Chapter 13

Three-State Atoms

13.1 The Three-State Rotating-Wave Approximation

approximation: it gives the equation

$$\frac{\mathrm{d}}{\mathrm{d}t}C_1 = -i\Delta_1 C_1 - \frac{i}{2}\Omega_1^* C_2 \tag{13.1-6}$$

where the coefficients are

$$\hbar\Delta_1 = E_1 - \hbar\dot{\zeta_1}\hbar\Omega_1^* \quad = -\mathrm{d}_{12}\cdot\epsilon_1^*\mathcal{E}_1^* \qquad or \qquad \hbar\Omega_1 = \ -\mathrm{d}_{21}\cdot\epsilon_1\mathcal{E}_1. \tag{13.1-7a}$$

The appearance in this equation of Ω_1^*, rather than Ω_1, follows the convention of pairing Ω with the positive-frequency amplitude $\mathcal{E}$ and pairing Ω^* with $\mathcal{E}^*$.

Phase choice (13.1-4) is suitable only when $E_1 < E_2$. If, instead, state 2 has the lower energy of this pair, then we must choose

$$\dot{\zeta_2} = \dot{\zeta_1} - \omega_1, \tag{13.1-8}$$

from which we obtain the equation

$$\frac{\mathrm{d}}{\mathrm{d}t}C_1 = -i\Delta_1 C_1 - \frac{i}{2}\Omega_1' C_2. \tag{13.1-9}$$

The Rabi frequency Ω_1' that appears here differs from that of Eq.n (13.1-7a) by complex conjugation of the field [cf. Eqn. (10.1-22)]:

$$\hbar\Omega' = -\mathrm{d}_{12}\cdot\epsilon_1\mathcal{E}_1. \tag{13.1-10}$$

We may note in passing that, just as in the two-state RWA, we are at liberty to add to ζ_2 a time-independent constant such that Ω_1 is real and non-negative at some instant of time. This additional phase does not affect the coefficients nor does it alter the RWA. If the phase of $\mathcal{E}_1$ remains constant, Ω_1 remains real.

However, when we allow phase variations (as we must in treating propagation or fluctuations) then we cannot necessarily assume the Rabi frequencies remain real at all times.

The Second Equation. The second equation, for the time derivative of C_2, has on its right hand side four exponential terms as coefficients of C_1 and a second four as coefficients of C_3. The arguments of the C_1 coefficients are those considered above; the C_3 arguments are

$$\begin{aligned} &(\zeta_3 - \zeta_2 + \omega_1 t), && (\zeta_3 - \zeta_2 + \omega_2 t), \\ &(\zeta_3 - \zeta_2 - \omega_1 t), && (\zeta_3 - \zeta_2 - \omega_2 t). \end{aligned}$$

Again a suitable choice of phases can eliminate time variation from one exponential, leaving terms that oscillate rapidly and so average to zero in the RWA. The choice depends upon whether E_3 lies above or below E_3. If $E_2 < E_3$ then we choose

$$\dot{\zeta_3} = \dot{\zeta_2} + \omega_2 \tag{13.1-11}$$

so that the exponential arguments become

$$(\omega_2 + \omega_1)t, \qquad (\omega_2 - \omega_1)t, \qquad 2\omega_2 t, \qquad 0.$$

Upon replacing the exponentials with their time averages (zero or unity) we obtain the second-step RWA equation (under the assumption $E_1 < E_2$)

$$\frac{\mathrm{d}}{\mathrm{d}t}C_2 = \; -i\Delta_2 C_2 - \frac{i}{2}[\Omega_1 C_1 + \Omega_2^* C_3] \tag{13.1-12}$$

where the coefficients are

$$\hbar\Delta_2 = E_2 - E_1 - \hbar\omega_1 + \hbar\Delta_1 \tag{13.1-13a}$$

$$\hbar\Omega_2^* = -\mathrm{d}_{23}\cdot\epsilon_2^*\mathcal{E}_2^* \qquad or \qquad \hbar\Omega_2 = -\mathrm{d}_{32}\cdot\epsilon_2\mathcal{E}_2. \tag{13.1-13b}$$

This expression for Δ_2 applies when $E_1 < E_2$. When the energy ranking is instead $E_1 > E_2$ the diagonal element appears as

$$\hbar\Delta_2 = \hbar\omega_1 - (E_1 - E_2) + \hbar\Delta_1 \tag{13.1-14a}$$

and the coefficient of C_1 becomes $(\Omega_1{}')^*$. If $E_3 < E_2$ then the proper choice of phases is

$$\dot{\zeta_3} = \dot{\zeta_2} - \omega_2 \tag{13.1-14b}$$

with corresponding modification of the formula for Δ_3 and Ω_2; in place of Eq.n (13.1-13b) we have the formula

$$\hbar\Omega_2' = -\mathrm{d}_{23}\cdot\epsilon_2\mathcal{E}_2. \tag{13.1-15}$$

The Third Equation. The foregoing phase choices produce, without further phase assignment, the third and final RWA equation. Along with Eq.n (**??**) we obtain the equation

$$\frac{\mathrm{d}}{\mathrm{d}t}C_3 = -i\Delta_3 C_3 + \frac{i}{2}\Omega_2 C_2. \tag{13.1-16}$$

Here, with the ordering $E_3 > E_2 > E_1$, the diagonal element is

$$\hbar\Delta_3 = E_3 - \hbar\dot{\zeta_3} = E_3 - E_1 - \hbar\omega_1 - \hbar\omega_2 + \hbar\Delta_1. \tag{13.1-17}$$

As with the first-step equation, a suitable constant term added to the phase ζ_3 can make Ω_2 real and nonnegative at time $t = 0$.

