Quantum statistical theory of optical-resonance phenomena in fluctuating laser fields*

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A quantum statistical theory of resonant optical phenomena is developed for the case when the fluctuations of the laser, used for exciting transitions, are important. The general equations describing the dynamics of a relaxing two-level atom (TLA) are solved *exactly* for different types of mean values and the two-time amplitude and intensity correlations. The phase-diffusion model is adopted for laser fluctuations. The exact results are used to analyze the effect of laser fluctuations on a number of optical effects—optical free-induction decay, adiabatic following, the spectrum of the scattered light from a relaxing TLA, the energy-absorption spectrum from a weak field, Hanle resonances, etc. In each case, the laser fluctuations are found to affect in an important way the characteristics of the above optical-resonance phenomena. For example, the spectrum of scattered radiation from a TLA has the usual three-peak structure for fields at resonance and with strengths α above the threshold and for $\alpha \gg \gamma_c$; however, now the peak heights (widths) are in the ratio $3x$ $(2/3x)$, $x = [\gamma_c + (T_1^{-1} + T_2^{-1})/3]/ [2\gamma_c + T_2^{-1}]$, where γ_c^{-1} is the correlation time for laser phase fluctuations. For $\gamma_c \gg \alpha$, one gets a single-peak spectrum. The laser field is treated as a second-quantized field with excitation in either a coherent state or a Fock state. The results, obtained by straightforward perturbation theory, but valid for arbitrary values of the relaxation parameters, detuning, and the laser correlation time, are also presented.

I. INTRODUCTION

Atomic systems in the presence of resonant optical fields are known to exhibit a variety of phenomena[1] depending on the interactions of the atomic system with its surroundings, i.e., the type of relaxation mechanism, the relations of different relaxation parameters among themselves as well as the relaxation of the applied field strength to the relaxation parameters. If the incident field is monochromatic (or nearly monochromatic) and if its strength is much smaller than the atomic energy-level differences (Rabi frequency ≪ atomic frequency), then one can obtain a number of exact solutions, and these have been used to predict a number of optical effects and to analyze the experimental data. The optical effects most extensively studied, both experimentally and theoretically, include optical nutation, free-induction decay,[2,3] resonance fluorescence,[4-15] optical double resonance,[16] Hanle resonances,[17] photon echoes,[18] self-induced transparency,[1] etc. In the case when the amplitude of the monochromatic field is a varying function of time such that its rate of change is much less than the rate of change of the Bloch vector, then the adiabatic solutions[19-22] to Bloch equations can still be obtained. When the field envelope varies in an arbitrary manner, the solution of the Bloch equations is far from being known (see, however, the case of self-induced transparency). The problem becomes worse if the laser field amplitude is a fluctuating variable which is indeed the case in practice. Of course if the laser correlation time is very large compared to the other characteristic times in the problem, then the known solutions can still be used. The trouble arises only when the laser correlation time is of the order of the characteristic times in the problem.

In this paper we analyze the effects of laser fluctuations on the outcome of resonance experiments. A number of *exact* results are presented. Our exact calculations require the knowledge of the laser correlations of arbitrary order. Since the phase-diffusion model[23,24] of the laser is probably the only one for which all order correlation functions are known in closed form, we have naturally used this model in our exact solutions. This is in contrast to approximate calculations in which case one needs to know only the second-order correlation functions of the field and hence the calculations there are independent of the laser model. In an earlier note[25] we have already described how drastically the spectrum of resonance fluorescence changes due to laser fluctuations.[26,27] In the meantime a study of the effect of laser fluctuations on level-crossing experiments[17] has also appeared.

The calculations presented here have important bearings on the experimental work on absorption,[28] the emission spectrum of a laser-driven two-level system, Hanle resonances, etc. In the development given in the succeeding sections we give the general formulation and then study a number of specific effects. From the point of view of statistical mechanics, the systems studied in Secs. III–V provide us with some samples of exactly soluble models. The outline of this paper is as follows. In Sec. II we formulate the general equations describing the dynamics of a relaxing two-level atom in the presence of a laser field. The study of the dynamics of an atom in a fluctuating field is essentially a study of stochastic Liouville operators. Sections III–V are devoted to exact results. The laser phase fluctuations are assumed to undergo diffusion. Using the techniques of multiplicative stochastic processes,[29] we obtain in Sec. III explicit results for the ensemble average of one- and two-time expectation values. The special optical effects we treat in Sec. III are (i) optical free-induction decay, (ii) adiabatic following, (iii) antibunching effects in fluorescence, (iv) the spectrum of the fluorescence (scattered light), and (v) the absorption spectrum. In all of the above cases analytical results are presented, these results being valid for arbitrary values of the field strengths, laser correlation time, etc. In Sec. IV we undertake a study of multilevel systems interacting with a fluctuating laser field. The case of Hanle resonances for transitions between $J=0$ and $J=1$ is analyzed in detail. In Sec. V we present a treatment of the laser field and its fluctuations which makes second quantization of the laser field quite transparent. The equivalence with the results of Sec. III is established. The paper concludes with two appendixes. In Appendix A the steady-state and transient spectra are calculated using second-order perturbation theory and the results valid for arbitrary values of relaxation parameters, laser correlation time, and detuning are presented. In Appendix B we discuss some important results on multiplicative stochastic processes. The results of this appendix have wide applications not only in the present context but also in other fields such as exciton diffusion.[30]

II. DYNAMICS OF A TWO-LEVEL ATOM IN A FLUCTUATING LASER FIELD AND IN THE PRESENCE OF VARIOUS RELAXATION MECHANISMS

The most general equations describing the Markovian dynamics of a two-level atom (having energy separation ω) in the presence of an external laser field are given by

$$\begin{aligned}\dot{\rho}_{11} &= -2\Gamma_1\rho_{11} + 2\Gamma_2^{(0)}\rho_{22} \\ &\quad + ig\mathcal{E}(t)(\rho_{21}e^{-i\varphi(t)} - \text{c.c.}), \\ \dot{\rho}_{22} &= -2\Gamma_2\rho_{22} + 2\Gamma_1^{(0)}\rho_{11} \\ &\quad + ig\mathcal{E}(t)(\rho_{12}e^{i\varphi(t)} - \text{c.c.}) + 2p, \\ \dot{\rho}_{12} &= -(i\Delta + \Gamma)\rho_{12} - ig\mathcal{E}(t)(\rho_{11} - \rho_{22})e^{-i\varphi(t)},\end{aligned} \tag{2.1}$$

where $2\Gamma_1$ ($2\Gamma_2$) represents the total transition rate per unit time from level 1 (2); $2\Gamma_1^{(0)}$ ($2\Gamma_2^{(0)}$) is the transition rate to level 2 (1) from level 1 (2); $2p$ is the rate at which the atoms are pumped to level $|2\rangle$ by some external source. The off-diagonal decay rate usually also has a contribution from phase-interrupting collisions. Note that $\rho_{11}+\rho_{22}\neq 1$, since we have allowed external pumping and decay. In deriving (2.1) the usual rotating-wave approximation has been made and ρ is in a frame rotating with the central frequency ω_0 of the applied field and Δ is the detuning ($\Delta=\omega-\omega_0$).

The complex field amplitude $\mathcal{E}(t)e^{-\varphi(t)}$ represents the laser field. The field may be treated either as a semiclassical field or a fully quantized field. In the latter case, one has to assume that the field is described by a coherent state whose amplitude is a stochastic function of time.[14,4(b)] If the initial state of the field is different from a coherent state, then a similar treatment is still possible (given in Sec. V).

In special cases, Eqs. (2.1) reduce to the usual equations if we use the following relations among relaxation parameters. (A) Radiative relaxation:

$$\Gamma_1=\Gamma_1^{(0)}=\gamma,\quad \Gamma_2=\Gamma_2^{(0)}=p=0,\quad \Gamma=\gamma, \tag{2.2}$$

(B) Collisional relaxation (cf. Ref. 31):

$$\Gamma_1=\Gamma_1^{(0)}=\tfrac{1}{2}\kappa(1-\beta),\quad \Gamma_2=\Gamma_2^{(0)}=\kappa\tfrac{1}{2}\beta,\quad \Gamma=\kappa, \tag{2.3}$$

(C) Torrey's case:

$$\begin{aligned}&\Gamma_1-\Gamma_1^{(0)}=\Gamma_2-\Gamma_2^{(0)}=p,\\ &T_1^{-1}=2(p+\Gamma_1^{(0)}+\Gamma_2^{(0)}),\quad T_2^{-1}=\Gamma,\\ &\langle S^z\rangle_{eq}=-\tfrac{1}{2}+\Gamma_2^{(0)}(p+\Gamma_1^{(0)}+\Gamma_2^{(0)})^{-1}.\end{aligned} \tag{2.4}$$

In all of the above three cases, Eqs. (2.1) reduce to Bloch equations:

$$\begin{aligned}\langle\dot{S}^+\rangle &= (i\Delta-1/T_2)\langle S^+\rangle + 2ig\langle S^z\rangle\mathcal{E}(t)e^{i\varphi(t)},\\ \langle\dot{S}^z\rangle &= -(1/T_1)\{\langle S^z\rangle-\langle S^z\rangle_{eq}\} + ig\mathcal{E}(t)(\langle S^+\rangle e^{-i\varphi(t)}-\text{c.c.})\end{aligned} \tag{2.5}$$

When $\Gamma_2^{(0)}=0$, the set of equations (2.1) reduces to that of Schenzle and Brewer.[32]

If the incident field is monochromatic [$\mathcal{E}(t)$, $\varphi(t)$ time independent, deterministic objects] then one can solve (2.1) by straightforward Laplace-transformation techniques. The detailed solutions are well known in the special cases mentioned above.[1,32] If $\mathcal{E}$, φ are time independent but fluctuating quantities, then the final results can be obtained by averaging the Laplace-transformed solutions of (2.1) over the distribution of $\mathcal{E}$, φ.

In this paper we study how the temporal fluctuations of the laser beam affect the outcome of the resonance experiments. In this case $\mathcal{E}$, φ are stochastic variables and thus ρ_{11}, ρ_{22}, ρ_{12} also acquire stochastic character. Equations (2.1) can be cast in the form of Langevin equations[33] if we assume that the laser field amplitude $b(t) = \mathcal{E}(t)e^{-i\varphi(t)}$ can be considered as undergoing a Markovian process,[18] i.e.,

$$\dot{b} = f(b, b^*) + F(t), \tag{2.6}$$

where $F(t)$ is a δ-correlated Gaussian random process. Most of the laser theories[18] and the experiments show that to a good approximation laser dynamics can be described by (2.6). It is clear from Eqs. (2.1) and (2.6) that the set (ρ_{11}, ρ_{22}, ρ_{12}, ρ_{21}, b, b^*) has a Markovian behavior and one can write the corresponding Fokker-Planck equation[33] with drift and diffusion coefficients which are nonlinear functions of (ρ_{11}, ρ_{22}, ρ_{12}, ρ_{21}, b, b^*). It seems highly unlikely that analytical solutions of such a Fokker-Planck equation can be obtained except in special cases.[34] In the case of the phase-diffusion model[23,24] of the laser beam, exact results can be obtained. For this model $\mathcal{E}(t)$ is taken to be a deterministic variable independent of time. The phase undergoes diffusion:

$$\dot{\varphi} = \mu(t), \quad \varphi(0) = \varphi_0, \tag{2.7}$$

where φ_0 is uniformly distributed between 0 and 2π, and $\mu(t)$ is a δ-correlated Gaussian random process with

$$\langle \mu(t)\rangle = 0, \quad \langle \mu(t_1)\mu(t_2)\rangle = 2\gamma_c \delta(t_1 - t_2), \tag{2.8}$$

where the single brackets $\langle\ \rangle$ denote the ensemble average with respect to the distribution of the random process $\mu(t)$. The next few sections are devoted to the consideration of these exact results.

