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TITLE: RESPONSE OF TWO-LEVEL ATOMS TO INTENSE AMPLITUDE MODULATED
LASER BEAMS

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The optical Bloch equations for a two-level atom in a laser beam with an amplitude of arbitrary time dependence are derived in the rotating wave approximation. The light scattered by the atom is investigated for the special case of an intense, resonant laser beam with a sinusoidally modulated amplitude. The coherent part and the incoherent part of the spectrum and the coherent part and the incoherent part of the total scattered intensity are calculated. The results for this modulated amplitude dynamic Stark effect and for the constant amplitude dynamic Stark effect are compared. The modulation introduces new peaks in the frequency distribution. The total intensities when plotted as functions of the average Rabi frequency exhibit parametric resonances. The results in the limit of zero modulation depth are in agreement with the results of the constant amplitude dynamic Stark effect.

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RESPONSE OF TWO-LEVEL ATOMS TO INTENSE AMPLITUDE MODULATED LASER BEAMS

1. INTRODUCTION

The spectral distribution of the light scattered by a two-level atom in an intense laser beam of constant amplitude has been termed the dynamic Stark effect. It has been the subject of a number of theoretical papers, as well as experimental ones.

We treat the case of a two-level atom in an intense, resonant, amplitude modulated laser beam. We refer to our case as the modulated amplitude dynamic Stark effect and to avoid confusion we name the dynamic Stark effect more specifically the constant amplitude dynamic Stark effect.

In Chapter 2 we derive the equations of motion for the elements of the density matrix of a two-level atom which interacts with a laser beam of arbitrary time dependent amplitude and at the same time undergoes spontaneous decay. We obtain these equations from the equations of motion for a system consisting of the atom and a radiation field by tracing over the field states. The laser-atom interaction is treated classically from the start.

In Chapter 3 we rewrite the equations of motion of the previous chapter in terms of a new set of variables, namely the atomic inversion and the components of the atomic dipole moment in phase and $\frac{\pi}{2}$ out of phase with the electric field of the laser. The equations in the new form are known as the optical Bloch equations. We solve them for the particular case of a sinusoidally modulated laser amplitude.

In Chapter 4 we derive the relation between the atomic variables and the light emitted by the atom. We subsequently calculate the coherently and incoherently scattered intensities as well as their spectral distributions.

In Chapter 5 we graph our results, interpret them and compare them to the constant amplitude dynamic Stark effect.

2. EQUATIONS OF MOTION FOR THE ELEMENTS OF THE ATOMIC DENSITY MATRIX OF THE TWO-LEVEL ATOM

2.1 Two-Level Atom Undergoing Spontaneous Decay: Time Dependence of the Amplitude of the Excited State

Whenever a Hamiltonian can be written as $H = H_0 + H_I$, where H_0 is the unperturbed part of the Hamiltonian and H_I is the perturbation, we can go from the Schrödinger representation with

$$H |\Phi\rangle = i\hbar \frac{\partial}{\partial t} |\Phi\rangle \quad (2-1)$$

to the interaction representation where

$$H_I' |\Phi'\rangle = i\hbar \frac{\partial}{\partial t} |\Phi'\rangle \quad (2-2)$$

with

$$|\Phi'\rangle = e^{\frac{i}{\hbar} H_0 t} |\Phi\rangle \quad (2-3)$$

and

$$H_I' = e^{\frac{i}{\hbar} H_0 t} H_I e^{-\frac{i}{\hbar} H_0 t} \quad (2-4)$$

We write $|\Phi'\rangle$ as a linear combination of states $|n\rangle$, which are orthonormal eigenfunctions of H_0 . Thus

$$|\Phi'\rangle = \sum_n b_n(t) |n\rangle \quad (2-5)$$

and we obtain the following differential equations for the amplitudes $b_n(t)$:

$$i\hbar \frac{\partial}{\partial t} b_m(t) = \sum_n b_n(t) \langle m | H_I' | n \rangle \quad (2-6)$$

We consider a two-level atom with an excited state $|j\rangle$ of energy $\hbar\omega_j$ and a ground state $|k\rangle$ of energy $\hbar\omega_k$. We write the vacuum state of the radiation field as $|0\rangle$ and the one photon states as

$|\lambda_e\rangle$ where λ_e stands for all the quantities which describe a photon in this mode, namely the frequency ω_{λ_e} , the polarization direction \hat{e}_{λ_e} and the propagation direction \vec{k}_{λ_e} .

The annihilation operator a_{λ_e} and the creation operator $a_{\lambda_e}^+$ for a photon in mode λ_e obey the commutation relation for bosons,

$[a_{\lambda_e}, a_{\lambda_m}^+] = \delta_{e,m}$. We can write some basic relations for the field states $|0\rangle$ and $|\lambda_e\rangle$ in terms of these operators as

$$a_{\lambda_e}|0\rangle = 0 \quad \langle 0|a_{\lambda_e}^+ = 0 \quad (2-7)$$

$$a_{\lambda_e}^+|0\rangle = |\lambda_e\rangle \quad \langle 0|a_{\lambda_e} = \langle \lambda_e| \quad (2-8)$$

$$\langle 0|\lambda_e\rangle = \langle 0|a_{\lambda_e}^+|0\rangle = 0 \quad (2-9)$$

$$\langle \lambda_e|\lambda_m\rangle = \langle 0|a_{\lambda_e}a_{\lambda_m}^+|0\rangle = \langle 0|a_{\lambda_m}^+a_{\lambda_e} + \delta_{e,m}|0\rangle = \delta_{e,m} \quad (2-10)$$

The unperturbed Hamiltonian H_0 consists of an atomic part H_A and a radiation Hamiltonian H_R , such that

$$H_A|j\rangle = \hbar\omega_j|j\rangle \quad (2-11)$$

$$H_A|k\rangle = \hbar\omega_k|k\rangle \quad (2-12)$$

$$H_R|0\rangle = 0 \quad (2-13)$$

$$H_R|\lambda_e\rangle = \hbar\omega_{\lambda_e}|\lambda_e\rangle \quad (2-14)$$

The transition of the system of atom plus radiation field (from now on simply called the system) from the initial state $|i\rangle = |j\rangle|0\rangle$ to the final states $|f_e\rangle = |k\rangle|\lambda_e\rangle$ is called spontaneous decay. Spontaneous decay has been treated extensively in the literature^{1,2,3,4}. We review it here to present our notation, to demonstrate our method of introducing the decay rate and to provide some means of comparison between the approximations made here and in later sections of this

chapter.

The states $|i\rangle$ and $|f_e\rangle$ are eigenstates of the unperturbed Hamiltonian H_0 with

$$H_0|i\rangle = (H_A + H_R)|i\rangle = \hbar\omega_j|i\rangle = \hbar\omega_i|i\rangle \quad (2-15)$$

and

$$H_0|f_e\rangle = (H_A + H_R)|f_e\rangle = \hbar(\omega_k + \omega_{\lambda_e})|f_e\rangle = \hbar\omega_{fe}|f_e\rangle \quad (2-16)$$

The interaction Hamiltonian is

$$H_1 = -\frac{e}{m} \vec{p} \cdot \vec{A} \quad (2-17)$$

with the quantized vector potential

$$\vec{A} = \sum_{\ell} \sqrt{\frac{\hbar}{2\omega_{\lambda_{\ell}}\epsilon_0}} \{ \hat{e}_{\lambda_{\ell}} a_{\lambda_{\ell}} e^{i\vec{k}_{\lambda_{\ell}} \cdot \vec{r}} + \hat{e}_{\lambda_{\ell}}^* a_{\lambda_{\ell}}^+ e^{-i\vec{k}_{\lambda_{\ell}} \cdot \vec{r}} \} \quad (2-18)$$

Using Equations (2-4), (2-7)-(2-10) and (2-15)-(2-17) we find that

$$\langle i | H_1' | i \rangle = 0 \quad (2-19)$$

$$\langle f_e | H_1' | f_m \rangle = 0 \quad \forall \ell, m \quad (2-20)$$

$$\begin{aligned} \langle i | H_1' | f_e \rangle &= \sum_m \langle j | \langle 0 | e^{\frac{i}{\hbar} H_0 t} \left(-\frac{e}{m} \vec{p} \cdot \right) \\ &\quad \sqrt{\frac{\hbar}{2\omega_{\lambda_m}\epsilon_0}} \{ \hat{e}_{\lambda_m} a_{\lambda_m} e^{i\vec{k}_{\lambda_m} \cdot \vec{r}} + \hat{e}_{\lambda_m}^* a_{\lambda_m}^+ e^{-i\vec{k}_{\lambda_m} \cdot \vec{r}} \} e^{-\frac{i}{\hbar} H_0 t} | k \rangle | \lambda_e \rangle \\ &\stackrel{\text{Def.}}{=} \hbar V_{ife} e^{i(\omega_i - \omega_{fe})t} \end{aligned} \quad (2-21)$$

where

$$\hbar V_{ife} = - \langle j | \sqrt{\frac{\hbar}{2\omega_{\lambda_e}\epsilon_0}} \frac{e}{m} \vec{p} \cdot \hat{e}_{\lambda_e} e^{i\vec{k}_{\lambda_e} \cdot \vec{r}} | k \rangle \quad (2-22)$$

and

$$\begin{aligned} \langle f_e | H_1' | i \rangle &= \langle i | H_1' | f_e \rangle^* = \hbar V_{ife}^* e^{-i(\omega_i - \omega_{fe})t} \\ &\stackrel{\text{Def.}}{=} \hbar V_{fei} e^{-i(\omega_i - \omega_{fe})t} \end{aligned} \quad (2-23)$$

where

$$\hbar V_{fei} = - \langle k | \sqrt{\frac{\hbar}{2\omega_{\lambda_e}\epsilon_0}} \frac{e}{m} \vec{p} \cdot \hat{e}_{\lambda_e}^* e^{-i\vec{k}_{\lambda_e} \cdot \vec{r}} | j \rangle \quad (2-24)$$

Before making use of Equation (2-6) a peculiarity of bound state problems has to be pointed out³: In problems involving discrete bound states the transition probability per unit time between states is finite, so that it is not possible to fix the initial conditions for $t = -\infty$; instead they have to be fixed for a finite time, say $t = 0$. Then Equation (2-6) is physically meaningful only for $t > 0$. We would like these equations to hold for all times, however. If we assume $b_i(t) = 0$ and $b_{fe}(t) = 0$ for $t < 0$, these amplitudes will satisfy Equation (2-6) for negative times.

It was pointed out earlier that in the case of spontaneous decay initially only state $|i\rangle$ is populated. This implies that

$$b_i(0) = 1 \quad ; \quad b_{fe}(0) = 0 \quad \forall l \quad (2-25)$$

There is a discontinuity in $b_i(t)$ at $t = 0$, so that the integral of $\frac{db_i(t)}{dt}$ over a small time interval around $t = 0$ is

$$\int_{0-\epsilon}^{0+\epsilon} \frac{db_i(t)}{dt} dt = b_i(0+\epsilon) - b_i(0-\epsilon) = 1$$

We conclude that we have to add a delta function term in the differential equation for $b_i(t)$ to achieve this. Therefore, with the use of Equations (2-6) and (2-19)-(2-24) we arrive at the following set of differential equations, which are valid for all times:

$$i\dot{b}_i(t) = \sum_l V_{ife} e^{i(\omega_i - \omega_{fe})t} b_{fe}(t) + i\delta(t) \quad (2-26)$$

$$i\dot{b}_{fe}(t) = V_{fei} e^{-i(\omega_i - \omega_{fe})t} b_i(t) \quad (2-27)$$

We solve these equations by Fourier transforming the amplitudes.

We define $b_m(t) = -\frac{1}{2\pi i} \int_{-\infty}^{\infty} B_m(\omega) e^{-i\omega t} e^{i\omega_m t} d\omega$, so that

$$b_i(t) = -\frac{1}{2\pi i} e^{i\omega_i t} \int_{-\infty}^{\infty} B_i(\omega) e^{-i\omega t} d\omega \quad (2-28)$$

$$b_{fe}(t) = -\frac{1}{2\pi i} e^{i\omega_{fe} t} \int_{-\infty}^{\infty} B_{fe}(\omega) e^{-i\omega t} d\omega \quad (2-29)$$

We write the delta function as

$$i\delta(t) = -\frac{1}{2\pi i} e^{i\omega_i t} \int_{-\infty}^{\infty} e^{-i\omega t} d\omega \quad (2-30)$$

where the factor $e^{i\omega_i t}$ is introduced in the definition of the delta function since it is also present in the definition of $b_i(t)$ and can be factored out of the equation which we obtain by substituting Equations (2-28)-(2-30) into Equation (2-26), namely

$$\begin{aligned} i \int_{-\infty}^{\infty} i(\omega_i - \omega) e^{i\omega_i t} e^{-i\omega t} B_i(\omega) d\omega \\ = \sum_l V_{ife} e^{i(\omega_i - \omega_{fe})t} \int_{-\infty}^{\infty} e^{i(\omega_{fe} - \omega)t} B_{fe}(\omega) d\omega \\ + \int_{-\infty}^{\infty} e^{i\omega_i t} e^{-i\omega t} d\omega \end{aligned}$$

or

$$\int_{-\infty}^{\infty} e^{i\omega_i t} \left[(\omega - \omega_i) B_i(\omega) - \sum_l V_{ife} B_{fe}(\omega) - 1 \right] e^{-i\omega t} d\omega = 0$$

which implies that

$$(\omega - \omega_j) B_i(\omega) = \sum_l V_{ife} B_{fe}(\omega) + 1 \quad (2-31)$$

Similarly, Equation (2-27) becomes

$$(\omega - \omega_K - \omega_{\lambda e}) B_{fe}(\omega) = V_{fei} B_i(\omega) \quad (2-32)$$

We replace $B_{fe}(\omega)$ in Equation (2-31) by the expression obtained from Equation (2-32) and get

$$(\omega - \omega_j - \sum_l \frac{V_{ife} V_{fei}}{\omega - \omega_k - \omega_{\lambda_l}}) B_i(\omega) = 1 \quad (2-33)$$

For photons in a cavity the density of modes in the frequency range $\omega_{\lambda_l} \dots \omega_{\lambda_l} + d\omega_{\lambda_l}$ is

$$g(\omega_{\lambda_l}) d\omega_{\lambda_l} = \frac{\omega_{\lambda_l}^2 d\omega_{\lambda_l}}{\pi^2 c^3}$$

We can replace the sum over states $|f_l\rangle$ in Equation (2-33) by an integral over frequencies, an integral over propagation directions and a sum over polarizations if we introduce the following density of states:

$$g(\omega_{\lambda_l}) d\omega_{\lambda_l} d\Omega = \frac{\omega_{\lambda_l}^2 d\omega_{\lambda_l} d\Omega}{8\pi^3 c^3} \quad (2-34)$$

Then

$$\sum_l \frac{V_{ife} V_{fei}}{\omega - \omega_k - \omega_{\lambda_l}} = \frac{1}{8\pi^3 c^3} \sum_{pol} \int_0^\infty d\omega_{\lambda_l} \int_0^{4\pi} d\Omega \frac{|V_{ife}|^2 \omega_{\lambda_l}^2}{\omega - \omega_k - \omega_{\lambda_l}} \quad (2-35)$$

where V_{ife} is given by Equation (2-22).

In the dipole approximation we can set $e^{i\vec{k}_{\lambda_l} \cdot \vec{r}} \approx 1$, so that

$$V_{ife} \approx -\frac{1}{\hbar} \frac{e}{m} \sqrt{\frac{\hbar}{2\omega_{\lambda_l} \epsilon_0}} \hat{e}_{\lambda_l} \cdot \langle j | \vec{p} | k \rangle \quad (2-36)$$

The following relations between matrix elements can be derived⁵:

$$\langle j | \vec{p} | k \rangle = i m (\omega_j - \omega_k) \langle j | \vec{r} | k \rangle \quad (2-37)$$

and

$$\overline{|\langle j | \vec{r} \cdot \hat{e} | k \rangle|^2} = \frac{1}{3} |\langle j | \vec{r} | k \rangle|^2 \quad (2-38)$$

where the bar denotes an average over the polarization directions \hat{e} .

We calculate $|V_{ife}|^2$ using Equations (2-36) and (2-37) and subsequently average over polarizations. We obtain

$$|V_{ife}|^2 = \frac{e^2}{2\omega_{\lambda e} \epsilon_0 \hbar} (\omega_j - \omega_k)^2 \frac{1}{3} |\langle j | \vec{r} | k \rangle|^2$$

We substitute this into Equation (2-35), sum over the polarization directions and integrate over the propagation directions. Then

$$\sum_e \frac{V_{ife} V_{fei}}{\omega - \omega_k - \omega_{\lambda e}} = \frac{e^2 (\omega_j - \omega_k)^2}{6\pi^2 c^3 \epsilon_0 \hbar} |\langle j | \vec{r} | k \rangle|^2 \int_0^\infty \frac{\omega_{\lambda e} d\omega_{\lambda e}}{\omega - \omega_k - \omega_{\lambda e}} \quad (2-39)$$

In Equation (2-39) we can extend the limit of integration to $-\infty$. This introduces a negligible error since the integrand is strongly peaked at $\omega_{\lambda e} = \omega_j - \omega_k$ as we shall see later and thus is essentially zero for negative values of $\omega_{\lambda e}$. Since

$$\int_{-\infty}^{\infty} \frac{f(\omega) d\omega}{\omega} = \lim_{\epsilon \rightarrow 0} \int_{-\infty}^{\infty} \frac{f(\omega) d\omega}{\omega + i\epsilon} = P \int_{-\infty}^{\infty} \frac{f(\omega) d\omega}{\omega} - i\pi \int_{-\infty}^{\infty} \delta(\omega) f(\omega) d\omega$$

where P denotes the Cauchy principal value, we can replace $\frac{1}{\omega - \omega_k - \omega_{\lambda e}}$ by the zeta function $\zeta(\omega - \omega_k - \omega_{\lambda e}) = P\left(\frac{1}{\omega - \omega_k - \omega_{\lambda e}}\right) - i\pi \delta(\omega - \omega_k - \omega_{\lambda e})$ so that Equation (2-39) becomes

$$\begin{aligned} \sum_e \frac{V_{ife} V_{fei}}{\omega - \omega_k - \omega_{\lambda e}} &= \\ &= \frac{e^2 (\omega_j - \omega_k)^2}{6\pi^2 c^3 \epsilon_0 \hbar} |\langle j | \vec{r} | k \rangle|^2 \left\{ P \int_{-\infty}^{\infty} \frac{\omega_{\lambda e} d\omega_{\lambda e}}{\omega - \omega_k - \omega_{\lambda e}} - i\pi \int_{-\infty}^{\infty} \omega_{\lambda e} \delta(\omega - \omega_k - \omega_{\lambda e}) d\omega_{\lambda e} \right\} \\ &= \eta(\omega) - \frac{i}{2} \gamma(\omega) \end{aligned} \quad (2-40)$$

where

$$\gamma(\omega) = \frac{e^2 (\omega_j - \omega_k)^2}{3\pi c^3 \epsilon_0 \hbar} |\langle j | \vec{r} | k \rangle|^2 (\omega - \omega_k) \equiv D(\omega - \omega_k) \quad (2-41)$$

and

$$\begin{aligned}\eta(\omega) &= \frac{e^2(\omega_j - \omega_K)^2}{6\pi^2 c^3 \epsilon_0 \hbar} |\langle j | \vec{r} | k \rangle|^2 \rho \int_{-\infty}^{\infty} \frac{\omega_{\lambda e} d\omega_{\lambda e}}{\omega - \omega_K - \omega_{\lambda e}} \\ &\equiv \frac{D}{2\pi} \rho \int_{-\infty}^{\infty} \frac{\omega_{\lambda e} d\omega_{\lambda e}}{\omega - \omega_K - \omega_{\lambda e}}\end{aligned}\quad (2-42)$$

We substitute Equation (2-40) into Equation (2-33) and get

$$(\omega - \omega_j - \eta(\omega) + \frac{i}{2} \gamma(\omega)) B_i(\omega) = 1 \quad (2-43)$$

We obtain the amplitude $b_i(t)$ by evaluating the integral in Equation (2-28). This integral can be replaced by a contour integral, where the contour consists of the real axis and an infinite semi-circle in the lower half-plane. There is no contribution from the semi-circle since t is positive in Equation (2-28). We make use of the residue theorem which states that

$$\oint_C f(z) dz = 2\pi i \lim_{z \rightarrow z_0} (z - z_0) f(z)$$

where $f(z)$ is a function of the complex variable z with a simple pole at $z = z_0$ and \oint_C stands for an integral around the contour C in the positive sense (counterclockwise). The integral described above is such an integral except for the direction of the integration which is clockwise. This introduces a negative sign. We need to know the pole of $B_i(\omega)$. For optical frequencies ($\omega_{\lambda e} \approx 10^{15}$ Hz) the order of magnitude of the quantity D defined by Equation (2-41) is 10^{-10} , which means $D \ll 1$. The function $\eta(\omega)$ is essentially independent of ω within a reasonable range of ω -values and to a first approximation we can replace the functions $\gamma(\omega)$ and $\eta(\omega)$ in Equation (2-43) by $\gamma(\omega_j)$ and $\eta(\omega_j)$, respectively. Then we find that

$$B_i(\omega) = \frac{1}{\omega - \omega_j - \eta(\omega_j) + \frac{i}{2} \gamma(\omega_j)}$$

with a pole at $\omega = \omega_j + \eta(\omega_j) - \frac{i}{2}\gamma(\omega_j)$ and

$$b_i(t) = e^{-i\eta(\omega_j)t} e^{-\frac{\gamma(\omega_j)}{2}t} \quad (2-44)$$

The wave function is therefore

$$\begin{aligned} |\phi\rangle &= e^{-\frac{i}{\hbar}H_0t} \left\{ b_i(t)|i\rangle + \sum_{\ell} b_{f\ell}(t)|f\ell\rangle \right\} \\ &= e^{-i\omega_j t} e^{-i\eta(\omega_j)t} e^{-\frac{\gamma(\omega_j)}{2}t} |i\rangle + \sum_{\ell} e^{-i\omega_{f\ell}t} b_{f\ell}(t) |f\ell\rangle \end{aligned} \quad (2-45)$$

The state $|i\rangle$ oscillates with a frequency $\omega_j + \eta(\omega_j)$; we can interpret $\eta(\omega_j)$ as an energy shift of the upper atomic level. It represents the so-called self energy of that level which arises because the bound electron is accompanied by, and interacts with, a field of virtual photons. We define

$$S \equiv \eta(\omega_j) = \frac{D}{2\pi} P \int_{-\infty}^{\infty} \frac{\omega_{\lambda e} d\omega_{\lambda e}}{\omega_j - \omega_{\kappa} - \omega_{\lambda e}} \quad (2-46)$$

In our representation of the system $S \neq 0$.

The correct representation is one in which the virtual photon states are included in the unperturbed eigenstates and the self energies are included in the eigenvalues of H_0 . In such a representation $\eta(\omega)$ vanishes³.

Let us drop $\eta(\omega)$ in Equation (2-43). By using Equation (2-41) we can write

$$B_i(\omega) = \frac{1}{\omega(1 + \frac{i}{2}D) - \omega_j - \frac{i}{2}D\omega_{\kappa}} \quad (2-47)$$

The pole of the function $B_i(\omega)$ is at

$$\omega = \frac{(\omega_j + \frac{i}{2}D\omega_{\kappa})(1 - \frac{i}{2}D)}{1 + (\frac{D}{2})^2}$$

or, approximately, at

$$\omega = \omega_j - \frac{i}{2} D(\omega_j - \omega_k) \quad (2-48)$$

where terms of the order of D^2 have been neglected, and Equation (2-28) yields

$$b_i(t) = e^{-\frac{1}{2} D(\omega_j - \omega_k)} \stackrel{\text{Def.}}{=} e^{-\frac{1}{2} \gamma t} \quad (2-49)$$

The decay rate γ is defined by Equation (2-49) as

$$\gamma = D(\omega_j - \omega_k) \quad (2-50)$$

Incidentally, $D(\omega_j - \omega_k) = \gamma(\omega_j)$, which is another justification for replacing $\gamma(\omega)$ in Equation (2-43) by $\gamma(\omega_j)$.

2.2 Two-Level Atom Undergoing Spontaneous Decay: Time Dependence of the Probability of the Excited State

As before, we work in the interaction representation where

$$i\hbar \frac{\partial}{\partial t} |\Phi'\rangle = H_I' |\Phi'\rangle \quad (2-51)$$

$$-i\hbar \frac{\partial}{\partial t} \langle\Phi'| = \langle\Phi'| H_I'$$

The definition of $|\Phi'\rangle$ and H_I' are given by Equations (2-3) and (2-4), respectively. The density operator $S' = |\Phi'\rangle\langle\Phi'|$ obeys the differential equation

$$\begin{aligned} i\hbar \frac{\partial}{\partial t} S' &= i\hbar \left[\left(\frac{\partial}{\partial t} |\Phi'\rangle \right) \langle\Phi'| + |\Phi'\rangle \left(\frac{\partial}{\partial t} \langle\Phi'| \right) \right] \\ &= H_I' |\Phi'\rangle\langle\Phi'| - |\Phi'\rangle\langle\Phi'| H_I' = [H_I', S'] \end{aligned} \quad (2-52)$$

which is equivalent to

$$i\hbar \frac{\partial}{\partial t} [S'] = [H_I'] [S'] - [S'] [H_I'] \quad (2-53)$$

where $[S']$ is the density matrix with elements $[S']_{mn} = \langle m | S' | n \rangle = S_{mn}$ and $[H_I']$ stands for a matrix with elements $[H_I']_{mn} = \langle m | H_I' | n \rangle$.

The system under consideration has states $|i\rangle = |j\rangle|0\rangle$ and $|f_e\rangle = |k\rangle|1\rangle$, its density matrix is

$$[S'] = \begin{pmatrix} S_{ii} & S_{if_1} & S_{if_2} & \cdots \\ S_{fi_1} & & & \\ S_{fi_2} & & & \\ \vdots & & & \end{pmatrix} \quad (2-54)$$

and its interaction Hamiltonian is given by Equation (2-17) so that

$$[H_I'] = \hbar \begin{pmatrix} 0 & e^{i(\omega_i - \omega_{f_1})t} V_{if_1} & \times & \\ e^{-i(\omega_i - \omega_{f_1})t} V_{fi_1} & & & \\ e^{-i(\omega_i - \omega_{f_2})t} V_{fi_2} & & 0 & \\ \times & & & \\ \times & & & \end{pmatrix} \quad (2-55)$$

We substitute Equations (2-54) and (2-55) into Equation (2-53) and get

$$i\dot{S}_{ii} = \sum_{\ell} e^{i(\omega_i - \omega_{f_\ell})t} V_{if_\ell} S_{f_\ell i} - \sum_{\ell} S_{if_\ell} e^{-i(\omega_i - \omega_{f_\ell})t} V_{f_\ell i} + i\delta(t) \quad (2-56)$$

$$i\dot{S}_{if_e} = \sum_m e^{i(\omega_i - \omega_{f_m})t} V_{if_m} S_{f_m f_e} - S_{ii} e^{i(\omega_i - \omega_{f_e})t} V_{if_e} \quad (2-57)$$

$$i\dot{S}_{f_e i} = e^{-i(\omega_i - \omega_{f_e})t} V_{f_e i} S_{ii} - \sum_m S_{f_e f_m} e^{-i(\omega_i - \omega_{f_m})t} V_{f_m i} \quad (2-58)$$

$$i\dot{S}_{f_e f_m} = e^{-i(\omega_i - \omega_{f_e})t} V_{f_e i} S_{if_m} - S_{f_e i} e^{i(\omega_i - \omega_{f_m})t} V_{if_m} \quad (2-59)$$

where we added a term $i\delta(t)$ in Equation (2-56) for reasons similar

to the ones stated in Section 2.1. These equations are valid for

all times and contain the initial conditions $S_{ii}(0) = 1$,

$S_{if_e}(0) = S_{f_e i}(0) = S_{f_e f_m}(0) = 0 \quad \forall \ell, m$. Probability is put

into the system at $t = 0$ and all matrix elements are set to zero

for negative times. We transform

$$S_{mn}(t) = -\frac{1}{2\pi i} \int_{-\infty}^{\infty} e^{i(\omega_m - \omega_n)t} e^{-i\omega t} R_{mn}(\omega) d\omega \quad (2-60)$$

so that

$$S_{ii}(t) = -\frac{1}{2\pi i} \int_{-\infty}^{\infty} e^{-i\omega t} R_{ii}(\omega) d\omega \quad (2-61)$$

$$S_{ife}(t) = -\frac{1}{2\pi i} \int_{-\infty}^{\infty} e^{i(\omega_i - \omega_{fe})t} e^{-i\omega t} R_{ife}(\omega) d\omega \quad (2-62)$$

$$S_{fei}(t) = -\frac{1}{2\pi i} \int_{-\infty}^{\infty} e^{-i(\omega_i - \omega_{fe})t} e^{-i\omega t} R_{fei}(\omega) d\omega \quad (2-63)$$

$$S_{fefm}(t) = -\frac{1}{2\pi i} \int_{-\infty}^{\infty} e^{i(\omega_{fe} - \omega_{fm})t} e^{-i\omega t} R_{fefm}(\omega) d\omega \quad (2-64)$$

and let

$$i\delta(t) = -\frac{1}{2\pi i} \int_{-\infty}^{\infty} e^{-i\omega t} d\omega \quad (2-65)$$

We substitute Equations (2-61)-(2-65) into Equations (2-56)-(2-59)

and get

$$\omega R_{ii}(\omega) = \sum_l (V_{ife} R_{fei}(\omega) - R_{ife}(\omega) V_{fei}) + 1 \quad (2-66)$$

$$(\omega - \omega_i + \omega_{fe}) R_{ife}(\omega) = \sum_m V_{ifm} R_{fmfe}(\omega) - R_{ii}(\omega) V_{ife} \quad (2-67)$$

$$(\omega + \omega_i - \omega_{fe}) R_{fei}(\omega) = V_{fei} R_{ii}(\omega) - \sum_m R_{fefm}(\omega) V_{fmi} \quad (2-68)$$

$$(\omega + \omega_{fm} - \omega_{fe}) R_{fefm}(\omega) = V_{fei} R_{ifm}(\omega) - R_{fei}(\omega) V_{ifm} \quad (2-69)$$

We replace $R_{ife}(\omega)$ and $R_{fei}(\omega)$ in Equation (2-66) by the expressions obtained from Equations (2-67) and (2-68), respectively, and get

$$\begin{aligned} \omega R_{ii}(\omega) = & \sum_{\ell} \frac{V_{ife} V_{fei}}{\omega - \omega_{fe} + \omega_i} R_{ii}(\omega) - \sum_{\ell m} \frac{V_{ife} V_{fmi} R_{fefm}(\omega)}{\omega - \omega_{fe} + \omega_i} \\ & - \sum_{\ell m} \frac{V_{ifm} V_{fei} R_{fmfe}(\omega)}{\omega + \omega_{fe} - \omega_i} + \sum_{\ell} \frac{V_{ife} V_{fei}}{\omega + \omega_{fe} - \omega_i} R_{ii}(\omega) + 1 \quad (2-70) \end{aligned}$$

The single sums become, in complete analogy with previous calculations,

$$\sum_{\ell} \frac{V_{ife} V_{fei}}{\omega + \omega_j - \omega_k - \omega_{\lambda e}} = \eta_1(\omega) - \frac{i}{2} \gamma_1(\omega) \quad (2-71)$$

where

$$\gamma_1(\omega) = D(\omega + \omega_j - \omega_k) \quad (2-72)$$

$$\eta_1(\omega) = \frac{D}{2\pi} \rho \int_{-\infty}^{\infty} \frac{\omega_{\lambda e} d\omega_{\lambda e}}{\omega + \omega_j - \omega_k - \omega_{\lambda e}} \quad (2-73)$$

and

$$\sum_{\ell} \frac{V_{ife} V_{fei}}{\omega + \omega_k - \omega_j + \omega_{\lambda e}} = \eta_2(\omega) - \frac{i}{2} \gamma_2(\omega) \quad (2-74)$$

where

$$\gamma_2(\omega) = D(\omega_j - \omega_k - \omega) \quad (2-75)$$

$$\eta_2(\omega) = \frac{D}{2\pi} \rho \int_{-\infty}^{\infty} \frac{\omega_{\lambda e} d\omega_{\lambda e}}{\omega + \omega_k - \omega_j + \omega_{\lambda e}} \quad (2-76)$$

We assume for the moment that we can neglect the double sums in Equation (2-70). We then get

$$\omega R_{ii}(\omega) = [\eta_1(\omega) + \eta_2(\omega) - i D(\omega_j - \omega_k)] R_{ii}(\omega) + 1 \quad (2-77)$$

We can eliminate $\eta_1(\omega)$ and $\eta_2(\omega)$ from Equation (2-77) by noting that to a very good approximation $\eta_1(\omega)$ and $\eta_2(\omega)$ can be replaced by $\eta_1(0)$ and $\eta_2(0)$ and that

$$\eta_1(0) = -\eta_2(0) \quad (2-78)$$

Thus Equation (2-77) becomes

$$R_{ii}(\omega) = \frac{1}{\omega + i\gamma} \quad (2-79)$$

and by using Equation (2-61) we finally get

$$S_{ii}(t) = e^{-\gamma t} \quad (2-80)$$

We compare Equations (2-80) and (2-49) and find that

$$S_{ii}(t) = b_i^*(t) b_i(t) \quad (2-81)$$

From Equation (2-81) we conclude that the approximation made by neglecting the double sums in Equation (2-70) is of the same order as the approximation made by dropping the D^2 terms.

