PHYSICAL REVIEW A VOLUME 5, NUMBER 5 MAY 1972

# Stimulated Emission and Absorption near Resonance for Driven Systems

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(Received 20 September 1971)

The rate of absorption of energy from a weak signal field by an atom driven by a strong pump field is evaluated. The pump field and the signal field are assumed to induce transitions between the same pair of states, and their frequencies are both assumed to lie near the atomic resonance frequency for the transition in question. We find that the signal-field absorption line-shape function takes on negative values, representing stimulated emission rather than absorption, even though population inversion does not occur. This amplification of the signal field, which is most pronounced at high pump intensities, is shown to occur primarily at the expense of the pump field, which suffers an increased rate of attenuation. The results are discussed in the context of a theorem which expresses the absorption line-shape function for general atomic systems in terms of a suitable atomic correlation function.

## I. INTRODUCTION

In two previous papers[1,2] we have evaluated the frequency spectrum of the radiation emitted during transitions between a particular pair of levels by an atom which is driven by a (possibly) strong harmonically oscillating external field.[3] In this paper we consider the complementary process, that of the absorption of radiation by a similarly driven system. The atom is thus assumed to be excited simultaneously by two fields of nearly equal frequency, a strong pump field and a weak perturbing or "signal" field, and the rate of absorption of energy from the signal field is found as a function of its frequency.[3a]

Our analysis is carried out within the context of a simple two-level model, in which atomic relaxation is treated in general terms. The pump field and the signal field are both treated classically, and both are assumed to oscillate harmonically at frequencies near the resonance frequency of the atom.[4,5] The absorption line-shape function is found by first finding the effect of the signal field on the equilibrium atomic density matrix, and then using this solution to evaluate the rate of absorption of quanta from the signal field.

A striking feature of the expression we find for the absorption line-shape function is that in some cases it takes on negative values, even though population inversion never occurs. The negative values, which are appreciable for intense pump fields, correspond to stimulated emission rather than to absorption, and imply amplification rather than attenuation of the signal field. Energy balance is discussed, and it is shown that for very intense pump fields the amplification of the signal field occurs primarily at the expense of the pump field, which suffers an increased rate of attenuation due to the presence of the signal field.

Our analysis is discussed in the context of a general relation which expresses the absorption spectrum of an atom in terms of a certain atomic correlation function, which is similar to, but not identical to, the one which determines the emission spectrum for the same system.[1,2] The possibility of extending our results to include more general pumping mechanisms emerges naturally from this discussion.

The basic model of a pumped two-level atom with general relaxation coefficients is introduced in Sec. II. In Sec. III the absorption line-shape function is evaluated, and the limiting cases of interest are treated. The results are then discussed in Sec. IV in the context of the general correlation-function theory of absorption.

## II. DRIVEN TWO-LEVEL SYSTEM WITH GENERAL RELAXATION COEFFICIENTS

Let us consider an atom with two energy eigenstates $|0\rangle$ and $|1\rangle$ and corresponding eigenvalues $E_0$ and $E_1$, where

$$E_1 - E_0 \equiv \hbar\omega_{10} \; . \qquad (2.1)$$

It is convenient to represent atomic relaxation by introducing separate off-diagonal and diagonal relaxation coefficients $\kappa'$, $\kappa_{01}$, and $\kappa_{10}$, in terms of which the equations of motion for the matrix elements $\alpha(t)=\rho_{10}(t)$, $\alpha^*(t)=\rho_{01}(t)$, $\bar{n}(t)=\rho_{11}(t)$, and $\bar{m}(t)=\rho_{00}(t)$ may be expressed, in the absence of external excitation, as

$$\left(\frac{d}{dt}+\kappa'+i\omega_{10}\right)\alpha(t)=0 \; , \qquad (2.2a)$$

$$\left(\frac{d}{dt}+\kappa'-i\omega_{10}\right)\alpha^*(t)=0 \; , \qquad (2.2b)$$

$$\left(\frac{d}{dt}+\kappa_{01}\right)\bar{n}(t)-\kappa_{10}\bar{m}(t)=0 \; , \qquad (2.2c)$$

$$\left(\frac{d}{dt}+\kappa_{10}\right)\bar{m}(t)-\kappa_{01}\bar{n}(t)=0 \; . \qquad (2.2d)$$

The equilibrium solution to these equations and the trace relation

$$\bar{n}(t)+\bar{m}(t)=1 \tag{2.3}$$

is represented by vanishing off-diagonal matrix elements, and by the diagonal elements

$$\bar{n}^{(0)}=\kappa_{10}/\kappa , \tag{2.4a}$$

$$\bar{m}^{(0)}=\kappa_{01}/\kappa , \tag{2.4b}$$

where the parameter $\kappa$ is defined as

$$\kappa \equiv \kappa_{10}+\kappa_{01} . \tag{2.5}$$

We may note that in the strong collision model of atomic relaxation,[6] $\kappa$ is the mean collision rate, $\bar{n}^{(0)}$ and $\bar{m}^{(0)}$ are the mean thermal occupation numbers for the states $|1\rangle$ and $|0\rangle$, respectively, and $\kappa'=\kappa$. In the case of (zero-temperature) radiative relaxation,[1] on the other hand, $\kappa$ is the spontaneous emission rate, $\bar{n}^{(0)}=0$, $\bar{m}^{(0)}=1$, and $\kappa'=\frac{1}{2}\kappa$.

