

**V. PHOTON ABSORPTION AND EMISSION**

**"POOR MAN'S" SECOND QUANTIZATION OF MATERIAL SYSTEM:**

In treating the complete quantum mechanical problem, it is useful to recast the material (atomic) Hamiltonian in terms of an appropriate set of *creation* and *destruction* operators. To that end we make the following definition

$$\mathcal{H}_A |x\rangle = \hbar \omega_x |x\rangle \tag{ V-1 }$$

Using the ubiquitous identity operation  $\int_x |x\rangle\langle x| = 1$ , we may write the material Hamiltonian in *second quantized* form -- viz.

$$\begin{aligned} \mathcal{H}_A &= \int_x |x\rangle\langle x| \mathcal{H}_A \int_y |y\rangle\langle y| = \int_x \int_y |x\rangle \hbar \omega_x \langle x|y\rangle\langle y| \\ &= \int_x \hbar \omega_x |x\rangle\langle x| \end{aligned} \tag{ V-2 }$$

In general, the operator  $\int_x |x\rangle\langle y| b_x^\dagger b_y$  applied to any state  $|z\rangle$  yields

$$\int_x |x\rangle\langle y| b_x^\dagger b_y |z\rangle = |x\rangle \delta_{yz} \tag{ V-3 }$$

-- *i.e.* the operator changes a state **z** to a state **x** if the state is **y** otherwise it produces zero. In other words, the operator **destroys** the state **y** and **creates** a state **x**. The second quantization viewpoint is particularly useful in treating the interaction of a two-level material system with the radiation field. This case, is most conveniently formulate in two-vector notation with the use of *Pauli spin matrices* -- viz.

$$|a\rangle = \begin{pmatrix} 1 \\ 0 \end{pmatrix} \quad \text{and} \quad |b\rangle = \begin{pmatrix} 0 \\ 1 \end{pmatrix} \tag{ V-4a }$$

$$\langle a| = [1 \quad 0] \quad \text{and} \quad \langle b| = [0 \quad 1] \tag{ V-4b }$$

$$|a\rangle\langle b| = \frac{1}{0} \begin{bmatrix} 0 & 1 \\ 0 & 0 \end{bmatrix} = \frac{0}{0} \frac{1}{0} = + \quad [ V-4c ]$$

$$|b\rangle\langle a| = \frac{0}{1} \begin{bmatrix} 1 & 0 \\ 0 & 0 \end{bmatrix} = \frac{0}{1} \frac{0}{0} = -$$

$$|a\rangle\langle a| = \frac{1}{0} \begin{bmatrix} 1 & 0 \\ 0 & 0 \end{bmatrix} = \frac{1}{0} \frac{0}{0} \quad [ V-4d ]$$

$$|b\rangle\langle b| = \frac{0}{1} \begin{bmatrix} 0 & 1 \\ 0 & 1 \end{bmatrix} = \frac{0}{0} \frac{0}{1}$$

Consequently, the atomic Hamiltonian may be written

$$\mathcal{H}_A = \hbar \begin{matrix} 1 & 0 \\ a & 0 \\ 0 & 0 \end{matrix} + \hbar \begin{matrix} 0 & 0 \\ b & 0 \\ 0 & 1 \end{matrix} \quad [ V-5a ]$$

$$= \frac{1}{2} \hbar \begin{pmatrix} a & -b \\ 0 & -1 \end{pmatrix} + \frac{1}{2} \hbar \begin{pmatrix} a & +b \\ 0 & 1 \end{pmatrix}$$

and if we neglect the mean energy of the states

$$\mathcal{H}_A = \frac{1}{2} \hbar \begin{matrix} 1 & 0 \\ ab & 0 \\ 0 & -1 \end{matrix} = \frac{1}{2} \hbar \begin{matrix} ab & z \\ ab & \end{matrix} \quad [ V-5b ]$$

and the electric dipole interaction Hamiltonian becomes

$$\begin{aligned}
 \mathcal{H}_{ED} &= e \vec{\mathcal{D}} \cdot \vec{\mathbf{E}}_T(0) \\
 &= [ |a\rangle\langle a| + |b\rangle\langle b| ] [ e \vec{\mathcal{D}} ] [ |a\rangle\langle a| + |b\rangle\langle b| ] \vec{\mathbf{E}}_T(0) \\
 &= \left\{ |a\rangle\langle a| [ e \vec{\mathcal{D}} ] |b\rangle\langle b| + |b\rangle\langle b| [ e \vec{\mathcal{D}} ] |a\rangle\langle a| \right\} \vec{\mathbf{E}}_T(0) \\
 &= \left\{ e \langle a | \vec{\mathcal{D}} | b \rangle + e \langle b | \vec{\mathcal{D}} | a \rangle \right\} \vec{\mathbf{E}}_T(0) \\
 &= - \left\{ \dots \right\} \vec{\mathbf{E}}_T(0)
 \end{aligned}
 \tag{V-6}$$