It should be noted that if the field strength is much less than the saturation field, then the results for the scattered spectrum can be obtained by iterating Eqs. (2.1). The approximate results are then model independent. We present these approximate results in Appendix A. Some of these approximate results in the special cases are known; however, their discussion is included because it is interesting from the viewpoint of the correlation functions and that we present expressions for arbitrary values of γ_c, T_1, T_2, Δ without using any *ad hoc* assumption, and also the initial state of the atom can be arbitrary.

III. EXACT SOLUTIONS FOR THE EFFECT OF LASER PHASE FLUCTUATIONS ON THE DYNAMICS OF A TWO-LEVEL ATOM

A. Ensemble average of one-time expectation values

Equations (2.1) involve the phase φ in a nonlinear fashion. By a redefinition of the variables, Eqs. (2.7) and (2.8) can be cast into a linearized set of equations. On introducing the variables

$$\chi_1 = \rho_{21}, \quad \chi_2 = \rho_{12}e^{2i\varphi}, \quad \chi_3 = \rho_{11}e^{i\varphi}, \quad \chi_4 = \rho_{22}e^{i\varphi}, \quad \chi_5 = e^{i\varphi}, \tag{3.1}$$

we find the equations for χ

$$\dot{\chi} = i\mu(t)D\chi + \begin{bmatrix} -\Gamma + i\Delta & 0 & i\alpha & -i\alpha & 0 \\ 0 & -\Gamma - i\Delta & -i\alpha & i\alpha & 0 \\ i\alpha & -i\alpha & -2\Gamma_1 & 2\Gamma_2^{(0)} & 0 \\ -i\alpha & +i\alpha & 2\Gamma_1^{(0)} & -2\Gamma_2 & 2p \\ 0 & 0 & 0 & 0 & 0 \end{bmatrix} \chi, \tag{3.2}$$

$$D_{ij} = \delta_{ij}D_{ii}, \quad D_{11} = 0, D_{22} = 2, \quad D_{33} = D_{44} = D_{55} = 1, \quad \alpha = g\mathcal{E}_0.$$

This equation has the form of the standard equation of the multiplicative stochastic processes. From the theory of the multiplicative stochastic processes (discussed briefly in Appendix B) it follows that the average of χ over the distribution of φ satisfies

$$\langle\dot{\chi}\rangle = \begin{bmatrix} -\Gamma + i\Delta & 0 & i\alpha & -i\alpha & 0 \\ 0 & -\Gamma - i\Delta - 4\gamma_c & -i\alpha & i\alpha & 0 \\ i\alpha & -i\alpha & -(2\Gamma_1 + \gamma_c) & 2\Gamma_2^{(0)} & 0 \\ -i\alpha & +i\alpha & 2\Gamma_1^{(0)} & -(2\Gamma_2 + \gamma_c) & 2p \\ 0 & 0 & 0 & 0 & -\gamma_c \end{bmatrix} \langle\chi\rangle. \tag{3.3}$$

The solution of (3.3) yields the mean values $\langle\rho_{21}\rangle$, $\langle\rho_{11}e^{i\varphi}\rangle$, etc. In order to calculate $\langle\rho_{11}\rangle$, $\langle\rho_{22}\rangle$ we have to introduce another set of variables for $\langle\rho_{11}e^{i\varphi}\rangle \neq \langle\rho_{11}\rangle\langle e^{i\varphi}\rangle$. The following set is found useful:

$$\psi(t) = \chi(t)e^{-i\varphi(t)}, \tag{3.4}$$

and hence

$$\dot{\psi} = i\mu(t)(D-1)\psi + \begin{bmatrix} -\Gamma+i\Delta & 0 & i\alpha & -i\alpha & 0 \\ 0 & -\Gamma-i\Delta & -i\alpha & i\alpha & 0 \\ i\alpha & -i\alpha & -2\Gamma_1 & 2\Gamma_2^{(0)} & 0 \\ -i\alpha & i\alpha & 2\Gamma_1^{(0)} & -2\Gamma_2 & 2p \\ 0 & 0 & 0 & 0 & 0 \end{bmatrix} \psi . \tag{3.5}$$

Using Eq. (B.7) of the appendix, we immediately find that the ensemble average ψ satisfies

$$\langle\dot{\psi}\rangle = \begin{bmatrix} -\Gamma-\gamma_c+i\Delta & 0 & i\alpha & -i\alpha & 0 \\ 0 & -\Gamma-\gamma_c-i\Delta & -i\alpha & i\alpha & 0 \\ i\alpha & -i\alpha & -2\Gamma_1 & 2\Gamma_2^{(0)} & 0 \\ -i\alpha & i\alpha & 2\Gamma_1^{(0)} & -2\Gamma_2 & 2p \\ 0 & 0 & 0 & 0 & 0 \end{bmatrix} \langle\psi\rangle . \tag{3.6}$$

Using Eqs. (3.3), and (3.6), we can calculate all the relevant mean values (single time) determining the dynamics of a two-level atom. We note that equations for $\langle\psi\rangle$ are identical to the equations for the old set provided we replace Γ by $\Gamma+\gamma_c$. γ_c appears in a more complicated way in the equations for $\langle\chi\rangle$. Using the techniques of Laplace transforms, one finds that

$$\langle\hat{\chi}_i(z)\rangle = \sum_{j=1}^{4} (z-A)^{-1}_{ij} \langle\chi_j(0)\rangle + (z+\gamma_c)^{-1} 2p (z-A)^{-1}_{i4} \langle\chi_5(0)\rangle , \tag{3.7}$$

$$\langle\hat{\psi}_i(z)\rangle = \sum_{j=1}^{4} (z-B)^{-1}_{ij} \langle\psi_j(0)\rangle + z^{-1}(z-B)^{-1}_{i4}(2p) , \tag{3.8}$$

with

$$A = \begin{bmatrix} -\Gamma+i\Delta & 0 & i\alpha & -i\alpha \\ 0 & -\Gamma-i\Delta-4\gamma_c & -i\alpha & i\alpha \\ i\alpha & -i\alpha & -2\Gamma_1-\gamma_c & 2\Gamma_2^{(0)} \\ -i\alpha & i\alpha & 2\Gamma_1^{(0)} & -2\Gamma_2-\gamma_c \end{bmatrix}, \tag{3.9}$$

$$B = \begin{bmatrix} -\Gamma+i\Delta-\gamma_c & 0 & i\alpha & -i\alpha \\ 0 & -\Gamma-i\Delta-\gamma_c & -i\alpha & i\alpha \\ i\alpha & -i\alpha & -2\Gamma_1 & 2\Gamma_2^{(0)} \\ -i\alpha & i\alpha & 2\Gamma_1^{(0)} & -2\Gamma_2 \end{bmatrix} . \tag{3.10}$$

In the steady state one has [provided $(z-B)^{-1}_{ij}$ does not have a zero at $z=0$]

$$\langle\psi_i(t)\rangle \to 2p(-B)^{-1}_{i4}, \qquad \langle\chi(t)\rangle \to 0 \text{ as } t\to\infty . \tag{3.11}$$

It is clear from (3.11) that in the steady state the average value of the dipole moment vanishes because of the phase fluctuations. The matrices appearing in (3.7) and (3.8) are easily found to be (we are quoting only the elements needed in further calculations)

$$(z-A)^{-1}_{11} = P_2^{-1}\{(z+\Gamma+4\gamma_c+i\Delta) \times [(z+\gamma_c+2\Gamma_1)(z+\gamma_c+2\Gamma_2) - 4\Gamma_1^{(0)}\Gamma_2^{(0)}] + 2\alpha^2(z+\gamma_c+\Gamma_1+\Gamma_2-\Gamma_1^{(0)}-\Gamma_2^{(0)})\} , \tag{3.12}$$

$$(z-A)^{-1}_{12} = 2\alpha^2 P_2^{-1}(z+\gamma_c+\Gamma_1+\Gamma_2-\Gamma_1^{(0)}-\Gamma_2^{(0)}) , \tag{3.13}$$

$$(z-A)^{-1}_{13} = i\alpha P_2^{-1}(z+\gamma_c+2\Gamma_2-2\Gamma_1^{(0)}) \times (z+4\gamma_c+\Gamma+i\Delta) , \tag{3.14}$$

$$(z-A)^{-1}_{14} = -i\alpha P_2^{-1}(z+\Gamma+4\gamma_c+i\Delta) \times (z+\gamma_c+2\Gamma_1-2\Gamma_2^{(0)}), \tag{3.15}$$

$$P_2(z) = 4\alpha^2(z+\gamma_c+\Gamma_1+\Gamma_2-\Gamma_1^{(0)}-\Gamma_2^{(0)}) \times (z+\Gamma+2\gamma_c) + (z+\Gamma-i\Delta)(z+\Gamma+4\gamma_c+i\Delta) \times [(z+\gamma_c+2\Gamma_1)(z+\gamma_c+2\Gamma_2) - 4\Gamma_1^{(0)}\Gamma_2^{(0)}] , \tag{3.16}$$

$$(z-B)^{-1}_{44} = P_1^{-1}\{2\alpha^2(z+\Gamma+\gamma_c) + (z+2\Gamma_1)[\Delta^2+(z+\Gamma+\gamma_c)^2]\} , \tag{3.17}$$

$$(z-B)^{-1}_{24} = i\alpha P_1^{-1}(z+2\Gamma_1-2\Gamma_2^{(0)}) \times (z+\Gamma+\gamma_c-i\Delta) , \tag{3.18}$$

$$(z-B)^{-1}_{34} = P_1^{-1}\{2\alpha^2(z+\Gamma+\gamma_c) + 2\Gamma_2^{(0)}[\Delta^2+(z+\Gamma+\gamma_c)^2]\} , \tag{3.19}$$

$$(z-B)^{-1}_{33} = P_1^{-1}\{2\alpha^2(z+\Gamma+\gamma_c) + (z+2\Gamma_2)[\Delta^2+(z+\Gamma+\gamma_c)^2]\} , \tag{3.20}$$

$$P_1(z) = 4\alpha^2(z+\Gamma_1+\Gamma_2-\Gamma_1^{(0)}-\Gamma_2^{(0)})(z+\Gamma+\gamma_c) + [\Delta^2+(z+\Gamma+\gamma_c)^2] \times [(z+2\Gamma_1)(z+2\Gamma_2) - 4\Gamma_1^{(0)}\Gamma_2^{(0)}] . \tag{3.21}$$

It is clear from the above that the dynamics of the population in each level is governed by the roots of P_1, whereas that of the dipole moment is governed by the roots of P_2. In the absence of laser fluctuations one, of course, has $P_1=P_2$. Note that in Torrey's case [Eq. (2.4)], we have (at resonance $\Delta=0$)

$$P_2=(z+\gamma_c+2p)[4\alpha^2(z+2\gamma_c+1/T_2)+(z+1/T_2+4\gamma_c)\times(z+1/T_2)(z+\gamma_c+1/T_1)], \quad (3.22)$$

$$P_1=(z+2p)(z+\gamma_c+1/T_2)\times[4\alpha^2+(z+1/T_1)(z+1/T_2+\gamma_c)]. \quad (3.23)$$

It is clear from (3.23) that the threshold of oscillations in the population inversion is determined by

$$16\alpha_{\text{th}}^2=(1/T_2+\gamma_c-1/T_1)^2.$$

In the limit of strong fields ($\alpha \gg$ all the damping constants in the problem) and at resonance the roots of P_1 and P_2 are

$$P_1(z)=0 \Rightarrow z=-(\Gamma+\gamma_c),\ -(\Gamma_1+\Gamma_2-\Gamma_1^{(0)}-\Gamma_2^{(0)}),\ \pm 2i\alpha-\tfrac{1}{2}(\gamma_c+\Gamma+\Gamma_1+\Gamma_2+\Gamma_1^{(0)}+\Gamma_2^{(0)}), \quad (3.24)$$

$$P_2(z)=0 \Rightarrow z=-(\Gamma+2\gamma_c),\ -(\gamma_c+\Gamma_1+\Gamma_2-\Gamma_1^{(0)}-\Gamma_2^{(0)}),\ \pm 2i\alpha-\tfrac{1}{2}(3\gamma_c+\Gamma+\Gamma_1+\Gamma_2+\Gamma_1^{(0)}+\Gamma_2^{(0)}). \quad (3.25)$$

