Susceptibility ($\chi$)

Linear case

induced polarization $\propto$ applied field

$$\vec{P} = \epsilon_0 \chi^{(1)} \vec{E}$$

↳ scalar if isotropic

tensor if anisotropic (birefringent)

$$\Rightarrow \vec{P} \nparallel \vec{E}$$

$$\vec{D} = \epsilon_0 \vec{E} + \vec{P}$$

$$= \epsilon_0 \underbrace{\left(1 + \chi^{(1)}\right)}_{\epsilon = n^2} \vec{E}$$

$n$ = refractive index

in a gas (low density) $\vec{P} = N_a \vec{p}$ w/ $\vec{p}$ = dipole moment of molecule

in condensed medium, apply local field corrections

$\chi^{(1)}$ is dependent on frequency $\rightarrow \chi^{(1)}(\omega)$

- controlled by resonances

Maxwell eqns no free charges, non magnetic

$$\nabla \cdot \vec{E} = 0 \qquad \nabla \times \vec{E} = -\frac{\partial}{\partial t} \vec{B}$$

$$\nabla \cdot \vec{B} = 0 \qquad \nabla \times \vec{B} = \mu_0 \frac{\partial}{\partial t} \vec{D} = \underbrace{\mu_0 \epsilon_0}_{1/c^2} \frac{\partial \vec{E}}{\partial t} + \mu_0 \frac{\partial \vec{P}}{\partial t}$$

$$\underbrace{\nabla \times (\nabla \times \vec{E})}_{\nabla(\nabla \cdot \vec{E}) - \nabla^2 \vec{E}} = -\frac{\partial}{\partial t}(\nabla \times \vec{B}) = -\frac{1}{c^2} \frac{\partial^2 \vec{E}}{\partial t^2} - \mu_0 \frac{\partial^2 \vec{P}}{\partial t^2}$$

inhomogeneous wave eqn: $\nabla^2 \vec{E} - \frac{1}{c^2} \frac{\partial^2 \vec{E}}{\partial t^2} = \underbrace{\mu_0 \frac{\partial^2 \vec{P}}{\partial t^2}}$ source term when $\vec{P} = \varepsilon_0 \chi^{(1)} \vec{E}$ (linear case)

$$\nabla^2 \vec{E} - \frac{1}{c^2}\frac{\partial^2 \vec{E}}{\partial t^2} = \frac{1}{c^2}\chi^{(1)}\frac{\partial^2 \vec{E}}{\partial t^2} \rightarrow \nabla^2 \vec{E} - \frac{n^2}{c^2}\frac{\partial^2 \vec{E}}{\partial t^2} = 0$$

if $\vec{E} = \vec{E}_1 + \vec{E}_2$ wave eqn separates into two independent eq

$\therefore$ no coupling.

<u>Nonlinear case</u>

$\vec{P}$ has a more complicated dependence on $\vec{E}$

often can expand in a Taylor series

$$P = \varepsilon_0 \left( \chi^{(1)} E + \chi^{(2)} E^2 + \chi^{(3)} E^3 + \cdots \right)$$ ignoring vector quantities for now

any $1/n!$ factors are included in $\chi$'s

separate into linear and NL parts

$$\vec{P} = \varepsilon_0 \chi^{(1)} \vec{E} + P^{NL}$$

$$\rightarrow \nabla^2 E - \frac{n^2}{c^2}\frac{\partial^2 E}{\partial t^2} = \underbrace{\mu_0 \frac{\partial^2}{\partial t^2} P^{NL}}_{\text{source term}}$$

Typically, we concentrate the analysis on one (or just a few) processes.

