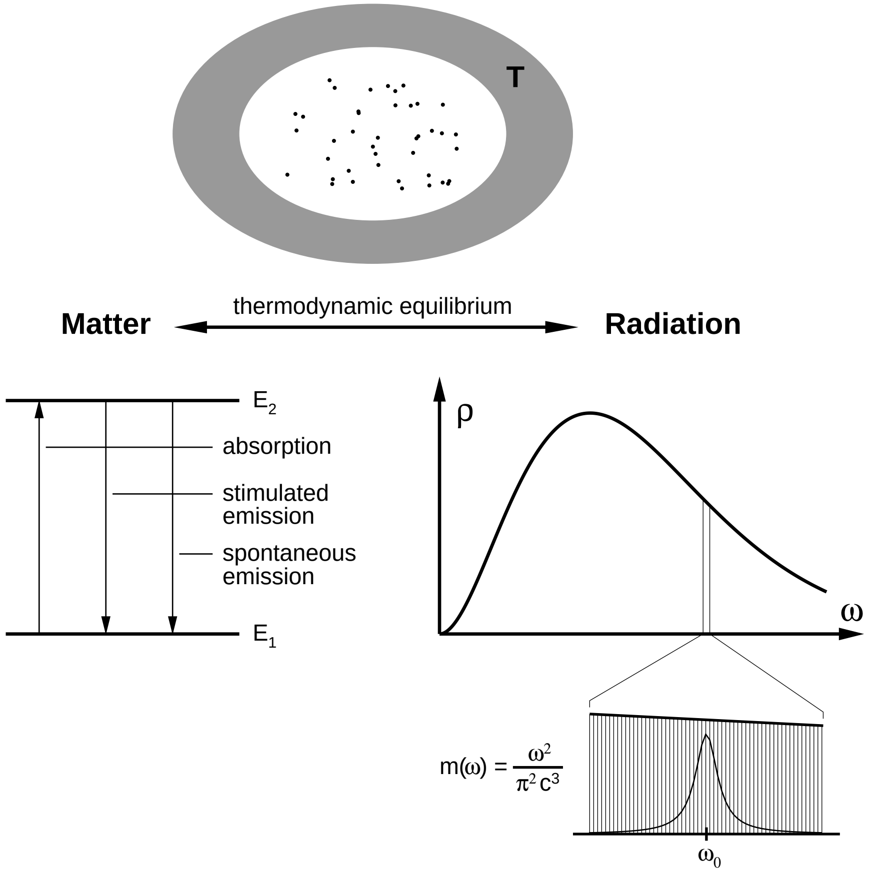
1 Interaction of radiation and matter

To describe the interaction of radiation and matter we seek an expression for field induced transition rates W_{ij} between energi levels of a tvo level system of atoms or molecules. We can approch the problem along three paths:

- QED picture: with both the radiation field and the matter quantized.
- Semiclassical picture: with the matter quantized but a classical description of the field.
- Thermodynamic picture: describing equilibrium between an atomic system and thermal radiation.

The first path needs the full arsenal of quantum mechanics and gives the most detailed information. It is too advanced for this course and we will limit the discussion to the other two paths.

1.1 Thermodynamic picture



$$\underbrace{\rho(\omega)}_{\text{Radiative energy density (pr volume and frequency)}} = \underbrace{m(\omega)}_{\text{Mode density (pr volume and frequency)}} \cdot \underbrace{\hbar\omega}_{\text{Energy pr photon}} \cdot \underbrace{\frac{1}{e^{\hbar\omega/kT} - 1}}_{\text{Average occupation per mode}}$$

