Advanced Experimental Methods

Non-linear optics in crystals

Wavelength conversion of laser light

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Part A
Nonlinear Optics

The first non-linear optical laser experiment

The Nonlinear Susceptibility

\[ \tilde{P} = \chi^{(1)} \tilde{E} + \chi^{(2)} \tilde{E} \tilde{E} + \chi^{(3)} \tilde{E} \tilde{E} \tilde{E} + ... \]

The \( \chi^{(n)}_{ij} \) are tensors even for lowest order

\[ P_i = \chi^{(n)}_{ij} E_j \]

Polarization not directed along electric field vector

In media with inversion symmetry:

\[ I_{op} \tilde{P} = -\tilde{P} = -\chi^{(1)} \tilde{E} - \chi^{(2)} \tilde{E} \tilde{E} - \chi^{(3)} \tilde{E} \tilde{E} \tilde{E} - ... \]

\[ I_{op} \tilde{E} = -\tilde{E} \]

Hence:

\[ I_{op} \tilde{P} = -\chi^{(1)} \tilde{E} + \chi^{(2)} \tilde{E} \tilde{E} - \chi^{(3)} \tilde{E} \tilde{E} \tilde{E} + ... \]

Conclude for centro-symmetric media:

\[ \chi^{(2n)} = 0 \]

Note that in principle there exist also nonlinear magnetic susceptibilities
Nonlinear Optics; graphically

Linear response:
\[ \bar{P} = \chi^{(1)} E \]

Nonlinear response:
\[ \bar{P} = \chi^{(1)} E + \chi^{(2)} EE \]

Nonlinear response evaluated in terms of Fourier series

\[ P = \sum a_n \sin(n \omega t + \phi_n) \]
Lorentz model of linear optics: classical oscillator

Lorentz Equation

\[ \frac{d^2}{dt^2} \mathbf{r} + 2\gamma \frac{d}{dt} \mathbf{r} + \omega_0^2 \mathbf{r} = -\frac{e}{m} \mathbf{E} \]

Write electric field and position vector:

\[ \mathbf{E} = \text{Re} [E e^{i\omega t}] \quad \mathbf{r} = \text{Re} [r e^{i\omega t}] \]

\[ (\omega_0^2 - \omega^2) r + 2i\omega \gamma r = -\frac{e}{m} E \]

Solution

\[ r = \frac{-eE}{m[\omega_0^2 - \omega^2 + 2i\omega \gamma]} \approx \frac{-eE}{2m[\omega_0(\omega_0 - \omega) + i\omega \gamma]} \]

Near resonance \( \omega = \omega_0 \)

\[ r = \frac{Ne^2}{2m[\omega_0(\omega_0 - \omega) + i\omega \gamma]} E = \varepsilon_0 \chi(\omega) E \]
Intermezzo

Lorentz model of linear optics: classical oscillator

Classical polarization of the medium

\[ P(\omega) = -N_{\text{er}}(\omega) \]

and the complex susceptibility

\[ \chi(\omega) = \chi'(\omega) - i\chi''(\omega) \]

yields expressions for the susceptibility

Real part, connected to the index of refraction

\[ \chi'(\omega) = \frac{Ne^2}{2m\omega_0\gamma_0} \frac{(\omega_0 - \omega) / \gamma}{1 + (\omega_0 - \omega)^2 / \gamma^2} \]

Imaginary part, connected to the absorption coefficient

\[ \chi''(\omega) = \frac{Ne^2}{2m\omega_0\gamma_0} \frac{1}{1 + (\omega_0 - \omega)^2 / \gamma^2} \]

Result:

Resonance features of driven electron

Dispersion and absorption

Note also the Kramers-Kronig relation

\[ \chi'(\omega) = P \int_0^{+\infty} \frac{d\nu}{\pi} \chi''(\nu) \frac{2\nu}{\nu^2 - \omega^2} \]
Lorentz model of nonlinear optics: classical oscillator

Motion of electron with anharmonic term:
\[
\frac{d^2}{dt^2} r + 2\gamma \frac{d}{dt} r + \omega_0^2 r - \xi r^2 = -\frac{e}{m} E
\]

Try a solution in power series
\[
r = r_1 + r_2 + r_3 + \ldots
\]
with:
\[
r_i = a_i E^i
\]

Collect terms in same order of \( E \)

First order
\[
\frac{d^2}{dt^2} r_1 + 2\gamma \frac{d}{dt} r_1 + \omega_0^2 r_1 = -\frac{e}{m} E
\]

Second order
\[
\frac{d^2}{dt^2} r_2 + 2\gamma \frac{d}{dt} r_2 + \omega_0^2 r_2 = \xi r_1^2
\]

General form of the field:
\[
E = \sum E(\omega_n)e^{-i\omega_n t}
\]

Calculate:
\[
\frac{d}{dt} r_1 = \frac{d^2}{dt^2} r_1
\]

Insert in (**):
\[
r_1 = -\frac{e}{m} \sum E(\omega_n)e^{-i\omega_n t}
\frac{m}{(\omega_0 - \omega_n)^2 - 2i\gamma\omega_n}
\]

Calculate \( r_1 \) and insert in (**): use
\[
(\sum E(\omega_n)e^{i\omega_n t})^2 = \sum \sum E(\omega_n)E(\omega_m)e^{-i(\omega_n + \omega_m)t}
\]

\[
r_2 = -\frac{e \xi}{m^2} \frac{1}{(\omega_0 - \omega_n)^2 - 2i\gamma\omega_n} 1
\frac{m}{(\omega_0 - \omega_n)(\omega_0 - \omega_m)^2 - 2i\gamma(\omega_n + \omega_m)}
\]

Write polarization:
\[
P = \sum P_k \quad P_k = -N r_k
\]

\[
P_{linear} = \sum \chi^{(1)}(\omega_n)E(\omega_n)e^{-i\omega_n t}
\]

\[
P_{second} = \sum \sum \chi^{(2)}(\omega_n, \omega_m)E(\omega_n)E(\omega_m)e^{-i(\omega_n + \omega_m)t}
\]