Let us now suppose that the atom is driven by an external field, with positive- and negative-frequency parts $\mathcal{E}(t)$ and $\mathcal{E}^*(t)$, respectively, and polarization specified by the unit vector $\hat{e}_0$,

$$\vec{\mathrm{E}}(t)=(1/\sqrt{2})\hat{e}_0[\mathcal{E}(t)+\mathcal{E}^*(t)] . \tag{2.6}$$

We assume that the field oscillates harmonically

$$\mathcal{E}(t)=\mathcal{E}_0 e^{-i\omega t} \tag{2.7}$$

at a frequency nearly equal to the atomic resonance frequency,

$$\omega \approx \omega_{10} . \tag{2.8}$$

In the presence of the driving field the equations of motion for the atomic matrix elements, in the resonant approximation, may be expressed with the aid of Eqs. (2.4) as

$$\left(\frac{d}{dt}+\kappa'+i\omega_{10}\right)\alpha(t)=i\lambda\mathcal{E}(t)\,[\bar{m}(t)-\bar{n}(t)] , \tag{2.9a}$$

$$\left(\frac{d}{dt}+\kappa'-i\omega_{10}\right)\alpha^*(t)=-i\lambda^*\mathcal{E}^*(t)\,[\bar{m}(t)-\bar{n}(t)] , \tag{2.9b}$$

$$\left(\frac{d}{dt}+\kappa\right)\bar{n}(t)-\kappa\bar{n}^{(0)}[\bar{n}(t)+\bar{m}(t)] = -i\lambda^*\mathcal{E}^*(t)\alpha(t)+i\lambda\mathcal{E}(t)\alpha^*(t) , \tag{2.9c}$$

$$\left(\frac{d}{dt}+\kappa\right)\bar{m}(t)-\kappa\bar{m}^{(0)}[\bar{n}(t)+\bar{m}(t)] = i\lambda^*\mathcal{E}^*(t)\alpha(t)-i\lambda\mathcal{E}(t)\alpha^*(t) , \tag{2.9d}$$

where the parameter $\lambda$ is defined in terms of the dipole matrix element $\vec{\mu}_{10}$ as

$$\lambda \equiv (\hat{e}_0\cdot\vec{\mu}_{10})/\hbar\sqrt{2} . \tag{2.10}$$

The equilibrium solution to Eqs. (2.9), (2.7), and (2.3) is characterized by constant diagonal elements $\bar{n}$ and $\bar{m}$, and by off-diagonal elements $\alpha(t)=\bar{\alpha}e^{-i\omega t}$ and $\alpha^*(t)=\bar{\alpha}^*e^{i\omega t}$. The parameters $\bar{\alpha}$, $\bar{n}$, and $\bar{m}$ may be obtained from the relations

$$\bar{\alpha}=i\lambda\mathcal{E}_0(\bar{m}-\bar{n})/z^* , \tag{2.11a}$$

$$\bar{m}-\bar{n}=(\bar{m}^{(0)}-\bar{n}^{(0)})\kappa\,|z|^2/(\kappa'\Omega^2+\kappa\,|z|^2) , \tag{2.11b}$$

$$\bar{m}+\bar{n}=1 , \tag{2.11c}$$

where the parameters $\Omega$ and $z$ are defined as

$$\Omega \equiv 2\,|\lambda\mathcal{E}_0| , \tag{2.12a}$$

$$z \equiv \kappa'+i\Delta\omega , \tag{2.12b}$$

$$\Delta\omega \equiv \omega-\omega_0 . \tag{2.12c}$$

The rate at which quanta are absorbed from the field is

$$\mathcal{W}=-i\lambda^*\mathcal{E}_0^*\bar{\alpha}+i\lambda\mathcal{E}_0\bar{\alpha}^* = \tfrac{1}{2}\Omega^2\kappa\kappa'(\bar{m}^{(0)}-\bar{n}^{(0)})/(\kappa'\Omega^2+\kappa\,|z|^2) . \tag{2.13}$$

The time-dependent solutions to the linear homogeneous Eqs. (2.9) for the matrix elements $\alpha(t)$, $\alpha^*(t)$, $\bar{n}(t)$, and $\bar{m}(t)$ have the form of linear functions of the initial values $\alpha(0)$, $\alpha^*(0)$, $\bar{n}(0)$, and $\bar{m}(0)$. The solution for $\alpha(t)$, for example, takes the form

$$\alpha(t)=\mathfrak{U}_{\alpha\alpha}(t)\alpha(0)+\mathfrak{U}_{\alpha\alpha^*}(t)\alpha^*(0) +\mathfrak{U}_{\alpha n}(t)\bar{n}(0)+\mathfrak{U}_{\alpha m}(t)\bar{m}(0) . \tag{2.14}$$

