

$\hookrightarrow$  density  $n$

$$\frac{dI}{dz} = -\alpha I(z)$$

$$I(z) = I_0 e^{-\alpha z} \quad \text{Beer's Law}$$

$\alpha$  = absorption coefficient

$\alpha L$  = absorption length

$$\alpha = n \sigma(\omega)$$

$\hookrightarrow$  cross section

for single resonance @  $\omega_0$

$$\sigma(\omega) = \sigma_0 \frac{\gamma/2\pi}{(\omega - \omega_0)^2 + (\gamma/2)^2}$$

complex index of refraction  $\equiv n = n' - iK$

consider EM <sup>wave</sup> passing through medium of index  $n$

$$\omega_n = \omega_0 \quad k_n = k_0 n$$

$$\begin{aligned} E &= E_0 e^{-k_0 K z} e^{i(\omega t - k_0 n' z)} \\ &= E_0 e^{-2\pi K z / \lambda} e^{i k_0 (c t - n' z)} \end{aligned}$$

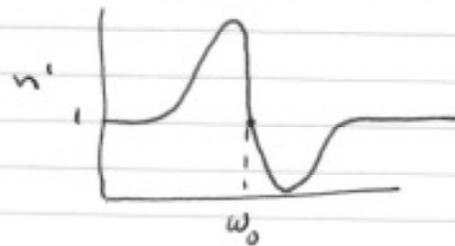
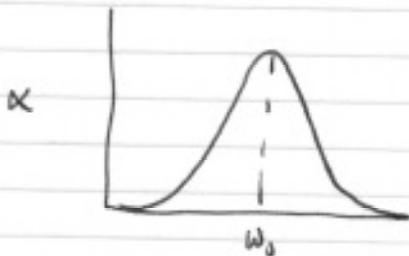
$$I \propto |E|^2 = I_0 e^{-2K k_0 z}$$

$$\Rightarrow \alpha = 2K k_0 = 4\pi K / \lambda$$

$n'$  represents dispersion

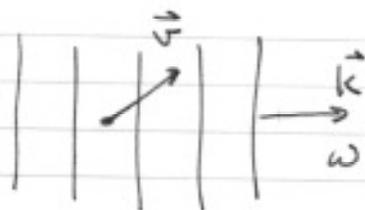
$$n' = 1 + \frac{c}{\omega_0} \left( \frac{\omega_0 - \omega}{\gamma} \right) \alpha(\omega)$$

for  $\omega_0 - \omega \ll \omega_0$



## Doppler broadening

$\omega_0 \equiv$  frequency of wave emitted by stationary atom



in the moving frame of the atom

$$\omega' = \omega - \vec{k} \cdot \vec{v}$$

atom will absorb at  $\omega_0$  in its frame, so the absorption frequency in the lab frame will be

$$\omega_a = \omega_0 + \vec{k} \cdot \vec{v}$$

- assume  $\vec{k} = k \hat{z}$ , so  $\omega_a = \omega_0 (1 + v_z/c)$

for a Boltzmann distribution of gas molecules probability of an atom having  $v_z$

$$P(v_z) dv_z = \sqrt{\frac{M}{2\pi kT}} e^{-Mv_z^2/2kT} dv_z$$

then

$$P(\omega) d\omega = \frac{2}{\sqrt{\pi} \cdot \Delta} e^{-4(\omega - \omega_0)^2/\Delta^2} d\omega$$

$$\Delta \equiv 2 \frac{\omega_0}{c} \sqrt{\frac{2kT}{M}}$$

(4)

$$P(\omega) d\omega = \mathcal{G}(\omega - \omega_0, \Delta) d\omega$$

↑ normalized Gaussian distribution

$$\text{FWHM} = 2 \frac{\omega_0}{c} \left[ \frac{2kT}{m} \ln 2 \right]^{1/2}$$

- this has assumed an arbitrarily narrow atomic response, but we know the atomic line should be a Lorentzian

$$\mathcal{L}(\omega - \omega_0, \gamma) \equiv \frac{\gamma/2\pi}{(\omega - \omega_0)^2 + (\gamma/2)^2}$$

to get full profile

$$\begin{aligned} I(\omega - \omega_0, \gamma, \Delta) &= \int_0^\infty \mathcal{L}(\omega - \omega'_0, \gamma) P(\omega'_0) d\omega'_0 \\ &= \int_0^\infty \mathcal{G}(\omega'_0 - \omega_0, \Delta) \mathcal{L}(\omega'_0 - \omega, \gamma) d\omega'_0 \end{aligned}$$

