Lecture Course

Advanced Experimental Methods

Non-linear optics in crystals

Wavelength conversion of laser light

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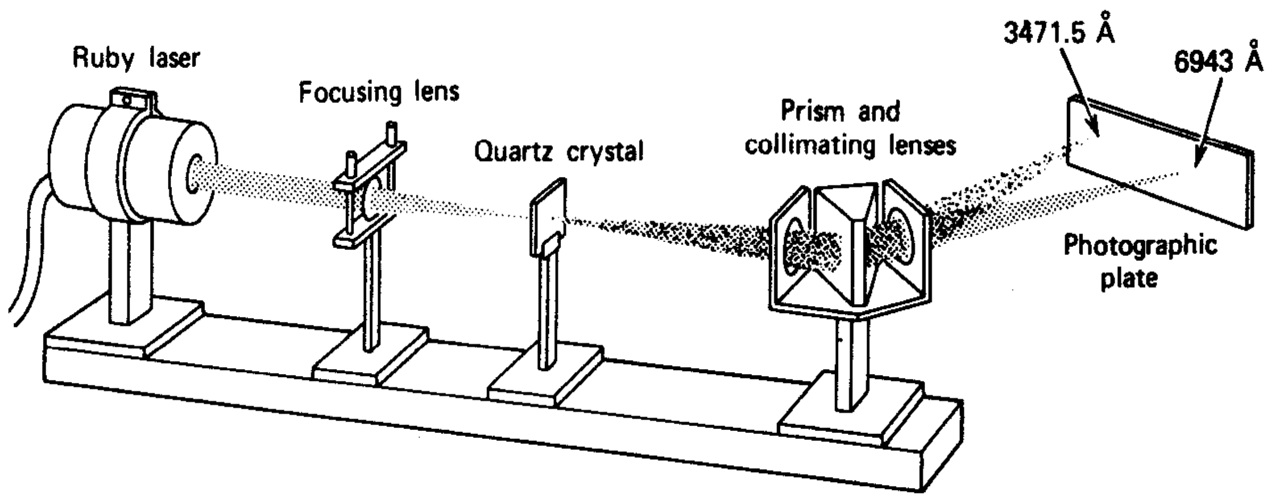
2013

Part A

Nonlinear Optics

Nicolaas Bloembergen
Nobel prize 1981

The first non-linear optical laser experiment



P.A. Franken, A.E. Hill, C.W. Peters and G. Weinreich, Phys. Rev. Lett. **7** (1961) 118

The Nonlinear Susceptibility

$$\vec{P} = \chi^{(1)}\vec{E} + \chi^{(2)}\vec{E}\vec{E} + \chi^{(3)}\vec{E}\vec{E}\vec{E} + \ldots$$

The $\chi_{ij}^{(n)}$ are tensors even for lowest order

$$P_i = \chi_{ij}^{(n)} E_j$$

Polarization not directed along electric field vector

In media with inversion symmetry:

$$I_{op}\vec{P} = -\vec{P} = -\chi^{(1)}\vec{E} - \chi^{(2)}\vec{E}\vec{E} - \chi^{(3)}\vec{E}\vec{E}\vec{E} - \ldots$$

$$I_{op}\vec{E} = -\vec{E}$$

Hence:

$$I_{op}\vec{P} = -\chi^{(1)}\vec{E} + \chi^{(2)}\vec{E}\vec{E} - \chi^{(3)}\vec{E}\vec{E}\vec{E} + \ldots$$

Conclude for centro-symmetric media:

$$\chi^{(2n)} = 0$$

Note that in principle there exist also nonlinear magnetic susceptibilities

Nonlinear Optics; graphically

Linear response:

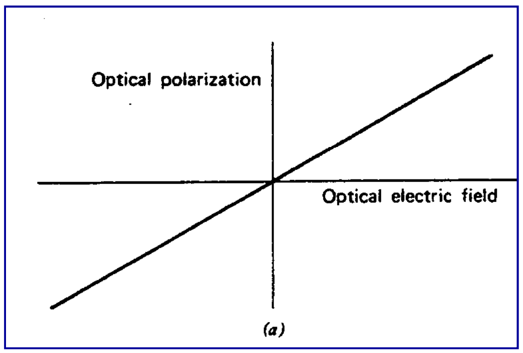
$$\vec{P} = \chi^{(1)} \vec{E}$$

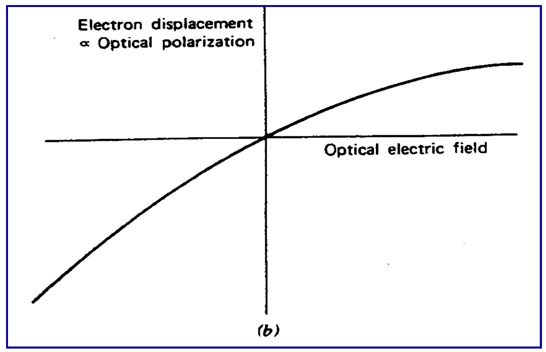
Nonlinear response:

$$\vec{P} = \chi^{(1)} \vec{E} + \chi^{(2)} \vec{E}\vec{E}$$

Nonlinear response evaluated in terms of Fourier series

DC





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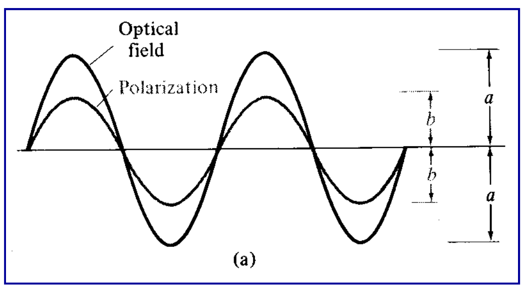

