Interaction of light with atoms: Line broadening and saturation

Collisional broadening

Doppler broadening

Saturation:

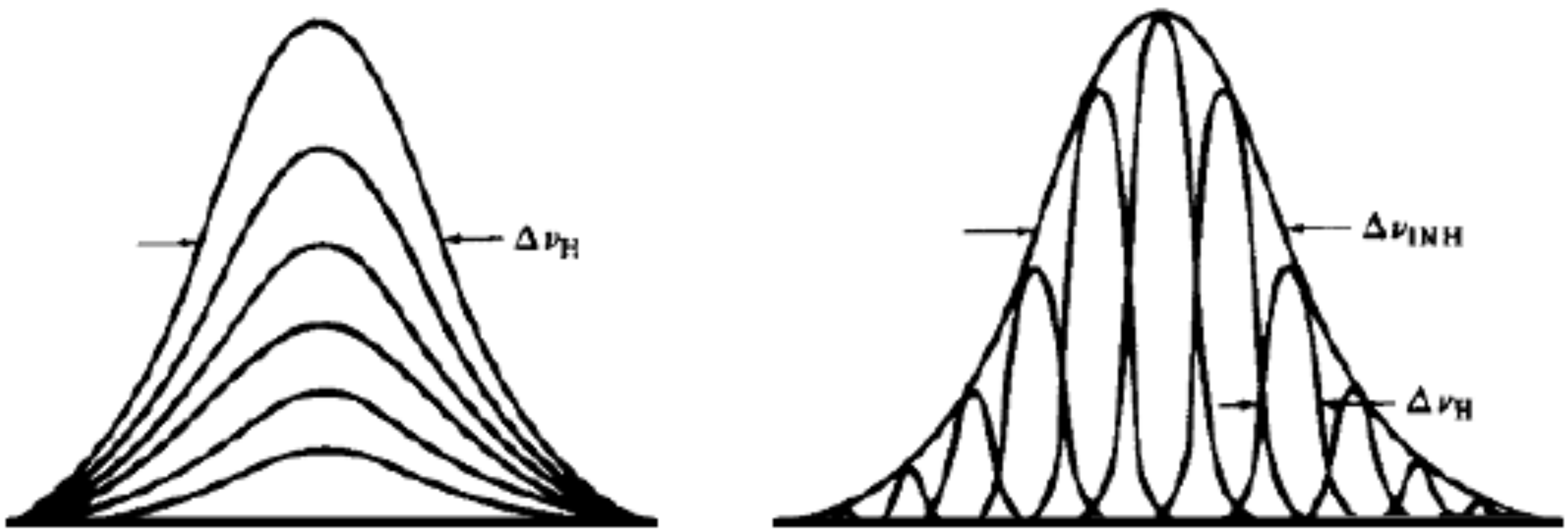
- solving CW equilibrium rate equations
- saturation with short pulses, saturation fluence

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

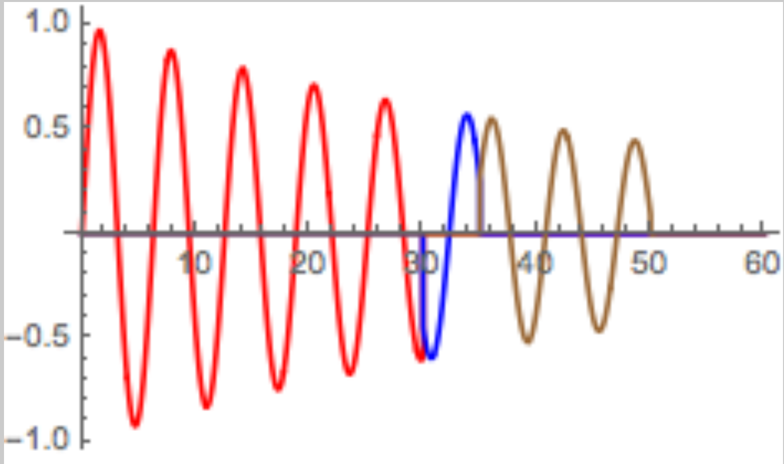
Types of broadening

- 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

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

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

$$\nu_0 = 4.74\times 10^{14}\,\mathrm{s}^{-1}$$

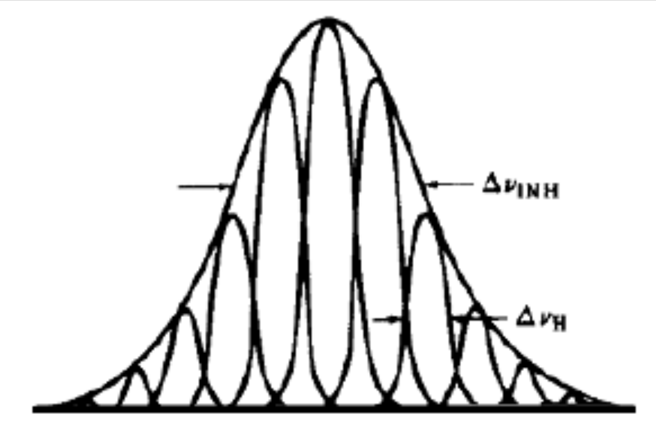
$$M = 20.12\ \mathrm{amu} = 3.34\times 10^{-26}\mathrm{kg}$$ For Neon

