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Elastic and inelastic collisional and radiative damping effects on saturated line shapes in the limit of well-separated spectral lines

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Resonant scattering of intense light is analyzed in the limit in which the separation of the spectral lines is large compared to their widths. The effect on the line intensities and widths of elastic and inelastic collisions in the impact approximation and of radiative damping is found. The limit of well-separated spectral lines allows simple solutions in a wide class of cases not easily treated outside this limit, in particular the case in which the upper laser-coupled state decays through a complicated cascade sequence to the lower laser-coupled ground state, or suffers collisional reorientation (depolarization) during the emission process.

The resonant scattering of intense light has been the subject of numerous experimental[1-6] and theoretical[7-25] studies. Recent experiments utilizing atomic-beam techniques[1-3] have effectively eliminated Doppler and collisional broadening, and have provided a convincing verification[2] of theoretical predictions of the emission line shape for the case of a two-level system governed by purely radiative relaxation.[10(a)]

The case in which collisional relaxation may exceed radiative relaxation by an order of magnitude has recently been investigated in an interesting series of experiments by Carlsten and Szöke,[5] who have observed the expected Rabi splitting of the (Doppler-broadened) spectrum into three components by a high-power laser with detuning much greater than the (homogeneous and inhomogeneous) widths of the lines. Under such circumstances the form of the spectral density at frequency ν of the light scattered from an incident field $E_0\cos\omega t$ by a two-level system with resonant frequency ω_{10} may be approximated as

$$\tilde{g}_e(\nu)\approx|\vec{\mu}_{10}|^2 2\pi[A_0\delta(\nu-\omega)+A_+\delta(\nu-\omega+\Omega')+A_-\delta(\nu-\omega-\Omega')], \tag{1}$$

where $\Omega'=(\Omega^2+\Delta^2)^{1/2}$, $\Delta=\omega-\omega_{10}$, $\Omega=|\vec{\mu}_{10}\cdot\vec{E}_0|/\hbar$, and $\vec{\mu}_{10}$ is the electric dipole matrix element connecting the two laser-coupled states $|0\rangle$ and $|1\rangle$. The widths of the lines are approximated as zero in Eq. (1), and are understood to be much smaller than the Rabi splitting frequency Ω'.

The spectrum of the light scattered by an atomic system described by a density operator governed by equations of the general form

$$\left(\frac{d}{dt}+i\omega_{jk}+\kappa'_{jk}\right)\rho_{jk}(t)=i\vec{E}(t)\cdot[\vec{\mu},\rho(t)]_{jk}\quad(j\neq k), \tag{2a}$$

$$\left(\frac{d}{dt}+\kappa_j\right)n_j(t)=\sum_k n_k(t)\kappa_{kj}+i\vec{E}(t)\cdot[\vec{\mu},\rho(t)]_{jj}, \tag{2b}$$

where $\omega_{jk}=(E_j-E_k)/\hbar$,

$$n_j\equiv\rho_{jj},\quad \kappa_j=\sum_k\kappa_{jk},\quad \kappa'_{jk}=\kappa'_{kj}, \tag{2c}$$

can be evaluated exactly by means of the quantum fluctuation-regression theorem.[26] Equations of the form (2) describe both radiative relaxation and collisional relaxation in the impact regime. For a two-level system, for example, which is deexcited and dephased by radiative decay with Einstein A-coefficient $\Gamma_{10}=\Gamma_1$ and by inelastic and elastic collisions occurring at mean rate Q_I and Q_E, respectively, the inverse longitudinal and transverse relaxation times $\kappa_{10}=\kappa_1$ and κ'_{10} are[27,28]

$$\kappa_1=\Gamma_{10}+Q_I,\qquad \kappa'_{10}=\tfrac{1}{2}(\Gamma_{10}+Q_I+Q_E). \tag{3}$$

