Lecture 11: Introduction to nonlinear optics I.

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Formulation of the nonlinear optics: nonlinear polarization

Classification of the nonlinear phenomena

- Propagation of weak optic signals in strong quasi-static fields (description using renormalized linear parameters)
  - ☞ Linear electro-optic (Pockels) effect
  - ☞ Quadratic electro-optic (Kerr) effect
  - ☞ Linear magneto-optic (Faraday) effect
  - ☞ Quadratic magneto-optic (Cotton-Mouton) effect
- Propagation of strong optic signals (proper nonlinear effects) — next lecture

Nonlinear optics

Experimental effects like

- Wavelength transformation
- Induced birefringence in strong fields
- Dependence of the refractive index on the field intensity

etc.

lead to the concept of the nonlinear optics

The principle of superposition is no more valid

The spectral components of the electromagnetic field interact with each other through the nonlinear interaction with the matter

Nonlinear polarization

Taylor expansion of the polarization in strong fields:

$$P_i = \varepsilon_0 \chi_{ij} E_j + \chi^{(2)}_{ijk} E_j E_k + \chi^{(3)}_{ijkl} E_j E_k E_l + \ldots$$

$$\begin{aligned} P_i(t) = {} & \varepsilon_0 \int \tilde{\chi}_{ij}(t-t') E_j(t')\, dt' + \\ & + \iint \chi^{(2)}_{ijk}(t-t', t-t'') E_j(t') E_k(t'')\, dt' dt'' + \\ & + \iiint \chi^{(3)}_{ijkl}(t-t', t-t'', t-t''') E_j(t') E_k(t'') E_l(t''')\, dt' dt'' + \\ & + \ldots \end{aligned}$$

$$\begin{aligned} P_i(\omega) = {} & \varepsilon_0 \chi_{ij}(\omega) E_j(\omega) + \int d\omega_1 \underbrace{\chi^{(2)}_{ijk}(\omega; \omega_1, \omega_2) E_j(\omega_1) E_k(\omega_2)}_{\omega=\omega_1+\omega_2} + \\ & + \iint d\omega_1 d\omega_2 \underbrace{\chi^{(3)}_{ijkl}(\omega; \omega_1, \omega_2, \omega_3) E_j(\omega_1) E_k(\omega_2) E_l(\omega_3)}_{\omega=\omega_1+\omega_2+\omega_3} + \ldots \end{aligned}$$

Linear electro-optic effect (Pockels effect)

Strong low-frequency field $\boldsymbol{E}_s$ (renormalization of optical constants due to second-order susceptibility)

Propagation of a weak high-frequency optical field $\boldsymbol{E}$ in such a disturbed linearized medium

$$P_i = \varepsilon_0 \chi_{ij} E_j + \chi^{(2)}_{ijk}\left(E_j E^S_k + E_k E^S_j\right)$$

New effective permittivity tensor:

$$\varepsilon_{ij} = \varepsilon_0\left(1+\chi_{ij}\right) + 2\chi^{(2)}_{ijk} E^S_k = \varepsilon^L_{ij} + 2\chi^{(2)}_{ijk} E^S_k$$

Wave equation (anisotropic media):

$$\boldsymbol{k}\left(\boldsymbol{k}\cdot\boldsymbol{E}\right) - k^2\boldsymbol{E} + \omega^2\mu_0\varepsilon\cdot\boldsymbol{E} = 0$$

Pockels effect: continued

Description using deformations of the indicatrix

We define the reciprocal dielectric tensor:

$$K_{ij} = \varepsilon_0 \left(\varepsilon^{-1}\right)_{ij}$$

Indicatrix in the system of the principal dielectric axes:

$$\frac{x^2}{n_x^2} + \frac{y^2}{n_y^2} + \frac{z^2}{n_z^2} = 1$$

Indicatrix in a general system of axes:

$$K_{ij} x_i x_j = 1$$

Now the external strong electric field is switched on…

Indicatrix in electro-optics

Reciprocal dielectric tensor is renormalized due to external field:

$$K'_{ij} = K_{ij} + r_{ijk} E_k^S \qquad r_{ijk} \ldots \text{ Pockels electro-optic tensor}$$

Deformed indicatrix: $K'_{ij} x_i x_j = 1$

$$K'_{11} x^2 + K'_{22} y^2 + K'_{33} z^2 + 2K'_{12} xy + 2K'_{13} xz + 2K'_{23} yz = 1$$

- Length of principal axes is modified
- Orientation of the ellipsoid is modified

The Pockels tensor and the second order susceptibility tensor are related by the following equation:

$$\chi_{ijk}^{(2)} = -\frac{\varepsilon_{ii}\varepsilon_{jj}}{2\varepsilon_0} r_{ijk}$$

