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Absorption and Emission Line-Shape Functions for Driven Atoms

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The effect of a driving or pump field on the emission and absorption line-shape functions for an atom is discussed. The pump field is assumed to oscillate at a frequency near the resonance frequency for transitions between a single pair of atomic states, and transitions between one of these states and any other state of the atom are analyzed. The pump field is treated classically, and atomic relaxation is treated in general terms. The emission and absorption line-shape function (the latter is defined as the rate of absorption of energy from a weak signal field, applied in addition to the pump field, as a function of the signal-field frequency) are found by evaluating the relevant two-time atomic correlation functions in the Markoff approximation. In the limit of high pump-field intensity, both the absorption and the emission spectra are doubly peaked at frequencies which differ from the usual resonance frequency by $\pm\frac{1}{2}\Omega$, where Ω is the frequency of the pump-field-induced oscillations in the populations of the two strongly coupled states. The absorption and the emission spectra are represented by essentially the same function in the limit of high pump-field intensity, as they are also in the limit of vanishing pump-field intensity. For intermediate pump-field intensities, however, the two functions are quite different in form, and no simple proportionality exists between them.

I. INTRODUCTION

When a strong external field is applied to an atom at a frequency near resonance for atomic transitions between a particular pair of states, its initial effect is to induce harmonic variations both in the populations for the states in question and in the absolute value of the associated off-diagonal matrix elements.[1-3] The frequency Ω of these variations is equal, in the limit of very intense driving fields, to the product of the field amplitude and the dipole matrix element connecting the states in question. After a period of time long compared to the atomic relaxation time, the oscillations in question damp out, and the atomic density matrix approaches a fixed equilibrium value.[4] In a very real physical sense, however, the oscillations continue to influence the dynamics of the system even after equilibrium is reached. One way of understanding this is to picture the atom in equilibrium as described by an ensemble of states, in each one of which the oscillations in the atomic populations continue, at the same frequency but with different phases. Mathematically, the effect of the field-induced modulation of the system appears in the solutions for the two-time correlation functions which describe its interaction with other systems. The correlation function which describes the emission of photons during transitions between the pair of states in question, for example, has been found,[5,6] and leads, in the limit of very strong driving fields, to a spectrum with peaks not only at the driving frequency but at frequencies displaced from the driving frequency by $\pm\frac{1}{2}\Omega$.[7]

In this paper we investigate the effect of the driving or "pump" field on transitions between pairs of states only one of which is a member of the original resonantly coupled pair.[7a] The pump field is

treated classically, and its intensity is allowed to assume arbitrary values. Atomic relaxation is treated in general terms by introducing phenomenological relaxation coefficients. The atomic correlation functions which determine the emission and the absorption line-shape functions (the latter is defined as the rate at which energy is absorbed from a weak signal field, applied in addition to the pump field, as a function of the signal-field frequency) are found by a method based on the Markoff approximation.[8]

In the limit of very intense pump fields, both the emission and the absorption line-shape functions are doubly peaked,[9,10] at frequencies displaced by $\pm \frac{1}{2}\Omega$ from the unperturbed resonance frequency. The two functions are essentially identical in this limit, as they are also in the limit of vanishing pump intensity. The two functions are nevertheless, in general, quite different from one another, exhibiting no simple proportionality for pump fields of intermediate strength. The difference between the emission and absorption spectra, which emerges directly from the basic method of evaluating the associated correlation functions in the Markoff approximation, is due to the presence of the off-diagonal density matrix elements which refer to the pair of states which are strongly coupled by the pump field.

The effect of the pump field on the equilibrium density matrix for an atom with general relaxation coefficients is discussed in Sec. II. The emission and absorption line-shape functions are then evaluated in Secs. III and IV, respectively, and the limiting cases of interest are discussed.

II. DRIVEN ATOM WITH GENERAL RELAXATION COEFFICIENTS

Let us consider an atom with energy eigenstates $|j\rangle$ and corresponding eigenvalues E_j, where $j = 0, 1, 2, \ldots$. We assume that the atom is driven by an external electric field, with positive- and negative-frequency parts $\mathcal{E}(t)$ and $\mathcal{E}^*(t)$, respectively, and polarization specified by the unit vector $\hat{e}_0$,

$$\vec{E}(t) = (1/\sqrt{2})\hat{e}_0[\mathcal{E}(t) + \mathcal{E}^*(t)] . \tag{2.1}$$