The emission spectrum as given by Eqs. (4.9), (2.11), and (2.16) of Ref. 10(c) is well approximated in the limit of well-separated spectral lines ($\Omega'\gg\kappa_1,\kappa'_{10}$) by the relation

$$\tilde{g}_e(\nu)=|\vec{\mu}_{10}|^2\left(|\bar{\rho}_{10}|^2 2\pi\delta(\nu-\omega)+\frac{2s_0A_0^{\rm inc}}{(\nu-\omega)^2+s_0^2}+\frac{2\sigma A_+}{(\nu-\omega+\Omega')^2+\sigma^2}+\frac{2\sigma A_-}{(\nu-\omega-\Omega')^2+\sigma^2}\right), \tag{4}$$

in which the widths are

$$s_0=(\kappa_1\Delta^2+\kappa'_{10}\Omega^2)/\Omega'^2,\qquad \sigma=[\kappa_1\Omega^2+\kappa'_{10}(\Omega^2+2\Delta^2)]/2\Omega'^2, \tag{5}$$

and the intensity coefficients are given by the relations

$$|\bar{\rho}_{10}|^2=\frac{\frac{1}{4}\Omega^2\Delta^2}{(\eta\Omega^2+\Delta^2)^2}, \tag{6a}$$

$$A_0^{\rm inc}=\frac{\frac{1}{4}\Omega^4[\eta^2\Omega^2+(2\eta-1)\Delta^2]}{\Omega'^2(\eta\Omega^2+\Delta^2)^2}, \tag{6b}$$

$$A_+=\frac{\frac{1}{8}\Omega^2(\Omega'+\Delta)[\eta(\Omega'+\Delta)-\Delta]}{\Omega'^2(\eta\Omega^2+\Delta^2)}, \tag{6c}$$

$$A_- = \frac{\frac{1}{8}\Omega^2(\Omega' - \Delta)[\eta(\Omega' - \Delta) + \Delta]}{\Omega'^2(\eta\Omega^2 + \Delta^2)}, \quad (6d)$$

where

$$\eta \equiv \kappa'_{10}/\kappa_1. \quad (6e)$$

If one approximates as zero all of the widths in Eq. (4) (and accordingly combines the coherent and incoherent terms at line center into a single term), one obtains a spectrum of the form given by Eq. (1), with

$$A_0 = |\bar{\rho}_{10}|^2 + A_0^{\text{inc}} = \tfrac{1}{4}\Omega^2/\Omega'^2, \quad (7)$$

and with A_+ and A_- given by Eqs. (6c) and (6d).

The central line in the spectrum in this limit is thus completely independent of the *type* of relaxation mechanism, as conjectured by Carlsten and Szöke.[5] The sideband strengths as given by Eqs. (6c) and (6d), on the other hand, depend critically upon the decay-constant ratio $\eta = \kappa'_{10}/\kappa_1$, and hence *are importantly dependent upon the type of relaxation mechanism even in the limit in which the spectral widths are ignored*. [Carlsten and Szöke have recently obtained improved agreement with theory by making use of Eqs. (6c), (6d), and (7).[29]]

As a means of understanding this result and of generalizing it to a somewhat wider class of cases, it is convenient to consider a simple way of deriving Eqs. (6c), (6d), and (7). The spectral density $\tilde{g}_e(\nu)$ is the Fourier transform of the atomic correlation function

$$g_e(t) = \text{tr}[\bar{\rho}\mu^{(-)}\mu^{(+)}(t)] \quad (8a)$$

$$= |\mu_{10}|^2 \text{tr}[\bar{\rho}a^\dagger a(t)], \quad (8b)$$

where

$$a^\dagger \equiv |1\rangle\langle 0|, \quad a = |0\rangle\langle 1| \quad (9)$$

are the atomic raising and lowering operators, respectively, and $\bar{\rho}$ is the steady-state atomic density operator.

The integrated line intensities discussed above can be obtained simply by ignoring the effect of atomic relaxation on the time evolution of $a(t)$ in Eq. (8), i.e., by putting

$$a(t) \approx e^{-i\omega t}(e^{i\mathcal{H}t}ae^{-i\mathcal{H}t}), \quad (10)$$

where $\mathcal{H}$ is the Hamiltonian for the laser-driven but undamped atom, evaluated in the rotating-wave representation,

$$\mathcal{H} = -\Delta a^\dagger a - \tfrac{1}{2}\Omega(a^\dagger + a). \quad (11)$$

