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

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

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

Intrinsic symmetry: \( \chi_{ijk} = \chi_{ikj} \)

For symmetric tensors Voigt notation can be introduced:

<table>
<thead>
<tr>
<th>indices ((ij))</th>
<th>11</th>
<th>22</th>
<th>33</th>
<th>23 or 32</th>
<th>13 or 31</th>
<th>12 or 21</th>
</tr>
</thead>
<tbody>
<tr>
<td>contraction ((l))</td>
<td>1</td>
<td>2</td>
<td>3</td>
<td>4</td>
<td>5</td>
<td>6</td>
</tr>
</tbody>
</table>

A 3×6 matrix \( \chi_{il} \) is introduced, where \( l = 1 \ldots 6 \) is a contracted index, and \( i = 1 \ldots 3 \).
Three-wave mixing

Coupling between two optical waves $\omega_1$ and $\omega_2$:

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

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

The total field:

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

Linear part of the polarization $P_L$:

$$P_L = \varepsilon_0 \left( \chi(\omega_1) E^{\omega_1}(t) + \chi(\omega_2) E^{\omega_2}(t) \right)$$

Nonlinear part of the polarization $P_{NL}$:

$$P_{NL} = \chi^{(2)} E E = \frac{1}{4} \chi^{(2)} \left( E^{\omega_1} E^{\omega_1} e^{2i\omega_1 t} + E^{\omega_2} E^{\omega_2} e^{2i\omega_2 t} + 2 E^{\omega_1} E^{\omega_2} e^{i(\omega_1+\omega_2)t} ight.$$  

$$\left. + 2 E^{\omega_1} (E^{\omega_2})^* e^{i(\omega_1-\omega_2)t} + E^{\omega_1} (E^{\omega_1})^* + E^{\omega_2} (E^{\omega_2})^* + \text{c.c.} \right)$$
Nonlinear polarization for three wave mixing

\[ P_{NL} = \chi^{(2)} E E = \frac{1}{4} \chi^{(2)} \left( E^{\omega_1} E^{\omega_1} e^{2i\omega_1 t} + E^{\omega_2} E^{\omega_2} e^{2i\omega_2 t} + 2E^{\omega_1} E^{\omega_2} e^{i(\omega_1 + \omega_2)t} \right. \]

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

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

The polarization \( P_{NL} \), 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 (\( \omega_1 \) and \( \omega_2 \)) give raise to a third one (\( \omega_3 \))

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 $\omega$ and $2\omega$ — other spectral components are omitted:

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

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

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

The wave equation in the time domain then reads:

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

Absorption can be taken into account in $\varepsilon$; however, we neglect it here.

The waves are supposed to propagate along $z$; their amplitudes do not depend on $x$ and $y$. 
SHG: continued

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

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

\[ \frac{dE_j^\omega}{dz} k_1 \gg \frac{d^2E_j^\omega}{dz^2} \quad \frac{dE_j^{2\omega}}{dz} k_2 \gg \frac{d^2E_j^{2\omega}}{dz^2} \]

Coupled wave equations:

\[
\left( \frac{(\omega n_0^2/c^2 - k_1^2)}{2} \frac{E_j^\omega}{dz} - ik_1 \frac{dE_j^\omega}{dz} \right) e^{i(\omega t - k_1 z)} + c.c. = \mu_0 \frac{\partial^2 P_{j,NL}^\omega}{\partial t^2} \\
\left( \frac{(2\omega n_0^2/c^2 - k_2^2)}{2} \frac{E_j^{2\omega}}{dz} - ik_2 \frac{dE_j^{2\omega}}{dz} \right) e^{i(2\omega t - k_2 z)} + c.c. = \mu_0 \frac{\partial^2 P_{j,NL}^{2\omega}}{\partial t^2}
\]
Coupled-wave equations

\[
\begin{align*}
\left( \left( \frac{\omega^2 n_{\omega}^2}{c^2} - k_1^2 \right) E_j^{\omega} + \frac{dE_j^{\omega}}{dz} \right) e^{i(\omega t - k_1 z)} + \text{c.c.} &= \mu_0 \frac{\partial^2 P_{j,\text{NL}}^\omega}{\partial t^2} \\
\left( \left( 2\omega \right)^2 n_{2\omega}^2 / c^2 - k_2^2 \right) E_j^{2\omega} + \frac{dE_j^{2\omega}}{dz} \right) e^{i(2\omega t - k_2 z)} + \text{c.c.} &= \mu_0 \frac{\partial^2 P_{j,\text{NL}}^{2\omega}}{\partial t^2}
\end{align*}
\]

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

\[
\frac{\omega^2 n_{\omega}^2}{c^2} - k_1^2 = 0 \quad \left( 2\omega \right)^2 \frac{n_{2\omega}^2}{c^2} - k_2^2 = 0
\]

