Interaction of light with atoms: Line broadening and saturation

Collisional broadening

Doppler broadening

Saturation:

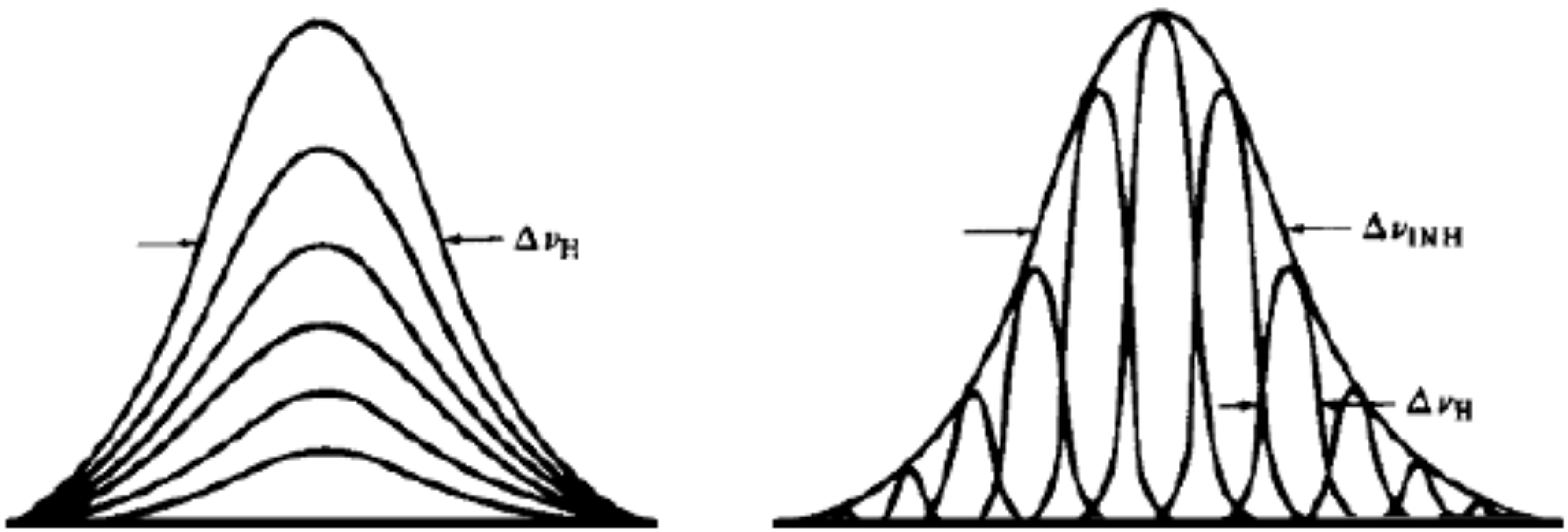
- solving CW equilibrium rate equations
- saturation with short pulses, saturation fluence

Additional effects found in real systems

- An atom or ion in influenced by the external surroundings in several ways:
  - blackbody EM background: radiatively establishes a thermal (Boltzmann) population distribution
  - Collisions (other atoms or electrons) broaden energy levels (and linewidths) and can also lead to population changes
  - Local fields from a lattice can broaden lines

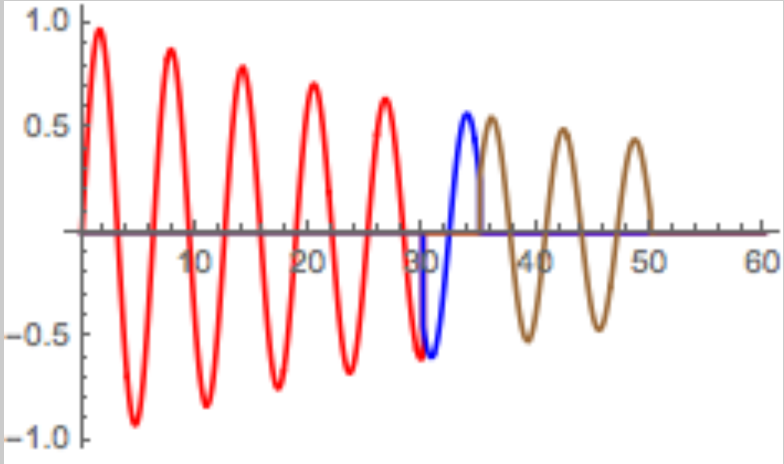
Types of broadening

- Homogeneous broadening: all individual atoms are broadened by the same amount
- Inhomogeneous broadening: each atom is shifted (e.g. Doppler) so that the ensemble has a broader spectrum