In Torrey's case the roots of P_2 are given approximately by

$$z=-(2p+\gamma_c),\ -(2\gamma_c+1/T_2),\ \pm 2i\alpha-\tfrac{1}{2}(3\gamma_c+1/T_1+1/T_2), \quad (3.26)$$

which amounts to the replacement in the usual Torrey solutions:

$$1/T_2 \to 1/T_2+2\gamma_c,\quad 1/T_1\to 1/T_1+\gamma_c. \quad (3.27)$$

Note that in the steady state the population distribution in the upper level is given by

$$\langle\rho_{11}(\infty)\rangle=\frac{p[\Gamma_2^{(0)}+\alpha_c^2\Gamma(\Delta^2+\Gamma^2)^{-1}]}{(\Gamma_1\Gamma_2-\Gamma_1^{(0)}\Gamma_2^{(0)})+\alpha_c^2\Gamma(\Delta^2+\Gamma^2)^{-1}(\Gamma_1+\Gamma_2-\Gamma_1^{(0)}-\Gamma_2^{(0)})},$$
$$\alpha_c^2=\Gamma^{-1}(\Delta^2+\Gamma^2)(\Gamma+\gamma_c)[\Delta^2+(\Gamma+\gamma_c)^2]^{-1}\alpha^2, \quad (3.28)$$

and that the dipole moment in the steady state is zero. It is seen that $\alpha_c=\alpha$ if $\gamma_c=0$. Moreover it is clear that to achieve a given amount of saturation the fields have to be stronger by the factor

$$\Gamma[\Delta^2+(\Gamma+\gamma_c)^2](\Gamma+\gamma_c)^{-1}(\Delta^2+\Gamma^2)^{-1}.$$

It should also be noticed that the state of the atom at time t is not a coherent state[42] even if we ignore all relaxation and pump mechanisms except laser fluctuations. We will now deal with specific optical effects.

1. *Optical induction decay*

The usual theory of optical induction[2,3] holds even in the presence of a fluctuating laser beam if we make the replacement $\Gamma\to\Gamma+\gamma_c$, showing that the laser linewidth simply adds to the linewidth associated with the decay of off-diagonal elements. This follows from the fact that the optical induction signal is determined by the interference of the laser fields and the scattered field. The interference term is proportional to $\langle\rho_{12}e^{i\varphi}\rangle$ whose approximate value is

$$\langle\rho_{12}e^{i\varphi(t)}\rangle=\langle\rho_{12}e^{i\varphi}\rangle\big|_{t=0}\exp[-(\Gamma+\gamma_c-i\Delta)t]. \quad (3.29)$$

Note that the atoms are shifted out of resonance by the Stark field. The average value appearing in (3.29) is given by (3.11) with $i=2$.

2. *Adiabatic following*

So far we have considered the laser amplitude to be independent of time. If $\mathcal{E}$ is a function of time, then explicit solutions can be obtained in the adiabatic limit,[19-22] i.e., when the rate of change of the field is much less than the rate of change of the Bloch vector $\langle\vec{S}\rangle$, as for example will be the case when the applied field is far off the resonance. When the applied field is close to the resonance, then the generalized adiabatic approximation of Ref. 21 is to be used. In what follows we assume that our two-level atom is driven by the "phase fluctuating" laser beam and we ignore for the sake of simplicity all the relaxation mechanisms. The ensemble average of

$$\Phi_1=\psi_1,\quad \Phi_2=\psi_2,\quad \Phi_3=\langle S^z\rangle=\tfrac{1}{2}(\psi_3-\psi_4),$$

satisfies the equation

$$\langle\dot{\Phi}\rangle=\begin{bmatrix}-\gamma_c+i\Delta & 0 & 2i\alpha(t)\\ 0 & -\gamma_c-i\Delta & -2i\alpha(t)\\ i\alpha(t) & -i\alpha(t) & 0\end{bmatrix}\langle\Phi\rangle. \quad (3.30)$$

On integrating the equation for Φ_1 we get in the usual manner

$$i\alpha(t)\langle\Phi_1\rangle=\sum_0^\infty\frac{2\alpha(t)}{(i\Delta-\gamma_c)^{n+1}}\frac{d^n}{dt^n}[\alpha(t)\langle\Phi_3(t)\rangle]. \quad (3.31)$$

We now substitute the first two terms of (3.31) in the equation for Φ_3 and obtain the analytical solution for $\langle\Phi_3\rangle$:

$$\langle\Phi_3(t)\rangle = -\frac{1}{2}\left(1-\frac{4(\gamma_c^2-\Delta^2)}{(\gamma_c^2+\Delta^2)^2}\alpha^2(t)\right)^{-1/2} \times \exp\left[\frac{-4\gamma_c}{(\gamma_c^2+\Delta^2)}\int_{-\infty}^{t} d\tau\, \alpha^2(\tau) \times\left(1-\frac{4(\gamma_c^2-\Delta^2)}{(\gamma_c^2+\Delta^2)^2}\alpha^2(\tau)\right)^{-1}\right]. \quad (3.32)$$

On substituting (3.32) in (3.31), we obtain the adiabatic solutions for the dipole moment in the instantaneous frame

$$\mathrm{Re}\langle\langle s^+(t)\rangle e^{-i\varphi(t)}\rangle = \frac{-\Delta}{(\gamma_c^2+\Delta^2)}\langle\alpha\Phi_3\rangle + \frac{2\gamma_c\Delta}{(\gamma_c^2+\Delta^2)^2}\frac{d}{dt}\langle\alpha\Phi_3\rangle, \quad (3.33)$$

$$\mathrm{Im}\langle\langle s^+(t)\rangle e^{-i\varphi(t)}\rangle = \frac{\gamma_c}{(\gamma_c^2+\Delta^2)}\langle\alpha\Phi_3\rangle - \frac{(\gamma_c^2-\Delta^2)}{(\gamma_c^2+\Delta^2)^2}\frac{d}{dt}\langle\alpha\Phi_3\rangle. \quad (3.34)$$

The adiabatic solutions for the dipole moments are similarly found. We start from the ensemble average of the equations for $f_1=\chi_1$, $f_2=\chi_2$, $f_3=\frac{1}{2}(\chi_3-\chi_4)$,

$$\langle\dot{f}\rangle = \begin{bmatrix} i\Delta & 0 & -2i\alpha(t) \\ 0 & -4\gamma_c - i\Delta & +2i\alpha(t) \\ i\alpha(t) & -i\alpha(t) & -\gamma_c \end{bmatrix}\langle f\rangle. \quad (3.35)$$

Note that if at $t=-\infty$, the atom is in the ground state, then $\chi_1(-\infty)=\chi_2(-\infty)=0$, $f_3(-\infty) = -\frac{1}{2}\langle e^{i\varphi_0}\rangle f_3(-\infty)$ will also be zero if we assume that φ_0 is randomly distributed. For such cases $\langle f_1(t)\rangle = 0$. Hence in what follows we leave the distribution of φ_0 arbitrary.

On integrating the $\langle f_1\rangle$ and $\langle f_2\rangle$ equations, we find

$$i\alpha(t)\langle f_1(t)\rangle = \sum_0^\infty 2(i\Delta)^{-n-1}\alpha(t)\frac{d^n}{dt^n}[\alpha(t)\langle f_3(t)\rangle], \quad (3.36)$$

$$-i\alpha(t)\langle f_2(t)\rangle = \sum_0^\infty 2(-1)^{n+1}(4\gamma_c+i\Delta)^{-n-1} \times\alpha(t)\frac{d^n}{dt^n}\langle\alpha(t)f_3(t)\rangle. \quad (3.37)$$

The $n=0$ and $n=1$ terms of (3.36) and (3.37) when inserted in (3.35) lead to

$$\langle f_3(t)\rangle = f_3(-\infty)/[1-2B\alpha^2(t)]^{1/2} \times\exp\left(\int_{-\infty}^{t} d\tau\,\frac{(2A\alpha^2(\tau)-\gamma_c)}{(1-2B\alpha^2(\tau))}\right), \quad (3.38)$$

where A and B are given by

$$A = 4\gamma_c(4i\Delta\gamma_c-\Delta^2)^{-1}, \quad B = \frac{\Delta^2-(i\Delta+4\gamma_c)^2}{(i\Delta+4\gamma_c)^2\Delta^2}. \quad (3.39)$$

The real and imaginary parts of the ensemble average of the dipole moment are obtained from (3.36) and (3.37). Note that since $\langle f_3\rangle$ is not real, the $n=1$ term of (3.36) also contributes to the real part of the dipole moment. In the limit $\gamma_c\to 0$, the adiabatic equations (3.32), (3.38), and (3.36) become the usual ones.

B. General structure of the ensemble average of the atomic correlation functions

We next discuss various types of correlation functions associated with a two-level atom. Such correlation functions describe, among other things, spectrum of resonance fluorescence, energy absorption from a weak external field, etc. Let us define

$$\begin{aligned} \Gamma_1^{(N)}(t+\tau,t) &= \langle\langle S^+(t+\tau)S^-(t)\rangle\rangle, \\ \Gamma_1^{(A)}(t+\tau,t) &= \langle\langle S^-(t)S^+(t+\tau)\rangle\rangle, \\ \Gamma_2^{(N)}(t+\tau,t) &= \langle\langle S^+(t)S^+(t+\tau)S^-(t+\tau)S^-(t)\rangle\rangle, \end{aligned} \quad (3.40)$$

where double brackets $\langle\langle\ \rangle\rangle$ indicate the quantum-mechanical averaging with respect to the density matrix of the system and the classical ensemble averaging with respect to the temporal distribution of the phase φ.

As discussed earlier our extended set of variables undergoes Markovian motion and hence all the correlation functions can be calculated by using quantum regression theorem.[43,44] Let us define Laplace transforms of the correlation functions by

$$\hat{\Gamma}(t,z) = \int_0^\infty d\tau\, e^{-z\tau}\,\Gamma(t+\tau,t). \quad (3.41)$$

We will indicate the use of quantum regression theorem in the case of $\Gamma_1^{(N)}$. Using the solution of (3.3) we find

$$\int_0^\infty d\tau\, e^{-z\tau}\langle\langle S^+(t+\tau)\rangle\rangle = (z-A)^{-1}_{11}\langle\langle S^+(t)\rangle\rangle + (z-A)^{-1}_{12}\langle\langle S^-(t)e^{2i\varphi(t)}\rangle\rangle + (z-A)^{-1}_{13}\langle\langle\,|1\rangle\langle 1|\,e^{i\varphi(t)}\rangle\rangle + (z-A)^{-1}_{14}\langle\langle\,|2\rangle\langle 2|\,e^{i\varphi(t)}\rangle\rangle + 2p(z+\gamma_c)^{-1}(z-A)^{-1}_{14}\langle\langle e^{i\varphi(t)}\rangle\rangle.$$

Using quantum regression theorem we then get for the correlation function

$$\begin{aligned}\hat{\Gamma}_1^{(N)}(t,z) &= (z-A)^{-1}_{11}\langle\langle S^+(t)S^-(t)\rangle\rangle + (z-A)^{-1}_{12}\langle\langle S^-(t)e^{2i\varphi(t)}S^-(t)\rangle\rangle + (z-A)^{-1}_{13}\langle\langle\,|1\rangle\langle 1|\,e^{i\varphi(t)}S^-(t)\rangle\rangle \\ &\quad + (z-A)^{-1}_{14}\langle\langle\,|2\rangle\langle 2|\,e^{i\varphi(t)}S^-(t)\rangle\rangle + 2p(z+\gamma_c)^{-1}(z-A)^{-1}_{14}\langle\langle e^{i\varphi(t)}S^-(t)\rangle\rangle \\ &= (z-A)^{-1}_{11}\langle\psi_3(t)\rangle + (z-A)^{-1}_{14}(z+\gamma_c)^{-1}(z+\gamma_c+2p)\langle\psi_2(t)\rangle. \end{aligned} \quad (3.42)$$

Using similar methods, we find for the antinormally ordered correlation function the expression

$$\hat{\Gamma}_1^{(A)}(t,z)=(z-A)^{-1}_{11}\langle\psi_4(t)\rangle+[(z-A)^{-1}_{13}+2p(z+\gamma_c)^{-1}(z-A)^{-1}_{14}]\times\langle\psi_2(t)\rangle\,, \tag{3.43}$$

and for the intensity correlation function the form

$$\hat{\Gamma}_2^{(N)}(t,z)=\langle\psi_3(t)\rangle[(z-B)^{-1}_{34}z^{-1}(z+2p)]=\langle\psi_3(t)\rangle\langle\hat{\psi}_3'(z)\rangle\,, \tag{3.44}$$

where ψ_3' represents the value of ψ_3 if at $t=0$ the atom is assumed to be in the ground state. Note that even in presence of the laser fluctuations the intensity correlations have the time factorization property of Ref. 45. In what follows we quote explicit expressions only for the steady-state correlation functions.

