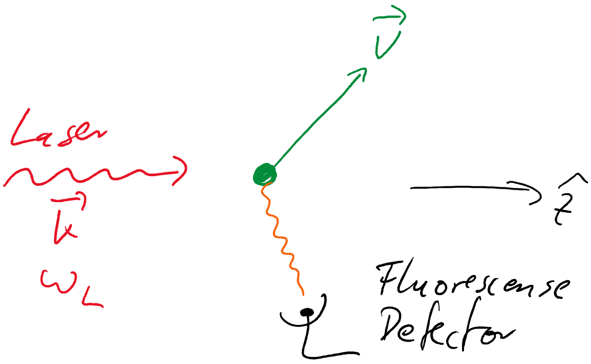
Doppler broadening & Doppler-free spectr.

Demtröder
Laser Spectroscopy Vol 2
Ch. 2: Nonlinear Spectroscopy



$\longrightarrow \hat{z}$ given by laser direction

Laser is Doppler-shifted in the atom's reference frame

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

Thermal distribution of atoms

$$n_i(v_z)\,dz = \frac{N_i}{v_w\sqrt{\pi}}\, e^{-\left(\frac{v_z}{v_w}\right)^2} dv_z$$

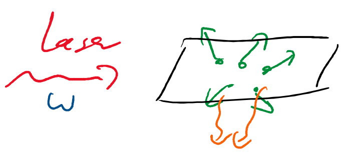
1D Maxwell-Boltzmann
= Gaussian (!)

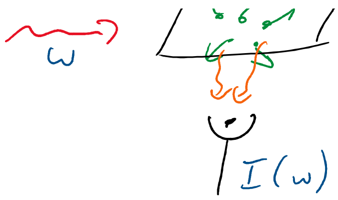
$$v_w = \sqrt{\frac{2kT}{M}} \quad \text{most probable velocity}$$

Number of atoms with resonance frequency within frequency in interval $[\omega, \ldots \omega + d\omega]$

$$n_i(\omega)\,d\omega = N_i \frac{c}{v_w\,\omega_0\sqrt{\pi}}\, e^{-\left(\frac{\omega-\omega_0}{\omega_0 \frac{v_w}{c}}\right)^2} d\omega$$

Measured fluorescence intensity profile $\propto n_i(\omega)$





$$I(\omega) = I(\omega_0)\, e^{-\left(\frac{\omega-\omega_0}{\omega_0 v_w / c}\right)^2} \qquad \text{Gaussian profile}$$

Doppler width $\delta\omega_D$ from $I(\omega) = \frac{I_0}{2}$

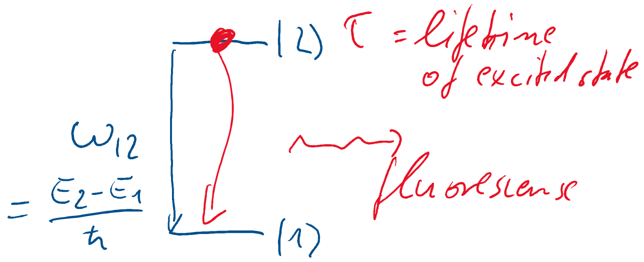
$$\delta\omega_D = 2(\ln 2)^{1/2}\, \omega_0 \frac{v_w}{c}$$

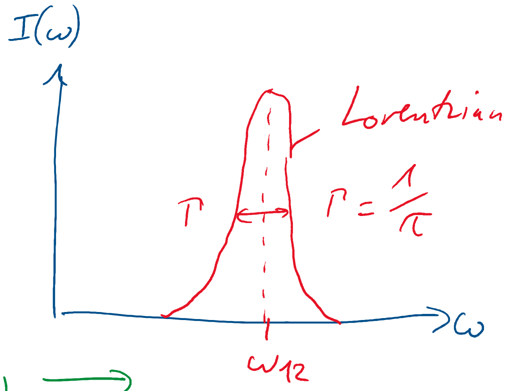
$$\delta\omega_D = \frac{\omega}{c}\sqrt{\frac{8kT\ln 2}{M}}$$

$$I(\omega) \approx I_0 \exp\left[-\frac{\omega-\omega_0}{0.6\,\omega_D}\right]^2$$

$$0.6 \approx \frac{1}{\sqrt{4\ln 2}}$$

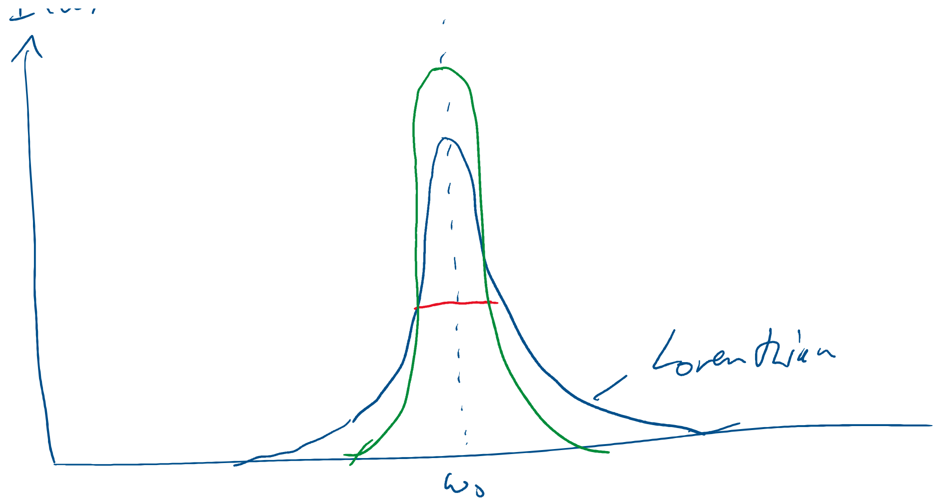
Natural line shape: Lorentzian (lifetime of states)





