagger, S a_L^\dagger a_S]) \\ &\quad + \gamma \bar{n} ([a_L a_S^\dagger, S], a_L^\dagger a_S). \end{aligned} \quad (4.3.5.)$$

We assume the reservoir spectrum is such that

$$|K(\omega)|^2 \bar{n}(\omega) g(\omega) \quad \text{is flat near} \quad \omega = \omega_L - \omega_S.$$

$$\bar{n} = \bar{n}(\omega_L - \omega_S), \quad \text{and} \quad \gamma = 2\pi |K(\omega_L - \omega_S)|^2 g(\omega_L - \omega_S).$$

To make the problem further tractable, we must make the parametric approximation:- We assume that the laser is undepleted. That is, we replace  $Q_L$  by its expectation value  $E_L e^{-i\omega_L t}$

With this approximation (4.3.5.) becomes

$$\frac{\partial S(t)}{\partial t} = \frac{K}{2} ([a_s^\dagger S, a_s] + [a_s^\dagger, S a_s]) + K \bar{n} [a_s^\dagger, S], a_s \quad (4.3.6.)$$

where  $K = |E_L|^2 \gamma$ . (4.3.6.) is in the interaction picture.

Some equations of Motion

(i) Destruction operator  $a_s$ :

$$\begin{aligned} \frac{\partial \langle a_s \rangle}{\partial t} &= \text{Tr} \left( a_s \frac{\partial S}{\partial t} \right) && \text{(see section 3.4)} \\ &= \frac{K}{2} \langle a_s \rangle. && \text{using (4.3.6.)} \end{aligned}$$

$$\therefore \langle a_s(t) \rangle = \langle a_s(0) \rangle e^{K/2 t} \quad (4.3.7.)$$

(ii) Number operator  $n_s$ :

$$\begin{aligned} \frac{\partial \langle n_s \rangle}{\partial t} &= \text{Tr} \left( n_s \frac{\partial S}{\partial t} \right) \\ &= K \langle n_s \rangle + K(\bar{n} + 1) \quad \text{by (4.3.6.)} \end{aligned}$$

$$\therefore \langle n_s(t) \rangle = \langle n_s(0) \rangle e^{Kt} + (\bar{n} + 1)(e^{Kt} - 1). \quad (4.3.8.)$$

The term  $\langle n_s(0) \rangle e^{Kt}$  represents stimulated emission, since  $K$  is +ve, so  $e^{Kt}$  gives exponential gain to  $\langle n_s \rangle$ .

The term  $(\bar{n} + 1)(e^{Kt} - 1)$  represents spontaneous emission noise, since this term exists even if  $\langle n_s(0) \rangle = 0$  (i.e. if there are no stokes photons initially present.)

In practice, the exponential gain would be limited in time, since the fact there is gain will cause depletion of the laser, so the parametric approximation will not remain valid. In practice, a "steady state" situation will result eventually.

#### 4.4 Stokes Radiation Photon Statistics

##### Fokker-Planck Equation

If we insert  $S(t) = \int |\alpha\rangle\langle\alpha| P(\alpha, t) d^2\alpha$  into the master equation (4.3.6.), and proceed as in section (3.1), we obtain the following Fokker Planck equation for the stokes' P-function

$$\frac{\partial P}{\partial t} = -\frac{k}{2} \left( \frac{\partial}{\partial \alpha} \alpha + \frac{\partial}{\partial \alpha^*} \alpha^* \right) P + K(\bar{n}+1) \frac{\partial^2 P}{\partial \alpha \partial \alpha^*} \quad (4.4.1.)$$

If the stokes' mode is initially coherent (i.e.

$P(\alpha, t=0) = \delta^2(\alpha - \alpha_0)$  say), the solution is:

$$P(\alpha, t) = \frac{1}{\pi(\bar{n}+1)(e^{kt}-1)} \exp \left[ \frac{-|\alpha - \alpha_0 e^{k/2 t}|^2}{(\bar{n}+1)(e^{kt}-1)} \right]. \quad (4.4.2.)$$

(see appendix)

This is a Gaussian, with complex, time varying mean  $\alpha_0 e^{k/2 t}$  and time varying variance  $(\bar{n}+1)(e^{kt}-1)$

This is the P-representation for a chaotic state (i.e. one of maximum entropy). (Glauber 1963)

Thus the initially coherent state becomes chaotic, due to the noise of spontaneous emission.

