Notes for Lectures on Coherent Excitation

I. RATE EQUATIONS: 2-LEVEL SYSTEM

Consider the 2-level system:

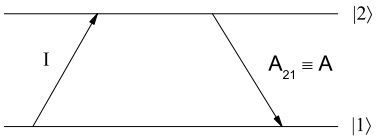


FIG. 1: Schematic of two-level system, showing excitation (left) and decay(right).

If we denote the relative populations of levels 1 and 2 by n_1 and n_2, respectively, then the equations for the rates of change of these populations are given by:

$$\dot{n}_1 = n_2 A + n_2 BI - n_1 BI \tag{1a}$$

$$\dot{n}_2 = -n_2 A - n_2 BI + n_1 BI \tag{1b}$$

$$n_1 + n_2 = 1. \tag{1c}$$

The constants A and B are the familiar Einstein A and B coefficients for spontaneous and stimulated emission/absorption. Here, $B_{ab} = \frac{\pi^2 c^2}{\hbar\omega^3 \Delta\nu} A_{ab}$, (for $g_1 = g_2 = 1$), and I is the intensity of light (in, say, mW/cm^2). In order to allow direct use of the more common laboratory definition of I, the linewidth of the light, $\Delta\nu$, has been included in the definition of B. The first two equations are clearly redundant. Combining the second two equations we get

$$\dot{n}_2 = -n_2(A + 2BI) + BI. \tag{2}$$

First, consider the steady-state solution of (2). That is, we set $\dot{n}_2 = 0$. Then, $n_2 = \frac{BI}{A+2BI}$. Note that if $BI \gg A$, then the steady-state excited population $\to 1/2$.

Now, back to equation (2): Try a solution like $n_2(t) = f(t)\exp[-(A + 2BI)t]$. Then,

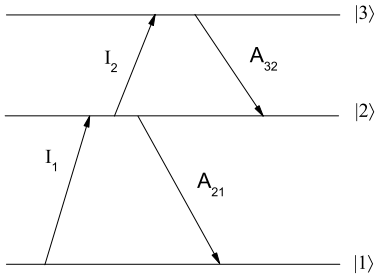
$$\dot{n}_2 = \left[\dot{f} - f(A + 2BI)\right] e^{-(A+2BI)t}. \tag{3}$$

Setting the right-hand side (RHS) of this expression equal to the RHS of equation (2), and applying the boundary condition $n_2(t = 0) = 0$, leads to

$$n_2(t) = \frac{BI}{A + 2BI}\left(1 - e^{-(A+2BI)t}\right). \tag{4}$$

Note that as $t \to \infty$, $n_2 \to \frac{BI}{A+2BI}$, as in the steady-state case.

HW1a: Set up the rate equations for the 3-level "ladder" system shown below. Here, $A_{31} = 0$, and for all levels, $B_{ab} = B_{ba}$. That is, $g_1 = g_2 = g_3 = 1$.



HW1b: Solve the equations analytically for steady-state conditions. What is the greatest fractional population that can be put into $|3>$? Here, $A_{31} = 0$, and for all levels, $B_{ab} = B_{ba}$. That is, $g_1 = g_2 = g_3 = 1$.

HW1c: Numerically solve the coupled differential equations. Assume various values for I_1 and I_2 (of order of a few mW/cm^2). For the purposes of this problem, set $A_{21} = 37.6 \times 10^6$ sec^{-1}, and $A_{32} = 12.5 \times 10^6$ sec^{-1}. Also assume $\lambda_1 = 780$ nm, and $\lambda_2 = 1.5\mu$m. (These are parameters consistent with the 5s, 5p, and 4d levels of Rb.)

II. COHERENCE: 2-LEVEL SYSTEM: THE RABI PROBLEM

It is natural to ask "can we ever do better than putting only 1/2 of the population into the excited state of a 2-level system? If we had a 3-level system, could we ever put more than 1/3 of the population into the highest level?" Clearly, from our study of these systems based on rate equations, the answer is "no!" However, as we shall soon see, in these studies we left out a very important resource available to us: the possibility of making use of the coherent property of light. From here on, we consider the use of coherent light in exciting atoms or molecules.

Consider again the 2-level system above but now, for the moment, set $A = 0$. The most appropriate vehicle for studying excitation of this system by coherent light is the time-dependent Schrödinger equation. That is, $i\hbar\frac{\partial\Psi}{\partial t} = H'\Psi$, $H' = H_0 + \hat{V}(t)$, where H', H_0, and $\hat{V}$ are Hermitian. H_0 is the Hamiltonian of the atomic (or molecular) system in the absence of external fields, and $\hat{V}$ represents the time-dependent external radiation field, treated here as a classical electromagnetic field. Note that, in general, near resonant, narrow linewidth

radiation can *not* be treated as a perturbation.

Set $\Psi(t) = \sum_n c_n(t)\psi_n e^{-i\xi_n(t)}$, where ψ_n satisfy the time-independent SE: $H_0\psi_n = E_n^0\psi_n$.

The $\xi_n(t)$ are time-dependent phases. Their values are arbitrary since, being just phases, they have no effect on any observable.

Then,

$$\frac{\partial\Psi}{\partial t} = \sum_n \psi_n \left[\dot{c}_n - i\dot{\xi}_n c_n\right] e^{-i\xi_n},$$

and

$$H'\Psi = \sum_n \left(H_0 + \hat{V}\right) c_n\psi_n e^{-i\xi_n}$$

Now, $\hat{V}$ acting on ψ re-distributes the probability. That is,

$$\hat{V}\psi_n = \psi_1 V_{1n} + \psi_2 V_{2n} + \cdots = \sum_m \psi_m V_{mn}.$$

Or, upon multiplying from the left by ψ_q^* and integrating over space, $V_{qn} = \langle\psi_q|V|\psi_n\rangle \equiv \langle q|V|n\rangle$, where we have made use of the orthonormality of ψ. Then, $H'\Psi = \sum_n \left[c_n E_n^0\psi_n + c_n\sum_m \psi_m V_{mn}\right] e^{-i\xi_n}$.