By substituting Eq. (10) into Eq. (8b) and making use of Eqs. (9), one finds that[10(d)]

$$g_e(t) \approx |\vec{\mu}_{10}|^2 e^{-i\omega t}[\bar{n}_1\psi_1^{(1)}(t) + \bar{\rho}_{01}\psi_1^{(0)}(t)]\psi_0^{(0)*}(t), \quad (12)$$

where $\psi_j^{(k)}(t)$ is the pure-state amplitude for finding the driven atom in state $|j\rangle$ at time t if it was in state $|k\rangle$ initially,

$$\psi_j^{(k)}(t) = \langle j|e^{-i\mathcal{H}t}|k\rangle. \quad (13)$$

These amplitudes are easily evaluated with the aid of Eqs. (11) and (9), and when substituted into Eq. (12) lead to the relation

$$g_e(t) \approx |\vec{\mu}_{10}|^2[A_0e^{-i\omega t} + A_+e^{-i(\omega-\Omega')t} + A_-e^{-i(\omega+\Omega')t}], \quad (14)$$

with the coefficients A_0 and $A_\pm$ given as

$$A_0 = (\bar{n}_1 - \bar{\rho}_{01}\Delta/\Omega)\tfrac{1}{2}\Omega^2/\Omega'^2, \quad (15a)$$

$$A_\pm = \tfrac{1}{4}[\bar{n}_1(\Omega' \pm \Delta)^2 \pm \bar{\rho}_{01}\Omega(\Omega' \pm \Delta)]/\Omega'^2. \quad (15b)$$

Hence one finds a solution dependent only upon the atomic populations $\bar{n}_0$ and $\bar{n}_1$,

$$A_0 = (\bar{n}_0 + \bar{n}_1)\tfrac{1}{4}\Omega^2/\Omega'^2, \quad (15c)$$

$$A_\pm = \tfrac{1}{8}(\Omega' \pm \Delta)^2[\bar{n}_0 + \bar{n}_1 \mp (\bar{n}_0 - \bar{n}_1)\Omega'/\Delta]/\Omega'^2, \quad (15d)$$

as a consequence of the steady-state relation

$$\bar{\rho}_{01} \approx -\tfrac{1}{2}(\bar{n}_0 - \bar{n}_1)\Omega/\Delta, \quad (16)$$

which follows from Eqs. (2). When the steady-state solution

$$\bar{n}_0 - \bar{n}_1 \approx \Delta^2/(\eta\Omega^2 + \Delta^2) \quad (17a)$$

[which also follows from Eqs. (2)] is used along with the relation

$$\bar{n}_0 + \bar{n}_1 = 1 \quad (17b)$$

in Eqs. (15), one finds exactly the line intensities given by Eqs. (6c), (6d), and (7), and hence, on taking the Fourier transform of Eq. (14), one obtains the previously noted three-component sharp-line spectrum.

When energy-increasing transitions take place at rate $\bar{n}_0\kappa_{01}$ from the state $|0\rangle$ to the state $|1\rangle$, the zero-field population difference

$$\bar{n}_0^0 - \bar{n}_1^0 = (\kappa_{10} - \kappa_{01})/(\kappa_{10} + \kappa_{01}) \equiv d^0$$

appears as a factor in Eq. (17a),[10(c)] and the solution for $A_\pm$ is then

$$A_\pm = \tfrac{1}{8}(\Omega' \pm \Delta)^2(\Omega')^{-2}[1 \mp d^0\Omega'\Delta/(\eta\Omega^2 + \Delta^2)], \quad (18)$$

where $\eta = \kappa'_{10}/(\kappa_{10} + \kappa_{01})$. [Note that κ_{jk} in Ref. 10(c) corresponds to κ_{kj} in this paper.]

The procedure of ignoring the effect of damping on the time evolution of $a(t)$ in Eq. (8) must yield good values for the integrated line intensities A_0 and $A_\pm$ as long as many Rabi oscillations take place within the homogeneous atomic lifetime ($\Omega' \gg \kappa_1, \kappa'_{10}$). This is true simply because the resolution of $g(t)$ into its Fourier components then takes place before damping begins to have an effect.