##### Number State Equations for $\rho$

Taking the  $m, n$ th matrix element of each side of the master equation (4.3.6.) yields the following equation:

$$\begin{aligned} \frac{\partial \rho_{mn}}{\partial t} = & A(mn)^{\frac{1}{2}} \rho_{m-1, n-1} - \frac{1}{2} A(m+n+2) \rho_{mn} \\ & - \frac{1}{2} C(m+n) \rho_{mn} + C[(m+1)(n+1)]^{\frac{1}{2}} \rho_{m+1, n+1} \end{aligned} \quad (4.4.3.)$$

where  $A = K(\bar{n}+1)$  and  $C = K\bar{n}$

If the stokes mode is initially in the vacuum state, (i.e.

$\rho_{mn}(0) = \delta_{mn} \delta_{n0}$ ) the solution of the equation for the diagonal elements, viz

$$\frac{\partial \rho_{nn}}{\partial t} = A n \rho_{n-1, n-1} - A(n+1) \rho_{nn} - C n \rho_{nn} + C(n+1) \rho_{n+1, n+1} \quad (4.4.4.)$$

$$\text{is: } \rho_{nn}(t) = \frac{[(1+\bar{n})(e^{kt}-1)]^n}{[(1+\bar{n})e^{kt}-\bar{n}]^{n+1}} \quad (4.4.5.)$$

(e.g. Bailey 1964).

Using the result (3.3.1.), it is possible to obtain the elements  $\rho_{mn}(t)$  from the P-function:-

Using (3.3.1.) and (4.4.2.) we have, for an initially coherent mode,

$$\rho_{mn}(t) = \frac{1}{\sqrt{m!n!}} \frac{1}{\pi(\bar{n}+1)(e^{kt}-1)} \int \beta^m \beta^{*n} e^{-|\beta|^2} \exp\left[\frac{-|\beta - \alpha_0 e^{k/2} t|^2}{(\bar{n}+1)(e^{kt}-1)}\right] d^2\beta.$$

This integral is evaluated in the appendix. The result is:-

$$\rho_{mn}(t) = \frac{\sqrt{m!} e^{i(m-n)(\theta_0 + \pi)} e^{-A r_0^2}}{\sqrt{n!} (\bar{n}+1)(e^{kt}-1)} \frac{(A r_0)^{m-n}}{(1+A)^{m+1} (m-n)!} {}_1F_1\left(m+1, m-n+1, \frac{A^2 r_0^2}{1+A}\right) \quad (4.4.6.)$$

where  ${}_1F_1$  is the confluent hypergeometric function, and

$$A = \frac{1}{(\bar{n}+1)(e^{kt}-1)}$$

$$r_0 = |\alpha_0|, \quad \theta_0 = \arg \alpha_0$$

Since the particular coherent state  $\alpha_0 = 0$  coincides with the vacuum state, (4.4.6.) can give us  $\rho_{mn}(t)$  for an initial vacuum state.

We set  $\alpha_0 = 0$  in (4.4.6.) (i.e.  $r_0 = \theta_0 = 0$ ). Noticing that

$$(r_0)^{m-n} = \delta_{mn} \text{ if } r_0 \rightarrow 0 \quad \text{and using } {}_1F_1(x, y, 0) = 1 \quad \forall x, y$$

we obtain

$$\rho_{mn}(t) = \frac{[(1+\bar{n})(e^{kt}-1)]^n}{[(1+\bar{n})e^{kt}-\bar{n}]^{n+1}} \delta_{mn}. \quad (4.4.7.)$$

We see that the result (4.4.5.) for  $\rho_{nn}(t)$  is regained. Also, we see that the density matrix remains diagonal as the

interaction proceeds. This is not unexpected, as we see from (4.4.3.):— the elements  $\rho_{mn}$  are only coupled along lines parallel to the diagonal. Hence an initially diagonal density matrix (which is the case for a vacuum state initially) will remain diagonal.

#### Expression for $\langle \alpha | \rho | \alpha \rangle$

Using the P-function we may obtain an expression for  $\langle \alpha | \rho | \alpha \rangle$  the diagonal elements of  $\rho$  in the  $|\alpha\rangle$  representation. Using (3.5.1.) and (4.4.2.), we have, for an initially coherent state,

$$\langle \alpha | \rho | \alpha \rangle = \frac{1}{\pi(\bar{n}+1)(e^{kt}-1)} \int \exp(-|\alpha-\beta|^2) \exp\left[\frac{-|\beta-\alpha_0 e^{k/2t}|^2}{(\bar{n}+1)(e^{kt}-1)}\right] d^2\beta.$$

Using the integral relation  $\frac{1}{\pi} \int d^2\beta \exp[-\mu|\beta|^2 + \nu\beta + \lambda\beta^*]$ .

