Advanced Vitreous State – The Physical Properties of Glass

Active Optical Properties of Glass

Lecture 20: Nonlinear Optics in Glass-Fundamentals

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Active Optical Properties of Glass

1. Light emission ➡ Optical amplification and lasing
   (fluorescence, luminescence)

   Optical transitions, spontaneous emission, lifetime, line broadening, stimulated emission, population inversion, gain, amplification and lasing, laser materials, role of glass

2. Nonlinear Optical Properties

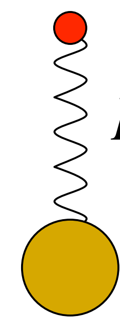
   Fundamentals: nonlinear polarization, 2nd-order nonlinearities, 3rd-order nonlinearities
   Applications: thermal poling, nonlinear index, pulse broadening, stimulated Raman effect, multiphoton ionization

Linear optics-classical electron oscillator

Equation of motion for bound electron:

$$m_e \frac{d^2 x}{dt^2} = \underbrace{-m_e \gamma \frac{dx}{dt}}_{\text{damping force}} \underbrace{- Kx}_{\text{binding force}} \underbrace{- \mathrm{e} E_0 e^{-i\omega t}}_{\text{driving force}}$$

electron



ion core

$$K = m_e \omega_0^2$$

resonance frequency

Solution: electron oscillates at driving frequency

$$x(t) = x_0 e^{-i\omega t} = \frac{-\mathrm{e}E_0}{m} \frac{1}{\omega_0^{\,2} - \omega^2 - i\omega\gamma} e^{-i\omega t}$$

Oscillating dipole: $p = -\mathrm{e}x(t)$

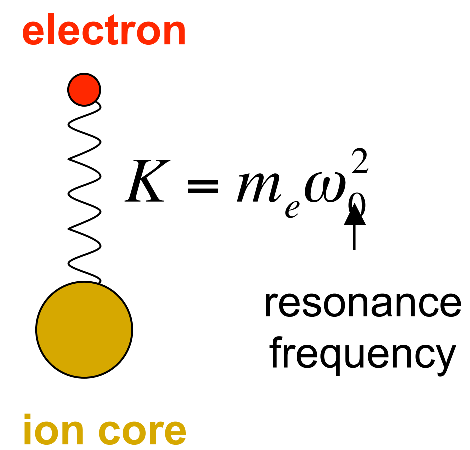
Polarization: $P(t) = \Sigma p_i = -N\mathrm{e}x(t) = \dfrac{\mathrm{Ne}^2 E_0}{m} \dfrac{1}{\omega_0^{\,2} - \omega^2 - i\omega\gamma} e^{-i\omega t}$

