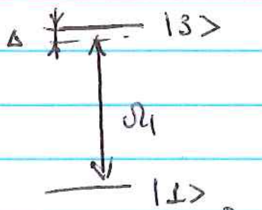
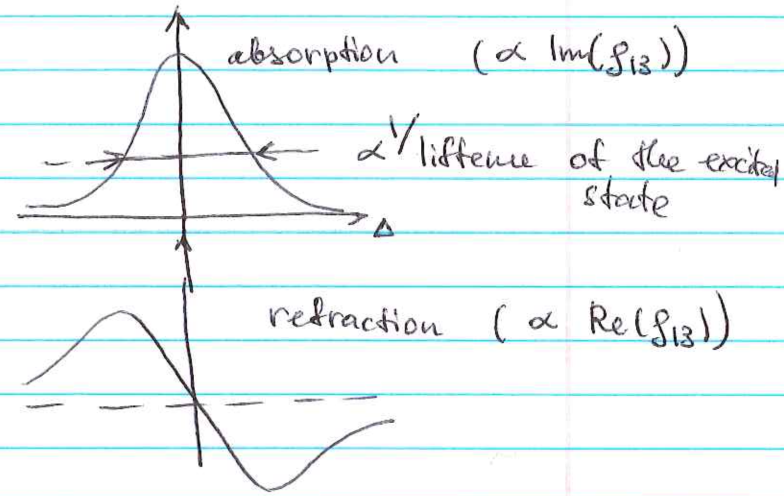
Two-photon resonances - physical meaning

~~Far-detuned~~ Two-level atom

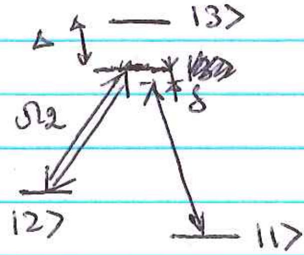


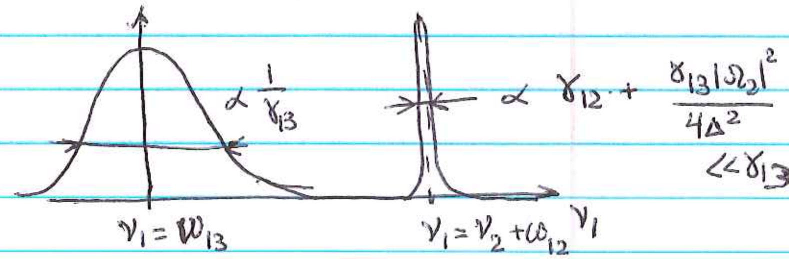
$$\chi(\Delta) = \frac{\wp_{13}^2}{\varepsilon_0 \hbar} \frac{\rho_{21}}{\Omega_1}$$

$\Delta = \nu_1 - \omega_{13}$



Far-detuned three-level system





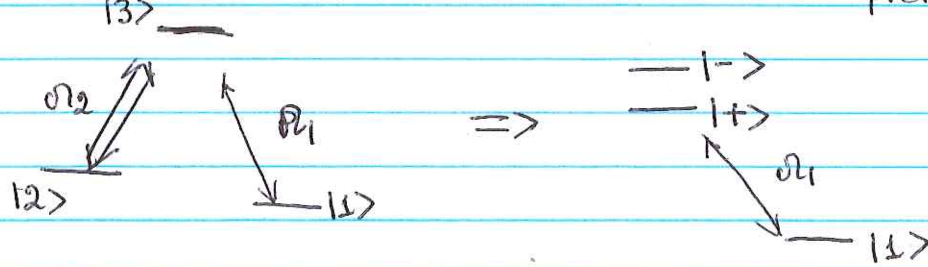
$\Delta = \nu_1 - \omega_{13}$

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

"normal" resonant absorption

two-photon transitions b/w ~~st~~ long-lived states $|1\rangle$ and $|2\rangle$

~~Physical~~ Intuitive explanation - dress-state picture



small!