From Equation [ II-24a ] in this lecture set we can write

$$\vec{\mathbf{E}}_T(0) = \sum_{\{l\}} \hat{\mathbf{e}}_{\{l\}} \mathcal{E}_{\{l\}} i a_{\{l\}}(t) \exp[i \vec{\mathbf{k}}_{\{l\}} \cdot \vec{\mathbf{r}}_A] - i a_{\{l\}}^\dagger(t) \exp[-i \vec{\mathbf{k}}_{\{l\}} \cdot \vec{\mathbf{r}}_A]
 \tag{V-7}$$

where  $\mathcal{E}_{\{l\}} = \sqrt{\frac{\hbar}{2 \epsilon_0 V}}$  is the so called the *electric field per photon* and  $\vec{\mathbf{r}}_A$  is the

location of the center of the atom under consideration. Thus Equation [ V-6 ] may be written quite generally for a two level atom as

$$\begin{aligned}
 \mathcal{H}_{ED} &= \left\{ e \langle a | \vec{\mathcal{D}} | b \rangle + e \langle b | \vec{\mathcal{D}} | a \rangle \right\} \\
 &\quad \times \sum_{\{l\}} \hat{\mathbf{e}}_{\{l\}} \mathcal{E}_{\{l\}} \left\{ i a_{\{l\}}(t) \exp[i \vec{\mathbf{k}}_{\{l\}} \cdot \vec{\mathbf{r}}_A] - i a_{\{l\}}^\dagger(t) \exp[-i \vec{\mathbf{k}}_{\{l\}} \cdot \vec{\mathbf{r}}_A] \right\} \\
 &= \hbar \sum_{\{l\}} \left\{ g_{\{l\}} + g_{\{l\}} \right\} \left\{ i a_{\{l\}}(t) \exp[i \vec{\mathbf{k}}_{\{l\}} \cdot \vec{\mathbf{r}}_A] + adj. \right\}
 \end{aligned}
 \tag{V-8}$$

where the coupling constant is given by

$$g_{\{\ell\}s} = \hbar^{-1} e \mathcal{E}_{\{\ell\}A} \hat{\mathbf{e}}_{\{\ell\}s} = e \sqrt{\frac{\{\ell\}}{2\hbar \epsilon_0}} \langle a | \vec{\mathcal{D}} | b \rangle \hat{\mathbf{e}}_{\{\ell\}s} \quad [ V-9 ]$$

**Interaction of a Two-level Atom and a Single Mode Field -- Rabi Flopping:**

Let us first consider the interaction of a two-level system with a single photon state to make contact with the discussion in Section III of the lecture set entitled *The Interaction of Radiation and Matter: Semiclassical Theory*. Equation [ V-8 ] then reduces to

$$\mathcal{H}_{ED} = i \hbar \left\{ g_{\mathbf{k}^+} + g_{\mathbf{k}^-} \right\} \left\{ a_{\mathbf{k}^-}(t) \exp[i \mathbf{k}^- \cdot \mathbf{r}_A] - a_{\mathbf{k}^+}^\dagger(t) \exp[-i \mathbf{k}^- \cdot \mathbf{r}_A] \right\} \quad [ V-10a ]$$

where  $g_{\mathbf{k}^-} = \hbar^{-1} e \mathcal{E}_{\mathbf{k}^-A} \hat{\mathbf{e}}_{\mathbf{k}^-} = e \sqrt{\frac{\mathbf{k}^-}{2\hbar \epsilon_0}} \langle a | \vec{\mathcal{D}} | b \rangle \hat{\mathbf{e}}_{\mathbf{k}^-}$ . If we neglect any inter-atomic interference effects by taking  $\mathbf{r}_A = 0$ , we may simplify Equation [ V-10a ] to

$$\begin{aligned} \mathcal{H}_{ED} &= \hbar \left\{ g_{\mathbf{k}^+} + g_{\mathbf{k}^-} \right\} \left\{ i a_{\mathbf{k}^-}(t) - i a_{\mathbf{k}^+}^\dagger(t) \right\} \\ &= \hbar \left\{ g_{\mathbf{k}^+} + g_{\mathbf{k}^-} \right\} \left\{ i a_{\mathbf{k}^-}(t) + adj. \right\} \end{aligned} \quad [ V-10b ]$$

Thus, we may then write the complete effective Hamiltonian of the composite system as

$$\mathcal{H} = \mathcal{H}_0 + \mathcal{H}_I = \frac{1}{2} \hbar \omega_{ab} \sigma_z + \hbar \omega_{\mathbf{k}^-} a^\dagger a + \hbar \left\{ g_{\mathbf{k}^+} + g_{\mathbf{k}^-} \right\} \left\{ i a_{\mathbf{k}^-}(t) - i a_{\mathbf{k}^+}^\dagger(t) \right\} \quad [ V-11a ]$$

In the **rotating wave approximation** this reduces to

$$\mathcal{H} = \mathcal{H}_0 + \mathcal{H}_I = \frac{1}{2} \hbar \omega_{ab} \sigma_z + \hbar \omega_{\mathbf{k}^-} a^\dagger a + \hbar \left\{ i g_{\mathbf{k}^+} a_{\mathbf{k}^+} - i g_{\mathbf{k}^-} a_{\mathbf{k}^-}^\dagger \right\} \quad [ V-11b ]$$

In the spirit of the discussion in the Section VII, *Semiconductor Photonics* in the lecture set entitled *The Interaction of Radiation and Matter: Semiclassical Theory*, this effective Hamiltonian may be adapted to provide a fully quantum mechanical treatment of optical interactions in semiconductors.<sup>26</sup>

**"DRESSED" ATOMIC STATES:**

We know that the unperturbed Hamiltonian satisfies the following eigenvalue equations

$$\begin{aligned} \mathcal{H}_0 |a n\rangle &= \hbar \left( \frac{1}{2} \omega_{ab} + n \omega_{\vec{k}} \right) |a n\rangle \\ \mathcal{H}_0 |b n\rangle &= \hbar \left( -\frac{1}{2} \omega_{ab} + n \omega_{\vec{k}} \right) |b n\rangle \end{aligned} \quad [ V-12 ]$$

-- where  $|a n\rangle = |a\rangle |n_{\vec{k}}\rangle$  and  $|b n\rangle = |b\rangle |n_{\vec{k}}\rangle$  -- and the electric dipole perturbation couples the states  $|a n\rangle$  and  $|b (n+1)\rangle$ . It is useful to resolve the complete

