

Origins of n_2

- electronic: fastest ($\leq fs$)
- molec. rotation $\sim ps$
- thermal
- semiconductor
- relativistic

Electronic

classical: $U(\omega) = \frac{1}{2} m \omega_b^2 |r|^2 - \frac{1}{4} m b |r|^4$

$$\rightarrow \chi_{ijkl}^{(3)} (\omega = \omega + \omega - \omega) = \frac{N b e^4}{3 m^3} \frac{(\delta_{ij} \delta_{kl} + \delta_{ik} \delta_{jl} + \delta_{il} \delta_{jk})}{D(\omega)^3 D(-\omega)}$$

recall 2 pairs must be the same

$$\therefore \left. \begin{aligned} A &= 6 \chi_{1122} \\ B &= 6 \chi_{1221} \end{aligned} \right\} A = B$$

for electronic NL $n_2 \sim 3 \times 10^{-16} \text{ cm}^2/\text{W}$ in Al_2O_3
 $n_2 \sim 1.8$

for $B=1$ $L=5 \text{ mm}$ $\lambda=0.8 \mu\text{m}$

$$I = \frac{1 \cdot \lambda}{2\pi n_2 L} = 8.4 \times 10^{10} \text{ W/cm}^2$$

if $w=1 \text{ mm}$ $P=2.6 \text{ GW}$

$\tau=100 \text{ ps}$ $E=260 \text{ mJ}$

quantum

sum over all atomic states.

$$\chi_{kjih}^{(3)}(\omega_\sigma, \omega_r, \omega_q, \omega_p) = \frac{N}{\hbar^3} \rho_F \sum_{lmn} \frac{\mu_{kl}^k \mu_{lm}^j \mu_{mn}^i \mu_{nj}^h}{(\omega_{mj} - \omega_\sigma)(\omega_{mj} - \omega_q - \omega_p)(\omega_{lj} - \omega_p)}$$

decode k_i - cont. index of output
 i, j, h - " " " inputs

ex. $M_{nm}^{j=2} = -e \langle n | \hat{y} | m \rangle$
 where $|m\rangle$ describes quantum state
 e.g. $|m\rangle \Rightarrow |n, l, m_l\rangle$ in atom.

if selection rule forces $|n\rangle$ to be one state

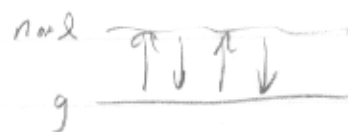
$M_{gn}^k \rightarrow \langle g | M^k | n \rangle$
 then M_{nm}^j has same $\langle n |$

For $\omega_r = \omega$ $\omega_g = \omega$ $\omega_p = -\omega$
 $\omega_{mg} = 0$ sometimes.
 $\rightarrow 0 - \omega - (-\omega)$ in denom.

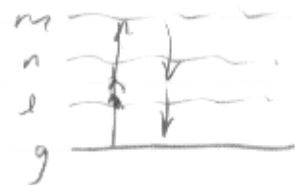
but permutations show this term is not there:
 see Math notebook for permutations.

terms that remain:

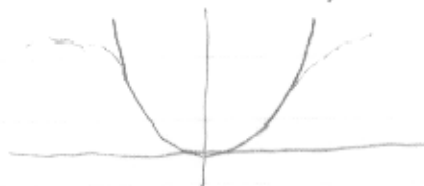
either $|m\rangle = |g\rangle$



or $|m\rangle =$ (second) excited state



$n_2 > 0$ almost always.



large ampl. \rightarrow weaker binding
 \rightarrow lower resonance freq
 \rightarrow higher index.

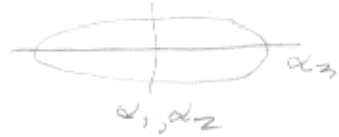
Molecular rotation

- CS_2 , nitrobenzene.

$$p = \alpha E \quad \alpha = \text{polarizability} \quad \chi = N\alpha$$

α is large w/ weak electron binding

\therefore w/ long molecule



$$\alpha_3 \gg \alpha_1, \alpha_2$$

Alignment:

$$\text{torque } \vec{\tau} = \vec{p} \times \vec{E}$$

energy in field

$$dU = -\vec{p} \cdot d\vec{E} = -p_3 dE_3 - p_1 dE_1$$

w/ permanent dipole p 's are constant.

induced dipole

$$dU = -\alpha_3 E_3 dE_3 - \alpha_1 E_1 dE_1$$

$$\rightarrow U = -\frac{1}{2}(\alpha_3 E_3^2 + \alpha_1 E_1^2)$$

$$\text{for } E_3 = E \cos \theta \quad E_1 = E \sin \theta$$

$$U = -\frac{E^2}{2}(\alpha_3 \cos^2 \theta + \alpha_1 \sin^2 \theta)$$

$$= -\frac{1}{2} \alpha_1 E^2 - \frac{1}{2} (\alpha_3 - \alpha_1) E^2 \cos^2 \theta$$

Refractive index

low density (no local field corr.)

$$n^2 = 1 + 4\pi\chi = 1 + 4\pi N \langle \alpha \rangle$$

we must do an avg. over all orientations using a thermal distribution

Thermal averaging:

Boltzmann distrib: $e^{-U/kT}$

$$\langle f(\theta) \rangle = \frac{\int f(\theta) e^{-U(\theta)/kT} d\Omega}{\int e^{-U(\theta)/kT} d\Omega}$$