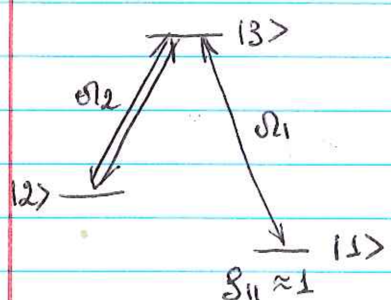
Dressed States $|+\rangle \approx \frac{\Omega_2^*}{2\Delta}|2\rangle + |3\rangle$ mainly $|3\rangle$

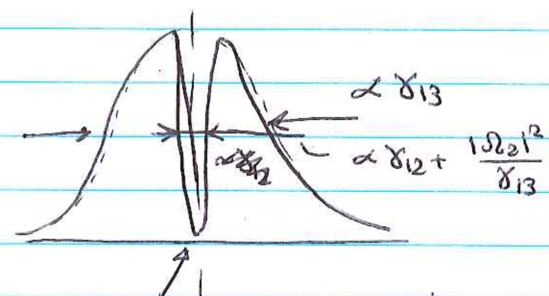
Transitions b/w $|1\rangle$ and $|+\rangle$ are basically equivalent to a normal interaction of resonant light with the 1-3 transition

$|-\rangle \approx |2\rangle - \frac{\Omega_2}{2\Delta}|3\rangle$ mainly $|2\rangle$

Thus, the short lifetime of $|3\rangle$ plays only a small role, and the width of the resonance is basically determined by the lifetime of state $|2\rangle$ (that can be quite long), giving rise to a <u>sub-natural</u> width of absorption resonance.

Resonant two-level system (EIT)





almost perfect transmission

residual absorption suppression $\frac{\gamma_{13}\gamma_{12}}{|\Omega_2|^2}$

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

Such hamiltonian has a non-interacting eigenstate $\hat{H}_{int}|D\rangle = 0$

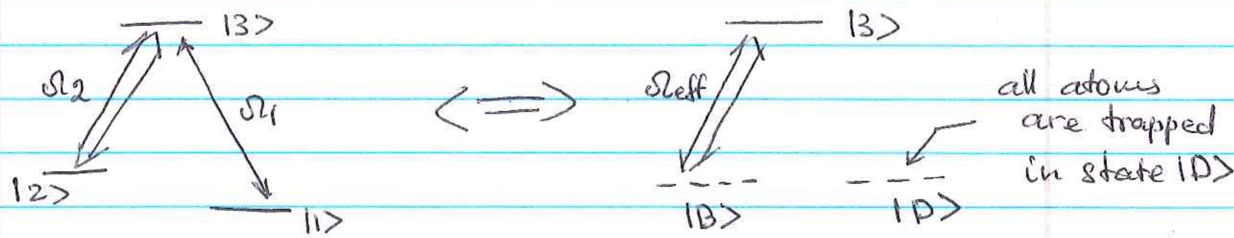
$$|D\rangle = \frac{1}{\sqrt{\Omega_1^2+\Omega_2^2}}\left(\Omega_1|2\rangle - \Omega_2|1\rangle\right)$$ no $|3\rangle$!

long-lived non-interacting state

Orthogonal state — bright state

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

$$\hat{H}_{int} = -\sqrt{\Omega_1^2+\Omega_2^2}\;|B\rangle\langle 3| + c.c.$$



Since $|D\rangle$ is decoupled from light, these atoms do not absorb light

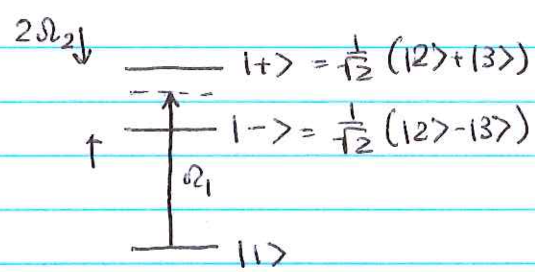
No population in state $|3\rangle$ → no loss