2.3 Two-Level Atom Undergoing Spontaneous Decay: Differential Equations for the Elements of the Reduced (Atomic) Density Matrix

We derive a set of differential equations for the elements of the atomic density matrix $[S_a']$ of a two-level atom undergoing spontaneous decay. The atom has the two eigenstates $|j\rangle$ and $|k\rangle$; $[S_a']$ consequently has four matrix elements: S_{jj} , S_{jk} , S_{kj} , S_{kk} . These are not all independent. We have

$$S_{jj} + S_{kk} = 1 \quad (2-82)$$

$$S_{kj} = S_{jk}^* \quad (2-83)$$

Calculations, which lead to the same result as the ones below are done by Mollow and Miller⁶. They consider a two-level atom coupled to a zero-temperature bath of harmonic oscillators. Such a zero-temperature bath can be assumed to always be in the ground state as far as its

effect on the atom is concerned (Markoff approximation). They derive a time development operator $U(\Delta t)$, which is expanded to second order in the coupling parameters which appear in the interaction Hamiltonian and apply $U(\Delta t)$ to $S'(t)$ to get $S'(t+\Delta t)$. Then they take traces over the states of the bath to obtain $S'_a(t)$ and $S'_a(t+\Delta t)$ and finally arrive at an expression for $\frac{S'_a(t+\Delta t) - S'_a(t)}{\Delta t}$, the "coarse-grained" time derivative of $S'_a(t)$, from which they are able to extract the following differential equations:

$$\dot{S}_{jj} = -\gamma S_{jj} \quad (2-84)$$

$$\dot{S}_{jk} = -\frac{\gamma}{2} S_{jk} \quad (2-85)$$

Compared to this approach our method is straightforward and the implications of the approximations we make are more transparent as well. We use the very general Equation (2-53), which holds for any quantum mechanical system and apply it to a suitable representation of the system of atom plus radiation field. This results in a set of differential equations for the elements of the density matrix of that system. We also derive the relations between the elements of the density matrix of the system and the elements of the reduced density matrix. With the help of these relations we arrive at the differential equations for the elements of the reduced density matrix.

First we note that the solutions of Equations (2-84) and (2-85) are

$$S_{jj}(t) = S_{jj}(0) e^{-\gamma t} \quad (2-86)$$

and

$$S_{jk}(t) = S_{jk}(0) e^{-\frac{\gamma}{2} t} \quad (2-87)$$

or, with the initial conditions $S_{jj}(0) = 1$ and $S_{jk}(0) = 0$

$$S_{jj}(t) = e^{-\gamma t} \quad (2-88)$$

and

$$S_{jk}(t) = 0 \quad \forall t \quad (2-89)$$

This should caution us against representing the system by states $|i\rangle = |j\rangle|0\rangle$ and $|f_e\rangle = |k\rangle|\lambda_e\rangle$ only, when trying to derive Equations (2-84) and (2-85). Let us pick such a representation (which implies that at some time t_0 all the probability is in state $|i\rangle$ and we have conditions as in Equations (2-88) and (2-89)). Then we expect to:

- a) get the equation $\dot{S}_{jj} = -\gamma S_{jj}$ and
- b) get the information that $S_{jk} = 0$.

We will now verify the claims made above. We write the most general wavefunction of the system as

$$|\Phi'\rangle = b_i |i\rangle + \sum_e b_{fe} |f_e\rangle \quad (2-90)$$

and calculate the density operator:

$$\begin{aligned} S' = |\Phi'\rangle\langle\Phi'| &= b_i b_i^* |i\rangle\langle i| + \sum_e b_i b_{fe}^* |i\rangle\langle f_e| \\ &+ \sum_e b_{fe} b_i^* |f_e\rangle\langle i| + \sum_{em} b_{fe} b_{fm}^* |f_e\rangle\langle f_m| \end{aligned} \quad (2-91)$$

We use the definition $[S']_{mn} = \langle m|S'|n\rangle = S_{mn}$ to rewrite Equation (2-91) as

$$\begin{aligned} S' &= S_{ii} |j\rangle|0\rangle\langle j| \langle 0| + \sum_e S_{ife} |j\rangle|0\rangle\langle k| \langle \lambda_e| \\ &+ \sum_e S_{fe i} |k\rangle|\lambda_e\rangle\langle j| \langle 0| + \sum_{em} S_{fefm} |k\rangle|\lambda_e\rangle\langle k| \langle \lambda_m| \end{aligned} \quad (2-92)$$

We "reduce" the density operator S' by taking the trace over the field

states; S_a' stands for the reduced (atomic) density operator. Then

$$S_a' = \text{tr}_{\text{field states}} S' = \langle 0 | S' | 0 \rangle + \sum_e \langle \lambda_e | S' | \lambda_e \rangle \quad (2-93)$$

and, with Equation (2-92)

$$S_a' = S_{ii} |j\rangle\langle j| + \sum_e S_{fefe} |k\rangle\langle k| \quad (2-94)$$

The matrix elements of $[S_a']$ are defined as follows:

$$S_{jj} = \langle j | S_a' | j \rangle \quad (2-95)$$

$$S_{jk} = \langle j | S_a' | k \rangle \quad (2-96)$$

$$S_{kj} = \langle k | S_a' | j \rangle \quad (2-97)$$

$$S_{kk} = \langle k | S_a' | k \rangle \quad (2-98)$$

We therefore find that in the case of the reduced density operator of Equation (2-94) we have

$$S_{jj} = S_{ii} \quad (2-99)$$

$$S_{jk} = 0 \quad (2-100)$$

$$S_{kj} = 0 \quad (2-101)$$

$$S_{kk} = \sum_e S_{fefe} \quad (2-102)$$

which proves the statement b) above.

In Section 2.2 we treat a system with states $|i\rangle$ and $|f_e\rangle$ and given initial conditions. Now we look at the same system without specifying the initial conditions, since we don't want a solution for $S_{ii}(t)$ but only a reduced differential equation. We therefore use Equations (2-56)-(2-59), but drop the term "+idf(t)" in Equation (2-56). This leads to Equations (2-66)-(2-69) but without the term "+I" in Equation (2-66). In a calculation analogous to the one above we obtain

$$\omega R_{ii}(\omega) = -i\tau R_{ii}(\omega) \quad (2-103)$$

We multiply Equation (2-103) by $-\frac{1}{2\pi i} e^{-i\omega t}$ and integrate over ω .

$$\begin{aligned} -\frac{1}{2\pi i} \int_{-\infty}^{\infty} \omega R_{ii}(\omega) e^{-i\omega t} d\omega &= \frac{i\tau}{2\pi i} \int_{-\infty}^{\infty} R_{ii}(\omega) e^{-i\omega t} d\omega \\ &= -\frac{1}{2\pi i} \int_{-\infty}^{\infty} i \frac{\partial}{\partial t} R_{ii}(\omega) e^{-i\omega t} d\omega \end{aligned} \quad (2-104)$$

We use Equation (2-61) and get

$$\dot{S}_{ii} = -\tau S_{ii} \quad (2-105)$$

or, because of Equation (2-99)

$$\dot{S}_{jj} = -\tau S_{jj} \quad (2-106)$$

thus proving the statement a) above.

We conclude that we have to pick the most general representation of the system in order to derive Equations (2-84) and (2-85) and take

$$|\Phi'\rangle = b_i |i\rangle + b_g |g\rangle + \sum_e b_{fe} |fe\rangle \quad (2-107)$$

where

$$\begin{aligned} |i\rangle &= |j\rangle |0\rangle \\ |g\rangle &= |K\rangle |0\rangle \\ |fe\rangle &= |K\rangle |2_e\rangle \end{aligned} \quad (2-108)$$

We use Equation (2-53) with

$$[S'] = \begin{pmatrix} S_{ii} & S_{ig} & S_{if_1} & S_{if_2} & \dots \\ S_{gi} & & & & \\ S_{fi} & & & & \\ \vdots & & & & \end{pmatrix} \quad (2-109)$$

and

$$[H_I'] = \hbar \begin{pmatrix} 0 & 0 & V_{if,i} e^{i(\omega_0 - \omega_{\lambda e})t} & \times & \times \\ 0 & & & & \\ V_{f,i} e^{-i(\omega_0 - \omega_{\lambda e})t} & & & & \\ \times & & & \sigma & \\ \times & & & & \end{pmatrix} \quad (2-110)$$

where $\omega_0 = \omega_j - \omega_k$ is the atomic frequency and find

$$i \dot{S}_{ii} = \sum_{\ell} e^{i(\omega_0 - \omega_{\lambda e})t} V_{if\ell} S_{f\ell i} - \sum_{\ell} e^{-i(\omega_0 - \omega_{\lambda e})t} V_{f\ell i} S_{if\ell} \quad (2-111)$$

$$i \dot{S}_{ig} = \sum_{\ell} e^{i(\omega_0 - \omega_{\lambda e})t} V_{if\ell} S_{f\ell g} \quad (2-112)$$

$$i \dot{S}_{ife} = \sum_m e^{i(\omega_0 - \omega_{\lambda m})t} V_{ifm} S_{fme} - S_{ii} e^{i(\omega_0 - \omega_{\lambda e})t} V_{ife} \quad (2-113)$$

$$i \dot{S}_{gi} = - \sum_{\ell} S_{gfe} e^{-i(\omega_0 - \omega_{\lambda e})t} V_{f\ell i} \quad (2-114)$$

$$i \dot{S}_{gg} = 0 \quad (2-115)$$

$$i \dot{S}_{gfe} = - S_{gi} e^{i(\omega_0 - \omega_{\lambda e})t} V_{ife} \quad (2-116)$$

$$i \dot{S}_{fei} = e^{-i(\omega_0 - \omega_{\lambda e})t} V_{f\ell i} S_{ii} - \sum_m S_{fem} e^{-i(\omega_0 - \omega_{\lambda m})t} V_{fmi} \quad (2-117)$$

$$i \dot{S}_{feg} = e^{-i(\omega_0 - \omega_{\lambda e})t} V_{f\ell i} S_{ig} \quad (2-118)$$

$$i \dot{S}_{fefm} = e^{-i(\omega_0 - \omega_{\lambda e})t} V_{f\ell i} S_{ifm} - S_{fei} e^{i(\omega_0 - \omega_{\lambda m})t} V_{ifm} \quad (2-119)$$

We Fourier transform the matrix elements using Equation (2-60) and find

$$\omega R_{ii} = \sum_{\ell} V_{i\ell e} R_{\ell e i} - \sum_{\ell} R_{i\ell e} V_{\ell e i} \quad (2-120)$$

$$(\omega - \omega_0) R_{ig} = \sum_{\ell} V_{i\ell e} R_{\ell e g} \quad (2-121)$$

$$(\omega - \omega_0 + \omega_{\lambda e}) R_{i\ell e} = \sum_m V_{i\ell m} R_{m\ell e} - R_{ii} V_{i\ell e} \quad (2-122)$$

$$(\omega + \omega_0) R_{gi} = - \sum_{\ell} R_{g\ell e} V_{\ell e i} \quad (2-123)$$

$$(\omega + \omega_{\lambda e}) R_{g\ell e} = - R_{gi} V_{i\ell e} \quad (2-124)$$

$$(\omega + \omega_0 - \omega_{\lambda e}) R_{\ell e i} = V_{\ell e i} R_{ii} - \sum_m R_{\ell e m} V_{m i} \quad (2-125)$$

$$(\omega - \omega_{\lambda e}) R_{\ell e g} = V_{\ell e i} R_{ig} \quad (2-126)$$

$$(\omega + \omega_{\lambda m} - \omega_{\lambda e}) R_{\ell e f m} = V_{\ell e i} R_{i f m} - R_{\ell e i} V_{i f m} \quad (2-127)$$

We replace $R_{i\ell e}(\omega)$ and $R_{\ell e i}(\omega)$ in Equation (2-120) by the expressions obtained from Equations (2-122) and (2-125), respectively, and following the procedure outlined above we again derive

$$\omega R_{ii}(\omega) = -i\gamma R_{ii}(\omega) \quad (2-128)$$

and

$$\dot{S}_{ii} = -\gamma S_{ii} \quad (2-129)$$

Next we replace $R_{\ell e g}(\omega)$ in Equation (2-121) by the expression obtained from Equation (2-126) and find

$$\begin{aligned} (\omega - \omega_0) R_{ig}(\omega) &= \sum_{\ell} \frac{V_{i\ell e} V_{\ell e i}}{\omega - \omega_{\lambda e}} R_{ig}(\omega) \\ &= (\tilde{\eta}(\omega) - \frac{i}{2} \tilde{\gamma}(\omega)) R_{ig}(\omega) \end{aligned} \quad (2-130)$$

where

$$\tilde{\eta}(\omega) = \frac{D}{2\pi} \rho \int_{-\infty}^{\infty} \frac{\omega_{\lambda e} d\omega_{\lambda e}}{\omega - \omega_{\lambda e}} \quad (2-131)$$

and

$$\tilde{\tau}(\omega) = D\omega \quad (2-132)$$

The pole of $R_{ig}(\omega)$ is approximately at $\omega = \omega_0$. We can therefore replace $\tilde{\eta}(\omega)$ in Equation (2-130) by

$$\tilde{\eta}(\omega_0) = \frac{D}{2\pi} P \int_{-\infty}^{\infty} \frac{\omega_{\lambda e} d\omega_{\lambda e}}{\omega_0 - \omega_{\lambda e}} = S$$

This turns out to be the self energy S again, which can be added to ω_j to give the renormalized ω_j . Similarly we can substitute $\tilde{\tau}(\omega_0)$ for $\tilde{\tau}(\omega)$ where $\tilde{\tau}(\omega_0) = D\omega_0 = \tau$. Thus Equation (2-130) becomes

$$\left[\omega - \underbrace{(\omega_j + S)}_{\omega_j} + \omega_K \right] R_{ig}(\omega) = -\frac{i}{2} \tau R_{ig}(\omega) \quad (2-133)$$

or

$$(\omega - \omega_0) R_{ig}(\omega) = -\frac{i}{2} \tau R_{ig}(\omega) \quad (2-134)$$

which leads to

$$\dot{S}_{ig} = -\frac{\tau}{2} S_{ig} \quad (2-135)$$

Similarly we get

$$(\omega + \omega_0) R_{gi}(\omega) = \left(\bar{\eta}(\omega) - \frac{i}{2} \bar{\tau}(\omega) \right) R_{gi}(\omega) \quad (2-136)$$

The pole of $R_{gi}(\omega)$ is at $\omega \approx -\omega_0$ and we can replace ω by $-\omega_0$ in $\bar{\eta}(\omega)$ and $\bar{\tau}(\omega)$. Then

$$\bar{\eta}(-\omega_0) = -S \quad (2-137)$$

and

$$\bar{\tau}(-\omega_0) = D\omega_0 = \tau \quad (2-138)$$

and we find

$$\left[\omega + \underbrace{(\omega_j + S)}_{\omega_j} - \omega_K \right] R_{gi}(\omega) = -\frac{i}{2} \tau R_{gi}(\omega) \quad (2-139)$$

or

$$(\omega + \omega_0) R_{gi}(\omega) = -\frac{i}{2} \tau R_{gi}(\omega) \quad (2-140)$$

which gives

$$\dot{S}_{gi} = -\frac{\gamma}{2} S_{gi} \quad (2-141)$$

The relations between the matrix elements of $[S'_a]$ and $[S']$

are

$$S_{jj} = S_{ii} \quad (2-142)$$

$$S_{jk} = S_{ig} \quad (2-143)$$

$$S_{kj} = S_{gi} \quad (2-144)$$

$$S_{kk} = S_{gg} + \sum_l S_{fele} \quad (2-145)$$

We use Equations (2-142), (2-143) and (2-144) to rewrite Equations (2-129), (2-135) and (2-141), respectively, as

$$\dot{S}_{jj} = -\gamma S_{jj} \quad (2-146)$$

$$\dot{S}_{jk} = -\frac{\gamma}{2} S_{jk} \quad (2-147)$$

$$\dot{S}_{kj} = -\frac{\gamma}{2} S_{kj} \quad (2-148)$$

From Equations (2-145) and (2-142) together with Equations (2-111), (2-115) and (2-119) we get

$$\dot{S}_{ii} + \dot{S}_{gg} + \sum_l \dot{S}_{fele} = \dot{S}_{jj} + \dot{S}_{kk} = 0 \quad (2-149)$$

2.4 Two-Level Atom in a Laser Beam: Differential Equations for the Elements of the Atomic Density Matrix

We are concerned with electric dipole transitions and describe the interaction between the atom and the laser beam by the Hamiltonian

$$H_2 = -\vec{d} \cdot \vec{E} \quad (2-150)$$

where $\vec{d} = e\vec{r}$ is the electric dipole moment of the atom. We write the electric field as a classical quantity, namely

$$\vec{E}(t) = E(t) \text{Re} \{ \hat{e} e^{-i\omega_L t} \} \quad (2-151)$$

where $E(t)$ is the (possibly time dependent) amplitude of the laser

beam, ω_L is its frequency and \hat{e} its polarization vector.

In the case of a left circularly polarized wave propagating in the z -direction we have

$$\hat{e} = \frac{1}{\sqrt{2}} (\hat{x} + i\hat{y}) \quad (2-152)$$

and Equation (2-150) becomes

$$\begin{aligned} H_2 &= -\frac{eE(t)}{2\sqrt{2}} \{ (r_x + ir_y) e^{-i\omega_L t} + (r_x - ir_y) e^{i\omega_L t} \} \\ &= \sqrt{\frac{4\pi}{3}} \frac{eE(t)}{2} \{ y_1^-(r) e^{-i\omega_L t} - y_1^+(r) e^{i\omega_L t} \} \end{aligned} \quad (2-153)$$

where

$$y_1^-(r) = \sqrt{\frac{3}{8\pi}} (r_x - ir_y) = r Y_1^-(\theta, \phi) \quad (2-154)$$

$$y_1^0(r) = \sqrt{\frac{3}{4\pi}} r_z = r Y_1^0(\theta, \phi) \quad (2-155)$$

and

$$y_1^+(r) = \sqrt{\frac{3}{8\pi}} (-r_x - ir_y) = r Y_1^+(\theta, \phi) \quad (2-156)$$

are the components of the solid spherical harmonic of first rank and

$Y_1^{\pm 1}(\theta, \phi)$ and $Y_1^0(\theta, \phi)$ are spherical harmonics.

We assume that the atomic states are states of good angular momentum. The atomic state $|j\rangle$ has angular momentum quantum numbers ℓ_j , m_j , and $|k\rangle$ has angular momentum quantum numbers ℓ_k, m_k , respectively.

The Wigner-Eckart theorem gives us some information about matrix elements involving states of good angular momentum (definite ℓ_1, m_1 and ℓ_2, m_2) and tensor operators T_ℓ^m , namely

$$\langle \ell_2 m_2 | T_\ell^m | \ell_1 m_1 \rangle = \langle \ell_1 \ell_2 m_1 m_2 | \ell_1 \ell_2 \ell_2 m_2 \rangle \langle \ell_2 || T_\ell || \ell_1 \rangle$$

where $\langle \ell_1 \ell_2 m_1 m_2 | \ell_1 \ell_2 \ell_2 m_2 \rangle$ is a Clebsch-Gordan coefficient which is zero unless $m_1 + m_2 = m_2$ and $|\ell_1 - \ell_2| \leq \ell_2 \leq \ell_1 + \ell_2$ and $\langle \ell_2 || T_\ell || \ell_1 \rangle$

is the reduced matrix element of the irreducible spherical tensor operator T_ℓ and does not depend on the magnetic quantum numbers. The set of the $2K+1$ operators $y_K^m = r^K Y_K^m$ is an irreducible spherical tensor operator of rank K . Thus the three operators y_1^{-1} , y_1^0 and y_1^1 form an irreducible spherical tensor operator of rank 1. Using the Wigner-Eckart theorem we find that

$$\begin{aligned} \langle j | y_1^{\pm 1} | j \rangle &= 0 \\ \langle k | y_1^{\pm 1} | k \rangle &= 0 \end{aligned} \quad (2-157)$$

and if

$$\langle j | y_1^1 | k \rangle \neq 0 \quad (2-158)$$

which implies $m_j - m_k = 1$ then

$$\langle j | y_1^{-1} | k \rangle = 0 \quad (2-159)$$

Thus

$$\langle j | H_2' | j \rangle = 0 \quad (2-160)$$

$$\langle k | H_2' | k \rangle = 0 \quad (2-161)$$

$$\langle j | H_2' | k \rangle = e^{i(\omega_0 - \omega_L)t} \hbar \frac{\Omega(t)}{2} \quad (2-162)$$

where

$$\hbar \frac{\Omega(t)}{2} = \sqrt{\frac{4\pi}{3}} \frac{e E(t)}{2} \langle j | y_1^1 | k \rangle \quad (2-163)$$

and

$$\langle k | H_2' | j \rangle = e^{-i(\omega_0 - \omega_L)t} \hbar \frac{\Omega^*(t)}{2} \quad (2-164)$$

where

$$\hbar \frac{\Omega^*(t)}{2} = \sqrt{\frac{4\pi}{3}} \frac{e E(t)}{2} \langle k | -y_1^{-1} | j \rangle \quad (2-165)$$

We use Equation (2-53) with

$$[S'] = [S_a'] = \begin{pmatrix} S_{jj} & S_{jk} \\ S_{kj} & S_{kk} \end{pmatrix} \quad (2-166)$$

and

$$[H_I'] = [H_2'] = \hbar \begin{pmatrix} 0 & e^{i(\omega_0 - \omega_L)t} \frac{\Omega(t)}{2} \\ e^{-i(\omega_0 - \omega_L)t} \frac{\Omega^*(t)}{2} & 0 \end{pmatrix} \quad (2-167)$$

and find

$$i \dot{S}_{jj} = \frac{1}{2} e^{i(\omega_0 - \omega_L)t} \Omega(t) S_{jk} - \frac{1}{2} e^{-i(\omega_0 - \omega_L)t} \Omega^*(t) S_{jk} \quad (2-168)$$

$$i \dot{S}_{jk} = \frac{1}{2} e^{i(\omega_0 - \omega_L)t} \Omega(t) [S_{kk} - S_{jj}] \quad (2-169)$$

$$i \dot{S}_{kj} = \frac{1}{2} e^{-i(\omega_0 - \omega_L)t} \Omega^*(t) [S_{jj} - S_{kk}] \quad (2-170)$$

$$i \dot{S}_{kk} = \frac{1}{2} e^{-i(\omega_0 - \omega_L)t} \Omega^*(t) S_{jk} - \frac{1}{2} e^{i(\omega_0 - \omega_L)t} \Omega(t) S_{jk} \quad (2-171)$$

Adding Equations (2-168) and (2-171) we get

$$\dot{S}_{jj} + \dot{S}_{kk} = 0$$

which implies that $S_{jj} + S_{kk} = 1$.

2.5 Two-Level Atom Interacting with a Laser Beam and Undergoing Spontaneous Decay: First Approximation

We consider a two-level atom interacting with a laser beam and with the radiation field. The interaction Hamiltonian is therefore

$$H_I = H_1 + H_2 \quad (2-172)$$

where H_1 is defined by Equation (2-17) and H_2 by Equation (2-150).

To a first approximation we describe the system by the wave function of Equation (2-107) and by the density matrix of Equation (2-109).

In Figure (2-1) we schematically show the states and interactions of the system.

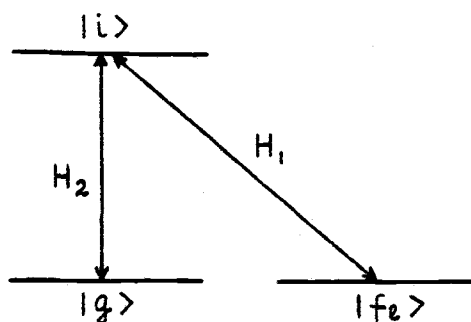


Figure (2-1). The states and interactions of the system in the first approximation.

We find

$$[H_I'] = \hbar \begin{pmatrix} 0 & \frac{\Omega}{2} e^{i(\omega_0 - \omega_1)t} & V_{if_1} e^{i(\omega_0 - \omega_2)t} & \times \dots \\ \frac{\Omega^*}{2} e^{-i(\omega_0 - \omega_1)t} & & & \\ V_{fi} e^{-i(\omega_0 - \omega_2)t} & & & \\ \times & & & \\ \vdots & & & \end{pmatrix} \quad (2-173)$$

and

$$i\dot{S}_{ii} = \Omega' S_{gi} + \sum_{\ell} V_{if_{\ell}}' S_{f_{\ell}i} - \Omega'^* S_{ig} - \sum_{\ell} V_{f_{\ell}i}' S_{ife} \quad (2-174)$$

$$i\dot{S}_{ig} = \Omega' S_{gg} + \sum_{\ell} V_{if_{\ell}}' S_{f_{\ell}g} - \Omega' S_{ii} \quad (2-175)$$

$$i\dot{S}_{ife} = \Omega' S_{gfe} + \sum_m V_{if_m}' S_{f_mfe} - V_{ife}' S_{ii} \quad (2-176)$$

$$i\dot{S}_{gi} = \Omega'^* S_{ii} - \Omega'^* S_{gg} - \sum_{\ell} V_{f_{\ell}i}' S_{gfe} \quad (2-177)$$

$$i\dot{S}_{gg} = \Omega'^* S_{ig} - \Omega' S_{gi} \quad (2-178)$$

$$i\dot{S}_{gfe} = \Omega'^* S_{ife} - V_{ife}' S_{gi} \quad (2-179)$$

$$i \dot{S}_{fei} = V'_{fei} S_{ii} - \Omega'^* S_{feg} - \sum_m V'_{fmi} S_{fefm} \quad (2-180)$$

$$i \dot{S}_{feg} = V'_{fei} S_{ig} - \Omega' S_{fei} \quad (2-181)$$

$$i \dot{S}_{fefm} = V'_{fei} S_{ifm} - V'_{ifm} S_{fei} \quad (2-182)$$

In Equations (2-174)-(2-182) we used the abbreviations

$$\begin{aligned} \Omega' &= \frac{\Omega(t)}{2} e^{i(\omega_0 - \omega_L)t} \\ V'_{ife} &= V_{ife} e^{i(\omega_0 - \omega_{Le})t} \\ V'_{fei} &= V_{fei} e^{-i(\omega_0 - \omega_{Le})t} \end{aligned} \quad (2-183)$$

These equations reduce to

$$i \dot{S}_{ii} = -i\gamma S_{ii} + \Omega' S_{gi} - \Omega'^* S_{ig} \quad (2-184)$$

$$i \dot{S}_{ig} = -\frac{i}{2}\gamma S_{ig} + \Omega' (S_{gg} - S_{ii}) \quad (2-185)$$

$$i \dot{S}_{gi} = -\frac{i}{2}\gamma S_{gi} + \Omega'^* (S_{ii} - S_{gg}) \quad (2-186)$$

$$i \dot{S}_{gg} = \Omega'^* S_{ig} - \Omega' S_{gi} \quad (2-187)$$

$$i \sum_{\ell} \dot{S}_{fefe} = i\gamma S_{ii} \quad (2-188)$$

or, when using Equations (2-142)-(2-145), to

$$i \dot{S}_{jj} = -i\gamma S_{jj} + \frac{\Omega(t)}{2} e^{i(\omega_0 - \omega_L)t} S_{kj} - \frac{\Omega^*(t)}{2} e^{-i(\omega_0 - \omega_L)t} S_{jk} \quad (2-189)$$

$$i \dot{S}_{jk} = -i\frac{\gamma}{2} S_{jk} + \frac{\Omega(t)}{2} e^{i(\omega_0 - \omega_L)t} (S_{kk} - S_{jj} - \sum_{\ell} S_{fefe}) \quad (2-190)$$

$$i \dot{S}_{kj} = -i\frac{\gamma}{2} S_{kj} + \frac{\Omega^*(t)}{2} e^{-i(\omega_0 - \omega_L)t} (S_{jj} - S_{kk} + \sum_{\ell} S_{fefe}) \quad (2-191)$$

$$i \dot{S}_{kk} = i\gamma S_{jj} + \frac{\Omega^*(t)}{2} e^{-i(\omega_0 - \omega_L)t} S_{jk} - \frac{\Omega(t)}{2} e^{i(\omega_0 - \omega_L)t} S_{kj} \quad (2-192)$$

These differential equations should reduce to Equations (2-146)-(2-149) for $\Omega = 0$, and Equations (2-168)-(2-171) for $\gamma = 0$. We set $\Omega = 0$ in Equations (2-189)-(2-192) and get a set of differential equations, which are identical with Equations (2-146)-(2-149). We set $\gamma = 0$ in Equations (2-189)-(2-192) and get

$$i \dot{S}_{jj} = \frac{\Omega(t)}{2} e^{i(\omega_0 - \omega_L)t} S_{kj} - \frac{\Omega^*(t)}{2} e^{-i(\omega_0 - \omega_L)t} S_{jk}$$

$$i \dot{S}_{jk} = \frac{\Omega(t)}{2} e^{i(\omega_0 - \omega_L)t} [S_{kk} - S_{jj} - \sum_l S_{fefe}]$$

$$i \dot{S}_{kj} = \frac{\Omega^*(t)}{2} e^{-i(\omega_0 - \omega_L)t} [S_{jj} - S_{kk} + \sum_l S_{fefe}]$$

$$i \dot{S}_{kk} = \frac{\Omega^*(t)}{2} e^{-i(\omega_0 - \omega_L)t} S_{jk} - \frac{\Omega(t)}{2} e^{i(\omega_0 - \omega_L)t} S_{kj}$$

These equations differ from Equations (2-168)-(2-171) by the term $\sum_l S_{fefe}$ in the equations for the off-diagonal matrix elements.

To sum up, the spontaneous decay is described correctly by Equations (2-189)-(2-192), the interaction with the laser beam is described incorrectly. The reason lies in our choice of states to represent the system, which makes it impossible to take into account that the atom can reach the excited state starting from states $|f_e\rangle$.

2.6 Two-Level Atom Interacting with a Laser Beam and Undergoing Spontaneous Decay: Second Approximation

We describe the system by the "improved" wavefunction

$$|\phi'\rangle = b_i |i\rangle + b_g |g\rangle + \sum_j b_{h_j} |h_j\rangle + \sum_i b_{f_i} |f_i\rangle \quad (2-193)$$

where

$$\begin{aligned}
 |i\rangle &= |j\rangle|0\rangle \\
 |q\rangle &= |k\rangle|0\rangle \\
 |h_j\rangle &= |j\rangle|\lambda_j\rangle \\
 |f_i\rangle &= |k\rangle|\lambda_i\rangle
 \end{aligned}
 \tag{2-194}$$

and by Figure (2-2).

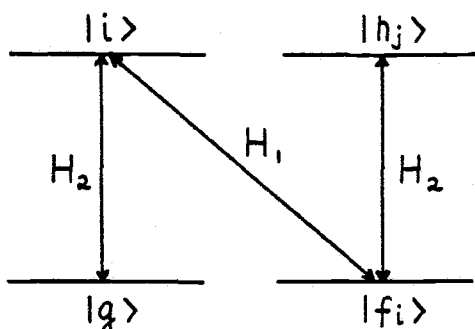


Figure (2-2). The states and interactions of the system in the second approximation.

