Nonlinear Optics

Basic concept of optical nonlinearity

The polarization density in the presence of an electric field:
$P \propto E$

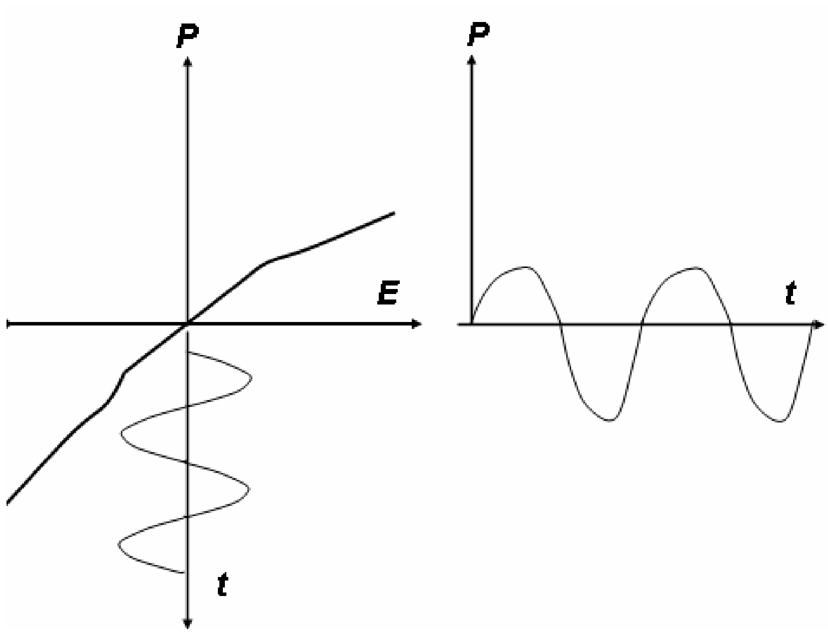
Here we assume that the medium is isotropic. When the field is strong, the displacement of electrons from their equilibrium point may deviate from linearity:

$$P = \varepsilon_0 \chi E + 2dE^2 + 4\chi^{(3)} E^3 \tag{1}$$

The coefficients describe the first order, second order and third order nonlinearity.

If the medium has an inversion symmetry, the second order nonlinearity, characterized by *d*, is zero. Why?

Physical pictures: When the potential as a function of displacement deviates from a perfect parabola, the electrons driven by an electric field may create a polarization. For example: a DC polarization can be created by an electric in an asymmetric potential. An asymmetric shape in the polarization-electric field relation results in a distorted polarization which, in turn, generates a wave which contains the harmonic waves whose frequencies are two times the fundamental.



The polarization created by the radiation then acts as a source for the generation of new waves.

$$P = \varepsilon_0 \chi E + 2dE^2 + 4\chi^{(3)} E^3$$

Second order nonlinearity

$$P^{(2)} = 2dE^2 = 2d(E_1 e^{j\omega_1 t} + E_2 e^{j\omega_2 t} + cc)^2$$

$\omega_1 = \omega_2$ ⇨
- Second harmonic generation
- Optical rectification
-

$\omega_1 = 0$ ⇨
- Electrooptic effect

$\omega_1 \neq \omega_2$ ⇨
- Frequency conversion
- Optical parametric process

$$P = \varepsilon_0 \chi E + 2dE^2 + 4\chi^{(3)} E^3$$

Third order nonlinearity

$$P^{(3)} = 4\chi(E_1 e^{j\omega_1 t} + E_2 e^{j\omega_2 t} + E_3 e^{j\omega_3 t} + cc)^3$$

$\omega_1 = \omega_2 = \omega_3$ ⇨
- Third harmonic generation
- Degenerate four-wave mixing
- Optical phase conjugation

$\omega_1 = \omega_2$ ⇨
- Optical Kerr effect
- Self-phase modulation
- Self- focusing

$\omega_1 \neq \omega_2 \neq \omega_3$ ⇨
- Raman gain
- Four-wave mixing

I

In general, the polarization density and electric field is related by a tensor. For example,

$$P_i = \varepsilon_0 \chi_{ij} E_j + 2d_{ijk} E_j E_k + 4\chi^{(3)}_{ijkl} E_j E_k E_l + ... \quad (1.1)$$

where each of the i,j,k,l are can be x, y, or z. The coordinate axis does not need to be one that diagonalizes the dielectric tensor.

