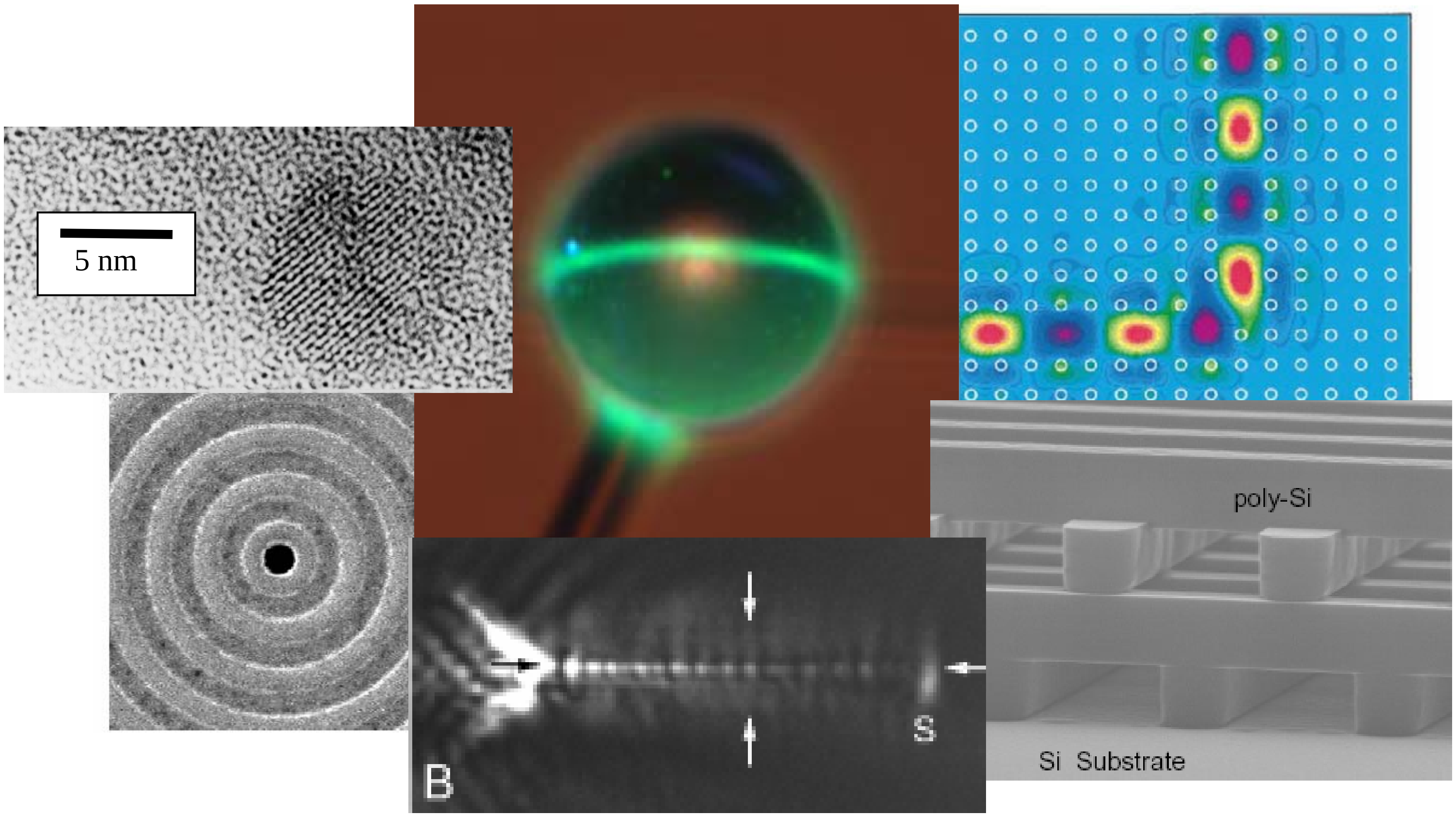
Lecture 2: Dispersion in Materials



Course Info

Course webpage

- Is now up and running
  http://shay.ecn.purdue.edu/~ece695s

Let me know what you think!

- Direct questions
- Topics ?
- Format ?
- **Big** comments on the nanocourse are most welcome!
- Ask questions any time……….

What Happened in the Previous Lecture ?