<sup>26</sup> In *Semiconductor Photonics* we noted that Chow, Koch and Sargent in their *Semiconductor-Laser Physics* (Springer-Verlag - 1994) treat semiconductor problems in terms of the following semiclassical Hamiltonian for an inhomogeneous two-level system:

$$\mathcal{H}_{\text{eff}} = \sum_{\vec{k}} \left[ \frac{\hbar^2 k^2}{2m_{ec}} a_{\vec{k}}^\dagger a_{\vec{k}} + \frac{\hbar^2 k^2}{2m_{hv}} b_{-\vec{k}}^\dagger b_{-\vec{k}} - \left[ \mu_{\vec{k}} a_{\vec{k}}^\dagger b_{-\vec{k}}^\dagger + \mu_{\vec{k}} a_{\vec{k}} b_{-\vec{k}} \right] E(z, t) \right]$$

where  $\{a_{\vec{k}}^\dagger, a_{\vec{k}}\}$  and  $\{b_{\vec{k}}^\dagger, b_{\vec{k}}\}$  are, respectively, electron and hole **{creation destruction}** operators.  $\mu_{\vec{k}}$  is the dipole matrix element between vertical states in the valence and conduction bands. In the fully quantal treatment the effective Hamiltonian becomes

$$\mathcal{H}_{\text{eff}} = \sum_{\vec{k}} \left[ \frac{\hbar^2 k^2}{2m_{ec}} a_{\vec{k}}^\dagger a_{\vec{k}} + \frac{\hbar^2 k^2}{2m_{hv}} b_{-\vec{k}}^\dagger b_{-\vec{k}} - \hbar \left[ g_{\vec{k}} a_{\vec{k}}^\dagger b_{-\vec{k}}^\dagger a + g_{\vec{k}} a^\dagger a_{\vec{k}} b_{-\vec{k}} \right] \right]$$

where  $g_{\vec{k}} = \sqrt{2} \mu_{\vec{k}} \mathcal{E}_r \sin(k_r z_0) / \hbar$

Hamiltonian into a sum of component Hamiltonians  $\mathcal{H} = \sum_n \mathcal{H}_n$  where the component  $\mathcal{H}_n$ 's act only within the  $\{|a, n\rangle, |b, (n+1)\rangle\}$  coupled manifold of states and can be written<sup>27</sup>

$$\mathcal{H}_n = \begin{pmatrix} \hbar(n + \frac{1}{2}) & 0 \\ 0 & \hbar(n + \frac{1}{2}) \end{pmatrix} + \begin{pmatrix} 0 & 2i g_{\mathbf{k}} a \\ -2i g_{\mathbf{k}} a^\dagger & 0 \end{pmatrix} \quad [ V-13 ]$$

where  $\mathcal{H}_n = \mathcal{H}_{ab} - \hbar \omega_{\mathbf{k}}$ . The second term in this equation is of the same form as the coupling matrix in Equation [ III-8c ] of the lecture set entitled *The Interaction of Radiation and Matter: Semiclassical Theory* where the semiclassical Rabi frequency  $\omega_{\mathbf{k}} = E_0/\hbar$  is replaced by its quantum equivalent -- viz.  $2i g_{\mathbf{k}} \sqrt{n+1}$ . Diagonalizing this matrix we find the eigenvalues of the so call **dressed atomic states** -- viz.<sup>28</sup>

<sup>27</sup> In particular, using the **identity operator** -- i.e.,  $I = \sum_n |n\rangle\langle n|$

$$\begin{aligned} \mathcal{H}_n &= \left\{ |a, n\rangle\langle a, n| + |b, (n+1)\rangle\langle b, (n+1)| \right\} \mathcal{H} \left\{ |a, n\rangle\langle a, n| + |b, (n+1)\rangle\langle b, (n+1)| \right\} \\ &= |a, n\rangle\langle a, n| \langle a, n| \mathcal{H} |a, n\rangle + |a, n\rangle\langle a, n| \langle a, n| \mathcal{H} |b, (n+1)\rangle \\ &\quad + |b, (n+1)\rangle\langle a, n| \langle b, (n+1)| \mathcal{H} |a, n\rangle + |b, (n+1)\rangle\langle b, (n+1)| \langle b, (n+1)| \mathcal{H} |b, (n+1)\rangle \end{aligned}$$

<sup>28</sup> It should be noted that the rotation matrix

$$= \begin{pmatrix} \cos \theta_n & -\sin \theta_n \\ \sin \theta_n & \cos \theta_n \end{pmatrix}$$

diagonalizes the Hamiltonian in Equation [ V-13 ] through the transformation  $\mathcal{H}_n = U_n^{-1} \mathcal{H}_n U_n$ . Further, relates the **dressed** and **bare** probability amplitudes as

$$\begin{aligned}
 E_{1n} &= \hbar \left( n + \frac{1}{2} \right) \omega_k + \frac{1}{2} \hbar \omega_n^R \\
 E_{2n} &= \hbar \left( n + \frac{1}{2} \right) \omega_k - \frac{1}{2} \hbar \omega_n^R
 \end{aligned}
 \tag{V-14}$$

(see energy level diagram below) where  $\omega_n^R = \sqrt{\omega_k^2 + 4|g_k|^2(n+1)}$  is the **quantized field generalization** of the **Rabi flopping frequency**.