See Yariv's.

Nonlinear coefficients- the anharmonic oscillator model

Consider the bound electron model with an electron in a anharmonic potential well driven by electromagnetic wave of frequency ω. The displacement of electron from equilibrium follows the following relations:

$$\ddot{x} + \gamma \dot{x} + \omega_0^2 + Dx^2 = \frac{eE}{2m}(e^{j\omega t} + cc) \qquad \textbf{(1.2)}$$

Assuming that the solution of this equation has the following form:

$$X = \frac{1}{2}(q_1 e^{j\omega t} + q_2 e^{j2\omega t} + q_3 e^{j3\omega t} + cc)$$

Solve for the q's

$$q_1 = \left(\frac{eE}{m}\right)\frac{1}{(\omega_0^2 - \omega^2) + j\omega\gamma} \qquad (1.3)$$

$$q_2 = \frac{-De^2E_0^2}{2m^2\left[(\omega_0^2 - \omega^2) + j\omega\gamma\right]^2(\omega_0^2 - \omega^2 + 2j\omega\gamma)} \qquad (1.4)$$

The nonlinear coefficients are larger in the vicinity of resonance lines..

Nonlinear wave equation

From Lecture 2 Eq. (7), the Helmholtz equation with a source is given by

$$\nabla^2 E - \frac{1}{c_0^2}\frac{\partial^2 E}{\partial t^2} = \mu_0 \frac{\partial^2 P}{\partial t^2} \qquad (2)$$

The polarization has a linear and nonlinear components.

$$P = \varepsilon_0 \chi E + P_{NL} \qquad (3)$$

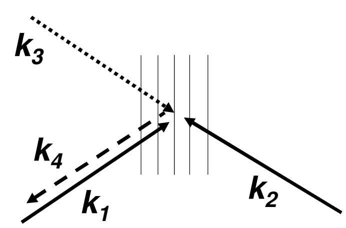
The linear part can be combined with the other time derivative term to give rise to a speed of light $c = c_0/n$. Eq. (2) can be rewritten as

$$\nabla^2 E - \frac{1}{c^2}\frac{\partial^2 E}{\partial t^2} = -S$$

$$S = -\mu_0 \frac{\partial^2 P_{NL}}{\partial t^2} \qquad (4)$$

Four-wave mixing :

beam deflection by an oscillating grating



The directions and frequencies of the $\boldsymbol{k}$'

Second order nonlinearity

Consider the case of two sinusoidal electric fields in a nonlinear medium

$$P^{(2)} = 2d(\left|E_{\omega_1}\right|^2 e^{j2\omega_1 t} + E_{\omega_2} e^{j\omega_2 t} + cc)^2$$

(5)

In general, the two electric fields can be in different direction and the polarization is related to the individual fields through a tensor. The mixing of the field results in four nonlinear polarization densities oscillating at $2\omega_1$, $2\omega_2$, $\omega_1+\omega_2$, and $\omega_1-\omega_2$, representing the sources for second harmonic generation of ω_1 and ω_2 , sum and difference frequency generation. Note the for second harmonic generation, only one wave is needed.

Pockels effect

A special case is when one of the waves is a DC electric field with $\omega_2=0$, the polarization density is given by

$$P^{(2)} = 2dE^2 = 2d\ E_{DC}E_{\omega_1}e^{j\omega_1 t} \tag{6}$$

This term contribute to a change in the refractive index

$$\Delta n = \frac{2d}{n\varepsilon_0}E_{DC} \tag{7}$$

The change of refractive index by applied voltage is the Pockels effect, which has wide applications in optics as optical shutter or modulators.