exponential decay (time domain) ⟶ Fourier Lorentzian frequency domain

Dopple line shape: Gaussian



Observed line shape : Voigt profile

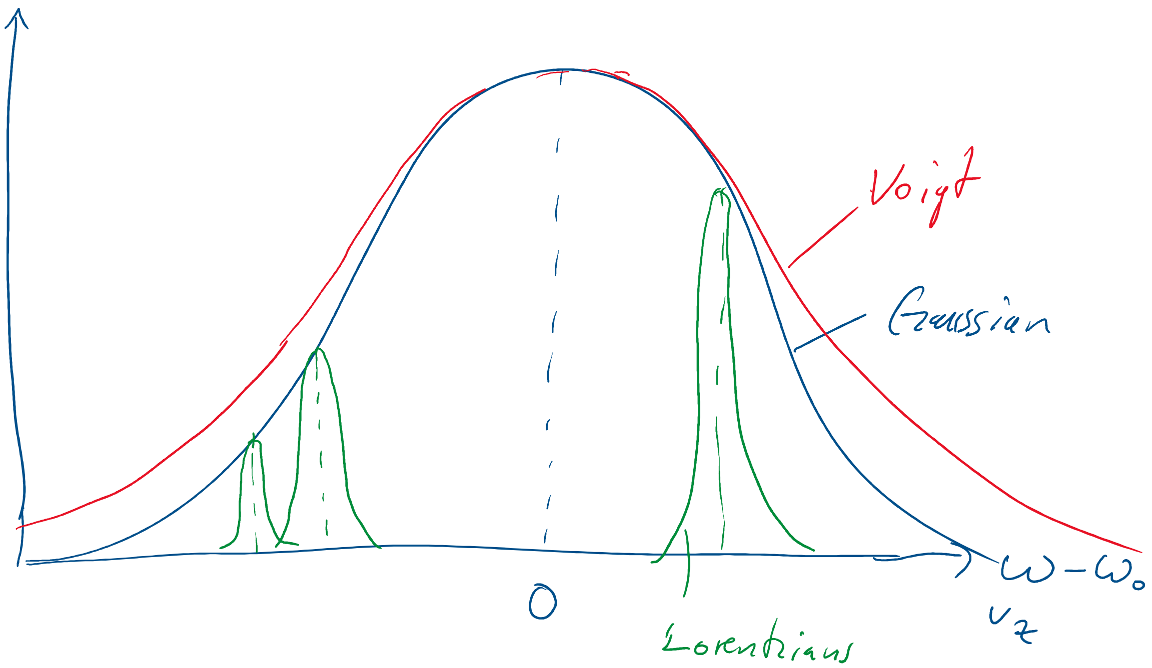
= convolution of Lorentzian & Gaussian

$$I(\omega) = \frac{\gamma I_0 N_1 c}{2\pi^{3/2}\omega_0 v_w} \int_0^\infty \frac{e^{-\frac{c^2(\omega-\omega')^2}{\omega'^2 v_w^2}}}{(\omega-\omega')^2 + \left(\frac{\gamma}{2}\right)^2}\, d\omega'$$

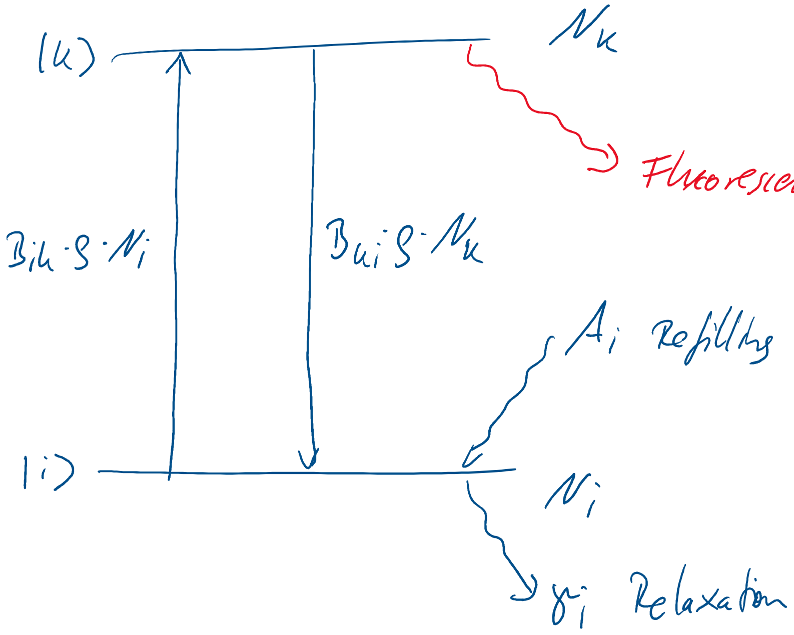
Don't use this to fit a line shape!