The density matrix is

$$[S'] = \begin{pmatrix}
 \begin{matrix} S_{ii} \\ S_{gi} \\ S_{hi} \\ S_{fi} \\ \vdots \end{matrix} &
 \begin{matrix} S_{iq} \\ S_{gg} \\ S_{hg} \\ S_{fg} \\ \vdots \end{matrix} &
 \begin{matrix} S_{ih_i} \\ S_{gh_i} \\ S_{hh_i} \\ S_{fh_i} \\ \vdots \end{matrix} &
 \begin{matrix} S_{if_i} \\ S_{gf_i} \\ S_{hf_i} \\ S_{ff_i} \\ \vdots \end{matrix}
 \end{pmatrix}
 \tag{2-195}$$

The interaction Hamiltonian is $H_I = H_1 + H_2$ again and $[H_I']$ is given in Equation (2-196) on the next page. The sole purpose of the dotted lines appearing in the matrices of Equations (2-195) and (2-196) is to make their structure more apparent.

$$\frac{1}{\hbar} [H_I'] =$$

(2-196)

$$\begin{pmatrix} 0 & \frac{\Omega}{2} e^{i(\omega_0 - \omega_L)t} & 0 \dots \dots \dots 0 & V_{fi} e^{i(\omega_0 - \omega_L)t} \dots \dots \dots \\ \frac{\Omega^*}{2} e^{-i(\omega_0 - \omega_L)t} & 0 & \sigma^{**} & 0 \dots \dots \dots 0 \\ 0 & \vdots & \vdots & \frac{\Omega}{2} e^{i(\omega_0 - \omega_L)t} \\ \vdots & \sigma^* & \sigma & \begin{matrix} \times & & \sigma \\ & \times & \\ & & \times \\ & & & \times \\ \sigma & & & \times \end{matrix} \\ 0 & \vdots & \vdots & \vdots \\ V_{fi} e^{-i(\omega_0 - \omega_L)t} & 0 & \frac{\Omega^*}{2} e^{-i(\omega_0 - \omega_L)t} & \begin{matrix} \times & & \sigma \\ & \times & \\ & & \times \\ & & & \times \\ \sigma & & & \times \end{matrix} \\ \vdots & \vdots & \vdots & \sigma \\ \vdots & 0 & \vdots & \vdots \end{pmatrix}$$

The matrix elements appearing in slots * and ** are

$$\begin{aligned}
 \langle h_i | H_I' | g \rangle &= \langle j | \langle \lambda_i | e^{i(\omega_j + \omega_{\lambda_i} - \omega_k)t} H_I | k \rangle | 0 \rangle \\
 &= e^{i(\omega_0 + \omega_{\lambda_i})t} \langle j | -\frac{e}{m} \sqrt{\frac{\hbar}{2\omega_{\lambda_i}\epsilon_0}} \vec{p} \cdot \hat{e}_{\lambda_i}^* e^{-i\vec{k}_{\lambda_i} \cdot \vec{r}} | k \rangle \quad (2-197) \\
 &= \hbar V_{hiq} e^{i(\omega_0 + \omega_{\lambda_i})t}
 \end{aligned}$$

and

$$\langle g | H_I' | h_i \rangle = \hbar V_{hiq}^* e^{-i(\omega_0 + \omega_{\lambda_i})t} \quad (2-198)$$

respectively. We set them to zero because they oscillate in time much more rapidly than the other matrix elements. This approximation is similar to the rotating wave approximation, which is covered extensively in the literature⁷.

The relation between the matrix elements of $[S_a']$ and $[S']$ is

$$S_{jj} = S_{ii} + \sum_i S_{hi} h_i \quad (2-199)$$

$$S_{jk} = S_{ig} + \sum_i S_{hi} f_i \quad (2-200)$$

$$S_{kj} = S_{gi} + \sum_i S_{fi} h_i \quad (2-201)$$

$$S_{kk} = S_{gg} + \sum_i S_{fi} f_i \quad (2-202)$$

We use the method outlined above and find

$$i \dot{S}_{jj} = \frac{\Omega(t)}{2} e^{i(\omega_0 - \omega_i)t} S_{kj} - \frac{\Omega^*(t)}{2} e^{-i(\omega_0 - \omega_i)t} S_{jk} - i\tau (S_{jj} - \sum_i S_{hi} h_i) \quad (2-203)$$

$$i \dot{S}_{jk} = \frac{\Omega(t)}{2} e^{i(\omega_0 - \omega_i)t} (S_{kk} - S_{jj}) - \frac{i}{2} \tau (S_{jk} - \sum_i S_{hi} f_i) \quad (2-204)$$

$$i \dot{S}_{kj} = \frac{\Omega^*(t)}{2} e^{-i(\omega_0 - \omega_i)t} (S_{jj} - S_{kk}) - \frac{i}{2} \tau (S_{kj} - \sum_i S_{fi} h_i) \quad (2-205)$$

$$i \dot{S}_{kk} = -i \dot{S}_{jj} \quad (2-206)$$

To check these equations in the known limiting cases we set $\mathcal{T} = 0$ in Equations (2-203)-(2-206) and get a set of differential equations which are identical with Equations (2-168)-(2-171). Then we set $\Omega = 0$ in Equations (2-203)-(2-206) and get

$$\begin{aligned} i \dot{S}_{jj} &= -i \mathcal{T} (S_{jj} - \sum_i S_{h_i h_i}) \\ i \dot{S}_{jk} &= -\frac{i}{2} \mathcal{T} (S_{jk} - \sum_i S_{h_i f_i}) \\ i \dot{S}_{kj} &= -\frac{i}{2} \mathcal{T} (S_{kj} - \sum_i S_{f_i h_i}) \\ i \dot{S}_{kk} &= i \mathcal{T} S_{jj} \end{aligned}$$

These equations differ from Equations (2-146)-(2-149) by the term $\sum_i S_{h_i h_i}$ in the equations for S_{jj} and S_{kk} , the term $\sum_i S_{h_i f_i}$ in the equation for S_{jk} and the term $\sum_i S_{f_i h_i}$ in the equation for S_{kj} .

This time the interaction with the laser beam is described correctly and the spontaneous decay is described incorrectly. The reason for this is that with our choice of states we cannot take the spontaneous decay of states $|h_j\rangle$ into account.

For the n^{th} approximation we expect the following:

For even n the spontaneous decay will be described incorrectly;
for odd n the interaction with the laser beam will be described incorrectly.

The deficiencies of the two kinds of approximations are illustrated by Figures (2-3) and (2-4). In Figure (2-3) the spontaneous decay is cut off, which results in an inaccurate spontaneous decay term. In Figure (2-4) the laser interaction is cut off, which results in an incorrect description of the interaction with the laser beam.

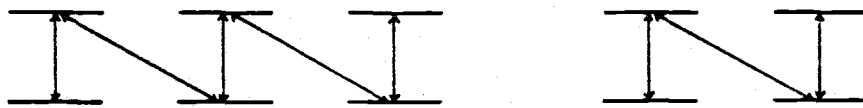


Figure (2-3). Diagram of the states and interactions for even n .

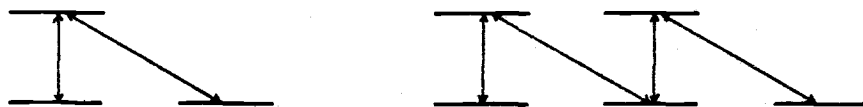


Figure (2-4). Diagram of the states and interactions for odd n .

From the above calculations it follows that

$$i\dot{S}_{jj} = -i\gamma S_{jj} + \frac{\Omega(t)}{2} e^{i(\omega_0 - \omega_L)t} S_{kj} - \frac{\Omega^*(t)}{2} e^{-i(\omega_0 - \omega_L)t} S_{jk} \quad (2-207)$$

$$i\dot{S}_{jk} = -\frac{i}{2}\gamma S_{jk} + \frac{\Omega(t)}{2} e^{i(\omega_0 - \omega_L)t} (S_{kk} - S_{jj}) \quad (2-208)$$

$$i\dot{S}_{kj} = -\frac{i}{2}\gamma S_{kj} + \frac{\Omega^*(t)}{2} e^{-i(\omega_0 - \omega_L)t} (S_{jj} - S_{kk}) \quad (2-209)$$

$$i\dot{S}_{kk} = -i\dot{S}_{jj} \quad (2-210)$$

are the correct differential equations. Mollow and Miller⁶ derive these equations for the special case of a resonant laser beam, where the atomic frequency ω_0 and the laser frequency ω_L are equal.

3. THE OPTICAL BLOCH EQUATIONS AND THEIR SOLUTIONS

3.1 The Optical Bloch Equations for a Two-Level Atom Interacting with a Laser Beam and Undergoing Spontaneous Decay

The optical Bloch equations are a set of differential equations which describe the time dependence of the expectation values of the Pauli spin matrices

$$\tilde{\sigma}_x = \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix} \quad \tilde{\sigma}_y = \begin{pmatrix} 0 & -i \\ i & 0 \end{pmatrix} \quad \tilde{\sigma}_z = \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix} \quad (3-1)$$

in a frame which rotates with the electric field vector of the laser beam. In our case this frame is one which rotates with frequency ω_L around the z -axis (see Equations (2-151) and (2-152)).

We can derive the Bloch equations from Equations (2-207)-(2-210) after establishing the connections between the matrix elements of $[S_a']$ and the expectation values of $\tilde{\sigma}_x$, $\tilde{\sigma}_y$, and $\tilde{\sigma}_z$. We know that $[S_a'] = \begin{pmatrix} S_{jj} & S_{jk} \\ S_{kj} & S_{kk} \end{pmatrix}$ is the atomic density matrix in the interaction representation. Since expectation values are independent of the representation we can write

$$\langle \tilde{\sigma}_i \rangle = \text{tr} [S_a] \tilde{\sigma}_i = \text{tr} [S_a'] \tilde{\sigma}_i' \quad i = x, y, z \quad (3-2)$$

where $[S_a]$ is the atomic density matrix in the Schrödinger representation and

$$\tilde{\sigma}_i' = e^{\frac{i}{\hbar} H_0 t} \tilde{\sigma}_i e^{-\frac{i}{\hbar} H_0 t} \quad (3-3)$$

are the Pauli spin matrices in the interaction representation. Using Equations (3-1) and (3-3) we find

$$\sigma_x' = \begin{pmatrix} 0 & e^{i\omega_0 t} \\ e^{-i\omega_0 t} & 0 \end{pmatrix} \quad (3-4)$$

$$\sigma_y' = \begin{pmatrix} 0 & -ie^{i\omega_0 t} \\ ie^{-i\omega_0 t} & 0 \end{pmatrix} \quad (3-5)$$

$$\sigma_z' = \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix} \quad (3-6)$$

and using Equations (3-2) and (3-4)-(3-6) we get

$$x_0 \equiv \langle \sigma_x \rangle = \text{tr} [\rho_a'] \sigma_x' = \rho_{jk} e^{-i\omega_0 t} + \rho_{kj} e^{i\omega_0 t} \quad (3-7)$$

$$y_0 \equiv \langle \sigma_y \rangle = \text{tr} [\rho_a'] \sigma_y' = i \rho_{jk} e^{-i\omega_0 t} - i \rho_{kj} e^{i\omega_0 t} \quad (3-8)$$

$$z_0 \equiv \langle \sigma_z \rangle = \text{tr} [\rho_a'] \sigma_z' = \rho_{jj} - \rho_{kk} \quad (3-9)$$

We want to observe the vector $\begin{pmatrix} x_0 \\ y_0 \\ z_0 \end{pmatrix}$ in a frame which, in a time t , rotates around the z -axis by an angle $\Phi_1 = \omega_L t$. Our observations are equivalent to the ones made in the lab frame of a vector being rotated by an angle $\Phi_2 = -\omega_L t$ around the same axis. This vector is

$$\begin{aligned} \begin{pmatrix} x \\ y \\ z \end{pmatrix} &= \begin{pmatrix} \cos \omega_L t & \sin \omega_L t & 0 \\ -\sin \omega_L t & \cos \omega_L t & 0 \\ 0 & 0 & 1 \end{pmatrix} \begin{pmatrix} x_0 \\ y_0 \\ z_0 \end{pmatrix} \\ &= \begin{pmatrix} x_0 \cos \omega_L t + y_0 \sin \omega_L t \\ -x_0 \sin \omega_L t + y_0 \cos \omega_L t \\ z_0 \end{pmatrix} \end{aligned} \quad (3-10)$$

We now calculate the replacements we have to make for ρ_{jj} , ρ_{jk} , ρ_{kj} , and ρ_{kk} in Equations (2-207)-(2-210) to obtain the corresponding equa-

tions for x , y and z . From Equations (3-7)-(3-9) we obtain

$$S_{jk} \rightarrow e^{i\omega_0 t} \frac{x_0 - iy_0}{2} \quad (3-11)$$

$$S_{kj} \rightarrow e^{-i\omega_0 t} \frac{x_0 + iy_0}{2} \quad (3-12)$$

$$S_{jj} - S_{kk} \rightarrow z_0 \quad (3-13)$$

and from Equation (3-10) we derive that

$$x_0 = x \cos \omega_L t - y \sin \omega_L t \quad (3-14)$$

$$y_0 = x \sin \omega_L t + y \cos \omega_L t \quad (3-15)$$

$$z_0 = z \quad (3-16)$$

so that

$$S_{jk} \rightarrow e^{i(\omega_0 - \omega_L)t} \frac{x - iy}{2} \quad (3-17)$$

$$S_{kj} \rightarrow e^{-i(\omega_0 - \omega_L)t} \frac{x + iy}{2} \quad (3-18)$$

$$S_{jj} - S_{kk} \rightarrow z \quad (3-19)$$

We substitute Equations (3-17)-(3-19) into Equations (2-207)-(2-210)

and, after some manipulations, find that

$$\dot{x} = -\frac{\gamma}{2} x - (\omega_0 - \omega_L) y - \frac{\Omega - \Omega^*}{2i} z \quad (3-20)$$

$$\dot{y} = -\frac{\gamma}{2} y + (\omega_0 - \omega_L) x - \frac{\Omega + \Omega^*}{2} z \quad (3-21)$$

$$\dot{z} = -\gamma(z + 1) + \frac{\Omega}{2}(y - ix) + \frac{\Omega^*}{2}(y + ix) \quad (3-22)$$

The quantity $\Omega(t)$ is defined by Equation (2-163). We can write

$$\frac{\hbar}{2} \Omega(t) = -\sqrt{\frac{4\pi}{3}} \frac{eE(t)}{2} \langle j | y_i^- - y_i^+ | k \rangle \quad (3-23)$$

since by Equation (2-159) $\langle j | Y_1^{-1} | k \rangle = 0$. The components of the solid spherical harmonic of rank 1 are given by Equations (2-154)-(2-156) and we find

$$r_x = \sqrt{\frac{4\pi}{3}} \frac{1}{\sqrt{2}} (Y_1^{-1} - Y_1^1) \quad (3-24)$$

$$r_y = i \sqrt{\frac{4\pi}{3}} \frac{1}{\sqrt{2}} (Y_1^{-1} + Y_1^1) \quad (3-25)$$

$$r_z = \sqrt{\frac{4\pi}{3}} Y_1^0 \quad (3-26)$$

The quantity $E(t)$ is the (real) amplitude of the electric field of the laser beam and $|j\rangle$ and $|k\rangle$ are eigenstates of good angular momentum. We can write these states in spherical polar coordinates as

$$|j\rangle = e^{i\varphi_j} R_{n_j \ell_j}(r) Y_{\ell_j}^{m_j}(\theta, \phi) \quad (3-27)$$

and

$$|k\rangle = e^{i\varphi_k} R_{n_k \ell_k}(r) Y_{\ell_k}^{m_k}(\theta, \phi) \quad (3-28)$$

where the $R_{n\ell}(r)$ are the (real) radial parts of the wave functions and the spherical harmonics $Y_{\ell}^m(\theta, \phi)$ are eigenstates of the angular momentum operator \vec{L} with

$$Y_{\ell}^m(\theta, \phi) = \sqrt{\frac{(2\ell+1)(\ell-m)!}{4\pi(\ell+m)!}} (-1)^m e^{im\phi} P_{\ell}^m(\cos\theta) \quad (3-29)$$

for $0 \leq m \leq \ell$

and

$$Y_{\ell}^m(\theta, \phi) = (-1)^m [Y_{\ell}^{-m}(\theta, \phi)]^* \quad (3-30)$$

The associated Legendre functions $P_{\ell}^m(\cos\theta)$ are real and thus $e^{im_j\phi}$ and $e^{im_k\phi}$ are the only complex quantities in $|j\rangle$ and $|k\rangle$ aside from the arbitrary phase factors $e^{i\varphi_j}$ and $e^{i\varphi_k}$. Using Equations (3-24), (3-29) and (3-30) we find

$$\frac{\hbar}{2} \Omega(t) = -\frac{eE(t)}{\sqrt{2}} \int_0^\infty \int_0^{2\pi} \int_0^{2\pi} e^{-iJ_j} R_{n_j l_j}(r) [Y_{l_j}^{m_j}(\theta, \phi)]^* r \sin \theta \cos \phi \cdot \\ \cdot e^{iJ_k} R_{n_k l_k}(r) Y_{l_k}^{m_k}(\theta, \phi) r^2 dr \sin \theta d\theta d\phi \quad (3-31)$$

The integrals over the variables r and θ are real numbers. Thus

$$\frac{\hbar}{2} \Omega(t) = E(t) R_0 \int_0^{2\pi} e^{i(m_k - m_j)\phi} \cos \phi d\phi e^{i(J_k - J_j)} \quad (3-32)$$

where R_0 is a real constant. From Equation (2-158) we know that

$m_j - m_k = 1$. Thus the integral over ϕ is real and we can adjust the arbitrary phases of the states so that $\Omega(t)$ becomes real.

We let $\Omega^*(t) = \Omega(t)$ in Equations (3-20)-(3-22) and obtain a simplified set of equations, namely

$$\dot{x} = -\frac{\gamma}{2} x - (\omega_0 - \omega_L) y \quad (3-33)$$

$$\dot{y} = -\frac{\gamma}{2} y + (\omega_0 - \omega_L) x - \Omega(t) z \quad (3-34)$$

$$\dot{z} = -\gamma(z+1) + \Omega(t) y \quad (3-35)$$

These are the optical Bloch equations for a two-level atom in a laser beam. The decay rate of the upper level is γ , $\Omega(t)$ is called the Rabi frequency. At resonance ($\omega_0 = \omega_L$) Equations (3-33)-(3-35) simplify considerably. They become

$$\dot{x} = -\frac{\gamma}{2} x \quad (3-36)$$

$$\dot{y} = -\frac{\gamma}{2} y - \Omega(t) z \quad (3-37)$$

$$\dot{z} = -\gamma(z+1) + \Omega(t) y \quad (3-38)$$

where the first equation is now uncoupled from the others.

3.2 Interpretation of the Quantities x , y and z

The original Bloch equations were phenomenological differential equations which describe the time dependence of the spin of a nucleus which is under the influence of a magnetic field $\vec{H}(t) = \vec{H}_0 + \vec{H}_1(t)$ where \vec{H}_0 is a strong, static field in the z -direction and $\vec{H}_1(t)$ is a weak, transverse field of arbitrary time dependence. These equations were first proposed by Bloch⁸. In later calculations, which are reminiscent of the approach taken by Mollow and Miller in deriving the optical Bloch equations, they were shown to be rigorously true for spin $\frac{1}{2}$ nuclei⁹.

The original Bloch equations are very similar to the optical Bloch equations, but instead of the factors γ and $\frac{\gamma}{2}$ they have two unrelated constants $\frac{1}{T_1}$ and $\frac{1}{T_2}$, where T_1 and T_2 are called the longitudinal (parallel to the static field) and transverse relaxation times respectively. This similarity is not surprising since formally a spin $\frac{1}{2}$ system in a static magnetic field and a two-level atom can both be described by operators which are 2×2 matrices and wave functions which are spinors.

In the case of a spin $\frac{1}{2}$ system, the operators σ_x , σ_y and σ_z represent the three components of the spin of the system, so that the vector $\begin{pmatrix} x \\ y \\ z \end{pmatrix}$ is the spin vector in the frame rotating with frequency ω_L around the z -axis. The physical significance of x , y and z in the case of a two-level atom is less transparent.

We use Equations (3-16) and (3-9) and find that

$$Z = Z_0 = S_{jj} - S_{kk} \quad (3-39)$$

where S_{jj} and S_{kk} are the probabilities of states $|j\rangle$ and $|k\rangle$ respectively. The quantity Z is called the inversion; it is a measure of the difference in population of the upper and lower atomic levels.

For an interpretation of X and Y we have to compare the electric field of the laser beam with the electric dipole moment of the atom.

We can write the electric field of the laser beam as

$$\begin{aligned} \vec{E}(t) &= \frac{E(t)}{2} \{ \hat{e} e^{-i\omega_L t} + \hat{e}^* e^{i\omega_L t} \} \\ &= \frac{E(t)}{\sqrt{2}} \{ \hat{x} \cos \omega_L t + \hat{y} \sin \omega_L t \} \end{aligned} \quad (3-40)$$

We derive a matrix $[\vec{d}']$ which represents the atomic dipole moment operator $\vec{d} = e\vec{r}$ in the interaction representation. Using Equations (3-24)-(3-26), (2-157)-(2-159) and the relations

$$\langle j | y_0 | k \rangle = 0$$

$$\langle j | y_1 | k \rangle = A$$

and

$$\langle j | y_1 | k \rangle^* = -\langle k | y_1^\dagger | j \rangle = A^*$$

where $A^* = A$ since we assume that $\Omega(t) = \Omega^*(t)$, we find that

$$\vec{d}' = e \sqrt{\frac{4\pi}{3}} \frac{1}{\sqrt{2}} e^{\frac{i}{\hbar} H_0 t} (y_1^\dagger - y_1, i y_1^\dagger + i y_1, \sqrt{2} y_0) e^{-\frac{i}{\hbar} H_0 t} \quad (3-41)$$

and

$$\begin{aligned} [\vec{d}'] &= -\hat{x} e \sqrt{\frac{4\pi}{3}} \frac{1}{\sqrt{2}} A \begin{pmatrix} 0 & e^{i\omega_0 t} \\ e^{-i\omega_0 t} & 0 \end{pmatrix} - \hat{y} e \sqrt{\frac{4\pi}{3}} \frac{1}{\sqrt{2}} A \begin{pmatrix} 0 & -ie^{i\omega_0 t} \\ ie^{-i\omega_0 t} & 0 \end{pmatrix} \\ &= \hat{x} d_0 \sigma_x' + \hat{y} d_0 \sigma_y' \end{aligned} \quad (3-42)$$

where

$$d_0 = -e \sqrt{\frac{4\pi}{3}} \frac{1}{\sqrt{2}} A \quad (3-43)$$

The expectation value of the dipole moment operator is the atomic dipole moment

$$\begin{aligned} \vec{D} &= \langle \vec{d} \rangle = \text{tr} [S_a'] [\vec{d}'] \\ &= \hat{x} d_0 \text{tr} [S_a'] \sigma_x' + \hat{y} d_0 \text{tr} [S_a'] \sigma_y' \\ &= \hat{x} d_0 \langle \sigma_x \rangle + \hat{y} d_0 \langle \sigma_y \rangle \end{aligned} \quad (3-44)$$

where

$$\langle \sigma_x \rangle = x_0$$

and

$$\langle \sigma_y \rangle = y_0$$

Using Equations (3-14) and (3-15) we find that

$$\begin{aligned} \vec{D} &= d_0 [x (\hat{x} \cos \omega_L t + \hat{y} \sin \omega_L t) \\ &\quad - y (\hat{x} \sin \omega_L t - \hat{y} \cos \omega_L t)] \\ &= d_0 [x (\hat{x} \cos \omega_L t + \hat{y} \sin \omega_L t) \\ &\quad - y (\hat{x} \cos(\omega_L t - \frac{\pi}{2}) + \hat{y} \sin(\omega_L t - \frac{\pi}{2}))] \end{aligned} \quad (3-45)$$

We compare Equation (3-45) with Equation (3-40) and see that x is the amplitude of that part of the electric dipole moment which is in phase with the laser beam, and y is the amplitude of the part which is $\frac{\pi}{2}$ out of phase.

3.3 The Time Dependence of the Rabi Frequency $\Omega(t)$

We consider a two-level atom which is exactly at resonance with a left circularly polarized laser beam so that Equations (3-36)-(3-38) hold. The Rabi frequency $\Omega(t)$ in these equations is proportional to the amplitude $E(t)$ of the electric field of the laser beam. We treat here the case of a sinusoidally modulated amplitude which can be written as

$$E(t) = E_0 + E_1 \cos \omega_1 t = E_0 (1 + a \cos \omega_1 t) \quad (3-46)$$

so that the Rabi frequency becomes

$$\Omega(t) = \Omega_0 + \Omega_1 \cos \omega_1 t = \Omega_0 (1 + a \cos \omega_1 t) \quad (3-47)$$

where Ω_0 is the average Rabi frequency, ω_1 is the modulation frequency and a is the modulation depth which is limited to $0 \leq a \leq 1$.

For $a = 0$ the amplitude and the Rabi frequency are constant in time. The spectral distribution of the light scattered by a two-level atom in an intense laser beam of constant amplitude is treated extensively in the literature in both theoretical^{10,11,12,13,14,15,16} and experimental^{17,18,19,20} papers.

The spectral distribution of the light scattered by a two-level atom in an intense, resonant and amplitude modulated laser beam has not been previously calculated. To determine this spectral distribution we need the solutions of both the optical Bloch equations

$$\dot{x} = -\frac{\gamma}{2} x \quad (3-48)$$

$$\dot{y} = -\frac{\gamma}{2} y - \Omega_0 z - \Omega_1 \cos \omega_1 t z \quad (3-49)$$

$$\dot{z} = -\gamma(z+1) + \Omega_0 y + \Omega_1 \cos \omega_1 t y \quad (3-50)$$

and their associated homogeneous equations

$$\dot{x} = -\frac{\gamma}{2} x \quad (3-51)$$

$$\dot{y} = -\frac{\gamma}{2} y - \Omega_0 z - \Omega_1 \cos \omega_1 t z \quad (3-52)$$

$$\dot{z} = -\gamma z + \Omega_0 y + \Omega_1 \cos \omega_1 t y \quad (3-53)$$

3.4 Solution of the Associated Homogeneous Equations

Any set of n linear first order differential equations of the form

$$\dot{V}_1 = m_{11}(t) V_1 + m_{12}(t) V_2 + \dots + m_{1n}(t) V_n$$

$$\vdots$$

$$\dot{V}_n = m_{n1}(t) V_1 + m_{n2}(t) V_2 + \dots + m_{nn}(t) V_n$$

can be written in matrix notation as $\dot{\vec{V}} = M(t) \vec{V}$

where $\vec{V} = \begin{pmatrix} V_1 \\ \vdots \\ V_n \end{pmatrix}$ and $M(t) = \begin{pmatrix} m_{11}(t) & m_{12}(t) & \dots & m_{1n}(t) \\ \vdots & \vdots & \ddots & \vdots \\ m_{n1}(t) & m_{n2}(t) & \dots & m_{nn}(t) \end{pmatrix}$

We can form an $n \times n$ matrix whose columns are solutions of $\dot{\vec{V}} = M(t) \vec{V}$.

Such a matrix is called a solution matrix²¹. A solution matrix $\Phi(t)$

whose columns are linearly independent is called a fundamental matrix

of $\dot{\vec{V}} = M(t) \vec{V}$ and has a determinant, which is non-zero for all times,

$\det \Phi(t) \neq 0 \quad \forall t$. We can write every solution $\vec{\psi}$ of

$\dot{\vec{V}} = M \vec{V}$ as

$$\vec{\psi} = \Phi(t) \vec{c}$$

where

$$\vec{C} = \begin{pmatrix} C_1 \\ C_2 \\ \vdots \\ C_n \end{pmatrix}$$

with the constants C_1, C_2, \dots, C_n determined by the initial conditions.

