Physics 566 - Lecture 9

Three-Level Atoms: Dark States and Electromagnetically Induced Transparency (EIT)

Dark States: Let us return to the lamba configuration studied in the last lecture. The solution for the excited probability amplitude c_3 shows an important quantum interference phenomenon: Dark States $\equiv$ States which don't absorb to $|3\rangle$.

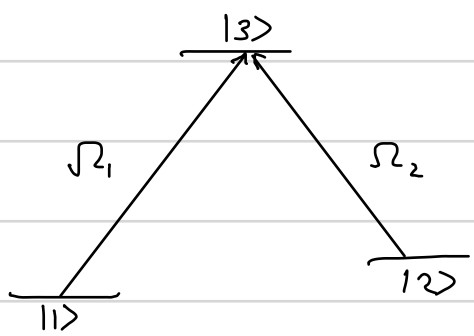
Look for solutions with $c_3 = 0$

$$c_3 = \frac{\Omega_1/2}{\Delta + i\Gamma/2} c_1 + \frac{\Omega_2/2}{\Delta + i\Gamma/2} c_2 = 0 \Rightarrow \Omega_1 c_1 + \Omega_2 c_2 = 0$$

$$\Rightarrow \boxed{\text{When } \frac{c_1}{c_2} = -\frac{\Omega_2}{\Omega_1}, \text{ the state is dark}}$$

True even on resonance.

The dark-state phenomenon can be understood as destructive interference between two absorption processes. Consider the resonant case



The two absorption paths occur with probability amplitudes. The transition rates have amplitudes $c_1\Omega_1$ and $c_2\Omega_2$ respectively. When $c_1\Omega_1 = -c_2\Omega_2 \Rightarrow$ destructive interference.

More quantatively: Transition rate $R \propto |\langle 3|\hat{H}_{AL}|\psi\rangle|^2$, $|\psi\rangle = c_1|1\rangle + c_2|2\rangle$

In rotating frame

$$\langle 3|\hat{H}_{AL}|\psi\rangle = -\frac{\hbar}{2}(\Omega_1 c_1 + \Omega_2 c_2) = 0 \quad \text{for dark state}$$

$$\boxed{|\psi_{Dark}\rangle = \frac{1}{\sqrt{\Omega_1^2+\Omega_2^2}}\left(\Omega_2|1\rangle - \Omega_1|2\rangle\right)}$$

For the case $\Omega_1 = \Omega_2 \Rightarrow |\psi_{Dark}\rangle = \frac{1}{\sqrt{2}}(|1\rangle - |2\rangle)$ (antisymmetric combination)

Said another way, the lack of absorption is because the two dipoles induced on $|1\rangle \to |3\rangle$ and $|2\rangle \to |3\rangle$ are equal in magnitude but 180° out of phase $\Rightarrow$ destructive interference. This kind of interference is known as Fano interference who studied this first in the context of atomic ionization to different excitation channels in atoms

Coherent Population Trapping (CPT)

The existence of dark states in two-photon spectroscopy were first discoversed experimentally in the absorption spectrum of atomic vapors. The system is "optically pumped" into the dark state. One the system is pumped into the dark state it no longer absorbs and is thus "trapped" in that state until some decoherence process causes the atom to leave that state. This was dubbed "coherent population trapping" (CPT), as the atom is trapped in a particular coherent superposition of state that is dark due to Fano interference.

To see how this might be possible, consider the equation of motion for the coherence ρ_{12}.

$$\frac{d}{dt}\rho_{12} = -\frac{i}{\hbar}\langle 1|[\hat{H}_{eff},\hat{\rho}]|2\rangle$$

(note: the "feeding term" in the Master equation does not contribute to the coherences).

