OPTICAL RESONANCE IN PARTIALLY COHERENT FIELDS

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1. Introduction :

In this lecture, I will deal with a class of problems in optical resonance and nonlinear optics in situations where the exciting sources are not necessarily monochromatic and deterministic functions of space and time. In such a case the exciting source can be modelled by a stochastic process, so that both statistics and the temporal coherence properties of the source will be important. We will see that the dynamical characteristics of the source will be quite crucial if the time scales of the fluctuations of the exciting source are of the same order as other time scales which determine the dynamical evolution of the system. The effects treated in this lecture are not only important as fundamental physical effects but also for the interpretation of the experimental results in the high resolution spectroscopy.

In the usual class room problems, we know that the interaction of radiation and matter can be described by the perturbation theory, for example, the transition rate R, say for stimulated emission or absorption in a monochromatic field of frequency ω_ℓ is

$$R = R(\omega_\ell)\, I \quad , \tag{1}$$

where I in the intensity of the field. If the exciting field is not strictly monochromatic but characterized by some spectrum $\Gamma(\omega_\ell)$, then one can get the transition rates by averaging (1)

$$R = \int d\omega_\ell R(\omega_\ell)\ \Gamma(\omega_\ell) \tag{2}$$

and thus the bandwidth and the shape of the spectrum will determine the transition rates. An important consequence of (2) can be seen in the resonance fluorescence studies. The spectrum of the scattered radiation by a two level atom with energy separation ω_o, of monochromatic radiation in the lowest order of perturbation, is [1]

$$S(\omega, \omega_\ell) = \frac{I g^2}{[\gamma^2 + (\omega_o - \omega_\ell)^2]}\ \delta(\omega - \omega_\ell) \quad , \tag{3}$$

which for the non-monochromatic fluctuating field generalizes to [2]

$$S(\omega) = \int S(\omega, \omega_\ell)\ \Gamma(\omega_\ell) d\omega_\ell$$

$$= \frac{I g^2}{[\gamma^2 + (\omega_o - \omega)^2]}\ \Gamma(\omega_o) \quad . \tag{4}$$

Expression (4) shows that the spectrum of the scattered radiation will consist of peaks at $\omega = \omega_o$ with width γ and the ones determined by the characteristics of the

shape of the spectrum of the exciting radiation. In particular for a broad band source $\Gamma(\omega_o) \sim$ constant, the spectrum would consist of a single peak determined by the atomic parameters ω_o and γ which is in contrast to the monochromatic result (3) where the scattered spectrum simply reflects the spectrum of the exciting source. This is, of course, the celebrated result of Heitler [1] and verified by Gibbs and Venkatesan [3] few years back.

Higher order processes such as two photon absorption depend nonlinearly on intensity of the exciting monochromatic source and that is when the effects of the source statistics first start appearing. For example a Gaussian random process has the property

$$\langle I^n \rangle = n! \langle I \rangle^n \qquad (5)$$

which implies that n^{th} order process in a Gaussian monochromatic field would be n! times more efficient than that in a deterministic field [4] ; provided the average intensity is same. Experiments[5] on multiphoton processes have verified such kinematical enhancement factors. If the field is <u>not</u> monochromatic, then the situation is far from simple. In such a case the spectral characteristics of arbitrary order [4,6] start entering. Expressions like (5) also assume that the perturbation theory is valid. Such an assumption starts breaking down when a resonance is involved and when the saturation effects are important. To describe such situations, we have to have a complete stochastic description of the exciting source since the physical quantities will depend on the time correlation functions of field of arbitrarily high order. This itself leads to difficulties as there are very few models in the theory of random processes where the correlations of arbitrary orders are known. These include:-

(i) The complex field $\vec{E}(t)$ is a Gaussian random processes.

(ii) The complex field $\vec{E}(t)$ has a time independent deterministic amplitude but whose phase $\phi(t)$ is such that its derivative is a Gaussian delta correlated random process [4] .

(iii) The field $\vec{E}(t)$ is a real dichotomic Markov process [cf.7].

(iv) The field $\vec{E}(t)$ has a time independent frequency ω_ℓ with ω_ℓ being a Gaussian random variable.

The model (iv) is relatively simple because the calculations for any nonlinear process can be done assuming ω_ℓ is fixed and then the end result can be averaged with respect to the probability distribution of ω_ℓ. The situation is similar to the Doppler broadening which one encounters in optical resonance in atomic vapours since in case of Doppler broadening the effective frequency is $\omega_\ell - \vec{k}.\vec{v}$ where $\vec{v}$ is a Gaussian random variable. The subsequent discussion is divided in two parts - one dealing with the perturbative results and the other dealing with the exact results for the models mentioned above.