We assume that we need to solve a system of n linear first order differential equations of the form

$$\dot{\vec{V}} = (M_1 + M_2)\vec{V} \quad (3-54)$$

and that we already know the fundamental matrix Φ_0 of the system of equations

$$\dot{\vec{V}} = M_1 \vec{V} \quad (3-55)$$

The solution of $\dot{\vec{V}} = M_1 \vec{V}$ can then be written as

$$\vec{\Psi}_0 = \Phi_0 \vec{C} \quad (3-56)$$

and obeys the differential equation

$$\dot{\vec{\Psi}}_0 = M_1 \vec{\Psi}_0 \quad (3-57)$$

We differentiate Equation (3-56) and make use of Equation (3-57). We find that

$$\dot{\Phi}_0 \vec{C} = M_1 \Phi_0 \vec{C} \quad (3-58)$$

We now assume that the solution of $\dot{\vec{V}} = (M_1 + M_2)\vec{V}$ is of the form

$$\vec{\Psi} = \Phi_0 \vec{C}(t) \quad (3-59)$$

where the constant vector \vec{C} of Equation (3-56) has been replaced by a time dependent vector $\vec{C}(t)$. We differentiate Equation (3-59) and find that

$$\dot{\vec{\Psi}} = \dot{\Phi}_0 \vec{C}(t) + \Phi_0 \dot{\vec{C}}(t) = (M_1 + M_2) \Phi_0 \vec{C}(t)$$

We use Equation (3-58) and obtain the relation

$$\Phi_0 \dot{\vec{C}}(t) = M_2 \Phi_0 \vec{C}(t) \quad (3-60)$$

Since Φ_0 is a fundamental matrix and thus $\det \Phi_0 \neq 0$, an inverse Φ_0^{-1} exists so that $\Phi_0 \Phi_0^{-1} = \Phi_0^{-1} \Phi_0 = 1$ where 1 is the $n \times n$ unit matrix. We multiply Equation (3-60) from the left by Φ_0^{-1} and arrive at the following differential equation for $\vec{C}(t)$:

$$\dot{\vec{C}}(t) = \Phi_0^{-1} M_2 \Phi_0 \vec{C}(t) \quad (3-61)$$

We now solve Equations (3-51)-(3-53) with the "known" initial conditions $x(t')$, $y(t')$ and $z(t')$. The solution of Equation (3-51) is

$$x(t, t') = x(t') e^{-\frac{\tau}{2}(t-t')} \quad (3-62)$$

The solutions of Equations (3-52) and (3-53) are obtained in the two step process outlined above. We write Equations (3-52) and (3-53) in the form of Equation (3-54) where

$$\vec{V} = \begin{pmatrix} y \\ z \end{pmatrix} \quad (3-63)$$

$$M_1 = \begin{pmatrix} -\frac{\tau}{2} & -\Omega_0 \\ \Omega_0 & -\tau \end{pmatrix} \quad (3-64)$$

and

$$M_2 = \Omega_1 \cos \omega_1 t \begin{pmatrix} 0 & -1 \\ 1 & 0 \end{pmatrix} \quad (3-65)$$

The matrix

$$\Phi_0 = \begin{pmatrix} e^{r+t} & e^{r-t} \\ A_- e^{r+t} & A_+ e^{r-t} \end{pmatrix} \quad (3-66)$$

with

$$r_{\pm} = -\frac{3}{4}\gamma \pm i\Omega_0 \sqrt{1 - \left(\frac{\gamma}{4\Omega_0}\right)^2} \quad (3-67)$$

and

$$A_{\pm} = \frac{\gamma}{4\Omega_0} \pm i \sqrt{1 - \left(\frac{\gamma}{4\Omega_0}\right)^2} \quad (3-68)$$

is a solution matrix of $\dot{\vec{V}} = M_1 \vec{V}$ and a fundamental matrix since

$$\begin{aligned} \det \Phi_0 &= e^{(r_+ + r_-)t} (A_+ - A_-) \\ &= e^{-\frac{3}{2}\gamma t} 2i \sqrt{1 - \left(\frac{\gamma}{4\Omega_0}\right)^2} \\ &\neq 0 \quad \forall t \end{aligned}$$

The inverse of Φ_0 is

$$\Phi_0^{-1} = \frac{1}{A_+ - A_-} \begin{pmatrix} A_+ e^{-r_+ t} & -e^{-r_+ t} \\ -A_- e^{-r_- t} & e^{-r_- t} \end{pmatrix} \quad (3-69)$$

The matrix $\Phi_0^{-1} M_2 \Phi_0$ can be calculated by using Equations (3-65), (3-66) and (3-69). We find that

$$\Phi_0^{-1} M_2 \Phi_0 = \frac{\Omega_0 \cos \omega_1 t}{A_+ - A_-} \begin{pmatrix} -1 - A_+ A_- & (1 + A_+^2) e^{-(r_+ - r_-)t} \\ (1 + A_-^2) e^{(r_+ - r_-)t} & 1 + A_+ A_- \end{pmatrix} \quad (3-70)$$

We have

$$A_+ A_- = 1$$

$$A_+ - A_- = 2i \sqrt{1 - s^2}$$

$$A_{\pm}^2 = 2s^2 \pm 2is \sqrt{1 - s^2} - 1$$

and

$$r_+ - r_- = 2i\Omega_0 \sqrt{1 - s^2}$$

where $S = \frac{\sigma}{4\Omega_0}$. Thus

$$\Phi_0^{-1} M_2 \Phi_0 = \frac{\Omega_1 \cos \omega_1 t}{i\sqrt{1-S^2}} \begin{pmatrix} -1 & e^{-2i\Omega_0\sqrt{1-S^2}t} (s^2 + is\sqrt{1-S^2}) \\ e^{2i\Omega_0\sqrt{1-S^2}t} (s^2 - is\sqrt{1-S^2}) & 1 \end{pmatrix} \quad (3-71)$$

We obtain the following differential equations by using Equations (3-61) and (3-71)

$$\dot{\vec{C}} = \frac{i\Omega_1 \cos \omega_1 t}{\sqrt{1-S^2}} \begin{pmatrix} 1 & -e^{-2i\Omega_0\sqrt{1-S^2}t} (s^2 + is\sqrt{1-S^2}) \\ -e^{2i\Omega_0\sqrt{1-S^2}t} (s^2 - is\sqrt{1-S^2}) & -1 \end{pmatrix} \vec{C} \quad (3-72)$$

In cases where the off-diagonal elements of the matrix $\Phi_0^{-1} M_2 \Phi_0$ are much smaller than the diagonal elements, we can find an approximate solution of Equation (3-72) by setting the off-diagonal elements to zero. This approximate solution is

$$\vec{C}(t) = \begin{pmatrix} C_+ e^{i\frac{\Omega_1}{\omega_1\sqrt{1-S^2}} \sin \omega_1 t} \\ C_- e^{-i\frac{\Omega_1}{\omega_1\sqrt{1-S^2}} \sin \omega_1 t} \end{pmatrix} \quad (3-73)$$

where C_+ and C_- are determined by the initial conditions. The approximation is valid for $S \ll 1$ and we restrict ourselves to this case. In Equation (3-73) we can replace $\sqrt{1-S^2}$ by 1 and in Equation (3-66) we let $\Gamma_{\pm} \approx -\frac{3}{4}\sigma \pm i\Omega_0$ and $A_{\pm} \approx \pm i$. Thus Equations (3-66) and (3-73) reduce to

$$\phi_0 \approx \begin{pmatrix} e^{-\frac{3}{4}\gamma t + i\Omega_0 t} & e^{-\frac{3}{4}\gamma t - i\Omega_0 t} \\ -ie^{-\frac{3}{4}\gamma t + i\Omega_0 t} & ie^{-\frac{3}{4}\gamma t - i\Omega_0 t} \end{pmatrix} \quad (3-74)$$

and

$$\vec{c}(t) \approx \begin{pmatrix} c_+ e^{i\frac{\Omega_1}{\omega_1} \sin \omega_1 t} \\ c_- e^{-i\frac{\Omega_1}{\omega_1} \sin \omega_1 t} \end{pmatrix} \quad (3-75)$$

respectively. We obtain the approximate solutions of Equations (3-52) and (3-53) from Equation (3-59) together with Equations (3-74) and (3-75). They are

$$y(t) = e^{(-\frac{3}{4}\gamma + i\Omega_0)t} c_+ e^{i\frac{\Omega_1}{\omega_1} \sin \omega_1 t} + e^{(-\frac{3}{4}\gamma - i\Omega_0)t} c_- e^{-i\frac{\Omega_1}{\omega_1} \sin \omega_1 t} \quad (3-76)$$

and

$$z(t) = -ie^{(-\frac{3}{4}\gamma + i\Omega_0)t} c_+ e^{i\frac{\Omega_1}{\omega_1} \sin \omega_1 t} + ie^{(-\frac{3}{4}\gamma - i\Omega_0)t} c_- e^{-i\frac{\Omega_1}{\omega_1} \sin \omega_1 t} \quad (3-77)$$

We determine the constants c_+ and c_- by using the initial conditions $y(t')$ and $z(t')$. We find

$$c_{\pm} = e^{(\frac{3}{4}\gamma \mp i\Omega_0)t'} e^{\mp i\frac{\Omega_1}{\omega_1} \sin \omega_1 t'} \frac{1}{2} [y(t') \pm iz(t')] \quad (3-78)$$

Thus

$$y(t, t') = e^{if_+(t, t')} \frac{1}{2} [y(t') + iz(t')] + e^{if_-(t, t')} \frac{1}{2} [y(t') - iz(t')] \quad (3-79)$$

and

$$\begin{aligned}
 z(t, t') = & -i e^{i f_+(t, t')} \frac{1}{2} [y(t') + i z(t')] \\
 & + i e^{i f_-(t, t')} \frac{1}{2} [y(t') - i z(t')]
 \end{aligned} \quad (3-80)$$

where

$$i f_{\pm}(t, t') = (-\frac{3}{4}\gamma \pm i\Omega_0)(t-t') \pm i\frac{\Omega_0}{\omega_1} (\sin \omega_1 t - \sin \omega_1 t') \quad (3-81)$$

3.5 Long Term Solution of the Optical Bloch Equations

The general solution of an inhomogeneous system of linear first-order differential equations is obtained by adding a particular solution of the inhomogeneous system to the general solution of the associated homogeneous system. The method of variation of parameters can be used to determine a particular solution of an inhomogeneous system whenever the general solution of the homogeneous system is known²².

We want to obtain the general solution of the optical Bloch equations. The general solution of their associated homogeneous equations is given by Equations (3-62), (3-76) and (3-77) as

$$X_{hom}(t) = c_0 e^{-\frac{\gamma}{2}t} \quad (3-82)$$

$$y_{hom}(t) = C_+ F_+(t) + C_- F_-(t) \quad (3-83)$$

and

$$z_{hom}(t) = -i C_+ F_+(t) + i C_- F_-(t) \quad (3-84)$$

where

$$F_{\pm}(t) = e^{-\frac{3}{4}\gamma t \pm i\Omega_0 t \pm i\frac{\Omega_0}{\omega_1} \sin \omega_1 t} \quad (3-85)$$

We choose a particular solution of the optical Bloch equations of the form

$$y_{part}(t) = C_+(t) F_+(t) + C_-(t) F_-(t) \quad (3-86)$$

and

$$z_{part}(t) = -i C_+(t) F_+(t) + i C_-(t) F_-(t) \quad (3-87)$$

We substitute the homogeneous solutions of Equations (3-83) and (3-84) into Equations (3-52) and (3-53) and the particular solutions of Equations (3-86) and (3-87) into Equations (3-49) and (3-50). The resulting equations can be reduced to

$$\dot{C}_+ F_+ + \dot{C}_- F_- = 0 \quad (3-88)$$

and

$$-i \dot{C}_+ F_+ + i \dot{C}_- F_- = -\gamma \quad (3-89)$$

which we can solve for \dot{C}_+ and \dot{C}_- . We find that

$$\dot{C}_{\pm} = \mp \frac{i}{2} \gamma \frac{1}{F_{\pm}} \quad (3-90)$$

We integrate Equation (3-90) and obtain $C_+(t)$ and $C_-(t)$. The integration is possible after replacing the functions $e^{\pm i \frac{\Omega_1}{\omega_1} \sin \omega_1 t}$ by their Fourier development²³

$$e^{\pm i \frac{\Omega_1}{\omega_1} \sin \omega_1 t} = \sum_{n=-\infty}^{\infty} J_n\left(\frac{\Omega_1}{\omega_1}\right) e^{\pm i n \omega_1 t} \quad (3-91)$$

where $J_n\left(\frac{\Omega_1}{\omega_1}\right)$ is the n^{th} order Bessel function. Then

$$C_{\pm}(t) = \mp \frac{i}{2} \gamma \sum_n \frac{J_n\left(\frac{\Omega_1}{\omega_1}\right) e^{\frac{3}{4} \gamma t \mp i \Omega_0 t \mp i n \omega_1 t}}{\frac{3}{4} \gamma \mp i \Omega_0 \mp i n \omega_1} \quad (3-92)$$

We substitute Equation (3-92) into Equations (3-86) and (3-87) and use Equation (3-91) one more time. We find that

$$\begin{aligned}
y_{\text{part}}(t) &= -\frac{i}{2}\gamma \sum_n \frac{J_n(\frac{\Omega_1}{\omega_1}) e^{-in\omega_1 t}}{\frac{3}{4}\gamma - i(\Omega_0 + n\omega_1)} \sum_m J_m(\frac{\Omega_1}{\omega_1}) e^{im\omega_1 t} \\
&\quad + \frac{i}{2}\gamma \sum_n \frac{J_n(\frac{\Omega_1}{\omega_1}) e^{in\omega_1 t}}{\frac{3}{4}\gamma + i(\Omega_0 + n\omega_1)} \sum_m J_m(\frac{\Omega_1}{\omega_1}) e^{-im\omega_1 t} \\
&= -\gamma \text{Re} \sum_{nm} i \frac{J_n(\frac{\Omega_1}{\omega_1}) J_m(\frac{\Omega_1}{\omega_1}) e^{-i(n-m)\omega_1 t}}{\frac{3}{4}\gamma - i(\Omega_0 + n\omega_1)} \\
&= -\gamma \text{Re} \sum_{np} i \frac{J_n(\frac{\Omega_1}{\omega_1}) J_{n-p}(\frac{\Omega_1}{\omega_1}) e^{-ip\omega_1 t}}{\frac{3}{4}\gamma - i(\Omega_0 + n\omega_1)} \quad (3-93)
\end{aligned}$$

and, by a similar calculation, that

$$z_{\text{part}}(t) = -\gamma \text{Re} \sum_{np} \frac{J_n(\frac{\Omega_1}{\omega_1}) J_{n-p}(\frac{\Omega_1}{\omega_1}) e^{-ip\omega_1 t}}{\frac{3}{4}\gamma - i(\Omega_0 + n\omega_1)} \quad (3-94)$$

The general solution of the optical Bloch equations is thus

$$x(t) = c_0 e^{-\frac{\gamma}{2}t} \quad (3-95)$$

$$\begin{aligned}
y(t) &= y_{\text{hom}}(t) + y_{\text{part}}(t) \\
&= c_+ e^{-\frac{3}{4}\gamma t + i\Omega_0 t + i\frac{\Omega_1}{\omega_1} \sin \omega_1 t} + c_- e^{-\frac{3}{4}\gamma t - i\Omega_0 t - i\frac{\Omega_1}{\omega_1} \sin \omega_1 t} \\
&\quad - \gamma \text{Re} \sum_{np} i \frac{J_n(\frac{\Omega_1}{\omega_1}) J_{n-p}(\frac{\Omega_1}{\omega_1}) e^{-ip\omega_1 t}}{\frac{3}{4}\gamma - i(\Omega_0 + n\omega_1)} \quad (3-96)
\end{aligned}$$

$$\begin{aligned}
z(t) &= z_{\text{hom}}(t) + z_{\text{part}}(t) \\
&= -ic_+ e^{-\frac{3}{4}\gamma t + i\Omega_0 t + i\frac{\Omega_1}{\omega_1} \sin \omega_1 t} + ic_- e^{-\frac{3}{4}\gamma t - i\Omega_0 t - i\frac{\Omega_1}{\omega_1} \sin \omega_1 t} \\
&\quad - \gamma \text{Re} \sum_{np} \frac{J_n(\frac{\Omega_1}{\omega_1}) J_{n-p}(\frac{\Omega_1}{\omega_1}) e^{-ip\omega_1 t}}{\frac{3}{4}\gamma - i(\Omega_0 + n\omega_1)} \quad (3-97)
\end{aligned}$$

For times which are large compared to the lifetime $\tau = \gamma^{-1}$ of the upper atomic level we find that $\gamma t \gg \gamma \tau$ and thus $\gamma t \gg 1$, $e^{-\frac{3}{4}\gamma t} \ll 1$ and $e^{-\frac{1}{2}\gamma t} \ll 1$. We obtain the long term solution of the optical Bloch equations from Equations (3-95)-(3-97) by dropping the terms which contain the functions $e^{-\frac{3}{4}\gamma t}$ or $e^{-\frac{1}{2}\gamma t}$. We label the long term solution with a subscript " ∞ ". Thus

$$x_{\infty}(t) = 0 \quad (3-98)$$

$$y_{\infty}(t) = -\gamma \operatorname{Re} \sum_{np} i \frac{J_n(\frac{\Omega_i}{\omega_i}) J_{n-p}(\frac{\Omega_i}{\omega_i}) e^{-ip\omega_i t}}{\frac{3}{4}\gamma - i(\Omega_0 + n\omega_i)} \quad (3-99)$$

$$z_{\infty}(t) = -\gamma \operatorname{Re} \sum_{np} \frac{J_n(\frac{\Omega_i}{\omega_i}) J_{n-p}(\frac{\Omega_i}{\omega_i}) e^{-ip\omega_i t}}{\frac{3}{4}\gamma - i(\Omega_0 + n\omega_i)} \quad (3-100)$$

In the long term limit only the particular solution survives. Thus the long term solution is independent of the initial conditions which are contained in the homogeneous solution.

3.6 The Optical Bloch Equations and Their Solutions in Terms of the Variables X_+ , X_- and z

For the purpose of solving the optical Bloch equations and their associated homogeneous equations it is most convenient to use the variables x , y and z . For the calculations in the next chapter it is more convenient to switch to the new variables X_+ , X_- and z , where

$$X_{\pm} = \frac{1}{2}(x \pm iy) \quad (3-101)$$

In terms of these new variables the optical Bloch equations are

$$\dot{X}_{\pm} = -\frac{\gamma}{2} X_{\pm} \mp \frac{i}{2} \Omega(t) z \quad (3-102)$$

$$\dot{z} = -\gamma(z+1) + i\Omega(t)[x_- - x_+] \quad (3-103)$$

with the associated homogeneous equations

$$\dot{x}_{\pm} = -\frac{\gamma}{2} x_{\pm} \mp \frac{i}{2} \Omega(t) z \quad (3-104)$$

$$\dot{z} = -\gamma z + i\Omega(t)[x_- - x_+] \quad (3-105)$$

The long term solutions of Equations (3-102) and (3-103) are

$$x_{\pm\infty} = \frac{1}{2} (x_{\infty} \pm iy_{\infty}) ; \quad z_{\infty} \quad (3-106)$$

where x_{∞} , y_{∞} and z_{∞} are given by Equations (3-98), (3-99) and (3-100), respectively. The solutions of Equations (3-104) and (3-105) with the initial conditions $x_{\pm}(t') = \frac{1}{2}(x(t') \pm iy(t'))$ and $z(t')$ can be obtained by using Equations (3-62), (3-79), (3-80) and (3-101).

We find

$$\begin{aligned} x_{\pm}(t, t') &= \frac{1}{2} [x_+(t') + x_-(t')] e^{-\frac{\gamma}{2}(t-t')} \\ &\quad \pm \frac{1}{4} [x_+(t') - x_-(t') - z(t')] e^{if_+(t, t')} \\ &\quad \pm \frac{1}{4} [x_+(t') - x_-(t') + z(t')] e^{if_-(t, t')} \end{aligned} \quad (3-107)$$

and

$$\begin{aligned} z(t, t') &= \frac{1}{2} [x_-(t') - x_+(t') + z(t')] e^{if_+(t, t')} \\ &\quad + \frac{1}{2} [x_+(t') - x_-(t') + z(t')] e^{if_-(t, t')} \end{aligned} \quad (3-108)$$

where $if_+(t, t')$ and $if_-(t, t')$ are defined in Equation (3-81).

4. THE LIGHT EMITTED BY A TWO-LEVEL ATOM IN A STRONG, RESONANT, AMPLITUDE MODULATED LASER BEAM

4.1 The Spectral Distribution of the Emitted Light: A General Formula

In Figure (4-1) we illustrate the setup of an experiment to observe the light emitted by a two-level atom in a laser beam.

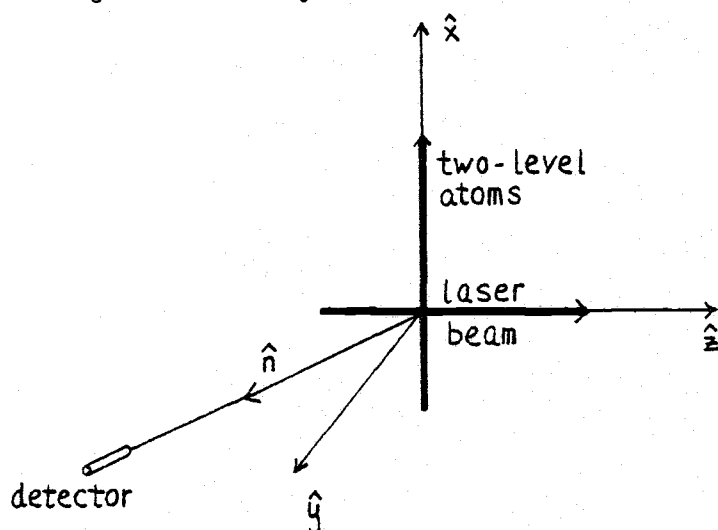


Figure (4-1). An experiment to observe the light emitted by a two-level atom in a laser beam.

Here the laser beam is propagating along the z -axis and the beam of two-level atoms is propagating along the x -axis, so that the two beams cross at the origin of the coordinate system. The unit vector \hat{n} points in the direction of the detector.

In the last two chapters we investigated the time dependence of the atomic variables of a two-level atom in a (strong, resonant, amplitude modulated) laser beam. Now we calculate the frequency distribution of

the light emitted by such an atom. We do not want to predict the readings of a real detector; we only want to derive an expression proportional to the spectral distribution of the light. In our calculations we can therefore replace the detector by a detecting atom and the beam of two-level atoms by one radiating atom^{16,24}. The detecting atom is a two-level atom with a ground state $|a\rangle$, an excited state $|b\rangle$ and an atomic frequency $\omega = \omega_b - \omega_a$. It is exposed to the radiation field between $t=0$ and $t=T$. At $t=0$ the atom is in its ground state. The probability $P(\omega, T)$ to find the detecting atom of a particular atomic frequency ω in its excited state at $t=T$ is proportional to the energy of the radiation in a frequency interval $\omega \dots \omega + d\omega$ and the function $P(\omega, T)$ is proportional to the frequency distribution $I(\omega)$. The observation time T has to be much larger than the lifetime of the radiating atom and much shorter than the lifetime of the detecting atom. The detecting atom is at $\vec{x} = r \hat{n}$ and the distance r between the detecting atom and the radiating atom is large in comparison with optical wavelengths. We assume that the detecting atom influences neither the radiating atom nor the radiation field.

The system we consider consists of the radiating atom plus radiation field (referred to as R) and the detecting atom (referred to as D) and has the Hamiltonian

$$H = H_R + H_D + H_I = H_0 + H_I \quad (4-1)$$

We write the interaction Hamiltonian in the electric dipole approximation as $H_I = -\vec{D} \cdot \vec{E}$, which, in the interaction representation, becomes

$$H_I' = e^{\frac{i}{\hbar} H_0 t} H_I e^{-\frac{i}{\hbar} H_0 t} = -\vec{D}(t) \cdot \vec{E}(\vec{x}, t) \quad (4-2)$$

Here $\vec{D}(t)$ is the electric dipole operator of the detecting atom in the interaction representation and can be written as

$$\vec{D}(t) = \vec{\sigma} |a\rangle\langle b| e^{-i\omega t} + \vec{\sigma}^* |b\rangle\langle a| e^{i\omega t} \quad (4-3)$$

where $\vec{\sigma} = \langle a | \vec{D} | b \rangle$ and $\vec{E}(\vec{x}, t)$ is the Heisenberg electric field operator of R and obeys the Maxwell equations. We rewrite the Hermitian operator $\vec{E}(\vec{x}, t)$ in terms of a pair of Hermitian adjoint operators as

$$\vec{E}(\vec{x}, t) = \vec{E}^+(\vec{x}, t) + \vec{E}^-(\vec{x}, t) \quad (4-4)$$

where $\vec{E}^+(\vec{x}, t)$ and $\vec{E}^-(\vec{x}, t)$ are the positive and negative frequency parts respectively of the electric field operator. Thus

$$\begin{aligned} H_I' &= -[\vec{\sigma} |a\rangle\langle b| e^{-i\omega t} + \vec{\sigma}^* |b\rangle\langle a| e^{i\omega t}] \cdot [\vec{E}^+ + \vec{E}^-] \\ &\approx -\vec{\sigma} \cdot \vec{E}^-(\vec{x}, t) e^{-i\omega t} |a\rangle\langle b| - \vec{\sigma}^* \cdot \vec{E}^+(\vec{x}, t) e^{i\omega t} |b\rangle\langle a| \end{aligned} \quad (4-5)$$

where we dropped the rapidly oscillating terms.

At $t = 0$, when D is in the ground state, the density matrix in the interaction representation is

$$\rho(0) = |a\rangle\langle a| \rho_R \quad (4-6)$$

where ρ_R is the density matrix of R at $t = 0$. This density matrix obeys the differential equation

$$i\hbar \frac{\partial}{\partial t} \rho(t) = [H_I'(t), \rho(t)] \quad (4-7)$$

which, to the second approximation, has the solution

$$\begin{aligned}
S(T) &= S(0) - \frac{i}{\hbar} \int_0^T dt [H_I'(t), S(0)] \\
&\quad - \frac{1}{\hbar^2} \int_0^T dt \int_0^t dt' [H_I'(t), [H_I'(t'), S(0)]]
\end{aligned} \tag{4-8}$$

The probability to find D in its excited state at $t = T$ is

$$P(\omega, T) = \text{tr}_{R,D} |b\rangle\langle b| S(T) \tag{4-9}$$

We use Equations (4-5), (4-6), (4-8) and (4-9) and take the trace over D . We find

$$\begin{aligned}
P(\omega, T) &= \text{tr}_R \frac{1}{\hbar^2} \int_0^T dt \int_0^t dt' \{ \vec{\sigma}^* \cdot \vec{E}^+(\vec{x}, t) S_R \vec{\sigma} \cdot \vec{E}^-(\vec{x}, t') e^{i\omega(t-t')} \\
&\quad + \vec{\sigma}^* \cdot \vec{E}^+(\vec{x}, t') S_R \vec{\sigma} \cdot \vec{E}^-(\vec{x}, t) e^{i\omega(t'-t)} \}
\end{aligned}$$

We exchange the variables t and t' in the first integral and use the relations

$$\int_0^T dt \int_0^t dt' + \int_0^T dt' \int_0^{t'} dt = \int_0^T dt \int_0^T dt' \tag{4-10}$$

$$\vec{\sigma} \cdot \vec{E}^-(\vec{x}, t) = \sum_{\mu} \sigma_{\mu} E_{\mu}^-(\vec{x}, t)$$

$$\vec{\sigma}^* \cdot \vec{E}^+(\vec{x}, t') = \sum_{\nu} \sigma_{\nu}^* E_{\nu}^+(\vec{x}, t')$$

and

$$\langle E_{\mu}^-(\vec{x}, t) E_{\nu}^+(\vec{x}, t') \rangle = \text{tr}_R S_R E_{\mu}^-(\vec{x}, t) E_{\nu}^+(\vec{x}, t')$$

We then find

$$P(\omega, T) = \frac{1}{\hbar^2} \sum_{\mu\nu} \delta_{\mu} \delta_{\nu}^* \int_0^T dt \int_0^T dt' \langle E_{\mu}^{-}(\vec{x}, t) E_{\nu}^{+}(\vec{x}, t') \rangle e^{-i\omega(t-t')} \quad (4-11)$$

The probability $P(\omega, T)$ in Equation (4-11) is expressed in terms of electric field operators which can, in turn, be related to the dipole moment operators of the radiating atom. In classical electrodynamics, the time dependent charge and current distributions are treated by making a Fourier analysis and handling each Fourier component separately²⁵. A charge distribution $\rho(\vec{x}, t) = \rho(\vec{x}) e^{-i\omega_0 t}$ has a dipole moment $\vec{p}(t) = \int \rho(\vec{x}', t) \vec{x}' d^3x' = \vec{p}_0 e^{-i\omega_0 t}$ and its fields at a position $\vec{x} = r \hat{n}$ in the radiation zone are

$$\vec{H}(\vec{x}, t) = \frac{\omega_0 R_0}{4\pi r} e^{iR_0 r} (\hat{n} \times \vec{p}_0) e^{-i\omega_0 t}$$

and

$$\vec{E}(\vec{x}, t) = \frac{R_0^2}{4\pi \epsilon_0} \frac{1}{r} e^{iR_0 r} [(\hat{n} \times \vec{p}_0) \times \hat{n}] e^{-i\omega_0 t}$$

where $R_0 = \frac{\omega_0}{c}$ is the wave number of the radiation. We can write the electric field as

$$\vec{E}(\vec{x}, t) = \frac{R_0^2}{4\pi \epsilon_0} \frac{1}{r} [(\hat{n} \times \vec{p}(t - \frac{r}{c})) \times \hat{n}] \quad (4-12)$$

The complex field $\vec{E}(\vec{x}, t)$ in Equation (4-12) depends on time like $e^{-i\omega_0 t}$, thus its quantum mechanical analog is the field operator $\vec{E}^{+}(\vec{x}, t)$. The classical dipole moment $\vec{p}(t)$ also goes with $e^{-i\omega_0 t}$. The atomic dipole operator of the radiating atom is given by Equation (3-42). We rewrite it in a slightly different form, namely as

$$[\vec{d}'] = d_0 (\hat{x} - i\hat{y}) e^{i\omega_0 t} \begin{pmatrix} 0 & 1 \\ 0 & 0 \end{pmatrix} + d_0 (\hat{x} + i\hat{y}) e^{-i\omega_0 t} \begin{pmatrix} 0 & 0 \\ 1 & 0 \end{pmatrix} \\ = \vec{D}^+(t) + \vec{D}^-(t) \quad (4-13)$$

where $\vec{D}^+(t)$ and $\vec{D}^-(t)$ are the raising and lowering parts of $[\vec{d}']$ respectively. By comparison we see that the classical quantity $\vec{p}(t - \frac{\epsilon}{\tau})$ in Equation (4-12) corresponds to the quantum operator $\vec{D}^-(t - \frac{\epsilon}{\tau})$ because of its time dependence. We thus find the following relations between operators:

$$\vec{E}^+(\vec{x}, t) = \frac{k_0^2}{4\pi\epsilon_0} \frac{1}{\tau} (\hat{n} \times \vec{D}^-(t - \frac{\epsilon}{\tau})) \times \hat{n} \quad (4-14)$$

and

$$\vec{E}^-(\vec{x}, t) = \frac{k_0^2}{4\pi\epsilon_0} \frac{1}{\tau} (\hat{n} \times \vec{D}^+(t - \frac{\epsilon}{\tau})) \times \hat{n} \quad (4-15)$$

The latter equation is the adjoint of the former one.

We now express the operators $\vec{D}^+(t)$ and $\vec{D}^-(t)$ in terms of those operators whose expectation values appear in the optical Bloch equations. As we noted earlier, the optical Bloch equations are a set of differential equations for the components of the fictitious spin- $\frac{1}{2}$ particle in a frame rotating with frequency ω_L around the \mathbf{z} -axis. In that frame any operator \mathcal{O} becomes

$$e^{-\frac{i}{2}\sigma_z\omega_L t} \mathcal{O} e^{\frac{i}{2}\sigma_z\omega_L t} \quad (4-16)$$

The operators σ_x' , σ_y' and σ_z' are the Pauli spin matrices in the interaction representation. They are given by Equations (3-4)-(3-6). The

operator $\tilde{\sigma}_z'$ is transformed into the operator

$$S_z(t) = e^{-\frac{i}{2}\tilde{\sigma}_z\omega_L t} \tilde{\sigma}_z' e^{\frac{i}{2}\tilde{\sigma}_z\omega_L t} = \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix} \quad (4-17)$$

in the rotating frame. The expectation value of $S_z(t)$ is the function $z(t)$, which we obtain from the optical Bloch equations;

$$\langle S_z(t) \rangle = z(t) \quad (4-18)$$

We note that

$$\tilde{\sigma}_+' \equiv \frac{1}{2}(\tilde{\sigma}_x' + i\tilde{\sigma}_y') = e^{i\omega_0 t} \begin{pmatrix} 0 & 1 \\ 0 & 0 \end{pmatrix} \quad (4-19)$$

and

$$\tilde{\sigma}_-' \equiv \frac{1}{2}(\tilde{\sigma}_x' - i\tilde{\sigma}_y') = e^{-i\omega_0 t} \begin{pmatrix} 0 & 0 \\ 1 & 0 \end{pmatrix} \quad (4-20)$$

In the rotating frame these operators become

$$S_+(t) = e^{-\frac{i}{2}\tilde{\sigma}_z\omega_L t} \tilde{\sigma}_+' e^{\frac{i}{2}\tilde{\sigma}_z\omega_L t} = e^{-i\omega_L t} \tilde{\sigma}_+' \quad (4-21)$$

and

$$S_-(t) = e^{-\frac{i}{2}\tilde{\sigma}_z\omega_L t} \tilde{\sigma}_-' e^{\frac{i}{2}\tilde{\sigma}_z\omega_L t} = e^{i\omega_L t} \tilde{\sigma}_-' \quad (4-22)$$

The expectation values of $S_+(t)$ and $S_-(t)$ are

$$\langle S_+(t) \rangle = \frac{1}{2} \langle S_x(t) + i S_y(t) \rangle = \frac{1}{2} [x(t) + i y(t)] = X_+(t) \quad (4-23)$$

and

$$\langle S_-(t) \rangle = \frac{1}{2} \langle S_x(t) - i S_y(t) \rangle = \frac{1}{2} [x(t) - i y(t)] = X_-(t) \quad (4-24)$$

respectively and $X_+(t)$ and $X_-(t)$ are solutions of the optical Bloch equations. We compare Equations (4-21) and (4-22) with Equation (4-13) and find

$$\vec{D}^+(t) = d_o (\hat{x} - i\hat{y}) e^{i\omega_L t} S_+(t) \quad (4-25)$$

$$\vec{D}^-(t) = d_o (\hat{x} + i\hat{y}) e^{-i\omega_L t} S_-(t) \quad (4-26)$$

We can substitute Equations (4-25) and (4-26) into Equations (4-14) and (4-15) and those in turn into Equation (4-11). We obtain an expression for the spectral distribution $I(\omega)$ in terms of two operators, $S_+(t)$ and $S_-(t)$, whose expectation values are known from the optical Bloch equations.