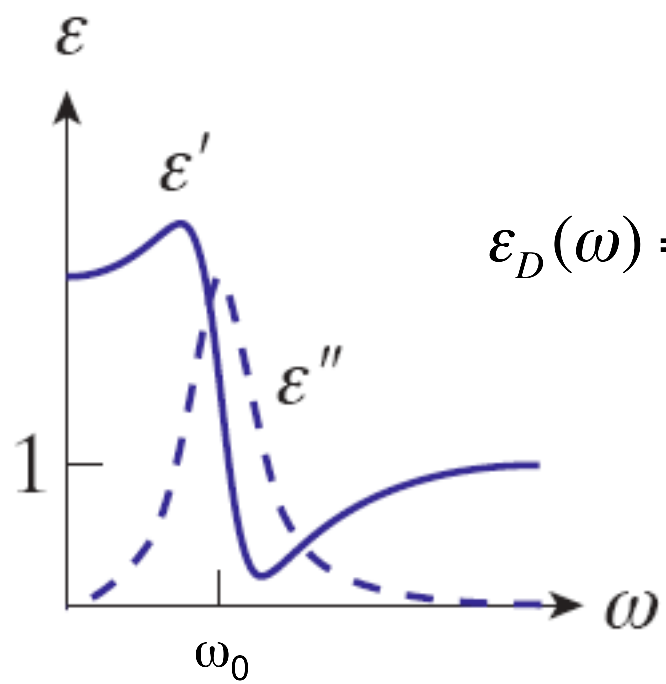
Frequency dependent dielectric constant

$$P(t) = \varepsilon_0 \chi E(t)$$

linear susceptibility

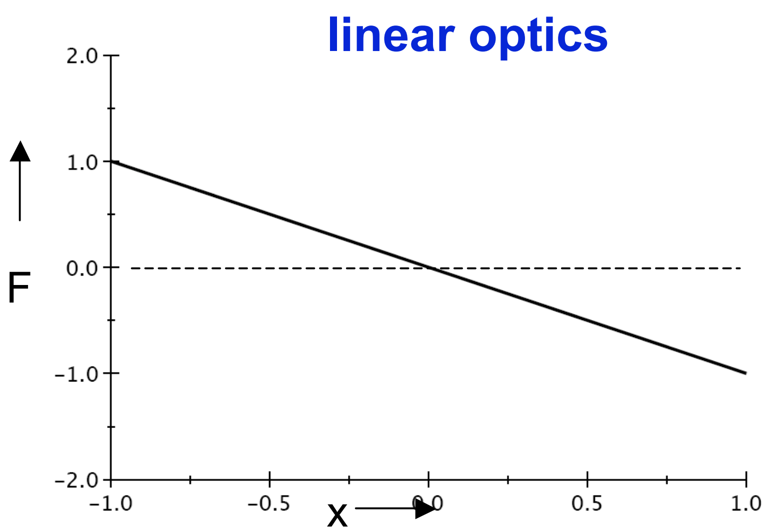
$$\chi(\omega) = \frac{P(t)}{\varepsilon_o E(t)} = \frac{Ne^2}{\varepsilon_o m} \frac{1}{\omega_0^2 - \omega^2 - i\omega\gamma}$$

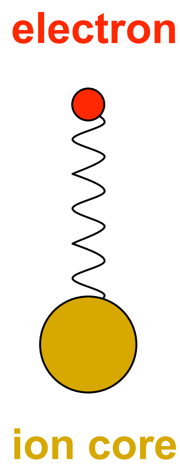


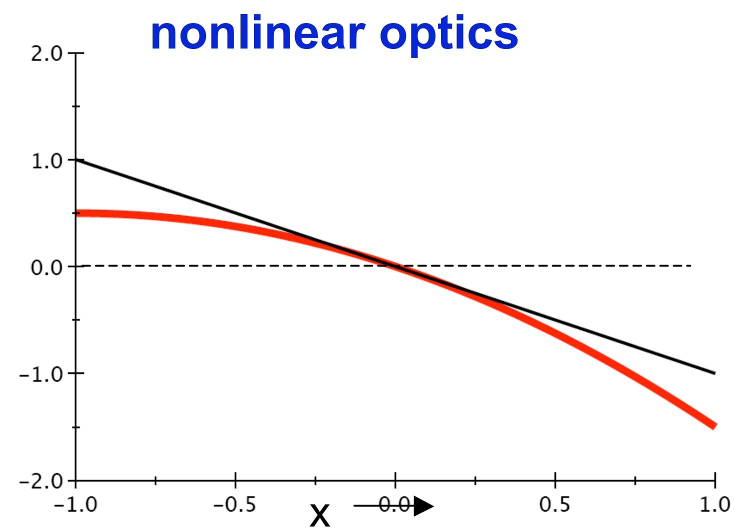


$$\varepsilon_D(\omega) = 1 + \chi(\omega) = \frac{P(t)}{\varepsilon_o E(t)} = \frac{Ne^2}{\varepsilon_o m} \frac{1}{\omega_0^2 - \omega^2 - i\omega\gamma}$$

Nonlinear optics-anharmonic binding force







binding force $$F = -Kx$$

$$F = -Kx - \boxed{ax^2}$$

(+higher order terms)

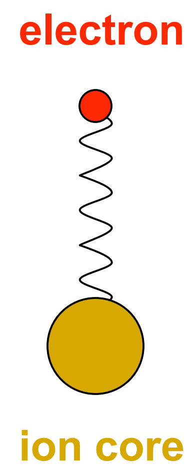
Equation of motion for bound electron:

$$m_e \frac{d^2x}{dt^2} = -m_e \gamma \frac{dx}{dt} - \underbrace{Kx - ax^2 + (.....)}_{\text{binding force}} - eE_0 e^{-i\omega t}$$

damping force   binding force   driving force

Nonlinear optics-classical electron oscillator

Equation of motion for bound electron:



$$m_e \frac{d^2 x}{dt^2} = -m_e \gamma \frac{dx}{dt} - \underbrace{Kx + ax^2} - \mathrm{e}\left[E_0 e^{-i\omega t} + cc\right]$$

damping force   binding force   driving force

This equation has no general solution, we will solve it by using a perturbation expansion. We replace E(t) by λE(t) and seek a solution in the form:

$$x = \lambda x^{(1)} + \lambda^2 x^{(2)} + \lambda^3 x^{(3)} + \ldots$$

The parameter λ is a parameter that characterizes the strength of the perturbation.
It ranges continuously between 0 and 1 and we will set it to 1 at the end of the calculation.