We shall describe the atomic relaxation process in general terms by introducing separate off-diagonal decay rates $\kappa'_{jk} = \kappa'_{kj}$ and diagonal decay rates κ_{jk} for transitions from the state $|k\rangle$ to the state $|j\rangle$. We may note that the strong-collision model[4] of atomic relaxation can be described in these terms by setting

$$\kappa'_{jk} = \kappa , \tag{2.2a}$$

$$\kappa_{jk} = \kappa \bar{n}_j^{(0)}(1 - \delta_{jk}) , \tag{2.2b}$$

where κ is the collision frequency and $\bar{n}_j^{(0)}$ is the mean thermal occupation number for the state $|j\rangle$. In the case of radiative relaxation,[11] on the other hand, the parameters κ_{jk} are simply the spontaneous-emission rates, and $\kappa'_{jk} = \frac{1}{2}(\kappa_j + \kappa_k)$, where

$$\kappa_j \equiv \sum_k \kappa_{kj} . \tag{2.3}$$

The equations of motion for the diagonal matrix elements $\bar{n}_j(t)$ and the off-diagonal matrix elements $\alpha_{jk}(t)$ of the atomic density operator are, in the dipole approximation,

$$\left(\frac{d}{dt} + \kappa_j\right)\bar{n}_j(t) - \sum_k \kappa_{jk}\bar{n}_k(t) = i[\mathcal{E}(t) + \mathcal{E}^*(t)]\sum_k [\lambda_{jk}\alpha_{kj}(t) - \alpha_{jk}(t)\lambda_{kj}] \tag{2.4a}$$

and

$$\left(\frac{d}{dt} + \kappa'_{jk} + i\omega_{jk}\right)\alpha_{jk}(t) = i[\mathcal{E}(t) + \mathcal{E}^*(t)]\{\lambda_{jk}[\bar{n}_k(t) - \bar{n}_j(t)] + \sum_{m,(m\neq j,k)} [\lambda_{jm}\alpha_{mk}(t) - \alpha_{jm}(t)\lambda_{mk}]\} , \tag{2.4b}$$

where κ_j is defined by Eq. (2.3); the parameter λ_{jk} is defined in terms of the dipole matrix element $\vec{\mu}_{jk}$ as

$$\lambda_{jk} \equiv (\vec{\mu}_{jk}\cdot\hat{e}_0)/\hbar\sqrt{2} , \tag{2.5}$$

and the parameter ω_{jk} is defined as

$$\omega_{jk} \equiv (E_j - E_k)/\hbar . \tag{2.6}$$

Let us now assume that the incident field oscillates harmonically,

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

at a frequency which very nearly coincides with the resonant frequency for transitions between the states $|0\rangle$ and $|1\rangle$,

$$\omega \simeq \omega_{10} , \tag{2.8}$$

but not with the resonant frequency for transitions between any other pair of states. The equilibrium solution for the atomic density matrix is then characterized, in the resonant approximation, by constant diagonal elements $\bar{n}_j$ and by off-diagonal elements [all of which vanish except $\alpha_{10}(t)$ and $\alpha_{01}(t)$] which have the form

$$\alpha_{10}(t) = \alpha^*_{01}(t) = \bar{\alpha}_{10}e^{-i\omega t} , \tag{2.9}$$

where $\bar{\alpha}_{10}$ is a constant. The quantities $\bar{n}_j$ and $\bar{\alpha}_{10}$ are the solutions to the equations

$$\kappa_j\bar{n}_j - \sum_k \kappa_{jk}\bar{n}_k = (\delta_{j1} - \delta_{j0})[-i\lambda^*_{10}\mathcal{E}^*_0\bar{\alpha}_{10} + i\lambda_{10}\mathcal{E}_0\bar{\alpha}^*_{10}] , \tag{2.10a}$$

$$z^*\bar{\alpha}_{10} = i\lambda_{10}\mathcal{E}_0(\bar{n}_0 - \bar{n}_1) , \tag{2.10b}$$

$$z\bar{\alpha}^*_{10} = -i\lambda^*_{10}\mathcal{E}^*_0(\bar{n}_0 - \bar{n}_1) , \tag{2.10c}$$

where

$$z \equiv \kappa'_{10} + i\Delta\omega , \tag{2.11a}$$

$$\Delta\omega \equiv \omega - \omega_{10} . \tag{2.11b}$$

The rate $\mathcal{W}$ at which quanta are absorbed from

the driving field may be found by calculating the rate at which the field does work and then dividing by the energy $\hbar\omega$ of a field quantum. We find directly from Eqs. (2.10)

$$\mathcal{W} = -i\lambda_{10}^{*}\mathcal{E}_0^{*}\bar{\alpha}_{10} + i\lambda_{10}\mathcal{E}_0\bar{\alpha}_{10}^{*} \tag{2.12a}$$