Maxwell's Equations

Bold face letters are vectors!

$$\nabla \cdot \boldsymbol{D} = \rho_f \qquad \nabla \cdot \boldsymbol{B} = 0 \qquad \nabla \times \boldsymbol{E} = -\frac{\partial \boldsymbol{B}}{\partial \boldsymbol{t}} \qquad \nabla \times \boldsymbol{H} = \frac{\partial \boldsymbol{D}}{\partial \boldsymbol{t}} + \boldsymbol{J}$$

Curl Equations lead to

$$\nabla^2 \boldsymbol{E} = \mu_0 \varepsilon_0 \frac{\partial^2 \boldsymbol{E}}{\partial t^2} + \mu_0 \frac{\partial^2 \boldsymbol{P}}{\partial t^2}$$

(under certain conditions)

Linear, Homogeneous, and Isotropic Media

$$\boldsymbol{P} = \varepsilon_0 \chi \boldsymbol{E}$$

Wave Equation

$$\nabla^2 \boldsymbol{E}(\boldsymbol{r}, t) = \frac{n^2}{c^2} \frac{\partial^2 \boldsymbol{E}(\boldsymbol{r}, t)}{\partial \boldsymbol{t}^2}$$

Solutions: EM waves

$$\boldsymbol{E}(z,t) = \mathrm{Re}\left\{ \boldsymbol{E}(z,\omega) \exp\left(-i\beta z - \frac{\alpha}{2} z + i\omega t \right) \right\}$$

where $\underbrace{\beta = k_0 n'}_{\text{Phase propagation}}$ and $\underbrace{\alpha = -2k_0 n''}_{\text{absorption}}$

In real life: Response of matter ($\boldsymbol{P}$) is not instantaneous

$$\boldsymbol{P}(\boldsymbol{r},t) = \varepsilon_0 \int_{-\infty}^{+\infty} dt'\, x(t-t') \boldsymbol{E}(\boldsymbol{r},t') \Rightarrow \left. \begin{array}{l} \chi' = \chi'(\omega) \\ \chi'' = \chi''(\omega) \end{array} \right\} \Leftrightarrow \left\{ \begin{array}{l} n' = n'(\omega) \\ n'' = n''(\omega) \end{array} \right.$$

Today: Microscopic Origin ω-Response of Matter

Origin frequency dependence of χ in real materials

- Lorentz model (harmonic oscillator model)
- Insulators (Lattice absorption, color centers…)
- Semiconductors (Energy bands, Urbach tail, excitons …)
- Metals (AC conductivity, Plasma oscillations, interband transitions…)

Real and imaginary part of χ are linked

- Kramers-Kronig

But first…..

- When should I work with χ, ε, or n ?

They all seem to describe the optical properties of materials!

n' and n'' vs χ' and χ'' vs ε' and ε''

All pairs (n' and n'', χ' and χ'', ε' and ε'') describe the same physics

For some problems one set is preferable for others another

n' and n'' used when discussing wave propagation

$$\boldsymbol{E}(z,t)=\mathrm{Re}\left\{\boldsymbol{E}(z,\omega)\exp\left(-i\beta z-\frac{\alpha}{2}z+i\omega t\right)\right\}$$ where $\underline{\beta = k_0 n'}$ and $\underline{\alpha = -2k_0 n''}$

Phase propagation absorption

$\left.\begin{array}{l}\chi' \text{ and } \chi'' \\ \varepsilon' \text{ and } \varepsilon''\end{array}\right\}$ used when discussing microscopic origin of optical effects

As we will see today…

Inter relationships

Example: n and ε

From $n=\sqrt{\varepsilon_r}$

$$n'+in''=\sqrt{\varepsilon_r'+i\varepsilon_r''}$$

$$\boxed{\begin{aligned}\varepsilon_r' &= (n')^2-(n'')^2\\ \varepsilon_r'' &= 2n'n''\end{aligned}}$$

and

$$\boxed{\begin{aligned}n' &= \sqrt{\frac{\sqrt{(\varepsilon_r')^2+(\varepsilon_r'')^2}+\varepsilon_r'}{2}}\\ n'' &= \sqrt{\frac{\sqrt{(\varepsilon_r')^2+(\varepsilon_r'')^2}-\varepsilon_r'}{2}}\end{aligned}}$$

