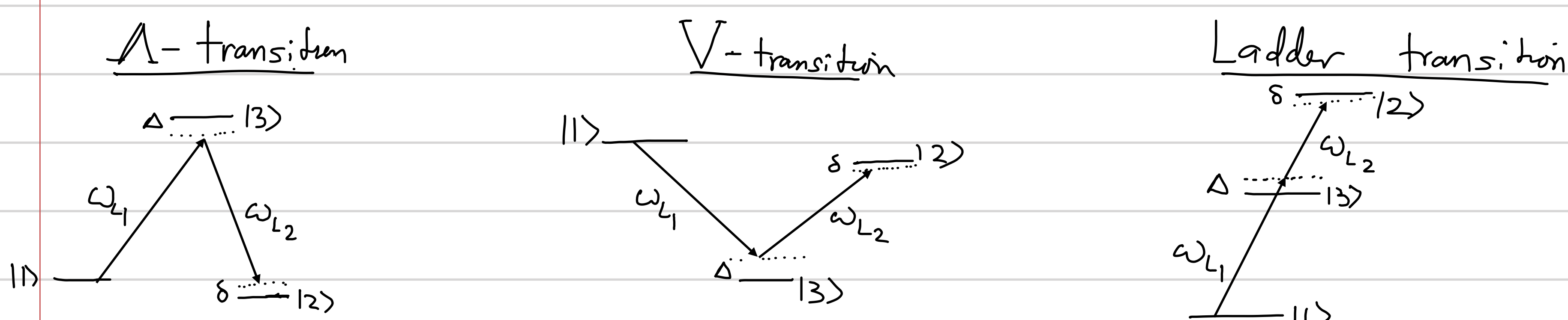
Physics 566 – Lecture 8

Three-Level Atoms: Adiabatic Elimination and Raman Transitions

Two-photon Transitions and Three-Level Atoms

We have focused thus far on near resonance interaction between a monochromatic optical field an atom transition $|e\rangle \to |g\rangle$. Often we would like to drive coherence between two levels $|1\rangle \leftrightarrow |2\rangle$, but they are not connected by an electric dipole transition at optical frequencies. Two achieve to transition, often we employ a "two-photon" transition, $|1\rangle \to |3\rangle \to |2\rangle$, where $|3\rangle$ is a "intermediate level" such that $|1\rangle \leftrightarrow |3\rangle$ and $|3\rangle \leftrightarrow |2\rangle$ are allowed optical transitions. We divide these into three categories

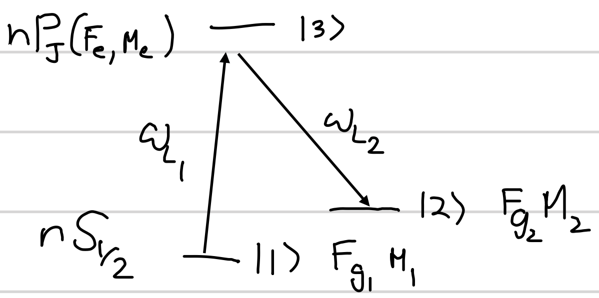


In all cases, the transition $|1\rangle \to |2\rangle$ is two-photon-resonant when $\delta = 0$. We have allowed the intermediate level, $|3\rangle$, to be off one-photon resonance when $\Delta \neq 0$.

- Λ transition: Two-photon resonance when $\omega_{L_1} - \omega_{L_2} = \frac{E_1 - E_2}{\hbar}$ (absorption $\omega_{L_1} \to$ emission ω_{L_2})
- V-transition: Two photon resonance when $\omega_{L_1} - \omega_{L_2} = \frac{E_1 - E_2}{\hbar}$ (emission $\omega_{L_1} \to$ absorption ω_2)
- Ladder-Transition: Two photon resonance when $\omega_{L_1} + \omega_{L_2} = \frac{E_2 - E_1}{\hbar}$ (absorption $\omega_{L_1} \to$ absorption ω_{L_2})

Note: The two laser modes, ω_{L_1} and ω_{L_2}, can be degenerate. In the context of the Λ or V-transitions, the $|1\rangle \leftrightarrow |3\rangle$ and $|2\rangle \leftrightarrow |3\rangle$ transitions are distinguished by the polarization of the laser, in which case levels $|1\rangle$ and $|2\rangle$ are distinguished by magnetic quantum numbers.