The dressed eigenstates are given by

$$\begin{aligned}
 |1n\rangle &= \sin \theta_n |a n\rangle + \cos \theta_n |b (n+1)\rangle \\
 |2n\rangle &= \cos \theta_n |a n\rangle - \sin \theta_n |b (n+1)\rangle
 \end{aligned}
 \tag{V-15a}$$

where

$$\cos \theta_n = \frac{\omega_n^R - \omega_k}{\sqrt{(\omega_n^R - \omega_k)^2 + 4|g_k|^2(n+1)}}
 \tag{V-15b}$$

and

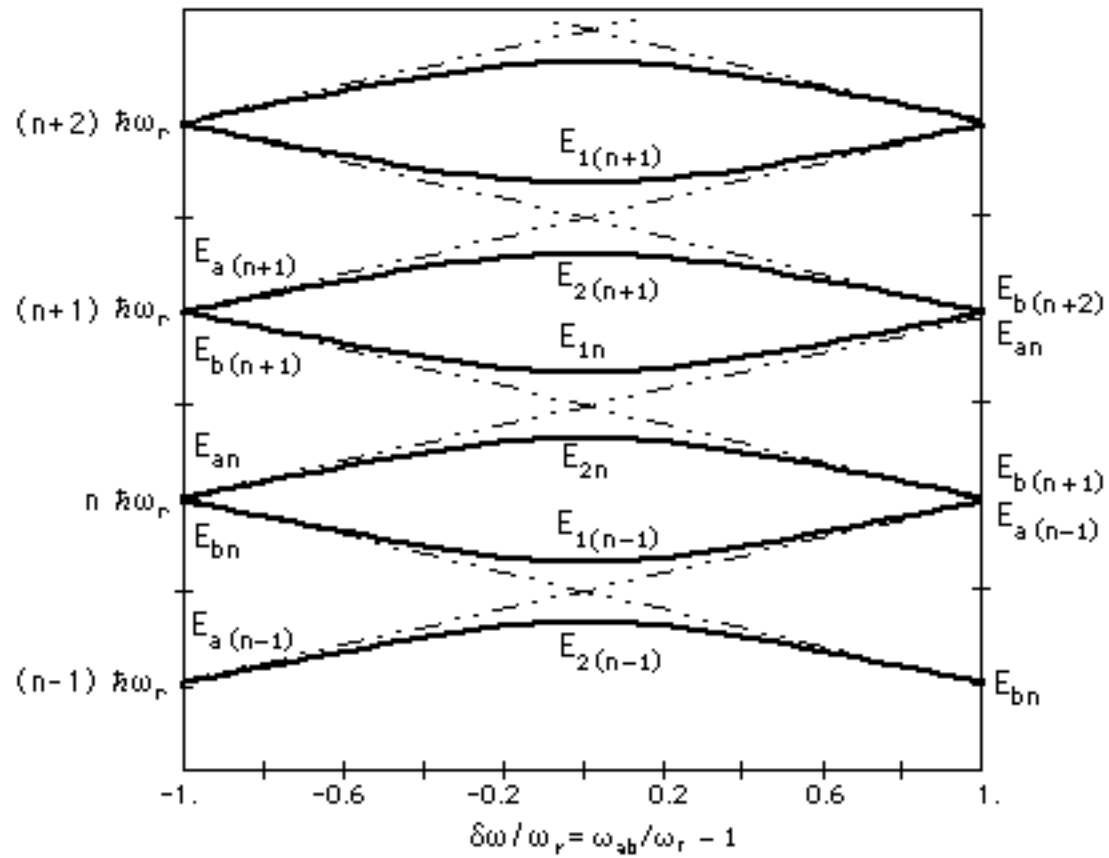
$$\sin \theta_n = \frac{2g_k \sqrt{n+1}}{\sqrt{(\omega_n^R - \omega_k)^2 + 4|g_k|^2(n+1)}}
 \tag{V-15c}$$

$$\begin{aligned}
 C_{2n}(t) &= C_{an}(t) \\
 C_{1n}(t) &= C_{b(n+1)}(t)
 \end{aligned}$$

where

$$\begin{aligned}
 | \rangle_n &= [C_{an}(t) |a n\rangle + C_{b(n+1)}(t) |b (n+1)\rangle] \\
 &= [C_{1n}(t) |1n\rangle + C_{2n}(t) |2n\rangle]
 \end{aligned}$$

**DRESSED ENERGY LEVELS OF TWO-STATE ATOM**



The time evolution of a state is directly represented in terms of these dressed states --  
*viz.*

$$\begin{aligned}
 | (t) \rangle &= \exp(-i \mathcal{H} t / \hbar) | (0) \rangle \\
 &= \sum_{n=0}^{\infty} \exp(-i E_{s_n} t / \hbar) |s n\rangle \langle s n| (0) \rangle \quad [ V-16a ] \\
 &= \sum_{n=0}^{\infty} \exp(-i [n + \frac{1}{2}] \frac{\mathbf{R}}{\hbar} t) \left\{ \exp(-\frac{1}{2} i \frac{\mathbf{R}}{\hbar} t) |1 n\rangle C_{1n}(0) + \exp(\frac{1}{2} i \frac{\mathbf{R}}{\hbar} t) |2 n\rangle C_{2n}(0) \right\}
 \end{aligned}$$

or more explicitly

$$\begin{pmatrix} C_{2n}(t) \\ C_{1n}(t) \end{pmatrix} = \begin{pmatrix} \exp(\frac{1}{2} i \frac{\mathbf{R}}{\hbar} t) & 0 \\ 0 & \exp(-\frac{1}{2} i \frac{\mathbf{R}}{\hbar} t) \end{pmatrix} \begin{pmatrix} C_{2n}(0) \\ C_{1n}(0) \end{pmatrix} \quad [ V-16b ]$$