We look at a tvo level system in thermodynamic equilibrium at a temperature T. The system is swamped with thermal radiation with an energy density (Planck)

$$\rho(\omega,T) = m(\omega)\cdot\hbar\omega\cdot\frac{1}{e^{\hbar\omega/kT}-1} \tag{1}$$

where the first term $m(\omega)$ is the mode density in the thermal radiation

$$m(\omega) = \frac{\omega^2}{\pi^2 c^3}, \tag{2}$$

the second term the photon energy and the third the average occupation in the modes. The equilibrium level population in the material is assumed to follow the Boltzmann distribution

$$\frac{N_2^e}{N_1^e} = \frac{g_2}{g_1}\, e^{-\hbar\omega_0/kT} \tag{3}$$

where the superscript e marks the equilibrium state, the g's are level degeneracy counters and $\hbar\omega_0 = \Delta E$ is the level separation. Let W_{xy} be the transition rate between levels x and y. Define rate coefficients A and B (Einstein coefficients) with

$$W_{21} = B_{21}\rho + A \qquad \text{and} \qquad W_{12} = B_{12}\rho \tag{4}$$

The A and B coefficients describe the three optical transition processes; spontaneous emission, stimulated emission and absorption. The population steady state condition can now be expressed as

$$\dot{N}_2 = \underset{\text{absorption}}{\underset{|}{B_{12}\rho(\omega_0)N_1^e}} - \underset{\substack{\text{stimulated}\\ \text{emission}}}{\underset{|}{B_{21}\rho(\omega_0)N_2^e}} - \underset{\substack{\text{spontaneous}\\ \text{emission}}}{\underset{|}{AN_2^e}} = 0 \tag{5}$$

The A and B coefficient can now be related to the Planck distribution

$$\rho(\omega_0) = \frac{A}{B_{21}}\;\frac{1}{\frac{B_{12}}{B_{21}}\,\frac{g_1}{g_2}\,e^{\hbar\omega_0/kT}-1} \;\equiv\; \frac{m(\omega_0)\,\hbar\omega_0}{e^{\hbar\omega_0/kT}-1} \tag{6}$$

leading to the ratio conditions

$$\frac{B_{12}}{B_{21}}\cdot\frac{g_1}{g_2} = 1 \qquad \text{and} \qquad \frac{A}{B_{21}} \;=\; m(\omega_0)\,\hbar\omega_0 \tag{7}$$

The Einstein coefficients refer to the all the radiation modes that can interact with the two level system. Level broadening or other mechanisms that lead to line broadening can be incorporated by considering the coefficients as integrals over a normalized lineshape function $G(\omega-\omega_0)$. This is equivalent to defining frequency dependent coefficients

$$A(\omega) \;=\; A\,G(\omega-\omega_0) \qquad \text{and} \qquad B(\omega) \;=\; B\,G(\omega-\omega_0) \tag{8}$$

with the normalizing condition

$$\int_{-\infty}^{\infty} G(\omega-\omega_0)\,d\omega = 1 \tag{9}$$

1.2 Semiclassical picture

Here we treat the field $\mathcal{E}$ classically but the material is quantized and described with the wavefunctions ψ_1 and ψ_2 for the two levels.

$$\psi_1 = u_1(\bar{r})\; e^{-iE_1 t/\hbar} \qquad\qquad \psi_2 = u_2(\bar{r})\; e^{-iE_2 t/\hbar}$$

The field—matter interaction is described with the perturbation operator $\mathbf{H}' = \bar{\mu}\cdot\bar{\mathcal{E}}$, where $\bar{\mu} = e\bar{r}$ is the dipole moment of the atom/molecule, and $\bar{\mathcal{E}} = \bar{\mathcal{E}_0}\; e^{-i\omega t}$ is a plane polarized radiation field. The interaction $\mathbf{H}'$ couples the wavefunctions into a time dependent mixture.

$$\psi = a_1(t)\psi_1 + a_2(t)\psi_2 \qquad\qquad |a_1|^2 + |a_2|^2 = 1 \tag{10}$$

The probability of finding the systen in the excited state is $|a_2|^2$ and the transition rate is given by

$$W_{12} = \frac{d}{dt}\;|a_2|^2 \tag{11}$$

First order perturbation calculations with application of Fermis golden rule give

$$W_{12} = \frac{2\pi}{\hbar^2}|\mu_{12}|^2\;|\mathcal{E}_0|^2\;\delta(\omega-\omega_0) \tag{12}$$

where $|\mu_{12}| =< u_2^*|\,\mu\,|u_1 >$ is the dipole matrix element and $\delta(\omega-\omega_0)$ is the Kronecker delta function. By symmetry we can conclude that $|\mu_{12}| = |\mu_{21}|^*$ so that the transition rate is the same for absorption and stimulated emission.