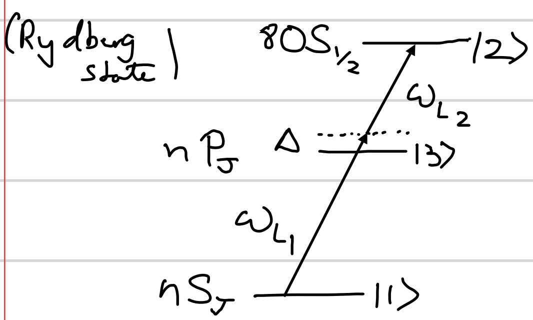
Example Λ transition: $|1\rangle$ and $|2\rangle$ are sublevels of the electronic ground state, e.g. two hyperfine states



This is a very common configuration in quantum optics. Ground state coherence is very long live – Controlled optically! This requires good phase-coherence between ω_{L_1} + ω_{L_2}

$\Rightarrow$ Coherent modulators at $\omega_{L_2} - \omega_{L_1}$ (rf/microwaves)

Example of Ladder Transition $|1\rangle$ is a ground S-state; $|2\rangle$ is a metastable, highly excited S-state in the "Rydberg series" $n > 40$



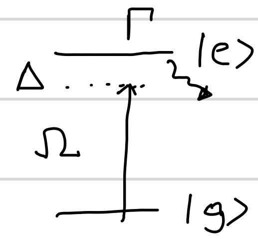
Rydberg states are interesting because they are very long lived and because the electron orbit is so far from the nucleus a huge dipole moment can be induced

Adiabatic Elimination

By choosing the detuning to the intermediate level, Δ, sufficiently large one can "adiabatically eliminate" $|3\rangle$ from the description of the dynamics and reduced to an effective two-level system $|1\rangle \Leftrightarrow |2\rangle$. Qualitatively, by the time-energy uncertainty principle, the atom makes a "virtual transition" to $|3\rangle$ for a time $\sim \frac{1}{\Delta}$. When $\Delta \gg \Omega_1, \Omega_2, \delta$, where $\hbar\Omega_i = d_{i3} E_{L_i}$ (the Rabi frequency), the time spent in $|3\rangle$ is much shorter than the dynamical evolution $|1\rangle \to |2\rangle$. The fast dynamics of $|3\rangle$ are then "slaved" to the slow dynamics of $|1\rangle, |2\rangle \Rightarrow$ the probability amplitude in $|3\rangle$ "adiabatically follows" that in levels $|1\rangle$ and $|2\rangle$.

We can see this quantitatively by studying approximate solutions to the time-dependent Schrödinger equation. We begin with the simple two-level case

Adiabatic elimination for two-level system: Ground-state dynamics



Consider a two-level system

Rabi frequency Ω, detuning Δ, Excited lifetime $\frac{1}{\Gamma}$

$\Delta, \Gamma \gg \Omega$.

If the spontaneous decay of $|e\rangle$ is only to levels other than $|g\rangle$, then we know that we can describe the dynamics by non-unitary evolution by a non-Hermitian Hamiltonian.

In the rotating frame and with $E_g = 0$, $E_e = \hbar\omega_{eg}$

$$\hat{H}_{eff} = \left(-\hbar\Delta - i\frac{\Gamma}{2}\right)|e\rangle\langle e| + \frac{\hbar\Omega}{2}\left(|e\rangle\langle g| + |g\rangle\langle e|\right)$$

The nonunitary Schrödinger equation:

$$\frac{d}{dt}c_e = \left(i\Delta - \frac{\Gamma}{2}\right)c_e - \frac{i}{2}\Omega\, c_g \; ; \qquad \frac{d}{dt}c_g = -i\frac{\Omega}{2}c_e$$

We can formally integrate the excited state amplitude, with the initial condition $c_e(0)$

$$c_e(t) = c_e(0)\, e^{(i\Delta - \frac{\Gamma}{2})t} - i\frac{\Omega}{2}\int_0^t dt'\, e^{(i\Delta - \frac{\Gamma}{2})(t-t')}\, c_g(t') \; ;$$

Let us take $c_e(0) = 0$

Atom in ground state @ $t = 0$

From this expression, it is clear that $c_e(t)$ changes on time scale Δ^{-1} and Γ^{-1}. In contrast, c_g changes on the time scale no faster than Ω^{-1} once c_e reaches steady state. When $|\Delta - i\frac{\Gamma}{2}| \gg \Omega$, we can treat c_g as unchanging inside the integral expression for $c_e(t)$.