In fact, if there is no transfer b/w $|B\rangle$ and $|D\rangle$ — perfect transparency ($\gamma_{12}=0$)

Only atoms that ~~transition~~ are transfered to $|B\rangle$ via dephasing, for example, can absorb light and end up at $|3\rangle$

Another way to understand EIT — quantum interference

Dressed-state picture



When the probe field is detuned precisely between the two dressed states (EIT conditions) the two possible excitation probability destructively interfere — no absorption

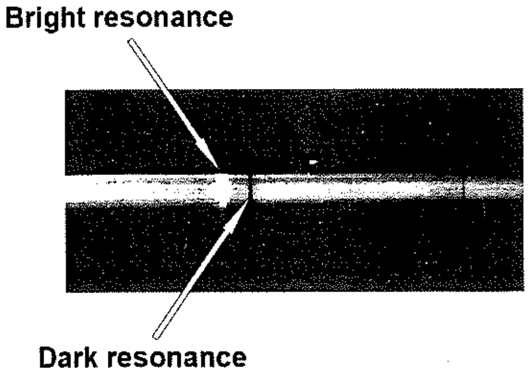


Figure 1. Bright and dark resonances observed in the fluorescence light emitted by a sodium vapor put in a spatially inhomogeneous magnetic field. The dark resonance appears at a point where the frequency difference $\omega_1 - \omega_2$ between two laser modes matches the frequency splitting between two Zeeman sublevels g_1 and g_2.

cooling of molecules. We try in this paper to give a review of the various developments having occurred in this research field, putting the emphasis on the interpretation of the physical phenomena. We will not enter into technical details and our bibliography cannot be considered as exhaustive.

2. Early works on dark resonances

The Pisa experiment [1] was using an optically pumped sodium vapor put in a spatially inhomogeneous magnetic field along the z-axis. The splitting between Zeeman sublevels is thus z-dependent. If one applies a radiofrequency field with frequency $\omega_{\rm RF}$, it induces resonant transitions between two Zeeman sublevels g_1 and g_2 only at the point z where $E_{g_2} - E_{g_1} = \hbar\omega_{\rm RF}$. The population difference between g_1 and g_2 is modified by the RF resonant transitions and this results in a modification of the fluorescence light (generally an increase) at the points where the resonance condition is fulfilled. One observes a series of bright lines in the fluorescence emitted along the path of the laser beam in the cell (see figure 1), forming a spatially resolved RF spectrum.

The important result of this experiment was the appearance of dark resonances (no fluorescence) at certain points along the path of the laser beam (see figure 1), which remain present if the RF field is switched off, provided however that the laser beam is multimode. It was readily found that dark resonances appear for values of z such that the frequency difference between two laser modes is equal to the frequency splitting between two Zeeman sublevels g_1 and g_2 belonging to the hyperfine states of the sodium atom:

$$E_{g_2} - E_{g_1} = \hbar\omega_1 - \hbar\omega_2. \tag{1}$$

This equation is a resonance condition for the stimulated Raman transitions between g_1 and g_2. One photon is absorbed in one laser mode, another one emitted in a stimulated way in another mode, the atom going from one Zeeman sublevel to another one with conservation of the total energy.

The first theoretical treatment of dark resonances [2] was using optical Bloch equations (OBE). A three-level atom $\{g_1, g_2, e\}$ is interacting with two laser beams: $\vec{E}_1 \exp\left(\mathrm{i}\,\vec{k}_1 \cdot \vec{r} - \omega_1 t\right)$ exciting only the transition $g_1 \leftrightarrow e$ and $\vec{E}_2 \exp\left(\mathrm{i}\,\vec{k}_2 \cdot \vec{r} - \omega_2 t\right)$ exciting only the transition $g_2 \leftrightarrow e$. Within the rotating wave approximation, OBE become a set of first order coupled differential equations with time-independent coefficients which can be solved exactly. One finds that, when the detuning Δ from the resonance Raman condition is equal to 0, the population σ_{ee} of the excited state e vanishes: there is no fluorescence. Simultaneously, the off diagonal element $\sigma_{g_1g_2}$ of the atomic density matrix σ between g_1 and g_2 takes a large value. This means that atoms are put in a linear combination of the two lower states g_1 and g_2. The OBE approach gives a quantitative description of the dark resonances but the physical reason why condition (1) is essential for the quenching of fluorescence does not appear clearly. We present in the following section a simple physical interpretation of this Raman resonance condition. Let us also mention that dark resonances can be simply related to the radiative cascade of the atom 'dressed' by the two types of photons ω_1 and ω_2 (see for example [3], chapter VI and [4]).

