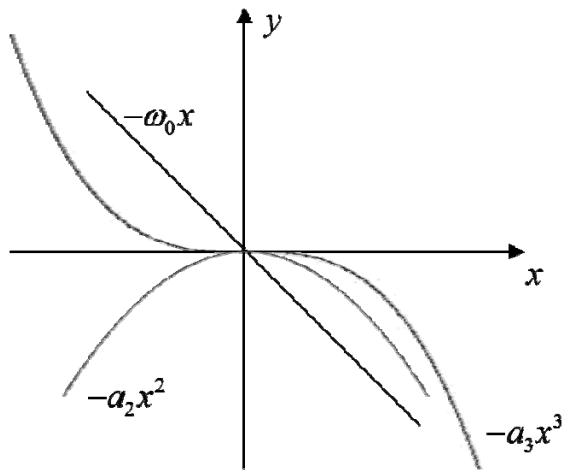


# 8. Propagation in Nonlinear Media

# 8.1. Microscopic Description of Nonlinearity.

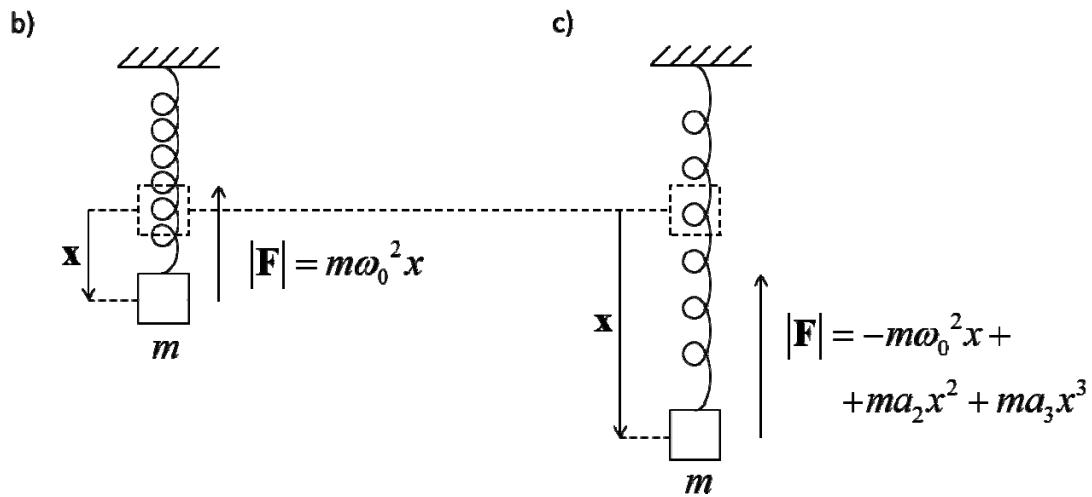
## 8.1.1. Anharmonic Oscillator.



Use Lorentz model (electrons on a spring) but with nonlinear response, or anharmonic spring

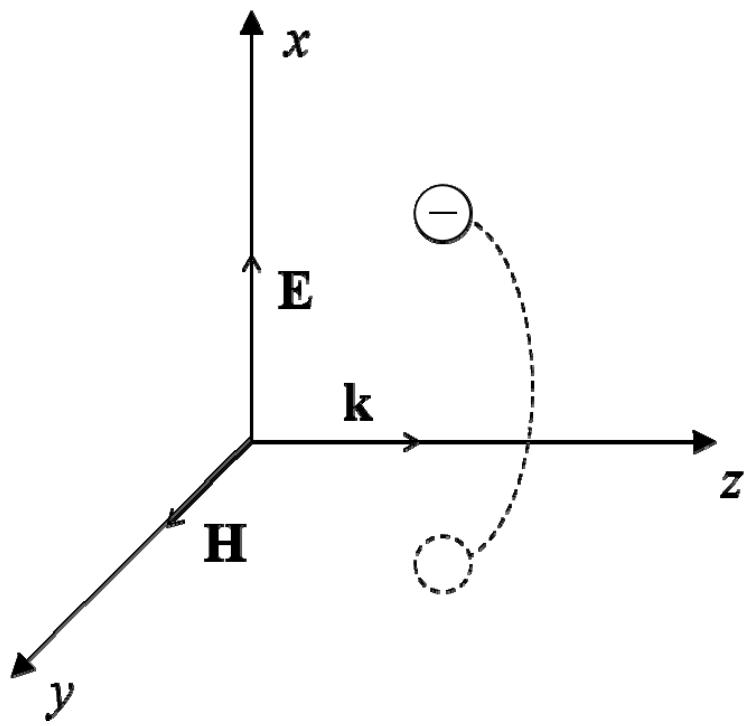
$$\frac{d^2x(t)}{dt^2} + \gamma \frac{dx(t)}{dt} + \omega_0^2 x(t) + \frac{f_{NL}[x(t)]}{m} = -\frac{eE(t)}{m}$$

$$f_{NL}/m = a_2 x(t)^2 + a_3 x(t)^3 + \dots$$



b) Harmonic spring; Low intensity

c) Anharmonic spring (overstretched); High intensity



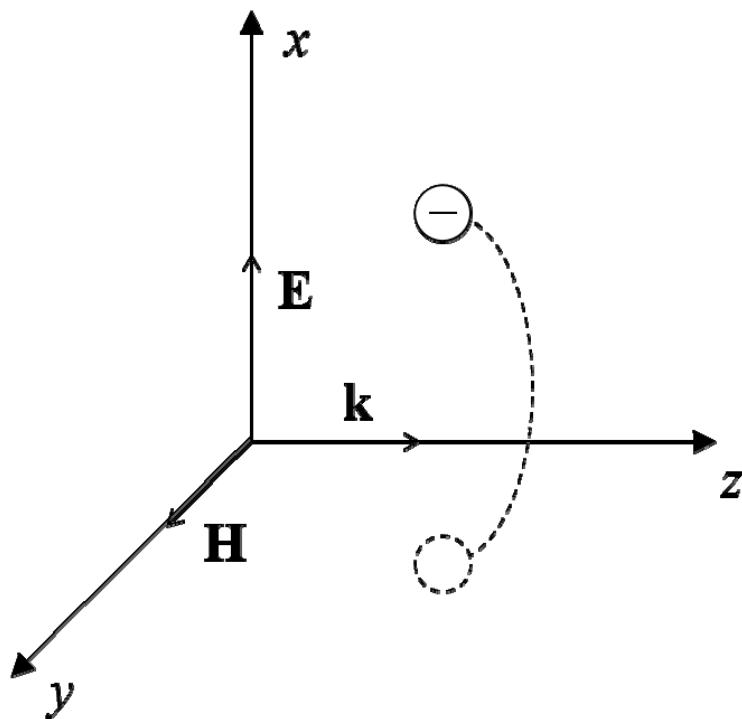
At strong incident linearly polarized fields electron trajectories no longer straight lines

## 8.1.2. Lorentz Force as Source of Nonlinearity

At strong incident linearly polarized fields electron trajectories no longer straight lines

Molecule oscillates over other degrees of freedom

Lorentz force  $-e\mathbf{v} \times \mathbf{B}$  becomes significant



Simple case where electric field polarized along x, magnetic field lies on y

$$\mathbf{E} = (E_x, 0, 0) \quad \mathbf{H} = (0, H_y, 0)$$

$$f_{em} = -e(\mathbf{E} + \mathbf{v} \times \mathbf{B})$$

$$= -e(E_x + \dot{z}B_y)\hat{x} + 0 \cdot \hat{y} - e\dot{x}B_y\hat{z}$$

$$\frac{d^2x(t)}{dt^2} + \gamma \frac{dx(t)}{dt} + \omega_0^2 x(t) = -\frac{e}{m} [E_x^{(t)} + \dot{z}(t)B_y(t)]$$

$$\frac{d^2y(t)}{dt^2} + \gamma \frac{dy(t)}{dt} + \omega_0^2 y(t) = 0$$

$$\frac{d^2z(t)}{dt^2} + \gamma \frac{dz(t)}{dt} + \omega_0^2 z(t) = -\frac{e}{m} \dot{x}(t)B_y(t)$$

Equations of motion from E and H fields, no force acting in y direction. Solve for x and z displacements.  
Take Fourier transform, obtain following equations:

$$x(\omega) [\omega_0^2 - \omega^2 - i\omega\gamma] = -\frac{eE_r(\omega)}{m} + i\frac{e}{m} [\omega z(\omega)] \cdot B_y(\omega)$$

$$z(\omega) [\omega_0^2 - \omega^2 - i\omega\gamma] = i\frac{e}{m} [\omega x(\omega)] \cdot B_y(\omega).$$

$$x(\omega) = \frac{D(\omega)}{D^2(\omega) - b^2(\omega)} \cdot \frac{-eE_x(\omega)}{m}$$

$$z(\omega) = \frac{b(\omega)}{D^2(\omega) - b^2(\omega)} \cdot \frac{-eE_x(\omega)}{m}.$$

$$D(\omega) = \omega_0^2 - \omega^2 - i\omega\gamma$$

$$b(\omega) = i \frac{e}{m} \omega B_y(\omega).$$

Equations obtained from method in 5.1  
Ignore magnetic field, we get linear response

Express magnetic field in terms of electric field

$$\mathbf{k} \times \mathbf{E}(\omega) = i\omega \mathbf{B}(\omega)$$

$$B_y(\omega) = \frac{E_x}{ic}$$

Order of magnitude relation between x and z displacements

$$\frac{z(\omega)}{x(\omega)} \approx \frac{eE_x}{me\omega} = \frac{eE_x \lambda}{2\pi mc^2}$$

Solve for when electric field and associated irradiance where x and z are comparable

$$E_x|_{x(\omega)=z(\omega)} = \frac{2\pi mc^2}{e\lambda}$$

$$\approx 10^{12} V/m,$$

$$I = \frac{1}{2\eta} E_x^2$$

$$\approx 10^{21} W/m^2$$

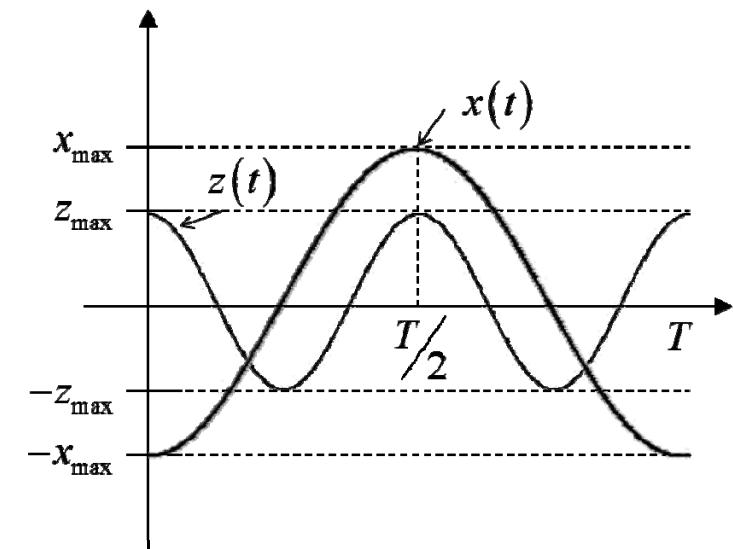
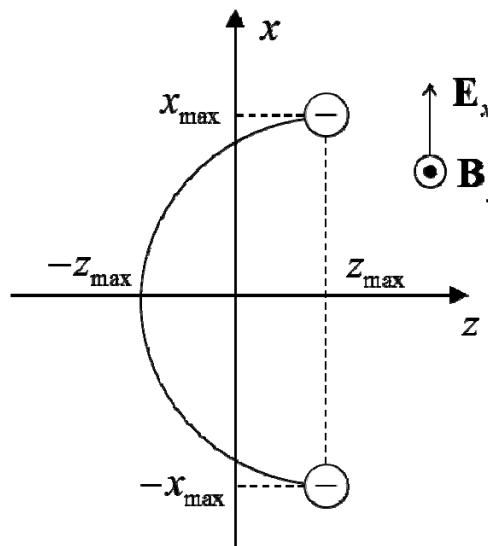
Using  $b(\omega) = \frac{e\omega E_x}{mc} \ll D(\omega)$  we have:

$$\begin{aligned} z(\omega) &= -\frac{e^2}{m^2 c} E_x^2(\omega) \frac{\omega}{D^2(\omega) - b^2(\omega)} \\ &\approx -\frac{e^2}{m^2 c} E_x^2(\omega) \frac{\omega}{D^2(\omega)} \end{aligned}$$

$$\begin{aligned} z(\omega) &\propto (E_x \cdot e^{i\omega t} + E_x e^{-i\omega t})^2 \\ &= 2E_x^2 (1 + \cos 2\omega t). \end{aligned}$$

Electron oscillates at  $2\omega$  on z axis, DC term is called optical rectification

$$\begin{aligned} x(\omega) &= -\frac{eE_x}{m} \cdot \frac{D(\omega)}{D^2(\omega) - b^2(\omega)} \\ &\approx -\frac{eE_x}{m} \cdot \frac{1}{D(\omega)}. \end{aligned}$$



### 8.1.3. Dropping the Complex Analytic Signal Representation of Real Fields.

Real field      Complex analytic signal

$$U_r = A \cos \omega t \quad U = A e^{i\omega t}$$

$$\begin{aligned} U_r &= \frac{1}{2} A^2 (e^{i\omega t} + e^{-i\omega t})^2 \\ &= A^2 (1 + \cos 2\omega t) \\ U &= A^2 e^{i2\omega t}. \end{aligned}$$

$$\text{Re}[U] = A^2 \cos(2\omega t) \neq U_r$$

Complex analytic signal does not capture DC term.

Whenever we deal with fields raised to powers higher than one, we use

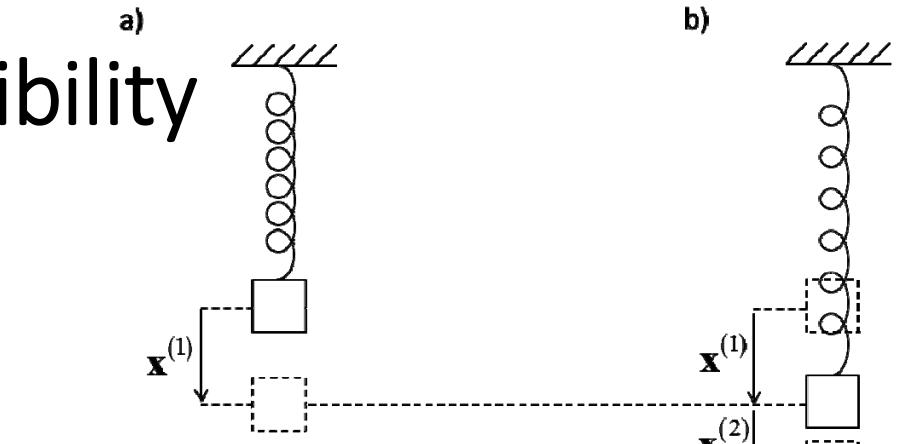
$$U_r = \frac{1}{2}(U + U^*)$$

## 8.2. Second-Order Susceptibility

$$\frac{d^2x(t)}{dt^2} + \gamma \frac{dx(t)}{dt} + \omega_0^2 x(t) + a_2 x(t)^2 = -\frac{2E(t)}{m}$$

Want solution of form  $x = x^{(1)} + x^{(2)}$  where  $x^{(2)}$  is the nonlinear perturbation to linear solution.

$$\begin{aligned} & \left[ \frac{d^2x^{(1)}(t)}{dt^2} + \gamma \frac{dx^{(1)}(t)}{dt} + \omega_0^2 x^{(1)}(t) + \frac{eE(t)}{m} \right] + \\ & + \frac{d^2x^{(2)}(t)}{dt^2} + \gamma \frac{dx^{(2)}(t)}{dt} + \omega_0^2 x^{(2)}(t) + \\ & + a_2 [x^{(1)}(t)]^2 + 2a_2 x^{(1)}(t)x^{(2)}(t) + a_2 [x^{(2)}(t)]^2 = 0. \end{aligned}$$