$\Rightarrow \frac{d}{dt}\rho_{12} = c_1^* \dot{c}_2 + \dot{c}_1^* c_2$, where c_j evolves according to $\hat{H}_{eff}$ as given in lecture 9.

$$\Rightarrow \frac{d}{dt}\rho_{12} = \left\{-\frac{i}{\hbar}\left(\delta E_2 - \delta E_1 - \delta\right) - \frac{\gamma_1+\gamma_2}{2}\right\}\rho_{12} - i\frac{\Omega_{Ram}}{2}\left(\rho_{11}-\rho_{22}\right) - \frac{\Gamma_{Ram}}{2}$$

where $\delta = \omega_{L_2} - \omega_{L_1} - \left(\frac{E_2-E_1}{\hbar}\right)$ (Raman detuning), $\delta E_j = \frac{s_j}{2}\hbar\Delta$ (light shift), $\gamma_j = \frac{s_j}{2}\Gamma$ (scattering rate)

$\Omega_{Ram} = \sqrt{s_1 s_2}\,\Delta$ (Raman Rabi freq), $\Gamma_{Ram} = \frac{\sqrt{s_1 s_2}}{2}\Gamma$ (Raman decay), $s_j = \frac{\Omega_j^2/2}{\Delta^2+\Gamma^2/4}$

The coherences decay to steady state $\rho_{12}^{s.s.} \neq 0$. The coherence is "trapped."

Consider the special case of resonance $\Delta = 0$ and $\delta = 0 \Rightarrow \delta E_j = 0$, $\Omega_{Ram} = 0$.

$$\Rightarrow \rho_{12}^{s.s.} = \frac{-\Gamma_{Ram}}{\gamma_1+\gamma_2} = \frac{-\Omega_1\Omega_2}{\Omega_1^2+\Omega_2^2}$$

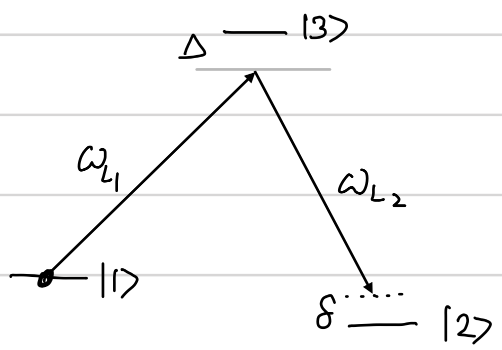
Exactly what we expect for the dark state $|\psi_{Dark}\rangle = \frac{\Omega_2|1\rangle - \Omega_1|2\rangle}{\sqrt{\Omega_1^2+\Omega_2^2}}$.

Thus, in steady state, the atom is optically pumped to the dark state.

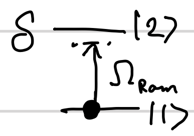
Note: In general, there is an additional source of decay of coherence γ_{12} due to, e.g. collisions. Typically, this limits the steady state coherence ρ_{12}, and thus the coherent population trapping.

STIRAP and Dark State Preparation via Counterintuitive Pulse Sequence

Suppose we seek to transfer population from level $|1\rangle$ to level $|2\rangle$ through a two photon transition connected through an intermediate level $|3\rangle$



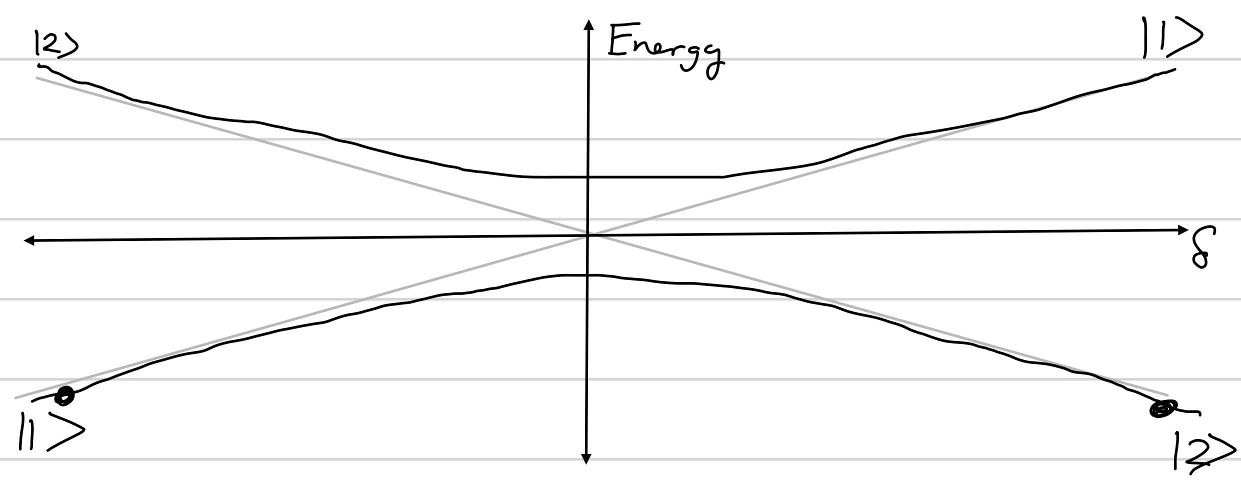
Once we adiabatically eliminate level $|3\rangle$, we can use what we have learned about two-level transition