Putting everything together gives

$$i\hbar\sum_n \psi_n \left[\dot{c}_n - i\dot{\xi}_n c_n\right] e^{-i\xi_n} = \sum_n c_n E_n^0\psi_n e^{-i\xi_n} + \sum_n\sum_m c_n\psi_m V_{mn} e^{-i\xi_n}.$$

Multiplying through by ψ_l^*, and integrating over space gives:

$$i\hbar\int\sum_n \psi_l^*\psi_n \left[\dot{c}_n - i\dot{\xi}_n c_n\right] e^{-i\xi_n} d\vec{r} = \int\sum_n c_n E_n^0\psi_l^*\psi_n e^{-i\xi_n} d\vec{r} + \int\sum_n\sum_m c_n\psi_l^*\psi_m V_{mn} e^{-i\xi_n} d\vec{r}.$$

Making use of the orthonormality of ψ yields $i\hbar\left(\dot{c}_l - i\dot{\xi}_l c_l\right) e^{-i\xi_l} = c_l E_l^0 e^{-i\xi_l} + \sum_n c_n V_{ln} e^{-i\xi_n}$, or, $i\hbar\dot{c}_l = -\hbar\dot{\xi}_l c_l + E_l^0 c_l + \sum_n c_n V_{ln} e^{-i(\xi_n - \xi_l)}$.

Finally,

$$\hbar\dot{c}_l = -i\left[\left(E_l^0 - \hbar\dot{\xi}_l\right) c_l + \sum_n c_n V_{ln} e^{-i(\xi_n - \xi_l)}\right]. \tag{5}$$

All of this is exact so far. Now, for the first approximation, we truncate the sum in Eq. (5) to just the number of levels in the system, here, 2. That is, we neglect the population of states far from resonance. Under this approximation we end up with:

$$\begin{aligned}
\hbar\dot{c}_1 &= -i\left[\left(E_1^0 - \hbar\dot{\xi}_1 + V_{11}\right) c_1 + V_{12} c_2 e^{-i(\xi_2 - \xi_1)}\right] \\
\hbar\dot{c}_2 &= -i\left[\left(E_2^0 - \hbar\dot{\xi}_2 + V_{22}\right) c_2 + V_{21} c_1 e^{-i(\xi_2 - \xi_1)}\right],
\end{aligned}$$

or, more compactly, as:

$$\hbar\dot{\mathbf{c}} = -i \begin{pmatrix} E_1^0 + V_{11} - \hbar\dot{\xi}_1 & V_{12}e^{-i(\xi_2-\xi_1)} \\ V_{12}^* e^{i(\xi_2-\xi_1)} & E_2^0 + V_{22} - \hbar\dot{\xi}_2 \end{pmatrix} \mathbf{c}, \tag{6}$$

with

$$\mathbf{c} \equiv \begin{pmatrix} c_1 \\ c_2 \end{pmatrix}.$$

Now, in the dipole approximation, $\hat{V} = -e\vec{E}\,(\vec{r},t)\cdot\vec{r}$, $\vec{E} = \hat{\varepsilon}E_0\left(e^{i\omega t} + e^{-i\omega t}\right)/2$.

So, $V_{mn} = -e\vec{E}\cdot\langle\psi_m|\vec{r}|\psi_n\rangle = -\frac{e}{2}E_0\left(e^{i\omega t} + e^{-i\omega t}\right)\langle m|r|n\rangle$. Then, defining $\Omega \equiv \frac{-eE_0}{\hbar}\langle 1|r|2\rangle$, we get $V_{12} = \frac{\hbar\Omega}{2}\left(e^{i\omega t} + e^{-i\omega t}\right)$. Note that because E_0 can, in general, have a phase term, Ω is, in general, a complex quantity. In many cases we can choose the phase of our electric field to be 0, in which case, Ω will be pure real.

Equation (6) then becomes:

$$\hbar\dot{\mathbf{c}} = -i \begin{pmatrix} E_1 - \hbar\dot{\xi}_1 & \frac{\hbar\Omega}{2}\left(e^{-i(\xi_2-\xi_1-\omega t)} + e^{-i(\xi_2-\xi_1+\omega t)}\right) \\ \frac{\hbar\Omega^*}{2}\left(e^{i(\xi_2-\xi_1-\omega t)} + e^{i(\xi_2-\xi_1+\omega t)}\right) & E_2 - \hbar\dot{\xi}_2 \end{pmatrix}, \tag{7}$$

where $E_1 \equiv E_1^0 + V_{11}$ and $E_2 \equiv E_2^0 + V_{22}$.

At this point, we could just solve the coupled differential equations represented by Eq. (7). First, however, we take advantage of the arbitrariness in the values of the time-dependent phases, ξ_n; we assign values to them in such a way as to render Eq. (7) into as simple a form as possible. That is, we set $\xi_2 - \xi_1 = \omega t \Rightarrow \dot{\xi}_2 - \dot{\xi}_1 = \omega$.

We now make our third approximation. We say that if a term oscillates at a frequency significantly higher than the optical frequency, ω , we can ignore that term since it averages to 0. This is called the *rotating wave approximation* or RWA. Applying our definitions of phase, and the RWA to Eq. (7) (whereupon we set terms $\exp(\pm 2i\omega) = 0$) we obtain:

$$\hbar\dot{\mathbf{c}} = \frac{-i}{2} \begin{pmatrix} 2\hbar\Delta_1 & \hbar\Omega \\ \hbar\Omega^* & 2\hbar\Delta_2 \end{pmatrix} \mathbf{c}, \tag{8}$$

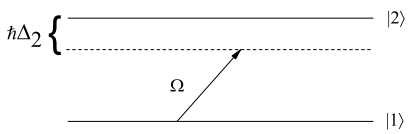
where we have defined $\hbar\Delta_1 \equiv E_1 - \hbar\dot{\xi}_1$ and $\hbar\Delta_2 \equiv E_2 - \hbar\dot{\xi}_2 = E_2 - E_1 + \hbar\Delta_1 - \hbar\omega$. We can then set the 0-point of our potential energy scale by setting $\Delta_1 = 0$.

Putting all of this together with Eq. (8) we finally obtain

$$\dot{\mathbf{c}} = \frac{-i}{2} \begin{pmatrix} 0 & \Omega \\ \Omega^* & 2\Delta_2 \end{pmatrix} \mathbf{c}, \tag{9}$$

with $\hbar\Delta_2 = E_2 - E_1 - \hbar\omega$. (Throughout these notes I have chosen to use the unfortunate convention that *red* detuning is *positive*, and *blue* detuning is *negative*.