Simplify by neglecting  $2a_2 x^{(1)} x^{(2)}$  and  $a_2 [x^{(2)}]^2$

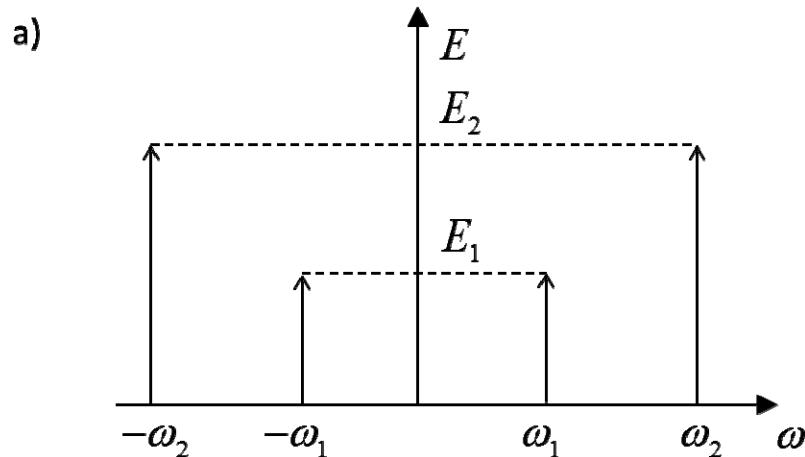
$$\dot{x}^{(2)}(t) + \gamma \dot{x}^{(2)}(t) + \omega_0^2 x^{(2)}(t) = -a_2 [x^{(1)}(t)]^2$$

$$\begin{aligned} D(\omega) x^{(2)}(\omega) &= -a_2 \tilde{x}^{(1)}(\omega) \nabla \tilde{x}^{(1)}(\omega) \\ &= -a_2 \left( \frac{e}{m} \right)^2 \left[ \frac{E(\omega)}{D(\omega)} \nabla \frac{E(\omega)}{D(\omega)} \right], \end{aligned}$$

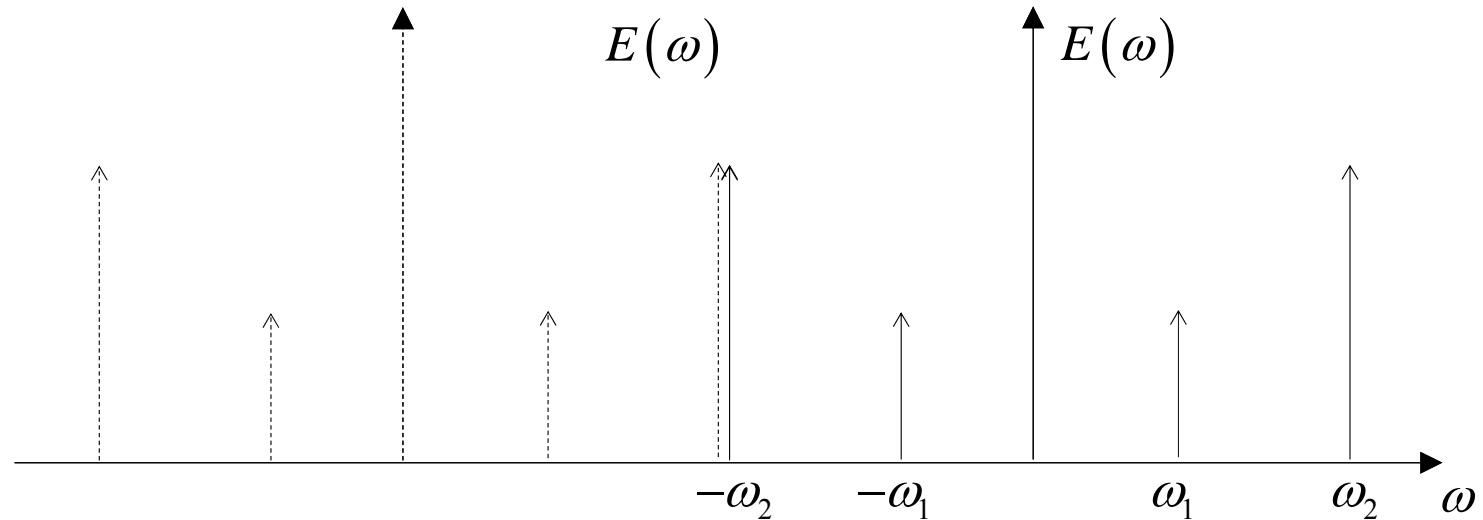
Illuminating the nonlinear crystal with two monochromatic fields of different frequencies

$$E(\omega) = E_1 \delta(\omega - \omega_1) + E_1^* \delta(\omega + \omega_1) + \\ + E_2 \delta(\omega - \omega_2) + E_2^* \delta(\omega + \omega_2)$$

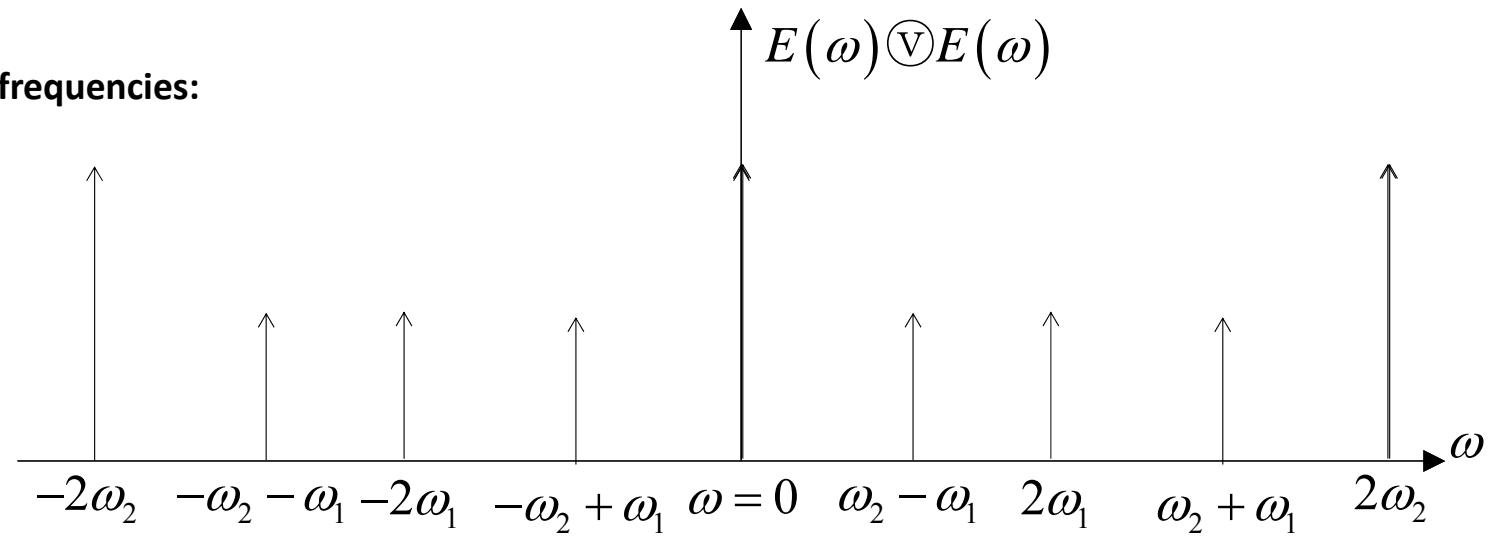
$$x^{(2)}(\omega) = -a_2 \left( \frac{e}{m} \right)^2 \frac{1}{D(\omega)} \left[ \frac{E(\omega)}{D(\omega)} \textcircled{V} \frac{E(\omega)}{D(\omega)} \right] \\ = -a_2 \left( \frac{e}{m} \right)^2 \frac{1}{D(\omega)} \int_{-\infty}^{\infty} \frac{E(\omega')}{D(\omega')} \cdot \frac{E(\omega - \omega')}{D(\omega - \omega')} d\omega'.$$



$$\delta(\omega + a) \textcircled{V} \delta(\omega + b) = \delta(\omega + a - b)$$



**All possible frequencies:**



$$x^{(2)}(\omega) = -a_2 \left(\frac{e}{m}\right)^2 \frac{1}{D(\omega)} [f(\omega_1) + f(-\omega_1) + f(\omega_2) + f(-\omega_2)]$$

$$f(\omega_1) = \frac{1}{D(\omega_1)D(\omega - \omega_1)} \begin{bmatrix} E_1^2 \delta(\omega - 2\omega_1) + |E_1|^2 \delta(\omega) + \\ + E_1 E_2 \delta[\omega - (\omega_1 + \omega_2)] + \\ + E_1 E_2^* \delta[\omega - (\omega_1 - \omega_2)] \end{bmatrix}$$

$$f(\omega_2) = \frac{1}{D(\omega_2)D(\omega - \omega_2)} \begin{bmatrix} E_2^2 \delta(\omega - 2\omega_2) + |E_2|^2 \delta(\omega) + \\ + E_1 E_2 \delta[\omega - (\omega_1 + \omega_2)] + \\ + E_1 E_2^* \delta[\omega - (\omega_1 - \omega_2)] \end{bmatrix}$$

Fourier  
Transform       $\rightarrow$

$$x^{(2)}(t) = -a_2 \left(\frac{e}{m}\right)^2 [g_1(t) + c.c. + g_2(t) + c.c.]$$

$$g_1(t) = \frac{E_1^2 e^{i2\omega_1 t}}{D(2\omega_1)D^2(\omega_1)} + \frac{|E_1|^2}{D(0)|D(\omega_1)|^2} + \frac{E_1 E_2 e^{i(\omega_1 + \omega_2)t}}{D(\omega_1 + \omega_2)D(\omega_1)D(\omega_2)} +$$

$$+ \frac{E_1 E_2^* e^{i(\omega_1 - \omega_2)t}}{D(\omega_1 - \omega_2)D(\omega_1)D^*(\omega_2)} + c.c.$$

$$g_2(t) = \frac{E_2^2 e^{i2\omega_2 t}}{D(2\omega_2)D^2(\omega_2)} + \frac{|E_2|^2}{D(0)|D(\omega_2)|^2} + \frac{E_1 E_2 e^{i(\omega_1 + \omega_2)t}}{D(\omega_1 + \omega_2)D(\omega_1)D(\omega_2)} +$$

$$+ \frac{E_1 E_2^* e^{i(\omega_1 - \omega_2)t}}{D(\omega_1 - \omega_2)D(\omega_1)D^*(\omega_2)} + c.c.$$

**Equation 8.26 indicates that, as the result of the second-order nonlinear interaction, the resulting field has components that oscillate at frequencies  $2\omega_2$  (second harmonic of  $\omega_2$ ),  $2\omega_1$  (second harmonic of  $\omega_1$ ),  $\omega_1 + \omega_2$  (sum frequency),  $\omega_1 - \omega_2$  (difference frequency) and 0 (optical rectification terms).**

## Nonlinear susceptibility from induced polarization

$$P^{(2)}(\pm\omega_i \pm \omega_j) = \varepsilon_0 \chi^{(2)}(\pm\omega_i \pm \omega_j; \pm\omega_i, \pm\omega_j) E^*(\pm\omega_i) E^*(\pm\omega_j) \rightarrow \chi^{(2)}(\pm\omega_i \pm \omega_j; \pm\omega_i, \pm\omega_j) = -\frac{Ne}{\varepsilon_0} x^{(2)}(\pm\omega_i \pm \omega_j)$$

$$P^{(2)}(\pm\omega_i \pm \omega_j) = -N e x^{(2)}(\pm\omega_i \pm \omega_j), \quad i, j = 1, 2$$

$$= \frac{a_2 N e^3}{\varepsilon_0 m^2} [g_1(t) + g_2(t) + c.c.]$$

Importantly,  $\chi^{(2)}$  vanishes in *centrosymmetric media*  $\chi^{(2)}(\mathbf{r}) = \chi^{(2)}(-\mathbf{r})$

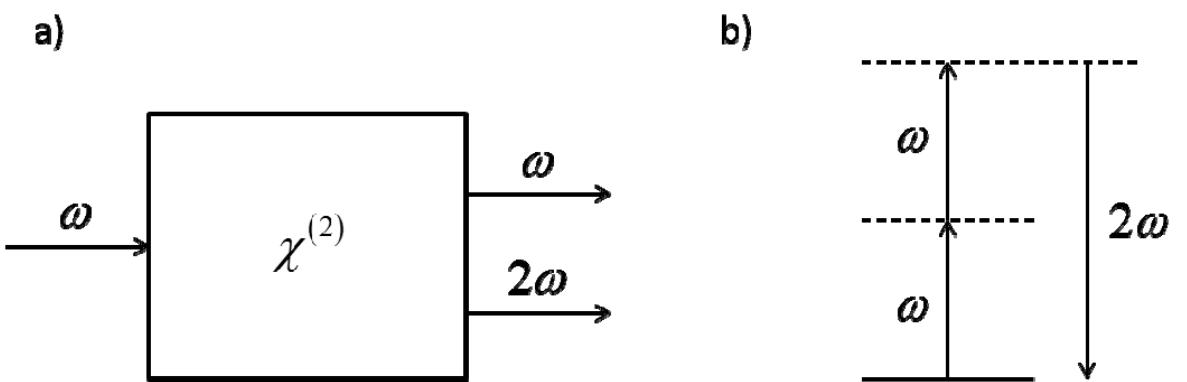
$$\begin{aligned} \mathbf{P}^{(2)}(\mathbf{r}) &= \varepsilon_0 \chi^{(2)}(\mathbf{r}) \mathbf{E}(\mathbf{r}) \mathbf{E}(\mathbf{r}) \\ &= \varepsilon_0 \chi^{(2)}(-\mathbf{r}) \mathbf{E}(-\mathbf{r}) \mathbf{E}(-\mathbf{r}) \\ &= \mathbf{P}(-\mathbf{r}). \end{aligned} \quad \begin{aligned} \mathbf{P}^{(2)}(\mathbf{r}) &= -Ner \\ &= Ne(-\mathbf{r}) \\ &= -\mathbf{P}^{(2)}(\mathbf{r}). \end{aligned} \quad \begin{aligned} &\text{Fulfilled simultaneously only if} \\ &\mathbf{P}^{(2)}(\mathbf{r}) = 0 \end{aligned}$$

**So second-order nonlinear processes require *noncentrosymmetric media***

### 8.2.1. Second Harmonic Generation (SHG)

$$\chi(2\omega_1; \omega_1, \omega_1) = \frac{a_2 Ne^3}{\epsilon_0 m^2} \cdot \frac{1}{D(2\omega_1) D^2(\omega_1)}$$

$$\chi(2\omega_2; \omega_2, \omega_2) = \frac{a_2 Ne^3}{\epsilon_0 m^2} \cdot \frac{1}{D(2\omega_2) D^2(\omega_2)}.$$



**Nonlinear susceptibility as function of linear chi:**

**Linear:**

$$\chi^{(1)}(\omega) = \frac{Ne^2}{\epsilon_0 m} \cdot \frac{1}{D(\omega)}.$$

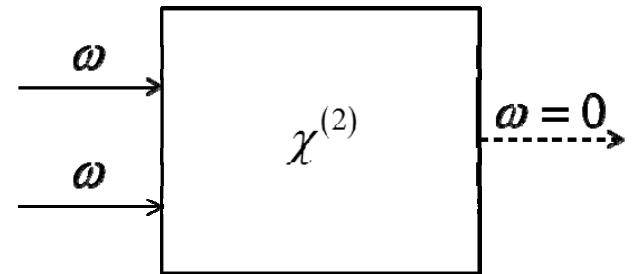
- a) SHG: pumping the chi(2) material at omega yields both the fundamental frequency (omega) and its second harmonic (2omega).
- b) Description in terms of virtual energy levels.

**Nonlinear:**

$$\chi^{(2)}(2\omega_1; \omega_1, \omega_1) = \frac{a_2 \epsilon_0^2 m}{N^2 e^3} \chi^{(1)}(2\omega_1) [\chi^{(1)}(\omega_1)]^2.$$

### 8.2.2. Optical Rectification (OR)

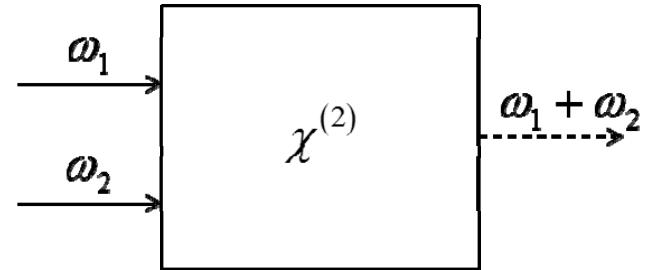
$$\begin{aligned}\chi^{(2)}(0; \omega_1, -\omega_1) &= \frac{a_2 Ne^3}{\epsilon_0 m^2} \cdot \frac{1}{D(0) D(\omega_1) D(-\omega_1)} \\ &= \frac{a_2 \epsilon_0^2 m}{N^2 e^3} \chi^{(1)}(0) \chi^{(1)}(\omega_1) \chi^{(1)}(-\omega_1).\end{aligned}$$



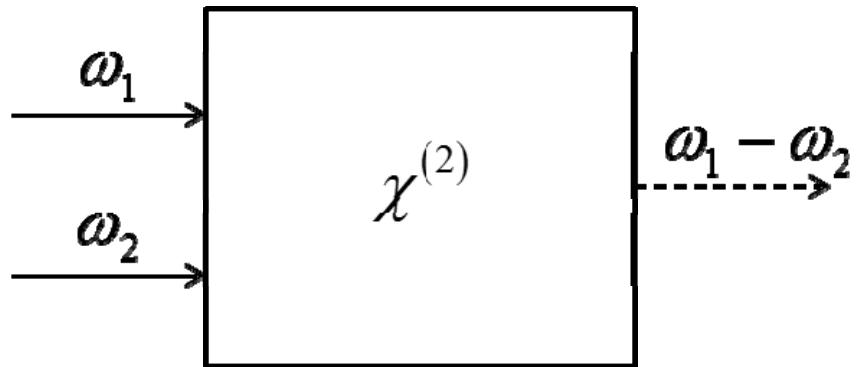
OR: a DC polarization is created in a chi(2) material.