Where damping cannot be ignored is in the expressions for the steady-state atomic density matrix elements. The solution in Eq. (17a) in particular depends importantly upon the damping ratio $\eta = \kappa'_{10}/\kappa_1$, no matter how small the damping constants are compared to the other frequency parameters (Ω and Δ) of the problem. [The assumption of steady-state conditions, on the other hand, requires the atomic lifetime to be short compared to the length of time the atom spends in the field. The correlation function $\langle \mu^{(-)}(t')\mu^{(+)}(t'+t)\rangle$ in the presently considered approximation is thus in effect evaluated in the short-time limit for t, but in the long-time limit for t'.]

It is clear from the method of their derivation that the Eqs. (15) are quite general. Their validity depends only upon the sharp-line assumption $\Omega' \gg \kappa_1, \kappa'_{10}$ and upon the assumption that the two states $|0\rangle$ and $|1\rangle$ are the only states directly coupled by the laser field. Thus $|0\rangle$ need not be the ground state of the atom, while decay out of and incoherent repopulation back into the $|0\rangle - |1\rangle$ subspace may take place via arbitrarily complicated sequences involving the other states of the atom. The Eqs. (15) for the integrated line intensities remain valid, though of course the density matrix elements in them must be determined from Eqs. (2) rather than from Eqs. (17). The solution so obtained naturally depends upon the steady-state repopulation rates into the states $|0\rangle$ and $|1\rangle$, but is otherwise independent of the details of the decay-repopulation sequence, even when the repopulation rates are appreciable compared to the other decay constants of the problem. [The sharp-line solution under discussion is in this respect more general than the exact solution found in Ref. 10(f). That solution was not limited by a sharp-line assumption, but did require vanishingly small repopulation rates, and hence was valid only for $\bar{n}_0 + \bar{n}_1 \ll 1$.]

An important case which can easily be treated in this way occurs when $|0\rangle$ is the ground state of the atom and $|1\rangle$ is an upper excited state which can decay to other excited states $|j\rangle$ of lower energy. A complicated cascade process then takes place, ultimately returning the atom to its ground state $|0\rangle$. In steady state, the populations $\bar{n}_j$ (where $j = 1, 2, \ldots$) of all of the states in the cascade sequence except $|0\rangle$ bear fixed ratios to one another, dependent only upon the lifetime ratios of the states involved. Hence one may put

$$1 - (\bar{n}_0 + \bar{n}_1) = \sum_{j\,(j\neq 0,1)} \bar{n}_j = x\bar{n}_1, \tag{19}$$

where the parameter x is characteristic of the atom but independent of the laser field. Another case that can be similarly described is that of *collisional reorientation*, in which collisions induce transitions back and forth between the upper laser-coupled state $|1\rangle$ and other members of the same Zeeman multiplicity (with the latter assumed uncoupled to the laser). Here too the ratios between $\bar{n}_1$ and the populations of the other excited levels are dependent only upon the (collisional and radiative) relaxation rates, and are thus independent of the laser field. [The fundamental assumption necessary for Eq. (19) is simply that no relaxation-induced transition take place from the atomic ground state $|0\rangle$ to any other atomic state.]

By making use of Eq. (19) in Eqs. (2) to evaluate the density matrix elements which appear in Eqs. (15), one finds that the spectral line intensities corresponding to the $|1\rangle - |0\rangle$ transition may be expressed in terms of the single parameter x by means of the relations

$$A_0 = \frac{\frac{1}{4}\Omega^2(\eta\Omega^2 + \Delta^2)}{\Omega'^2[(1+\frac{1}{2}x)\eta\Omega^2 + \Delta^2]} \tag{20a}$$

$$A_\pm = \frac{\frac{1}{8}\Omega^2(\Omega' \pm \Delta)[\eta(\Omega' \pm \Delta) \mp \Delta]}{\Omega'^2[(1+\frac{1}{2}x)\eta\Omega^2 + \Delta^2]}, \tag{20b}$$

where $\eta = \kappa'_{10}/\kappa_1$, with κ_1 the full width of the state $|1\rangle$. The coherent contribution to the central term in the spectrum is

$$|\bar{\rho}_{10}|^2 = \tfrac{1}{4}\Omega^2\Delta^2/[(1+\tfrac{1}{2}x)\eta\Omega^2 + \Delta^2]^2. \tag{20c}$$

In a general two-level problem with decay and repopulation of the kind under discussion, the detailed structure of the *incoherent* contribution to A_0 cannot be evaluated, even in the presently considered limit of well-separated spectral lines, if the repopulation rate is appreciable (i.e., if $\bar{n}_0 + \bar{n}_1$ is not small compared to unity) except by means of a detailed solution in which all of the participating states play an important role. The solution in question is not in general expressible simply as a Lorentzian function, nor would it be expressible in the example discussed above in terms of the single parameter x.