II. Cross Sections for NonLinear Processes Expressed in Terms of the Convolutions involving the Spectral Tensors of the Field:

Let us first assume that the exciting sources are such that their frequencies are far detuned from any of the atomic resonances, so that one can use the n^{th} order perturbation theory to describe n^{th} order nonlinear process. In the steady state, the n^{th} order nonlinear response of the system can be written as

$$P_{\mu}^{(n)}(\omega) = \int\cdots\int d^n\{\omega\}\ \delta(\omega - \Sigma\omega_i)\ \chi^{(n)}_{\mu\{\alpha_n\}}(\{\omega\}) \prod_{i=1}^{n} E_{\alpha_i}(\omega_i) \quad , \tag{6}$$

where $\chi^{(n)}_{\mu\{\alpha_n\}}(\{\omega\})$ stands for the n^{th} order nonlinear susceptibility $\chi^{(n)}(\omega_1, \omega_2, \omega_3 \cdots \omega_n)$. Here $P(\omega)$ and $E(\omega)$ are respectively, the Fourier transform of $P(t)$ and $E(t)$. If E_α is stochastic in nature, then $P_{\mu}^{(n)}(\omega)$ also becomes stochastic. The cross section for the n^{th} order process will be given in terms of a functional involving the polarization fluctuations, the spectrum of such fluctuations will be given by

$$\langle P_{\mu}^{(n)}(\omega)\, P_{\nu}^{(n)}(\omega')\rangle = \delta(\omega+\omega')\ S^{(P)}_{\mu\nu}(\omega) \ ,$$

$$S^{P}_{\mu\nu}(\omega) = \int\cdots\int d^n(\omega_i)\ d^n(\omega_i')\ \delta(\omega - \sum_i \omega_i)\ \delta(\omega + \Sigma\omega_i')$$

$$\chi^{(n)}_{\mu\{\alpha_n\}}(\{\omega_i\})\ \chi^{(n)}_{\nu\{\beta_n\}}(\{\omega_i'\}) \left\langle \prod_{i=1}^{n} E_{\alpha_i}(\omega_i) \prod_{i=1}^{n} E_{\beta_i}(\omega_i') \right\rangle \tag{7}$$

The first line of (7) follows from the stationarity and the Wiener-Khintchine theorem. Relation (7) shows that the cross section for an n^{th} order process will depend on the $2n^{th}$ order correlation function of the field[4,6]. As generally happens in the study of the nonlinear response, one uses sources with different frequencies and so a reasonable assumption to use will be the uncorrelated nature of sources. Then the $2n^{th}$ order correlation can be expressed in terms of the lower ordered correlations of the fields produced by different sources. Such higher order correlations are known only for certain specific models of the sources.

Expression (7) has been recently used to predict the existence of fluctuation induced resonances[8] in the field of a random pump in the context of four wave mixing. For the purpose of calculating the nonlinear gain one has to proceed differently for the gain is directly related to the induced nonlinear polarization. For example the rate of absorption of energy σ from an external field $\vec{E}^{(s)} = 2\mathrm{Re}\ \vec{E}^{(s)} e^{-i\omega_s t}$ per unit incident flux is

$$\sigma = \frac{4\pi\omega_s}{c\,|E^{(s)}|^2}\ \mathrm{Im}\ (\vec{P}^{(s)} \cdot \vec{E}^{(s)*}) \quad , \tag{8}$$

where $\vec{P}^{(s)}$ is the nonlinear polarization induced at ω_s. As an application of (8), consider the case of stimulated Raman gain in the field of a random pump wave $\vec{E}^{(\ell)}(t)$. Writing $\vec{E}(t) = \vec{E}^{(\ell)}(t) + \vec{E}^{(s)}(t)$, using

$$\langle E_\mu^{(\ell)}(\omega_1) \; E_\upsilon^{(\ell)}(\omega_2) \rangle = \delta(\omega_1 + \omega_2) \quad \Gamma_{\mu\upsilon}(\omega_1) \quad , \tag{9}$$

and (6), we find that (8) reduces to

$$\sigma = \frac{12\pi\omega_s}{c|E^{(s)}|^2} \; \mathrm{Im} \int \chi^{(3)}_{\mu\alpha\beta\gamma}(\omega_1, -\omega_1, +\omega_s) \quad \Gamma_{\alpha\beta}(\omega_1) \quad \varepsilon_\gamma^{(s)} \quad \varepsilon_\mu^{(s)*} \; d\omega_1 \tag{10}$$