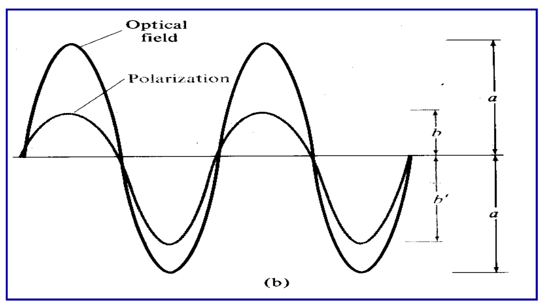

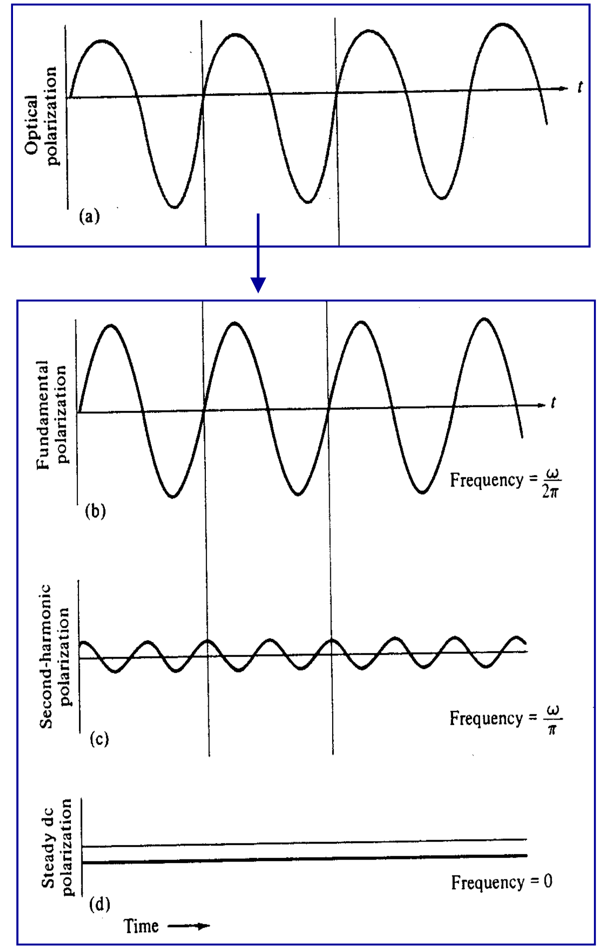

$$P = \sum a_n \sin(n\omega t + \phi_n)$$

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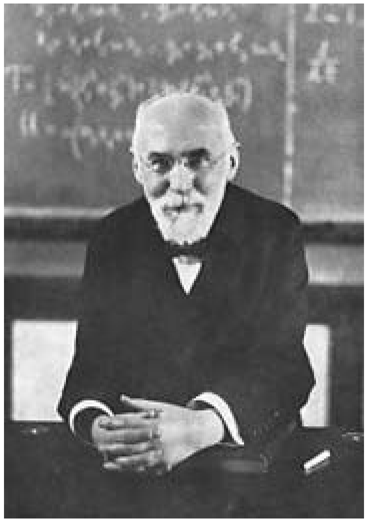
Intermezzo

Lorentz model of linear optics: classical oscillator



Equation of motion for a damped electronic oscillator in one dimension

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:

$$\mathbf{E} = \mathrm{Re}\left[E e^{i\omega t}\right] \qquad \mathbf{r} = \mathrm{Re}\left[r e^{i\omega t}\right]$$

$$\longrightarrow \quad \left(\omega_0^2 - \omega^2\right)r + 2i\omega\gamma r = -\frac{e}{m}E$$

Solution

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

Near resonance $\omega = \omega_0$

$$\longrightarrow$$

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

Intermezzo

Lorentz model of linear optics: classical oscillator

Classical polarization of the medium

$$P(\omega) = -Ner(\omega)$$

and the complex susceptibility

$$\chi(\omega) = \chi'(\omega) - i\chi''(\omega)$$

yields expressions for the susceptibility

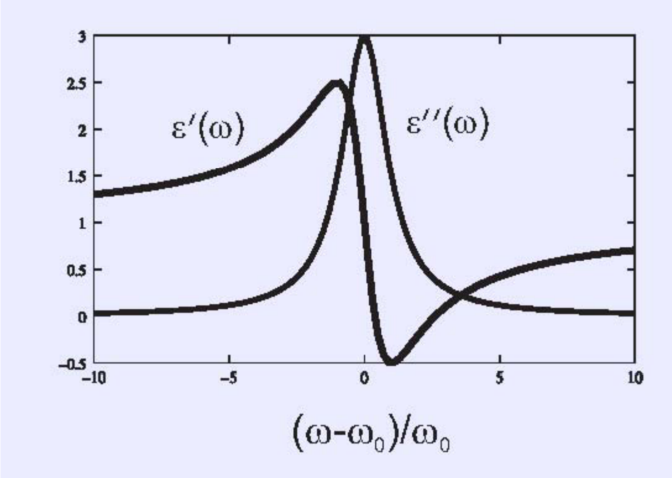
Real part,
connected to the index of refraction

$$\chi'(\omega) = \frac{Ne^2}{2m\omega_0\gamma\varepsilon_0} \frac{(\omega_0 - \omega)/\gamma}{\left[1 + (\omega_0 - \omega)^2/\gamma^2\right]}$$

Imaginary part,
connected to the absorption coefficient

$$\chi''(\omega) = \frac{Ne^2}{2m\omega_0\gamma\varepsilon_0} \frac{1}{\left[1 + (\omega_0 - \omega)^2/\gamma^2\right]}$$

Result:
Resonance features of driven electron



Dispersion and absorption

Note also the Kramers-Kronig relation

$$\chi'(\omega) = P\int_0^{+\infty} \frac{d\nu}{\pi}\chi''(\nu)\frac{2\nu}{\nu^2 - \omega^2}$$

Intermezzo

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

$$r = r_1 + r_2 + r_{3+\dots}$$

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: $\frac{d}{dt}r_1 \quad \frac{d^2}{dt^2}r_1$

Insert in (*)

$$r_1 = -\frac{e}{m}\frac{\sum E(\omega_n)e^{-i\omega_n t}}{(\omega_0 - \omega_n)^2 - 2i\gamma\omega_n}$$

Calculate r_1 and insert in (**); use

$$\left(\sum E(\omega_n)e^{i\omega_n t}\right)^2 = \sum\sum E(\omega_n)E(\omega_m)e^{-i(\omega_n+\omega_m)t}$$

$$r_2 = -\frac{e\xi}{m^2}\frac{\sum\sum E(\omega_n)E(\omega_m)e^{-i(\omega_n+\omega_m)t}}{\left[\omega_0^2 - \omega_n^2 - 2i\gamma\omega_n\right]\left[\omega_0^2 - \omega_m^2 - 2i\gamma\omega_m\right]\left[\omega_0^2 - (\omega_n+\omega_m)^2 - 2i\gamma(\omega_n+\omega_m)\right]}$$

Write polarization;

$$P = \sum P_k \qquad P_k = -Ner_k$$

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

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

Linear and nonlinear sucseptibilities:

$$\chi^{(1)}(\omega_n) = \frac{Ne^2}{m}\frac{1}{(\omega_0 - \omega_n)^2 - 2i\gamma\omega_n}$$

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

Verify: $\chi^{(2)}(\omega_n,\omega_m) = \frac{-m\xi}{N^2e^3}\chi^{(1)}(\omega_n)\chi^{(1)}(\omega_m)\chi^{(1)}(\omega_n+\omega_m)$

Maxwell's equations for nonlinear optics

Starting point:

$$\vec{\nabla}\times\vec{E} = -\frac{\partial\vec{B}}{\partial t} \qquad \vec{\nabla}\times\vec{H} = \vec{j} + \frac{\partial\vec{D}}{\partial t}$$

$$\vec{\nabla}\cdot\vec{D} = \rho \qquad \vec{\nabla}\cdot\vec{B} = 0$$

with

$$\vec{D} = \varepsilon_0\vec{E} + \vec{P} \qquad \vec{j} = \sigma\vec{E}$$

Induced polarization:

$$\vec{P} = \varepsilon_0\chi\vec{E} + \vec{P}^{NL}$$

Insert in Maxwells equation

$$\varepsilon = \varepsilon_0(1+\chi)$$

$$\vec{\nabla}\times\vec{H} = \sigma\vec{E} + \varepsilon\frac{\partial\vec{E}}{\partial t} + \frac{\partial\vec{P}^{NL}}{\partial t}$$