$$= \frac{1}{\mu} \exp\left(\frac{\nu\lambda}{\mu}\right), \quad \text{Re}(\mu) > 0, \quad \text{we have}$$

$$\langle \alpha | \rho | \alpha \rangle = \frac{1}{[(\bar{n}+1)(e^{kt}-1)]} \left[ \exp\left[-|\alpha|^2 - A|\alpha_0|^2 e^{kt} + \frac{|\alpha + A\alpha_0 e^{k/2t}|^2}{(A+1)}\right] \right]. \quad (4.4.8.)$$

where  $A = [(\bar{n}+1)(e^{kt}-1)]^{-1}$

Note that for an initially vacuum state stokes mode,

$$(\alpha_0 = 0), \quad (4.4.5.) \text{ becomes a gaussian.}$$

#### 4.5 Comparison: Damped and Stimulated Processes

It is interesting to compare damped processes, such as the damped oscillator, with stimulated processes, such as the stokes radiation as discussed above. If we compare the appropriate master equations, (2.4.7.) and (4.3.6.), we see that (4.3.6.) is obtained by replacing  $-\gamma$  by  $k$  and  $T$  by  $-T$  in (2.4.7.). (The change  $T \rightarrow -T$  means  $\bar{n} \rightarrow (\bar{n} + 1)$ ). Thus the stimulated process is described formally in the same manner as the damped process, except now the coupling

is of opposite sign, and the heatbath is now a reservoir at negative temperature.

#### 4.6 The Coupled Stokes and Antistokes Fields

The build up of stokes radiation means increasing excitation of phonons, so that antistokes emission becomes more likely. For this reason, we should include the antistokes' field in our calculations.

We thus consider the following problem, which is the field  $E = E_L + E_S + E_A$ , with free Hamiltonian  $\hbar\omega_L(a_L^\dagger a_L + \frac{1}{2}) + \hbar\omega_S(a_S^\dagger a_S + \frac{1}{2}) + \hbar\omega_A(a_A^\dagger a_A + \frac{1}{2})$  interacting with the phonon reservoir, with free Hamiltonian  $\sum_j \hbar\Omega_j(b_j^\dagger b_j + \frac{1}{2})$ . The subscript A denotes antistokes operators. Inserting the expression  $\underline{E} = i\sqrt{\frac{\hbar}{2\omega_L}} a_L \underline{u}_L(r) e^{-i\omega_L t} + i\sqrt{\frac{\hbar}{2\omega_S}} a_S \underline{u}_S(r) e^{-i\omega_S t} + i\sqrt{\frac{\hbar}{2\omega_A}} a_A \underline{u}_A(r) e^{-i\omega_A t} + \text{h.c.}$  into the interaction part of the Hamiltonian (4.2.4.) gives our interaction Hamiltonian in terms of the field and reservoir operators. As before, we make the rotating wave approximation:

$$V = \hbar a_L a_S^\dagger \sum_j K_j^S b_j^\dagger + \hbar a_L a_A^\dagger \sum_j K_j^{*A} b_j + \text{h.c.} \quad (4.6.1.)$$

where, similarly to (4.3.3.)

$$K_j^{*A} = \left( \frac{\hbar\omega_L\omega_A}{V^2 \epsilon^2 \Omega_j M_j \epsilon_L \epsilon_A} \right)^{\frac{1}{2}} \underline{e}_L \cdot \underline{\alpha}_j \underline{e}_A \int_V e^{i(\underline{k}_A - \underline{k}_j - \underline{k}_L) \cdot \underline{r}} d^3 r.$$

$$K_j^S = K_j \quad \text{of (4.3.3.)}$$

We proceed to derive the master equation as before, again making the parametric approximation for tractability. The master equation in the interaction picture is:

$$\frac{\partial \rho(t)}{\partial t} = \frac{K_{SS}}{2} ([a_S^\dagger \rho_S a_S] + [\rho_S^\dagger, S a_S]) + \frac{K_{AA}}{2} ([a_A \rho_A a_A^\dagger] + [\rho_A, S a_A^\dagger])$$

$$\begin{aligned}
& + \frac{k_{SA}}{2} e^{-2i\Delta\omega t} ([a_s^\dagger, a_A^\dagger] + [a_s^\dagger, s a_A^\dagger]) \\
& + \frac{k_{AS}}{2} e^{2i\Delta\omega t} ([a_{AS}, a_s] + [a_{AS}, s a_s]) \\
& + \bar{n} k_{SS} [a_s^\dagger, s], a_s] + \bar{n} k_{AA} [a_{AA}, s], a_A^\dagger] \\
& + \bar{n} k_{SA} e^{-2i\Delta\omega t} [a_s^\dagger, s], a_A^\dagger] + \bar{n} k_{AS} e^{2i\Delta\omega t} [a_{AS}, s], a_s].
\end{aligned} \tag{4.6.2.}$$

where:

$$\begin{aligned}
k_{SS} &= 2\pi g(\omega_0) |K^S(\omega_0)|^2 |E_L|^2 \\
k_{AA} &= 2\pi g(\omega_0) |K^A(\omega_0)|^2 |E_L|^2 \\
k_{SA}^* &= k_{AS} = 2\pi g(\omega_0) K^A(\omega_0) K^{*S}(\omega_0) |E_L|^2 \\
\bar{n} &= \bar{n}(\omega_0)
\end{aligned}$$

where  $\omega_L - \omega_s \approx \omega_0 \approx \omega_A - \omega_L$ .