There are polynomial approximations

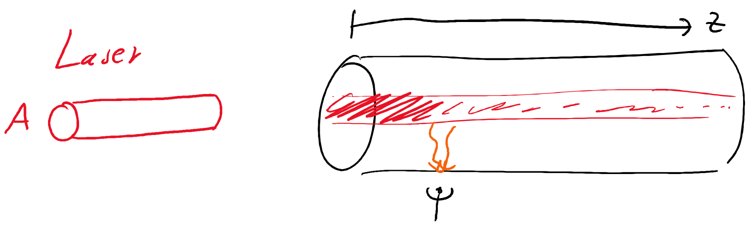
$\hat{=}$ much faster and correct to 10^{-4}



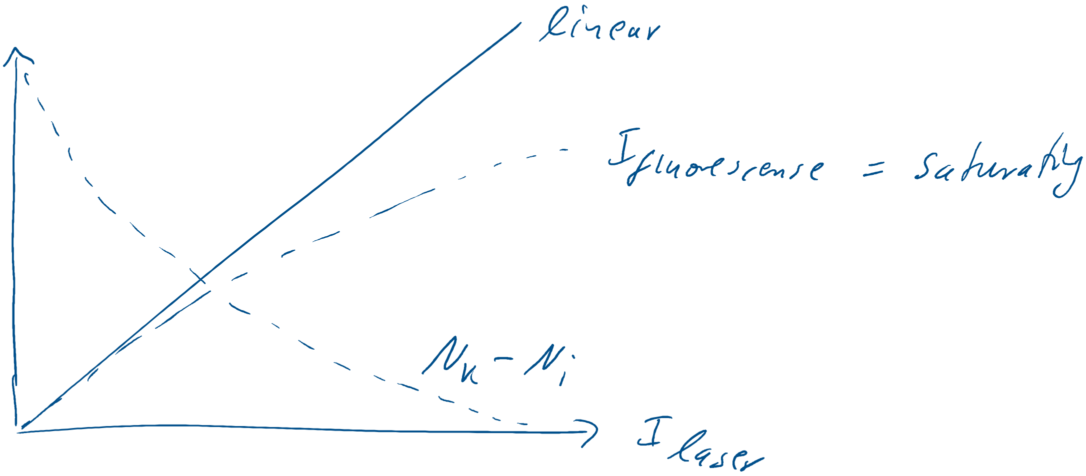
Doppler-free spectroscopy again Demtröder



Linear absorption



$$I_{laser} = \frac{c\varepsilon_0}{2} E_0^2 \qquad E = E_0 \cos(\omega t - kz)$$



Fluorescence in Volume element $dV = A \cdot dz$

Atoms absorb power $dP = A \cdot dI =$

$$dP = A\, I\, \sigma_{ik} \left[N_i - \left(\frac{g_i}{g_k}\right) N_k \right] dz \qquad (*)$$

↑ statistical weights

Rate equations

$$\frac{dN_i}{dt} = B_{ik}\, \rho \left[\frac{g_i}{g_k} N_k - N_i \right] - N_i \gamma + A_i$$

$\rho = \frac{I(\omega)}{c}$ spectral energy density of laser

steady-state solution $\frac{dN_i}{dt} = 0$

$$\boxed{N_i = \frac{A_i}{B_{ik}\rho + \gamma_i} + N_k \frac{\frac{g_i}{g_k} B_{ik}\, \rho}{B_{ik}\rho + \gamma_i}}$$

small laser intensity

$$B_{ik} \frac{I}{c} \ll \gamma_i$$

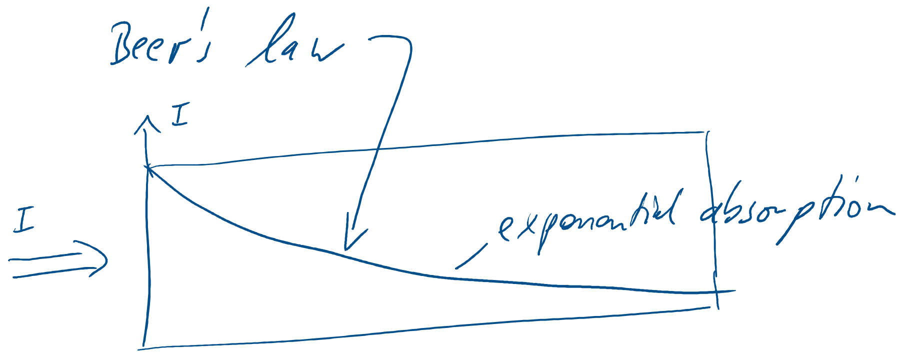
usually $N_k \ll N_i$

$\Rightarrow$ solution

atoms in the g.s. $N_i = \frac{A_i}{\gamma_i}$

Integrate $(*)$ $\Rightarrow$

$$P = P_0\, e^{-\left[N_i - \frac{g_i}{g_k} N_k\right] \sigma_{ik} \cdot z} = P_0\, e^{-\text{const} \cdot z}$$



Large I (Laser)

$$N_i = \frac{N_i^{(0)}}{1 + aI} \approx N_i^{(0)} (1 - aI)$$

$$a = \frac{B_{ik}}{c \gamma_i}$$

$\Rightarrow$ $N_i(I)$ decreases with increasing laser power

$$dP = A \sigma_{ik} N_i^{(0)} (I - aI^2)\, dz$$

lin   quadratic in I

very large intensities $B_{ik} \frac{I}{c} \gg \gamma_i$

$$N_i \simeq \frac{A_i}{B_{ik} I/c} + \frac{g_i}{g_k} N_k$$

increasing intensity reduces 1st term

For $I \longrightarrow \infty$ the absorption goes to zero!