Use the equation for $\vec{E}$

$$\vec{\nabla}\times\vec{\nabla}\times\vec{E} = -\frac{\partial}{\partial t}\left(\vec{\nabla}\times\vec{B}\right) = -\mu\frac{\partial}{\partial t}\left(\vec{\nabla}\times\vec{H}\right)$$

$$= -\mu\frac{\partial}{\partial t}\left(\sigma\vec{E} + \varepsilon\frac{\partial\vec{E}}{\partial t} + \frac{\partial}{\partial t}\vec{P}^{NL}\right)$$

Use the vector relation:

$$\vec{\nabla}\times\vec{\nabla}\times\vec{E} = \vec{\nabla}\left(\vec{\nabla}\cdot\vec{E}\right) - \nabla^2\vec{E}$$

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

$$\nabla^2\vec{E} = \mu\sigma\frac{\partial\vec{E}}{\partial t} + \mu\varepsilon\frac{\partial^2\vec{E}}{\partial t^2} + \mu\frac{\partial^2}{\partial t^2}\vec{P}^{NL}$$

Maxwells 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

$$\frac{\chi_{SI}^{(n)}}{\chi_{esu}^{(n)}} = 4\pi/\left(10^{-4}c\right)^{n-1}$$

$$\frac{P_{SI}^{(n)}}{P_{esu}^{(n)}} = \frac{10^3}{c}$$

Coupled Wave Equations

Input waves, *plane waves*, at frequencies

$\omega_1 \qquad \omega_2$

$$\vec{E}(t) = \mathrm{Re}[E(\omega_1)\exp(i\omega_1 t) + E(\omega_2)\exp(i\omega_2 t)]$$

Polarization at the <u>sum</u>-frequency:

$$P_i(\omega_1+\omega_2) = \mathrm{Re}[\chi_{ijk}(\omega=\omega_1+\omega_2)E_j(\omega_1)E_k(\omega_2)\exp[i(\omega_1+\omega_2)t]]$$

and at the <u>difference</u>-frequency:

$$P_i(\omega_1-\omega_2) = \mathrm{Re}[\chi_{ijk}(\omega=\omega_1-\omega_2)E_j(\omega_1)E_k{}^*(\omega_2)\exp[i(\omega_1-\omega_2)t]]$$

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

$$\nabla^2\vec{E} = \mu\sigma\frac{\partial\vec{E}}{\partial t} + \mu\varepsilon\frac{\partial^2\vec{E}}{\partial t^2} + \mu\frac{\partial^2}{\partial t^2}\vec{P}^{NL}$$

- 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 subsituted into Maxwell's equation

and $\nabla^2 E_3(z,t) = \dfrac{d^2}{dz^2}E_3(z)$

Coupled Wave Equations - 2

Again

$$\nabla^2 \vec{E} - \mu\sigma \frac{\partial \vec{E}}{\partial t} - \mu\varepsilon \frac{\partial^2 \vec{E}}{\partial t^2} = \mu \frac{\partial^2}{\partial t^2} \vec{P}^{NL}$$

Substitute left side:

$$\frac{d^2}{dz^2} E_3(z,t) - \mu\sigma \frac{d}{dt} E_3(z,t) - \mu\varepsilon \frac{d^2}{dt^2} E_3(z,t) =$$

$$\frac{d^2}{dz^2} E_3(z,t) + 2ik_3 \frac{d}{dz} E_3(z,t) - k_3^2 E_3(z,t)$$

$$+ i\omega_3 \mu\sigma E_3(z,t) + \mu\varepsilon\omega_3^2 E_3(z,t)$$

Slowly varying amplitude approximation

$$\left| \frac{d^2}{dz^2} E_3(z,t) \right| << \left| 2ik_3 \frac{d}{dz} E_3(z,t) \right|$$

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

$$\frac{d^2}{dz^2} E_3(z,t) + 2ik_3 \frac{d}{dz} E_3(z,t) - k_3^2 E_3(z,t)$$

$$+ i\omega_3 \mu\sigma E_3(z,t) + \mu\varepsilon\omega_3^2 E_3(z,t)$$

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

$$\mu \frac{\partial^2}{\partial t^2} \vec{P}^{NL} =$$

$$\mu \frac{d^2}{dt^2} dE_1(z) E_2(z) \exp[i(\omega_1 + \omega_2)t - i(k_1 + k_2)z] =$$

$$-\mu(\omega_1 + \omega_2)^2 dE_1(z) E_2(z) \exp[i(\omega_1 + \omega_2)t - i(k_1 + k_2)z]$$

Coupled Wave Equations - 3

Equate left and right side and use:

$$\omega_3 = ck_3 \qquad \omega_3 = \omega_1 + \omega_2$$

Then:

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

This is a coupled-wave equation.
Also reverse processes occur: $\omega_3 - \omega_2 \to \omega_1$

Leading to other coupled equations

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

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

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 σ=0
- Only little production of wave ω_3, so no back-conversion
- Wave vector mismatch is

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

The coupled wave equation can be integrated:

$$E^{(2\omega)}(z) = -i\omega\sqrt{\frac{\mu}{\varepsilon^{(2\omega)}}}dE^2(\omega)\int e^{i\Delta kz}dz$$

Conditions

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

Result of integration:

$$E^{(2\omega)}(L) = -\omega\sqrt{\frac{\mu}{\varepsilon^{(2\omega)}}}dE^2(\omega)\frac{e^{i\Delta kL}-1}{\Delta k}$$

Output of second harmonic is:

$$E^{(2\omega)}(L)E^{(2\omega)}(L)^* =$$

$$\frac{\omega^2\mu}{n^2\varepsilon_0}d^2|E(\omega)|^4L^2\frac{\sin^2\left(\frac{\Delta kL}{2}\right)}{\left(\frac{\Delta kL}{2}\right)^2}$$

Power at second harmonic:

$$P^{(2\omega)} \propto \omega^2 d^2 L^2 \frac{\sin^2\left(\frac{\Delta kL}{2}\right)}{\left(\frac{\Delta kL}{2}\right)^2}\frac{P^{(\omega)2}}{A}$$

Second harmonic power; conditions

Conversion efficiency:

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

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

$$\left|\chi^{(2)}\right|^2$$

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

$$\eta_{SHG} \propto L^2 \operatorname{sinc}\left(\frac{\Delta k L}{2}\right)$$

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: $k = \frac{n\omega}{c}$

$$k^{(2\omega)} = \frac{2n^{(2\omega)}\omega}{c} \qquad 2k^{(\omega)} = \frac{2n^{(\omega)}\omega}{c}$$

And dispersion in the medium:

$$n^{(2\omega)} > n^{(\omega)}$$

So always $\Delta k \neq 0$

Physics: two waves with

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

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

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:

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

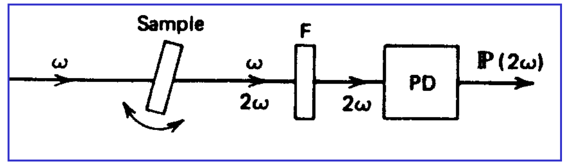
Typical values

$$\lambda = 1\mu m$$

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

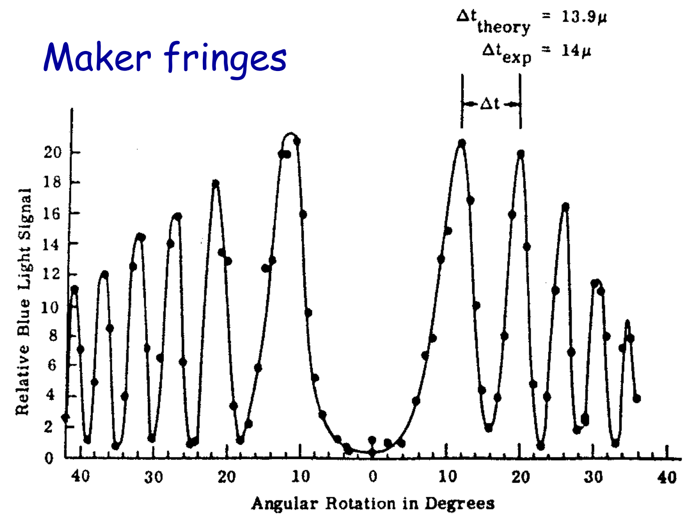
$$L_c = 25\mu m$$

Experiment:



P.D. Maker, R.W. Terhune, M. Nisenoff, and C. M. Savage, Phys. Rev. Lett. **8**, 19 (1962).

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

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

Elements of tensor depend on coordinate frame;

$$\vec{D} = \varepsilon_0 \vec{E} + \vec{P} = \varepsilon_0 \left(1 + \chi_{ij} \right) \vec{E} = \varepsilon_{ij} \vec{E}$$

With permitivity tensor $\vec{\vec{\varepsilon}}_{ij}$

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)

$$\vec{\nabla} \times \vec{E} = -\frac{\partial \vec{B}}{\partial t} \qquad \vec{\nabla} \times \vec{H} = \frac{\partial \vec{D}}{\partial t}$$

Derivatives:

$$\vec{\nabla} \rightarrow -i\vec{k} = -i\frac{n\omega}{c}\vec{s} \qquad \frac{\partial}{\partial t} \rightarrow i\omega$$

For the plane waves:

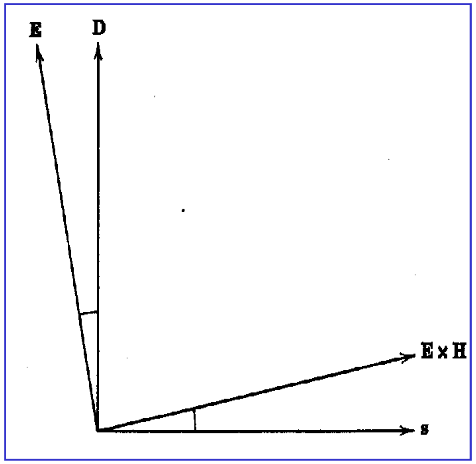
$$\vec{k} \times \vec{E} = \mu_0 \omega \vec{H} \qquad \vec{k} \times \vec{H} = -\omega \vec{D}$$

Two vectors orthogonal to k

$$\vec{k} \perp \vec{H} \qquad \vec{k} \perp \vec{D}$$

Intermezzo

Group and Phase velocity



H and D perpendicular to wave vector
Verify:

$$\vec{E} \perp \vec{H}$$

Further

$$\vec{D} = \vec{\vec{\varepsilon}}\vec{E}$$

If ε is a scalar then D and E parallel,
but this is not the case in general

Poynting vector: $\vec{S} = \vec{E} \times \vec{H}$

Is not along k-vector

→

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

Intermezzo

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: $\vec{k}\times\vec{k}\times\vec{E} = \vec{k}\left(\vec{k}\cdot\vec{E}\right) - \vec{E}\left(\vec{k}\cdot\vec{k}\right)$

$$\longrightarrow \quad \vec{D} = n^2\varepsilon_0\left[\vec{E} - \vec{s}\left(\vec{s}\cdot\vec{E}\right)\right]$$

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

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

$\longrightarrow$ Fresnel's equation

$$\frac{s_x^2}{\frac{1}{n^2} - \frac{\varepsilon_0}{\varepsilon_x}} + \frac{s_y^2}{\frac{1}{n^2} - \frac{\varepsilon_0}{\varepsilon_y}} + \frac{s_z^2}{\frac{1}{n^2} - \frac{\varepsilon_0}{\varepsilon_z}} = 0$$

Equation is quadratic in n and will have two solutions *n'* and *n"*

Two waves *D'(n')* and *D"(n")* obey the equation

$$\mathbf{D}'\cdot\mathbf{D}'' = \varepsilon_0^2\left(\mathbf{s}\cdot\mathbf{E}\right)^2\left\langle \sum_{x,y,z} \frac{s_\alpha^2}{\left(\frac{1}{n'^2} - \frac{\varepsilon_0}{\varepsilon_\alpha}\right)\left(\frac{1}{n''^2} - \frac{\varepsilon_0}{\varepsilon_\alpha}\right)} \right\rangle$$

$$= \varepsilon_0^2\left(\mathbf{s}\cdot\mathbf{E}\right)^2 \frac{\left(n'n''\right)^2}{\left(n'^2 - n''^2\right)} \left\langle \sum_{x,y,z}\left[\frac{s_\alpha^2}{\left(\frac{1}{n'^2} - \frac{\varepsilon_0}{\varepsilon_\alpha}\right)} + \frac{s_\alpha^2}{\left(\frac{1}{n''^2} - \frac{\varepsilon_0}{\varepsilon_\alpha}\right)} \right] \right\rangle$$

Summation α is over x,y,z

$$\longrightarrow \quad \vec{D}'\cdot\vec{D}'' = 0$$

Anisotropic crystal can transmit two waves with perpendicular parallel polarizations (and any linear combination of these two)

Refraction at boundary of anisotropic crystal

Incident beam is always decomposed into two eigenmodes of the anisotropic crystal

$$\vec{D}'(n') \qquad \vec{D}''(n'')$$

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

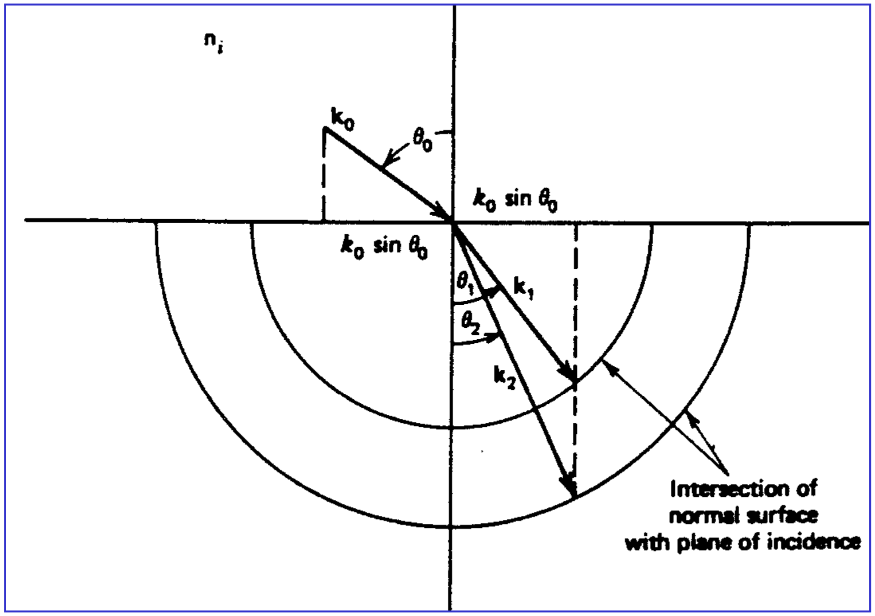
Hence:

$$k_0 \sin\theta_0 = k_1 \sin\theta_1 = k_2 \sin\theta_2$$

This is:

Double refraction

Birefringence



Intermezzo

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:

$$\vec{r} = \vec{D}\sqrt{2U_e}$$

Index ellipsoid:

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

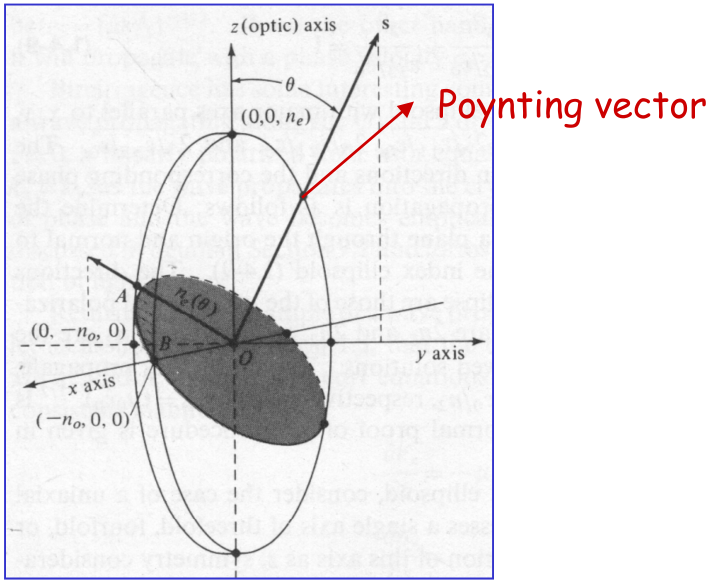
Three-dimensional body to find two indices of refraction for the two waves D

Uni-axial crystal:

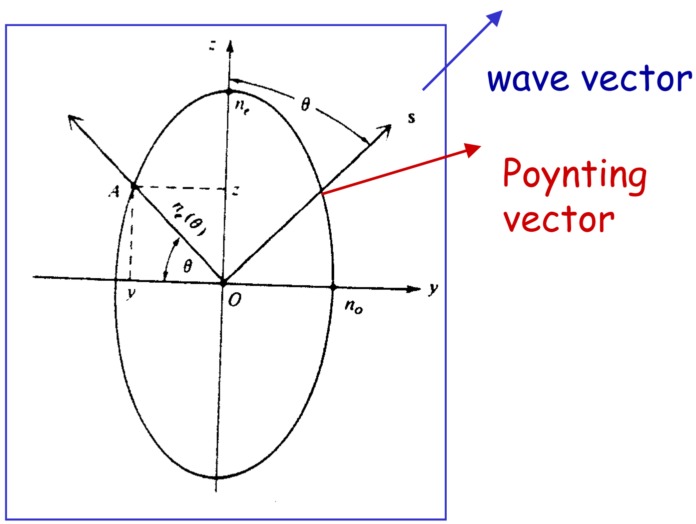
$$n_0^2 = \frac{\varepsilon_x}{\varepsilon_0} = \frac{\varepsilon_y}{\varepsilon_0} \qquad n_e^2 = \frac{\varepsilon_z}{\varepsilon_0}$$

Index becomes:

$$\frac{x^2}{n_0^2} + \frac{y^2}{n_0^2} + \frac{z^2}{n_e^2} = 1$$



Birefringent media



Two allowed polarization directions
-one polarized along the x-axis;
polarization vector perpendicular to the optic axis
ordinary wave; it transmits with index *no*.
-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*.

For an arbitrary angle:

$$x = n_0 \qquad y = n_e(\theta)\cos\theta \qquad z = n_e(\theta)\sin\theta$$

Projection of the ellipsoid on x=0

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

Insert:

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

So index depends on propagation
of wave vector (θ)

Birefringence	$n_e > n_0$	positive
	$n_e < n_0$	negative

Phase matching in Birefringent media

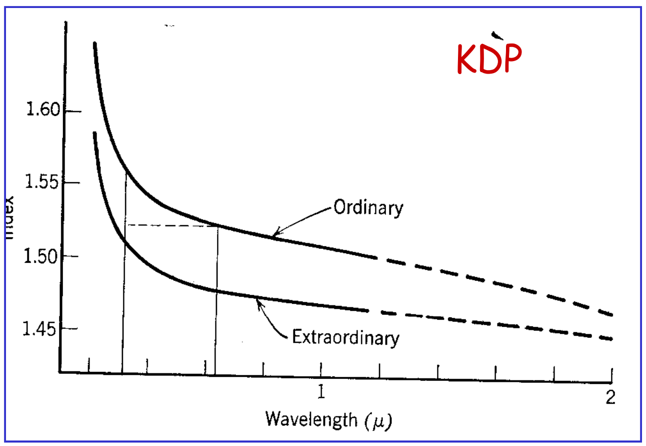
There exists an ordinary wave with

$$n_0$$

And an extra-ordinary wave with

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

Both undergo dispersion



Phase-matching, or Δk=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:

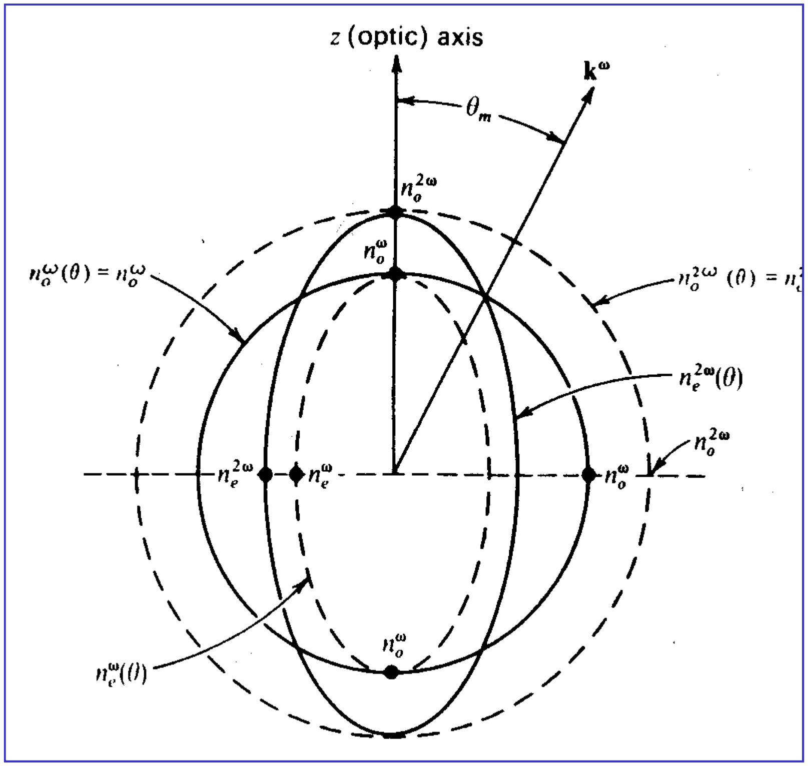
$$n_e^{2\omega}(\theta_m) = \frac{n_e^{2\omega} n_o^{2\omega}}{\sqrt{\left(n_o^{2\omega}\right)^2 \sin^2\theta_m + \left(n_e^{2\omega}\right)^2 \cos^2\theta_m}}$$

