Lecture 12: Introduction to nonlinear optics II.

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Propagation of strong optic signals (proper nonlinear effects)

- Second order effects
  - ☞ Three-wave mixing
    - Phase matching condition
  - ☞ Second harmonic generation
  - ☞ Sum frequency generation
  - ☞ Parametric generation
- Third order effects
  - ☞ Four-wave mixing
  - ☞ Optical Kerr effect

Nonlinear polarization

$$P_i(\omega) = \varepsilon_0 \chi_{ij}(\omega) E_j(\omega) + \int d\omega_1 \underbrace{\chi^{(2)}_{ijk}(\omega;\omega_1,\omega_2) E_j(\omega_1) E_k(\omega_2)}_{\omega=\omega_1+\omega_2} + \ldots$$

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

Intrinsic symmetry: $\chi_{ijk} = \chi_{ikj}$

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 3×6 matrix χ_{il} is introduced,
where $l = 1\ldots6$ is a contracted index,
and $i = 1\ldots3$.

$$\chi_{il} = \begin{pmatrix} \chi_{11} & \chi_{12} & \chi_{13} & \chi_{14} & \chi_{15} & \chi_{16} \\ \chi_{21} & \chi_{22} & \chi_{23} & \chi_{24} & \chi_{25} & \chi_{26} \\ \chi_{31} & \chi_{32} & \chi_{33} & \chi_{34} & \chi_{35} & \chi_{36} \end{pmatrix}$$

Three-wave mixing

Coupling between two optical waves ω_1 and ω_2:

$$\boldsymbol{E}^{\omega_1}(t) = \mathrm{Re}\left\{\boldsymbol{E}^{\omega_1}\, e^{i\omega_1 t}\right\} = \frac{1}{2}\left(\boldsymbol{E}^{\omega_1}\, e^{i\omega_1 t} + (\boldsymbol{E}^{\omega_1})^*\, e^{-i\omega_1 t}\right) = \frac{1}{2}\left(\boldsymbol{E}^{\omega_1}\, e^{i\omega_1 t} + c.c.\right)$$

$$\boldsymbol{E}^{\omega_2}(t) = \mathrm{Re}\left\{\boldsymbol{E}^{\omega_2}\, e^{i\omega_2 t}\right\} = \frac{1}{2}\left(\boldsymbol{E}^{\omega_2}\, e^{i\omega_2 t} + (\boldsymbol{E}^{\omega_2})^*\, e^{-i\omega_2 t}\right) = \frac{1}{2}\left(\boldsymbol{E}^{\omega_2}\, e^{i\omega_2 t} + c.c.\right)$$

The total field:

$$\boldsymbol{E} = \boldsymbol{E}^{\omega_1}(t) + \boldsymbol{E}^{\omega_2}(t) = \frac{1}{2}\left(\boldsymbol{E}^{\omega_1}\, e^{i\omega_1 t} + \boldsymbol{E}^{\omega_2}\, e^{i\omega_2 t} + c.c.\right)$$

Linear part of the polarization $\boldsymbol{P}_L$:

$$\boldsymbol{P}_L = \varepsilon_0\left(\chi(\omega_1)\, \boldsymbol{E}^{\omega_1}(t) + \chi(\omega_2)\, \boldsymbol{E}^{\omega_2}(t)\right)$$

Nonlinear part of the polarization $\boldsymbol{P}_{NL}$:

$$\boldsymbol{P}_{NL} = \chi^{(2)}\boldsymbol{E}\,\boldsymbol{E} = \frac{1}{4}\chi^{(2)}\left(\boldsymbol{E}^{\omega_1}\boldsymbol{E}^{\omega_1}\, e^{2i\omega_1 t} + \boldsymbol{E}^{\omega_2}\boldsymbol{E}^{\omega_2}\, e^{2i\omega_2 t} + 2\boldsymbol{E}^{\omega_1}\boldsymbol{E}^{\omega_2}\, e^{i(\omega_1+\omega_2)t}\right.$$

$$\left. + 2\boldsymbol{E}^{\omega_1}(\boldsymbol{E}^{\omega_2})^*\, e^{i(\omega_1-\omega_2)t} + \boldsymbol{E}^{\omega_1}(\boldsymbol{E}^{\omega_1})^* + \boldsymbol{E}^{\omega_2}(\boldsymbol{E}^{\omega_2})^* + c.c.\right)$$

