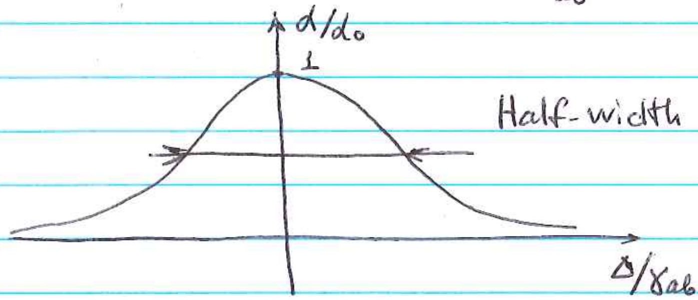
Inhomogeneous broadening

Previously we have calculated the absorption coefficient

$$\alpha = \alpha_0 \cdot \frac{L(\Delta)}{1 + I\,L(\Delta)} \qquad L(\Delta) = \frac{\gamma_{ab}^2}{\gamma_{ab}^2 + \Delta^2}$$

Unsaturated regime $I \ll 1$

$$\alpha = \alpha_0 \cdot L(\Delta) = \alpha_0 \frac{\gamma_{ab}^2}{\gamma_{ab}^2 + \Delta^2}$$



Half-width half maximum (HWHM) γ_{ab}

Saturated regime

$\Delta = 0$ $\quad \alpha = \frac{\alpha_0}{1+I}$ $\qquad$ HWHM width: $\gamma_{ab}\sqrt{1+I}$

power broadening

This is <u>homogeneous</u> broadening: it is the same for all atoms

Inhomogeneous broadening: due to difference in different groups of atoms

Example: Doppler broadening, due to atomic velocity distribution

$$n(v_z) = \frac{N}{V} e^{-v_z^2/v_T^2} \frac{1}{\sqrt{\pi} \cdot v_T}$$

A moving atom experiences a Doppler shift
$\omega \longrightarrow \omega - kv_z$
this is equivalent shifting an atomic resonance
$\omega_{ab} \rightarrow \omega_{ab} + kv_z$

Thus, atoms moving with different velocities absorb light differently

$$\alpha = \alpha_0 \int_{-\infty}^{+\infty} \underbrace{\frac{L(\Delta - kv_z)}{1 + I L(\Delta - kv_z)}}_{\text{width } \propto \gamma_{ab}(1+I)} \underbrace{n(v_z)}_{\substack{\text{width} \\ \sim kv_T}} \, dv_z$$

Two limiting cases : $\gamma_{ab} >> kv_T$ (cold atoms)
negligible inhomogeneous broadening

$\gamma_{ab} << kv_T$ hot atoms
Lorentzian function is very narrow $\longrightarrow$ almost like δ- function at $kv_z \approx \Delta$

$$\alpha = \alpha_0^{(inhom)} e^{-\Delta^2/\Delta_D^2} \qquad \alpha_0^{(inhom)} = \alpha_0 \frac{\gamma^{(hom)}}{\Delta_D}$$

$\Delta_D = kv_T$

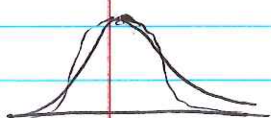
less absorption than that of a homogeneously - broadened ensemble of the same atomic depth

In general, there is no analytical expression for the convolution integral (sometimes it is referred as a Voight profile)

Theorists' life hack for calculations

Sometimese it is possible to obtain more or less accutate analytical results with Doppler breadening, if Gaussian Doppler profile is replaced with Lorentzian

$$\alpha(\Delta) = \int \alpha^{(hom)}(\Delta - kv_z) \cdot \underbrace{e^{-(kv_z)^2/\Delta_D^2} \frac{d(kv_z)}{\sqrt{\pi}\,\Delta_D}}_{\frac{1}{\pi}\frac{\Delta_D^2}{(kv_z)^2 + \Delta_D^2}\frac{d(kv_z)}{\Delta_D}}$$