Collisional broadening

- Elastic collisions don't cause transition, but interrupt the phase
- Timescales:
  - Period of EM cycle much less than radiative lifetime $\frac{2\pi}{\omega_0} \ll \tau$
  - Avg time btw collisions < lifetime $\tau_c < \tau$
  - Duration of a collision << time btw coll, lifetime $\Delta\tau_c \ll \tau_c, \tau$



Calculating collisional broadening linewidth

- Calculation:
  – Fourier Transform over time 0 to τ_1 to get lineshape for a specific oscillation length
  – Average over probability of a given time between collisions:

$$P(\tau_1)d\tau_1 = \frac{1}{\tau_c} e^{-\tau_1/\tau_c} d\tau_1$$

Result:
Lorentzian shape with new width

$$\Delta\nu = \gamma / 2\pi + 1/\pi\tau_c$$

- All atoms see the same collision rate, so collisional broadening is **homogeneous**

Doppler broadening

- From relative velocity of atom to input beam, Doppler shift:

$$\nu_0' = \frac{\nu_0}{1 - \mathrm{v}_z / c}$$

Beam propagating in *z* direction

  - Each atom in distribution is shifted according to its velocity

- Boltzmann distribution

$$P(\mathrm{v}_z) \sim \exp\left[-\tfrac{1}{2} M v_z^2 / k_B T\right]$$

- Average over distribution to get effective lineshape:

$$g^*(\nu - \nu_0) = \frac{1}{\nu_0}\left(\frac{Mc^2}{2\pi k_B T}\right)^{1/2} \exp\left\{\frac{Mc^2}{2k_B T}\frac{(\nu - \nu_0)^2}{\nu_0{}^2}\right\}$$

FWHM:

$$\Delta\nu_0{}^* = 2\nu_0\left[\frac{2k_B T \ln 2}{Mc^2}\right]^{1/2}$$

Doppler broadening in HeNe lasers

$$\Delta\nu_0{}^* = 2\nu_0\left[\frac{2k_BT\ln 2}{Mc^2}\right]^{1/2}$$

$$\lambda_0 = 632.8\text{nm}$$

$$\nu_0 = 4.74\times10^{14}\,\text{s}^{-1}$$

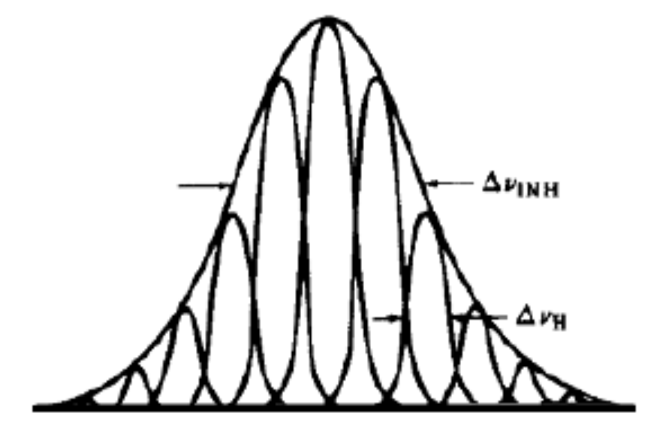
$$M = 20.12\text{ amu}=3.34\times10^{-26}\text{kg}$$ For Neon