### 8.2.3. Sum Frequency Generation (SFG)

$$\begin{aligned}\chi^{(2)}(\omega_1 + \omega_2; \omega_1, \omega_2) &= \frac{a_2 Ne^3}{\varepsilon_0 m^2} \cdot \frac{1}{D(\omega_1 + \omega_2) D(\omega_1) D(\omega_2)} \\ &= \frac{a_2 \varepsilon_0^2 m}{N^2 e^3} \chi^{(1)}(\omega_1 + \omega_2) \chi^{(1)}(\omega_1) \chi^{(1)}(\omega_2).\end{aligned}$$



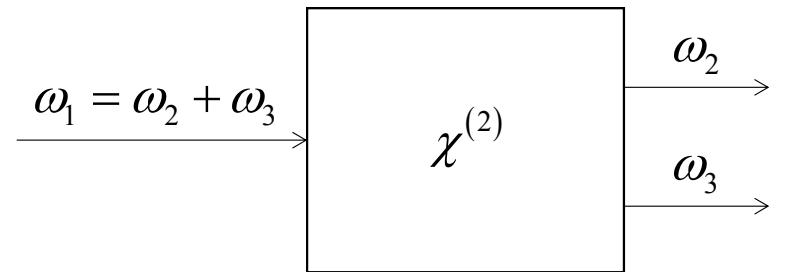
#### 8.2.4. Difference Frequency Generation (DFG)



$$\begin{aligned}\chi^{(2)}(\omega_1 - \omega_2; \omega_1, -\omega_2) &= \frac{a_2 Ne^3}{\varepsilon_0 m^2} \frac{1}{D(\omega_1 - \omega_2) D(\omega_1) D(-\omega_2)} \\ &= \frac{a_2 \varepsilon_0^2 m}{N^2 e^3} \chi^{(1)}(\omega_1 - \omega_2) \chi^{(1)}(\omega_1) \chi^{(1)}(-\omega_2).\end{aligned}$$

### 8.2.5. Optical Parametric Generation (OPG)

The time reverse process of SFG



## 8.3. Third-Order Susceptibility

Anharmonic oscillator

$$\ddot{x}(t) + \gamma \dot{x}(t) + \omega_0^2 x(t) + a_3 x^3(t) = -\frac{eE(t)}{m}.$$

Solve using perturbation theory, solution of form  $x(t) = x^{(1)}(t) + x^{(3)}(t)$

$$\begin{aligned} & \left[ \dot{x}^{(1)}(t) + \gamma \dot{x}^{(1)}(t) + \omega_0^2 x^{(1)}(t) + \frac{eE(t)}{m} \right] + \\ & + \ddot{x}^{(3)}(t) + \gamma \dot{x}^{(3)}(t) + \omega_0^2 x^{(3)}(t) + a_3 [x^{(1)}(t) + x^{(3)}(t)]^3 = 0. \end{aligned}$$

First term vanishes, approximate  $a_3 (x^{(1)} + x^{(3)})^3 \approx a_3 (x^{(1)})^3$   $\rightarrow$   $\ddot{x}^{(3)}(t) + \gamma \dot{x}^{(3)}(t) + \omega_0^2 x^{(3)}(t) = -a_3 [x^{(1)}(t)]^3$ ,

$$x^{(1)}(t) \leftrightarrow x^{(1)}(\omega) = -\frac{e}{m} \frac{E(\omega)}{D(\omega)}, \quad E(\omega) = E_1 \delta(\omega - \omega_1) + E_1^* \delta(\omega + \omega_1) + \\ + E_2 \delta(\omega - \omega_2) + E_2^* \delta(\omega + \omega_2) + \\ + E_3 \delta(\omega - \omega_3) + E_3^* \delta(\omega + \omega_3).$$

So,

$$x^{(1)}(t) = -\frac{e}{m} \sum_{n=1}^3 \frac{E_n e^{-i\omega_n t}}{D(\omega_n)} + c.c.$$

For scalar fields perturbation displacement is:

$$\ddot{x}^{(3)}(t) + \gamma \dot{x}^{(3)}(t) + \omega_0^2 x(t) = -a_3 \left( \frac{e}{m} \right)^3 \sum_{m,n,p=-3}^3 \frac{E_m E_n E_p e^{-i(\omega_m + \omega_n + \omega_p)}}{D(\omega_m) D(\omega_n) D(\omega_p)}$$

Induced polarization both for electromagnetic fields in terms of displacement and susceptibility

$$P_i^{(3)}(\omega_q) = -N e x^{(3)}(\omega_q)$$

$$\begin{aligned} P_i^{(3)}(\omega_q) &= \varepsilon_0 \sum_{j,k,l=1}^3 \sum_{m,n,p=-3}^3 \chi_{ijkl}^{(3)}(\omega_q; \omega_m, \omega_n, \omega_p) E_j(\omega_m) E_k(\omega_n) E_l(\omega_p) \\ &= \varepsilon_0 d \sum_{jkl} \chi_{ijkl}^{(3)}(\omega_q; \omega_m, \omega_n, \omega_p) E_j(\omega_m) E_k(\omega_n) E_l(\omega_p), \end{aligned}$$

Where  $d$  is the degeneracy factor

General expression for  $\chi^{(3)}$

$$\chi_{ijkl}^{(3)}(\omega_q; \omega_m, \omega_n, \omega_p) = \frac{a_3 Ne^*}{d\varepsilon_0 m^3} \cdot \frac{1}{D_i(\omega_q) D_j(\omega_m) D_k(\omega_n) D_l(\omega_p)}, \quad D_a(\omega) = \omega_{0a}^2 - \omega^2 - i\gamma\omega$$

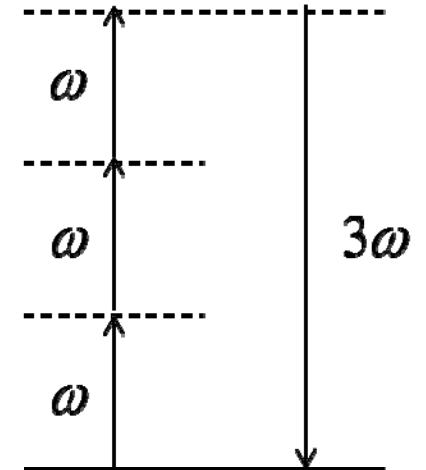
Express  $\chi^{(3)}$  in terms of  $\chi^{(1)}$

$$\chi_{ijkl}^{(3)}(\omega_q; \omega_m, \omega_n, \omega_p) = \frac{a_3 m \varepsilon_0^3}{d N^3 e^4} \left[ \chi_i^{(1)}(\omega_q) \chi_j^{(1)}(\omega_m) \chi_k^{(1)}(\omega_n) \chi_l^{(1)}(\omega_p) \right], \quad \chi_a^{(1)}(\omega) = \frac{Ne^2}{\varepsilon_0 m} \cdot \frac{1}{D_a(\omega)}$$

### 8.3.1. Third Harmonic Generation (THG) signal contained by terms that oscillate at $3\omega$

The THG susceptibility for one component of the electric field (scalar case) is

$$\begin{aligned}\chi^{(3)}(3\omega; \omega, \omega, \omega) &= \frac{a_3 Ne^4}{d\varepsilon_0 m^3} \cdot \frac{1}{D(3\omega)[D(\omega)]^3} \\ &= \frac{a_3 m \varepsilon_0^3}{N^3 e^4} \chi^{(1)}(3\omega) [\chi^{(1)}(\omega)]^3,\end{aligned}\quad D(\omega) = \omega_0^2 - \omega^2 - i\gamma\omega$$



### 8.3.2. Two-Photon Absorption (TPA) and Intensity-Dependent Refractive Index

If  $\omega_1 = \omega$ ,  $\omega_2 = \omega$ ,  $\omega_3 = -\omega$ ,

$$\begin{aligned}\chi^{(3)}(\omega; \omega, \omega, -\omega) &= \frac{a_3 Ne^4}{d\varepsilon_0 m^3} \cdot \frac{1}{D^2(\omega) |D(\omega)|^2} \\ &= \frac{a_3 m \varepsilon_0^3}{N^3 e^4} \left\{ [\chi^{(1)}(\omega)]^2 |\chi^{(1)}(\omega)|^2 \right\}\end{aligned}$$

$$\begin{aligned}\chi^{(3)}(\omega) &= \frac{a_3 m \varepsilon_0^3}{N^3 e^4} |\chi^{(1)}(\omega)|^2 \left\{ [\chi_R^{(1)}(\omega)]^2 - [\chi_I^{(1)}(\omega)]^2 + i2\chi_R^{(1)}(\omega)\chi_I^{(1)}(\omega) \right\} \\ &= \chi_R^{(3)}(\omega) + i\chi_I^{(3)}(\omega),\end{aligned}$$

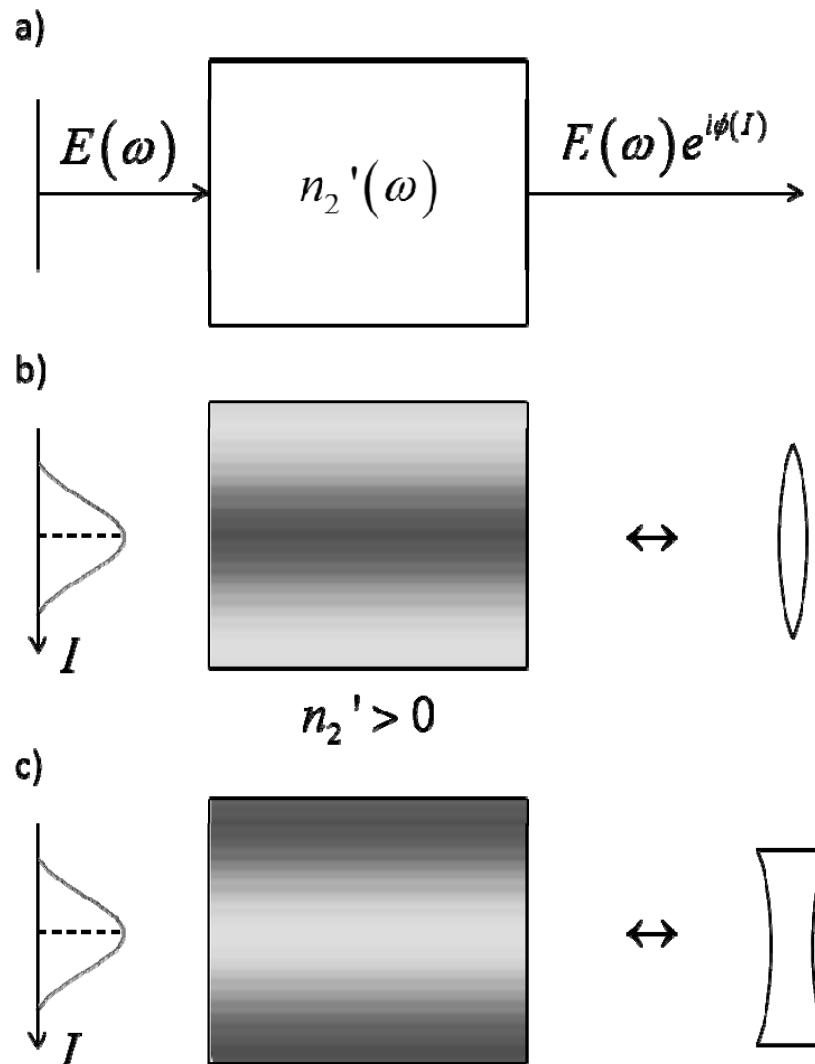
Define effective refractive index

$$\begin{aligned}\chi(\omega) &= \chi^{(1)}(\omega) + 3\chi^{(3)}(\omega)|E(\omega)|^2 \\ &= n_0^2 - 1 + 3\chi^{(3)}(\omega)|E(\omega)|^2 \\ &= n^2 - 1,\end{aligned}$$

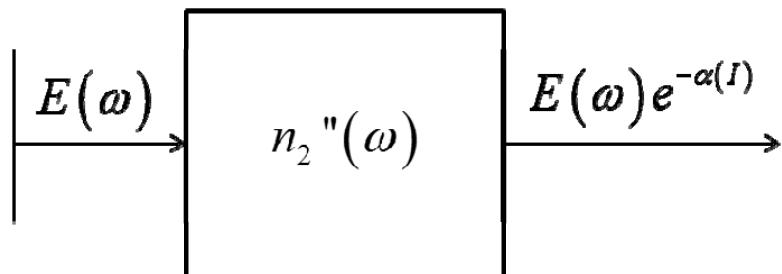
$$n = n_0 + n_2 I$$

Find expression for  $n_2$

$$\begin{aligned}n_0^2 + 2n_2 I &= 1 + \chi(\omega) \quad n_2(\omega) = \frac{3\chi^{(3)}}{4n_0^2 \epsilon_0 c} \\ &= \frac{3}{4n_0^2 \epsilon_0 c} \left[ \chi_R^{(3)}(\omega) + i\chi_I^{(3)}(\omega) \right] \\ &= n_2'(\omega) + i n_2''(\omega).\end{aligned}$$

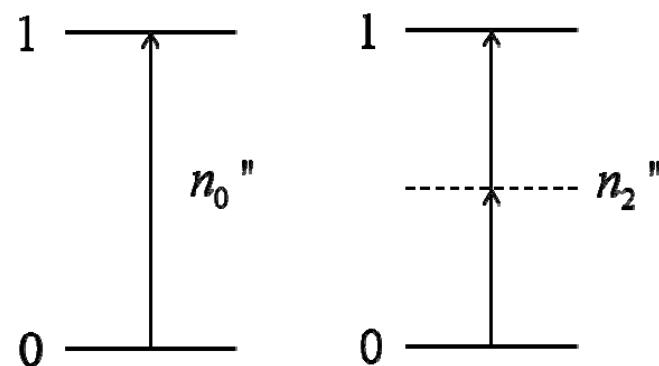


a)



- A plane wave undergoes an intensity-dependent loss of factor  $e^{-\alpha(I)}$ .
- b) Energy level diagram for single photon absorption (left) and two-photon absorption (right).

b)



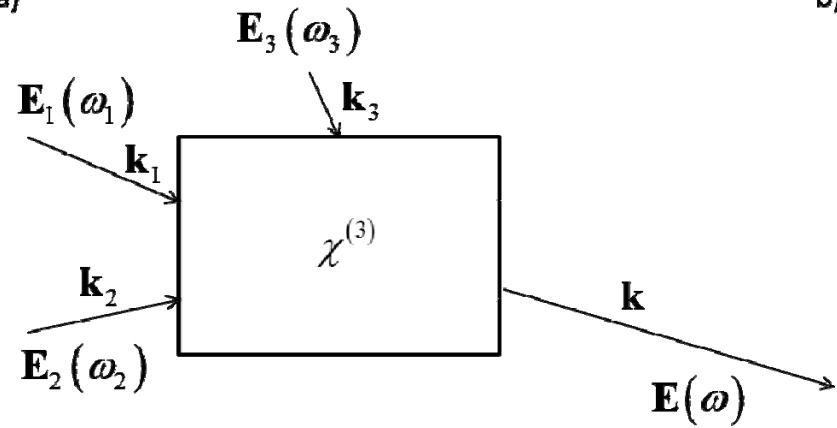
### 8.3.3. Four Wave Mixing

$$\chi^{(3)}(\omega; \omega_1, \omega_2, \omega_3) = \frac{a_3 Ne}{d\varepsilon_0 m^3} \cdot \frac{1}{D(\omega) D(\omega_1) D(\omega_2) D(\omega_3)}$$

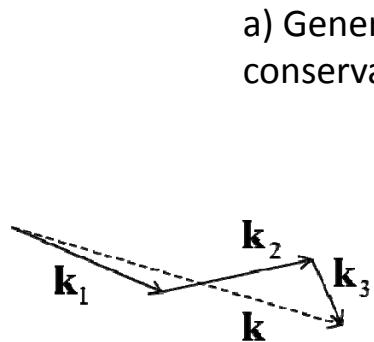
$$\omega = \omega_1 + \omega_2 + \omega_3 \quad \mathbf{k} = \mathbf{k}_1 + \mathbf{k}_2 + \mathbf{k}_3$$

k-vector conservation, phase matching condition

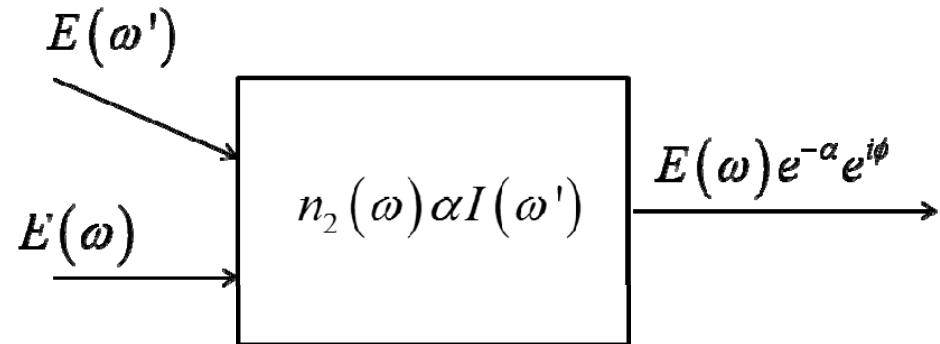
a)



b)



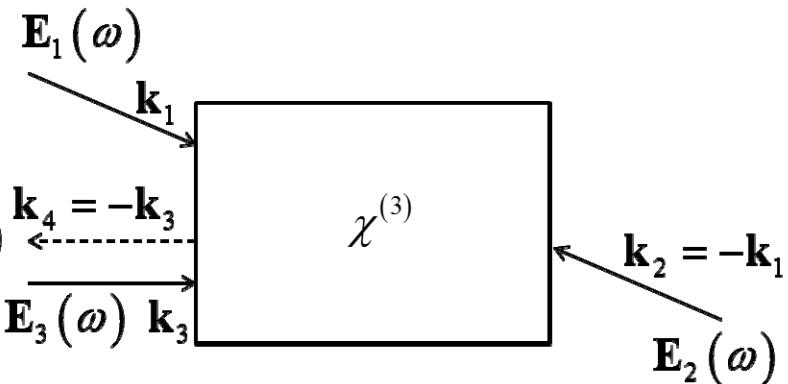
a) Generic four-wave mixing process. b) Momentum conservation.



