Chapter 6

Nonlinear Optics in the Two-Level Approximation

6.1 Introduction

Our treatment of nonlinear optics in the previous chapters has for the most part made use of power series expansions to relate the response of a material system to the strength of the applied optical field. In simple cases, this relation can be taken to be of the form

$$\tilde{P}(t) = \epsilon_0 \chi^{(1)} \tilde{E}(t) + \epsilon_0 \chi^{(2)} \tilde{E}(t)^2 + \epsilon_0 \chi^{(3)} \tilde{E}(t)^3 + \cdots. \tag{6.1.1}$$

However, there are circumstances under which such a power series expansion does not converge, and under such circumstances different methods must be employed to describe nonlinear optical effects. One example is that of a saturable absorber, where the absorption coefficient α is related to the intensity $I = 2n\epsilon_0 c|E|^2$ of the applied optical field by the relation

$$\alpha = \frac{\alpha_0}{1 + I/I_s}, \tag{6.1.2}$$

where α_0 is the weak-field absorption coefficient and I_s is an optical constant called the saturation intensity. We can expand this equation in a power series to obtain

$$\alpha = \alpha_0 \big[1 - (I/I_s) + (I/I_s)^2 - (I/I_s)^3 + \cdots\big]. \tag{6.1.3}$$

However, this series converges only for $I < I_s$, and thus only in this limit can saturable absorption be described by means of a power series of the sort given by Eq. (6.1.1).

It is primarily under conditions such that a transition of the material system is resonantly excited that perturbation techniques fail to provide an adequate description of the response of the system to an applied optical field. However, under such conditions it is usually adequate to deal only with the two atomic levels that are resonantly connected by the optical field. The increased complexity entailed in describing the atomic system in a nonperturbative manner

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is thus compensated in part by the ability to make the two-level approximation. When only two levels are included in the theoretical analysis, there is no need to perform the sums over *all* atomic states that appear in the general quantum-mechanical expressions for $\chi^{(3)}$ given in Chapter 3.

In the present chapter, we shall for the most part concentrate on the situation in which a monochromatic beam of frequency ω interacts with a collection of two-level atoms. The treatment is thus an extension of that of Chapter 4, which treated the interaction of a monochromatic beam with a nonlinear medium in terms of the third-order susceptibility $\chi^{(3)}(\omega = \omega + \omega - \omega)$. In addition, in the last two sections of this chapter we generalize the treatment by studying nondegenerate four-wave mixing involving a collection of two-level atoms.

Even though the two-level model ignores many of the features present in real atomic systems, there is still an enormous richness in the physical processes that are described within the two-level approximation. Some of the processes that can occur and that are described in the present chapter include saturation effects, power broadening, Rabi oscillations, and optical Stark shifts. Parallel treatments of optical nonlinearities in two-level atoms can be found in the books of Allen and Eberly (1975) and Cohen-Tannoudji et al. (1989) and in the reviews of Sargent (1978) and Boyd and Sargent (1988).

6.2 Density Matrix Equations of Motion for a Two-Level Atom

We first consider the density matrix equations of motion for a two-level system in the absence of damping effects. Since damping mechanisms can be very different under different physical conditions, there is no *unique* way to include damping in the model. The present treatment thus serves as a starting point for the inclusion of damping by any mechanism.

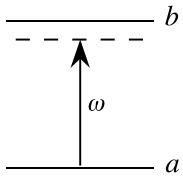

FIGURE 6.2.1: Near-resonant excitation of a two-level atom.

The interaction we are treating is illustrated in Fig. 6.2.1. The lower atomic level is denoted a and the upper level b. We represent the Hamiltonian for this system as

$$\hat{H} = \hat{H}_0 + \hat{V}(t), \tag{6.2.1}$$

where $\hat{H}_0$ denotes the atomic Hamiltonian and $\hat{V}(t)$ denotes the energy of interaction of the atom with the electromagnetic field. We denote the energies of the states a and b as

$$E_a = \hbar\omega_a \quad \text{and} \quad E_b = \hbar\omega_b. \tag{6.2.2}$$

The Hamiltonian $\hat{H}_0$ can thus be represented by the diagonal matrix whose elements are given by

$$H_{0,nm} = E_n \delta_{nm}. \tag{6.2.3}$$

We assume that the interaction energy can be adequately described in the electric dipole approximation, in which case the interaction Hamiltonian has the form

$$\hat{V}(t) = -\hat{\mu}\tilde{E}(t). \tag{6.2.4}$$

We also assume that the atomic wave functions corresponding to states a and b have definite parity so that the diagonal matrix elements of $\hat{\mu}$ vanish—that is, we assume that $\mu_{aa} = \mu_{bb} = 0$ and thus that

$$V_{aa} = V_{bb} = 0. \tag{6.2.5}$$

The only nonvanishing elements of $\tilde{V}$ are hence V_{ba} and V_{ab}, which are given explicitly by

$$V_{ba} = V_{ab}^* = -\mu_{ba}\tilde{E}(t). \tag{6.2.6}$$

We describe the state of this system by means of the density matrix, which is given explicitly by

$$\hat{\rho} = \begin{bmatrix} \rho_{aa} & \rho_{ab} \\ \rho_{ba} & \rho_{bb} \end{bmatrix}, \tag{6.2.7}$$

where $\rho_{ba} = \rho_{ab}^*$. The time evolution of the density matrix is given, still in the absence of damping effects, by Eq. (3.3.21) as

$$\begin{aligned} \dot{\rho}_{nm} &= \frac{-i}{\hbar}\left[\hat{H}, \hat{\rho}\right]_{nm} = \frac{-i}{\hbar}\left[\left(\hat{H}\hat{\rho}\right)_{nm} - \left(\hat{\rho}\hat{H}\right)_{nm}\right] \\ &= \frac{-i}{\hbar}\sum_{\nu}(H_{n\nu}\rho_{\nu m} - \rho_{n\nu}H_{\nu m}). \end{aligned} \tag{6.2.8}$$

We now introduce the decomposition of the Hamiltonian into atomic and interaction parts (Eq. (6.2.1)) into this expression to obtain

$$\dot{\rho}_{nm} = -i\omega_{nm}\rho_{nm} - \frac{i}{\hbar}\sum_{\nu}(V_{n\nu}\rho_{\nu m} - \rho_{n\nu}V_{\nu m}), \tag{6.2.9}$$

where we have introduced the transition frequency $\omega_{nm} = (E_n - E_m)/\hbar$. For the case of the two-level atom, the indices n, m, and ν can take on the values a or b only, and the equations of

motion for the density matrix elements are given explicitly as

$$\dot{\rho}_{ba} = -i\omega_{ba}\rho_{ba} + \frac{i}{\hbar}V_{ba}(\rho_{bb} - \rho_{aa}), \tag{6.2.10a}$$

$$\dot{\rho}_{bb} = \frac{-i}{\hbar}(V_{ba}\rho_{ab} - \rho_{ba}V_{ab}), \tag{6.2.10b}$$

$$\dot{\rho}_{aa} = \frac{-i}{\hbar}(V_{ab}\rho_{ba} - \rho_{ab}V_{ba}). \tag{6.2.10c}$$

It can be seen by inspection that

$$\dot{\rho}_{bb} + \dot{\rho}_{aa} = 0, \tag{6.2.11}$$

which shows that the total population $\rho_{bb} + \rho_{aa}$ is a conserved quantity. From the definition of the density matrix, we know that the diagonal elements of $\hat{\rho}$ represent probabilities of occupation, and hence that

$$\rho_{aa} + \rho_{bb} = 1. \tag{6.2.12}$$

No separate equation of motion is required for ρ_{ab}, because of the relation $\rho_{ab} = \rho_{ba}^*$.

Eqs. (6.2.10) constitute the density matrix equations of motion for a two-level atom in the absence of relaxation processes. These equations provide an adequate description of resonant nonlinear optical processes under conditions where relaxation processes can be neglected, such as excitation with short pulses whose duration is much less than the material relaxation times. We next see how these equations are modified in the presence of relaxation processes.

6.2.1 Closed Two-Level Atom

Let us first consider relaxation processes of the sort illustrated schematically in Fig. 6.2.2. We assume that the upper level b decays to the lower level a at a rate Γ_{ba} and therefore that the lifetime of the upper level is given by $T_1 = 1/\Gamma_{ba}$. Typically, the decay of the upper level would be due to spontaneous emission. This system is called closed, because any population that leaves the upper level enters the lower level. We also assume that the atomic dipole moment is dephased in the characteristic time T_2, leading to a transition linewidth (for weak applied fields) of characteristic width $\gamma_{ba} = 1/T_2$.*

* In fact, one can see from Eq. (6.3.25) that the full width at half maximum in angular frequency units of the absorption line in the limit of weak fields is equal to $2\gamma_{ba}$.

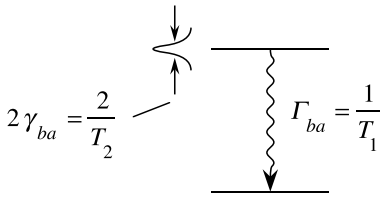

FIGURE 6.2.2: Relaxation processes of the closed two-level atom.

We can describe these relaxation processes mathematically by adding decay terms phenomenologically to Eqs. (6.2.10); the modified equations are given by

$$\dot{\rho}_{ba} = -\left(i\omega_{ba} + \frac{1}{T_2}\right)\rho_{ba} + \frac{i}{\hbar}V_{ba}(\rho_{bb} - \rho_{aa}), \tag{6.2.13a}$$

$$\dot{\rho}_{bb} = \frac{-\rho_{bb}}{T_1} - \frac{i}{\hbar}(V_{ba}\rho_{ab} - \rho_{ba}V_{ab}), \tag{6.2.13b}$$

$$\dot{\rho}_{aa} = \frac{\rho_{bb}}{T_1} + \frac{i}{\hbar}(V_{ba}\rho_{ab} - \rho_{ba}V_{ab}). \tag{6.2.13c}$$

The forms of the relaxation terms included in these equations will be justified in the discussion given below. One can see by inspection of Eqs. (6.2.13) that the condition

$$\dot{\rho}_{bb} + \dot{\rho}_{aa} = 0 \tag{6.2.14}$$

is still satisfied.

Since Eq. (6.2.13a) depends on the populations ρ_{bb} and ρ_{aa} only in terms of the population difference, $\rho_{bb} - \rho_{aa}$, it is useful to consider the equation of motion satisfied by this difference itself. We subtract Eq. (6.2.13c) from Eq. (6.2.13b) to find that

$$\frac{d}{dt}(\rho_{bb} - \rho_{aa}) = \frac{-2\rho_{bb}}{T_1} - \frac{2i}{\hbar}(V_{ba}\rho_{ab} - \rho_{ba}V_{ab}). \tag{6.2.15}$$

The first term on the right-hand side can be rewritten using the relation $2\rho_{bb} = (\rho_{bb} - \rho_{aa}) + (\rho_{bb} + \rho_{aa}) = (\rho_{bb} - \rho_{aa}) + 1$, where we have made use of Eq. (6.2.12), to obtain

$$\frac{d}{dt}(\rho_{bb} - \rho_{aa}) = -\frac{(\rho_{bb} - \rho_{aa}) + 1}{T_1} - \frac{2i}{\hbar}(V_{ba}\rho_{ab} - \rho_{ba}V_{ab}). \tag{6.2.16}$$

This relation is often generalized by allowing the possibility that the population difference $(\rho_{bb} - \rho_{aa})^{(\mathrm{eq})}$ in thermal equilibrium can have some value other than -1, the value taken above by assuming that all of the population resides in the ground state in thermal equilibrium. This generalized version of Eq. (6.2.16) is given by

$$\frac{d}{dt}(\rho_{bb} - \rho_{aa}) = -\frac{(\rho_{bb} - \rho_{aa}) - (\rho_{bb} - \rho_{aa})^{(\mathrm{eq})}}{T_1} - \frac{2i}{\hbar}(V_{ba}\rho_{ab} - \rho_{ba}V_{ab}). \tag{6.2.17}$$

We therefore see that for a closed two-level system the density matrix equations of motion reduce to just two coupled equations, Eqs. (6.2.13a) and (6.2.17).

In order to justify the choice of relaxation terms used in Eqs. (6.2.13a) and (6.2.17), let us examine the nature of the solutions to these equations in the absence of an applied field—that is, for $V_{ba} = 0$. The solution to Eq. (6.2.17) is

$$\begin{aligned}\left[\rho_{bb}(t) - \rho_{aa}(t)\right] &= (\rho_{bb} - \rho_{aa})^{(\mathrm{eq})} \\ &\quad + \left\{\left[\rho_{bb}(0) - \rho_{aa}(0)\right] - (\rho_{bb} - \rho_{aa})^{(\mathrm{eq})}\right\} e^{-t/T_1}. \end{aligned} \tag{6.2.18}$$

This equation shows that the population inversion $[\rho_{bb}(t) - \rho_{aa}(t)]$ relaxes from its initial value $\rho_{bb}(0) - \rho_{aa}(0)$ to its equilibrium value $(\rho_{bb} - \rho_{aa})^{(\mathrm{eq})}$ in a time of the order of T_1. For this reason, T_1 is called the population relaxation time.

Similarly, the solution to Eq. (6.2.13a) for the case $V_{ba} = 0$ is of the form

$$\rho_{ba}(t) = \rho_{ba}(0) e^{-(i\omega_{ba} + 1/T_2)t}. \tag{6.2.19}$$

We can interpret this result more directly by considering the expectation value of the induced dipole moment, which is given by (see also Eq. (3.3.36))

$$\begin{aligned}\left\langle \tilde{\mu}(t) \right\rangle &= \mathrm{Tr}(\hat{\rho}\hat{\mu}) = \mu_{ab}\rho_{ba}(t) + \mu_{ba}\rho_{ab}(t) = \mu_{ab}\rho_{ba}(0) e^{-(i\omega_{ba} + 1/T_2)t} + \text{c.c.} \\ &= \left[\mu_{ab}\rho_{ba}(0) e^{-i\omega_{ba}t} + \text{c.c.}\right] e^{-t/T_2}. \end{aligned} \tag{6.2.20}$$

This result shows that, for an undriven atom, the dipole moment oscillates at frequency ω_{ba} and decays to zero in the characteristic time T_2, which is hence known as the dipole dephasing time.

For reasons that were discussed in relation to Eq. (3.3.25), T_1 and T_2 are related to the collisional dephasing rate γ_c by

$$\frac{1}{T_2} = \frac{1}{2T_1} + \gamma_c. \tag{6.2.21a}$$

For an atomic vapor, γ_c is usually described accurately by the formula

$$\gamma_c = C_s N + C_f N_f, \tag{6.2.21b}$$

where N is the number density of atoms having resonance frequency ω_{ba}, and N_f is the number density of any "foreign" atoms of a different atomic species having a different resonance frequency. The parameters C_s and C_f are coefficients describing self-broadening and foreign-gas broadening, respectively. As an example, for the resonance line (i.e., the $3s \rightarrow 3p$ transition) of atomic sodium, T_1 is equal to 16 nsec, $C_s = 1.50 \times 10^{-7}$ cm^3/sec, and for the case of foreign-gas broadening by collisions with argon atoms, $C_f = 2.53 \times 10^{-9}$ cm^3/sec. The values of T_1, C_s, and C_f for other transitions are tabulated, for example, by Miles and Harris (1973).

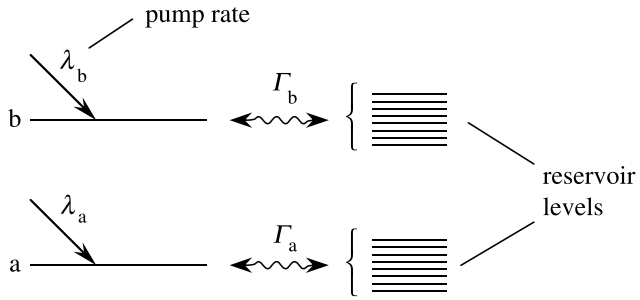

FIGURE 6.2.3: Relaxation processes for the open two-level atom.