$$= \tfrac{1}{2}\Omega^2\kappa'_{10}(\bar{n}_0 - \bar{n}_1)/|z|^2 , \tag{2.12b}$$

where

$$\Omega \equiv 2|\lambda_{10}\mathcal{E}_0| . \tag{2.13}$$

We note the identity

$$-i\lambda_{10}^{*}\mathcal{E}_0^{*}\bar{\alpha}_{10} = \tfrac{1}{2}\mathcal{W}z/\kappa'_{10} , \tag{2.14}$$

which follows directly from Eqs. (2.10b), (2.12b), and (2.13).

In the case of the strong-collision model [Eqs. (2.2)], the equations (2.10) lead directly to the relations[4]

$$\bar{n}_0 + \bar{n}_1 = \bar{n}_0^{(0)} + \bar{n}_1^{(0)} , \tag{2.15a}$$

$$\bar{n}_0 - \bar{n}_1 = (\bar{n}_0^{(0)} - \bar{n}_1^{(0)})|z|^2/(\Omega^2 + |z|^2) , \tag{2.15b}$$

$$\bar{\alpha}_{10} = (\bar{n}_0^{(0)} - \bar{n}_1^{(0)})\, i\lambda_{10}\mathcal{E}_0 z/(\Omega^2 + |z|^2) , \tag{2.15c}$$

and

$$\bar{n}_j = \bar{n}_j^{(0)} \quad \text{for } j \geq 2. \tag{2.15d}$$

The quantity $\mathcal{W}$ in this case is given by the relation

$$\mathcal{W} = (\bar{n}_0^{(0)} - \bar{n}_1^{(0)})\tfrac{1}{2}\kappa\Omega^2/(\Omega^2 + |z|^2) . \tag{2.16}$$

III. EMISSION SPECTRUM

Our assumption that the driving field induces resonant transitions only between the states $|0\rangle$ and $|1\rangle$ has led directly to the conclusion that only the elements of the associated 2×2 submatrix of the full atomic density matrix are directly affected *in equilibrium* by the driving field.[12] It is important to realize, however, that in the equations (2.4) governing the time evolution of the full density matrix, resonant couplings exist between other matrix elements as well. In particular, the off-diagonal matrix elements $\alpha_{j1}(t)$ and $\alpha_{j0}(t)$ are coupled for all $j \geq 2$. These functions obey the equations

$$\left(\frac{d}{dt} + \kappa'_{j1} + i\omega_{j1}\right)\alpha_{j1}(t) = -i\lambda_{10}^{*}\mathcal{E}^{*}(t)\alpha_{j0}(t) , \tag{3.1a}$$

$$\left(\frac{d}{dt} + \kappa'_{j0} + i\omega_{j0}\right)\alpha_{j0}(t) = -i\lambda_{10}\mathcal{E}(t)\alpha_{j1}(t) . \tag{3.1b}$$

It is not difficult to show with the aid of these relations that the driving field affects the emission spectrum for transitions between any state $|j\rangle$ (for $j \geq 2$) and either of the states $|0\rangle$ or $|1\rangle$. We shall consider directly only the case of transitions between the states $|j\rangle$ and $|1\rangle$, and we shall assume $E_j > E_1 > E_0$; other energy orderings may be treated in a straightforward manner.

We begin by introducing (Schrödinger) operators a_{jk} and $a_{jk}^{\dagger}$, for $j \neq k$, by means of the definitions

$$a_{jk} \equiv |k\rangle\langle j| , \quad a_{jk}^{\dagger} \equiv |j\rangle\langle k| . \tag{3.2}$$

The elements of the density matrix may be expressed in terms of these operators as

$$\alpha_{jk} = \langle a_{jk}\rangle , \tag{3.3a}$$

$$\bar{n}_j = \langle a_{jk}^{\dagger}a_{jk}\rangle , \tag{3.3b}$$

the latter relation holding for all $k \neq j$. The spectral density of the radiation emitted at the (angular) frequency ν during atomic transitions between the states $|j\rangle$ and $|1\rangle$ can be shown[5] to be proportional to the function

$$\tilde{g}_e(\nu) = \int_{-\infty}^{\infty} dt\, e^{i\nu t} g_e(t) , \tag{3.4}$$

where the atomic correlation function $g_e(t)$ is defined in terms of the Heisenberg operators $a_{j1}^{\dagger}(t)$ and $a_{j1}(t)$ as

$$g_e(t) \equiv \langle a_{j1}^{\dagger}(t')a_{j1}(t'+t)\rangle = \langle a_{j1}^{\dagger}a_{j1}(t)\rangle , \tag{3.5}$$

the latter relation following from the stationarity of the process under consideration and the relation $a_{j1}^{\dagger}(0) = a_{j1}^{\dagger}$. We may note that the total intensity of the emitted radiation is proportional to

$$(2\pi)^{-1}\int d\nu\, \tilde{g}_e(\nu) = g_e(0) = \langle a_{j1}^{\dagger}a_{j1}\rangle = \bar{n}_j , \tag{3.6}$$

and hence is not directly affected by the driving field.[12]