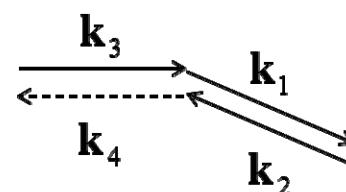
$$P_{NL} = 6\epsilon_0 \chi^{(3)} A_1 A_2 A_3^* e^{i(\mathbf{k}_1 + \mathbf{k}_2 + \mathbf{k}_3) \cdot \mathbf{r}}.$$

#### 8.3.4. Phase Conjugation via Degenerate (all $\omega$ are the same) Four-Wave Mixing

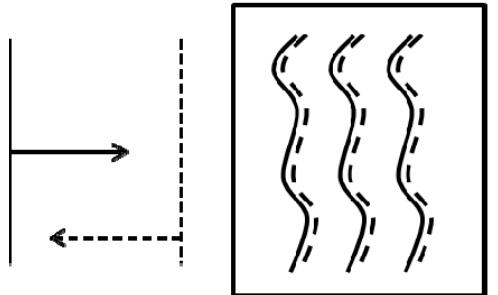
a)



b)



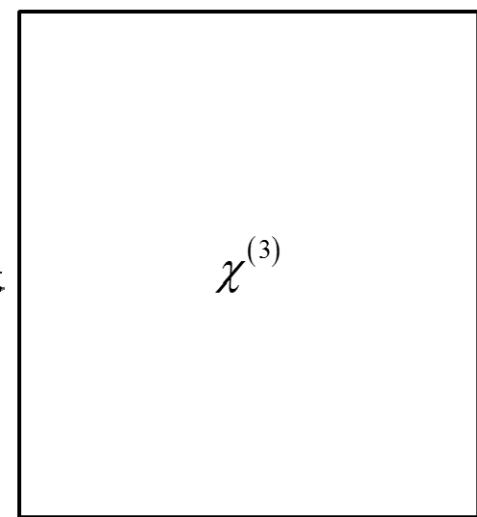
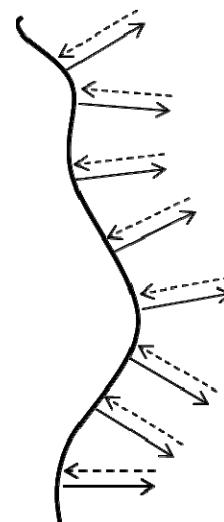
Phase conjugation via degenerated four wave mixing: field  $E_4$  emerges as the phase conjugate of  $E_3$ , i.e.  $E_4=E_3^*$ .



Distorting medium



Phase conjugator

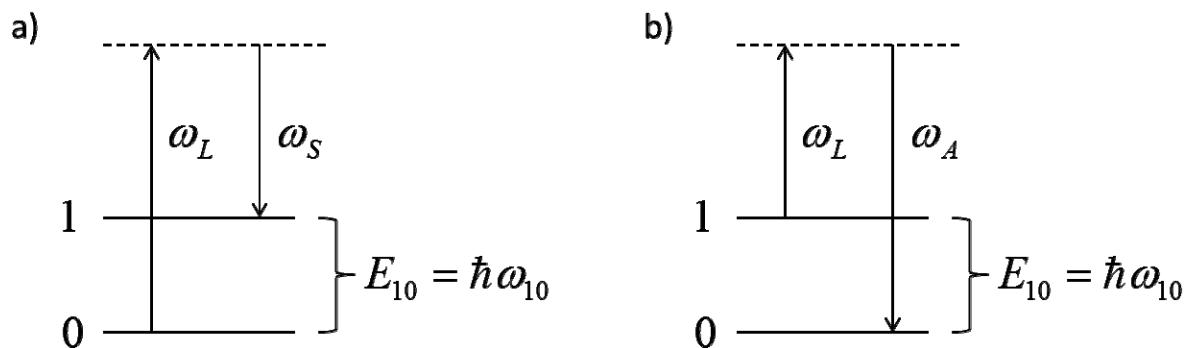
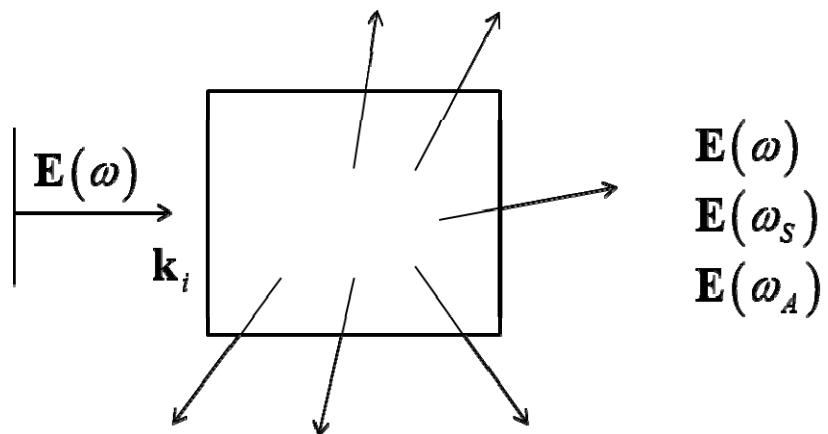


### 8.3.5. Stimulated Raman Scattering (SRS).

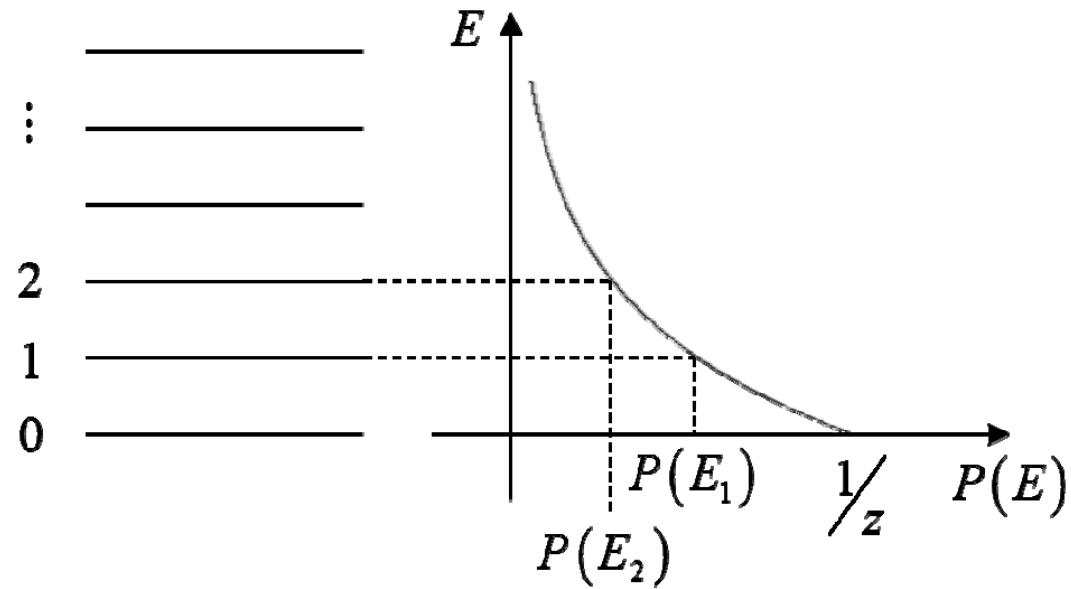
#### 8.3.5.1. Spontaneous Raman Scattering

Population of the excited vibrational levels  
obeys the Maxwell-Boltzmann distribution

$$P(E) = \frac{1}{z} e^{-\frac{E}{k_B T}},$$



Spontaneous Raman scattering:  
 -a) Stokes shift  
 -b) anti-Stokes shift



$$\frac{P(E_1)}{P(E_0)} = e^{\frac{-\bar{h}\omega_{10}}{k_B T}} \ll 1$$

**Thus, the Stokes component is typically orders of magnitude stronger !**

$\mathbf{p} = \alpha \mathbf{E}$ . the molecular *optical polarizability*,  $\alpha$

$$\boxed{\alpha(t) = \alpha_0 + \frac{\partial \alpha}{\partial x_v} \Big|_{x_v=0} x_v(t).}$$

$$P(t) = N \langle P \rangle \\ = N \left[ \alpha_0 + \frac{\partial \alpha}{\partial x_v} \Big|_{x_v=0} x_v(t) \right] E(t).$$

$$x_v(t) = A_v e^{-i\omega_v t} + A_v^* e^{i\omega_v t},$$

**Harmonic vibration**  
occurs spontaneously due  
to Brownian motion

$$P(t) = P_L(t) + P_{NL}(t)$$

$$P_L(t) = N \alpha_0 \left[ A_L e^{-i\omega_L t} + c.c. \right]$$

$$P_{NL}(t) = N \frac{\partial \alpha}{\partial x_v} \Big|_0 \left[ A_v^* A_L e^{-i(\omega_L - \omega_v)t} + c.c. + A_v A_L e^{-i(\omega_L + \omega_v)t} + c.c. \right].$$

### 8.3.5.2. Stimulated Raman Scattering

To derive expression for SRS susceptibility, solve equation of motion for vibrational mode of resonant frequency and damping  $\omega_v, \gamma$

$$\frac{d^2x_v(t)}{dt^2} + \gamma \frac{dx_v(t)}{dt} + \omega_v^2 x_v(t) = \frac{F(t)}{m},$$

$$\begin{aligned} W &= \frac{1}{2} \langle p(t) E(t) \rangle_t & F(t) &= \frac{dW}{dx_v} \\ dW &= F dx_v. & &= \frac{1}{2} \frac{d\alpha}{dx_v} \Big|_0 \langle E^2(t) \rangle_t \\ &= \frac{1}{2} \alpha \langle E^2(t) \rangle_t & &= \frac{1}{2} \frac{d\alpha}{dx_v} \Big|_0 \langle E^2(t) \rangle_t \\ &= \frac{1}{2} \left( \alpha_0 + \frac{d\alpha}{dx_v} \Big|_0 x_v \right) \langle E^2(t) \rangle_t & \tilde{F}(\omega) &= \frac{1}{2} \frac{d\alpha}{dx_v} \Big|_0 E(\omega) \circledast E(\omega) \end{aligned}$$

$$E(t) = A_L e^{-i\omega_L t} + A_S e^{-i\omega_S t} + c.c.$$

$$E(\omega) = A_L \delta(\omega - \omega_L) + A_S \delta(\omega - \omega_S) + A_L^* \delta(\omega + \omega_L) + A_S^* \delta(\omega + \omega_S)$$

$$\begin{aligned}x_v(\omega) &= \frac{\tilde{F}(\omega)}{m} \frac{1}{\omega_v^2 - \omega^2 - i\gamma\omega} \\&= \frac{1}{2m} \frac{d\alpha}{dx_v} \Bigg|_0 \frac{A_L^* A_S \delta(\omega - \omega_L) \circledcirc \delta(\omega + \omega_S)}{\omega_v^2 - \omega^2 - i\gamma\omega}\end{aligned}$$

$$\begin{aligned}P(\omega) &= N \left[ \alpha_0 + \frac{\partial \alpha}{\partial x_v} \Bigg|_0 x_v(\omega) \right] E(\omega) \\&= N \alpha_0 E(\omega) + N \frac{\partial \alpha}{\partial x_v} \Bigg|_0 x_v(\omega) E(\omega) \\&= P_L(\omega) + P_{NL}(\omega).\end{aligned}$$

$$\begin{aligned}P_{NL}(\omega) &= \frac{N}{2m} \left( \frac{d\alpha}{dx_v} \Bigg|_0 \right)^2 \frac{A_L^* A_S \delta(\omega - \omega_L) \circledcirc \delta(\omega + \omega_S)}{\omega_v^2 - \omega^2 - i\gamma\omega} \left[ A_L \delta(\omega + \omega_L) + A_L^* \delta(\omega - \omega_L) \right] \\&= \varepsilon_0 d \chi^{(R)}(\omega) A_L^* A_S \delta[\omega - (\omega_L - \omega_S)] \left[ A_L \delta(\omega + \omega_L) + A_L^* \delta(\omega - \omega_L) \right]\end{aligned}$$

nonlinear contribution

$$P_{NL}(t) = \frac{N}{2m} \left( \frac{d\alpha}{dx_v} \Bigg|_0 \right)^2 \frac{A_L^* A_S \left( A_L e^{-i\omega_S t} + A_L^* e^{i(2\omega_L - \omega_S)t} \right)}{\omega_v^2 - (\omega_L - \omega_S)^2 - i\gamma(\omega_L - \omega_S)}$$

Induced polarization for SRS

$$\chi^{(s)}(t) = \frac{N}{12m\epsilon_0} \left( \frac{dx}{dx_v} \Big|_0 \right)^2 \cdot \frac{1}{\omega_v^2 - (\omega_L - \omega_s)^2 + i\gamma(\omega_L - \omega_s)}$$

$$\boxed{\omega_L = \omega_v + \omega_s} \rightarrow \chi^{(s)}(t) = -i \frac{N}{12m\epsilon_0} \left( \frac{dx}{dx_v} \Big|_0 \right)^2 \frac{1}{\gamma(\omega_L - \omega_s)} \in \mathbb{C} - \Re$$

**By resonantly enhancing the vibration mode the Stokes component can be amplified significantly; in practice, this amplification can be many orders of magnitude higher than for the spontaneous Raman.**

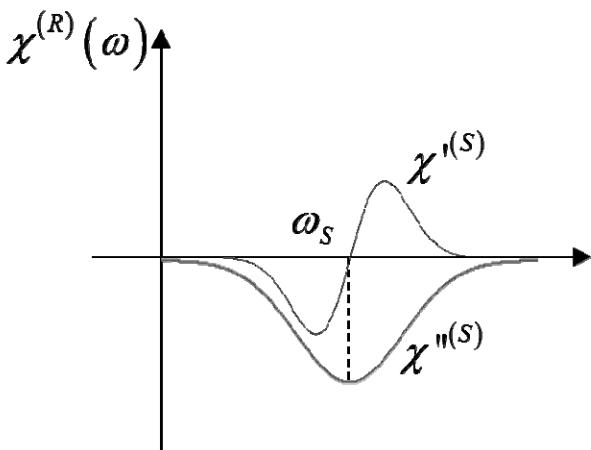
susceptibility associated with the *anti-Stokes component*

$$\omega_L - \omega_{AS} = -\omega_v = -(\omega_L - \omega_S)$$

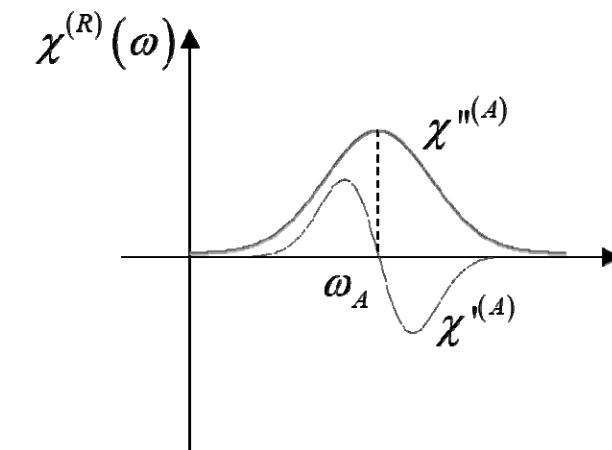
$$\begin{aligned}\chi^{(AS)}(t) &= i \frac{N}{12m\epsilon_0} \left( \frac{d\alpha}{dx_v} \Big|_0 \right)^2 \frac{1}{\gamma(\omega_L - \omega_S)} \\ &= \chi^{(S)}(t)^*\end{aligned}$$

**Strong attenuation**

a)



b)



- a) Raman susceptibility at Stokes frequency; indicates amplification.
- b) Raman susceptibility at anti-Stokes frequency; indicates absorption.