Linear and nonlinear susceptibilities:
\[
\chi^{(1)}(\omega_n) = \frac{Ne^2}{m} \frac{1}{(\omega_0 - \omega_n)^2 - 2i\gamma\omega_n}
\]

\[
\chi^{(2)}(\omega_n, \omega_m) = \frac{Ne^3 \xi}{m^2} \frac{1}{(\omega_0 - \omega_n)^2 - 2i\gamma\omega_n}
\frac{1}{(\omega_0 - \omega_m)^2 - 2i\gamma(\omega_n + \omega_m)}
\]

Verify:
\[
\chi^{(2)}(\omega_n, \omega_m) = -\frac{m}{N^2e^3} \chi^{(1)}(\omega_n)\chi^{(1)}(\omega_n)\chi^{(1)}(\omega_n + \omega_m)
\]
Maxwell’s equations for nonlinear optics

Starting point:
\[ \nabla \times \vec{E} = \frac{\partial \vec{B}}{\partial t} \quad \nabla \times \vec{H} = \vec{j} + \frac{\partial \vec{D}}{\partial t} \]
\[ \nabla \cdot \vec{D} = \rho \quad \nabla \cdot \vec{B} = 0 \]

with
\[ \vec{D} = \varepsilon_0 \vec{E} + \vec{P} \quad \vec{j} = \sigma \vec{E} \]

Induced polarization:
\[ \vec{P} = \varepsilon_0 \chi \vec{E} + \vec{P}^{NL} \]

Insert in Maxwell's equation
\[ \varepsilon = \varepsilon_0 (1 + \chi) \]
\[ \nabla \times \vec{H} = \sigma \vec{E} + \varepsilon \frac{\partial \vec{E}}{\partial t} + \frac{\partial \vec{P}^{NL}}{\partial t} \]

Use the equation for \( \vec{E} \)
\[ \nabla \times \nabla \times \vec{E} = -\frac{\partial}{\partial t} \left( \nabla \times \vec{B} \right) = -\mu \frac{\partial}{\partial t} \left( \nabla \times \vec{H} \right) \]
\[ = -\mu \frac{\partial}{\partial t} \left( \sigma \vec{E} + \varepsilon \frac{\partial \vec{E}}{\partial t} + \frac{\partial \vec{P}^{NL}}{\partial t} \right) \]

Use the vector relation:
\[ \nabla \times \nabla \times \vec{E} = \nabla \left( \nabla \cdot \vec{E} \right) - \nabla^2 \vec{E} \]

And (no charges in medium) \( \nabla \cdot \vec{E} = 0 \)

Maxwell’s wave equation in nonlinear optics

This equation for SI units
\[ \vec{P}(n) = \varepsilon_0 \chi^{(n)} \vec{E}^{(n)} \]
in C/m²

Often used esu units
\[ \vec{P}(n) = \chi^{(n)} \vec{E}^{(n)} \]
in statvolt/cm

\[ \frac{\chi^{(n)}_{SI}}{\chi^{(n)}_{esu}} = 4\pi / \left( 10^{-4} c \right)^{n-1} \]
\[ \frac{P^{(n)}_{SI}}{P^{(n)}_{esu}} = \frac{10^3}{c} \]
Input waves, *plane waves*, at frequencies
\[ \omega_1 \quad \omega_2 \]
\[ \tilde{E}(t) = \text{Re}[E(\omega_1)\exp(i\omega_1 t) + E(\omega_2)\exp(i\omega_2 t)] \]

Polarization at the **sum**-frequency:
\[ P_i(\omega_1 + \omega_2) = \]
\[ \text{Re}[\chi_{ijk}(\omega = \omega_1 + \omega_2)E_j(\omega_1)E_k(\omega_2)\exp[i(\omega_1 + \omega_2)t]] \]

and at the **difference**-frequency:
\[ P_i(\omega_1 - \omega_2) = \]
\[ \text{Re}[\chi_{ijk}(\omega = \omega_1 - \omega_2)E_j(\omega_1)E_k^*(\omega_2)\exp[i(\omega_1 - \omega_2)t]] \]

Notation: \( E_k(-\omega_2) = E_k^*(\omega_2) \)
\( \chi_{ijk}(\omega = \omega_1 + \omega_2) \) and \( \chi_{ijk}(\omega = \omega_1 - \omega_2) \)
are material properties of the medium

Use Maxwell’s equation
\[ \nabla^2 \tilde{E} = \mu \sigma \frac{\partial \tilde{E}}{\partial t} + \mu \varepsilon \frac{\partial^2 \tilde{E}}{\partial t^2} + \mu \frac{\partial^2 \tilde{P}_{NL}}{\partial t^2} \]
- take one component of linear polarization
- propagate plane wave along \( z \)-axis
\[ E_1(z,t) = E_1(z)\exp(i\omega_1 t - ik_1 z) \]
\[ E_2(z,t) = E_2(z)\exp(i\omega_2 t - ik_2 z) \]

Producing a non-linear polarization at sum.
\[ P_{NL}(z,t) = dE_1(z)E_2(z) \times \exp[i(\omega_1 + \omega_2)t - i(k_1 + k_2)z] \]

A new field is created at \( \omega_3 = \omega_1 + \omega_2 \)
\[ E_3(z,t) = E_3(z)\exp(i\omega_3 t - ik_3 z) \]

All this is substituted into Maxwell’s equation
\[ \nabla^2 E_3(z,t) = \frac{d^2}{dz^2} E_3(z) \]
Again
\[
\nabla^2 \ddot{E} - \mu \sigma \frac{\partial \ddot{E}}{\partial t} - \mu \varepsilon \frac{\partial^2 \ddot{E}}{\partial t^2} = \mu \frac{\partial^2}{\partial t^2} \bar{p}_{NL}
\]

Substitute left side:
\[
\begin{align*}
\frac{d^2}{dz^2} E_3(z,t) - \mu \sigma \frac{d}{dt} E_3(z,t) - \mu \varepsilon \frac{d^2}{dt^2} E_3(z,t) &= \\
\frac{d^2}{dz^2} E_3(z,t) + 2ik_3 \frac{d}{dz} E_3(z,t) - k_3^2 E_3(z,t) &= \\
+ i \omega_3 \mu \sigma E_3(z,t) + \mu \varepsilon \omega_3^2 E_3(z,t)
\end{align*}
\]

**Slowly varying amplitude approximation**
\[
\left| \frac{d^2}{dz^2} E_3(z,t) \right| \ll \left| 2ik_3 \frac{d}{dz} E_3(z,t) \right|
\]