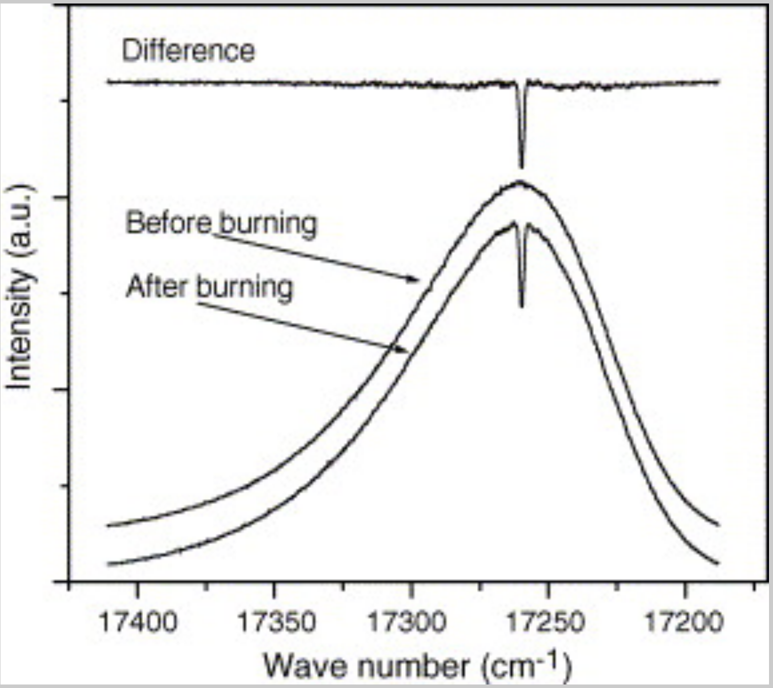
$$k_B T = 1/40 eV = 4\times 10^{-21} J$$

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

Inhomogeneous vs homogeneous broadening

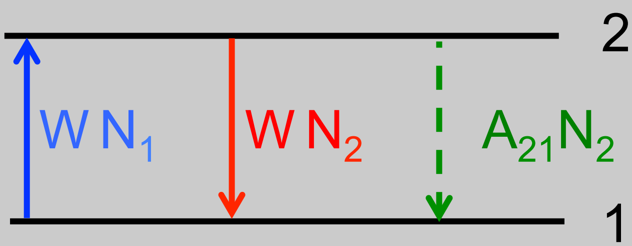
- Homogeneous broadening: every atom is broadened by same shape
  - Radiative, collisional, phonon
  - All atoms participate in absorption or gain
- Inhomogeneous broadening:
  - Doppler broadening
  - Absorption or gain only by atoms in resonance
  - Leads to “spectral hole burning”





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 $W\tau_{21}$

$$W_{21} = \rho_\nu B_{21}$$

  - 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)

Input power Absorbed by atoms → 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*.

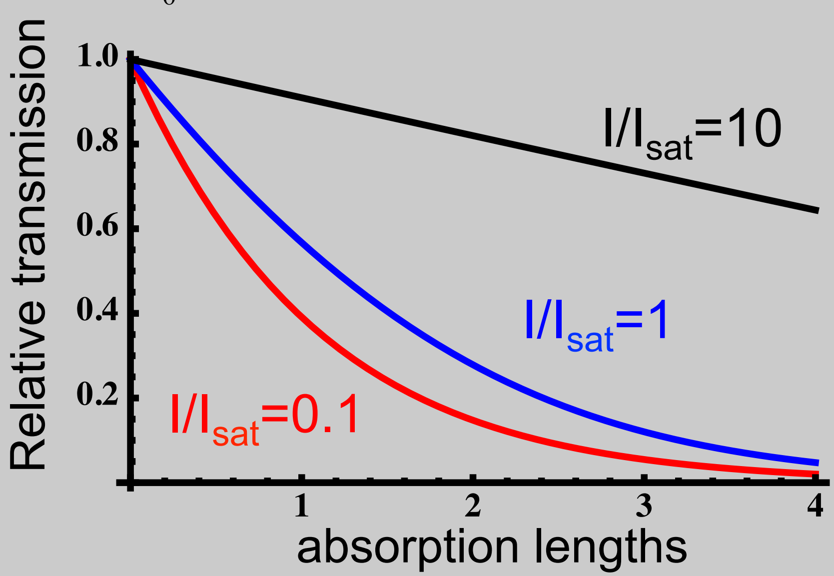
Saturable absorbers are used for pulsed lasers: Q-switching and mode-locking

Saturated CW propagation through absorbing medium

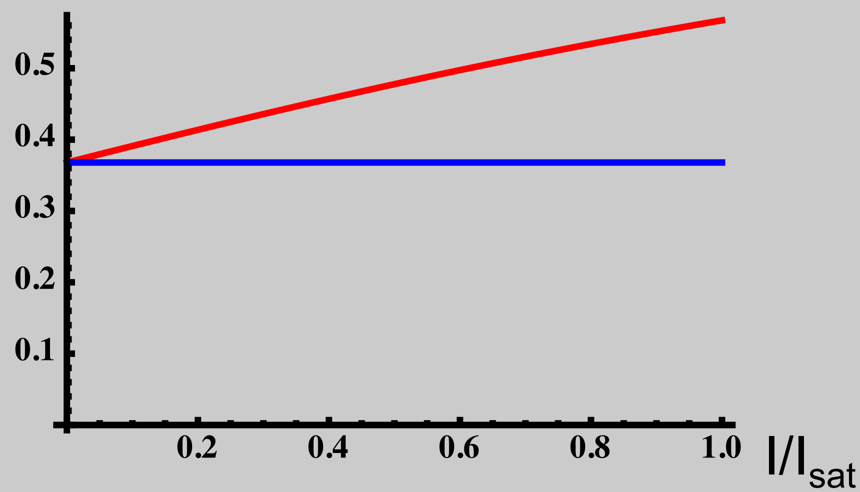
- For a given thickness for an absorbing medium, the transmission will increase with intensity

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

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



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: $\frac{1}{\Delta N}\frac{d}{dt}\Delta N \approx -\frac{1}{\Gamma_{sat}\tau_p}I(t)$

