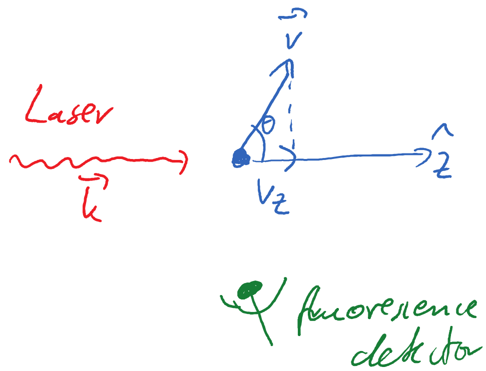
Doppler broadening and Doppler-free spectroscopy

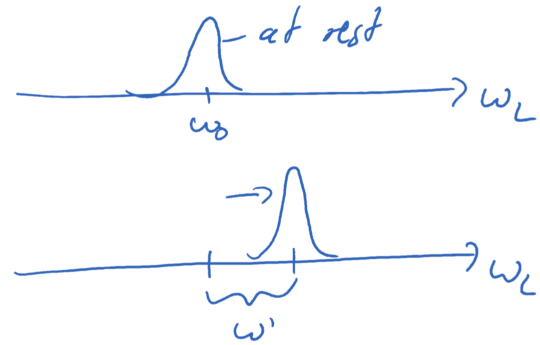
→ Demtröder



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

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

upper picture: laser appears red-shifted



thermal distribution of atoms

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

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

number of atoms with resonance frequency within interval $\omega \ldots \omega + d\omega$

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

measured intensity profile $\propto n_i(\omega)$

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

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

FWHM

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

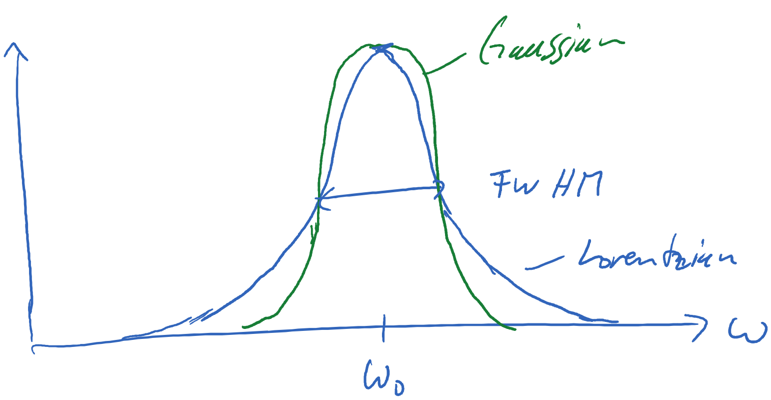
$$= \boxed{\frac{\omega_0}{c}\sqrt{\frac{8kT\ \ln 2}{M}}}$$

Doppler broadening from thermal motion of atoms

$$I(\omega) = I_0 \exp\left[-\left(\frac{\omega-\omega_0}{0.6\,\omega_D}\right)^2\right] \qquad 0.6 \approx \frac{1}{\sqrt{4\ln 2}}$$