By directly solving[7] Eqs. (2.9) we find that the Laplace transform functions

$$\hat{\mathfrak{U}}(s)\equiv\int_0^\infty dt\,e^{-st}\,\mathfrak{U}(t) \tag{2.15}$$

of $\mathfrak{U}_{\alpha\alpha}$, $\mathfrak{U}_{\alpha n}$, and $\mathfrak{U}_{\alpha m}$ may be obtained from the relations

$$\hat{\mathfrak{U}}_{\alpha\alpha}(s-i\omega)=\frac{(s+z)(s+\kappa)+\frac{1}{2}\Omega^2}{f(s)} \tag{2.16a}$$

$$\hat{\mathfrak{U}}_{\alpha n}(s-i\omega)=\frac{-i\lambda\mathcal{E}_0(s+z)[s-\kappa(\bar{m}^{(0)}-\bar{n}^{(0)})]}{s f(s)} , \tag{2.16b}$$

$$\hat{\mathfrak{U}}_{\alpha m}(s-i\omega)=\frac{i\lambda\mathcal{E}_0(s+z)[s+\kappa(\bar{m}^{(0)}-\bar{n}^{(0)})]}{s f(s)} , \tag{2.16c}$$

where $f(s)$ is the third-degree polynomial

$$f(s)\equiv(s+\kappa)(s+z)(s+z^*)+\Omega^2(s+\kappa') . \tag{2.17}$$

We note that in the case of collisional relaxation, the three roots $s_0$, $s_+$, and $s_-$ of $f(s)$ are

$$s_0=-\kappa , \tag{2.18a}$$

$$s_\pm=-\kappa\pm i\Omega' , \tag{2.18b}$$

where

$$\Omega'\equiv[\Omega^2+(\Delta\omega)^2]^{1/2} . \tag{2.19}$$

In the case of strong driving fields, the roots are well approximated for general relaxation coefficients by the relations

$$s_0=-\kappa' , \tag{2.20a}$$

$$s_{\pm}=-\tfrac{1}{2}(\kappa+\kappa')\pm i\,\Omega \qquad \text{for } \Omega\gg\kappa,\ |z|. \tag{2.20b}$$

## III. LINE-SHAPE FUNCTION FOR ABSORPTION AND STIMULATED EMISSION

Let us now suppose that a weak perturbing or "signal" field, with positive frequency part

$$\mathcal{E}'(t)=\mathcal{E}_0'\,e^{-i\nu t} \tag{3.1}$$

and polarization specified by the unit vector $\hat{e}_0'$, is applied in addition to the "pump" field given by Eq. (2.7). We assume that the signal-field frequency $\nu$ nearly coincides with the atomic resonance frequency, and hence also with the pump frequency, so that the frequency difference

$$\Delta\nu\equiv\nu-\omega \tag{3.2}$$

is a small quantity. The equations of motion for the atomic matrix elements, in the resonant approximation, may then be obtained simply by making the substitution

$$\lambda\mathcal{E}(t)\to\lambda\mathcal{E}(t)+\lambda'\mathcal{E}'(t) \tag{3.3}$$

in Eqs. (2.9). The parameter $\lambda'$ in this relation is defined as

$$\lambda'\equiv(\hat{e}_0'\cdot\vec{\mu}_{10})/\hbar\sqrt{2}\ . \tag{3.4}$$

The effect of the weak signal field on the equilibrium solution for the atomic density matrix is to induce small components oscillating at frequencies which differ by $\pm\Delta\nu$ from the frequencies of the unperturbed components, as well as small corrections in the unperturbed components themselves. The atomic matrix elements thus have the form

$$\alpha(t)=(\overline{\alpha}+\delta\alpha_0)e^{-i\omega t}+\delta\alpha_+e^{-i\nu t}+(\delta\alpha_-)^*e^{-i(\omega-\Delta\nu)t}\ , \tag{3.5a}$$

$$\alpha^*(t)=(\overline{\alpha}+\delta\alpha_0)^*e^{i\omega t}+(\delta\alpha_+)^*e^{i\nu t}+\delta\alpha_-e^{i(\omega-\Delta\nu)t}\ , \tag{3.5b}$$

$$\overline{n}(t)=(\overline{n}+\delta\overline{n})+\eta e^{-i\Delta\nu t}+\eta^*e^{i\Delta\nu t}\ , \tag{3.5c}$$

$$\overline{m}(t)=(\overline{m}-\delta\overline{n})-\eta e^{-i\Delta\nu t}-\eta^*e^{i\Delta\nu t}\ , \tag{3.5d}$$