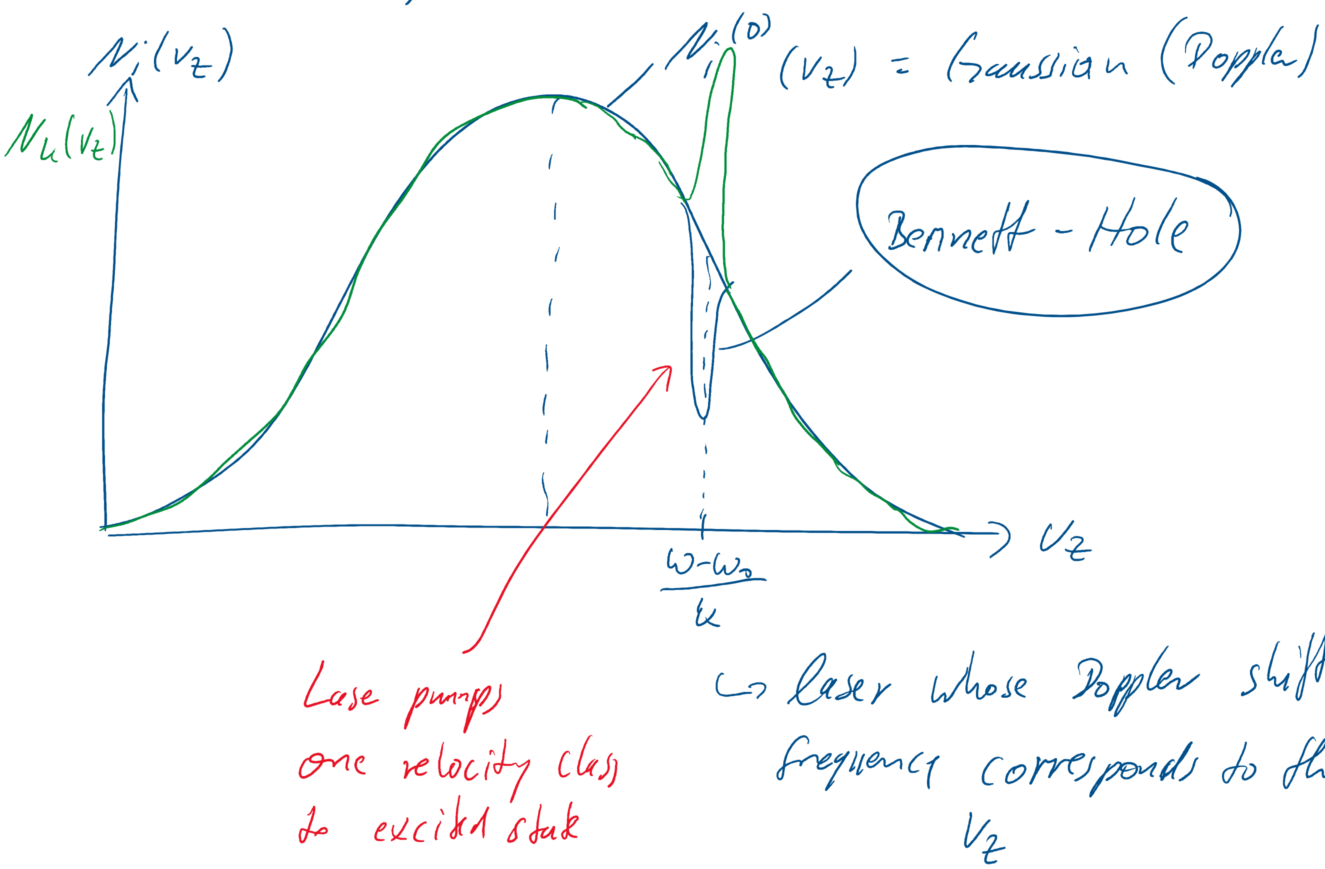
Atomic sample becomes transparent.

Absorption reduces g.s. occupation $N_i(v_z)$

enhances e.s. " $N_k(v_z)$

$$N_i(v_z) = N_i^{(0)}(v_z) - \Delta N^0 \frac{v_z}{\gamma_i \tau} \underbrace{\frac{S_0 \left(\frac{\gamma}{2}\right)^2}{(\omega - \omega_0 - k v_z)^2 + \left(\frac{\gamma_s}{2}\right)^2}}$$

$$N_k(v_z) = N_k^{(0)}(v_z) + \ldots\ldots$$



↳ laser whose Doppler shifted frequency corresponds to this v_z

Bennet Hole = depletion of atoms in the ground state with $v_z = \frac{\omega - \omega_0}{k}$