Linear Dielectric Response of Matter

Behavior of bound electrons in an electromagnetic field

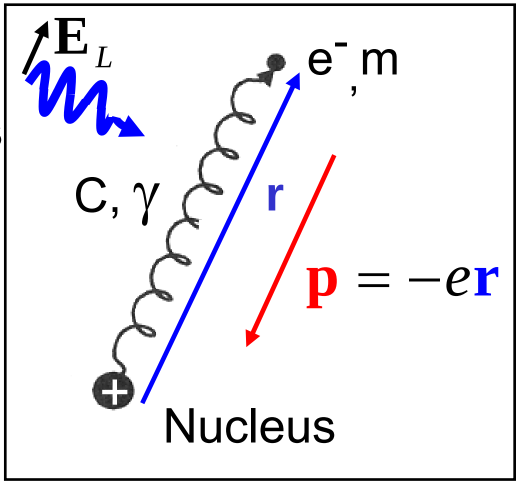
- Optical properties of insulators are determined by bound electrons

Lorentz model

- Charges in a material are treated as harmonic oscillators

$$m\mathbf{a}_{el} = \mathbf{F}_{E,Local} + \mathbf{F}_{Damping} + \mathbf{F}_{Spring} \qquad \text{(one oscillator)}$$

$$m\frac{d^2\mathbf{r}}{dt^2} + m\gamma\frac{d\mathbf{r}}{dt} + C\mathbf{r} = -e\mathbf{E}_L \exp(-i\omega t)$$



- The electric dipole moment of this system is: $\mathbf{p} = -e\mathbf{r}$

$$m\frac{d^2\mathbf{p}}{dt^2} + m\gamma\frac{d\mathbf{p}}{dt} + C\mathbf{p} = e^2\mathbf{E}_L \exp(-i\omega t)$$

- Guess a solution of the form:

$$\mathbf{p} = \mathbf{p}_0 \exp(-i\omega t)\ ; \frac{d\mathbf{p}}{dt} = -i\omega\mathbf{p}_0 \exp(-i\omega t)\ ; \frac{d^2\mathbf{p}}{dt^2} = -\omega^2\mathbf{p}_0 \exp(-i\omega t)$$

$\Rightarrow -m\omega^2\mathbf{p}_0 - im\gamma\omega\mathbf{p}_0 + C\mathbf{p}_0 = e^2\mathbf{E}_L$ $\Rightarrow$ Solve for $\mathbf{p}_0(\mathbf{E}_L)$

Atomic Polarizability

Determination of atomic polarizability

- Last slide: $-m\omega^2\mathbf{p}_0 - im\gamma\omega\mathbf{p}_0 + C\mathbf{p}_0 = e^2\mathbf{E}_L$

$$\Rightarrow \quad -\omega^2\mathbf{p}_0 - i\gamma\omega\mathbf{p}_0 + \frac{C}{m}\mathbf{p}_0 = \frac{e^2}{m}\mathbf{E}_L$$ (Divide by m)

Define as ω_0^2 (turns out to be the resonance ω)

$$\Rightarrow \quad \mathbf{p}_0 = \frac{e^2}{m}\frac{1}{\omega_0^2 - \omega^2 - i\gamma\omega}\mathbf{E}_L$$

Atomic polarizability (in SI units)

- Define atomic polarizability: $\alpha(\omega) \equiv \dfrac{p_0}{\varepsilon_0 \mathrm{E}_L} = \dfrac{e^2}{\varepsilon_0 m}\dfrac{1}{\omega_0^2 - \omega^2 - i\gamma\omega}$

Resonance frequency

Damping term

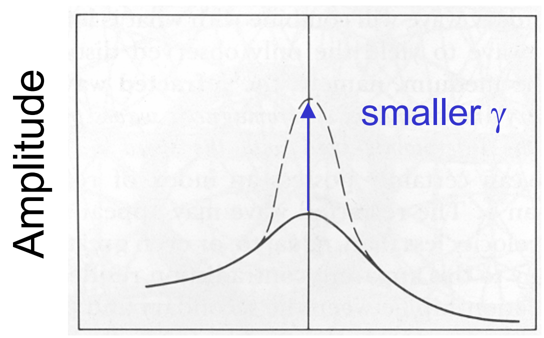
Characteristics of the Atomic Polarizability