6.2.2 Open Two-Level Atom

The open two-level atom is shown schematically in Fig. 6.2.3. Here the upper and lower levels are allowed to exchange population with associated reservoir levels. These levels might, for example, be magnetic sublevels or hyperfine levels associated with states a and b. The system is called open because the population that leaves the upper level does not necessarily enter the lower level. This model is often encountered in connection with laser theory, in which case the upper level or both levels are assumed to acquire population at some controllable pump rates, which we take to be λ_b and λ_a for levels b and a, respectively. As previously, we assume that the induced dipole moment relaxes in a characteristic time T_2. In order to account for relaxation and pumping processes of the sort just described, the density matrix equations (6.2.10) are modified to become

$$\dot{\rho}_{ba} = -\left(i\omega_{ba} + \frac{1}{T_2}\right)\rho_{ba} + \frac{i}{\hbar}V_{ba}(\rho_{bb} - \rho_{aa}), \tag{6.2.22a}$$

$$\dot{\rho}_{bb} = \lambda_b - \Gamma_b(\rho_{bb} - \rho_{bb}^{(\mathrm{eq})}) - \frac{i}{\hbar}(V_{ba}\rho_{ab} - \rho_{ba}V_{ab}), \tag{6.2.22b}$$

$$\dot{\rho}_{aa} = \lambda_a - \Gamma_a(\rho_{aa} - \rho_{aa}^{(\mathrm{eq})}) + \frac{i}{\hbar}(V_{ba}\rho_{ab} - \rho_{ba}V_{ab}). \tag{6.2.22c}$$

Note that in this case the total population contained in the two levels a and b is not conserved and that in general all three equations must be considered. The relaxation rates are related to the collisional dephasing rate γ_c and population rates Γ_b and Γ_a by

$$\frac{1}{T_2} = \tfrac{1}{2}(\Gamma_b + \Gamma_a) + \gamma_c. \tag{6.2.23}$$

6.2.3 Two-Level Atom with a Non-Radiatively Coupled Third Level

We next consider the energy level scheme shown in Fig. 6.2.4, which is often used to model a saturable absorber. Population spontaneously leaves the optically excited level b at a rate

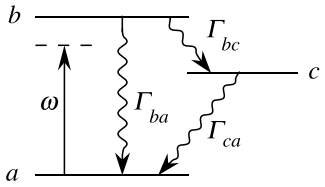

FIGURE 6.2.4: Relaxation processes for a two-level atom with a nonradiatively coupled third level.

$\Gamma_{ba} + \Gamma_{bc}$, where Γ_{ba} is the rate of decay to the ground state a, and Γ_{bc} is the rate of decay to level c. Level c acts as a trap level; population decays from level c back to the ground state at a rate Γ_{ca}. The lifetime T_c of population in level c is given by $T_c = 1/\Gamma_{ca}$ and is usually assumed to be the longest time scale in the problem. In addition, any dipole moment associated with the transition between levels a and b is damped at a rate γ_{ba}. These relaxation processes are modeled by modifying Eqs. (6.2.10) to take on the form

$$\dot{\rho}_{ba} = -(i\omega_{ba} + \gamma_{ba})\rho_{ba} + \frac{i}{\hbar}V_{ba}(\rho_{bb} - \rho_{aa}), \tag{6.2.24a}$$

$$\dot{\rho}_{bb} = -(\Gamma_{ba} + \Gamma_{bc})\rho_{bb} - \frac{i}{\hbar}(V_{ba}\rho_{ab} - \rho_{ba}V_{ab}), \tag{6.2.24b}$$

$$\dot{\rho}_{cc} = \Gamma_{bc}\rho_{bb} - \Gamma_{ca}\rho_{cc}, \tag{6.2.24c}$$

$$\dot{\rho}_{aa} = \Gamma_{ba}\rho_{bb} + \Gamma_{ca}\rho_{cc} + \frac{i}{\hbar}(V_{ba}\rho_{ab} - \rho_{ba}V_{ab}). \tag{6.2.24d}$$

It can be seen by inspection that the total population in the three levels is conserved, that is, that

$$\dot{\rho}_{aa} + \dot{\rho}_{bb} + \dot{\rho}_{cc} = 0.$$

6.3 Steady-State Response of a Two-Level Atom to a Monochromatic Field

We next examine the nature of the solution to the density matrix equations of motion for a two-level atom in the presence of a monochromatic, steady-state field. For definiteness, we treat the case of a closed two-level atom, although our results would be qualitatively similar for any of the models described above (see Problem 1 at the end of this chapter). For the closed two-level atomic system, the density matrix equations were shown above (Eqs. (6.2.13a) and (6.2.17)) to be of the form

$$\frac{d}{dt}\rho_{ba} = -\left(i\omega_{ba} + \frac{1}{T_2}\right)\rho_{ba} + \frac{i}{\hbar}V_{ba}(\rho_{bb} - \rho_{aa}), \tag{6.3.1}$$

$$\frac{d}{dt}(\rho_{bb} - \rho_{aa}) = -\frac{(\rho_{bb} - \rho_{aa}) - (\rho_{bb} - \rho_{aa})^{(\mathrm{eq})}}{T_1} - \frac{2i}{\hbar}(V_{ba}\rho_{ab} - \rho_{ba}V_{ab}). \tag{6.3.2}$$

In the electric dipole approximation, the interaction Hamiltonian for an applied field in the form of a monochromatic wave of frequency ω is given by

$$\hat{V} = -\hat{\mu}\tilde{E}(t) = -\hat{\mu}\left(Ee^{-i\omega t} + E^* e^{i\omega t}\right), \tag{6.3.3}$$

and the matrix elements of the interaction Hamiltonian are then given by

$$V_{ba} = -\mu_{ba}\left(Ee^{-i\omega t} + E^* e^{i\omega t}\right). \tag{6.3.4}$$

Eqs. (6.3.1) and (6.3.2) cannot be solved exactly for V_{ba} given by Eq. (6.3.4). However, they can be solved in an approximation known as the rotating-wave approximation. We recall from the discussion of Eq. (6.2.20) that, in the absence of a driving field, ρ_{ba} tends to evolve in time as $\exp(-i\omega_{ba}t)$. For this reason, when ω is approximately equal to ω_{ba}, the part of V_{ba} that oscillates as $e^{-i\omega t}$ acts as a far more effective driving term for ρ_{ba} than does the part that oscillates as $e^{i\omega t}$. It is thus a good approximation to take V_{ba} not as Eq. (6.3.4) but instead as

$$V_{ba} = -\mu_{ba}Ee^{-i\omega t}. \tag{6.3.5}$$

This approximation is called the rotating-wave approximation. Within this approximation, the density matrix equations of motion (6.3.1) and (6.3.2) become

$$\frac{d}{dt}\rho_{ba} = -\left(i\omega_{ba} + \frac{1}{T_2}\right)\rho_{ba} - \frac{i}{\hbar}\mu_{ba}Ee^{-i\omega t}(\rho_{bb} - \rho_{aa}), \tag{6.3.6}$$

$$\frac{d}{dt}(\rho_{bb} - \rho_{aa}) = -\frac{(\rho_{bb} - \rho_{aa}) - (\rho_{bb} - \rho_{aa})^{(\mathrm{eq})}}{T_1} + \frac{2i}{\hbar}\left(\mu_{ba}Ee^{-i\omega t}\rho_{ab} - \mu_{ab}E^* e^{i\omega t}\rho_{ba}\right). \tag{6.3.7}$$

Note that, in the rotating-wave approximation, ρ_{ba} is driven only at nearly its resonance frequency ω_{ba}, and $\rho_{bb} - \rho_{aa}$ is driven only at nearly zero frequency, which is its natural frequency.

We next find the steady-state solution to Eqs. (6.3.6) and (6.3.7), that is, the solution which is valid long after the transients associated with the turn-on of the driving field have died out. We do so by introducing the slowly varying quantity σ_{ba}, defined by

$$\rho_{ba}(t) = \sigma_{ba}(t)e^{-i\omega t}. \tag{6.3.8}$$

Eqs. (6.3.6) and (6.3.7) then become

$$\frac{d}{dt}\sigma_{ba} = \left[i(\omega - \omega_{ba}) - \frac{1}{T_2}\right]\sigma_{ba} - \frac{i}{\hbar}\mu_{ba}E(\rho_{bb} - \rho_{aa}), \tag{6.3.9}$$

$$\frac{d}{dt}(\rho_{bb} - \rho_{aa}) = -\frac{(\rho_{bb} - \rho_{aa}) - (\rho_{bb} - \rho_{aa})^{(\mathrm{eq})}}{T_1} + \frac{2i}{\hbar}\left(\mu_{ba}E\sigma_{ab} - \mu_{ab}E^*\sigma_{ba}\right). \tag{6.3.10}$$

The steady-state solution can now be obtained by setting the left-hand sides of Eqs. (6.3.9) and (6.3.10) equal to zero. We thereby obtain two coupled equations, which we solve algebraically to obtain

$$\rho_{bb} - \rho_{aa} = \frac{(\rho_{bb} - \rho_{aa})^{(\mathrm{eq})}[1 + (\omega - \omega_{ba})^2 T_2^2]}{1 + (\omega - \omega_{ba})^2 T_2^2 + (4/\hbar^2)|\mu_{ba}|^2 |E|^2 T_1 T_2}, \tag{6.3.11}$$

$$\rho_{ba} = \sigma_{ba} e^{-i\omega t} = \frac{\mu_{ba} E e^{-i\omega t}(\rho_{bb} - \rho_{aa})}{\hbar(\omega - \omega_{ba} + i/T_2)}. \tag{6.3.12}$$

We now use this result to calculate the polarization (i.e., the dipole moment per unit volume), which is given in terms of the off-diagonal elements of the density matrix by (see also Eq. (3.3.36))

$$\tilde{P}(t) = N\langle\tilde{\mu}\rangle = N\,\mathrm{Tr}\big(\hat{\tilde{\rho}}\hat{\tilde{\mu}}\big) = N(\mu_{ab}\rho_{ba} + \mu_{ba}\rho_{ab}), \tag{6.3.13}$$

where N is the number density of atoms. We introduce the complex amplitude P of the polarization through the relation

$$\tilde{P}(t) = P e^{-i\omega t} + \text{c.c.}, \tag{6.3.14}$$

and we define the susceptibility χ as the constant of proportionality relating P and E according to

$$P = \epsilon_0 \chi E. \tag{6.3.15}$$

We hence find from Eqs. (6.3.12) through (6.3.15) that the susceptibility is given by

$$\chi = \frac{N|\mu_{ba}|^2(\rho_{bb} - \rho_{aa})}{\epsilon_0 \hbar(\omega - \omega_{ba} + i/T_2)}, \tag{6.3.16}$$

where $\rho_{bb} - \rho_{aa}$ is given by Eq. (6.3.11). We introduce this expression for $[\rho_{bb} - \rho_{aa}]$ into Eq. (6.3.16) and rationalize the denominator to obtain the result

$$\chi = \frac{N(\rho_{bb} - \rho_{aa})^{(\mathrm{eq})}|\mu_{ba}|^2(\omega - \omega_{ba} - i/T_2)T_2^2/\epsilon_0\hbar}{1 + (\omega - \omega_{ba})^2 T_2^2 + (4/\hbar^2)|\mu_{ba}|^2|E|^2 T_1 T_2}. \tag{6.3.17}$$

Note that this expression gives the total susceptibility, including both its linear and nonlinear contributions.

We next introduce new notation to simplify this expression. We introduce the quantity

$$\Omega = 2|\mu_{ba}|\,|E|/\hbar, \tag{6.3.18}$$

which is known as the on-resonance Rabi frequency, and the quantity

$$\Delta = \omega - \omega_{ba}, \tag{6.3.19}$$

which is known as the detuning factor, so that the susceptibility can be expressed as

$$\chi = \left[N(\rho_{bb} - \rho_{aa})^{(\mathrm{eq})} |\mu_{ba}|^2 \frac{T_2}{\epsilon_0 \hbar} \right] \frac{\Delta T_2 - i}{1 + \Delta^2 T_2^2 + \Omega^2 T_1 T_2}. \tag{6.3.20}$$

Next, we express the combination of factors set off by square brackets in this expression in terms of the normal (i.e., linear) absorption coefficient of the material system, which is a directly measurable quantity. The absorption coefficient is given in general by*

$$\alpha = \frac{2\omega}{c} \operatorname{Im} n = \frac{2\omega}{c} \operatorname{Im}\left[(1 + \chi)^{1/2} \right], \tag{6.3.21a}$$

and, whenever the condition $|\chi| \ll 1$ is valid, the absorption coefficient can be expressed by

$$\alpha = \frac{\omega}{c} \operatorname{Im} \chi. \tag{6.3.21b}$$

If we let $\alpha_0(\Delta)$ denote the absorption coefficient experienced by a *weak* optical wave detuned from the atomic resonance by an amount Δ, we find by ignoring the contribution $\Omega^2 T_1 T_2$ to the denominator of Eq. (6.3.20) that $\alpha_0(\Delta)$ can be expressed as

$$\alpha_0(\Delta) = \frac{\alpha_0(0)}{1 + \Delta^2 T_2^2}, \tag{6.3.22a}$$

where the unsaturated, line-center absorption coefficient is given by

$$\alpha_0(0) = -\frac{\omega_{ba}}{c} \left[N(\rho_{bb} - \rho_{aa})^{(\mathrm{eq})} |\mu_{ba}|^2 \frac{T_2}{\epsilon_0 \hbar} \right]. \tag{6.3.22b}$$

By introducing this last expression into Eq. (6.3.20), we find that the susceptibility can be expressed as

$$\chi = -\frac{\alpha_0(0)}{\omega_{ba}/c} \frac{\Delta T_2 - i}{1 + \Delta^2 T_2^2 + \Omega^2 T_1 T_2}. \tag{6.3.23}$$

In order to interpret this result, it is useful to express the susceptibility as $\chi = \chi' + i\chi''$ with its real and imaginary parts given by

$$\chi' = -\frac{\alpha_0(0)}{\omega_{ba}/c} \frac{1}{\sqrt{1 + \Omega^2 T_1 T_2}} \frac{\Delta T_2 / \sqrt{1 + \Omega^2 T_1 T_2}}{1 + \Delta^2 T_2^2 / (1 + \Omega^2 T_1 T_2)}, \tag{6.3.24a}$$

* To justify the first equality of Eq. (6.3.21a), we recall that the absorption coefficient α is implicitly defined by the relation $I(z) = I(0) \exp(-\alpha z)$, where $I(z) = 2n\epsilon_0 c |E(z)|^2$ is the optical intensity. We also recall that the propagation of a plane-wave field in the z direction is described by $E(z) = E(0) \exp(ikz) = E(0) \exp(in\omega z/c)$. We thus find that $I(z) = 2n\epsilon_0 c E(z) E^*(z) = I(0) \exp(-2 \operatorname{Im} n\, \omega z/c)$. By comparison of the two expressions for $I(z)$, find that $\alpha = 2 \operatorname{Im} n\, \omega/c$.

$$\chi'' = \frac{\alpha_0(0)}{\omega_{ba}/c}\left(\frac{1}{1+\Omega^2 T_1 T_2}\right)\frac{1}{1+\Delta^2 T_2^2/(1+\Omega^2 T_1 T_2)}. \tag{6.3.24b}$$

We see from these expressions that, even in the presence of an intense laser field, χ' has a standard dispersive lineshape and χ'' has a Lorentzian lineshape. However, each of these lines has been broadened with respect to its weak-field width by the factor $(1+\Omega^2 T_1 T_2)^{1/2}$. In particular, the width of the absorption line (full width at half maximum) is given by

$$\Delta\omega_{\text{FWHM}} = \frac{2}{T_2}\left(1+\Omega^2 T_1 T_2\right)^{1/2}. \tag{6.3.25}$$

The tendency of spectral lines to become broadened when measured using intense optical fields is known as power broadening. We also see (e.g., from Eq. (6.3.24b)) that the line center value of χ'' (and consequently of the absorption coefficient α) is decreased with respect to its weak-field value by the factor $(1+\Omega^2 T_1 T_2)^{1/2}$. The tendency of the absorption to decrease when measured using intense optical fields is known as saturation. This behavior is illustrated in Fig. 6.3.1.

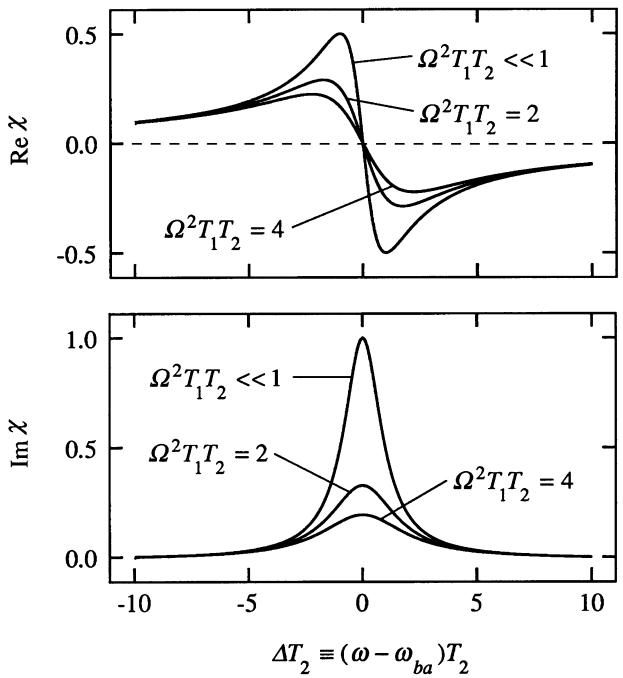

FIGURE 6.3.1: Real and imaginary parts of the susceptibility χ (in units of $\alpha_0 c/\omega_{ba}$) plotted as functions of the optical frequency ω for several values of the saturation parameter $\Omega^2 T_1 T_2$.