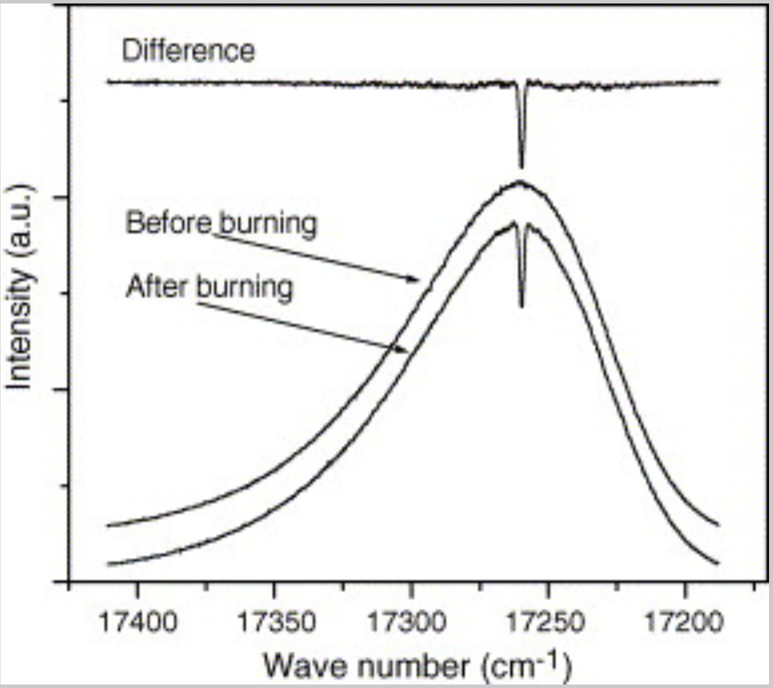
$$k_BT = 1/40eV = 4\times10^{-21}J$$

$$\Delta\nu_0{}^* = 1.55GHz$$

Inhomogeneous vs homogeneous broadening

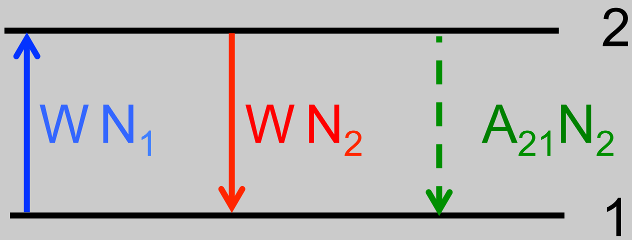
- Homogeneous broadening: every atom is broadened by same shape
  - Radiative, collisional, phonon
  - All atoms participate in absorption or gain
- Inhomogeneous broadening:
  - Doppler broadening
  - Absorption or gain only by atoms in resonance
  - Leads to "spectral hole burning"





Population dynamics of absorption

- Closed 2 level system, assume $g_1 = g_2$



$$\frac{dN_2}{dt} = W\,N_1 - W\,N_2 - A_{21}N_2$$

$$\frac{dN_1}{dt} = -\frac{dN_2}{dt}$$

- Since system is closed, reduce to one equation for population difference:

$$\Delta N = N_1 - N_2 \qquad \rightarrow \frac{dN_2}{dt} = W\,\Delta N - A_{21}N_2$$

$$\frac{d}{dt}\Delta N = \frac{dN_1}{dt} - \frac{dN_2}{dt} = -2\frac{dN_2}{dt} = -2\left(W\,\Delta N - A_{21}N_2\right)$$

$$N_t = N_1 + N_2 \qquad N_t = \Delta N + 2N_2 \qquad \rightarrow N_2 = \tfrac{1}{2}\left(N_t - \Delta N\right)$$

$$\frac{d}{dt}\Delta N = -2W\,\Delta N + A_{21}\left(N_t - \Delta N\right) = -\Delta N\left(A_{21} + 2W\right) + A_{21}N_t$$

Steady-state (CW) solutions for saturated absorption

- For continuous wave operation – all transients have damped out:

$$\frac{d}{dt}\Delta N = -\Delta N\left(A_{21} + 2W\right) + A_{21}N_t$$

$$0 = -\Delta N\left(A_{21} + 2W\right) + A_{21}N_t$$

$$\Delta N = \frac{A_{21}N_t}{A_{21} + 2W} \qquad A_{21} = 1/\tau_{21}$$

$$\Delta N = \frac{N_t}{1 + 2W\tau_{21}}$$

Saturation of absorption

- The key parameter in this situation is W τ_{21}

$$W_{21} = \rho_v B_{21}$$

  – Low intensity, $2W\tau_{21} << 1$, $\Delta N \approx N_t$
  – High intensity, $2W\tau_{21} >> 1$, $\Delta N \approx 0$. Here $N_1 \approx N_2$

$$\Delta N = \frac{N_t}{1 + 2W\tau_{21}}$$

$$\Delta N = N_1 - N_2$$

- Energy balance:

Input power
Absorbed by atoms

→ Radiated power (into 4π)

→ Stimulated emission
(back into beam)

- Radiated power per unit volume:

$$\frac{dP}{dV} = h\nu_{21} W \Delta N(W) = h\nu_{21} \frac{N_t W}{1 + 2W\tau_{21}} \rightarrow h\nu_{21} \frac{N_t}{2\tau_{21}}$$

For W $\tau_{21} >> 1$

Power radiated in high intensity limit: half of atoms are radiating

Saturation intensity

- Absorbed power per atom: $\sigma_{12} I$
- Absorption rate: $W = \dfrac{\sigma_{12} I}{h\nu_{21}}$
- In steady state: $\dfrac{\Delta N}{N_t} = \dfrac{1}{1+2W\tau_{21}} = \dfrac{1}{1+2\dfrac{\sigma_{12} I}{h\nu_{21}}\tau_{21}} \equiv \dfrac{1}{1+\dfrac{I}{I_{sat}}}$
- Saturation intensity for absorption: $I_{sat} = \dfrac{h\nu_{21}}{2\sigma_{12}\tau_{21}}$
  - 2: transition affects both levels at once
  - At I = I_{sat}, stimulated and spontaneous emission rates are equal.
- Intensity-dependent absorption coefficient:

$$\alpha(I) = \frac{\alpha_0}{1 + I/I_{sat}}$$

At high intensity, material absorbs *less*.

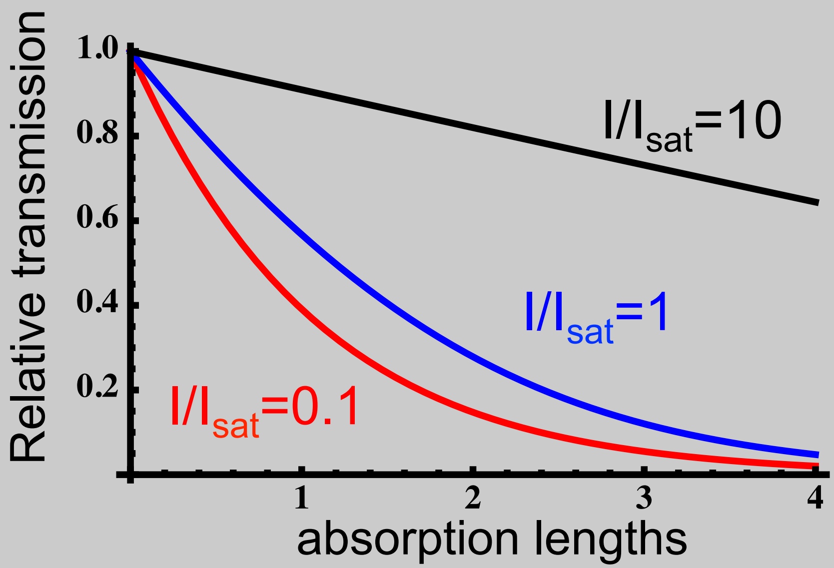
Saturable absorbers are used for pulsed lasers: Q-switching and mode-locking