As we studied, there are two ways to transfer population from $|1\rangle$ to $|2\rangle$ in a two level scenario:

(i) Resonant Rabi π-pulse, $\delta = 0$, $\Omega_{Ram} T = \pi$.

(ii) Adiabatic transfer from $|1\rangle \to |2\rangle$

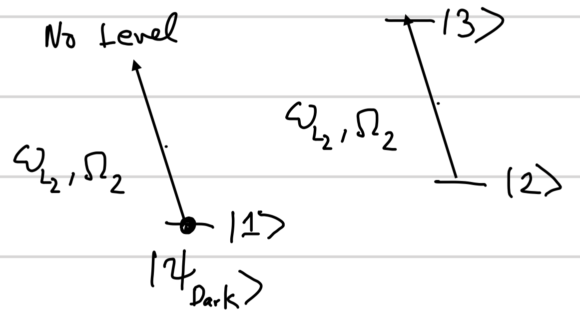


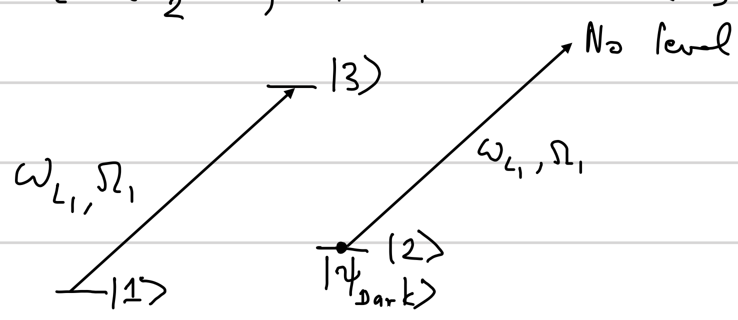
By adiabatically sweeping δ, we transfer $|1\rangle$ to $|2\rangle$ in a manner that is robust to the energy difference between $|1\rangle$ & $|2\rangle$. This is effective when $\Omega_{Ram} \gg \gamma_s$, the scattering rate.

For the three level transition, there is yet another method: Dark state adiabatic transfer via the counter-intuitive pulse sequence. This is known as Stimulated Raman Adiabatic Passage (STIRAP).