Variation of the amplitude of the distance of a wavelength is small

For plane waves in a medium;
\[
\mu \varepsilon \omega_3^2 - k_3^2 = \frac{\omega_3^2}{c^2} - k_3^2 = 0
\]

So left side of wave equation;
\[
2ik_3 \frac{d}{dz} E_3(z,t) + i \omega_3 \mu \sigma E_3(z,t)
\]

**Right side of wave equation**
\[
\mu \frac{\partial^2}{\partial t^2} \bar{p}_{NL} =
\]
\[
\mu \frac{d^2}{dt^2} dE_1(z) E_2(z) \exp[i(\omega_1 + \omega_2)t - i(k_1 + k_2)z] =
\]
\[
- \mu (\omega_1 + \omega_2)^2 dE_1(z) E_2(z) \exp[i(\omega_1 + \omega_2)t - i(k_1 + k_2)z]
\]
Equate left and right side and use:

\[ \omega_3 = ck_3 \quad \omega_3 = \omega_1 + \omega_2 \]

Then:

\[ \frac{d}{dz} E_3(z) = -\frac{\sigma}{2} \sqrt{\frac{\mu}{\varepsilon_3}} E_3(z) - \frac{i\omega_3}{2} \sqrt{\frac{\mu}{\varepsilon_3}} dE_1(z)E_2(z) \exp[-i(k_1 + k_2 - k_3)z] \]

This is a coupled-wave equation.

Also reverse processes occur: \( \omega_3 - \omega_2 \rightarrow \omega_1 \)

Leading to other coupled equations

\[ \frac{d}{dz} E_1(z) = -\frac{\sigma}{2} \sqrt{\frac{\mu}{\varepsilon_1}} E_1(z) - \frac{i\omega_1}{2} \sqrt{\frac{\mu}{\varepsilon_1}} dE_3(z)E_2(z) \ast \exp[-i(k_3 - k_2 - k_1)z] \]

\[ \frac{d}{dz} E_2(z) = -\frac{\sigma}{2} \sqrt{\frac{\mu}{\varepsilon_2}} E_2(z) + \frac{i\omega_2}{2} \sqrt{\frac{\mu}{\varepsilon_2}} dE_1(z)E_3(z) \ast \exp[-i(k_1 + k_2 - k_3)z] \]

Three differential equations describe the couplings of the fields.

Note that we used cancellation of the frequency terms via:

\[ \omega_3 = \omega_1 + \omega_2 \]

But this does not hold for the spatial phase factors, because:

\[ \omega_i = \frac{k_i}{\sqrt{\mu \varepsilon(\omega_i)}} = \frac{ck_i}{n(\omega_i)} \]

Hence:

\[ k_1 + k_2 - k_3 \neq 0 \]

There is a phase-mismatch because of dispersion in the medium.

Define the wave vector mismatch:

\[ \Delta\vec{k} = \vec{k}_3 - \vec{k}_1 - \vec{k}_2 \]

This relation pertains to plane waves; Later we will use focused beams.
Second harmonic generation

Use a single input field:

\[ E_1(z) = E_2(z) \]

Then:

\[ \frac{d}{dz} E_3(z) = -\frac{\sigma}{2} \sqrt{\frac{\mu}{\varepsilon_3}} E_3(z) - \frac{i \omega_3}{2} \sqrt{\frac{\mu}{\varepsilon_3}} dE_1^2(z) e^{-i(2k_1-k_3)z} \]

Assume now:

- There is a nonlinearity \( d \) (only for certain symmetry)
- No absorption in the medium, so \( \sigma = 0 \)
- Only little production of wave \( \omega_3 \), so no back-conversion
- Wave vector mismatch is

\[ \Delta k = k^{(2\omega)} - 2k^{(\omega)} \]

The coupled wave equation can be integrated:

\[ E^{(2\omega)}(z) = -i \omega \sqrt{\frac{\mu}{\varepsilon^{(2\omega)}}} dE^2(\omega) | e^{i\Delta k z} dz \]

Conditions
1) Integration for 0 to \( L \) (length of medium)
2) And boundary \( E^{(2\omega)}(0) = 0 \)

Result of integration:

\[ E^{(2\omega)}(L) = -\omega \sqrt{\frac{\mu}{\varepsilon^{(2\omega)}}} dE^2(\omega) e^{i2kL} - 1 \]

Output of second harmonic is:

\[ E^{(2\omega)}(L) E^{(2\omega)}(L)^* = \frac{\omega^2 \mu}{n^2 \varepsilon_0} d^2 | E(\omega) |^4 L^2 \frac{\sin^2 \left( \frac{\Delta k L}{2} \right)}{\left( \frac{\Delta k L}{2} \right)^2} \]

Power at second harmonic:

\[ P^{(2\omega)} \propto \omega^2 d^2 L^2 \frac{\sin^2 \left( \frac{\Delta k L}{2} \right)}{\left( \frac{\Delta k L}{2} \right)^2} P(\omega)^2 \]

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Second harmonic power; conditions

Conversion efficiency:

\[ \eta_{SHG} = \frac{P(2\omega)}{P(\omega)} \propto \omega^2 d^2 L^2 \sin^2 \left( \frac{\Delta k L}{2} \right) \frac{p(\omega)}{A} \]

1) Second harmonic produced is proportional to

\[ P(2\omega) \propto P(\omega)^2 \]

nonlinear power production

2) Efficiency is proportional to \( d^2 \) or

\[ |\chi^{(2)}|^2 \]

3) Efficiency is proportional to \( L^2 \)
and a sinc function

\[ \eta_{SHG} \propto L^2 \sin \left( \frac{\Delta k L}{2} \right) \]

4) Efficiency is optimal if

\[ \Delta k = 0 \]

This is the “phase-matching condition”
cannot be met, because:

\[ k^{(2\omega)} \neq 2k^{(\omega)} \]

Use:

\[ k = \frac{n\omega}{c} \]

\[ k^{(2\omega)} = \frac{2n^{(2\omega)}\omega}{c}, \quad 2k^{(\omega)} = \frac{2n^{(\omega)}\omega}{c} \]

And dispersion in the medium:

\[ n^{(2\omega)} > n^{(\omega)} \]

So always \( \Delta k \neq 0 \)