Saturated CW propagation through absorbing medium

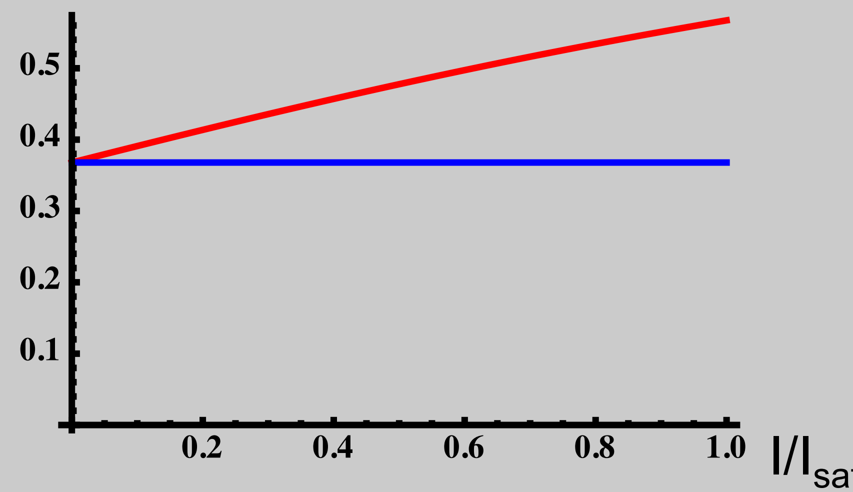
- For a given thickness for an absorbing medium, the transmission will increase with intensity

$$\alpha(I) = \frac{\alpha_0}{1 + I/I_{sat}} \qquad \frac{dI}{dz} = -\alpha(I)I = -\frac{\alpha_0}{1 + I/I_{sat}} I$$

$$\int_{I_0}^{I} \left(\frac{1}{I} + \frac{1}{I_{sat}} \right) dI = -\int_0^L \alpha_0 \, dz \rightarrow \ln\left[\frac{I(z)}{I(0)}\right] + \frac{I(z) - I(0)}{I_{sat}} = -\alpha_0 z$$



Transmission over 1 absorption length



Dynamic saturation: pulsed input

- Rewrite equation using intensity:

$$\frac{d}{dt}\Delta N = -\Delta N\left(A_{21} + \frac{2\sigma}{h\nu_{21}} I(t)\right) + A_{21}N_t \equiv -\Delta N\left(A_{21} + \frac{I(t)}{\Gamma_{sat}}\right) + A_{21}N_t$$

Define Γ_{sat} = saturation fluence

- Scaling of equation: determine relative importance of terms
  - Input intensity can be rewritten as: $I_{in} = \Gamma_{in} / \tau_p$
  - Rewrite equation:

$$\frac{d}{dt}\Delta N = -\Delta N\left(\frac{1}{\tau_{21}} + \frac{\Gamma_{in}}{\Gamma_{sat}}\frac{1}{\tau_p}\right) + \frac{1}{\tau_{21}}N_t = \frac{2N_2}{\tau_{21}} - \frac{\Gamma_{in}}{\Gamma_{sat}}\frac{1}{\tau_p}\Delta N \qquad \Delta N = N_1 - N_2$$

  - Note there are two timescales: τ_p and τ_{21}
  - The weighting of the two controls which we might ignore.

Saturation with short pulse input

$$\frac{d}{dt}\Delta N = 2N_2 \frac{1}{\tau_{21}} - \frac{\Gamma_{in}}{\Gamma_{sat}} \Delta N \frac{1}{\tau_p}$$

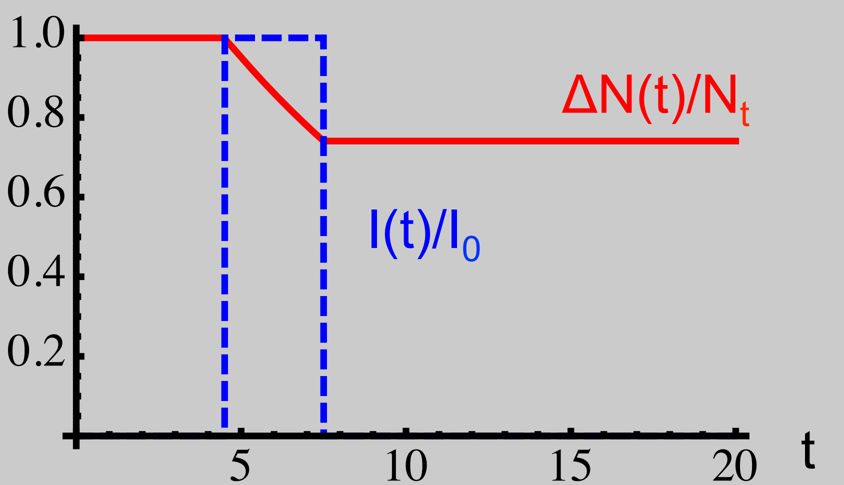
- For short pulse input: ignore stimulated emission and fluorescence