where $\overline{\alpha}$, $\overline{n}$, and $\overline{m}$ as given by Eqs. (2.11) are determined in the presence of the pump field alone, and $\delta\alpha_0$, $\delta\overline{n}$, $\delta\alpha_\pm$, and $\eta$ are small constant parameters. By substituting Eqs. (3.5) into Eqs. (2.9) and (2.3) and making the substitution (3.3), we find that to lowest order in the signal-field strength the parameters in Eqs. (3.5) satisfy the equations

$$(-i\Delta\nu+z^*)\delta\alpha_++2i\lambda\mathcal{E}_0\eta=i\lambda'\mathcal{E}_0'(\overline{m}-\overline{n})\ , \tag{3.6a}$$

$$(-i\Delta\nu+z)\,\delta\alpha_--2i\lambda^*\mathcal{E}_0^*\eta=0\ , \tag{3.6b}$$

$$(-i\Delta\nu+\kappa)\eta+i\lambda^*\mathcal{E}_0^*\delta\alpha_+-i\lambda\mathcal{E}_0\delta\alpha_-=i\lambda'\mathcal{E}_0'\overline{\alpha}^*\ , \tag{3.6c}$$

and

$$z^*\delta\alpha_0+2i\lambda\mathcal{E}_0\delta\overline{n}=-2i\lambda'\mathcal{E}_0'\eta^*\ , \tag{3.7a}$$

$$z(\delta\alpha_0)^*-2i\lambda^*\mathcal{E}_0^*\delta\overline{n}=2i\lambda'^*\mathcal{E}_0'^*\eta\ , \tag{3.7b}$$

$$\kappa\delta\overline{n}+i\lambda^*\mathcal{E}_0^*\delta\alpha_0-i\lambda\mathcal{E}_0(\delta\alpha_0)^* =-i\lambda'^*\mathcal{E}_0'^*\delta\alpha_++i\lambda'\mathcal{E}_0'(\delta\alpha_+)^*\ . \tag{3.7c}$$

The rate $\mathcal{W}'$ at which quanta are absorbed from the signal field is

$$\mathcal{W}'=-i\lambda'^*\mathcal{E}_0'^*\delta\alpha_++i\lambda'\mathcal{E}_0'(\delta\alpha_+)^*\ . \tag{3.8}$$

Similarly, the increase in the rate at which quanta are absorbed from the pump field is

$$\delta\mathcal{W}=-i\lambda^*\mathcal{E}_0^*\,\delta\alpha_0+i\lambda\mathcal{E}_0(\delta\alpha_0)^*\ . \tag{3.9}$$

The sum of these two quantities, according to Eq. (3.7c), is

$$\kappa\delta\overline{n}=\delta\mathcal{W}+\mathcal{W}'\ , \tag{3.10}$$

and is equal to the increase in the net rate of dissipative transitions from the atomic state $|1\rangle$ to the state $|0\rangle$.

By solving the three coupled equations (3.6) for the quantities $\delta\alpha_+$, $\delta\alpha_-$, and $\eta$ in terms of the quantities on the right-hand sides and then making use of Eq. (2.11a) for $\overline{\alpha}$, we find the relations

$$\delta\alpha_+=\frac{i\lambda'\mathcal{E}_0'(\overline{m}-\overline{n})[(-i\Delta\nu+\kappa)(-i\Delta\nu+z)+\frac{1}{2}i\Omega^2\Delta\nu/z\,]}{f(-i\Delta\nu)} \tag{3.11a}$$

$$\delta\alpha_-=\frac{2i\lambda'\mathcal{E}_0'(\overline{m}-\overline{n})(\lambda^*\mathcal{E}_0^*)^2(-i\Delta\nu+2\kappa')}{zf(-i\Delta\nu)}\ , \tag{3.11b}$$

$$\eta=\frac{\lambda'\mathcal{E}_0'(\overline{m}-\overline{n})\lambda^*\mathcal{E}_0^*(-i\Delta\nu+z)(-i\Delta\nu+2\kappa')}{zf(-i\Delta\nu)}\ , \tag{3.11c}$$

where $f$ is the polynomial defined by Eq. (2.17).

The absorption line-shape function $\mathcal{W}'(\nu)$ for the atom in the presence of the pump field is thus given quite generally by Eqs. (3.8) and (3.11a), in terms of the parameters defined by Eqs. (2.11b), (2.12), and (3.2).