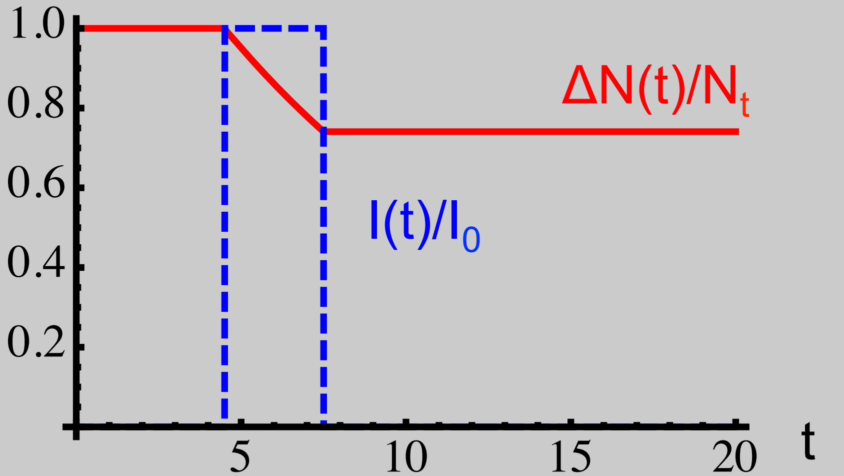
  – Solve equation by integrating $\rightarrow \ln\left[\frac{\Delta N(t)}{\Delta N(0)}\right] \approx -\frac{1}{\Gamma_{sat}}\int_0^t I(t')dt'$

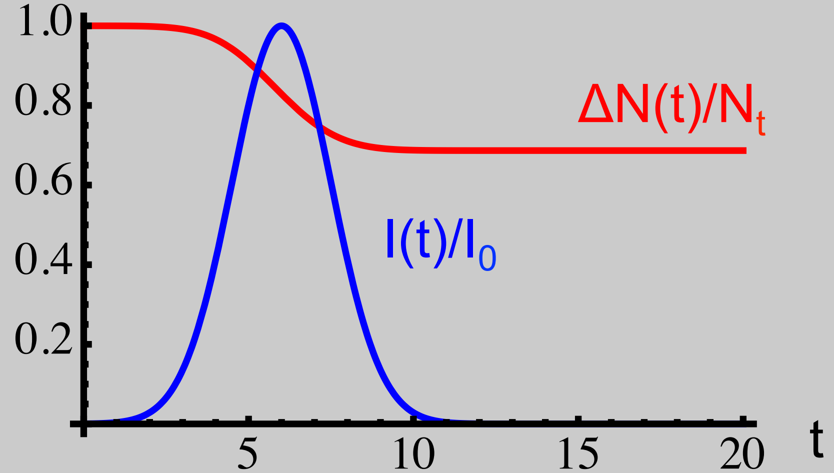
  – If all in ground state initially, $\Delta N(0) = N_t$

  – 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

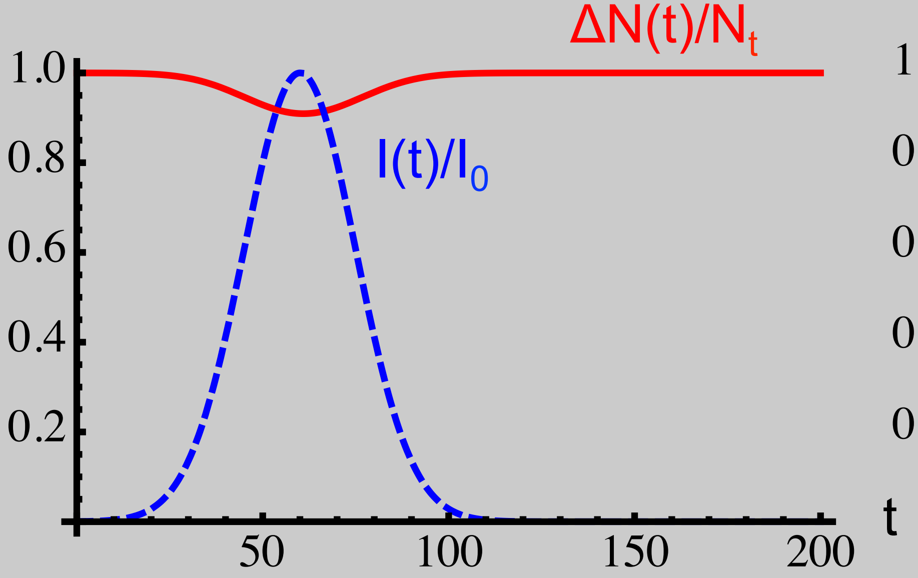
Long pulse limit

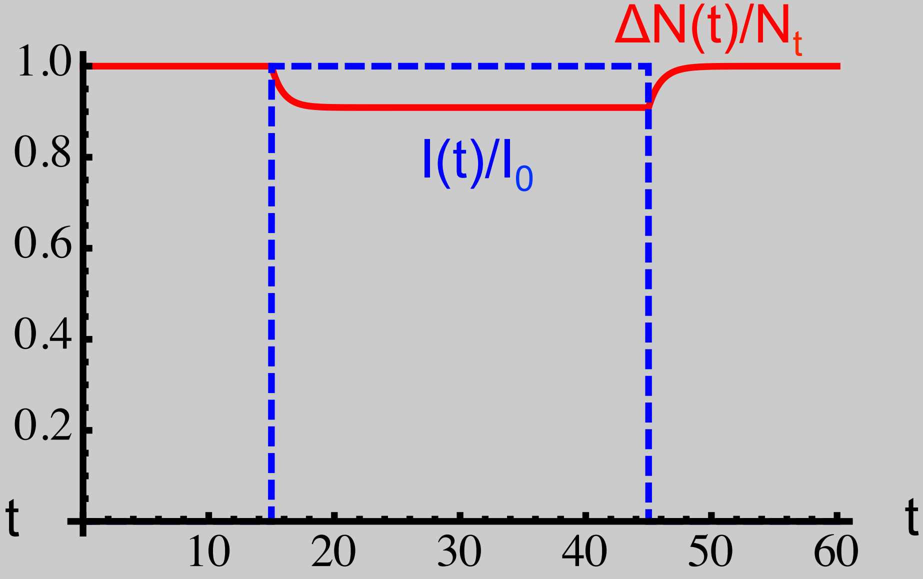
- For *long* pulse input: $\tau_p >> \tau_{21}$, and peak $I << I_{sat}$, $\Delta N(t)$ follows $I(t)$