The sidebands, on the other hand, are simply Lorentzian functions in the general case under discussion, and their widths can be accurately evaluated by means of relations which make no reference whatever to states of the atom other than $|0\rangle$ and $|1\rangle$. Here it is useful to adapt to the case of general relaxation coefficients a procedure which Cohen-Tannoudji and Reynaud[30] have developed to treat the case of purely radiative relaxation in the limit of well-separated resonance lines when many levels are coupled by the laser field, particularly when (Zeeman or hyperfine) degeneracy or near-degeneracy exists within the optically separated levels. It is a simple matter to show (by using operator radiation-reaction theory,[10(e),21-24] for example) that even when degeneracy is present, the radiative damping of the atomic density operator is governed by the general equation

$$\left(\frac{d}{dt}\right)_{\rm rad}\rho=-\tfrac{1}{2}(\vec{d}^{\dagger}\cdot\vec{d}\rho+\rho\vec{d}^{\dagger}\cdot\vec{d})+\vec{d}\cdot\rho\vec{d}^{\dagger}, \tag{21a}$$

where the optical energy-lowering operator $\vec{d}$ is defined by the relation

$$\vec{d}_{jk}\equiv\vec{\mu}_{jk}^{(+)}(\omega_{jk}^{3}/3\pi\hbar c^{3})^{1/2} \tag{21b}$$

in rationalized units.

Following Cohen-Tannoudji and Reynaud, one may find the eigenstates $|\alpha\rangle$ of the rotating-wave Hamiltonian $\mathcal{H}$,

$$\mathcal{H}|\alpha\rangle=\omega_{\alpha}|\alpha\rangle \tag{22}$$

[with $\mathcal{H}$ defined by a suitable multistate generalization of Eq. (11)], and think of the spectral lines at frequency $\omega+\omega_{\alpha\beta}$ as resulting from transitions from the state $|\alpha\rangle$ to the state $|\beta\rangle$. (In the "dressed-atom" approach adopted in Ref. 30, an infinite hierachy of states $|\alpha\rangle_n$ appears, with energy separation ω between $|\alpha\rangle_n$ and $|\alpha\rangle_{n-1}$.) When the spectral lines are well separated (as they are, for example, when an intense laser removes the initial degeneracy of the system), an off-diagonal density matrix element $\rho_{\alpha\beta}(t)$ is effectively decoupled in its time evolution from all other density matrix elements, and consequently by virtue of Eq. (21a) obeys the equation[30]

$$\left(\frac{d}{dt}+i\omega_{\alpha\beta}+\sigma_{\alpha\beta}\right)\rho_{\alpha\beta}(t)\approx 0, \tag{23}$$

where

$$\sigma_{\alpha\beta}=\sigma_{\beta\alpha}=\tfrac{1}{2}(\Gamma_{\alpha}+\Gamma_{\beta})-\mathrm{Re}(\vec{d}_{\alpha\alpha}\cdot\vec{d}_{\beta\beta}^{*}), \tag{24a}$$

$$\Gamma_{\alpha}=(\vec{d}^{\dagger}\cdot\vec{d})_{\alpha\alpha}. \tag{24b}$$

The populations n_α, on the other hand, are coupled to one another by rate equations involving the transition rates

$$\Gamma_{\alpha\beta}=|\vec{d}_{\beta\alpha}|^{2}, \tag{25}$$

but are effectively decoupled from the off-diagonal density matrix elements in the limit in question.

