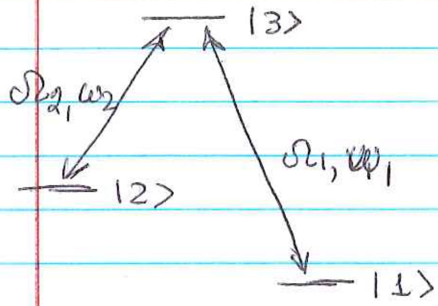
Classification of multy-photon processes

1. Incoherent interactions: atomic coherences (off-diagonal elements of the density matrix) are not important
   Examples: saturation spectroscopy

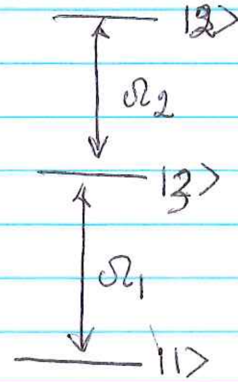
2. Coherent interactions: atomic coherences are important, and two or more photons are required to generate them
   Examples: two-photon Raman transitions, electromagnetically induced transparency

3. Parametric interactions: no energy exchange between light and atom, but atoms ~~do~~ mediate interactions between multiple light fields
   Examples: four-wave mixing, parametric down conversion, second harmonic generation

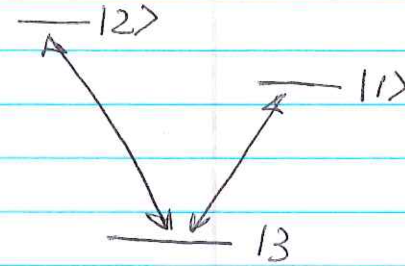
Two-photon Raman transitions



Λ - system



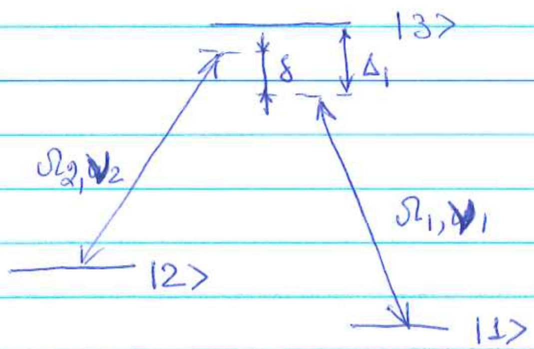
Cascade, or ladder system



V system

Common properties: optical transitions b/w states 1 & 3 and 2 & 3 are allowed, but transitions b/w 1 & 2 are forbidden

Let's concentrate on a Λ-system



long-lived ground states

Δ_1 - one photon detuning $\quad \Delta_1 = \nu_1 - \omega_{13}$

$\Delta_2 = \nu_2 - \omega_{23}$

δ - two photon detuning $\quad \delta = \Delta_1 - \Delta_2 = (\nu_1 - \nu_2) - (\omega_{13} - \omega_{23})$

$= (\nu_1 - \nu_2) - \omega_{12}$

Closed three-level system

$$|\psi(t)\rangle = c_1(t)|1\rangle + c_2(t)|2\rangle + c_3(t)|3\rangle$$

Hamiltonian

$$\hat{H} = \hbar\omega_1 |1\rangle\langle 1| + \hbar\omega_2 |2\rangle\langle 2| + \hbar\omega_3 |3\rangle\langle 3| -$$
$$- \frac{\hbar\Omega_1}{2}|3\rangle\langle 1| - \frac{\hbar\Omega_1^*}{2}|1\rangle\langle 3| - \frac{\hbar\Omega_2}{2}|3\rangle\langle 2| - \frac{\hbar\Omega_2^*}{2}|2\rangle\langle 3|$$