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

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

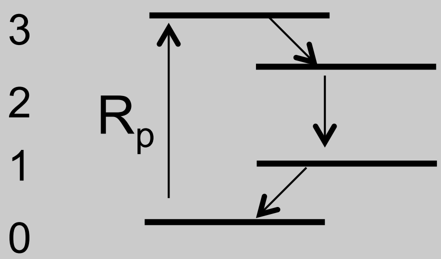
$$\frac{\Delta N}{N_t} = \frac{1}{1 + I(t)/I_{sat}}$$





Gain saturation

- Consider a 4-level system:



Assume: τ_{32} and $\tau_{10} << \tau_{21}$ 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: $$N_2 = \frac{R_P\tau_{21}}{1+W\tau_{21}} = \frac{R_P\tau_{21}}{1+\frac{\sigma_{21}\tau_{21}}{h\nu_{21}}I} = \frac{R_P\tau_{21}}{1+\frac{I}{I_{sat}}}$$

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

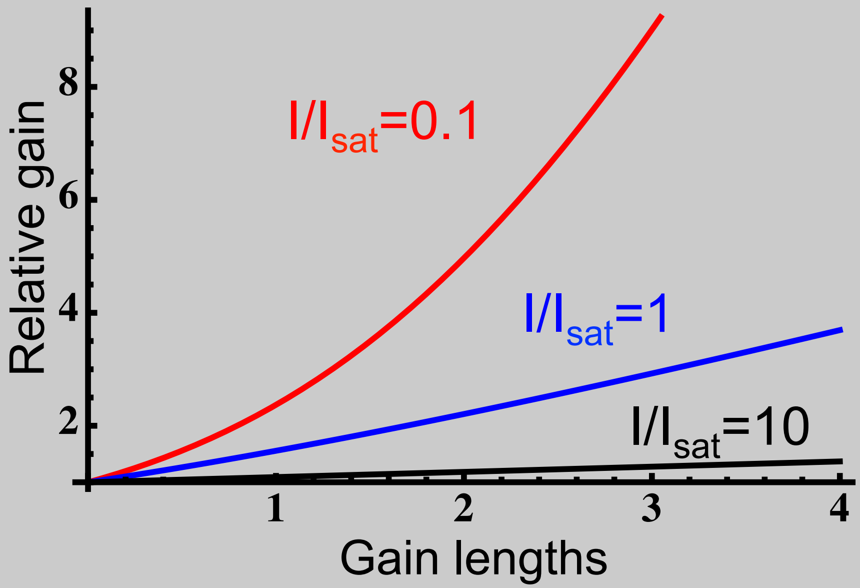
$$I_{sat} = \frac{h\nu_{21}}{\sigma_{21}\tau_{21}} = \frac{\Gamma_{sat}}{\tau_{21}}$$

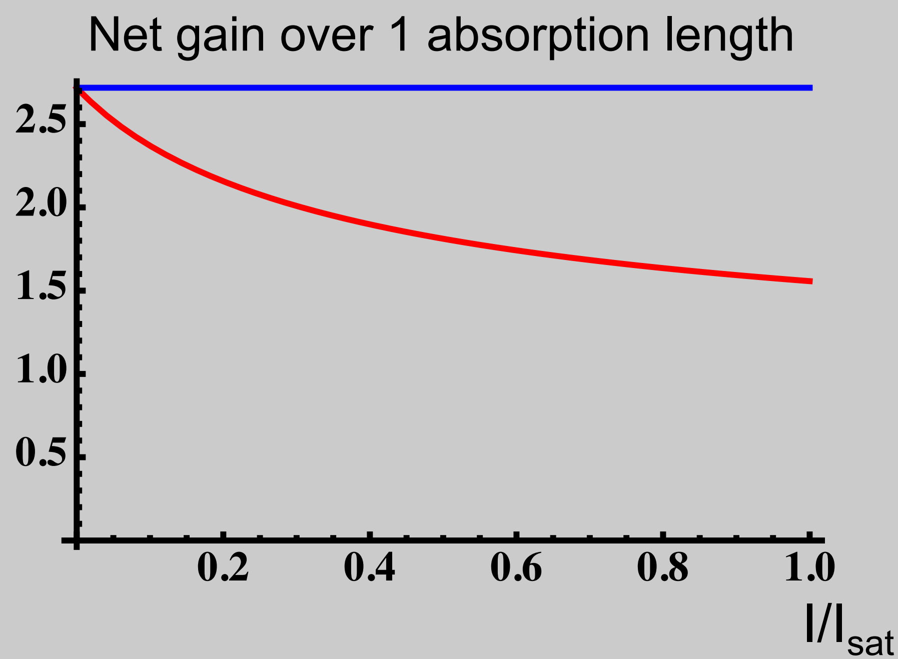
$$g(I) = \frac{g_0}{1+I/I_{sat}}$$

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**

Spatial dependence

- Gain follows distribution of pump intensity
- Spatial variation of gain affects beam profile
- Examples:
  – longitudinal pumping with Gaussian beam leads to gain narrowing of spatial profile. More gain in center, less at edges
  – Saturated absorption by a Gaussian beam: saturation in center suppresses intensity there. Leads to widening of output beam.