natural line shape: Lorentzian

Doppler line shape: Gaussian



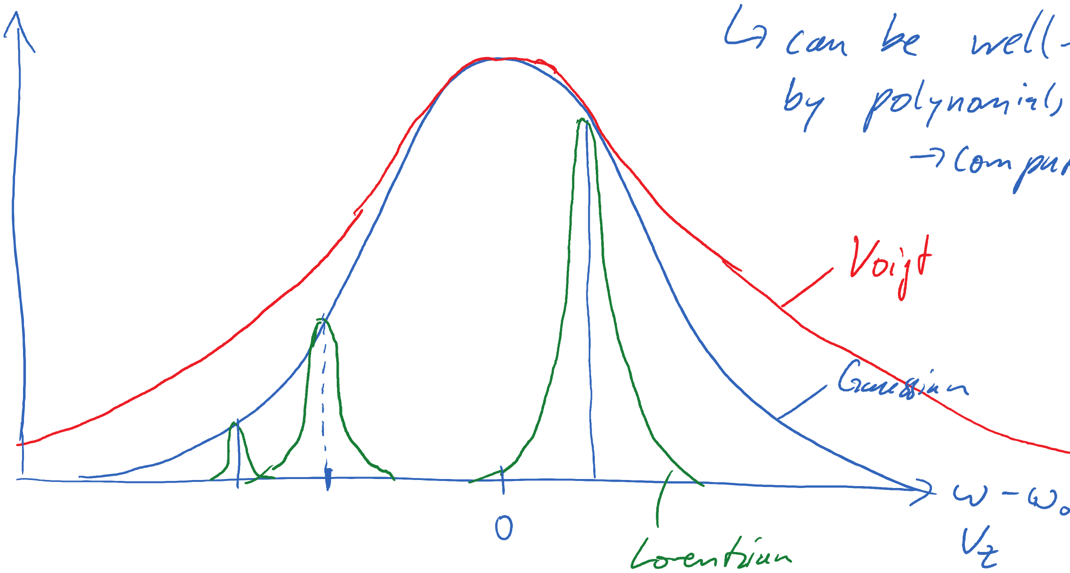
observed line shape: Voigt profile

= convolution of Lorentzian (natural) and Gaussian (Doppler)

$$I(\omega) = \frac{I_0 \gamma N_i c}{2\pi} \int^{\infty} e^{-\frac{c^2(\omega-\omega')}{\omega'^2 v_w^2}}$$

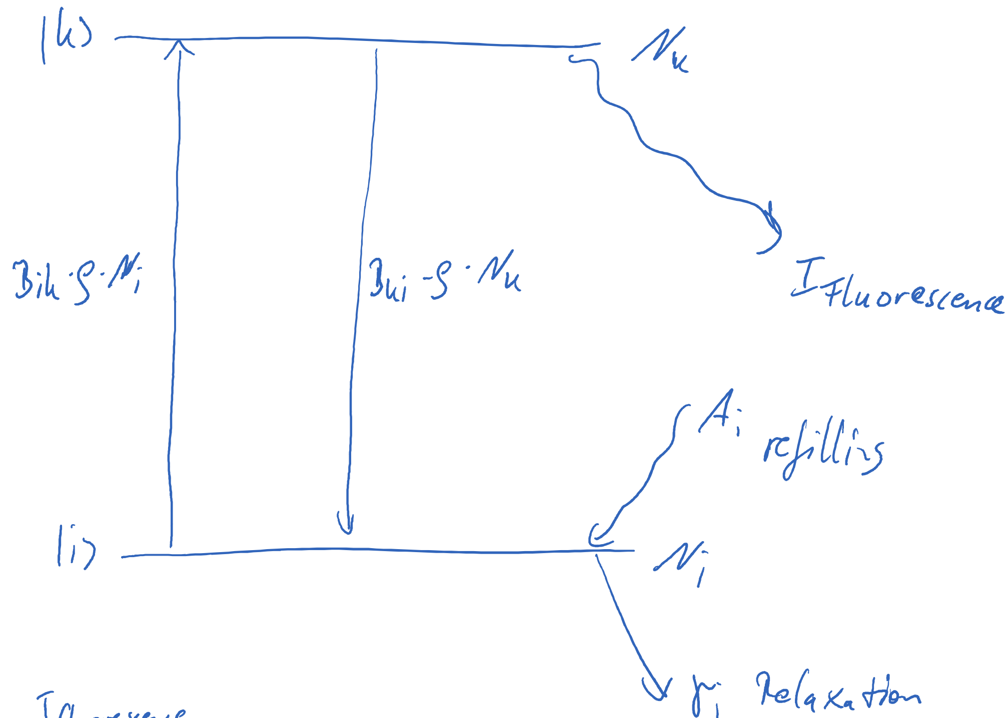
$$I(\omega) = \frac{I_0 \gamma N_i c}{2\pi^{3/2} \omega_0 v_w} \int\limits_{\substack{0 \\ -\infty}} \frac{e^{-\frac{c^2(\omega-\omega')}{\omega'^2 v_w^2}}}{(\omega-\omega')^2 + \left(\frac{\gamma}{2}\right)^2} \, d\omega'$$