The uncomfortable presence of the delta function is a consequence of disregarding level broading due to collisions and finite radiative lifetime of levels and the spread of the apparent field frequencies due to thermal velocity dependent Doppler shifts. The delta function will be replaced by appropriate lineshape functions later.

For comparison with the thermodynamic picture we need to tie the field amplitude to the energy density with

$$\rho = \frac{1}{2}\varepsilon_0\mathcal{E}_0^2 \tag{13}$$

With equation 4 we can now express the A and B coefficients in terms of material parameters

$$B_{12}(\omega) = \frac{4\pi}{\varepsilon_0\hbar^2}\;\frac{g_2}{g_1}\;|\mu_{12}|^2\;G(\omega-\omega_0) \tag{14}$$

$$A(\omega) = \frac{4\omega_0^3}{\pi c^3\varepsilon_0\hbar}\;|\mu_{12}|^2\;G(\omega-\omega_0) \tag{15}$$

where the δ functions have been replaced by a normalized lineshape function.

Warning: *This has been calculated for angular frequency* ω. *Transformation to circular frequency* ν *can be tricky as the B's contain the distribution function m where* $m(\omega)d\omega = m(\nu)d\nu$. *We have also assumed the dipole to be induced by the field and therefore aligned to it. For permanent dipole moments and unpolarized light we need a directional cosine averaged over space directions,* $< \cos^2\theta >= 1/3$.

Without an external field the $A = \int A(\omega) d\omega$ coefficient determines the radiative lifetime of level 2, and we can define the spontaeous emission lifetime as

$$\tau_{sp} = 1/A \tag{16}$$

The radiative lifetime τ_{sp} is proportional to λ^3 making fluorescence to dominant decay mechanism in the UV and visible ranges, while collisions are often dominating in the IR range.

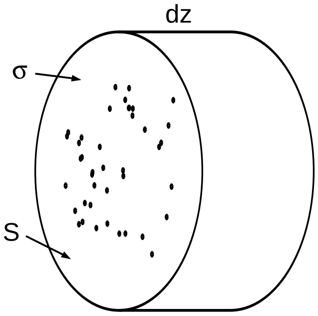
1.3 Relationships to other macroscopic parameters

In the thermodynamic picture the radiation field was described with the energy density ρ, and in the semiclassical picture with the field amplitude $\mathcal{E}_0$. We often need to tie the transition rate to other paramers like power density, photon flux density, absorption cross section or absorption coefficient. In the table below the polarizability of the material has been incorporated into the formulas through the refractive index n.

$$n = c/v = \sqrt{\varepsilon/\varepsilon_0} \tag{17}$$

where v is the light speed in the material and ε its dielectric constant.

Parameter name	Parameter connection	Transition rate W
Field amplitude	$\mathcal{E}_0$	$\frac{2\pi}{\hbar^2}\, \lvert\mu_{12}\rvert^2\, \mathcal{E}_0^2$
Energy density	$\rho = \frac{1}{2} n^2 \varepsilon_0 \mathcal{E}_0^2$	$\frac{4\pi}{n^2\, \varepsilon_0\, \hbar^2}\, \lvert\mu_{12}\rvert^2\, \rho$
Power density	$I = \frac{c}{n}\, \rho$	$\frac{4\pi}{n\, c\, \varepsilon_0\, \hbar^2}\, \lvert\mu_{12}\rvert^2\, I$
Photon flux density	$F = \frac{I}{\hbar\omega}$	$\frac{4\pi\, \omega}{n\, c\, \varepsilon_0\, \hbar}\, \lvert\mu_{12}\rvert^2\, F$



We define an absorption cross-section, σ, through flux decay with beam travel distance. Let $\Delta N = (N_2 - \frac{g_2}{g_1} N_1)$.

$$dF = -W \Delta N\, c dt = -\sigma F \Delta N\, dz \tag{18}$$

$$\frac{dF}{F} = -\frac{\sigma \Delta N\, dz S}{S} = -\frac{\sigma \Delta N\, dV}{S} \tag{19}$$

Referring to the figure we can interpret the right side of eq. 19 as the ratio of the shadow area of all the active atoms in the slice to the cross-section of the slice. We picture the atoms as opaque spheres, each with a cross-section σ.