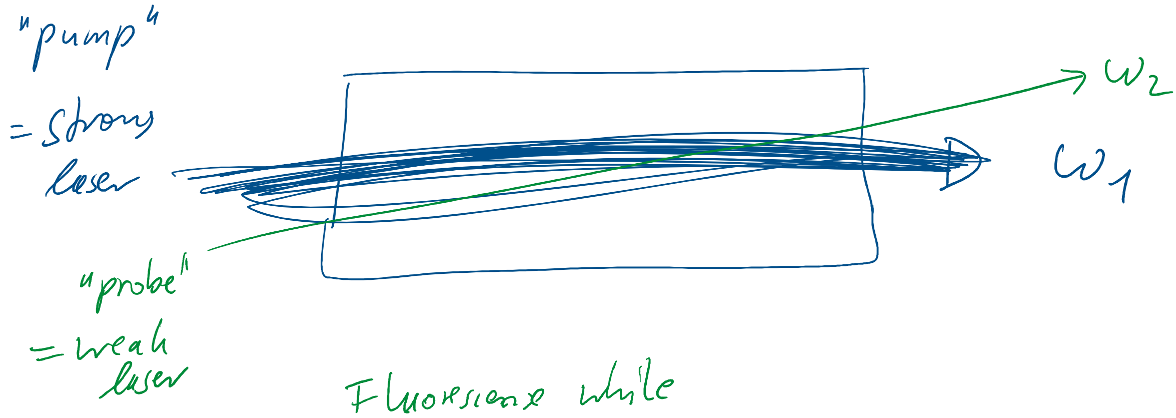
width $\gamma_s = \gamma_0 (1 + S_0)^{\frac{1}{2}}$

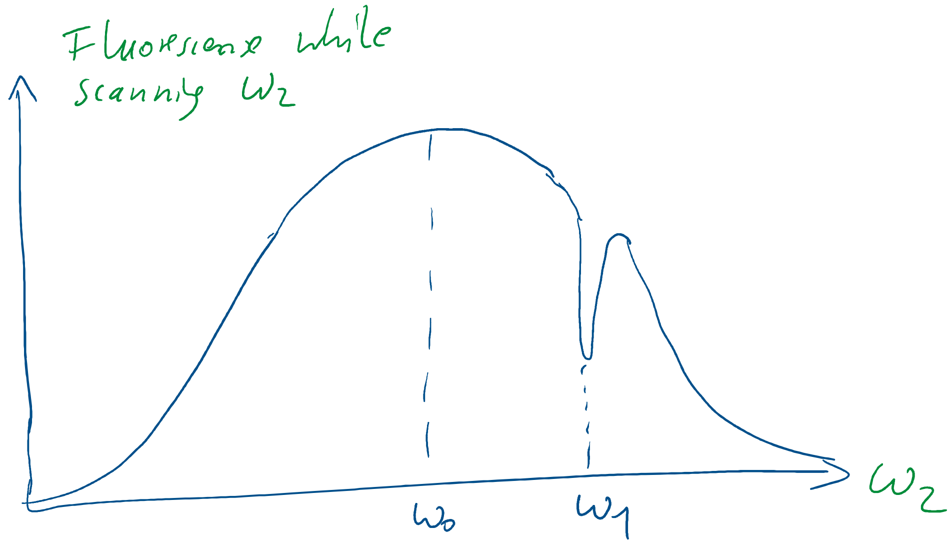
depth $\Delta N(v_z) \propto \frac{S_0}{1 + S_0}$