The RWA Equations. These phase choices, together with the neglect of exponentials having arguments

$$2\omega_1 t, \qquad 2\omega_2 t, \qquad (\omega_1 \pm \omega_2)t,$$

produce from the Schrödinger equation a set of three coupled equations

$$\frac{\mathrm{d}}{\mathrm{d}t}C_n(t) = -i\sum_m W_{nm}C_m(t) \qquad \text{or} \qquad \frac{\mathrm{d}}{\mathrm{d}t}C(t) = -iWC(t). \tag{13.1-18}$$

We can *supplement the detunings and Rabi frequencies that provide the elements of the matrix W with non-Hermitian diagonal elements to model probability loss.* When the energy ordering is $E_1 < E_2 < E_3$ the 3×3 RWA Hamiltonian matrix W is

$$[W] = \frac{1}{2}\begin{bmatrix} 2\Delta_1 - i\Gamma_1 & \Omega_1^* & 0 \\ \Omega_1 & 2\Delta_2 - i\Gamma_2 & \Omega_2^* \\ 0 & \Omega_2 & 2\Delta_3 - i\Gamma_3 \end{bmatrix}. \tag{13.1-19}$$

The quantity Γ_n is the *probability loss rate* from level n. Other orderings of the energy values alter the patterns of Ω and Ω^* asdiscussed above. With any ordering the validity of these RWA equations requires that exponentials of frequencies

$$2\omega_1, \qquad 2\omega_2, \qquad \omega_1 + \omega_2, \qquad \omega_1 - \omega_2$$

must oscillate many times during the progress of excitation. As with the two-state atom, the speed with which probability amplitudes change is governed by the eigenvalues of W; the largest eigenvalue governs the most rapid response. In turn, the eigenvalues of W cannot greatly exceed the size of the elements of W. Thus we *require*, **for the RWA, that all of the detunings Δ_n and Rabi frequencies Ω_n must be smaller in magnitude than the several sum and difference frequencies listed above:**

$$\max\{|\Delta_n|, |\Omega_n|\} \ll \omega_p \pm \omega_q \qquad \text{for } p, q = 1, 2. \tag{13.1-20}$$

The most stringent of these conditions is obviously posed by the difference frequency $\omega_2 - \omega_1$ being much larger than the detunings and Rabi frequencies.

For this condition to hold the two lasers must differ in frequency by many Rabi frequencies, so that each laser can be identified unambiguously as near-resonant with a particular transition. If the laser frequencies differ by less than this amount then confusion may occur and the RWA fails.

Three-State Configurations. The three-state RWA equations above describe excitation in the linkage pattern $1 \leftrightarrow 2 \leftrightarrow 3$ for any ranking of the relative energy values. When we incorporate probability loss by augmenting the diagonal elements of W with negative imaginary terms $-i\gamma_n$ three cases have particular significance.

First and simplest is the *ladder configuration* in which the energies increases with index $n, E_1 < E_2 < E_3$, population initially resides in state 1, and loss occurs only from the uppermost state 3. Figure 13.1-1a illustrates this configuration. The convenient choice $\Delta_1 = 0$ *is equivalent to setting the lowest energy* E_1 *to zero*.

A second possibility, the *lambda configuration*, occurs when the central energy of the sequence exceed either end-point energy, probability loss occurs again from the uppermost state, and initial population again resides in the lowest state. 13.1b illustrates this configuration. Again the convenient choice $\Delta_1 = 0$ fixeds the energy zero point. The detuning Δ_3 now involves differences between photon energies. More significantly (as we shall see in section §13.7), loss occurs from the middle state of the sequence.

A third possibility the *vee configuration*, occurs when the central state lies lowest in energy. 13.1c illustrates this case. Here the convetional choice $\Delta_2 = 0$ fixes E_2 as the zero point of energy.

