

## Absorption spectra of a system of atoms under cooperative conditions

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The problem of the energy absorption from a weak probe field by a system of atoms driven by a coherent laser field is considered with special reference to the collective behavior of the atomic system. The changes in the nature of the absorption spectra as the intensity of the coherent laser field is changed are examined explicitly for a system of two atoms. Cooperative effects are shown to be important for weak laser fields. For strong fields, it is shown that the absorption spectra under cooperative conditions differ from those without cooperative conditions only by a scale factor.

### I. INTRODUCTION

Saturation absorption spectroscopy is a powerful tool for measuring homogeneous linewidths, relaxation times, level shifts, etc., in gases.<sup>1-3</sup> The absorbing medium is saturated by a strong laser field and the absorption of electromagnetic energy from a weak probe field, propagating either in the same direction as or in an opposite direction to the strong laser field, is measured. In theoretical calculations the interaction of an atom with the weak probe field is treated perturbatively and that with the strong pump field exactly.<sup>4-6</sup>

Previously, Mollow<sup>4</sup> and Haroche and Hartmann<sup>5</sup> calculated the spectrum of the energy absorbed from the probe field by a single two-level atom, in a strong laser field. They found that the absorption spectrum changes drastically with the intensity of the pump field. For low pump intensities peak absorption occurs when the probe is in resonance with the atomic transition frequency. At high-field intensities the absorption is negative in a certain range of frequencies, indicating that stimulated emission takes place, amplifying the probe beam at the expense of the pump field. A recent experiment<sup>7</sup> has confirmed some of these predictions.

In all these calculations, however, it was assumed that different atoms interact with the external field independently of each other. Such an assumption may be expected to hold for high-field intensities, when the field reradiated by the atom is small in comparison to the pump field, so that the atom-atom interaction is insignificant compared to the atom-field interaction.

A study of the cooperative effects in resonance fluorescence was undertaken by Agarwal *et al.*,<sup>8</sup> who studied the interaction of an external electromagnetic field with one-, two-, and three-atom systems. They found that as far as the first-order correlation function  $\langle S^+(t+\tau)S^-(t) \rangle$  is concerned, the light scattered by a system of two-

level atoms driven by a laser field is strongly influenced by atom-atom interactions only at low-field strengths. On the other hand, for high-field intensities, the laser-atom interaction predominates. However, in the case of the second-order correlation function  $\langle S^+(t)S^+(t+\tau)S^-(t+\tau)S^-(t) \rangle$  the collective effects play an important role both at low- and high-field intensities.

It would be interesting then, to study the effect of collective behavior of the atoms on the absorption spectrum of two-level atoms for various field strengths.

We have studied the absorption spectrum of a system of two two-level atoms, taking into account the vacuum field-induced cooperative effects and we have compared the results with the case where collective effects are ignored. An outline of this paper is as follows. In Sec. II we present the mathematical formulation of the problem, and show how the absorption spectra can be computed. In Sec. III we present analytical but approximate results for the absorption spectra in the limit of intense laser fields (the probe field is always taken to be weak). In this limit, we show that there is scaling of the response functions. Finally, we give the results of our numerical analysis.

### II. MATHEMATICAL FORMULATION

We consider the system of two-level atoms in a strong laser field that saturates the system. A *weak* probe beam is introduced after the system has attained steady-state conditions. The time-average rate of absorption of energy by the system from the probe field is given by

$$\left\langle \frac{dU}{dt} \right\rangle_{av} = -\frac{2\omega}{\hbar} |\vec{d} \cdot \vec{E}|^2 \times \text{Re} \int_0^\infty d\tau e^{-i\omega\tau} \langle [S^+(\tau), S^-(0)] \rangle_{st}, \quad (2.1)$$

where  $\omega$  is the frequency of the probe field and  $\vec{E}$  is its amplitude;  $\vec{d}$  is the dipole-moment matrix

element;  $S^{\pm} = \sum_i S_i^{\pm}$  are the collective dipole operators for the system in the Heisenberg picture. The steady-state expectation values are defined as

$$\langle S^+(\tau)S^-(0) \rangle_{st} = \lim_{t \rightarrow \infty} \langle S^+(t+\tau)S^-(t) \rangle. \quad (2.2)$$

The time-correlation functions are calculated in the absence of probe field. The laser field, however, is always present.