S_0 = Saturation parameter $\propto$ laser intensity

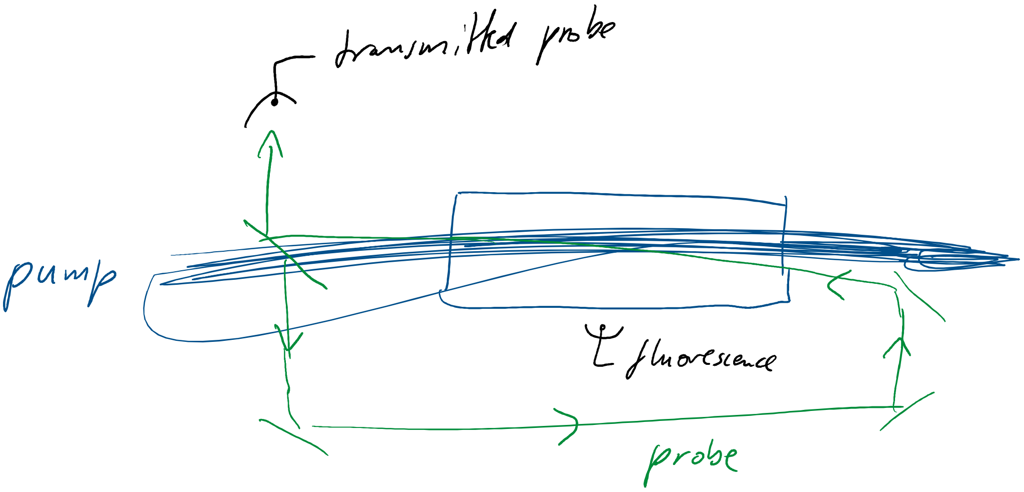
$\propto$ laser intensity

You can observe the Bennet Hole
with a 2nd laser

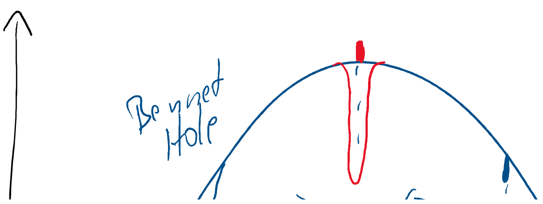


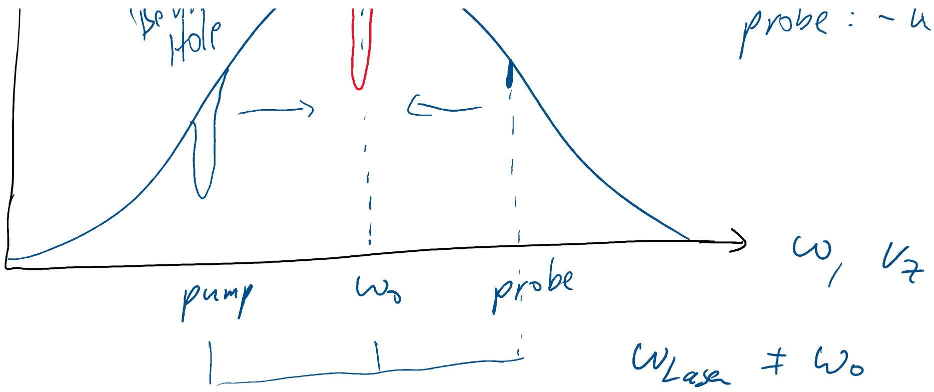


Lamb-dip



Doppler : $\omega' = \omega_0 - k \cdot v_z$
pump : k
probe : $-k$

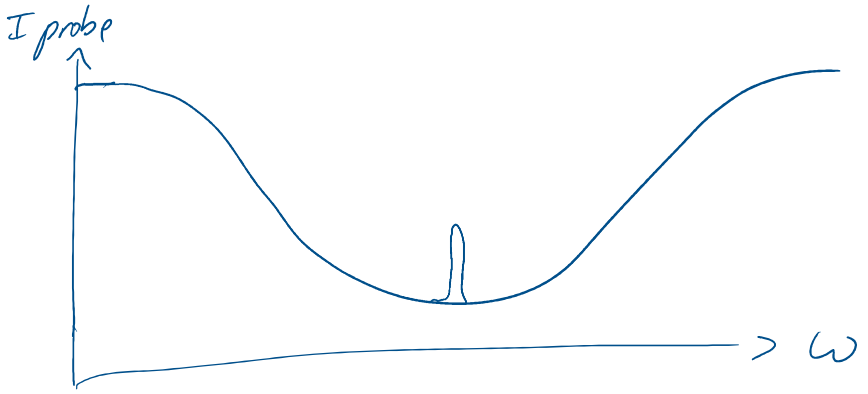




$\omega_{Laser} \neq \omega_0$
Bennet hole at $v_z \neq 0$
probe samples atoms at $-v_z$

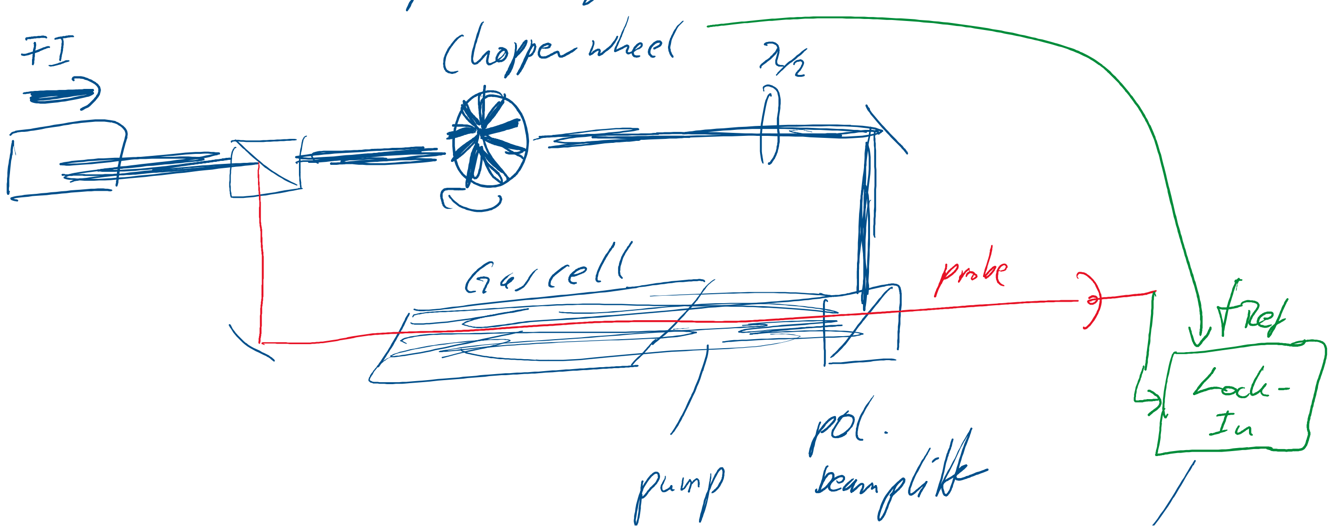
laser on resonance:
pump creates Bennet-Hole for $v_z = 0$
probe samples atoms at $v_z = 0$