That $\sigma_{\alpha\beta}$ is the width of the spectral line at frequency $\omega+\omega_{\alpha\beta}$ follows at once from an expansion of the correlation function in Eq. (8a) in the basis set $\{|\alpha\rangle\}$. One also finds in this way the simple expression[30]

$$A_{\alpha\beta}=\bar{n}_{\alpha}|\vec{\mu}_{\beta\alpha}^{(+)}|^{2}\propto\bar{n}_{\alpha}\Gamma_{\alpha\beta}. \tag{26}$$

for the intensity of the line in question.

In the nondegenerate two-level case, where the transformed basis set $\{|\alpha\rangle\}$ consists of the states $|+\rangle$ and $|-\rangle$ with eigenfrequencies $-\frac{1}{2}\Omega'$ and $+\frac{1}{2}\Omega'$, respectively, the radiative transition rates are

$$\Gamma_{+-}=\tfrac{1}{4}\Gamma_{10}(\Omega'+\Delta)^{2}/\Omega'^{2} \tag{27}$$
$$\Gamma_{-+}=\tfrac{1}{4}\Gamma_{10}(\Omega'-\Delta)^{2}/\Omega'^{2},$$

and the integrated line intensities A_+ and A_- at the frequencies $\omega-\Omega'$ and $\omega+\Omega'$, respectively, are simply[31]

$$A_{+}\propto n_{+}\Gamma_{+-},\quad A_{-}\propto n_{-}\Gamma_{-+}. \tag{28}$$

When collisional as well as radiative damping is present, an equation of the form (23) is obtained in the limit of well-separated spectral lines by a direct expansion of Eqs. (2) in the transformed basis set $\{|\alpha\rangle\}$. In the two-level case, one finds that the (equal) widths of the two sidebands are

$$\sigma=\kappa_{10}'+\tfrac{1}{4}(\kappa_{1}+\kappa_{10}+\kappa_{01}+\kappa_{0}-2\kappa_{10}')\Omega^{2}/\Omega'^{2}. \tag{29}$$

Here decay out of and (possibly appreciable) repopulation into the $|0\rangle-|1\rangle$ subspace has been allowed, and for the sake of generality the effect of an upward transition rate κ_{01} has been included.

It should be apparent from the foregoing discussion that the formula (29) for the sideband widths, like the formulas (15) for the line intensities, is quite generally valid, provided only that the states $|0\rangle$ and $|1\rangle$ are the only atomic states directly coupled by the laser field.

The method of Cohen-Tannoudji and Reynaud also provides a way of understanding how the symmetry of the emission spectrum, which is present in the case of purely radiative damping, can be removed by collisions. In steady state, the populations $\bar{n}_\alpha$ obey the relations

$$\bar{n}_{\alpha}\kappa_{\alpha\beta}=\bar{n}_{\beta}\kappa_{\beta\alpha} \tag{30}$$

if "detailed balance" is present, as it of course must be in the two-level case. If the transition rates are purely radiative ($\kappa_{\alpha\beta}=\Gamma_{\alpha\beta}$), then the steady-state relation (30) together with Eq. (26) immediately implies symmetry of the spectrum,[30] at least in the presently considered limit of well-separated spectral lines.

When collisions are present, on the other hand, the proportionality between the total transition rate $\kappa_{\alpha\beta}$ and $|\vec{\mu}_{\beta\alpha}^{(+)}|^2$ may be lost, and Eqs. (30) and (26) then imply an *asymmetrical* spectrum. This is exactly what happens in the two-level case when elastic collisions are present.[28] If there is no decay out of or repopulation into the $|0\rangle-|1\rangle$ subspace, one finds with the aid of Eqs. (2) and (3) the transition rates

$$\kappa_{+-}=\tfrac{1}{4}[(\Gamma_{10}+Q_{I})(\Omega'+\Delta)^{2}+Q_{E}\Omega^{2}]/\Omega'^{2},$$
$$\kappa_{-+}=\tfrac{1}{4}[(\Gamma_{10}+Q_{I})(\Omega'-\Delta)^{2}+Q_{E}\Omega^{2}]/\Omega'^{2}, \tag{31}$$

which are proportional to the *radiative* transition rates given by Eqs. (27) only if the elastic collision rate Q_E vanishes. Thus the spectrum is asymmetrical unless $Q_E = 0$.[28]