Substitute expression for x in above equation of motion and group terms with equal powers of λ

Nonlinear optics-classical electron oscillator

$$\lambda: \quad \ddot{x}^{(1)} + 2\gamma\dot{x}^{(1)} + \omega_0^2 x^{(1)} = -eE(t)/m$$

$$\lambda^2: \quad \ddot{x}^{(2)} + 2\gamma\dot{x}^{(2)} + \omega_0^2 x^{(2)} + a\left[x^{(1)}\right]^2 = 0$$

$$\lambda^3: \quad \ddot{x}^{(3)} + 2\gamma\dot{x}^{(3)} + \omega_0^2 x^{(3)} + 2ax^{(1)}x^{(2)} = 0$$

$$x^{(1)}(t) \propto E(t) \qquad x^{(1)}(t) = \frac{-\mathrm{e}E_0}{m}\frac{1}{{\omega_0}^2 - \omega^2 - i\omega\gamma}e^{-i\omega t}$$

$$x^{(2)}(t) \propto \left[E(t)\right]^2 \qquad x^{(2)}(t) = \frac{-ae^2E_0^2}{m^2}\left[\frac{1}{D(2\omega)D^2(\omega)}e^{-i2\omega t} + \frac{1}{D(0)D(\omega)D(-\omega)}\right]$$

$$D(\omega_j) = (\omega_0^2 - {\omega_j}^2) - i\omega_j\gamma$$

oscillates at 2ω **“oscillates” at 0**

$\chi^{(2)}$ resonances

$$x^{(2)}(t) \propto \left[E(t)\right]^2 \qquad x^{(2)}(t) = \frac{-ae^2E_0^2}{m^2}\left[\frac{1}{D(2\omega)D^2(\omega)}e^{-i2\omega t} + \frac{1}{D(0)D(\omega)D(-\omega)}\right]$$

oscillates at 2ω **"oscillates" at 0**

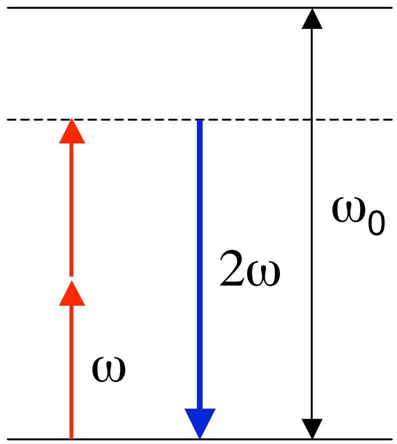
2nd-order nonlinear polarization

$$P^{(2)}(t) = -Nex^{(2)}(t)$$

2nd-order susceptibility (for SHG)

$$\chi^{(2)}(2\omega) = \frac{P^{(2)}(2\omega)}{E(\omega)^2} = \frac{aNe^3E_0^2}{m^2}\frac{1}{D(2\omega)D^2(\omega)}$$

$$D(\omega_j) = (\omega_0^2 - \omega_j{}^2) - i\omega_j\gamma$$



2nd-order susceptibility is enhanced when either ω or 2ω are equal to ω_0

$\chi^{(2)}$ frequency terms

When the input field has 2 frequency components:

$$E(t) = E_1 e^{-i\omega_1 t} + E_2 e^{-i\omega_2 t} + C.C$$

the 2nd-order polarization has many frequency components (mixing terms):

$$P^{(2)}(t) = \chi^{(2)}[E_1^2 e^{-i2\omega_1 t} + E_2^2 e^{-i2\omega_2 t} + 2E_1E_2 e^{-i(\omega_1+\omega_2)t} + 2E_1E_2^* e^{-i(\omega_1-\omega_2)t} + c.c]$$

$$+ 2\chi^{(2)}[E_1E_1^* + E_2E_2^*]$$

$$= \sum_n P(\omega_n) e^{-i\omega_n t}.$$

$$P(2\omega_1) = \chi^{(2)} E_1^2 \quad (SHG),$$

$$P(2\omega_2) = \chi^{(2)} E_2^2 \quad (SHG),$$

Second harmonic generation

$$P(0) = 2\chi^{(2)}(E_1E_1^* + E_2E_2^*) \quad (OR),$$

Optical rectification

$$P(\omega_1+\omega_2) = 2\chi^{(2)} E_1 E_2 \quad (SFG),$$

Sum-frequency generation

$$P(\omega_1-\omega_2) = 2\chi^{(2)} E_1 E_2^* \quad (DFG).$$

Difference-frequency generation

Nonlinear optics: general formalism

$$\vec{P}(t) = \underbrace{\chi^{(1)} \cdot \vec{E}}_{\textbf{linear}}(t) + \underbrace{\chi^{(2)} \cdot \vec{E}(t)^2 + \chi^{(3)} \cdot \vec{E}(t)^3 + \cdots}_{\textbf{nonlinear}}$$