Frequency conversion

$$P^{(2)} = 2d(\left|E_{\omega_1}\right|^2 e^{j2\omega_1 t} + \left|E_{\omega_2}\right|^2 e^{j2\omega_2 t} + 2E_{\omega_1}E_{\omega_2}^* e^{j(\omega_1-\omega_2)t} + 2E_{\omega_1}E_{\omega_2}e^{j(\omega_1+\omega_2)t} \tag{8}$$

Thus the nonlinear medium can be used to mix two optical waves of difference frequencies and generate a third wave at the sum and difference frequencies. Although the waves produce polarization densities at various frequencies, not all of them are generated in a meaningful way unless the following conditions are simultaneously satisfied.

Frequency matching : $\omega_3 = \omega_1 + \omega_2$ (9)

Phase matching : $\vec{k}_3 = \vec{k}_1 + \vec{k}_2$ (10)

Physical meaning: Using propagation delay of SHG in one dimension as an example. The waves must have the same speed (phase velocity) to ensure constructive interference along the path.

Coupled-wave theory with second-order nonlinearity

$$\begin{aligned} &\nabla^2 E - \frac{1}{c^2}\frac{\partial^2 E}{\partial t^2} = -S \\ &S = -\mu_0 \frac{\partial^2 P_{NL}}{\partial t^2} = -2d\mu_0 \frac{\partial^2}{\partial t^2}E^2 \end{aligned} \tag{11}$$

From (4)
The field is a superposition of three waves

$$E = \sum_{q=1,2,3} \frac{1}{2}(E_q \exp(j\omega_q t) + cc) = \sum_{q=\pm1,\pm2,\pm3} \frac{1}{2} E_q \exp(j\omega_q t) \qquad (12)$$

where E_{-q} is defined to be E_q^*
By substitution the electric filed into (11), and carrying out the time derivative

$$S = \frac{1}{2}\mu_0 d \sum_{q,r=\pm1,\pm2,\pm3} (\omega_q + \omega_r)^2 E_q E_r \exp[(j\omega_q + \omega_r)t]$$

The Hemlholtz equations with sources are

$$(\nabla^2 + k_s^2)E_s = -S_s \quad s = 1,2,3 \qquad (13)$$

where S_s is the component of S with a frequency, and $k_s=n_s\omega/c$. Among the frequencies in the three equation, the interaction takes place only when

$$\omega_3 = \omega_1 + \omega_2 \qquad (14)$$

$$(\nabla^2 + k_1^2)E_1 = -2\mu_0\omega_1^2 dE_3 E_2^* \qquad (15\text{-}1)$$
$$(\nabla^2 + k_2^2)E_2 = -2\mu_0\omega_2^2 dE_3 E_1^* \qquad (15\text{-}2)$$
$$(\nabla^2 + k_3^2)E_3 = -2\mu_0\omega_3^2 dE_1 E_2 \qquad (15\text{-}3)$$

With this constraint,

This is the Helmholtz equations for three-wave mixing.

Special case: Collinear plane waves

In the normalized form

$$E_q = (2\pi\hbar\omega_q)^{1/2} a_q \exp(-jk_q z) \qquad q = 1,2,3 \qquad (16)$$

The coupled mode equations are, from (15) and (16) and using slow varying amplitude approximation.

$$\frac{da_1}{dz} = -jga_3 a_2^* \exp(-j\Delta kz) \qquad (17)$$
$$\frac{da_2}{dz} = -jga_3 a_1^* \exp(-j\Delta kz) \qquad (18)$$
$$\frac{da_3}{dz} = -jga_1 a_2^* \exp(j\Delta kz) \qquad (19)$$

where $g^2 = 2\hbar\omega_1\omega_2\omega_3\eta^3 d^2$ and $\eta = (\mu_0 / \varepsilon_0)^{1/2}$, and $\Delta k = k_3 - k_2 - k_1$ is the phase mismatch of the three waves.