Therefore, the **flopping** of the undressed states is given by

$$\begin{aligned}
 \begin{pmatrix} C_{an}(t) \\ C_{b(n+1)}(t) \end{pmatrix} &= \begin{pmatrix} \exp(\frac{1}{2} i \frac{\mathbf{R}}{\hbar} t) & 0 \\ 0 & \exp(-\frac{1}{2} i \frac{\mathbf{R}}{\hbar} t) \end{pmatrix} \begin{pmatrix} C_{an}(0) \\ C_{b(n+1)}(0) \end{pmatrix} \\
 &= \begin{pmatrix} \cos(\frac{1}{2} \frac{\mathbf{R}}{\hbar} t) + i \sin(\frac{1}{2} \frac{\mathbf{R}}{\hbar} t) \cos 2_n & -i \sin(\frac{1}{2} \frac{\mathbf{R}}{\hbar} t) \sin 2_n \\ -i \sin(\frac{1}{2} \frac{\mathbf{R}}{\hbar} t) \sin 2_n & \cos(\frac{1}{2} \frac{\mathbf{R}}{\hbar} t) - i \sin(\frac{1}{2} \frac{\mathbf{R}}{\hbar} t) \cos 2_n \end{pmatrix} \begin{pmatrix} C_{an}(0) \\ C_{b(n+1)}(0) \end{pmatrix} \quad [ V-17 ] \\
 &= \begin{pmatrix} \cos(\frac{1}{2} \frac{\mathbf{R}}{\hbar} t) - i \left( \frac{\mathbf{R}}{\hbar} \right) \sin(\frac{1}{2} \frac{\mathbf{R}}{\hbar} t) & -i \left( 2g_r \sqrt{n+1} / \frac{\mathbf{R}}{\hbar} \right) \sin(\frac{1}{2} \frac{\mathbf{R}}{\hbar} t) \\ -i \left( 2g_r \sqrt{n+1} / \frac{\mathbf{R}}{\hbar} \right) \sin(\frac{1}{2} \frac{\mathbf{R}}{\hbar} t) & \cos(\frac{1}{2} \frac{\mathbf{R}}{\hbar} t) + i \left( \frac{\mathbf{R}}{\hbar} \right) \sin(\frac{1}{2} \frac{\mathbf{R}}{\hbar} t) \end{pmatrix} \begin{pmatrix} C_{an}(0) \\ C_{b(n+1)}(0) \end{pmatrix}
 \end{aligned}$$

Perhaps the most revealing application of this result is for the case of a resonant coupled system -- *i.e.*  $\omega = \omega_0$  -- which is prepared so that  $C_{b(n+1)}(0) = 0$ . In this instance, Equation [ V-17 ] yields

$$\begin{aligned} |C_{an}(t)|^2 &= \cos^2[g_k \sqrt{n+1} t] |C_{an}(0)|^2 \\ |C_{b(n+1)}(t)|^2 &= \sin^2[g_k \sqrt{n+1} t] |C_{an}(0)|^2 \end{aligned} \quad [ V-18 ]$$

which clearly exhibits the simplest manifestation of *spontaneous emission* -- *i.e.* Rabi flopping in the absence of an applied field!!!

**Interaction of a Two-level Atom and a Multi Mode Field -- Spontaneous Emission:**

To broaden (make more realistic) our treatment of spontaneous emission we return to Equation [ V-8 ] to include the interaction with many modes with a two-level atom -- *viz.*

$$\mathcal{H}_{ED} = \hbar \sum_{\{l\}} \left\{ g_{\{l\}s}^+ + g_{\{l\}s}^- \right\} \left\{ i a_{\{l\}s}(t) \exp[i \vec{k}_{\{l\}} \cdot \vec{r}_A] + adj. \right\} \quad [ V-8' ]$$

This equation may be easily generalized to encompass multi-level material systems.<sup>29</sup>

If we are dealing with situation in which the locations of the atoms are **uncorrelated** we may, for simplicity, dispense with the  $\exp[\pm i \vec{k}_{\{l\}} \cdot \vec{r}_0]$  factors -- *i.e.* we will neglect, for the present, any possible *interference effects* -- and write

<sup>29</sup> For a multilevel atomic system the effective interaction Hamiltonian in second quantized form becomes

$$\mathcal{H}_{ED} = e \sum_{\{l\}} \sqrt{\frac{\hbar}{2 \epsilon_0}} \left\{ \hat{\mathbf{e}}_{\{l\}s} \langle | \mathcal{D} | \rangle \right\} \left\{ i a_{\{l\}s}(t) \exp[i \vec{k}_{\{l\}} \cdot \vec{r}_0] + adj. \right\} | \chi \rangle$$

$$\mathcal{H}_{ED} = \hbar \sum_{\{\ell\}} \sum_{s=1}^2 \left[ g_{\{\ell\}s}^+ + g_{\{\ell\}s}^- \right] \left[ i a_{\{\ell\}s}(t) - i a_{\{\ell\}s}^\dagger(t) \right] \quad [ V-19 ]$$

Transforming to the Schrödinger picture and taking the unperturbed ground state  $|g\rangle = |b\rangle|0\rangle|0\rangle \cdots |0\rangle$  as the zero energy reference point we may write the complete effective Hamiltonian as

$$\mathcal{H} = \hbar \sum_{ab} \left( \epsilon_a - \epsilon_b \right) |a\rangle \langle b| + \sum_{\{\ell\}} \sum_{s=1}^2 \left\{ \hbar a_{\{\ell\}s}^\dagger a_{\{\ell\}s} + \hbar \left[ g_{\{\ell\}s}^+ + g_{\{\ell\}s}^- \right] \left[ i a_{\{\ell\}s} - i a_{\{\ell\}s}^\dagger \right] \right\} \cdot [ V-20a ]$$

If we include only **energy-conserving** terms -- *i.e.* in the rotating wave approximation --

$$\mathcal{H} = \hbar \sum_{ab} \left( \epsilon_a - \epsilon_b \right) |a\rangle \langle b| + \sum_{\{\ell\}} \sum_{s=1}^2 \left\{ \hbar a_{\{\ell\}s}^\dagger a_{\{\ell\}s} + \hbar \left[ i g_{\{\ell\}s}^+ a_{\{\ell\}s} - i g_{\{\ell\}s}^- a_{\{\ell\}s}^\dagger \right] \right\} \cdot [ V-20b ]$$