Rotating wave approximation $\Omega_{1,2} \to \Omega_{1,2} e^{-i\nu_{1,2} t}$

$|3\rangle \to |3\rangle e^{-i\omega_{13} t}$

$\omega_1 = 0$ $\qquad |2\rangle \to |2\rangle e^{-i\omega_{12} t}$

$$\hat{H}_{RWA} = -\hbar\Delta_1 |3\rangle\langle 3| - \hbar\delta |2\rangle\langle 2| - \frac{\hbar}{2}(\Omega_1 |3\rangle\langle 1| + \Omega_1^* |1\rangle\langle 3|) -$$
$$- \frac{\hbar}{2}(\Omega_2 |3\rangle\langle 2| + \Omega_2^* |2\rangle\langle 3|)$$

Shrodinger equation $i\hbar \frac{\partial \psi}{\partial t} = \hat{H}\psi$

$$\begin{cases} \dot{c}_1 = \frac{i}{2}\Omega_1^* c_3 \\ \dot{c}_2 = i\delta c_2 + \frac{1}{2} i \Omega_2^* c_3 \\ \dot{c}_3 = i\Delta_1 c_3 + \frac{i}{2}\Omega_1 c_1 + \frac{i}{2}\Omega_2 c_2 \end{cases}$$

One situation when we can neglect decays is a far-detuned case $\Delta_1 \gg \Omega_1, \Omega_2, \gamma_3, \delta$

(then $\Delta_1 \simeq \Delta_2$)

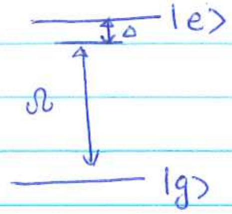
Small parameter $\frac{\Omega_{1,2}}{\Delta}$ and $\frac{\delta}{\Delta}$

Then $c_3^{(0)} = - \frac{\Omega_1 c_1 + \Omega_2 c_2}{2\Delta}$

$$\dot{c}_1 = -i\frac{|\Omega_1|^2}{4\Delta} c_1 - i\frac{\Omega_2 \Omega_1^*}{4\Delta} c_2$$

$$\dot{c}_2 = i\left(\delta - \frac{|\Omega_2|^2}{4\Delta}\right) c_2 - i\frac{\Omega_1 \Omega_2^*}{4\Delta} c_1$$

Compare these equations to those for a Two-level system



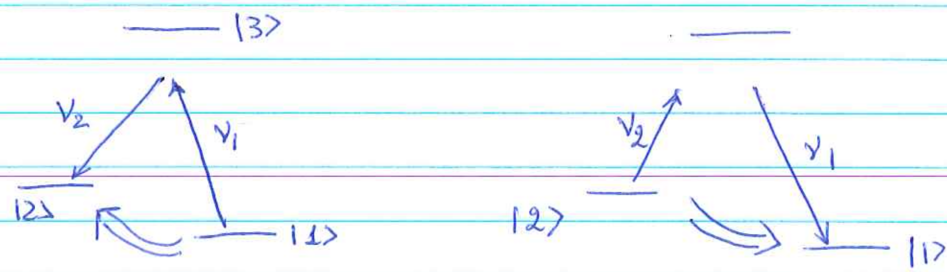
$$\dot{c}_g(t) = -\frac{i\Omega}{2}\, c_e$$

$$\dot{c}_e(t) = \underbrace{i\Delta c_e}_{\text{detuning}} - \frac{i\Omega}{2}\, c_g$$

Ω – Rabi frequency

Our far-detuned three-level system effectively behaves as a two level system, Rabi-flopping between states $|1\rangle$ and $|2\rangle$ with an effective two-photon Rabi ~~fre~~ frequency $\Omega_{eff} = \frac{\Omega_2^* \Omega_1}{\Delta}$

Two-photon process

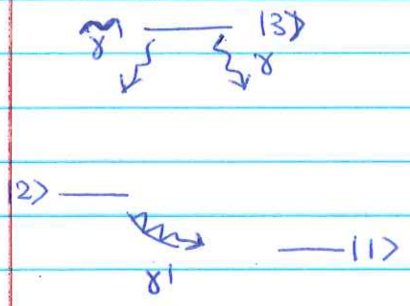


coherent transfer

two-photon Rabi flopping

Atoms are not excited into state $|3\rangle$, but are transfered between states $|1\rangle$ and $|2\rangle$ via a two-photon process