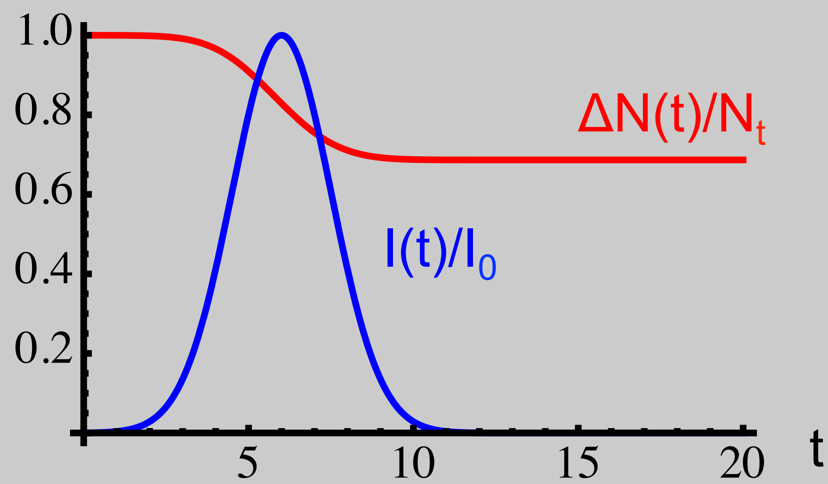
$$\frac{\Gamma_{in}}{\Gamma_{sat}} \frac{1}{\tau_p} |\Delta N| \gg \frac{2N_2}{\tau_{21}} \qquad \rightarrow \frac{d}{dt}\Delta N \approx -\frac{\Gamma_{in}}{\Gamma_{sat}} \frac{1}{\tau_p} \Delta N$$

  - Put back in terms of time-dependent intensity: $\frac{1}{\Delta N} \frac{d}{dt}\Delta N \approx -\frac{1}{\Gamma_{sat}\tau_p} I(t)$
  - Solve equation by integrating $\rightarrow \ln\left[\frac{\Delta N(t)}{\Delta N(0)}\right] \approx -\frac{1}{\Gamma_{sat}} \int_0^t I(t')dt'$
  - If all in ground state initially, $\Delta N(0) = N_t$
  - At end of pulse: $\int_0^\infty I(t')dt' = \Gamma_{in}$ $\qquad \rightarrow \Delta N(\infty) = N_t \exp\left[-\frac{\Gamma_{in}}{\Gamma_{sat}}\right]$

Short pulse limit

- For short pulse input: $\tau_p << \tau_{21}$, so ignore fluorescence
  - Medium just integrates energy of pulse.
  - Example: Ti:sapphire: τ_{21}=3.2μs, τ_p=10ns or 200ns for Q-switched Nd:YAG lasers pumped with flashlamps or CW arc lamps
- Square input pulse   Gaussian input pulse
  - τ = 3, I_0/I_{sat} = 0.1, (no fluorescence)