As in section (2.4), we must assume that

$|K^S(\omega)|^2 \bar{n}(\omega) g(\omega)$  and  $|K^A(\omega)|^2 \bar{n}(\omega) g(\omega)$  are flat functions of  $\omega$  near  $\omega = \omega_0$ .

$2\Delta\omega = 2\omega_L - \omega_s - \omega_A$  is an allowed frequency mismatch, obtained by varying the laser input frequency, and/or the cavity resonances.

#### 4.7 Some Equations of Motion

(i) Coupled equations for  $\langle a_s \rangle$  and  $\langle a_A^\dagger \rangle$ .

$$\begin{aligned}
\frac{d\langle a_s \rangle}{dt} &= \text{Tr}(a_s \frac{\partial \rho}{\partial t}) = \frac{k_{SS}}{2} \langle a_s \rangle + \frac{k_{SA}}{2} \langle a_A^\dagger \rangle e^{-2i\Delta\omega t} \\
\frac{d\langle a_A^\dagger \rangle}{dt} &= \text{Tr}(a_A^\dagger \frac{\partial \rho}{\partial t}) = -\frac{k_{AA}}{2} \langle a_A^\dagger \rangle - \frac{k_{AS}}{2} \langle a_s \rangle e^{2i\Delta\omega t}.
\end{aligned} \tag{4.7.1.}$$

We may ignore small dispersion effects in the gain constants, and set  $k_{SS} = k_{AA} = k_{AS} = k_{SA} = k$ . say.

Setting  $A = \langle a_s \rangle e^{i\Delta\omega t}$  and  $B = \langle a_A^\dagger \rangle e^{-i\Delta\omega t}$ , (4.7.1.)

becomes

$$\frac{d}{dt} \begin{pmatrix} A \\ B \end{pmatrix} = \begin{pmatrix} k/2 + i\Delta\omega & k/2 \\ -k/2 & -(k/2 + i\Delta\omega) \end{pmatrix} \begin{pmatrix} A \\ B \end{pmatrix} \tag{4.7.2.}$$

(4.7.2.) may be solved in the standard manner to yield



solutions which are linear combinations of time varying exponentials. The time variation is given by the eigenvalues of the coefficient matrix in (4.7.2.) - i.e.:

$$\lambda = \pm i(\Delta\omega^2 - ik\Delta\omega)^{1/2} \quad (4.7.3.)$$

The solution is:

$$\begin{pmatrix} A \\ B \end{pmatrix} = c_1 \begin{pmatrix} k/2 \\ \lambda - (k/2 + i\Delta\omega) \end{pmatrix} e^{\lambda t} + c_2 \begin{pmatrix} k/2 \\ -\lambda - (k/2 + i\Delta\omega) \end{pmatrix} e^{-\lambda t} \quad (4.7.4.)$$

where  $\lambda = +i(\Delta\omega^2 - ik\Delta\omega)^{1/2}$ .

and  $c_1 = \{[\lambda + (k/2 + i\Delta\omega)]A(0) + k/2 B(0)\} / k\lambda$ .

$$c_2 = \{[\lambda - (k/2 + i\Delta\omega)]A(0) - k/2 B(0)\} / k\lambda$$

If  $\Delta\omega \gg k$ , (4.7.3.) reduces to

$$\lambda \approx \pm(i\Delta\omega + k/2). \quad (4.7.5.)$$

In this case, A and B become uncoupled, (4.7.4.) reducing

$$\text{to} \quad A(t) \approx A(0)e^{\lambda t} = A(0)\exp[(i\Delta\omega + k/2)t]. \quad (4.7.6.)$$

$$\text{i.e: } \langle a_s(t) \rangle = \langle a_s(0) \rangle e^{k/2 t}$$

$$B(t) \approx B(0)e^{-\lambda t} = B(0)\exp[-(i\Delta\omega + k/2)t]. \quad (4.7.7.)$$

$$\text{i.e: } \langle a_A^\dagger(t) \rangle = \langle a_A^\dagger(0) \rangle e^{-k/2 t}$$

We have two waves, one of gain constant  $\frac{k}{2}$ , the other of  $-\frac{k}{2}$ . If we compare (4.7.6.) with (4.3.7.), we see that the first wave is almost wholly stokes in character. The second, attenuated, wave is almost wholly antistokes in character. Thus in the case of a large frequency mismatch, the waves uncouple into separate stokes and antistokes waves.