### 8.3.6. Coherent Anti-Stokes Raman Scattering (CARS) and Coherent Stokes Raman Scattering (CSRS)

- Coherent Anti-Stokes Raman Scattering (CARS) and Coherent Stokes Raman Scattering (CSRS) are also established methods for amplifying Raman scattering.
- These techniques involve two laser frequencies for excitation

$$\chi_{CARS}^{(3)} (\omega_A = 2\omega_1 - \omega_2; \omega_1, \omega_1, -\omega_2)$$

$$\chi_{CSRS}^{(3)} (\omega_S = 2\omega_2 - \omega_1; \omega_2, \omega_2, -\omega_1)$$

CARS is a powerful method currently used in microscopy → we will hear about it during student presentations.

## 8.4. Solving the Nonlinear Wave Equation.

### 8.4.1. Nonlinear Helmholtz Equation

$$\nabla \times \mathbf{E}(\mathbf{r}, t) = -\frac{\partial \mathbf{B}(\mathbf{r}, t)}{\partial t}$$

$$\nabla \times \mathbf{H}(\mathbf{r}, t) = \frac{\partial \mathbf{D}(\mathbf{r}, t)}{\partial t} + \mathbf{j}(\mathbf{r}, t) \quad \rho = 0$$

$$\nabla \cdot \mathbf{D}(\mathbf{r}, t) = \rho \quad j = 0.$$

$$\nabla \cdot \mathbf{B}(\mathbf{r}, t) = 0.$$

follow the standard procedure of eliminating  $\mathbf{B}$  and  $\mathbf{H}$  from the equations

$$\nabla \times \nabla \times \mathbf{E}(\mathbf{r}, t) + \mu_0 \frac{\partial^2 \mathbf{D}(\mathbf{r}, t)}{\partial t^2} = 0,$$

$$\mathbf{D}(\mathbf{r}, t) = \epsilon_0 \mathbf{E}(\mathbf{r}, t) + \mathbf{P}(\mathbf{r}, t)$$

$$= \epsilon_0 \mathbf{E}(\mathbf{r}, t) + \mathbf{P}_L(\mathbf{r}, t) + \mathbf{P}_{NL}(\mathbf{r}, t)$$

$$= \epsilon_0 \epsilon_r \mathbf{E}(\mathbf{r}, t) + \mathbf{P}_{NL}(\mathbf{r}, t)$$

$$\nabla \times \nabla \times \mathbf{E}(\mathbf{r}, t) = \nabla(\nabla \cdot \mathbf{E}) - \nabla^2 \mathbf{E}$$

$$\simeq -\nabla^2 \mathbf{E}.$$

$$\nabla^2 \mathbf{E}(\mathbf{r}, t) - \frac{n^2}{c^2} \frac{\partial^2 \mathbf{E}(\mathbf{r}, t)}{\partial t^2} = \mu_0 \frac{\partial^2 \mathbf{P}_{NL}(\mathbf{r}, t)}{\partial t^2},$$

Nonlinear wave equation after approximation,

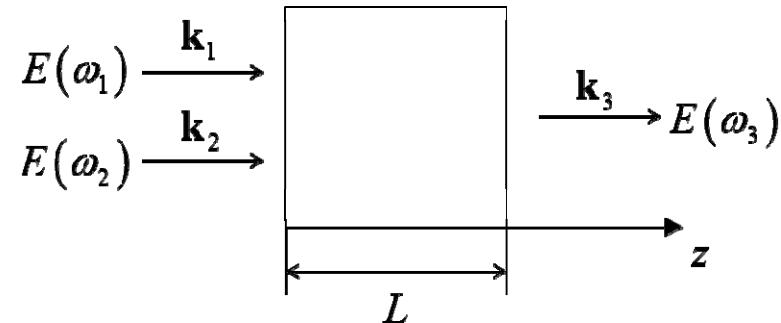
$$\nabla \cdot \mathbf{D} = \nabla (\varepsilon \mathbf{E}) \quad \text{term negligible}$$

$$\nabla^2 \mathbf{E}(\mathbf{r}, \omega) + \beta^2 \mathbf{E}(\mathbf{r}, \omega) = -\frac{1}{\varepsilon_0} \beta_0^2 \mathbf{P}_{NL}(\mathbf{r}, \omega),$$

## Propagation of the Sum Frequency Field

SFG nonlinear polarization has the form

$$P_{NL}^{(2)}(\mathbf{r}; \omega_3 = \omega_1 + \omega_2; \omega_1, \omega_2) = \epsilon_0 \chi^{(2)}(\omega_3; \omega_1, \omega_2) \cdot E_1(\mathbf{r}, t) E_2(\mathbf{r}, t)$$



$$E_1(\mathbf{r}, t) = A_1(\mathbf{r}) e^{-i(\omega_1 t - \beta_1 z)} + c.c.$$

$$E_2(\mathbf{r}, t) = A_2(\mathbf{r}) e^{-i(\omega_2 t - \beta_2 z)} + c.c. \quad \nabla^2 E_3(\mathbf{r}, t) + n^2(\omega_3) \frac{\omega_3^2}{c^2} E_3(\mathbf{r}, t) = -\mu_0 \omega_3^2 \epsilon_0 \chi^{(2)} E_1(\mathbf{r}, t) E_2(\mathbf{r}, t)$$

$$E_3(\mathbf{r}, t) = A_3(\mathbf{r}) e^{-i(\omega_3 t - \beta_3 z)} + c.c.$$

$$\beta_1 = n(\omega_1) \frac{\omega_1}{c}$$

$$\partial E_3 / \partial x = \partial E_3 / \partial y = 0$$

$$\nabla^2 E_3(\mathbf{r}, t) = \frac{d^2}{dt^2} \left[ A_3^{(z)} e^{-i(\omega_3 t - \beta_3 z)} \right]$$

$$= \left( \frac{d^2}{dz^2} A_3^{(z)} + 2i\beta_3 \frac{dA_3}{dz} - \beta_3^2 A_3 \right) e^{-i(\omega_3 t - \beta_3 z)}.$$

$$\frac{d^2 A_3(t)}{dt^2} + 2i\beta_3 \frac{dA_3(z)}{dz} = -\mu_0 \omega_3^2 \epsilon_0 \chi^{(2)} A_1 A_2 e^{i(\beta_1 + \beta_2 - \beta_3)z}.$$

Simplifying approximation,  $A_1$  and  $A_2$ , do not change with z (do not *deplete*).

$$\frac{d^2 A_3(t)}{dt^2} + 2i\beta_3 \frac{dA_3(t)}{dt} = -Be^{i\Delta kz}$$

$$B = \beta_3^2 \chi^{(2)} A_1 A_2$$

$$\Delta k = \beta_1 + \beta_2 - \beta_3,$$

amplitude is *slowly varying*

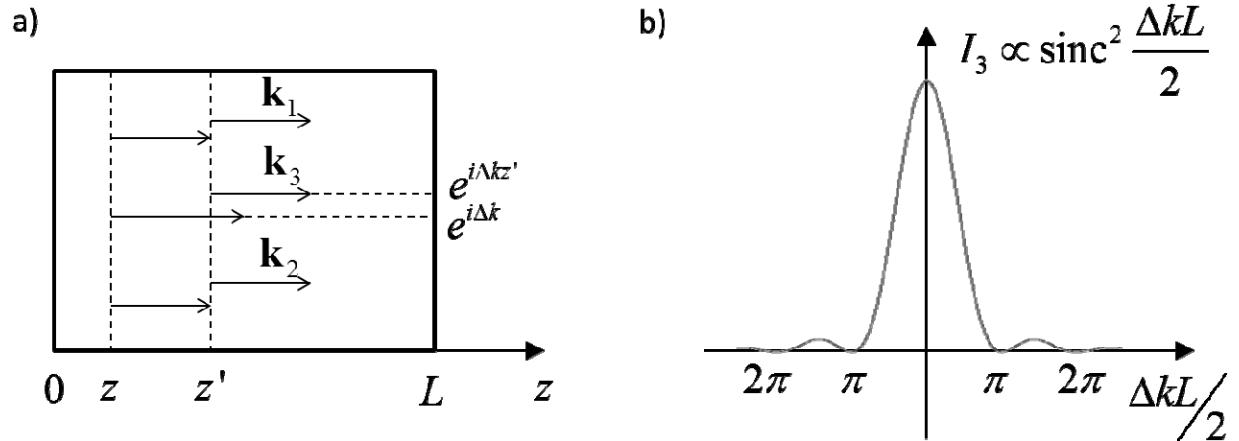
$$\left| \frac{d^2 A_3(z)}{dz^2} \right|^2 \ll \left| \beta_3 \frac{dA_3(t)}{dz} \right|. \quad dA_3(t) = \frac{i\beta}{2\beta_3} e^{i\Delta kz} dz$$

$$\begin{aligned}
A_3(L) &= \frac{iB}{2\beta_3} \int_0^L e^{i\Delta kz} dz \\
&= \frac{iB}{2\beta_3} \left( \frac{e^{i\Delta k L} - 1}{i\Delta k} \right) \\
&= \frac{iB}{2\beta_3} e^{\frac{i\Delta k L}{2}} \left( \frac{2 \sin \frac{\Delta k L}{2}}{\Delta k} \right) \\
&= \frac{iBL}{2\beta_3} e^{\frac{i\Delta k L}{2}} \left( \frac{\sin \frac{\Delta k L}{2}}{\frac{\Delta k L}{2}} \right) \\
&= \frac{iB}{2\beta_3} e^{\frac{i\Delta k L}{2}} \text{sinc}\left(\frac{\Delta k L}{2}\right).
\end{aligned}$$

The intensity of SFG field is

$$\begin{aligned}
I_3(z) &= \frac{1}{2\zeta} |A_3(z)|^2 \quad \zeta = \sqrt{\mu/\epsilon} = \zeta_0/n \\
&= \frac{B^2 L^2}{8\zeta \beta_3^2} \text{sinc}^2\left(\frac{\Delta k L}{2}\right),
\end{aligned}$$

Net output power can vanish at  $\frac{\Delta k L}{2} = \pi, 2\pi, \dots$



- a) The SFG output field has a phase that depends on the position where the conversion took place.
- b) The overall SFG intensity oscillates with respect to  $\Delta k L/2$ .

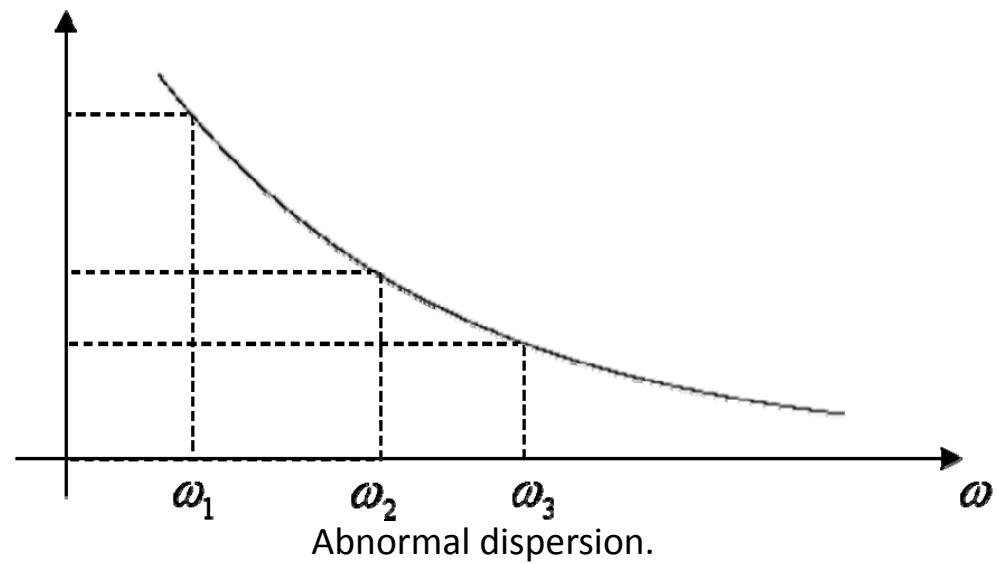
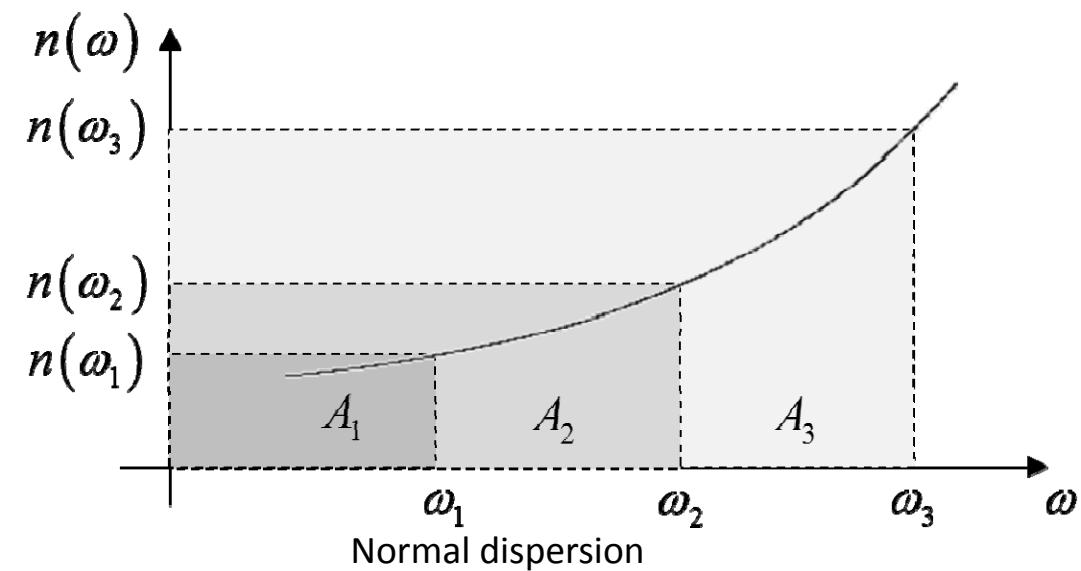
## Parametric Processes: Phase Matching

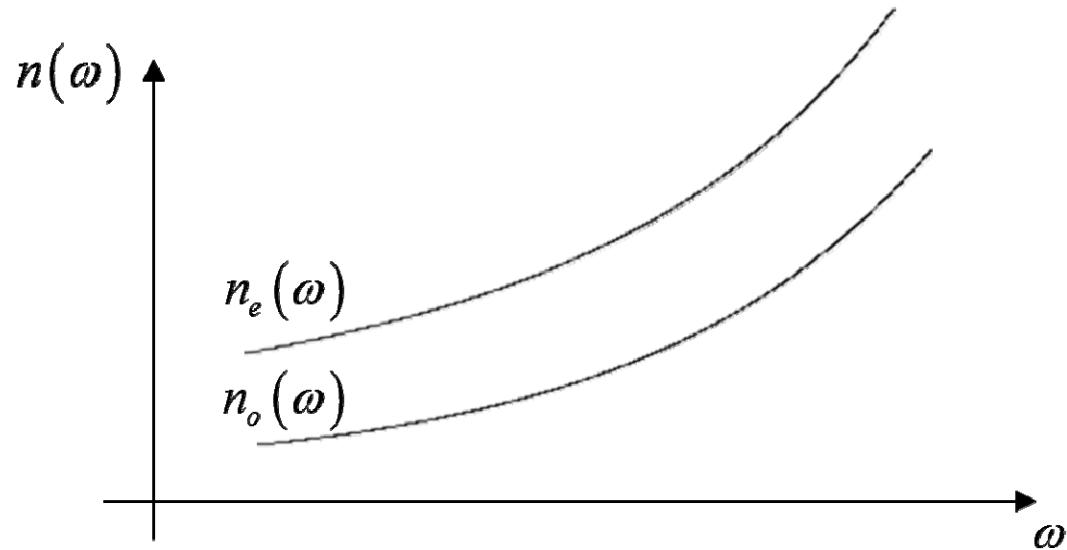
$$\Delta k = 0$$

$$\beta_3 = \beta_1 + \beta_2$$

$$n(\omega_3) \frac{\omega_3}{c} = \frac{n(\omega_1)\omega_1}{c} + n(\omega_2) \frac{\omega_2}{c}, \quad \omega_3 = \omega_1 + \omega_2$$

$$n(\omega_3) = \frac{n(\omega_1)\omega_1 + n(\omega_2)\omega_2}{\omega_1 + \omega_2}.$$





Normal dispersion curves for a *positive* uniaxial crystal.