Physics: two waves with

\[ E_\omega(z,t) = E_\omega \exp[i\omega t - ik^{(\omega)}z] \]
\[ E_{2\omega}(z,t) = E_{2\omega} \exp[2i\omega t - ik^{(2\omega)}z] \]

will run out of phase
Coherence length and Maker fringes

After a distance the waves will run out of phase

$$\Delta k l = \pi$$

Then the amplitude is at maximum. The wave will die out in:

$$L_c = 2l$$

The coherence length:

$$L_c = \frac{2\pi}{\Delta k} = \frac{2\pi}{k(2\omega) - 2k(\omega)} = \frac{\pi c}{2\omega (n(2\omega) - n(\omega))} = \frac{\lambda}{4(n(2\omega) - n(\omega))}$$

Typical values

$$\lambda = 1\mu m$$

$$n(2\omega) - n(\omega) \approx 10^{-2}$$

$$L_c = 25\mu m$$

Experiment:


Maker fringes

Only effective length of $L_c$ can be used (Note: non-sinusoidal behavior due to “non-critical phase matching”)

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Intermezzo

Solution to problem: anisotropic media

Induced polarization in a medium:

\[ \vec{P} = \varepsilon_0 \chi \vec{E} \]

Susceptibility is tensor of rank 2, causing the \( P \) and \( E \) vectors to have different directions

\[ P_1 = \varepsilon_0 (\chi_{11} E_1 + \chi_{12} E_2 + \chi_{13} E_3) \]
\[ P_2 = \varepsilon_0 (\chi_{21} E_1 + \chi_{22} E_2 + \chi_{23} E_3) \]
\[ P_3 = \varepsilon_0 (\chi_{31} E_1 + \chi_{32} E_2 + \chi_{33} E_3) \]

Elements of tensor depend on coordinate frame;

\[ \vec{D} = \varepsilon_0 \vec{E} + \vec{P} = \varepsilon_0 (1 + \chi_{ij}) \vec{E} = \varepsilon_{ij} \vec{E} \]

With permittivity tensor \( \varepsilon_{ij} \)

Monochromatic plane wave with perpendicular:

\[ \vec{E} \exp[i \omega t - i \vec{k} \cdot \vec{r}] \]
\[ \vec{H} \exp[i \omega t - i \vec{k} \cdot \vec{r}] \]

Wavefront vector

\[ \vec{k} = \frac{n \omega}{c} \vec{s} \]

Maxwell's equations (non-magnetic media)

\[ \vec{\nabla} \times \vec{E} = -\frac{\partial \vec{B}}{\partial t} \]
\[ \vec{\nabla} \times \vec{H} = \frac{\partial \vec{D}}{\partial t} \]

Derivatives:

\[ \vec{\nabla} \rightarrow -i \vec{k} = -i \frac{n \omega}{c} \vec{s} \quad \quad \frac{\partial}{\partial t} \rightarrow i \omega \]

For the plane waves:

\[ \vec{k} \times \vec{E} = \mu_0 \omega \vec{H} \quad \vec{k} \times \vec{H} = -\omega \vec{D} \]

Two vectors orthogonal to \( k \)

\[ \vec{k} \perp \vec{H} \quad \vec{k} \perp \vec{D} \]
Intermezzo

**Group and Phase velocity**

\[ \vec{S} = \vec{E} \times \vec{H} \]

The Poynting vector is not along the \( k \)-vector.

Group Velocity is not equal to Phase Velocity
- in magnitude
- in direction

\( H \) and \( D \) are perpendicular to the wave vector.
Verify:
\[ \vec{E} \perp \vec{H} \]
Further
\[ \vec{D} = \varepsilon \vec{E} \]

If \( \varepsilon \) is a scalar then \( D \) and \( E \) are parallel, but this is not the case in general.
Fresnel equations

Verify: \(-\vec{k} \times \vec{k} \times \vec{E} = \vec{\nabla} \times \vec{\nabla} \times \vec{E} = \omega^2 \mu \vec{D}\)

Use: \(\vec{k} \times \vec{k} \times \vec{E} = \vec{k}(\vec{k} \cdot \vec{E}) - \vec{E}(\vec{k} \cdot \vec{k})\)

\(\vec{D} = n^2 \varepsilon_0 [\vec{E} - \vec{s}(\vec{s} \cdot \vec{E})]\)

Choose coordinate frame \((x,y,z)\) along principal dielectric axes

\[
\begin{pmatrix}
D_x \\
D_y \\
D_z
\end{pmatrix} =
\begin{pmatrix}
\varepsilon_x & 0 & 0 \\
0 & \varepsilon_y & 0 \\
0 & 0 & \varepsilon_z
\end{pmatrix}
\begin{pmatrix}
E_x \\
E_y \\
E_z
\end{pmatrix}
\]

Permittivities \(\varepsilon_i\) differ along axes

\(D_i = n^2 \varepsilon_0 \left[ \frac{D_i}{\varepsilon_i} - \left( \vec{s} \cdot \vec{E} \right) \right] \)

Hence: \(D_i = \frac{\varepsilon_0 (\vec{s} \cdot \vec{E})}{\frac{1}{n^2} - \frac{\varepsilon_0}{\varepsilon_i}}\)

Form the scalar product \(\vec{s} \cdot \vec{D} = 0\)

Fresnel's equation

\[
\frac{s_x^2}{n^2 - \varepsilon_x} + \frac{s_y^2}{n^2 - \varepsilon_y} + \frac{s_z^2}{n^2 - \varepsilon_z} = 0
\]

Equation is quadratic in \(n\) and will have two solutions \(n'\) and \(n''\)

Two waves \(D'(n')\) and \(D''(n'')\) obey the equation

\[
D'^2 = \varepsilon_0^2 (\vec{s} \cdot \vec{E})^2 \left( \sum_{x,y,z} \frac{s^2}{\frac{1}{n^2} - \frac{\varepsilon_0}{\varepsilon_x}} \right)
\]

\[
= \varepsilon_0^2 (\vec{s} \cdot \vec{E})^2 \left( \frac{(n'n'')^2}{n^2 - n'^2} \right) \left( \sum_{x,y,z} \left[ \frac{s^2}{\frac{1}{n^2} - \frac{\varepsilon_0}{\varepsilon_x}} + \frac{s^2}{\frac{1}{n^2} - \frac{\varepsilon_0}{\varepsilon_y}} \right] \right)
\]