According to (4.7.3.), if  $\Delta\omega = 0$  there is no gain. This is because the laser depletion has not been taken into account. A classical analysis (Walls 1970) which takes the depletion into account shows that there is a non-zero gain even with  $\Delta\omega = 0$ . However, this gain is still small, and the main gain occurs for large  $\Delta\omega$ .

(ii) Photon Numbers and Correlations

We can obtain equations of motion for the photon numbers  $\langle n_s(t) \rangle = \langle a_s^\dagger a_s \rangle$ ,  $\langle n_A(t) \rangle = \langle a_A^\dagger a_A \rangle$  and for the correlation functions  $\langle a_A a_s \rangle$ ,  $\langle a_A^\dagger a_s^\dagger \rangle$ .

$$\frac{d}{dt} \langle a_A^\dagger a_A \rangle = -K_{AA} \langle a_A^\dagger a_A \rangle + K_{AA} \bar{n} - \frac{K_{SA}}{2} e^{-2i\Delta\omega t} \langle a_A^\dagger a_s^\dagger \rangle - \frac{K_{AS}}{2} e^{2i\Delta\omega t} \langle a_A a_s \rangle.$$

$$\frac{d}{dt} \langle a_s^\dagger a_s \rangle = K_{SS}(\bar{n}+1) + K_{SS} \langle a_s^\dagger a_s \rangle + \frac{K_{SA}}{2} e^{-2i\Delta\omega t} \langle a_A^\dagger a_s^\dagger \rangle + \frac{K_{AS}}{2} e^{2i\Delta\omega t} \langle a_A a_s \rangle.$$

$$\frac{d}{dt} \langle a_A a_s \rangle = \frac{K_{SS}}{2} \langle a_A a_s \rangle - \frac{K_{AA}}{2} \langle a_A a_s \rangle + \frac{K_{SA}}{2} e^{-2i\Delta\omega t} \langle a_A^\dagger a_A - a_s^\dagger a_s \rangle - K_{SA} \bar{n} e^{-2i\Delta\omega t}.$$

$$\frac{d}{dt} \langle a_A^\dagger a_s^\dagger \rangle = \frac{K_{SS}}{2} \langle a_A^\dagger a_s^\dagger \rangle - \frac{K_{AA}}{2} \langle a_A^\dagger a_s^\dagger \rangle + \frac{K_{AS}}{2} e^{2i\Delta\omega t} \langle a_A^\dagger a_A - a_s^\dagger a_s \rangle - K_{AS} \bar{n} e^{2i\Delta\omega t}.$$

As before we shall ignore dispersion in the K's, setting all equal to K. Let  $A = \langle a_A^\dagger a_A \rangle$ ,  $B = \langle a_s^\dagger a_s \rangle$

$$C = \langle a_A a_s \rangle e^{2i\Delta\omega t}, \quad D = \langle a_A^\dagger a_s^\dagger \rangle e^{-2i\Delta\omega t}.$$

We then obtain the non-homogenous system

$$\frac{d}{dt} \begin{pmatrix} A \\ B \\ C \\ D \end{pmatrix} = \begin{pmatrix} -K & 0 & -\frac{K}{2} & -\frac{K}{2} \\ 0 & K & \frac{K}{2} & \frac{K}{2} \\ \frac{K}{2} & -\frac{K}{2} & 2i\Delta\omega & 0 \\ \frac{K}{2} & -\frac{K}{2} & 0 & -2i\Delta\omega \end{pmatrix} \begin{pmatrix} A \\ B \\ C \\ D \end{pmatrix} + \begin{pmatrix} K\bar{n} \\ K(\bar{n}+1) \\ -K(\bar{n}+\frac{1}{2}) \\ -K(\bar{n}+\frac{1}{2}) \end{pmatrix} \quad (4.7.8.)$$

We can make this an homogenous system by defining

$A' = A - \bar{n}$ ,  $B' = B + (\bar{n}+1)$ , to get:-

$$\frac{d}{dt} \begin{pmatrix} A' \\ B' \\ C \\ D \end{pmatrix} = \begin{pmatrix} -K & 0 & -\frac{K}{2} & -\frac{K}{2} \\ 0 & +K & \frac{K}{2} & \frac{K}{2} \\ \frac{K}{2} & -\frac{K}{2} & 2i\Delta\omega & 0 \\ \frac{K}{2} & -\frac{K}{2} & 0 & -2i\Delta\omega \end{pmatrix} \begin{pmatrix} A' \\ B' \\ C \\ D \end{pmatrix} \quad (4.7.9.)$$

