Absorption spectrum of optically bistable systems*

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The quantum-mechanical theory of optical bistability developed in an earlier paper is generalized to calculate the absorption spectrum (gain coefficient) of an optically bistable system in the presence of a weak probe field. The behavior of the gain coefficient on the cooperative branch and the single-atom branch is analyzed in detail.

The rate of absorption of energy from a weak probe field propagating through a medium of strongly driven atoms was considered by Mollow, Haroche, and Hartmann and others[1-3] in the limiting case of vanishing atomic density. Their predictions have been tested recently by Wu *et al.* in a very impressive set of measurements.[4] In the experiments described in Ref. 4 a beam of independent atoms was driven by an external resonant source at different power levels. A weak probe field was sent through the atomic beam, and the transmitted power was monitored for different values of the probe carrier frequency. The probe line shape exhibited regions of negative absorption (gain) in the limit of strong driving fields. On the surface, this situation is quite surprising especially if we consider that, under strong driving conditions, the resonant atomic levels are almost equally populated. The process can be viewed as a parametric transfer of energy between the pump and the probe field with the atomic beam providing the nonlinear susceptibility that couples the waves. In the calculations it was assumed that the atoms interact independently with various radiation fields, viz., the laser field, the probe field, and the vacuum of the fields. Recently, several manifestations of the cooperative phenomena have been investigated both theoretically and experimentally (Refs. 5–8 and references therein). In this paper we examine the gain coefficient at the probe frequency for a system exhibiting optical bistability. We use the quantum-mechanical model of bistability of Ref. 7 and discuss the hysteresis behavior of the gain coefficient.[9] In our model we consider a collection of two-level atoms inside an effective single-mode Fabry-Perot resonator; the mode frequency is supposed to be on resonance with the atomic system. The entire system interacts with a strong laser field, also at resonance. Such a system has been shown to exhibit bistable behavior for a certain range of parameters. In what follows we examine the absorption of energy of such a system from a weak probe field of variable frequencies.

The work done by the external fields on a system of currents and charges is given by $\int \vec{J}\cdot\vec{E}\,d^3r$, where $\vec{J}$ is the total current in the system, which can be written $d\vec{p}/dt$, where $\vec{p}$ is the polarization of the system. Hence if we write the external field as

$$\vec{E}_{ex}=\sum_i \vec{\mathcal{E}}_i^{(+)}e^{-i\omega_i t}+\text{c.c.}\,, \tag{1}$$

and if we write the induced polarization as

$$\vec{p}=\sum_i \vec{p}_i^{(+)}e^{-i\omega_i t}+\text{c.c.}\,, \tag{2}$$

then the rate at which the energy of the system changes becomes

$$\frac{d\bar{u}}{dt}=\sum_i(-i\omega_i)(\vec{p}_i^{(+)}\cdot\vec{\mathcal{E}}_i^{(-)}-\text{c.c.}), \tag{3}$$

where a time averaging with respect to the rapidly oscillating terms has been done. Expression (3) is valid in general, i.e., it holds both for linear and nonlinear absorption. In what follows we consider only the linear absorption and we write

$$\vec{p}_i^{(+)}=\chi^{(i)}\vec{\mathcal{E}}_i^{(+)}, \tag{4}$$

then the time-averaged rate of change of the energy becomes

$$\frac{d\bar{u}}{dt}=\sum_i 2\omega_i|\vec{\mathcal{E}}_i^{(+)}|^2\,\mathrm{Im}\chi^{(i)}. \tag{5}$$

Equation 5 will now be applied to our system of two-level atoms interacting with a strong laser field and a weak probe field at frequency Ω. The

total polarization in the system can be written in terms of the dipole moment matrix element $\vec{d}$ and the collective operators as

$$\langle\vec{p}\rangle = \vec{d}(\langle S^+(t)\rangle + \langle S^-(t)\rangle)$$

and hence the change in the polarization due to the probe field will be

$$\begin{aligned}\langle\Delta\vec{p}\rangle &= \vec{d}(\langle\Delta S^+(t)\rangle + \text{c.c.}) \\ &= \vec{d}(e^{i\omega_L t}\langle\Delta S^+(t)\rangle + \text{c.c.}),\end{aligned} \tag{6}$$

and $\langle\Delta S^+\rangle$ represents the difference in the expectation value of S^+ in the presence and the absence of the probe field. Note that in the second line in (6) the response is in the rotating frame, rotating with frequency ω_L of the laser field. We will see that the linear response has the structure [cf. Eq. (21)]

$$\langle\Delta S^+(t)\rangle = Re^{it(\Omega-\omega_L)} + Te^{-it(\Omega-\omega_L)}. \tag{7}$$