The physical picture of this system is:



That is, $\hbar\Delta_2$ is the energy by which the laser is detuned from resonance. Because Ω is related to the electric field amplitude it is, in that sense, related to the laser intensity. However, since it is also related to the dipole matrix element coupling two states, the value of Ω for two different transitions are in general different, even if the two laser intensities are identical. Without going through the (trivial) derivation, Ω is related to more easily obtained system parameters by:

$$\Omega = \sqrt{\frac{3\lambda^3 I\gamma}{2\pi hc}} \text{ , or } \Omega[\text{MHz}] = 1.55\times10^{-7}\sqrt{\lambda^3[\text{nm}]I[\text{mW/cm}^2]\gamma[\text{s}^{-1}]},$$

where γ is the spontaneous decay rate from the upper state. For example, in the case of the rubidium 5s-5p transition, $\lambda = 780$ nm, and $1/\gamma = 26.6$ nsec. Then, for a laser intensity of 1.0 mW/cm^2, the Rabi frequency is 21 MHz. Note that Ω is an *angular* frequency.

Equation (9) is pretty simple, so let's try to solve it. Breaking up the matrix equation into its constituent parts, then taking the derivative of each of these we obtain:

$$\dot{c}_1 = \frac{-i}{2}\Omega c_2 \Rightarrow \ddot{c}_1 = \frac{-i}{2}\dot{c}_1 \tag{10a}$$

$$\dot{c}_2 = \frac{-i}{2}\left(\Omega^* c_1 + 2\Delta c_2\right) \Rightarrow \ddot{c}_2 = \frac{-i}{2}\left(\Omega^*\dot{c}_1 + 2\Delta\dot{c}_2\right). \tag{10b}$$

Combining these equations gives the single, second order differential equation:

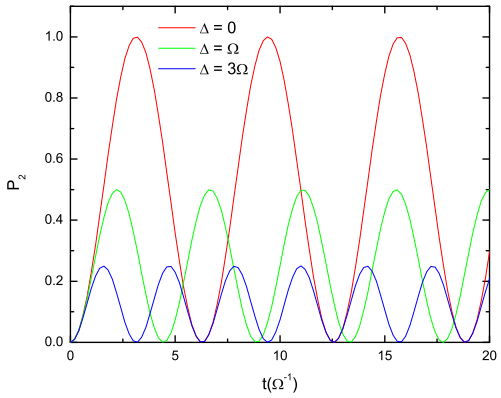
$$\ddot{c}_2 + i\Delta\dot{c}_2 + \frac{\Omega^2}{4}c_2 = 0.$$

With the boundary condition $c_2(0) = 0$, we can readily show that this has the solution

$$c_2 = -i\frac{\Omega}{\Omega'}\sin\frac{\Omega' t}{2}e^{-i\frac{\Delta t}{2}} \ , \ \ \Omega' \equiv \sqrt{\Omega^2 + \Delta^2}. \tag{11}$$

Now, the probability of finding the atom in state $|2\rangle$ is simply

$$P_2(t) = c_2^* c_2 = \left(\frac{\Omega}{\Omega'}\right)^2 \sin^2\frac{\Omega' t}{2} = \frac{\Omega^2}{2\left(\Omega^2 + \Delta^2\right)}\left(1 - \cos\Omega' t\right). \tag{12}$$



Plots of $P_2(t)$ are shown for a few different values of Δ.

Note that for $\Delta = 0$ (the case for the laser being exactly on resonance) the excited-state population oscillates between 0 and 1. That is, for some period of time, 100% of the population resides in the excited state. This is to be contrasted with the 50% result obtained using rate equations. The population oscillates with (angular) frequency Ω, which is known as the *Rabi frequency*. For non-zero detuning, the oscillations are faster, occurring at (angular) frequency Ω', a sort of generalized Rabi frequency. Also, if the detuning is non-zero, one never obtains 100% population inversion.

While for some systems of interest, particularly those involving microwave transitions, where the Rabi period is often short compared to the radiative lifetime, and where one can actually control the phase and timing of the coherent radiation to fractions of a Rabi period, one can use a "π-pulse" to efficiently transfer populations.

As a final note, if one were to take the time average of the resonant case, one would obtain the same 50% excited state fraction as we obtained using the incoherent approach from before - as one should expect.

Dressed State Approach

Consider the time-independent Schrödinger equation: $H\phi = \lambda\phi$, where λ and ϕ are the eigenvalue and eigenvector, respectively, of the Hamiltonian, H. If this Hamiltonian already contains the effects of the applied laser radiation, then the stationary state is said to be "dressed".

To obtain these "dressed states", expressed in terms of the "naked" or field-free basis, we

simply solve the eigenvalue equation: $H\vec{\phi} = \lambda\vec{\phi}$, or

$$\frac{\hbar}{2}\begin{pmatrix} 0 & \Omega \\ \Omega & 2\Delta \end{pmatrix}\begin{pmatrix} \phi_1 \\ \phi_2 \end{pmatrix} = \lambda\begin{pmatrix} \phi_1 \\ \phi_2 \end{pmatrix}.$$

Solving the characteristic equation

$$\begin{vmatrix} -\lambda & \Omega \\ \Omega & 2\Delta - \lambda \end{vmatrix} = 0 \Rightarrow \lambda'^2 - 2\Delta\lambda' - \Omega^2 = 0,$$

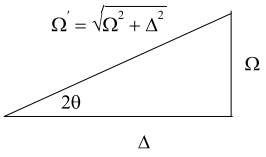
or

$$\lambda_\pm = \frac{\hbar}{2}\left(\Delta \pm \Omega'\right). \tag{13}$$

$$\text{Then, } \frac{\hbar}{2}\begin{pmatrix} 0 & \Omega \\ \Omega & 2\Delta \end{pmatrix}\begin{pmatrix} \phi_1 \\ \phi_2 \end{pmatrix} = \frac{\hbar}{2}\left(\Delta \pm \Omega'\right)\begin{pmatrix} \phi_1 \\ \phi_2 \end{pmatrix}$$

$$\text{From whence, } \phi_\pm = \phi_\pm^0\left(1, \frac{\Delta \pm \Omega'}{\Omega}\right).$$

We can re-parameterize this expression through the substitutions inspired from the following right triangle:



$$\text{Then, } \phi_\pm = \phi_\pm^0\left(1, \frac{\cos 2\theta \pm 1}{\sin 2\theta}\right)$$

Normalizing ϕ gives

$$\phi_+ = (\sin\theta, \cos\theta) \tag{14a}$$

$$\phi_- = (\cos\theta, -\sin\theta). \tag{14b}$$

That is,

$$\phi_+ = \psi_1\sin\theta + \psi_2\cos\theta \Rightarrow P_1^{(+)} = \sin^2\theta;\;\; P_2^{(+)} = \cos^2\theta \tag{15a}$$

$$\phi_- = \psi_1\cos\theta - \psi_2\sin\theta \Rightarrow P_1^{(-)} = \cos^2\theta;\;\; P_2^{(-)} = \sin^2\theta, \tag{15b}$$

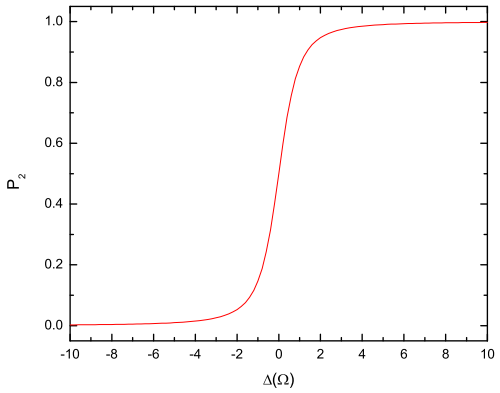
where ψ_1 and ψ_2 are the naked basis eigenvectors.