The atomic correlation function $g_e(t)$ may be directly evaluated in the Markoff approximation.[5] The method, due to Lax,[8] consists essentially of substituting for the operator $a_{j1}(t)$ in Eq. (3.5) an expression formally analogous to the solution to Eqs. (3.1) for the matrix element $\alpha_{j1}(t) = \langle a_{j1}(t)\rangle$ (where $t > 0$) in terms of the initial functions $\alpha_{j1}(0)$ and $\alpha_{j0}(0)$. The solution in question takes the form

$$\alpha_{j1}(t) = \mathfrak{U}_{j1;j1}(t)\,\alpha_{j1}(0) + \mathfrak{U}_{j1;j0}(t)\,\alpha_{j0}(0) . \tag{3.7}$$

The function $g_e(t)$ for $t > 0$ may then be expressed in the Markoff approximation as

$$g_e(t) = \mathfrak{U}_{j1;j1}(t)\langle a_{j1}^{\dagger}a_{j1}\rangle + \mathfrak{U}_{j1;j0}(t)\langle a_{j1}^{\dagger}a_{j0}\rangle = \bar{n}_j\mathfrak{U}_{j1;j1}(t) , \tag{3.8}$$

where the latter relation follows from Eq. (3.3b) and the identity $a_{j1}^{\dagger}a_{j0} = 0$.

The functions $\mathfrak{U}(t)$ in Eq. (3.7) can be found directly by solving the linear coupled Eqs. (3.1) [with $\mathcal{E}(t)$ given by Eq. (2.7)] for $\alpha_{j1}(t)$ and making the appropriate identification of coefficients in the solution. We find that the Laplace transform functions

$$\hat{\mathfrak{U}}(s) \equiv \int_0^{\infty} dt\, e^{-st}\mathfrak{U}(t) \tag{3.9}$$

are given by the relations

$$\hat{\mathfrak{U}}_{j1;j1}(s) = [s + i(\omega_{j1} - \Delta\omega) + \kappa'_{j0}]/f(s) , \tag{3.10a}$$

$$\hat{\mathfrak{U}}_{j1;j0}(s) = -i\lambda_{10}^{*}\mathcal{E}_0^{*}/f(s) , \tag{3.10b}$$

where the function $f(s)$ is the second-degree polynomial

$$f(s) \equiv (s + i\omega_{j1} + \kappa'_{j1})(s + i\omega_{j1} - i\Delta\omega + \kappa'_{j0}) + \tfrac{1}{4}\Omega^2 . \tag{3.11}$$

We note that the two roots s_+ and s_- of $f(s)$ are given in the case of collisional relaxation by the relation

$$s_\pm = -\kappa - i(\omega_{j1} - \tfrac{1}{2}\Delta\omega \mp \tfrac{1}{2}\Omega') \quad \text{for} \quad \kappa'_{jk} = \kappa , \tag{3.12}$$

where Ω' is the Rabi[1] frequency of population inversion (for the levels $|0\rangle$ and $|1\rangle$) in the absence of damping,

$$\Omega' \equiv [\Omega^2 + (\Delta\omega)^2]^{1/2} . \tag{3.13}$$

In the case of general relaxation coefficients, in the limit in which Ω' is much greater than κ'_{jk}, the roots of $f(s)$ are well approximated by the relation

$$s_\pm = -\kappa'_\pm - i(\omega_{j1} - \tfrac{1}{2}\Delta\omega \mp \tfrac{1}{2}\Omega') , \tag{3.14a}$$

where

$$\kappa'_\pm = \tfrac{1}{2}(\kappa'_{j1} + \kappa'_{j0}) \mp (\kappa'_{j1} - \kappa'_{j0})\,\Delta\omega/\Omega' . \tag{3.14b}$$