The steady-state probabilities $\bar{n}_+$ and $\bar{n}_-$ of finding the atomic pseudospin parallel and antiparallel, respectively, to the equivalent field $(\Omega, 0, \Delta)$ are found from Eqs. (30), (31), (3), and (6e) to have the values

$$\bar{n}_\pm = \tfrac{1}{2} \mp \tfrac{1}{2}\Omega'\Delta/(\eta\Omega^2 + \Delta^2), \quad (32)$$

which of course depend through the parameter η on the type of relaxation mechanism which operates, in particular upon the ratio $Q_E/(\Gamma_{10}+Q_I)$. The steady-state line intensities given by Eqs. (6c) and (6d) of course follow directly upon substituting Eq. (32) into Eqs. (28) and (27).

In the case in which the laser field consists of a pulse short compared to the (possibly collision-reduced) atomic lifetime but long compared to the Rabi period Ω'^{-1}, it is clear that Eqs. (15) and (28) are still valid, but with occupation numbers determined from adiabatic relaxation-free equations[32] rather than from relaxation-determined steady-state equations. For $\Delta > 0$, the atomic pseudospin is then antiparallel to the effective field, i.e., $n_+ = 0$ and $n_- = 1$, and hence one finds

$$A_+ = 0, \; A_0 = \tfrac{1}{4}\Omega^2/\Omega'^2, \; A_- = \tfrac{1}{4}(\Omega' - \Delta)^2/\Omega'^2 \; (\Delta > 0). \quad (33)$$

The sideband with frequency nearer to the atomic resonance frequency is thus entirely absent in this limit, while the other sideband is correspondingly enhanced.[29,33-35]

An analysis of the absorption of a weak probe field by the pumped atoms can be carried out by suitably modifying the above discussion of emission so as to evaluate the absorption line-shape function, which is the Fourier transform of the function[10(c)]

$$g_a(t) = [\mu^{(+)}(t), \mu^{(-)}]. \quad (34)$$

The analysis in the case of absorption is somewhat simpler than for emission, and in the two-level case leads directly in the limit of well-separated spectral lines to a steady-state spectrum with two components[30,36]—one absorbing and one amplifying—with collision-modified widths given by Eq. (29), and with integrated line intensities B_+ and B_- given by the relations

$$B_\pm = \pm\tfrac{1}{4}(\bar{n}_0 - \bar{n}_1)(\Omega' \pm \Delta)^2/\Omega'\Delta. \quad (35)$$

Precisely because of the absence of the central component, the absorption spectrum, unlike the emission spectrum, is thus accurately and completely represented, even when decay out of and appreciable repopulation back into the laser-coupled subspace $|0\rangle - |1\rangle$ take place, by formulas which make no detailed reference to the decay-repopulation sequence. Indeed, the precise nature of the relaxation mechanism in operation has no effect whatever, in the sharp-line limit (where widths are ignored), upon the *shape* of the absorption spectrum, and is important only in its effect upon the over-all laser-dependent factor $\bar{n}_0 - \bar{n}_1$.

The case in which emissive or absorptive transitions take place between a laser-coupled state and another, uncoupled state of the atom has been treated in detail in Refs. 12, which contain explicit and general formulas describing the limit of well-separated resonance lines.[37] [If the system decays completely out of the laser-coupled $|0\rangle - |1\rangle$ subspace, the formulas of Ref. 12 become valid under the substitution[10(f)]

$$\bar{\rho}_{jk} \to R\hat{\rho}_{jk}(0) \equiv R\int_0^\infty \rho_{jk}(t)\,dt \quad (j,k=0,1), \quad (36)$$

where R is the rate of preparation of systems in the $|0\rangle - |1\rangle$ subspace.]

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[35]The pulse length in Ref. 29 was comparable to the atomic lifetimes, and hence steady-state conditions were satisfied only roughly. The regime investigated was in fact intermediate between the steady-state regime and the undamped-adiabatic regime.

[36]Near $\Delta = 0$ (for Δ comparable to or smaller than κ), a more accurate evaluation of the absorption lineshape function leads to dispersionlike curves rather than to simple Lorentzian functions. See, for example, Ref. 10(c), Eq. (3.19b) or Fig. 1.

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