It is important to note that, in general, the interaction terms in this Hamiltonian include contributions from the coupling of the material system (atom) to any externally excited mode(s) (the incident electromagnetic field) and to all available electromagnetic cavity modes. For the present, we ignore the coupling to externally excited modes: we treat the external interaction later as a perturbation. Our goal at this point is to diagonalize the Hamiltonian of the complete unperturbed system which may be written

$$\begin{aligned} \hbar^{-1}\mathcal{H} = & \begin{bmatrix} \omega_0 & ig_1 & ig_2 & \cdots & ig_N & 0 \\ -ig_1 & \omega_1 & 0 & \cdots & 0 & a_1 \\ -ig_2 & 0 & \omega_2 & \cdots & 0 & a_2 \\ \vdots & \vdots & \vdots & \ddots & \vdots & \vdots \\ -ig_N & 0 & 0 & \cdots & \omega_N & a_N \end{bmatrix} \cdot \text{ [ V-21 ]} \\ & = \bar{\mathbf{a}}^\dagger \left( \bar{\omega} + \bar{\mathbf{g}} \right) \bar{\mathbf{a}} \end{aligned}$$

We are looking for the eigenstates and eigenvalues of this Hamiltonian which we write as

$$\mathcal{H} |s\rangle = \hbar \omega_s |s\rangle. \quad \text{ [ V-22 ]}$$

Since the square matrix is Hermitian, it is possible, **in principle**, to diagonalize it via a unitary transformation of the form

$$\bar{\mathbf{U}} \left( \bar{\omega} + \bar{\mathbf{g}} \right) \bar{\mathbf{U}}^{-1} = \bar{\omega} \quad \text{ [ V-23 ]}$$

where  $\bar{\omega}$  is a diagonal matrix whose elements are the  $N+1$  eigenvalues of the **dressed** states of the coupled system. If we define the row vector  $\bar{\mu}^\dagger = \bar{\mathbf{a}}^\dagger \bar{\mathbf{U}}^{-1}$  and the column vector  $\bar{\mu} = \bar{\mathbf{U}} \bar{\mathbf{a}}$  then Equation [ V-22 ] becomes

$$\hbar^{-1}\mathcal{H} = \bar{\mathbf{a}}^\dagger \left( \bar{\omega} + \bar{\mathbf{g}} \right) \bar{\mathbf{a}} = \bar{\mu}^\dagger \bar{\omega} \bar{\mu} = \sum_{t=1}^{N+1} \mu_t^\dagger \mu_t \quad \text{ [ V-24 ]}$$

For consistency,  $\langle r | \mu_t^\dagger \mu_t | s \rangle = \delta_{rs} \delta_{ts}$  and, hence,  $\bar{\mu}^\dagger$  and  $\bar{\mu}$  are, respectively, paired creation and destruction operators in the sense of the operators defined in Equation [ V-3 ] above -- *i.e.*  $|s\rangle = \mu_s^\dagger |g\rangle$  and  $|g\rangle = \mu_s |s\rangle$ . From Equation [ V-23 ] we may write

$$\vec{U} (\vec{r} + \vec{g}) = \vec{r} \vec{U} \quad [ V-25a ]$$

which can be written explicitly as

$$\begin{matrix} U_{11} & U_{12} & U_{13} & \cdots & U_{1(N+1)} & 0 & ig_1 & ig_2 & \cdots & ig_N \\ U_{21} & U_{22} & U_{23} & \cdots & U_{2(N+1)} & -ig_1 & 1 & 0 & \cdots & \\ U_{31} & U_{32} & U_{33} & \cdots & U_{3(N+1)} & -ig_2 & 0 & 2 & \cdots & \\ \vdots & \vdots & \vdots & \vdots & \vdots & \vdots & \vdots & \vdots & \vdots & \\ U_{(N+1)1} & U_{(N+1)2} & U_{(N+1)3} & \cdots & U_{(N+1)(N+1)} & -ig_N & 0 & 0 & \cdots & N \end{matrix}$$

$$= \begin{matrix} {}_1 U_{11} & {}_1 U_{12} & {}_1 U_{13} & \cdots & {}_1 U_{1(N+1)} \\ {}_2 U_{21} & {}_2 U_{22} & {}_2 U_{23} & \cdots & {}_2 U_{2(N+1)} \\ {}_3 U_{31} & {}_3 U_{32} & {}_3 U_{33} & \cdots & {}_3 U_{3(N+1)} \\ \vdots & \vdots & \vdots & \vdots & \vdots \\ {}_{(N+1)} U_{(N+1)1} & {}_{(N+1)} U_{(N+1)2} & {}_{(N+1)} U_{(N+1)3} & \cdots & {}_{(N+1)} U_{(N+1)(N+1)} \end{matrix} \quad [ V-25b ]$$