The time-average rate of absorption per unit flux of the incident field is

$$\begin{aligned} \left\langle \frac{dU'}{dt} \right\rangle_{av} &= -\frac{4\pi\omega}{3c\hbar} |\vec{d}|^2 \\ &\times \text{Re} \int_0^{\infty} d\tau e^{-i\omega\tau} \langle [S^+(\tau), S^-(0)] \rangle_{st}, \end{aligned} \quad (2.3)$$

since the dipole moment  $\vec{d}$  is usually randomly oriented and hence the average value of  $|\vec{d} \cdot \vec{E}|^2 / |\vec{E}|^2$  is  $1/3 |\vec{d}|^2$ . Since we are considering the absorption in the vicinity of the atomic transition frequency,  $\omega$  varies slowly so that the time-average rate of absorption of energy per unit flux of the incident field varies according to the function

$$R(\omega) \equiv \text{Re} \int_0^{\infty} d\tau e^{-i\omega\tau} \langle [S^+(\tau), S^-(0)] \rangle_{st}, \quad (2.4)$$

and hence in what follows we only calculate  $R(\omega)$ .

We start with the master equation for the reduced density operator for the system, taking into account the radiative decay of the atoms into the vacuum state of the radiation field and the interac-

tion of the atoms with the strong pump field. The reduced density operator for the atomic system can be shown to satisfy<sup>9</sup>

$$\begin{aligned} \frac{\partial \bar{\rho}}{\partial t} &= 2\gamma(S^-\bar{\rho}S^+ - \frac{1}{2}S^+S^-\bar{\rho} - \frac{1}{2}\bar{\rho}S^+S^-) \\ &\quad - ig[S^+ + S^-, \bar{\rho}] - i\Delta[S^z, \bar{\rho}], \end{aligned} \quad (2.5)$$

where  $\Delta = (\omega_0 - \omega_L)$ ,  $\omega_0$  being the atomic transition frequency and  $\omega_L$  is the pump-field frequency;  $2\gamma$  is the Einstein  $A$  coefficient for the single atom;  $g = -\vec{d} \cdot \vec{E}_{0L}$  is the coupling constant with the coherent laser field,  $\vec{E}_{0L}$  being the amplitude of the pump field,  $\vec{E} = 2\vec{E}_{0L} \cos\omega_L t$ , and  $\vec{d}$  is the atomic dipole-moment matrix element;  $S^z = \sum_i S_i^z$  gives the difference in the population of the two levels. In deriving Eq. (2.5) the interaction of the system with the laser field has been treated exactly. However, in treating the interaction with the vacuum of the field, Born, Markoff, and rotating-wave approximations have been made. These approximations are justified as this interaction is relatively weak and the vacuum is a continuum of modes. The reduced-density operator  $\bar{\rho}$  evolves in a frame rotating with the frequency of the pump field  $\omega_L$ . The time-correlation functions can be calculated using the solution of (2.5).

Projecting Eq. (2.5) into the Hilbert space of energy eigenvalues  $|j, m\rangle$  with  $j=1$ ,  $m=0, \pm 1$ , we get the equations of motion for the matrix elements  $\bar{\rho}_{m', m}(t)$  of the atomic density operator as

$$\begin{aligned} \frac{\partial \bar{\rho}_{m', m}}{\partial \tau} &= (\nu_{m'+1} \nu_{m+1})^{1/2} \bar{\rho}_{m'+1, m+1} - \left( \frac{1}{2}(\nu_{m'} + \nu_m) + \frac{i\Delta}{2\gamma} (m' - m) \right) \bar{\rho}_{m', m} \\ &\quad - \frac{ig}{2\gamma} (\nu_{m'}^{1/2} \bar{\rho}_{m'-1, m} + \nu^{1/2} \bar{\rho}_{m'+1, m} - \nu_{m+1}^{1/2} \bar{\rho}_{m', m+1} - \nu_m^{1/2} \bar{\rho}_{m', m-1}), \end{aligned} \quad (2.6)$$

where  $\tau = 2\gamma t$ ,  $\nu_m = (j+m)(j-m+1)$ , so that  $\nu_1 = \nu_0 = 2$ ;  $\nu_{-1} = 0$ . Equation (2.6) differs from that in Paper I by the presence of the detuning term. It should be noted that  $j=1$  corresponds to a cooperative pair of two two-level ( $j = \frac{1}{2}$ ) atoms.