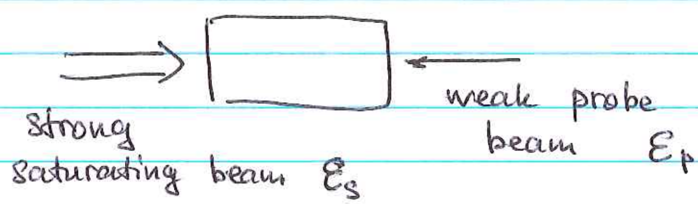
This integral can be calulated using residee theorem

Saturation spectroscopy

Incoherent nonlinear multy-photon interaction

Allows measuring the homogeneous linewidth within an inhomogeneously-broadened medium

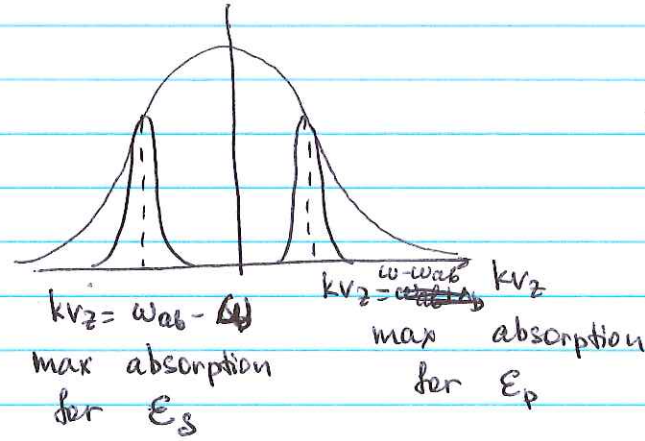
Two counter-propagating beams



$$E_{total} = \mathcal{E}_s e^{ikz - i\omega t} + \mathcal{E}_p e^{-i\omega t - ikz} + c.c.$$

The strong saturating field modifies population distribution of atoms.
Probe is weak so by itself it does not produse saturation (linear regime), but its absorption is affected by the strong field.

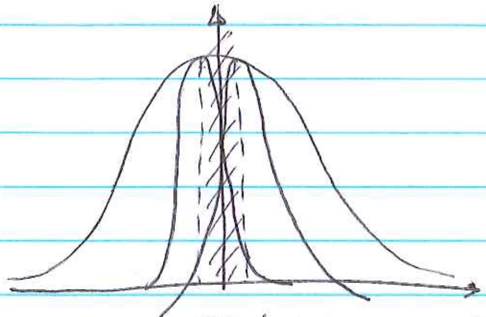
In an inhomogeneously-broadened medium two counter-propagating fields may interact with different velocity groups of atoms



~~peaks~~
resonance conditions

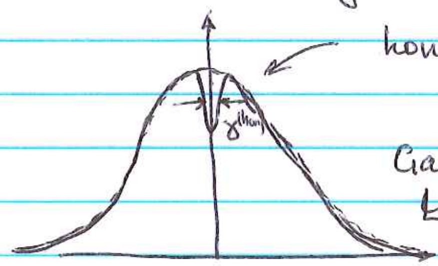
$$\omega \pm kv_z = \omega_{ab}$$

However, if $|\omega - \omega_{ab}| \lesssim \gamma^{(hom)}$, same atoms "see" both optical fields



overlapping velocity groups (around $kv_z = 0$)

We expect that for this case the absorption of the probe field is affected (reduced) by the saturating optical field



homogeneously broadened dip (Lamb dip)

Gaussian profile (for $\gamma_{ab} \ll \Delta_D$)

~~Lorentzian profile~~

$\omega - \omega_{ab}$
laser detuning

Let's conduct the more quantitative calculations

Probe absorption

$$\alpha_p = k \, \mathrm{Im}(\chi_p) = k \, \mathrm{Im}\left(\frac{P_-}{\varepsilon_0 \mathcal{E}_p}\right)$$

$$P = \underbrace{P_+ e^{-i\omega t + ikz}}_{\text{along } \mathcal{E}_s} + \underbrace{P_- e^{-i\omega t - ikz}}_{\text{along } \mathcal{E}_p} + c.c$$