Using the known form of $\chi^{(3)}$ [9] and of the spectral shape of the pump, the nonlinear gain can be obtained. The integrals can be done analytically for the Lorentzian model of the pump i.e. for

$$\Gamma_{\alpha\beta}(\omega_1) = \frac{\gamma_\ell/\pi}{(\omega_1 - \omega_\ell)^2 + \gamma_\ell^2} \quad I_{\alpha\beta}^{(\ell)} \quad . \tag{11}$$

For Raman scattering in a three level model [9] with g and f representing the initial and final states and n the intermediate state, we find that the integral in (10) reduces to ($\vec{P}_{ij}$ represents the dipole matrix element)

$$(\vec{P}_{fn} \cdot \vec{\varepsilon}^{(s)*}) \; | \vec{P}_{ng}^{*} \cdot \vec{\varepsilon}^{(\ell)} |^2 \; (\vec{P}_{nf}^{*} \cdot \vec{\varepsilon}^{(s)}) \; (\omega_s - \omega_{nf} + i\,\Gamma_{nf})^{-1}$$

$$\Big\{ (\omega_\ell - \omega_s - \omega_{fg} - i\,\Gamma_{fg} - i\,\gamma_\ell)^{-1} \; (\omega_\ell - \omega_{ng} - i\,\Gamma_{ng} - i\,\gamma_\ell)^{-1}$$

$$- 2 \, \frac{(\Gamma_{ng} + \gamma_\ell)}{\Gamma_{nn}} \left[(\omega_\ell - \omega_{ng})^2 + (\Gamma_{ng} + \gamma_\ell)^2 \right]^{-1} \Big\}$$

which clearly shows the effect of γ_ℓ on various relaxation coefficients. The linewidth of the Raman transition is now $(\Gamma_{fg} + \gamma_\ell)$. An exactly similar result holds for spontaneous Raman scattering[10]. It is obvious that similar considerations will apply to higher order gain coefficients for example to Hyper Raman gain which can be shown to depend on $\chi^{(5)}$ and the fourth order correlation function of the pump wave.

III. Methods of Solution for Specific Models :

The general problem, with which one has to deal, can be written in the form

$$\frac{\partial \rho}{\partial t} = L_o \rho + E(t) \; L_+ \rho + E^*(t) \; L_- \rho \quad , \tag{12}$$

where E(t) is the random field. The physical quantities will be given by the ensemble average of ρ and of the two time correlation function of the dipole moment operators. In the interaction picture, one can write (12) as

$$\frac{\partial \rho}{\partial t} = iE(t)\, L_+(t)\rho + iE^*(t)\, L_-(t)\rho \quad , \tag{13}$$

which in some cases may be reduced to

$$\frac{\partial \rho}{\partial t} = i\xi(t)\, \mathcal{L}(t)\rho, \tag{14}$$

which has a formal solution in terms of the time ordering operator

$$\rho(t) = T\, e^{i\int^t \xi(\tau)\mathcal{L}(\tau)d\tau}\rho(o) \tag{15}$$

A. Gaussian model for E(t):

Even if $\xi(t)$ is Gaussian, we can not evaluate the ensemble averages of (15) due to the appearance of the time ordering operator in (15). Only if $\mathcal{L}(\tau)$ is such that

$$[\mathcal{L}(\tau_1),\ \mathcal{L}(\tau_2)] = 0 \quad , \tag{16}$$

then (15) simplifies to

$$\langle \rho(t)\rangle = \exp\left[-\frac{1}{2}\int_o^t dt_1 \int_o^t dt_2 \langle \xi(t_1)\ \xi(t_2)\rangle \mathcal{L}(t_1)\mathcal{L}(t_2)\right]\rho(o) \tag{17}$$

As an example of the type of situation (16) consider optical Bloch equations in a fluctuating electric field for the special case when the transverse and longitudinal relaxation times are equal

$$\frac{\partial}{\partial t}\begin{bmatrix}\langle s^+\rangle\\ \langle s^-\rangle\\ \langle s^z\rangle\end{bmatrix} = \begin{bmatrix} i\Delta - \frac{1}{T_2} & 0 & +2i\alpha^*(t)\\ 0 & -i\Delta - \frac{1}{T_2} & -2i\,\alpha(t)\\ i\alpha(t) & -i\alpha^*(t) & -\frac{1}{T_1}\end{bmatrix}\begin{bmatrix}\langle s^+\rangle\\ \langle s^-\rangle\\ \langle s^z\rangle\end{bmatrix} + \begin{bmatrix}0\\0\\ \eta/T_1\end{bmatrix} \tag{18}$$