Expanding out the matrix product on the left we see that

$$U_{s1} - i \sum_t U_{s(t+1)} g_t = {}_s U_{s1} \quad [ V-26a ]$$

for elements **in the first column** and

$$i U_{s1} g_{t-1} + U_{st} - {}_{t-1} U_{st} = {}_s U_{st} \quad s \neq t \quad [ V-26b ]$$

for elements **not in the first column**. Hence

$$U_{st} = i U_{s1} g_{t-1} / ({}_s - {}_{t-1}) \quad s \neq t \quad [ V-26c ]$$

and all elements of the unitary matrix can be expressed in terms of the first-column elements as

$$\vec{U} = \begin{matrix} U_{11} & \frac{i U_{11} g_1}{1^{-} 1} & \frac{i U_{11} g_2}{1^{-} 2} & \dots & \frac{i U_{11} g_N}{1^{-} N} \\ U_{21} & \frac{i U_{21} g_1}{2^{-} 1} & \frac{i U_{21} g_2}{2^{-} 2} & \dots & \frac{i U_{21} g_N}{2^{-} N} \\ U_{31} & \frac{i U_{31} g_1}{3^{-} 1} & \frac{i U_{31} g_2}{3^{-} 2} & \dots & \frac{i U_{31} g_N}{3^{-} N} \\ \vdots & \vdots & \vdots & \vdots & \vdots \\ U_{(N+1)1} & \frac{i U_{(N+1)1} g_1}{(N+1)^{-} 1} & \frac{i U_{(N+1)1} g_2}{(N+1)^{-} 2} & \dots & \frac{i U_{(N+1)1} g_N}{(N+1)^{-} N} \end{matrix} \quad [ V-27 ]$$

Quite generally the columns of any unitary matrix satisfy an orthonormal condition -- viz.

$$\sum_t U_{tr} U_{ts} = \delta_{rs} \quad [ V-28a ]$$

so the normalization of the first column gives

$$\sum_t |U_{t1}|^2 = 1 \quad [ V-28b ]$$

and the orthogonality of the first column with any other column yields

$$\sum_t \frac{|U_{t1}|^2}{t^{-} q} = 0 \quad [ V-29c ]$$

for each one of the cavity frequencies  $q$ .

Further we may multiply Equation [ V-28c ] by a product of factors

$$\frac{|U_{t1}|^2}{\omega_q - \omega_t - \omega_s} = 0 \quad [ V-28d ]$$

It is obvious from this expression the  $\omega_q$ 's are roots of the left side of the equation if the  $\omega_q$ 's are replaced by  $-\omega_q$ . Thus

$$\frac{|U_{t1}|^2}{-\omega_t - \omega_s} = \frac{|U_{t1}|^2}{\omega_q} \quad [ V-28e ]$$

Finally, we see that

$$\frac{|U_{t1}|^2}{\omega_t - \omega_s} = \frac{\omega_q}{\omega_s} \quad [ V-29 ]$$

We can make use of this expression to obtain the time varying polarization induced by an externally excited field. The Hamiltonian associated with this perturbation may be written

$$\mathcal{H}_{ED} = \hbar \{ g_{ext}^+ + g_{ext}^- \} \{ i a_{ext} - i a_{ext}^\dagger \} \quad [ V-30 ]$$

Since  $\vec{a}^\dagger = \vec{\mu}^\dagger \vec{U}$  and  $\vec{a} = \vec{U}^{-1} \vec{\mu}$  we see that

$$g_{ext}^+ = \mu_t^\dagger U_{t1} \quad \text{and} \quad g_{ext}^- = U_{t1}^{-1} \mu_t = U_{t1} \mu_t \quad [ V-31 ]$$

and from Equation [ V-6 ] we may write

$$\begin{aligned} \vec{\mathcal{D}} &= \langle a | \vec{\mathcal{D}} | b \rangle^+ + \langle b | \vec{\mathcal{D}} | a \rangle^- \\ &= \left\{ \langle a | \vec{\mathcal{D}} | b \rangle U_{t1} \mu_t^\dagger + \langle b | \vec{\mathcal{D}} | a \rangle U_{t1} \mu_t \right\} \end{aligned} \quad [ V-32 ]$$

In light of Equation [ V-29 ], the *standardized form* for the frequency dependent susceptibility (see Equation [ VA-12 ] becomes

$$\chi''(\omega) = \frac{N e^2}{\hbar \omega_0} \left| \langle a | \vec{\mathcal{D}} | b \rangle \right|^2 \left[ \frac{1}{\omega_s - \omega} + \frac{1}{\omega_s + \omega} \right] \quad [ V-33 ]$$

By eliminating the U's from Equations [ V-24a ] and [ V-24b ] we see that

$$\sum_t |g_t|^2 (\omega_s - \omega_t)^{-1} = 0 \quad [ V-34 ]$$

Again

$$\sum_t |g_t|^2 (\omega_s - \omega_t)^{-1} \sum_q (\omega_s - \omega_q) = 0 \quad [ V-35 ]$$

so that we can write

$$\sum_t |g_t|^2 (\omega_s - \omega_t)^{-1} \sum_q (\omega_s - \omega_q) = \sum_s (\omega_s - \omega_s) \quad [ V-36 ]$$

Therefore

$$\chi''(\omega) = \frac{N e^2}{\hbar \omega_0} \left| \langle a | \vec{\mathcal{D}} | b \rangle \right|^2 \left[ \frac{1}{\sum_t |g_t|^2 (\omega_s - \omega_t)^{-1}} + \frac{1}{\sum_t |g_t|^2 (\omega_s + \omega_t)^{-1}} \right] \quad [ V-37 ]$$

where the sum  $\sum_t |g_t|^2 (\omega_s - \omega_t)^{-1}$  gives an explicit, non-phenomenological accounting of interactions with the cavity modes and hence of spontaneous emission!

