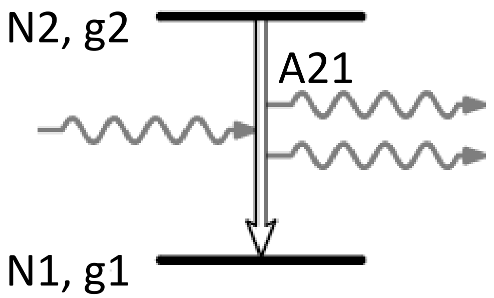
Lasers

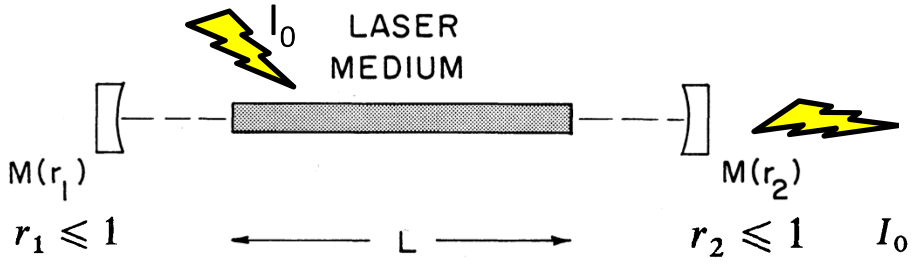
Requirements:

- metastable excited state
- population inversion



The absorption coefficient of the laser medium:

$$\alpha = \frac{c^2 A_{21}}{8\pi \nu^2 n^2}\left(\frac{g_2}{g_1}\right)\left(N_1 - \frac{g_1}{g_2} N_2\right) \qquad N_1 < \frac{g_1}{g_2} N_2 \qquad \alpha < 0 \qquad \text{Amplification, } I > I_0 \qquad (1)$$



$r_1 \leqslant 1$ $\qquad$ $r_2 \leqslant 1$

$$I_0 e^{-2\alpha L} e^{-2\gamma} > I_0 \qquad (2)$$

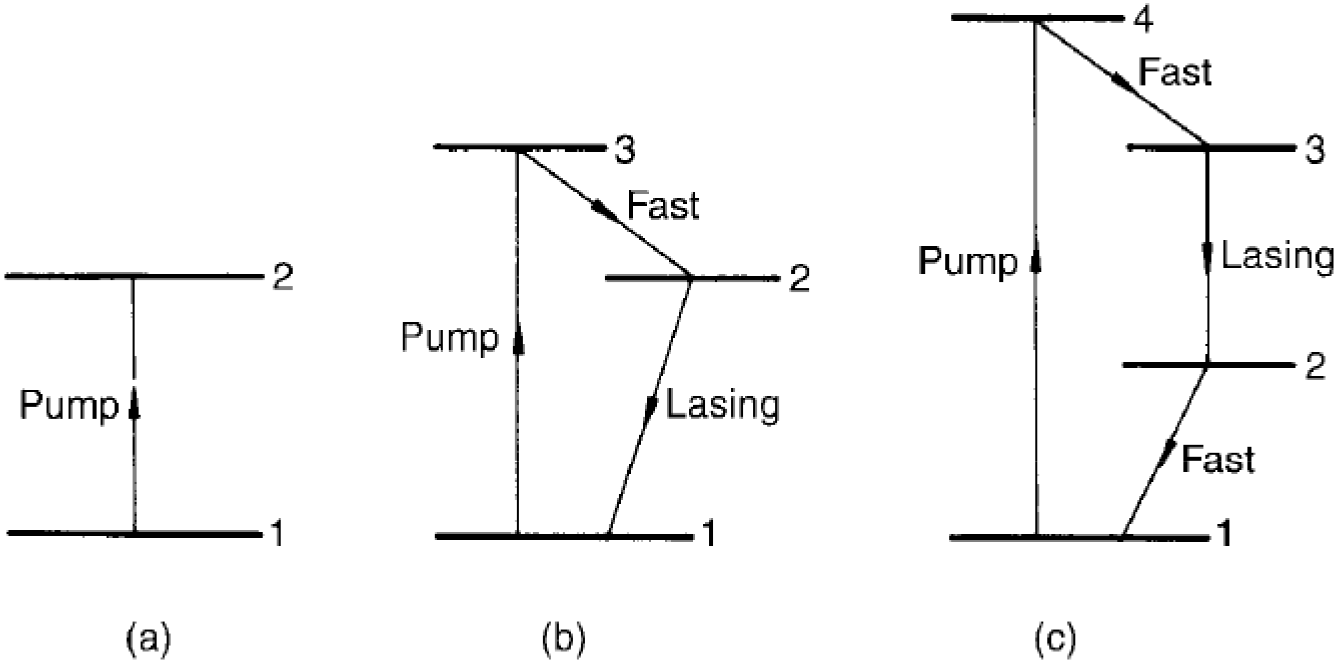
$I_0 \exp(-2\alpha L)$

$r_1 r_2 \equiv \exp(-2\gamma)$

$$-\alpha(\nu) > \gamma / L$$

$$\left(\frac{g_1}{g_2}\right) N_2 - N_1 > \frac{8\pi n^2 \nu^2}{c^2 A_{21}} \left(\frac{g_1}{g_2}\right) \frac{\gamma}{L} \qquad (3)$$

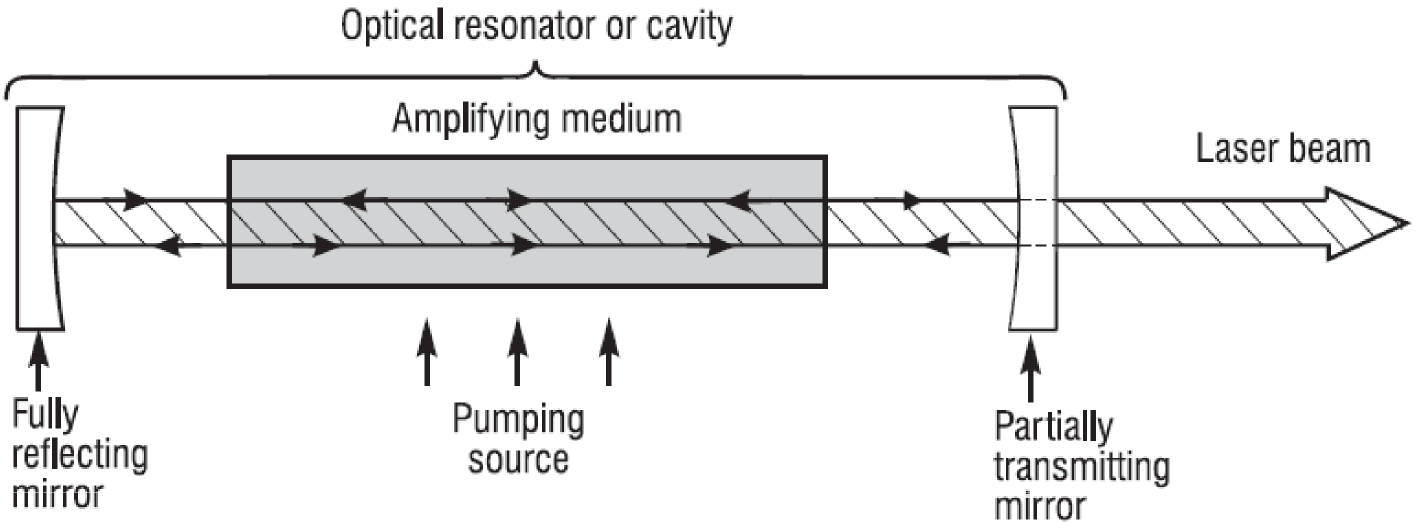
The **threshold condition** for the population difference $\qquad \Delta N = N_2 - N_1 > \frac{8\pi n^2 \nu^2}{c^2 A_{21}} \frac{\gamma}{L}$ $\qquad$ (4)

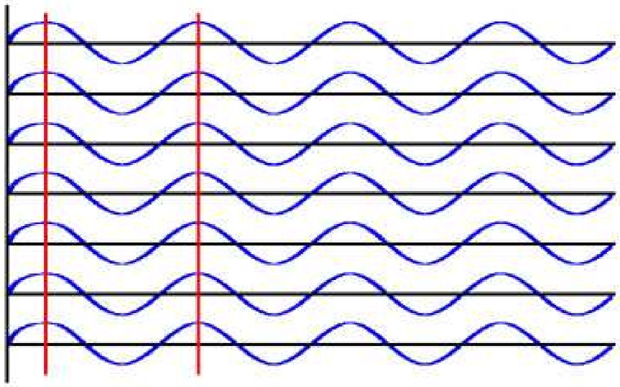


Methods of pumping:

- Optical pumping: solid and liquid lasers
- Electrical pumping: gas and semiconductor lasers

Laser properties





Laser radiation has four remarkable properties:

1. Directionality: divergence only few milliradians

2. Monochromaticity: laser cavity is resonant only for particular freque

3. Brightness: 10^8–10^9 W/cm^2

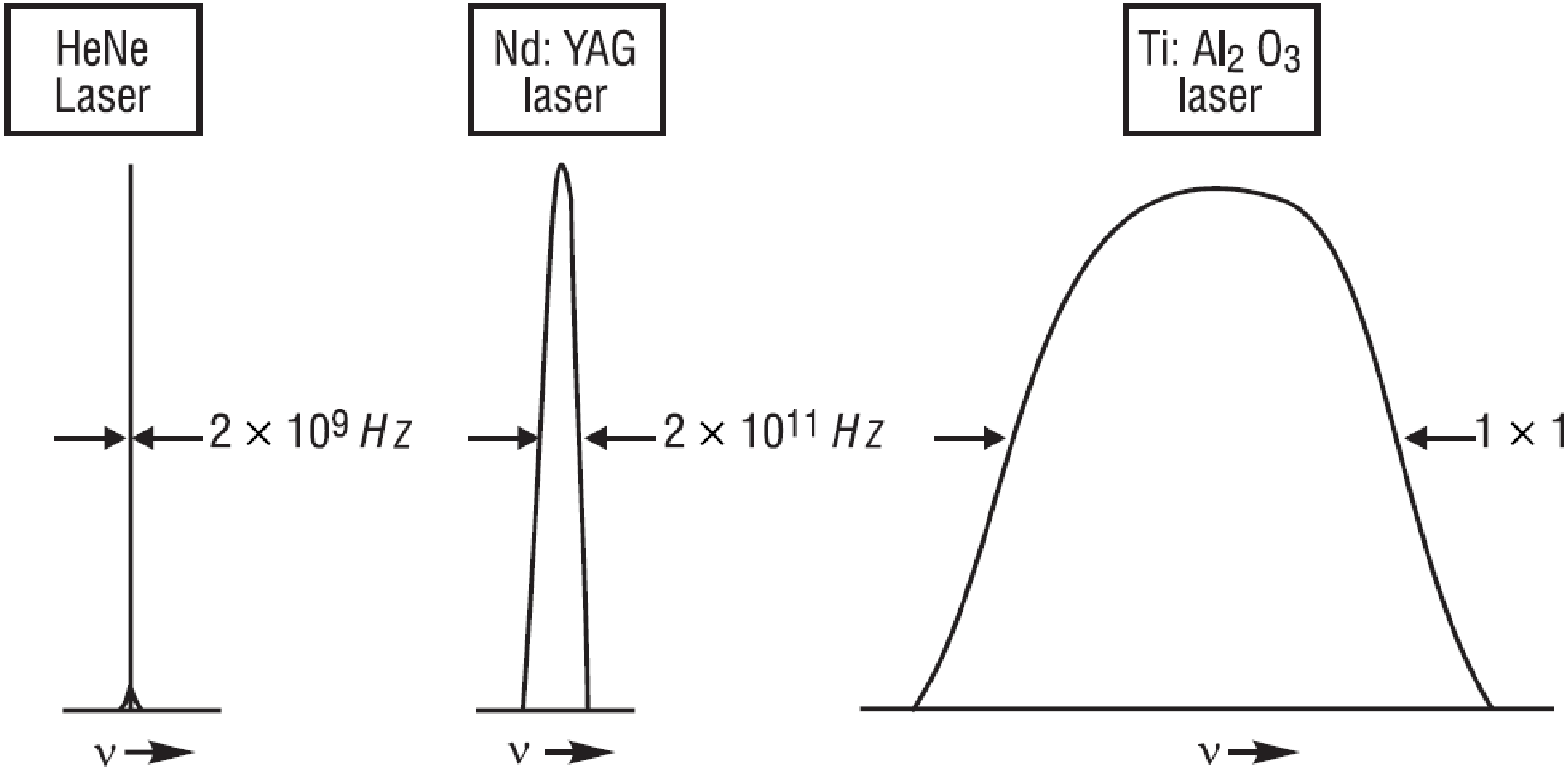
4. Coherence: spatial
temporal

$$l_C = \frac{\lambda^2}{2\Delta\lambda}$$

Light bulb: l_c = 2 pm
He-Ne laser: l_c = 10 m

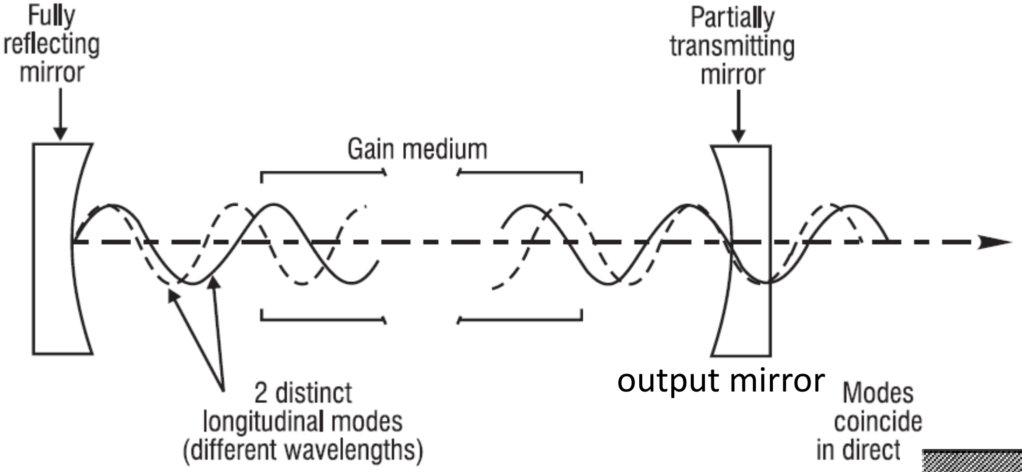
Bandwidth of laser gain medium

$$\Delta\lambda_G = \frac{\lambda^2}{c}\Delta\nu_G$$



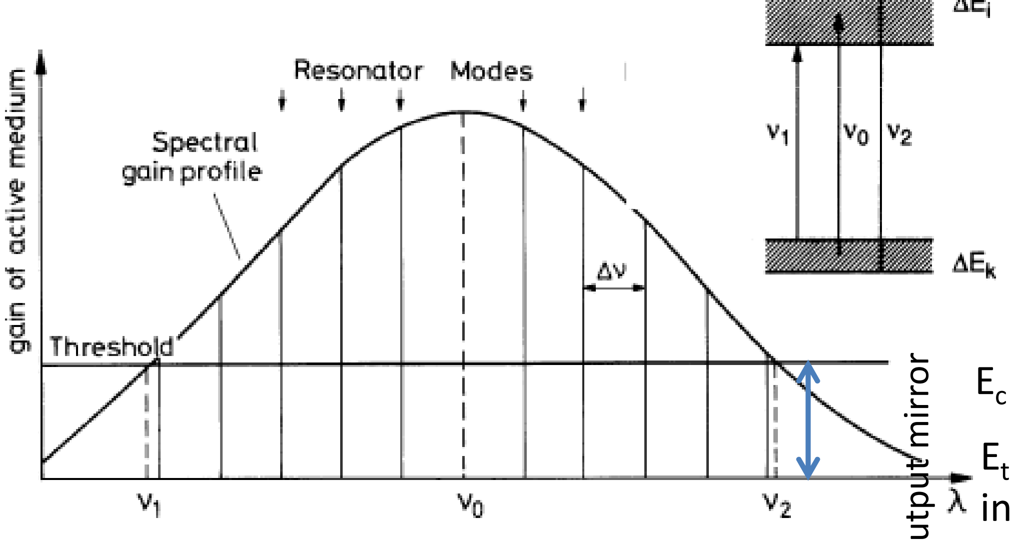
Laser gain bandwidths for the HeNe, Nd:YAG, and Ti:Al_2O_3 lasers

Laser Cavity Resonance Modes: longitudinal modes



output mirror

$$n \times \tfrac{1}{2}\lambda = L$$



loss at output mirror

$$\Delta\nu = c/\,2L$$

Quality factor of a laser cavity:

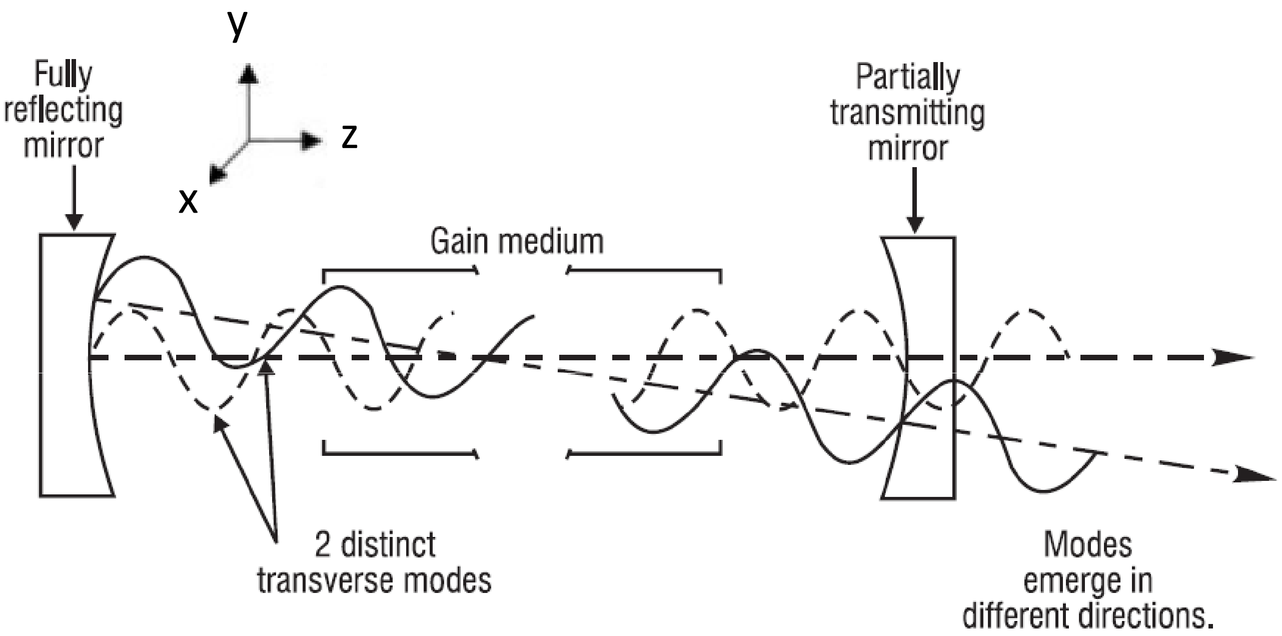
$$Q = \frac{\nu}{\Delta\nu} \qquad (5)$$