Summation \(\alpha\) is over \(x,y,z\)

\(\vec{D'} \cdot \vec{D''} = 0\)

Anisotropic crystal can transmit two waves with perpendicular parallel polarizations (and any linear combination of these two)
Incident beam is always decomposed into two eigenmodes of the anisotropic crystal

\[ \vec{D}'(n') \quad \vec{D}''(n'') \]

These modes are orthogonal to each other. Each of the two modes undergoes refraction with its index \( n' \) or \( n'' \)

Hence:

\[ k_0 \sin \theta_0 = k_1 \sin \theta_1 = k_2 \sin \theta_2 \]

This is:

Double refraction

Birefringence
The index ellipsoid

Energy stored in an electric field in a medium:

\[ U_e = \frac{1}{2} (\mathbf{E} \cdot \mathbf{D}) \]

With: \( D_i = \varepsilon_i E_i \)

\[ \frac{D_x^2}{\varepsilon_x} + \frac{D_y^2}{\varepsilon_y} + \frac{D_z^2}{\varepsilon_z} = 2U_e \]

This is a surface (ellipsoid) of constant energy

Define a normalized polarization vector:

\[ \vec{r} = \hat{D} \sqrt{2U_e} \]

Index ellipsoid:

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

Three-dimensional body to find two indices of refraction for the two waves \( \mathbf{D} \)

Uni-axial crystal:

\[ n_0^2 = \frac{\varepsilon_x}{\varepsilon_0} = \frac{\varepsilon_y}{\varepsilon_0} \quad n_e^2 = \frac{\varepsilon_z}{\varepsilon_0} \]

Index becomes:

\[ \frac{x^2}{n_0^2} + \frac{y^2}{n_0^2} + \frac{z^2}{n_e^2} = 1 \]
Birefringent media

For an arbitrary angle:

\[ x = n_0 \quad y = n_e(\theta)\cos\theta \quad z = n_e(\theta)\sin\theta \]

Projection of the ellipsoid on x=0

\[ \frac{y^2}{n_0^2} + \frac{z^2}{n_e^2} = 1 \]

Insert:

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

So index depends on propagation of wave vector (\(\theta\))

Birefringence  

- \(n_e > n_0\) positive
- \(n_e < n_0\) negative

Two allowed polarization directions
- one polarized along the x-axis; polarization vector perpendicular to the optic axis ordinary wave; it transmits with index \(n_0\).
- one polarized in the x-y plane but perpendicular to s; polarization vector in the plane with the optic axis is called the extraordinary wave.
Phase matching in Birefringent media

There exists an ordinary wave with $n_0$

And an extra-ordinary wave with

$$n_e(\theta) = \frac{n_enn_0}{\sqrt{n_o^2 \sin^2 \theta + n_e^2 \cos^2 \theta}}$$

Both undergo dispersion

Phase-matching, or $\Delta k = 0$ can be reached now; required is

$$n^\omega = n^{2\omega}$$

In case of (for KDP) $n_e < n_0$

$$n_e^{2\omega}(\theta_m) = n_o^\omega$$

Equation to find the phase-matching angle:

$$n_e^{2\omega}(\theta_m) = \frac{n_e^{2\omega}n_o^{2\omega}}{\sqrt{(n_o^{2\omega})^2 \sin^2 \theta_m + (n_e^{2\omega})^2 \cos^2 \theta_m}}$$

Solve for $\sin \theta$

$$\sin^2 \theta_m = \frac{(n_o^\omega)^{-2} - (n_o^{2\omega})^{-2}}{(n_e^{2\omega})^{-2} - (n_o^{2\omega})^{-2}}$$
Phase matching in Birefringent media

Graphical: index ellipsoid including dispersion

\[
\sin^2 \theta_m = \frac{(n_\omega^2 - n_o^2)^2}{(n_e^2 - n_o^2)^2}
\]

**TYPE I phase matching**
- \(E_o^\omega + E_o^\omega \rightarrow E_e^{2\omega}\) negative birefringence
- \(E_e^\omega + E_e^\omega \rightarrow E_o^{2\omega}\) positive birefringence

**TYPE II phase matching**
- \(E_o^\omega + E_e^\omega \rightarrow E_e^{2\omega}\) negative birefringence
- \(E_o^\omega + E_e^\omega \rightarrow E_o^{2\omega}\) positive birefringence

Type I \(\rightarrow\) polarization of second harmonic is perpendicular to fundamental
Type II \(\rightarrow\) can be understood as sumfrequency mixing
Phase matching and the "opening angle"

Consider Type I phase-matching and a negatively birefringent crystal. Phase matching

\[ \Delta k = \frac{2\omega}{c} [n_e^{2\omega}(\theta) - n_o^{2\omega}] = 0 \]

This works for a certain angle \( \theta_m \). Near this angle a Taylor series

\[
\frac{d\Delta k}{d\theta} = \frac{2\omega}{c} \frac{d}{d\theta} \left[ n_e^{2\omega}(\theta) - n_o^{2\omega} \right] = \\
\frac{2\omega}{c} \frac{d}{d\theta} \frac{n_e n_o}{\sqrt{n_o^2 \sin^2 \theta + n_e^2 \cos^2 \theta}} = \\
- \frac{\omega}{c} \frac{n_e n_o}{\left(n_o^2 \sin^2 \theta + n_e^2 \cos^2 \theta\right)^{3/2}} \left(n_o^2 - n_e^2\right) \sin 2\theta \\
= - \frac{\omega}{c} \frac{n_e^{2\omega}(\theta)}{n_o^2 n_e^2} \left(n_o^2 - n_e^2\right) \sin 2\theta
\]

with: \( n_e^{2\omega}(\theta) = n_o^{2\omega} \)

\[
\left| \frac{d\Delta k}{d\theta} \right| \left| \theta_m \right| = - \frac{\omega}{c} n_0^3 \left(n_e^{-2} - n_o^{-2}\right) \sin 2\theta_m
\]