**EVALUATION OF SPONTANEOUS EMISSION RATE:**

Recall the discussion of phenomenologically defined damping in semiclassical models of the dielectric response function in the lecture set entitled *The Interaction of Radiation and Matter: Semiclassical Theory*. Recollect, in particular, Equation [ III-19c ] in those notes. In reconciling that discussion with the content of Equation [ V-37 ] above, we see that the summation  $\sum_t |g_t|^2 (\omega_t - \omega)^{-1}$  replaces the simple damping parameter  $\gamma$ . Our task here is evaluate this integral which we write as

$$i^{-1} \gamma(\omega) = \lim_{\epsilon \rightarrow 0} \sum_t \frac{|g_t|^2}{\omega_t - \omega - i\epsilon} = \lim_{\epsilon \rightarrow 0} \int \frac{|g(\omega')|^2}{\omega' - \omega - i\epsilon} d\omega' \quad [ V-38 ]$$

where  $\int \frac{|g(\omega')|^2}{\omega' - \omega - i\epsilon} d\omega'$  is number of cavity modes with frequency between  $\omega'$  and  $\omega' + d\omega'$ . Following arguments best explicated long ago by Heitler,<sup>30</sup> it may be shown that

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<sup>30</sup> W. Heitler, in Chapter II, Section 8 of *The Quantum Theory of Radiation* (3rd edition), Oxford Press (1954) uses contour integral arguments to show that

$$\lim_{\epsilon \rightarrow 0} \frac{1}{x - i\epsilon} = \mathcal{P}r \frac{1}{x} + i\pi \delta(x).$$

To quote W. H. Louisell’s summary in Chapter 5 of *Radiation and Noise in Quantum Electronics*,

“...if  $f(x)$  is well behaved and has no poles at  $x = a$ , then

$$\int \frac{f(x)}{x - a} dx = \mathcal{P}r \int \frac{f(x)}{x - a} dx + i\pi f(a)$$

where  $\mathcal{P}r$  means the Cauchy principle part and is defined by

$$\mathcal{P}r \int \frac{f(x)}{x - a} dx = \lim_{\mu \rightarrow 0} \int_{a-\mu}^{a-\mu} \frac{f(x)}{x - a} dx + \int_{a+\mu}^{a+\mu} \frac{f(x)}{x - a} dx$$

provided the limit on the right side exists.”

$$i \dot{\rho}_{ii} = \mathcal{P}r \int_0^\infty \frac{|g_t|^2}{t} \rho_{ii}(t) d t + i \rho_{ii} - |g_t|^2 \rho_{ii}(t) d t \dots [V-39]$$

which we write as  $i \dot{\rho}_{ii} = \rho_{ii} + i \Delta \rho_{ii}$ . **This is an extremely important result!!** It shows that the interaction between the atom and the cavity modes leads to a frequency shift or correction in the atomic splitting  $\Delta \omega_{ab}$

$$\Delta \omega_{ab} = \mathcal{P}r \int_0^\infty \frac{|g_t|^2}{t} \rho_{ii}(t) d t \quad [V-40a]$$

and a spontaneous emission decay rate

$$\Gamma = |g_t|^2 \rho_{ii} \quad [V-40b]$$

If we assume that the cavity modes defined for the blackbody calculation in Section IV of the lecture set entitled *The Interaction of Radiation and Matter: Semiclassical Theory* (see Equation [IV-5] in those notes) are the appropriate modes, we know that

$$\rho_{ii}(t) = \frac{2}{c^3} \omega^2$$

and from Equation [V-9] we know that

$$|g_t|^2 = \frac{e^2}{2 \hbar} \left| \langle a | \vec{D} | b \rangle \cdot \hat{e}_t \right|^2$$

Treating the shift  $\Delta \omega_{ab}$ , the radiative correction to atomic energy level separation, is a very complex and much studied matter. The simple interpretation of Equation [V-38a] is problematic since the integrand is proportional to  $\omega^2$  at large  $\omega$  and, thus, the correction significantly diverges!!

The divergence in  $(\dots)$  was for many years an unresolved discrepancy between the quantum theory of radiation and observational spectroscopy. The difficulty was overcome by Bethe in 1947<sup>31</sup> using a technique known as mass renormalization. Bethe pointed out that the divergence can mainly be associated with the mass of the electron. It is found that the energy of a free electron has an infinite contribution arising from the interaction of the electron with the electromagnetic field. In other words, the apparent mass of the electron is shifted by an infinite amount from the mass of an electron which is not in interaction with the radiation field. However, the former mass is the one measured experimentally, since it is never possible to isolate an electron from the radiation field. Identification of the measured electron mass with the theoretical mass, after renormalization to take account of the energy of interaction with the radiation field, removes most of the divergence from  $(\dots)$ .....

Calculations for the hydrogen atom show that  $(\dots)$  vanish unless one of the states in the transition is an S state. Even when it does not vanish, the renormalized  $(\dots)$  is always very small compared with the excitation frequency  $\omega_0$ , and varies slowly with  $\omega_0$ . For example, the magnitude of  $(\dots)$  for the  $2^2S_{1/2}$  state of hydrogen is about  $10^9$  Hz, or roughly six orders of magnitude smaller than the  $n = 2$  state excitation energy..... The existence of level shifts was first demonstrated by Lamb and Retherford in experiments on radiative transition between the  $2^2S_{1/2}$  state of hydrogen and the unshifted  $2^2P_{1/2}$  state. The splitting between these states is known as the Lamb shift.<sup>32</sup>

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<sup>31</sup> Bethe, H. A., *Phys. Rev.* **72**, 339 (1947)

<sup>32</sup> From Chapter 8, Rodney Loudon, *Quantum Theory of Light* (1st edition), Oxford (1973)

However Equation [ V-38b ] is not complicated by divergences and, consequently, we easily obtain the famous Weisskopf-Wigner formul<sup>33</sup> for the spontaneous emission decay rate into the modes of a three-dimensional cavity

$$\Gamma = \frac{1}{4\pi} \frac{2}{3} \frac{e^2}{\hbar c^3} |\langle a | \vec{\mathcal{D}} | b \rangle \cdot \hat{\mathbf{e}}_i|^2 \quad [ V-41 ]$$

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<sup>33</sup> V. Weisskopf and E. Wigner, Z. Phys., **63**, 54 (1930).