Response of matter (***P***) is not instantaneous ⇨ ω-dependent response

- Atomic polarizability: $\alpha(\omega) = \dfrac{p_0}{\varepsilon_0 \mathrm{E}_L} = \dfrac{e^2}{\varepsilon_0 m} \dfrac{1}{\omega_0^2 - \omega^2 - i\gamma\omega} = A\exp\left[i\theta(\omega)\right]$

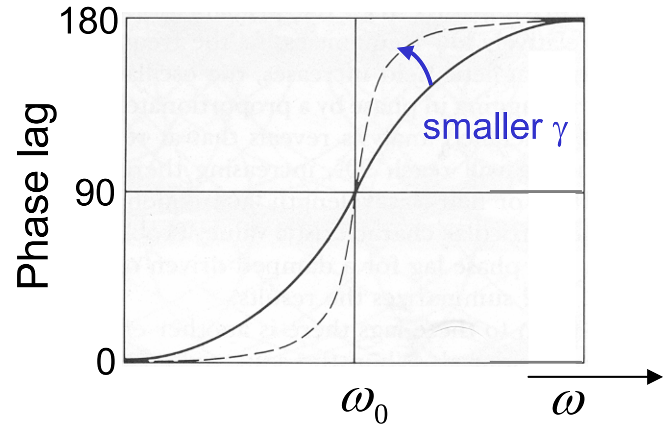
- Amplitude

$$A = \frac{e^2}{\varepsilon_0 m} \frac{1}{\left[\left(\omega_0^2 - \omega^2\right)^2 + \gamma^2\omega^2\right]^{1/2}}$$



- Phase lag of α with ***E:***

$$\theta = \tan^{-1} \frac{\gamma\omega}{\omega_0^2 - \omega^2}$$



Relation Atomic Polarizability (α) and χ: 2 cases

Case 1: Rarified media (.. gasses)

E-field photon

- Dipole moment of one atom, j : $\mathbf{p}_j = \varepsilon_0 \alpha_j(\omega)\mathbf{E}_L$

Density

- Polarization vector: $\mathbf{P} = \underbrace{\frac{1}{V}\sum_j \mathbf{p}_j}_{\text{sum over all atoms}} = \frac{\varepsilon_0}{V}\sum_j \alpha_j \mathbf{E}_L = \varepsilon_0 N \alpha_j \mathbf{E}_L$

Occurs in Maxwell's equation..

$$\alpha_j(\omega) = \frac{e^2}{\varepsilon_0 m}\frac{1}{\omega_0^2 - \omega^2 - i\gamma\omega}$$

$$\Rightarrow \mathbf{P} = \frac{Ne^2}{m}\frac{1}{\omega_0^2 - \omega^2 - i\gamma\omega}\mathbf{E}_L \quad \left(= \varepsilon_0 \chi \mathbf{E}_L\right)$$

$\Rightarrow$ Microscopic origin susceptibility: $\boxed{\chi(\omega) = \frac{Ne^2}{\varepsilon_0 m}\frac{1}{\omega_0^2 - \omega^2 - i\gamma\omega}}$

- Plasma frequency defined as: $\omega_p^2 = \frac{Ne^2}{\varepsilon_0 m}$ $\Rightarrow$ $\chi(\omega) = \frac{\omega_p^2}{\omega_0^2 - \omega^2 - i\gamma\omega}$