It is convenient to define, by means of the relation

$$\Omega^2 T_1 T_2 = \frac{|E|^2}{|E_s^0|^2}, \tag{6.3.26}$$

the quantity E_s^0, which is known as the line-center saturation field strength. Through the use of Eq. (6.3.18), we find that E_s^0 is given explicitly by

$$\left|E_s^0\right|^2 = \frac{\hbar^2}{4|\mu_{ba}|^2 T_1 T_2}. \tag{6.3.27}$$

The expression (6.3.23) for the susceptibility can be rewritten in terms of the saturation field strength as

$$\chi = \frac{-\alpha_0(0)}{\omega_{ba}/c} \frac{\Delta T_2 - i}{1 + \Delta^2 T_2^2 + |E|^2/|E_s^0|^2}. \tag{6.3.28}$$

We see from this expression that the significance of E_s^0 is that the absorption experienced by an optical wave tuned to line center (which is proportional to $\operatorname{Im}\chi$ evaluated at $\Delta = 0$) drops to one-half its weak-field value when the optical field has a strength of E_s^0. We can analogously define a saturation field strength for a wave of arbitrary detuning, which we denote E_s^Δ, by means of the relation

$$\left|E_s^\Delta\right|^2 = \left|E_s^0\right|^2 \left(1 + \Delta^2 T_2^2\right). \tag{6.3.29}$$

We then see from Eq. (6.3.28) that $\operatorname{Im}\chi$ drops to one-half its weak-field value when a field of detuning Δ has a field strength of E_s^Δ.

It is also useful to define the saturation *intensity* for a wave at line center (assuming that $|n-1| \ll 1$) as

$$I_s^0 = 2\epsilon_0 c \left|E_s^0\right|^2, \tag{6.3.30}$$

and the saturation intensity for a wave of arbitrary detuning as

$$I_s^\Delta = 2\epsilon_0 c \left|E_s^\Delta\right|^2 = I_s^0 \left(1 + \Delta^2 T_2^2\right). \tag{6.3.31}$$

In order to relate our present treatment of the nonlinear optical susceptibility to the perturbative treatment that we have used in the previous chapters, we next calculate the first- and third-order contributions to the susceptibility of a collection of two-level atoms. By performing

a power series expansion of Eq. (6.3.28) in the quantity $|E|^2/|E_s^0|^2$ and retaining only the first and second terms, we find that the susceptibility can be approximated as

$$\chi \simeq \frac{-\alpha_0(0)}{\omega_{ba}/c}\left(\frac{\Delta T_2 - i}{1+\Delta^2 T_2^2}\right)\left(1 - \frac{1}{1+\Delta^2 T_2^2}\frac{|E|^2}{|E_s^0|^2}\right). \tag{6.3.32}$$

We now equate this expression with the usual power series expansion $\chi = \chi^{(1)} + 3\chi^{(3)}|E^2|$ (where $\chi^{(3)} \equiv \chi^{(3)}(\omega = \omega + \omega - \omega)$) to find that the first- and third-order susceptibilities are given by

$$\chi^{(1)} = \frac{-\alpha_0(0)}{\omega_{ba}/c}\frac{\Delta T_2 - i}{1+\Delta^2 T_2^2}, \tag{6.3.33a}$$

$$\chi^{(3)} = \frac{\alpha_0(0)}{3\omega_{ba}/c}\left[\frac{\Delta T_2 - i}{(1+\Delta^2 T_2^2)^2}\right]\frac{1}{|E_s^0|^2}. \tag{6.3.33b}$$

The frequency dependence of $\chi^{(3)}$ as given by this expression is illustrated in Fig. 6.3.2. Note that the sign of $\chi^{(3)}$ is the opposite of that of $\chi^{(1)}$. One can understand this result by noting that $\chi^{(3)}$ represents a saturation of the optical response.

For some purposes, it is useful to express the nonlinear susceptibility in terms of the line-center saturation intensity as

$$\chi^{(3)} = \frac{\alpha_0(0)}{3\omega_{ba}/c}\left[\frac{\Delta T_2 - i}{(1+\Delta^2 T_2^2)^2}\right]\frac{2\epsilon_0 c}{I_s^0} \tag{6.3.34a}$$

or, through the use of Eqs. (6.3.22a) and (6.3.31), in terms of the saturation intensity and absorption coefficient at the laser frequency as

$$\chi^{(3)} = \frac{\alpha_0(\Delta)(\Delta T_2 - i)}{3\omega_{ba}/c}\frac{2\epsilon_0 c}{I_s^\Delta}. \tag{6.3.34b}$$

Note also that the third-order susceptibility can be related to the linear susceptibility by

$$\chi^{(3)} = \frac{-\chi^{(1)}}{3(1+\Delta^2 T_2^2)|E_s^0|^2} = \frac{-\chi^{(1)}}{3|E_s^\Delta|^2}. \tag{6.3.35}$$

Furthermore, through use of Eqs. (6.3.22b) and (6.3.27), the first- and third-order susceptibilities can be expressed in terms of microscopic quantities as

$$\chi^{(1)} = \left[N(\rho_{bb} - \rho_{aa})^{(\mathrm{eq})}|\mu_{ba}|^2\frac{T_2}{\epsilon_0\hbar}\right]\frac{\Delta T_2 - i}{1+\Delta^2 T_2^2}, \tag{6.3.36a}$$

$$\chi^{(3)} = -\tfrac{4}{3}N(\rho_{bb} - \rho_{aa})^{(\mathrm{eq})}|\mu_{ba}|^4\frac{T_1 T_2^2}{\epsilon_0\hbar^3}\frac{\Delta T_2 - i}{(1+\Delta^2 T_2^2)^2}. \tag{6.3.36b}$$

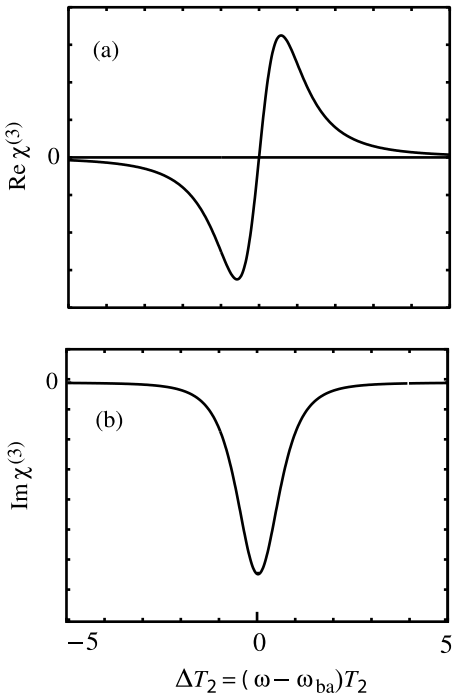

FIGURE 6.3.2: Real and imaginary parts of the third-order susceptibility $\chi^{(3)}$ plotted as functions of the optical frequency ω.

In the limit $\Delta T_2 \gg 1$, the expression for $\chi^{(3)}$ reduces to

$$\chi^{(3)} = -\tfrac{4}{3} N(\rho_{bb} - \rho_{aa})^{(\mathrm{eq})} |\mu_{ba}|^4 \frac{1}{\hbar^3 \Delta^3} \frac{T_1}{T_2}. \tag{6.3.37}$$

Let us consider the magnitudes of some of the physical quantities we have introduced in this section. Since (for $n = 1$) the intensity of an optical wave with field strength E is given by $I = 2\epsilon_0 c|E|^2$, the Rabi frequency of Eq. (6.3.18) can be expressed as

$$\Omega = \frac{2|\mu_{ba}|}{\hbar} \left(\frac{I}{2\epsilon_0 c} \right)^{1/2}. \tag{6.3.38}$$

Assuming that $|\mu_{ba}| = 2.5ea_0 = 2.0 \times 10^{-29}$ Cm (as it is for the $3s \rightarrow 3p$ transition of atomic sodium) and that I is measured in W/cm^2, this relationship gives the numerical result

$$\Omega[\mathrm{rad/sec}] = 2\pi \left(1 \times 10^9\right) \left(\frac{I[\mathrm{W/cm^2}]}{127} \right)^{1/2}. \tag{6.3.39}$$

Hence, whenever the intensity I exceeds 127 W/cm^2, $\Omega/2\pi$ becomes greater than 1 GHz, which is a typical value of the Doppler-broadened linewidth of an atomic transition. Intensities this large are available from the focused output of even low-power, cw lasers.

The saturation intensity of an atomic transition can be quite small. Again using $|\mu_{ba}| = 2.0 \times 10^{-29}$ Cm, and assuming that $T_1 = 16$ nsec (the value for the $3p \rightarrow 3s$ transition of atomic sodium) and that $T_2/T_1 = 2$ (the ratio for a radiatively broadened transition; see Eq. (6.2.21a)), we find from Eq. (6.3.30) that

$$I_s^0 = 5.27 \frac{\text{mW}}{\text{cm}^2} = 52.7 \frac{\text{W}}{\text{m}^2}. \tag{6.3.40}$$

Lastly, let us consider the magnitude of $\chi^{(3)}$ under conditions of the near-resonant excitation of an atomic transition. We take the typical values $N = 10^{14}$ cm^{-3}, $(\rho_{bb} - \rho_{aa})^{(\text{eq})} = -1$, $\mu_{ba} = 2.0 \times 10^{-29}$ Cm, $\Delta = \omega - \omega_{ba} = 2\pi c\ (1\ \text{cm}^{-1}) = 6\pi \times 10^{10}$ rad/sec, and $T_2/T_1 = 2$, in which case we find from Eq. (6.3.37) that $\chi^{(3)} = 2.1 \times 10^{-16}$ m^2/V^2. Note that this value is very much larger than the values of the nonresonant susceptibilities discussed in Chapter 4.

6.4 Optical Bloch Equations

In the previous two sections, we treated the response of a two-level atom to an applied optical field by working directly with the density matrix equations of motion. We chose to work with the density matrix equations in order to establish a connection with the calculation of the third-order susceptibility presented in Chapter 3. However, in theoretical quantum optics the response of a two-level atom is often treated through use of the optical Bloch equations or through related theoretical formalisms. Although these various formalisms are equivalent in their predictions, the equations of motion look different within different formalisms, and consequently different intuition regarding the nature of resonant optical nonlinearities is obtained. In this section, we review several of these formalisms.

We have seen (Eqs. (6.3.9) and (6.3.10)) that the density matrix equations describing the interaction of a closed two-level atomic system with the optical field

$$\tilde{E}(t) = E(t)e^{-i\omega t} + \text{c.c.}, \tag{6.4.1}$$

can be written in the rotating-wave approximation as

$$\frac{d}{dt}\sigma_{ba} = \left[i(\omega - \omega_{ba}) - \frac{1}{T_2}\right]\sigma_{ba} - \frac{i}{\hbar}\mu_{ba}E(\rho_{bb} - \rho_{aa}), \tag{6.4.2a}$$

$$\frac{d}{dt}(\rho_{bb} - \rho_{aa}) = -\frac{(\rho_{bb} - \rho_{aa}) - (\rho_{bb} - \rho_{aa})^{(\text{eq})}}{T_1} \tag{6.4.2b}$$

$$+ \frac{2i}{\hbar}(\mu_{ba}E\sigma_{ab} - \mu_{ab}E^*\sigma_{ba}), \tag{6.4.2c}$$

where the slowly varying, off-diagonal density matrix component $\sigma_{ba}(t)$ is defined by

$$\rho_{ba}(t) = \sigma_{ba}(t)e^{-i\omega t}. \tag{6.4.3}$$

The form of Eqs. (6.4.2) can be greatly simplified by introducing the following quantities:

1. The population inversions

$$w = \rho_{bb} - \rho_{aa} \quad \text{and} \quad w^{(\text{eq})} = (\rho_{bb} - \rho_{aa})^{(\text{eq})} \tag{6.4.4a}$$

2. The detuning of the optical field from resonance,*

$$\Delta = \omega - \omega_{ba} \tag{6.4.4b}$$

3. The atom–field coupling constant

$$\kappa = 2\mu_{ba}/\hbar. \tag{6.4.4c}$$

We also drop the subscripts on σ_{ba} for compactness. The density matrix equations of motion (6.4.2) then take the simpler form

$$\frac{d}{dt}\sigma = \left(i\Delta - \frac{1}{T_2}\right)\sigma - \tfrac{1}{2}i\kappa E w, \tag{6.4.5a}$$

$$\frac{d}{dt}w = -\frac{w - w^{(\text{eq})}}{T_1} + i(\kappa E\sigma^* - \kappa^* E^*\sigma). \tag{6.4.5b}$$

It is instructive to consider the equation of motion satisfied by the complex amplitude of the induced dipole moment. We first note that the expectation value of the induced dipole moment is given by

$$\langle\hat{\tilde{\mu}}\rangle = \text{Tr}(\hat{\rho}\hat{\mu}) = \rho_{ba}\mu_{ab} + \rho_{ab}\mu_{ba} = \sigma_{ba}\mu_{ab}e^{-i\omega t} + \sigma_{ab}\mu_{ba}e^{i\omega t}. \tag{6.4.6}$$

If we define the complex amplitude p of the dipole moment $\langle\hat{\tilde{\mu}}\rangle$ through the relation

$$\langle\hat{\tilde{\mu}}\rangle = pe^{-i\omega t} + \text{c.c.}, \tag{6.4.7}$$

we find by comparison with Eq. (6.4.6) that

$$p = \sigma_{ba}\mu_{ab}. \tag{6.4.8}$$

Eqs. (6.4.5) can hence be rewritten in terms of the dipole amplitude p as

$$\frac{dp}{dt} = \left(i\Delta - \frac{1}{T_2}\right)p - \frac{\hbar}{4}i|\kappa|^2 E w, \tag{6.4.9a}$$

$$\frac{dw}{dt} = -\frac{w - w^{(\text{eq})}}{T_1} - \frac{4}{\hbar}\text{Im}(Ep^*). \tag{6.4.9b}$$

* Note that some authors use the opposite sign convention for Δ.

These equations illustrate the nature of the coupling between the atom and the optical field. Note that they are linear in the atomic variables p and w and in the applied field amplitude E. However, the coupling is parametric: the dipole moment p is driven by a term that depends on the product of E with the inversion w, and likewise the inversion is driven by a term that depends on the product of E with p.

For those cases in which the field amplitude E can be taken to be a real quantity, the density matrix equations (6.4.5) can be simplified in a different way. We assume that the phase convention for describing the atomic energy eigenstates has been chosen such that μ_{ba} and hence κ are real quantities. It is then useful to express the density matrix element σ in terms of two real quantities u and v as

$$\sigma = \tfrac{1}{2}(u - iv). \tag{6.4.10}$$

The factor of one-half and the minus sign are used here to conform with convention (Allen and Eberly, 1975). This definition is introduced into Eq. (6.4.5b), which becomes

$$\frac{d}{dt}(u - iv) = \left(i\Delta - \frac{1}{T_2}\right)(u - iv) - i\kappa E w.$$

This equation can be separated into its real and imaginary parts as

$$\frac{d}{dt}u = \Delta v - \frac{u}{T_2}, \tag{6.4.11a}$$

$$\frac{d}{dt}v = -\Delta u - \frac{v}{T_2} + \kappa E w. \tag{6.4.11b}$$

Similarly, Eq. (6.4.5b) becomes

$$\frac{d}{dt}w = -\frac{w - w^{(\mathrm{eq})}}{T_1} - \kappa E v. \tag{6.4.11c}$$

The set (6.4.11) is known as the optical Bloch equations.