$$Q = \frac{2\pi\nu E_c t}{E_t}$$

E_c energy stored in the cavity

E_t energy allowed to leak out in time t

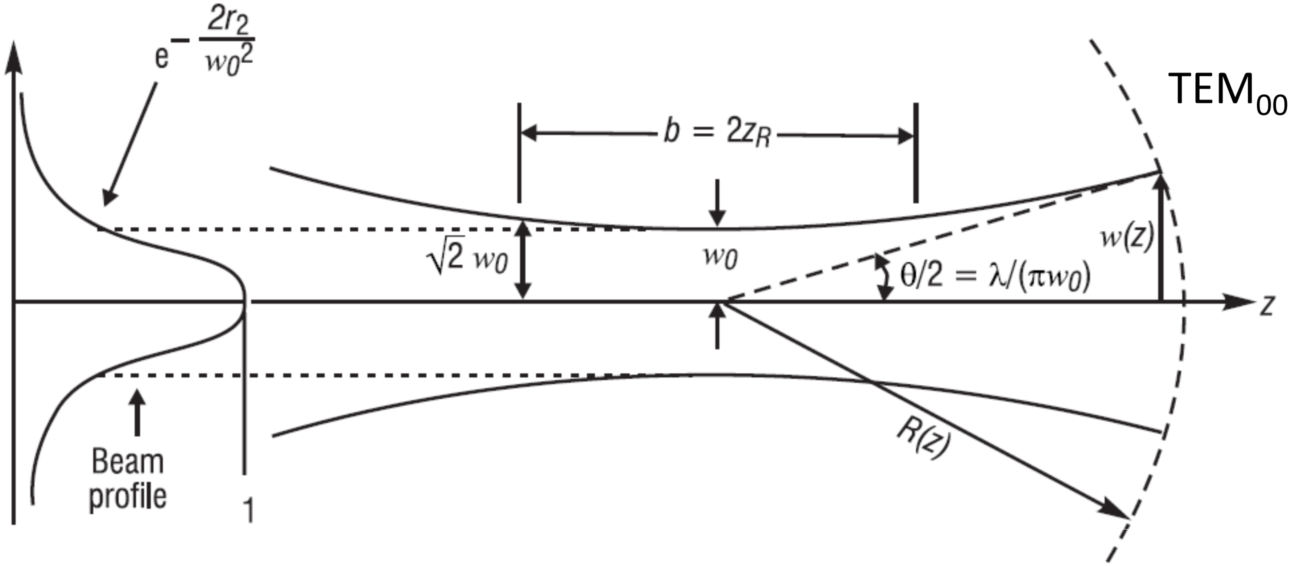
Laser Cavity Resonance Modes: transversal modes



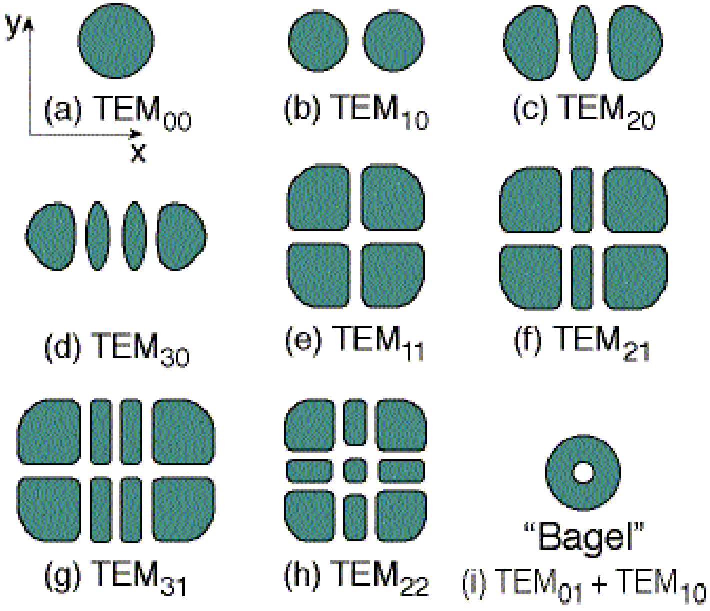
$$\mathrm{TEM}_{pq}(x,y) = H_p\left(\frac{\sqrt{2}x}{w}\right) H_q\left(\frac{\sqrt{2}y}{w}\right) e^{-\frac{(x^2+y^2)}{w^2}}$$

w = mode radius (radial intensity distribution)

Laser Cavity Resonance Modes: transversal modes



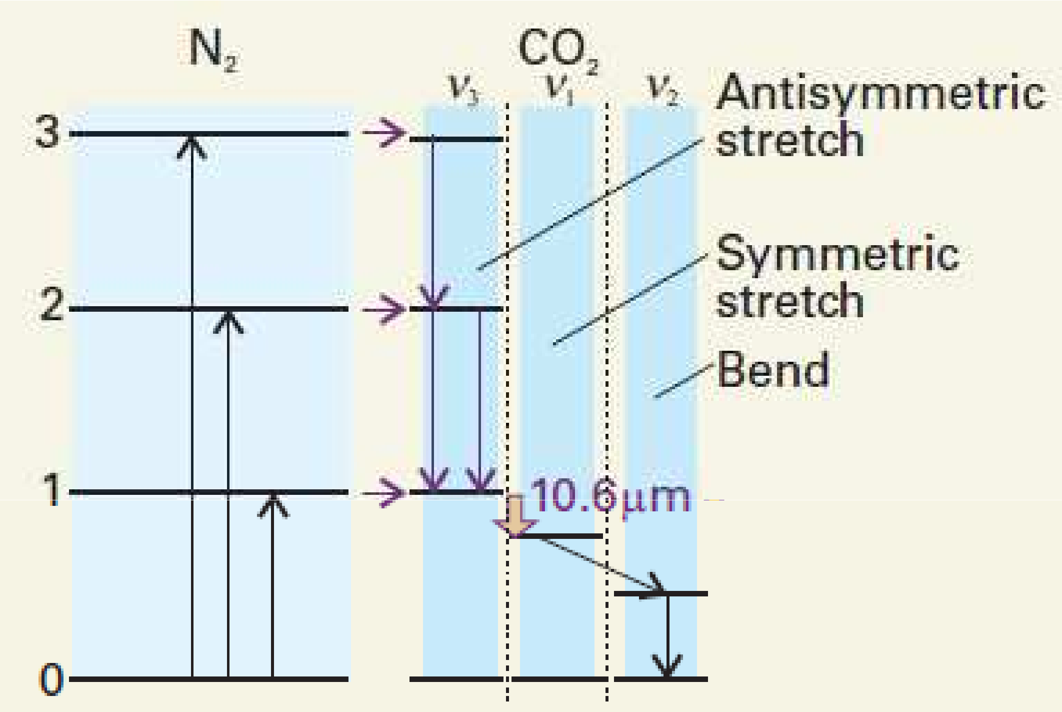
TEM$_{00}$: $I = I_0 e^{-\frac{2r^2}{w^2}}$