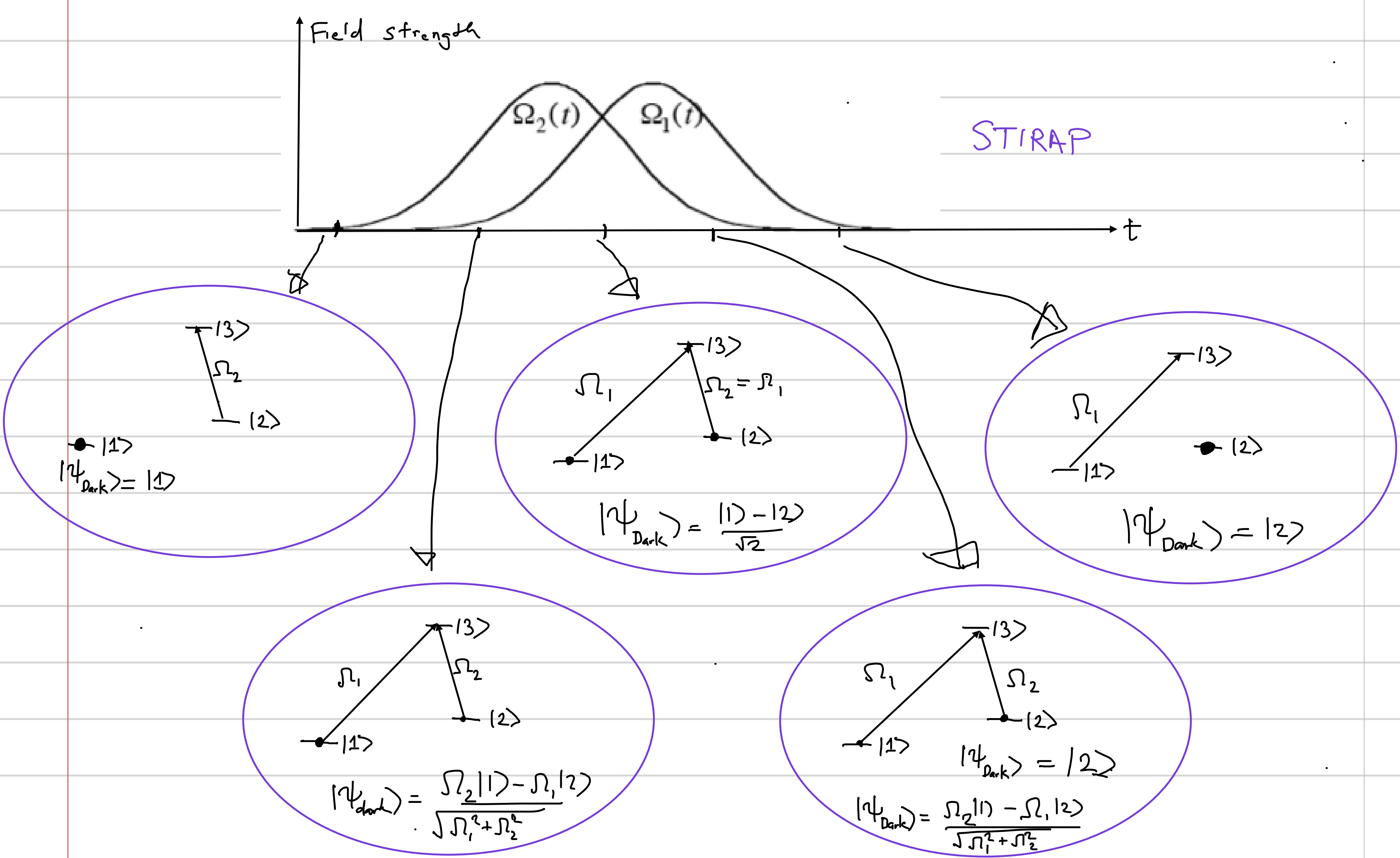
Recall the dark state on resonance: $|\psi_{Dark}\rangle = \dfrac{\Omega_2 |1\rangle - \Omega_1 |2\rangle}{\sqrt{\Omega_1^2 + \Omega_2^2}}$

Where $\Omega_1 = 0$, $|1\rangle$ is the dark state; when $\Omega_2 = 0$, $|2\rangle$ is the dark state.





Thus we can adiabatically transfer $|1\rangle \to |2\rangle$ by first turning on laser at ω_{L_2} and thus slowly turning on the laser at frequency ω_{L_1}, always staying in the dark state



We start in $|1\rangle$ and first turn on field $\vec{E}_2 \cos(\omega_{L_2} t)$ (Rabi freq $\Omega_2 = \vec{d}_{23} \cdot \vec{E}_2 / \hbar$).
As we slowly turn off Ω_2 and turn on field $\vec{E}_1 \cos(\omega_{L_1} t)$ $(\Omega_1 = \vec{d}_{13} \cdot \vec{E}_1 / \hbar)$
the state is adiabatically transferred from $|1\rangle \to \dfrac{\Omega_2(t)\,|1\rangle - \Omega_1(t)|2\rangle}{\sqrt{\Omega_1^2(t) + \Omega_2^2(t)}}$.