Nonlinear polarization for three wave mixing

$$\boldsymbol{P}_{NL} = \chi^{(2)} \boldsymbol{E}\,\boldsymbol{E} = \frac{1}{4}\chi^{(2)} \Big(\boldsymbol{E}^{\omega_1} \boldsymbol{E}^{\omega_1} e^{2i\omega_1 t} + \boldsymbol{E}^{\omega_2} \boldsymbol{E}^{\omega_2} e^{2i\omega_2 t} + 2\boldsymbol{E}^{\omega_1} \boldsymbol{E}^{\omega_2} e^{i(\omega_1+\omega_2)t}$$
$$+ 2\boldsymbol{E}^{\omega_1} (\boldsymbol{E}^{\omega_2})^* e^{i(\omega_1-\omega_2)t} + \boldsymbol{E}^{\omega_1} (\boldsymbol{E}^{\omega_1})^* + \boldsymbol{E}^{\omega_2} (\boldsymbol{E}^{\omega_2})^* + c.c. \Big)$$

If we take into account the dispersion, the susceptibility is weighted: $\chi^{(2)}(\omega_1, \omega_2)$

The polarization $\boldsymbol{P}_{NL}$, when introduced into the Maxwell equations, becomes the source of the radiation at frequencies $2\omega_1$, $2\omega_2$, $\omega_1 + \omega_2$ et $\omega_1 - \omega_2$

It causes an energy transfer between the fundamental and the mixed spectral components

Three wave mixing: two initial components (ω_1 and ω_2) give raise to a third one (ω_3)

A phase matching condition has to be fulfilled : at most one efficient energy transfer channel is in general possible

Second harmonic generation (SHG)

Coupling between ω and 2ω — other spectral components are omitted:

$$\boldsymbol{E} = \boldsymbol{E}^{\omega}(t) + \boldsymbol{E}^{2\omega}(t) = \frac{1}{2}\left(\boldsymbol{E}^{\omega} e^{i\omega t} + \boldsymbol{E}^{2\omega} e^{i2\omega t} + c.c.\right)$$

$$P^{\omega}_{i,NL} = \frac{1}{2}\chi^{(2)}_{ijk}\left(E^{2\omega}_{j}(E^{\omega}_{k})^{*} e^{i\omega t} + c.c.\right)$$

$$P^{2\omega}_{i,NL} = \frac{1}{4}\chi^{(2)}_{ijk}\left(E^{\omega}_{j}E^{\omega}_{k} e^{2i\omega t} + c.c.\right)$$

The wave equation in the time domain then reads:

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

Absorption can be taken into account in ε; however, we neglect it here

The waves are supposed to propagate along *z*; their amplitudes do not depend on *x* and *y*.

SHG: continued

$$E_j^{\omega}(z,t)=\frac{1}{2}\left(E_j^{\omega}(z)e^{i(\omega t-k_1 z)}+c.c.\right) \qquad E_j^{2\omega}(z,t)=\frac{1}{2}\left(E_j^{2\omega}(z)e^{i(2\omega t-k_2 z)}+c.c.\right)$$

The energy transfer between the two waves is assumed to be very small in the scale of the wavelength:

$$\frac{dE_j^{\omega}}{dz}k_1 \gg \frac{d^2E_j^{\omega}}{dz^2} \qquad \frac{dE_j^{2\omega}}{dz}k_2 \gg \frac{d^2E_j^{2\omega}}{dz^2}$$

Coupled wave equations:

$$\left(\left(\omega^2 n_\omega^2 / c^2 - k_1^2\right)\frac{E_j^{\omega}}{2} - ik_1\frac{dE_j^{\omega}}{dz}\right)e^{i(\omega t-k_1 z)}+c.c.=\mu_0\frac{\partial^2 P_{j,NL}^{\omega}}{\partial t^2}$$

$$\left(\left((2\omega)^2 n_{2\omega}^2 / c^2 - k_2^2\right)\frac{E_j^{2\omega}}{2} - ik_2\frac{dE_j^{2\omega}}{dz}\right)e^{i(2\omega t-k_2 z)}+c.c.=\mu_0\frac{\partial^2 P_{j,NL}^{2\omega}}{\partial t^2}$$