⇒ Voigt profile

convolution of Lorentzian & Gaussian

- no analytic form

Doppler broadening = inhomogeneous broadening  
 - summing up different conditions for each atom in the ensemble

Lorentzian linewidth = homogeneous broadening  
 - exact same effect on each individual atom

other examples of broadening

magnetic field gradient - inhomogeneous

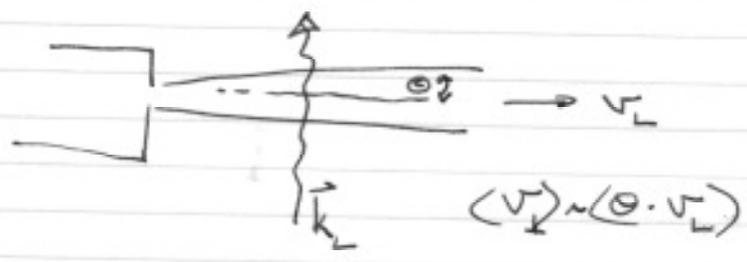
collisions - homogeneous \*

\* - averaged over many rapid collisions

transit time broadening - homogeneous

# Doppler-free spectroscopy

- Atomic beam



$$\omega_a = \omega_0 + \vec{k} \cdot \vec{v}$$

→ expect Doppler width reduced by factor  $\propto \theta$

$$G(\omega_a - \omega_0, \Delta) \text{ where } \Delta = 2 \frac{\omega_0}{c} \sqrt{\frac{2kT_{\perp}}{M}}$$

$$\frac{1}{2} k_B T_{\perp} = \frac{1}{2} m \langle v_{\perp}^2 \rangle$$

- also will have transit time broadening

Assume laser has intensity profile

$$I(r) = I_0 e^{-2r^2/w_0^2}$$

the atoms will see a time-dependent pulse of light

$$I(t) \propto e^{-t^2/2t_0^2}$$

where  $t_0 = \frac{w_0}{v_L}$

# Fast ion beam spectroscopy

- consider a beam of ions, emitted from a source with a thermal velocity spread  $v_{th}$ ,  $\frac{1}{2} M v_{th}^2 = E$
- accelerate them through a constant potential  $U$

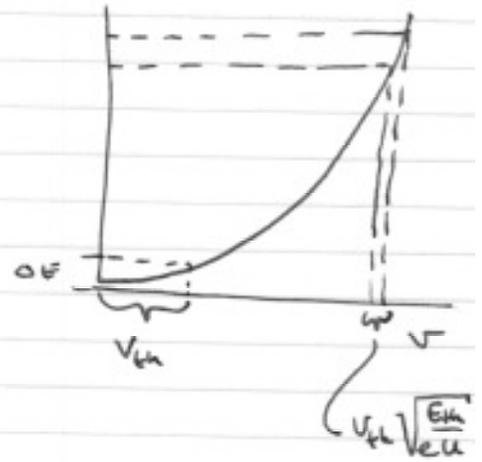
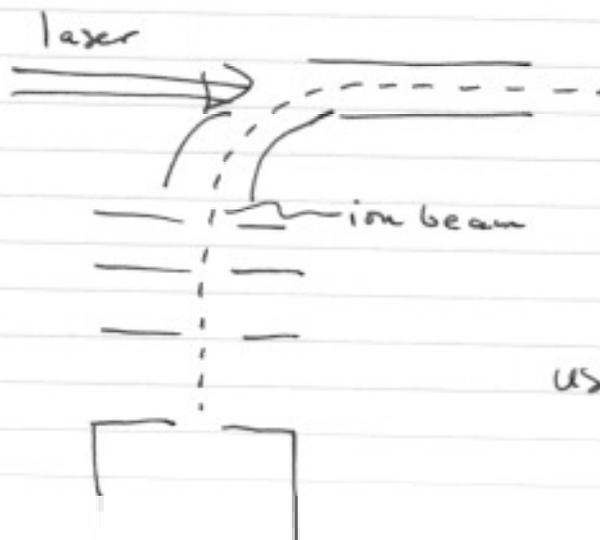
then  $E_f = eU \pm E_{th}$