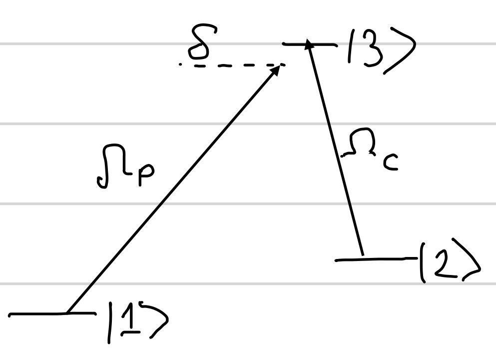
Eventually, we transition to field E_1 off and E_2 on $\Rightarrow$ we transition to $|2\rangle$.
This is known as the "counter-intuitive" pulse sequence because it is the reverse of a direct transition $|1\rangle \to |3\rangle \to |2\rangle$.

The counter-intuitive sequence requires a careful timing of the two-pulses and so it less prevalent than the simpler $|1\rangle \to |2\rangle$ transition through a Raman π-pulse. It is central, however, to the phenomenon of "electromagnetic induced transparency" (EIT) which we turn to next.

Electromagnetically Induced Transparency (EIT)

While the phenomenon of CPT dates back to 1976, it was seen mainly as a curiosity rather than a tool for controlling the atom-light interface. This changed in the 1990s, when there was intense study of manipulating the atomic coherence to "engineer" different from the Lorentz oscillator or saturable absorber. Typically, interesting nonlinear response occurs near resonance, where absorption is high. The goal, thus, is to use quantum coherence to enhance nonlinear response, while reducing absorption. The quintessential example is EIT.

Consider again the Λ-configuration on resonance



We drive the $|2\rangle \leftrightarrow |3\rangle$ transition with a strong "coupling laser," Rabi frequency Ω_c. The transition $|1\rangle \leftrightarrow |3\rangle$ is driven with a weak "probe laser," Rabi frequency $\Omega_p \ll \Omega_c$.

Our goal is to determine to dipole response to the probe field in the presence of the coupling field.

Because Ω_c is strong, we do not adiabatically eliminate level $|3\rangle$. We have the following equation of motion for the coherence as follows from the effective Hamiltonian:

$$\dot{\rho}_{31} = c_3\dot{c}_1^* + \dot{c}_3 c_1^* = \left(i\delta - \frac{\Gamma}{2}\right)\rho_{31} - i\frac{\Omega_p}{2}\left(\rho_{11} - \rho_{33}\right) - i\frac{\Omega_c}{2}\rho_{21}$$

When $\Omega_c >> \Omega_p$, we have $\rho_{33} << \rho_{11} \approx 1$, assuming $\rho_{11}(0) = 1$, and to linear order in Ω_p

$$\Rightarrow \rho_{31} \approx \frac{1}{\delta + i\Gamma/2}\left[\frac{\Omega_p}{2} + \frac{\Omega_c}{2}\rho_{21}\right]$$