1. Intensity correlations–photon antibunching effects

For the sake of simplicity we also restrict the discussion to the Torrey type of relaxation equations. Using (3.19) in (3.44), we find that the normalized intensity correlation function has the structure

$$\Gamma(t)=\Gamma_2^{(N)}(\infty,t)/(\langle\langle S^+S^-\rangle\rangle_\infty)^2=1+(z_+-z_-)^{-1}(z_-e^{-z_+t}-z_+e^{-z_-t})+\frac{z_+z_-\Gamma_2^{(0)}}{z_+-z_-}(e^{-z_+t}-e^{-z_-t})\times\left[\alpha^2+\Gamma_2^{(0)}\left(\gamma_c+\frac{1}{T_2}\right)\right]^{-1}, \tag{3.45a}$$

where $z_\pm$ are the roots of

$$(z+1/T_1)(z+\gamma_c+1/T_2)+4\alpha^2=0\,. \tag{3.45b}$$

For large fields (3.45) shows that the envelope of the oscillation decays at the rate $\frac{1}{2}(\gamma_c+1/T_1+1/T_2)$ showing that the laser linewidth adds simply to the linewidths associated with T_1 and T_2.

2. Normally ordered amplitude correlations–spectrum of the scattered radiation

The amplitude correlation is similarly obtained by using (3.12)–(3.16) in (3.42). The final result is

$$\hat{\Gamma}_1^{(N)}(\infty,z)=4pP_2^{-1}(z)P_1^{-1}(0)\big((z+\gamma_c)^{-1}(z+\gamma_c+2p)(\Gamma+\gamma_c-i\Delta)(\Gamma_1-\Gamma_2^{(0)})(z+\Gamma+i\Delta+4\gamma_c)(z+\gamma_c+2\Gamma_1-2\Gamma_2^{(0)})+\{\alpha^2(\Gamma+\gamma_c)+\Gamma_2^{(0)}[\Delta^2+(\Gamma+\gamma_c)^2]\}\{2\alpha^2(z+\gamma_c+\Gamma_1+\Gamma_2-\Gamma_1^{(0)}-\Gamma_2^{(0)})+(z+\Gamma+4\gamma_c+i\Delta)[(z+\gamma_c+2\Gamma_1)(z+\gamma_c+2\Gamma_2)-4\Gamma_1^{(0)}\Gamma_2^{(0)}]\}\big)\,, \tag{3.46}$$

which for the Torrey type of relaxation mechanism reduces to

$$\hat{\Gamma}_1^{(N)}(\infty,z)=2p\,P_1^{-1}(0)(z+\gamma_c+2p)P_2^{-1}(z)\,Y(z)\,,$$

$$Y(z)=\left[2\alpha^2+\left(z+\gamma_c+\frac{1}{T_1}\right)\left(z+4\gamma_c+\frac{1}{T_2}+i\Delta\right)\right]\left\{2\alpha^2\left(\gamma_c+\frac{1}{T_2}\right)+\frac{E+\frac{1}{2}}{T_1}\left[\Delta^2+\left(\gamma_c+\frac{1}{T_2}\right)^2\right]\right\}+\alpha^2(z+\gamma_c)^{-1}\times\left(z+4\gamma_c+i\Delta+\frac{1}{T_2}\right)\left(z+\gamma_c-\frac{2E}{T_1}\right)\left(\gamma_c+\frac{1}{T_2}-i\Delta\right)\left(\frac{-2E}{T_1}\right),\quad E\equiv\langle S^z\rangle_{eq}\,. \tag{3.47}$$

In the special cases of the radiative and collisional relaxations, (3.47) reduces to

$$Y(z)=2\alpha^2(\gamma+\gamma_c)[2\alpha^2+(z+\gamma_c+2\gamma)(z+4\gamma_c+\gamma+i\Delta)]+2\alpha^2(z+\gamma_c)^{-1}(z+4\gamma_c+i\Delta+\gamma)(z+\gamma_c+2\gamma)(\gamma_c+\gamma-i\Delta)\gamma\,, \tag{3.48}$$

$$Y(z)=\alpha^2(z+\gamma_c)^{-1}(z+4\gamma_c+i\Delta+\kappa)(z+\gamma_c-2E\kappa)(\gamma_c+\kappa-i\Delta)(-2E\kappa)+\{2\alpha^2(\gamma_c+\kappa)+\kappa(E+\tfrac{1}{2})[\Delta^2+(\gamma_c+\kappa)^2]\}\times[2\alpha^2+(z+\gamma_c+\kappa)(z+4\gamma_c+\kappa+i\Delta)],\quad E\equiv\beta-\tfrac{1}{2}\,. \tag{3.49}$$

The power spectrum of the radiation scattered from such a two-level atom is obtained from

$$S(\omega)=\mathrm{Re}\,\hat{\Gamma}_1^{(N)}(\infty,i\omega)\,. \tag{3.50}$$

For the case of incident laser fields at *resonance* several features of the spectrum are to be noted: (i) the spectrum is symmetric; (ii) the coherent part of the spectrum present in the theory with the nonfluctuating beam is now broadened by the amount of the laser linewidth; (iii) in the limit of strong laser fields $\alpha\gg 1/T_1,\ 1/T_2,\ \gamma_c$, one finds the three-peak spectrum. Let us introduce the parameter

$$x\equiv\frac{\gamma_c+\frac{1}{3}(1/T_1+1/T_2)}{2\gamma_c+1/T_2}\,; \tag{3.51}$$

then one gets the following results for the ratios of peak heights and widths: (i) center peak height per side peak height is $3x$; (ii) center peak width per side peak width is $2/3x$; Note further that in the special cases of (iii) radiative relaxation, $x=(\gamma_c+\gamma)/(2\gamma_c+\gamma)$; (iv) collisional relaxation, $x=(\gamma_c+\frac{2}{3}\kappa)/(2\gamma_c+\kappa)$.

In the limit when phase fluctuations are dominant over $1/T_1$, $1/T_2$ then $x\sim\frac{1}{2}$, which should be compared with the value $\frac{1}{3}(1+T_2/T_1)$ in the absence of laser phase fluctuations. In the absence of laser phase fluctuations and the two special cases of radiative and collisional relaxations, the above results agree with those of Mollow.[4,31] In the presence of laser phase fluctuations the results for the widths and peak heights are in agreement with those of Eberly[26] and the author's[25] as well as Avan and Cohen-Tannoudji's[27] results on the radiative relaxation case.[46]

We show in Figs. 1 and 2 the shape of the spectrum for different values of α and γ_c. Figure 2 clearly shows the three-peak spectrum—the widths and peak heights are in agreement with the ratios $2/3x$, $3x$. Figure 1 shows the behavior of the spectrum for field strengths less than the threshold value. The broadening of the spectrum due to laser phase fluctuations is to be noted. The very sharp peak in Fig. 2 corresponds to $\gamma_c=0.01$ and in the limit $\gamma_c\to 0$, this peak transforms into the δ function. Finally when $\gamma_c\gtrsim\alpha$ then the three-peak spectrum disappears and one finds a single broad peak. For the off-resonant case $\Delta\neq 0$, the spectrum $S(\omega)$ has the property $S(\Delta,\omega)=S(-\Delta,-\omega)$. The explicit form of the spectrum is obtained from (3.48). For the case of radiative relaxation $1/2T_1=\gamma$, $1/T_2=\gamma$, $\langle S^z\rangle_{eq}=-\frac{1}{2}$, the result obtained from (3.48) coincides with that of Kimble and Mandel.[47] Kimble and Mandel have studied the detuned spectrum in detail and have discussed many of the asymmetry properties of the spectrum. It should be remembered that the method leading to Eq. (3.48) is exact whereas the method of Kimble and Mandel[47] is seemingly an approximate one obtained by using a suitable decorrelation. The reasons for the coincidence of the results obtained by the two methods are given in Appendix B.

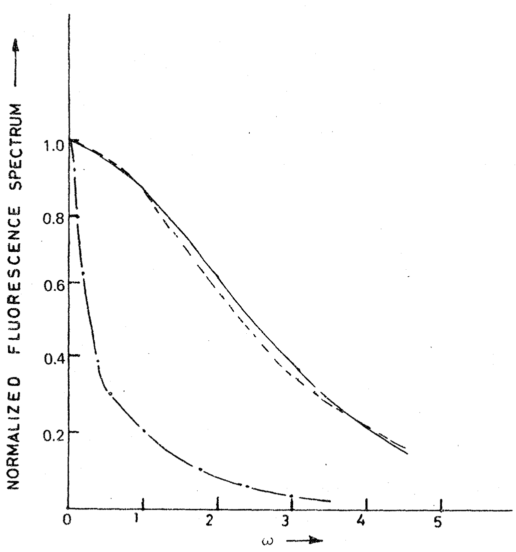

FIG. 1. Normalized fluorescence spectrum as a function of ω. γ has been set equal to 1. The curves correspond to (i) solid $\gamma_c=2$, $\alpha=2$. (ii) Dashed $\gamma_c=2$, $\alpha=0.5$ (iii) Dashed-dotted $\gamma_c=0.2$, $\alpha=0.5$. For the dashed and dashed-dotted curves one unit on x axis is equal to 0.4.

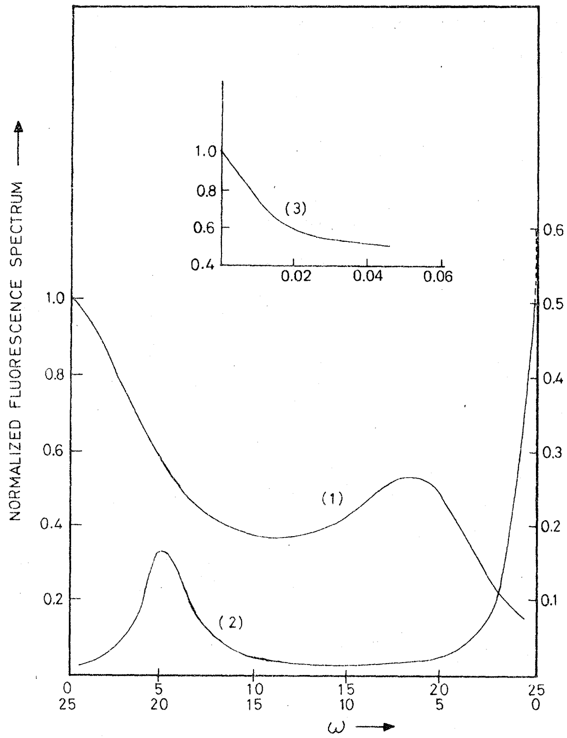

FIG. 2. Normalized fluorescence spectrum as a function of ω for $\alpha=10$ and for (1) $\gamma_c=2$ (2) $\gamma_c=0.01$. The behavior of the peak arising entirely due to laser fluctuations is shown by curve (3) for the case $\gamma_c=0.01$ $\alpha=10$.