Remember: ε and n follow directly from χ

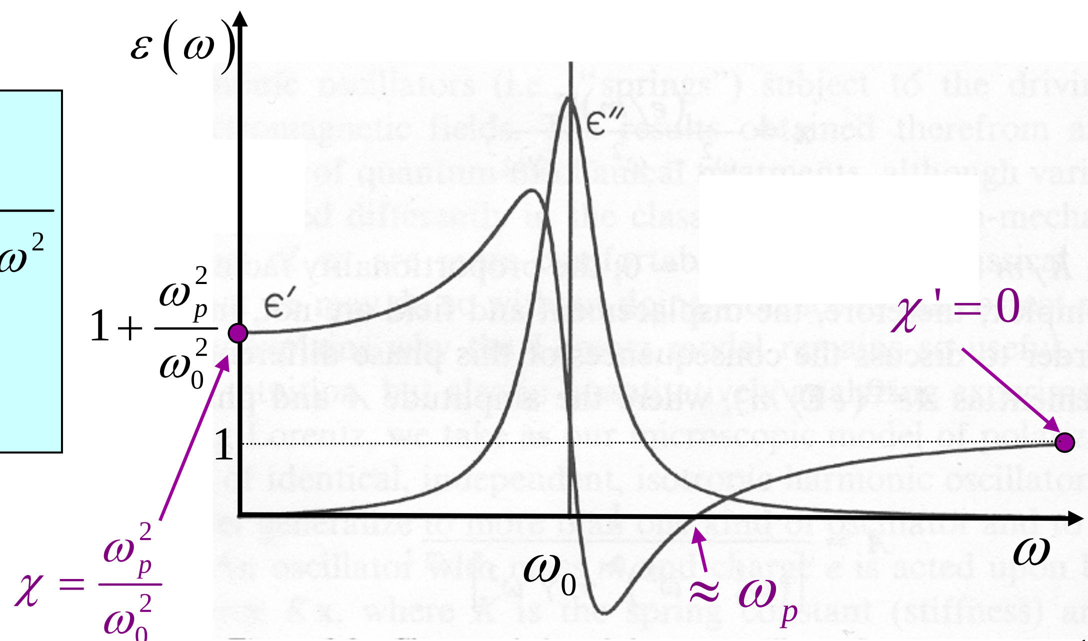
Frequency dependence ε

- Relation of ε to χ : $\varepsilon = 1 + \chi = 1 + \dfrac{\omega_p^2}{\omega_0^2 - \omega^2 - i\gamma\omega}$

$$\Rightarrow \quad \varepsilon' + i\varepsilon'' = 1 + \chi' + i\chi'' = 1 + \frac{\omega_p^2}{\omega_0^2 - \omega^2 - i\gamma\omega}$$

$$\varepsilon' = 1 + \chi'(\omega) = 1 + \frac{\omega_p^2\left(\omega_0^2 - \omega^2\right)}{\left(\omega_0^2 - \omega^2\right)^2 + \gamma^2\omega^2}$$

$$\varepsilon'' = \chi''(\omega) = \frac{\omega_p^2 \gamma\omega}{\left(\omega_0^2 - \omega^2\right)^2 + \gamma^2\omega^2}$$



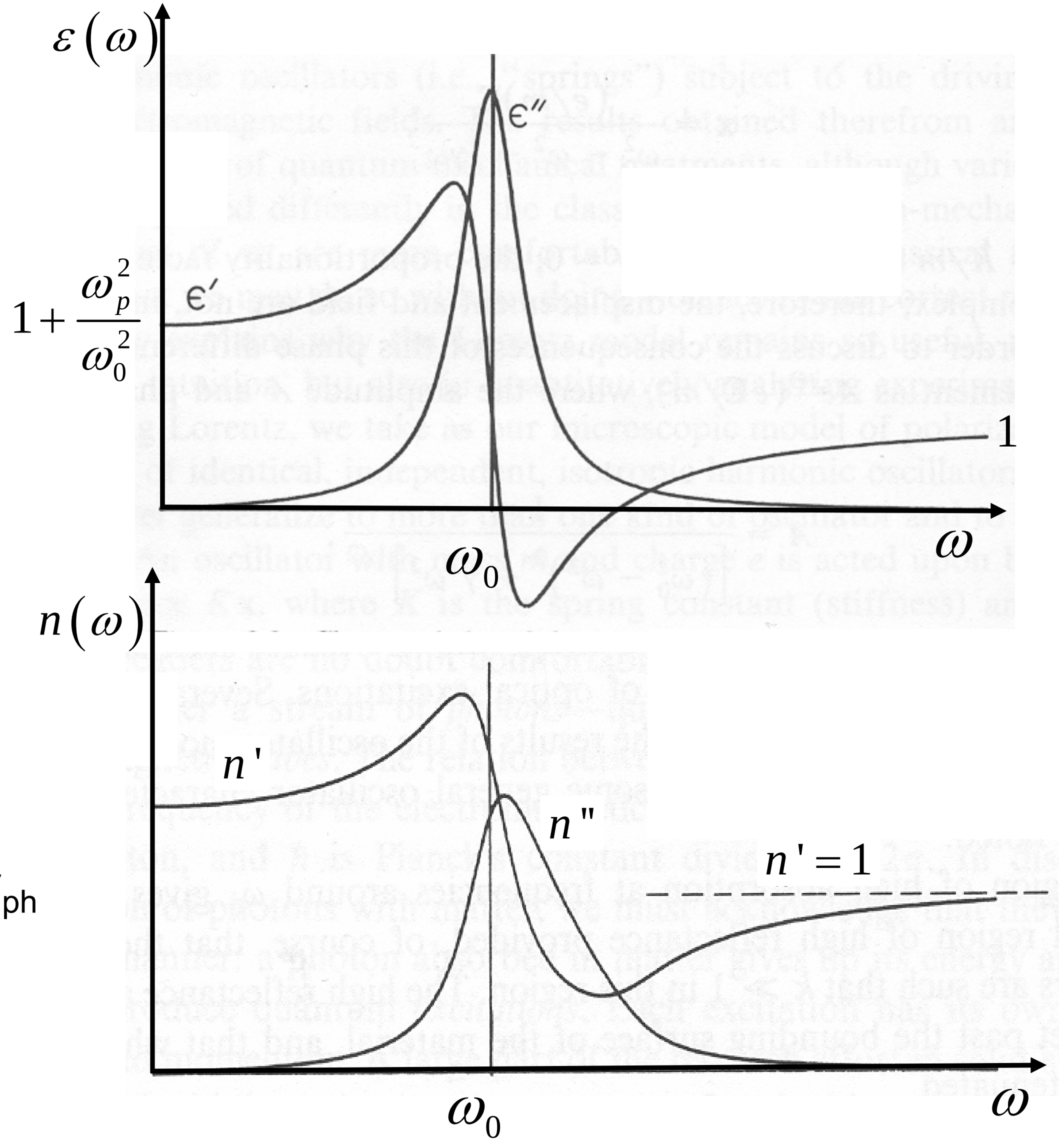
Propagation of EM-waves: Need n' and n''