The macroscopic quantity absorption coefficient, α, can now be expressed in terms of the microscopic material parameters.

$$-\alpha(\omega) = \sigma\, \Delta N \; = \frac{4\pi\, \omega}{n\, c\, \varepsilon_0\, \hbar}\, \lvert\mu_{12}\rvert^2\, \Delta N \;\; G(\omega - \omega_0) \tag{20}$$

Beer's law states the physical meaning the absorption coefficient; as a decay rate constant with beam travel distance.

$$dI = -\alpha I\, dz \qquad\qquad I = I_0\, e^{-\alpha\, z} \tag{21}$$

A negative value of α constitutes a gain coefficient and calls for the inversion condition $\Delta N > 0$.

1.4 Line broadening

Line broadening mechanisms fall in two categories; homogeneous broadening and inhomogeneous broadening, that divide lasers into two distinct classes. For a homogeneously broadened transition all the atoms or molecules respond in the same way to the external monocromatic radiation, whereas the the radiation can only interact with a subgroup of atoms in the inhomogeneous case.

1.4.1 Homogeneous broadening

Radiative decay and hard collisions inducing transitions lead to homogeneous broadening. So do soft collisions that only disturb the phase development of the interaction of an external field with atoms.

We will look at an ensamble of atoms with a N_2 population that is decaying through spontaneous emission with a time constant τ_{rad}.

$$N_2 = N_2^0\, e^{-t/\tau_{rad}} \tag{22}$$

The radiated elctric field amplitude is proportional to the square root of $-\dot{N}_2$ since $-\dot{N}_2 \sim I \sim \mathcal{E}_0^2$. The time devolopment of the radiation field at a fixed point in space is described with

$$\mathcal{E}(t) = \mathcal{E}_0\, e^{-t/2\tau_{rad}}\, \cos(\omega_0\, t) = \tfrac{1}{2}\mathcal{E}_0\, e^{-t/2\tau_{rad}}\, [e^{i\omega_0\, t} + e^{-i\omega_0\, t}] \tag{23}$$

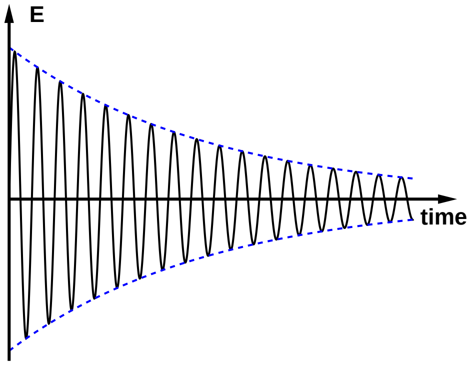
for $t > 0$. To get information about the frequency content in the field we take a Fourier transform

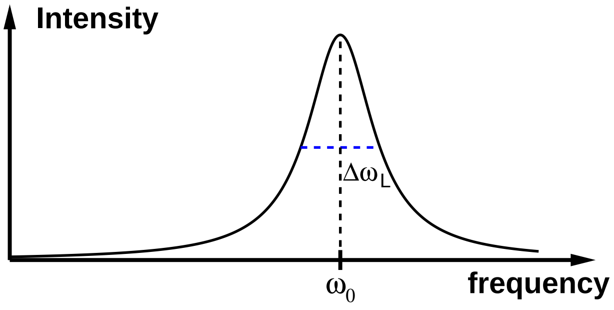
$$\mathcal{F}(\omega) = \frac{1}{2\pi}\int_0^\infty \mathcal{E}(t)\, e^{-i\omega\, t} dt = \frac{i\mathcal{E}_0}{4\pi}\left[\frac{1}{\omega - \omega_0 + i/2\tau_{rad}} - \frac{1}{\omega + \omega_0 - i/2\tau_{rad}}\right] \tag{24}$$

The power spectrum is proportional to $|\mathcal{F}(\omega)|^2$ so dropping the small sum frequency term in eq. 24 we get the lineshape function

$$G_L = \frac{1}{\pi\Delta\omega_L}\,\frac{1}{1 + \left(\frac{\omega-\omega_0}{\Delta\omega_L}\right)^2} \tag{25}$$

where $\Delta\omega_L = 1/2\tau_{rad}$ is the halfwidth (HWHM). The profile G_L has been area normalized. This lineshape, which is common for all homogeneous broadened systems, is called a Lorentzian.