It follows directly from Eqs. (3.8) and (3.10a) that the Laplace transform of $g_e(t)$,

$$\hat{g}_e(s) \equiv \int_0^\infty dt\, e^{-st} g_e(t) , \tag{3.15}$$

is given by

$$\hat{g}_e(s) = \bar{n}_j [s + i(\omega_{j1} - \Delta\omega) + \kappa'_{j0}]/f(s) . \tag{3.16}$$

The spectral density $\tilde{g}_e(\nu)$ defined by Eq. (3.4) may be expressed, with the aid of the Hermiticity relation $g_e(-t) = g_e^{*}(t)$, in terms of the Laplace transform function $\hat{g}_e(s)$ defined by Eq. (3.15), as

$$\tilde{g}_e(\nu) = 2\,\mathrm{Re}[\hat{g}_e(-i\nu)] . \tag{3.17}$$

By making use of Eq. (3.16) in (3.17), we find that

$$\tilde{g}_e(\nu) = 2\bar{n}_j \left[\frac{(\Delta\nu + \Delta\omega)^2 \kappa'_{j1} + (\tfrac{1}{4}\Omega^2 + \kappa'_{j1}\kappa'_{j0})\kappa'_{j0}}{|f(-i\nu)|^2}\right] , \tag{3.18a}$$

where

$$\Delta\nu \equiv \nu - \omega_{j1} . \tag{3.18b}$$

The denominator in Eq. (3.18a) is

$$|f(-i\nu)|^2 = [\Delta\nu(\Delta\nu + \Delta\omega) - \tfrac{1}{4}\Omega^2 - \kappa'_{j1}\kappa'_{j0}]^2 + [\Delta\nu(\kappa'_{j1} + \kappa'_{j0}) + \Delta\omega\kappa'_{j1}]^2 . \tag{3.18c}$$

The function $\tilde{g}_e(\nu)$, which is proportional to the power spectrum of the field radiated during atomic transitions between the states $|j\rangle$ and $|1\rangle$, is thus given by Eqs. (3.18) in terms of the relaxation constants κ'_{j1} and κ'_{j0} and the parameters $\Delta\omega$ and Ω defined by Eqs. (2.11b) and (2.13), respectively.

The limiting cases of interest may be treated in a straightforward manner. In the case in which the driving field vanishes identically, the spectral density is the familiar spontaneous emission field

$$\tilde{g}_e(\nu) = \frac{2\bar{n}_j \kappa'_{j1}}{(\Delta\nu)^2 + \kappa'^2_{j1}} \quad \text{for} \quad \Omega = 0 . \tag{3.19}$$

For $\Omega' \gg \kappa'_{jk}$, the function $\tilde{g}_e(\nu)$ is sharply peaked at the two displaced frequencies

$$\nu = \omega_{j1} - \tfrac{1}{2}\Delta\omega - \tfrac{1}{2}\Omega' \quad \text{and} \quad \nu = \omega_{j1} - \tfrac{1}{2}\Delta\omega + \tfrac{1}{2}\Omega' .$$

The function is well approximated in the domain in which it is appreciable in this limit by the relation

$$\tilde{g}_e(\nu) = \frac{\tfrac{1}{2}\bar{n}_j}{\Omega'^2}\left[\frac{(\Delta\omega - \Omega')^2\kappa'_{j1} + \Omega^2\kappa'_{j0}}{(\Delta\nu + \tfrac{1}{2}\Delta\omega + \tfrac{1}{2}\Omega')^2 + \kappa'^2_+} + \frac{(\Delta\omega + \Omega')^2\kappa'_{j1} + \Omega^2\kappa'_{j0}}{(\Delta\nu + \tfrac{1}{2}\Delta\omega - \tfrac{1}{2}\Omega')^2 + \kappa'^2_-}\right] \quad \text{for} \quad \Omega' \gg \kappa'_{jk} , \tag{3.20}$$

where κ'_+ and κ'_- are defined by Eq. (3.14b).

In the limit of very intense driving fields, the peaks in the spectral density occur at $\nu = \omega_{j1} \pm \tfrac{1}{2}\Omega$, and the function takes the limiting form

$$\tilde{g}_e(\nu) = \bar{n}_j \kappa' \left[\frac{1}{(\Delta\nu + \tfrac{1}{2}\Omega)^2 + \kappa'^2} + \frac{1}{(\Delta\nu - \tfrac{1}{2}\Omega)^2 + \kappa'^2}\right] \quad \text{for} \quad \Omega \gg \kappa'_{jk}, \ |\Delta\omega| , \tag{3.21}$$