	Negative ( $n_e < n_o$ )	Positive $n_e > n_o$
Type I	$n_e(\omega_3)\omega_3 = n_o(\omega_1)\omega_1 + n_o(\omega_2)\omega_2$	$n_o(\omega_3)\omega_3 = n_e(\omega_1)\omega_1 + n_o(\omega_2)\omega_2$
Type II	$n_e(\omega_3)\omega_3 = n_e(\omega_1)\omega_1 + n_o(\omega_2)\omega_2$	$n_o(\omega_3)\omega_3 = n_o(\omega_1)\omega_1 + n_e(\omega_2)\omega_2$

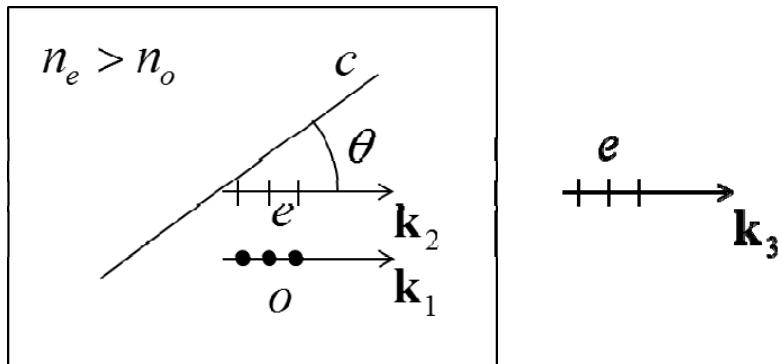
Table 8-I.

$$\frac{1}{n^2(\theta)} = \frac{\cos^2 \theta}{n_0^2} + \frac{\sin^2 \theta}{n_e^2}$$

phase matching can be achieved by *angle tuning*, that is, selecting the angle  $\theta$  that ensures  $\Delta k = 0$

Type I

$$n_o(\omega_3)\omega_3 = n(\omega_1, \theta)\omega_1 + n(\omega_2, \theta)\omega_2$$



Type II

$$n_o(\omega_3)\omega_3 = n_o(\omega_1)\omega_1 + n(\omega_2, \theta)\omega_2$$

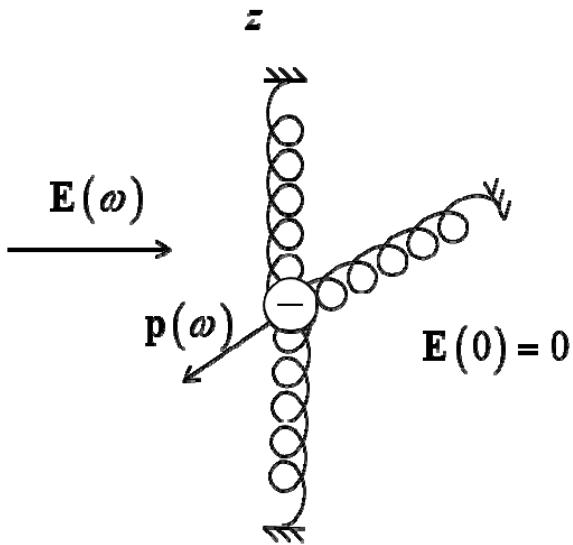
Type II phase matching by angle tuning in a positive uniaxial crystal: o-ordinary wave, e-extraordinary wave, c-optical axis.

## Electro-Optic Effect

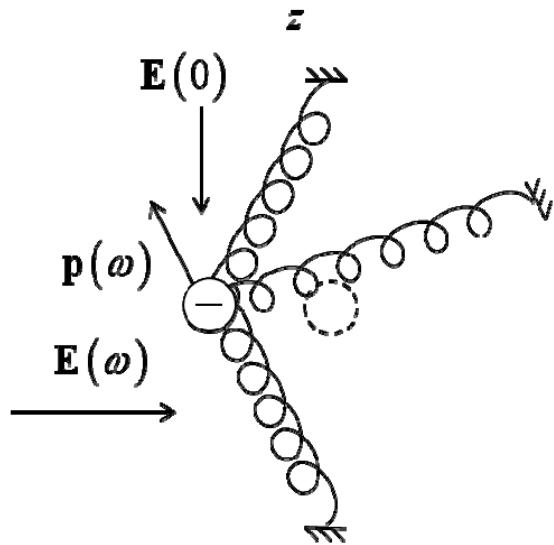
The *electro-optic effect* is the change in optical properties of a material due to an applied electric field that oscillates at much lower frequencies than the optical frequency.

## Electro-Optic Tensor

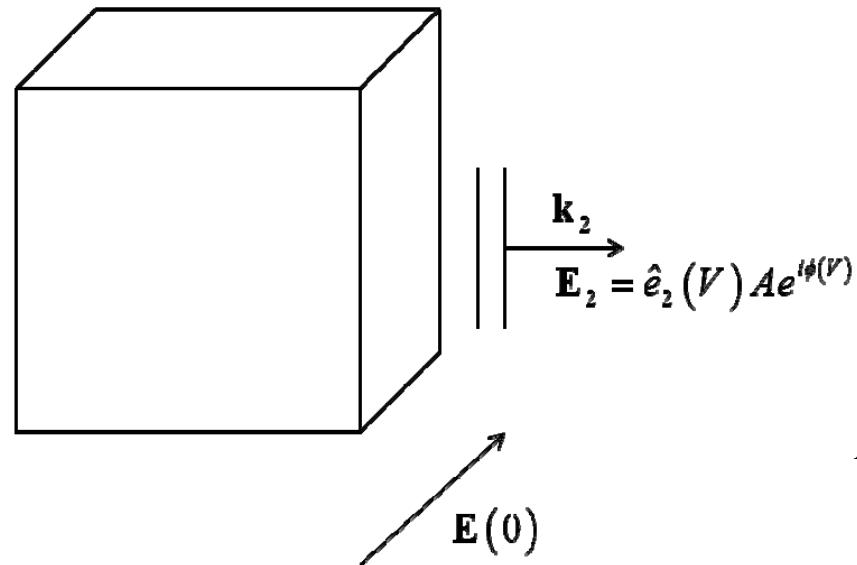
a)



b)



- a) Linear interaction with a birefringent crystal.
- b) Electro-optic (nonlinear) interaction.  $p$  is the induced dipole,  $E(\omega)$  is the optical field, and  $E(0)$  is the static field. These sketches should be interpreted as 3D representations.



The induced polarization for the Pockels effect

$$P_i(\omega; \omega, 0) = \epsilon_0 \chi_{ijk}^{(2)}(\omega; \omega, 0) E_j(\omega) E_k(0).$$

Kerr effect

$$P_i(\omega; \omega, 0, 0) = \epsilon_0 \chi_{ijkl}^{(3)}(\omega; \omega, 0, 0) E_j(\omega) E_k(\omega) E_l(\omega),$$

Due to the electro-optic effect, an optical field can suffer  
*voltage-dependent* polarization and phase changes.

Pockles effect

$$P_i^{(2)}(\omega; \omega, 0) = -\frac{1}{\epsilon_0} \epsilon_{ii} \epsilon_{jj} r_{ijk} E_j(\omega) E_k(0).$$

$$r_{ijk} = \frac{-\epsilon_0^2 \chi_{ijk}}{\epsilon_{ii} \epsilon_{jj}} = \frac{-\chi_{ijk}}{n_i^2 n_j^2}$$

$$r_{ijk} = r_{jik}$$

Change in the rank of the tensor, from 3 to 2

$$r_{11k} = r_{1k}$$

$$r_{22k} = r_{2k}$$

$$r_{33k} = r_{3k}$$

$$r_{12k} = r_{21k} = r_{6k}$$

$$r_{13k} = r_{31k} = r_{5k}$$

$$r_{23k} = r_{32k} = r_{4k}.$$

Electro-optic tensor can be represented by a 3x6 matrix. This tensor contraction, allowed by the permutation symmetry, reduces the number of independent elements from

$$3^2 = 27 \text{ to } 3 \times 6 = 18.$$

## Electro-Optic Effect in Uniaxial Crystals

Use KDP ( $\text{KH}_2\text{PO}_4$ , or potassium dihydrogen phosphate) as a specific example of uniaxial crystal

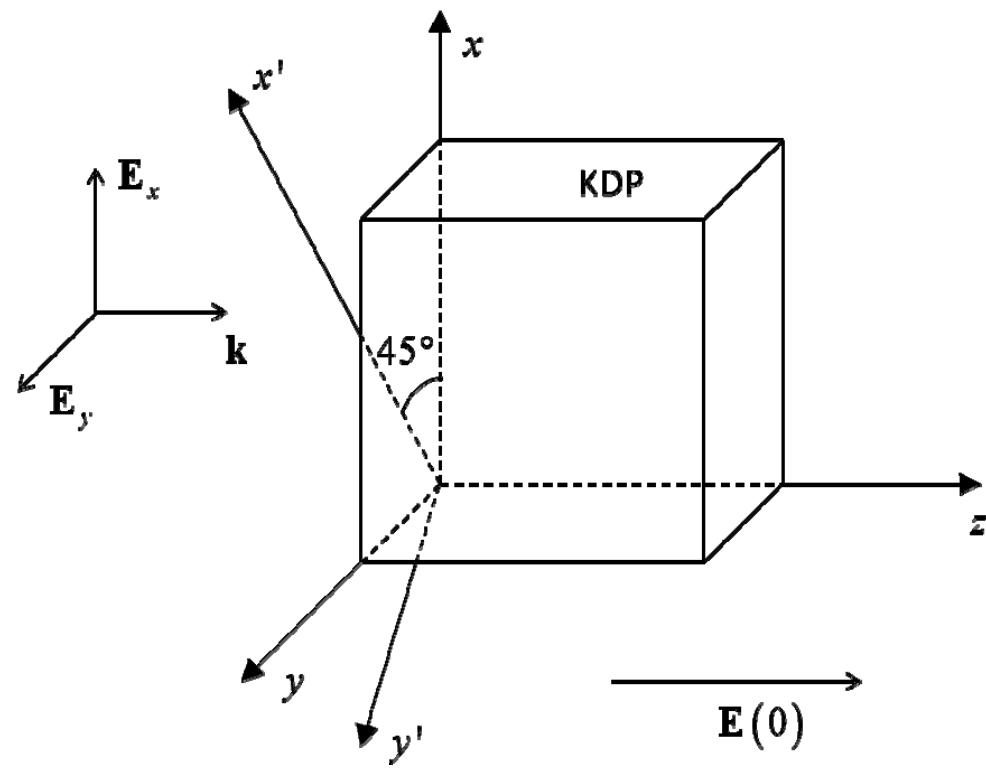
$$r = \begin{pmatrix} 0 & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & 0 \\ r_{41} & 0 & 0 \\ 0 & r_{41} & 0 \\ 0 & 0 & r_{63} \end{pmatrix}$$

The KDP refractive index tensor is (in the normal coordinate system of interest)

$$n = \begin{pmatrix} n_o & 0 & 0 \\ 0 & n_o & 0 \\ 0 & 0 & n_e \end{pmatrix}$$

assume that the voltage is applied only along z, such that only  $r_{63}$  is relevant.

$$\mathbf{D} = \epsilon_0 \overline{n}^2 \mathbf{E} + \mathbf{P}$$



Electro-optic effect in a KDP crystal. For  $\mathbf{E}(0)$  parallel to  $z$ , the normal axes rotate by 45 degrees around the  $z$ -axis.

$$D_i = \epsilon_0 n_i^2 E_i - \epsilon_0 n_i^2 n_j^2 r_{ijk} E_j(\omega) E_k(0) \\ = \epsilon_{ij} E_j(\omega),$$

electric displacement can be expressed for each component as  
(i=x, j=y, k=z)

$$D_x = \epsilon_0 n_o^2 E_x(\omega) - \epsilon_0 n_o^4 r_{63} E_y(\omega) E_z(0)$$

$$D_y = \epsilon_0 n_o^2 E_y(\omega) - \epsilon_0 n_o^4 r_{63} E_x(\omega) E_z(0)$$

$$D_z = \epsilon_0 n_e^2 E_z(\omega).$$

$$\overline{\epsilon} = \epsilon_0 \begin{pmatrix} n_o^2 & -n_o^4 r_{63} E_z(0) & 0 \\ -n_o^4 r_{63} E_z(0) & n_o^2 & 0 \\ 0 & 0 & n_e^2 \end{pmatrix}.$$

express  $\varepsilon_{ij}$  in a reference system rotated about the z-axis by an arbitrary angle, say  $\theta$

$$\bar{\bar{\varepsilon}}'(\theta) = R(-\theta) \bar{\bar{\varepsilon}} R(\theta)$$

$$= \varepsilon_0 \begin{pmatrix} \cos \theta & -\sin \theta \\ \sin \theta & \cos \theta \end{pmatrix} \begin{pmatrix} n_o^2 & -D \\ -D & n_o^2 \end{pmatrix} \begin{pmatrix} \cos \theta & \sin \theta \\ -\sin \theta & \cos \theta \end{pmatrix}$$

$$= \varepsilon_0 \begin{pmatrix} n_o^2 + D \sin 2\theta & -D \cos 2\theta \\ -D \cos 2\theta & n_o^2 - D \sin 2\theta \end{pmatrix}, \quad D = n_o^4 r_{63} E_z(0)$$

$$\bar{\bar{\varepsilon}}' = \varepsilon_0 \begin{pmatrix} n_o^2 + D & 0 & 0 \\ 0 & n_o^2 - D & 0 \\ 0 & 0 & n_e^2 \end{pmatrix}.$$

## Electro-Optic Modulators (EOMs)

defining the normal refractive indices

$$n_{ij}' = \sqrt{\epsilon_{ij}'}$$

$$\bar{n}' = \begin{pmatrix} n'_x & 0 & 0 \\ 0 & n'_y & 0 \\ 0 & 0 & n'_z \end{pmatrix}$$

$$n'_x = \sqrt{n_o^2 + D}$$

$$\approx n_o + \frac{1}{2} \frac{D}{n_o}$$

$$= n_o + \frac{1}{2} n_o^3 r_{63} E_z(0)$$

$$n'_y \approx n_o - \frac{1}{2} n_o^3 r_{63} E_z(0)$$

$$n'_z = n_e.$$

phase retardation

$$\Gamma = (n'_x - n'_y) \frac{\omega}{c} L$$

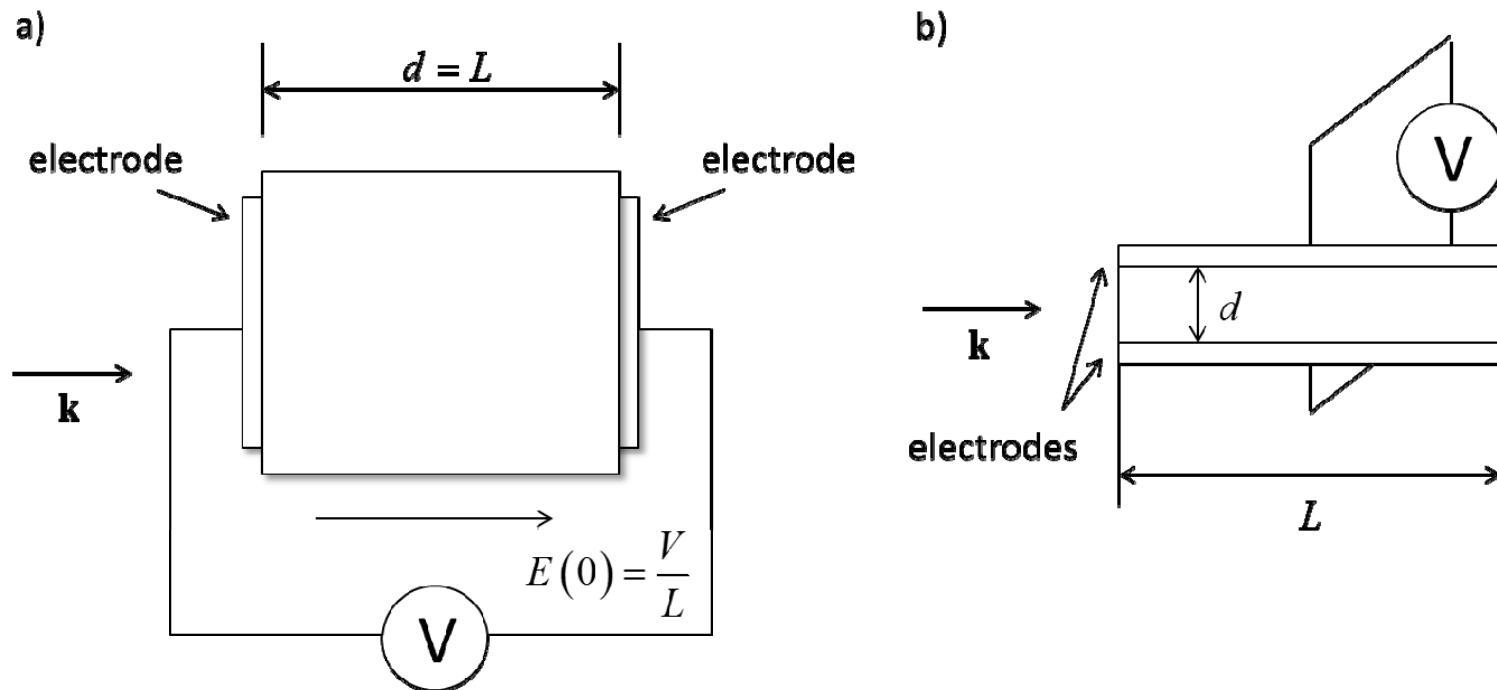
$$= \frac{2\pi}{\lambda_0} n_o^3 r_{63} E_z(0) L,$$