We next show that in the absence of relaxation processes (i.e., in the limit $T_1, T_2 \to \infty$) the variables u, v, and w obey the conservation law

$$u^2 + v^2 + w^2 = 1. \tag{6.4.12}$$

First, we note that the time derivative of $u^2 + v^2 + w^2$ vanishes:

$$\begin{aligned}\frac{d}{dt}\left(u^2 + v^2 + w^2\right) &= 2u\frac{du}{dt} + 2v\frac{dv}{dt} + 2w\frac{dw}{dt} \\ &= 2u\Delta v - 2v\Delta u + 2v\kappa E w - 2w\kappa E v \\ &= 0,\end{aligned} \tag{6.4.13}$$

where we have used Eqs. (6.4.11) in obtaining expressions for the time derivatives. We hence see that $u^2 + v^2 + w^2$ is a constant. Next, we note that before the optical field is applied the atom must be in its ground state and hence that $w = -1$ and $u = v = 0$ (as there can be no probability amplitude to be in the upper level). In this case we see that $u^2 + v^2 + w^2$ is equal to 1, but since the quantity $u^2 + v^2 + w^2$ is conserved, it must have this value at all times. We also note that since all of the damping terms in Eqs. (6.4.11) have negative signs associated with them, it must generally be true that

$$u^2 + v^2 + w^2 \leq 1. \tag{6.4.14}$$

6.4.1 Harmonic Oscillator Form of the Density Matrix Equations

Still different intuition regarding the nature of resonant optical nonlinearities can be obtained by considering the equation of motion satisfied by the expectation value of the dipole moment induced by the applied field (rather than considering the equation satisfied by its complex amplitude). This quantity is given by

$$\tilde{M} \equiv \langle \hat{\tilde{\mu}} \rangle = \rho_{ba}\mu_{ab} + \text{c.c.} \tag{6.4.15}$$

For simplicity of notation, we have introduced the new symbol $\tilde{M}$ rather than continuing to use $\langle \hat{\tilde{\mu}} \rangle$. Note that $\tilde{M}$ is a real quantity that oscillates at an optical frequency.

We take the density matrix equations of motion in the form

$$\dot{\rho}_{ba} = -\left(i\omega_{ba} + \frac{1}{T_2}\right)\rho_{ba} - \frac{i}{\hbar}\mu_{ba}\tilde{E}w, \tag{6.4.16a}$$

$$\dot{w} = -\frac{w - w^{(\text{eq})}}{T_1} + \frac{4\tilde{E}}{\hbar}\,\text{Im}(\mu_{ab}\rho_{ba}), \tag{6.4.16b}$$

where the dot denotes a time derivative. These equations follow from Eqs. (6.2.6), (6.2.13a), and (6.2.17) and the definition $w = \rho_{bb} - \rho_{aa}$. Here $\tilde{E}$ is the real, time-varying optical field; note that we have not made the rotating-wave approximation. We find by direct time differentiation of Eq. (6.4.15) and subsequent use of Eq. (6.4.16a) that the time derivative of $\tilde{M}$ is given by

$$\begin{aligned}\dot{\tilde{M}} &= \dot{\rho}_{ba}\mu_{ab} + \text{c.c.} \\ &= -\left(i\omega_{ba} + \frac{1}{T_2}\right)\rho_{ba}\mu_{ab} - \frac{i}{\hbar}|\mu_{ba}|^2\tilde{E}w + \text{c.c.} \\ &= -\left(i\omega_{ba} + \frac{1}{T_2}\right)\rho_{ba}\mu_{ab} + \text{c.c.}\end{aligned} \tag{6.4.17}$$

We have dropped the second term in the second-to-last form because it is imaginary and disappears when added to its complex conjugate. Next, we calculate the second time derivative of $\tilde{M}$ by taking the time derivative of Eq. (6.4.17) and introducing expression (6.4.16a) for $\dot{\rho}_{ba}$:

$$\begin{aligned}\ddot{\tilde{M}} &= -\left(i\omega_{ba} + \frac{1}{T_2}\right)\dot{\rho}_{ba}\mu_{ab} + \text{c.c.} \\ &= \left(i\omega_{ba} + \frac{1}{T_2}\right)^2 \rho_{ba}\mu_{ab} + \frac{i}{\hbar}\left(i\omega_{ba} + \frac{1}{T_2}\right)|\mu_{ba}|^2\tilde{E}w + \text{c.c.}\end{aligned}$$

or

$$\ddot{\tilde{M}} = \left(-\omega_{ba}^2 + \frac{2i\omega_{ba}}{T_2} + \frac{1}{T_2^2}\right)\rho_{ba}\mu_{ab} - \frac{\omega_{ba}}{\hbar}|\mu_{ba}|^2\tilde{E}w + \text{c.c.} \tag{6.4.18}$$

If we now introduce Eqs. (6.4.15) and (6.4.17) into this expression, we find that $\tilde{M}$ obeys the equation

$$\ddot{\tilde{M}} + \frac{2}{T_2}\dot{\tilde{M}} + \omega_{ba}^2\tilde{M} = \frac{-\tilde{M}}{T_2^2} - \frac{2\omega_{ba}}{\hbar}|\mu_{ba}|^2\tilde{E}w. \tag{6.4.19}$$

Since ω_{ba}^2 is much larger than $1/T_2^2$ in all physically realistic circumstances, we can drop the first term on the right-hand side of this expression to obtain the result

$$\ddot{\tilde{M}} + \frac{2}{T_2}\dot{\tilde{M}} + \omega_{ba}^2\tilde{M} = -\frac{2\omega_{ba}}{\hbar}|\mu_{ba}|^2\tilde{E}w. \tag{6.4.20}$$

This is the equation of a damped, driven harmonic oscillator. Note that the driving term is proportional to the product of the applied field strength $\tilde{E}(t)$ with the inversion w.

We next consider the equation of motion satisfied by the inversion w. In order to simplify Eq. (6.4.16b), we need an explicit expression for $\text{Im}(\rho_{ba}\mu_{ab})$. To find such an expression, we rewrite Eq. (6.4.17) as

$$\begin{aligned}\dot{\tilde{M}} &= -\left(i\omega_{ba} + \frac{1}{T_2}\right)\rho_{ba}\mu_{ab} + \text{c.c.} \\ &= -i\omega_{ba}(\rho_{ba}\mu_{ab} - \text{c.c.}) - \frac{1}{T_2}(\rho_{ba}\mu_{ab} + \text{c.c.}) \\ &= 2\omega_{ba}\,\text{Im}(\rho_{ba}\mu_{ab}) - \frac{\tilde{M}}{T_2},\end{aligned} \tag{6.4.21}$$

which shows that

$$\text{Im}(\rho_{ba}\mu_{ab}) = \frac{1}{2\omega_{ba}}\left(\dot{\tilde{M}} + \frac{\tilde{M}}{T_2}\right). \tag{6.4.22}$$

This result is now introduced into Eq. (6.4.16b), which becomes

$$\dot{w} = -\frac{w - w^{(\mathrm{eq})}}{T_1} + \frac{2\tilde{E}}{\hbar\omega_{ba}}\left(\dot{\tilde{M}} + \frac{\tilde{M}}{T_2}\right). \tag{6.4.23}$$

Since $\dot{\tilde{M}}$ oscillates at an optical frequency (which is much larger than $1/T_2$), the term $\tilde{M}/T_2$ can be omitted, yielding the result

$$\dot{w} = -\frac{w - w^{(\mathrm{eq})}}{T_1} + \frac{2}{\hbar\omega_{ba}}\tilde{E}\dot{\tilde{M}}. \tag{6.4.24}$$

We see that the inversion w is driven by the product of $\tilde{E}$ with $\dot{\tilde{M}}$, which is proportional to the part of $\tilde{M}$ that is 90 degrees out of phase with $\tilde{E}$. We also see that w relaxes to its equilibrium value $w^{(\mathrm{eq})}$ (which is typically equal to -1) in a time of the order of T_1.

Eqs. (6.4.20) and (6.4.24) provide a description of the two-level atomic system. Note that each equation is linear in the atomic variables $\tilde{M}$ and w. The origin of the nonlinear response of atomic systems lies in the fact that the coupling to the optical field depends parametrically on the atomic variables. A linear harmonic oscillator, for example, would be described by Eq. (6.4.20) with the inversion w held fixed at the value -1. The fact that the coupling depends on the inversion w, whose value depends on the applied field strength as described by Eq. (6.4.24), leads to nonlinearities.

6.4.2 Adiabatic-Following Limit

The treatment of Section 6.3 considered the steady-state response of a two-level atom to a cw laser field. The adiabatic-following limit (Grischkowsky, 1970) is another limit in which it is relatively easy to obtain solutions to the density matrix equations of motion. The nature of the adiabatic-following approximation is as follows: We assume that the optical field is in the form of a pulse whose length τ_p obeys the condition

$$\tau_p \ll T_1, T_2; \tag{6.4.25}$$

we thus assume that essentially no relaxation occurs during the optical pulse. In addition, we assume that the laser is detuned sufficiently far from resonance that

$$|\omega - \omega_{ba}| \gg T_2^{-1}, \tau_p^{-1}, \mu_{ba}E/\hbar; \tag{6.4.26}$$

that is, we assume that the detuning is greater than the transition linewidth, that no Fourier component of the pulse extends to the transition frequency, and that the transition is not power-broadened into resonance with the pulse. These conditions ensure that no appreciable population is excited to the upper level by the laser pulse.

To simplify the following analysis, we introduce the (complex) Rabi frequency

$$\Omega(t) = 2\mu_{ba} E(t)/\hbar, \tag{6.4.27}$$

where $E(t)$ gives the time evolution of the pulse envelope. The density matrix equations of motion (6.4.5) then become, in the limit $T_1 \to \infty$, $T_2 \to \infty$,

$$\frac{d\sigma}{dt} = i\Delta\sigma - \tfrac{1}{2} i\Omega w, \tag{6.4.28a}$$

$$\frac{dw}{dt} = -i(\Omega^*\sigma - \Omega\sigma^*). \tag{6.4.28b}$$

We note that the quantity $w^2 + 4\sigma\sigma^*$ is a constant of the motion whose value is given by

$$w^2(t) + 4\left|\sigma(t)\right|^2 = 1. \tag{6.4.29}$$

This conclusion is verified by means of a derivation analogous to that leading to Eq. (6.4.12).

We now make the adiabatic-following approximation, that is, we assume that for all times the atomic response is nearly in steady state with the applied field. We thus set $d\sigma/dt$ and dw/dt equal to zero in Eqs. (6.4.28). The simultaneous solution of these equations (which in fact is just the solution to (6.4.28a)) is given by

$$\sigma(t) = \frac{w(t)\Omega(t)}{2\Delta}. \tag{6.4.30}$$

Since $w(t)$ is a real quantity, this result shows that $\sigma(t)$ is always in phase with the driving field $\Omega(t)$. We now combine Eqs. (6.4.29) and (6.4.30) to obtain the equation

$$w(t)^2 + \frac{w(t)^2|\Omega|^2}{\Delta^2} = 1, \tag{6.4.31}$$

which can be solved for $w(t)$ to obtain

$$w(t) = \frac{-|\Delta|}{\sqrt{\Delta^2 + |\Omega(t)|^2}}. \tag{6.4.32}$$

This expression can now be substituted back into Eq. (6.4.30) to obtain the result

$$\sigma(t) = -\frac{\Delta}{|\Delta|} \frac{\frac{1}{2}\Omega(t)}{\sqrt{\Delta^2 + |\Omega(t)|^2}}. \tag{6.4.33}$$

We now use these results to deduce the value of the nonlinear susceptibility. As in Eqs. (6.3.11) through (6.3.17), the polarization P is related to $\sigma(t)$ (recall that $\sigma = \sigma_{ba}$) through

$$P = N\mu_{ab}\sigma, \tag{6.4.34}$$

which through use of Eq. (6.4.33) becomes

$$P = -\frac{\Delta}{|\Delta|}\frac{\frac{1}{2}N\mu_{ab}\Omega(t)}{\sqrt{\Delta^2 + |\Omega(t)|^2}}. \tag{6.4.35}$$

Our derivation has assumed that the condition $|\Delta| \gg |\Omega|$ is valid. We can thus expand Eq. (6.4.35) in a power series in the small quantity $|\Omega|/\Delta$ to obtain

$$P = \frac{\Delta\Omega}{\Delta^2}\frac{-\frac{1}{2}N\mu_{ab}}{\left(1 + |\Omega|^2/\Delta^2\right)^{1/2}} = -\frac{\Delta\Omega}{\Delta^2}\frac{1}{2}N\mu_{ab}\left(1 - \frac{1}{2}\frac{|\Omega|^2}{\Delta^2} + \cdots\right). \tag{6.4.36}$$

The contribution to P that is third-order in the applied field is thus given by

$$P^{(3)} = \frac{|\Omega|^2\Omega\Delta n\mu_{ab}}{4\Delta^4} = \frac{2N|\mu_{ab}|^4}{\hbar^3\Delta^3}|E|^2E, \tag{6.4.37}$$

where, in obtaining the second form, we have used the fact that $\Omega = 2\mu_{ba}E/\hbar$. By convention, the coefficient of $|E|^2E$ is $3\epsilon_0\chi^{(3)}$, and hence we find that

$$\chi^{(3)} = \frac{2N|\mu_{ba}|^4}{3\epsilon_0\hbar^3\Delta^3}. \tag{6.4.38}$$

Note that this prediction is identical to that of the steady-state theory (Eq. (6.3.37)) in the limit $\Delta T_2 \gg 1$ for the case of a radiatively broadened transition (i.e., $T_2/T_1 = 2$) and for which $(\rho_{bb} - \rho_{aa})^{(\mathrm{eq})} = -1$.

6.5 Rabi Oscillations and Dressed Atomic States

In this section we consider the response of a two-level atom to an optical field sufficiently intense to remove a significant fraction of the population from the atomic ground state. One might think that the only consequence of a field this intense would be to lower the overall response of the atom. Such is not the case, however. Stark shifts induced by the laser field profoundly modify the energy-level structure of the atom, leading to new resonances in the optical susceptibility. In the present section, we explore some of the processes that occur in the presence of a strong driving field.

The development will proceed primarily at the level of the atomic wavefunction, rather than at the level of the density matrix, as our interest is primarily in determining how the atomic energy level structure is modified by an intense driving field. Some brief comments regarding damping effects are included at the end of this section.

6.5.1 Rabi Solution of the Schrödinger Equation

Let us consider the solution to the Schrödinger equation for a two-level atom in the presence of an intense optical field.* We describe the state of the system in terms of the atomic wave function $\psi(\mathbf{r}, t)$, which obeys the Schrödinger equation

$$i\hbar\frac{\partial\psi}{\partial t} = \hat{H}\psi \tag{6.5.1}$$

with the Hamiltonian operator $\hat{H}$ given by

$$\hat{H} = \hat{H}_0 + \hat{V}(t). \tag{6.5.2}$$

Here $\hat{H}_0$ represents the Hamiltonian of a free atom, and $\hat{V}(t)$ represents the energy of interaction with the applied field. In the electric dipole approximation, $\hat{V}(t)$ is given by

$$\hat{V}(t) = \hat{\mu}\tilde{E}(t), \tag{6.5.3}$$

where the dipole moment operator is given by $\hat{\mu} = -e\hat{r}$.

We assume that the applied field is given by $\tilde{E}(t) = Ee^{-i\omega t} + \text{c.c.}$ with E constant, and that the field is nearly resonant with an allowed transition between the atomic ground state a and some other level b, as shown in Fig. 6.2.1. Since the effect of the interaction is to mix states a and b, the atomic wavefunction in the presence of the applied field can be represented as

$$\psi(\mathbf{r}, t) = C_a(t)u_a(\mathbf{r})e^{-i\omega_a t} + C_b(t)u_b(\mathbf{r})e^{-i\omega_b t}. \tag{6.5.4}$$

Here $u_a(\mathbf{r})e^{-i\omega_a t}$ represents the wavefunction of the atomic ground state a, and $u_b(\mathbf{r})e^{-i\omega_b t}$ represents the wavefunction of the excited state b. We assume that these wavefunctions are orthonormal in the sense that

$$\int d^3r\, u_i^*(\mathbf{r})u_j(\mathbf{r}) = \delta_{ij}. \tag{6.5.5}$$

The quantities $C_a(t)$ and $C_b(t)$ that appear in Eq. (6.5.4) can be interpreted as the probability amplitudes that at time t the atom is in state a or state b, respectively.