The variables $\langle s^{\pm}\rangle$, $\langle s^z\rangle$ correspond respectively to the atomic polarization and inversion. In the special case when $\Delta=0$, $T_1=T_2$, $\alpha(t)$=real, the time evolution operator $U(t,\tau)$ reduces to

$$U(t,\tau) = e^{-\frac{(t-\tau)}{T_2} + i\int_\tau^t \alpha(t')dt' A}, \quad A = \begin{bmatrix}0 & 0 & 2\\ 0 & 0 & -2\\ 1 & -1 & 0\end{bmatrix} \tag{19}$$

and therefore the ensemble average of U can be obtained in closed form

$$\langle U(t,\tau)\rangle = e^{-\frac{t-\tau}{T_2} - \frac{1}{2}A^2\int_\tau^t dt' \int_\tau^t dt''\langle\alpha(t')\ \alpha(t'')\rangle} . \tag{20}$$

However some of these assumptions are quite restrictive and some essential physics is lost by assuming $\alpha(t)$ to be a real field. For example, the character of Stark splittings changes. To see this in its simplest form, consider

$$I(\omega) = \delta(\omega-\omega_\ell-\alpha) + \delta(\omega-\omega_\ell+\alpha) \tag{21}$$

If α is a real Gaussian random variable, then

$$\langle I(\omega)\rangle = \frac{2}{\sqrt{\pi\langle\alpha^2\rangle^{1/2}}} \; e^{-(\omega-\omega_\ell)^2/\sqrt{\langle\alpha^2\rangle}} \tag{22}$$

showing that the ac Stark effect is completely smeared out. However in the same problem if α had been treated as a complex variable, then

$$I(\omega) = \delta(\omega-\omega_\ell-|\alpha|) + \delta(\omega-\omega_\ell+|\alpha|) \tag{23}$$

The averaging of I over the complex Gaussian distribution leads to

$$I(\omega) = \frac{2|\omega-\omega_\ell|}{\langle|\alpha|^2\rangle} \; e^{-(\omega-\omega_\ell)^2/\langle|\alpha|^2\rangle} \tag{24}$$

thus preserving ac stark effect.

In the general case, since (16) is not valid, the progress has been quite slow even when E(t) is a complex Gaussian random process. However if E(t) is also Markovian, then it is possible to use the methods from the theory of Markov processes to obtain continued fraction expansions for the ensemble average of the physical quantities : we will now briefly present this formulation, the details can be found in the work of Lambropoulos and coworkers[11]. Let us denote the set $\langle S^+\rangle, \langle S^-\rangle, \langle S^z\rangle$ by ψ, and let $P(\psi,\mathcal{E})$ be the joint distribution for ψ and the field $\mathcal{E}$. The field $\mathcal{E}(t)$ obeys the Fokker-Planck equation

$$\frac{\partial P(\mathcal{E})}{\partial t} = +2b\,\frac{\partial}{\partial I}(I-I_o)P + 2b\,I_o\,\frac{\partial^2}{\partial I^2}IP$$

$$+\frac{b\,I_o}{2I}\,\frac{\partial^2 P}{\partial\phi^2} = KP\,, \quad \mathcal{E} = \sqrt{I}\,e^{-i\phi} \tag{25}$$

so that

$$\langle\mathcal{E}(t)\;\mathcal{E}^*(t')\rangle = I_o\,e^{-b|t-t'|} \tag{26}$$

Then a simple exercise using (18) and (25) shows that ($\alpha(t) = \alpha_o\,\mathcal{E}(t) = \alpha_o\sqrt{I}\,e^{-i\phi}$)

$$\frac{\partial P(\mathcal{E},\psi)}{\partial t} = -\frac{\partial}{\partial\psi_1}\left[(i\Delta-\frac{1}{T_2})\,\psi_1 P + 2i\alpha_o^*\,\mathcal{E}^*\,\psi_3 P\right]$$

$$+\frac{\partial}{\partial\psi_2}\left[(+i\Delta+\frac{1}{T_2})\,\psi_2 P + 2i\alpha_o\,\mathcal{E}\,\psi_3 P\right]$$