but  $v_f = \frac{1}{2} \sqrt{\frac{2E_f}{M}} \approx \sqrt{\frac{2eU}{M}} \pm \frac{E_{th}}{\sqrt{\frac{2eU}{M}}}$

so velocity spread has been reduced

← typical 300-1000x reduce

$v_{th} \rightarrow v_{th} \sqrt{\frac{E_{th}}{eU}}$



use longitudinal excitation!

(7)

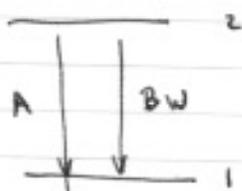
The Fourier width of this time-dependent pulse yields an additional width

$$\Delta\omega_{\text{FF}} = 2 \frac{v}{w_0} \sqrt{2 \ln 2}$$

— make smaller with lower velocities or larger beams

→ see (6A)

— Saturation spectroscopy



$$\alpha = \sigma (N_2 - N_1)$$

recall from earlier lecture,  $N_2 - N_1 \rightarrow 0$  as intensity is increased.  $\Rightarrow$  saturation

$$\alpha = \frac{\alpha_0}{1+S}$$

$S =$  saturation parameter

$$= \frac{2\sigma I(\omega)}{h\nu A_{12}} \quad (W_s(\omega) B_{21} = A_{12})$$

time rate of change of energy density

$$\begin{aligned} \frac{dE}{dt} &= h\nu B_{12} W(\omega) \Delta N = h\nu B_{12} W(\omega) \frac{N_1}{1+S} \\ &= h\nu A_{21} \frac{N_1}{1+S} \end{aligned}$$

introduce

$$S_\omega = \frac{B_{12} W(\omega)}{A_{21}} \mathcal{L}(\omega, \omega_0)$$

→ assume

$$\text{then } S_{\omega} = S_0 \frac{(\gamma/2)^2}{(\omega - \omega_0)^2 + (\gamma/2)^2} \quad \text{where } S_0 = S_{\omega}(\omega_0)$$

$$\text{then } \frac{dE}{dt} = \frac{h \omega A_{21} N_1 S_0 (\gamma/2)^2}{(\omega - \omega_0)^2 + (\gamma/2)^2 (1 + S_0)} = \frac{C}{(\omega - \omega_0)^2 + (\gamma_s/2)^2}$$

$$= \text{Lorentzian with width } \gamma_s = \gamma \sqrt{1 + S_0}$$

= power broadening

- exactly as expected - lifetime of state 2, normally given by  $A_{21}^{-1}$  is shortened by stimulated emission,  $B_{21} W(\omega)$ , increasing the observed linewidth.

we can write

$$\alpha_s(\omega) = \alpha_0(\omega_0) \frac{(\gamma/2)^2}{(\omega - \omega_0)^2 + (\gamma_s/2)^2} = \frac{\alpha_0(\omega)}{1 + S_0}$$

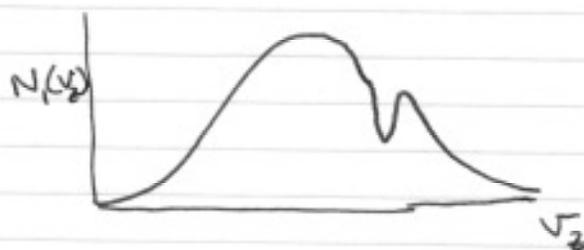
- now consider a Doppler-broadened profile:  
and  $\vec{k} = k \hat{z}$

$$\sigma(\omega, v_z) = \sigma_0 \frac{(\gamma/2)^2}{(\omega - \omega_0 - k v_z)^2 + (\gamma/2)^2}$$