We next derive the equations of motion for $C_a(t)$ and $C_b(t)$, using methods analogous to those used in Section 3.2. By introducing Eq. (6.5.4) into the Schrödinger equation (6.5.1), multiplying the resulting equation by u_a^*, and integrating this equation over all space, we find that

$$\dot{C}_a = \frac{1}{i\hbar}C_b V_{ab}e^{-i\omega_{ba}t}, \tag{6.5.6}$$

* See also Sargent et al. (1974, p. 26), or Dicke and Wittke (1960, p. 203).

where we have introduced the resonance frequency $\omega_{ba} = \omega_b - \omega_a$ and the interaction matrix element

$$V_{ab} = V_{ba}^* = \int d^3r\, u_a^* \hat{V} u_b. \tag{6.5.7}$$

Similarly, by multiplying instead by u_b^* and again integrating over all space, we find that

$$\dot{C}_b = \frac{1}{i\hbar} C_a V_{ba} e^{i\omega_{ba}t}. \tag{6.5.8}$$

We now explicitly introduce the form of the interaction Hamiltonian and represent the interaction matrix elements as

$$V_{ab}^* = V_{ba} = -\mu_{ba}\tilde{E}(t) = -\mu_{ba}\left(Ee^{-i\omega t} + E^* e^{i\omega t}\right). \tag{6.5.9}$$

Eqs. (6.5.6) and (6.5.8) then become

$$\dot{C}_a = \frac{-\mu_{ab}}{i\hbar} C_b \left(E^* e^{-i(\omega_{ba}-\omega)t} + Ee^{-i(\omega_{ba}+\omega)t}\right) \tag{6.5.10a}$$

and

$$\dot{C}_b = \frac{-\mu_{ba}}{i\hbar} C_a \left(Ee^{i(\omega_{ba}-\omega)t} + E^* e^{i(\omega_{ba}+\omega)t}\right). \tag{6.5.10b}$$

We next make the rotating-wave approximation, that is, we drop the rapidly oscillating second terms in these equations and retain only the first terms.* We also introduce the detuning factor

$$\Delta = \omega - \omega_{ba}. \tag{6.5.11}$$

The coupled equations (6.5.10) then reduce to the set

$$\dot{C}_a = i\frac{\mu_{ab}E^*}{\hbar} C_b e^{i\Delta t}, \tag{6.5.12a}$$

$$\dot{C}_b = i\frac{\mu_{ba}E}{\hbar} C_a e^{-i\Delta t}. \tag{6.5.12b}$$

This set of equations can be readily solved by adopting a trial solution of the form

$$C_a = Ke^{-i\lambda t}. \tag{6.5.13}$$

This expression is introduced into Eq. (6.5.12a), which shows that C_b must be of the form

$$C_b = \frac{-\hbar\lambda K}{\mu_{ab}E^*} e^{-i(\lambda+\Delta)t}. \tag{6.5.14}$$

* See also the discussion preceding Eq. (6.3.5).

This form for C_b and the trial solution (6.5.13) for C_a are now introduced into Eq. (6.5.12b), which shows that the characteristic frequency λ must obey the equation

$$\lambda(\lambda + \Delta) = \frac{|\mu_{ba}|^2 |E|^2}{\hbar^2}. \tag{6.5.15}$$

The solutions of this equation are

$$\lambda_\pm = -\tfrac{1}{2}\Delta \pm \tfrac{1}{2}\Omega', \tag{6.5.16}$$

where we have introduced the generalized (or detuned) Rabi frequency

$$\Omega' = \left(|\Omega|^2 + \Delta^2\right)^{1/2} \tag{6.5.17}$$

and where, as before, $\Omega = 2\mu_{ba}E/\hbar$ denotes the complex Rabi frequency. The general solution to Eqs. (6.5.12) for $C_a(t)$ can thus be expressed as

$$C_a(t) = e^{(1/2)i\Delta t}\left(A_+ e^{-(1/2)i\Omega' t} + A_- e^{(1/2)i\Omega' t}\right), \tag{6.5.18a}$$

where A_+ and A_- are constants of integration whose values depend on the initial conditions. The corresponding expression for $C_b(t)$ is obtained by introducing this result into Eq. (6.5.12a):

$$\begin{aligned} C_b(t) &= \frac{-\hbar \dot{C}_a}{\mu_{ab} E^*} e^{-i\Delta t} \\ &= e^{-(1/2)i\Delta t}\left(\frac{\Delta - \Omega'}{\Omega^*} A_+ e^{-(1/2)i\Omega' t} + \frac{\Delta + \Omega'}{\Omega^*} A_- e^{(1/2)i\Omega' t}\right). \end{aligned} \tag{6.5.18b}$$

Eqs. (6.5.18) give the general solution to Eqs. (6.5.12). Next, we find the specific solution for two different sets of initial conditions.

6.5.2 Solution for an Atom Initially in the Ground State

One realistic set of initial conditions is that of an atom known to be in the ground state at time $t = 0$ so that

$$C_a(0) = 1 \quad \text{and} \quad C_b(0) = 0. \tag{6.5.19}$$

Eq. (6.5.18a) evaluated at $t = 0$ then shows that

$$A_+ + A_- = 1, \tag{6.5.20}$$

while Eq. (6.5.18b) evaluated at $t = 0$ shows that

$$(\Delta - \Omega')A_+ + (\Delta + \Omega')A_- = 0. \tag{6.5.21}$$

These equations are solved algebraically to find that

$$A_+ = 1 - A_- = \frac{\Omega' + \Delta}{2\Omega'}. \tag{6.5.22}$$

The probability amplitudes $C_a(t)$ and $C_b(t)$ are now determined by introducing these expressions for A_+ and A_- into Eqs. (6.5.18), to obtain

$$\begin{aligned} C_a(t) &= e^{(1/2)i\Delta t}\left[\left(\frac{\Omega' + \Delta}{2\Omega'}\right)e^{-(1/2)i\Omega' t} + \left(\frac{\Omega' + \Delta}{2\Omega'}\right)e^{(1/2)i\Omega' t}\right] \\ &= e^{(1/2)i\Delta t}\left[\cos\left(\tfrac{1}{2}\Omega' t\right) - \frac{i\Delta}{\Omega'}\sin\left(\tfrac{1}{2}\Omega' t\right)\right] \end{aligned} \tag{6.5.23}$$

and

$$\begin{aligned} C_b(t) &= e^{-(1/2)i\Delta t}\left(\frac{-\Omega}{2\Omega'}e^{-(1/2)i\Omega' t} + \frac{\Omega}{2\Omega'}e^{(1/2)i\Omega' t}\right) \\ &= ie^{-(1/2)i\Delta t}\left[\frac{\Omega}{\Omega'}\sin\left(\tfrac{1}{2}\Omega' t\right)\right]. \end{aligned} \tag{6.5.24}$$

The probability that the atom is in level a at time t is hence given by

$$|C_a|^2 = \cos^2\left(\tfrac{1}{2}\Omega' t\right) + \frac{\Delta^2}{\Omega'^2}\sin^2\left(\tfrac{1}{2}\Omega' t\right), \tag{6.5.25}$$

while the probability of being in level b is given by

$$|C_b|^2 = \frac{|\Omega|^2}{\Omega'^2}\sin^2\left(\tfrac{1}{2}\Omega' t\right). \tag{6.5.26}$$

Note that (since $\Omega'^2 = |\Omega|^2 + \Delta^2$)

$$|C_a|^2 + |C_b|^2 = 1, \tag{6.5.27}$$

which shows that probability is conserved.

For the case of exact resonance ($\Delta = 0$), Eqs. (6.5.25) and (6.5.26) reduce to

$$|C_a|^2 = \cos^2\left(\tfrac{1}{2}|\Omega| t\right), \tag{6.5.28a}$$

$$|C_b|^2 = \sin^2\left(\tfrac{1}{2}|\Omega| t\right), \tag{6.5.28b}$$

and the probabilities oscillate between zero and one in the simple manner illustrated in Fig. 6.5.1. Note that, since the probability amplitude C_a oscillates at angular frequency $|\Omega|/2$, the probability $|C_a|^2$ oscillates at angular frequency $|\Omega|$, that is, at the Rabi frequency. As the detuning Δ is increased, the angular frequency at which the population oscillates increases,

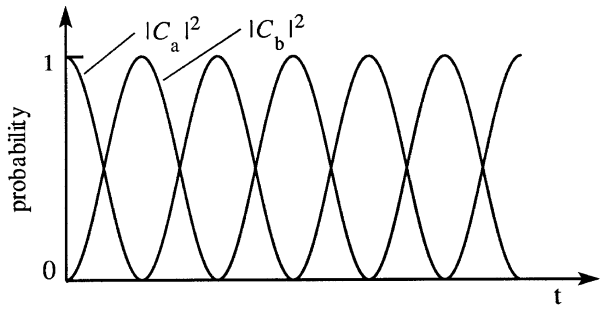

FIGURE 6.5.1: Rabi oscillations of the populations in the ground ($|C_a|^2$) and excited ($|C_b|^2$) states for the case of exact resonance ($\Delta = 0$).

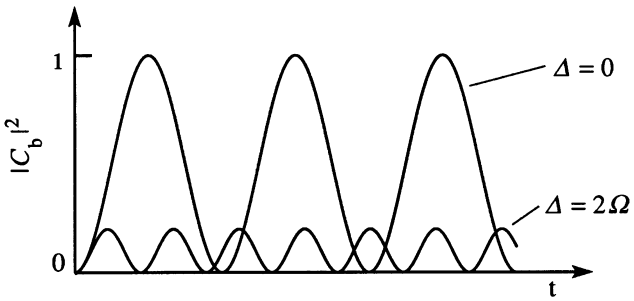

FIGURE 6.5.2: Rabi oscillations of the excited state population for two values of $\Delta \equiv \omega - \omega_{ba}$.

since the generalized Rabi frequency is given by $\Omega' = [|\Omega|^2 + \Delta^2]^{1/2}$, but the amplitude of the oscillation decreases, as shown in Fig. 6.5.2.

Next, we calculate the expectation value of the atomic dipole moment for an atom known to be in the atomic ground state at time $t = 0$. This quantity is given by

$$\langle \tilde{\mu} \rangle = \langle \psi | \tilde{\mu} | \psi \rangle, \tag{6.5.29}$$

where $\psi(\mathbf{r}, t)$ is given by Eq. (6.5.4). We assume as before that $\langle a|\tilde{\mu}|a\rangle = \langle b|\tilde{\mu}|b\rangle = 0$, and we denote the nonvanishing matrix elements of $\tilde{\mu}$ by

$$\mu_{ab} = \langle a|\tilde{\mu}|b\rangle = \langle b|\tilde{\mu}|a\rangle^* = \mu_{ba}^*. \tag{6.5.30}$$

We thus find that the induced dipole moment is given by

$$\langle \tilde{\mu} \rangle = C_a^* C_b \mu_{ab} e^{-i\omega_{ba}t} + \text{c.c.} \tag{6.5.31}$$

or, by introducing Eqs. (6.5.23) and (6.5.24) for C_a and C_b, by

$$\langle\tilde{\mu}\rangle = \mu_{ab}\frac{\Omega}{\Omega'}\left[\frac{-\Delta}{2\omega'}e^{-i\omega t} + \frac{1}{4}\left(\frac{\Delta}{\Omega'} - 1\right)e^{-i(\omega+\Omega')t} + \frac{1}{4}\left(\frac{\Delta}{\Omega'} + 1\right)e^{-i(\omega-\Omega')t}\right] + \text{c.c.} \tag{6.5.32}$$

This result shows that the atomic dipole oscillates not only at the driving frequency ω but also at the Rabi sideband frequencies $\omega + \Omega'$ and $\omega - \Omega'$. We can understand the origin of this effect by considering the frequencies that are present in the atomic wavefunction. We recall that the wavefunction is given by Eq. (6.5.4), where (according to Eqs. (6.5.23) and (6.5.24)) $C_a(t)$ contains frequencies $-\frac{1}{2}(\Delta \pm \Omega')$ and $C_b(t)$ contains frequencies $\frac{1}{2}(\Delta \pm \Omega')$. Fig. 6.5.3 shows graphically the frequencies that are present in the atomic wavefunction. Note that the frequen-

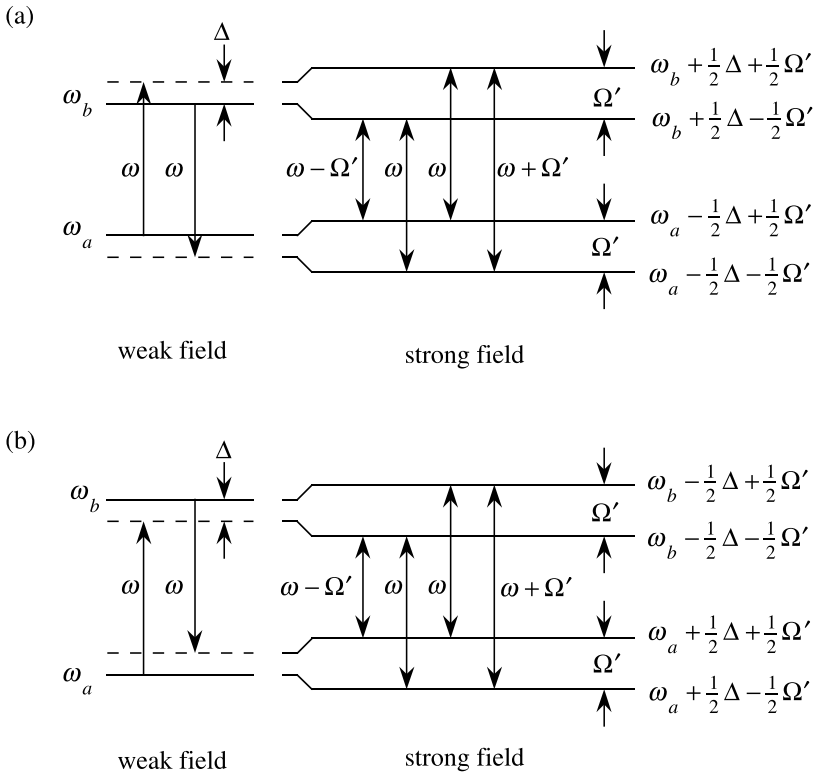

FIGURE 6.5.3: Frequency spectrum of the atomic wavefunction given by Eq. (6.5.4) (with $C_a(t)$ and $C_b(t)$ given by Eqs. (6.5.28) and (6.5.29)) for the case of (a) positive detuning ($\Delta > 0$) and (b) negative detuning ($\Delta < 0$).

cies at which the atomic dipole oscillates correspond to differences of the various frequency components of the wavefunction.

6.5.3 Dressed States

Another important solution to the Schrödinger equation for a two-level atom is that corresponding to the dressed atomic states (Autler and Townes, 1955; Cohen-Tannoudji and Reynaud, 1977). The characteristic feature of these states is that the probability to be in atomic level a (or b) is constant in time. As a consequence, the probability amplitudes $C_a(t)$ and $C_b(t)$ can depend on time only in terms of exponential phase factors. Recall, however, that in general (that is, for states that are not dressed states) $C_a(t)$ and $C_b(t)$ are given by the more complicated expressions (6.5.18).

There are two ways in which the solution of Eqs. (6.5.18) can lead to time-independent probabilities of occupancy for levels a and b. One such solution, which we designate as ψ_+, corresponds to the case in which the integration constants A_+ and A_- have the values

$$A_+ = 1, \qquad A_- = 0 \quad (\text{for } \psi_+); \tag{6.5.33a}$$

the other solution, which we designate as ψ_-, corresponds to the case in which

$$A_+ = 0, \qquad A_- = 1 \quad (\text{for } \psi_-). \tag{6.5.33b}$$

Explicitly, the atomic wavefunction corresponding to each of these solutions is given, through use of Eqs. (6.5.4), (6.5.18), and (6.5.33), as

$$\begin{aligned}\psi_\pm = N_\pm \Bigg\{ & u_a(\mathbf{r}) \exp\left[-i\left(\omega_a - \tfrac{1}{2}\Delta \pm \tfrac{1}{2}\Omega'\right)t\right] \\ & + \frac{\Delta \mp \Omega'}{\Omega^*} u_b(\mathbf{r}) \exp\left[-i\left(\omega_b + \tfrac{1}{2}\Delta \pm \tfrac{1}{2}\Omega'\right)t\right] \Bigg\},\end{aligned} \tag{6.5.34}$$

where $N_\pm$ is a normalization constant. The value of this constant is determined by requiring that

$$\int |\psi_\pm|^2 \, d^3r = 1. \tag{6.5.35}$$

By introducing Eq. (6.5.34) into this expression and performing the integrations, we find that

$$|N_\pm|^2 \left[1 + \frac{(\Delta \mp \Omega')^2}{|\Omega|^2}\right] = 1. \tag{6.5.36}$$

For future convenience, we choose the phases of $N_\pm$ such that $N_\pm$ are given by

$$N_\pm = \frac{\Omega^*}{\Omega'} \left[\frac{\Omega'}{2(\Omega' \mp \Delta)}\right]^{1/2}. \tag{6.5.37}$$

The normalized dressed-state wavefunctions are hence given by

$$\psi_{\pm} = \frac{\Omega^*}{\Omega'}\left[\frac{\Omega'}{2(\Omega' \mp \Delta)}\right]^{1/2} u_a(\mathbf{r}) \exp\left[-i\left(\omega_a - \tfrac{1}{2}\Delta \pm \tfrac{1}{2}\Omega'\right)t\right]$$
$$\mp \left[\frac{\Omega' \mp \Delta}{2\Omega'}\right]^{1/2} u_b(\mathbf{r}) \exp\left[-i\left(\omega_b + \tfrac{1}{2}\Delta \pm \tfrac{1}{2}\Omega'\right)t\right]. \tag{6.5.38}$$

We next examine some of the properties of the dressed states. The probability amplitude for an atom in the dressed state $\psi_{\pm}$ to be in the atomic level a is given by

$$\langle a|\psi_{\pm}\rangle = \frac{\Omega^*}{\Omega'}\left[\frac{\Omega'}{2(\Omega' \mp \Delta)}\right]^{1/2} \exp\left[-i\left(\omega_a - \tfrac{1}{2}\Delta \pm \tfrac{1}{2}\Omega'\right)t\right], \tag{6.5.39}$$

and thus the probability of finding the atom in the state a is given by

$$\left|\langle a|\psi_{\pm}\rangle\right|^2 = \frac{|\Omega|^2}{\Omega'^2}\frac{\Omega'}{2(\Omega' \mp \Delta)} = \frac{|\Omega|^2}{2\Omega'(\Omega' \mp \Delta)}. \tag{6.5.40}$$

Similarly, the probability amplitude of finding the atom in state b is given by

$$\langle b|\psi_{\pm}\rangle = \mp\left(\frac{\Omega' \mp \Delta}{2\Omega'}\right)^{1/2} \exp\left[-i\left(\omega_b + \tfrac{1}{2}\Delta \pm \tfrac{1}{2}\Omega'\right)t\right], \tag{6.5.41}$$

and thus the probability of finding the atom in the state b is given by

$$\left|\langle b|\psi_{\pm}\rangle\right|^2 = \frac{\Omega' \mp \Delta}{2\Omega'}. \tag{6.5.42}$$

Note that these probabilities of occupancy are indeed constant in time; in this sense the dressed states constitute the stationary states of the coupled atom–field system.