The detecting atom may be anywhere in the radiation zone of the field as long as it is outside the beams. Thus \hat{n} can point in any direction other than the X -axis or the Z -axis. For convenience we assume that \hat{n} is a unit vector in the y -direction. Thus

$$\hat{n} = \hat{y} \quad (4-27)$$

$$\vec{E}^+(\vec{x}, t) = \frac{k_o^2}{4\pi\epsilon_o} \frac{d_o}{T} e^{-i\omega_L(t-\frac{r}{c})} S_-(t-\frac{r}{c}) \hat{x} \quad (4-28)$$

and

$$\vec{E}^-(\vec{x}, t) = \frac{k_o^2}{4\pi\epsilon_o} \frac{d_o}{T} e^{i\omega_L(t-\frac{r}{c})} S_+(t-\frac{r}{c}) \hat{x} \quad (4-29)$$

It is common to neglect the transit time i.e. to replace $t - \frac{r}{c}$ by t . We substitute Equations (4-28) and (4-29) into Equation (4-11) and find

$$P(\omega, T) = \frac{\delta_x \delta_x^*}{\hbar^2} \int_0^T dt \int_0^T dt' \left(\frac{k_o^2 d_o}{4\pi\epsilon_o T} \right)^2 \langle S_+(t) S_-(t') \rangle e^{-i(\omega - \omega_L)(t-t')}$$

Thus we arrive at the following relations for $I(\omega)$:

$$I(\omega) \sim \frac{1}{T} \int_0^T dt \int_0^T dt' \langle S_+(t) S_-(t') \rangle e^{-i(\omega - \omega_L)(t-t')} \quad (4-30)$$

or

$$I(\omega) \sim \frac{2}{T} \operatorname{Re} \int_0^T dt \int_0^t dt' \langle S_+(t) S_-(t') \rangle e^{-i(\omega - \omega_L)(t-t')} \quad (4-31)$$

where we used Equation (4-10) and the relation

$\langle S_+(t') S_-(t) \rangle = \langle S_+(t) S_-(t') \rangle^*$ to derive Equation (4-31) from Equation (4-30).

4.2 Average Motion and Fluctuations

There are several approaches to the quantum theory of damping^{26,27}. In the density operator methods we deal with a quantum mechanical system coupled to a "reservoir" (the radiation field). We trace all equations of motion over both the system and reservoir since we are only interested in the statistical properties of the system. This results in equations like the optical Bloch equations which describe the average motion of the atomic operators $S_+(t)$, $S_-(t)$ and $S_z(t)$ and which are listed here one more time in terms of the variables $\langle S_+(t) \rangle$, $\langle S_-(t) \rangle$ and $\langle S_z(t) \rangle$:

$$\frac{d}{dt} \langle S_{\pm}(t) \rangle = -\frac{\gamma}{2} \langle S_{\pm}(t) \rangle \mp \frac{i}{2} \Omega(t) \langle S_z(t) \rangle \quad (4-32)$$

$$\frac{d}{dt} \langle S_z(t) \rangle = -\gamma [\langle S_z(t) \rangle + 1] + i\Omega(t) [\langle S_-(t) \rangle - \langle S_+(t) \rangle] \quad (4-33)$$

A quantum system experiences damping and fluctuations when inter-

acting with the reservoir. In the Langevin method we take the reservoir into account by introducing damping terms, which describe the cumulative effect of the reservoir and give the correct statistical properties of the system and by introducing a random force $\vec{K}(t)$, with an extremely short correlation time and a zero average value to create fluctuations. We then obtain the following equations of motion for the atomic operators $S_+(t)$, $S_-(t)$ and $S_z(t)$:

$$\frac{d}{dt} S_{\pm}(t) = -\frac{\gamma}{2} S_{\pm}(t) \mp \frac{i}{2} \Omega(t) S_z(t) + K_{\pm}(t) \quad (4-34)$$

$$\frac{d}{dt} S_z(t) = -\gamma [S_z(t) + 1] + i \Omega(t) [S_-(t) - S_+(t)] + K_z(t) \quad (4-35)$$

Here

$$\langle K_{\pm}(t) \rangle = 0 \quad (4-36)$$

and

$$\langle K_z(t) \rangle = 0 \quad (4-37)$$

so that Equations (4-34) and (4-35) reduce to Equations (4-32) and (4-33) when we take the average.

We need all the above equations to calculate the two-time average $\langle S_+(t) S_-(t') \rangle$, which appears in Equations (4-30) and (4-31) for $I(\omega)$. We express the operators $S_+(t)$, $S_-(t)$ and $S_z(t)$ in terms of their average values and fluctuations as¹⁶

$$S_{\pm}(t) = \langle S_{\pm}(t) \rangle + \delta S_{\pm}(t) \quad (4-38)$$

$$S_z(t) = \langle S_z(t) \rangle + \delta S_z(t) \quad (4-39)$$

where

$$\langle \delta S_{\pm}(t) \rangle = 0 \quad (4-40)$$

$$\langle \delta S_z(t) \rangle = 0 \quad (4-41)$$

and find that

$$\langle S_+(t) S_-(t') \rangle = \langle S_+(t) \rangle \langle S_-(t') \rangle + \langle \delta S_+(t) \delta S_-(t') \rangle \quad (4-42)$$

Thus

$$\begin{aligned} I(\omega) \sim & \frac{2}{T} \operatorname{Re} \int_0^T dt \int_0^t dt' \langle S_+(t) \rangle \langle S_-(t') \rangle e^{-i(\omega - \omega_L)(t-t')} \\ & + \frac{2}{T} \operatorname{Re} \int_0^T dt \int_0^t dt' \langle \delta S_+(t) \delta S_-(t') \rangle e^{-i(\omega - \omega_L)(t-t')} \end{aligned} \quad (4-43)$$

Both the average motion and the fluctuations contribute to the spectral distribution of the emitted light; their contributions are the coherent and incoherent parts of the spectrum, respectively.

We now derive equations of motion for the functions $g_+(t, t')$,

$g_-(t, t')$ and $g_z(t, t')$, which are defined as

$$g_{\pm}(t, t') = \langle \delta S_{\pm}(t) \delta S_{\pm}(t') \rangle \quad (4-44)$$

and

$$g_z(t, t') = \langle \delta S_z(t) \delta S_z(t') \rangle \quad (4-45)$$

We note that

$$\delta S_{\pm}(t) = S_{\pm}(t) - \langle S_{\pm}(t) \rangle \quad (4-46)$$

$$\delta S_z(t) = S_z(t) - \langle S_z(t) \rangle \quad (4-47)$$

$$\frac{d}{dt} \delta S_{\pm}(t) = -\frac{\gamma}{2} \delta S_{\pm}(t) \mp \frac{i}{2} \Omega(t) \delta S_z(t) + K_{\pm}(t) \quad (4-48)$$

$$\frac{d}{dt} \delta S_z(t) = -\gamma \delta S_z(t) + i \Omega(t) [\delta S_-(t) - \delta S_+(t)] + K_z(t) \quad (4-49)$$

We multiply Equations (4-48) and (4-49) by $\delta S_{\pm}(t')$ from the right and then take the average. We find

$$\frac{d}{dt} q_{\pm}(t, t') = -\frac{\gamma}{2} q_{\pm}(t, t') \mp \frac{i}{2} \Omega(t) q_{\pm}(t, t') \quad (4-50)$$

and

$$\frac{d}{dt} q_{\pm}(t, t') = -\gamma q_{\pm}(t, t') + i \Omega(t) [q_{-}(t, t') - q_{+}(t, t')] \quad (4-51)$$

since

$$\langle K_{\pm}(t) \delta S_{-}(t') \rangle = 0 \quad (4-52)$$

and

$$\langle K_{\pm}(t) \delta S_{-}(t') \rangle = 0 \quad (4-53)$$

because of the extremely short correlation time of the random force

$\vec{K}(t)$. Thus $q_{+}(t, t')$, $q_{-}(t, t')$ and $q_{\pm}(t, t')$ obey Equations (3-104) and (3-105), the solutions of which are given by Equations (3-107) and (3-108).

We are particularly interested in $q_{+}(t, t') = \langle \delta S_{+}(t) \delta S_{-}(t') \rangle$ and find that

$$\begin{aligned} q_{+}(t, t') &= \frac{1}{2} [q_{+}(t') + q_{-}(t')] e^{-\frac{\gamma}{2}(t-t')} \\ &+ \frac{1}{4} [q_{+}(t') - q_{-}(t') - q_{\pm}(t')] e^{i f_{+}(t, t')} \\ &+ \frac{1}{4} [q_{+}(t') - q_{-}(t') + q_{\pm}(t')] e^{i f_{-}(t, t')} \end{aligned} \quad (4-54)$$

where

$$q_{\pm}(t') = \langle \delta S_{\pm}(t') \delta S_{-}(t') \rangle \quad (4-55)$$

and

$$q_{\pm}(t') = \langle \delta S_{\pm}(t') \delta S_{-}(t') \rangle \quad (4-56)$$

Instead of the two-time average $\langle \delta S_{+}(t) \delta S_{-}(t') \rangle$ we now have to evaluate the one-time averages $q_{+}(t')$, $q_{-}(t')$ and $q_{\pm}(t')$. This is easier, since for equal times there exist some fundamental relations between the operators, like

$$S_x^2(t') = S_y^2(t') = S_z^2(t') = 1 \quad (4-57)$$

$$S_x(t') S_y(t') = i S_z(t') \quad (\text{cyclic}) \quad (4-58)$$

and

$$S_y(t') S_x(t') = -i S_z(t') \quad (\text{cyclic}) \quad (4-59)$$

The time T during which the "detector" is exposed to the radiation is the time during which the atom radiates and is approximately equal to the time the atom spends in the laser beam. For a beam crossing experiment like the one illustrated in Figure (4-1) T is large compared to the lifetime of the radiating atom. Thus the atom is essentially at all times in a state described by the long term solutions, where

$\langle S_{\pm}(t') \rangle = X_{\pm\infty}(t')$ and $\langle S_z(t') \rangle = z_{\infty}(t')$. We use Equations (4-46) and (4-47) and find

$$g_+(t') = \frac{1}{2} [1 + z_{\infty}(t')] - \frac{1}{4} y_{\infty}^2(t') \quad (4-60)$$

$$g_-(t') = \frac{1}{4} y_{\infty}^2(t') \quad (4-61)$$

$$g_z(t') = \frac{i}{2} y_{\infty}(t') + \frac{i}{2} y_{\infty}(t') z_{\infty}(t') \quad (4-62)$$

Here we made use of Equation (3-98) ($X_{\infty} = 0$). We substitute Equations (4-60)-(4-62) into Equation (4-54) and get

$$\begin{aligned} \langle \delta S_+(t) \delta S_-(t') \rangle &= \frac{1}{4} (1 + z_{\infty}(t')) e^{-\frac{\gamma}{2}(t-t')} \\ &+ \frac{1}{8} (1 + z_{\infty}(t') - y_{\infty}^2(t') - i y_{\infty}(t') - i y_{\infty}(t') z_{\infty}(t')) e^{i f_+(t,t')} \\ &+ \frac{1}{8} (1 + z_{\infty}(t') - y_{\infty}^2(t') + i y_{\infty}(t') + i y_{\infty}(t') z_{\infty}(t')) e^{i f_-(t,t')} \end{aligned} \quad (4-63)$$

4.3 The Spectral Distribution of the Emitted Light: The Coherent Part and the Incoherent Part of the Spectrum

We now calculate the coherent part and the incoherent part of the spectrum using

$$I_{\text{coh}}(\omega) \sim \frac{1}{T} \int_0^T dt \int_0^T dt' \langle S_+(t) \rangle \langle S_-(t') \rangle e^{-i(\omega - \omega_L)(t-t')} \quad (4-64)$$

and

$$I_{\text{inc}}(\omega) \sim \frac{2}{T} \text{Re} \int_0^T dt \int_0^t dt' \langle \delta S_+(t) \delta S_-(t') \rangle e^{-i(\omega - \omega_L)(t-t')} \quad (4-65)$$

respectively.

We rewrite the long term solutions in a more transparent form as

$$\begin{aligned} y_{\infty}(t) &= -\frac{i}{2} \sigma \sum_p \sum_n \left\{ \frac{J_n J_{n-p}}{\frac{3}{4}\sigma - i(\Omega_0 + n\omega_L)} - \frac{J_n J_{n+p}}{\frac{3}{4}\sigma + i(\Omega_0 + n\omega_L)} \right\} e^{-ip\omega_L t} \\ &= -\frac{i}{2} \sigma \sum_p y_p e^{-ip\omega_L t} \end{aligned} \quad (4-66)$$

$$x_{\pm\infty}(t) = \pm \frac{i}{2} y_{\infty}(t) = \pm \frac{\sigma}{4} \sum_p y_p e^{-ip\omega_L t} \quad (4-67)$$

and

$$\begin{aligned} z_{\infty}(t) &= -\frac{\sigma}{2} \sum_p \sum_n \left\{ \frac{J_n J_{n-p}}{\frac{3}{4}\sigma - i(\Omega_0 + n\omega_L)} + \frac{J_n J_{n+p}}{\frac{3}{4}\sigma + i(\Omega_0 + n\omega_L)} \right\} e^{-ip\omega_L t} \\ &= -\frac{\sigma}{2} \sum_p z_p e^{-ip\omega_L t} \end{aligned} \quad (4-68)$$

where y_p and z_p are time independent factors which obey the relations

$$y_{-p} = -y_p^* \quad (4-69)$$

and

$$z_{-p} = z_p^* \quad (4-70)$$

Thus the coherent part of the spectrum is

$$I_{\text{coh}}(\omega) \sim \frac{1}{T} \int_0^T dt \int_0^T dt' \left(\frac{1}{4} \sum_p y_p e^{-ip\omega_1 t} \right) \left(-\frac{1}{4} \sum_q y_q e^{-iq\omega_1 t'} \right) e^{-i(\omega - \omega_1)(t-t')} \quad (4-71)$$

A common representation of the Dirac delta function is

$$\delta(k-k') = \frac{1}{2\pi} \int_{-\infty}^{\infty} e^{i(k-k')x} dx \quad (4-72)$$

We can derive an equivalent representation for the Kronecker delta function, $\delta_{p,p'}$. We note that

$$\frac{1}{L} \int_0^L e^{i(p-p')x} dx = 1 \quad \text{for } p = p'$$

and

$$\frac{1}{L} \int_0^L e^{i(p-p')x} dx \rightarrow 0 \quad \text{for } p \neq p' \text{ and } L \gg 1$$

Thus we have

$$\delta_{p,p'} = \lim_{L \rightarrow \infty} \frac{1}{L} \int_0^L e^{i(p-p')x} dx$$

and from this it follows that

$$\int_0^L e^{i(p-p')x} dx \approx L \delta_{p,p'} \quad (4-73)$$

for large L .

With Equations (4-72) and (4-73) in mind we change the integration

variable t' in Equation (4-71) to $t'' = t - t'$ and get

$$I_{coh}(\omega) \sim -\frac{1}{T} \left(\frac{\tau}{4}\right)^2 \sum_{pq} y_p y_q \int_0^T dt e^{-i(p+q)\omega_1 t} \int_{t-T}^t dt'' e^{-i(\omega - \omega_L - q\omega_1)t''} \quad (4-74)$$

where the integrand of the integral over t has the form of the one in Equation (4-73) and the integrand of the integral over t'' has the form of the one in Equation (4-72). The values of the variable t lie in the range $[0, T]$; thus the values of the lower limit of the t'' -integral are in the range $[-T, 0]$ and the values of the upper limit are in the range $[0, T]$. Since $T \gg \tau$ and $\omega - \omega_L - q\omega_1$ is of the order of τ we can replace the function $f(t) = \int_{t-T}^T dt'' e^{-i(\omega - \omega_L - q\omega_1)t''}$ by $f_0(t) = \int_{-\infty}^{\infty} dt'' e^{-i(\omega - \omega_L - q\omega_1)t''}$ for all t except in a small region near $t=0$ and $t=T$. We can thus write to a very good approximation

$$I_{coh}(\omega) \sim -\frac{1}{T} \left(\frac{\tau}{4}\right)^2 \sum_{pq} y_p y_q \int_0^T dt e^{-i(p+q)\omega_1 t} 2\pi \delta(\omega - \omega_L - q\omega_1) \quad (4-75)$$

In the limit of large measuring times T we can use Equation (4-73) to solve the remaining integral. This, together with Equation (4-69), yields

$$I_{coh}(\omega) \sim \frac{\pi \tau^2}{8} \sum_p y_p y_p^* \delta(\omega - \omega_L + p\omega_1) \quad (4-76)$$

We substitute the expressions for y_p and y_p^* into Equation (4-76) and get

$$\begin{aligned}
I_{coh}(\omega) \sim & \frac{\pi \sigma^2}{8} \sum_p \left[\sum_{nn'} \frac{J_n(\frac{\Omega_i}{\omega_i})}{(\frac{3}{4}\sigma)^2 + (\Omega_o + n\omega_i)^2} \cdot \frac{J_{n'}(\frac{\Omega_i}{\omega_i})}{(\frac{3}{4}\sigma)^2 + (\Omega_o + n'\omega_i)^2} \right. \\
& \cdot \left\{ [(\frac{3}{4}\sigma)^2 + (\Omega_o + n\omega_i)(\Omega_o + n'\omega_i)] [J_{n-p}(\frac{\Omega_i}{\omega_i}) J_{n'-p}(\frac{\Omega_i}{\omega_i}) + J_{n+p}(\frac{\Omega_i}{\omega_i}) J_{n'+p}(\frac{\Omega_i}{\omega_i})] \right. \\
& \left. + [(\Omega_o + n\omega_i)(\Omega_o + n'\omega_i) - (\frac{3}{4}\sigma)^2] [J_{n-p}(\frac{\Omega_i}{\omega_i}) J_{n'+p}(\frac{\Omega_i}{\omega_i}) + J_{n+p}(\frac{\Omega_i}{\omega_i}) J_{n'-p}(\frac{\Omega_i}{\omega_i})] \right\} \\
& \cdot \delta(\omega - \omega_L + p\omega_i)
\end{aligned} \tag{4-77}$$

The incoherent part of the spectrum is

$$\begin{aligned}
I_{inc}(\omega) \sim & \frac{2}{T} \operatorname{Re} \int_0^T dt \int_0^t dt' \frac{1}{4} (1 + z_\infty(t')) e^{-\frac{T}{2}(t-t')} e^{-i(\omega - \omega_L)(t-t')} \\
& + \frac{2}{T} \operatorname{Re} \int_0^T dt \int_0^t dt' \frac{1}{8} (1 + z_\infty(t') - i y_\infty(t')) e^{if_+(t,t')} e^{-i(\omega - \omega_L)(t-t')} \\
& + \frac{2}{T} \operatorname{Re} \int_0^T dt \int_0^t dt' \frac{1}{8} (-y_\infty^2(t') - i y_\infty(t') z_\infty(t')) e^{if_+(t,t')} e^{-i(\omega - \omega_L)(t-t')} \\
& + \frac{2}{T} \operatorname{Re} \int_0^T dt \int_0^t dt' \frac{1}{8} (1 + z_\infty(t') + i y_\infty(t')) e^{if_-(t,t')} e^{-i(\omega - \omega_L)(t-t')} \\
& + \frac{2}{T} \operatorname{Re} \int_0^T dt \int_0^t dt' \frac{1}{8} (-y_\infty^2(t') + i y_\infty(t') z_\infty(t')) e^{if_-(t,t')} e^{-i(\omega - \omega_L)(t-t')} \\
& = I_1 + I_2^+ + I_3^+ + I_2^- + I_3^-
\end{aligned} \tag{4-78}$$

To evaluate the integral I_1 , we express $z_\infty(t')$ by Equation (4-68) and change the variable from t' to $t'' = t - t'$. We get

$$I_1 = \frac{2}{T} \operatorname{Re} \int_0^T dt \int_0^t dt'' \frac{1}{4} \left(1 - \frac{T}{2} \sum_p z_p e^{-ip\omega_1 t} e^{ip\omega_1 t''} \right) e^{-(\frac{T}{2} + i(\omega - \omega_L))t''} \quad (4-79)$$

Because of the factor $e^{-\frac{T}{2}t''}$ in the integrands the integrals over t'' are independent of t except for a small region near $t = 0$. Thus we can find an approximate expression for I_1 by replacing the upper limits of the t'' -integrals by infinity and subsequently using Equation (4-73) to evaluate the integral over t . We find that

$$I_1 = 2 \operatorname{Re} \left\{ \frac{1}{4} \frac{1}{\frac{T}{2} + i(\omega - \omega_L)} - \frac{T}{8} \frac{z_0}{\frac{T}{2} + i(\omega - \omega_L)} \right\} \quad (4-80)$$

We now evaluate the integrals I_2^+ and I_2^- . We use Equations (3-87) and (3-97) and find

$$e^{if_\pm(t, t')} = e^{(-\frac{3}{4}T \pm i\Omega_0)(t - t')} \sum_{k, m} J_k\left(\frac{\Omega_1}{\omega_1}\right) J_m\left(\frac{\Omega_1}{\omega_1}\right) e^{\pm i k \omega_1 t} e^{\mp i m \omega_1 t'} \quad (4-81)$$

From Equations (4-66) and (4-68) we get

$$z_\infty(t') \mp i y_\infty(t') = -\frac{T}{2} \sum_p (z_p \pm y_p) e^{-ip\omega_1 t'} \quad (4-82)$$

We manipulate the integration variables and limits as before and find that

$$I_2^\pm = 2 \operatorname{Re} \left\{ \frac{1}{8} \sum_m \frac{J_m^2(\frac{\Omega_1}{\omega_1})}{i(\omega - \omega_L \mp \Omega_0 \mp m\omega_1) + \frac{3}{4}\gamma} \right. \\ \left. - \frac{\gamma}{16} \sum_{km} \frac{J_k(\frac{\Omega_1}{\omega_1}) J_m(\frac{\Omega_1}{\omega_1}) [z_{\pm(k-m)} \pm y_{\pm(k-m)}]}{i(\omega - \omega_L \mp \Omega_0 \mp k\omega_1) + \frac{3}{4}\gamma} \right\} \quad (4-83)$$

We note that

$$z_p \pm y_p = 2 \sum_n \frac{J_n(\frac{\Omega_1}{\omega_1}) J_{n \mp p}(\frac{\Omega_1}{\omega_1})}{\frac{3}{4}\gamma \mp i(\Omega_0 + n\omega_1)} \quad (4-84)$$

and²⁸

$$\sum_m J_{n \mp m}(a) J_m(b) = J_n(a \pm b) \quad (4-85)$$

The triple sums in I_2^+ and I_2^- reduce to single sums, since executing the sum over m gives $J_{n-k}(0) = \delta_{n,k}$ in both cases. Thus

$$I_2^\pm = \frac{1}{4} \operatorname{Re} \left\{ \sum_m \frac{J_m^2(\frac{\Omega_1}{\omega_1})}{i(\omega - \omega_L \mp \Omega_0 \mp m\omega_1) + \frac{3}{4}\gamma} \left(1 - \frac{\gamma}{\frac{3}{4}\gamma \mp i(\Omega_0 + m\omega_1)} \right) \right\} \quad (4-86)$$

The evaluation of the integrals I_3^+ and I_3^- is straightforward; the result is

$$I_3^+ + I_3^- = \left(\frac{\gamma}{4}\right)^2 \operatorname{Re} \sum_{npp'} J_{n+p+p'}(\frac{\Omega_1}{\omega_1}) J_n(\frac{\Omega_1}{\omega_1}) \cdot \\ \cdot \left\{ \frac{y_p(z_{p'} + y_{p'})}{\frac{3}{4}\gamma + i(\omega - \omega_L - \Omega_0 - (n+p+p')\omega_1)} + \frac{y_{p'}^*(z_p^* + y_p^*)}{\frac{3}{4}\gamma + i(\omega - \omega_L + \Omega_0 + (n+p+p')\omega_1)} \right\} \quad (4-87)$$

We replace \mathbf{z}_0 in Equation (4-80) and \mathbf{z}_p and \mathbf{y}_p in Equation (4-87) by their corresponding sums and determine the real parts of the resulting expressions. Then the incoherent part of the spectrum becomes

$$\begin{aligned}
 I_{inc}(\omega) \sim & \frac{\frac{\pi}{4}}{(\frac{\pi}{2})^2 + (\omega - \omega_L)^2} \left(1 - \frac{3}{4} \gamma^2 \sum_m \frac{J_m^2(\frac{\Omega_1}{\omega_1})}{(\frac{3}{4} \gamma)^2 + (\Omega_0 + m\omega_1)^2} \right) \\
 & + \frac{3}{16} \gamma \sum_m \left(\frac{J_m^2(\frac{\Omega_1}{\omega_1})}{(\frac{3}{4} \gamma)^2 + (\omega - \omega_L - \Omega_0 - m\omega_1)^2} + \frac{J_m^2(\frac{\Omega_1}{\omega_1})}{(\frac{3}{4} \gamma)^2 + (\omega - \omega_L + \Omega_0 + m\omega_1)^2} \right) \\
 & - \frac{1}{4} \gamma \sum_m \frac{J_m^2(\frac{\Omega_1}{\omega_1}) [(\frac{3}{4} \gamma)^2 + (\Omega_0 + m\omega_1)(\omega - \omega_L - \Omega_0 - m\omega_1)]}{[(\frac{3}{4} \gamma)^2 + (\Omega_0 + m\omega_1)^2][(\frac{3}{4} \gamma)^2 + (\omega - \omega_L - \Omega_0 - m\omega_1)^2]} \\
 & - \frac{1}{4} \gamma \sum_m \frac{J_m^2(\frac{\Omega_1}{\omega_1}) [(\frac{3}{4} \gamma)^2 - (\Omega_0 + m\omega_1)(\omega - \omega_L + \Omega_0 + m\omega_1)]}{[(\frac{3}{4} \gamma)^2 + (\Omega_0 + m\omega_1)^2][(\frac{3}{4} \gamma)^2 + (\omega - \omega_L + \Omega_0 + m\omega_1)^2]} \\
 & \left[+ 2 \left(\frac{\gamma}{4} \right)^2 \sum_{npp'} J_{n+p+p'} \left(\frac{\Omega_1}{\omega_1} \right) J_n \left(\frac{\Omega_1}{\omega_1} \right) \cdot \right. \\
 & \cdot \left\{ \frac{\frac{3}{4} \gamma C_{pp'}^I + (\omega - \omega_L - \Omega_0 - (n+p+p')\omega_1) C_{pp'}^II}{(\frac{3}{4} \gamma)^2 + (\omega - \omega_L - \Omega_0 - (n+p+p')\omega_1)^2} \right. \\
 & \left. + \frac{\frac{3}{4} \gamma C_{pp'}^I - (\omega - \omega_L + \Omega_0 + (n+p+p')\omega_1) C_{pp'}^II}{(\frac{3}{4} \gamma)^2 + (\omega - \omega_L + \Omega_0 + (n+p+p')\omega_1)^2} \right\} \left. \right]
 \end{aligned}$$

where

$$C_{pp'}^I = \sum_{mm'} \frac{J_m(\frac{\Omega_1}{\omega_1}) J_{m'}(\frac{\Omega_1}{\omega_1}) J_{m'-p'}(\frac{\Omega_1}{\omega_1})}{[(\frac{3}{4}\gamma)^2 + (\Omega_o + m\omega_1)^2][(\frac{3}{4}\gamma)^2 + (\Omega_o + m'\omega_1)^2]} \cdot$$

$$\cdot \left\{ (\frac{3}{4}\gamma)^2 (J_{m-p}(\frac{\Omega_1}{\omega_1}) - J_{m+p}(\frac{\Omega_1}{\omega_1})) \right.$$

$$\left. - (\Omega_o + m\omega_1)(\Omega_o + m'\omega_1)(J_{m-p}(\frac{\Omega_1}{\omega_1}) + J_{m+p}(\frac{\Omega_1}{\omega_1})) \right\}$$

and

$$C_{pp'}^II = \sum_{mm'} \frac{J_m(\frac{\Omega_1}{\omega_1}) J_{m'}(\frac{\Omega_1}{\omega_1}) J_{m'-p'}(\frac{\Omega_1}{\omega_1})}{[(\frac{3}{4}\gamma)^2 + (\Omega_o + m\omega_1)^2][(\frac{3}{4}\gamma)^2 + (\Omega_o + m'\omega_1)^2]} \cdot$$

$$\cdot \left\{ (\frac{3}{4}\gamma)(\Omega_o + m'\omega_1)(J_{m-p}(\frac{\Omega_1}{\omega_1}) - J_{m+p}(\frac{\Omega_1}{\omega_1})) \right.$$

$$\left. + (\frac{3}{4}\gamma)(\Omega_o + m\omega_1)(J_{m-p}(\frac{\Omega_1}{\omega_1}) + J_{m+p}(\frac{\Omega_1}{\omega_1})) \right\}$$

(4-88)

It turns out that for most frequencies I_3^+ and I_3^- contribute very little to the spectrum and the approximate formula obtained by dropping the terms in the square bracket is generally quite accurate.

4.4 The Total Intensity of the Emitted Light; Coherent and Incoherent Parts of the Total Intensity

By integrating Equation (4-30) for $I(\omega)$ over all (positive) frequencies ω we obtain an expression which is proportional to the total intensity of the emitted light. We can extend the integral to include the negative frequencies as well, since the spectrum is non-zero only

at optical frequencies and thus $\int_0^\infty I(\omega) d\omega = \int_{-\infty}^\infty I(\omega) d\omega$. We use the definition of the delta function and find that the total scattered intensity is

$$I_{\text{tot}} \sim \frac{2\pi}{T} \int_0^T dt \langle S_+(t) S_-(t) \rangle = \frac{2\pi}{T} \int_0^T dt \frac{1}{2} (1 + \langle S_z(t) \rangle) \quad (4-89)$$

We use Equation (4-42) with $t = t'$ and get the coherent and incoherent parts of the total scattered intensity. They are

$$I_{\text{coh}} \sim \frac{2\pi}{T} \int_0^T dt \langle S_+(t) \rangle \langle S_-(t) \rangle \quad (4-90)$$

and

$$\begin{aligned} I_{\text{inc}} &\sim \frac{2\pi}{T} \int_0^T dt \langle \delta S_+(t) \delta S_-(t) \rangle \\ &= \frac{2\pi}{T} \int_0^T dt \left\{ \frac{1}{2} (1 + \langle S_z(t) \rangle) - \langle S_+(t) \rangle \langle S_-(t) \rangle \right\} \end{aligned} \quad (4-91)$$

respectively.

In the limit of long measuring times T , we get the following results:

$$I_{\text{tot}} \sim \pi \left(1 - \frac{3}{4} \sigma^2 \sum_n \frac{J_n^2(\frac{\Omega_1}{\omega_1})}{(\frac{3}{4} \sigma)^2 + (\Omega_0 + n\omega_1)^2} \right) \quad (4-92)$$

$$\begin{aligned} I_{\text{coh}} &\sim \pi \frac{\sigma^2}{4} \left[\sum_n \frac{J_n^2(\frac{\Omega_1}{\omega_1})}{(\frac{3}{4} \sigma)^2 + (\Omega_0 + n\omega_1)^2} \right. \\ &\quad \left. - \sum_{nn'} \frac{J_n(\frac{\Omega_1}{\omega_1}) J_{n'}(\frac{\Omega_1}{\omega_1}) J_{n+n'}(\frac{2\Omega_1}{\omega_1}) \left[(\frac{3}{4} \sigma)^2 - (\Omega_0 + n\omega_1)(\Omega_0 + n'\omega_1) \right]}{\left[(\frac{3}{4} \sigma)^2 + (\Omega_0 + n\omega_1)^2 \right] \left[(\frac{3}{4} \sigma)^2 + (\Omega_0 + n'\omega_1)^2 \right]} \right] \end{aligned} \quad (4-93)$$

$$\begin{aligned}
I_{inc} \sim \pi \left[1 - \sigma^2 \sum_n \frac{J_n^2\left(\frac{\Omega_1}{\omega_1}\right)}{\left(\frac{3}{4}\sigma\right)^2 + (\Omega_0 + n\omega_1)^2} \right. \\
\left. + \frac{\sigma^2}{4} \sum_{nn'} \frac{J_n\left(\frac{\Omega_1}{\omega_1}\right) J_{n'}\left(\frac{\Omega_1}{\omega_1}\right) J_{n+n'}\left(\frac{2\Omega_1}{\omega_1}\right) \left[\left(\frac{3}{4}\sigma\right)^2 - (\Omega_0 + n\omega_1)(\Omega_0 + n'\omega_1) \right]}{\left[\left(\frac{3}{4}\sigma\right)^2 + (\Omega_0 + n\omega_1)^2 \right] \left[\left(\frac{3}{4}\sigma\right)^2 + (\Omega_0 + n'\omega_1)^2 \right]} \right]
\end{aligned}
\tag{4-94}$$

It is straightforward to show that Equations (4-93) and (4-94) can also be obtained by directly integrating Equations (4-77) and (4-88) respectively over ω . This fact justifies once again the approximation which we make in evaluating the integrals of Equation (4-64) and (4-65).

4.5 Why the Terminology "Coherent-Incoherent"?

The total scattered intensity of Equation (4-89) is proportional to the time average of the time dependent quantity $\langle S_+(t) S_-(t) \rangle$ which, in turn, is proportional to the counting rate of an ideal photo-detector placed in the field of the radiating atom²⁹.