Writing the elements  $\bar{\rho}_{m', m}(t)$  of the density matrix as elements  $\Psi_i(t)$  of a column vector  $\vec{\Psi}$ , the equations can be cast into a matrix form

$$\frac{d\vec{\Psi}}{d\tau} = \mathcal{L}\vec{\Psi} + \vec{I}, \quad (2.7)$$

where  $\mathcal{L}$  is an  $8 \times 8$  matrix defined by

$$\mathcal{L} = \begin{bmatrix} -2 & 0 & i\beta\sqrt{2} & -i\beta\sqrt{2} & 0 & 0 & 0 & 0 \\ 2 & -2 & -i\beta\sqrt{2} & i\beta\sqrt{2} & 0 & 0 & i\beta\sqrt{2} & -i\beta\sqrt{2} \\ i\beta\sqrt{2} & -i\beta\sqrt{2} & -(2+i\Delta/2\gamma) & 0 & i\beta\sqrt{2} & 0 & 0 & 0 \\ -i\beta\sqrt{2} & i\beta\sqrt{2} & 0 & -(2-i\Delta/2\gamma) & 0 & -i\beta\sqrt{2} & 0 & 0 \\ 0 & 0 & i\beta\sqrt{2} & 0 & -(1+2i\Delta/2\gamma) & 0 & -i\beta\sqrt{2} & 0 \\ 0 & 0 & 0 & -i\beta\sqrt{2} & 0 & -(1-2i\Delta/2\gamma) & 0 & i\beta\sqrt{2} \\ i\beta\sqrt{2} & 2i\beta\sqrt{2} & 2 & 0 & -i\beta\sqrt{2} & 0 & -(1+i\Delta/2\gamma) & 0 \\ -i\beta\sqrt{2} & -2i\beta\sqrt{2} & 0 & 2 & 0 & i\beta\sqrt{2} & 0 & -(1-i\Delta/2\gamma) \end{bmatrix}, \quad (2.8)$$

where  $\beta = g/2\gamma$ , and  $\vec{\Psi}$  is the column vector

$$\vec{\Psi} = \begin{bmatrix} \Psi_1 \\ \Psi_2 \\ \Psi_3 \\ \Psi_4 \\ \Psi_5 \\ \Psi_6 \\ \Psi_7 \\ \Psi_8 \end{bmatrix} = \begin{bmatrix} \bar{\rho}_{1,1} \\ \bar{\rho}_{0,0} \\ \bar{\rho}_{1,0} \\ \bar{\rho}_{0,1} \\ \bar{\rho}_{1,-1} \\ \bar{\rho}_{-1,1} \\ \bar{\rho}_{0,-1} \\ \bar{\rho}_{-1,0} \end{bmatrix}; \text{ and } \vec{I} = \begin{bmatrix} 0 \\ 0 \\ 0 \\ 0 \\ 0 \\ 0 \\ -i\beta\sqrt{2} \\ i\beta\sqrt{2} \end{bmatrix}. \quad (2.9)$$

The Laplace transform of Eq. (2.7) is

$$\hat{\Psi}(z) = M\Psi(0) + z^{-1}M\vec{I}, \quad (2.10)$$

where  $M = (z - \mathcal{L})^{-1}$ . The steady-state solutions are given by

$$\Psi(\infty) = \Psi(t) = \lim_{z \rightarrow 0} z \hat{\Psi}(z) = (-\mathcal{L})^{-1} \vec{I}. \quad (2.11)$$

We now proceed to calculate the function

$$R(\omega) = \text{Re} \int_0^\infty d\tau e^{-i\omega\tau} \lim_{t \rightarrow \infty} \langle [S^*(t+\tau), S^-(t)] \rangle. \quad (2.12)$$