~~[illegible]~~ Correspondingly $\quad P_\pm^{(v_z)} = n(v_z) \, \wp_{ab} \, \rho_{ab}^{(v_z)\pm}$

$$\rho_{ab}^{(v_z)} = \underbrace{\rho_{ab}^{(v_z)+}}_{\sim \mathcal{E}_s} + \underbrace{\rho_{ab}^{(v_z)-}}_{\sim \mathcal{E}_p}$$

$$\rho_{ab}^{(v_z)-} \approx \frac{1}{2} i\Omega_p \frac{(\rho_{aa}-\rho_{bb})^{(v_z)}}{\gamma_{ab} - i(\Delta + kv_z)} \qquad \Delta = \omega - \omega_{ab}$$

We assume that $(\rho_{aa}-\rho_{bb})^{(v_z)}$ is controlled by the strong saturating field

$$(\rho_{aa}-\rho_{bb})^{(v_z)} = \frac{1}{1 + I_s L(\Delta - kv_z)}$$

I_s - dimentionless intensity of $\mathcal{E}_s$

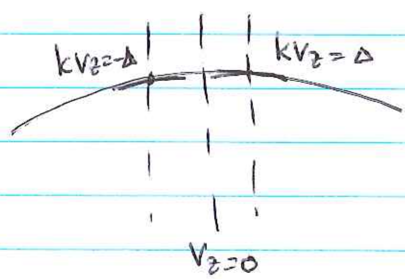
Thus, $$\rho_{ab}^{(v_z)-} \approx \underbrace{\frac{i\Omega_p}{2} \frac{1}{\gamma_{ab} - i(\Delta + kv_z)}}_{\text{centered at } \Delta = -kv_z} \underbrace{\frac{\gamma_{ab}^2 + (\Delta - kv_z)^2}{\gamma_{ab}^2 + (\Delta - kv_z)^2 + |\Omega_s|^2}}_{\text{centered at } \Delta = kv_z}$$

Susceptibility

$$\chi_-(\Delta) = \frac{|\rho_{ab}|^2}{\varepsilon_0 \hbar} \int dv_z \, n(v_z) \, \rho_{ab}^{(v_z)-}(\Delta)$$

If $|\Delta| >> \gamma_{ab}$, the effect of the saturating field is negligeable around the probe absorption resonance;

~~Near~~ For $|\Delta| < \gamma_{ab} \Rightarrow v_z \approx 0$, we can neglect slow variation of atomic density $n(v_z)$



$n(v_z) \approx n$

also, assuming relatively weak saturation

$$\frac{\gamma_{ab}^2 + (\Delta - kv_z)^2}{\gamma_{ab}^2 + (\Delta - kv_z)^2 + |\Omega_s|^2} \simeq 1 - \frac{|\Omega_s|^2}{\gamma_{ab}^2 + (\Delta - kv_z)^2 + |\Omega_s|^2}$$

$$\chi_-(\Delta) \approx \frac{|\rho_{ab}|^2}{\varepsilon_0 \hbar} n \int dv_z \frac{i}{\gamma_{ab} - i(\Delta + kv_z)} \left(1 - \frac{|\Omega_s|^2}{\gamma_{ab}^2 + (\Delta - kv_z)^2 + |\Omega_s|^2}\right)$$

First term – "regular" unsaturated inhomogeneously broadened absorption $\alpha_p(\Delta)$; second term – ~~eff~~ change due to the presence of the saturating beam

$$\alpha = k\, \mathrm{Im}(\chi_-(\Delta)) = \alpha_p(\Delta) -$$

$$- \frac{|\wp_{ab}|^2}{\varepsilon_0 \hbar} n \int d(kv_z) \frac{\gamma_{ab}}{\gamma_{ab}^2 + (\Delta + kv_z)^2} \frac{|\Omega_s|^2}{\gamma_{ab}^2 + (\Delta + kv_z)^2} =$$