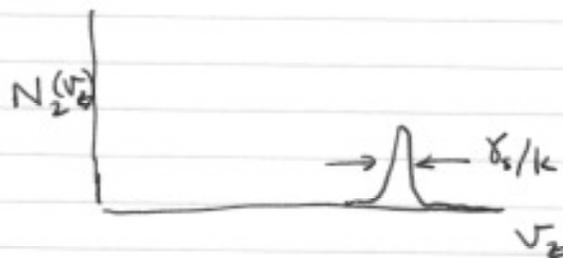
due to saturation, we expect  $N_1(v_z) dv_z$  to be reduced in an interval  $dv_z = \delta/k$  while  $N_2(v_z) dv_z$  is increased.

The population difference can be written as:

$$\Delta N(\omega, v_z) = N_1(v_z) \left[ 1 - \frac{S_0 (\delta/2)^2}{(\omega - \omega_0 - kv_z)^2 + (\delta_s/2)^2} \right]$$



⇒ Hole burning



we can write velocity-dependent contribution to absorption

$$\frac{d\alpha(\omega, v_z)}{dv_z} dv_z = \Delta N(v_z) \sigma(\omega, v_z) dv_z$$

then

$$\alpha(\omega) = \int \Delta N(v_z) \sigma(\omega, v_z) dv_z$$

we find:

$$\alpha(\omega) = \frac{N_1 \sigma_0}{\sqrt{\frac{2\pi kT}{m}}} \int \frac{e^{-\frac{1}{2} m v_z^2 / kT} dv_z}{(\omega - \omega_0 - k v_z)^2 + (\gamma/2)^2}$$

$\Rightarrow$  a Voigt profile with width <sup>parameters</sup>  $\gamma, \Delta$

— even though we burn a hole that is much less than the Doppler width, it does not show up in the absorption profile!

solution: use 2 lasers

A) saturating laser with  $\vec{k}_1$  at  $\omega_1$ ,  
burns hole at  $v_z = \omega_0 - \omega_1 / k_1$

B) weak probe laser with  $\vec{k}_2$  and  $\omega_2$ ,  
which does not effect saturation ( $S_2 \ll 1$ )

then

$$\alpha_p(\omega_1, \omega_2) = \frac{\sigma N_1}{\langle v \rangle \sqrt{\pi}} \int \frac{e^{-(\gamma/2)^2}}{(\omega_0 - \omega_2 - k_2 v_z)^2 + (\gamma/2)^2} \left[ 1 - \frac{S_0 (\gamma/2)^2}{(\omega_0 - \omega_1 - k_1 v_z)^2 + (\gamma/2)^2} \right] dv_z$$

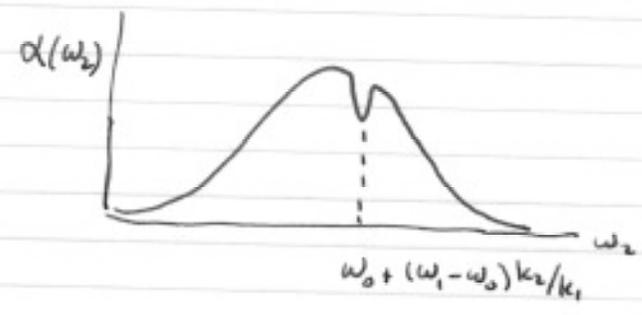
integrating over  $dv_z$  yields:

$$\alpha_p(\omega_1, \omega_2) = \alpha(\omega) \left[ 1 - \frac{S_0 (\gamma/2)^2}{(\omega_0 - \omega_1 - k_1 v_z)^2 + (\gamma/2)^2} \right]$$