For induced dipole

$$U(\theta) = - (J \cos^2 \theta) kT = \frac{1}{2} (\alpha_3 - \alpha_1) \bar{E}^2 / kT$$

use time avg \bar{E}^2 : molecule can't respond at 2ω

$$f(\theta) = \alpha(\theta) = \alpha_3 \cos^2 \theta + \alpha_1 \sin^2 \theta \\ = \alpha_1 + (\alpha_3 - \alpha_1) \cos^2 \theta$$

$$n^2 = 1 + 4\pi N [(\alpha_3 - \alpha_1) \langle \cos^2 \theta \rangle + \alpha_1]$$

$$w/ J \rightarrow 0 \quad \langle \cos^2 \theta \rangle = 1/3$$

i. find value of n_0^2 for small J (small I)

$$\text{get } n = n_0 + n_2 I \text{ by approx. small } n_2 I$$

For permanent dipole:

$$U(\theta) = - p_0 E \cos \theta$$

$$P = \frac{N p_0 \langle \cos \theta \rangle}{E}$$

$$n^2 = 1 + 4\pi N \frac{p_0 \langle \cos \theta \rangle}{E}$$

in this case the refr. index decreases w/ field as alignment saturates it.

All this assumes rotational response is fast enough to respond to changes in E or $|E|$.

- if not, consider it as a delayed Debye response.

Thermal NL effects.

$$n = n_0 + \left(\frac{dn}{dT}\right) T_L \quad T_L = \Delta T \text{ from laser.}$$

$\frac{dn}{dT}$ can be + (typ) or -

Here we're working with a coupled eqn: \rightarrow abs. coefft

$$(\rho_0 C) \frac{\partial T_L}{\partial t} - K \nabla^2 T_L = Q = \alpha I(r)$$

spec. heat $\frac{\partial T}{\partial t}$ \hookrightarrow therm. conductivity \hookrightarrow heat source

time response: scale eqn.

$$\rho_0 C \frac{T_L}{\tau} \approx \frac{K T_L}{R^2} \quad \rightarrow \tau \sim \frac{\rho_0 C R^2}{K}$$

typ. 1 sec. for macroscopic beams \sim 1 mm
100 μ s for small beams 10 μ m.

Notes: $\frac{dn}{dT}$, C , K vary with temperature in most materials.

\rightarrow cryo cooling sapphire

decrease ΔT and increase time response

$>$ $n(r)$ again leads to thermally induced lens.

- imp. in amplifiers.

For CW beams, assume steady state.

$$-K \nabla^2 T_L = \alpha I(r) \quad I(r)$$

$$\frac{K T_L}{R^2} \approx \alpha I_0 \quad T_L \sim \frac{\alpha I_0 R^2}{K}$$

$$\Delta n \sim T_L \frac{dn}{dT} \rightarrow \text{effective } n_2$$

• very strong NL effects for CW

• must solve coupled eqns in general

- can use limiting forms of eqn for fast or slow t .

ionization changes to refr. index.

plasma refr. index.

$$n = \left[1 - \frac{\omega_p^2}{\omega^2} \right]^{1/2}$$

$$\omega_p^2 = \frac{4\pi N_e e^2}{m_e}$$

$$n = \left[1 - \frac{4\pi N_e e^2}{m_e \omega^2} \right]^{1/2} = \left(1 - \frac{N_e}{N_{cr}} \right)^{1/2} \quad N_{cr} = \frac{m_e \omega^2}{4\pi e^2}$$

so $n(N_e)$ for fixed ω

critical density.

small $N_e \rightarrow n \approx 1 - \frac{1}{2} N_e / N_{cr}$.

Result n w/ bound electrons:

$$n \approx (1 + 4\pi\chi)^{1/2} \approx 1 + 2\pi N_a \alpha \quad \text{bound only.}$$

w/ both:

$$n \approx 1 + 2\pi N_a \alpha - \frac{1}{2} N_e / N_{cr}$$

Ionization

$$\frac{dN_e}{dt} = W \quad \text{ioniz. rate}$$

$W(I)$ given by mechanism

tunnelling \rightarrow ADK rates

multiphoton

avalanche/collisional

But note: on short timescale, ionization is irreversible.

- recombination is slow (direct recomb, 3-body).

Also, conservation of species.

$$n \approx 1 + 2\pi N_a \alpha \cdot (1 - \eta_e) = \frac{1}{2} \frac{N_a}{N_{cr}} \cdot N_e$$

can also add NL response of bound electrons: $+ (1 - \eta_e) n_2 I$

Relativistic NLO

recall ponderomotive energy:

$$U_p = \frac{1}{4} \frac{e^2 E^2}{m_e \omega^2} = \text{time average KE of electron in field } E = \text{peak field.}$$

when

$$U_p \approx m_e c^2 \rightarrow \text{relativistic effects.}$$

in terms of field:

$$E_c^2 = \frac{4 m_e^2 c^2 \omega^2}{e^2}$$

or Intensity:

$$U_p \sim 9 \times 10^{14} I \lambda^2 \text{ eV}$$

$$I_c = \frac{500 \text{ keV} \cdot 10^3}{9 \times 10^{-14} \lambda^2} \sim \frac{5 \times 10^{18}}{\lambda^2}$$

index changes due to rel. mass shift:

$$n^2 = 1 - \frac{\omega_p^2}{\omega^2}$$

$$\gamma^2 = 1 + \frac{e^2 E_0^2}{m \omega^2 c^2} = 1 + \frac{4 U_p}{m c^2}$$

$$\rightarrow n_2 = \frac{2\pi \omega_p^2 e^2}{n_0^2 m^2 c^3 \omega^4}$$

factoring in proportionality const in $U_p(I)$

$$\approx \frac{1}{2\pi^2 n_0^2} \left(\frac{\omega_p}{\omega} \right)^2 \left[\frac{\lambda^2}{m c^2 / (e_0 / c)} \right]$$

$$e_0 = \text{classical el radius} = \frac{e^2}{m c^2}$$



large ampl. \rightarrow weaker binding
 \rightarrow lower resonance freq
 \rightarrow higher index.