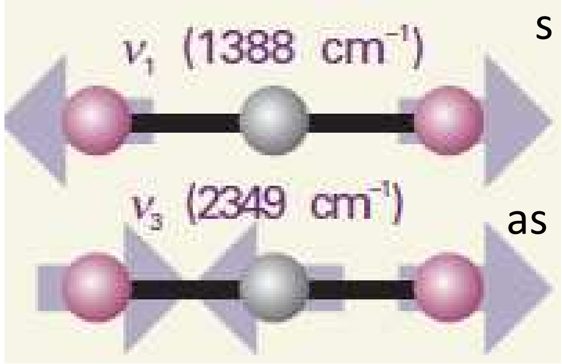


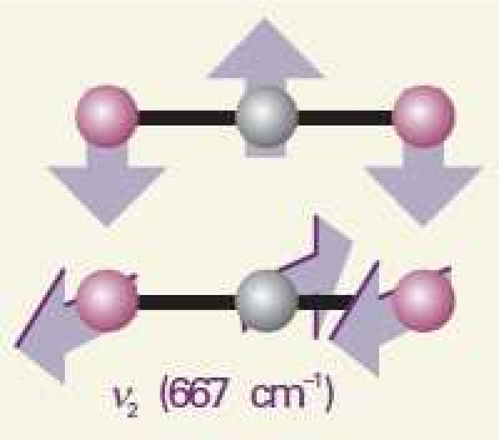
CO_2 laser

cw powers : 100 kW
pulsed energies : 10 kJ
λ_{em}: 10.6 µm

$CO_2 : N_2 : He$ gas mixture





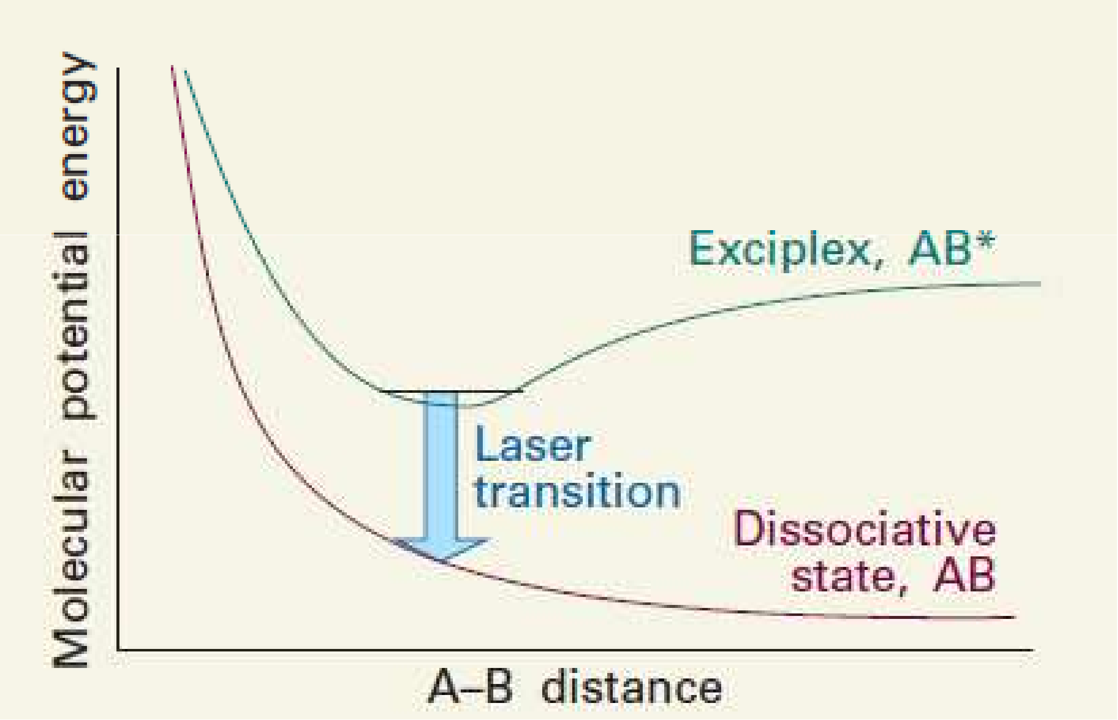


CO2 vibrations

Excimer / Exciplex lasers

Ex:

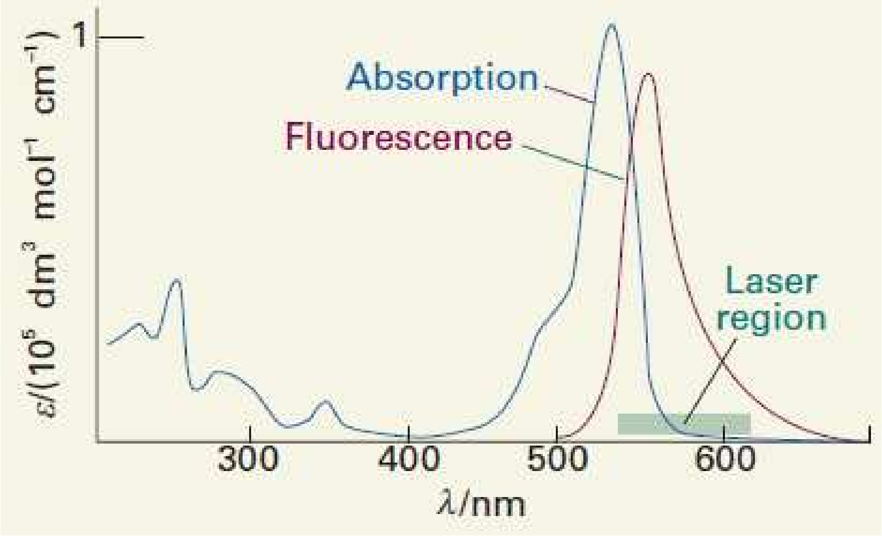
ArF (193 nm), KrF (248 nm), XeF (351 nm), KrCl (222 nm), XeCl (308 nm) XeBr (282 nm)



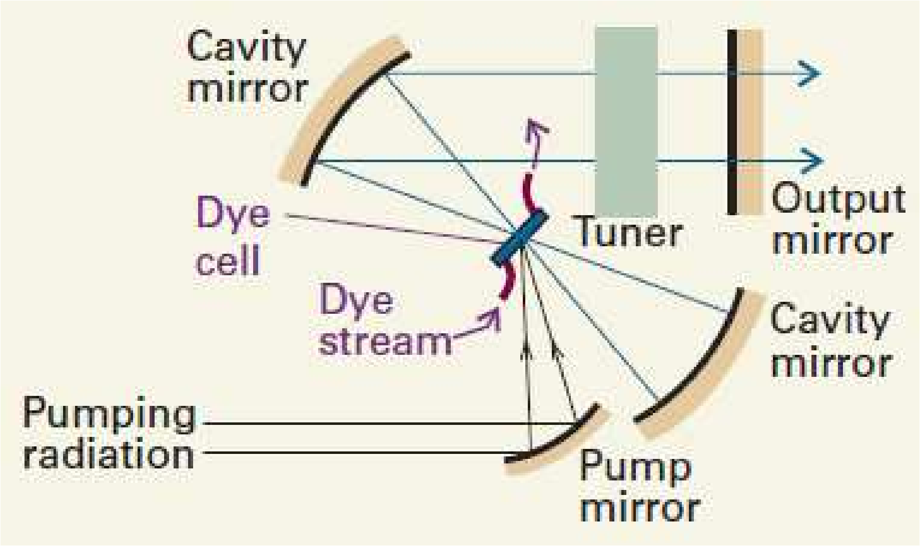
10–50-ns pulses of 0.2 to 1 J/pulse at repetition rates of up to 1 kHz