To include decoherence and decay processes we have to go to the density matrix approach



γ – population decay $3 \to 1$
$\tilde{\gamma}$ – " $2 \to 1$
γ' – " $3 \to 2$

$\gamma_{12}, \gamma_{23}, \gamma_{13}$ – decoherence rates

$$\dot{\rho}_{11} = \gamma\rho_{33} + \gamma'\rho_{22} - \frac{i}{2}\Omega_1\rho_{13} + \frac{i}{2}\Omega_1^*\rho_{31}$$
$$\dot{\rho}_{22} = \tilde{\gamma}\rho_{33} - \gamma'\rho_{22} - \frac{i}{2}\Omega_2\rho_{23} + \frac{i}{2}\Omega_2^*\rho_{32}$$

$$\dot{\rho}_{21} = -\overbrace{(\gamma_{12} - i\delta)}^{\Gamma_{12}}\rho_{21} + \frac{1}{2}i\Omega_2^*\rho_{31} - \frac{1}{2}i\Omega_1\rho_{23}$$

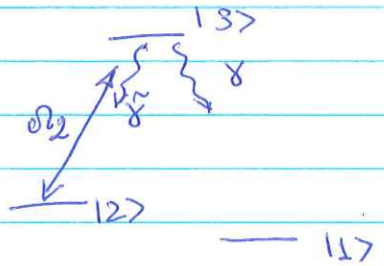
$$\dot{\rho}_{31} = -\overbrace{(\gamma_{13} - i\Delta)}^{\Gamma_{13}}\rho_{31} + \frac{i\Omega_2}{2}\rho_{21} + \frac{i}{2}\Omega_1(\rho_{11} - \rho_{33})$$
$$\dot{\rho}_{32} = -\underbrace{(\gamma_{23} - i(\Delta - \delta))}_{\Gamma_{23}}\rho_{32} + \frac{i}{2}\Omega_2(\rho_{22} - \rho_{33}) + \frac{i\Omega_1}{2}\rho_{12}$$

$$\rho_{11} + \rho_{22} + \rho_3 = 1$$

Rather cumbersome (yet solvable) system of equations $\longrightarrow$ only numerical in general case, even for the steady state!

Special case #1 $\rightarrow$ far-detuned case $\Delta \gg \Omega_1, \Omega_2, \gamma_{ij}, \delta$, and $\Omega_1 \gg \Omega_2, \gamma_{ij}, \delta$

Polarizations $\propto$ field amplitude, so for the first-order effect in Ω_1 we need to know populations, as affected by Ω_2 only



$\rho_{11} = 1$

$\rho_{22} = \rho_{33} = 0$

$\rho_{23} = 0$ (b/w empty levels)

Then we can easily find the remaining density matrix elements

$$\begin{cases} -\Gamma_{12}\rho_{21} + \frac{i}{2}\Omega_2^*\rho_{31} = 0 \\ -\Gamma_{13}\rho_{31} + \frac{i}{2}\Omega_2\rho_{21} + \frac{i\Omega_1}{2} = 0 \end{cases}$$

$$\rho_{21} = -\frac{\Omega_1\Omega_2^*}{4\Gamma_{12}\Gamma_{13} + |\Omega_2|^2}$$ ground-state coherence

$$\rho_{31} = i\Omega_1 \frac{2\Gamma_{12}}{4\Gamma_{12}\Gamma_{13} + |\Omega_2|^2}$$ optical coherence at the ω_1 (probe) transition

$$\chi_p = \frac{\wp_{\cancel{13}}^2}{\hbar\varepsilon_0}\frac{\rho_{31}}{\Omega_1} = i\frac{\wp_{\cancel{13}}^2}{\hbar\varepsilon_0\Gamma_{13}}\left(\frac{\Gamma_{12}\Gamma_{13}}{\Gamma_{12}\Gamma_{13} + |\Omega_2|^2/4}\right) =$$

$$= i\frac{\wp_{\cancel{13}}^2}{\hbar\varepsilon_0\Gamma_{13}} - i\frac{\wp_{\cancel{13}}^2}{\hbar\varepsilon_0\Gamma_{13}}\frac{|\Omega_2|^2/4}{\Gamma_{12}\Gamma_{13} + |\Omega_2|^2/4}$$