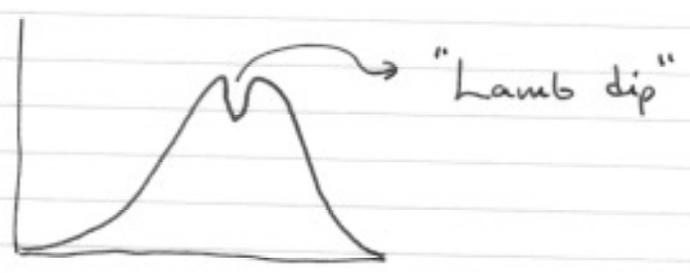
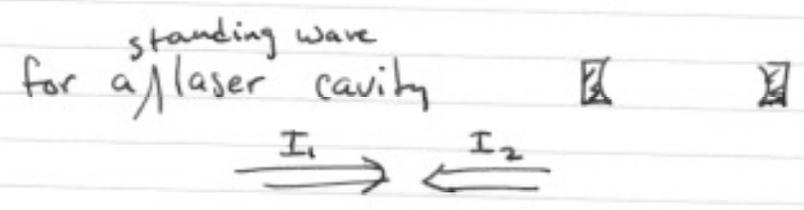
- an unsaturated Doppler profile  $\alpha_0(\omega)$  with a dip at  $\omega = \omega_0 \pm (\omega_1 - \omega_0) k_1/k_2$

where + = colinear - counterpropagating

and  $\gamma_s'' = \gamma + \gamma_s$



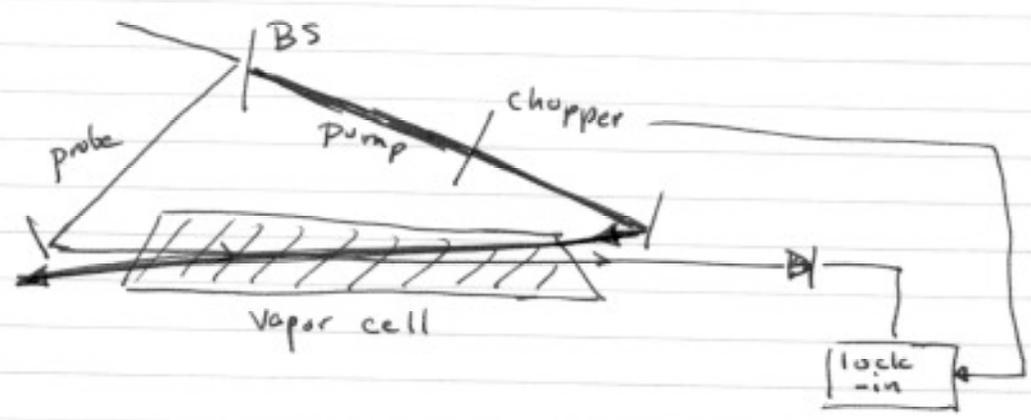
=> allows observation of sub-Doppler linewidth



dip comes from atoms moving  $\perp$  to laser beams, which interact with both beams - because of non-linearity of saturation, dip appears.