The normalized form of the first order correlation function of the radiation field is

$$g^{(1)}(t, t') = \frac{G^{(1)}(t, t')}{[G^{(1)}(t, t) G^{(1)}(t', t')]^{\frac{1}{2}}} \tag{4-95}$$

with the first order correlation function $G^{(1)}(t, t') \sim \langle S_+(t) S_-(t') \rangle$.

A necessary condition for radiation which is coherent to first order is

$$|g^{(1)}(t, t')| = 1 \quad \forall t, t' \tag{4-96}$$

Apart from laser light even the most coherent fields in the optical range lack second and higher order coherence²⁷ and thus we only need to

check for first order coherence.

We have the relation $\langle S_+(t) S_-(t') \rangle = \langle S_+(t) \rangle \langle S_-(t') \rangle + \langle \delta S_+(t) \delta S_-(t') \rangle$. Thus $\langle S_+(t) \rangle \langle S_-(t) \rangle$ and $\langle \delta S_+(t) \delta S_-(t) \rangle$ are proportional to the counting rates of ideal photodetectors due to the parts of the field, which we termed "coherent" and "incoherent" respectively; and $\langle S_+(t) \rangle \langle S_-(t') \rangle$ and $\langle \delta S_+(t) \delta S_-(t') \rangle$ are proportional to the first-order correlation functions of the "coherently" and "incoherently" scattered radiation respectively.

For the "coherent" part of the radiation we find

$$g_{coh}^{(1)}(t, t') = \frac{\langle S_+(t) \rangle \langle S_-(t') \rangle}{[\langle S_+(t) \rangle \langle S_-(t) \rangle \langle S_+(t') \rangle \langle S_-(t') \rangle]^{\frac{1}{2}}} \\ = \frac{X_+(t) X_-(t')}{[X_+(t) X_-(t) X_+(t') X_-(t')]^{\frac{1}{2}}} \quad (4-97)$$

In the limit where $t, t' > \tau^{-1}$ this becomes

$$g_{coh}^{(1)}(t, t') = \frac{y_{\infty}(t) y_{\infty}(t')}{[y_{\infty}(t) y_{\infty}(t) y_{\infty}(t') y_{\infty}(t')]^{\frac{1}{2}}} = 1 \quad (4-98)$$

Thus this part of the radiation is indeed coherent to first order as long as the radiating atom is in the long term regime.

For the "incoherent" part of the radiation we find

$$g_{inc}^{(1)}(t, t') = \frac{\langle \delta S_+(t) \delta S_-(t') \rangle}{[\langle \delta S_+(t) \delta S_-(t) \rangle \langle \delta S_+(t') \delta S_-(t') \rangle]^{\frac{1}{2}}} \quad (4-99)$$

or

$$\begin{aligned}
g_{inc}^{(1)}(t, t') &= \frac{\frac{1}{2}}{[(1 + z_{\infty}(t) - \frac{1}{2} y_{\infty}^2(t))(1 + z_{\infty}(t') - \frac{1}{2} y_{\infty}^2(t'))]^{\frac{1}{2}}} \cdot \\
&\cdot \{ (1 + z_{\infty}(t')) e^{-\frac{\gamma}{2}(t-t')} \\
&+ \frac{1}{2} (1 + z_{\infty}(t') - y_{\infty}^2(t') - i y_{\infty}(t') - i y_{\infty}(t') z_{\infty}(t')) e^{i f_+(t, t')} \\
&+ \frac{1}{2} (1 + z_{\infty}(t') - y_{\infty}^2(t') + i y_{\infty}(t') + i y_{\infty}(t') z_{\infty}(t')) e^{i f_-(t, t')} \} \quad (4-100)
\end{aligned}$$

in the steady state. Here $|g_{inc}^{(1)}(t, t')| \neq 1$; in fact $g_{inc}^{(1)}(t, t') \rightarrow 0$ for $t - t' > \tau^{-1}$ and there is no coherence in time beyond an interval of the order of the atomic lifetime.

We expect the total radiation to exhibit partial coherence:

$$\begin{aligned}
g_{tot}^{(1)}(t, t') &= \frac{\langle S_+(t) S_-(t') \rangle}{[\langle S_+(t) S_-(t) \rangle \langle S_+(t') S_-(t') \rangle]^{\frac{1}{2}}} \\
&\approx \frac{\frac{1}{4} y_{\infty}(t) y_{\infty}(t')}{[\frac{1}{2} (1 + z_{\infty}(t)) \cdot \frac{1}{2} (1 + z_{\infty}(t'))]^{\frac{1}{2}}} \quad \begin{array}{l} t, t' > \tau^{-1} \\ t - t' > \tau^{-1} \end{array} \\
&= \sqrt{\frac{\frac{1}{4} y_{\infty}^2(t)}{\frac{1}{2} (1 + z_{\infty}(t))} \cdot \frac{\frac{1}{4} y_{\infty}^2(t')}{\frac{1}{2} (1 + z_{\infty}(t'))}} \quad (4-101)
\end{aligned}$$

Under the square root we have the ratio of the coherent counting rate to the total counting rate at the times t and t' .

5. RESULTS

5.1 The Approximations in the Numerical Calculations

The results of our calculations, namely Equations (4-77), (4-88), (4-93) and (4-94), contain single sums, double sums, triple sums and even fivefold sums over all the indices m of the Bessel functions $J_m(\frac{\Omega_1}{\omega_1})$ and $J_m(\frac{2\Omega_1}{\omega_1})$.

The relation $\Omega_0 < 3\omega_1$ expresses an experimental limitation. In the graphs which follow, the variables are taken to be multiples of ω_1 . We restrict ourselves to $0.5 \leq \frac{\Omega_0}{\omega_1} \leq 2.5$ for $\frac{\tau}{\omega_1} = 0.2$, which means $0.02 \leq \frac{\tau}{4\Omega_0} \leq 0.1$ and $0 \leq \frac{\Omega_1}{\omega_1} \leq 2.5$.

Thus, in order to obtain the data for our graphs, we have to evaluate the sums for $0 \leq \frac{\Omega_1}{\omega_1} \leq 2.5$.

For $0 \leq x \leq 5$ $|J_m(x)|$ decreases rapidly with increasing $|m|$ beyond a certain small value of $|m|$ as illustrated by Table (5-1). Because of the structure of the summands we can approximate all the infinite sums by finite sums with only a few terms.

The constant M in each figure caption indicates how many terms were kept in obtaining the data for the graph. For $M = 2$, for example, the single sums contain the terms with J_0 , $J_{\pm 1}$ and $J_{\pm 2}$. The multiple sums contain products of J_m 's such that all m 's go at least through the full range of values $0, \pm 1, \pm 2$. Some of them may go beyond that range however, due to the relations among the indices. For example, to evaluate the double sum of Equation (4-95) with the product $J_n J_{n'} J_{n+n'}$ we let $n = 0, \pm 1, \pm 2$ and $n' = 0, \pm 1, \pm 2, \pm 3, \pm 4$ to insure that

$n+n'$ takes on all the "important" values ($0, \pm 1, \pm 2$ in this case) for each fixed value of n as n' covers the chosen range.

We determine the appropriate value of M for a particular plot by listing some sample data for various values of M and finding the M for which the approximation is "good enough". This means that the data obtained when using $M+1$ and those obtained when using M differ by a factor of 10^{-4} or less of the maximum value in the set of data to be plotted. We illustrate the method with an example. Let us assume that we want to plot the coherently and incoherently scattered total intensities versus $\frac{\Omega_0}{\omega_i}$ for $\frac{\tau}{\omega_i} = 0.2$ and $\alpha = 1$ (i.e. $\Omega_0 = \Omega_i$). We compute some sample data as shown in Tables (5-2) and (5-3), namely I_{coh} and I_{inc} respectively for $\frac{\Omega_0}{\omega_i} = 0.5, 0.75, 1.0, 1.25, 1.5, 1.75, 2.0, 2.25, \text{ and } 2.5$, when $M = 0, 1, 2, 3, 4, \text{ and } 5$. (The values are $\frac{1}{2\pi}$ of the right-hand side of Equations (4-93) and (4-94) respectively.) The maximum value in the set of data is ≈ 0.5 . Thus we allow an error of no more than 0.00005. In order to get this accuracy we need to use $M = 4$ when calculating the data for the graph.

Table (5-1). Table of the Bessel functions $J_m(x)$ up to $|m| = 10$ and with arguments $x = 0, 1, 2, 3, 4, 5$. Note that $\sum_{p=-\infty}^{\infty} J_p^2(x) = 1$ for all values of x . Thus all the J_m 's for $|m| > 10$ are essentially 0 for $0 \leq x \leq 5$.

	$x = 0.0$	$x = 1.0$	$x = 2.0$	$x = 3.0$	$x = 4.0$	$x = 5.0$
$J_0(x)$	1.0	.76520	.22389	-.26005	-.39715	-.17760
$J_{\pm 1}(x)$	0.0	$\pm .44005$	$\pm .57672$	$\pm .33906$	$\mp .06604$	$\mp .32758$
$J_{\pm 2}(x)$	0.0	.11490	.35283	.48609	.36413	.04657
$J_{\pm 3}(x)$	0.0	$\pm .01956$	$\pm .12894$	$\pm .30906$	$\pm .43017$	$\pm .36483$
$J_{\pm 4}(x)$	0.0	.00248	.03400	.13203	.28113	.39123
$J_{\pm 5}(x)$	0.0	$\pm .00025$	$\pm .00704$	$\pm .04303$	$\pm .13209$	$\pm .26114$
$J_{\pm 6}(x)$	0.0	.00002	.00120	.01139	.04909	.13105
$J_{\pm 7}(x)$	0.0	$\pm .00000$	$\pm .00017$	$\pm .00255$	$\pm .01518$	$\pm .05338$
$J_{\pm 8}(x)$	0.0	.00000	.00002	.00049	.00403	.01841
$J_{\pm 9}(x)$	0.0	$\pm .00000$	$\pm .00000$	$\pm .00008$	$\pm .00094$	$\pm .00552$
$J_{\pm 10}(x)$	0.0	.00000	.00000	.00001	.00020	.00147
$\sum_{m=-10}^{10} J_m^2(x)$	1.0	.99999	.99998	.99999	1.00000	1.00000

Table (5-2). The coherently scattered total intensity
for $\frac{I}{\omega_i} = 0.2$, $\alpha = 1.0$ and $0.5 \leq \frac{\Omega_i}{\omega_i} \leq 2.5$
when $M = 0, 1, 2, 3, 4$, and 5 .

$\frac{\Omega_i}{\omega_i}$	$M = 0$	$M = 1$	$M = 2$	$M = 3$	$M = 4$	$M = 5$
0.50	.02648	.02587	.02590	.02590	.02590	.02590
0.75	.00940	.01279	.01296	.01296	.01296	.01296
1.00	.00348	.03079	.03078	.03078	.03078	.03078
1.25	.00125	.02444	.02343	.02334	.02334	.02334
1.50	.00043	.01080	.01006	.00994	.00993	.00993
1.75	.00014	.00541	.00795	.00784	.00782	.00782
2.00	.00004	.00285	.02202	.02184	.02182	.02182
2.25	.00000	.00151	.01517	.01452	.01449	.01449
2.50	.00000	.00079	.00703	.00662	.00655	.00655

Table (5-3). The incoherently scattered total intensity
for $\frac{I}{\omega_i} = 0.2$, $\alpha = 1.0$ and $0.5 \leq \frac{\Omega_i}{\omega_i} \leq 2.5$
when $M = 0, 1, 2, 3, 4$, and 5 .

$\frac{\Omega_i}{\omega_i}$	$M = 0$	$M = 1$	$M = 2$	$M = 3$	$M = 4$	$M = 5$
0.50	.42504	.42203	.42200	.42200	.42200	.42200
0.75	.47145	.44594	.44572	.44572	.44572	.44572
1.00	.48793	.33080	.33060	.33060	.33060	.33060
1.25	.49480	.42484	.42505	.42513	.42513	.42513
1.50	.49784	.46959	.46730	.46739	.46740	.46740
1.75	.49920	.48463	.46675	.46677	.46679	.46679
2.00	.49978	.49153	.38925	.38917	.38919	.38919
2.25	.49998	.49520	.45250	.45238	.45240	.45240
2.50	.49999	.49727	.47993	.47774	.47777	.47777

5.2 The Population Inversion and the Atomic Dipole Moment in the Steady State

The results for the constant amplitude dynamic Stark effect are listed in the Appendix. They are well-established theoretically and have been verified experimentally to some extent. They can be used to check our results since the constant amplitude case is contained in our more general calculations. When making comparisons we have to keep in mind that our results are only valid for $\frac{\gamma}{4\Omega_0} \ll 1$. In addition, comparison of the constant amplitude dynamic Stark effect and the modulated amplitude dynamic Stark effect reveals which features stem from the modulation.

At resonance, the first one of the optical Bloch equations does not depend on the applied field and thus has the same solution regardless of the time dependence of the amplitude. The long term result $x_\infty = 0$ is easy to understand: A classical dipole oscillator, which is allowed to radiate and which is driven by a field whose frequency equals its natural frequency, is $\frac{\pi}{2}$ out of phase with that field.

For the constant amplitude dynamic Stark effect the long term solutions y_∞ and z_∞ are time independent. The function y_∞ of Equation (A-5) is plotted in Figure (5-1) for $0 \leq \frac{\Omega_0}{\omega_1} \leq 2.5$ and $\frac{\gamma}{\omega_1} = 0.2$. Similarly, the function z_∞ of Equation (A-6) is plotted in Figure (5-2) for $0 \leq \frac{\Omega_0}{\omega_1} \leq 2.5$ and $\frac{\gamma}{\omega_1} = 0.2$.

All our results simplify considerably when $a = 0$, since

$J_n(0) = \delta_{0,n}$. In particular

$$y_\infty(a=0) = \frac{\gamma \Omega_0}{(\frac{3}{4}\gamma)^2 + \Omega_0^2} \quad (5-1)$$

and

$$z_{\infty}(a=0) = - \frac{\frac{3}{4}\gamma^2}{(\frac{3}{4}\gamma)^2 + \Omega_o^2} \quad (5-2)$$

In the limit where $\frac{\gamma}{4\Omega_o} \ll 1$ Equations (5-1) and (5-2) agree with Equations (A-5) and (A-6) if terms of the order of $(\frac{\gamma}{\Omega_o})^2$ are dropped. Then

$y_{\infty} \approx \frac{\gamma}{\Omega_o}$ and $z_{\infty} \approx 0$ in both cases. The degree of accuracy of our results for $a = 0$ is demonstrated by Figures (5-3) and (5-4). In Figure (5-3) the functions y_{∞} of Equation (5-1) (solid line) and y_{∞} of Equation (A-5) (dashed line) are plotted versus $\frac{\Omega_o}{\omega_1}$. Similarly, in Figure (5-4) the functions z_{∞} of Equation (5-2) and z_{∞} of Equation (A-6) are plotted versus $\frac{\Omega_o}{\omega_1}$. As expected the agreement is much better for $\frac{\Omega_o}{\omega_1} = 2.5$ where $\frac{\gamma}{4\Omega_o} = 0.02$ than for $\frac{\Omega_o}{\omega_1} = 0.5$ where $\frac{\gamma}{4\Omega_o} = 0.1$.

For $a \neq 0$ the functions $y_{\infty}(t)$ and $z_{\infty}(t)$ of Equations (3-99) and (3-100) respectively become time dependent. They oscillate with all harmonics of the modulation frequency. This is illustrated by Figure (5-5) where $z_{\infty}(t)$ is plotted as a function of time for various values of a ; $\frac{\gamma}{\omega_1} = 0.2$ and $\frac{\Omega_o}{\omega_1} = 1.0$ are kept constant. The amplitude of the oscillations increases with increasing a , otherwise the features of the curves are rather similar. It seems reasonable then to choose $a = 1$ for the modulation depth, since the effects of the modulation are most obvious in that case. The function $y_{\infty}(t)$ is plotted for $a = 1$, $\frac{\gamma}{\omega_1} = 0.2$ and various values of $\frac{\Omega_o}{\omega_1}$ in Figure (5-6). The corresponding plots for $z_{\infty}(t)$ are shown in Figure (5-7). With increasing values of $\frac{\Omega_o}{\omega_1}$ a greater number of the harmonics contribute significantly to the time dependence, which becomes increasingly complicated.

The amplitude of the oscillations goes through a maximum when $\Omega_o = n\omega_1$ (n integer). This is attributed to parametric resonances^{30,31}.

The time average of the atomic dipole moment $y_\infty(t)$ is just the static component of $y_\infty(t)$, the time average of the inversion $z_\infty(t)$ is the static component of $z_\infty(t)$. These static components are the quantities which should be compared with y_∞ and z_∞ of the constant amplitude dynamic Stark effect. The static components of $y_\infty(t)$ and $z_\infty(t)$ as functions of $\frac{\Omega_o}{\omega_1}$ are shown as solid lines in Figures (5-8) and (5-9) respectively; the dashed lines are once again the corresponding curves of Figures (5-1) and (5-2). In the constant amplitude case the inversion steadily approaches zero as Ω_o increases toward saturating values. In the modulated amplitude case, the static component of the inversion exhibits parametric resonances, i.e. it reaches zero over a set of maxima and minima. Similar remarks hold true for the atomic dipole moment.

FIGURE CAPTIONS

Figure (5-1): The atomic dipole moment y_{∞} as a function of $\frac{\Omega_0}{\omega_1}$ for $\frac{\tau}{\omega_1} = 0.2$ in the case of the constant amplitude dynamic Stark effect.

Figure (5-2): The atomic inversion z_{∞} as a function of $\frac{\Omega_0}{\omega_1}$ for $\frac{\tau}{\omega_1} = 0.2$ in the case of the constant amplitude dynamic Stark effect.

Figure (5-3): The atomic dipole moment for the constant amplitude dynamic Stark effect (dashed line) and for the modulated amplitude dynamic Stark effect (solid line) with $Q = 0$. Both are plotted as functions of $\frac{\Omega_0}{\omega_1}$ for $\frac{\tau}{\omega_1} = 0.2$.

Figure (5-4): The atomic inversion for the constant amplitude dynamic Stark effect (dashed line) and for the modulated amplitude dynamic Stark effect (solid line) with $Q = 0$. Both are plotted as functions of $\frac{\Omega_0}{\omega_1}$ for $\frac{\tau}{\omega_1} = 0.2$.

Figure (5-5): The inversion $z_{\infty}(t)$ of the modulated amplitude dynamic Stark effect as a function of time in the interval $0 \leq \frac{\omega_1 t}{2\pi} \leq 3$, for $\frac{\tau}{\omega_1} = 0.2$, $M = 4$ and various values of the modulation depth Q .

Figure (5-6): The atomic dipole moment $y_{\infty}(t)$ of the modulated amplitude dynamic Stark effect as a function of time in the interval $0 \leq \frac{\omega_1 t}{2\pi} \leq 2$, for $Q = 1$, $\frac{\tau}{\omega_1} = 0.2$, $M = 4$ and

various values of $\frac{\Omega_0}{\omega_1}$. The y-axes go from -1 to +1, the numbers at the bottom of the graphs give the respective values of $\frac{\Omega_0}{\omega_1}$.

Figure (5-7): The atomic inversion $z_\infty(t)$ of the modulated amplitude dynamic Stark effect as a function of time in the interval $0 \leq \frac{\omega_1 t}{2\pi} \leq 2$, for $\alpha = 1$, $\frac{\tau}{\omega_1} = 0.2$, $M = 4$ and various values of $\frac{\Omega_0}{\omega_1}$. The y-axes go from -1 to +1, the numbers at the bottom of the graphs give the respective values of $\frac{\Omega_0}{\omega_1}$.

Figure (5-8): The static component of the atomic dipole moment $y_\infty(t)$ of the modulated amplitude dynamic Stark effect (solid line; $\alpha = 1$, $M = 4$) and the atomic dipole moment y_∞ of the constant amplitude dynamic Stark effect (dashed line). Both are plotted as a function of $\frac{\Omega_0}{\omega_1}$ for $\frac{\tau}{\omega_1} = 0.2$.

Figure (5-9): The static component of the atomic inversion $z_\infty(t)$ of the modulated amplitude dynamic Stark effect (solid line; $\alpha = 1$, $M = 4$) and the atomic inversion z_∞ of the constant amplitude dynamic Stark effect (dashed line). Both are plotted as a function of $\frac{\Omega_0}{\omega_1}$ for $\frac{\tau}{\omega_1} = 0.2$.

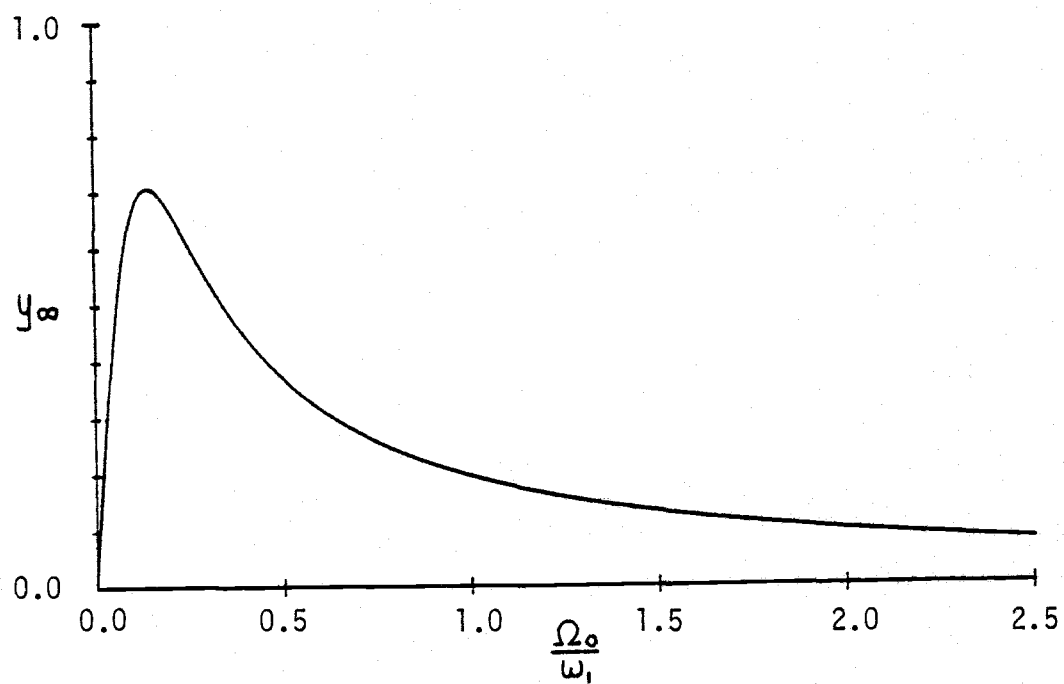


Figure (5-1)

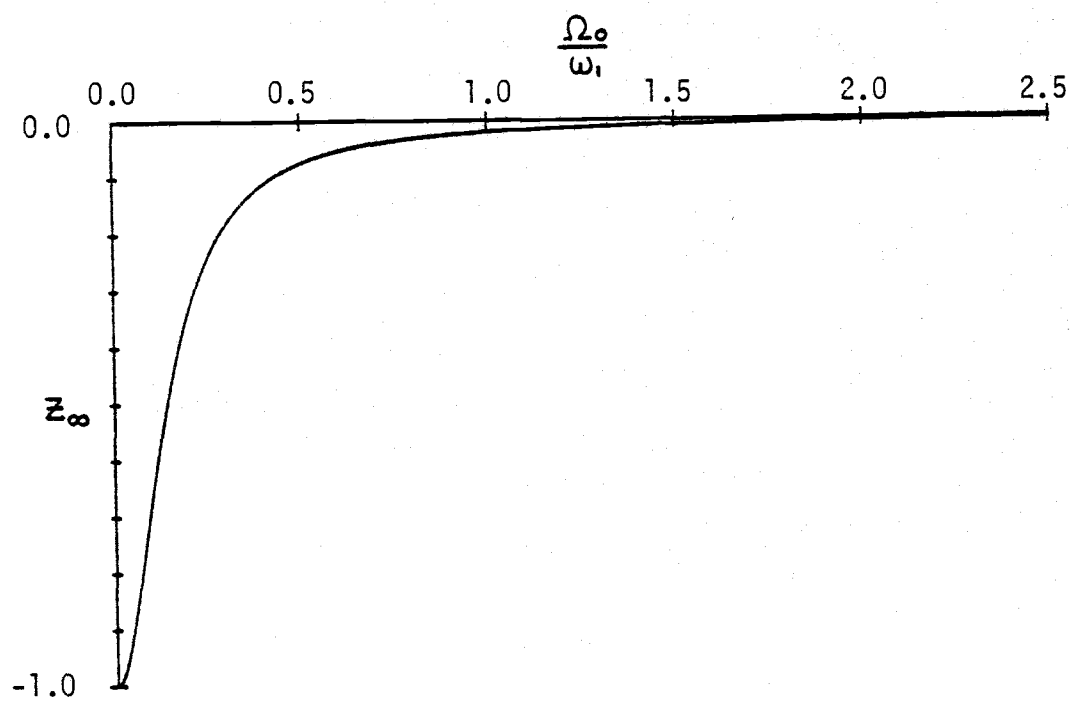


Figure (5-2)

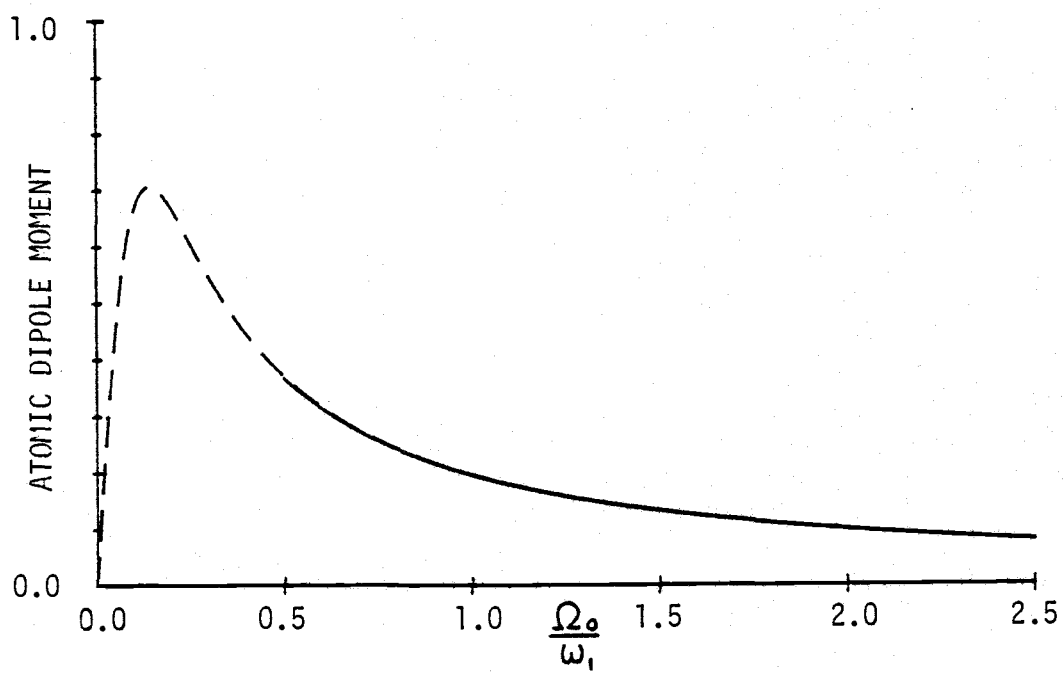


Figure (5-3)

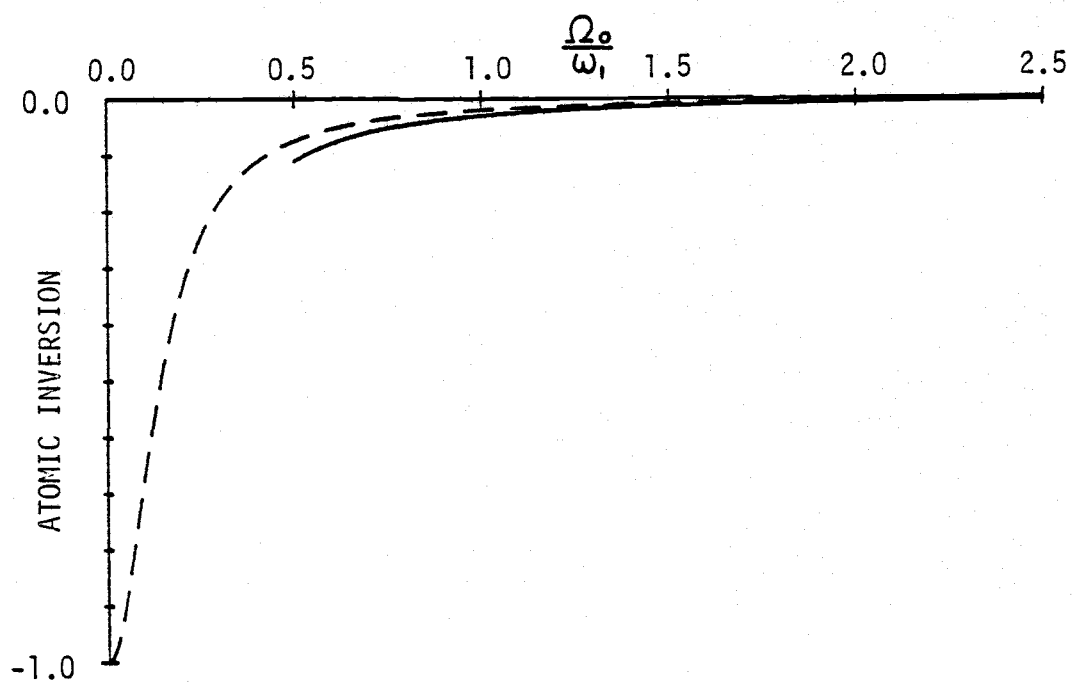


Figure (5-4)

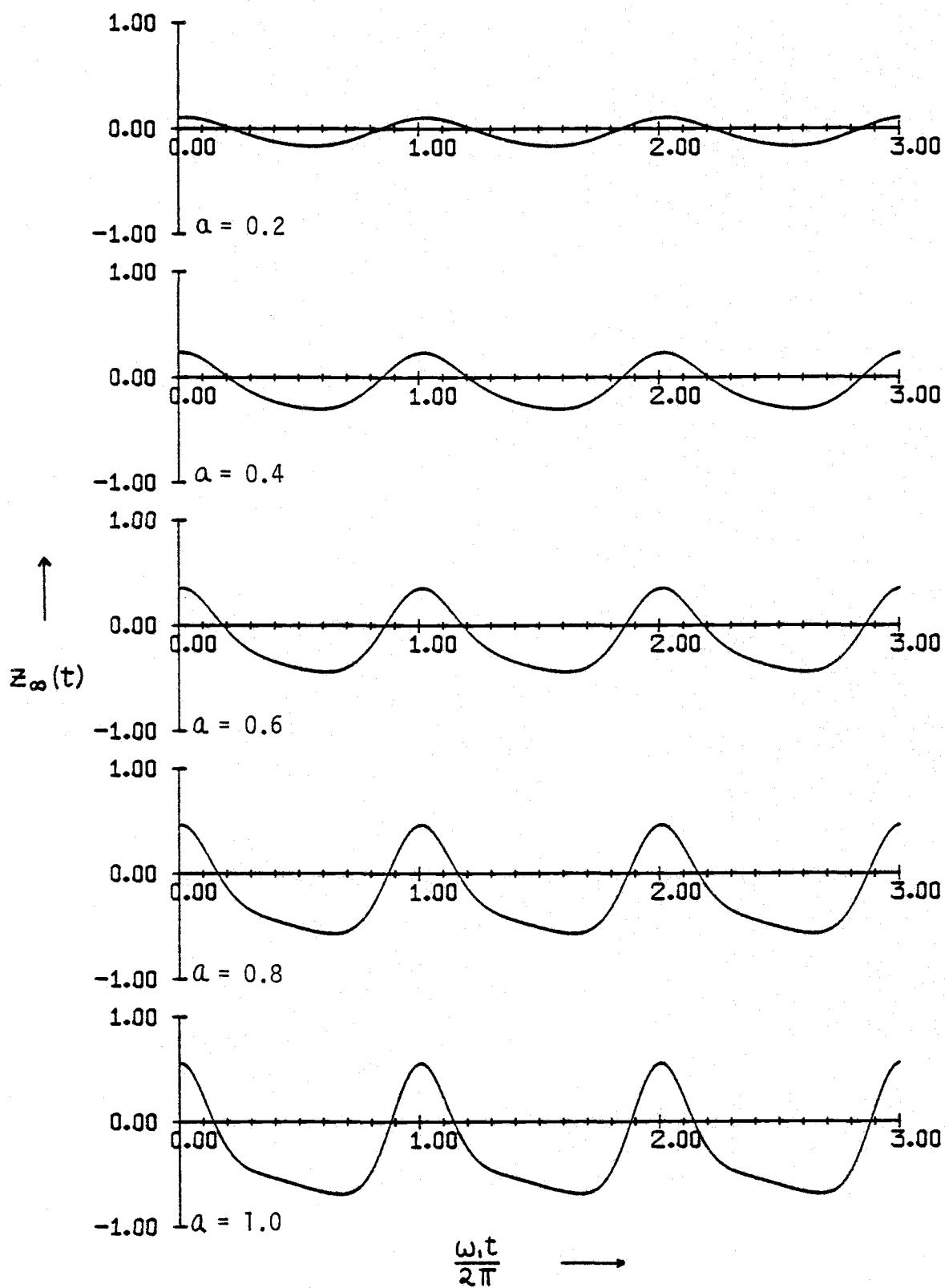


Figure (5-5)