$\Gamma_{13} = \gamma_{13} - i\Delta$

$\Gamma_{12} = \gamma_{23} - i\delta$

$$\chi_p = i\frac{\wp_{13}^2}{\hbar\varepsilon_0}\underbrace{\frac{\gamma_{13}+i\Delta}{\gamma_{13}+\Delta^2}}_{\substack{\text{"regular" one-photon}\\ \text{absorption, centered}\\ \text{at } \Delta=0 \ (\nu_1=\omega_{13})\\ \chi_p^{(1)}(\Delta)}} - i\underbrace{\frac{\wp_{13}^2}{\hbar\varepsilon_0}\frac{\gamma_{13}+i\Delta}{\gamma_{13}^2+\Delta^2}\frac{|\Omega_2|^2/4}{(\gamma_{12}+i\delta)(\gamma_{13}+i\Delta)+|\Omega_2|^2/4}}_{\substack{\text{two-photon}\\ \text{contribution}}}$$

$$\frac{\chi_p-\chi_p^{(1)}(\Delta)}{(\wp_{13}^2/\hbar\varepsilon)} \approx \frac{i|\Omega_2|^2/4\Delta^2}{\gamma_{12}-i\delta+\frac{|\Omega_2|^2}{4\Delta^2}(\gamma_{13}+i\Delta)} \approx$$

assuming $\gamma_{13}^2 \ll \Delta^2$

$$\approx \frac{i|\Omega_2|^2/4\Delta^2}{\underbrace{\left(\frac{\gamma_{12}}{2}+\frac{\gamma_{13}}{2}\cdot\frac{|\Omega_2|^2}{4\Delta^2}\right)}_{\gamma_{eff}} - i\underbrace{\left(\delta-\frac{|\Omega_2|^2}{\Delta}\right)}_{\delta_{eff}}}$$

Lorentzian lineshape

Narrow absorption resonance

width $\gamma_{eff} = \frac{\gamma_{12}}{2} + \frac{\gamma_{13}}{2}\frac{|\Omega_2|^2}{4\Delta^2} \ll \gamma_{13}$

position $\delta = \frac{|\Omega_2|^2}{4\Delta} = (\nu_1-\nu_2)-\omega_{12}$

$\nu_1-\nu_2 = \omega_{12}+\frac{|\Omega_2|^2}{4\Delta}$

~~Elec~~ Special case #2 — Electromagnetically induced transparency

We consider that $\Delta_2 = 0$ $(\nu_2 = \omega_{23})$, $\Delta_1 = \delta$

The espression for the susceptibility is still the same

$$\chi_p = i\frac{\cancel{\wp_{13}}^2}{\hbar\varepsilon_0\Gamma_{13}}\left(\frac{\Gamma_{12}}{\Gamma_{12} + |\Omega_2|^2/4\Gamma_{13}}\right)$$

if $\delta = 0$ $\quad \Gamma_{12} = \gamma_{12} \quad \Gamma_{13} = \gamma_{13}$

$$\chi_p = i\frac{\cancel{\wp_{13}}^2}{\hbar\varepsilon_0\gamma_{13}}\frac{\gamma_{12}}{\gamma_{12} + |\Omega_2|^2/4\gamma_{13}} \longrightarrow i\frac{\wp_{ab}^2}{\hbar\varepsilon_0\gamma_{13}}\frac{\gamma_{12}}{|\Omega_2|^2/4\gamma_{13}}$$

For $\gamma_{12} \to 0$ ($|1\rangle$ and $|2\rangle$ — stable states)
absorption disappears

$$\chi_p^{EIT} = \chi_p^{(resonant)}\frac{\gamma_{12}}{|\Omega_2|^2/4\gamma_{13}} \xrightarrow{|\Omega_2|\to\infty} 0$$