Multiple homogeneous broadening mechanisms characterized with the time constants τ_k still give a Lorenzian with the halfwidth

$$\Delta\omega_L = \sum_k \frac{1}{2\tau_k} = \sum_k \Delta\omega_{L,k} \tag{26}$$

1.4.2 Inhomogeneous broadening

If transition frequency of subcomponents of an atomic system is spread over an interval we talk of inhomogeneous broadening. The inhomogeneous width is a property of the system, while each atom only possesses a homogeneous width.

The most common inhomogeneous broadening mechanism is the Doppler shift due to the thermal motion of atoms. The normalized thermal distribution of velocities is given with the Maxwell distribution

$$p_v = \sqrt{\frac{M}{2\pi kT}}\, e^{-\frac{M\ v^2}{2kT}} \tag{27}$$

where M is the atomic mass, v the velocity and T the temperature. An atom with a velocity v against the propagation direction of a monocromatic beam feels an upshifted frequency

$$\omega' = \omega\,[1 + v/c] \tag{28}$$

where ω and v are measured in the same frame of reference. The resonance condition $\omega' = \omega_0$ is thus downshifted

$$\omega = \frac{\omega_0}{1+v/c} \qquad \text{giving} \qquad dv = -\frac{c}{\omega_0}(1+v/c)^2 d\omega \simeq -\frac{c}{\omega_0} d\omega \tag{29}$$

The spectral distribution G_D is tied to the thermal distribution with

$$G_D(\omega - \omega_0)d\omega = -p_v dv = \frac{c}{\omega_0}\sqrt{\frac{M}{2\pi kT}}\, e^{-\frac{M\ c^2}{2kT}\left(\frac{\omega-\omega_0}{\omega_0}\right)^2} d\omega \tag{30}$$

Here it is convenient to define a halfwidth (HWHM) with

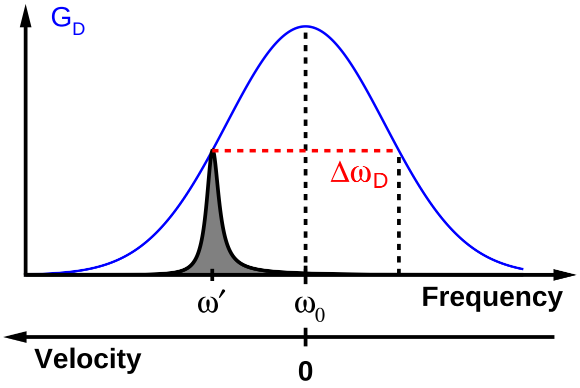
$$\Delta\omega_D = \frac{\omega_0}{c}\sqrt{\frac{2\ln 2\, kT}{M}} \tag{31}$$

leaving the area normalized lineshape function expressed as

$$G_D(\omega - \omega_0) = \sqrt{\frac{\ln 2}{\pi}}\, \frac{1}{\Delta\omega_D}\, e^{-\ln 2\left(\frac{\omega-\omega_0}{\Delta\omega_D}\right)^2} \tag{32}$$

This lineshape is called a Doppler profile. The Doppler linewidth scales as $\Delta\omega_D \sim \omega_0\sqrt{T/M}$. At the wavelength λ =500 nm, temperature T =300 K and mass M =20 amu it takes the value $\Delta\omega_D \simeq 2\pi\cdot 1$ GHz.