where

$$\kappa' \equiv \tfrac{1}{2}(\kappa'_{j1} + \kappa'_{j0}) . \tag{3.22}$$

IV. ABSORPTION SPECTRUM

The analysis of Sec. III of the effect of the driving field on atomic transitions between the state $|j\rangle$ and the driven state $|1\rangle$ was concerned with the emission spectrum for those transitions. We may extend our analysis so as to treat the absorption spectrum for transitions between the same pair of states by supposing that in addition to the pump field (2.7), a weak signal field is applied

$$\vec{\mathbf{E}}'(t) = (1/\sqrt{2})\hat{e}'_0[\mathcal{E}'(t) + \mathcal{E}'^{*}(t)] , \tag{4.1a}$$

$$\mathcal{E}'(t) = \mathcal{E}'_0 e^{-i\nu t} , \tag{4.1b}$$

oscillating at a frequency satisfying the resonant condition

$$\nu \approx \omega_{j1} \tag{4.2}$$

for a particular value of $j \geq 2$.

To determine the rate of absorption of energy from the perturbing or signal field, we must first find the change it produces in the equilibrium density operator. It is not difficult to show that to lowest order in the signal field strength this change may be expressed in terms of the interaction Hamiltonian $H'(t)$ associated with the perturbation and the unperturbed equilibrium density operator ρ as

$$\Delta\rho(t) = \frac{1}{i\hbar}\int_{-\infty}^{t} dt' \,[H'(t'), \rho] . \tag{4.3}$$

The operators in this relation are evaluated in the interaction picture, where the (time-dependent) unperturbed Hamiltonian must be understood to include the effect of the pump field. The operator ρ is hence a time-independent quantity, and corresponds to the Heisenberg density operator for the system in the absence of the perturbation.

It follows directly from Eq. (4.3) that the rate at which the perturbation does work on the system, since it does no work in the unperturbed state of the system, is[13]

$$W'(t)=\mathrm{tr}\left[\frac{\partial H'(t)}{\partial t}\,\Delta\rho(t)\right]$$

$$=\frac{1}{i\hbar}\int_{-\infty}^{t}dt'\,\mathrm{tr}\left\{\rho\left[\frac{\partial H'(t)}{\partial t}\,,\,H'(t')\right]\right\}. \quad (4.4)$$

In the resonant approximation the Hamiltonian $H'(t)$ corresponding to the signal field (4.1) and its partial time derivative are given, according to Eq. (4.2), by the relations

$$H'(t)=-\hbar\lambda'^{*}_{j1}\mathcal{E}'^{*}(t)a_{j1}(t)-\hbar\lambda'_{j1}\mathcal{E}'(t)a^{\dagger}_{j1}(t)\,, \quad (4.5a)$$

$$\frac{\partial H'(t)}{\partial t}=-i\hbar\nu\left[\lambda'^{*}_{j1}\mathcal{E}'^{*}(t)a_{j1}(t)-\lambda'_{j1}\mathcal{E}'(t)a^{\dagger}_{j1}(t)\right]. \quad (4.5b)$$

When these relations are used in Eq. (4.4) and the stationarity of the unperturbed system is taken into account, it is found that in the resonant approximation, the rate at which the perturbing or signal field does work is

$$W'=\hbar\nu\left|\lambda'_{j1}\mathcal{E}'_{0}\right|^{2}\tilde{g}_{a}(\nu)\,,$$

where

$$\tilde{g}_{a}(\nu)=\int_{-\infty}^{\infty}dt\,e^{i\nu t}\langle[a_{j1}(t),\,a^{\dagger}_{j1}]\rangle\,. \quad (4.6)$$

The time-dependent expectation value in this relation is evaluated in the presence of the pump field alone.

It follows immediately from Eqs. (4.6), (3.2), and (3.3b) that the integral of the absorption line-shape function $\tilde{g}_a(\nu)$ over all frequencies is

$$(2\pi)^{-1}\int d\nu\,\tilde{g}_{a}(\nu)=\bar{n}_1-\bar{n}_j\,, \quad (4.7)$$

and hence, as in the case of the emission spectrum, is not directly affected by the pump field.[12]

The absorption line-shape function as given by Eq. (4.6) is the Fourier transform of the atomic correlation function

$$g_{a}(t)=\langle[a_{j1}(t),\,a^{\dagger}_{j1}]\rangle \quad (4.8)$$

$$=g_{d}(t)-g_{e}(t)\,, \quad (4.9)$$

where

$$g_{d}(t)=\langle a_{j1}(t)a^{\dagger}_{j1}\rangle\,, \quad (4.10)$$

and $g_e(t)$ is the function defined by Eq. (3.5). The functions $g_d(t)$ and $g_e(t)$ in Eq. (4.9) may be thought of as representing direct absorption and stimulated emission, respectively, the difference between them representing the net rate of absorption.