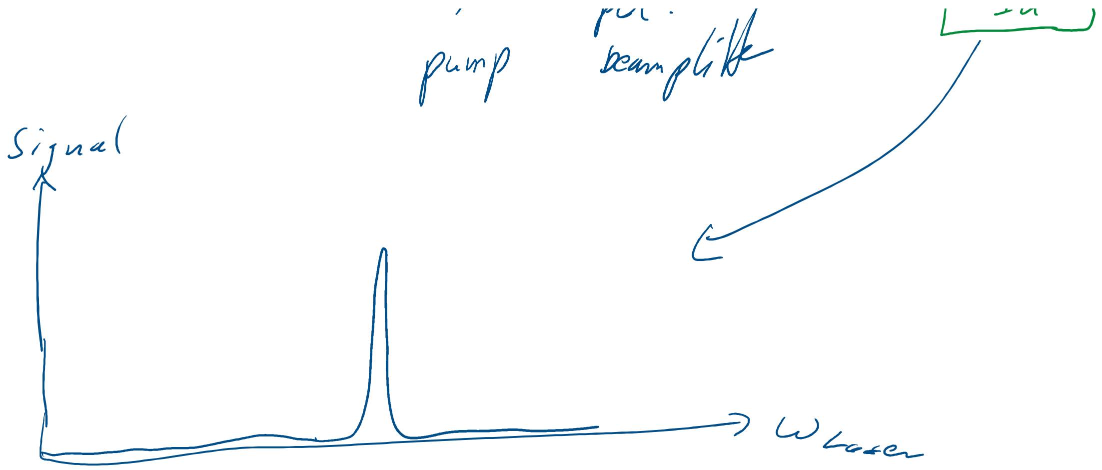
absorption of probe beam



Doppler-free signal
at atomic resonance frequency

Saturated absorption spectroscopy



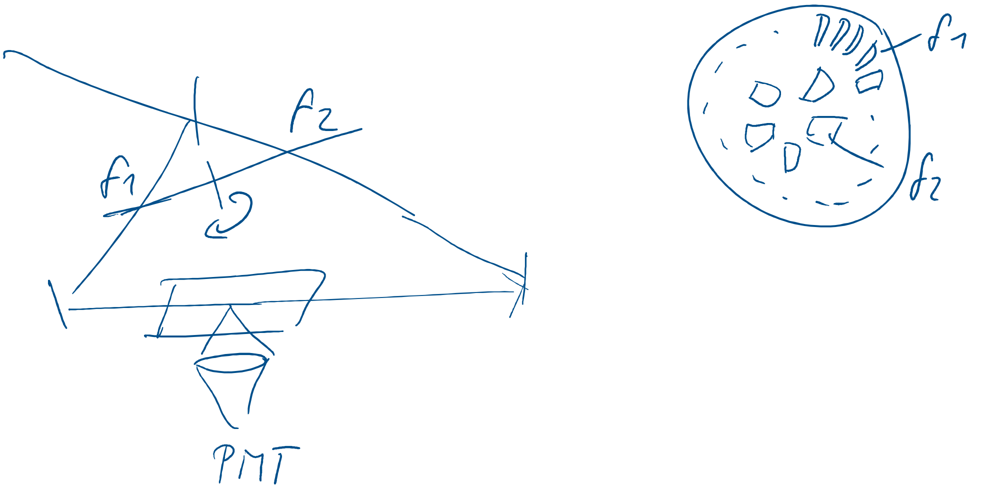


Chopper on pump change Bennett Hole

→ probe absorption is modified

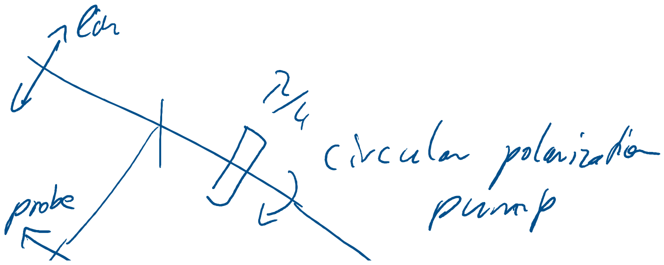
Lock-In Amplifier gives good signal/noise

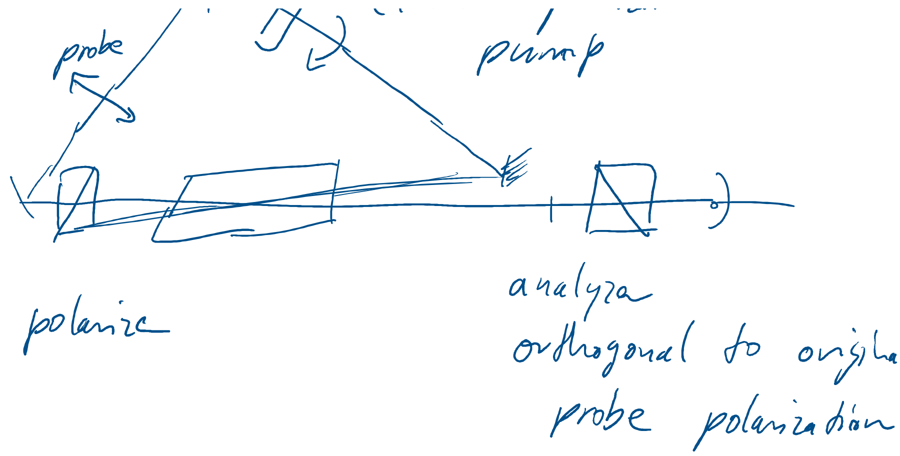
Alternative chop both pump at f_1
probe at f_2