Hence the polarization response becomes

$$\langle\Delta\vec{p}\rangle = \vec{d}(Re^{i\Omega t} + Te^{-it(\Omega-2\omega_L)} + \text{c.c.}), \tag{8}$$

and hence (3) reduces to

$$\frac{d\bar{u}}{dt} = -i\Omega\{R^*(\vec{d}\cdot\vec{g}^{(-)}) - \text{c.c.}\}. \tag{9}$$

In rotating-wave approximation and in a frame rotating with frequency ω_L, the interaction of the atomic system with the probe field $(\vec{g}^{(+)}e^{-i\Omega t} + \text{c.c.})$ has the form

$$H_{ex} = -(\vec{d}\cdot\vec{g}^{(+)}e^{-i(\Omega-\omega_L)t}S^+ + \text{H.c.}). \tag{10}$$

On defining

$$\begin{aligned}y_1^*(t) &= y_1^* e^{it(\Omega-\omega_L)} \\ &= (\sqrt{2}/\gamma_\perp)(-\vec{d}\cdot\vec{g}^{(-)}e^{i(\Omega-\omega_L)t}),\end{aligned} \tag{11}$$

the quantum Langevin equations (3.1) of I for the operators $X^{(\pm)} = S^\pm/N, X^{(3)} = S^z/N$ are now modified to

$$\frac{dX^{(+)}}{d\tau} = -X^{(+)} + 4CX^{(+)}X^{(3)} - \sqrt{2}\,iX^{(3)}(y + y_1^*) + \frac{F_+}{\sqrt{N}}, \tag{12}$$

$$\frac{dX^{(-)}}{d\tau} = -X^{(-)} + 4CX^{(3)}X^{(-)} + \sqrt{2}\,iX^{(3)}(y + y_1) + \frac{F_-}{\sqrt{N}}, \tag{13}$$

$$\frac{dX^{(3)}}{d\tau} = -(2X^{(3)} + 1) - 4CX^{(+)}X^{(-)} - \frac{iy}{\sqrt{2}}(X^{(+)} - X^{(-)}) - \frac{i}{\sqrt{2}}(X^{(+)}y_1 - \text{c.c.}) + \frac{i}{\sqrt{N}}F_3, \tag{14}$$

where all the symbols have the same meaning as in I, i.e., $\tau = \gamma_\perp t$, $C = g^2N/2\kappa\gamma_\perp$, $y = \sqrt{2}\,\Omega/\gamma_\perp$, where $\gamma_\perp$ is the single-atom decay rate, g is the atom-vacuum field coupling constant, κ is the inverse photon lifetime in cavity, and 2Ω is the Rabi frequency of the strong (pump) laser field. The fluctuating forces are δ correlated. The diffusion coefficients for these fluctuating forces are now different than (3.8) of I owing to the coupling with the probe field. Since in what follows we do not need the explicit form of these diffusion coefficients, we do not list them here. We can again carry out the system size expansion[10,11] and we find that to lowest order in $1/N$, $\langle X^{(\pm)}\rangle = X_{op}^{(\pm)} + O(1/N)$, $\langle X^{(3)}\rangle = X_{op}^{(3)} + O(1/N)$, etc., and that $X_{op}^{(\pm)}, X_{op}^{(3)}$ satisfy same equations as (12)–(14) without fluctuating forces and with $X \to X_{op}$. Since we are only interested in the linear response of the system, we write

$$X_{op}^{(\pm)} = X_0^{(\pm)} + \Delta X^{(\pm)}, \quad X_{op}^{(3)} = X_0^{(3)} + \Delta X^{(3)}, \tag{15}$$

where ΔX's are of first order in y_1. It is obvious that ΔX's in the steady state satisfy

$$\frac{d}{d\tau}\begin{pmatrix}\Delta X^{(+)}\\ \Delta X^{(-)}\\ \Delta X^{(3)}\end{pmatrix} = M\begin{pmatrix}\Delta X^{(+)}\\ \Delta X^{(-)}\\ \Delta X^{(3)}\end{pmatrix} + \begin{pmatrix}0 & 0 & -i\sqrt{2}\,y_1^*(t)\\ 0 & 0 & i\sqrt{2}\,y_1(t)\\ (-i/\sqrt{2})y_1(t) & (i/\sqrt{2})y_1^*(t) & 0\end{pmatrix}\begin{pmatrix}X_0^{(+)}\\ X_0^{(-)}\\ X_0^{(3)}\end{pmatrix}, \tag{16}$$