$$-\frac{\partial}{\partial\psi_3}\left[i\alpha_o\,\mathcal{E}\,\psi_1 P - i\alpha_o^*\,\mathcal{E}^*\,\psi_2 P - \frac{\psi_3}{T_1} P\right] + KP \tag{27}$$

$$+\frac{\eta}{T_1}P$$

This multi-dimensional Fokker-Planck equation is not known to have any analytical solution. The physical quantities which we need to know are

$$\langle\psi\rangle = \int P(\mathcal{E},\psi)\,\psi\;d^2\mathcal{E}\,d\psi$$

$$= \int \chi\,d^2\mathcal{E} \tag{28}$$

The elements χ obviously satisfy (M being the square matrix of (18))

$$\frac{\partial \chi}{\partial t} = M\chi + \begin{bmatrix} 0 \\ 0 \\ \frac{\eta}{T_1} \end{bmatrix} + K\chi \quad , \tag{29}$$

where M is linear in ε . The next step in the calculation consists of expanding χ in terms of the eigenfunctions of K which are explicitly known

$$K\Phi_{nm} = \lambda_{nm}\Phi_{nm}, \quad \Phi_{nm} = \frac{e^{in\phi}}{2\pi I_o}\left(\frac{m!}{(m+|n|)!}\right)^{\frac{1}{2}}$$

$$e^{-x}\, x^{|n|/2}\, L_m^{|n|}(x), \quad x = I/I_o, \quad \lambda_{nm} = b(2m+|n|) \quad . \tag{30}$$

Once χ is expanded in terms of Φ_{nm}, the coupled equations for the expansion coefficients can be obtained in the usual manner. It has been possible to convert some of these recursion relations for the expansion coefficients into continued fractions, which can be then numerically evaluated. Lambropoulos and coworkers[11] have investigated a wide class of problems in optical physics using this technique.

If the field $\varepsilon(t)$ is Gaussian but nonmarkovian, then no general techniques appear to be known for the solution to the optical resonance equations in such fields. However some special cases can be handled. For example the rate equation say for atomic population obeys the equation of the form

$$\dot{n} = -a - c|\varepsilon(t)|^2 n - bn \tag{31}$$

Thus the ensemble average of n(t) requires the knowledge of the functional

$$F = \left\langle e^{-c\int_o^t |\varepsilon(\tau)|^2 d\tau} \right\rangle \tag{32}$$

Using the Karhunen-Lo'eve expansion for the Gaussian process (which can be markovian or non-markovian), one can show that

$$F = \prod_k (1 + c\,\lambda_k)^{-1} \quad , \tag{33}$$

where λ_k's are the eigenvalues of the integral equation

$$\int_o^t \langle \varepsilon^*(t)\, \varepsilon(t+\tau)\rangle \Phi_k(\tau)\, d\tau = \lambda_k \Phi_k(t) \quad . \tag{34}$$

For Lorentzian spectrum (markovian case) the functional F can be evaluated in closed form [12]

$$F = e^{bt}\left[\cosh(\beta bt) + \frac{1}{2\beta}(\beta^2+1)\sinh(\beta bt)\right], \quad \beta^2 = \left(1 + \frac{2I_o c}{b}\right) \tag{35}$$

whereas for other types of spectrum such as rectangular, F can only be evaluated numerically[12] . This method has been applied to some problems in Raman scattering and multiphoton ionization[13] .

B.Random Telegraphical Signal Model for E(t) :

Another theoretical model of E for which the optical resonance equations are exactly soluble corresponds to

$$E(t) = \mathcal{E} e^{-i\beta\Phi(t) - i\omega_\ell t} \quad , \tag{36}$$

where $\Phi(t)$ is a dichotomic markov process

$$\langle\Phi(t)\rangle = 0, \langle\Phi(t)\ \Phi(t')\rangle = e^{-2\Gamma|t-t'|} \quad , \Phi^2 = 1 \tag{37}$$

leading to

$$E(t) = \mathcal{E}\cos\beta - i\,\mathcal{E}\sin\beta\,\Phi(t) \tag{38}$$

The auto correlation function of E(t) has an interesting form

$$\langle E(t)\ E^*(t')\rangle = \mathcal{E}^2\left[\cos^2\beta + \sin^2\beta\ e^{-\Gamma|t-t'|}\right] e^{-i\omega_\ell(t-t')} \tag{39}$$

i.e. the spectrum contains a coherent component and a Lorentzian, the relative strength of the two contributions depends on the parameter β . In view of (38), the original density matrix equation (12) can be written in the component form as a matrix equation