$$E_z(0) = \frac{V}{d}.$$

half-wave voltage  $\Gamma = \pi$

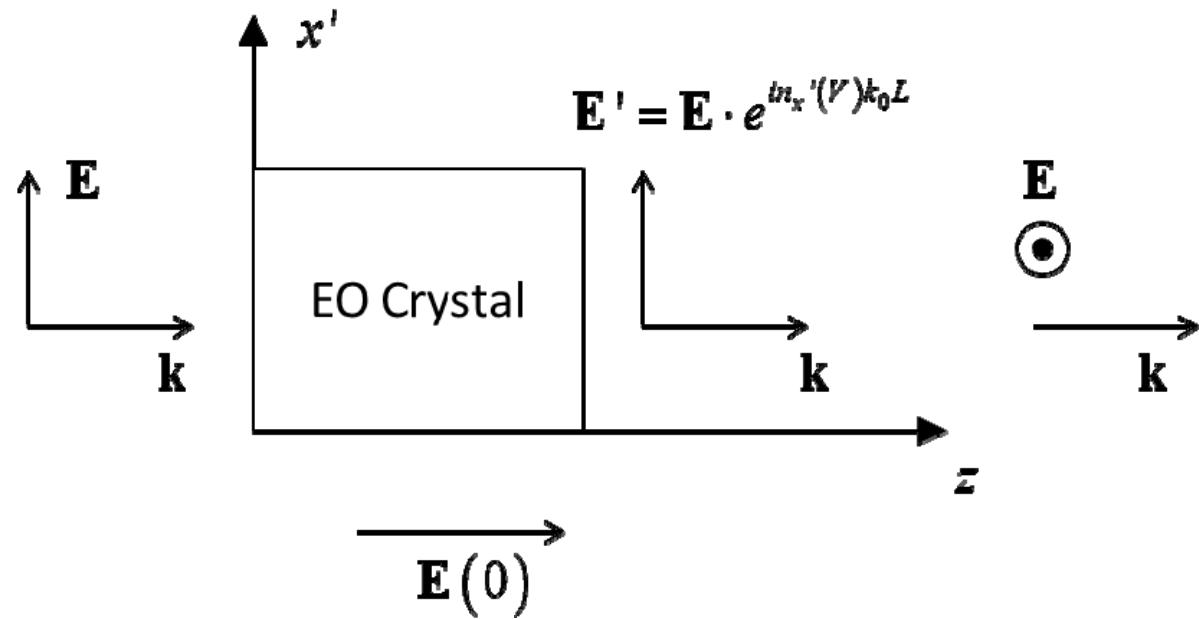
$$V_\pi = \frac{\lambda_0}{2n_o^3 r_{63}} \cdot \frac{d}{L}.$$

$$\Gamma(V) = \pi \frac{V}{V_\pi}.$$

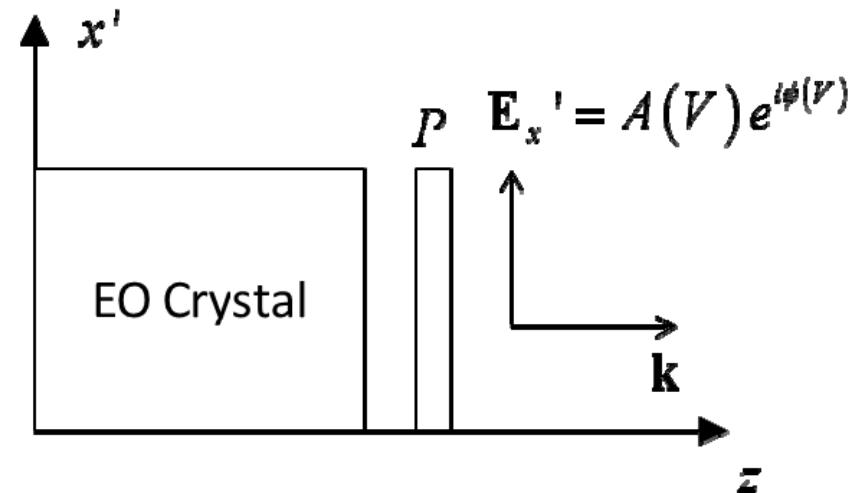


a) Longitudinal modulators (electrodes are made of transparent material),  $d=L$ . b) Transverse modulator,  $d < L$ .

a)



b)



a) Electro-optic (EO) crystal operating as phase modulator: incident polarization parallel to the new normal axis. b) Amplitude modulation: input polarization parallel to original (i.e. when  $V=0$ ) normal axis;  $P$  is a polarizer with its axis parallel to  $x$ .

KDP *longitudinal* modulator reads

$$\begin{aligned} E'(V) &= E e^{in_o k_0 L} \cdot e^{\frac{1}{2} k_0 n_o^3 r_{63} V} \\ &= E e^{in_o k_0 L} \cdot e^{\frac{i\Gamma(V)}{2}}. \end{aligned}$$

$(x',y')$  is rotated by  $45^\circ$  with respect to  $(x,y)$

$$\begin{pmatrix} E_x' \\ E_y' \end{pmatrix} = \bar{R}(-45^\circ) \cdot \bar{W}_0(V) \cdot \bar{R}(45^\circ) \begin{pmatrix} 0 \\ 1 \end{pmatrix}$$

$$W_0(V) = \begin{pmatrix} e^{\frac{i\Gamma}{2}} & 0 \\ 0 & e^{\frac{-i\Gamma}{2}} \end{pmatrix}$$

$$R(45^\circ) = \begin{pmatrix} \frac{\sqrt{2}}{2} & \frac{\sqrt{2}}{2} \\ -\frac{\sqrt{2}}{2} & \frac{\sqrt{2}}{2} \end{pmatrix}.$$

$$E_x' = -i \sin \left[ \frac{\Gamma(V)}{2} \right]$$

$$\begin{aligned} I_x' &= |E_x'|^2 \\ &= \sin^2 \left[ \frac{\Gamma(V)}{2} \right], \end{aligned}$$

voltage is modulated sinusoidally, at frequencies,  $\Omega$ , much lower than the optical frequency,

$$V(t) = V_0 \sin \Omega t$$

$$\Gamma(t) = \pi \frac{V_0}{V_\pi} \cdot \sin \Omega t.$$

phase and intensity modulations

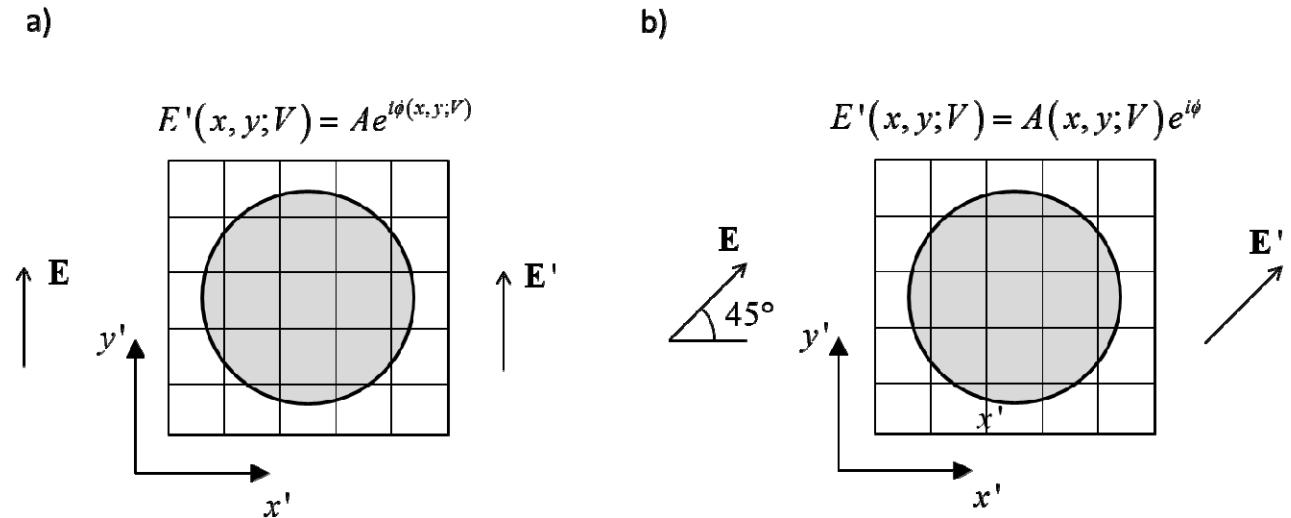
$$\phi_x'(t) = \frac{1}{2}\pi \frac{V_0}{V_\pi} \sin \Omega t$$

$$I_x'(t) = \sin^2 \left[ \frac{1}{2}\pi \frac{V_0}{V_\pi} \sin \Omega t \right].$$

$$\begin{aligned} I_x'(t) &= \frac{1}{2} \left( 1 - \cos \left[ \frac{\pi}{2} + \pi \frac{V_0}{V_\pi} \sin \Omega t \right] \right) \\ &= \frac{1}{2} \left[ 1 + \sin \left( \pi \frac{V_0}{V_\pi} \sin \Omega t \right) \right] \\ &\approx \frac{1}{2} + \frac{\pi}{2} \frac{V_0}{V_\pi} \sin \Omega t. \end{aligned}$$

phase modulators also modulate the frequency of the light

$$\begin{aligned} \Omega'(t) &= \frac{d\phi_x'(t)}{dt} \\ &= \frac{1}{2} \frac{\pi}{V_\pi} \cdot V(t). \end{aligned}$$

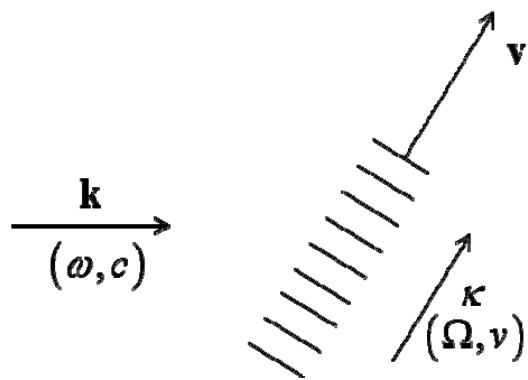


Liquid crystal spatial light modulator: a) phase mode; b) amplitude mode.

## Acousto-Optic Effect

The *acousto-optic effect* is the change in optical properties of a material due to the presence of an acoustic wave

## Elasto-Optic Tensor



the medium acts as a (phase) grating, which is capable of diffracting the light

$$\Lambda = \frac{2\pi}{k} = \frac{\Omega}{v},$$

Light wave (wavevector  $k$ , frequency  $\omega$ , speed  $c$ ) interacts with a travelling grating induced by a sound wave (wavevector  $\kappa$ , frequency  $\Omega$ , speed  $v$ ).

induced polarization in the normal coordinate system

$$P_i^{(3)}(\omega) = -\epsilon_0 \sum_{j,k,l=1}^3 n_i^2 n_j^2 P_{ijkl} S_{kl} E_j(\omega).$$

strain tensor

$$S_{kl} = \frac{1}{2} \left( \frac{\partial u_k}{\partial x_l} + \frac{\partial u_l}{\partial x_k} \right).$$

Kerr *electro-optic* effect, induced polarization

$$P_i^{(3)}(\omega) = -\epsilon_0 \sum_{ijkl=1}^3 n_i^2 n_j^2 S_{ijkl} E_k(0) E_l(0) E_j(\omega),$$

$$P_{ijkl} S_{kl} = S_{ijkl} E_k(0) E_l(0).$$

electro-optic tensor, due to symmetry, the elasto-optic tensor can also be used using the following (Voigt) *contraction*

$$P_{ijkl} = P_{mn}, \quad i, j, k, l = 1, 2, 3; \quad m, n = 1, 2, \dots, 6$$

$$ij = 11, \quad kl = 11; \quad m = 1, \quad n = 1$$

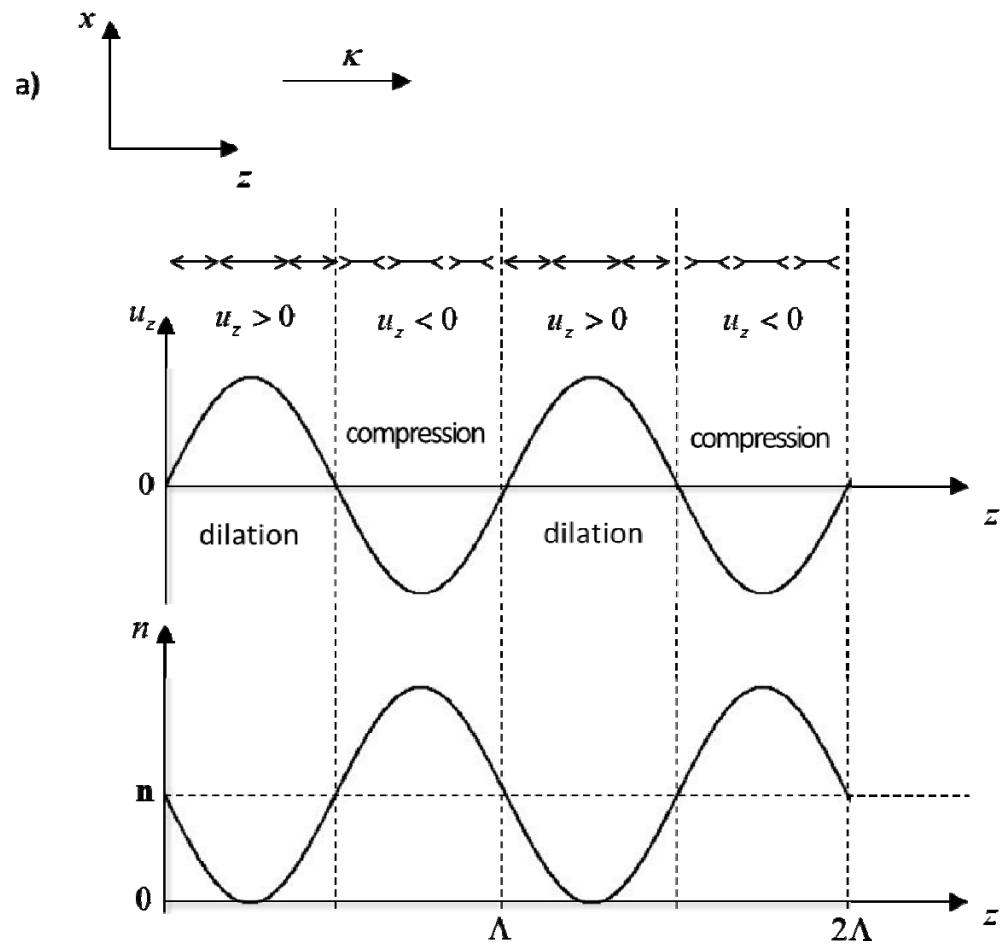
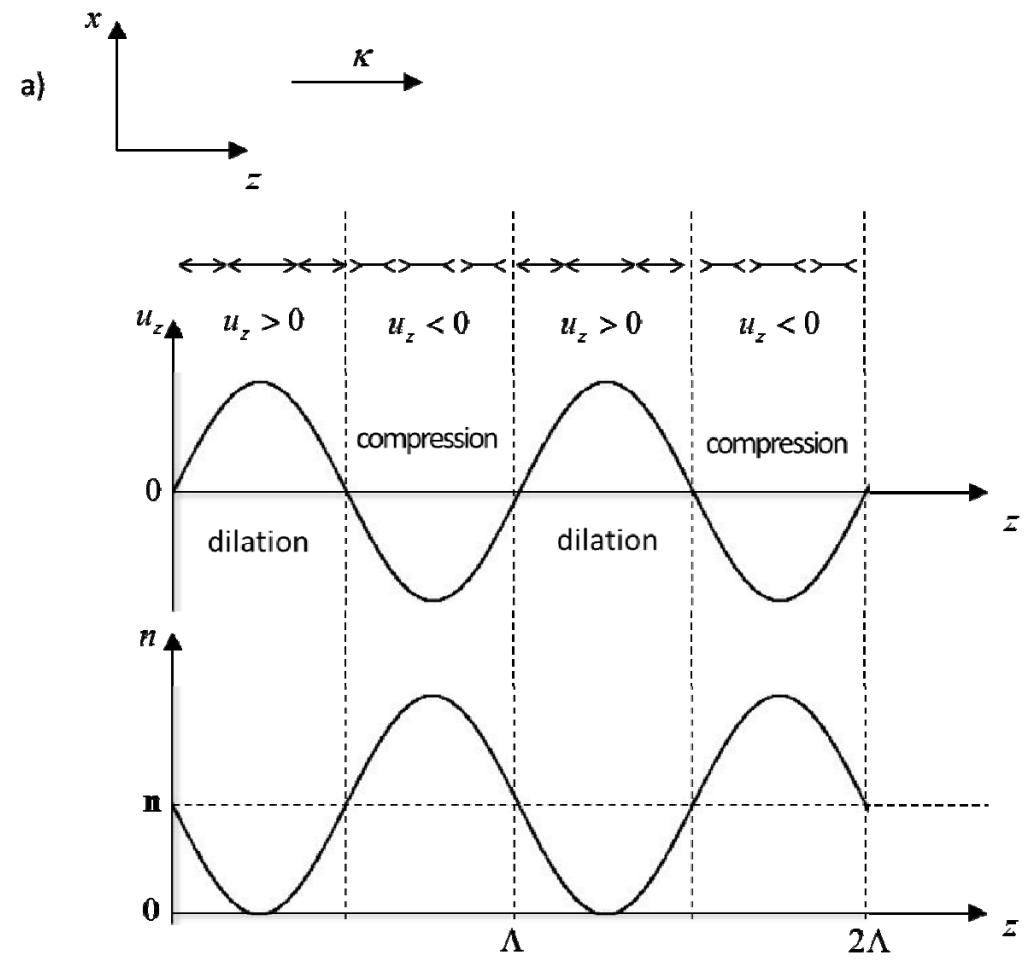
$$ij = 22, \quad kl = 22; \quad m = 2, \quad n = 2$$