$$c_e(t) \cong -i\frac{\Omega}{2}\int_0^t dt'\, e^{(i\Delta - \frac{\Gamma}{2})(t-t')}\, c_g(t) \approx \frac{-i\Omega/2}{-i\Delta + \Gamma/2}\left[1 - e^{(i\Delta - \frac{\Gamma}{2})t}\right]c_g(t)$$

$$\Rightarrow \dot{c}_g = \frac{-\Omega^2/4}{-i\Delta + \frac{\Gamma}{2}}\left[1 - \underbrace{e^{(i\Delta - \frac{\Gamma}{2})t}}_{\text{rapidly varying}}\right]c_g(t)$$

$$\Rightarrow \dot{c}_g \cong \left(-i\frac{s}{2}\Delta - \frac{s}{4}\Gamma\right)c_g = \left(-i\frac{\delta E_{LS}}{\hbar} - \frac{\gamma_s}{2}\right)c_g$$

where $\delta E_{LS} = \frac{s}{2}\hbar\Delta$ is the "light shift" on the ground state energy

$\gamma_s = \frac{s}{2}\Gamma = N_e\Gamma$ is the photon scattering rate

In other words, adiabatically eliminating the excited state, the "coherent" evolution of the ground state is governed by the light shift energy δE_{LS}, whereas the decay of the ground state is set by the scattering rate.

Note: $\delta E_{LS} \propto s\Delta \propto \frac{\Omega^2 \Delta}{\Delta^2 + \Gamma^2/4} \sim \frac{\Omega^2}{\Delta} \propto \frac{I}{\Delta}$ when $\Delta \gg \Gamma$

$\gamma_s \propto s\Gamma \propto \frac{\Omega^2 \Gamma}{\Delta^2 + \Gamma^2/4} \propto \frac{\Omega^2 \Gamma}{\Delta^2} \propto \frac{I\Gamma}{\Delta^2}$

Thus, by increasing detuning and intensity, one can make δE_{LS} arbitrarily large compared to $\hbar\gamma_s$ $\Rightarrow$ At large detuning and low saturation, one can have coherent dynamics dominate over dissipative dynamics.

Light-shift and dressed states

The ac-Stark shift can be seen as the shift in the eigenvalue of the perturbed or "dressed state." The dressed state is the eigenvector of the total Hamilton in the rotating frame

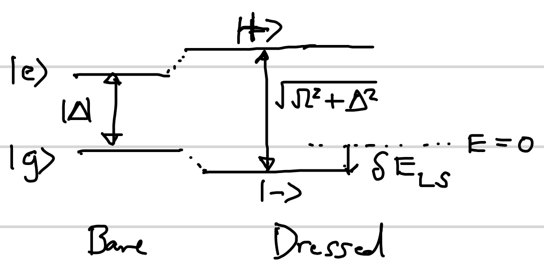
$$\hat{H} = \underbrace{\hat{H}_A}_{\text{"Bare Hamiltonian"}} + \underbrace{\hat{H}_{AL}}_{\text{"Dressing Hamiltonian"}} = -\hbar\Delta|e\rangle\langle e| + \frac{\hbar\Omega}{2}(|e\rangle\langle g| + |g\rangle\langle e|) = -\frac{\hbar\Delta}{2}\hat{\mathbb{1}} - \frac{\hbar\Delta}{2}\hat{\sigma}_z + \frac{\hbar\Omega}{2}\hat{\sigma}_x$$

Diagonalize: Eigenvalues $E_\pm = -\frac{\hbar\Delta}{2} \pm \frac{\hbar\sqrt{\Omega^2+\Delta^2}}{2}$

Eigenvectors $|+\rangle = |\uparrow_n\rangle = \cos\frac{\theta}{2}|e\rangle + \sin\frac{\theta}{2}|g\rangle$, $|-\rangle = |\downarrow_n\rangle = \sin\frac{\theta}{2}|e\rangle - \cos\frac{\theta}{2}|g\rangle$

$\cos\theta = \frac{-\Delta}{\sqrt{\Omega^2+\Delta^2}}$, $\sin\theta = \frac{\Omega}{\sqrt{\Delta^2+\Omega^2}}$, $\theta = \tan^{-1}\left(-\frac{\Omega}{\Delta}\right)$