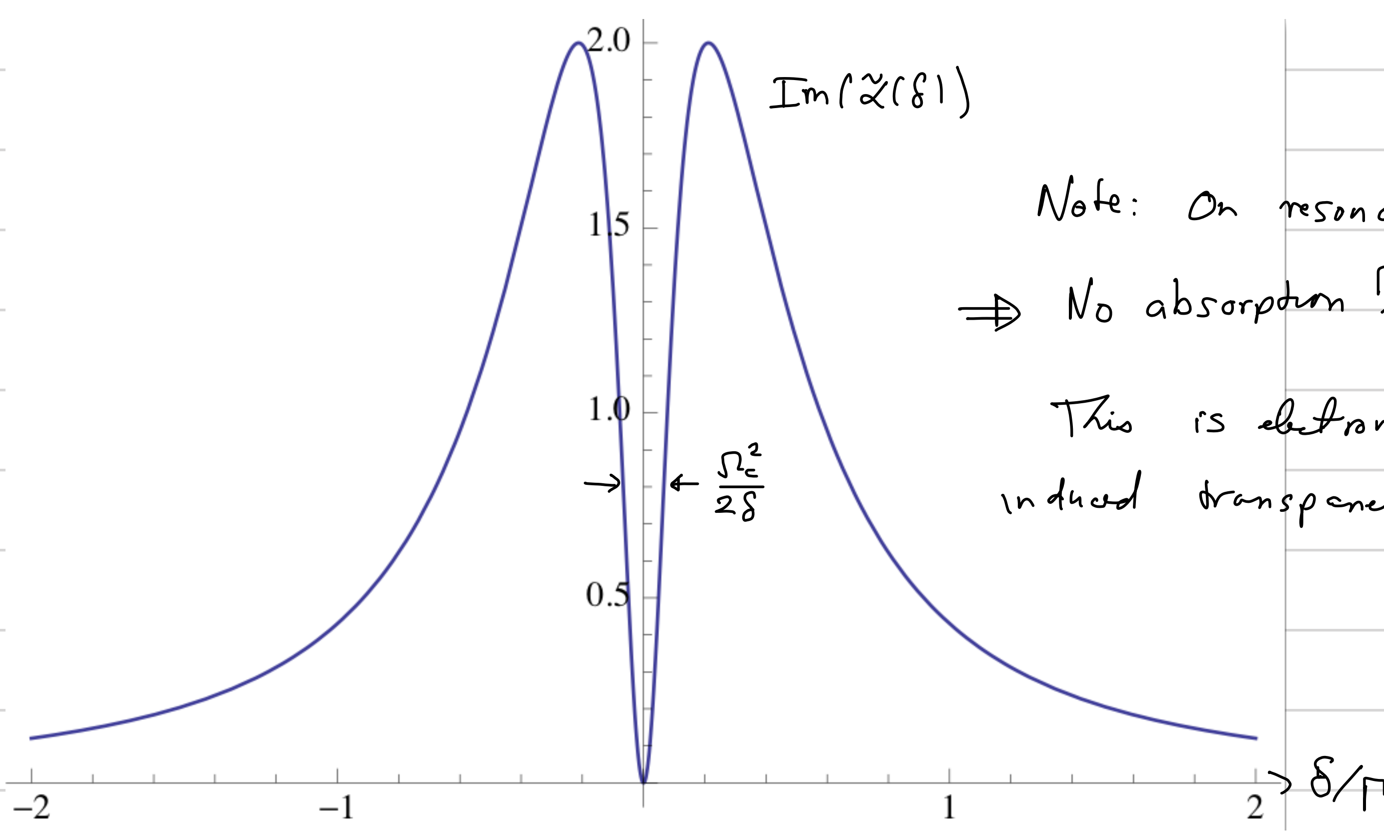
$$\dot{\rho}_{21} = c_2\dot{c}_1^* + \dot{c}_2 c_1^* = +i\delta\rho_{21} + \underbrace{\cancel{i\frac{\Omega_p}{2}\rho_{23}}}_{\text{neglect for } \Omega_c >> \Omega_p} - \frac{i\Omega_c}{2}\rho_{31} \quad \Rightarrow \quad \rho_{21} \approx \frac{\Omega_c}{2\delta}\rho_{31}$$

$$\Rightarrow \rho_{31} \approx \frac{\Omega_p/2}{\delta + i\frac{\Gamma}{2} - \frac{\Omega_c^2}{2\delta}} = \frac{(-d_{31}/2\hbar)}{\delta - i\frac{\Gamma}{2} - \frac{\Omega_c^2}{2\delta}} E_p$$

$\Rightarrow$ Induced dipole @ ω_p

$$\langle d\rangle_{\text{induced}} = \underbrace{\frac{-|d_{31}|^2/\hbar}{\delta + i\frac{\Gamma}{2} - \frac{\Omega_c^2}{2\delta}}}_{\tilde{\alpha}(\delta) \leftarrow \text{polarizability}} E_p$$