Pulse amplification: saturated gain algorithm

Frantz-Nodvick Equation:

$$G = \frac{\Gamma_{sat}}{\Gamma_{seed}} \ln[1 + (e^{\Gamma_{seed}/\Gamma_{sat}} - 1)e^{\Gamma_{Pump}/\Gamma_{Sat}}]$$

No Spatial Dependence:

$$F_{Pump} = \frac{E_{Abs}}{\pi w_P^{\,2}}$$

$$F_{Seed} = \frac{E_{Seed}}{\pi w_S^{\,2}}$$

→ **Frantz-Nodvick** → Gain

Assumptions:

- Thermal Equilibrium within Stark Manifolds
- Square Temporal Profile of Seed

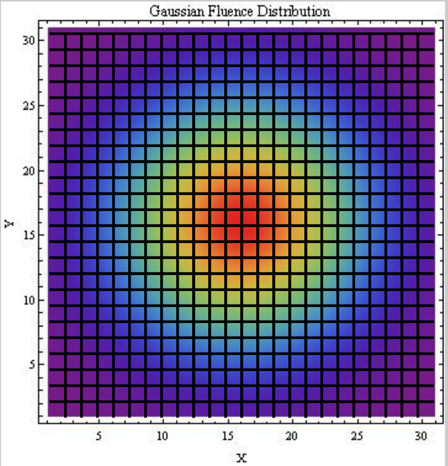
Transverse dependence: super-Gaussian

$$\Gamma(x,y) = \Gamma_0 e^{\frac{-[(\frac{x}{w_x})^{nx} + (\frac{y}{w_y})^{ny}]}{2}}$$

where:- ***nx,ny*** = 2 (Gaussian),
Even > 2 (Super-Gaussian)

-F_0 is defined via the Total Energy and integration of the distribution

(Δx, Δy) →



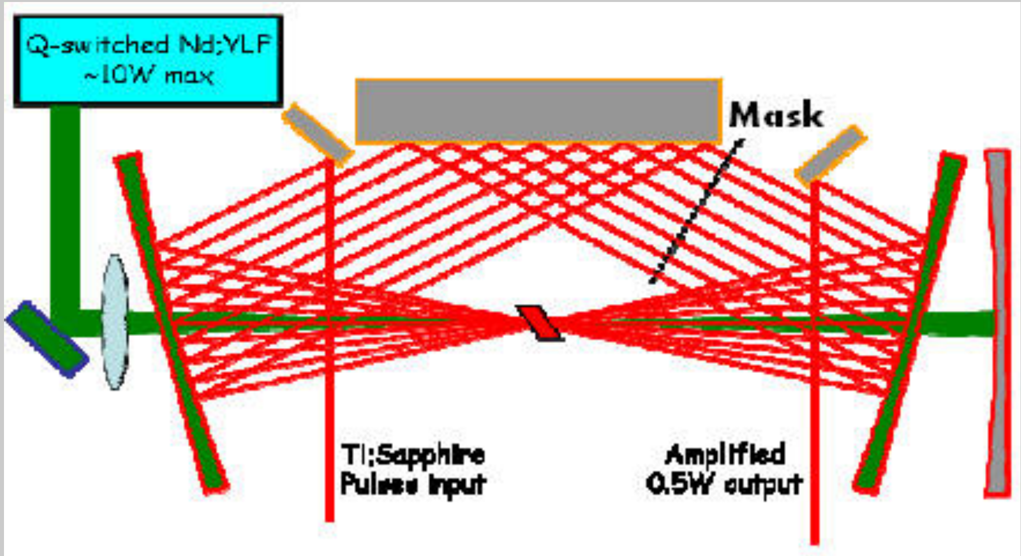
Define Spatial Arrays → **Define Gain Array** *(Frantz-Nodvick for each element in spatial array)* → $F'_{Seed} = G \bullet F_{Seed}$ → **Compute Post-Gain Seed Energy** (Integrate Spatial Array) →

$$Gain = \frac{E'_{Seed}}{E_{Seed}}$$

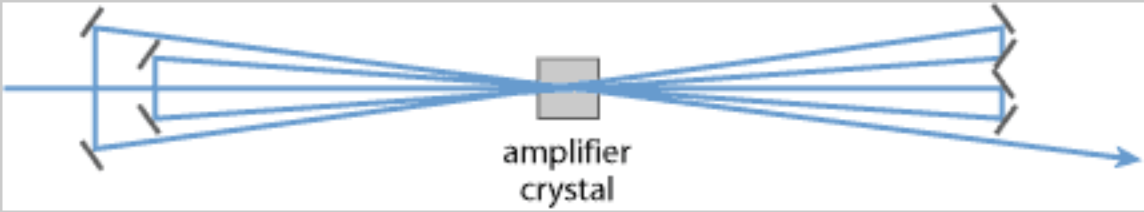
Example: Ti:sapphire multipass amp

- Seed pulse from pulsed laser oscillator: 1nJ (800nm)
- Amplify to 1mJ, use 7mJ of pump energy (532nm)
- Multipass designs: spatially separate beams

Three-mirror ring preamp:
- Up to 12 passes
- Focused beam in crystal
- 2 mirror alignment



Bowtie power amp:
- Collimated beam
- 8 mirrors



Multipass design

- Assume uniform pumping with round beams
- Calculate stored fluence and small signal gain
- Use saturated gain expression to calculate new energy after 1st pass
- Subtract extracted energy from stored energy (over seed spot area)
- Repeat for N passes

Conditions: 1nJ seed, 7mJ pump energy, 95% absorption, 10% loss/pass

Stored energy:

$$E_{stor} = E_{pump}\eta_{abs}\frac{h\nu_{seed}}{h\nu_{pump}} = 4.4mJ$$

Small signal gain estimate:

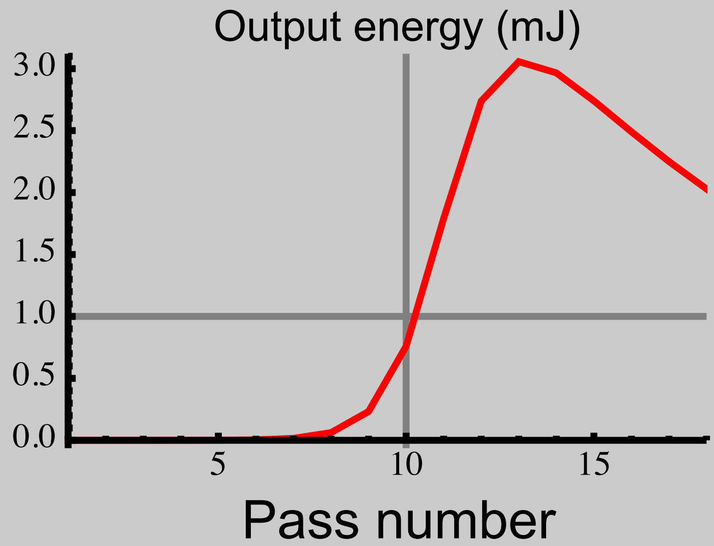
$$G_0 = \left(\frac{E_{\text{target}}}{E_{\text{seed}}}\right)^{1/N}\frac{1}{1-L} = 4.42$$

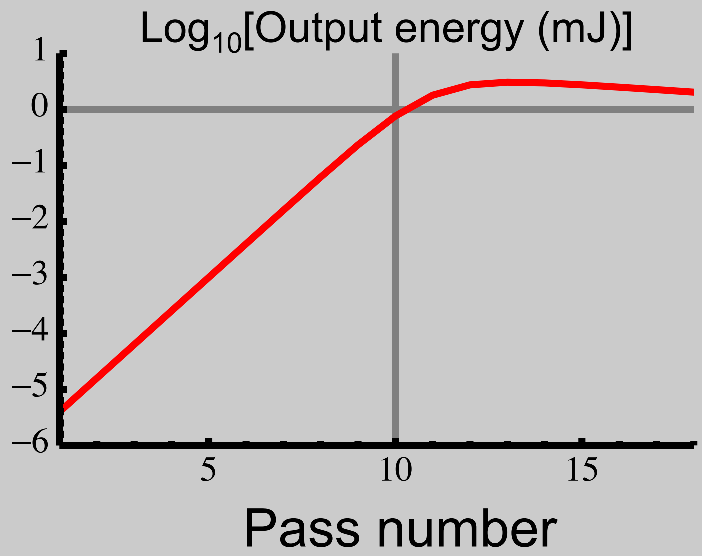
Estimated spot size:

$$A_{pump} = \frac{E_{stor}}{\Gamma_{sat}\ln\left[G_0\right]}, \quad w_p = 300\mu m$$