$$ij = 33, \quad kl = 33; \quad m = 3, \quad n = 3$$

$$ij = 12, \quad kl = 12; \quad m = 6, \quad n = 6$$

$$ij = 13, \quad kl = 13; \quad m = 5, \quad n = 5$$

$$ij = 23, \quad kl = 23; \quad m = 4, \quad n = 4.$$



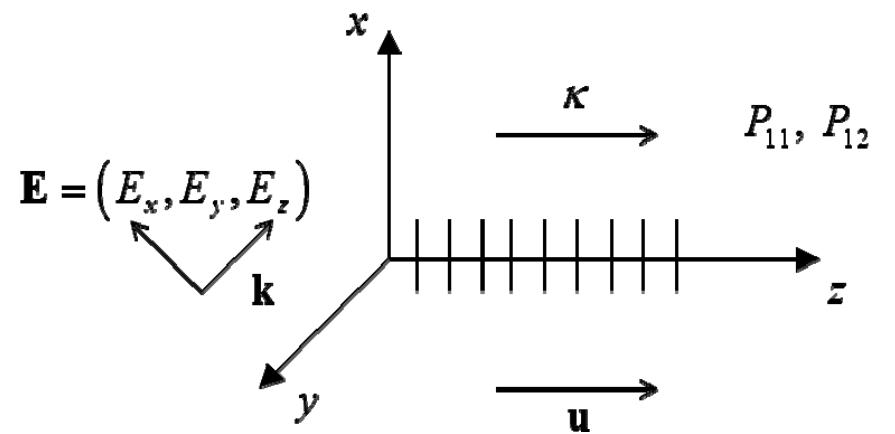
Photoelastic effect: the sound wave induces strain, which in turn modulates the refractive index. a) *Longitudinal* sound wave; b) *Transverse (shear)* sound wave. Lambda is the sound wavelength, with Omega the frequency and v the propagation speed.

## Acousto-Optic Effect in Isotropic Media

Let us investigate in more detail the acousto-optic effect in *isotropic media*

elasto-optic tensor for isotropic media

$$p_{mn} = \begin{pmatrix} p_{11}' & p_{12}' & p_{12}' & 0 & 0 & 0 \\ p_{12}' & p_{11}' & p_{12}' & 0 & 0 & 0 \\ p_{12}' & p_{12}' & p_{11}' & 0 & 0 & 0 \\ 0 & 0 & 0 & \frac{1}{2}(p_{11}' - p_{12}') & 0 & 0 \\ 0 & 0 & 0 & 0 & \frac{1}{2}(p_{11}' - p_{12}') & 0 \\ 0 & 0 & 0 & 0 & 0 & \frac{1}{2}(p_{11}' - p_{12}') \end{pmatrix}$$



$$S_{kl} = S_{33} = S_3$$

expressions for the polarization

$$P_x^{(3)}(\omega) = -\epsilon_0 n^4 S_3 [p_{13} E_x(\omega) + p_{63} E_y(\omega) + p_{53} E_z(\omega)]$$

$$P_y^{(3)}(\omega) = -\epsilon_0 n^4 S_3 [p_{63} E_x(\omega) + p_{23} E_y(\omega) + p_{43} E_z(\omega)]$$

$$P_z^{(3)}(\omega) = -\epsilon_0 n^4 S_3 [p_{53} E_x(\omega) + p_{43} E_y(\omega) + p_{33} E_z(\omega)].$$

Dielectric displacement

$$\mathbf{D}(\omega) = \epsilon_0 n^2 \mathbf{E}(\omega) + P^{(3)}(\omega) \\ = \bar{\epsilon} \mathbf{E}(\omega),$$

$$p_{43} = p_{53} = p_{63} = 0$$

$$P_x^{(3)}(\omega) = -\epsilon_0 n^4 S_3 p_{13} E_x(\omega)$$

$$P_y^{(3)}(\omega) = -\epsilon_0 n^4 S_3 p_{23} E_y(\omega)$$

$$P_z^{(3)}(\omega) = -\epsilon_0 n^4 S_3 p_{33} E_z(\omega).$$

dielectric tensor

$$= \bar{\epsilon} \epsilon_0 n^2 S_3 \begin{pmatrix} 1-n^2 & p_{12} & 0 & 0 \\ 0 & 1-n^2 & p_{12} & 0 \\ 0 & 0 & 1-n^2 & p_{11} \end{pmatrix}.$$

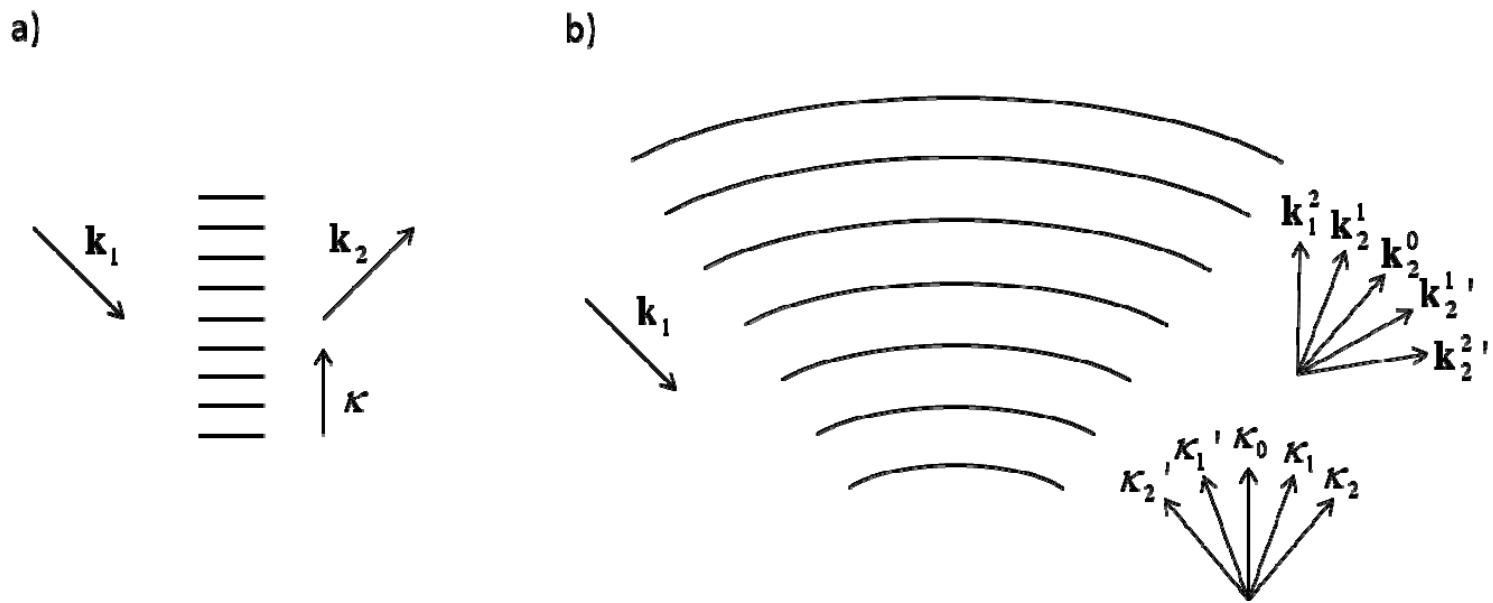
medium becomes *uniaxial*, i.e.  $\epsilon_{xx} = \epsilon_{yy} \neq \epsilon_{zz}$

$$S_3 = \frac{1}{2} \frac{\partial u_z}{\partial z}$$

$$= \frac{1}{2} i k A e^{-i(\Omega t - kz)}$$

plane acoustic wave of amplitude A

$$u_z = A e^{-i(\Omega t - kz)}.$$



a) Brogg regime; b) Raman-Nath regime.

## Bragg Diffraction Regime

wave equation for the total field (incident plus diffracted) reads

$$\nabla^2 E(\mathbf{r}, t) - \frac{n^2}{c^2} \frac{\partial E(\mathbf{r}, t)}{\partial t^2} = \mu_0 \frac{\partial^2 P^{(3)}(\mathbf{r}, t)}{\partial t^2}$$

$$P^{(3)}(\mathbf{r}, t) = -BA e^{-i(\Omega t - \mathbf{k} \cdot \mathbf{r})} E_1(\mathbf{r}, t)$$

$$E(\mathbf{r}, t) = E_1(\mathbf{r}, t) + E_2(\mathbf{r}, t),$$

Fourier transform

$$\nabla^2 E_2(\mathbf{r}, \omega) + n^2 \beta_0^2 E_2(\mathbf{r}, \omega) = F(\mathbf{r}, \omega) E_1(\mathbf{r}, \omega - \Omega)$$

$$F(\mathbf{r}, \omega) = \omega^2 B A e^{i\mathbf{k} \cdot \mathbf{r}}.$$

$$\nabla^2 E_1(\mathbf{r}, \omega) + n^2 \beta_0^2 E_1(\mathbf{r}, \omega) = 0$$

$$e^{-i\Omega t} E_1(\mathbf{r}, t) \leftrightarrow E_1(\mathbf{r}, \omega + \Omega).$$

$$\begin{aligned} E_2(\mathbf{q}, \omega) &\propto \int_V F(\mathbf{r}, \omega) e^{i\mathbf{qr}} d^3\mathbf{r} \\ &\propto A_l(\omega - \Omega) \cdot \partial(\mathbf{q} - \mathbf{k}) \\ &\propto \partial(\omega - \omega_l - \Omega) \cdot \delta(\mathbf{k} - \mathbf{k}_l - \mathbf{k}), \end{aligned}$$

$$A_l(\omega) \propto \delta(\omega - \omega_l)$$

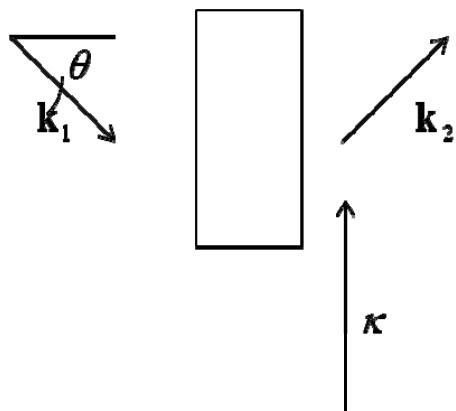
$$\mathbf{k}_2 = \mathbf{k}_1 + \mathbf{k}$$

$$\omega_2 = \omega_l + \Omega.$$

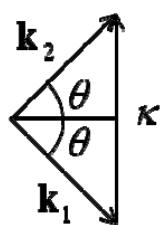
*Bragg condition*

$$\begin{aligned}k_2 &= \frac{2\pi}{\omega_l + \Omega} \\&\approx \frac{2\pi}{\omega_l} \\&= k_1.\end{aligned}$$

a)



b)



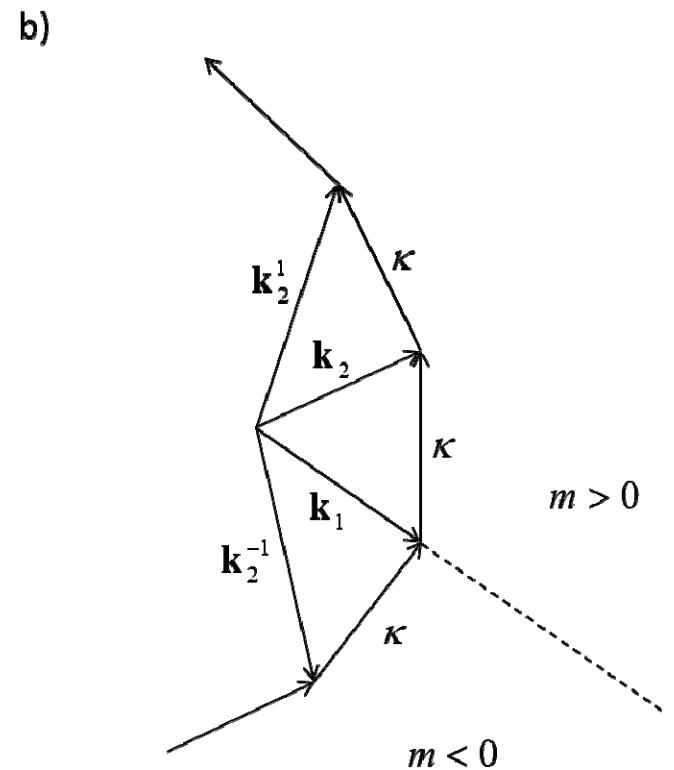
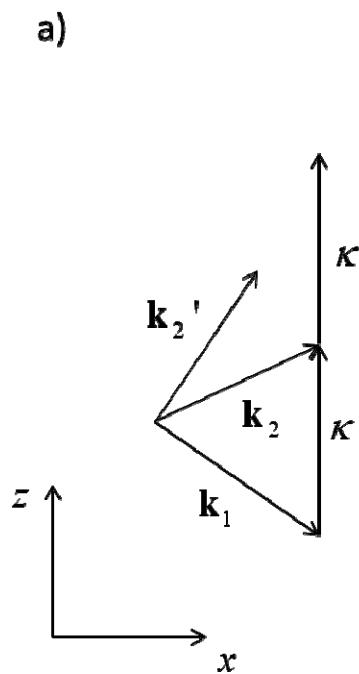
$$2k_1 \sin \theta = \kappa$$

$$\theta = \sin^{-1} \left[ \frac{\lambda}{2n\Lambda} \right]$$

The Bragg condition: a)  $k_1$ ,  $k_2$  and  $\kappa$  are, respectively, the incident, diffracted and acoustic wavevectors. b) Triangle that illustrates geometrically  $k_2 = k_1 + \kappa$ .

## Raman-Nath Diffraction Regime

- a) Bragg diffraction (see solution for  $k_2$ ).
- b) Raman-Nath diffraction when sound wave is curved (multiple solutions for  $k_2$ ).



scattering potential in the  $(x, z)$  coordinates     $F(x, z, \omega) = \omega^2 B A e^{i\kappa_0 z} e^{\frac{i\kappa_0 x^2}{2z}}$

diffracted field is the Fourier transform of F

$$E_2(q_x, q_z, \omega) = \delta \left[ q_z - \left( \kappa_0 - \frac{q_x^2}{2\kappa_0} \right) \right].$$

$\mathbf{q}$  is the scattering wavevector,  $\mathbf{q} = \mathbf{k}_2 - \mathbf{k}_1$ .

$$k_{2z} - k_{1z} - \kappa_0 + \frac{(k_{2x} - k_{1x})^2}{2\kappa_0}.$$

if the sound beam curvature satisfies

$$\frac{(k_{2x} - k_{1x})^2}{2\kappa_0} = m \cdot \kappa_0,$$

Where  $m=0,1,2,\dots$ , the diffracted beam has multiple solutions

$$k_{2z} - k_{1z} = \kappa_0 + m\kappa_0.$$