The function $g_d(t)$ for $t>0$ may be evaluated in the Markoff approximation by methods analogous to those used in Sec. III to evaluate $g_e(t)$. We find that

$$g_{d}(t)=\mathfrak{U}_{j1;j1}(t)\langle a_{j1}a^{\dagger}_{j1}\rangle+\mathfrak{U}_{j1;j0}(t)\langle a_{j0}a^{\dagger}_{j1}\rangle$$

$$=\bar{n}_1\mathfrak{U}_{j1;j1}(t)+\bar{\alpha}_{10}\mathfrak{U}_{j1;j0}(t)\,, \quad (4.11)$$

where the latter relation follows from Eqs. (3.2) and (3.3) and the identity $a_{j0}a^{\dagger}_{j1}=a_{10}$.

By substituting Eqs. (4.11) and (3.8) into Eq. (4.9), we find that

$$g_{a}(t)=(\bar{n}_1-\bar{n}_j)\mathfrak{U}_{j1;j1}(t)+\bar{\alpha}_{10}\mathfrak{U}_{j1;j0}(t)\,. \quad (4.12)$$

The Laplace transform of g_a is thus

$$\hat{g}_{a}(s)=(\bar{n}_1-\bar{n}_j)\hat{\mathfrak{U}}_{j1;j1}(s)+\bar{\alpha}_{10}\hat{\mathfrak{U}}_{j1;j0}(s)$$

$$=\frac{(\bar{n}_1-\bar{n}_j)[s+i(\omega_{j1}-\Delta\omega)+\kappa'_{j0}]-i\lambda^{*}_{10}\mathcal{E}^{*}_{0}\bar{\alpha}_{10}}{f(s)}\,, \quad (4.13)$$

the latter relation following from Eqs. (3.10).

We find directly from Eqs. (4.13), (3.11), and (2.14) that the function $\tilde{g}_a(\nu)=2\,\mathrm{Re}\hat{g}_a(-i\nu)$ is given by the relation

$$\tilde{g}_{a}(\nu)=[1/|f(-i\nu)|^{2}]\{[2(\bar{n}_1-\bar{n}_j)\kappa'_{j1}-\mathcal{W}](\Delta\nu+\Delta\omega)^{2}$$

$$+[2(\bar{n}_1-\bar{n}_j)\kappa'_{j0}+\mathcal{W}](\tfrac{1}{4}\Omega^{2}+\kappa'_{j1}\kappa'_{j0})$$

$$+\mathcal{W}\Delta\omega[(\Delta\nu+\Delta\omega)(\kappa'_{10}-\kappa'_{j1})-\Delta\nu\kappa'_{j0}]/\kappa'_{10}\}\,, \quad (4.14)$$

where $\Delta\nu=\nu-\omega_{j1}$. The absorption line-shape function for the signal field is thus given, quite generally, in terms of the equilibrium occupation numbers, the rate $\mathcal{W}$ of absorption of quanta from the pump field (both evaluated in the absence of the signal field), and the parameters defined by Eqs. (2.13) and (2.11b). [The function $|f(-i\nu)|^2$ in Eq. (4.14) is given by Eq. (3.18c).]

In the case of collisional relaxation, we find with the aid of Eqs. (2.15), (2.16), and (3.12) that $\tilde{g}_a(\nu)$ is given by the expression

$$\tilde{g}_{a}(\nu)=\left(\frac{1}{[(\Delta\nu+\frac{1}{2}\Delta\omega+\frac{1}{2}\Omega')^{2}+\kappa^{2}][(\Delta\nu+\frac{1}{2}\Delta\omega-\frac{1}{2}\Omega')^{2}+\kappa^{2}]}\right)$$

$$\times\{2(\bar{n}_1^{(0)}-\bar{n}_j^{(0)})\kappa[(\Delta\nu+\Delta\omega)^{2}+\tfrac{1}{4}\Omega^{2}+\kappa^{2}]+\mathcal{W}[(\Delta\nu+\tfrac{1}{2}\Delta\omega)^{2}+\tfrac{3}{4}\Omega'^{2}+3\kappa^{2}]\}\,, \quad (4.15)$$

which is manifestly positive for $\bar{n}_0^{(0)} > \bar{n}_1^{(1)} > \bar{n}_j^{(0)}$.