Spread in \( k \)-values relates to spread in \( \Delta \theta \)

\[
\Delta k = \frac{2\beta}{L} \Delta \theta \quad \text{with} \quad \beta \propto \sin 2\theta_m
\]

\[
P^{(2\omega)}(\theta) \propto \frac{\sin^2 \frac{\Delta k L}{2}}{\left( \frac{\Delta k L}{2} \right)^2} \propto \frac{\sin^2 [\beta(\theta - \theta_m)]}{[\beta(\theta - \theta_m)]^2}
\]

Opening angle:
1) Interpret as angle - 0.1° - of collimated beam
2) As a divergence (convergence) of a laser beam
3) As a wavelength spread

\[
\frac{\Delta k}{k} = - \frac{\Delta \lambda}{\lambda}
\]
phase matching by angle tuning

For the example of LiIO₃

Dispersion:

<table>
<thead>
<tr>
<th>λ</th>
<th>n₀</th>
<th>nₑ</th>
</tr>
</thead>
<tbody>
<tr>
<td>4000</td>
<td>1.948</td>
<td>1.780</td>
</tr>
<tr>
<td>4360</td>
<td>1.931</td>
<td>1.766</td>
</tr>
<tr>
<td>5000</td>
<td>1.908</td>
<td>1.754</td>
</tr>
<tr>
<td>5300</td>
<td>1.901</td>
<td>1.750</td>
</tr>
<tr>
<td>5780</td>
<td>1.888</td>
<td>1.742</td>
</tr>
<tr>
<td>6900</td>
<td>1.875</td>
<td>1.731</td>
</tr>
<tr>
<td>8000</td>
<td>1.868</td>
<td>1.724</td>
</tr>
<tr>
<td>10600</td>
<td>1.860</td>
<td>1.719</td>
</tr>
</tbody>
</table>

Use dispersion and phase-matching relation:

\[ \sin^2 \theta_m = \frac{\left(n_\omega^2\right)^{-2} - \left(n_{2\omega}^2\right)^{-2}}{\left(n_e^2\right)^{-2} - \left(n_{2\omega}^2\right)^{-2}} \]

Calculate phase matching angle

Practical issue of limitation:

LiIO₃ starts absorbing at 295 nm
Non-critical phase matching and temperature tuning

Opening angle for wave vectors:

$$\Delta k = \frac{2\beta}{L} \Delta \theta \quad \beta \propto \sin 2\theta_m$$

Best if $\theta_m = 90^\circ$

Calculation of Type I for temperatures

Advantages of 90° phase matching

1) Poynting vector coincides with phase vector so no "walk-off"

2) The first order derivative in Taylor expansion

$$\frac{d\Delta k}{d\theta} = -\frac{\omega}{c} \left( n_e^2 \omega(\theta) \right)^3 \left( n_o^2 - n_e^2 \right) \sin 2\theta_m$$

$\longrightarrow 0$

Hence non-critical phase matching:

$$\Delta k \propto (\Delta \theta)^2$$

3) In many cases $d$ is larger at $\theta_m = 90^\circ$
Quasi phase matching by periodic poling

Fundamental and harmonic run out of phase in conversion processes. 
→ Coherence length is limited

Periodic poling

Manufacturing of segments by external fields During/after growth

Stick segments of material together with opposite optical axes- crystal modulation. 
Change of sign of polarization in each $L_c$ 
→ Coherence “runs back”
Quasi phase matching: analysis

Coupled wave equation, with \( \Gamma = i \omega E_1^2 / n_2 c \)

\[
\frac{d}{dz} E_2 = \Gamma d(z) \exp[-i \Delta k' z]
\]

Integrate for second harmonic

\[
E_2(L) = \Gamma \int_0^L d(z) \exp[-i \Delta k' z] dz
\]

d(z) consists of domains with alternating signs

\[
E_2 = \frac{i \Gamma d_{\text{eff}}}{\Delta k'} \sum_{k=1}^N g_k \left[ \exp(-i \Delta k' z_k) - \exp(-i \Delta k' z_{k-1}) \right]
\]

Sign changes (should) occur at: \( e^{-i \Delta k_0} z_{k,0} = (-1)^k \)

\( \Delta k_0' \) wave vector mismatch at design wavelength

For m\textsuperscript{th} order QPM: \( z_{k,0} = mk l_c \)

\[
E_{2,\text{ideal}} \approx i \Gamma d_{\text{eff}} \frac{2}{m \pi} L
\]

\[
E_2(L) = \Gamma d_{\text{eff}} L \quad \text{for perfect phase matching}
\]

Loss factor: \( \frac{2}{m \pi} \)

A: perfect phase matching
C: phase mismatch for non-poling
B\textsubscript{1}: poling at \( L_c \)
B\textsubscript{3}: poling after \( 3 L_c \)
Intermezzo

Pump depletion in SHG

In case of high conversion also reverse processes play a role:

\[ \omega_1 + \omega_2 \rightarrow \omega_3 \quad \omega_3 - \omega_1 \rightarrow \omega_2 \]

\[ \omega_3 - \omega_2 \rightarrow \omega_1 \]

Define amplitudes and assume no absorption

\[ A_i = \frac{n_i}{\omega_i} E_i \quad \kappa = d \frac{1}{2} \sqrt{\frac{\mu_0}{\varepsilon_0} \sqrt{\omega_1 \omega_2 \omega_3}} \]

Then coupled wave equations turn to coupled amplitude equations

\[ \frac{d}{dz} A_1 = -i \kappa A_2 A_2^* e^{-i \Delta k z} \]

\[ \frac{d}{dz} A_2 = +i \kappa A_1 A_3^* e^{i \Delta k z} \]

\[ \frac{d}{dz} A_3 = -i \kappa A_1 A_2 e^{i \Delta k z} \]

Assume second harmonic generation \( \Delta k = 0 \); no field with \( A_2 \); field \( A_1 \) is degenerate \( A_1 A_2 = \frac{1}{2} A_1^2 \)

Rewrite: \( A_3' = -i A_3 \)

Then:

\[ \frac{d}{dz} A_1 = -\kappa A_3' A_1 \quad \frac{d}{dz} A_3' = \frac{1}{2} \kappa A_1^2 \]

Calculate:

\[ \frac{d}{dz} \left[ A_1^2 + 2(A_3'(z))^2 \right] = 2 A_1 \frac{d}{dz} A_1 + 4 A_3' \frac{d}{dz} A_3' = 0 \]

So in crystal: \( A_1^2 + 2(A_3'(z))^2 = \text{constant} = A_1^2(0) \)