Solve for sinθ

$$\sin^2\theta_m = \frac{\left(n_o^{\omega}\right)^{-2} - \left(n_o^{2\omega}\right)^{-2}}{\left(n_e^{2\omega}\right)^{-2} - \left(n_o^{2\omega}\right)^{-2}}$$

Phase matching in Birefringent media

Graphical: index ellipsoid including dispersion



$$\sin^2\theta_m = \frac{\left(n_o^{\omega}\right)^{-2} - \left(n_o^{2\omega}\right)^{-2}}{\left(n_e^{2\omega}\right)^{-2} - \left(n_o^{2\omega}\right)^{-2}}$$

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 → polarization of second harmonic is perpendicular to fundamental

Type II → can be understood as sumfrequency mixing

Phase matching and the "opening angle"

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

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

This works for a certain angle θ_m.
Near this angle a Taylor series

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

$$\frac{2\omega}{c}\frac{d}{d\theta}\frac{n_e n_o}{\sqrt{n_o^2 \sin^2\theta + n_e^2 \cos^2\theta}} =$$

$$-\frac{\omega}{c}\frac{n_e n_o}{\left(n_o^2 \sin^2\theta + n_e^2 \cos^2\theta\right)^{3/2}}\left(n_o^2 - n_e^2\right)\sin 2\theta$$

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

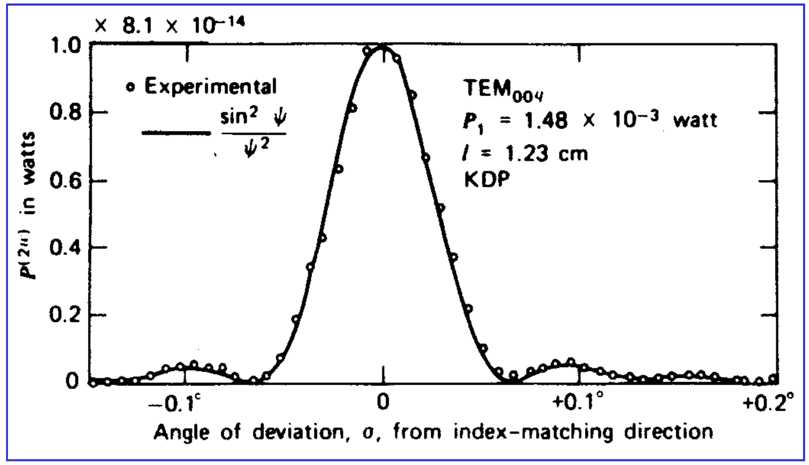
with: $n_e^{2\omega}(\theta) = n_o^{\omega}$

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

Spread in k-values relates to spread in $\Delta\theta$

$$\Delta k = \frac{2\beta}{L}\Delta\theta \qquad \text{with} \qquad \beta \propto \sin 2\theta_m$$

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



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 $LiIO_3$

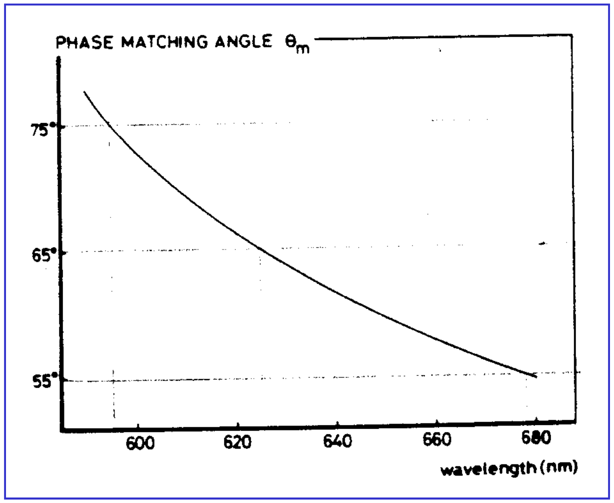
Dispersion:

A	n_o	n_e
4000	1.948	1.780
4360	1.931	1.766
5000	1.908	1.754
5300	1.901	1.750
5780	1.888	1.742
6900	1.875	1.731
8000	1.868	1.724
10600	1.860	1.719

Use dispersion and phase-matching relation:

$$\sin^2\theta_m = \frac{\left(n_o^{\omega}\right)^{-2} - \left(n_o^{2\omega}\right)^{-2}}{\left(n_e^{2\omega}\right)^{-2} - \left(n_o^{2\omega}\right)^{-2}}$$

Calculate phase matching angle



Practical issue of limitation:

$LiIO_3$ starts absorbing at 295 nm

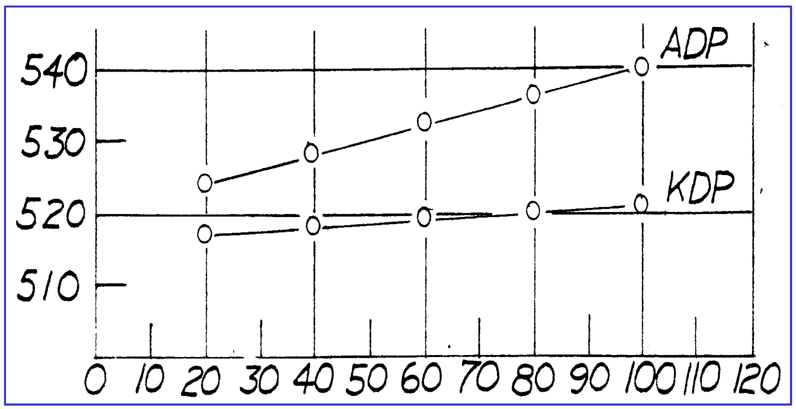
Non-critical phase matching and temperature tuning

Opening angle for wave vectors:

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

Best if $\theta_m = 90^o$

Calculation of Type I for temperatures



→ Temperature tuning

Advantages of 90° phase matching

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

2) The first order derivative in Taylor expansion

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

→ 0

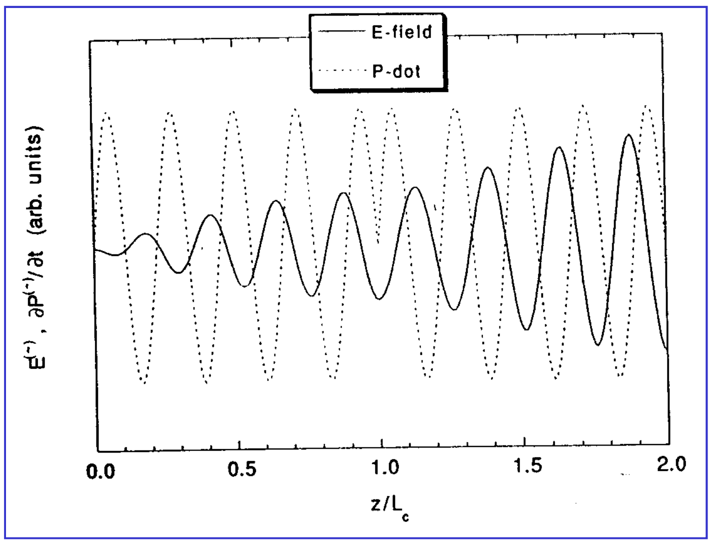
Hence non-critical phase matching:

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

3) In many cases d is larger at θ_m=90°

Quasi phase matching by periodic poling

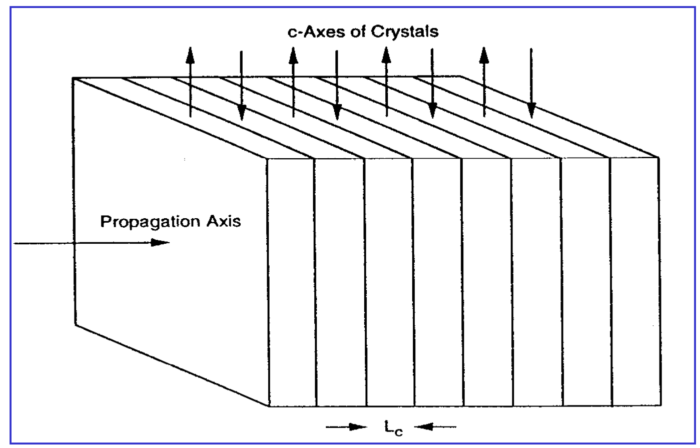
Fundamental and harmonic run out of phase
in conversion processes.
→ Coherence length is limited



Stick segments of material together with
opposite optical axes- crystal modulation.
Change of sign of polarization in each L_c
→ Coherence "runs back"

Periodic poling

Manufacturing of segments by external fields
During/after growth



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

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

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

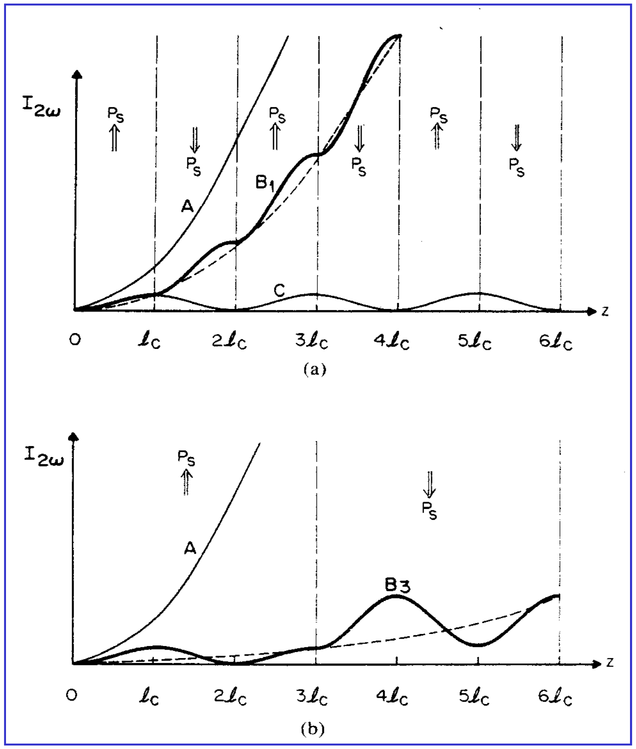
$\Delta k_0'$ wave vector mismatch at design wavelength

For m^th order QPM: $z_{k,0} = mkl_c$

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

$E_2(L) = \Gamma d_{eff} L$ for perfect phase matching

Loss factor: $\frac{2}{m\pi}$



A: perfect phase matching
C: phase mismatch for non-poling
B_1: poling at L_c
B3: poling after $3 L_c$

Intermezzo

Pump depletion in SHG

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

$$\omega_1 + \omega_2 \rightarrow \omega_3 \qquad \omega_3 - \omega_1 \rightarrow \omega_2$$

$$\omega_3 - \omega_2 \rightarrow \omega_1$$

Define amplitudes and assume no absorption

$$A_i = \frac{\sqrt{n_i}}{\omega_i} E_i \qquad \kappa = d\frac{1}{2}\sqrt{\frac{\mu_0}{\varepsilon_0}}\sqrt{\frac{\omega_1\omega_2\omega_3}{n_1 n_2 n_3}}$$

Then coupled wave equations turn to Coupled amplitude equations

$$\frac{d}{dz}A_1 = -i\kappa A_3 A_2^* e^{-i\Delta kz}$$

$$\frac{d}{dz}A_2 = +i\kappa A_1 A_3^* e^{i\Delta kz}$$

$$\frac{d}{dz}A_3 = -i\kappa A_1 A_2 e^{i\Delta kz}$$

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

Rewrite: $\quad A_3' = -iA_3$

Then:

$$\frac{d}{dz}A_1 = -\kappa A_3' A_1 \qquad \frac{d}{dz}A_3' = \frac{1}{2}\kappa A_1^2$$

Calculate:

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

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

Consider:

$$I_i = \frac{1}{2}\sqrt{\frac{\mu_0}{\varepsilon_0}}n_i\left|E_i\right|^2 = \frac{1}{2}\sqrt{\frac{\mu_0}{\varepsilon_0}}\omega_i\left|A_i\right|^2 \qquad I_i \propto N_i \hbar\omega_i$$

Hence: #photons(ω_1) + 2#photons(ω_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:

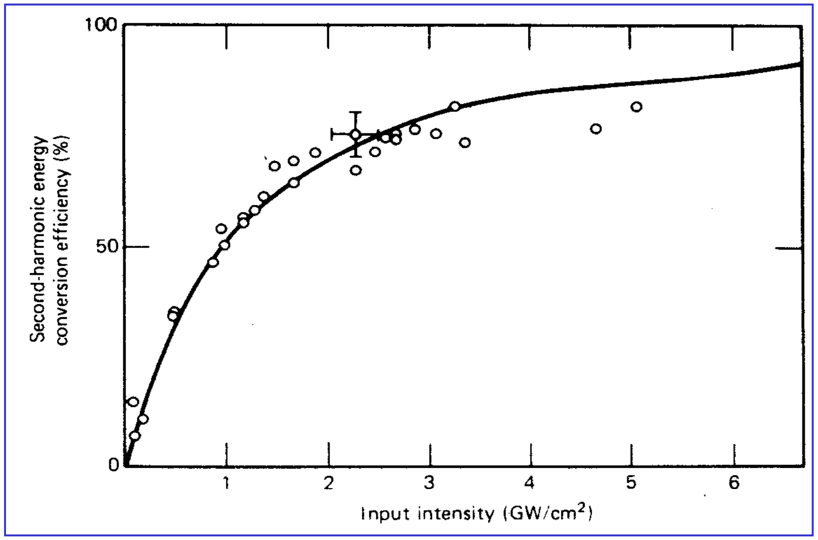
$$A_3'(z) = \frac{A_1(0)}{\sqrt{1/2}}\tanh\left[\frac{A_1(0)\kappa z}{\sqrt{1/2}}\right]$$