Red detuning $\Delta < 0$



Ground-state atom attracted to regions of higher intensity

$$\delta E_{LS} = -\frac{\hbar\Delta}{2} - \frac{\hbar}{2}\sqrt{\Omega^2+\Delta^2} = \frac{\hbar|\Delta|}{2} - \frac{\hbar|\Delta|}{2}\sqrt{1+\frac{\Omega^2}{\Delta^2}}$$

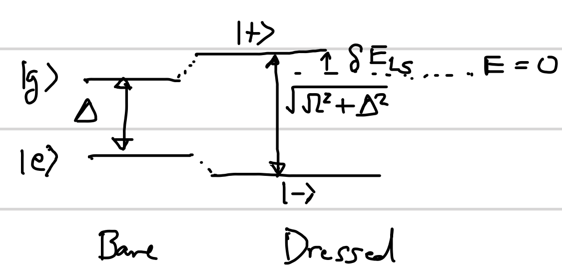
$$\approx -\frac{\hbar\Omega^2}{4|\Delta|} = \frac{\hbar\Omega^2}{4\Delta} = \frac{\hbar}{2}s\Delta \quad \left(\frac{\Omega}{|\Delta|} \ll 1\right)$$

$$\theta = \tan^{-1}\left(\frac{\Omega}{|\Delta|}\right) \approx \frac{\Omega}{|\Delta|} \quad \text{(low saturation)}$$

$$|-\rangle \approx \frac{\theta}{2}|e\rangle - |g\rangle$$

$$\approx -\left(|g\rangle - \frac{\theta}{2}|e\rangle\right) \Rightarrow |g\rangle - \frac{\Omega}{2|\Delta|}|e\rangle = \underbrace{|g\rangle - \frac{d_{ge}E_0}{2\hbar\Delta}|e\rangle}_{\text{dressed ground state}}$$

Blue detuning $\Delta > 0$



Ground-state atom repelled from regions of higher intensity

$$\delta E_{LS} = -\frac{\hbar\Delta}{2} + \frac{\hbar}{2}\sqrt{\Omega^2+\Delta^2} \approx \frac{\hbar\Omega^2}{4\Delta} = \frac{\hbar}{2}s\Delta$$

$$\theta = \tan\left(\frac{\Omega}{-\Delta}\right) \approx \pi - \frac{\Omega}{\Delta}$$

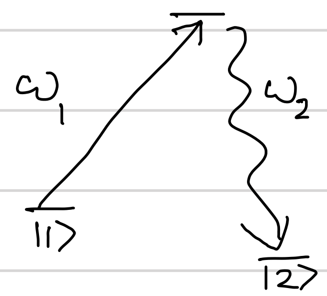
$$|+\rangle = \cos\left(\frac{\pi}{2} - \frac{\Omega}{2\Delta}\right)|e\rangle + \sin\left(\frac{\pi}{2} - \frac{\Omega}{2\Delta}\right)|g\rangle$$

$$= \sin\left(\frac{\Omega}{2\Delta}\right)|e\rangle + \cos\left(\frac{\Omega}{2\Delta}\right)|g\rangle$$

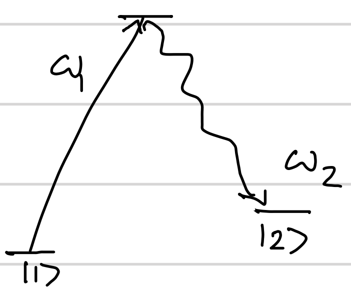
$$\approx |g\rangle + \frac{\Omega}{2\Delta}|e\rangle = |g\rangle - \frac{d_{eg}E_0}{2\hbar\Delta}|e\rangle$$

Raman Scattering

In traditional optics, the inelastic scattering a light, absorption of a photon at frequency ω_1 followed by emission of a photon with a different frequency, and transfer of the system from one electronic ground level to another is know as Raman Scattering. Typically, this is one resonance and the scattering is via spontaneous emission between two "ro-vibrational" states of a ground-electronic molecule. In AMO physics, we tend to call any two-photon transition from $|1\rangle \leftrightarrow |2\rangle$ through an intermediate $|3\rangle$ that is higher energy that $|1\rangle$ and $|2\rangle$ a "Raman transition." If $|1\rangle$ and $|2\rangle$ are degenerate, then the two photons are distinguished by their polarization, rather than their frequency.