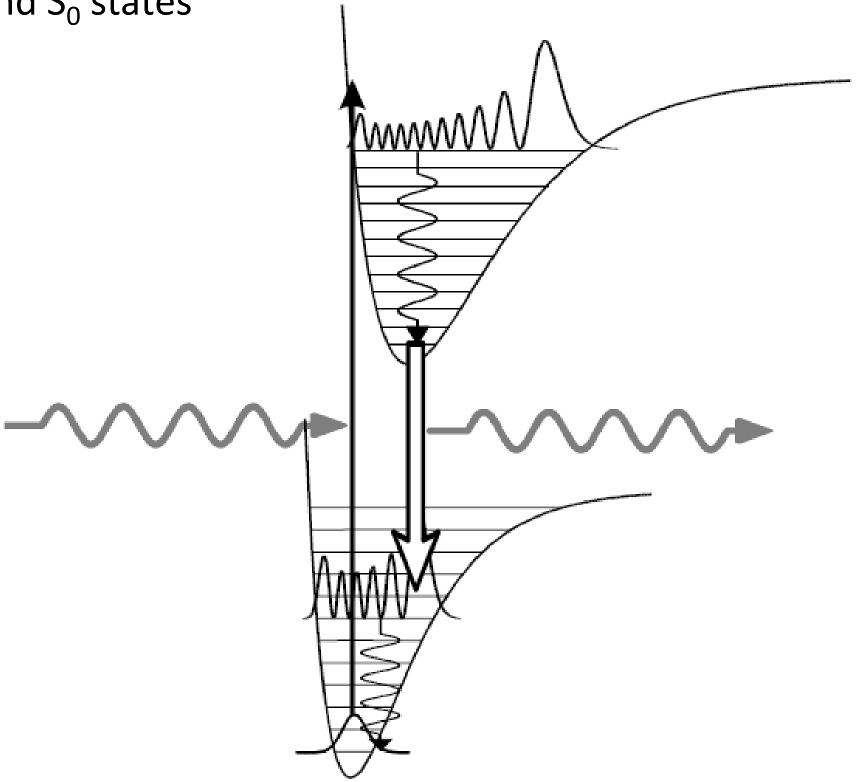
Dye lasers

Lasing region: 320 - 1500 nm
pulsed mode: 50–100 mJ/pulse
cw mode: few W

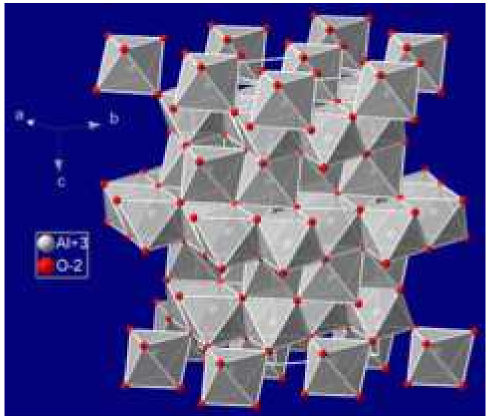


laser dyes: significant geometry change between the S_1 and S_0 states



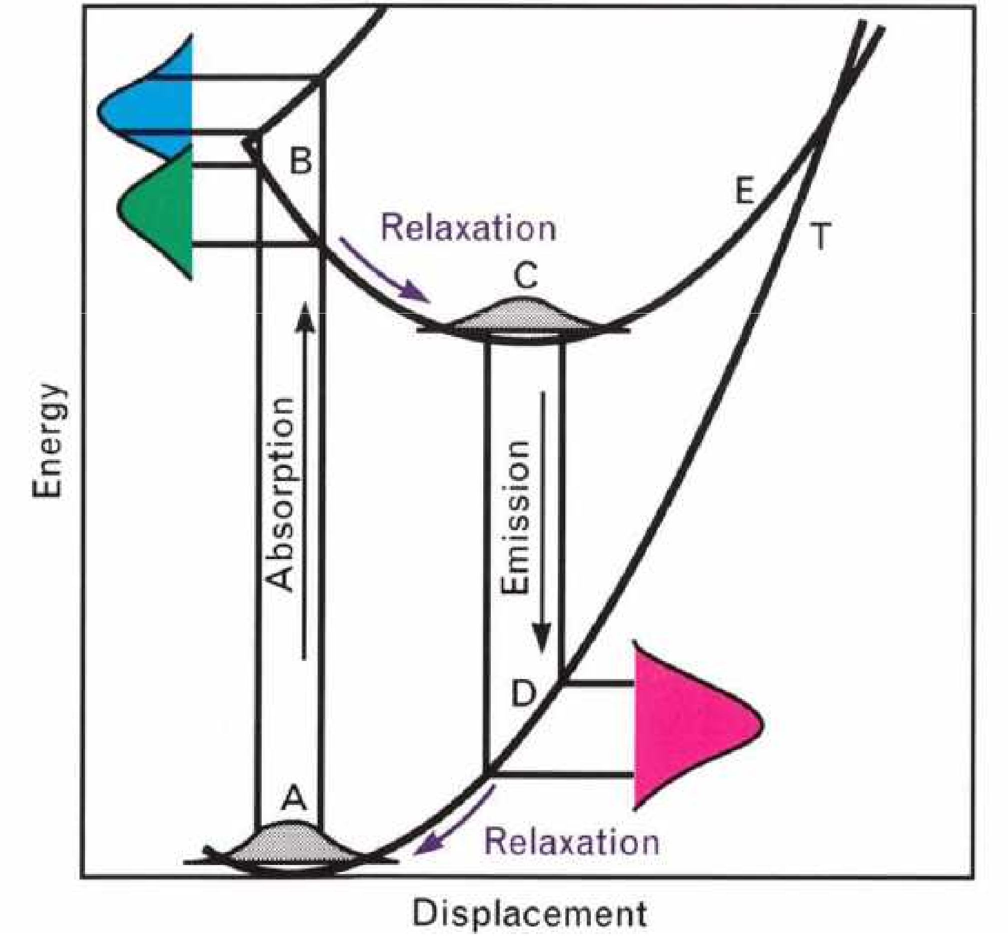


Tunable solid-state lasers

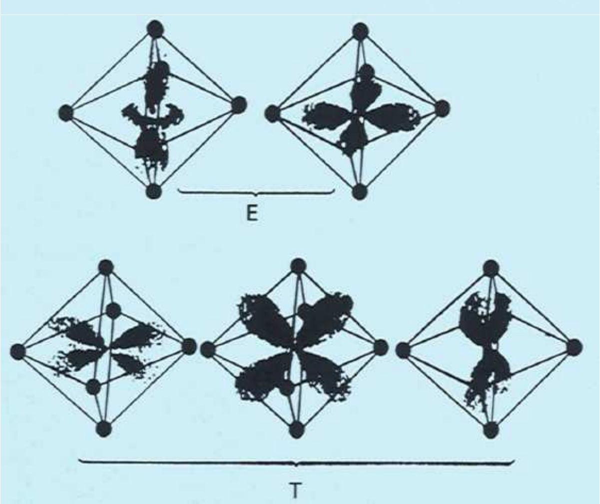


Ti:Al_2O_3

660-1180 nm when pumped by argon laser
10 fs pulse length



Ti [Ar] $3d^2\ 4s^2$



Ti^{3+} in sapphire lattice

Generation of short pulses (ps-fs): Mode locking

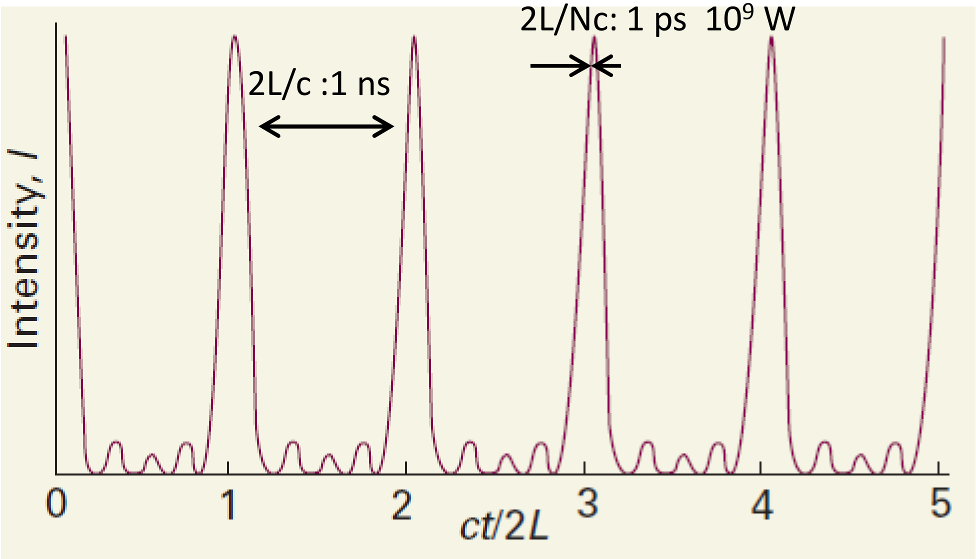
$$\mathcal{E}_n(t) = \mathcal{E}_0 e^{2\pi i(\nu + nc/2L)t} \qquad (6)$$

Superimposing *N modes:*

$$\mathcal{E}(t) = \Sigma_n \mathcal{E}_n(t) = \mathcal{E}_0 e^{2\pi i\nu t} \sum_{n=0}^{N-1} e^{i\pi nct/L} \qquad (7)$$

$$\sum_{n=0}^{N-1} e^{i\pi nct/L} = 1 + e^{i\pi ct/L} + e^{2i\pi ct/L} + \cdots = \frac{\sin(N\pi ct/2L)}{\sin(\pi ct/2L)} \times e^{(N-1)i\pi ct/2L}$$

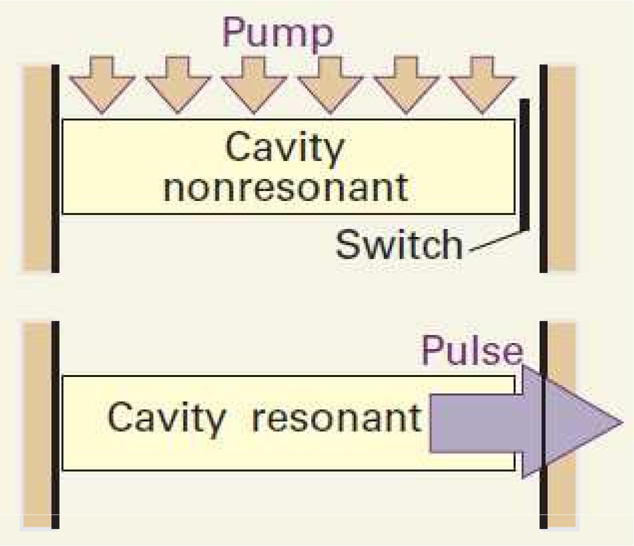
The intensity: $I \propto \mathcal{E}^*\mathcal{E} = \mathcal{E}_0^2 \dfrac{\sin^2(N\pi ct/2L)}{\sin^2(\pi ct/2L)}$ pulse duration: 2L/cN (8)