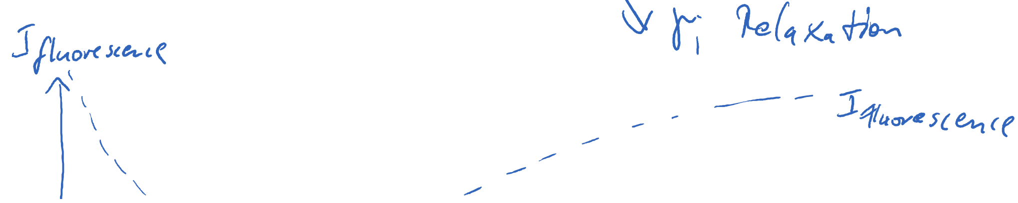
↳ can be well-approximated by polynomials
→ computer code

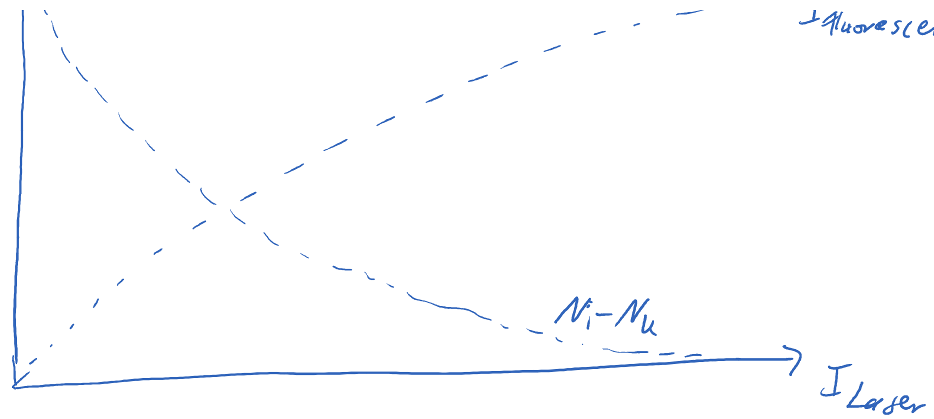


Methods of Doppler-free spectroscopy

(Demtröder)







Linear absorption

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

in volume element $dV = A \cdot dz$

atoms absorb power $dP = A \cdot dI$

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

stat. weights

rate equations

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

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

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

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

$$N_i = \frac{\dots}{B_{ik}\rho + \gamma_i} + N_k \frac{\dots g_k \dots B_{ik}\rho}{B_{ik}\rho + \gamma_i}$$

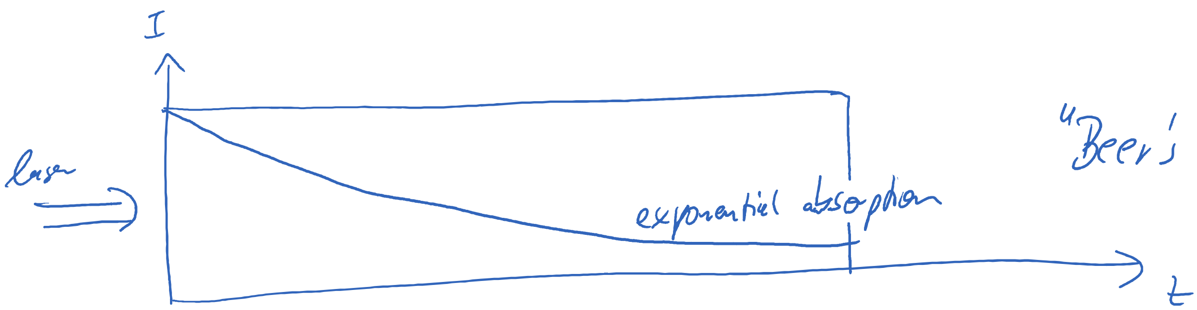
small laser intensity $\quad B_{ik}\frac{I_c}{c} \ll \gamma_i$

usually $\quad N_k \ll N_i$

$$\rightarrow \quad N_i = \frac{A}{\gamma_i}$$

integrate $(*)$

$$P = P_0 \exp\left[-\left(N_i - \frac{g_i}{g_k} N_k\right)\sigma_{ik} \cdot z\right.$$



"Beer's law"

I large (strong laser)