Coupled-wave equations

$$\left(\left(\omega^2 n_\omega^2 / c^2 - k_1^2\right)\frac{E_j^\omega}{2} - ik_1 \frac{dE_j^\omega}{dz}\right)e^{i(\omega t - k_1 z)} + c.c. = \mu_0 \frac{\partial^2 P_{j,NL}^\omega}{\partial t^2}$$

$$\left(\left((2\omega)^2 n_{2\omega}^2 / c^2 - k_2^2\right)\frac{E_j^{2\omega}}{2} - ik_2 \frac{dE_j^{2\omega}}{dz}\right)e^{i(2\omega t - k_2 z)} + c.c. = \mu_0 \frac{\partial^2 P_{j,NL}^{2\omega}}{\partial t^2}$$

The wave equations without coupling define the wave vectors k_1 and k_2:

$$\omega^2 n_\omega^2 / c^2 - k_1^2 = 0 \qquad (2\omega)^2 n_{2\omega}^2 / c^2 - k_2^2 = 0$$

We finally obtain:

$$\frac{dE_j^\omega}{dz} = -\frac{i\omega\eta_0}{2n_\omega}\chi_{jkl}^{(2)} E_k^{2\omega} (E_l^\omega)^* e^{-i(k_2 - 2k_1)z}$$

$$\frac{dE_j^{2\omega}}{dz} = -\frac{i\omega\eta_0}{2n_{2\omega}}\chi_{jkl}^{(2)} E_k^\omega E_l^\omega e^{-i(2k_1 - k_2)z}$$

Constant field approximation

The fundamental wave is supposed not to be depleted:

$$\frac{dE_j^{2\omega}}{dz} = -\frac{i\omega\eta_0}{2n_{2\omega}} \chi_{jkl}^{(2)} E_k^{\omega} E_l^{\omega} e^{-i(2k_1-k_2)z}$$

Solution:

$$E_j^{2\omega}(z) = B - A e^{i\Delta kz}$$

with

$$\Delta k = k_2 - 2k_1$$

$$A = \frac{\omega\eta_0 \chi_{jkl}^{(2)} E_k^{\omega} E_l^{\omega}}{2 n_{2\omega} \Delta k}$$

B determined from the boundary condition: $E_j^{2\omega}(z=0) = 0$

SHG solution

$$E_j^{2\omega}(L) = \frac{\omega\eta_0}{2n_{2\omega}} \frac{1-e^{i\Delta kL}}{\Delta k} \chi_{jkl}^{(2)} E_k^{\omega} E_l^{\omega}$$

$$I_{2\omega} = \frac{n_{2\omega}}{2\eta_0} \left| E_j^{2\omega}(L) \right|^2 = \frac{1}{2} \eta_0^3 \frac{\omega^2 \left(\chi_{eff}^{(2)}\right)^2 L^2}{n_{2\omega} n_{\omega}^2} I_{\omega}^2 \frac{\sin^2(\frac{1}{2}\Delta kL)}{\left(\frac{1}{2}\Delta kL\right)^2}$$

Character of the solution depends critically on the value of Δk

$\Delta k \neq 0$

Both waves do not propagate with the same phase velocity: they are not constantly in phase, but become periodically out-of-phase. This leads to a modulation of $I_{2\omega}$ with the period (called coherence length):

$$l_c = \frac{2\pi}{\Delta k} = \frac{2\pi}{k_2 - 2k_1} = \frac{\lambda}{2\left(n_{2\omega} - n_{\omega}\right)}$$

Typically: $n_{2\omega} - n_{\omega} \approx 10^{-2}$, $l_c \approx 100$ μm. This is the maximum crystal length that can efficiently participate to SHG.