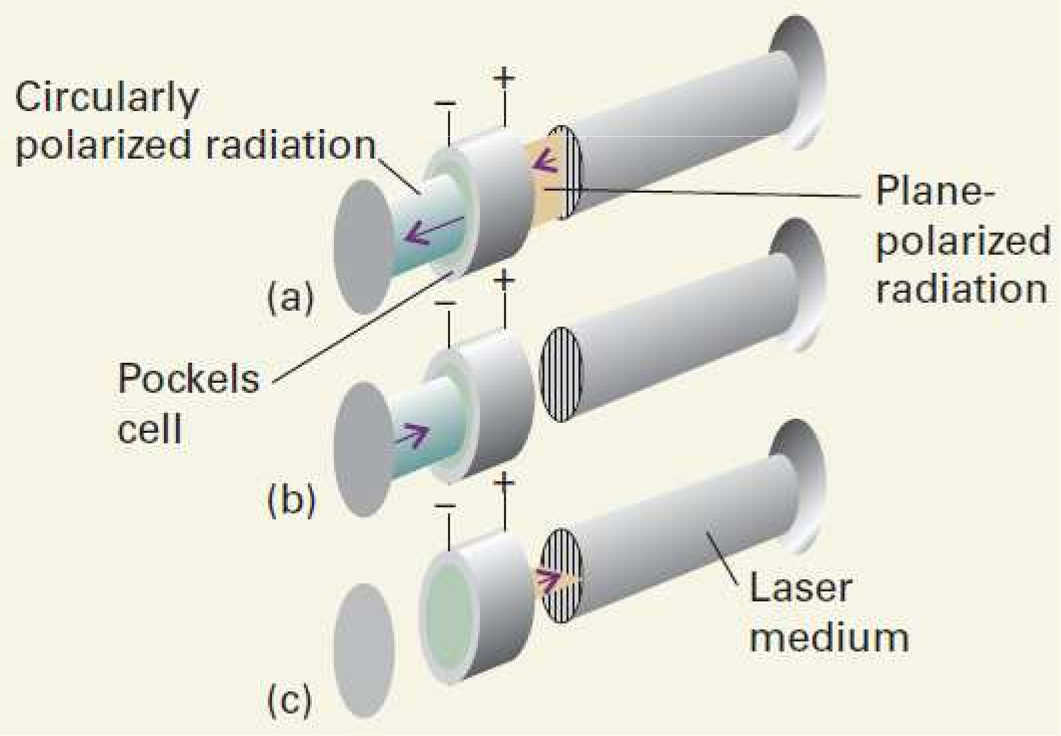
Generation of short pulses:Q-switching

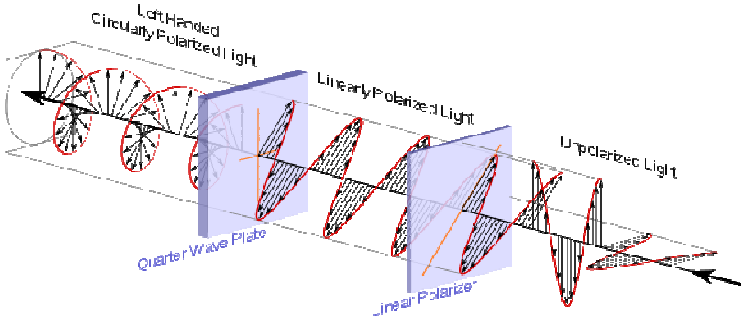
$$P_p = \frac{E_p}{\Delta t}$$

P_p pulse power
E_p pulse energy
Δt pulse duration



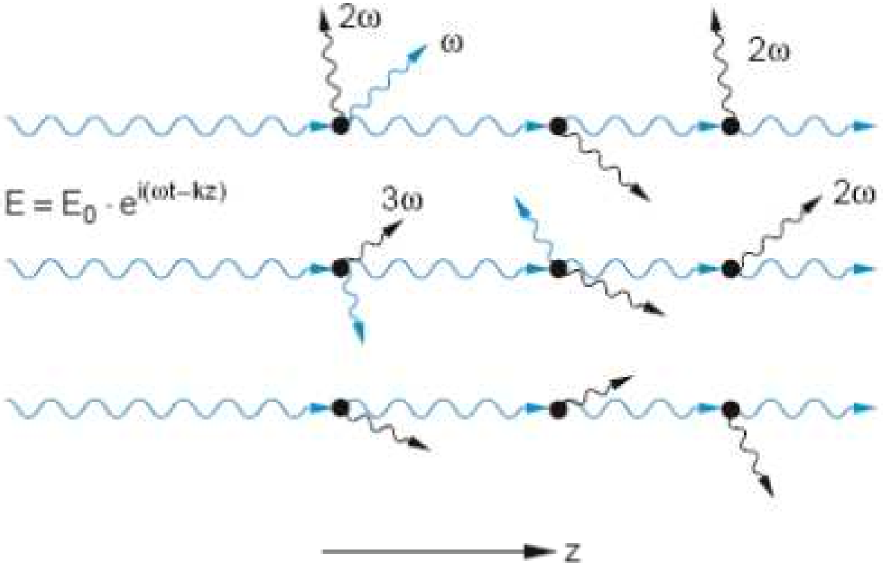
Pockels cell: potassium dihydrogenphosphate (KH_2PO_4)





giant pulses: 1−20 ns peak powers up to 10^9W

Non-linear optical phenomena



Second harmonic generation:

low light intensity: $\mu = \alpha E$

high light intensity (MW / cm^2): $\mu = \alpha E + \frac{1}{2} \beta E^2 + \ldots$

β hyperpolarizability

$E = E_0 \cos \omega t$

$$\beta\mathcal{E}^2 = \beta\mathcal{E}_0^2 \cos^2 \omega t = \frac{1}{2}\beta\mathcal{E}_0^2(1 + \cos 2\omega t)$$

$I(2\omega) \sim I^2(\omega)$

Non-linear optical phenomena

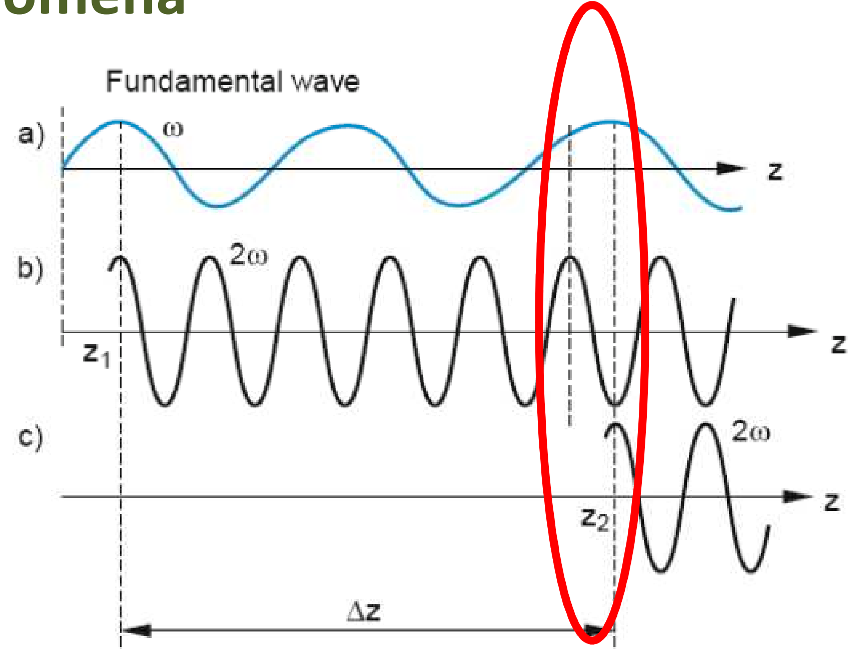
Common materials: KH_2PO_4 $LiNbO_3$ $\beta\text{-}BaB_2O_4$

Plane wave: $E = E_0 \cos(\omega t - kz)$

Second harmonic waves: $v_{ph}(2\omega) = c/n(2\omega)$

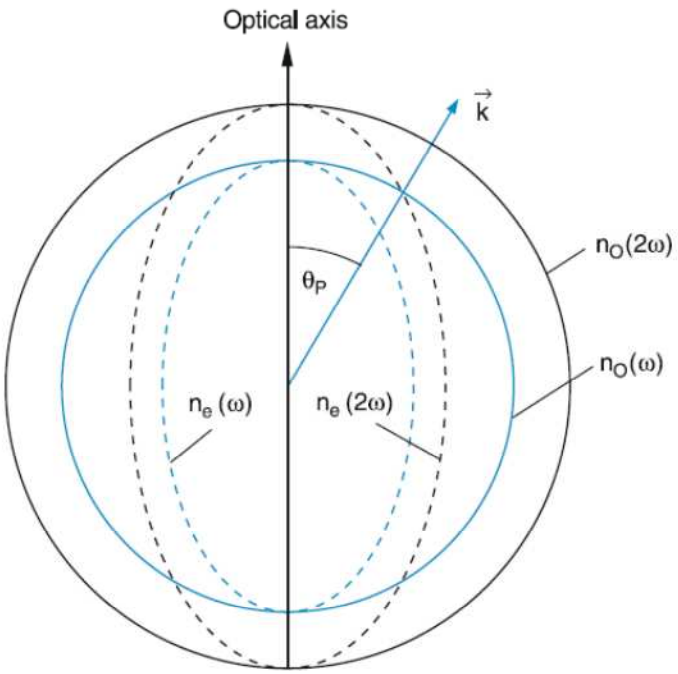
Incident waves: $v_{ph}(\omega) = c/n(\omega)$

$$n(2\omega) \neq n(\omega) \qquad \Delta z = (\lambda/2)/[n(\omega) - n(2\omega)]$$



Phase matching:

$$n_e(2\omega) = n_0(\omega) \Rightarrow v_{ph}(\omega) = v_{ph}(2\omega)$$
$$\Rightarrow \boldsymbol{k}(2\omega) = 2\boldsymbol{k}(\omega) .$$



The neodymium –YAG laser

Nd^{3+} $Y_3Al_5O_{12}$ = yttrium aluminium garnet

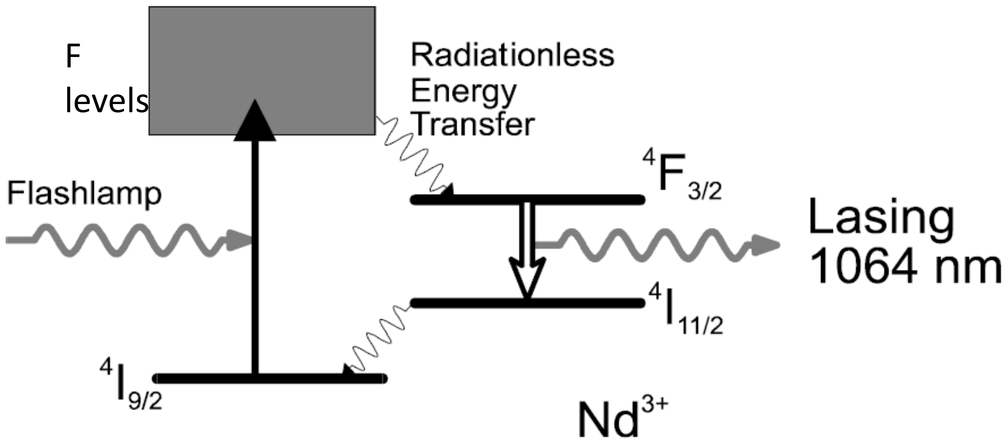
Ground configuration of Nd^{3+} $\ldots 4d^{10}4f^3 5s^2 5p^6$

^4I , ^4F terms are involved in lasing.