We use the quantum regression theorem<sup>10,11</sup> to evaluate the two-time commutator in Eq. (2.12). The theorem states that for a quantum-mechanical system, which is Markovian, if the one-time expectation values can be written

$$\langle A(t) \rangle = \sum_\mu f_\mu(t, t') \langle A_\mu(t') \rangle, \quad (2.13)$$

where  $f_\mu(t, t')$  is a  $c$ -number function and  $t > t'$ , then the two-time expectation values can be written

$$\langle A(t)B(t') \rangle = \sum_\mu f_\mu(t, t') \langle A_\mu(t')B(t') \rangle. \quad (2.14)$$

In this case, the one-time expectation values of  $\langle S^*(t) \rangle$  are obtained in terms of  $\Psi_i$ 's as

$$\begin{aligned} \langle S^*(t) \rangle &= \text{Tr} \{ \rho(t) S^* \} = e^{i\omega_L t} \text{Tr} \{ \bar{\rho}(t) S^* \} \\ &= \sqrt{2} (\Psi_4(t) + \Psi_8(t)) e^{i\omega_L t}. \end{aligned} \quad (2.15)$$

To Eq. (2.15) we now apply the quantum regression theorem to evaluate the correlations  $\langle S^*(t+\tau)S^-(t) \rangle$  and  $\langle S^-(t)S^*(t+\tau) \rangle$  and obtain the following expression for the energy absorbed by the cooperative system of two atoms:

$$\begin{aligned} R_2(\omega - \omega_L) &= [(M_{44} + M_{84})(\psi_1(\infty) - \psi_2(\infty)) \\ &\quad + (M_{46} + M_{86})(\psi_4(\infty) - \psi_8(\infty)) \\ &\quad + (M_{42} + M_{82})(\psi_3(\infty) - \psi_7(\infty)) \\ &\quad + (M_{48} + M_{88})(2\psi_2(\infty) + \psi_1(\infty) - 1) \\ &\quad + (M_{47} + M_{87})\psi_5(\infty) - (M_{41} + M_{81})\psi_3(\infty) \\ &\quad - (M_{43} + M_{83})\psi_5(\infty)] \Big|_{z=i(\omega-\omega_L)} + \text{c. c.} \end{aligned} \quad (2.16)$$

Since we would like to examine the influence of collective behavior on the absorption, we compare the absorption when collective effects are included, which is given by Eq. (2.16), with the corresponding absorption when collective effects are ignored. If the atoms absorb energy independently of each other, then the total energy absorbed by  $N$  atoms is  $N$  times the energy absorbed by one atom. The energy absorbed by a single atom from the probe field is a well established result and is given by

$$\begin{aligned} R_1(\omega - \omega_L) &= [\langle S^z(\infty) \rangle / P(z)] \{ (z + 2\gamma)(z + \gamma + i\Delta) + 2g^2 \\ &\quad - 2g^2(z + \gamma + i\Delta) / (\gamma + i\Delta) \} \Big|_{z=i(\omega-\omega_L)} + \text{c. c.}, \end{aligned} \quad (2.17)$$

where

$$\langle S^z(\infty) \rangle = -\frac{1}{2} [1 + 2g^2 / (\gamma^2 + \Delta^2)]^{-1} \quad (2.18)$$

and

$$P(z) = 4g^2(z + \gamma) + (z + 2\gamma)(z + \gamma)^2 + \Delta^2(z + 2\gamma). \quad (2.19)$$

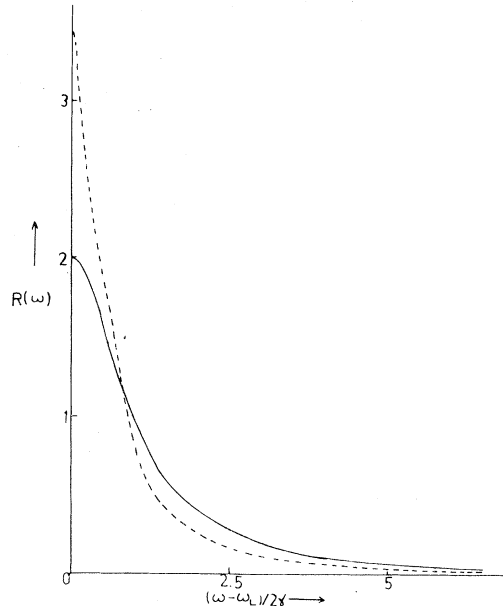


FIG. 1. Behavior of  $R_2$  (solid curve) and  $2R_1$  (dashed curve) as a function of  $\omega$  for  $\beta=0.1$  and  $\Delta=0$ .