3. *Linear response to an applied weak field–absorption spectrum*

It is well known that the power absorbed from a weak field by a system is proportional to the imaginary part of the appropriate linear susceptibility which is related the correlation function[48] of the form $\langle[S^+(t),S^-(0)]\rangle$. Hence the absorption spectrum is essentially proportional to Re $[\hat{\Gamma}_1^{(A)}(\infty, i\omega)-\hat{\Gamma}_1^{(N)}(\infty, i\omega)]$. Using (3.41), (3.42), and (3.12)–

(3.21), one finds that the absorption spectrum has the form

$$S(\omega)=\mathrm{Re}[\hat{\Gamma}_1^{(A)}(\infty, i\omega)-\hat{\Gamma}_1^{(N)}(\infty, i\omega)]$$
$$=4p(\Gamma+\gamma_c)(\Gamma_1-\Gamma_2^{(0)})P_1^{-1}(0)\,\mathrm{Re}(z+\gamma_c+2p)P_2^{-1}(z)$$
$$\times\left[\left(z+4\gamma_c+\frac{1}{T_2}\right)\left(z+\gamma_c+\frac{1}{T_1}\right)\left(\gamma_c+\frac{1}{T_2}\right)-2\alpha^2(z+3\gamma_c)\right], \quad (3.52)$$

where we have specialized to Torrey type of relaxation mechanism. Thus the absorption spectrum is determined by

$$x(\omega)=(A_1A_2+B_1B_2)(A_2^2+B_2^2)^{-1}, \quad (3.53)$$

with

$$A_1=\left(\gamma_c+\frac{1}{T_2}\right)\left[\left(\gamma_c+\frac{1}{T_1}\right)\left(4\gamma_c+\frac{1}{T_2}\right)-\omega^2\right]-6\gamma_c\alpha^2,$$
$$B_1=\omega\left[\left(\gamma_c+\frac{1}{T_2}\right)\left(5\gamma_c+\frac{1}{T_1}+\frac{1}{T_2}\right)-2\alpha^2\right],$$
$$A_2=4\alpha^2\left(2\gamma_c+\frac{1}{T_2}\right)+\frac{1}{T_2}\left(\gamma_c+\frac{1}{T_1}\right)\left(4\gamma_c+\frac{1}{T_2}\right)-\omega^2\left(5\gamma_c+\frac{1}{T_1}+\frac{2}{T_2}\right),$$
$$B_2=\omega\left[4\alpha^2-\omega^2+\left(\frac{2}{T_2}+4\gamma_c\right)\left(\gamma_c+\frac{1}{T_1}\right)+\frac{1}{T_2}\left(\frac{1}{T_2}+4\gamma_c\right)\right]. \quad (3.54)$$

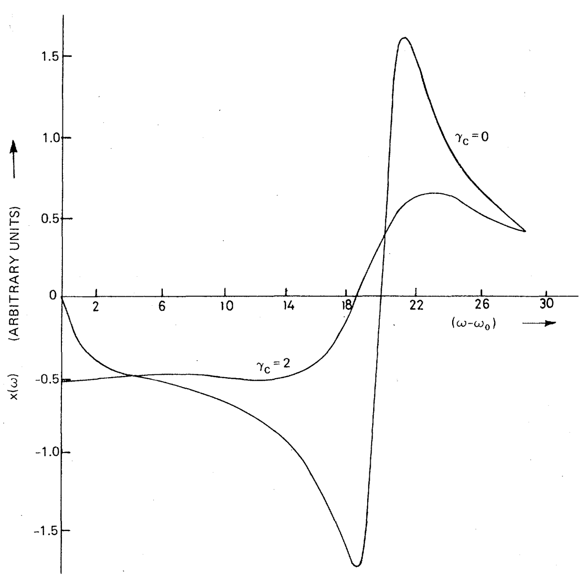

FIG. 3. Absorption spectrum as a function of $\omega-\omega_0$ for the radiative relaxation of the atom.

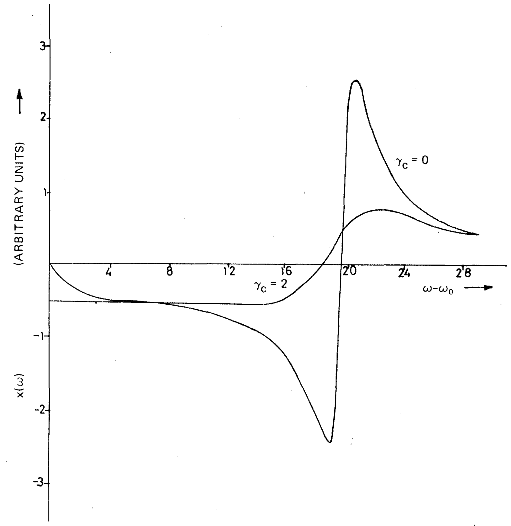

FIG. 4. Same as in Fig. 3 but for the collisional relaxation of the atom.

Note that the spectrum $x(\omega)$ is symmetric. In the special case when $\gamma_c=0$, the result (3.53) is equivalent to that of Mollow.[28] We have plotted (3.53) for two special cases $1/T_1=1/T_2=1$ (collisional relaxation) and $1/T_1=2$, $1/T_2=1$ (radiative relaxation) in Figs. 3 and 4 for the case of strong fields and for $\gamma_c \ll \alpha$. It is seen that the laser fluctuations reduce considerably the strength of attenuation and amplification of the external weak field. The spectrum changes very slowly from the negative values to positive values compared to the case when the nonfluctuating laser beam is used to drive the two-level atom. Note also that the spectrum $x(\omega)$ is independent of temperature which is in contrast to the case of scattered field spectrum, given by Eq. (3.49), which depends in an important way on temperature (β).

IV. INTERACTION OF A MULTILEVEL SYSTEM WITH FLUCTUATING LASER FIELDS–HANLE RESONANCES

Multilevel systems show a variety of interesting optical effects[16,17,49] with laser fields. Such optical effects include optical double resonance experiments and Hanle resonances. It would be interesting to study how the laser fluctuations affect the spectrum of optical double resonance as well as the intensity of level-crossing signals. It should be noted that the problem of resonance fluorescence from a collection of atoms is also an exercise in the interaction of a multilevel system with a resonance field. It will be clear from the treatment given in Sec. III that the laser fluctuations can be incorporated in the theory. One has to find the set of variables such that the set satisfies equations of multiplicative stochastic processes. For example let us discuss briefly the resonance fluorescence from a collective atomic system. The master equation describing the atomic dynamics[14,15] is

$$\dot{\rho} = -\gamma(S^+S^-\rho - 2S^-\rho S^+ + \rho S^+S^-) + i\alpha[S^+e^{-i\varphi} + \text{c.c.}, \rho], \tag{4.1}$$

where $\varphi(t)$ is the fluctuating phase variable (2.7). It is clear from (4.1) that if we introduce variables

$$\rho_{nm}^{(1)} = \rho_{nm}\exp[i(n-m)\varphi(t)], \tag{4.2}$$

then

$$\dot{\rho}^{(1)} = -\gamma(S^+S^-\rho^{(1)} - 2S^-\rho^{(1)}S^+ + \rho^{(1)}S^+S^-) + i\alpha[S^+ + S^-, \rho^{(1)}] + i\mu(t)[S^z, \rho^{(1)}], \tag{4.3}$$

and hence the ensemble average of $\rho^{(1)}$ satisfies

$$\langle\dot{\rho}^{(1)}\rangle = -\gamma(S^+S^-\langle\rho^{(1)}\rangle - 2S^-\langle\rho^{(1)}\rangle S^+ + \langle\rho^{(1)}\rangle S^+S^-) + i\alpha[S^+ + S^-, \langle\rho^{(1)}\rangle] - \gamma_c[S^z, [S^z, \langle\rho^{(1)}\rangle]]. \tag{4.4}$$

It is clear that the ensemble average of $\rho^{(1)}$ enables one to calculate the ensemble average of the diagonal elements of ρ. The ensemble average of the correlation functions involving the atomic operators diagonal in $|n\rangle$ representation can be calculated from (4.4) and the quantum regression theorem. In order to calculate the dipole moment and the dipole moment correlation functions, one can introduce the variables

$$\rho_{nm}^{(2)} = \rho_{nm}\exp[i(n-m+1)\varphi(t)], \tag{4.5}$$

since

$$\langle S^+(t)\rangle = \sum \rho_{nm}S_{mn}^+ = \sum \rho_{n,n+1}\langle n+1|S^+|n\rangle = \mathrm{Tr}[\rho^{(2)}(t)S^+], \tag{4.6}$$

and thus $\langle\langle S^+(t)\rangle\rangle$ can be calculated from the ensemble average of $\rho^{(2)}$. The ensemble average of $\rho^{(2)}$ satisfies the *exact* master equation

$$\langle\dot{\rho}^{(2)}\rangle = -\gamma(S^+S^-\langle\rho^{(2)}\rangle - 2S^-\langle\rho^{(2)}\rangle S^+ + \langle\rho^{(2)}\rangle S^+S^-) + i\alpha[S^+ + S^-, \langle\rho^{(2)}\rangle] - \gamma_c\{[S^z, [S^z, \langle\rho^{(2)}\rangle]] + \rho^{(2)} + 2[S^z, \langle\rho^{(2)}\rangle]\}. \tag{4.7}$$

It is thus clear that in the presence of laser fluctuations one has to solve different types of master equations [cf. analogous relations (3.3) and (3.6)]. Of course, master equations like (4.4) and (4.7) have to be solved on a computer (cf. calculations in Refs. 15 and 50).

A similar analysis can be carried out for optical double resonance[16] experiments—one has to solve numerically 9×9 matrices. As this becomes a problem in computational physics, we do not discuss it here. We will however present the analysis of Hanle resonances for $J=0$ to $J=1$ transition excited by a fluctuating laser field. This system has been analyzed quite recently by Avan and Cohen-Tannoudji.[17] We will show how the theory of the multiplicative stochastic processes can be applied to obtain exact results. The analysis is rather simple as the following statement will show. The model for the Hanle resonance is same as that of Avan and Cohen-Tannoudji. In the equivant spin-1 space the density matrix ρ obeys the equation

$$\dot{\rho} = -i[H, \rho] + L_{\text{incoh}}\rho, \tag{4.8}$$

where

$$H = \Omega\begin{bmatrix}1 & 0 & 0\\ 0 & 0 & 0\\ 0 & 0 & -1\end{bmatrix} + \begin{bmatrix}0 & \alpha_0 & 0\\ \alpha_0^* & 0 & \alpha_0^*\\ 0 & \alpha_0 & 0\end{bmatrix}, \quad \alpha_0 = \alpha e^{-i\varphi(t)}, \tag{4.9}$$

and $L_{\text{incoh}}\rho$ represents the damping part

$$(L_{\text{incoh}}\rho)_{ij} = -\Gamma\rho_{ij} + \Gamma\delta_{ij}\delta_{i2} + \tfrac{1}{2}\Gamma_{ij}^1\rho_{ij}, \tag{4.10}$$

where Γ^1 is a symmetric matrix and its only nonvanishing elements are $\Gamma_{12}^1 = \Gamma_{23}^1 = 1$ and 2Ω represents the splitting between the two Zeeman levels. As is well known the level-crossing signal is determined by the steady-state behavior of $\rho_{11}+\rho_{33}$, $\mathrm{Re}\,\rho_{13}$. We now introduce the set of variables

$$\psi_1 = \rho_{13}, \quad \psi_2 = \psi_1^*, \quad \psi_3 = \rho_{23}e^{-i\varphi}, \quad \psi_4 = \psi_3^*, \quad \psi_5 = e^{i\varphi}\rho_{12}, \quad \psi_6 = \psi_5^*, \quad \psi_7 = \rho_{11}, \quad \psi_8 = \rho_{33}. \tag{4.11}$$