Relation between n and ε

$$n = \sqrt{\varepsilon}$$

$$\varepsilon_r' = (n')^2 - (n'')^2$$

$$\varepsilon_r'' = 2n'n''$$



- $\omega \ll \omega_0$: High n' ⇨ low $v_{ph} = c/n'$
- $\omega \approx \omega_0$: Strong ω dependence v_{ph}

  Large absorption (~ n'')
- $\omega \gg \omega_0$: n' = 1 ⇨ $v_{ph} = c$

Realistic Rarefied Media

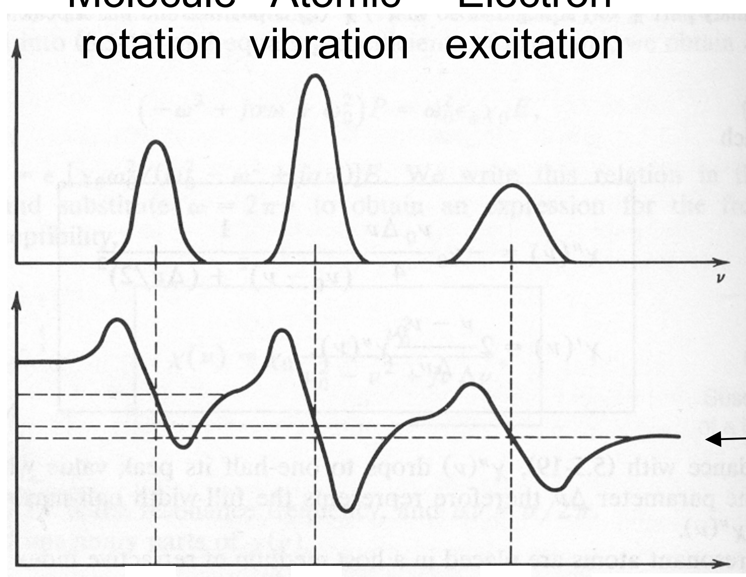
Realistic atoms have many resonances

- Resonances occur due to motion of the atoms (low ω) and electrons (high ω)

$$\Rightarrow \quad \chi = \sum_k \frac{N_k e^2}{\varepsilon_0 m} \frac{1}{\omega_k^2 - \omega^2 - i\gamma\omega}$$