By solving the Eqs. (3.7) for the quantities $\delta\alpha_0$, $(\delta\alpha_0)^*$, and $\delta\overline{n}$ in terms of the quantities on the right-hand sides, we find, with the aid of the relation (3.8),

$$\delta\alpha_0=\frac{1}{f(0)}\,[-2i\lambda'\mathcal{E}_0'(\kappa z+\tfrac{1}{2}\Omega^2)\eta^*+4i\lambda'^*\mathcal{E}_0'^*(\lambda\mathcal{E}_0)^2\eta -2i\lambda\mathcal{E}_0z\,\mathcal{W}'\,]\ , \tag{3.12a}$$

$$\delta\overline{n}=\frac{1}{f(0)}\,\{[\,-2\lambda'^*\mathcal{E}_0'^*\lambda\mathcal{E}_0z^*\eta+\text{c.c.}\,]+|z|^2\mathcal{W}'\}\ , \tag{3.12b}$$

where $f(0)$, according to Eq. (2.17), is just

$$f(0)=\kappa'\Omega^2+\kappa\,|z|^2\ . \tag{3.13}$$

By substituting Eq. (3.12a) and its complex conjugate into Eq. (3.9), we find that

$$\delta\mathcal{W}=\frac{1}{f(0)}\,\{[\,-2\lambda'^*\mathcal{E}_0'^*\lambda\mathcal{E}_0\kappa z^*\eta+\text{c.c.}\,]-\kappa'\Omega^2\mathcal{W}'\}\ . \tag{3.14}$$

Thus the increase $\delta\bar{n}$ in the population of the upper atomic state and the increase $\delta\mathcal{W}$ in the rate of absorption of quanta from the pump field are given by Eqs. (3.12b) and (3.14), respectively, where $\eta$ is given by Eq. (3.11c). It is worth noting that $\mathcal{W}'$, $\delta\mathcal{W}$, $\delta\bar{n}$, and $\delta\alpha_0$ are all proportional to the square of the signal-field amplitude $\mathcal{E}_0'$, while $\delta\alpha_\pm$ and $\eta$ are simply proportional to $\mathcal{E}_0'$.

Before proceeding further with our discussion, it is convenient to check the consistency of our approximations by considering the limit in which the signal-field frequency $\nu$ approaches the frequency $\omega$ of the pump field. In that limit the atom is effectively driven by a single monochromatic field, with the slowly varying complex amplitude $\hat{e}_0\mathcal{E}_0 + \hat{e}_0'\mathcal{E}_0' e^{-i\Delta\nu t}$. We should therefore expect the atomic matrix elements to adiabatically maintain the values which have been found for them in the presence of a constant pump field, i.e., to be obtainable by making the substitution

$$\lambda\mathcal{E}_0 \to \lambda\mathcal{E}_0 + \lambda'\mathcal{E}_0' e^{-i\Delta\nu t} \tag{3.15}$$

in Eqs. (2.11) and (2.12a). When this substitution is made and the resulting expressions are expanded to second order in $\mathcal{E}_0'$ (and first order in harmonics of $\Delta\nu$), it is found that the atomic matrix elements indeed have the form given by Eqs. (3.5), where the small parameters $\delta\alpha_\pm$, $\eta$, $\delta\alpha_0$, and $\delta\bar{n}$ are given as

$$\delta\alpha_+ = i\lambda'\mathcal{E}_0'(\bar{m}-\bar{n})\kappa z/f(0)\,, \tag{3.16a}$$

$$\delta\alpha_- = 4i\lambda'\mathcal{E}_0'(\bar{m}-\bar{n})\kappa'(\lambda^*\mathcal{E}_0^*)^2/zf(0)\,, \tag{3.16b}$$

$$\eta = 2\lambda'\mathcal{E}_0'(\bar{m}-\bar{n})\kappa'\lambda^*\mathcal{E}_0^*/f(0)\,, \tag{3.16c}$$

$$\delta\alpha_0 = -8i\,|\lambda'\mathcal{E}_0'|^2(\bar{m}-\bar{n})\kappa\kappa' z\lambda\mathcal{E}_0/[f(0)]^2\,, \tag{3.16d}$$

$$\delta\bar{n} = -2\,|\lambda'\mathcal{E}_0'|^2(\bar{m}-\bar{n})\kappa'(\kappa'\Omega^2-\kappa|z|^2)/[f(0)]^2\,. \tag{3.16e}$$

These are exactly the values given by Eqs. (3.11), (3.12), and (3.8) in the limit $\Delta\nu \to 0$.

The absorption line-shape function $\mathcal{W}'(\nu)$ as given by Eqs. (3.8) and (3.11a) may be expressed in the case of the strong collision model of atomic relaxation ($\kappa'=\kappa$) in the form

$$\mathcal{W}'(\nu) = |\lambda'\mathcal{E}_0'|^2\left\{\frac{M_0-N_0\Delta\nu}{(\Delta\nu)^2+\kappa^2}+\frac{M_+-N_+(\Delta\nu+\Omega')}{(\Delta\nu+\Omega')^2+\kappa^2}+\frac{M_--N_-(\Delta\nu-\Omega')}{(\Delta\nu-\Omega')^2+\kappa^2}\right\}, \tag{3.17}$$

where the parameters $M_0$, $N_0$, $M_\pm$, and $N_\pm$ are given in terms of the quantities defined by Eqs. (2.12) and and (2.19) by the relations

$$M_0 = (\bar{m}-\bar{n})\Omega^2\kappa^3/\Omega'^2\,|z|^2\,, \tag{3.18a}$$

$$N_0 = -(\bar{m}-\bar{n})\Omega^2\kappa\Delta\omega/\Omega'^2\,|z|^2\,, \tag{3.18b}$$

$$M_\pm = \tfrac{1}{2}(\bar{m}-\bar{n})\kappa(1\pm\Delta\omega/\Omega')\,[1\pm\Delta\omega(\Omega^2+|z|^2)/\Omega'\,|z|^2]\,, \tag{3.18c}$$

$$N_\pm = \pm\tfrac{1}{2}(\bar{m}-\bar{n})\kappa\Omega^2(\Omega'\pm\Delta\omega)/\Omega'^2\,|z|^2\,. \tag{3.18d}$$