Phase matching condition

$$\Delta k = 0 \Rightarrow \quad k_2 = 2k_1$$

$$n_{2\omega} = n_{\omega}$$

$$I_{2\omega} = \frac{1}{2}\eta_0^3 \frac{\omega^2 \left(\chi_{eff}^{(2)}\right)^2}{n_{2\omega} n_{\omega}^2} I_{\omega}^2 L^2$$

All the crystal length participates efficiently to the generation

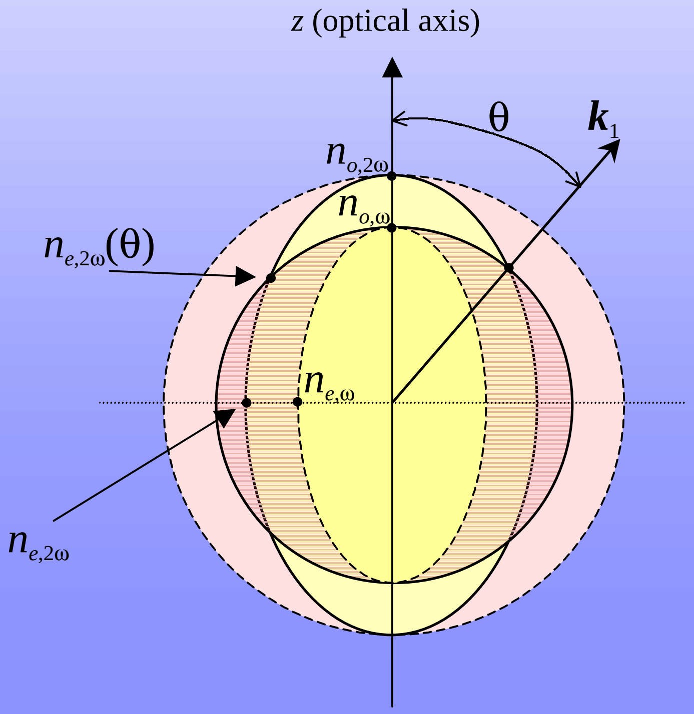
How to achieve the phase matching condition:

- Compensation of the birefringence and the dispersion

OO-E interaction

$$n_{e,2\omega}(\theta) = n_{o,\omega}$$

$$\sin^2\theta = \frac{n_{o,\omega}^{-2} - n_{o,2\omega}^{-2}}{n_{e,2\omega}^{-2} - n_{o,2\omega}^{-2}}$$

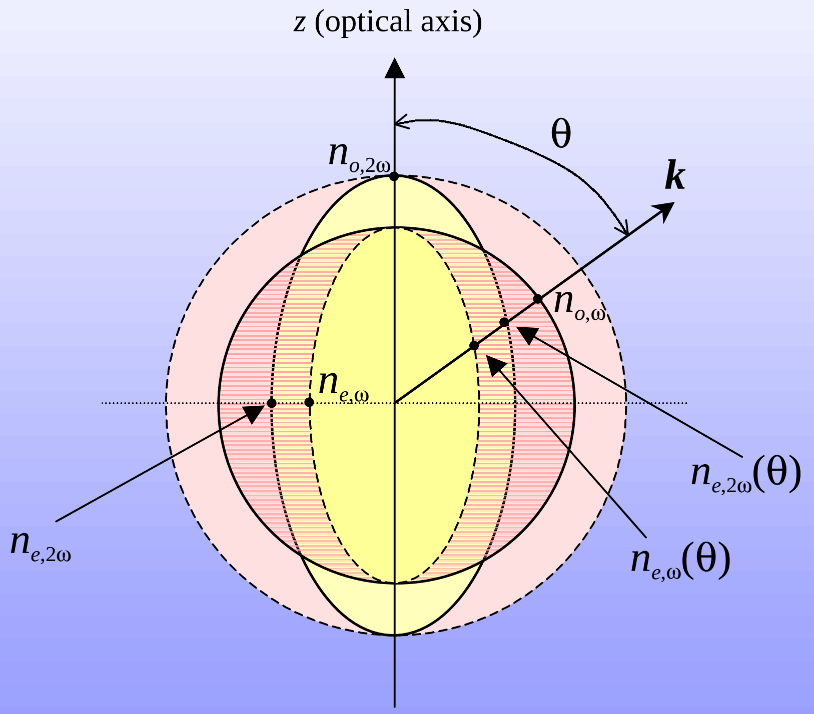


Phase matching condition: continued

EO-E interaction:

$$\Delta k = 0 \;\Rightarrow\; \quad k_{2\omega,e} = k_{\omega,o} + k_{\omega,e}$$

$$n_{e,2\omega}(\theta) = \frac{1}{2}\left(n_{o,\omega} + n_{e,\omega}(\theta)\right)$$