where M is the matrix determining the relaxation of the system [Eq. (3.22) of I], given by

$$M = \begin{pmatrix}-A & 0 & -\sqrt{2}\,ix\\ 0 & -A & \sqrt{2}\,ix\\ -i\sqrt{2}\,x\Lambda & i\sqrt{2}\,x\Lambda & -2\end{pmatrix},$$

$$A = \frac{y}{x}, \quad y = x + \frac{2cx}{1+x^2}, \quad \Lambda = \left(1 - \frac{y}{2x}\right); \tag{17}$$

the steady-state values of X_0's are given by

$$\begin{aligned}X_0^{(\pm)} &= \pm(i/\sqrt{2})x/(1+x^2). \\ X_0^{(3)} &= -1/2(1+x^2).\end{aligned} \tag{18}$$

It is clear from (16) that $\Delta X^{(\pm)}$, $\Delta X^{(3)}$ are given by

$$\begin{pmatrix}\Delta X^{(+)}\\ \Delta X^{(-)}\\ \Delta X^{(3)}\end{pmatrix} = f_+e^{-it(\Omega-\omega_L)} + f_-e^{it(\Omega-\omega_L)}, \tag{19}$$

where

$$f_+ = \frac{iy_1}{\sqrt{2}}\Big[-Z-M\Big]^{-1}\begin{bmatrix}0\\ 2X_0^{(3)}\\ -X_0^{(+)}\end{bmatrix},$$

$$f_- = \frac{-iy_1^*}{\sqrt{2}}\Big[Z-M\Big]^{-1}\begin{bmatrix}2X_0^{(3)}\\ 0\\ -X_0^{(-)}\end{bmatrix}, \qquad (20)$$

$$Z = \frac{i(\Omega-\omega_L)}{\gamma_\perp},$$

and hence

$$\langle \Delta S^+\rangle = N\Delta X^{(+)}$$

$$= \frac{iN}{\sqrt{2}}\sum_j\Big[y_1(-Z-M)^{-1}_{ij}\begin{bmatrix}0\\ 2X_0^{(3)}\\ -X_0^{(+)}\end{bmatrix}_j e^{-it(\Omega-\omega_L)}$$

$$-y_1^*(Z-M)^{-1}_{ij}\begin{bmatrix}2X_0^{(3)}\\ 0\\ -X_0^{(-)}\end{bmatrix}_j e^{it(\Omega-\omega_L)}\Big]. \qquad (21)$$

On comparison with (7) we get

$$R = \Big(+\frac{iN}{\sqrt{2}}\Big)\Big(\frac{\sqrt{2}}{\gamma_\perp}\Big)(\vec{d}\cdot\vec{g}^{(-)})$$

$$\times\sum_j (Z-M)^{-1}_{ij}\begin{bmatrix}2X_0^{(3)}\\ 0\\ -X_0^{(-)}\end{bmatrix}_j \qquad (22)$$

and hence on using the steady-state values of $X_0^{(\pm)}$, $X_0^{(3)}$ we get

$$\frac{d\bar{u}}{dt} = \frac{|\vec{d}\cdot\vec{g}^{(-)}|^2 N\Omega}{\gamma_\perp(1+x^2)}\,\mathrm{Re}\Big((Z-M)^{-1}_{11} - \frac{ix}{\sqrt{2}}(Z-M)^{-1}_{13}\Big) \qquad (23)$$

$$= |\vec{d}\cdot\vec{g}^{(-)}|^2\frac{N\Omega}{\gamma_\perp}S(\omega), \qquad (24)$$

where

$$S(\omega) = \mathrm{Re}\,\alpha(Z), \quad Z = i(\Omega-\omega_L)/\gamma_\perp, \qquad (25)$$

and where on simplification α is found to be

$$\alpha(Z) = \frac{(Z+2)(Z+y/x)+x^2\{2-Z-2y/x\}}{(1+x^2)(Z+y/x)[(Z+2)(Z+y/x)+4x^2(1-y/2x)]} \qquad (26)$$