$$\frac{\partial\psi}{\partial t} = (C_o + C_1\ \Phi(t))\psi + g \quad , \tag{40}$$

where both C_o and C_1 depend on the strength of the applied field. As has been shown elsewhere[14] this model is exactly soluble i.e. ensemble averages of ψ and of the correlations can be calculated in closed form for example $\langle\psi\rangle$ obeys the equation

$$\frac{\partial\langle\psi\rangle}{\partial t} = C_o\langle\psi\rangle + \int_o^t d\tau\ e^{-2\Gamma(t-\tau)}\ C_1\, e^{C_o(t-\tau)}\ C_1\langle\psi(\tau)\rangle + g \tag{41}$$

The detailed consequences of this for $\beta = \pi/2$ are given in [7] , however the more interesting case $\beta \neq \pi/2$ is under investigation. It may be noted that the variables ψ are non-markovian for this model.

C. Phase Diffusion Model :

We next consider the model of the field where

$$E(t) = \mathcal{E} e^{-i\omega_\ell t - i\Phi(t)} \quad , \dot{\Phi} = \mu(t) \quad ,$$

$$\langle\mu(t)\rangle = 0, \langle\mu(t)\ \mu(t')\rangle = 2\gamma_c\ \delta(t-t') \tag{42}$$

and μ is Gaussian. For this model the general optical resonance equations are exactly soluble [15-17]. Agarwal and coworkers[15,16] have used this model in the context of a large class of optical resonance phenomena such as in resonance fluorescence from two level and three level systems, optical Hanle effect, modulation studies, coherent antistokes Raman scattering, four wave mixing, parametric amplification etc. Rather than give these various applications, we only present the elements of the method used to obtain exact solutions.

A major simplification in optical resonance problems occurs due to the use of the rotating wave approximation i.e. the interaction part of the Hamiltonian is approximated as

$$\mathcal{H} = \sum_{i>j} g_{ij} e^{-i\omega_{ij}t - i\Phi_{ij}(t)} |i\rangle\langle j| + \text{c.c.} \tag{43}$$

and the sum in (43) is over all the allowed transitions. The notation used in (43) implies that the level $|i>$ is above the level $|j>$ by an amount Ω_{ij}. The field with frequency ω_{ij} and phase Φ_{ij} couples only the transition $|i> \leftrightarrow |j>$. Thus the density matrix equation will be

$$\frac{\partial \rho}{\partial t} = L_o \rho - i \left[\sum_{i>j} g_{ij} e^{-i\omega_{ij}t - i\Phi_{ij}(t)} |i\rangle\langle j| + \text{h.c.} \;, \rho \right] \tag{44}$$

where L_o stands for other coherent interactions as well as other sources of relaxation such as due to collisions, radiative emission etc. The basic idea is to transform (44) into an equation of the form

$$\frac{\partial \tilde{\rho}}{\partial t} = L_o \tilde{\rho} - i \left[\sum_{i>g} g_{ij} |i\rangle\langle j| + \text{h.c}, \tilde{\rho} \right] + \sum \mu_{ij}(t) L_{ij} \tilde{\rho} \tag{45}$$

so that the Gaussian delta correlated random processes appear linearly in the dynamical equations. Such dynamical equations are exactly soluble and we quote the result [15]. Write the basic equation as

$$\dot{x}_i = \sum_j (M_{ij} x_j + \mu_{ij}(t) x_j) \;, \tag{46}$$

$$\langle \mu_{ij} \rangle = 0, \langle \mu_{ij}(t) \; \mu_{kl}(t') \rangle = 2Q_{ijkl} \; \delta(t-t') \tag{47}$$

then

$$\langle \dot{x}_i \rangle = \sum_j M_{ij} \langle x_j \rangle + \sum_{jk} Q_{ikkj} \langle x_j \rangle = \langle A_i \rangle \tag{48}$$

The second moments also satisfy the closed set of equations

$$\frac{\partial}{\partial t} \langle x_k x_l \rangle = \langle A_k x_l \rangle + \langle A_l x_k \rangle + \sum_{ij} Q_{kilj} \langle x_i x_j \rangle \tag{49}$$

The variables x_i's are also markovian, so that the time correlations can be obtained from (48)

$$\frac{d}{dt} \langle x_i(t) \; x_k(t') \rangle = \sum_j (M_{ij} + \sum_k Q_{ikkj}) \langle x_j(t) \; x_k(t') \rangle \;. \tag{50}$$