The dressed states $\psi_{\pm}$ are solutions of Schrödinger's equation in the presence of the total Hamiltonian $\hat{H} = \hat{H}_0 + \hat{V}(t)$. Thus, if the system is known to be in state ψ_+ (or ψ_-) at the time $t = 0$, the system will remain in this state, even though the system is subject to the interaction Hamiltonian $\hat{V}$. They are stationary states in the sense just mentioned that the probability of finding the atom in either of the atomic states a or b is constant in time. Although the states $\psi_{\pm}$ are stationary states, they are not energy eigenstates, because the Hamiltonian $\hat{H}$ depends explicitly on time.

It is easy to demonstrate that the dressed states are orthogonal—that is, that

$$\langle \psi_+|\psi_-\rangle = 0. \tag{6.5.43}$$

The expectation value of the induced dipole moment for an atom in a dressed state is given by

$$\left\langle \psi_{\pm}|\hat{\mu}|\psi_{\pm}\right\rangle = \mp\frac{\Omega}{2\Omega'}\mu_{ab}e^{-i\omega t} + \text{c.c.} \tag{6.5.44}$$

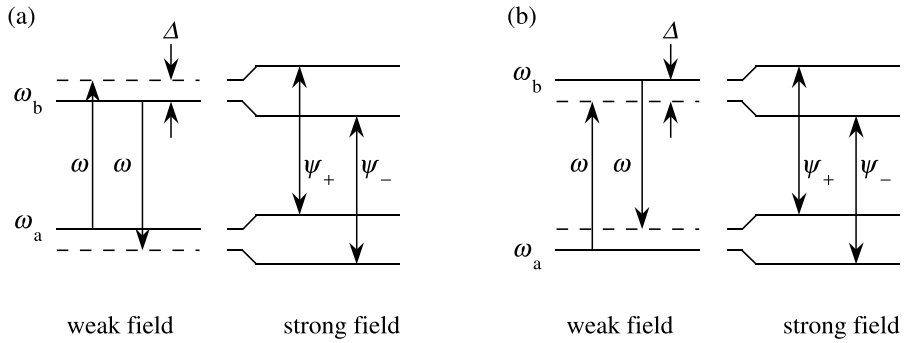

FIGURE 6.5.4: The dressed atomic states ψ_+ and ψ_- for Δ positive (a) and negative (b).

Thus, the induced dipole moment of an atom in a dressed state oscillates only at the driving frequency. However, the dipole transition moment between the dressed states is nonzero:

$$\langle \psi_\pm | \hat{\mu} | \psi_\mp \rangle = \pm \mu_{ab} \frac{\Omega}{2\Omega'} \left(\frac{\Omega' \pm \Delta}{\Omega' \mp \Delta} \right)^{1/2} e^{-i(\omega \mp \Omega')t} \mp \mu_{ba} \frac{\Omega^*}{2\Omega'} \left(\frac{\Omega' \mp \Delta}{\Omega' \pm \Delta} \right)^{1/2} e^{i(\omega \pm \Omega')t}. \tag{6.5.45}$$

The properties of the dressed states are summarized in the frequency level diagram shown for the case of positive Δ in Fig. 6.5.4(a) and for the case of negative Δ in Fig. 6.5.4(b).

Next, we consider the limiting form of the dressed states for the case of a weak applied field—that is, for $|\Omega| \ll |\Delta|$. In this limit, we can approximate the generalized Rabi frequency Ω' as

$$\Omega' = \left(|\Omega|^2 + \Delta^2 \right)^{1/2} = |\Delta| \left(1 + \frac{|\Omega|^2}{\Delta^2} \right)^{1/2} \simeq |\Delta| \left(1 + \frac{1}{2} \frac{|\Omega|^2}{\Delta^2} \right). \tag{6.5.46}$$

Using this result, we can approximate the dressed-state wavefunctions of Eq. (6.5.38) for the case of positive Δ as

$$\psi_+ = \frac{\Omega^*}{|\Omega|} u_a e^{-i\omega_a t} - \frac{|\Omega|}{2\Delta} u_b e^{i(\omega_b + \Delta)t}, \tag{6.5.47a}$$

$$\psi_- = \frac{\Omega^*}{2\Delta} u_a e^{-i(\omega_a - \Delta)t} + u_b e^{-i\omega_b t}. \tag{6.5.47b}$$

We note that in this limit ψ_+ is primarily ψ_a and ψ_- is primarily ψ_b. The smaller contribution to ψ_+ can be identified with the virtual level induced by the transition. For the case of negative Δ,

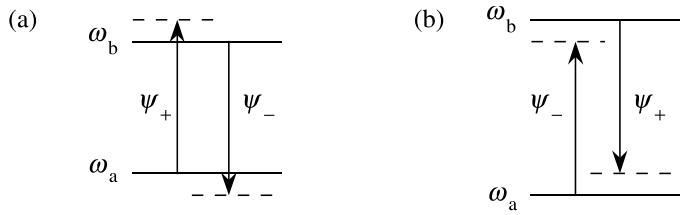

FIGURE 6.5.5: The weak field limit of the dressed states ψ_+ and ψ_- for the case of (a) positive and (b) negative detuning Δ.

we obtain

$$\psi_+ = -\frac{\Omega^*}{2\Delta} u_a e^{-i(\omega_a - \Delta)t} - u_b e^{-i\omega_b t}, \tag{6.5.48a}$$

$$\psi_- = \frac{\Omega^*}{|\Omega|} u_a e^{-i\omega_a t} - \frac{|\Omega|}{2\Delta} u_b e^{-i(\omega_b + \Delta)t}. \tag{6.5.48b}$$

Now ψ_+ is primarily ψ_b, and ψ_- is primarily ψ_a. These results are illustrated in Fig. 6.5.5. Note that these results have been anticipated in drawing certain of the levels as dashed lines in the weak-field limit of the diagrams shown in Figs. 6.5.3 and 6.5.4.

6.5.4 Inclusion of Relaxation Phenomena

In the absence of damping phenomena, it is adequate to treat the response of a two-level atom to an applied optical field by solving Schrödinger's equation for the time evolution of the wave-function. We have seen that under such circumstances the population inversion oscillates at the generalized Rabi frequency $\Omega' = (\Omega^2 + \Delta^2)^{1/2}$. If damping effects are present, we expect that these Rabi oscillations will eventually become damped out and that the population difference will approach some steady-state value. In order to treat this behavior, we need to solve the density matrix equations of motion with the inclusion of damping effects. We take the density matrix equations in the form (essentially Eqs. (6.4.9) with $w_{eq} = -1$ and $\kappa = 2\mu E/\hbar$)

$$\dot{p} = \left(i\Delta - \frac{1}{T_2}\right) p - \frac{i}{\hbar} |\mu|^2 E w, \tag{6.5.49a}$$

$$\dot{w} = -\frac{w+1}{T_1} - \frac{2i}{\hbar}(pE^* - p^* E), \tag{6.5.49b}$$

and we assume that at $t = 0$ the atom is in its ground state—that is, that

$$p(0) = 0, \quad w(0) = -1, \tag{6.5.50}$$

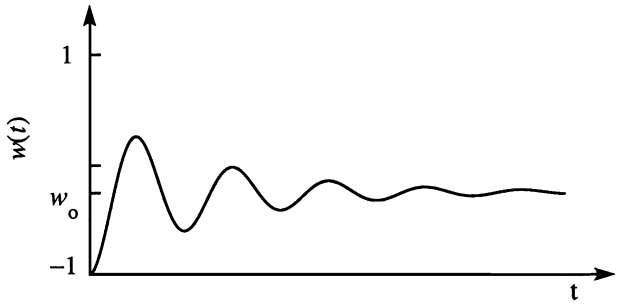

FIGURE 6.5.6: Damped Rabi oscillations.

and that the field $\tilde{E}(t)$ is turned on at $t = 0$ and oscillates harmonically thereafter (i.e., $E = 0$ for $t < 0$, $E =$ constant for $t \geq 0$).

Eqs. (6.5.49) can be solved in general under the conditions given above (see Problem 4 at the end of this chapter). For the special case in which $T_1 = T_2$, the form of the solution is considerably simpler than in the general case. The solution to Eqs. (6.5.49) for the population inversion for this special case is given by

$$w(t) = w_0 - (1 + w_0)e^{-t/T_2}\left[\cos\Omega' t + \frac{1}{\Omega' T_2}\sin\Omega' t\right], \tag{6.5.51a}$$

where

$$w_0 = \frac{-(1 + \Delta^2 T_2^2)}{1 + \Delta^2 T_2^2 + \Omega^2 T_1 T_2}. \tag{6.5.51b}$$

The nature of this solution is shown in Fig. 6.5.6. Note that the Rabi oscillations are damped out in a time of the order of T_2. Once the Rabi oscillations have damped out, the system enters one of the dressed states of the coupled atom–field system.

In summary, we have seen that, in the absence of damping effects, the population inversion of a strongly driven two-level atom oscillates at the generalized Rabi frequency $\Omega' = (\Omega^2 + \Delta^2)^{1/2}$ and that consequently the induced dipole moment oscillates at the applied frequency ω and also at the Rabi sideband frequencies $\omega \pm \Omega'$. In the presence of dephasing processes, the Rabi oscillations die out in a characteristic time given by the dipole dephasing time T_2. Hence, Rabi oscillations are not present in the steady state.

In the following section, we explore the nature of the response of the atom to a strong field at frequency ω and a weak field at frequency $\omega + \delta$. If the frequency difference δ (or its negative $-\delta$) between these two fields is nearly equal to the generalized Rabi frequency Ω', the beat frequency between the two applied fields can act as a source term to drive the Rabi oscillation. We shall find that, in the presence of such a field, the population difference oscillates at the beat frequency δ, and that the induced dipole moment contains the frequency components ω and $\omega \pm \delta$.

6.6 Optical Wave Mixing in Two-Level Systems

In the present section we consider the response of a collection of two-level atoms to the simultaneous presence of a strong optical field (which we call the pump field) and one or more weak optical fields (which we call probe fields). These latter fields are considered weak in the sense that they alone cannot saturate the response of the atomic system.

An example of such an occurrence is saturation spectroscopy, using a setup of the sort shown in Fig. 6.6.1. In such an experiment, one determines how the response of the medium to the probe wave is modified by the presence of the pump wave. Typically, one might measure the transmission of the probe wave as a function of the frequency ω and intensity of the pump wave and of the frequency detuning δ between the pump and probe waves. The results of such experiments can be used to obtain information regarding the dipole transition moments and the relaxation times T_1 and T_2.

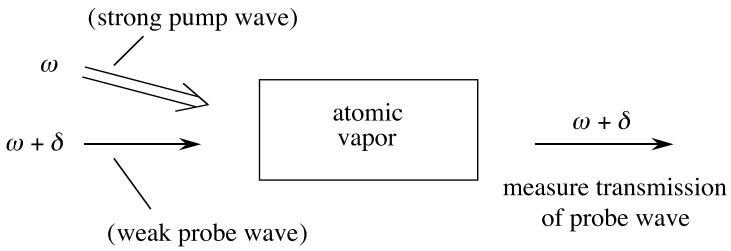

FIGURE 6.6.1: Saturation spectroscopy setup.

Another example of the interactions considered in this section is the multiwave mixing experiment shown in part (a) of Fig. 6.6.2. Here the pump wave at frequency ω and the probe wave at frequency $\omega + \delta$ are copropagating (or nearly copropagating) through the medium. For this geometry, the four-wave mixing process shown in part (b) of the figure becomes phase-matched (or nearly phase-matched), and this process leads to the generation of the symmetric sideband at frequency $\omega - \delta$.

At low intensities of the pump laser, the response of the atomic system at the frequencies $\omega + \delta$ and $\omega - \delta$ can be calculated using perturbation theory of the sort developed in Chapter 3. In this limit, one finds that the absorption (and dispersion) experienced by the probe wave in the geometry of Fig. 6.6.1 is somewhat reduced by the presence of the pump wave. One also finds that, for the geometry of Fig. 6.6.2, the intensity of the generated sideband at frequency $\omega - \delta$ increases quadratically as the pump intensity is increased.

In this section we show that the character of these nonlinear processes is profoundly modified when the intensity of the pump laser is increased to the extent that perturbation theory is not sufficient to describe the interaction. These higher-order processes become important when the Rabi frequency Ω associated with the pump field is greater than both the detuning Δ of the

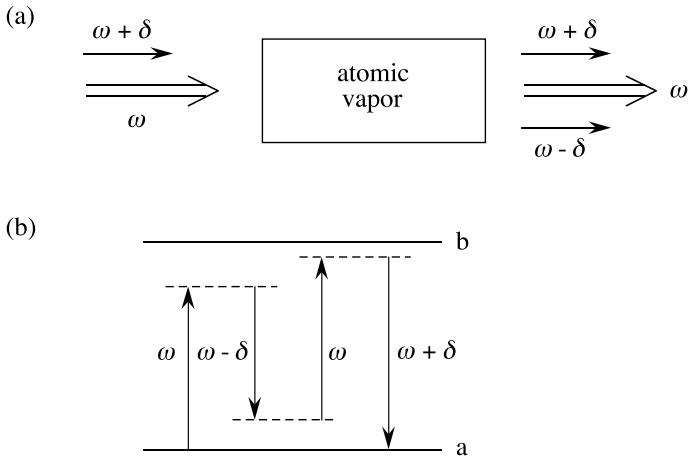

FIGURE 6.6.2: (a) Forward four-wave mixing. (b) Energy-level description of the four-wave mixing process, drawn for clarity for the case in which δ is negative.

pump wave from the atomic resonance and the transition linewidth $1/T_2$. Under this condition, the atomic energy levels are strongly modified by the pump field, leading to new resonances in the absorptive and mixing responses. In particular, we shall find that these new resonances can be excited when the pump–probe detuning δ is approximately equal to $\pm\Omega'$, where Ω' is the generalized Rabi frequency.

6.6.1 Solution of the Density Matrix Equations for a Two-Level Atom in the Presence of Pump and Probe Fields

We have seen in Section 6.4 that the dynamical behavior of a two-level atom in the presence of the optical field

$$\tilde{E}(t) = Ee^{-i\omega t} + \text{c.c.} \tag{6.6.1}$$

can be described in terms of equations of motion for the population inversion $w = \rho_{bb} - \rho_{aa}$ and the complex dipole amplitude $p = \mu_{ab}\sigma_{ba}$, which is related to the expectation value $\tilde{p}(t)$ of the atomic dipole moment by

$$\tilde{p}(t) = pe^{-i\omega t} + \text{c.c.} \tag{6.6.2}$$

The equations of motion for p and w are given explicitly by (see Eqs. (6.4.9) and take $\kappa = 2\mu E/\hbar$)

$$\frac{dp}{dt} = \left(i\Delta - \frac{1}{T_2}\right)p - \frac{i}{\hbar}|\mu_{ba}|^2 Ew, \tag{6.6.3}$$

$$\frac{dw}{dt} = -\frac{w - w^{(\mathrm{eq})}}{T_1} + \frac{4}{\hbar}\,\mathrm{Im}(pE^*), \tag{6.6.4}$$

where $\Delta = \omega - \omega_{ba}$. For the problem at hand, we represent the amplitude of the applied optical field as

$$E = E_0 + E_1 e^{-i\delta t}, \tag{6.6.5}$$

where we assume that $|E_1| \ll |E_0|$. By introducing Eq. (6.6.5) into Eq. (6.6.1), we find that the electric field can alternatively be expressed as

$$\tilde{E}(t) = E_0 e^{-i\omega t} + E_1 e^{-i(\omega+\delta)t} + \text{c.c.}; \tag{6.6.6}$$

hence, E_0 and E_1 represent the complex amplitudes of the pump and probe waves, respectively.