It is easily seen that the set (4.11) satisfies equations of multiplicative stochastic processes:

$$\begin{aligned}
\dot{\psi}_1 &= -(\Gamma + 2i\Omega)\psi_1 - i\alpha(\psi_3 - \psi_5),\\
\dot{\psi}_7 &= -\Gamma\psi_7 - i\alpha(\psi_6 - \psi_5),\\
\dot{\psi}_8 &= -\Gamma\psi_8 - i\alpha(\psi_3 - \psi_4),\\
\dot{\psi}_3 &= -(\Gamma/2 + i\Omega)\psi_3 - i\dot{\varphi}\,\psi_3 - i\alpha(\psi_1 + 2\psi_8 - 1 + \psi_7),\\
\dot{\psi}_5 &= -(\Gamma/2 + i\Omega)\psi_5 + i\dot{\varphi}\,\psi_5 - i\alpha(1 - 2\psi_7 - \psi_8 - \psi_1),
\end{aligned} \tag{4.12}$$

which can be written in matrix form (ψ a column matrix having eight components)

$$\dot{\psi} = A\psi - i\mu(t)I\psi + i\alpha I\beta, \quad I_{33} = -I_{44} = -I_{55} = I_{66} = 1, \quad \beta_i = 1 \text{ for every } i, \tag{4.13}$$

the remaining elements of I being zero. Hence the ensemble average of ψ satisfies the exact equations

$$\langle\dot{\psi}\rangle = A\langle\psi\rangle + i\alpha I\beta - \gamma_c\Lambda\langle\psi\rangle, \quad \Lambda_{ii} = 1 \text{ for } i = 3, 4, 5, 6, \tag{4.14}$$

and where the remaining elements of Λ are zero.

Eq. (4.14) and the regression theorem leads to the complete dynamics of the system. In the steady state one will have

$$\langle\psi\rangle = i\alpha(\gamma_c \Lambda - A)^{-1} I\beta . \quad (4.15)$$

Note that one need not invert the 8×8 matrix $(\gamma_c\Lambda - A)$. Equations (4.14) can be solved directly for the steady-state values. A simple analysis shows that

$$\langle\psi_7\rangle = \langle\psi_8\rangle, \quad \langle\psi_5\rangle = -\langle\psi_3\rangle, \quad \langle\psi_6\rangle = -\langle\psi_4\rangle, \quad (4.16)$$

$$\langle\psi_3\rangle = i\mathfrak{D}^{-1}(\tfrac{1}{2}\gamma + i\Omega)^{-1}[1 + (\tfrac{1}{2}\gamma - i\Omega)^{-1}(\tfrac{1}{2}\Gamma - i\Omega)^{-1}], \quad (4.17)$$

$$\langle\psi_1\rangle = \langle\rho_{13}\rangle = \mathfrak{D}^{-1}(\tfrac{1}{4}\Gamma^2 + \Omega^2)^{-1}(\tfrac{1}{4}\gamma^2 + \Omega^2)^{-1} \times [1 - \Omega^2 + \tfrac{1}{4}\Gamma\gamma - \tfrac{1}{2}i\Omega(\Gamma + \gamma)], \quad (4.18)$$

$$\langle\psi_8\rangle = \langle\rho_{33}\rangle = \mathfrak{D}^{-1}(\tfrac{1}{4}\gamma^2 + \Omega^2)^{-1}(\tfrac{1}{4}\Gamma^2 + \Omega^2)^{-1} \times [1 + (\gamma/\Gamma)(\Omega^2 + \tfrac{1}{4}\Gamma^2)], \quad (4.19)$$

where

$$\gamma = \Gamma + 2\gamma_c,$$
$$\mathfrak{D} = 1 + (3\gamma/\Gamma)(\Omega^2 + \tfrac{1}{4}\gamma^2)^{-1} + (\Omega^2 + \tfrac{1}{4}\Gamma^2)^{-1} \times (\Omega^2 + \tfrac{1}{4}\gamma^2)^{-1}(4 + \tfrac{1}{2}\gamma\Gamma - 2\Omega^2). \quad (4.20)$$

Note that in the limit of $\Omega \gg \Gamma, \gamma$ one gets

$$\mathrm{Re}\langle\rho_{13}\rangle \approx \frac{\Omega^2 - 1}{2\Omega^2 - \Omega^4 - 4 - (3\gamma\Omega^2/\Gamma)} . \quad (4.21)$$

In this limit the results are in agreement with Avan and Cohen-Tannoudji.[17] Since the curves giving the behavior for different values are given in Ref. 17, we do not pursue this topic any further. We would like to emphasize that our results (4.16)–(4.19) are valid for arbitrary values of the field strength and γ_c, Ω, Γ, etc., whereas Avan and Cohen-Tannoudji have presented only the limiting results (4.21). The simplicity of our method should also be noted.

V. QUANTUM ELECTRODYNAMIC TREATMENT OF THE EFFECT OF LASER PHASE FLUCTUATIONS

We have so far treated the external laser field as either a classical field or a coherent field with a fluctuating amplitude and phase. In this section we present a treatment which makes the second-quantized nature of the field explicit. We assume that the initial state of the field is a Fock state. The phase fluctuations appear in a dynamic form in equations. For simplicity of the analysis, we ignore the terms arising from the relaxation of the atomic system. It is well known that the phase-diffusion model of the laser is described by the interaction Hamiltonian:

$$H = \omega_0 a^\dagger a + \mu(t) a^\dagger a , \quad (5.1)$$

where $\mu(t)$ has the properties given by (2.8). The total interaction can now be written

$$H = \omega_0 a^\dagger a + \mu(t) a^\dagger a + \omega S^z - g(S^+ a + S^- a^\dagger) . \quad (5.2)$$

Thus the density operator in a frame rotating with the frequency ω_0 satisfies the equation

$$\dot{\rho} = -i(\omega - \omega_0)[S^z, \rho] + ig[S^+ a + S^- a^\dagger, \rho] - i\mu(t)[a^\dagger a, \rho] . \quad (5.3)$$

We now eliminate the stochastic element from (5.3). This can be done by using again the theory of the multiplicative stochastic processes (see Appendix B) and the exact result is

$$\langle\dot{\rho}\rangle = -i\Delta[S^z, \langle\rho\rangle] + ig[S^+ a + S^- a^\dagger, \langle\rho\rangle] - \gamma_c[a^\dagger a, [a^\dagger a, \langle\rho\rangle]] \quad (5.4)$$

The solutions of (5.4) describe the time-dependent behavior of the states of the field as well as the atom. The solutions of (5.4) can be obtained by using Fock space representation of the radiation field. Let us assume an initial state of the form

$$\rho(0) = |\chi\rangle\langle\chi| ,$$
$$|\chi\rangle = \alpha_1 |n, 1\rangle + \alpha_2 |n, 2\rangle . \quad (5.5)$$

Note that for the system (5.4) $(a^\dagger a + S^z)$ is a constant of motion and hence the density matrix at time t will be of the form

$$\langle\rho(t)\rangle = \sum_{\mu,\nu=1}^{4} \rho^{(\mu,\nu)} |\Phi_\mu\rangle\langle\Phi_\nu| , \quad (5.6)$$

where

$$|\Phi_1\rangle = |n, 1\rangle , \quad |\Phi_2\rangle = |n+1, 2\rangle ,$$
$$|\Phi_3\rangle = |n, 2\rangle , \quad |\Phi_4\rangle = |n-1, 1\rangle . \quad (5.7)$$

The reduced density operators for the atomic system and the radiation field will be

$$\langle\rho_A\rangle = (\rho^{(1,1)} + \rho^{(4,4)})|1\rangle\langle 1| + (\rho^{(2,2)} + \rho^{(3,3)})|2\rangle\langle 2| + \rho^{(1,3)}|1\rangle\langle 2| + \rho^{(3,1)}|2\rangle\langle 1| , \quad (5.8)$$

$$\langle\rho_R\rangle = (\rho^{(1,1)} + \rho^{(3,3)})|n\rangle\langle n| + \rho^{(2,2)}|n+1\rangle\langle n+1| + \rho^{(4,4)}|n-1\rangle\langle n-1| + \{\rho^{(1,4)}|n\rangle\langle n-1| + \rho^{(2,3)}|n+1\rangle\langle n| + \mathrm{H.c.}\} . \quad (5.9)$$

Equation (5.8) clearly shows the relation of $\rho^{(\mu,\nu)}$ to the atomic expectation values such as dipole moments. Equations for $\rho^{(\mu,\nu)}$ follow from (5.4). On defining

$$\langle\chi_1\rangle = \rho^{(1,3)} = \langle\langle S^-\rangle\rangle , \quad \langle\chi_2\rangle = \rho^{(2,4)} ,$$
$$\langle\chi_3\rangle = \rho^{(1,4)} , \quad \langle\chi_4\rangle = \rho^{(2,3)} , \quad (5.10)$$

we find the equations

$$\langle\dot{\chi}\rangle = \begin{bmatrix} -i\Delta & 0 & -ig\sqrt{n} & ig\sqrt{n+1} \\ 0 & i\Delta-4\gamma_c & ig\sqrt{n+1} & -ig\sqrt{n} \\ -ig\sqrt{n} & ig\sqrt{n+1} & -\gamma_c & 0 \\ ig\sqrt{n+1} & -ig\sqrt{n} & 0 & -\gamma_c \end{bmatrix}\langle\chi\rangle. \tag{5.11}$$

We thus see that the mean value of the dipole moment satisfies the same equation (3.3) provided we assume that the number of photons in the field is large enough so as to justify the replacement $n+1 \cong n$. We next show that a similar result holds for the population distribution in the lower and upper levels for example, we obtain for the atomic density matrix the equation

$$\frac{d}{dt}\begin{bmatrix}\langle\rho_{21}\rangle\\ \langle\rho_{12}\rangle\\ \langle\rho_{11}\rangle\\ \langle\rho_{22}\rangle\end{bmatrix} = \begin{bmatrix} i\Delta-\gamma_c & 0 & ig\sqrt{n+1} & -ig\sqrt{n+1} \\ 0 & -i\Delta-\gamma_c & -ig\sqrt{n+1} & ig\sqrt{n+1} \\ ig\sqrt{n+1} & -ig\sqrt{n+1} & 0 & 0 \\ -ig\sqrt{n+1} & ig\sqrt{n+1} & 0 & 0 \end{bmatrix}\begin{bmatrix}\langle\rho_{21}\rangle\\ \langle\rho_{12}\rangle\\ \langle\rho_{11}\rangle\\ \langle\rho_{22}\rangle\end{bmatrix}, \tag{5.12}$$

which compares very well with Eq. (3.6). We have thus established the equivalence of the results of Sec. II with the results of the present section provided $n \gg 1$. Since in Sec. III one has assumed the excitation of the field in a coherent state whereas in the present section in the Fock state, one expects the equivalence on the basis of the correspondence principle, i.e., in the limit of large n and that is what we have explicitly shown. For an initial Fock state in which n is of the order of 1, then the equations like (5.11) and (5.12) are to be used. In the case $\gamma_c \to 0$, the solutions of (5.11) and (5.12) are equivalent to the Cummings and Jaynes'[51] solutions.

We have considered so far only the phase fluctuations of the laser beam. In principle the amplitude also fluctuates. In many cases, for a laser operating well above threshold, the amplitude can be written as

$$\mathcal{E}(t) = \mathcal{E}_0 + \mathcal{E}_1(t),$$

where the part $\mathcal{E}_1(t)$ is fluctuating and is much smaller compared to $\mathcal{E}_0$. Since $\langle\mathcal{E}_1^2\rangle \ll \mathcal{E}_0^2$, the effect of $\mathcal{E}_1(t)$ can be incorporated in the theory by doing a second-order perturbation calculation with respect to $\mathcal{E}_1$, in a way analogous to the one in Appendix A. The results on the effect of laser amplitude fluctuations will be discussed in a future publication.