The em field is expressed as sum of frequency components, for example

$$E(t) = E_1 e^{-i\omega_1 t} + E_2 e^{-i\omega_2 t} + c.c.$$

The induced polarization can be written as:

$$P(t) = \sum_n P(\omega_n) e^{-i\omega_n t}$$

The frequency components ω_n are determined by the order of the interaction and the input frequencies

<u>2nd-order interaction:</u>

	input frequencies, E	induced polarization frequencies, P
One	ω_1	$2\omega_1$, 0
Two	ω_1, ω_2	$2\omega_1$, $2\omega_2$, 0, $\omega_1+\omega_2$, $\omega_1-\omega_2$
Three	ω_1, ω_2, ω_3	$2\omega_1$, $2\omega_2$, $2\omega_3$,0, $\omega_1+\omega_2$, $\omega_1+\omega_3$, $\omega_2+\omega_3$, $\omega_1-\omega_2$, $\omega_1-\omega_3$, $\omega_2-\omega_3$

3rd-order polarization and nonlinear index

Applied field, one input frequency $E = E_0 e^{-i\omega t}$

3rd order nonlinear polarization $P^{(3)}(t) = \chi^{(3)} E(t)^3$

has frequency components:

$$P(3\omega) = \chi^{(3)} E_0^3$$

$$P(\omega) = 3\chi^{(3)} E_0^2 E_0^*$$

The total polarization at ω has linear and nonlinear contributions

$$P(\omega) = \chi^{(1)} E_0 + 3\chi^{(3)} E_0^2 E_0^*$$

$$P(\omega) = (\chi^{(1)} + 3\chi^{(3)} E_0 E_0^*) E_0 = \chi^{(eff)} E_0$$

$$\chi^{(eff)} = \chi^{(1)} + 3\chi^{(3)} |E_0|^2$$

$$|E_0|^2 \propto I$$

intensity

$$n = n_0 + n_2 I \qquad n_2 \propto \chi^{(3)}$$

n_2 = nonlinear index coefficient

$\chi^{(3)}$ - frequency terms

With three input fields: $\omega_1, \omega_2, \omega_3$

$P^{(3)}$ has the following frequency components:

$3\omega_1, 3\omega_2, 3\omega_3$

$\omega_1, \omega_2, \omega_3$

$2\omega_1 \pm \omega_2, 2\omega_1 \pm \omega_3, 2\omega_2 \pm \omega_1, 2\omega_2 \pm \omega_3, 2\omega_3 \pm \omega_1, 2\omega_3 \pm \omega_2$

$\omega_1 + \omega_2 + \omega_3, \omega_1 + \omega_2 - \omega_3, \omega_1 + \omega_3 - \omega_2, \omega_2 + \omega_3 - \omega_1$