We can further simplify by setting

$W = A'+B'$ ,  $X = A'-B'$ ,  $Y = C+D$ ,  $Z = C-D$ , to obtain

$$\frac{d}{dt} \begin{pmatrix} W \\ X \\ Y \\ Z \end{pmatrix} = \begin{pmatrix} 0 & -k & 0 & 0 \\ -k & 0 & -k & 0 \\ 0 & k & 0 & 2i\Delta\omega \\ 0 & 0 & 2i\Delta\omega & 0 \end{pmatrix} \begin{pmatrix} W \\ X \\ Y \\ Z \end{pmatrix} \quad (4.7.10)$$

The solutions of (4.7.10) are exponentials, with time dependence given by the eigenvalues  $\lambda$  of the matrix of coefficients. i.e:

$$\lambda = \pm i\sqrt{2} \Delta\omega \left[ 1 \mp \sqrt{1 + k^2/\Delta\omega^2} \right]^{1/2} \quad (4.7.11.)$$

For  $\Delta\omega \gg k$ , the solutions again separate into waves of almost wholly stokes or antistokes character.

#### 4.8 Fokker Planck Equation

This is derived in an analogous manner to the previous cases:-

We assume the joint stokes-antistokes density matrix has a P-representation

$$\text{i.e: } \rho(t) = \int P(\alpha, \beta, t) |\alpha\rangle\langle\alpha| |\beta\rangle\langle\beta| d^2\alpha d^2\beta. \quad (4.8.1.)$$

where  $|\alpha\rangle$  is an eigenstate of  $a_A$  and  $|\beta\rangle$  is an eigenstate of  $a_S$ .

Substituting (4.8.1.) into the master equation and proceeding as in Chapter 3, we obtain the following Fokker-Planck equation for the coupled stokes and antistokes fields:

$$\begin{aligned} \frac{\partial P}{\partial t} = & \left\{ -(k/2 + i\Delta\omega) \frac{\partial}{\partial \tilde{\beta}} \tilde{\beta} + (k/2 - i\Delta\omega) \frac{\partial}{\partial \tilde{\alpha}} \tilde{\alpha} \right. \\ & + k/2 \left( \tilde{\beta} \frac{\partial}{\partial \tilde{\alpha}^*} - \tilde{\alpha} \frac{\partial}{\partial \tilde{\beta}^*} \right) + k/2 \bar{n} \frac{\partial^2}{\partial \tilde{\alpha} \partial \tilde{\alpha}^*} \\ & + k/2 (\bar{n}+1) \frac{\partial^2}{\partial \tilde{\beta} \partial \tilde{\beta}^*} - k(\bar{n} + \frac{1}{2}) \frac{\partial^2}{\partial \tilde{\alpha} \partial \tilde{\beta}} \\ & \left. + \text{complex conjugate} \right\} P(\tilde{\alpha}, \tilde{\beta}, t). \end{aligned} \quad (4.8.2.)$$

where  $\tilde{\alpha} = \alpha e^{i\Delta\omega t}$ ,  $\tilde{\beta} = \beta e^{i\Delta\omega t}$ .

The solution of this equation, subject to both modes being initially coherent is outlined in the appendix.

## 5. CONCLUSIONS

The quantum theory of damping and stimulation is a self-consistent theory, applicable to systems perturbed by small interactions. We see that the results it yields agree with those expected classically (e.g. an exponential decay or increase in average photon number is the quantum version of an exponential decay or increase in a classical field intensity). Further, in non-linear optical processes, the effects due to spontaneous emission are described self-consistently by the quantum formalism. In classical theory, we have to add spontaneous emission in an ad hoc manner.

The approximations made in the theory are in most cases highly valid ones. A main short coming is the necessity to make the parametric approximation in order that a problem may become tractable. However, even this approximation is valid for processes of low efficiency, and for times  $t < \frac{1}{K}$ , the rise time constant.

Application of the Louisell damping and stimulation model to the Raman scattering process yields results which agree closely with the classical standing wave results. The model successfully describes the coupling of the stokes and antistokes fields through the phonon bath. Furthermore, the quantum analysis yields the spontaneous Raman effect, as well as the stimulated effect, in a self-consistent manner. The classical analysis describes only the stimulated effect, and hence is only applicable in situations

where the stimulated effect dominates. (e.g: in a Raman laser well above threshold.)