- Shape of *transmitted* pulse is affected

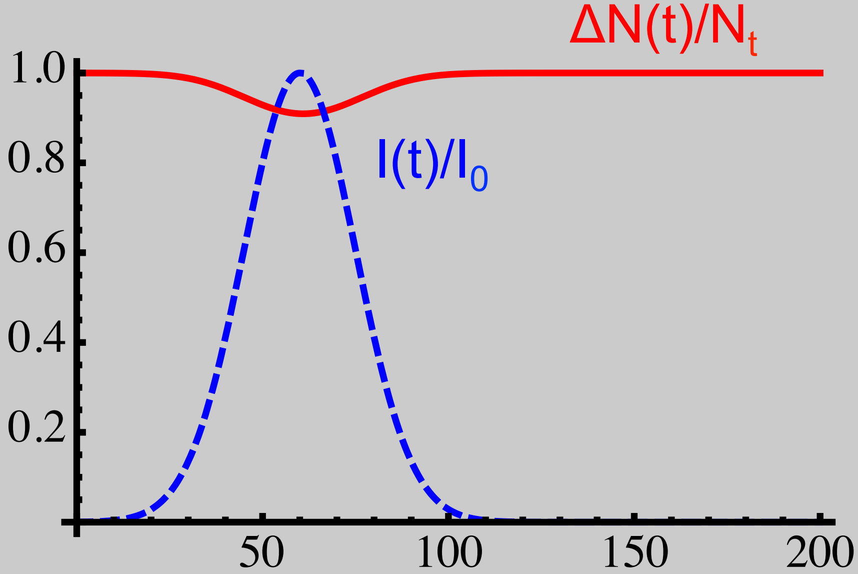
Long pulse limit

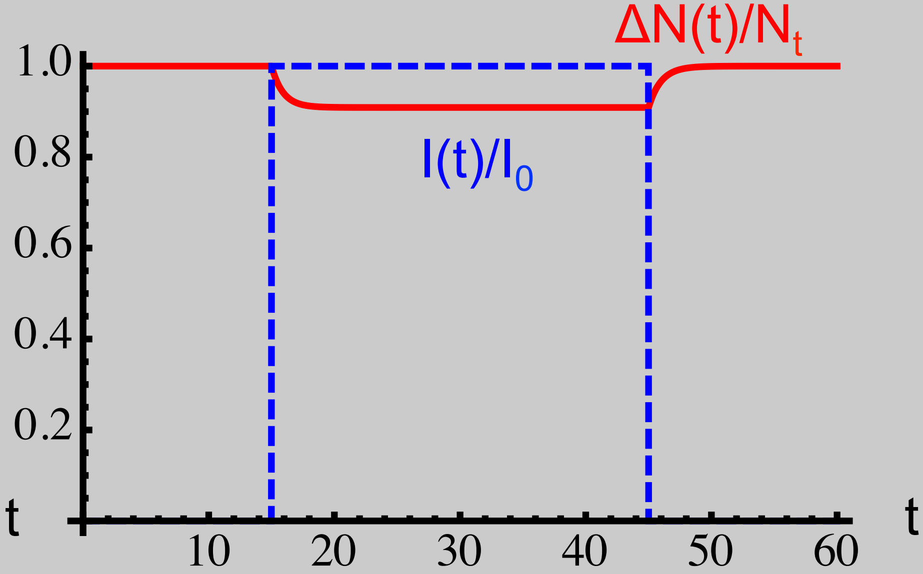
- For *long* pulse input: $\tau_p \gg \tau_{21}$, and peak $I \ll I_{sat}$, $\Delta N(t)$ follows $I(t)$

$$\rightarrow \frac{d}{dt}\Delta N \ll A_{21} N_t$$

Quasi-static, quasi-CW limit
N_t adiabatically follows I(t)

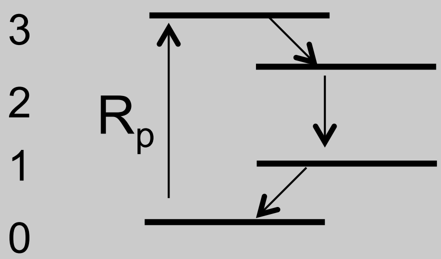
$$\frac{\Delta N}{N_t} = \frac{1}{1 + I(t)/I_{sat}}$$





Gain saturation

- Consider a 4-level system:



Assume: τ_{32} and τ_{10} << τ_{21} and $W_{21}N_2$

- Look at level 2 only:

$$\frac{dN_2}{dt} = R_P - W\,N_2 - N_2/\tau_{21}$$

Low intensity: $N_2 = R_P\tau_{12}$

τ_{12} is called "storage time"

- Steady state: $$N_2 = \frac{R_P\tau_{21}}{1+W\tau_{21}} = \frac{R_P\tau_{21}}{1+\dfrac{\sigma_{21}\tau_{21}}{h\nu_{21}}I} = \frac{R_P\tau_{21}}{1+\dfrac{I}{I_{sat}}}$$

