Two-level and three-level atoms with a strongly coupled resonance

References: Boyd, Nonlinear Optics (3rd Ed.); Levensen and Kano, Introduction to Nonlinear Laser Spectroscopy

Until now, our treatment of multiphoton and nonlinear optical effects has assumed that lowest-order perturbation theory is applicable. This is not the case if the optical field is large enough, or close enough to resonance, to strongly couple at least one pair of atomic or molecular states. To initiate our brief overview of strong-field effects of light on atoms and molecules, consider an atom in which two levels $|a\rangle$ and $|b\rangle$ are strongly electric-dipole coupled by a monochromatic radiation field at frequency $\omega$. We will also allow for the possible existence of additional levels to which transitions are possible from the coupled $|a\rangle - |b\rangle$ system. However, it’s important to note that the general three-level atom is an extremely complicated problem (see Vol. 2 of Shore, The Theory of Coherent Atomic Excitation, and Stenholm, Foundations of Laser Spectroscopy). We focus here on effects that occur when only two of the levels are strongly coupled.

I. Two-level system with strong coupling

The starting point is identical to p. 2LA-12 in the Physics 6110 notes, from which the relevant equations are repeated here for convenience, with a few minor changes:

Neglecting spontaneous decay rates, which can be introduced approximately by using complex energies (the Bethe-Lamb prescription), we want to find the solution of the time-dependent Schrödinger equation

$$i\hbar \frac{\partial \psi}{\partial t} = H \psi,$$

as a superposition of the two unperturbed eigenstates,

$$\psi = c_a e^{-i\omega t} |a\rangle + c_b e^{-i\omega t} |b\rangle.$$ (2)

We must include the effects of the radiation field. We do this in the electric dipole approximation by adding to the Hamiltonian the electric dipole interaction of pp. 2LA-8 and 2LA-9, assuming for simplicity that $E$ is along $z$: 
\[ H_{E1} = e \mathbf{E} \cdot \mathbf{r} = \frac{e\omega}{2} \left( E_0 e^{-i\omega t} + E_0^* e^{i\omega t} \right). \] (3)

Taking into account that the diagonal matrix elements are zero due to the electric dipole parity selection rule, the time-dependent Schrödinger equation leads to two coupled equations,

\[
\dot{c}_a = -\frac{\langle a | z | b \rangle}{2\imath \hbar} c_b \left( E_0^* e^{-(i\omega_b-a)\imath t} + E_0 e^{-(i\omega_b+a)\imath t} \right) \quad \text{and} \quad \dot{c}_b = -\frac{\langle b | z | a \rangle}{2\imath \hbar} c_a \left( E_0 e^{(i\omega_b-a)\imath t} + E_0^* e^{(i\omega_b+a)\imath t} \right). \] (4)

Here it’s convenient to introduce the Rabi frequency, which may be complex:

\[ \omega_R = \frac{eE_0}{\hbar} \langle b | z | a \rangle. \] (5)

We make the rotating wave approximation (RWA), dropping the \( \omega_{ba}+\omega \) terms. Defining the detuning \( \delta \) in the usual way,

\[ \delta = \omega - \omega_{ba} \] (6)
we find,

\[
i\dot{c}_a = \frac{\omega_R}{2} c_b e^{i\delta \imath t}, \]
\[
i\dot{c}_b = \frac{\omega_R}{2} e^{-i\delta \imath t} c_a. \] (7)

Unlike our earlier treatment, this time we need the general solution to Eqs. (7). Solve using trial solutions of the form \( c_a = ke^{-i\delta t} \) to obtain,

\[
c_a(t) = e^{i\delta t/2} \left( A_+ e^{-i\Omega' t/2} + A_- e^{i\Omega' t/2} \right),
\]
\[
c_b(t) = e^{-i\delta t/2} \left( -A_+ \frac{\Omega' - \delta}{\omega_R} e^{-i\Omega' t/2} + A_- \frac{\Omega' + \delta}{\omega_R} e^{i\Omega' t/2} \right), \] (8)

where \( A_+ \) and \( A_- \) are arbitrary constants of integration, and the \textit{generalized Rabi frequency} \( \Omega' \) is given by

\[ \Omega' \equiv \sqrt{|\omega_R|^2 + \delta^2} \] (9)

Clearly, Eqs. (8) contain new frequencies that have been generated by the intense optical interaction. Although the Schrödinger equation itself is linear, the behavior of the coupled two-level system is not.
Substituting Eqs. (8) into (2), we conclude that the wave functions contain four frequencies:

\[
\begin{align*}
    w_b & \quad w_b + \frac{1}{2} \Delta + \frac{1}{2} \Omega' \\
    w_b + \frac{1}{2} \Delta - \frac{1}{2} \Omega' & \quad w_b + \frac{1}{2} \Delta + \frac{1}{2} \Omega' \\
    w_a & \quad w_a - \frac{1}{2} \Delta + \frac{1}{2} \Omega' \\
    w_a - \frac{1}{2} \Delta - \frac{1}{2} \Omega' &
\end{align*}
\]

The drawing shows the case \( \Delta > 0 \). For \( \Delta < 0 \) the algebraic expressions are still correct, but the average shift is \( < 0 \) for \( |b \rangle \) and \( > 0 \) for \( |a \rangle \). Also, note that \( \Delta > 0 \) and \( \Delta > |\Delta| \) always.