Consider:

\[ I_i = \frac{1}{2} \sqrt{\frac{\mu_0}{\varepsilon_0} n_i |E_i|^2} = \frac{1}{2} \sqrt{\frac{\mu_0}{\varepsilon_0} \omega_i |A_i|^2} \quad I_i \propto N_i \hbar \omega_i \]

Hence: \#photons(\( \omega_1 \)) + 2\#photons(\( \omega_3 \)) = constant

Energy and photon numbers are conserved
Pump depletion in SHG - 2

Solve amplitude equation

\[ \frac{d}{dz} A_3' = -\frac{1}{2} \kappa \left[ A_1^2(0) - 2(A_3')^2 \right] = 0 \]

Solution:

\[ A_3'(z) = \frac{A_1(0)}{\sqrt{1/2}} \tanh \left[ \frac{A_1(0) \kappa z}{\sqrt{1/2}} \right] \]

Conversion efficiency

\[ \eta_{\text{SHG}} = \frac{P^{(2\omega)}}{P^{(\omega)}} = \frac{|A_3(z)|^2}{\frac{1}{2} |A_1(0)|^2} = \tanh^2 \left[ \frac{A_1(0) \kappa z}{\sqrt{1/2}} \right] \]

For:

\[ A_1(0) \kappa z \to \infty \quad |A_3'(z)|^2 \to \frac{1}{2} |A_1(0)|^2 \]
## Crystals and properties

<table>
<thead>
<tr>
<th>Material</th>
<th>Transparency range [nm]</th>
<th>Spectral range of phase matching of type I or II</th>
<th>Damage threshold [GW/cm²]</th>
<th>Relative doubling efficiency</th>
</tr>
</thead>
<tbody>
<tr>
<td>ADP</td>
<td>220–2000</td>
<td>500–1100</td>
<td>0.5</td>
<td>1.2</td>
</tr>
<tr>
<td>KD*P</td>
<td>200–2500</td>
<td>517–1500 (I)</td>
<td>8.4</td>
<td>1.0</td>
</tr>
<tr>
<td>KD*P</td>
<td></td>
<td>732–1500 (II)</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Urea</td>
<td>210–1400</td>
<td>473–1400 (I)</td>
<td>1.5</td>
<td>6.1</td>
</tr>
<tr>
<td>BBO</td>
<td>197–3500</td>
<td>410–3500 (I)</td>
<td>9.9</td>
<td>26.0</td>
</tr>
<tr>
<td>BBO</td>
<td></td>
<td>750–1500 (II)</td>
<td></td>
<td></td>
</tr>
<tr>
<td>LiJO₃</td>
<td>300–5500</td>
<td>570–5500 (I)</td>
<td>0.06</td>
<td>50.0</td>
</tr>
<tr>
<td>KTP</td>
<td>350–4500</td>
<td>1000–2500 (I)</td>
<td>1.0</td>
<td>215.0</td>
</tr>
<tr>
<td>LiNbO₃</td>
<td>400–5000</td>
<td>800–5000 (II)</td>
<td>0.05</td>
<td>105.0</td>
</tr>
<tr>
<td>LiB₃O₅</td>
<td>160–2600</td>
<td>550–2600</td>
<td>18.9</td>
<td>3</td>
</tr>
<tr>
<td>CdGeAs₂</td>
<td>1–20 µm</td>
<td>2–15 µm</td>
<td>0.04</td>
<td>9</td>
</tr>
<tr>
<td>AgGaSe₂</td>
<td>3–15 µm</td>
<td>3.1–12.8 µm</td>
<td>0.03</td>
<td>6</td>
</tr>
<tr>
<td>Tc</td>
<td>3.8–32 µm</td>
<td></td>
<td>0.045</td>
<td>270</td>
</tr>
</tbody>
</table>

\[ ADP = \text{Ammonium dihydrogen phosphate} \]
\[ KDP = \text{Potassium dihydrogen phosphate} \]
\[ KD*P = \text{Potassium dideuterium phosphate} \]
\[ KTP = \text{Potassium titanyl phosphate} \]
\[ KNO₃ = \text{Potassium niobate} \]
\[ LBO = \text{Lithium triborate} \]
\[ LiO₃ = \text{Lithium iodate} \]
\[ LiNbO₃ = \text{Lithium niobate} \]
\[ BBO = \text{Beta-barium borate} \]

---

### Lasers

**High power fixed wavelength Lasers**

- **Nd-YAG** 1064 nm
- **2nd** 532 nm
- **3rd** 355 nm
- **4th** 266 nm
- **5th** 212 nm

**Excimer lasers**

- **KrF** 248 nm
- **XeCl** 308 nm
- **ArF** 193 nm

**Dye Lasers**

- Tunable 400–750 nm

**Titanium:Sapphire Lasers**

- Tunable 760–900 nm
A tracking device for angle tuning based on the opening angle

This curve may be interpreted as SHG as a function of angle

Electronic scheme
Diodes in differential measurement

W. Ubachs - Advanced Experimental Methods; 2013 Part A
Optical Parametric Oscillation and Amplification
Optical Parametric Amplification

Consider a NLO-process
\[ \omega_3 \rightarrow \omega_1 + \omega_2 \]
Where a short-wavelength photon (pump) is converted into a photon at \( \omega_1 \) (signal) and a photon at \( \omega_2 \) (idler).

Start again from coupled wave equations:
- no absorption
- phase-matched \( \Delta k = 0 \)
- \( k \) defined
\[ \kappa = d \frac{1}{2} \sqrt{\frac{\mu_0}{\epsilon_0}} \sqrt{n_1 n_2 n_3} \]

This leads to coupled amplitudes
\[ \frac{dA_1}{dz} = -\frac{1}{2} i \kappa A_3 A_2^* e^{-i\Delta k z} \]
\[ \frac{dA_2^*}{dz} = \frac{1}{2} i \kappa A_1 A_3^* e^{i\Delta k z} \]

Assume no depletion of the pump:
\[ A_3(z) = A_3(0) \]

Define:
\[ g = \kappa A_3(0) \]

Coupled amplitudes:
\[ \frac{dA_1}{dz} = -\frac{1}{2} i g A_2^* \quad \frac{dA_2^*}{dz} = \frac{1}{2} i g A_1 \]

Boundary conditions: \( A_2(0) = 0 \) and \( A_1(0) = \text{small} \)
\[ A_1(z) = A_1(0) \cosh \left( \frac{gz}{2} \right) \]
\[ A_2^*(z) = A_1(0) \sinh \left( \frac{gz}{2} \right) \]

Approximation for \( gz > 0 \)
\[ |A_1(z)|^2 = |A_2(z)|^2 \propto e^{gz} \]

Both fields grow with gain factor: \( g \)
This parametric gain.