an experimental example of saturation spectroscopy

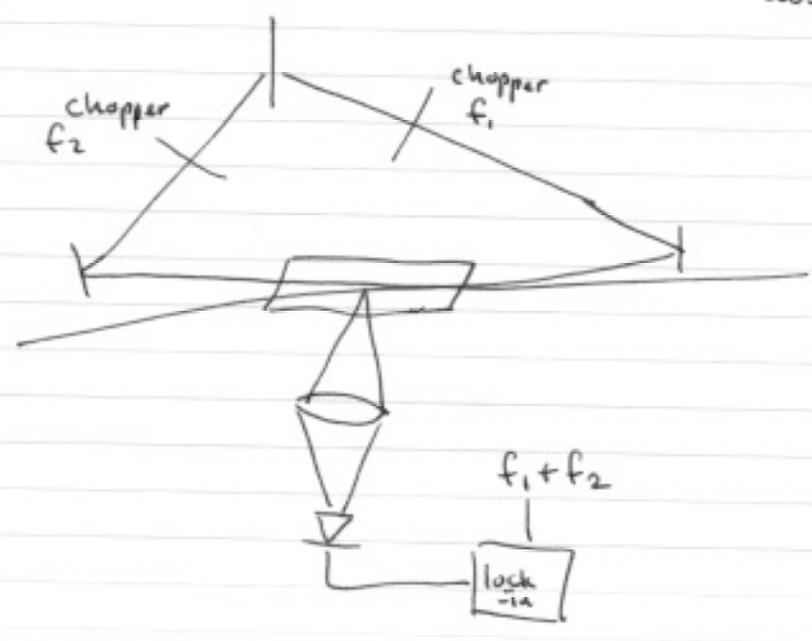


- chopping of pump & detecting synchronously the probe removes the Doppler background.

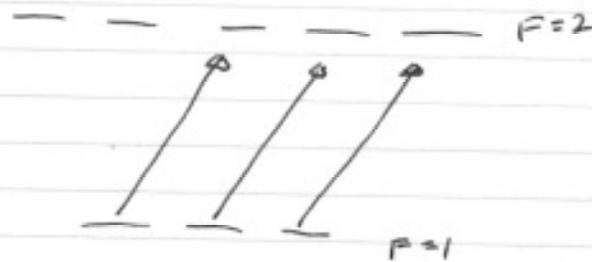
- a related technique

### Intermodulated fluorescence

(good for small absorption (weak signal)

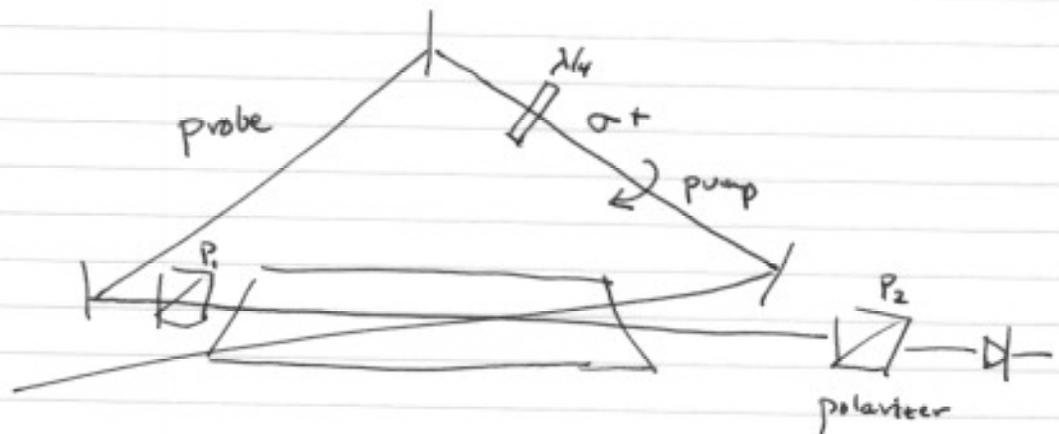


## Polarization Spectroscopy



apply strong  $\sigma^+$  pump  
to sample

- will create m-state population imbalances (optical pumping)
- probe with polarization-sensitive transition



probe is linearly-polarized

$$\vec{E} = E_0 \hat{x} = E_0 \left( \frac{1}{\sqrt{2}} \hat{\sigma}_+ + \frac{1}{\sqrt{2}} \hat{\sigma}_- \right)$$

$$(\hat{\sigma}_+ = \hat{x} + i\hat{y} \quad \hat{\sigma}_- = \hat{x} - i\hat{y})$$

set  $P_2$  to be crossed with  $P_1$  (no transmission)

two effects - differential absorption  $\alpha_+ \neq \alpha_-$   
 - differential index  $n_+ \neq n_-$

depending on setting of polarizers



$$\theta = 90^\circ$$

$\hookrightarrow$  angle between  $P_1$  &  $P_2$

$$\theta \neq 90^\circ$$

- again a sub-Doppler technique

pump creates an anisotropic  $m$ -distribution  
 for a particular  $v$ -class; probe measures  
 the anisotropy through polarization.