Multipass: Simple calculated results

- Small signal gain estimate works as long as stored energy is not depleted

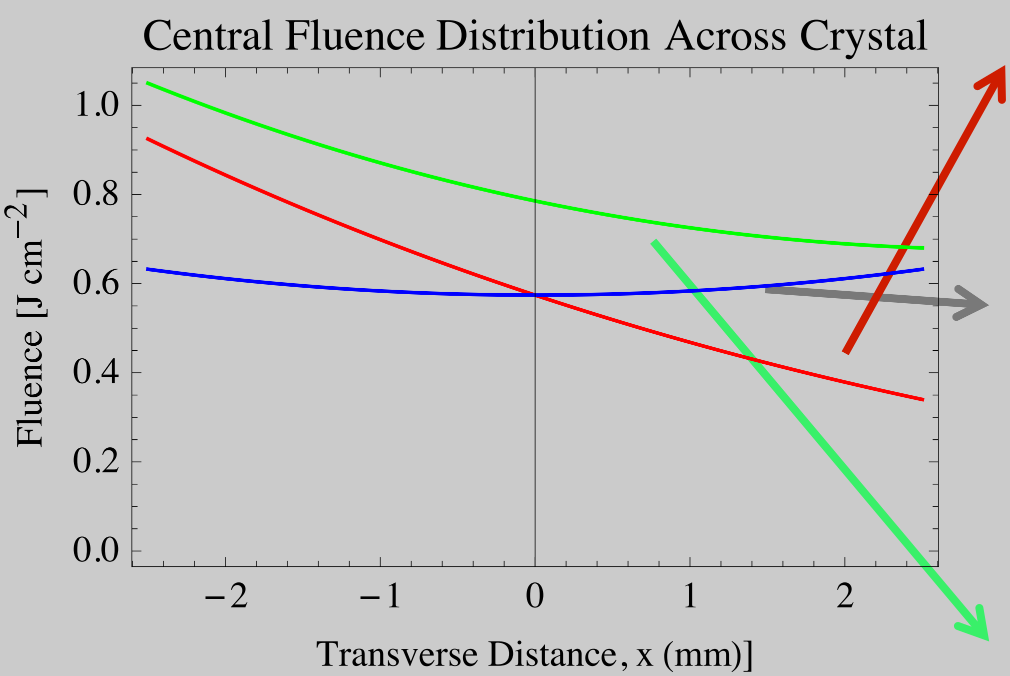


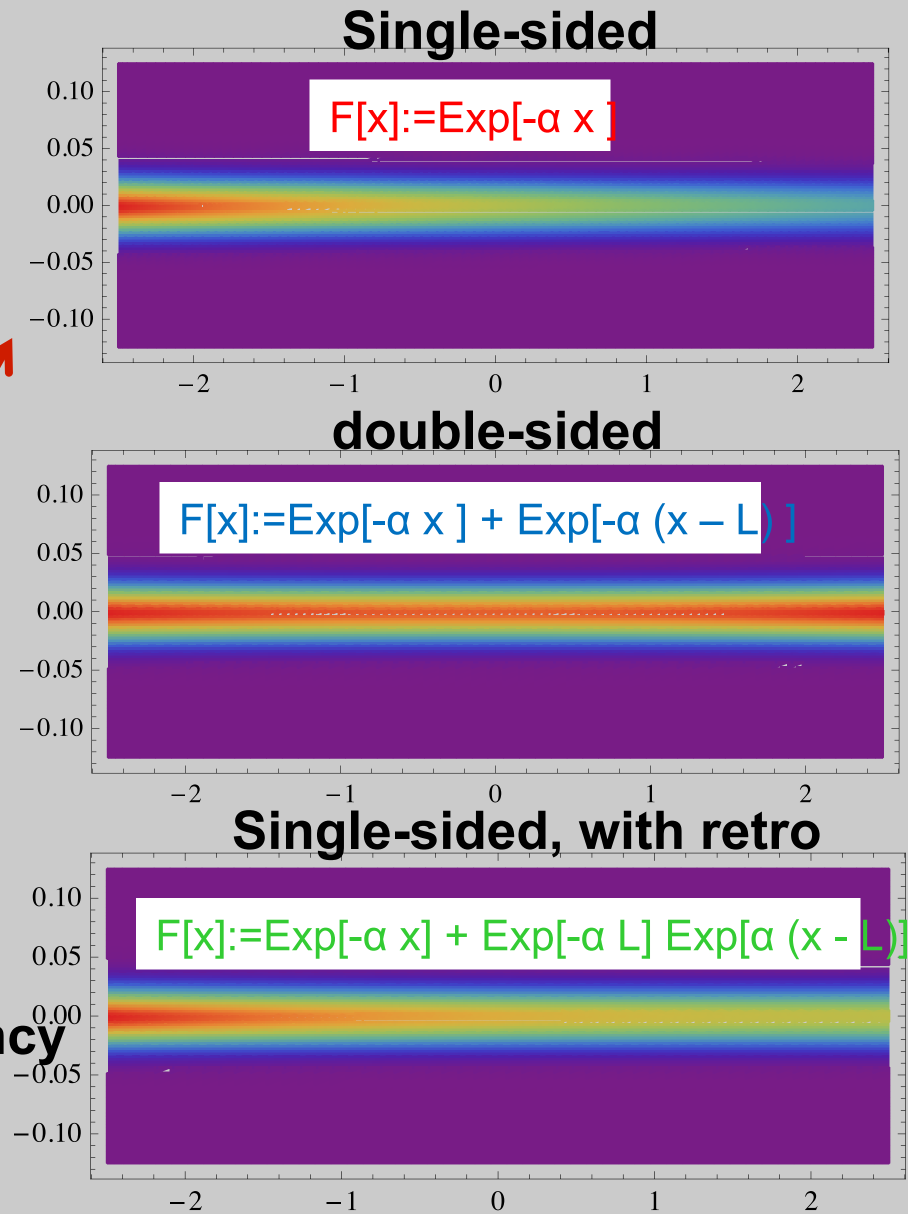


  - Smaller seed size to ensure full overlap with pump
  - Avoid damage thresholds for pump and seed
  - Saturate at desired energy to reduce noise
  - Account for size change in Brewster cut crystal