We finally obtain:

\[
\begin{align*}
\frac{dE_j^{\omega}}{dz} &= -i \omega \eta_0 \chi_{jkl}^{(2)} E_k^{2\omega} \left( E_l^{\omega} \right)^* e^{-i(k_2 - 2k_1)z} \\
\frac{dE_j^{2\omega}}{dz} &= -i \omega \eta_0 \chi_{jkl}^{(2)} E_k^{\omega} E_l^{\omega} e^{-i(2k_1 - k_2)z}
\end{align*}
\]
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 \( \Delta k = k_2 - 2k_1 \)

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

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

\[ E^{2\omega}_{j}(L) = \frac{\omega \eta_0}{2n_{2\omega}} \frac{1 - e^{i\Delta k L}}{\Delta k} \chi^{(2)}_{jkl} E^{\omega}_{k} E^{\omega}_{l} \]

\[ I_{2\omega} = \frac{n_{2\omega}}{2\eta_0} \left| E^{2\omega}_{j}(L) \right|^2 = \frac{1}{2} \eta_0 \frac{\omega^2 \left( \chi^{(2)}_{\text{eff}} \right)^2 L^2}{n_{2\omega} n_{\omega}^2} I_{\omega}^2 \frac{\sin^2 \left( \frac{1}{2} \Delta k L \right)}{\left( \frac{1}{2} \Delta k L \right)^2} \]

Character of the solution depends critically on the value of \( \Delta k \)

\( \Delta k \neq 0 \)

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):

\[ l_c = \frac{2\pi}{\Delta k} = \frac{2\pi}{k_2 - 2k_1} = \frac{\lambda}{2(n_{2\omega} - n_{\omega})} \]

Typically: \( n_{2\omega} - n_{\omega} \approx 10^{-2} \), \( l_c \approx 100 \mu \text{m} \). This is the maximum crystal length that can efficiently participate to SHG.
Phase matching condition

\[ \Delta k = 0 \Rightarrow k_2 = 2k_1 \]

\[ n_{2\omega} = n_{\omega} \]

\[ I_{2\omega} = \frac{1}{2} \eta_0^3 \frac{\omega^2 (\chi^{(2)}_{\text{eff}})^2}{n_{2\omega} n_{\omega}^2} I_\omega^2 L^2 \]

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:**

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

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

The choice of the polarizations depends on the available coefficients of $\chi_{ijk}$ (e.g. $\chi_{111}$ couples only parallel polarizations and thus can never allow the phase matching)
Three-wave mixing: summary

General equations of three-wave mixing
\[ \omega_1 \pm \omega_2 \pm \omega_3 = 0 \quad \text{(frequency transformation)} \]
\[ k_1 \pm k_2 \pm k_3 = 0 \quad \text{(phase matching condition)} \]

Sum and difference frequency generation (SFD, DFD):
- Input: two strong beams \( \omega_1 \) and \( \omega_2 \)
- Output: strong beam \( \omega_3 \)
\[
\omega = \omega_1 \pm \omega_2 = \omega_3 \\
k = k_1 \pm k_2 = k_3
\]

Parametric generation (amplification of weak beams):
- Input: strong \( \omega_3 \) + weak \( \omega_1 \)
- Output: medium \( \omega_2 \) + medium \( \omega_1 \)
\[
\omega = \omega_3 - \omega_1 = \omega_2 \\
k = k_3 - k_1 = k_2
\]

Up-conversion
- Input: strong \( \omega_1 \) + weak \( \omega_2 \)
- Output: weak \( \omega_3 \)
\[
\omega = \omega_1 + \omega_2 = \omega_3 \\
k = k_1 + k_2 = k_3
\]
Four-wave mixing

Third order effect:

\[ P_{NL}^{\omega_4} = \chi_{ijkl}^{(3)} E_j^{\omega_1} E_k^{\omega_2} E_l^{\omega_3} \]

Required conditions for the wavelength transformation:

\[ \omega_4 = \omega_1 + \omega_2 + \omega_3 \quad \text{or} \quad \omega_4 + \omega_3 = \omega_1 + \omega_2 \]
\[ k_4 = k_1 + k_2 + k_3 \quad \text{etc.} \]
\[ k_4 + k_3 = k_1 + k_2 \]

Degenerated cases are frequently used

Transient grating experiments
Propagation in Kerr-like media

Degenerated case (one very strong optical beam):

\[ P_{NL}^{\omega} = 3\chi^{(3)} E^{\omega} E^{\omega} \left( E^{\omega} \right)^* \]

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

The beam propagated along \( z \):

\[ E^{\omega}(z, t) = A(z) e^{i(\omega t - k z)} \]

Wave equation:

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

Linear wave equation: definition of \( k \)

Nonlinear polarization
Propagation in Kerr media: continued

Remaining terms in the wave equation

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

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

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

The wave vector is renormalized:

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

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

\[
n' = n + n_2 I \quad \left(n_2 = \frac{3\eta_0 \chi^{(3)}}{\varepsilon} \right)
\]
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