Verify that:
\[ -\frac{dA_3}{dz} = \frac{dA_1}{dz} = \frac{dA_2^*}{dz} \]
and
\[ -\Delta \left( \frac{P_3}{\omega_3} \right) = \Delta \left( \frac{P_1}{\omega_2} \right) = \Delta \left( \frac{P_2}{\omega_2} \right) \]

Manley-Rowe equations
Parametric oscillation

Principle: amplification starts from noise, as in laser

Assume again parametric gain with $\Delta k=0$

Steady state condition:

$$\frac{dA_1}{dz} = \frac{dA_2}{dz}$$

Retain losses (absorption or mirror losses)

$$-\frac{1}{2} \alpha_1 A_1 - \frac{1}{2} g A_2^* = 0$$

$$\frac{1}{2} g A_1 - \frac{1}{2} \alpha_2 A_2^* = 0$$

Nontrivial solution at threshold if:

$$g^2 = \alpha_1 \alpha_2$$

Above threshold if

$$g^2 > \alpha_1 \alpha_2$$

Gain > losses
Parameter is the phase-matching condition:

\[ \Delta \vec{k} = 0 \quad \vec{k}_3 = \vec{k}_1 + \vec{k}_2 \]

For co-linear beams this equals:

\[ n_3 \omega_3 = n_1 \omega_1 + n_2 \omega_2 \]

And energy conservation

\[ \omega_3 = \omega_1 + \omega_2 \]

Phase-matching again in birefringent crystals e.g., Type I

\[ n_3^e(\theta_m)\omega_3 = n_1 \omega_1 + n_2 \omega_2 \]

At each specific angle \( \theta_m \) the OPO will produce a combination of two frequencies \( \omega_1 \) and \( \omega_2 \)

Rotation of angle near \( \theta_m \) yields

\[ \theta_m \rightarrow \theta_m + \Delta \theta \]

\[ n_1 \rightarrow n_1 + \Delta n_1 \]
\[ n_2 \rightarrow n_2 + \Delta n_2 \]
\[ n_3 \rightarrow n_3 + \Delta n_3 \]

For fixed pump this gives

\[ \omega_1 \rightarrow \omega_1 + \Delta \omega_1 \quad \omega_2 \rightarrow \omega_2 + \Delta \omega_2 \]

Energy conservation:

\[ \Delta \omega_2 = -\Delta \omega_1 \]

Index \( n_3 \) changes if it is extra-ordinary

\[ \Delta n_3 = \frac{\partial n_3}{\partial \theta} \bigg|_{\theta_m} \Delta \theta \quad \text{angle dependence} \]

\[ \Delta n_1 = \frac{\partial n_1}{\partial \omega_1} \bigg|_{\omega_1} \Delta \omega_1 \quad \text{dispersion} \]

\[ \Delta n_2 = \frac{\partial n_2}{\partial \omega_2} \bigg|_{\omega_2} \Delta \omega_2 \quad \text{dispersion} \]

New phase-matching condition:

\[ (n_3 + \Delta n_3)\omega_3 = (n_1 + \Delta n_1)(\omega_1 + \Delta \omega_1) + (n_2 + \Delta n_2)(\omega_2 + \Delta \omega_2) \]

Use:

\[ \Delta \omega_2 = -\Delta \omega_1 \quad \text{and solve:} \]

\[ \Delta \omega_1 = \frac{\omega_3 \Delta n_3 - \omega_1 \Delta n_1 - \omega_2 \Delta n_2}{n_1 - n_2} \]
Then:

\[
\Delta \omega_1 = \frac{\omega_3 \frac{\partial n_3}{\partial \theta} \Delta \theta - \omega_1 \frac{\partial n_1}{\partial \omega_1} \Delta \omega_1 + \omega_2 \frac{\partial n_2}{\partial \omega_2} \Delta \omega_1}{n_1 - n_2}
\]

Solve for: \( \Delta \omega_1 \)

\[
\frac{\Delta \omega_1}{\Delta \theta} = \frac{\omega_3 \frac{\partial n_3}{\partial \theta}}{(n_1 - n_2) + [\omega_1 \frac{\partial n_1}{\partial \omega_1} - \omega_2 \frac{\partial n_2}{\partial \omega_2}]}
\]

Use the result for the calculation of the opening angle obtained previously (SHG)

\[
\left| \frac{\partial n_3}{\partial \theta} \right|_{\theta_m} = -\frac{1}{2} n_o^3 [n_e^{-2}(\omega_3) - n_o^{-2}(\omega_3)] \sin 2\theta_m
\]

This results in the angle tuning function:

\[
\frac{\partial \omega_1}{\partial \theta} = -\frac{1}{2} n_o^3 [n_e^{-2}(\omega_3) - n_o^{-2}(\omega_3)] \omega_3 \sin 2\theta_m
\]

\[
(n_1 - n_2) + \left[ \omega_1 \frac{\partial n_1}{\partial \omega_1} - \omega_2 \frac{\partial n_2}{\partial \omega_2} \right]
\]
SNLO - Public Domain Software for non-linear optics

http://www.as-photonics.com/SNLO

SNLO.Ink

[Image of a software interface for non-linear optics]
Practical problems

1) The $\text{EF}^1\Sigma_g^+$, $v=0$ state in the $\text{H}_2$ molecule can be excited via two-photon excitation. For this pulsed laser radiation at 202 nm is required. Devise schemes that make this possible using pulsed dye lasers in the visible domain. Devise a scheme based on a tunable titanium-sapphire laser delivering pulses in the range 780-850 nm.

2) The $\text{EF}^1\Sigma_g^+$, $v=6$ state in the $\text{H}_2$ molecule can be excited in two-photon at 193 nm. Devise a scheme to produce this radiation by taking one of colors from the fixed Nd-YAG laser and its harmonics.