Application of (48) to (45) would then lead to

$$\frac{\partial \langle \tilde{\rho} \rangle}{\partial t} = L_o \langle \tilde{\rho} \rangle - i \left[\sum_{i>j} g_{ij} |i\rangle\langle j| + \text{h.c.}, \langle \tilde{\rho} \rangle \right]$$

$$+ \sum Q_{ijkl} L_{ij} L_{kl} \langle \tilde{\rho} \rangle . \tag{51}$$

In order to see how the transformation from (44) to (45) is done and what is the physics of this new density matrix $\tilde{\rho}$, consider the case of resonant Raman scattering in fields of arbitrary intensity. As before let a laser of frequency ω_ℓ [ω_s]phase Φ_ℓ [Φ_s]act between the ground level $|g\rangle$[final level $|f\rangle$] and the resonant intermediate level $|n\rangle$. In such a case ρ and $\tilde{\rho}$ are related by the canonical transformation such that

$$\tilde{\rho}_{\alpha\alpha} = \rho_{\alpha\alpha}; \quad \tilde{\rho}_{gn} = e^{-i\Phi_\ell - i\omega_\ell t}\rho_{gn}$$

$$\tilde{\rho}_{fn} = e^{-i\Phi_s - i\omega_s t}\rho_{fn}, \quad \tilde{\rho}_{gf} = e^{-i(\Phi_\ell - \Phi_s) - it(\omega_\ell - \omega_s)} \tag{52}$$

Use of (51), then shows that the equations of motion for $\langle\tilde{\rho}\rangle$ can be obtained from those in the absence of any phase fluctuations, if we change the relaxation coefficients in the off-diagonal elements as follows

$$\Gamma_{ng} \rightarrow \Gamma_{ng} + \gamma_\ell, \; \Gamma_{nf} \rightarrow \Gamma_{nf} + \gamma_s \; , \; \Gamma_{fg} \rightarrow \Gamma_{fg} + (\gamma_\ell + \gamma_s - 2\gamma_{\ell s}) \tag{53}$$

where $\gamma_{\ell s}$ denotes the cross correlation between the lasers i.e.

$$2\gamma_{\ell s}\,\delta(t-t') = \langle \Phi_\ell(t)\,\Phi_s(t')\rangle \tag{54}$$

This simple substitution rule [cf.15,18] enables one to obtain the characteristics of the Raman system for example Raman gain which can be shown to be related to Im $\tilde{\rho}_{fn}$. An important point worth emphasizing is that in case the same laser is used on both the transitions, then $\gamma_\ell + \gamma_s - 2\gamma_{\ell s} = 0$ and hence Γ_{fg} remains unchanged. This has important consequences on the trapping of the population in the state $|f\rangle$[19] . This cross correlation has been successfully used by Thomas et al [20] in the observation of Ramsay fringes using a resonant Raman transition which will probably have important implications on frequency standards. For the description of the nonlinear optical processes, such as four wave mixing [6,8] which are characterized by the square of the induced polarization, we have to use equations like (49). Note further that if one wants to calculate the dipole moment e.g. ρ_{fn}, then we can not use $\tilde{\rho}_{fn}$, but we have to make another transformation $\tilde{\rho} \rightarrow \tilde{\tilde{\rho}}$ such that

$$\tilde{\tilde{\rho}} = \tilde{\rho}\, e^{i\Phi_s} \tag{55}$$

Equations for $\tilde{\tilde{\rho}}$ would be similar to (45) so that (48) can be applied directly. Now the equations for the diagonal elements of $\langle\tilde{\tilde{\rho}}\rangle$ will depend on γ_ℓ, γ_s etc. The dipole moment ρ_{fn} is needed to evaluate the spectrum of the spontaneously emitted radiation; which is related to the two time correlation function $\langle(|n\rangle\langle f|)_t \; (|f\rangle\langle n|)_{t'}\rangle$. Such two time correlations can be obtained by using the equations for $\langle\tilde{\tilde{\rho}}\rangle$ and the markov property of the variables $\tilde{\tilde{\rho}}_{\alpha\beta}$. Note also that the transformation (55) is not adequate if we want to calculate the dipole moment ρ_{gn} - in this case we should use

$$\bar{\rho} = \tilde{\rho}\, e^{i\Phi_\ell} \tag{56}$$

So far we have not put in any propagation effects. It turns out that even the propagation effects can be incorporated in the optical resonance equations in the fluctuating fields, which then could be solved for the phase diffusion model. To see this, let us consider the equations [cf.21] for the stimulated Raman scattering under the conditions that the saturation effects are not important :