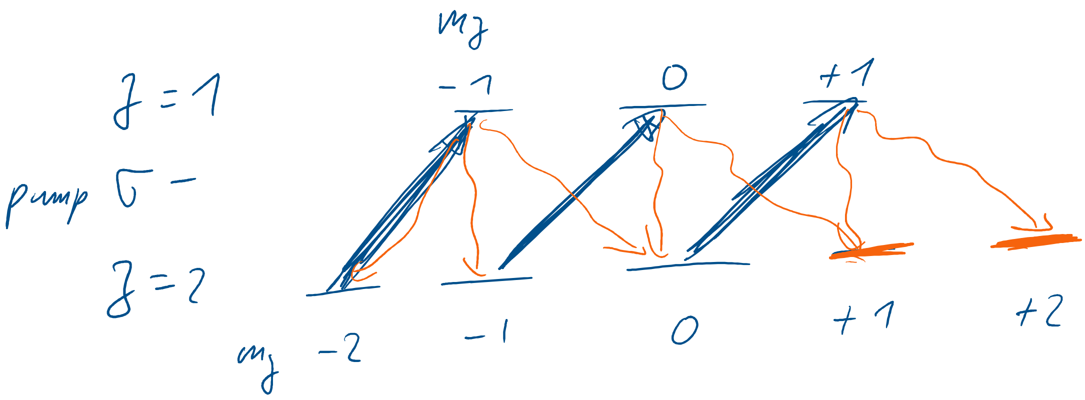


→ Lockin Amplifier @ $f_1 + f_2$

Polarization spectroscopy







pump beam pumps all atoms to $m_J = +1$ or $+2$

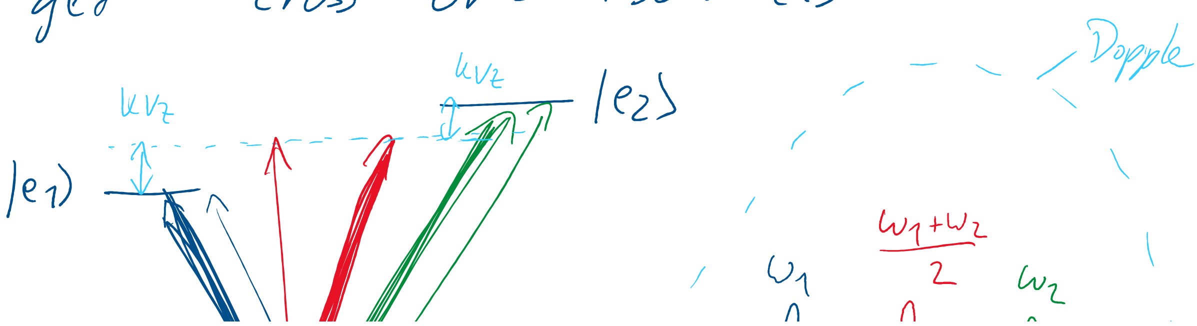
optical pumping creates <u>birefringence</u>

probe beam polarization is rotated

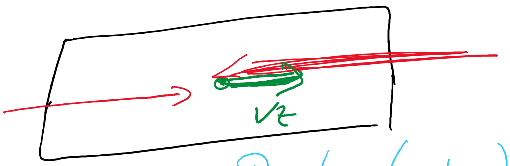
→ light on detector

Note: Saturated absorption etc. signals are non-trivial

for more than 1 transition you can get "cross-over resonances"

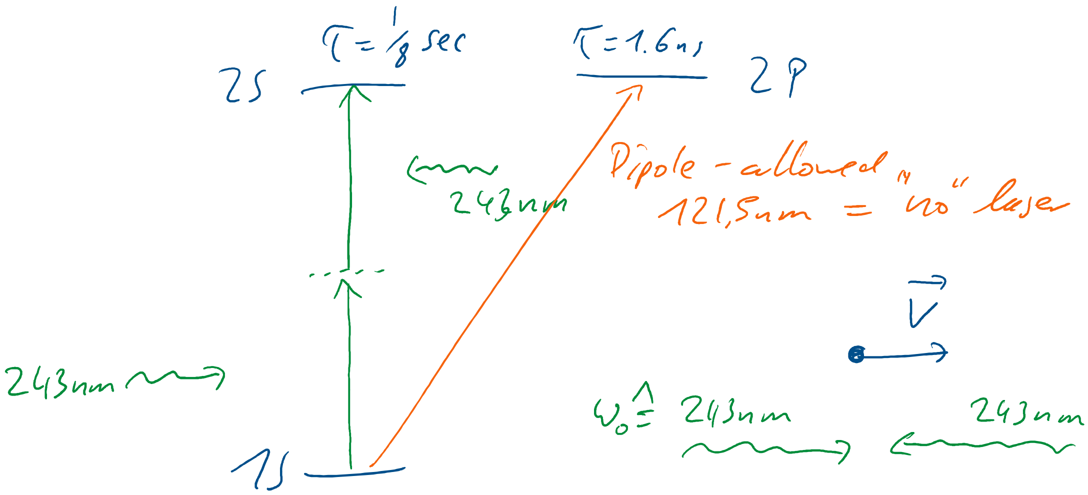


pump + Doppler $(+v_z)$ $= \omega_2$



probe − Doppler $(-v_z)$ $= \omega_1$

H: 1S-2S 2photon spectroscopy



$\omega_0 - k v_z$ $\qquad$ $\omega_0 + k v_z$

$\Sigma = 2\omega_0$ $\qquad$ Doppler-free 1st order