It is clear that if $S(\omega)$ is positive (negative) then the system absorbs (emits) energy from (into) the probe field. We examine numerically the behavior of S for different values of the laser field strengths y. We observe that in the limit of low concentration $C \to 0$, our result for the absorption spectrum goes over to the well-known spectrum[1-1]

$$\alpha \to \frac{(Z+2)(Z+1)-y^2Z}{(1+y^2)(Z+1)[(Z+2)(Z+1)+2y^2]}. \qquad (27)$$

The absorption spectrum is related to the correlation function of the atomic system $\langle[S^+(t), S^-(0)]\rangle$ in the steady state. In fact the fluctuation-dissipation theorem or the linear-response theory shows that (cf. Ref. 1)

$$\alpha(Z) = \frac{1}{N}\int_0^\infty d\tau\, e^{-Z\tau}\langle[S^-(0), S^+(\tau)]\rangle, \qquad (28)$$

where the steady-state correlation is to be calculated in the absence of the probe field. Using the system size expansion of Refs. 10 and 11, and following the procedure of I, we calculated both the correlation functions appearing in (28) and we checked that result (26) is obtained.

Typical absorption spectra are shown Figs. 1 and 2 and 3 and 4 along cooperative and single-atom branches, respectively. The parameter C is chosen so that the system exhibits bistability. From these figures the hysteresis characteristics of the absorption spectra are quite obvious. As expected, the main differences between the low-density limit and the present prediction is along the cooperative branch.

The absorption spectrum could be expressed in terms of the eigenvalues $\lambda_1, \lambda_2, \lambda_3$,

$$\lambda_1 = -\frac{y}{x}, \quad \lambda_{\frac{2}{3}} = -\frac{(2+A)}{2} \pm \frac{1}{2}$$

$$\times\Big((2+A)^2 - 8(1+x^2)\frac{dy}{dx}\Big)^{1/2}, \qquad (29)$$

of the matrix M. When this is done Eq. (26) then reduces to

$$\alpha = \frac{1}{1+x^2}\Big[\frac{1}{2(Z+y/x)} - \frac{x^2}{(\lambda_2-\lambda_3)}$$

$$\times\Big(\frac{2+\lambda_2}{(\lambda_2+y/x)(Z-\lambda_2)} - \frac{(2+\lambda_3)}{(\lambda_3+y/x)(Z-\lambda_3)}\Big)\Big]. \qquad (30)$$

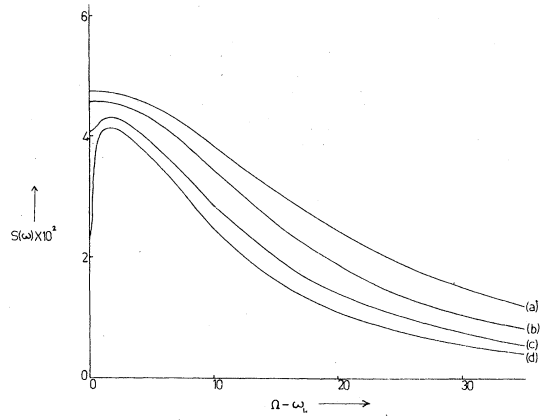

FIG. 1. Absorption spectrum on the cooperative branch for $c=10$ and for (a) $y=3$, (b) $y=9$, (c) $y=10.6$, (d) $y=11.0$.

The eigenvalue λ_2 becomes soft near the upper bistability threshold y_{max}, and $y \gg x$ in the neighborhood of y_{max} and hence we get approximately

$$\alpha \approx \frac{1}{2(Z+y/x)} - \frac{2x^2}{1+x^2}\left(\frac{x^2}{y^2}\right)\frac{1}{Z}. \tag{31}$$

On the cooperative branch the absorption spectrum is quite broad for weak pump fields. This follows from (30) since for weak fields we have $x \ll 1$ and one of the eigenvalues then is approximately equal to $-(1+2C)$; hence the spectrum is very broad. For increasing values of y, dy/dx decreases and hence the width decreases. As the system is driven near the upper bistability threshold, the absorption spectrum has a contribution which narrows significantly (Fig. 2) as is clear from (31) which shows an inverted spike whose weight is rather small since $y \gg x$.