For example, suppose we start out with the laser greatly detuned to the blue: ($|\Delta| \gg \Omega, \Delta < 0$) or $2\theta \approx \pi \Rightarrow \theta \approx \pi/2$. Also, suppose that at $t = 0$, $P_1 = 1$, and $P_2 = 0$. This corresponds to the ϕ_+ state, since $\sin^2(\pi/2) = 1$ and $\cos^2(\pi/2) = 0$.

If we then *adiabatically* allow Δ to move from greatly blue-detuned to greatly red-detuned, passing through 0-detuning along the way, we will still be in the ϕ_+ state (which is what we really meant by "adiabatically") but now we have ($|\Delta| \gg \Omega, \Delta > 0$) or $2\theta \approx 0$. So, $P_1 = \sin^2(0) = 0$, and $P_2 = \cos^2(0) = 1$. In other words, neglecting spontaneous emission, we have forced *all* of the population into the excited state – and without the need for micro-control of the phase and timing of the radiation.

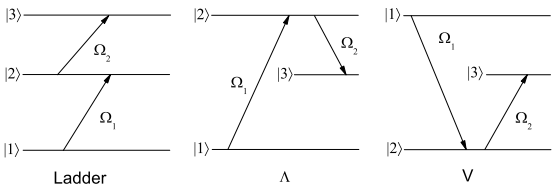
Note that if we had started out with a red-detuned laser and "chirped" to the blue, we would have had the same result, but using the ϕ_- state. This process is known by the seemingly oxymoronic phrase "adiabatic rapid passage", or ARP.

The following is a plot of P_2 versus detuning for a case of ARP. The detuning, measured in units of Rabi frequency, varies linearly in time from -10Ω to $+10\Omega$.



III. COHERENCE: 3-LEVEL SYSTEMS

Let's now try to apply the same sort of formalism to 3-level systems. Unlike the 2-level system, the 3-level system has 3 different basic configurations:



All three configurations have the property that levels 1 and 2 are connected by dipole matrix elements, levels 2 and 3 are connected by dipole matrix elements, and levels 1 and 3 are *not* connected by dipole matrix elements. As was the case with the 2-level system, we shall assume, for the time being, that we can neglect spontaneous emission from any of

the levels. We shall also assume that there is no "leakage" into any other state. That is, probability is conserved within the 3 levels.

In analogy with the 2-level system, we express the electric field of the applied radiation as

$$\vec{E}(t) = Re\left(\hat{\varepsilon}_1 E_1 e^{-i\omega_1 t} + \hat{\varepsilon}_2 E_2 e^{-i\omega_2 t}\right) = \frac{1}{2}\left[\hat{\varepsilon}_1 E_1\left(e^{-i\omega_1 t} + e^{i\omega_1 t}\right) + \hat{\varepsilon}_2 E_2\left(e^{-i\omega_2 t} + e^{i\omega_2 t}\right)\right].$$

The ε_n indicate the polarization of the two electric fields and, as before, $\Psi(t) = \sum_n c_n(t)\psi_n e^{-i\xi_n(t)}$. Also as before, plugging $\Psi(t)$ into the time-dependent Schrödinger equation, we obtain again Eq. (5) above,

$$\hbar\dot{c}_l = -i\left[\left(E_l^0 - \hbar\dot{\xi}_l\right)c_l + \sum_n c_n V_{ln} e^{-i(\xi_n - \xi_l)}\right].$$

Now, $V_{mn} = 0$ if $|n-m| > 1$ since, by hypothesis, levels 1 and 3 are not connected. As before, we approximate the infinite sums by truncating the number of terms to 3, the number levels in the system. We then obtain the following three equations:

$$\begin{aligned}
\hbar\dot{c}_1 &= -i\left[\left(E_1^0 - \hbar\dot{\xi}_1 + V_{11}\right)c_1 + V_{12}c_2 e^{-i(\xi_2-\xi_1)}\right] \\
\hbar\dot{c}_2 &= -i\left[\left(E_2^0 - \hbar\dot{\xi}_2 + V_{22}\right)c_2 + V_{12}c_1 e^{-i(\xi_1-\xi_2)} + V_{32}c_3 e^{-i(\xi_3-\xi_2)}\right] \\
\hbar\dot{c}_3 &= -i\left[\left(E_3^0 - \hbar\dot{\xi}_3 + V_{33}\right)c_3 + V_{23}c_2 e^{-i(\xi_2-\xi_3)}\right].
\end{aligned}$$

Or, in matrix form:

$$\hbar\dot{\mathbf{c}} = -i\begin{pmatrix} E_1 - \hbar\dot{\xi}_1 & V_{12}e^{-i(\xi_2-\xi_1)} & 0 \\ V_{12}^* e^{i(\xi_2-\xi_1)} & E_2 - \hbar\dot{\xi}_2 & V_{23}e^{-i(\xi_3-\xi_2)} \\ 0 & V_{23}^* e^{i(\xi_3-\xi_2)} & E_3 - \hbar\dot{\xi}_3 \end{pmatrix}\mathbf{c}, \tag{16}$$

where, as before, $E_n = E_n^0 + V_{nn}$.