For an input of two frequencies, the wave of the third frequency cannot build up unless the phase mismatch is zero. *Second harmonic generation*

Angle tuning

The electric displacement vector and the electric field are related through a dielectric tensor

$$D_k = \varepsilon_{kl} E_l \tag{1}$$

where k and l refer to the Cartesian coordinates (x,y,z). Using the principal axes as the coordinate system, the dielectric tensor is diagonal. The index ellipsoid is described by

$$\frac{x_1^2}{n_1^2} + \frac{x_2^2}{n_2^2} + \frac{x_3^3}{n_3^3} = 1 \tag{2}$$

where $1/n_1, 1/n_2, and\ 1/n_3$ are the principal values. The index ellipsoid of an isotropic medium is a sphere.

A wave propagating in the z-direction and linearly polarized in the x-direction travels with phase velocity c_0/n_1.

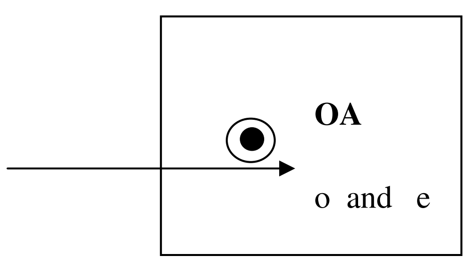
What about a wave traveling in the z-direction and an electric field on the z=0 plane and making a 45-degree angle with the x-axis?

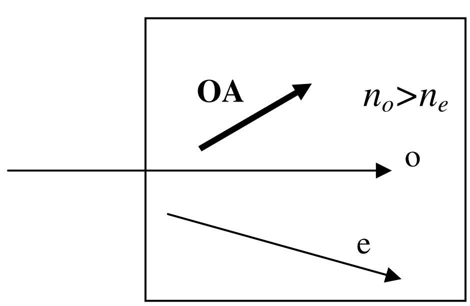
In an uniaxial crystal,

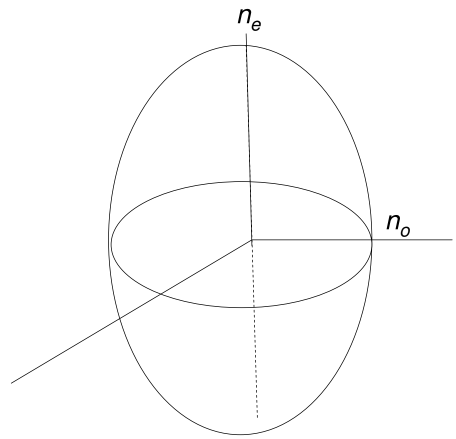
$$\frac{1}{n^2(\theta)} = \frac{\cos^2(\theta)}{n_0^2} + \frac{\sin^2\theta}{n_e^2}$$

The ray with index n_0 is the ordinary wave and $n(\theta)$ the extraordinary wave.

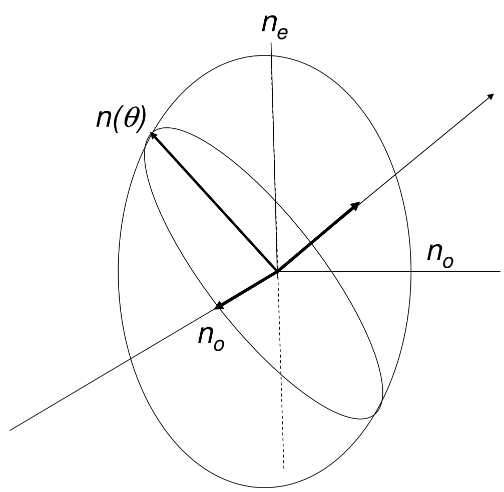
Walk-off







n_e
n_o



n_e
$n(\theta)$
n_o
n_o