Example with $\Omega_c = 0.3\Gamma$

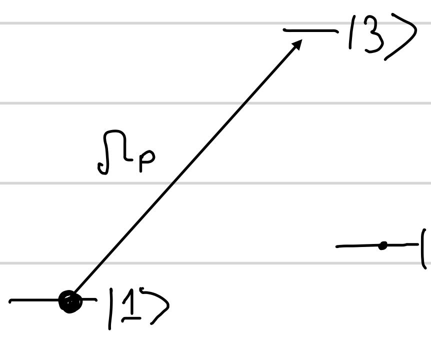


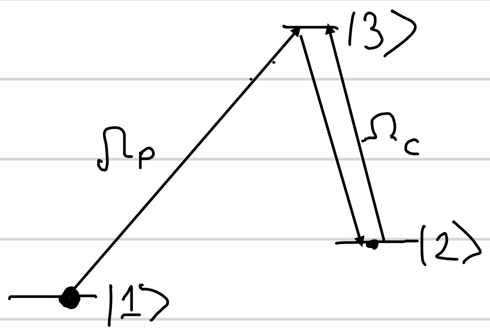
Note: On resonance $\delta = 0$

$\Rightarrow$ No absorption!

This is electromagnetically induced transparency

EIT is quantum interference phenomenon. Absorption vanishes on resonance because of destructive interference between different processes that lead to a transfer of population from $|1\rangle \to |3\rangle$ via removal of one laser photon @ ω_p



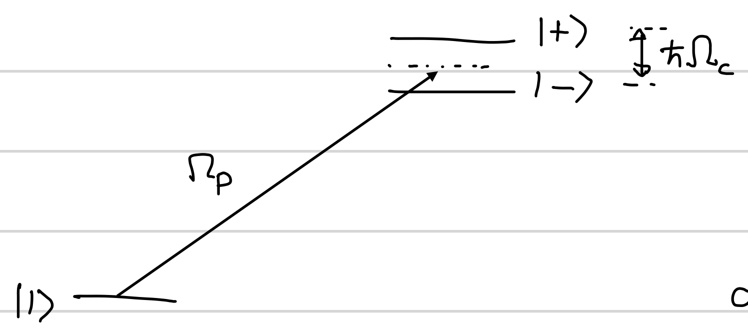


Process #1: Direct absorption $|1\rangle \to |3\rangle$ of $\vec{E}_p$

Process #2: Two-photon transition from $|1\rangle \to |2\rangle$ followed by absorption of a coupling photon $\vec{E}_c$

These two processes destructively interfere when we are in the dark state. Here, only a tiny fraction of the population is in $|2\rangle$ because $\Omega_c \gg \Omega_p$.

Another way to view this that goes beyond the perturbation theory to second order in Ω_c is to use the dressed states. The coupling laser strongly couples $|2\rangle$ and $|3\rangle$, hence the name. We thus consider the dressed states $|\pm\rangle = \frac{1}{\sqrt{2}}(|2\rangle \pm |3\rangle)$, eigenstates of the interaction Hamiltonian coupling this levels. The probe laser then probes these levels:

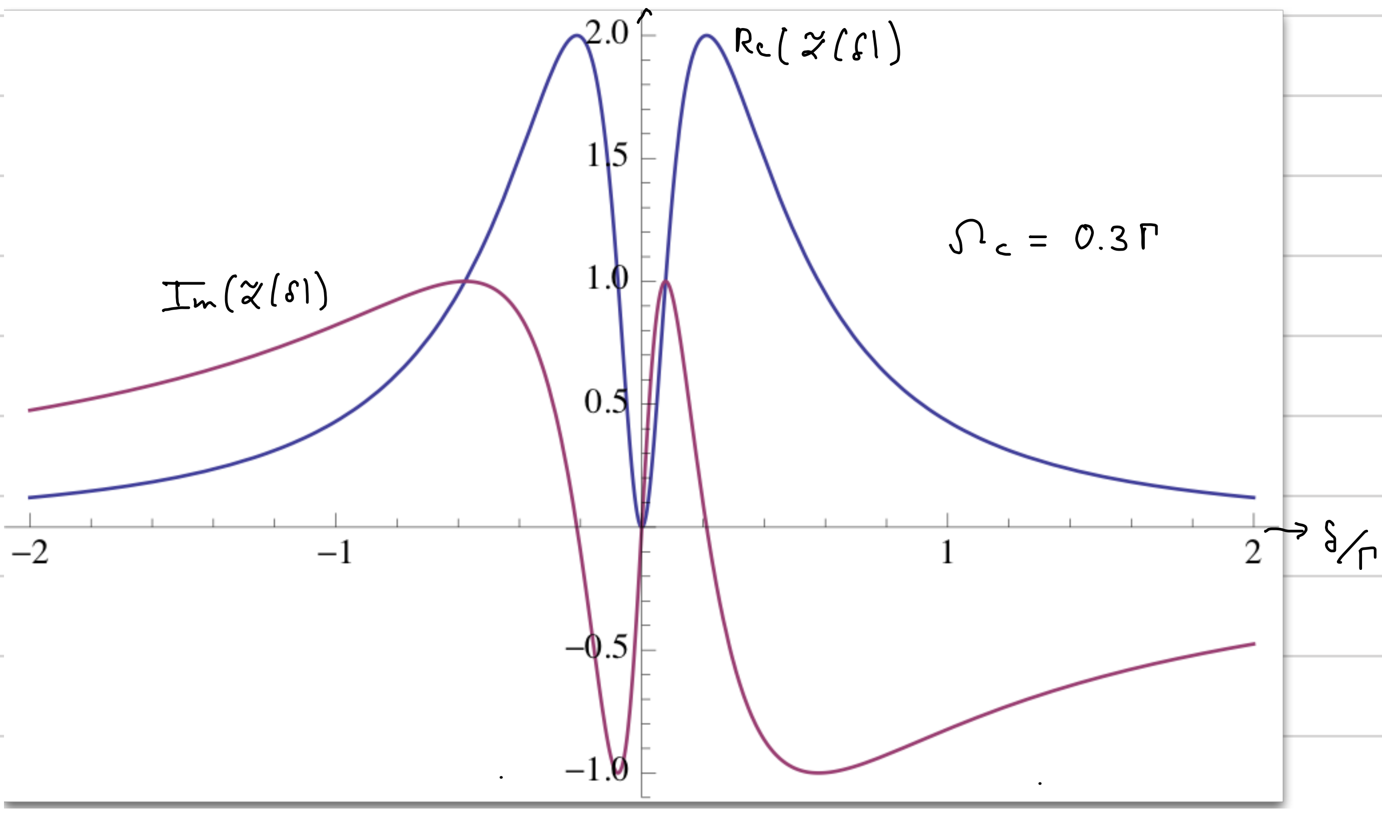


There is a (Fano) quantum interference for absorption when the probe is detuned half-way between $|+\rangle$ & $|-\rangle$ because the two dipoles are 180° out of phase

Quantatively, we can find the dressed state for the whole 3 level system. We find a perfect dark state at Raman resonance and an destructive interference between paths. (See homework)

EIT and Dispersion

In addition to a modification of the absorption properties of the atom, the coupling laser modifies the real part of $\tilde{\chi}(\delta)$ and thus the dispersion



The rapid variation in Re($\tilde{\chi}(\delta)$) right is the "transparency window" of EIT implies that there is a major difference in the propagation of a pulse in that frequency window compared to pulse propagation in free-space. In particular, the group velocity of pulse is strongly modified.

Recall the group velocity $v_g = \left(\frac{dk}{d\omega}\right)^{-1}$, where $k(\omega) = n(\omega)\frac{\omega}{c}$

$n(\omega) = \sqrt{\epsilon(\omega)} = \sqrt{1 + 4\pi \, Re(\chi(\omega))} \simeq 1 + 2\pi N \, Re(\tilde{\chi})$ is the index of (real) refraction of a gas w/ density N.

$$\Rightarrow \frac{1}{v_g} = \frac{1}{c} + \frac{N}{\lambda} \frac{d}{d\omega} Re(\tilde{\chi}(\omega))$$

Because $\frac{d}{d\omega} Re(\tilde{\chi}(\omega))$ is so huge right at the center of the EIT dip, $v_g \ll c$

In a famous experiment by using ultracold atoms in a BEC, Hau & Harris et al. (Nature 397, 594 (1999)) where able to slow the group velocity to 10^{-7} of c.
The lowest value measured was $v_g = 17\,m/s$