Beyond the upper bistable threshold Figs. 3 and 4 the absorption spectra become asymptotically identical to those discussed by Mollow[1] for very

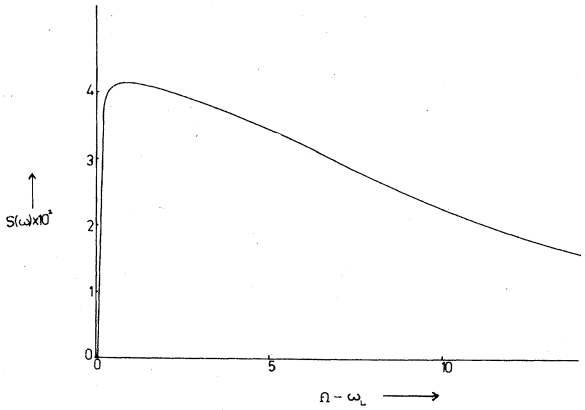

FIG. 2. Same as in Fig. 1 but for parameters very close to the bistability threshold $y=11.0558$, $x=1.11$.

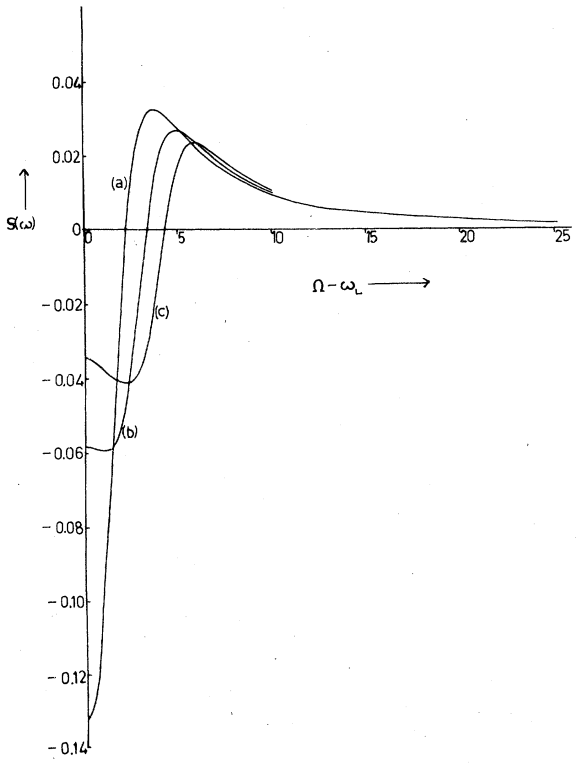

FIG. 3. Absorption spectrum on the single-atom branch for $c=10$ for (a) $y=8.736$, (b) $y=8.818$, (c) $y=8.944$.

large driving fields. This follows from (26) since for very large driving fields on the single-atom branch $y \sim x$ and hence (26) and (27) become identical. On decreasing the pump field towards the lower bistability point, a large gain peak develops

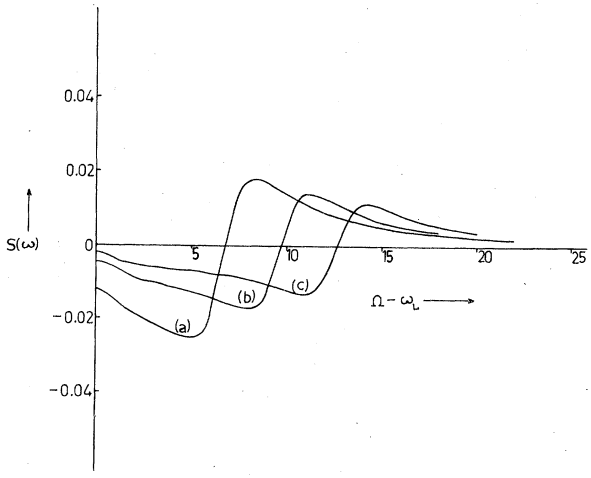

FIG. 4. Same as in Fig. 3 but for (a) $y=9.5$, (b) $y=10.6$, (c) $y=12$.

near $\Omega - \omega_L \sim \gamma_\perp x$. Just before the onset of the second bistable transition, we predict the largest gain coefficient for a resonant probe field. This follows from (26) since for the resonant case $z \to 0$ and the denominator in (26) becomes proportional to dy/dx which goes to zero as the bistability threshold is approached. Thus the gain behaves as $(y - y_{\min})^{-1/2}$, where $y_{\min}$ is the lower bistability threshold. The nature of the spectra near $y_{\min}$ is different from those for large values of y on the single atom branch because of the nature of the eigenvalue λ_2, λ_3 which are complex conjugates of each other for large values of y. However near $y_{\min}$, the imaginary parts of λ_2, λ_3 become zero.

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