We define the Rabi frequencies as before,

$$\Omega_1 = \frac{-eE_1}{\hbar}\langle 1|r|2\rangle \Rightarrow V_{12} = \frac{\hbar\Omega_1}{2}\left(e^{-i\omega_1 t} + e^{+i\omega_1 t}\right)$$

and

$$\Omega_2 = \frac{-eE_2}{\hbar}\langle 2|r|3\rangle \Rightarrow V_{23} = \frac{\hbar\Omega_2}{2}\left(e^{-i\omega_2 t} + e^{+i\omega_2 t}\right)$$

With the 3-level systems, the most convenient choice of phases depends on which of the three systems, ladder, Λ , or V, we are trying to describe. For the *ladder* system, we choose

$$\begin{aligned}
\xi_2 - \xi_1 &= \omega_1 t \Rightarrow \dot{\xi}_2 = \dot{\xi}_1 + \omega_1, \\
\xi_3 - \xi_2 &= \omega_2 t \Rightarrow \dot{\xi}_3 = \dot{\xi}_2 + \omega_2.
\end{aligned}$$

For the Λ system, we choose

$$\begin{aligned}\xi_2 - \xi_1 &= \omega_1 t \Rightarrow \dot{\xi}_2 = \dot{\xi}_1 + \omega_1, \\ \xi_3 - \xi_2 &= -\omega_2 t \Rightarrow \dot{\xi}_3 = \dot{\xi}_2 - \omega_2.\end{aligned}$$

For the V system, we choose

$$\begin{aligned}\xi_2 - \xi_1 &= -\omega_1 t \Rightarrow \dot{\xi}_2 = \dot{\xi}_1 - \omega_1, \\ \xi_3 - \xi_2 &= \omega_2 t \Rightarrow \dot{\xi}_3 = \dot{\xi}_2 + \omega_2.\end{aligned}$$

Here, we concentrate on the ladder system. Then, applying that choice of phase and applying the rotating wave approximation, we obtain

$$\hbar\dot{\mathbf{c}} = -i \begin{pmatrix} E_1 - \hbar\dot{\xi}_1 & \frac{\hbar\Omega_1}{2} & 0 \\ \frac{\hbar\Omega_1^*}{2} & E_2 - \hbar\dot{\xi}_1 - \hbar\omega_1 & \frac{\hbar\Omega_2}{2} \\ 0 & \frac{\hbar\Omega_2^*}{2} & E_3 - \hbar\dot{\xi}_1 - \hbar\omega_1 - \hbar\omega_2 \end{pmatrix} \mathbf{c},$$

where now the RWA has allowed us to set terms like $e^{\pm 2i\omega_1 t}$, $e^{\pm 2i\omega_2 t}$, and $e^{\pm i(\omega_1 \pm \omega_2)t}$ all approximately equal to 0. Note that in this last term, ω_1 and ω_2 must differ by many Rabi frequencies for this approximation to be valid. Therefore, the RWA is not strictly valid for 3 equally spaced levels.

We can once again define $\hbar\Delta_1 \equiv E_1 - \hbar\dot{\xi}_1$. Then we obtain,

$$\hbar\dot{\mathbf{c}} = -i \begin{pmatrix} \hbar\Delta_1 & \frac{\hbar\Omega_1}{2} & 0 \\ \frac{\hbar\Omega_1^*}{2} & E_2 - E_1 + \hbar\Delta_1 - \hbar\omega_1 & \frac{\hbar\Omega_2}{2} \\ 0 & \frac{\hbar\Omega_2^*}{2} & E_3 - E_1 + \hbar\Delta_1 - \hbar\omega_1 - \hbar\omega_2 \end{pmatrix} \mathbf{c}.$$

We can arbitrarily set the zero-point of our energy to be $\hbar\Delta_1$, giving us our final result:

$$\dot{\mathbf{c}} = -\frac{i}{2} \begin{pmatrix} 0 & \Omega_1 & 0 \\ \Omega_1^* & 2\Delta_2 & \Omega_2 \\ 0 & \Omega_2^* & 2\Delta_3 \end{pmatrix} \mathbf{c} \tag{17}$$

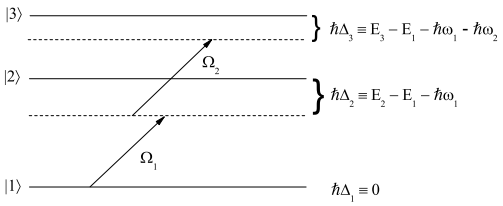
where we have defined

$$\hbar\Delta_2 \equiv E_2 - E_1 - \hbar\omega_1 \tag{18a}$$

and

$$\hbar\Delta_3 \equiv E_3 - E_1 - \hbar\omega_1 - \hbar\omega_2. \tag{18b}$$

Re-drawing the ladder diagram, we can see that the diagonal terms of the Hamiltonian indicate the detuning of the exciting lasers from resonance:



A. Special Case I: $\Omega_1 = \Omega_2 \equiv \Omega$, $\Delta_2 = \Delta_3 = 0$, (exact resonance)

Then,

$$H = \frac{\hbar}{2}\begin{pmatrix} 0 & \Omega & 0 \\ \Omega & 0 & \Omega \\ 0 & \Omega & 0 \end{pmatrix}.$$

Solving the eigen-equation gives us 3 eigenvalues, $\lambda_0 = 0$, and $\lambda_\pm = \pm\frac{\hbar\sqrt{2}\Omega}{2}$. The corresponding (normalized) eigenvectors are:

$$\Phi_- = \frac{1}{2}\begin{pmatrix} 1 \\ -\sqrt{2} \\ 1 \end{pmatrix}; \Phi_0 = \frac{\sqrt{2}}{2}\begin{pmatrix} 1 \\ 0 \\ -1 \end{pmatrix}; \Phi_+ = \frac{1}{2}\begin{pmatrix} 1 \\ \sqrt{2} \\ 1 \end{pmatrix}.$$

HW2: Verify that these eigenvalues and eigenvectors satisfy the eigenequation.

In the dressed state picture,

$$\Psi(t) = \left[k_-\Phi_- e^{-i\lambda_- t} + k_0\Phi_0 + k_+\Phi_+ e^{-i\lambda_+ t}\right],$$

where $k_-^2 + k_0^2 + k_+^2 = 1$, and initial conditions determine the choice of k_-, k_0, and k_+. For example, if $\Psi(t=0) = \psi_1$, then

$$\psi_1 = \frac{1}{2}\left[k_-\left(\psi_1 - \sqrt{2}\psi_2 + \psi_3\right) + k_0\sqrt{2}\left(\psi_1 - \psi_3\right) + k_+\left(\psi_1 + \sqrt{2}\psi_2 + \psi_3\right)\right]$$

or,

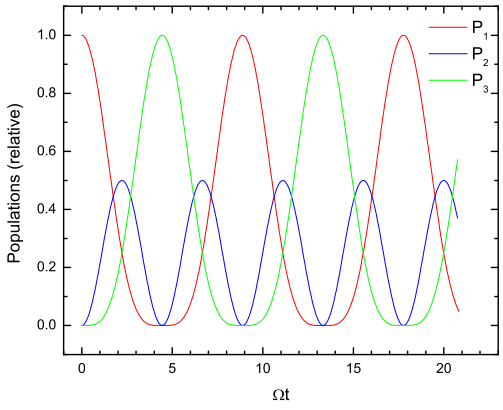
$$\psi_1\left(\frac{k_-}{2}+\frac{k_0}{\sqrt{2}}+\frac{k_+}{2}-1\right)=0$$
$$\psi_2\left(-\frac{k_-}{\sqrt{2}}+\frac{k_+}{\sqrt{2}}\right)=0$$
$$\psi_3\left(\frac{k_-}{2}-\frac{k_0}{\sqrt{2}}+\frac{k_+}{2}\right)=0.$$