Pockels tensor

Intrinsic symmetry: related to the fact that ε_{ij} is a hermitic tensor:

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

For symmetric tensors Voigt notation can be introduced:

indices (*ij*)	11	22	33	23 or 32	13 or 31	12 or 21
contraction (*l*)	1	2	3	4	5	6

A 6×3 matrix r_{lk} is introduced, where $l = 1\ldots6$ is a contracted index, and $k = 1\ldots3$.

$$r_{lk} = \begin{pmatrix} r_{11} & r_{12} & r_{13} \\ r_{21} & r_{22} & r_{23} \\ r_{31} & r_{32} & r_{33} \\ r_{41} & r_{42} & r_{43} \\ r_{51} & r_{52} & r_{53} \\ r_{61} & r_{62} & r_{63} \end{pmatrix}$$

Pockels effect: cubic media

In cubic non-centrosymmetric media (GaAs, InP, GaP, CdTe, ZnTe…):

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

$$r_{63} = r_{52} = r_{41}$$

For an applied electric field $\boldsymbol{E} = (E_x, E_y, E_z)$:

$$\frac{x^2}{n^2} + \frac{y^2}{n^2} + \frac{z^2}{n^2} + 2r_{41}E_x yz + 2r_{41}E_y xz + 2r_{41}E_z xy = 1$$

Example: ***E***//***c***

$$\frac{x^2}{n^2}+\frac{y^2}{n^2}+\frac{z^2}{n^2}+2r_{41}E\,xy=1$$

Rotation by 45° in the *xy* plane will diagonalize the equation:

$$x=\frac{1}{\sqrt{2}}(\eta+\xi) \qquad \eta=\frac{1}{\sqrt{2}}(x+y)$$

$$y=\frac{1}{\sqrt{2}}(\eta-\xi) \qquad \xi=\frac{1}{\sqrt{2}}(x-y)$$

One gets:

$$n_\xi=\frac{n}{\sqrt{1-n^2r_{41}E}}\approx n+\frac{1}{2}n^3r_{41}E$$

$$n_\eta=\frac{n}{\sqrt{1+n^2r_{41}E}}\approx n-\frac{1}{2}n^3r_{41}E$$

$$n_z=n$$

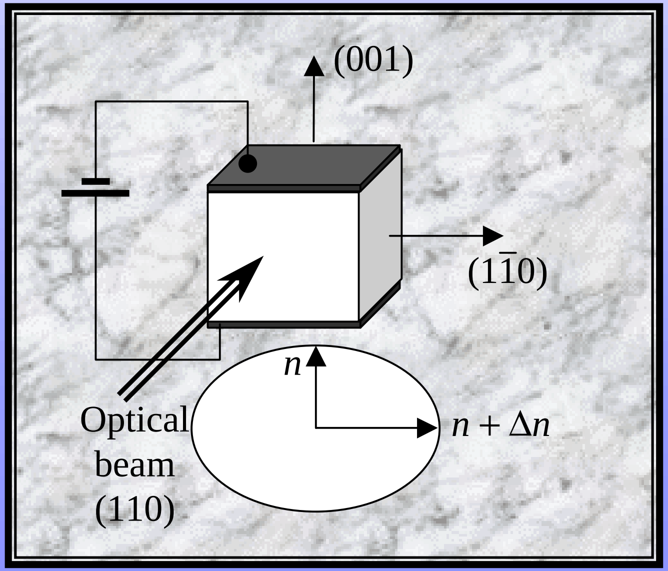
Example: ***E*//*c***, continued

$$n_\xi = \frac{n}{\sqrt{1-n^2 r_{41} E}} \approx n + \frac{1}{2} n^3 r_{41} E$$

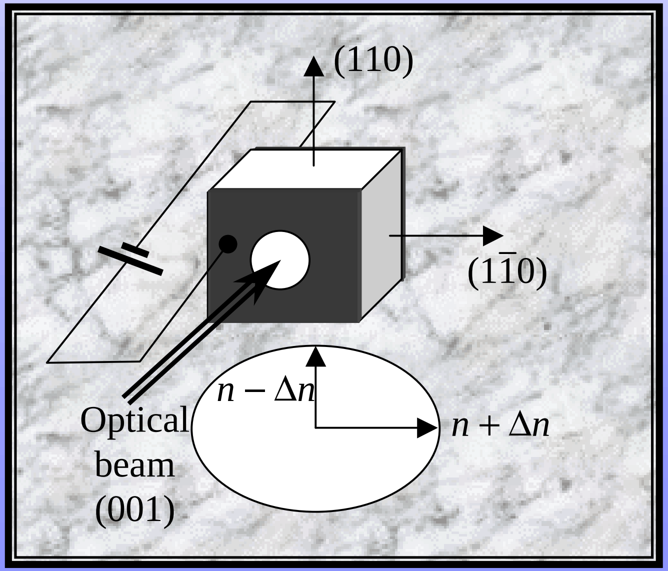
$$n_\eta = \frac{n}{\sqrt{1+n^2 r_{41} E}} \approx n - \frac{1}{2} n^3 r_{41} E$$

$$n_z = n$$

Transverse geometry:



Longitudinal geometry:



Example: ***E*** // (1,1,0)

$$\frac{x^2}{n^2}+\frac{y^2}{n^2}+\frac{z^2}{n^2}+\sqrt{2}r_{41}E\,yz+\sqrt{2}r_{41}E\,xz=1$$

1) Rotation by 45° in the *xy* plane (as previously); new variables ξ, η

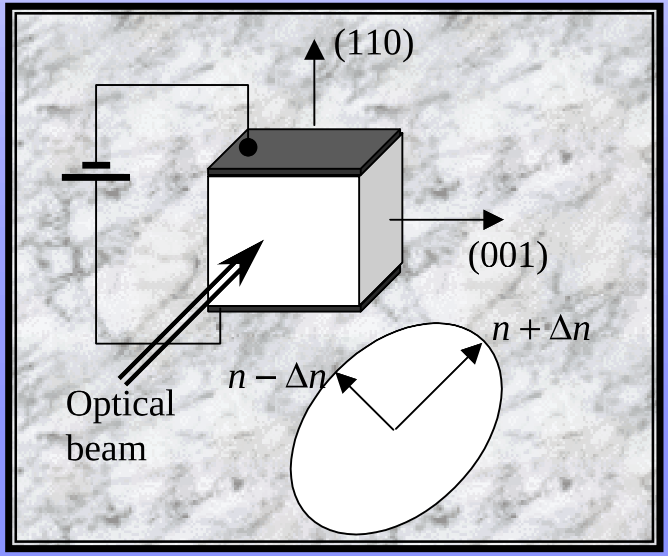
$$\frac{\xi^2}{n^2}+\frac{\eta^2}{n^2}+\frac{z^2}{n^2}+2r_{41}E\,\eta z=1$$

2) Rotation by 45° in the η*z* plane; new variables η', *z'*

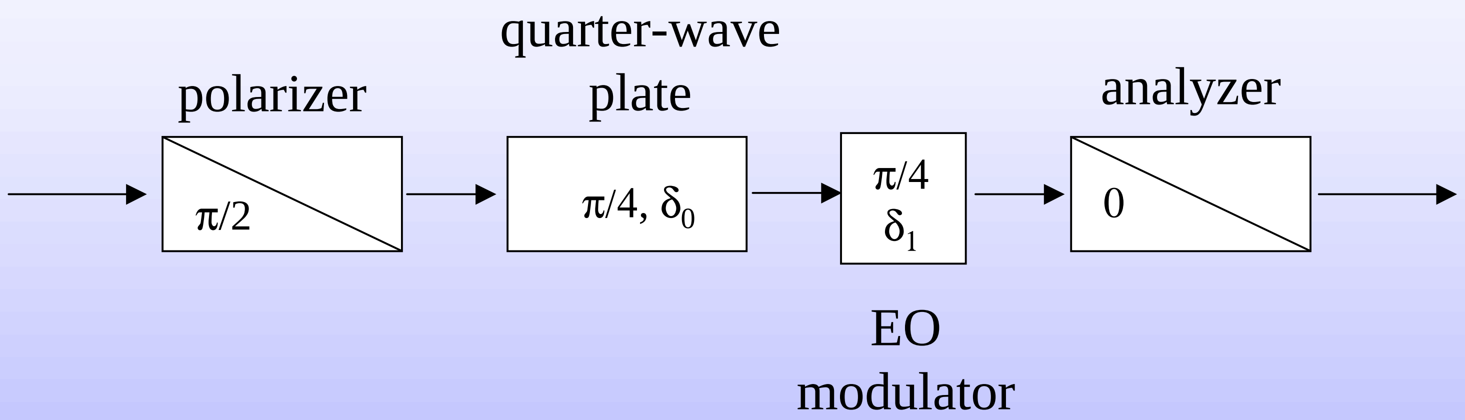
$$n_{z'}\approx n+\frac{1}{2}n^3r_{41}E$$

$$n_{\eta'}\approx n-\frac{1}{2}n^3r_{41}E$$

$$n_{\xi}=n$$



Electro-optic modulation



It was shown previously

$$I = \frac{1}{2}\left(1 - \cos(\delta_0 + \delta_1)\right) = \frac{1}{2}\left(1 + \sin\delta_1\right) \approx \frac{1+\delta_1}{2}$$