"Anti-Stokes" $\omega_2 > \omega_1$
Spontaneous Raman Scattering

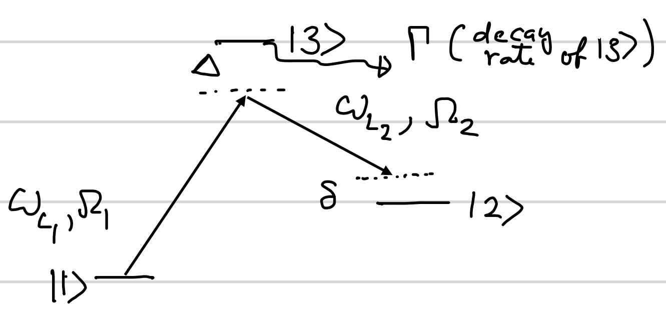


"Stokes" $\omega_2 < \omega_1$
Spontaneous Raman Scattering

When both fields at ω_1 and ω_2 are present, one speaks of "Stimulated Raman" transitions. Like the two-level system previously studied, the description in terms of spontaneous and stimulated transitions misses the important physics of interest to us: Creation and manipulation of coherence between levels $|1\rangle$ and $|2\rangle$. We can achieve such coherent manipulation by detuning from from excited resonance and adiabatically eliminating level $|3\rangle$. The dynamics is then described by coherent Raman-Rabi oscillations between $|1\rangle + |2\rangle$.

Raman-Rabi Flopping

Consider the Λ-configuration



We have chosen $\Delta_1 = \omega_{L_1} - \omega_{31} \equiv \Delta$ $\qquad \Delta_2 = \omega_{L_2} - \omega_{32}$

$\omega_{ij} = (E_i - E_j)/\hbar$

$$\delta = \Delta_1 - \Delta_2 = \omega_{L_1} - \omega_{L_2} - \left(\frac{E_2 - E_1}{\hbar}\right)$$

$\equiv$ Raman detuning

The effective Hamiltonian $\hat{H} = \hat{H}_A^{eff} + \hat{H}_{AL}$, $\quad \hat{H}_A = \sum_j (E_j - i\frac{\hbar\Gamma_j}{2}) |j\rangle\langle j|$

$$\hat{H}_{AL} = -\hat{\vec{d}} \cdot \mathrm{Re}\,(\vec{E}_1 e^{-i\omega_{L1} t} + \vec{E}_2 e^{i\omega_{L2} t})$$

Restricting to the 3-levels, go to an appropriate rotating frame, and making the RWA

$$\hat{H}_{eff} = -\hbar(\Delta + i\tfrac{\Gamma}{2})|3\rangle\langle 3| - \hbar\delta|2\rangle\langle 2| + \frac{\hbar\Omega_1}{2}(|3\rangle\langle 1| + |1\rangle\langle 3|) + \frac{\hbar\Omega_2}{2}(|3\rangle\langle 2| + |2\rangle\langle 3|)$$

where we have taken $E_1 = 0$, $\quad \hbar\Omega_i \simeq -\langle 3|\hat{\vec{d}}\cdot\vec{E}_1|i\rangle$, $\quad i = 1, 2$ (see homework)

The non-unitary evolution (neglecting "refeeding" by spontaneous decay back into $|1\rangle, |2\rangle$)

$$\frac{\partial}{\partial t}|\psi\rangle = \frac{-i}{\hbar}\hat{H}_{eff}|\psi\rangle \qquad |\psi\rangle = c_1|1\rangle + c_2|2\rangle + c_3|3\rangle$$

$$\Rightarrow \dot{c}_1 = -i\frac{\Omega_1}{2}c_3, \quad \dot{c}_2 = i\delta c_2 - i\frac{\Omega_2}{2}c_3, \quad \dot{c}_3 = i(\Delta + i\tfrac{\Gamma}{2})c_3 - i\frac{\Omega_1}{2}c_1 - i\frac{\Omega_2}{2}c_2$$

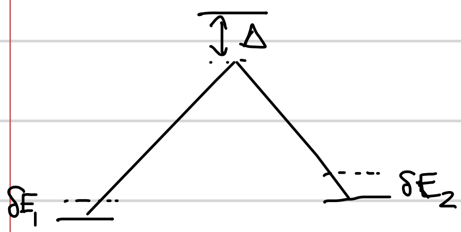
Adiabatic elimination: When $|\Delta + i\frac{\Gamma}{2}| \gg \Omega_1, \Omega_2, \delta$ $\quad c_3$ is "slaved" to c_1 + c_2