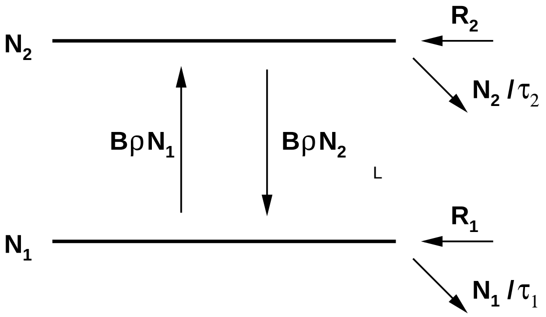
All transitions possess both a Doppler width and a Lorentz with. A monocromatic beam of frequency ω can only communicate with a velocity group of atoms that see a Doppler shifted beam to the resonance frequency ω_0, for the case $\Delta\omega_D \gg \Delta\omega_L$. The frequency width of the velocity group is $2\Delta\omega_L$. In the other extreme $\Delta\omega_L \gg \Delta\omega_D$ the beam can communicate with the whole ensamble if ω is within the profile.



1.5 Saturation

We talk of gain/absorption saturation when the radiation field modifies the level population differens ΔN from its weakfield value.

1.5.1 Saturation of a homogeneous profile



Now we consider a more general level system than before, assuming that the lower level is not the ground state. We introduce constant pump rates R_k and decay rates $1/\tau_k$ for both levels. The τ's can be due to radiative transitions or collisions.

In the low field limit (ρ=0) the steady state conditions are

$$\dot{N}_k = R_k - N_k^0/\tau_k = 0 \tag{33}$$

for $k = 1,2$. The low field inversion can now be written as

$$\Delta N^0 = N_2^0 - \frac{g_2}{g_1}N_1^0 = R_2\tau_2 - \frac{g_2}{g_1}R_1\tau_1 \tag{34}$$

With a radiation field at frequency ω' the steady state equations take the form

$$\begin{aligned} \dot{N}_1 &= R_1 - N_1/\tau_1 + B(\omega')\rho(\omega')\Delta N = 0 \\ \dot{N}_2 &= R_2 - N_2/\tau_2 - B(\omega')\rho(\omega')\Delta N = 0 \end{aligned} \tag{35}$$

leading to the field modified inversion

$$\Delta N = \frac{\Delta N^0}{1 + B(\omega')\rho(\omega')\tau} \tag{36}$$

where $\tau = \frac{g_2}{g_1}\tau_1 + \tau_2$. So far we have been looking at a fixed point in space. To tie the discussion to an absorption/gain coefficient we must follow the beam on its journey ($dz/dt = c$).

$$\rho = I/c \qquad \frac{d\rho}{dt}\frac{dt}{dz} = \frac{1}{c}\frac{dI}{dz} \quad \Longrightarrow \quad \dot{\rho} = \frac{dI}{dz} \tag{37}$$

The impact of the stimulated transitions on the field energy density is

$$2\dot{\rho}(\omega') = -\hbar\omega'(\dot{N}_2 - \dot{N}_1)_{field} = 2\hbar\omega' B(\omega')\rho(\omega')\Delta N \tag{38}$$

The subscript "field" is to stress that this is only the field induced (stimulated) part of the time derivative. The field intensity gradient can now be written as

$$\frac{dI(\omega')}{dz} = \frac{\hbar\omega'}{c} B(\omega')I(\omega')\Delta N = -\alpha(\omega', I)\, I(\omega') \tag{39}$$

where the absorption/gain coefficient α now has become intensity dependent, *saturated*.

$$-\alpha(\omega', I) = \frac{\hbar\omega' B}{c} \frac{\Delta N^0}{1 + IB(\omega')\tau/c} G_L(\omega' - \omega_0) \tag{40}$$

With the pump rates R_k we can control the sign of the inversion $\Delta N^0 = (N_2^0 - \frac{g_2}{g_1} N_1^0)$. With a negative sign we get absorption, $\alpha > 0$, whereas a positive inversion gives gain, $\alpha < 0$. In the last case it is customary to define a positive gain coefficient $g = -\alpha$. We introduce the concept of saturation intensity, I_s, by writing the intensity dependent part of eq. 40 as

$$IB\tau/c = I/I_S \tag{41}$$

$$I_S(\omega') \;=\; \frac{n c \varepsilon_0 \hbar^2}{4\pi |\mu_{12}|^2} \frac{g_1}{g_2} \frac{1}{\tau\, G_L(\omega' - \omega_0)} \;=\; I_{S0} \left[1 + \left(\frac{\omega' - \omega_0}{\Delta\omega_L} \right)^2 \right] \tag{42}$$

where

$$I_{S0} = I_S(\omega_0) = \frac{n c \varepsilon_0 \hbar^2}{4 |\mu_{12}|^2} \frac{g_1}{g_2} \frac{\Delta\omega_L}{\tau} \tag{43}$$