The choice of the polarizations depends on the available coefficients of χ_{ijk} (e.g. χ_{111} couples only parallel polarizations and thus can never allow the phase matching)

Three-wave mixing: summary

General equations of three-wave mixing

$\omega_1 \pm \omega_2 \pm \omega_3 = 0$ (frequency transformation)

$\boldsymbol{k}_1 \pm \boldsymbol{k}_2 \pm \boldsymbol{k}_3 = 0$ (phase matching condition)

Sum and difference frequency generation (SFD, DFD):

- Input: two strong beams ω_1 and ω_2
- Output: strong beam ω_3

$$\omega_1 \pm \omega_2 = \omega_3$$
$$\boldsymbol{k}_1 \pm \boldsymbol{k}_2 = \boldsymbol{k}_3$$

Parametric generation (amplification of weak beams):

- Input: strong ω_3 + weak ω_1
- Output: medium ω_2 + medium ω_1

$$\omega_3 - \omega_1 = \omega_2$$
$$\boldsymbol{k}_3 - \boldsymbol{k}_1 = \boldsymbol{k}_2$$

Up-conversion

- Input: strong ω_1 + weak ω_2
- Output: weak ω_3

$$\omega_1 + \omega_2 = \omega_3$$
$$\boldsymbol{k}_1 + \boldsymbol{k}_2 = \boldsymbol{k}_3$$

Four-wave mixing

Third order effect:

$$\boldsymbol{P}_{NL}^{\omega_4} = \chi_{ijkl}^{(3)} E_j^{\omega_1} E_k^{\omega_2} E_l^{\omega_3}$$

Required conditions for the wavelength transformation:

$$\omega_4 = \omega_1 + \omega_2 + \omega_3 \qquad \text{or} \qquad \omega_4 + \omega_3 = \omega_1 + \omega_2 \qquad \text{etc.}$$

$$\boldsymbol{k}_4 = \boldsymbol{k}_1 + \boldsymbol{k}_2 + \boldsymbol{k}_3 \qquad\qquad \boldsymbol{k}_4 + \boldsymbol{k}_3 = \boldsymbol{k}_1 + \boldsymbol{k}_2$$

Degenerated cases are frequently used

Transient grating experiments

Propagation in Kerr-like media

Degenerated case (one very strong optical beam):

$$\boldsymbol{P}_{NL}^{\omega} = 3\chi^{(3)} E^{\omega} E^{\omega} \left(E^{\omega}\right)^{*}$$

Indices are omitted (i.e. the beam is linearly polarized and it is an eigenmode of the medium

The beam propagated along z:

$$E^{\omega}(z,t) = A(z) e^{i(\omega t - kz)}$$

Wave equation:

$$\left(\left(\omega^2 n^2 / c^2 - k^2\right) A - 2ik \frac{dA}{dz}\right) e^{i(\omega t - kz)} = -3\mu_0 \omega^2 \chi^{(3)} A^2 A^{*} e^{i(\omega t - kz)}$$

Linear wave equation: definition of k

Nonlinear polarization

Propagation in Kerr media: continued

Remaining terms in the wave equation

$$\frac{dA}{dz} = -i\frac{3}{2}\sqrt{\frac{\mu_0}{\varepsilon_0}}\frac{\omega}{n}\chi^{(3)}\left|A\right|^2 A$$

if $\chi^{(3)}$ is real then

$$A = A_0 \exp\left(-i\frac{3\eta_0\omega\chi^{(3)}}{2n}\left|A_0\right|^2 z\right) = A_0 e^{-ik_1 z}$$

$$E^{\omega}(z,t) = A(z)e^{i(\omega t - kz - k_1 z)}$$

The wave vector is renormalized:

$$K = k + k_1 = \frac{\omega}{c}\left(n + \frac{3\chi^{(3)}}{2\varepsilon_0 n}\left|A_0\right|^2\right)$$

The effective refractive index depends on the intensity of the beam:

$$n' = n + n_2 I \qquad \left(n_2 = \frac{3\eta_0\chi^{(3)}}{\varepsilon}\right)$$

Propagation in Kerr media

Self-phase modulation (ultrashort pulses)

refractive index is time dependent

phase of the pulse is modulation

creation of new frequency components (bandwidth broadening)

pulse shortening

Self-focusation (intense beams)

Kerr lensing due to spatial profile of the beam