From these equations we find $k_- = k_+ = 1/2$; $k_0 = \sqrt{2}/2$. So that

$$\begin{aligned}
\Psi(t) &= \frac{1}{2}\left[\Phi_- e^{-i\lambda_- t}+\sqrt{2}\Phi_0+\Phi_+ e^{-i\lambda_+ t}\right]\\
&= \frac{1}{4}\left[\left(\psi_1-\sqrt{2}\psi_2+\psi_3\right)e^{+i\frac{\sqrt{2}\Omega t}{2}}+2\left(\psi_1-\psi_3\right)+\left(\psi_1+\sqrt{2}\psi_2+\psi_3\right)e^{-i\frac{\sqrt{2}\Omega t}{2}}\right]\\
&= \frac{1}{2}\left[\psi_1\left(\frac{e^{+i\frac{\sqrt{2}\Omega t}{2}}+e^{-i\frac{\sqrt{2}\Omega t}{2}}}{2}+1\right)+\psi_3\left(\frac{e^{+i\frac{\sqrt{2}\Omega t}{2}}+e^{-i\frac{\sqrt{2}\Omega t}{2}}}{2}-1\right)-i\sqrt{2}\psi_2\left(\frac{e^{+i\frac{\sqrt{2}\Omega t}{2}}-e^{-i\frac{\sqrt{2}\Omega t}{2}}}{2i}\right)\right]\\
&= \frac{1}{2}\left[\psi_1\left(\cos\frac{\sqrt{2}\Omega t}{2}+1\right)+\psi_3\left(\cos\frac{\sqrt{2}\Omega t}{2}-1\right)-\psi_2 i\sqrt{2}\sin\frac{\sqrt{2}\Omega t}{2}\right]\\
&= \psi_1\cos^2\frac{\sqrt{2}\Omega t}{4}-\psi_3\sin^2\frac{\sqrt{2}\Omega t}{4}-\frac{i}{\sqrt{2}}\psi_2\sin\frac{\sqrt{2}\Omega t}{2}.
\end{aligned}$$

Therefore,

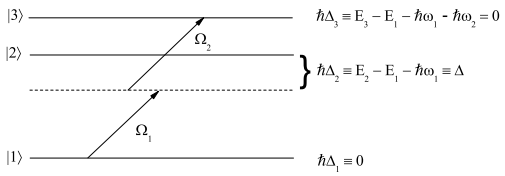
$$P_1=\cos^4\frac{\sqrt{2}\Omega t}{4}\ ;\ \ P_3=\sin^4\frac{\sqrt{2}\Omega t}{4}\ ;\ \ P_2=\frac{1}{2}\sin^2\frac{\sqrt{2}\Omega t}{2}.$$



These results are shown in the above figure where we plot the relative populations of the 3 levels as a function of time, with time in units of the Rabi period, Ω^{-1}. You can see both from the equations and from the figure that the oscillation frequency for P_2 is twice that for P_1 and P_3. This is because level 2 gets has "source terms" during both the excitation and de-excitation of P_3.

HW3: Calculate the population probabilities as a function of time (for this same degenerate case) and for the initial condition $\Psi(t=0)=\psi_1$, but for $\Omega_1 \neq \Omega_2$. For example, set $\Omega_2 = b\Omega_1$, and plot the populations versus time for $b = 0.5$, 0, and 2.0.

B. Special Case II: $\Omega_1 \neq \Omega_2$; $\Delta_1 = \Delta_3 = 0$; $\Delta_2 \equiv \Delta$. (2-photon resonance)



The relevant Hamiltonian is

$$H = \frac{\hbar}{2}\begin{pmatrix} 0 & \Omega_1 & 0 \\ \Omega_1^* & 2\Delta & \Omega_2 \\ 0 & \Omega_2^* & 0 \end{pmatrix}.$$

The characteristic equation is then

$$\begin{vmatrix} -\lambda' & \Omega_1 & 0 \\ \Omega_1^* & 2\Delta - \lambda' & \Omega_2 \\ 0 & \Omega_2^* & -\lambda' \end{vmatrix} = 0 \text{ , or } \lambda'\left[\lambda'^2 - 2\Delta\lambda' - \left(\Omega_1^2 + \Omega_2^2\right)\right] = 0,$$

with solutions

$$\lambda_0 = 0\ ,\ \ \lambda_\pm = \frac{\hbar}{2}\left(\Delta \pm \sqrt{\Delta^2 + \Omega_1^2 + \Omega_2^2}\right).$$

The *unnormalized* eigenvectors are then given by:

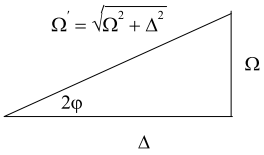
$$\Phi_- = \begin{pmatrix} \Omega_1 \\ \Delta - \sqrt{\Delta^2 + \Omega^2} \\ \Omega_2 \end{pmatrix};\quad \Phi_0 = \begin{pmatrix} -\Omega_2 \\ 0 \\ \Omega_1 \end{pmatrix};\quad \Phi_+ = \begin{pmatrix} \Omega_1 \\ \Delta + \sqrt{\Delta^2 + \Omega^2} \\ 1 \end{pmatrix},$$

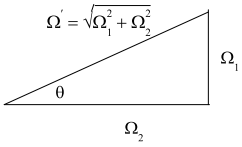
with $\Omega^2 \equiv \Omega_1^2 + \Omega_2^2$.