Notice that $\rho_{21} = -\frac{\Omega_1\Omega_2^*}{4\gamma_{12}\gamma_{13} + |\Omega_2|^2} \approx -\Omega_1/\Omega_2$
non-vanishing coherence
atoms are in a superposition of $|1\rangle$ & $|2\rangle$

$$\hat{H}_{int} = -\frac{\Omega_1^*}{2}|1\rangle\langle 3| - \frac{\Omega_1}{2}|3\rangle\langle 1| - \frac{\Omega_2^*}{2}|2\rangle\langle 3| - \frac{\Omega_2}{2}|3\rangle\langle 2|$$

~~Non interacting state~~ Eiguen-states $\hat{H}|\lambda\rangle = E_\lambda|\lambda\rangle$
Non-interacting state $\hat{H}|D\rangle = 0$
Dark state $|D\rangle = \frac{1}{\sqrt{\Omega_1^2+\Omega_2^2}}\left(\Omega_1|2\rangle - \Omega_2|1\rangle\right)$
orthogonal bright state $(\langle D|B\rangle = 0)$

$$|B\rangle = \frac{1}{\sqrt{\Omega_1^2+\Omega_2^2}}\left(\Omega_1^*|1\rangle + \Omega_2^*|2\rangle\right)$$

$$\hat{H}_{int} = \sqrt{\Omega_1^2+\Omega_2^2}\;|B\rangle\langle B|$$

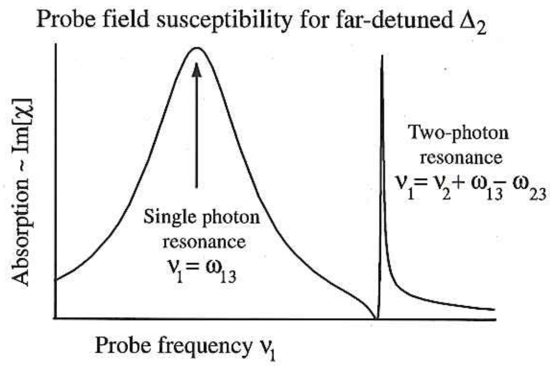


Figure 5.3: The imaginary part of the probe beam susceptibility as a function of the probe beam detuning. The absorption of Ω_1 exhibits a double resonance lineshape in the presence of a far-off resonant coupling beam Ω_2. In this calculation, $\gamma_{12} = 0$, so the single-photon and two-photon resonances have the same peak value, but the linewidth of the two-photon peak is significantly narrower.

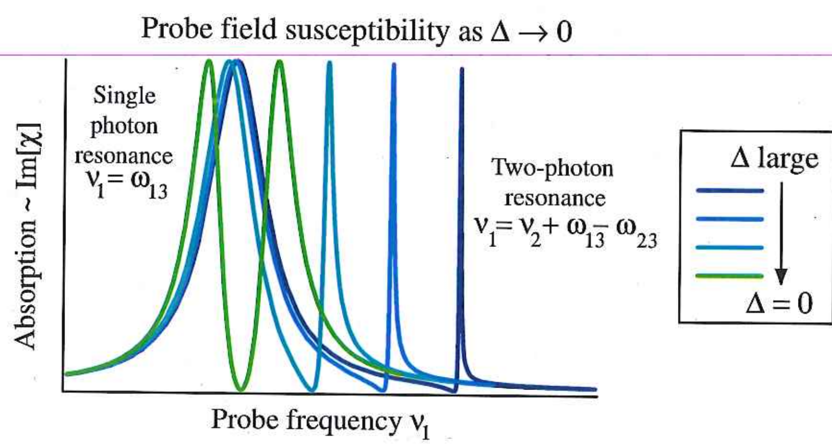


Figure 5.4: As the detuning Δ is reduced, the two-photon resonance moves towards the single-photon resonance and broadens. When the the two resonances begin to overlap, the system no longer behaves like a far-detuned system, and qualitatively new effects appear.