In the absence of the pump field ($\Omega = \mathcal{W} = 0$), the function $\tilde{g}_a(\nu)$ for the case of general relaxation coefficients takes the simple form

$$\tilde{g}_a(\nu) = \frac{2(\bar{n}_1 - \bar{n}_j)\kappa'_{j1}}{(\Delta\nu)^2 + \kappa'^2_{j1}} \quad \text{for } \Omega = 0. \tag{4.16}$$

In the limit of very intense pump fields,[14] the function is well approximated by the expression

$$\tilde{g}_a(\nu) = (\bar{n}_1 - \bar{n}_j)\kappa' \left(\frac{1}{(\Delta\nu + \frac{1}{2}\Omega)^2 + \kappa'^2} + \frac{1}{(\Delta\nu - \frac{1}{2}\Omega)^2 + \kappa'^2} \right) \quad \text{for } \Omega \gg \kappa'_{jk},\ |\Delta\omega|\ , \tag{4.17}$$

where κ' is defined by Eq. (3.22).

Comparison of Eqs. (4.16) and (4.17) with Eqs. (3.19) and (3.21), respectively, shows that both in the limit of very weak and of very intense pump fields, the absorption and emission spectra are essentially identical, differing only by a normalization factor. In the general case, on the other hand, the absorption and emission spectra as given by Eqs. (4.14) and (3.18a), respectively, are represented by quite different functions, and no simple proportionality exists between them.

It is clear from Eqs. (3.8) and (4.12) for the correlation functions for emission and absorption that the important difference between the two functions is due to the presence of the off-diagonal matrix element $\bar{\alpha}_{10}$ in Eq. (4.12). Although the term in question cannot affect the integral of the absorption line-shape function over frequencies [this is clear from the identity $\mathfrak{U}_{j1;j0}(t=0) = 0$, which follows from evaluating Eq. (3.7) at $t = 0$], it does importantly modify the shape of the absorption spectrum whenever $\bar{\alpha}_{10}$ is appreciable, i.e., for pump intensities of intermediate magnitude. It should also be noted that, inasmuch as the derivation of Eqs. (3.16) and (4.13) does not depend in a detailed way on the form of the equations of motion for the elements of the 2×2 density submatrix elements referring to the pair of strongly coupled states $|0\rangle$ and $|1\rangle$, the relations in question may be presumed to remain valid even when, e.g., the coupling between the states $|0\rangle$ and $|1\rangle$ (but not between either one of these states and the other weakly coupled states of the atom) is described by means of more general forms[11] of atomic relaxation theory.

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[1] I. I. Rabi, Phys. Rev. 51, 652 (1937).

[2] B. R. Mollow and M. M. Miller, Ann. Phys. (N.Y.) 52, 464 (1969).

[3] Similar variations are induced when the driving field causes transitions by means of multiphoton processes. See, for example, B. R. Mollow, Phys. Rev. A 4, 1666 (1971).

[4] See, for example, R. Karplus and J. Schwinger, Phys. Rev. 73, 1020 (1948).

[5] B. R. Mollow, Phys. Rev. 188, 1969 (1969); Phys. Rev. A 2, 76 (1970).

[6] M. Newstein, Phys. Rev. 167, 89 (1968).

[7] C. R. Stroud, Jr., [Phys. Rev. A 3, 1044 (1971)] has obtained a similar splitting of the emission line by means of a direct quantum-electrodynamical analysis.

[7a] For the absorption spectrum corresponding to transitions from one of the resonantly coupled states to the other see, B. R. Mollow, Phys. Rev. A (to be published).

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

[9] S. H. Autler and C. H. Townes, Phys. Rev. 100, 703 (1955).

[10] Relations similar to our relations for the absorption spectrum appear in analyses of the three-level maser. See, for example, A. M. Clogston, J. Phys. Chem. Solids 4, 271 (1958); and V. M. Fain and Ya. I. Khanin, *Quantum Electronics* (MIT Press, Cambridge, Mass., 1969), Vol. I, Chap. V.

[11] It should be noted that in the case of coupling to "soft" photon or phonon modes, the use of the familiar form of atomic relaxation theory has been criticized by R. H. Lehmberg [Phys. Letters 33A, 501 (1970)] who proposes a more complicated theory to describe relaxation in the presence of very strong driving fields.

[12] It should be noted, however, that the driving field *indirectly* affects the occupation numbers $\bar{n}_j$ for $j \geq 2$, since these quantities are coupled through Eq. (2.10a) to $\bar{n}_0$ and $\bar{n}_1$.

[13] R. Kubo, J. Phys. Soc. Japan 12, 570 (1957).

[14] The quantity $\mathcal{W}$ approaches a fixed value in this limit, and makes a vanishingly small contribution in Eq. (4.11).