$$\frac{\partial \mathcal{E}_s}{\partial z} + \frac{1}{v_s}\frac{\partial \mathcal{E}_s}{\partial t} + \Gamma_s \mathcal{E}_s = \sigma_1 \, Q^* |\mathcal{E}_\ell| \, e^{-i\Phi_\ell \, (t - \frac{z}{v_\ell})}$$

$$\frac{\partial Q^*}{\partial t} + \frac{Q^*}{T_2} = \sigma_2 |\mathcal{E}_\ell| \mathcal{E}_s \, e^{i\Phi_\ell (t - \frac{z}{v_\ell})} \qquad (57)$$

where $\mathcal{E}_s$ is the slowly varying envelope of the stokes field and Q is the Raman mode. A simple change of variables

$$z' = z, \quad \tau = t - z/v_s \; . \; P = Q^* e^{-i\Phi_\ell} \qquad (58)$$

brings the above equations in the form

$$\frac{\partial \mathcal{E}_s}{\partial z'} + \Gamma_s \mathcal{E}_s = \sigma_1 |\mathcal{E}_\ell| P \; ,$$

$$\frac{\partial P}{\partial \tau} + \frac{P}{T_2} + i\mu (\tau - \beta z') P = \sigma_2 |\mathcal{E}_\ell| \mathcal{E}_s \; , \; \beta = \frac{1}{v_s} - \frac{1}{v_\ell} \; . \qquad (59)$$

The Gaussian delta correlated noise appears in (59) in the multiplicative form and we can solve the equations for first and second moments by using (48), (49) and the Laplace transform of (59) with respect to the z' coordinate. The results so obtained are found to agree with those derived by using other methods [21].

Elliott et al[22]have considered a modification of the laser by putting a modulator so that the phase fluctuations can be controlled and they are planning extensive studies of the optical resonance phenomena in fluctuating fields. In their set up, one has

$$\langle \mu(t) \; \mu(t') \rangle = \frac{\mu^2}{\pi} \; \frac{\sin B/2 \, (t-t')}{(t-t')} \qquad (60)$$

which for large B, goes over to a delta correlated process. For small B, $\langle \mu(t) \; \mu(t') \rangle \sim \mu^2 B/2\pi$ which corresponds to an external frequency ω_ℓ being a Gaussian random variable and thus the situation is similar to Doppler broadening. For intermediate values of B no explicit solutions are known.

Finally we would like to mention that the methods discussed above are equally applicable[23]to study the effect of the velocity changing collisions on optical line shapes. In the classical model of weak collisions[24]the effective field acting on the atom can be written as

$$E(t) = \mathcal{E} \, e^{-i\omega_\ell t + i \vec{k}_\ell \cdot \vec{R}(t)} \; , \qquad (61)$$

where $\vec{R}(t)$ is the position of the atom, which is given by the familiar Brownian motion equations

$$\dot{\vec{R}} = \vec{v} \quad , \quad \dot{\vec{v}} + \gamma_v \vec{v} = \vec{F}(t) \quad ,$$

$$\langle F_\alpha(t)\, F_\beta(t') \rangle = 2\, \Gamma_v\, \delta(t-t')\, \delta_{\alpha\beta} \quad . \tag{62}$$

Notice the similarity of this situation to the phase diffusion model, the correspondence is exact in the limit of large γ_v . Hence the results obtained in the context of the phase diffusion model could be directly used to get the corresponding results for the case of velocity changing collisions. For example for the case of Raman gain, we will have[23]in place of (53)

$$\Gamma_{ng} \rightarrow \Gamma_{ng} + \hat{k}_\ell^2 \tilde{\Gamma}_v \quad , \quad \Gamma_{nf} \rightarrow \Gamma_{ng} + \hat{k}_s^2 \tilde{\Gamma}_v \quad ,$$

$$\Gamma_{fg} \rightarrow \Gamma_{fg} + \tilde{\Gamma}_v\, (\hat{k}_\ell - \hat{k}_s)^2 \quad , \qquad \tilde{\Gamma}_v = \Gamma_v / \gamma_v^2 \tag{63}$$

where $\hat{k}_\ell$ and $\hat{k}_s$ are, respectively, the unit vectors in the direction of propagation of the pump and Stokes radiation. The case of finite γ_v can also be handled by using methods similar to those discussed in connection with Gaussian model for the appropriate $\tilde{\rho}$ equation involves the general Gaussian stochastic variable v(t).

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