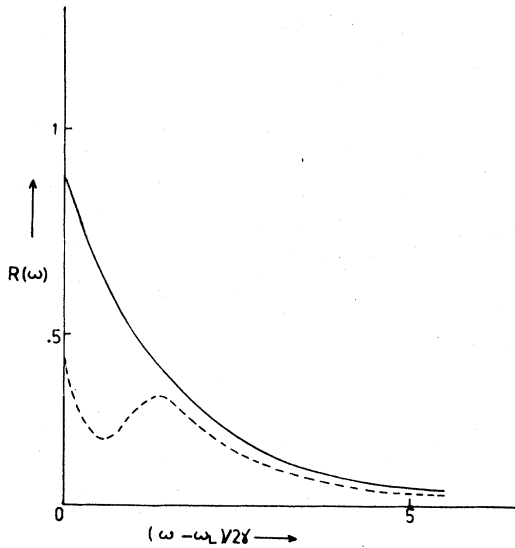


FIG. 2. Same as in Fig. 1 but with larger pump field  $\beta=0.5$ ,  $\Delta=0$ .

In Sec. III we present the results of our numerical analysis for  $R_2(\omega - \omega_L)$  and its comparison with  $2R_1(\omega - \omega_L)$ .

Since the spectrum of energy absorbed depends strongly on the steady-state inversion, we also study the effects produced by the collective behavior on the power broadening. In the two-atom case, we have

$$\langle S^z(\infty) \rangle = \text{Tr} \{ \rho(\infty) S^z \} = 2\psi_1(\infty) + \psi_2(\infty) - 1. \quad (2.20)$$

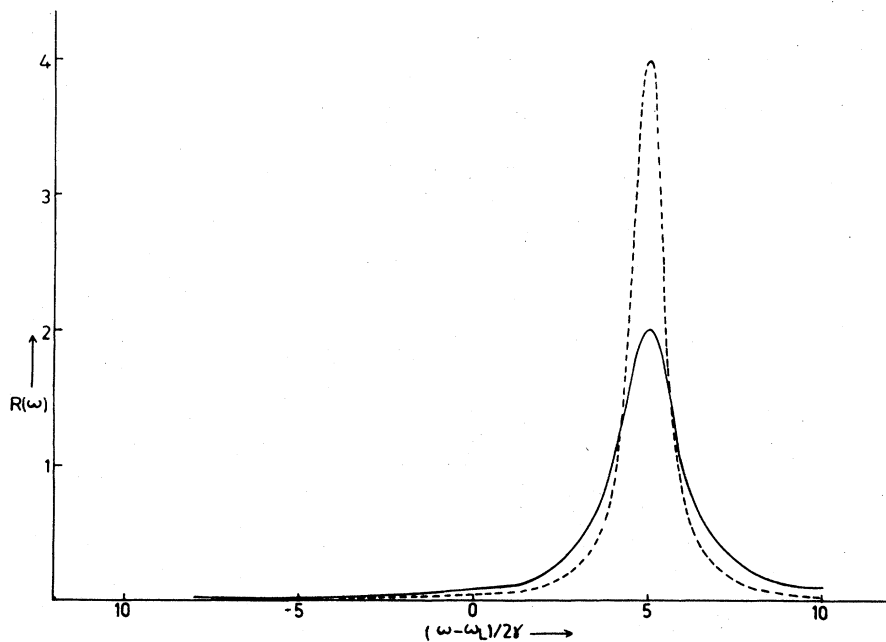


FIG. 4. Same as in Fig. 1 but with detuning  $\Delta=10\gamma$ ,  $\beta=0.1$ .