Transverse diode bar pumping

For good absorption, pump must have sufficient path length



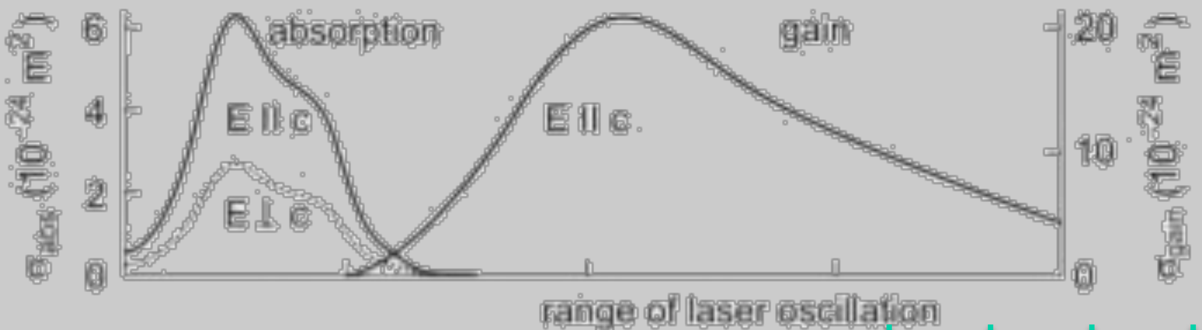


Using retro: better absorption efficiency

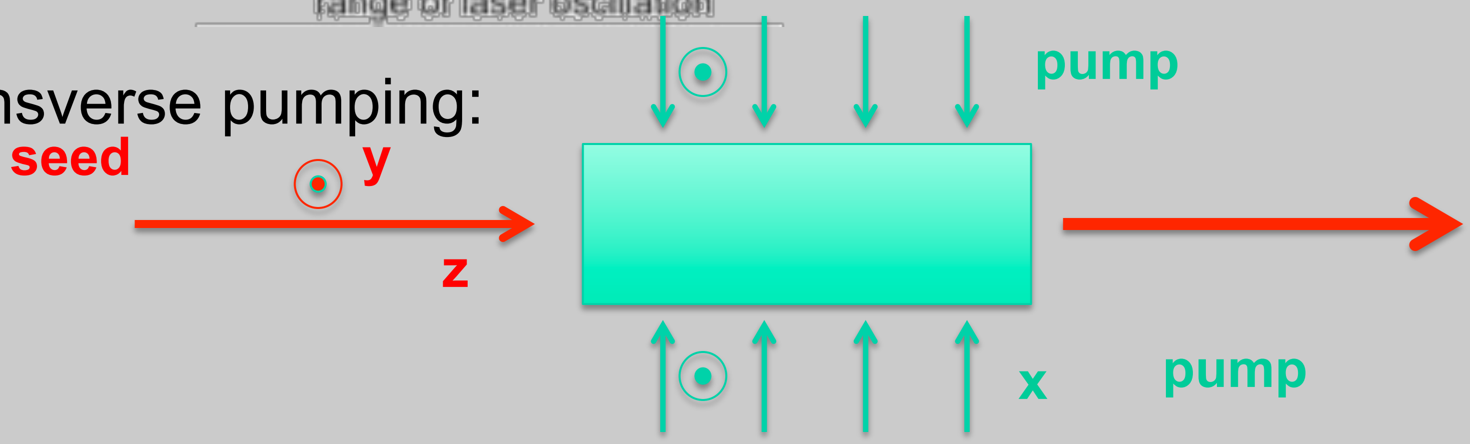
Double-sided: better uniformity

Polarization issues in pumping birefringent materials

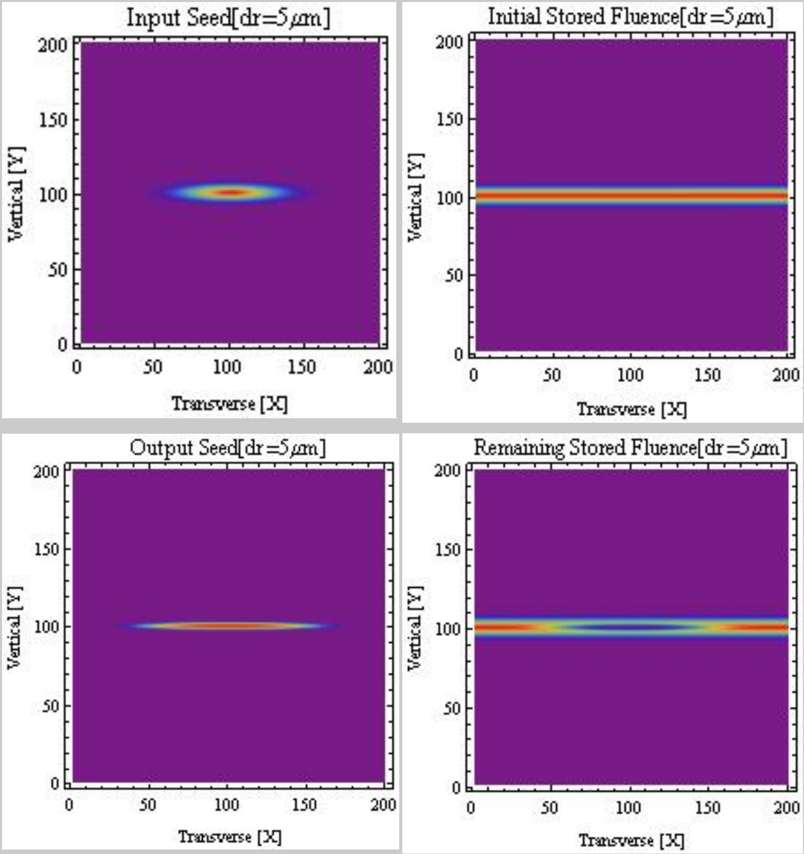
- For Ti:sapphire, both polarizations contribute to seed gain along c-axis
- Much higher pump absorption for E along c-axis
  - α across c-axis is about 40% lower than along c-axis



- Ex: transverse pumping:



Transverse Pumping Gain Estimates



- Seed: 2nJ
  - Cavity Losses: ~1%
  - τ_{pass}: 1ns
- Pump (CW): 1kW (Total: 2X .5kW Bars)
  - η_{Abs}=63.2%
  - η_{QD}=55.6%
  - $\eta_{Pump}=\eta_{Abs}\ \eta_{QD}$=35.1%
  - Heat: ~560 W
  - Significant (Cylindrical) Thermal Lens Expected
  - w=30um
- Single Pass Gain (small signal)
  - Astigmatic Seed: g≈1.64
    - w_x=200um, w_y=30μm
  - Spatially Chirped Seed: g≈1.64
    - w_x=2mm, w_y=30μm

•Multi-Pass Extraction: 37 Passes

-Astigmatic Mode: ~136uJ (small extraction area)

-Spatially Chirped:~.53mJ (46% extraction)

Central dip in gain: spatial gain mode *expansion*.
This could be used to counter gain narrowing for spatially-chirped seed

Frequency dependence: account for lineshapes

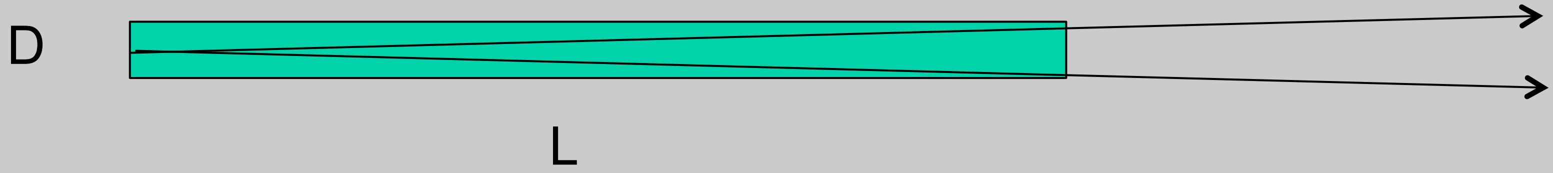
- Absorption and gain coefficients and saturation intensity both depends on frequency

$$\alpha(I,\nu)=\frac{\alpha_0(\nu-\nu_0)}{1+\frac{I(\nu)}{I_{sat}(\nu-\nu_0)}}$$

- For broadband input, saturation changes shape of transmitted spectrum
  - Absorption: power broadening
  - Gain: spectral gain narrowing

Amplified Spontaneous Emission (ASE)

- Spontaneous emission is emitted into 4π steradians, but is amplified on the way out if there is gain.



  - ASE can be considered to be a noise source
  - ASE is more directional than fluorescence, but not as directional as a coherent laser beam
  - Some high-gain lasers are essentially ASE sources (no mirrors)
- Implications for amplifier design
  - ASE can deplete stored energy before pulse extraction
  - Use timing and good seed energy to extract energy from medium before ASE
  - Ensure that transverse gain is smaller than longitudinal to avoid parasitic depletion.

Self-absorption and “optically-thick” media

- A related phenomenon for an absorbing medium is when radiation is *absorbed* along the way out.
- More absorption near the line center, so the transmitted light is broader in spectrum.
- For an extended luminous body (e.g. the Sun), the individual spectral lines get merged together to look like the blackbody.