The saturation intensity at line center is thus independent of the inversion and even of number density of absorbers in dilute mixtures but depends on total pressure as p^2 since both $\Delta\omega_L$ and $\frac{1}{\tau}$ are proportional to p. Off center the saturation intensity increases with detuning as a reciprocal Lorentzian.

The homogeneous saturated gain (or absorption) is then decribed with

$$g(\omega', I) \;=\; \frac{g(\omega', 0)}{1 + I(\omega')/I_S(\omega')} \tag{44}$$

Rearranging terms in eq. 44 collecting all ω' dependence to to one place and using the shorthand notation $g_0 = g(\omega = \omega_0, I = 0)$ for the linecenter small signal gain, result in

$$g(\omega', I) \;=\; \frac{g_0}{1 + I/I_{S0}} \frac{1}{1 + (\frac{\omega' - \omega_0}{\Delta\omega_L \sqrt{1 + I/I_{S0}}})^2} \tag{45}$$

Equation 45 implies that when the profile is scanned with a constant intensity saturating beam, the saturated gain (or absorption) still shows a Lorentzian shape but now with an increased linewidth $\Delta\omega_L \sqrt{1 + I/I_{S0}}$. This effect is called *power broadening*. It is a consequence of the offset dependence of the saturation intensity. The wings saturate less than the line center.

Before leaving the subject of homogeneous saturation we must look at the effect of a saturating beam at frequency ω' on the gain at other frequencies ω. We do this by observing the

gain/attenuation of a weak probe beam I_ω that is scanned over the profile, while the strong beam is saturating at a fixed frequency ω'.
Rewriting eq. 39 for this situation gives

$$\frac{dI_\omega}{dz} = \frac{\hbar\omega}{c} B(\omega) I_\omega \, \Delta N = -\alpha(\omega, I(\omega'))\, I_\omega \tag{46}$$

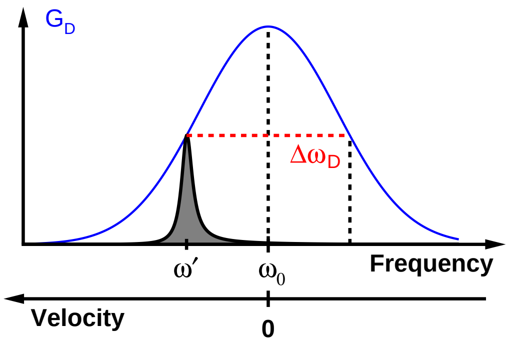
where the absorption/gain coefficient α now is also a function of ω' through the saturation effect described in eq. 36

$$\alpha(\omega, I(\omega')) = \frac{1}{1 + I(\omega')/I_S(\omega')} \, \frac{\alpha_0}{1 + \left(\frac{\omega-\omega_0}{\Delta\omega_L}\right)^2} \tag{47}$$

The equation shows that a monocromatic saturating beam at frequency ω' will saturate the whole line profile. This property has important consequences for the behavior of homogeneously broadened lasers.

1.5.2 Saturation of an inhomogeneous profile

We consider a Doppler line with $\Delta\omega_D \gg \Delta\omega_L$, and use similar procedure as for the homogeneous case. First we find the saturated lineform when scanning the saturation beam and then consider the impact of a stationary saturation beam on the profile. The gain (or absorption) is found by integrating over the velocitygroup that is Doppler shifted to resonance, corresponding to the shaded area on the figure. The Lorentzian form of the velocitygroup is power broadened.



The saturated gain is found with a convolution summing up contributions from different velocity groups.