The work in Chapter 4 on the Raman effect is applicable to standing wave problems only: the fields are taken to be monochromatic modes. To study travelling wave problems, we would have to represent the fields by wave-packets. An outline of this method is given in several papers on travelling wave frequency conversion (Tucker and Walls 1969, Von Foerster and Glauber, 1971).

In our description we ignored higher order stokes and antistokes fields. Experimentally, these can be suppressed (Bloembergen 1964) so this is valid. For Raman scattering by atoms, a similar approach to Chapter 4 may be used, replacing the phonons by atoms, and obtaining atomic matrix elements in the tracing out process (Shen 1969).

APPENDICESA. The P-Representations of Equation (3.1.3.)

1. Since  $a|\alpha\rangle = \alpha|\alpha\rangle$  it is obvious that

$$\int P(\alpha) a|\alpha\rangle\langle\alpha| a^\dagger d^2\alpha = \int \alpha^* \alpha P(\alpha) |\alpha\rangle\langle\alpha| d^2\alpha \quad (A1)$$

2. Consider  $(\frac{\partial}{\partial \alpha^*} + \alpha)|\alpha\rangle\langle\alpha|$ :-

$$|\alpha\rangle\langle\alpha| = e^{-\alpha^* a} e^{\alpha a^\dagger} |0\rangle\langle 0| e^{\alpha^* a}$$

$$\begin{aligned} \therefore (\frac{\partial}{\partial \alpha^*} + \alpha)|\alpha\rangle\langle\alpha| &= (\frac{\partial}{\partial \alpha^*} + \alpha) \{ e^{-\alpha^* a} e^{\alpha a^\dagger} |0\rangle\langle 0| e^{\alpha^* a} \} \\ &= e^{-\alpha^* a} e^{\alpha a^\dagger} |0\rangle\langle 0| e^{\alpha^* a} a \end{aligned}$$

$$= |\alpha\rangle\langle\alpha| a$$

$$\text{Hence } \int P(\alpha) |\alpha\rangle\langle\alpha| a d^2\alpha = \int P(\alpha) (\frac{\partial}{\partial \alpha^*} + \alpha) |\alpha\rangle\langle\alpha| d^2\alpha$$

$$\begin{aligned} \text{Now } \int P(\alpha) |\alpha\rangle\langle\alpha| a d\alpha^* &= [P(\alpha) |\alpha\rangle\langle\alpha|]_{-\infty}^{\infty} - \int |\alpha\rangle\langle\alpha| \frac{\partial P}{\partial \alpha^*} d\alpha^* \\ &= - \int |\alpha\rangle\langle\alpha| \frac{\partial P}{\partial \alpha^*} d\alpha^* \end{aligned}$$

since  $P = 0$  at infinity if  $\rho$  is to be a true density operator.

$$\therefore \int P(\alpha) |\alpha\rangle\langle\alpha| a d^2\alpha = \int |\alpha\rangle\langle\alpha| (\alpha - \frac{\partial}{\partial \alpha^*}) P(\alpha) d^2\alpha. \quad (A2)$$

$$\text{Now } \int P(\alpha) |\alpha\rangle\langle\alpha| a a^\dagger d^2\alpha = \int P(\alpha) |\alpha\rangle\langle\alpha| (1 + a^\dagger a) d^2\alpha.$$

(using the  $a, a^\dagger$  commutation relations)

$$= \int P(\alpha) |\alpha\rangle\langle\alpha| (1 + \alpha \alpha^*) d^2\alpha$$

$$\text{since } \langle\alpha| a^\dagger = \alpha^* \langle\alpha|.$$

$$\begin{aligned} \therefore \int P(\alpha) |\alpha\rangle\langle\alpha| a a^\dagger d^2\alpha &= \int P(\alpha) |\alpha\rangle\langle\alpha| d^2\alpha + \int (\alpha^* P) |\alpha\rangle\langle\alpha| a d^2\alpha \\ &= \int P(\alpha) |\alpha\rangle\langle\alpha| d^2\alpha + \int |\alpha\rangle\langle\alpha| (\alpha - \frac{\partial}{\partial \alpha^*}) (\alpha^* P) d^2\alpha \end{aligned}$$

(using (A2))

$$= \int |\alpha\rangle\langle\alpha| \{ 1 + \alpha \alpha^* - \frac{\partial}{\partial \alpha^*} \alpha^* \} P(\alpha) d^2\alpha \quad (A3)$$

The other results of (3.1.3.) are obtained in a similar manner.