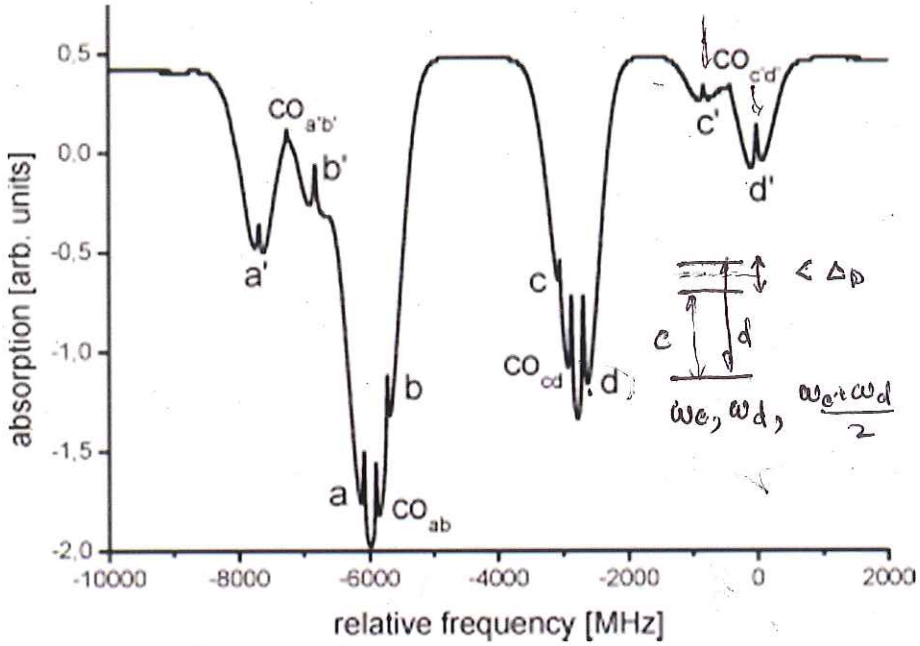
$$= \alpha_p(\Delta) - \frac{\pi |\wp_{ab}|^2}{2\varepsilon_0 \hbar} n \frac{|\Omega_s|^2}{\gamma_{ab}^2 + \Delta^2}$$

$$\left\{ \int_{-\infty}^{+\infty} \frac{1}{1+(x-a)^2} \frac{1}{1+(x+a)^2} dx = \frac{\pi}{2(1+a^2)} \right\}$$ handy math

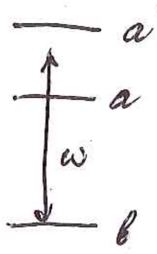
$$\alpha - \alpha_p = -\frac{\pi}{2} \frac{n |\wp_{ab}|^2}{\varepsilon_0 \hbar} \frac{|\Omega_s|^2}{\gamma_{ab}^2 + \Delta^2}$$

Reduction of absorption (~~de~~ Lamb dip)
= peak in transmission
HWHM of these extra feature – γ_{ab}
(homogeneous linewidth)

Saturation spectroscopy is an example of incoherent nonlinear interaction: the strong optical field affects the populations of the atomic levels, but a coherence, created by one field, does not affect the coherence ~~of~~ for the other. Thus, relative phase of two fields is not important.



Cross-over resonances occure when more than one optical transition is overlapped within the Doppler-broadened line.
In this case, the same group of atoms can interact with the saturating field at one transition, and with the probe field on the other.



If $kv_z = \omega - \omega_{ab}$ and $kv_z = \omega_{a'b} - \omega$

$\Rightarrow$ cross-over $\omega_{co} = \dfrac{\omega_{ab} + \omega_{a'b}}{2}$