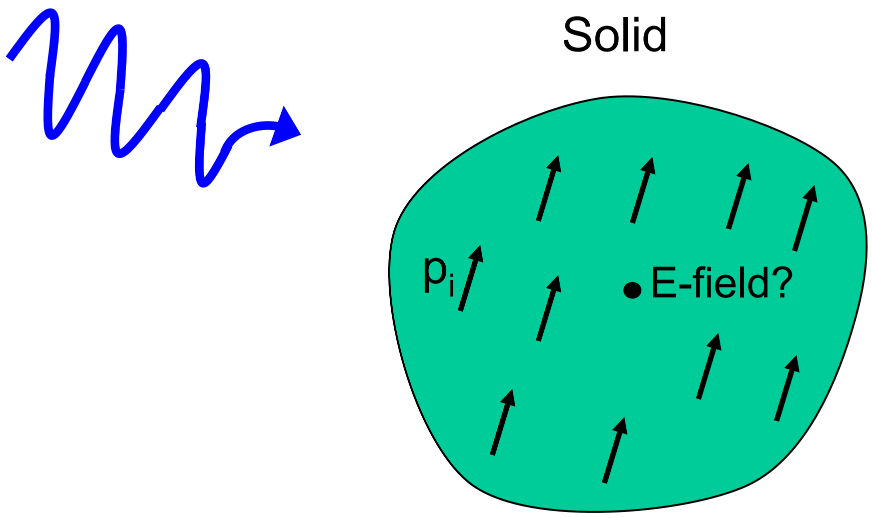
Where N_k is the density of the electrons/atoms with a resonance at ω_k

Example of a realistic dependence of n' and n''



Back to Relation Atomic Polarizability (α) and χ:

Case 2: Solids



- Atom "feels" field from: 1) Incident light beam

  2) Induced dipolar field from other atoms, p_i

- Local field:

$$\underbrace{\mathbf{E}_L}_{\text{Local field}} = \underbrace{\mathbf{E}_0}_{\text{Field without matter}} + \underbrace{\mathbf{E}_I}_{\text{Induced dipolar field from all the other atoms}}$$

Electric Susceptibility of a Solid

Local field

- Local field: $\mathbf{E}_L = \mathbf{E}_0 + \mathbf{E}_I$

Local field   Field without matter   All the other atoms

Induced dipolar field

- Example: For cubic symmetry: $\mathbf{E}_I = \dfrac{\mathbf{P}}{3\varepsilon_0}$ (Solid state Phys. Books, e.g. Kittel, Ashcroft..)

$$\Rightarrow \mathbf{E}_L = \mathbf{E}_0 + \frac{\mathbf{P}}{3\varepsilon_0}$$ (Similar relations can be derived for any solid)

Polarization of a solid

- Solid consists of atom type j at a concentration N_j

$$\mathbf{P} = \varepsilon_0 \sum_j N_j \underbrace{\alpha_j \mathbf{E}_L}_{p_j} = \varepsilon_0 \sum_j N_j \alpha_j \left(\mathbf{E}_0 + \frac{\mathbf{P}}{3\varepsilon_0} \right) = \varepsilon_0 \sum_j N_j \alpha_j \mathbf{E}_0 + \sum_j N_j \alpha_j \frac{\mathbf{P}}{3}$$

$$\Rightarrow \mathbf{P}\left(1 - \frac{1}{3}\sum_j N_j \alpha_j\right) = \varepsilon_0 \sum_j N_j \alpha_j \mathbf{E}_0 \Rightarrow \boxed{\chi = \frac{P}{\varepsilon_0 E} = \frac{\sum_j N_j \alpha_j}{1 - \frac{1}{3}\sum_j N_j \alpha_j}}$$

Clausius-Mossotti Relation

Polarization of a solid

- Susceptibility:

$$\chi = \frac{P}{\varepsilon_0 E} = \frac{\sum_j N_j \alpha_j}{1 - \frac{1}{3}\sum_j N_j \alpha_j} \qquad \text{I}$$

- Limit of low atomic concentration:

  ….or weak polarizability:

pretty good for gasses and glasses

$$\chi \approx \sum_j N_j \alpha_j \qquad \text{II}$$

Clausius-Mossotti

- By definition: $\varepsilon = 1 + \chi$
- Rearranging I gives

$$\frac{\varepsilon - 1}{\varepsilon + 2} = \frac{1}{3\varepsilon_0}\sum_j N_j \alpha_j \qquad \text{III}$$

- Conclusion: Dielectric properties of solids related to atomic polarizability
- This is very general!!