## B. Solution of Fokker Planck Equations

If we set  $\alpha = x_1 + ix_2, \beta = x_3 + ix_4, \dots$ , all  $x_i$  real,  
 $i = 1, 2, \dots, 2n$ , say,

we can always reduce a Fokker Planck equation to the form

$$\frac{\partial P}{\partial t} = - \sum_i \sum_j M_{ij} \frac{\partial(x_j P)}{\partial x_i} + \frac{1}{2} \sum_i \sum_j \tau_{ij} \frac{\partial^2 P}{\partial x_i \partial x_j} \quad (B1)$$

where the matrices  $M = (M_{ij})$  and  $\tau = (\tau_{ij})$  are real.

We diagonalize  $M$  with an orthogonal matrix  $S$  ( $M$  is skew symmetric in general) to get

$$\lambda = \text{diag}(\lambda_1, \lambda_2, \dots, \lambda_{2n}) = SMS^{-1}$$

Using the orthogonality of  $S$ , we obtain

$$\sum_i \sum_j M_{ij} \frac{\partial(x_j P)}{\partial x_i} = \sum_i \lambda_i \frac{\partial(u_i P)}{\partial u_i}$$

where

$$u = \begin{pmatrix} u_1 \\ \vdots \\ u_{2n} \end{pmatrix} = S \alpha = S \begin{pmatrix} x_1 \\ \vdots \\ x_{2n} \end{pmatrix}$$

Similarly,  $\sum_i \sum_j \tau_{ij} \frac{\partial^2 P}{\partial x_i \partial x_j} = \sum_i \sum_j \sigma_{ij} \frac{\partial^2 P}{\partial u_i \partial u_j}$ ,

where

$$(\sigma_{ij}) = S \tau S^T$$

Hence (B1) becomes

$$\frac{\partial P}{\partial t} = - \sum_i \lambda_i \frac{\partial(u_i P)}{\partial u_i} + \sum_i \sum_j \sigma_{ij} \frac{\partial^2 P}{\partial u_i \partial u_j} \quad (B2)$$

We wish to solve (B2) subject to the initial condition

$$\begin{aligned} P(x_1, \dots, x_{2n}, t=0) &= \delta(x_1 - x_1^0) \delta(x_2 - x_2^0) \dots \delta(x_{2n} - x_{2n}^0) \\ &= \delta(u_1 - u_1^0) \delta(u_2 - u_2^0) \dots \delta(u_{2n} - u_{2n}^0). \end{aligned}$$

Wang & Uhlenbeck (1945) show that the solution of this problem is a  $2n$  dimension Gaussian distribution in the variables  $v_1, v_2, \dots, v_{2n}$ , with

$$\text{averages } \bar{v}_i = v_i^0 \exp(\lambda_i t)$$

$$\text{variances } \overline{(v_i - \bar{v}_i)(v_j - \bar{v}_j)} = \frac{-\sigma_{ij}}{\lambda_i + \lambda_j} [1 - \exp(\lambda_i + \lambda_j)t].$$

We substitute for the  $v_j$  in terms of the  $x_k$  to obtain our final answer.



C. Calculation of  $\rho_{mn}$ , section (4.4)Set  $\alpha_0 = r_0 e^{i\theta_0}$ and  $\beta = r e^{i\theta}$ , so  $d^2\beta = r dr d\theta$ The  $\theta$  integration is

$$\int_0^{2\pi} e^{i(m-n)\theta} \exp\{-2A r r_0 \omega (\theta - \theta_0)\} d\theta \quad (C1)$$

$$\text{where } A = [(\bar{n}+1)(e^{kt}-1)]^{-1}$$

$$\text{Using } \int_0^{2\pi} e^{im\theta} \exp\{ix \cos\theta\} d\theta = i^m 2\pi J_m(x)$$

(see, e.g. Watson 1966)

$$(C1) \text{ becomes } 2\pi i^{(m-n)} J_{m-n}(i2A r r_0)$$

The  $r$  integration is then

$$\int_0^\infty r^{(m+n+1)} \exp[-(1+A)r^2] J_{m-n}(2iA r r_0) dr \quad (C2)$$

$$\text{Using } \int_0^\infty J_\nu(at) \exp(-p^2 t^2) t^{\mu-1} dt = \frac{\Gamma(\frac{\nu+\mu}{2}) (\frac{a}{2p})^\nu}{2p^\mu \Gamma(\nu+1)} {}_1F_2\left(\frac{\mu+\nu}{2}, \nu+1, -\frac{a^2}{p^2}\right)$$

(see, e.g. Watson 1966)

$$(C2) \text{ becomes } \frac{m! \left[ \frac{iA r_0}{(1+A)^{1/2}} \right]^{m-n}}{2(1+A)^{(m+n+2)/2} (m-n)!} {}_1F_2\left(m+1, m-n+1, \frac{A^2 r_0^2}{1+A}\right).$$

The result for  $\rho_{mn}$  then follows.

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