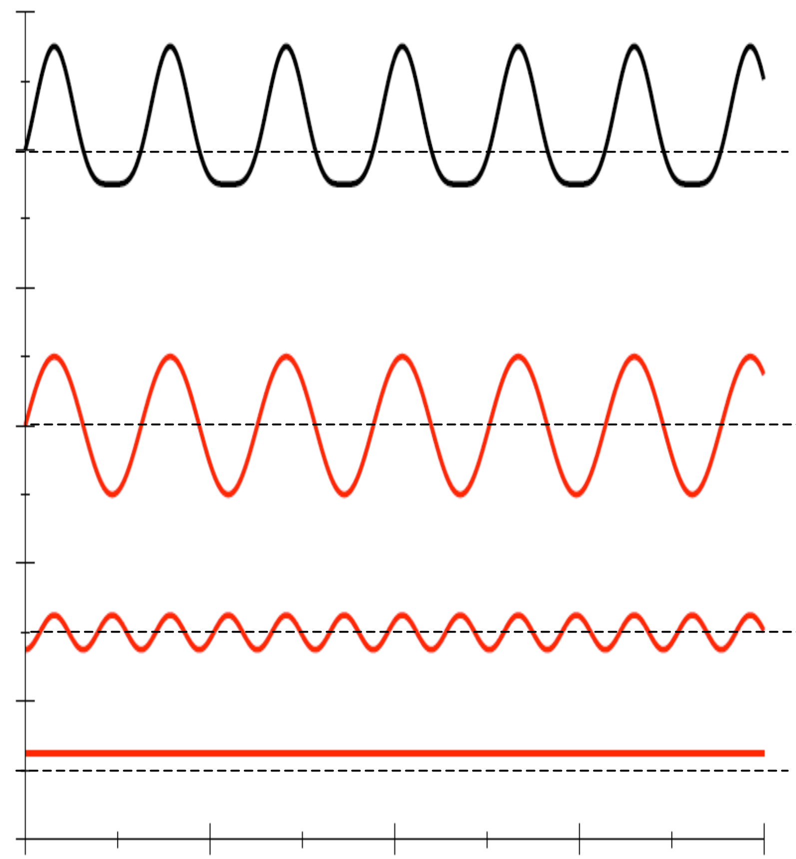
2nd-order polarization

$$P(t) = P^{(1)}(t) + P^{(2)}(t) = \chi^{(1)}E(t) + \chi^{(2)}E(t)^2$$

$$P(t) = \sin(\omega t) + 0.5\sin(\omega t)^2$$

linear polarization $\sin(\omega t)$

nonlinear polarization $\begin{cases} -0.25\cos(2\omega t) \\ 0.25 \end{cases}$



$\chi^{(2)}$ requires lack of inversion symmetry

$$P(t) = P^{(1)}(t) + P^{(2)}(t) = \chi^{(1)}E(t) + \chi^{(2)}E(t)^2$$

$$P^{(2)}(t) = \chi^{(2)}E(t)^2$$

In material with inversion symmetry

$$-P^{(2)}(t) = \chi^{(2)}\left[-E(t)\right]^2$$

$$P^{(2)}(t) = -P^{(2)}(t) \rightarrow \chi^{(2)} = 0$$

Glass is isotropic $\longrightarrow$ $\chi^{(2)} = 0$

$\chi^{(2)}$ tensor

P and E are vectors!
So $\chi^{(n)}$'s are tensors

n = order of χ	number of tensor elements	notation for one element
1	9	$\chi^{(1)}_{ij}$
2	27	$\chi^{(2)}_{ijk}$
3	81	$\chi^{(3)}_{ijkl}$

$$P_x(\omega_n + \omega_m) = \chi^{(2)}_{xxx}(\omega_n + \omega_m, \omega_n, \omega_m) E_x(\omega_n) E_x(\omega_m)$$

$$P_y(\omega_n + \omega_m) = \chi^{(2)}_{yxx}(\omega_n + \omega_m, \omega_n, \omega_m) E_x(\omega_n) E_x(\omega_m)$$

Nonlinear wave equation

Propogation of waves is described by nonlinear wave equation

$$-\nabla^2 \vec{E} + \frac{\varepsilon^{(1)}}{c^2} \frac{\partial^2 \vec{E}^{(1)}}{\partial t^2} = -\frac{4\pi}{c^2} \frac{\partial^2 \vec{P}^{NL}}{\partial t^2}$$

which leads to a set of coupled differential equations (example SHG)

$$\frac{\partial A_2}{\partial z} = \frac{4\pi i d_{eff} \omega_2^2}{k_2 c^2} A_1^2 e^{i\Delta k z} \qquad \Delta k = k_2 - 2k_1$$

$$\frac{\partial A_1}{\partial z} = \frac{8\pi i d_{eff} \omega_1^2}{k_1 c^2} A_2 A_1^* e^{-i\Delta k z}$$

Growth of SH wave depends on propagation length and is optimized when Δk=o (phase matching)

Nonlinear optics in glass

2nd-order nonlinearities

- In normal glasses $\chi^{(2)}=0$

3nd-order nonlinearities

- All materials, including glasses, have a $\chi^{(3)}$
- In glass there are only three independent $\chi^{(3)}$ tensor elements
- $\chi^{(3)}$ processes involve the interaction of 3 input waves to generate a polarization (4th wave) at a mixing frequency
  - -with 3 different input frequencies there are many possible output frequencies
- Strength of generated signal depends on propagation length
  - -optical fibers!
- Phase matching: $\Delta k=k_4-k_3-k_2-k_1=0$