As in the 2-level system, we next define "rotation" variables in order to more simply characterize the system. For ϕ and θ, respectively:

With these definitions, the eigenvalues with their corresponding normalized eigenvectors are:

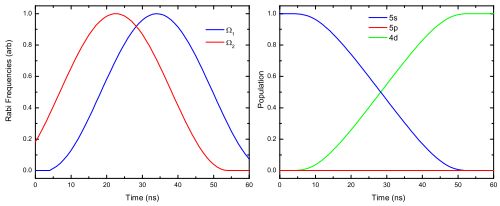
$$\lambda_- = \frac{\hbar}{2}\Omega\tan\phi,\quad \lambda_0 = 0,\quad \lambda_+ = \frac{\hbar}{2}\text{ctn }\phi,\ \text{and}$$





$$\Phi_- = \begin{pmatrix} \sin\theta\cos\phi \\ -\sin\phi \\ \cos\theta\cos\phi \end{pmatrix}, \quad \Phi_0 = \begin{pmatrix} -\cos\theta \\ 0 \\ \sin\theta \end{pmatrix}, \quad \Phi_+ = \begin{pmatrix} \sin\theta\sin\phi \\ \cos\phi \\ \cos\theta\sin\phi \end{pmatrix}.$$

Suppose we start out at $t \to -\infty$ with $\Omega_2 \gg \Omega_1$, which means $\theta \to 0$. Suppose we also require that at $t \to -\infty$, all of the population is in ψ_1. Then, we see that these two conditions also require that all of the population be in the single dressed state Φ_0. Now, if we *adiabatically* increase Ω_1 and decrease Ω_2, we will stay in Φ_0, but the entire population will have moved into ψ_3 as $\Omega_2 \ll \Omega_1$. Note also that because we stay at all times in Φ_0, and because Φ_0 never has any component of ϕ_2, *no population is ever in* ψ_2. In the following two figures, we plot the Rabi frequencies Ω_1 and Ω_2, and the relative populations of $|1\rangle$ and $|3\rangle$ versus time.



Thus, quantum mechanics somehow, "magically" allows us to transfer population from $|1\rangle$ to $|3\rangle$, with absolutely no population at any time in $|2\rangle$, in spite of the fact that there is no direct coupling between states $|1\rangle$ and $|3\rangle$, but strong coupling between both of these and $|2\rangle$! We will now try to, at least qualitatively understand the physics behind this "magic".

Consider a 3-level system in which $\Omega_2 \gg \Omega_1$. Let the Ω_1-field have angular frequency ω, and the Ω_2-field have angular frequency ω_2. For the moment, assume these fields are static. Furthermore, for simplicity we will require ω_2 to resonantly couple $|2\rangle$ and $|3\rangle$, while we will allow ω to slowly vary with time. Now, because the field associated with the $|1\rangle$ to $|2\rangle$ transition is weak, while the other is strong, we can consider the state of the system to be well described by $\Psi(t) = \psi_1 c_1 e^{-i\omega t} + \Phi_- B_-(t) + \Phi_+ B_+(t)$. That is, we "dress" the upper states, leaving the lowest state "undressed" or "naked".

Then, the RWA Hamiltonian describing this system is given by

$$H(t) = \frac{\hbar}{2} \begin{pmatrix} 2\Delta & \Omega_- & \Omega_+ \\ \Omega_-^* & -2\epsilon & 0 \\ \Omega_+^* & 0 & +2\epsilon \end{pmatrix}.$$

Recall from our treatment of 2-level systems that the 2 states in the "naked" basis, separated by an energy $E_2^0 - E_1^0$ are, in the "dressed" basis separated by $\lambda_+ - \lambda_-$. Furthermore, they are centered about the "0-point" of the potential, and that this 0-point was simply E_1. Thus, in the 3-level system, the dressed states are situated symmetrically about E_2, and are separated by an energy $\hbar\Omega_2$. If we were to scan ω, and measure the absorption of this light as a function of ω, we would find absorption structure at 2 values of ω, rather than the single absorption line of the naked system. This dual structure is known as Autler-Townes splitting and was discovered by Autler and Townes in 1955.

So now let us fix ω so that it would be resonant with the naked basis (placing $E_1 + \hbar\omega$ right at the mid-point of λ_1 and λ_-. In the naked basis, we would now be exactly 2-photon resonant with $|1\rangle \rightarrow |2\rangle \rightarrow |3\rangle$ excitation. (Actually, we'd be better off if we slightly detuned ω to, say, the red, and detuned ω_2 by the corresponding amount to the blue. But this is a detail. The key is to be 2-photon resonant.) Now we can see that direct excitation of $|3\rangle$ is possible since we can excite both Φ_1 and Φ_2, each of which has some $|3\rangle$ "character". It turns out that if ω really does go to the half-way point between the dressed states, destructive interference takes place which completely wipes out any population of $|2\rangle$. At this frequency, there is absolutely no absorption of the light associated with ω. That is, the material becomes completely transparent to the light associated with ω. This condition is referred to as "Electromagnetically-Induced Transparency" or EIT, and is currently a hot area of research due to its potential in quantum information applications.

Let's consider a step-by-step analysis (still quantitative) of how this three-level ARP process works.

1. Only Ω_2 is present, dressing the system (except for $|1\rangle$). For now, Ω_1 is negligible.

2. Now the light associated with Ω_1 begins to become important. This light is "scattered" off the dressed structure, populating $|3\rangle$, as described above.

3. Now the light associated with Ω_2 begins to drop a bit in intensity, while Ω_1 continues to grow in intensity. Here the population of $|3\rangle$ really "kicks in".

4. The population is now almost completely in $|3\rangle$. The intensity of Ω_2 is really dropping, and Ω_1 is maximal. So why does the population not revert back to the ground state? The answer is that there is now nearly no mixing of $|2\rangle$ and $|3\rangle$ going on, and so the light associated with ω can't "talk" to level 3.

5. Finally, the light associated with both lasers is off. In the absence of spontaneous emission, all the population is trapped in level 3.

IV. DENSITY MATRIX TREATMENT OF COHERENT EXCITATION

I won't go into much background here. I will only define what we mean by a density matrix, show that the formalism is equivalent to the time-dependent Schrödinger equation approach, and then, with *no justification at all* present you with the prescription for setting up the "equations of motion" for the system undergoing coherent excitation – but including, for the first time here, spontaneous emission.