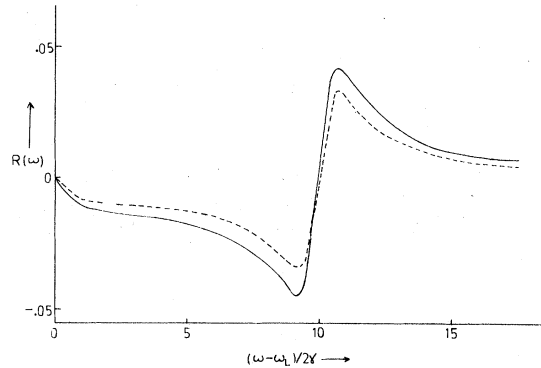


FIG. 3. Same as in Fig. 1 but with  $\beta=5.0$ ,  $\Delta=0$ .

As the detuning  $\Delta$  is increased,  $\langle S^z(\infty) \rangle$  decreases and the half-width at half the maximum height of the plot of  $\langle S^z(\infty) \rangle$  vs  $\Delta$ , gives the power broadening.<sup>12</sup> For the single atom, from Eq. (2.18), we readily obtain the power broadening  $\delta_P$  as

$$\delta_P = \gamma(1 + 2g^2/\gamma^2)^{1/2} \rightarrow \sqrt{2}g \text{ for } g \gg \gamma. \quad (2.21)$$

Thus  $\delta_P$  is proportional to  $g$  and hence to the intensity of the pump field.

### III. SCALING OF THE RESPONSE FUNCTIONS IN THE LIMIT OF STRONG COHERENT DRIVING FIELD

Here we analytically calculate the form of the absorption spectra in the limit when the driving field is very strong. We show that the absorption spectra is identical to that of a single two-level

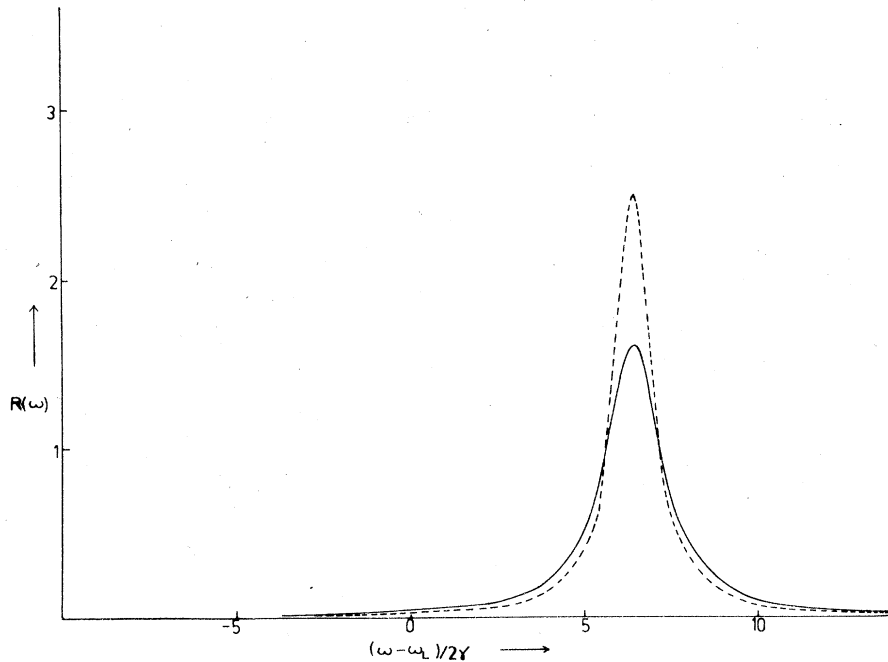


FIG. 5. Same as in Fig. 4 but with  $\beta=2$ ,  $\Delta=10\gamma$ .