- Saturation intensity for *gain:*
  - *No factor of 2*

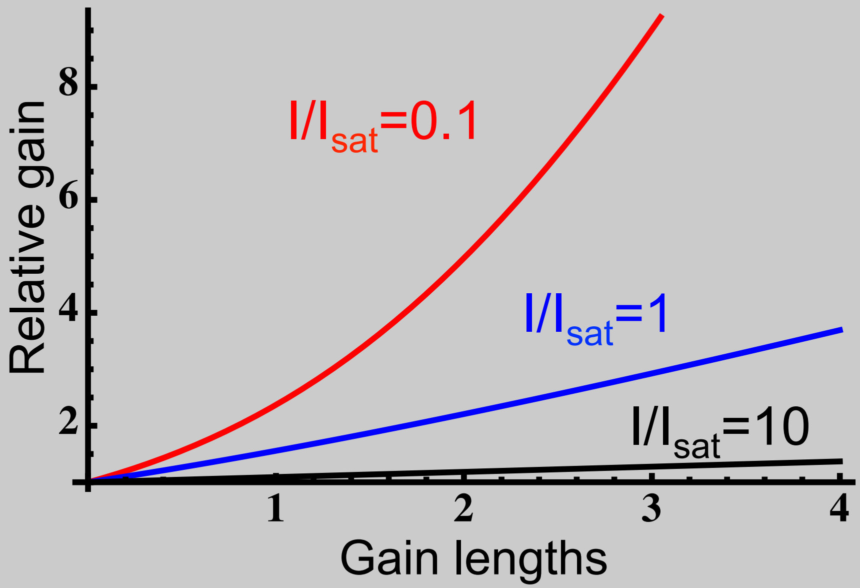
$$I_{sat} = \frac{h\nu_{21}}{\sigma_{21}\tau_{21}} = \frac{\Gamma_{sat}}{\tau_{21}}$$

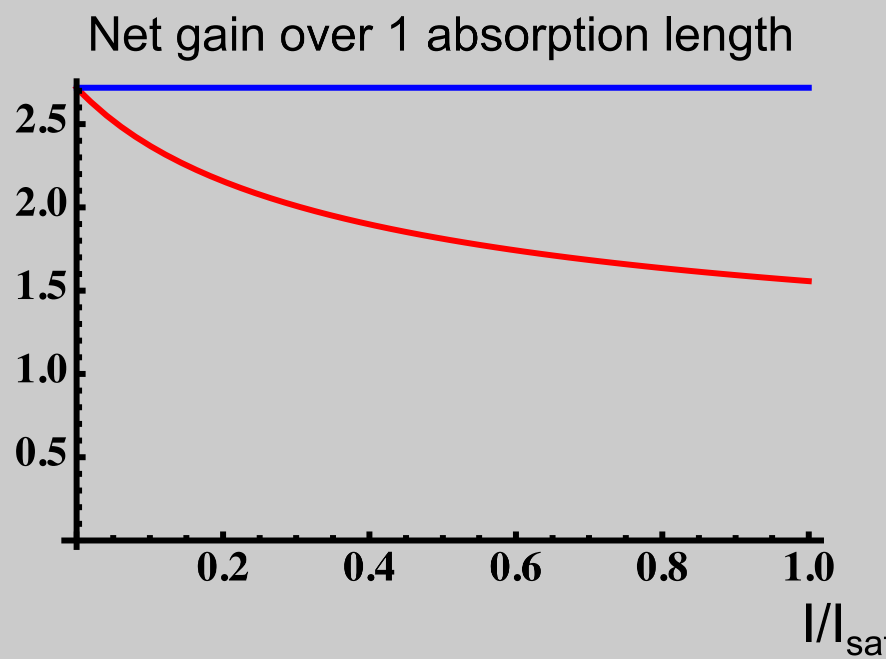
$$g(I) = \frac{g_0}{1+I/I_{sat}}$$

Beam growth during amplification

- Calculation just as with absorption

$$\int_{I_0}^{I}\left(\frac{1}{I}+\frac{1}{I_{sat}}\right)dI=+\int_0^L g_0\,dz\rightarrow \ln\left[\frac{I(z)}{I(0)}\right]+\frac{I(z)-I(0)}{I_{sat}}=+g_0 z$$





- **Even though saturated gain is low, it is efficient at extracting stored energy**