For the case of very intense pump fields exactly on resonance ($\Delta\omega=0$), we find that the value for $\mathcal{W}'(\nu)$ given by Eqs. (3.8) and (3.11a) is well approximated for general relaxation coefficients by the relation

$$\mathcal{W}'(\nu) = \frac{|\lambda'\mathcal{E}_0'|^2(\bar{m}-\bar{n})\Omega^2}{\kappa'}\times\left(\frac{(\Delta\nu)^4-\Omega^2(\Delta\nu)^2+2\kappa\kappa'^3}{|f(-i\Delta\nu)|^2}\right) \tag{3.19a}$$

$$\approx \frac{|\lambda'\mathcal{E}_0'|^2(\bar{m}-\bar{n})\Omega^2}{\kappa'}\times\left(\frac{[(\Delta\nu)^2-\Omega^2][(\Delta\nu)^2-2\kappa\kappa'^3/\Omega^2]}{|f(-i\Delta\nu)|^2}\right) \tag{3.19b}$$

for $\Delta\omega=0$ and $\Omega \gg \kappa,\ \kappa'$. The denominator in these relations, according to Eqs. (2.20), is given approximately by the relation

$$|f(-i\Delta\nu)|^2 = [(\Delta\nu)^2+\kappa'^2][(\Delta\nu-\Omega)^2+\tfrac{1}{4}(\kappa+\kappa')^2]\times[(\Delta\nu+\Omega)^2+\tfrac{1}{4}(\kappa+\kappa')^2]\,. \tag{3.20}$$

The function $\mathcal{W}'(\nu)$ is plotted for $\kappa'=\kappa$, $\Delta\omega=0$, and $\Omega=5\kappa$ in Fig. 1.

A noteworthy feature of the function $\mathcal{W}'(\nu)$ as given by Eq. (3.19b) is that it is negative for values of $\Delta\nu \equiv \nu-\omega$ satisfying the condition

$$2\kappa\kappa'^3/\Omega^2 < (\Delta\nu)^2 < \Omega^2\,. \tag{3.21}$$

This interval of signal-field frequencies thus corresponds to *stimulated emission* rather than to absorption, and the signal field is consequently amplified rather than attenuated by its interaction with the strongly driven atom.

As a means of discussing the question of energy balance in the process under consideration, let us evaluate the increase $\delta\mathcal{W}$ in the rate of absorption of quanta from the pump field and the increase $\kappa\delta\bar{n}$ in the rate of energy-dissipating transitions, in the limit described by Eqs. (3.19). We find from Eqs. (3.14), (3.12b), (3.11c), and (3.19a) that these quantities are well approximated in the limit in question by the relations

$$\delta\mathcal{W} = \frac{|\lambda'\mathcal{E}_0'|^2(\bar{m}-\bar{n})\Omega^2}{\kappa'}\left(\frac{-(\Delta\nu)^4+\Omega^2(\Delta\nu)^2-4\kappa\kappa'^3}{|f(-i\Delta\nu)|^2}\right) \tag{3.22}$$

and

$$\kappa\delta\bar{n} = \frac{|\lambda'\mathcal{E}_0'|^2(\bar{m}-\bar{n})\kappa}{\kappa'}\times\left(\frac{(2\kappa'-\kappa)(\Delta\nu)^4-3\Omega^2\kappa'(\Delta\nu)^2-2\Omega^2\kappa'^3}{|f(-i\Delta\nu)|^2}\right) \tag{3.23}$$

for $\Delta\omega=0$ and $\Omega \gg \kappa,\kappa'$.

Except for very small values of $\Delta\nu$, the quantity $\kappa\delta\bar{n}$ in the limit we are considering is small com-