Eqs. (6.6.3) and (6.6.4) cannot readily be solved exactly for the field given in Eq. (6.6.5). Instead, our strategy will be to find a solution that is exact in the amplitude E_0 of the strong field and is correct to lowest order in the amplitude E_1 of the weak field. We hence require that the steady-state solution of Eqs. (6.6.3) and (6.6.4) be of the form

$$p = p_0 + p_1 e^{-i\delta t} + p_{-1} e^{i\delta t} \tag{6.6.7}$$

and

$$w = w_0 + w_1 e^{-i\delta t} + w_{-1} e^{i\delta t}, \tag{6.6.8}$$

where p_0 and w_0 denote the solution for the case in which only the pump field E_0 is present, and where the other terms are assumed to be small in the sense that

$$|p_1|, |p_{-1}| \ll |p_0|, \quad |w_1|, |w_{-1}| \ll |w_0|. \tag{6.6.9}$$

Note that, to the lowest order in the amplitude E_1 of the probe field, 0 and $\pm\delta$ are the only frequencies that can be present in the solution of Eqs. (6.6.3) and (6.6.4). Note also that, in order for $w(t)$ to be a real quantity, w_{-1} must be equal to w_1^*. Hence, $w(t)$ is of the form $w(t) = w_0 + 2|w_1|\cos(\delta t + \phi)$, where ϕ is the phase of w. Thus, in the simultaneous presence of pump and probe fields, the population difference oscillates harmonically at the pump–probe frequency difference, and w_1 represents the complex amplitude of the population oscillation.

We now introduce the trial solution (6.6.7) and (6.6.8) into the density matrix equations (6.6.3) and (6.6.4) and equate terms with the same time dependence. In accordance with our perturbation assumptions, we drop any term that contains the product of more than one small quantity. Then, for example, the zero-frequency part of the equation of motion for dipole amplitude, Eq. (6.6.3), becomes

$$0 = \left(i\Delta - \frac{1}{T_2}\right) p_0 - \frac{i}{\hbar}|\mu_{ba}|^2 E_0 w_0,$$

whose solution is

$$p_0 = \frac{\hbar^{-1}|\mu_{ba}|^2 E_0 w_0}{\Delta + i/T_2}. \tag{6.6.10}$$

Likewise, the part of Eq. (6.6.3) oscillating as $e^{-i\delta t}$ is

$$-i\delta p_1 = \left(i\Delta - \frac{1}{T_2}\right)p_1 - \frac{i}{\hbar}|\mu_{ba}|^2(E_0 w_1 + E_1 w_0),$$

which can be solved algebraically to obtain

$$p_1 = \frac{\hbar^{-1}|\mu_{ba}|^2(E_0 w_1 + E_1 w_0)}{(\Delta - \delta) + i/T_2}; \tag{6.6.11}$$

the part of Eq. (6.6.3) oscillating as $e^{i\delta t}$ is similarly given by

$$i\delta p_{-1} = \left(i\Delta - \frac{1}{T_2}\right)p_{-1} - \frac{i}{\hbar}|\mu_{ba}|^2(E_0 w_{-1}),$$

which can be solved to obtain

$$p_{-1} = \frac{\hbar^{-1}|\mu_{ba}|^2 E_0 w_{-1}}{(\Delta - \delta) + i/T_2}. \tag{6.6.12}$$

Next, we consider the solution of the inversion equation (6.6.4). We introduce the trial solution (6.6.7) and (6.6.8) into this equation. The zero-frequency part of the resulting expression is

$$0 = -\frac{w_0 - w^{(\mathrm{eq})}}{T_1} + \frac{4}{\hbar}\,\mathrm{Im}\big(p_0 E_0^*\big). \tag{6.6.13}$$

We now introduce Eq. (6.6.10) for p_0 into this expression to obtain

$$\frac{w_0 - w^{(\mathrm{eq})}}{T_1} = \Omega^2 w_0\,\mathrm{Im}\left(\frac{\Delta - i/T_2}{\Delta^2 + 1/T_2^2}\right) = \frac{-\Omega^2 w_0/T_2}{\Delta^2 + 1/T_2^2}, \tag{6.6.14}$$

where we have introduced the on-resonance Rabi frequency $\Omega = 2|\mu_{ba}E|/\hbar$. We now solve Eq. (6.6.14) algebraically for w_0 to obtain

$$w_0 = \frac{w^{(\mathrm{eq})}(1 + \Delta^2 T_2^2)}{1 + \Delta^2 T_2^2 + \Omega^2 T_1 T_2}. \tag{6.6.15}$$

We next consider the oscillating part of Eq. (6.6.4). The part of $\mathrm{Im}(pE^*)$ oscillating at frequencies $\pm\delta$ is given by

$$\begin{aligned}\mathrm{Im}(pE^*) &= \mathrm{Im}\big(p_0E_1^*e^{i\delta t} + p_1E_0^*e^{-i\delta t} + p_{-1}E_0^*e^{i\delta t}\big)\\ &= \frac{1}{2i}\big(p_0E_1^*e^{i\delta t} + p_1E_0^*e^{-i\delta t} + p_{-1}E_0^*e^{i\delta t}\\ &\quad - p_0^*E_1e^{-i\delta t} - p_1^*E_0e^{i\delta t} - p_{-1}^*E_0e^{-i\delta t}\big),\end{aligned} \tag{6.6.16}$$

where in obtaining the second form we have used the identity $\mathrm{Im}\,z = (z - z^*)/2i$. We now introduce this result into Eq. (6.6.4). The part of the resulting expression that varies as $e^{-i\delta t}$ is

$$-i\delta w_1 = \frac{-w_1}{T_1} - \frac{2i}{\hbar}\big(p_1E_0^* - p_0^*E_1 - p_{-1}^*E_0\big).$$

This expression is solved for w_1 to obtain

$$w_1 = \frac{2\hbar^{-1}(p_1E_0^* - p_0^*E_1 - p_{-1}^*E_0)}{\delta + i/T_1}. \tag{6.6.17}$$

We similarly find from the part of Eq. (6.6.4) oscillating as $e^{i\delta t}$ that

$$w_{-1} = \frac{2\hbar^{-1}(p_1^*E_0 - p_0E_1^* - p_{-1}E_0^*)}{\delta - i/T_1}. \tag{6.6.18}$$

Note that $w_{-1} = w_1^*$, as required from the condition that $w(t)$ as given by Eq. (6.6.8) be real.

At this point we have a set of six coupled equations [(6.6.10), (6.6.11), (6.6.12), (6.6.15), (6.6.17), (6.6.18)] for the six quantities p_0, p_1, p_{-1}, w_0, w_1, w_{-1}. We note that w_0 is given by Eq. (6.6.15) in terms of known quantities. Our strategy is thus to solve next for w_1, since the other unknown quantities are simply related to w_0 and w_1. We thus introduce the expressions for p_1, p_0, and p_{-1} into Eq. (6.6.17), which becomes

$$\begin{aligned}\left(\delta + \frac{i}{T_1}\right)w_1 &= \frac{2|\mu_{ba}|^2}{\hbar^2}\\ &\quad\times\left(\frac{|E_0|^2w_1}{\Delta + \delta + i/T_2} + \frac{E_1E_0^*w_0}{\Delta + \delta + i/T_2} - \frac{E_1E_0^*w_0}{\Delta - i/T_2} - \frac{|E_0|^2w_1}{\Delta - \delta - i/T_2}\right).\end{aligned} \tag{6.6.19}$$

This equation is now solved algebraically for w_1, yielding

$$w_1 = -\frac{w_0^2|\mu_{ba}|^2E_1E_0^*\hbar^{-2}(\delta - \Delta + i/T_2)(\delta + 2i/T_2)(\Delta - i/T_2)^{-1}}{(\delta + i/T_1)(\delta - \Delta + i/T_2)(\Delta + \delta + i/T_2) - \Omega^2(\delta + i/T_2)}. \tag{6.6.20}$$

The combination of terms that appears in the denominator of this expression appears repeatedly in the subsequent equations. For convenience we denote this combination as

$$D(\delta) = \left(\delta + \frac{i}{T_1}\right)\left(\delta - \Delta + \frac{i}{T_2}\right)\left(\delta + \Delta + \frac{i}{T_2}\right) - \Omega^2\left(\delta + \frac{i}{T_2}\right), \tag{6.6.21}$$

so that Eq. (6.6.20) can be written as

$$w_1 = -2w_0|\mu_{ba}|^2 E_1 E_0^* \hbar^{-2} \frac{(\delta - \Delta + i/T_2)(\delta + 2i/T_2)}{(\Delta - i/T_2)D(\delta)}. \tag{6.6.22}$$

Note that w_1 (and consequently p_1 and p_{-1}) shows a resonance whenever the pump wave is tuned to line center so that $\Delta = 0$, or whenever a zero occurs in the function $D(\delta)$. We next examine the resonance nature of the function $D(\delta)$. We first consider the limit $\Omega^2 \to 0$—that is, the $\chi^{(3)}$ perturbation theory limit. In this limit $D(\delta)$ is automatically factored into the product of three terms as

$$D(\delta) = \left(\delta + \frac{i}{T_1}\right)\left(\delta - \Delta + \frac{i}{T_2}\right)\left(\Delta + \delta + \frac{i}{T_2}\right), \tag{6.6.23}$$

and we see by inspection that zeros of $D(\delta)$ occur at

$$\delta = 0, \pm\Delta. \tag{6.6.24}$$

The positions of these frequencies are indicated in part (a) of Fig. 6.6.3. However, inspection of Eq. (6.6.22) shows that no resonance occurs in w_1 at $\delta = \Delta$, because the factor $\delta - \Delta + i/T_2$ in the numerator exactly cancels the same factor in the denominator. However, a resonance occurs *near* $\delta = \Delta$ when the term containing Ω^2 in Eq. (6.6.21) is not ignored. $\chi^{(5)}$ is the lowest-order contribution to this resonance.

In the general case in which Ω^2 is not small, the full form of Eq. (6.6.21) must be used. In order to determine its resonance structure, we write $D(\delta)$ in terms of its real and imaginary parts as

$$\begin{aligned} D(\delta) = \delta&\left(\delta^2 - \Omega'^2 - \frac{1}{T_2^2} - \frac{2}{T_1 T_2}\right) \\ &+ i\left(\frac{\delta^2 - \Delta^2}{T_1} + \frac{2\delta^2}{T_2} - \frac{\Omega^2}{T_2} - \frac{1}{T_1 T_2^2}\right), \end{aligned} \tag{6.6.25}$$

where we have introduced the detuned Rabi frequency $\Omega' = (\Omega^2 + \Delta^2)^{1/2}$. We see by inspection that the real part of D vanishes for

$$\delta = 0, \quad \delta = \pm\left(\Omega'^2 + \frac{1}{T_2^2} + \frac{2}{T_1 T_2}\right)^{1/2}. \tag{6.6.26}$$

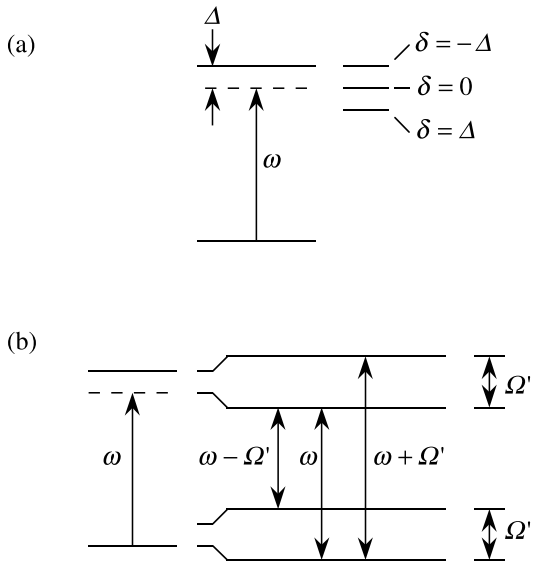

FIGURE 6.6.3: Resonances in the response of a two-level atom to pump and probe fields, as given by the function $D(\delta)$, (a) in the limit $\Omega^2 \to 0$, and (b) in the general case.

If we now assume that $\Omega' T_2$ is much greater than unity, these three resonances will be well separated, and we can describe their properties individually. In this limit, the function $D(\delta)$ becomes

$$D(\delta) = \delta\left(\delta^2 - \Omega'^2\right) + i\left(\frac{\delta^2 - \Delta^2}{T_1} + \frac{2\delta^2 - \Omega^2}{T_2}\right), \tag{6.6.27}$$

and the three resonances occur at

$$\delta = 0, \pm\Omega'. \tag{6.6.28}$$

Near the resonance at $\delta = 0$, and $D(\delta)$ can be approximated as

$$D(\delta) = -\Omega'^2(\delta + i\Gamma_0), \tag{6.6.29a}$$

where

$$\Gamma_0 = \frac{\Delta^2/T_1 + \Omega^2/T_2}{\Delta^2 + \Omega^2} \tag{6.6.29b}$$

represents the width of this resonance. Likewise, near the resonances at $\delta = \mp\Omega'$, $D(\delta)$ can be approximated as

$$D(\delta) = 2\Omega'^2\left[(\delta \pm \Omega') + i\Gamma_\pm\right], \tag{6.6.30a}$$

where

$$\Gamma_{\pm} = \frac{\Omega^2/T_1 + (2\Delta^2 + \Omega^2)/T_2}{2(\Omega^2 + \Delta^2)} \tag{6.6.30b}$$

represents the width of these resonances. Note that the positions of these resonances can be understood in terms of the energies of the dressed atomic states, as illustrated in Fig. 6.6.3(b). Note also that, for the case of weak optical excitation (i.e., for $\Omega^2 \ll \Delta^2$), Γ_0 approaches the population decay rate $1/T_1$, and $\Gamma_{\pm}$ approach the dipole dephasing rate $1/T_2$. In the limit of strong optical excitation (i.e., for $\Omega^2 \gg \Delta^2$), Γ_0 approaches the limit $1/T_2$ and $\Gamma_{\pm}$ approach the limit $\frac{1}{2}(1/T_1 + 1/T_2)$.

We next calculate the response of the atomic dipole at the sideband frequencies $\pm\delta$. We introduce the expression (6.6.20) for w_1 into Eq. (6.6.11) for p_1 and obtain

$$p_1 = \frac{\hbar^{-1}|\mu_{ba}|^2 w_0 E_1}{\Delta + \delta + i/T_2} \times \left[1 - \frac{\frac{1}{2}\Omega^2(\delta - \Delta + i/T_2)(\delta + 2i/T_2)/(\Delta - i/T_2)}{(\delta + i/T_1)(\delta - \Delta + i/T_2)(\Delta + \delta + i/T_2) - \Omega^2(\delta + i/T_2)}\right]. \tag{6.6.31}$$

Written in this form, we see that the response at the probe frequency $\omega + \delta$ can be considered to be the sum of two contributions. The first is the result of the zero-frequency part of the population difference w. The second is the result of population oscillations. The first term is resonant only at $\delta = -\Delta$, whereas the second term contains the additional resonances associated with the function $D(\delta)$. Sargent (1978) has pointed out that the second term obeys the relation

$$\int_{-\infty}^{\infty} \frac{-\frac{1}{2}\Omega^2(\delta - \Delta + i/T_2)(\delta + 2i/T_2)(\Delta - i/T_2)^{-1}}{(\delta + i/T_1)(\delta - \Delta + i/T_2)(\Delta + \delta + i/T_2) - \Omega^2(\delta + i/T_2)}\, d\delta = 0. \tag{6.6.32}$$

Thus, the second term, which results from population oscillations, does not modify the integrated absorption of the atom in the presence of a pump field; it simply leads to a spectral redistribution of probe-wave absorption.