In this paper we have treated exactly the effect of the laser phase fluctuations on the outcome of a number of optical-resonance experiments such as resonance fluorescence, energy absorption from a weak field, optical double resonance, Hanle resonances, free-induction decay, etc. The laser field can be either a semiclassical field or a full quantized field having excitation in coherent state or Fock state. In future publications, we will study the effect of the laser fluctuations on the experiments involving two-photon[52] type of processes, on the nonlinear susceptibilities[53] of a two-level atom in the presence of a strong laser field, coherent and incoherent pulse propagation, etc.

APPENDIX A: ATOMIC CORRELATION FUNCTIONS TO SECOND ORDER IN APPLIED FIELD AND THE SPECTRUM OF THE SCATTERED LIGHT

In this appendix we assume that the applied laser field strength is much less than the saturation field. In this case a perturbative solution[54] in powers of $\mathcal{E}$ can be obtained. From the perturbative solutions the correlation functions involving the ensemble averages over the distribution of the laser field can be calculated easily. Such approximate results involve, to lowest order in field strengths, only the second-order correlation functions of the field and hence are model independent.

In order to keep the analysis simple, we do only the case when the relaxation mechanisms are of the Torrey type. On integrating (2.5) once, we get

$$\langle S^z(t+\tau)\rangle = e^{-\tau/T_1}\langle S^z(t)\rangle + \int_0^\tau dt'\, e^{-(\tau-t')/T_1}\left(\frac{1}{T_1}\langle S^z\rangle_{eq} + \{ig\mathcal{E}(t+t')[\langle S^+(t+t')\rangle e^{-i\varphi(t+t')} - \text{c.c.}]\}\right), \tag{A1}$$

$$\langle S^+(t+\tau)\rangle = e^{i\Delta\tau}e^{-\tau/T_2}\langle S^+(t)\rangle + \int_0^\tau dt' \exp\left[-\left(\frac{1}{T_2} - i\Delta\right)(\tau - t')\right] 2ig\langle S^z(t+t')\rangle\mathcal{E}(t+t')e^{i\varphi(t+t')}. \tag{A2}$$

Using (A1) and (A2) in (2.5) we obtain

$$\frac{d}{d\tau}\langle S^+(t+\tau)\rangle = -\left(\frac{1}{T_2} - i\Delta\right)\langle S^+(t+\tau)\rangle + 2ig\mathcal{E}(t+\tau)\langle S^z(t)\rangle e^{i\varphi(t+\tau)}e^{-\tau/T_1} + 2ig\mathcal{E}(t+\tau)e^{i\varphi(t+\tau)}$$
$$\times\int_0^\tau dt'\, e^{-(\tau-t')/T_1}\left(\frac{1}{T_1}\langle S^z\rangle_{eq} + \{ig\mathcal{E}(t+t')[\langle S^+(t+t')\rangle e^{-i\varphi(t+t')} - \text{c.c.}]\}\right), \tag{A3}$$

$$\frac{d}{d\tau}\langle S^z(t+\tau)\rangle = -\frac{1}{T_1}(\langle S^z(t+\tau)\rangle - \langle S^z\rangle_{eq}) + i\Big(g\mathcal{E}(t+\tau)e^{-i\varphi(t+\tau)}\langle S^+(t)\rangle e^{-\tau(1/T_2 - i\Delta)}$$
$$+\int_0^\tau dt'\, e^{-(1/T_2 - i\Delta)(\tau-t')} 2ig\langle S^z(t+t')\rangle\mathcal{E}(t+t')e^{i\varphi(t+t')} - \text{c.c.}\Big). \tag{A4}$$

The equations for the correlation functions can be obtained from (A3) and quantum regression theorem:

$$\frac{d}{d\tau}\langle S^+(t+\tau)S^-(t)\rangle = -\left(\frac{1}{T_2} - i\Delta\right)\langle S^+(t+\tau)S^-(t)\rangle + 2ig\mathcal{E}(t+\tau)e^{i\varphi(t+\tau)}$$
$$\times[\langle S^z\rangle_{eq} - (\langle S^z\rangle_{eq} + \tfrac{1}{2})e^{-\tau/T_1}]\langle S^-(t)\rangle - 2g^2\mathcal{E}(t+\tau)e^{i\varphi(t+\tau)}$$
$$\times\int_0^\tau dt'\, e^{-(\tau-t')/T_1}\mathcal{E}(t+t')[\langle S^+(t+t')S^-(t)\rangle e^{-i\varphi(t+t')} - \langle S^-(t+t')S^-(t)\rangle e^{i\varphi(t+t')}]. \tag{A5}$$

Equation (A5) involves the correlation function $\langle S^-(t+t')S^-(t)\rangle$, which satisfies a similar equation

$$\frac{d}{d\tau}\langle S^-(t+\tau)S^-(t)\rangle = -\left(\frac{1}{T_2} + i\Delta\right)\langle S^-(t+\tau)S^-(t)\rangle - 2ig\mathcal{E}(t+\tau)e^{-i\varphi(t+\tau)}$$
$$\times[\langle S^z\rangle_{eq} - (\langle S^z\rangle_{eq} + \tfrac{1}{2})e^{-\tau/T_1}]\langle S^-(t)\rangle - 2g^2\mathcal{E}(t+\tau)e^{-i\varphi(t+\tau)}$$
$$\times\int_0^\tau dt'\, e^{-(\tau-t')/T_1}\mathcal{E}(t+t')[\langle S^-(t+t')S^-(t)\rangle e^{i\varphi(t+t')} - \langle S^+(t+t')S^-(t)\rangle e^{-i\varphi(t+t')}]. \tag{A6}$$

Note that the above equations for correlation functions involves terms like $\mathcal{E}(t+\tau)e^{-i\varphi(t+\tau)}\langle S^-(t)\rangle$. These can be simplified by using (A2), i.e.,

$$\langle S^-(t)\rangle = e^{-(1/T_2 + i\Delta)t}\langle S^-(0)\rangle + \int_0^t dt' e^{-(1/T_2 + i\Delta)(t-t')}[-2ig\mathcal{E}(t')e^{-i\varphi(t')}\langle S^z(t')\rangle], \tag{A7}$$

and hence

$$\mathcal{E}(t+\tau)e^{i\varphi(t+\tau)}\langle S^-(t)\rangle = \mathcal{E}(t+\tau)e^{i\varphi(t+\tau) - (1/T_2 + i\Delta)t}\langle S^-(0)\rangle$$
$$+\int_0^t dt'\, e^{-(1/T_2 + i\Delta)(t-t')}(-2ig)\mathcal{E}(t')\mathcal{E}(t+\tau)\langle S^z(t')\rangle e^{-i\varphi(t') + i\varphi(t+\tau)}. \tag{A8}$$

On substituting (A8) in (A5) we find that we have iterated in a manner so that $\mathcal{E}$ appears as second power in (A5) and (A6). Equations (A5) and (A6) are exact equations, i.e., we have not made any approximations concerning the strength of the laser field. If the fields are less than the saturation fields, then one can do a second-order calculation in $\mathcal{E}$. From (A5) and (A8) it is clear that to second order in $\mathcal{E}$ we have

$$\frac{d}{d\tau}\langle S^+(t+\tau)S^-(t)\rangle^{(2)} = -\left(\frac{1}{T_2} - i\Delta\right)\langle S^+(t+\tau)S^-(t)\rangle^{(2)} + 4g^2[\langle S^z\rangle_{eq} - (\langle S^z\rangle_{eq} + \tfrac{1}{2})e^{-\tau/T_1}]$$
$$\times\int_0^t dt'\, e^{-(1/T_2 + i\Delta)(t-t')}\mathcal{E}(t')\mathcal{E}(t+\tau)\langle S^z(t')\rangle^{(0)}e^{-i\varphi(t') + i\varphi(t+\tau)}$$
$$-2g^2\int_0^\tau dt'\, \mathcal{E}(t+\tau)\mathcal{E}(t+t')e^{i\varphi(t+\tau) - (\tau-t')/T_1}[\langle S^+(t+t')S^-(t)\rangle^{(0)}e^{-i\varphi(t+t')}$$
$$-\langle S^-(t+t')S^-(t)\rangle^{(0)}e^{i\varphi(t+t')}]. \tag{A9}$$

Note that

$$\langle S^+(t+t')S^-(t)\rangle^{(0)} = e^{-(1/T_2 - i\Delta)t'}\langle S^+(t)S^-(t)\rangle^{(0)}, \tag{A10}$$

$$\langle S^-(t+t')S^-(t)\rangle^{(0)} = e^{-(1/T_2 + i\Delta)t'}\langle S^-(t)S^-(t)\rangle^{(0)} = 0. \tag{A11}$$

We now assume that the correlation functions of the field are given by

$$\langle \mathcal{E}(t+\tau)\mathcal{E}(t+\tau')e^{i\varphi(t+\tau)-i\varphi(t+\tau')}\rangle = \mathcal{E}^2 e^{-\gamma_c|\tau-\tau|} ,$$
$$\langle \mathcal{E}(t)\mathcal{E}(\tau)e^{i\varphi(t)+i\varphi(\tau)}\rangle = 0. \tag{A12}$$

On using (A10)–(A12) in (A9) we get for the ensemble averaged correlations

$$\frac{d}{d\tau}\langle\langle S^+(t+\tau)S^-(t)\rangle\rangle^{(2)} = -\left(\frac{1}{T_2} - i\Delta\right)\langle\langle S^+(t+\tau)S^-(t)\rangle\rangle^{(2)} + 4g^2\mathcal{E}^2[\langle S^z\rangle_{eq} - (\langle S^z\rangle_{eq}+\tfrac{1}{2})e^{-\tau/T_1}]$$
$$\times\int_0^t dt'\langle\langle S^z(t')\rangle\rangle^{(0)}\exp[-(1/T_2+i\Delta)(t-t')-\gamma_c(t+\tau-t')]$$
$$-2g^2\mathcal{E}^2\int_0^\tau dt'\langle\langle S^+(t)S^-(t)\rangle\rangle^{(0)}\exp\left[-\left(\gamma_c+\frac{1}{T_1}\right)(\tau-t')-\left(\frac{1}{T_2}-i\Delta\right)t'\right]. \tag{A13}$$

Hence the Laplace transform of the correlation function defined by

$$\hat{\Gamma}^{(N)(2)}(t,z) = \int_0^\infty d\tau\, e^{-z\tau}\langle\langle S^+(t+\tau)S^-(t)\rangle\rangle^{(2)} \tag{A14}$$

is given by

$$\left[z+\left(\frac{1}{T_2}-i\Delta\right)\right]\hat{\Gamma}^{(N)(2)}(t,z) = \langle\langle S^+(t)S^-(t)\rangle\rangle^{(2)} - 2g^2\mathcal{E}^2\langle\langle S^+(t)S^-(t)\rangle\rangle^{(0)}\left(z+\gamma_c+\frac{1}{T_1}\right)^{-1}\left(z+\frac{1}{T_2}-i\Delta\right)^{-1}$$
$$+4g^2\mathcal{E}^2\left[\langle S^z\rangle_{eq}(z+\gamma_c)^{-1} - (\langle S^z\rangle_{eq}+\tfrac{1}{2})\left(z+\gamma_c+\frac{1}{T_1}\right)^{-1}\right]$$
$$\times\int_0^t dt'\langle\langle S^z(t')\rangle\rangle^{(0)}\exp[-(1/T_2+i\Delta+\gamma_c)(t-t')] . \tag{A15}$$

Note that (A4) shows that to second order in $\mathcal{E}$, $\langle S^z\rangle$ satisfies

$$\frac{d}{d\tau}\langle\langle S^z(t)\rangle\rangle^{(2)} = -\frac{1}{T_1}\langle\langle S^z(\tau)\rangle\rangle^{(2)} - 2g^2\mathcal{E}^2\int_0^\tau dt'\langle\langle S^z(t')\rangle\rangle^{(0)}\{\exp[-(1/T_2+\gamma_c-i\Delta)(\tau-t')]+\text{c.c.}\}. \tag{A16}$$