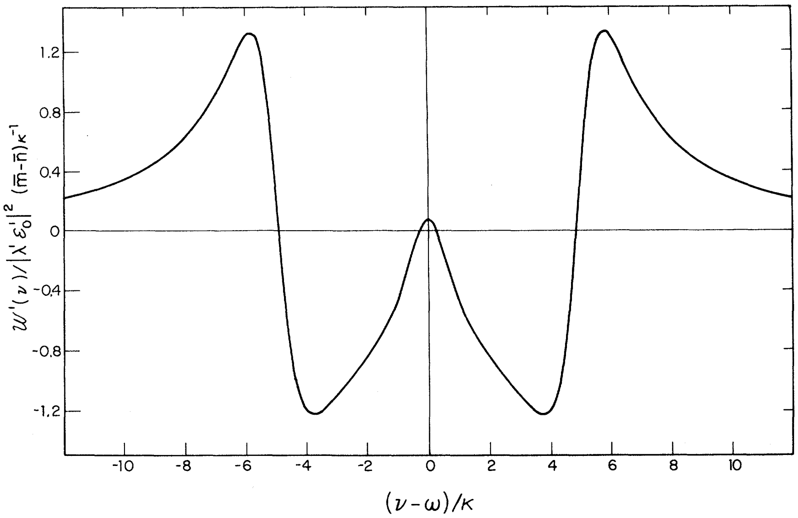


FIG. 1. Signal-field absorption line-shape function for an atom driven exactly on resonance by a pump field for which $\Omega = 5\kappa$. The negative values of the absorption function represent stimulated emission, i.e., amplification of the signal field.

pared either to $\mathcal{W}'$ or to $\delta\mathcal{W}$. The latter two quantities, as given by Eqs. (3.19a) and (3.22), thus obey, in accordance with the energy-balance equation (3.10), the approximate relation[8]

$$\mathcal{W}' \approx -\delta\mathcal{W} \tag{3.24}$$

throughout most of the frequency interval in question. It is clear from this relation that the amplification of the signal field in the frequency interval described by Eq. (3.21) occurs primarily as a result of quanta which are transferred to the signal field from the pump field. (For $|\Delta\nu| > \Omega$, it should be noted, the reverse process occurs: The signal field is attenuated, and the rate of attenuation of the pump field is correspondingly reduced.)

## IV. CORRELATION FUNCTIONS FOR ABSORPTION AND SPONTANEOUS EMISSION

Further discussion of the absorption of radiation by the driven system under consideration is greatly facilitated by formulating the problem of absorption in more general terms. The rate of absorption of energy from a weak perturbation may be expressed quite generally in terms of a certain atomic correlation function, which is evaluated in the absence of the perturbation. Let us introduce the operators

$$a \equiv |0\rangle\langle 1|, \quad a^\dagger \equiv |1\rangle\langle 0| \tag{4.1}$$

in terms of which the atomic matrix elements may be expressed as

$$\alpha = \langle a\rangle, \quad \alpha^* = \langle a^\dagger\rangle , \tag{4.2a}$$

$$\bar{n} = \langle a^\dagger a\rangle , \tag{4.2b}$$

$$\bar{m} = \langle a a^\dagger\rangle . \tag{4.2c}$$

The absorption line-shape function may be expressed in terms of the correlation function[9]

$$g_a(t) \equiv \langle [a(t), a^\dagger]\rangle \tag{4.3}$$

as

$$\mathcal{W}'(\nu) = |\lambda'\mathcal{E}_0'|^2 \int_{-\infty}^{\infty} dt\, e^{i\nu t} g_a(t) , \tag{4.4}$$

and hence satisfies the integral relation

$$\int \mathcal{W}'(\nu)\, d\nu = 2\pi\, |\lambda'\mathcal{E}_0'|^2 (\bar{m} - \bar{n}) . \tag{4.5}$$

The Heisenberg expectation value in Eq. (4.3) is evaluated in the presence of the pump field alone.

It is convenient to express the function $g_a(t)$ as

$$g_a(t) = g_d(t) - g_e(t) , \tag{4.6}$$

where

$$g_d(t) \equiv \langle a(t) a^\dagger\rangle , \tag{4.7}$$

$$g_e(t) \equiv \langle a^\dagger a(t)\rangle . \tag{4.8}$$

The function $g_e(t)$, the Fourier transform of which is proportional to the emission spectrum for the pumped atom for frequencies near $\omega_{10}$, has been evaluated separately for the cases of radiative and collisional relaxation in Refs. 1 and 2, respectively. The method, which is based on the Markoff approximation,[10] leads directly to the relation

$$g_e(t) = \bar{n}\,\mathfrak{U}_{\alpha\alpha}(t) + \bar{\alpha}^*\mathfrak{U}_{\alpha m}(t) , \tag{4.9}$$

where the functions $\mathfrak{U}(t)$ are the same functions that appear in Eqs. (2.14).

The function $g_d(t)$ defined by Eq. (4.7) is given in the Markoff approximation by the expression

$$g_d(t) = \mathfrak{U}_{\alpha\alpha}(t)\langle a a^\dagger\rangle + \mathfrak{U}_{\alpha\alpha^*}(t)\langle a^\dagger a^\dagger\rangle + \mathfrak{U}_{\alpha n}(t)\langle a^\dagger a a^\dagger\rangle + \mathfrak{U}_{\alpha m}(t)\langle a a^\dagger a^\dagger\rangle = \bar{m}\mathfrak{U}_{\alpha\alpha}(t) + \bar{\alpha}^*\mathfrak{U}_{\alpha n}(t) , \tag{4.10}$$

where the latter relation follows from Eqs. (4.2) and the identities $a^\dagger a^\dagger = 0$ and $a^\dagger a a^\dagger = a^\dagger$.