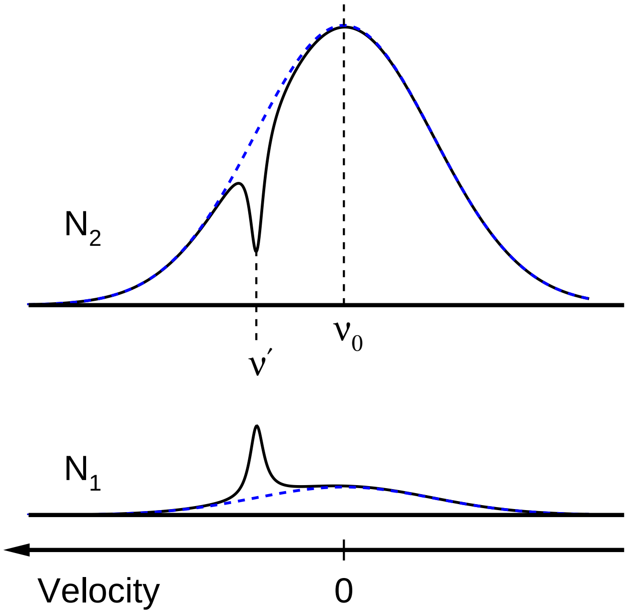
$$g(\omega', I(\omega')) = \int_{-\infty}^{\infty} \frac{g_0}{1 + I(\omega')/I_S(\omega - \omega')} \, G_D^*(\omega - \omega_0)\, G_L(\omega' - \omega)\, d\omega \tag{48}$$

(To keep g_0 clean we use the Doppler profile G_D^* with a peak value 1.)

$$g(\omega', I(\omega')) = \int_{-\infty}^{\infty} g_0 \, e^{-\ln 2 \left(\frac{\omega-\omega_0}{\Delta\omega_D}\right)^2} \frac{\frac{1}{\pi\Delta\omega_L} \, \frac{1}{1+(\frac{\omega'-\omega}{\delta\omega_L})^2} d\omega}{1 + \frac{I(\omega')}{I_{S0}} \, \frac{1}{1+\left(\frac{\omega-\omega'}{\Delta\omega_L}\right)^2}} \simeq \frac{g_0}{\sqrt{1 + I/I_{S0}}} \, e^{-\ln 2 \left(\frac{\omega'-\omega_0}{\Delta\omega_D}\right)^2} \tag{49}$$

Equation 48 shows that the saturated profile is still Gaussian and the width is unaffected. The square–rooot in the denominator makes the inhomogeneous saturation slower than in the homogeneous counterpart. The saturation intesity is now independent of position within the profile.

A saturating beam at a fixed frequency ω' will saturate at that frequency according to equation 48 but leave other velocitygroups outside the saturated width unaffected. This effect of shunting a selected velocitygroup to the other level is called a holeburning and is characteristic for saturation of inhomogeneous systems. The deformed gain profile is displayed in the next figure. The derivation of the analytical form is outside the scope of this course.



1.6 Amplifiers

The homogeneously saturated gain coefficient is given with eq. 44.

$$g(\omega, I) = \frac{g(\omega,0)}{1+I/I_S(\omega)} \tag{50}$$

The amplification of the intensity with distance inside the amplifier is described wit the differential equation

$$\frac{dI}{dz} = \frac{g(\omega,0)}{1+I/I_S(\omega)}\, I \tag{51}$$

With separation of variables and integration we get a formal solution

$$\ln I/I_0 + (I-I_0)/I_S = g(\omega,0)\, z \tag{52}$$

We can not solve this equation for I(z) in a closed form due to the saturation term, which slows the amplification down. The solution can always be found with iterative numerical methods as boundaries for the solution are known; $1 < I(z)/I_0 < e^{gz}$.

For the inhomogeneous case the differential equation takes the form

$$\frac{\sqrt{1+I/I_S(\omega)}}{I}\, dI = g(\omega,0)\, dz \tag{53}$$

With the substitutions $S = \sqrt{1+I/I_S}$ and $S_0 = \sqrt{1+I_0/I_S}$ the formal solution can be written as

$$2(S-S_0) + \ln\frac{(S-1)(S_0+1)}{(S_0-1)(S+1)} = g(\omega,0)\, z \tag{54}$$

It takes numerical methods to isolate I(z) as in the other case.