Equations (A15) and (A16) give the transient spectrum of the scattered light which is proportional to the real part of $\hat{\Gamma}^{(N)(2)}(t,i\omega)$. In the steady state one has from (A16)

$$\langle\langle S^z\rangle\rangle_{st}^{(2)} = -2g^2\mathcal{E}^2\langle S^z\rangle_{eq}T_1[(1/T_2+\gamma_c-i\Delta)^{-1}+\text{c.c.}] , \tag{A17}$$

$$\left(z+\frac{1}{T_2}-i\Delta\right)\hat{\Gamma}^{(N)(2)}(\infty,z) = \langle\langle S^z\rangle\rangle_{St}^{(2)} - 2g^2\mathcal{E}^2\left(z+\gamma_c+\frac{1}{T_1}\right)^{-1}\left(z+\frac{1}{T_2}-i\Delta\right)^{-1}(\tfrac{1}{2}+\langle S^z\rangle_{eq})$$
$$+4g^2\mathcal{E}^2\left(\frac{1}{T_2}+i\Delta+\gamma_c\right)^{-1}\langle S^z\rangle_{eq}\left[\langle S^z\rangle_{eq}(z+\gamma_c)^{-1} - (\tfrac{1}{2}+\langle S^z\rangle_{eq})\left(z+\gamma_c+\frac{1}{T_1}\right)^{-1}\right]. \tag{A18}$$

In the special case of radiative relaxation $\langle S^z\rangle_{eq} = -\frac{1}{2}$, (A18) reduces to

$$\hat{\Gamma}^{(N)(2)}(\infty,z) = \alpha^2[\gamma^2-(\gamma_c+i\Delta)^2]^{-1}(z+\gamma_c)^{-1}$$
$$+(\gamma_c\alpha^2/\gamma)[\gamma_c^2-(\gamma-i\Delta)^2]^{-1}$$
$$\times(z+\gamma-i\Delta)^{-1} . \tag{A19}$$

If one further specializes to the case of resonance, then (A19) leads to the well-known form of the spectrum

$$\hat{\Gamma}^{(N)(2)}(\infty,z) = \alpha^2(\gamma_c/\gamma)(\gamma_c^2-\gamma^2)^{-1}(z+\gamma)^{-1}$$
$$+\alpha^2(\gamma^2-\gamma_c^2)^{-1}(z+\gamma_c)^{-1} , \tag{A20}$$

or

$$Re\hat{\Gamma}^{(N)(2)}(\infty,i\omega) = \alpha^2\gamma_c[\gamma^2+\omega^2]^{-1}[\gamma_c^2+\omega^2]^{-1} . \tag{A21}$$

Equation (A21) leads to the standard result.[55] If $\gamma_c \ll \gamma$, then the spectrum of resonance fluorescence will be determined by the spectrum of laser radiation and hence the linewidth of the scattered radiation is much less than the natural width. Gibbs and Venkatesan[56] have measured linewidths much less than the natural widths. In the other extreme case $\gamma_c \gg \gamma$, the spectrum is same as that of spontaneously emitted radiation.

In this appendix we thus have obtained the most general forms of the transient and steady-state spectrum when the two-level atom is relaxing with relaxation parameters T_1 and T_2. The results are valid up to second order in fields, but for arbitrary values of γ_c, T_1, T_2, Δ, and the initial state of the atom. The iteration has been done self-consistently, i.e., we made *a priori* no *ad hoc* assumptions like linearization of the first two Bloch equations by substituting some form for $\langle S^z(t)\rangle$ such as $\langle S^z\rangle_{eq}$. For arbitrary field strengths one has to solve the exact equations like (A5) and (A6). It is clear that if the ensemble average of both sides is taken, then (A5) still involves the unknowns like the ensemble average of

$$\langle S^+(t+t')S^-(t)\rangle \times \exp[i\varphi(t+\tau)-i\varphi(t+t')]\mathcal{E}(t+\tau)\mathcal{E}(t+t')$$

A closed set of equations can be obtained by decoupling the hierarchy suitably. However, it is difficult to assert in this case the region of validity of the approximation. In the main text we obtain an exact solution for the phase-diffusion model of the laser. The exact result reduces to (A18) when one expands to second order in α. We finally also note that the structure of (A5) and (A6) is similar to the equations obtained by Eberly.[26]

APPENDIX B: MULTIPLICATIVE STOCHASTIC PROCESSES

Our exact results, in Secs. III–V are derived using the theory of multiplicative stochastic processes. For the sake of completeness we review the main results on such processes and also present a rather simplified derivation of such results. Fox has given a detailed derivation of the important results on multiplicative stochastic processes. He also has generalized many of the earlier results of Kubo.[57] Consider the Langevin equations of the form

$$\dot{x}_i=\sum_j M_{ij}x_j+\sum_j F_{ij}(t)x_j \quad , \tag{B1}$$

where F_{ij} are δ-correlated Gaussian random processes satisfying

$$\begin{aligned}&\langle F_{ij}(t)\rangle=0 \ ,\\ &\langle F_{ij}(t)F_{kl}(t^1)\rangle=2Q_{ijkl}\delta(t-t^1) \ .\end{aligned} \tag{B2}$$

One wants to study the behavior of the mean values of x and the correlations between $x_i(t)$. The system of variables has the Markovian property and hence it is sufficient to know the Fokker-Planck equation for the conditional distribution function. The Fokker-Planck equation can be obtained by using the general results of Lax[33] and Stratonivich.[58] For a more general Langevin equation of the form

$$\begin{aligned}&\dot{x}_i=B_i+\sum_j\sigma_{ij}f_j(t) \ ,\\ &\langle f_j(t)\rangle=0 \ ,\\ &\langle f_i(t)f_j(t^1)\rangle=2\delta_{ij}\delta(t-t^1) \ ,\end{aligned} \tag{B3}$$

the associated Fokker-Planck equation has the form

$$\frac{dP}{dt}=-\sum_i\frac{\partial}{\partial x_i}[A_iP]+\sum_{ij}\frac{\partial^2}{\partial x_i\partial x_j}[D_{ij}P] \quad , \tag{B4}$$

with

$$\begin{aligned}&A_i=B_i+\sum_{mk}\frac{\partial\sigma im}{\partial x_k}\sigma_{km} \ ,\\ &D_{ij}=\sum_m\sigma_{im}\sigma_{jm} \ .\end{aligned} \tag{B5}$$

The derivation (B4) does not involve any approximation. Thus for the system of Langevin equations (B1) one will get (B4) with the drift and diffusion coefficients given by

$$\begin{aligned}&A_i=\sum_j M_{ij}x_j+\sum_{kl}Q_{ikkl}x_l \quad ,\\ &D_{ij}=\sum_{lm}Q_{iljm}x_lx_m \ .\end{aligned} \tag{B6}$$

Fox has obtained (B6) through a laborious method using the moment theorem for Gaussian random processes.[29] Note that we have simple equations for the mean values

$$\langle\dot{x}_i\rangle=\sum_j\left(M_{ij}+\sum_k Q_{ikkj}\right)\langle x_j\rangle \quad . \tag{B7}$$

The classical equations like (B1) are applicable to quantum-mechanical systems. The density matrix ρ satisfies equations of the form

$$\dot{\rho}_{ij}=\sum_{kl}\{L_{0ijkl}\rho_{kl}+[L_1(t)]_{ijkl}\rho_{kl}\} \ , \tag{B8}$$

where the random forces (δ correlated and Gaussian) appear in the part $L_1(t)$:

$$\begin{aligned}&L_1(t)=\sum_\alpha F_\alpha(t)L_1^{(\alpha)} \quad ,\\ &\langle F_\alpha(t)F_\beta(t^1)\rangle=2D_{\alpha\beta}\delta(t-t^1) \ .\end{aligned} \tag{B9}$$

The result (B7) when applied to (B8) implies that

$$\begin{aligned}\langle\dot{\rho}\rangle&=L_0\langle\rho\rangle+\int_0^t\langle L_1(t)L_1(\tau)\rangle\langle\rho(t)\rangle\\ &=L_0\langle\rho\rangle+\sum_{\alpha\beta}D_{\alpha\beta}L_1^{(\alpha)}L_1^{(\beta)}\langle\rho(t)\rangle \ .\end{aligned} \tag{B10}$$

Equation (B10) is the master equation for the ensemble average of the density matrix interacting with stochastic perturbations. In the derivation of (B10) no approximation has been made and (B10) is exact. Note also that because of the δ-correlated nature of F_α, the term in (B10) arising due to the interaction with the fluctuating forces does not depend on L_0. Note also that the dynamics of the random forces is prescribed. The master equations which we have discussed in Secs. III–V follow from a straightforward application of (B10). Many other master equations, such as the Redfield equation,[35] Haken-Strobl-Reineker[30] equations, which have been derived in the literature by using very complicated methods, follow from a simple application of (B10). We illustrate the method for Haken-Strobl-Reineker equation describing the problem of exciton diffusion

$$\dot{\rho} = -i[H_1, \rho] - i[H_2(t), \rho] , \quad \text{(B11)}$$

where

$$H_2(t) = \sum_{nn'} h_{nn'}(t) S_n^+ S_{n'}^- \ , \langle h_{nn'} \rangle = 0 \ , \quad \text{(B12)}$$

$$\langle h_{nn'}(t) h_{n''n'''}(t') \rangle = 2\Lambda_{nn'n''n'''} \delta(t - t') \ . \quad \text{(B13)}$$

On applying (B10) to (B11) we immediately get the exact master equation

$$\langle \dot{\rho} \rangle = -i[H_1, \langle \rho \rangle] - \sum_{nn'n''n'''} \Lambda_{nn'n''n'''} [S_n^+ S_{n'}^-, [S_{n''}^+ S_{n'''}^-, \langle \rho \rangle] \ , \quad \text{(B14)}$$

which is the desired equation. It has been obtained in Ref. 30 by infinite-order perburbation theory.[59] We close this appendix with some remarks on the exact master equation (B10). If one uses the Born approximation (cf. Refs. 35–41) then (B8) leads to

$$\langle \dot{\rho} \rangle = L_0 \langle \rho \rangle + \int_0^t \langle L_1(t) e^{L_0(t-\tau)} L_1(\tau) \rangle \langle \rho(\tau) \rangle d\tau \ . \quad \text{(B15)}$$

On substituting the δ-correlated nature of the random forces in (B15), we find that (B15) reduces to (B10). We have thus shown that *when the random forces acting on the system characterized by the Langevin equation* (B1) *are Gaussian* δ *correlated, then the exact master equation is same as the one obtained by using the Born approximation.* We have mentioned in Sec. III that our results on resonance fluorescence are in agreement with those obtained by Mandel and Kimble[47] and by Eberly[26] on the laser phase fluctuations even though these authors employed a decorrelation technique, which is equivalent to the Born approximation. The above agreement, though at first sight mysterious, can be understood from the italic result cited above.

The results of this appendix can also be used to calculate the fluctuations in various dynamical variables for example, population inversion, dipole moment, etc., i.e., if one wants to calculate quantities like $\langle\langle (S^z)^2 \rangle - (\langle\langle S^z \rangle\rangle)^2$ then the Fokker-Planck equation (B4) can be used. This equation for example, leads to

$$\langle x_k x_l \rangle = \langle A_k x_l \rangle + \langle A_l x_k \rangle + \sum_{ij} Q_{kilj} \langle x_i x_j \rangle \ , \quad \text{(B16)}$$

where A_k's are defined by (B6). An analysis of these fluctuations and their experimental implications will be presented elsewhere.

*A previous account of this work was presented by G. S. Agarwal [Phys. Rev. Lett. 37, 1383 (1976)].

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