atom except for a scale factor. We restrict our analysis to the case when the driving field is exactly on resonance with the atomic frequency. We use the method of secular perturbations well known in the theory of nonlinear oscillations<sup>13</sup> and which has been applied<sup>14-16</sup> to the problem of the absorption spectra of a single two-level atom and to the master equation (2.5). If we introduce the new angular momentum operators defined by

$$\begin{aligned} R^x &= S^y, \quad R^y = S^z, \quad R^z = S^x, \\ R^+ &= iS^z - \frac{1}{2}i(S^+ - S^-), \quad R^- = \frac{1}{2}(S^+ + S^-), \end{aligned} \quad (3.1)$$

then in the limit of very strong fields  $g \gg \gamma$  one can show that<sup>16</sup>

$$\begin{aligned} \langle \dot{R}^{\pm} \rangle &= (-\frac{3}{2}\gamma \mp 2ig)\langle R^{\pm} \rangle, \\ \langle \dot{R}^z \rangle &= -\gamma\langle R^z \rangle. \end{aligned} \quad (3.2)$$

In deriving Eqs. (3.2) the rapidly oscillating terms (the oscillating frequency is now  $2g$ ) have been ignored. The above conclusion is valid for the single-atom situation as well as the two-atom situation. The interaction with the external field (probe field with frequency  $\omega$ ) has the form

$$H_{\text{ext}} = g_1 S^+ e^{-i\Omega t} + \text{H. c.}, \quad \Omega = \omega - \omega_L, \quad (3.3)$$

since the density matrix is taken in a frame rotating with frequency  $\omega_L$  of the laser field. It can now be shown that Eqs. (3.2) are modified to

$$\begin{aligned} \langle \dot{R}^+ \rangle &= (-\frac{3}{2}\gamma - 2ig)\langle R^+ \rangle \\ &+ \{g_1 e^{-i\Omega t}\langle S^+ \rangle - g_1^* e^{i\Omega t}\langle S^- \rangle \\ &- \langle S^z \rangle (g_1 e^{-i\Omega t} + g_1^* e^{i\Omega t})\}, \end{aligned} \quad (3.4)$$

$$\begin{aligned} \langle \dot{R}^z \rangle &= -\gamma\langle R^z \rangle \\ &+ i(g_1 e^{-i\Omega t} - g_1^* e^{i\Omega t})\langle S^z \rangle. \end{aligned} \quad (3.5)$$

The steady-state linear response of  $\langle R^{\pm} \rangle$ ,  $\langle R^z \rangle$  to the probe field,  $g_1$  can be obtained from Eqs. (3.4) and (3.5) by replacing  $\langle S^{\pm} \rangle$ ,  $\langle S^z \rangle$  in curly brackets by their steady-state values in the absence of the probe field. For example

$$\begin{aligned} \langle R^+ \rangle &= e^{-i\Omega t} g_1 \left\{ \frac{\langle S^+ \rangle_{(0)} - \langle S^z \rangle_{(0)}}{\frac{3}{2}\gamma + 2ig - i\Omega} \right\} \\ &- e^{i\Omega t} g_1^* \left\{ \frac{\langle S^- \rangle_{(0)} + \langle S^z \rangle_{(0)}}{\frac{3}{2}\gamma + 2ig + i\Omega} \right\}. \end{aligned} \quad (3.6)$$

The linear response of  $\langle S^z \rangle$  to  $g_1$  can be obtained by substituting expressions like (3.6) into

$$\langle S^+(t) \rangle = \langle R^+(t) \rangle + \frac{1}{2}i\langle R^-(t) \rangle + \frac{1}{2}i\langle R^-(t) \rangle. \quad (3.7)$$

It is clear from the above analysis that the linear response of a single two-level atom and the two two-level atoms can differ only by a scale factor and it is this scale factor that is determined by the equilibrium values of  $\langle S^{\pm} \rangle$ ,  $\langle S^z \rangle$ . From the results of the Appendix A of Paper I, it follows that

$$\langle S^+ \rangle \rightarrow -4\gamma/3ig + O((\gamma/g)^2), \quad \langle S^z \rangle \sim O((\gamma/g)^2). \quad (3.8)$$