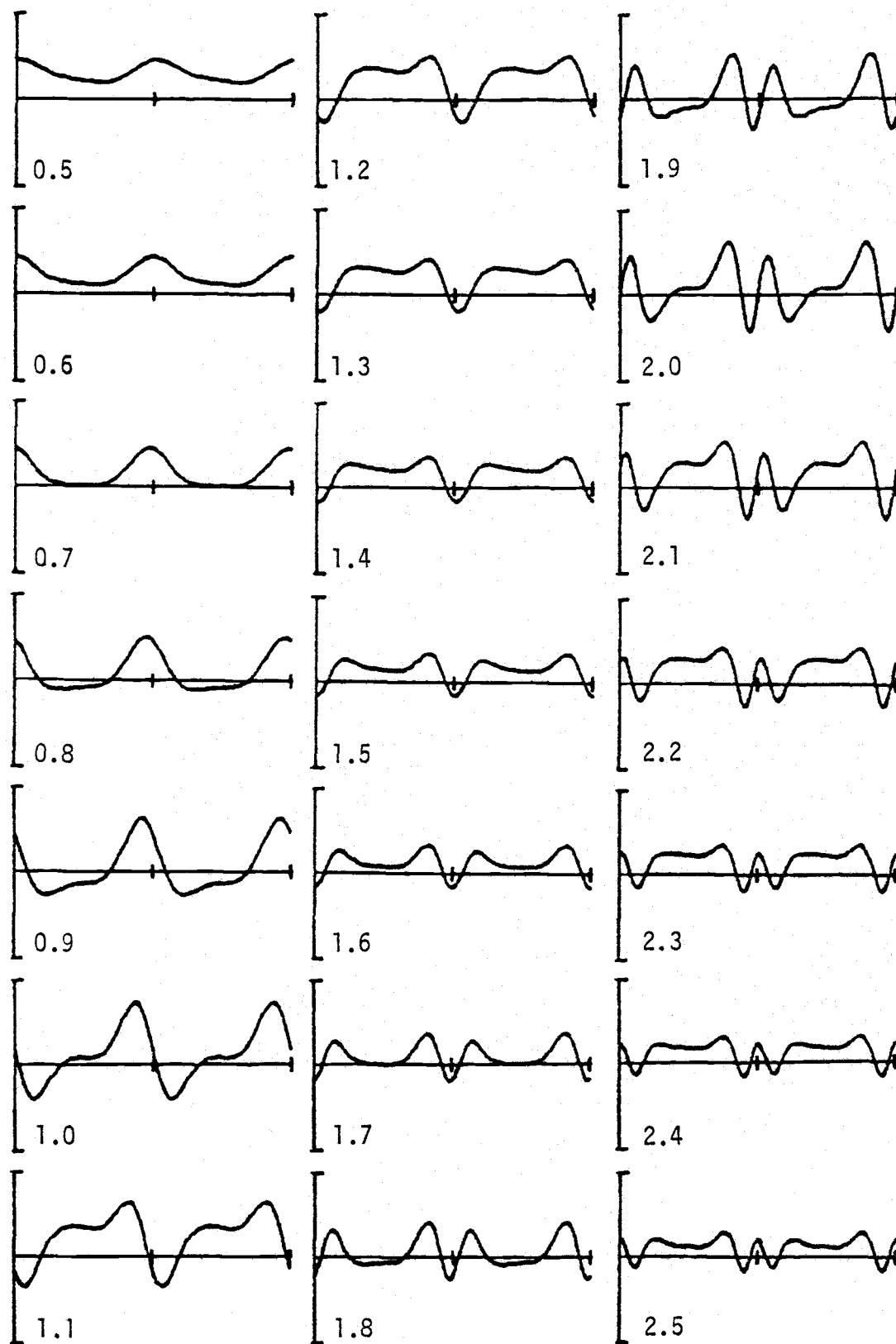


Figure (5-6)

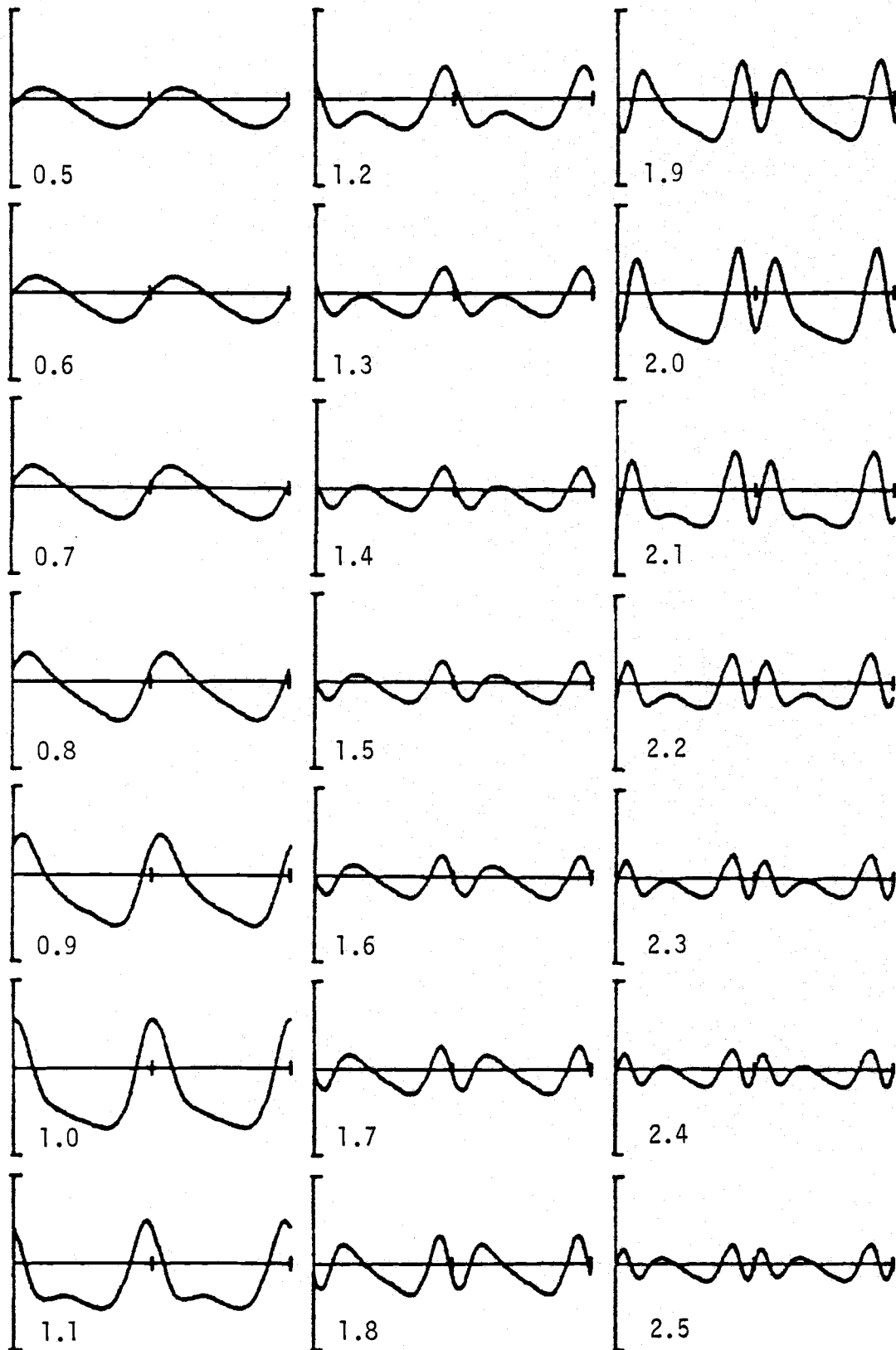


Figure (5-7)

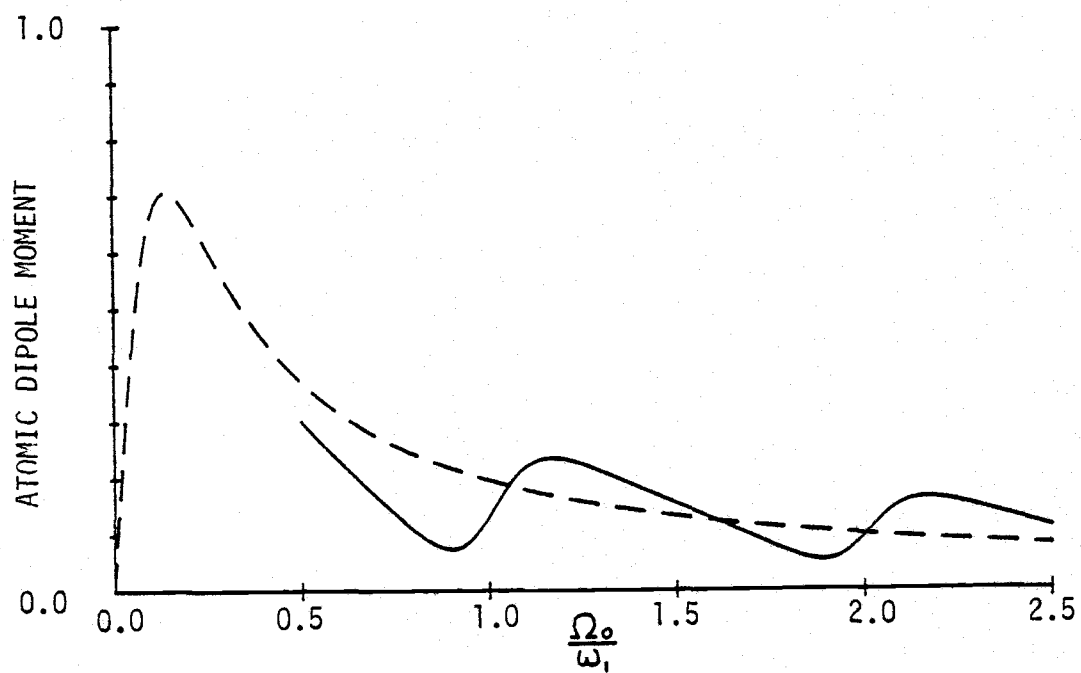


Figure (5-8)

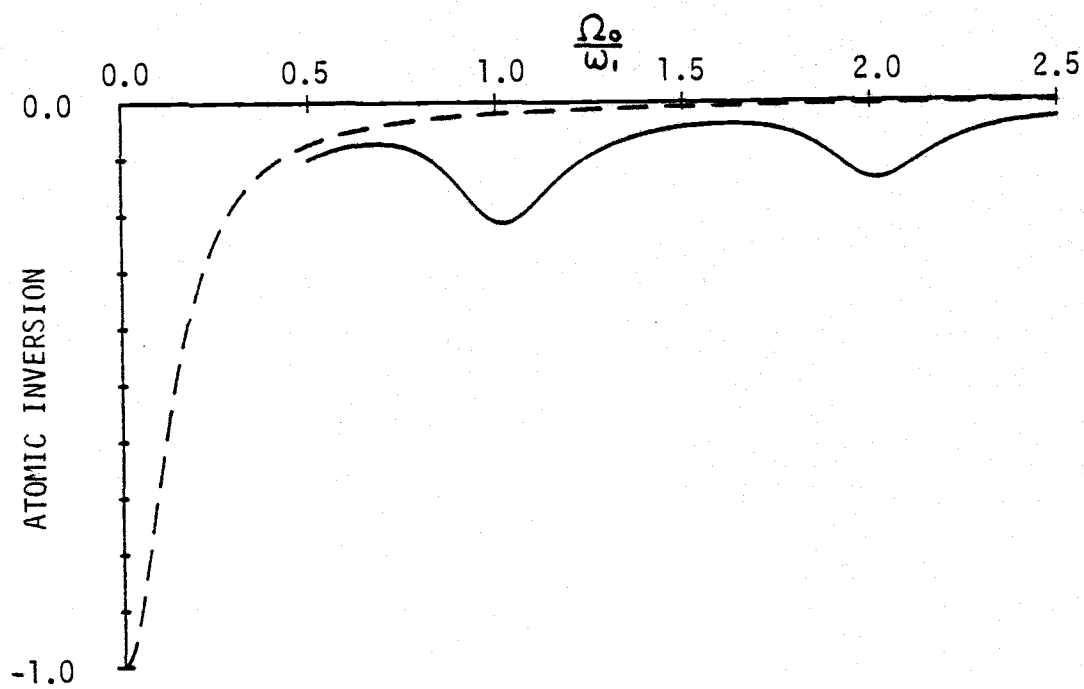


Figure (5-9)

5.3 The Intensities

The formulas which we use to calculate the scattered intensities are formally identical to the ones used for the constant amplitude dynamic Stark effect. The total scattered intensity, for example, is obtained from the relation $I_{\text{tot}} \sim \frac{2\pi}{T} \int_0^T dt \frac{1}{2}(1 + z_{\infty}(t))$ and will, in the modulated amplitude case, exhibit parametric resonances just like the static component of the inversion. This is illustrated in Figure (5-10) where the total intensity and its coherent and incoherent parts are plotted using Equations (4-92), (4-93) and (4-94) with $\alpha = 1$ (solid curves) and the corresponding equations of the Appendix (dashed curves). The decay rate is $\frac{\Gamma}{\omega_i} = 0.2$ in all cases. We can determine the average Rabi frequency Ω_0 and thus the strength of the atom-field interaction by measuring the total intensity of the scattered light as a function of the mean field strength and finding the minima of the resulting curve. For the constant amplitude dynamic Stark effect this is not possible. The coherently scattered intensity has the same functional dependence on $\frac{\Omega_0}{\omega_i}$ as the static component of $y_{\infty}^2(t)$ since $I_{\text{coh}} \sim \frac{2\pi}{T} \int_0^T dt \frac{1}{4} y_{\infty}^2(t)$; the incoherently scattered intensity is linked with the static components of both $z_{\infty}(t)$ and $y_{\infty}^2(t)$ since $I_{\text{inc}} \sim \frac{2\pi}{T} \int_0^T dt \left\{ \frac{1}{2}(1 + z_{\infty}(t)) - \frac{1}{4} y_{\infty}^2(t) \right\}$.

When Ω_0 reaches saturating values, almost all the light is scattered incoherently in both the constant amplitude and the modulated amplitude case. Moreover, the incoherently scattered intensity in the modulated amplitude case has minima at the same values of $\frac{\Omega_0}{\omega_i}$ as the static component of the inversion. We recall that the incoherently scattered light has its origin in the fluctuations of the atomic varia-

bles. It thus appears that the atoms fluctuate more when they are in the excited state.

We can rewrite I_{tot} as

$$I_{\text{tot}} \sim \frac{2\pi}{T} \int_0^T S_{jj}(t) dt \quad (5-3)$$

The time average of the probability $S_{jj}(t)$ cannot exceed 0.5 and thus there is an upper limit to the scattered intensity. The few authors who have treated aspects of the modulated amplitude dynamic Stark effect^{30,31,32,33} all make use of the fact that the intensity of the light scattered by an atom is proportional to the population of its upper level. Consequently, they have to derive the function $S_{jj}(t)$ or $z(t)$.

Armstrong and Feneuille³² treat the case of a two-level atom at resonance with a monochromatic, weakly modulated laser beam using perturbation theory with the modulation depth a as the expansion parameter. Their results are valid for arbitrary field strengths and for $a \ll 1$. Their density matrix element $S_{jj}(t)$, expressed in our notation, is

$$S_{jj}(t) = \frac{\Omega_o^2}{2\Omega_o^2 + \sigma^2} \left\{ 1 + \frac{4a\sigma}{(2\omega_1^2 - \sigma^2 - 2\Omega_o^2)^2 + 9\sigma^2\omega_1^2} \cdot \right. \\ \left. \cdot \left[\sigma \left(\frac{1}{2}\sigma^2 + \Omega_o^2 + \frac{1}{2}\omega_1^2 \right) \cos \omega_1 t - \omega_1 (\Omega_o^2 - \omega_1^2 - \sigma^2) \sin \omega_1 t \right] \right\} \quad (5-4)$$

In Figure (5-11) we compare Equation (5-4) to our function

$S_{jj}(t) = \frac{1}{2}(1 + z_\infty(t))$. We see that the time dependence of the two functions agrees quite well, but that they differ by a small constant term.

The assumptions made by Feneuille, Schweighofer and Oliver³³ are such that a comparison between their results and ours is not possible.

FIGURE CAPTIONS

Figure (5-10): The intensities I_{coh} , I_{inc} and I_{tot} for the constant amplitude dynamic Stark effect (dashed lines) and for the modulated amplitude dynamic Stark effect (solid lines, $Q = 1$, $M = 5$). They are plotted as functions of $\frac{\Omega_0}{\omega_1}$ for $\frac{\Gamma}{\omega_1} = 0.2$.

Figure (5-11): The density matrix element $S_{jj}(t)$ as a function of time in the interval $0 \leq \frac{\omega_1 t}{2\pi} \leq 2$ for our calculation (solid lines) and for the calculation done by Armstrong and Feneuille (dashed lines). The respective values of Q and $\frac{\Omega_0}{\omega_1}$ are given at the lower left hand corner of each graph.

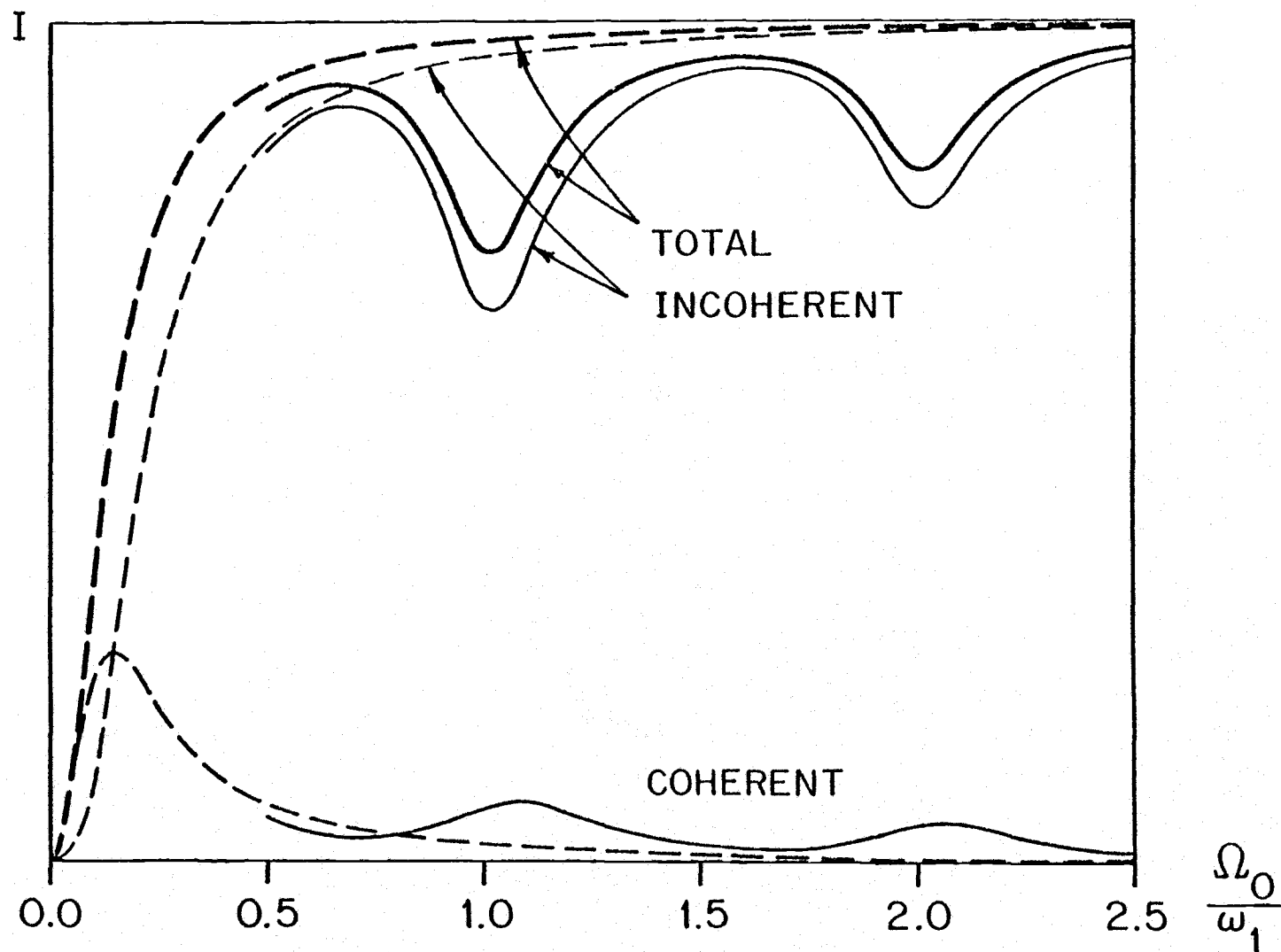


Figure (5-10)

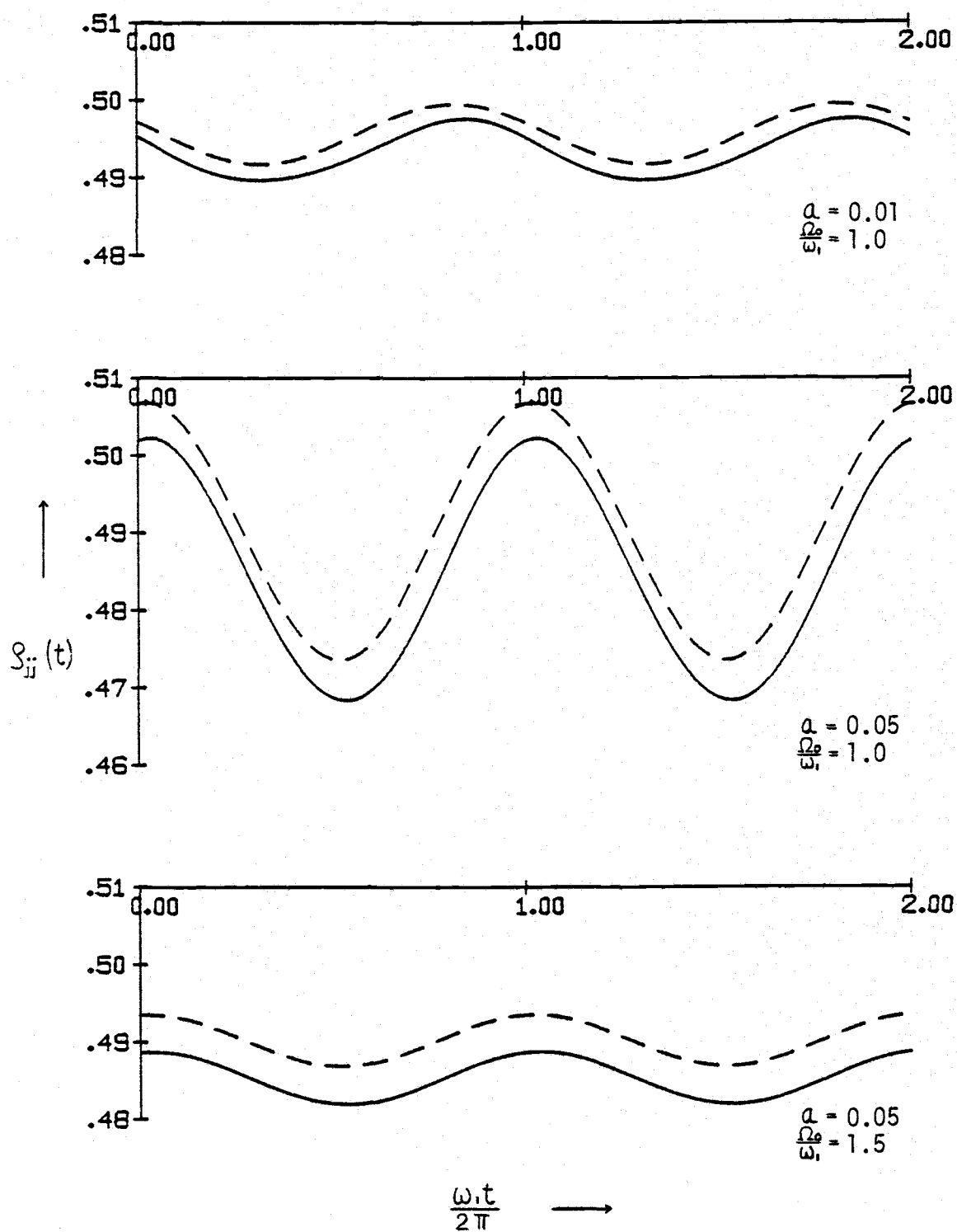


Figure (5-11)

5.4 The Coherent Spectrum

The coherent spectrum in the case of the constant amplitude dynamic Stark effect consists of a sharp (δ function) peak at $\omega = \omega_L$; the coherently scattered light is scattered elastically.

In the modulated amplitude case additional frequencies appear at $\omega = \omega_L + m\omega_1$, (m integer). This is illustrated by Figure (5-12), where the length of the line at $\omega = \omega_L + m\omega_1$ represents the factor $y_m y_m^*$ which appears with $\delta(\omega - \omega_L - m\omega_1)$ in Equation (4-76); i.e. it is proportional to the intensity of the light with frequency $\omega = \omega_L + m\omega_1$. We note that the coherent spectrum is symmetric about $\omega = \omega_L$ since $y_{-m} y_{-m}^* = y_m^* y_m$.

For $a = 0$ Equation (4-77) reduces to

$$I_{coh}(\omega) \sim \frac{\pi}{2} \frac{\sigma^2 \Omega_o^2}{((\frac{3}{4}\sigma)^2 + \Omega_o^2)^2} \delta(\omega - \omega_L) \quad (5-5)$$

which is in good agreement with Equation (A-14).

FIGURE CAPTION

Figure (5-12): The coherent part of the spectrum, $I_{\text{coh}}(\omega)$, of the modulated amplitude dynamic Stark effect as a function of $\frac{\omega - \omega_L}{\omega_L}$ for $Q = 1$, $\frac{\gamma}{\omega_L} = 0.2$, $M = 4$ and $\frac{\Omega_0}{\omega_L} = 0.5$, 1.0, 1.5, 2.0, and 2.5.

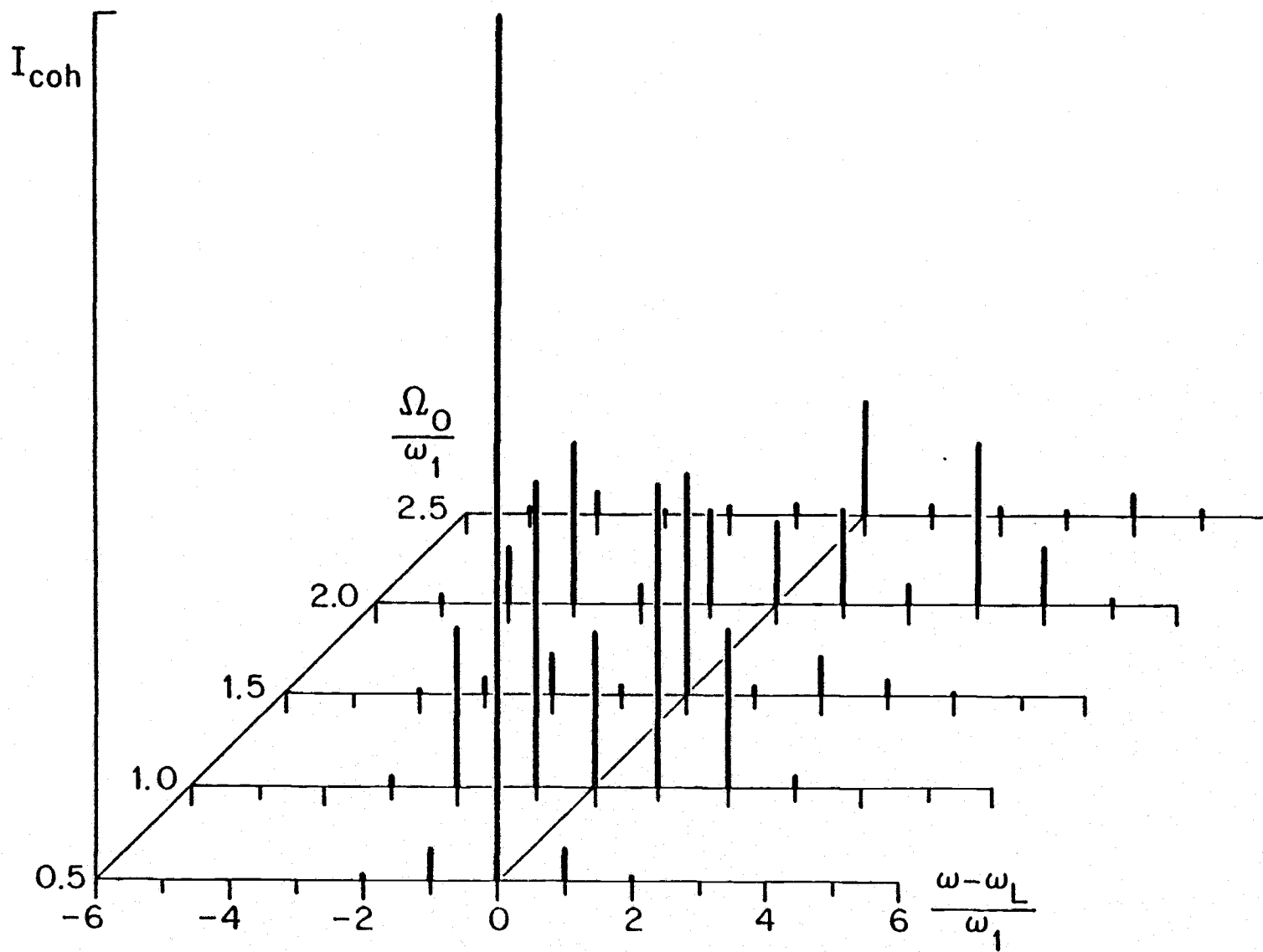


Figure (5-12)

5.5 The Incoherent Spectrum

The feature which is usually associated with the constant amplitude dynamic Stark effect is the spectrum of the incoherently scattered light. It is a three-peaked structure which is symmetric about ω_L . In the strong field limit the peaks are at $\omega = \omega_L$ and at $\omega = \omega_L \pm \Omega_0$ and have a half width of $\frac{\gamma}{2}$ and $\frac{3}{4}\gamma$ respectively. The height of the central peak is three times greater than that of each side band. A set of such curves is plotted in Figure (5-13) for $\frac{\Omega_0}{\omega_1} = 0.5, 1.0, 1.5, 2.0$, and 2.5 and $\frac{\gamma}{\omega_1} = 0.2$ using Equation (A-15).