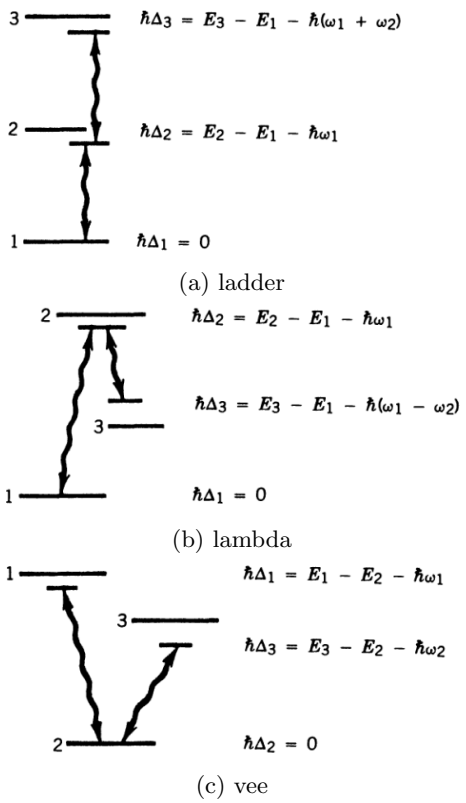

Figure 13.1: Schematic diagram of three-state configurations

We shall see that each of these configurations gives rise to characteristic behavior of population flow. It should be apeciated that the discussion, being based upon the Schrödinger equation, refers to coherent excitation, not to the conventional incoherent rate-equation regime. For example, the diagram o the linkage pattern for the lambda system is that of the *Raman scattering* process, in which a photon of one frequency is absorbed and a second frequency

is reemitted. This two-photon process is usually observed under steady-state conditions, corresponding to a limit best treated by density matrix methods or rate equations.

Summary of Rotating-Wave Phase Choices. The various choices of phases are summarized by the following relationships:

$$\dot{\zeta_1} = \begin{cases} \dot{\zeta_2} - \omega_1, \\ \dot{\zeta_2} + \omega_1, \end{cases} \qquad \hbar\Delta_1 \equiv E_1 - \hbar\dot{\zeta_1} = \begin{cases} \hbar\Delta_2 + E_1 - E_2 + \hbar\omega_1, & E_1 < E_2 \\ \hbar\Delta_2 + E_1 - E_2 - \hbar\omega_1, & E_1 > E_2 \end{cases}$$

$$\dot{\zeta_2} = \begin{cases} \dot{\zeta_1} - \omega_1, \\ \dot{\zeta_1} + \omega_1, \end{cases} \qquad \hbar\Delta_2 \equiv E_2 - \hbar\dot{\zeta_2} = \begin{cases} \hbar\Delta_1 + E_2 - E_1 - \hbar\omega_1, & E_1 < E_2 \\ \hbar\Delta_1 + E_2 - E_1 + \hbar\omega_1, & E_1 > E_2 \end{cases}$$

$$\dot{\zeta_3} = \begin{cases} \dot{\zeta_2} + \omega_2, \\ \dot{\zeta_2} - \omega_2, \end{cases} \qquad \hbar\Delta_3 \equiv E_3 - \hbar\dot{\zeta_3} = \begin{cases} \hbar\Delta_2 + E_3 - E_2 - \hbar\omega_2, & E_2 < E_3 \\ \hbar\Delta_2 + E_3 - E_2 - \hbar\omega_2, & E_2 > E_3 \end{cases}$$

For the ladder system, with $E_1 < E_2 < E_3$ and the choice $\Delta_1 = 0$ the phases are

$$\begin{aligned} \dot{\zeta_2} &= \dot{\zeta_1} + \omega_1, & \hbar\Delta_2 &= E_2 - E_1 - \hbar\omega_1, \\ \dot{\zeta_3} &= \dot{\zeta_1} + \omega_2, & \hbar\Delta_3 &= E_3 - E_1 - \hbar\omega_2 \end{aligned}$$

For the lambda system, with $E_1 < E_2, E_3 < E_2$ and the choice $\Delta_2 = 0$ the phase are

$$\begin{aligned} \dot{\zeta_1} &= \dot{\zeta_2} - \omega_1, & \hbar\Delta_1 &= -(E_2 - E_1) + \hbar\omega_1, \\ \dot{\zeta_3} &= \dot{\zeta_2} - \omega_2, & \hbar\Delta_3 &= -(E_2 - E_3) + \hbar\omega_2. \end{aligned}$$

For the vee system, with $E_1 > E_2, \quad E_3 > E_2$ and the choice $\Delta_2 = 0$ the phases are

$$\begin{aligned} \dot{\zeta_1} &= \dot{\zeta_2} + \omega_1, & \hbar\Delta_1 &= E_1 - E_2 - \hbar\omega_1, \\ \dot{\zeta_3} &= \dot{\zeta_2} + \omega_2, & \hbar\Delta_3 &= E_3 - E_2 - \hbar\omega_2. \end{aligned}$$

Closed Loop Configuration. I shall be discussing simple RWA systems involving two transitions amongst three states. It is also possible to consider three-state transitions combined with *three* near-resonant excitation frequencies, in a linkage pattern that forms a closed loop. For a free atom (as distinguished from an atom embedded in a solid or other external field) at least one of the transitions of such a loop must be a forbidden transition, produced by magnetic dipole or electric quadrupole transition. This arrangement offers interesting possibilities for observation of the relative phase of the radiation, as discussed by Buckle et al. (1986).