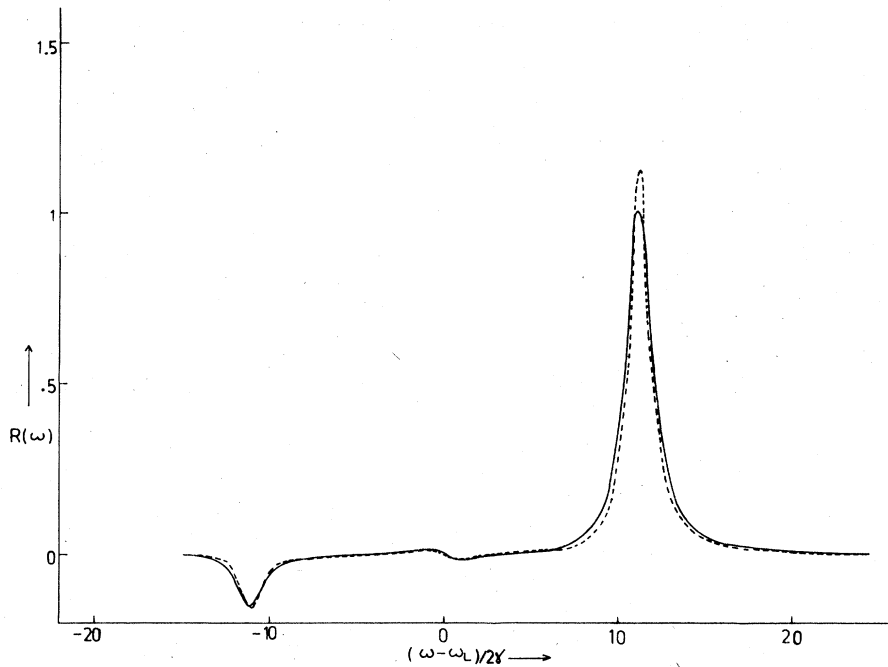


FIG. 6. Same as in Fig. 4 but with intense pump field  $\beta=5$ ,  $\Delta=10\gamma$ .

The corresponding result of the single-atom case is

$$\langle S^* \rangle \rightarrow -\gamma/2ig + O((\gamma/g)^2), \quad \langle S^* \rangle \sim O((\gamma/g)^2). \quad (3.9)$$

A comparison of Eqs. (3.6)–(3.9) gives the linear

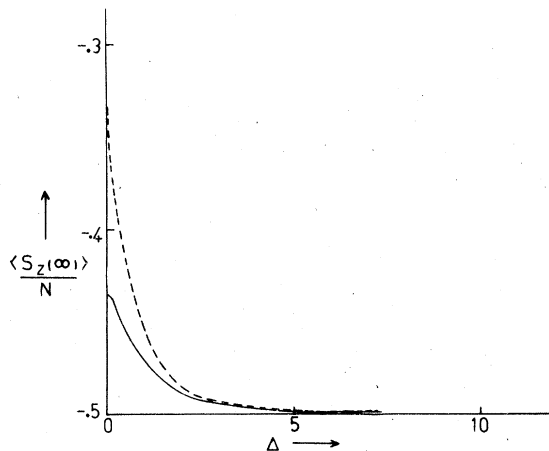


FIG. 7. Steady-state inversion as a function of detuning  $\Delta = (\omega_0 - \omega_L)$  for  $\beta=0.25$ .

response of two two-level atoms to be  $\frac{8}{3}$  of the linear response of a single two-level atom. This in turn implies that the energy absorbed per atom by a system of two atoms under cooperative conditions is  $\frac{4}{3}$  of the energy absorbed when cooperative effects are ignored.

The behavior of the absorption spectra with and without cooperative conditions for a system of two two-level atoms is shown in Figs. 1–6. The solid (dotted) lines in Figs. 1–6 denote the spectra under (without) cooperative conditions. It is seen from Figs. 1 and 2 that the cooperative effects play a dominant role for weak driving fields. As the laser intensity is increased, the two spectra acquire the same character (Fig. 3 and 4). We have checked with the actual numerical values for Fig. 3, the scaling obtained above. Figures 4–6 describe the behavior of the absorption spectra in the presence of detuning as the laser field strength is changed from very low to very high values. The peaks in Fig. 6 occur near  $\pm[(2g)^2 + \Delta^2]^{1/2}$ . Figure 5 also shows a power-dependent shift. Figure 7 describes the behavior of the steady-state inversion as a function of detuning parameter. It is seen that the cooperative effects compete with the power-dependent broadening effects. We have also checked that for strong fields (say  $\beta=10$ ) the steady-state inversion does not depend to a good approximation on the presence of cooperative effects.

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