Notation: treat the full field as the sum of components that have a well defined central freq. $\omega_n$ and direction $\vec{k}_n$

$$\vec{E}(\vec{r}, t) = \sum_{n>0} \vec{E}_n(\vec{r}, t)$$

$$\vec{E}_n(\vec{r}, t) = \hat{\mathcal{E}}_n(\vec{r}, t) \cos(\vec{k}_n \cdot \vec{r} - \omega_n t)$$

$$= \vec{A}_n(\vec{r}, t) e^{i(\vec{k}_n \cdot \vec{r} - \omega_n t)} + c.c.$$

with the convention $A_n = \frac{1}{2}\mathcal{E}_n$

$\therefore$ intensity $= I_n = \frac{1}{2}\varepsilon_0 n c |\mathcal{E}_n|^2 = 2\varepsilon_0 n c |A_n|^2$

Now we can write the total field

$$\vec{E}(\vec{r},t) = \sum_n \vec{A}_n(\vec{r},t)\, e^{i(\vec{k}_n\cdot\vec{r} - \omega_n t)}$$

→ positive and negative freq.

Same for the polarization

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

→ can depend on $\vec{r}$, but not $e^{i\vec{k}\cdot\vec{r}}$

Now extend definition of nonlinear polarization $\chi^{(2)}$ example

$$P_i(\omega_n+\omega_m) = \sum_{jk}\sum_{(nm)} \chi^{(2)}_{ijk}(\omega_n+\omega_m;\omega_n,\omega_m)\, E_j(\omega_n)\, E_k(\omega_m)$$

$(nm)$ → sum over frequencies, holding $\omega_n+\omega_m$ constant

$i,j,k = 1,2,3$ Cartesian component 1,2,3

Summing: $i,j$, ...

$\chi^{(2)}_{ijk}$ = tensor  $\chi^{(2)}_{123}$ → polariz. along $x$, input along $y,z$

many components of $\chi_{ijk}$ are zero, or equal due to crystal symmetry.

when sum over frequencies → degeneracy factors.

Review: non isotropic, linear medium (birefringent)
- different refractive index for different directions

isotropic: $\vec{D} = \epsilon \vec{E}$ $\quad \vec{E} \parallel \vec{P}$

$= (1 + 4\pi\chi)\vec{E}$

non isotropic

$$\vec{D} = \overleftrightarrow{\epsilon} \cdot \vec{E} = (1 + 4\pi\overleftrightarrow{\chi}) \cdot \vec{E}$$

$$= \begin{pmatrix} \epsilon_{xx} & \epsilon_{xy} & \epsilon_{xz} \\ \epsilon_{yx} & \epsilon_{yy} & \epsilon_{yz} \\ \epsilon_{zx} & \epsilon_{zy} & \epsilon_{zz} \end{pmatrix} \begin{pmatrix} E_x \\ E_y \\ E_z \end{pmatrix}$$

Now $\vec{D}$ isn't parallel to $\vec{E}$

so, since $\vec{D} = \vec{E} + 4\pi\vec{P}$

$= \vec{E} + 4\pi\overleftrightarrow{\chi} \cdot \vec{E}$

$\boxed{\vec{D} = \epsilon_0 \vec{E} + \vec{P} \text{ SI} = \epsilon_0 \vec{E} + \overleftrightarrow{\chi} \cdot \vec{E}, \text{ SI}}$

Also $\vec{E}$ response is along $\vec{P}$,
could write as:

$$P_i(\omega) = \sum_j 4\pi \chi_{ij}(\omega) E_j(\omega)$$

$\epsilon_0$

It is possible to find a basis (coord system) in which $\overleftrightarrow{\epsilon}$ or $\overleftrightarrow{\chi}$ is diagonal. These are the crystal axes.

in this basis:

$$\vec{D} = \begin{pmatrix} \epsilon_x & & \\ & \epsilon_y & \\ & & \epsilon_z \end{pmatrix} \begin{pmatrix} E_x \\ E_y \\ E_z \end{pmatrix}$$

note: crystal structure doesn't have to be anything special to diagonalize.

$\epsilon_x = \epsilon_y = \epsilon_z$ isotropic, otherwise biaxial

$\epsilon_x = \epsilon_y \neq \epsilon_z$ uniaxial