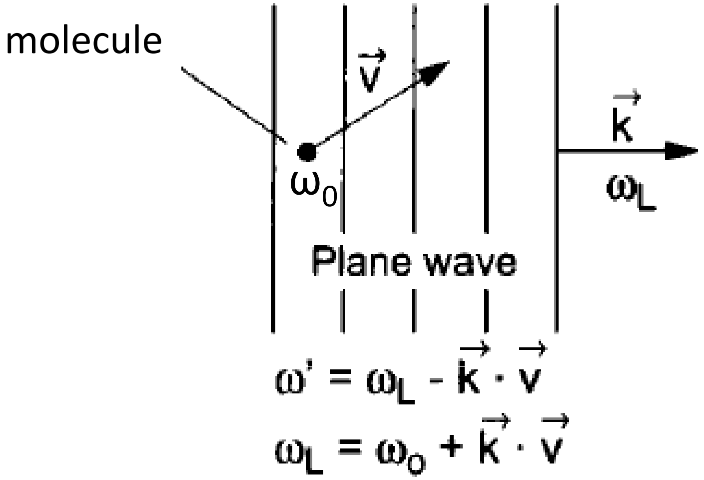
^4I term: L =6 $S = \frac{3}{2}$ $J = \frac{15}{2}, \frac{13}{2}, \frac{11}{2}, \frac{9}{2}$

^4F term: L =3 $S = \frac{3}{2}$ J = 9/2, 7/2, 5/2, 3/2

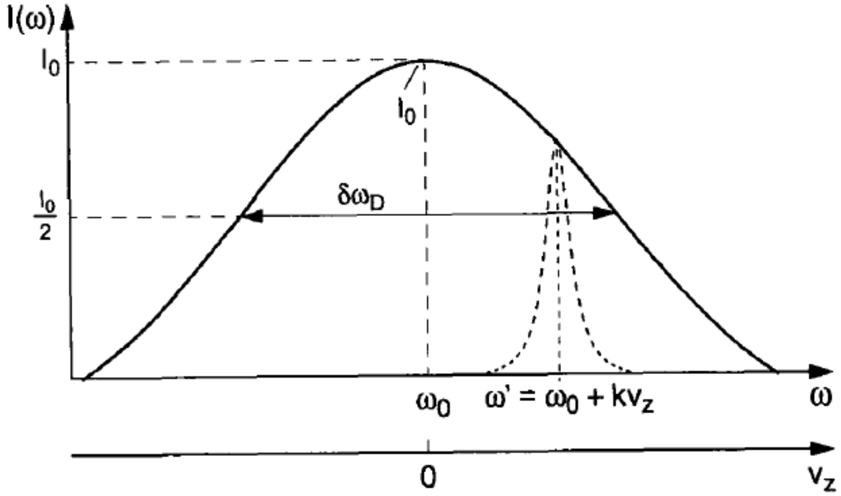
Laser action: $^4F_{3/2} - {}^4I_{11/2}$



Doppler-Free Multiphoton Spectroscopy



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



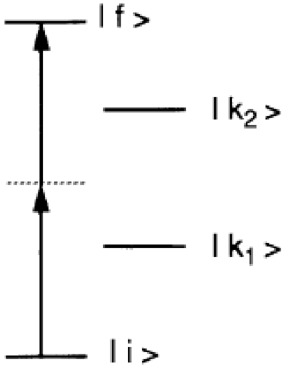
If the light wave propagates in the z direction:

$$(\boldsymbol{k} = \{0,0,k_z\})$$
$$\omega_\mathrm{L} = \omega_0 + k_z v_z = \omega_0\left(1 + \frac{v_z}{c}\right)$$

Intensity profile of the Doppler-broadened spectral line:

$$I(\omega) = I(\omega_0)\exp\left[-\left(c\frac{(\omega-\omega_0)}{(\omega_0 v^*)}\right)^2\right]$$
$$v^* = (2k_\mathrm{B}T/m)^{1/2}$$
$$\delta\omega_\mathrm{D} = |\omega_1 - \omega_2|$$
$$I(\omega_1) = I(\omega_2) = I(\omega_0)/2.$$

Doppler-Free two photon Spectroscopy



$$\omega' = \omega - \boldsymbol{k} \cdot \boldsymbol{v}$$

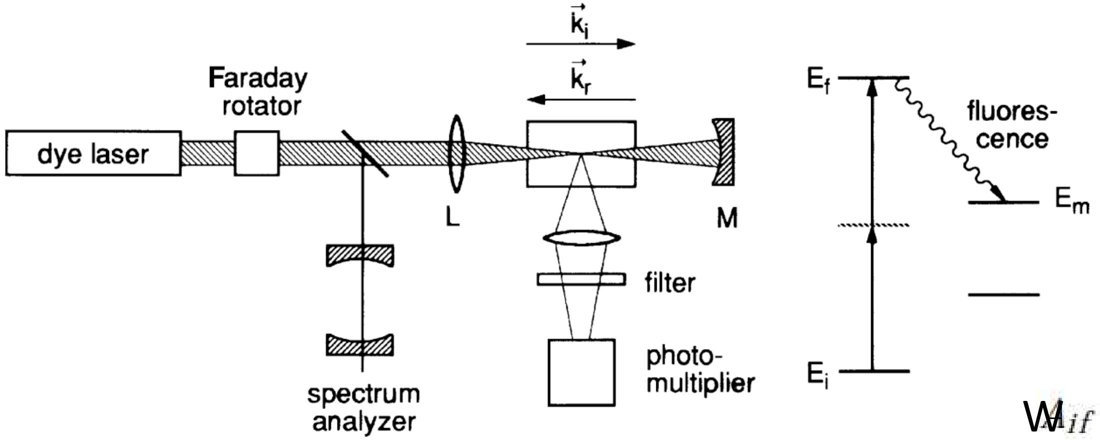
The resonance condition for the simultaneous absorption of two photons:

$$(E_f - E_i)/\hbar = (\omega_1' + \omega_2') = \omega_1 + \omega_2 - \boldsymbol{v} \cdot (\boldsymbol{k}_1 + \boldsymbol{k}_2)$$

ω1 =ω2 =ω:

k1 =−k2

All molecules absorb at the same sum frequency ω1+ω2 = 2ω.



The probability *Wif for a two photon* transition:

$$W_{if} \propto \frac{\gamma_{if} I_1 I_2}{[\omega_{if} - \omega_1 - \omega_2 - \boldsymbol{v} \cdot (\boldsymbol{k}_1 + \boldsymbol{k}_2)]^2 + (\gamma_{if}/2)^2} \times \left| \sum_k \frac{D_{ik} \cdot \hat{e}_1 \cdot D_{kf} \cdot \hat{e}_2}{\omega_{ki} - \omega_1 - \boldsymbol{v} \cdot \boldsymbol{k}_1} + \frac{D_{ik} \cdot \hat{e}_2 \cdot D_{kf} \cdot \hat{e}_1}{\omega_{ki} - \omega_2 - \boldsymbol{v} \cdot \boldsymbol{k}_2} \right|^2$$

Doppler-Free Two photon Spectroscopy

Absorption profile α:

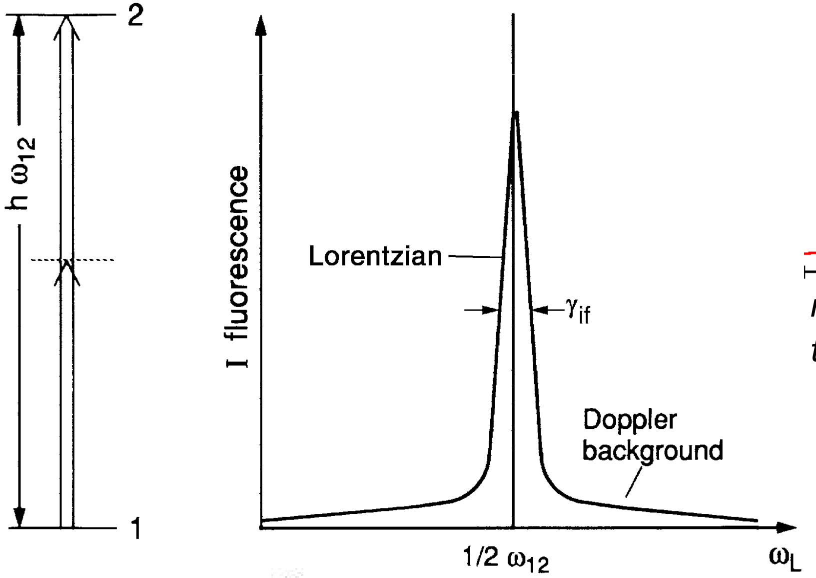
$$\alpha(\omega) \propto \Delta N^0 I^2 \left|\sum_m \frac{(D_{im}\hat{e})(D_{mf}\hat{e})}{\omega-\omega_{im}}\right|^2$$
$$\times \left\{\exp\left[-\left(\frac{\omega_{if}-2\omega}{2kv_\mathrm{p}}\right)^2\right] + \frac{kv_\mathrm{p}}{\sqrt{\pi}}\frac{\gamma_{if}/2}{(\omega_{if}-2\omega)^2+(\gamma_{if}/2)^2}\right\}$$