Steady state for $c_3 \Rightarrow$ $$c_3 = \frac{\Omega_1/2}{\Delta + i\Gamma/2}c_1 + \frac{\Omega_2/2}{\Delta + i\Gamma/2}c_2$$

$$\Rightarrow \dot{c}_1 = -i\frac{\Omega_1^2/4}{\Delta + i\Gamma/2}c_1 - i\frac{\Omega_1\Omega_2/4}{\Delta + i\Gamma/2}c_2 \;;\quad \dot{c}_2 = i\left(\delta - \frac{\Omega_2^2/4}{\Delta - i\Gamma/2}\right)c_2 - i\frac{\Omega_1\Omega_2/4}{\Delta - i\Gamma/2}c_1$$

$$\Rightarrow \boxed{\begin{aligned} \dot{c}_1 &= \frac{-i}{\hbar}\left(\delta E_1 - i\frac{\hbar\gamma_1}{2}\right)c_1 - i\left(\frac{\Omega_{Ram}}{2} - i\frac{\Gamma_{Ram}}{2}\right)c_2 \\ \dot{c}_2 &= \frac{-i}{\hbar}\left(-\delta + \delta E_2 - i\frac{\hbar\gamma_2}{2}\right)c_2 - i\left(\frac{\Omega_{Ram}}{2} - i\frac{\Gamma_{Ram}}{2}\right)c_1 \\ \delta E_j &= \frac{\hbar\Omega_j^2/4}{\Delta^2 + \frac{\Gamma^2}{4}} = \frac{s_j}{2}\hbar\Delta = \text{Light shift on level } |j\rangle, \quad \gamma_j = \frac{s_j}{2}\Gamma = \text{Photon scattering rate for field } \vec{E}_j \\ \Omega_{Ram} &= \frac{\Omega_1\Omega_2\Delta/2}{\Delta^2 + \frac{\Gamma^2}{4}} = \text{Raman-Rabi frequency}; \quad \Gamma_{Ram} = \frac{\Omega_1\Omega_2\Gamma/4}{\Delta^2 + \Gamma^2/4} \end{aligned}}$$

These are the equations of damped Rabi oscillations between levels $|1\rangle$ and $|2\rangle$



"Raman Resonance" when $\delta = \delta E_2 - \delta E_1$

$$\delta_{Ram} \equiv \delta - (\delta E_2 - \delta E_1)$$

When $\Delta >> \Gamma$ (typical case),

$$\gamma_j \simeq \frac{\Omega_j^2 \Gamma}{4\Delta^2}, \quad \Gamma_{Ram} \simeq \frac{\Omega_1 \Omega_2 \Gamma}{4\Delta^2} << \frac{\delta E_j}{\hbar} \simeq \frac{\Omega_j^2}{4\Delta}, \quad \Omega_{Ram} \simeq \frac{\Omega_1 \Omega_2}{4\Delta}$$

$$\boxed{\begin{aligned} &\Rightarrow \dot{c}_1 = -i \frac{\delta E_1}{\hbar} c_1 - i \frac{\Omega_{Ram}}{2} c_2, \qquad \dot{c}_2 = -i\left(\frac{\delta E_2}{\hbar} - \delta\right) c_2 - i \frac{\Omega_{Ram}}{2} c_1 \\ &\Rightarrow \dot{\rho}_{21} = c_2^* \dot{c}_1 + \dot{c}_2^* c_1 = -i \delta_{Ram} \rho_{21} - i \frac{\Omega_{Ram}}{2} (\rho_{22} - \rho_{11}) \end{aligned}}$$

Equations of coherent Rabi oscillations between $|1\rangle$ and $|2\rangle$

The coherence of the Raman transition is fundamentally limited by spontaneous emission from the excited level $|3\rangle$. However the probability to be excited can be very small compared to the probability to be in levels $|1\rangle$ and $|2\rangle$ $\Rightarrow$ Decoherence rate $\sim \gamma_s$ = photon scattering rate. When $\Delta >> \Gamma$, $\Omega_{Ram} >> \gamma_s, \Gamma_{Ram}$