A certain simplification of Eq. (6.6.31) can be obtained by combining the two terms algebraically so that p_1 can be expressed as

$$p_1 = \frac{\hbar^{-1}|\mu_{ba}|^2 w_0 E_1}{D(\delta)}\left[\left(\delta + \frac{i}{T_1}\right)\left(\delta - \Delta + \frac{i}{T_2}\right) - \frac{1}{2}\Omega^2 \frac{\delta}{\Delta - i/T_2}\right]. \tag{6.6.33}$$

Finally, we calculate the response at the sideband opposite to the applied probe wave through use of Eqs. (6.6.12) and (6.6.22) and the fact that $w_{-1} = w_1^*$, as noted in the discussion following Eq. (6.6.18). We obtain the result

$$p_{-1} = \frac{2w_0|\mu_{ba}|^4 E_0^2 E_1^*(\delta - \Delta - i/T_2)(-\delta + 2i/T_2)(\Delta + i/T_2)^{-1}}{\hbar^3(\Delta - \delta + i/T_2)D^*(\delta)}. \tag{6.6.34}$$

6.6.2 Nonlinear Susceptibility and Coupled-Amplitude Equations

Let us now use these results to determine the forms of the nonlinear polarization and the nonlinear susceptibility. Since p_1 is the complex amplitude of the dipole moment at frequency $\omega+\delta$ induced by a probe wave at this frequency, the polarization at this frequency is $P(\omega+\delta)=Np_1$. If we set $P(\omega+\delta)$ equal to $\epsilon_0\chi_{\text{eff}}^{(1)}(\omega+\delta)E_1$, we find that $\chi_{\text{eff}}^{(1)}(\omega+\delta)=Np_1/\epsilon_0E_1$, or through use of Eq. (6.6.33) that

$$\chi_{\text{eff}}^{(1)}(\omega+\delta)=\frac{N|\mu_{ba}|^2w_0}{\epsilon_0\hbar D(\delta)}\left[\left(\delta+\frac{i}{T_1}\right)\left(\delta-\Delta+\frac{i}{T_2}\right)-\tfrac{1}{2}\Omega^2\frac{\delta}{\Delta-i/T_2}\right]. \tag{6.6.35}$$

We have called this quantity an effective linear susceptibility because it depends on the intensity of the pump wave. Similarly, the part of the nonlinear polarization oscillating at frequency $\omega-\delta$ is given by $P(\omega-\delta)=Np_{-1}$. If we set this quantity equal to $3\epsilon_0\chi_{\text{eff}}^{(3)}[\omega-\delta=\omega+\omega-(\omega+\delta)]E_0^2E_1^*$, we find through use of Eq. (6.6.34) that

$$\begin{aligned}&\chi_{\text{eff}}^{(3)}\big[\omega-\delta=\omega+\omega-(\omega+\delta)\big]\\&\quad=\frac{2Nw_0|\mu_{ba}|^4(\delta-\Delta-i/T_2)(-\delta+2i/T_2)(\Delta+i/T_2)^{-1}}{3\epsilon_0\hbar^3(\Delta-\delta+i/T_2)D^*(\delta)}.\end{aligned} \tag{6.6.36}$$

We have called this quantity an effective third-order susceptibility, because it too depends on the laser intensity.

The calculation just presented has assumed that E_1 (the field at frequency $\omega+\delta$) is the only weak wave that is present. However, for the geometry of Fig. 6.6.2, a weak wave at frequency $\omega-\delta$ is generated by the interaction, and the response of the medium to this wave must also be taken into consideration. If we let E_{-1} denote the complex amplitude of this new wave, we find that we can represent the total response of the medium through the equations

$$P(\omega+\delta)=\epsilon_0\chi_{\text{eff}}^{(1)}(\omega+\delta)E_1+3\epsilon_0\chi_{\text{eff}}^{(3)}\big[\omega+\delta=\omega+\omega-(\omega-\delta)\big]E_0^2E_{-1}^*, \tag{6.6.37a}$$

$$P(\omega-\delta)=\epsilon_0\chi_{\text{eff}}^{(1)}(\omega-\delta)E_{-1}+3\epsilon_0\chi_{\text{eff}}^{(3)}\big[\omega-\delta=\omega+\omega-(\omega+\delta)\big]E_0^2E_1^*. \tag{6.6.37b}$$

Formulas for the new quantities $\chi_{\text{eff}}^{(1)}(\omega-\delta)$ and $\chi_{\text{eff}}^{(3)}[\omega+\delta=\omega+\omega-(\omega-\delta)]$ can be obtained by formally replacing δ by $-\delta$ in Eqs. (6.6.35) and (6.6.36).

The nonlinear response of the medium as described by Eqs. (6.6.37) will of course influence the propagation of the weak waves at frequencies $\omega\pm\delta$. We can describe the propagation of these waves by means of coupled-amplitude equations that we derive using methods described in Chapter 2. We introduce the slowly varying amplitudes $A_{\pm1}$ of the weak waves by means of the equation

$$E_{\pm1}=A_{\pm1}e^{ik_{\pm1}z}, \tag{6.6.38a}$$

where the propagation constant is given by

$$k_{\pm 1} = n_{\pm 1}(\omega \pm \delta)/c. \tag{6.6.38b}$$

Here $n_{\pm 1}$ is the real part of the refraction index experienced by each of the sideband frequencies and is given by

$$n_{\pm 1}^2 = 1 + \operatorname{Re} \chi_{\text{eff}}^{(1)}(\omega \pm \delta). \tag{6.6.38c}$$

We now introduce the nonlinear polarization of Eqs. (6.6.37) and the field decomposition of Eq. (6.6.38a) into the wave equation in the form of Eq. (2.1.22), and assume the validity of the slowly varying amplitude approximation. We find that the slowly varying amplitudes must obey the set of coupled equations

$$\frac{dA_1}{dz} = -\alpha_1 A_1 + \kappa_1 A_{-1}^* e^{i\Delta kz}, \tag{6.6.39a}$$

$$\frac{dA_{-1}}{dz} = -\alpha_{-1} A_{-1} + \kappa_{-1} A_1^* e^{i\Delta kz}, \tag{6.6.39b}$$

where we have introduced the nonlinear absorption coefficients

$$\alpha_{\pm 1} = -\frac{1}{2}\left(\frac{\omega \pm \delta}{n_{\pm 1} c}\right) \operatorname{Im} \chi_{\text{eff}}^{(1)}(\omega \pm \delta), \tag{6.6.40a}$$

the nonlinear coupling coefficients

$$\kappa_{\pm 1} = -i\frac{3}{2}\left(\frac{\omega \pm \delta}{n_{\pm 1} c}\right) \chi_{\text{eff}}^{(3)}\big[\omega \pm \delta = \omega + \omega - (\omega \mp \delta)\big] A_0^2, \tag{6.6.40b}$$

and the wavevector mismatch

$$\Delta k = 2k_0 - k_1 - k_{-1}, \tag{6.6.40c}$$

where k_0 is the magnitude of the wavevector of the pump wave.* The coupled wave equations given by Eqs. (6.6.37) can be solved explicitly for arbitrary boundary conditions. We shall not present the solution here; it is formally equivalent to the solution presented in Chapter 10 to the equations describing Stokes–anti-Stokes coupling in stimulated Raman scattering. The nature of the solution to Eqs. (6.6.37) for the case of a two-level atomic system has been described in detail by Boyd et al. (1981). These authors find that significant amplification of the A_1 and

* We have arbitrarily placed the real part of $\chi_{\text{eff}}^{(1)}$ into $n_{\pm 1}$ and the imaginary part into $\alpha_{\pm 1}$. We could equivalently have placed all of $\chi_{\text{eff}}^{(1)}$ in a complex absorption coefficient $\alpha_{\pm 1}$ and set Δk equal to zero, or could have placed all of $\chi_{\text{eff}}^{(1)}$ in a complex refraction index $n_{\pm 1}$ and set $\alpha_{\pm 1}$ equal to zero. We have chosen the present convention because it illustrates most clearly the separate effects of absorption and of wavevector mismatch.

A_{-1} waves can occur in the near-forward direction as a consequence of the four-wave mixing processes described by Eqs. (6.6.37). They also find that the gain is particularly large when the detuning δ (or its negative $-\delta$) is approximately equal to the generalized Rabi frequency Ω'. These effects have been studied experimentally by Harter et al. (1981).

Let us consider the nature of the solutions of Eqs. (6.6.37) for the special case of the geometry shown in Fig. 6.6.1. For this geometry, because of the large angle θ between the pump and probe beams, the magnitude Δk of the wavevector mismatch is very large, and as a result the coupled-amplitude equations (6.6.39a) and (6.6.39b) decouple into the two equations

$$\frac{dA_1}{dz} = -\alpha_1 A_1, \qquad \frac{dA_{-1}}{dz} = -\alpha_{-1} A_{-1}. \tag{6.6.41}$$

Recall that $\alpha_{\pm 1}$ denotes the absorption coefficient experienced by the probe wave at frequency $\omega \pm \delta$, and that $\alpha_{\pm 1}$ depends on the probe–pump detuning δ, on the detuning Δ of the pump wave from the atomic resonance, and on the intensity I of the pump wave.

The dependence of α_1 on the probe–pump detuning δ is illustrated for one representative case in part (a) of Fig. 6.6.4. We see that three features appear in the probe absorption spectrum. One of these features is centered on the laser frequency, and the other two occur at the *Rabi sidebands* of the laser frequency, that is, they occur at frequencies detuned from the laser frequency by the generalized Rabi frequency $\Omega' = (\Omega^2 + \Delta^2)^{1/2}$ associated with the driven atomic response. Note that α_1 can become negative for two of these features; the gain associated with these features was predicted by Mollow (1972) and has been observed experimentally by Wu et al. (1977) and by Gruneisen et al. (1988, 1989). The gain feature that occurs near $\delta = 0$ can be considered to be a form of stimulated Rayleigh scattering (see also Chapter 9). Alternatively, the origin of this feature can be traced to the harmonic temporal modulation of the population difference $w(t)$, as described by Eqs. (6.6.8) and (6.6.22)—that is, to coherent population oscillations. The gain associated with these features has been utilized to construct optical parametric oscillators (Grandclement et al., 1987). As demanded by Kramers–Kronig relations, there is a rapid spectral variation of the refractive index associated with this gain feature. This rapid frequency variation can lead to a significant modification of the group velocity v_g at which light pulses propagate through such a system in accordance with the standard relation $v_g = c/n_g$, where the group index n_g is given by $n_g = n + \omega(dn/d\omega)$. Physical situations occur for which $n_g \gg 1$; the term "slow light" is sometimes used to describe this situation. Analogously, the situation n_g positive with $n_g \ll 1$ corresponds to "fast light." Most intriguingly, physical situations can occur for which n_g is negative, which corresponds to "backward" light propagation. More detailed accounts of these possibilities have been presented by Bigelow et al. (2003a, 2003b) and Gehring et al. (2006).

Part (b) of Fig. 6.6.4 shows the origin of each of the features shown in part (a). The leftmost portion of this figure shows how the dressed states of the atom are related to the unperturbed

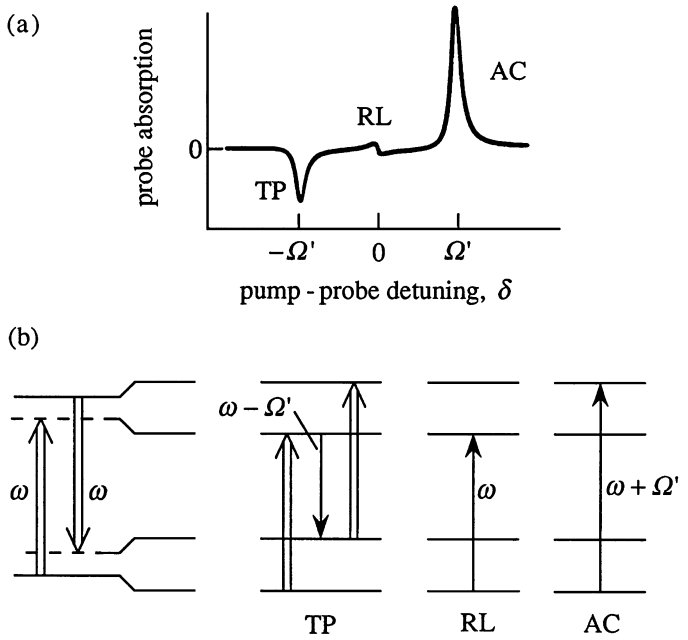

FIGURE 6.6.4: (a) Absorption spectrum of a probe wave in the presence of a strong pump wave for the case $\Delta T_2 = -3$, $\Omega T_2 = 8$, and $T_2/T_1 = 2$. (b) Each of the features in the spectrum shown in part (a) is identified by the corresponding transition between dressed states of the atom. TP denotes the three-photon resonance, RL denotes the Rayleigh resonance, and AC denotes the ac-Stark-shifted atomic resonance.

atomic energy states. The next diagram, labeled TP, shows the origin of the three-photon resonance. Here the atom makes a transition from the lowest dressed level to the highest dressed level by the simultaneous absorption of two pump photons and the emission of a photon at the Rabi sideband frequency $\omega - \Omega'$. This process can amplify a wave at the Rabi sideband frequency, as indicated by the region of negative absorption labeled TP in part (a). The third diagram of part (b), labeled RL, shows the origin of the stimulated Rayleigh resonance. The Rayleigh resonance corresponds to a transition from the lower level of the lower doublet to the upper doublet. Each of these transitions is centered on the frequency of the pump laser. The final diagram of part (b) of the figure, labeled AC, corresponds to the usual absorptive resonance of the atom as modified by the ac-Stark effect. For the sign of the detuning used in the diagram, the atomic absorption is shifted to higher frequencies. Note that this last feature can lead only to absorption, whereas the first two features can lead to amplification. The theory of optical wave mixing has been generalized by Agarwal and Boyd (1988) to treat the quantum nature of the optical field; this theory shows how quantum fluctuations can initiate the four-wave mixing process described in this section.

Problems

1. *Alternative relaxation models.* Determine how the saturated absorption of an atomic transition depends on the intensity of the incident (monochromatic) laser field for the case of an open two-level atom and for a two-level atom with a non-radiatively coupled intermediate level, and compare these results with those derived in Section 6.3 for a closed two-level atom.
2. $\chi^{(3)}$ *for an impurity-doped solid.* One is often interested in determining the third-order susceptibility of a collection of two-level atoms contained in a medium of constant (i.e., wavelength-independent and non-intensity-dependent) refractive index n_0. Show that the third-order susceptibility of such a system is given by Eq. (6.3.36b) in the form shown, or by Eq. (6.3.33b) with a factor of n_0 introduced in the numerator, or by Eq. (6.3.34a) or (6.3.34b) with a factor of n_0^2 introduced in the numerator. In cases in which I_s^0, I_s^Δ, or $\alpha_0(\Delta)$ appears in the expression, it is to be understood that the expressions (6.3.30) and (6.3.31) for I_s^0 and I_s^Δ should each be multiplied by a factor of n_0 and the expression (6.3.22b) for $\alpha_0(0)$ should be divided by a factor of n_0.
3. *Orthogonality of dressed states.* Verify Eq. (6.5.43).
4. *Damping of Rabi oscillations.* The intent of this problem is to determine the influence of T_1- and T_2-type relaxation processes on Rabi oscillations of the sort predicted by the solution to the Schrödinger equation for an atom in the presence of an intense, near-resonant driving field. In particular, you are to solve the Bloch equation in the form of Eqs. (6.5.49) for the time evolution of an atom known to be in the ground state at time $t = 0$ and subject to a field $Ee^{-i\omega t} + \text{c.c.}$ that is turned on at time $t = 0$. In addition, sketch the behavior of w and of p as functions of time.

   [Hint: At a certain point in the calculation, the mathematical complexity will be markedly reduced by assuming that $T_1 = T_2$. Make this simplification only when it becomes necessary.]
5. *Response times.* Consider the question of estimating the response time of nonresonant electronic nonlinearities of the sort described in Section 4.3. Student A argues that it is well known that the response time under such conditions is of the order of the reciprocal of the detuning of the laser field from the nearest atomic resonance. Student B argues that only relaxation processes can allow a system to enter the steady state and that consequently the response time is of the order of the longer of T_1 and T_2—that is, is of the order of T_1. Who is right, and in what sense is each of them correct?

   [Hint: Consider how the graph shown in Fig. 6.5.6 and the analogous graph of $p(t)$ would look in the limit of $\Delta \gg \Omega$, $\Delta T_2 \gg 1$.]

   [Partial answer: The nonlinearity turns on in a time Δ^{-1} but does not reach its steady-state value until a time of the order of T_1.]

6. *Identity pertaining to population oscillations.* Verify Eq. (6.6.32).
7. *Coupled wave equations for forward four-wave mixing.* Verify that Eqs. (6.6.37) follow from the referenced equations. Also, display explicitly the two alternative forms of these equations alluded to in the footnote to these equations.

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