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

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

$N_i(I)$ decreases with increasing laser power

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

lin $\qquad$ quadratic in I

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

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

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

increasing intensity reduces 1st term

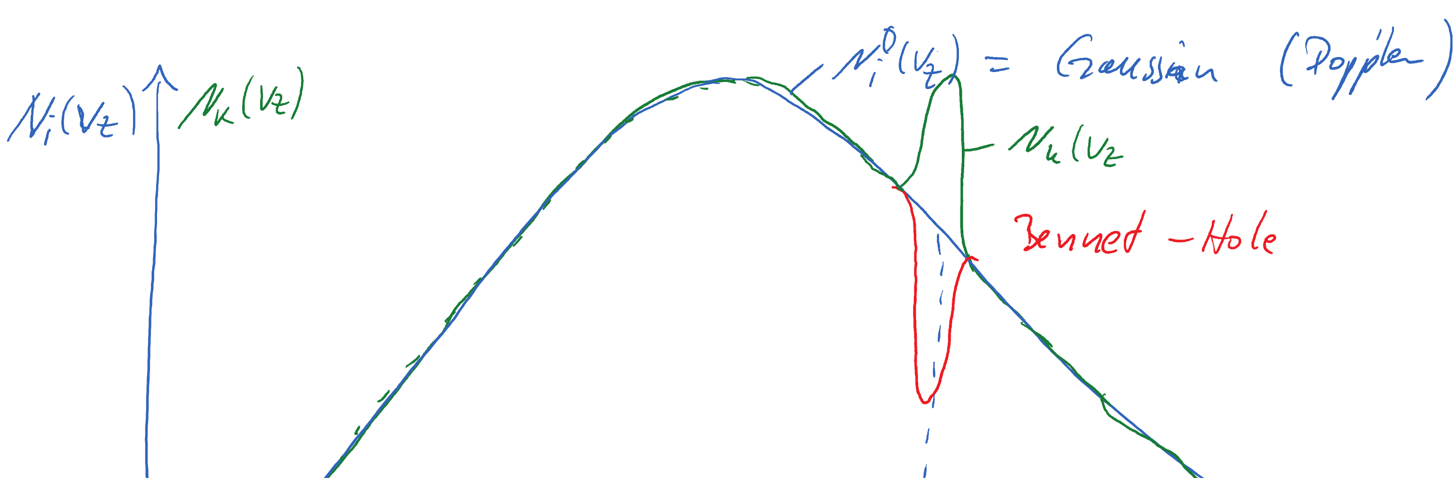
for $I \to \infty$ the absorption goes to 0!

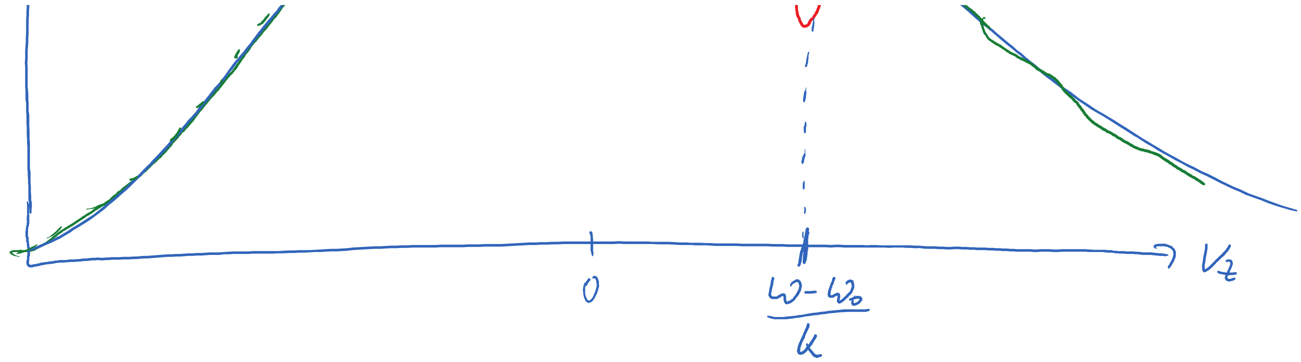
Sample becomes transparent!

Absorption reduces occupation $N_i(v_z)$

enhances $N_k(v_z)$

$$N_i(v_z) = N_i^0(v_z) - \underbrace{\Delta N^0 \frac{v_z}{\gamma_i T} \frac{S_0 \left(\frac{\gamma}{2}\right)^2}{(\omega - \omega_0 - k \cdot v_z)^2 + \left(\frac{\gamma_S}{2}\right)^2}}_{\text{Bennet-hole}}$$





Bennet-hole = local minimum of atoms with $v_z = \frac{\omega-\omega_0}{k}$