The induced dipole moment oscillates not only at \( \Omega \), but also at \( \Omega \pm \Delta \) as shown on the sketch. This is not directly observable by monitoring \( P_b(t) \), but it can be seen by introducing a weak "probe" beam to examine the shifted energy levels.

II. Three-level "pump-probe" system

\[
\begin{align*}
    |f\rangle & \quad |f\rangle \\
    w_2 & \quad w_2 \\
    |b\rangle & \quad |b\rangle \\
    w & \quad w \\
    |a\rangle & \quad |a\rangle
\end{align*}
\]

If \( w \) is fixed and \( w_2 \) is scanned, the wave functions of Eqs. (8) and (2) can be monitored. Let's look at the most obvious consequences:
A. The Autler-Townes doublet:

Assume \( w \) is resonant, \( w = w_{ba} \).

Then \( \delta = 0 \) and \( \Delta^* = w_{R} \). The level \( 1b \rangle \)
is split by the field,

We see a doublet split by \( w_{R} \), the Rabi frequency of the \( a \leftrightarrow b \) transition created by the field at \( w \).

The splitting is proportional to \( E_{1} \), or \( \sqrt{E_{1}} \)
(not to \( E_{2} \), which we assume is weak).

If \( \delta \neq 0 \), we still see a doublet split by \( \Delta^* \), but the intensities become asymmetric.

(If \( |w_{R}| < |\delta| \), can expand to get ordinary ac Stark shift.)

But how do we find the relative sizes? We need the "stationary-state" solutions, in the sense that \( \rho_{a} \) and \( \rho_{b} \) are time-independent:

B. Dressed states

A very helpful way to look at the system is to view the levels of the atom + field system.

For the two-level system, with no interaction,
If we turn on the dipole coupling, the crossings are avoided:

These dressed states correspond to the solutions that have stationary probability, $P_r(a) =$ constant and $P_r(b) =$ constant. There are two types:

1) $\psi^+ : \text{ set } A_+ = 1, \quad A_- = 0$
2) $\psi^- : \text{ set } A_+ = 0, \quad A_- = 1$

Substituting into (2) and normalizing,

$$\Psi_\pm = \frac{\omega^*_R}{\sqrt{2}} \sqrt{-\frac{\Delta'}{2(\Delta'^2 + \delta)}} \left| a \right> e^{-i(w_a - \frac{1}{2}\Delta' \pm \frac{1}{2}\Delta) t} + \sqrt{\frac{\Delta'^2 + \delta}{2\Delta'}} \left| b \right> e^{-i(w_b + \frac{1}{2}\Delta' \pm \frac{1}{2}\Delta) t}$$

Note $P_r(a) = |\langle a | \Psi_\pm \rangle|^2$ is constant, as specified.

Note we cannot see the Auster-Townes doublet or the ac Stark shift except by using a "probe" — absorption at $\omega$ is resonant only at $\omega_B$.

Expand $-\Delta'$ for $| \Delta' | \gg | \Delta |$ to show $\psi_+$ is mostly $| a \rangle$,

$\psi_-$ is mostly $| b \rangle$. 

Note: There is no $\Delta'$. It is assumed that $\Delta'$ is large compared to $\Delta$.
The probe transition can now be interpreted as excitation from \( |\Psi_+ \rangle \) or \( |\Psi_- \rangle \) to \( |c\rangle \), and the relative amplitudes can be found by solving for their amplitudes, if we know the history of the system. (At resonance, degeneracy guarantees equal populations.)

Note that \( |\Psi_+ \rangle \), \( |\Psi_- \rangle \) are not eigenstates. Instead they are stationary solutions for a time-dependent Hamiltonian.

**IV. Two-level atom with a near-resonant probe**

The induced dipole oscillates not only at \( \omega \), but also at \( \omega + \Delta \), \( \omega - \Delta \). Thus if we introduce a second field at \( \omega_2 \), we see three scattering peaks:

- "three-photon" resonance, \( \omega_2 = \omega - \Delta \)
- A.C.-Stark shifted resonance, \( \omega_2 = \omega + \Delta \)
  (transition from \( \Psi_+ \) to \( \Psi_- \))
- Rayleigh or elastic scattering.
Note the absorption can be negative — "parametric mixing". Also, this is a nonlinear two-level response — at strong fields, get both $X^{(1)}$ and $X^{(7)}$.

V. Coherent Dark States and Coherent Control

Here we take advantage of coherent excitation to change the optical behavior or to control product pathways. See the transparenties that follow for an introduction.