In Figures (5-14) and (5-15) our results for $Q = 0$ (solid curves) are plotted along with the constant amplitude results in the strong field limit (dashed curves) for $\frac{\gamma}{\omega_1} = 0.2$ and $\frac{\Omega_0}{\omega_1} = 0.5$ and 2.5 respectively. The agreement for $\frac{\Omega_0}{\omega_1} = 2.5$ is considerably better than that for $\frac{\Omega_0}{\omega_1} = 0.5$.

The spectral distribution of Equation (A-16) is correct for all Ω_0 provided $\Omega_0 \geq \frac{\gamma}{4}$. In Figures (5-16) and (5-17) we compare our results for $Q = 0$ (solid curves) with the right-hand side of Equation (A-20) for $\frac{\gamma}{\omega_1} = 0.2$ and $\frac{\Omega_0}{\omega_1} = 0.5$ and 2.5 respectively. The agreement for $\frac{\Omega_0}{\omega_1} = 0.5$ is better as before and for $\frac{\Omega_0}{\omega_1} = 2.5$ it is very good.

In the modulated amplitude case the incoherent spectrum has peaks at $\omega = \omega_L$ and $\omega = \omega_L \pm \Omega_0 + m\omega_1$ (m integer). Thus, the main effect of the modulation is that the sidebands are now accompanied by their own sidebands whereas the central peak is essentially unaffected. This can be justified by examining the origin of the different peaks in the incoherent spectrum. The width and position of the central peak

are determined by the transient behavior of X , which is independent of the applied field. On the other hand, the widths and positions of the sidebands are determined by the transient behavior of y , and z , which for $Q \neq 0$ contains the frequencies $\Omega_0 + m\omega_1$ and $-\Omega_0 + m\omega_1$.

In Figure (5-18) we show a set of spectra corresponding to $a = 1$, $\frac{\Gamma}{\omega_1} = 0.2$ and $\frac{\Omega_0}{\omega_1} = 0.5, 1.0, 1.5, 2.0$ and 2.5 . These curves exhibit the expected peaks at $\omega = \omega_L$ and $\omega = \omega_L \pm \Omega_0 + m\omega_1$, but some peculiarities do occur. For example, for $\frac{\Omega_0}{\omega_1} = 2.5$, we would expect peaks at $\omega = \omega_L \pm 2.5\omega_1$, but they are missing. The reason for this is that the factor $J_0^2(a \frac{\Omega_0}{\omega_1})$ which appears in the terms for the peaks at $\omega = \omega_L \pm \Omega_0$ is very small for this particular combination of modulation depth and Rabi frequency: $J_0(2.5) = -0.04838$,

$$J_0(2.40483) = 0.$$

The incoherent spectrum is also symmetric about $\omega = \omega_L$. This can be shown by replacing $\omega - \omega_L$ by $-(\omega - \omega_L)$ in Equation (4-90). This yields an expression which is identical with the original one.

The positions of the peaks in the incoherent spectrum can be determined using the dressed atom picture³⁵.

FIGURE CAPTIONS

Figure (5-13): The incoherent part of the spectrum of the constant amplitude dynamic Stark effect in the strong field limit as a function of $\frac{\omega - \omega_L}{\omega_1}$ for $\frac{\gamma}{\omega_1} = 0.2$ and $\frac{\Omega_0}{\omega_1} = 0.5, 1.0, 1.5, 2.0, \text{ and } 2.5$.

Figure (5-14): The incoherent part of the spectrum for the modulated amplitude dynamic Stark effect with $\alpha = 0$ (solid line) and for the constant amplitude dynamic Stark effect in the strong field limit (dashed line). Both are plotted as functions of $\frac{\omega - \omega_L}{\omega_1}$ for $\frac{\Omega_0}{\omega_1} = 0.5$ and $\frac{\gamma}{\omega_1} = 0.2$.

Figure (5-15): Same as Figure (5-14) but with $\frac{\Omega_0}{\omega_1} = 2.5$.

Figure (5-16): The incoherent part of the spectrum for the modulated amplitude dynamic Stark effect with $\alpha = 0$ (solid line) and for the constant amplitude dynamic Stark effect (dashed line). Both are plotted as functions of $\frac{\omega - \omega_L}{\omega_1}$ for $\frac{\Omega_0}{\omega_1} = 0.5$ and $\frac{\gamma}{\omega_1} = 0.2$.

Figure (5-17): Same as Figure (5-16) but with $\frac{\Omega_0}{\omega_1} = 2.5$.

Figure (5-18): The incoherent part of the spectrum of the modulated amplitude dynamic Stark effect as a function of $\frac{\omega - \omega_L}{\omega_1}$ for $\alpha = 1$, $M = 5$, $\frac{\gamma}{\omega_1} = 0.2$ and $\frac{\Omega_0}{\omega_1} = 0.5, 1.0, 1.5, 2.0, \text{ and } 2.5$.

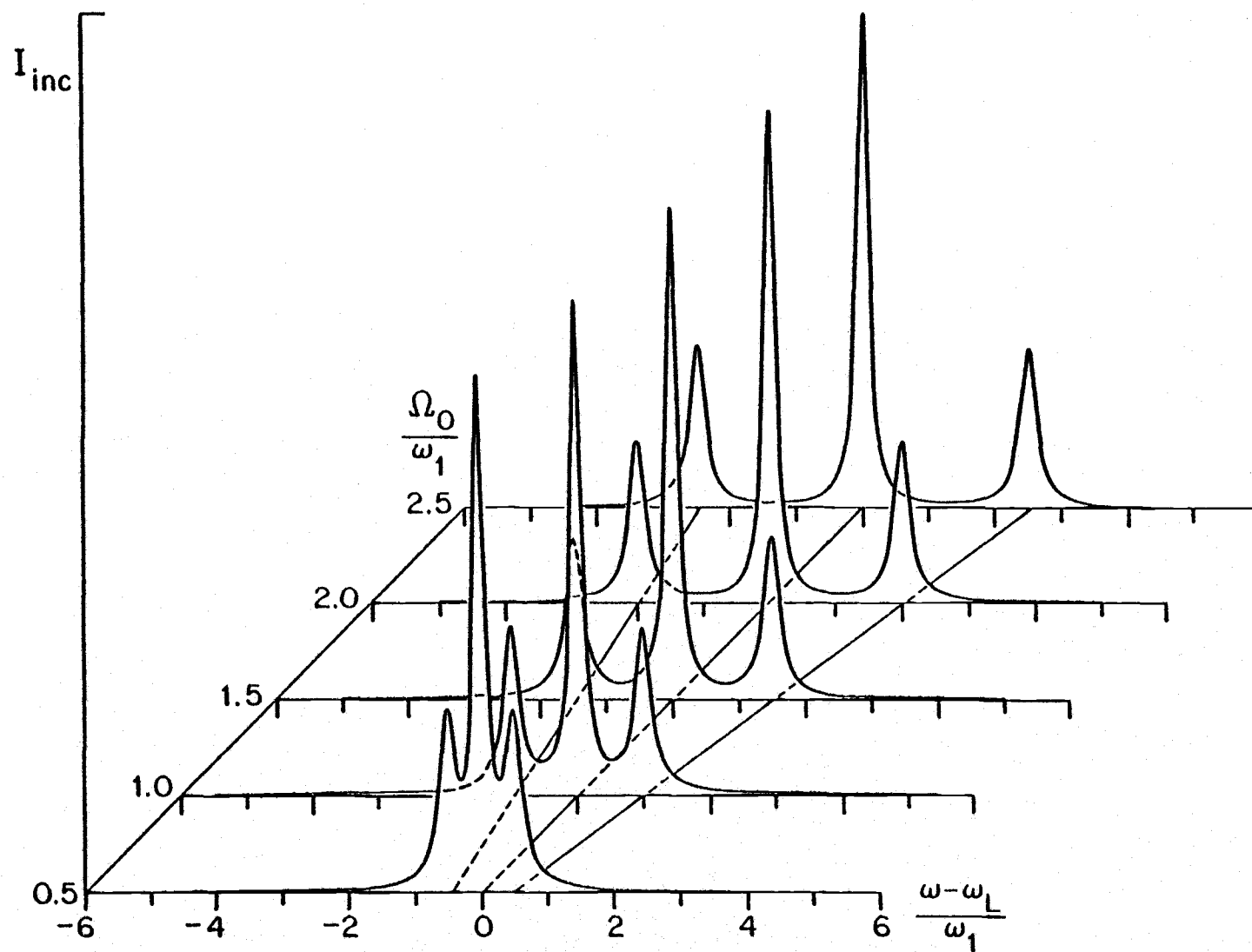


Figure (5-13)

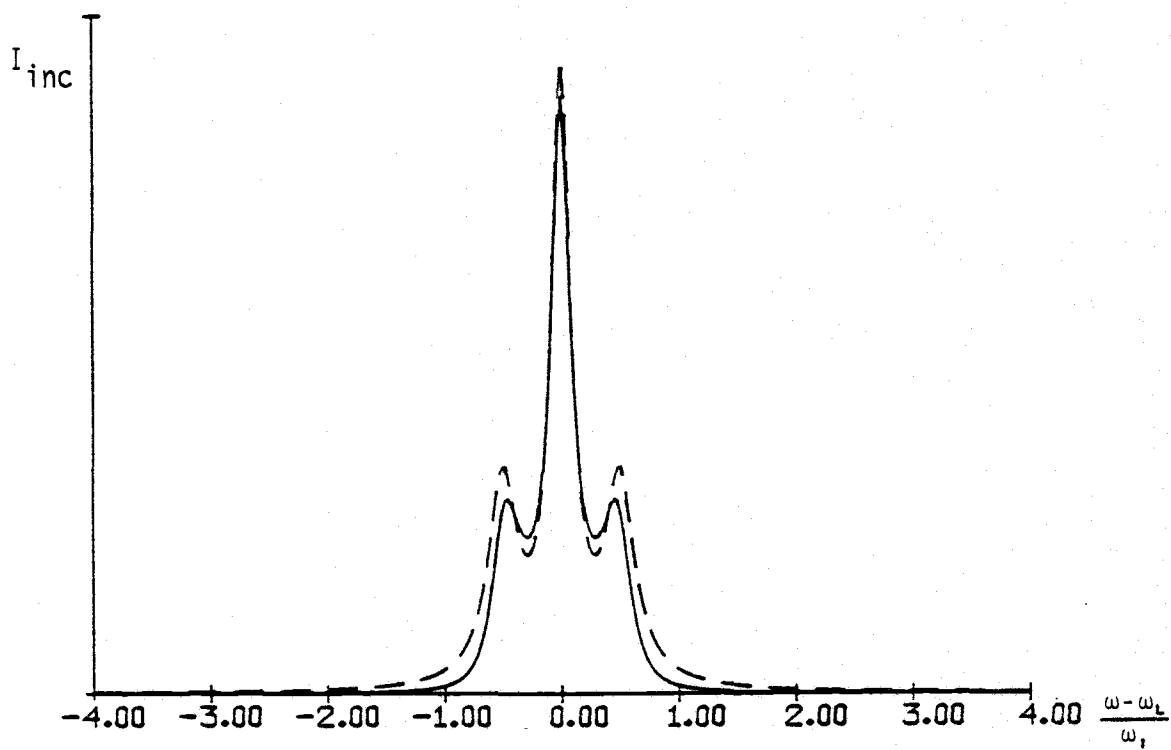


Figure (5-14)

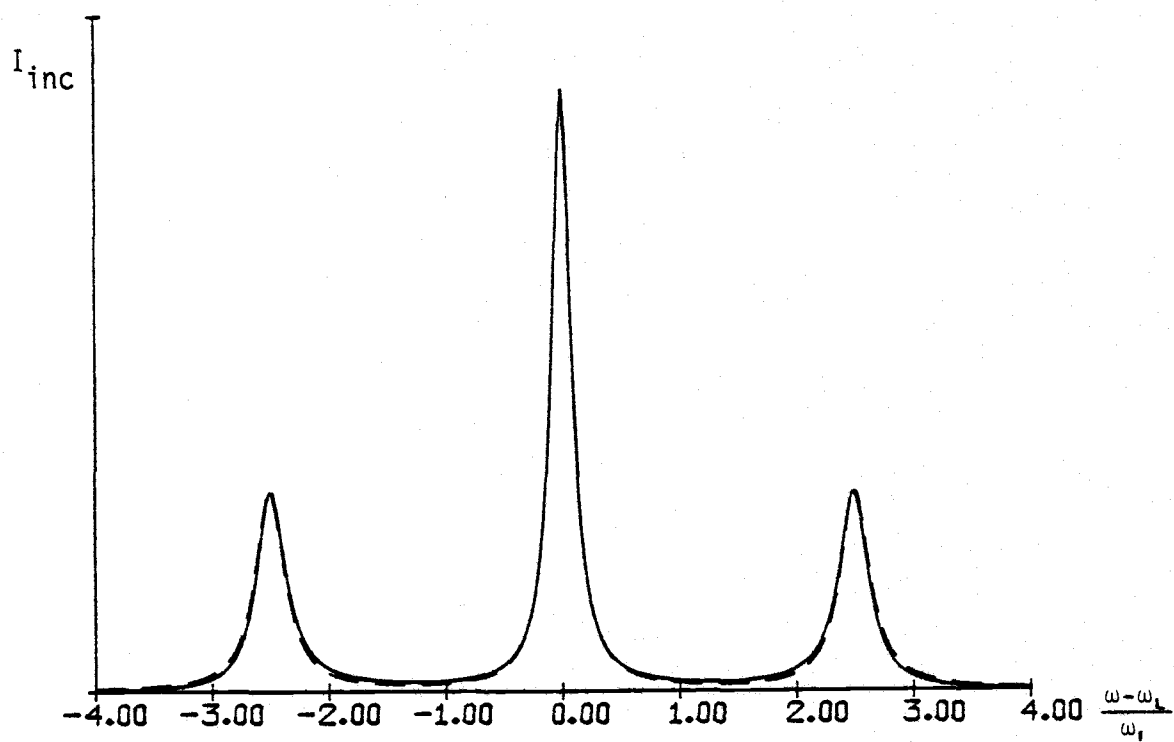


Figure (5-15)

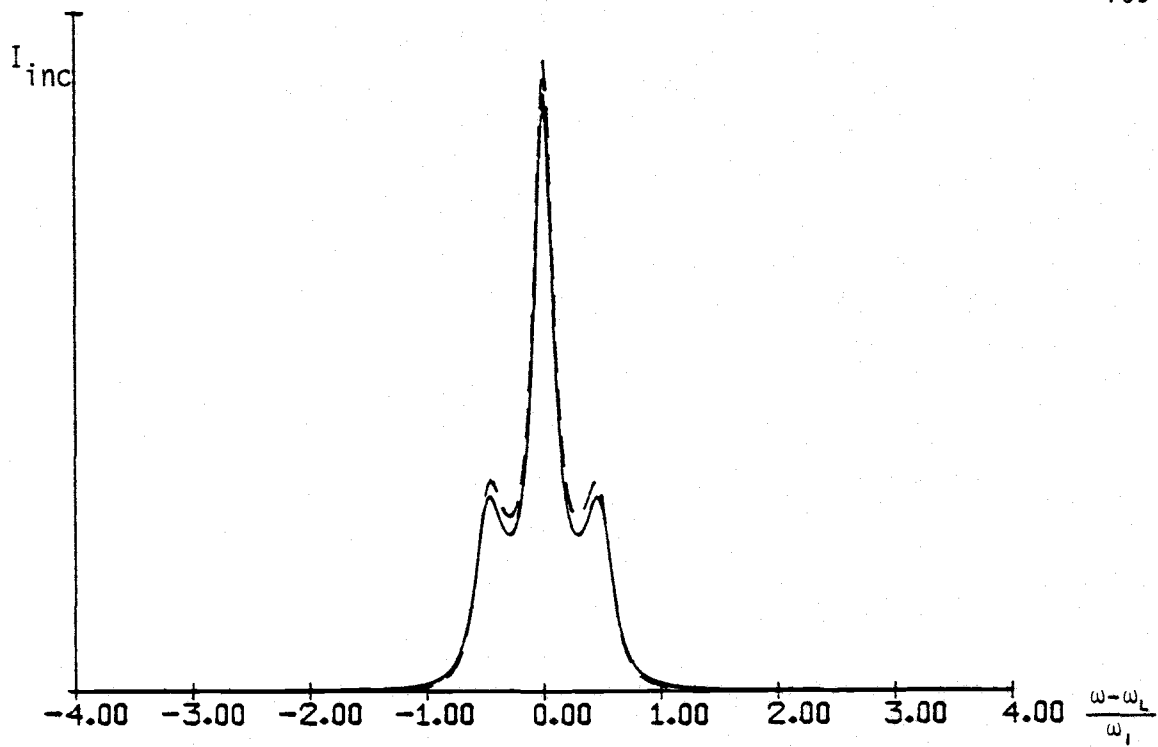


Figure (5-16)

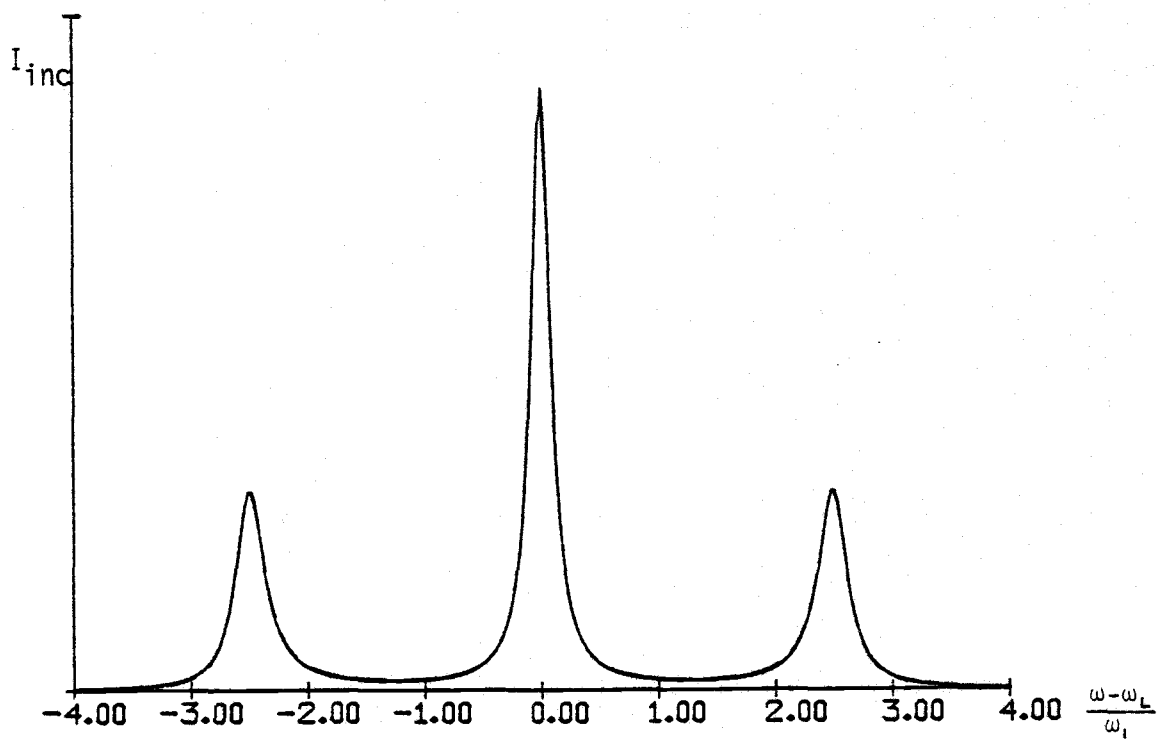


Figure (5-17)

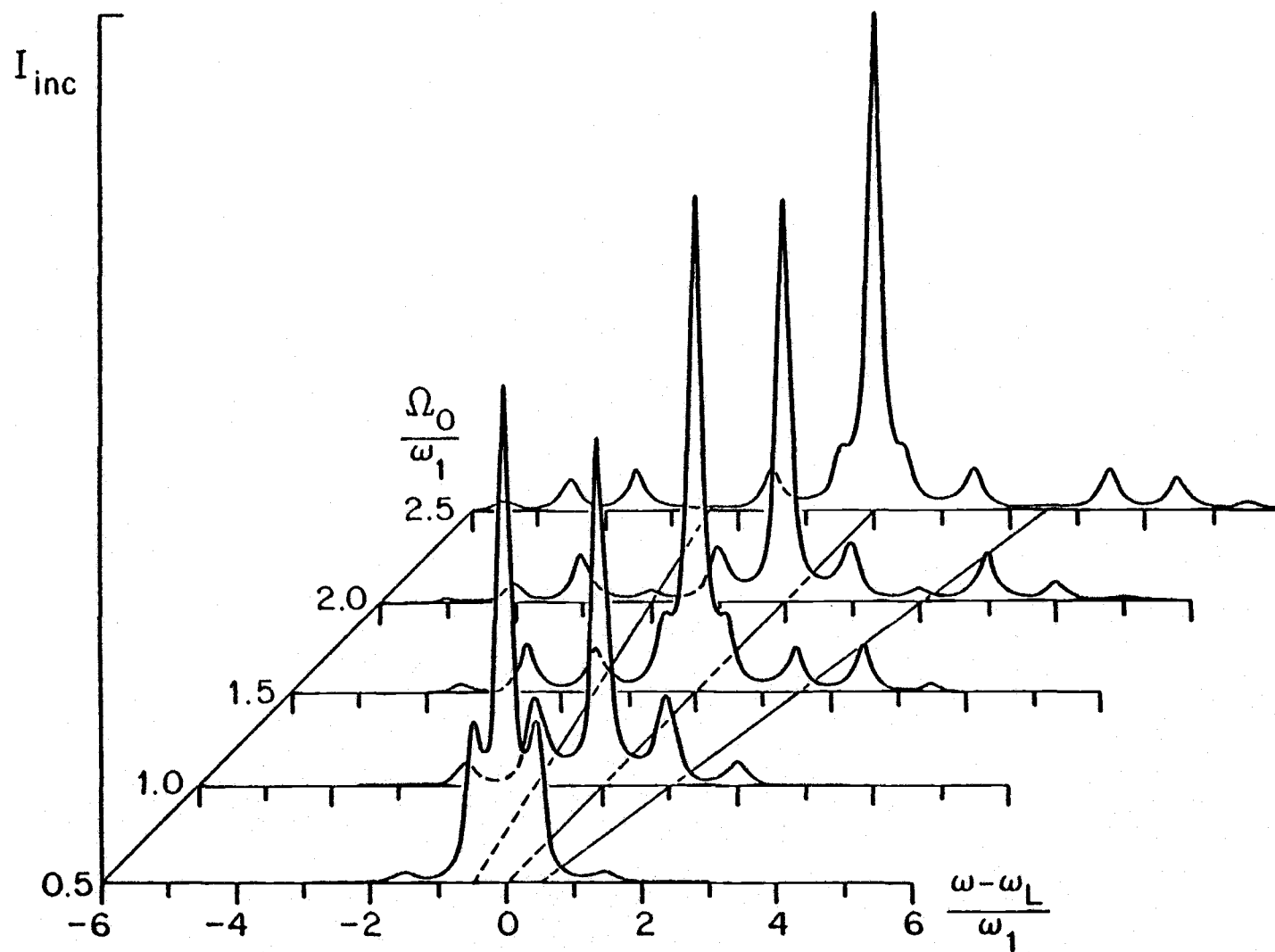


Figure (5-18)

5.6 An Estimate of the Order of the Effects Which are Neglected in the Rotating Wave Approximation

The optical Bloch equations of Chapter 3 describe a two-level atom of atomic frequency ω_0 interacting with a left circularly polarized wave of frequency ω_L . In the rotating frame there are, for the constant amplitude case and near resonance, no optical frequencies left in the problem.

A linear wave can be written as the sum of a left circularly polarized wave and a right circularly polarized wave. In the frame rotating with the left circularly polarized wave these are observed as a static field and a field which rotates clockwise with frequency $2\omega_L$. Neglecting the latter is called the rotating wave approximation. For optical frequencies ω_L this is a very good approximation and the neglected effects, notably the Bloch-Siegert Shift^{7,34}, are very small.

Our calculations enable us to make an estimate of the order of the effects which are neglected when we approximate a linear wave of constant amplitude and with frequency $\omega_L = \omega_0$ by a left circularly polarized wave. The linear field, which can be written as $\{E_0 \cos \omega_L t\} \hat{x}$ in the lab frame becomes $\left\{ \frac{E_0}{2} (1 + \cos 2\omega_L t) \right\} \hat{x}' - \left\{ \frac{E_0}{2} \sin 2\omega_L t \right\} \hat{y}'$ in the rotating frame. Instead of dropping the terms containing the optical frequencies altogether, as in the rotating wave approximation, we retain the component in the \hat{x}' direction and thus are left with a field of amplitude $E(t) = \frac{E_0}{2} (1 + \cos 2\omega_L t)$

This amplitude has the same time dependence as the one of Equation (3-46) with a modulation depth $Q = 1$ and a modulation frequency

$\omega_1 = 2\omega_L$. In our calculations we did not have to impose any re-

strictions on ω , even though we chose $\omega_1 = 5\tau$ for our graphs as a reasonable value for the modulation frequency. The results of our calculations are correct even for optical ω . The effect of the rapidly oscillating part of the \hat{x}' -component of the electric field is to introduce the terms containing $J_n(a \frac{\Omega_0}{\omega_1})$ and products thereof in the results. We have neglected the \hat{y}' -component of the electric field, which should have a similar effect. The right circularly polarized wave thus manifests itself in the spectrum in effects of the order of $J_n(\frac{\Omega_0}{2\omega_1})$ of the effects of the left circularly polarized wave. A typical value of the argument for a strong field ($\Omega_0 \approx 100$ MHz) is $\frac{\Omega_0}{2\omega_1} \approx 10^{-8}$. We use the relation $J_n(x) \approx \frac{(\frac{1}{2}x)^n}{n!}$ for $x \ll 1$ and conclude that the effects neglected in the rotating wave approximation are of the order of 0.5×10^{-8} and smaller of the main effects for optical frequencies.

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APPENDIX

The Spectral Distributions and Total Intensities in the Case of the Constant Amplitude Dynamic Stark Effect

The constant amplitude dynamic Stark effect has been treated by many authors^{10,11,12,13,14,15,16}. Their results can be obtained in many different ways, one of which is the one we use to solve the modulated amplitude dynamic Stark effect.

The optical Bloch equations

$$\dot{x} = -\frac{\sigma}{2} x \quad (A-1)$$

$$\dot{y} = -\frac{\sigma}{2} y - \Omega_0 z \quad (A-2)$$

$$\dot{z} = -\sigma(z+1) + \Omega_0 y \quad (A-3)$$

have the long term solution

$$x_\infty = 0 \quad (A-4)$$

$$y_\infty = \frac{\sigma \Omega_0}{\frac{1}{2} \sigma^2 + \Omega_0^2} \quad (A-5)$$

$$z_\infty = \frac{-\frac{1}{2} \sigma^2}{\frac{1}{2} \sigma^2 + \Omega_0^2} \quad (A-6)$$

and the transient solution (for $\Omega_0 \geq \frac{\sigma}{4}$)

$$x(t, t') = x(t') e^{-\frac{\sigma}{2}(t-t')} \quad (A-7)$$

$$y(t, t') = \frac{A_+ y(t') - z(t')}{A_+ - A_-} e^{\Gamma_+(t-t')} + \frac{A_- y(t') - z(t')}{A_- - A_+} e^{\Gamma_-(t-t')} \quad (A-8)$$

$$z(t, t') = \frac{y(t') - A_- z(t')}{A_+ - A_-} e^{\Gamma_+(t-t')} + \frac{y(t') - A_+ z(t')}{A_- - A_+} e^{\Gamma_-(t-t')} \quad (A-9)$$

where Γ_\pm and A_\pm are given by Equations (3-67) and (3-68) respectively.

The coherently and incoherently scattered total intensities can

be determined using Equations (4-90) and (4-91) respectively.

In the limit of large measuring times T one can replace $\langle S_z(t) \rangle$ by z_∞ and $\langle S_\pm(t) \rangle$ by $\pm i y_\infty$ and one finds

$$I_{coh} \sim \frac{\pi}{2} \frac{\Omega_o^2 \gamma^2}{(\frac{1}{2} \gamma^2 + \Omega_o^2)^2} \quad (A-10)$$

and

$$I_{inc} \sim \pi \frac{\Omega_o^4}{(\frac{1}{2} \gamma^2 + \Omega_o^2)^2} \quad (A-11)$$

The coherent and the incoherent parts of the spectrum can be determined using Equations (4-64) and (4-65) respectively. One can show that in the limit of large measuring times

$$\begin{aligned} \langle \delta S_+(t) \delta S_-(t') \rangle &= \frac{1}{4} (1 + z_\infty) e^{-\frac{\gamma}{2}(t-t')} \\ &+ \frac{1}{4} \frac{A_+(1 + z_\infty - y_\infty^2) + y_\infty(1 + z_\infty)}{A_+ - A_-} e^{r_+(t-t')} \\ &+ \frac{1}{4} \frac{A_-(1 + z_\infty - y_\infty^2) + y_\infty(1 + z_\infty)}{A_- - A_+} e^{r_-(t-t')} \end{aligned} \quad (A-12)$$

In the strong field limit ($\frac{\gamma}{4\Omega_o} \ll 1$)

$$\begin{aligned} \langle \delta S_+(t) \delta S_-(t') \rangle &\approx \frac{1}{4} e^{-\frac{\gamma}{2}(t-t')} \\ &+ \frac{1}{8} e^{(-\frac{3}{4}\gamma + i\Omega_o)(t-t')} + \frac{1}{8} e^{(-\frac{3}{4}\gamma - i\Omega_o)(t-t')} \end{aligned} \quad (A-13)$$

In the limit of large measuring times T one finds

$$I_{coh}(\omega) \sim 2\pi \left(\frac{y_\infty}{2}\right)^2 \delta(\omega - \omega_L) = \frac{\pi}{2} \frac{\gamma^2 \Omega_o^2}{(\frac{1}{2} \gamma^2 + \Omega_o^2)^2} \delta(\omega - \omega_L) \quad (A-14)$$

and in the limit of large measuring times T and strong fields one obtains

$$I_{inc}(\omega) \sim \frac{\tau}{16} \left\{ \frac{4}{(\frac{1}{2}\tau)^2 + (\omega - \omega_L)^2} + \frac{3}{(\frac{3}{4}\tau)^2 + (\omega - \omega_L - \Omega_0)^2} + \frac{3}{(\frac{3}{4}\tau)^2 + (\omega - \omega_L + \Omega_0)^2} \right\} \quad (A-15)$$

In the above references the incoherent frequency distribution is given in the strong field limit of Equation (A-15). We can calculate $I_{inc}(\omega)$ for arbitrary field strengths using Equations (4-65) and (A-12).

In the limit of large measuring times T we obtain the following result, which is valid for $\Omega_0 \geq \frac{\tau}{4}$:

$$I_{inc}(\omega) \sim \frac{\tau \Omega_0^2}{2(\tau^2 + 2\Omega_0^2)} \left\{ \frac{1}{(\frac{\tau}{2})^2 + (\omega - \omega_L)^2} + \frac{\frac{3}{4}(\Omega_0^2 - \frac{1}{2}\tau^2) - (\frac{5}{4}\Omega_0^2 - \frac{1}{8}\tau^2) \frac{\omega - \omega_L - \Omega_0 \sqrt{1 - (\frac{\tau}{4\Omega_0})^2}}{\Omega_0 \sqrt{1 - (\frac{\tau}{4\Omega_0})^2}}}{[\frac{1}{2}\tau^2 + \Omega_0^2][(\frac{3}{4}\tau)^2 + (\omega - \omega_L - \Omega_0 \sqrt{1 - (\frac{\tau}{4\Omega_0})^2})^2]} + \frac{\frac{3}{4}(\Omega_0^2 - \frac{1}{2}\tau^2) + (\frac{5}{4}\Omega_0^2 - \frac{1}{8}\tau^2) \frac{\omega - \omega_L + \Omega_0 \sqrt{1 - (\frac{\tau}{4\Omega_0})^2}}{\Omega_0 \sqrt{1 - (\frac{\tau}{4\Omega_0})^2}}}{[\frac{1}{2}\tau^2 + \Omega_0^2][(\frac{3}{4}\tau)^2 + (\omega - \omega_L + \Omega_0 \sqrt{1 - (\frac{\tau}{4\Omega_0})^2})^2]} \right\} \quad (A-16)$$