First of all, the density matrix, ρ, is defined as $\rho_{ij} = |\psi_i\rangle\langle\psi_j|$. Thus, for a 2-level system, we can write the density matrix as:

$$\rho = \begin{pmatrix} \rho_{11} & \rho_{12} \\ \rho_{21} & \rho_{22} \end{pmatrix}.$$

Now, the density matrix is Hermitian, meaning that $\rho_{ij} = \rho^*_{ji}$. Therefore, the diagonal elements of the density matrix are real, and in fact represent the populations of the respective levels. Thus, ρ_{11} is the population of level 1; ρ_{22} is the population of level 2, etc. For a

conservative system then, $Tr[\rho] = 1$ (assuming the total population is defined as 1). Note that since

$$\rho \equiv |\psi\rangle\langle\psi|,$$
$$i\hbar\frac{d\rho}{dt} = i\hbar\frac{d|\psi\rangle}{dt}\langle\psi| + i\hbar|\psi\rangle\frac{d\langle\psi|}{dt}.$$

But, the TDSE is

$$i\hbar\frac{d|\psi\rangle}{dt} = H|\psi\rangle \ , \ \text{and} - i\hbar\frac{\langle\psi|}{dt} = (H|\psi\rangle)^\dagger = \langle\psi|H^\dagger.$$

Then,

$$i\hbar\frac{d\rho}{dt} = H|\psi\rangle\langle\psi| \ - \ |\psi\rangle\langle\psi|H^\dagger.$$

or,

$$i\hbar\dot{\rho} = H\rho \ - \ \rho H^\dagger \equiv [H, \rho]\,.$$

This is the "equation of motion", or "equation of evolution" for the density matrix.

We can therefore simply solve this equation (or more correctly this system of N equations for an N-level system) and from the diagonal elements of ρ, write down our populations. (These are the optical Bloch equations you may have heard of.)

Now, with no derivation or justification at all I present a modification of this equation of evolution, in which we finally include spontaneous emission. (For details, I refer you to Shore's books, listed in the references.)

$$i\hbar\dot{\rho}_{ij} = [H, \rho]_{ij} - i\hbar\,[\Gamma\rho]_{ij} \ , \ \text{with}\,[\Gamma\rho]_{ij} \equiv \rho_{ij}\sum_k \frac{1}{2}\,(\gamma_{ik} + \gamma jk) - \delta_{ij}\sum_k \rho_{kk}\gamma_{ki}.$$

The γ's here are simply the decay rates or the Einstein A-coefficients (that we have until now been designating by "A". The use of A is usual in computations using incoherent sources of light, whereas γ is more commonly used in coherent excitation formulism. In fact, the two symbols are interchangeable.) In order to simplify the notation, we define $\gamma_{lm} = 0$ for $l \leq m$. Now at last we've got an expression than can truly simulate the "real world" situation in which spontaneous emission can not always be neglected. Let's try applying this expression to the simple 2-level system – including for the first time, realistic decay rates.

Our Hamiltonian and density matrices are:

$$H = \frac{\hbar}{2}\begin{pmatrix} 0 & \Omega \\ \Omega & 2\Delta \end{pmatrix} \ \text{and} \ \rho = \begin{pmatrix} \rho_{11} & \rho_{12} \\ \rho_{21} & \rho_{22} \end{pmatrix}.$$

where we choose our electric field phases such that the Rabi frequency is pure real. Then, the commutator is:

$$[H,\rho] = \frac{\hbar}{2}\begin{pmatrix} \Omega\rho_{21} - \Omega\rho_{12} & \Omega\rho_{22} - \Omega\rho_{11} - 2\Delta\rho_{12} \\ -\Omega\rho_{22} + \Omega\rho_{11} + 2\Delta\rho_{21} & \Omega\rho_{12} - \Omega\rho_{21} \end{pmatrix},$$

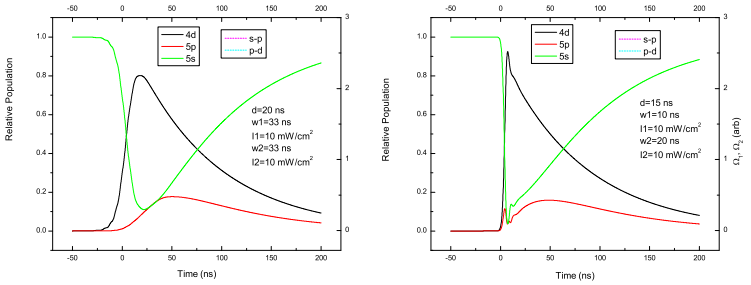
while

$$\Gamma\rho = \begin{pmatrix} -\rho_{22}\gamma_{21} & \rho_{12}\gamma_{21}/2 \\ -\rho_{21}\gamma_{21}/2 & \rho_{22}\gamma_{21} \end{pmatrix}.$$

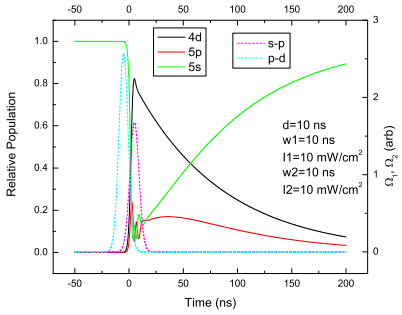
Now, because ρ is Hermitian, $\rho_{ij} = \rho^*_{ji}$. Furthermore, in general for any complex number $z = a + ib$, with a and b real, $z - z^* = 2ib$. Therefore, defining $\rho_{ij} = \rho^a_{ij} + i\rho^b_{ij}$, with ρ^a_{ij} and ρ^b_{ij} pure real for all values of i, j, we obtain:

$$\dot{\rho} = \begin{pmatrix} \Omega\rho^b_{12} + \rho_{22}\gamma_{21} & -i\left(\Omega\rho_{22} - \Omega\rho_{11} - 2\Delta\rho_{12}\right)/2 - \rho_{12}\gamma_{21}/2 \\ cc & \Omega\rho^b_{12} - \rho_{22}\gamma_{21} \end{pmatrix}.$$

where "cc" denotes complex conjugate of the 1,2 element. We are solving for the four elements of ρ. Because it is Hermitian, knowledge of ρ_{12} tells us ρ_{21}. Furthermore, the diagonal elements must be pure real, and must sum to 1. Therefore, this expression represents 3 coupled, first order differential equations. In general, for an N-level system, you will have to set up and solve $N^2 - 1$ coupled, first order differential equations. The following figures are examples of calculations carried out for a realistic 3-level system. In this case, the system of interest is 2-photon interaction with the 5s, 5p, and 4d levels of atomic rubidium. In the smorgasbord of figures, I have mainly just varied the pulse width, and relative timing of the two lasers, as indicated on the figures.



As these figures show, it is possible to place more than 90% of the atoms in the 4d state. Note that unlike our idealized situation in which we neglected the effect of spontaneous



emission, the intermediate state, here the 5p, does get populated. This is seen to be essentially exclusively from spontaneous emission from the 4d level, rather than due to direct excitation from the 5s.

This concludes the "theory" part of the lectures. Next we will consider experimental efforts to realize these results. We will also discuss applications. None of this will, however, appear in lecture notes.

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