Electro-optic dephasing:

Transverse setup: $\delta_1 = \frac{\pi}{\lambda} n^3 r_{41} L \frac{U}{d}$

Longitudinal setup: $\delta_1 = \frac{2\pi}{\lambda} n^3 r_{41} U$

Quadratic electro-optic effect (Kerr effect)

Kerr effect can be of a great importance in the media where the Pockels effect vanishes (centrosymmetric media):

$$P_i = \varepsilon_0 \chi_{ij} E_j + \chi^{(3)}_{ijkl} \left(E_j E^S_k E^S_l + E_k E^S_j E^S_l + E_l E^S_j E^S_k \right)$$

$$\varepsilon^{NL}_{ij} = 3\chi^{(3)}_{ijkl} E^S_k E^S_l$$

The same treatment can be used as for the linear Pockels effect; the following differences should be emphasized:

- it is a 3rd order nonlinear effect (the electric field should be very strong)
- it is a quadratic effect in ***E***, so the induced birefringence is proportional to $\Delta n \sim (E^S)^2$
- the effect depends on a 4th rank tensor, so the symmetry properties are different comparing to the linear EO effect

Magneto-optic effects (Faraday, Cotton-Mouton)

The polarization, and consequently, the permittivity can depend on the magnetic field:

$$P_i = \varepsilon_0 \chi_{ij} E_j + \chi^{(2)}_{ijk} E_j E_k + \chi^{(3)}_{ijkl} E_j E_k E_l + h^{(2)}_{ijk} B_k E_j + h^{(3)}_{ijkl} B_l B_k E_j \ldots$$

Faraday effect

Cotton-Mouton effect

$$\varepsilon_{ij}(\boldsymbol{B}) = h^{(2)}_{ijk} B_k + h^{(3)}_{ijkl} B_l B_k + \ldots$$

Intrinsic symmetry properties in the magnetic field (Onsager theorem):

$$\varepsilon_{ij}(\boldsymbol{B}) = \varepsilon_{ji}(-\boldsymbol{B}) = \varepsilon^*_{ji}(\boldsymbol{B}) = \varepsilon^*_{ij}(-\boldsymbol{B})$$

From this relation we find conditions for h_{ijk} and h_{ijkl}

Faraday effect

$$\varepsilon_{ij}(\boldsymbol{B}) = h_{ijk}^{(2)} B_k$$

$$\varepsilon_{ij}(\boldsymbol{B}) = \varepsilon_{ji}(-\boldsymbol{B}) = \varepsilon_{ji}^{*}(\boldsymbol{B}) = \varepsilon_{ij}^{*}(-\boldsymbol{B})$$

These conditions are similar as for the spatial dispersion and the optical activity

The tensor h_{ijk} is

- purely imaginary
- antisymmetric in first two indices

Similarly as for the optical activity, we can define the effective permittivity tensor:

$$\boldsymbol{\varepsilon} = \begin{pmatrix} \varepsilon_{xx}^{0} & ig_{12} & ig_{13} \\ -ig_{12} & \varepsilon_{yy}^{0} & ig_{23} \\ -ig_{13} & -ig_{23} & \varepsilon_{zz}^{0} \end{pmatrix}$$

Effect of a longitudinal field

Cubic non-centrosymmetric medium

Wave equation:

$$\begin{pmatrix} n_0^2 - n^2 & i\Delta B & 0 \\ -i\Delta B & n_0^2 - n^2 & 0 \\ 0 & 0 & n_0^2 \end{pmatrix} \begin{pmatrix} E_x \\ E_y \\ E_z \end{pmatrix} = 0$$

Eigenvalues
(effective refractive indices)

$$n_{I,II} \approx n_0 \pm \frac{\Delta B}{2n_0}$$

Eigenvectors
(Polarization direction)

$$\begin{pmatrix} 1 \\ -i \\ 0 \end{pmatrix}, \begin{pmatrix} 1 \\ i \\ 0 \end{pmatrix}$$

- Cumulative character of the Faraday effect

Effect of a transverse field

Wave equation:

$$\begin{pmatrix} n_0^2 - n^2 & 0 & 0 \\ 0 & n_0^2 - n^2 & i\Delta B \\ 0 & -i\Delta B & n_0^2 \end{pmatrix} \begin{pmatrix} E_x \\ E_y \\ E_z \end{pmatrix} = 0$$

Eigenvalues:

$$n_I = n_0$$

$$n_{II} \approx n_0 - \frac{\Delta^2 B^2}{2n_0^4}$$

Cotton-Mouton effect: very small correction to a linear birefringence