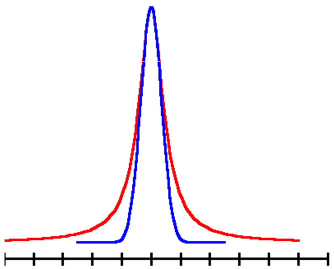
$$v_\mathrm{p} = (2kT/m)^{1/2}$$

ΔN^0 population difference

Doppler-broadened background

narrow Lorentzian profile





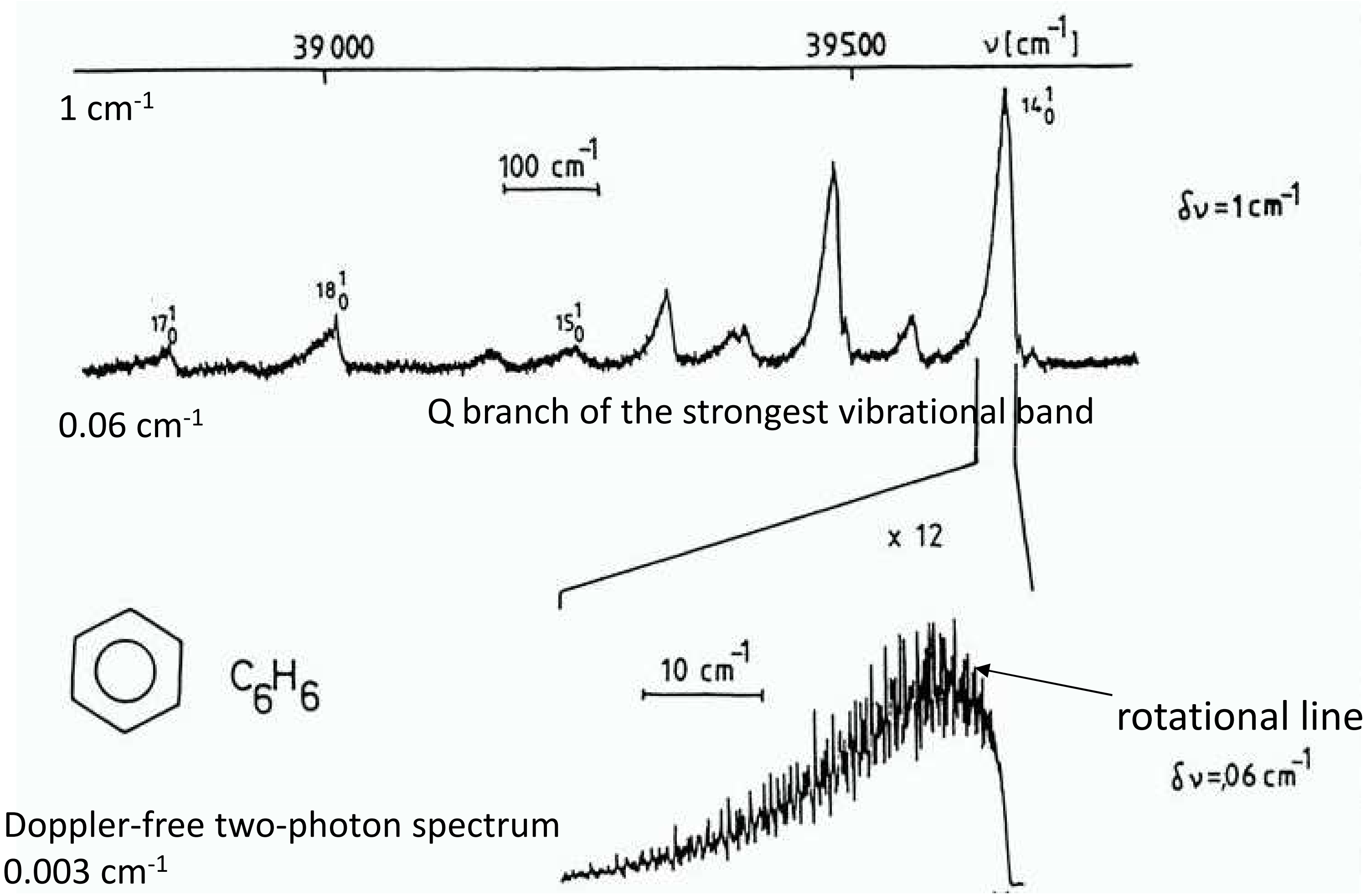
normalized
to equal maximum

Resonance case: $2\omega = \omega if$

second term: $2kv_\mathrm{p}/(\gamma_{if}\sqrt{\pi}) \gg 1$

$$\alpha(\omega) \propto I^2 \frac{\Delta N^0 kv_\mathrm{p}}{\sqrt{\pi}\gamma_{if}} \left|\sum_m \frac{(D_{im}\cdot\hat{e})\cdot(D_{mf}\cdot\hat{e})}{\omega-\omega_{im}}\right|^2$$

Applications: rotational structure of UV spectrum of benzene



1 cm^{-1}

0.06 cm^{-1}

Q branch of the strongest vibrational band

rotational lines

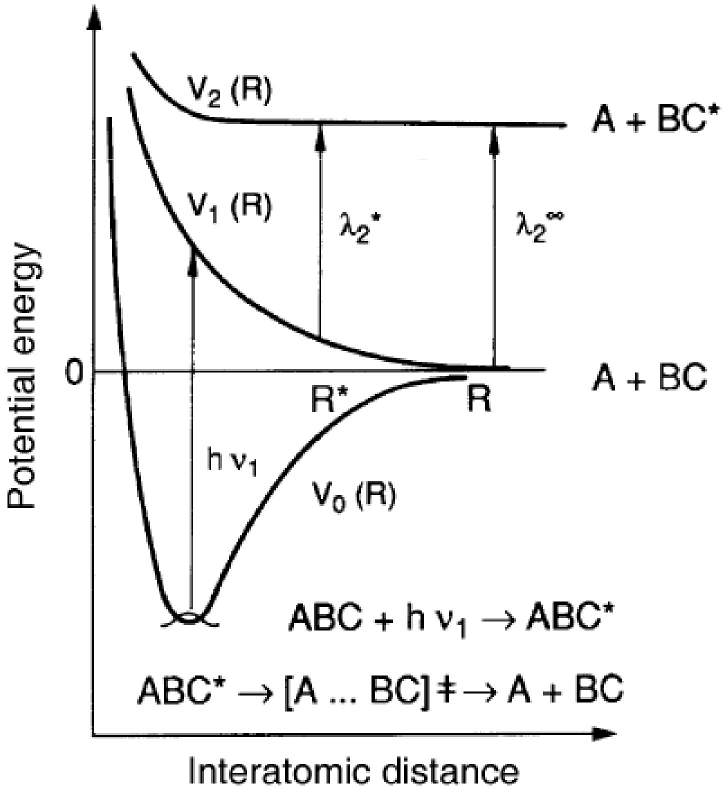
Doppler-free two-photon spectrum
0.003 cm^{-1}

Ultrashort Pulse Spectroscopy

Short pulses 10 fs:
- studies of molecular vibrations (ps)
- ultrafast chemical rections (10^{-11}-10^{-13} s)

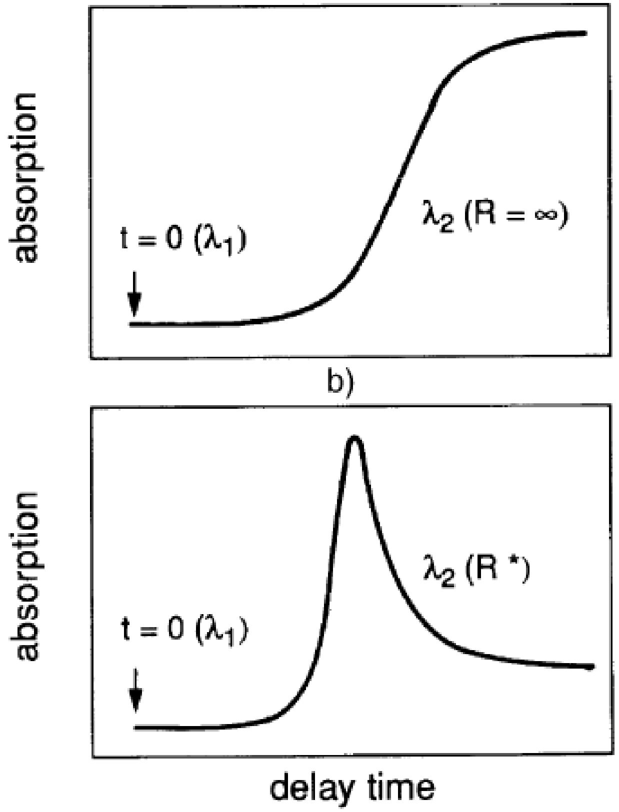
Photodissociation : $ABC + h\nu \rightarrow [ABC]^* \rightarrow A + BC\,,$

Pump (λ1)-probe (λ2) technique



a)

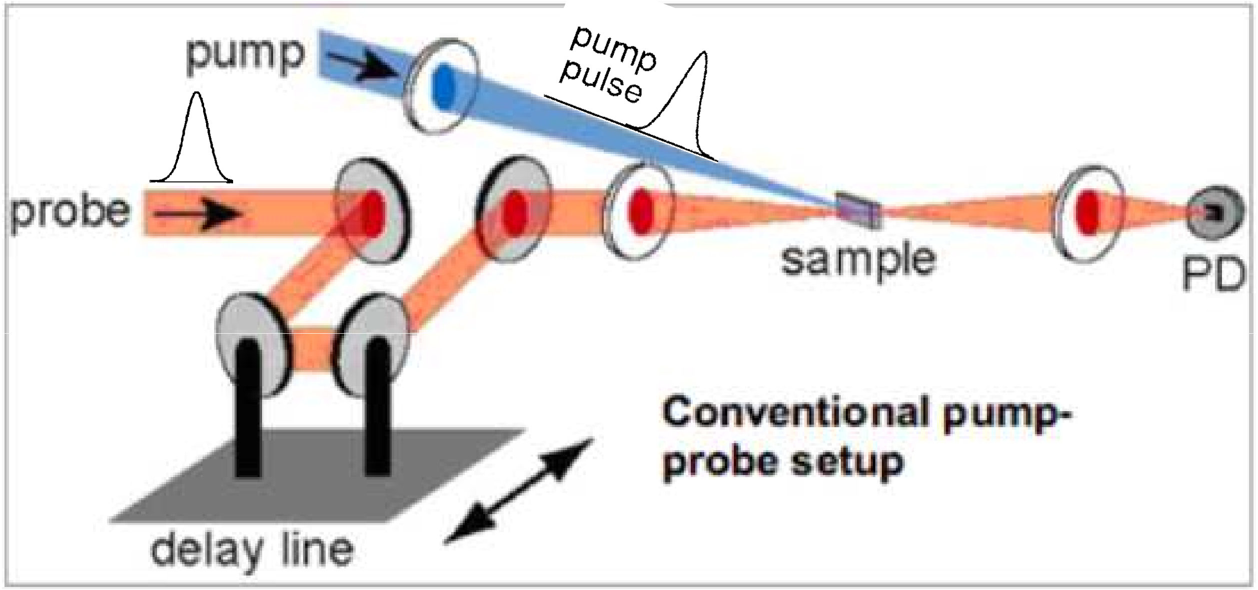
probe pulse



b)

c)

Pump-Probe Setup



$\Delta d = 3\ mm$ $\Delta t = \Delta d/c \approx 10\ ps$

Ultrashort Pulse Spectroscopy: ionization of the Na_2