By substituting Eqs. (4.9) and (4.10) into Eq.

(4.6) we find that the atomic correlation function which determines the absorption spectrum is given by the relation

$$g_a(t)=(\overline{m}-\overline{n})\,\mathfrak{U}_{\alpha\alpha}(t)+\overline{\alpha}^*[\,\mathfrak{U}_{\alpha n}(t)-\mathfrak{U}_{\alpha m}(t)\,]\ . \qquad (4.11)$$

We may note that the integral relation (4.5) follows directly from this relation and the identities $\mathfrak{U}_{\alpha\alpha}(t=0)=1$ and $\mathfrak{U}_{\alpha n}(t=0)=\mathfrak{U}_{\alpha m}(t=0)=0$, which may be obtained by evaluating Eq. (2.14) at $t=0$.

The absorption line-shape function as given by Eq. (4.4) may be expressed as

$$\mathcal{W}'(\nu)=2\,|\,\lambda'\mathcal{E}_0'\,|^2\,\mathrm{Re}[\,\hat{g}_a(-i\nu)\,]\ , \qquad (4.12)$$

where $\hat{g}_a(s)$, the Laplace-transform function of $g_a(t)$ [defined by a relation similar to Eq. (2.15)] is given, according to Eq. (4.11), by the relation

$$\hat{g}_a(s)=(\overline{m}-\overline{n})\,\hat{\mathfrak{U}}_{\alpha\alpha}(s)+\overline{\alpha}^*[\,\hat{\mathfrak{U}}_{\alpha n}(s)-\hat{\mathfrak{U}}_{\alpha m}(s)]\ . \qquad (4.13)$$

By substituting Eqs. (2.16) into this relation and then making use of the resulting expression in Eq. (4.12), we find, with the aid of Eq. (2.11a), exactly the value for $\mathcal{W}'(\nu)$ which is given by Eqs. (3.8) and (3.11a).

The absorption spectrum we have found is represented by a function quite different in form from the function which represents the emission spectrum evaluated in Refs. 1 and 2. This difference is most remarkable in the limit of intense pump fields, where the absorption line-shape function takes on appreciable negative values within a wide range of signal-field frequencies. The negative values are the result of the contribution of the term proportional to $\overline{\alpha}^*$ in Eq. (4.13), which, although it makes no net contribution to the integral in Eq. (4.5), importantly modifies the shape of the absorption spectrum.

It is worth noting, finally, that the general correlation-function approach described in this section enables us to see that the range of validity of our analysis extends well beyond the simple model we have discussed. Inasmuch as the signal field plays no direct role in our more general treatment, our results should apply whenever the two levels in question are driven by a suitable pumping mechanism. In the case in which the pump field induces transitions by means of two-photon processes, for example, it has been shown[11] that the atomic density matrix obeys equations analogous to Eqs. (2.9). It follows therefore that in this case the absorption line-shape function (evaluated for the case of one-photon absorption, i.e., for $\nu\approx\omega_{10}$) is similarly related to the function we have found.

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[1]B. R. Mollow, Phys. Rev. 188, 1969 (1969).

[2]B. R. Mollow, Phys. Rev. A 2, 76 (1970).

[3]See also M. Newstein, Phys. Rev. 167, 89 (1968).

[3a]This problem has been treated by the author [B. R. Mollow, Phys. Rev. A 5, 1522 (1972)] for the case in which the signal field oscillates at a frequency well separated from the pump frequency, thus inducing resonant transitions between pairs of states of which only one is a member of the pair resonantly coupled by the pump field.

[4]The signal and pump fields may be easily distinguished in an experimental situation by allowing them to propagate in different directions or to have different polarizations.

[5]An experiment along these lines has been carried out on spin systems by Bucci and Santucci [P. Bucci and S. Santucci, Phys. Rev. A 2, 1105 (1970)]. Both fields, however, are allowed to be intense in their experiment.

[6]R. Karplus and J. Schwinger, Phys. Rev. 73, 1020 (1948).

[7]It is important that the trace relation $\overline{n}(t)+\overline{m}(t)=1$ *not* be used in solving Eqs. (2.9) for the functions $\mathfrak{U}(t)$.

[8]The approximations in Eqs. (3.19), (3.22), and (3.23) are not sufficiently accurate to satisfy the energy-balance relation (3.10) near the zeroes of the functions in question.

[9]S. Bloom and H. Margenau, Phys. Rev. 90, 791 (1953). A more satisfactory derivation of this relation follows directly from methods developed by Kubo [R. Kubo, J. Phys. Soc. Japan 12, 570 (1957)]. See, for example, Ref. 3a.

[10]See M. Lax, Phys. Rev. 172, 350 (1968) and related references.

[11]B. R. Mollow, Phys. Rev. A 4, 1666 (1971).