3. Interpretation of the Raman resonance condition

We consider in this section an atom at rest in $\vec{r} = \vec{0}$. We will discuss later on the case of a moving atom.

(i) Expression of the dark state at time $t = 0$.

Consider an atom in the state:

$$|\psi(t = 0)\rangle = c_1\,|g_1\rangle + c_2|g_2\rangle \tag{2}$$

$c_1(c_2)$ is the amplitude for the atom to be in $g_1(g_2)$. Let us introduce the Rabi frequencies characterizing the interaction with the two laser fields:

$$\Omega_i(t = 0) = -\vec{D}_{eg_i} \cdot \vec{E}_i/\hbar \qquad i = 1, 2. \tag{3}$$

where $\vec{D}_{eg_i}$ is the matrix element of the dipole moment operator between e and g_i. The amplitude to have an absorption process from g_i to e is equal to the amplitude c_i for the atom to be in g_i times the amplitude to absorb a photon from g_i which is proportional to Ω_i. If the amplitude c_1 and c_2 appearing in equation (2) are such that:

$$c_1\,\Omega_1 + c_2\,\Omega_2 = 0 \tag{4}$$

the two absorption amplitudes interfere destructively and the atom cannot be excited. A state (2) obeying condition (4) is called a dark state.

(ii) If a state is dark at time $t = 0$, does it remain dark at a later time?

We suppose that the state remains dark as time goes on and we try to find the condition for this to happen. If the state remains dark, this means that there is no interaction between the atom and the laser fields and the time evolution of the atom is a free evolution as if the laser fields were switched

Cold vs hot atoms: the effect of the Doppler broadening

Moving atoms

$\Delta_1 \longrightarrow \Delta_1 - \vec{k}_1 \cdot \vec{v}$

$\Delta_2 \longrightarrow \Delta_2 - \vec{k}_2 \cdot \vec{v}$

$\delta \rightarrow \delta - (\vec{k}_1 - \vec{k}_2)\vec{v}$

Cold atoms $v \rightarrow 0$, no restrictions on mutual orientations of the two optical fields. Hot atoms $\vec{k}\cdot\vec{v} \sim 500\,MHz$

~~In general,~~ Typical EIT / Raman linewidth $\lesssim 1\,MHz$

In general, misalignment of the beams can completely wash out the EIT resonance unless $\vec{k}_1 - \vec{k}_2$ is minimized $\Rightarrow \vec{k}_1 \uparrow\uparrow \vec{k}_2$

Co-propagating geometry (for Λ and V system)

Residual Doppler broadening $(k_1 - k_2)v = (\nu_1 - \nu_2)\cdot\frac{v}{c}$

$\approx 10\,GHz \cdot \frac{300\,m/s}{3\cdot 10^8\,m/s} \sim 10\,kHz$

However, it is possible to obtain much narrower resonances by restricting the motion of atoms using a buffer gas - Dicky narrowing.

Ladder system

Two-photon resonance $\nu_1 + \nu_2 = \omega_{12}$

$\nu_1 + \nu_2 - \vec{k}_1\vec{v} - \vec{k}_2\vec{v} = \omega_{12}$

$(\vec{k}_1 + \vec{k}_2)\vec{v} \rightarrow 0 \qquad \vec{k}_1 = -\vec{k}_2$

Counter-propagating geometry for Doppler-free configuration