Conversion efficiency

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

For:

$$A_1(0)\kappa z \to \infty \qquad |A_3'(z)|^2 \to \frac{1}{2}|A_1(0)|^2$$



Crystals and properties

Material	Transparency range [nm]	Spectral range of phase matching of type I or II	Damage threshold [GW/cm^2]	Relative doubling efficiency
ADP	220–2000	500–1100	0.5	1.2
KD*P	200–2500	517–1500 (I)	8.4	1.0
		732–1500 (II)		8.4
Urea	210–1400	473–1400 (I)	1.5	6.1
BBO	197–3500	410–3500 (I)	9.9	26.0
		750–1500 (II)		
$LiJO_3$	300–5500	570–5500 (I)	0.06	50.0
KTP	350–4500	1000–2500 (II)	1.0	215.0
$LiNbO_3$	400–5000	800–5000 (II)	0.05	105.0
LiB_3O_5	160–2600	550–2600	18.9	3
$CdGeAs_2$	1–20 μm	2–15 μm	0.04	9
$AgGaSe_2$	3–15 μm	3.1–12.8 μm	0.03	6
Te	3.8–32 μm		0.045	270

ADP	= Ammonium dihydrogen phosphate	$NH_4H_2PO_4$
KDP	= Potassium dihydrogen phosphate	KH_2PO_4
KD*P	= Potassium dideuterium phosphate	KD_2PO_4
KTP	= Potassium titanyl phosphate	$KTiOPO_4$
$KNbO_3$	= Potassium niobate	$KNbO_3$
LBO	= Lithium triborate	LiB_3O_5
$LiIO_3$	= Lithium iodate	$LiIO_3$
$LiNbO_3$	= Lithium niobate	$LiNbO_3$
BBO	= Beta-barium borate	β-BaB_2O_4

Lasers

High power fixed wavelength Lasers

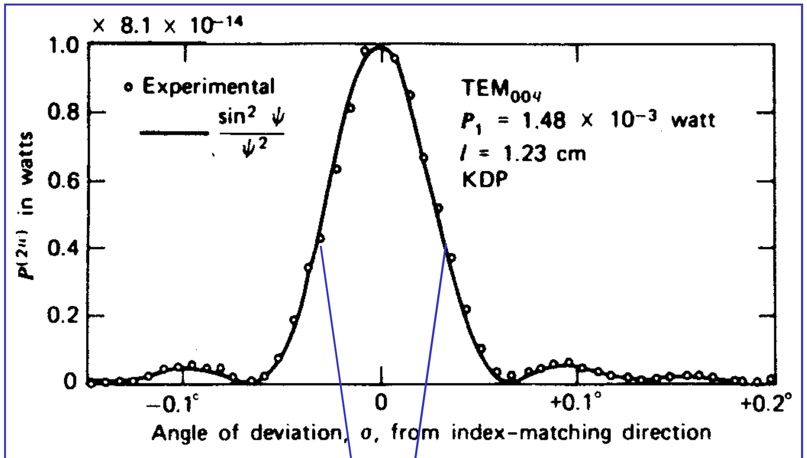
Nd-YAG 1064 nm
2nd 532 nm
3rd 355 nm
4th 266 nm
5th 212 nm

Excimer lasers
KrF 248 nm
XeCl 308 nm
ArF 193 nm

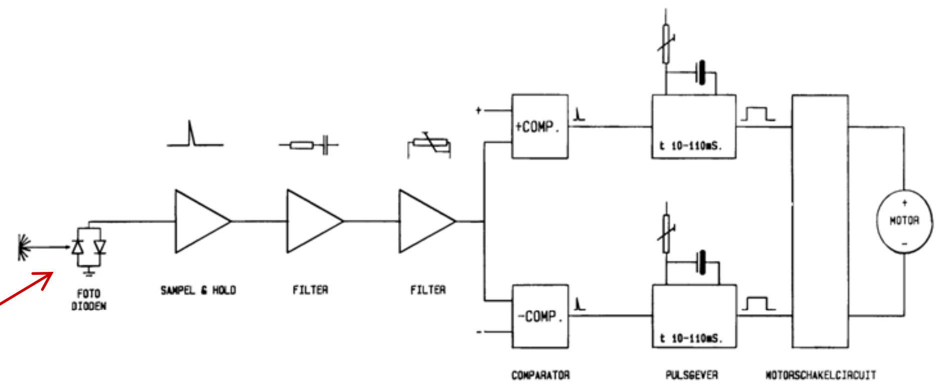
Dye Lasers
Tunable 400-750 nm

Titanium:Sapphire Lasers
Tunable 760-900 nm

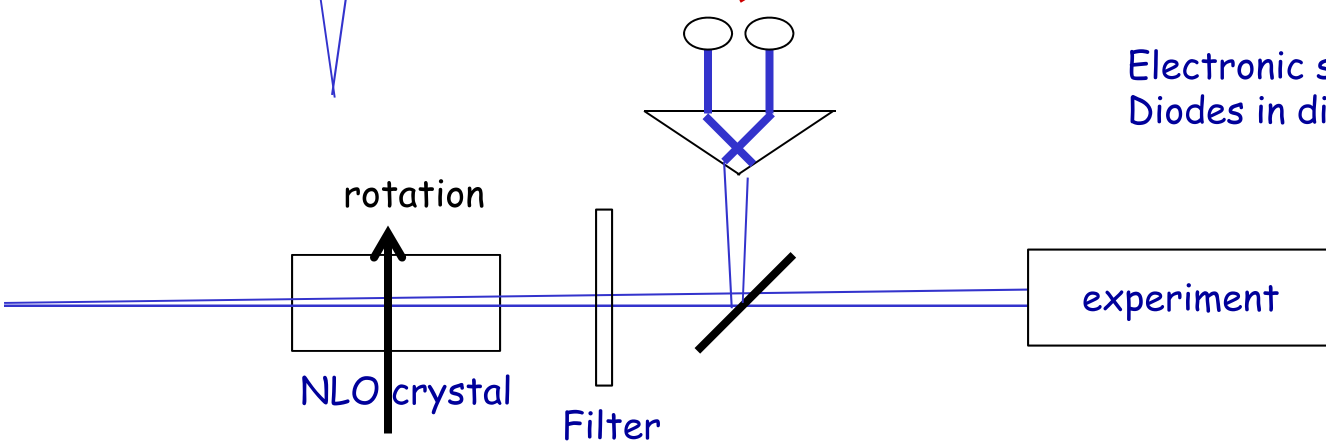
A tracking device for angle tuning based on the opening angle



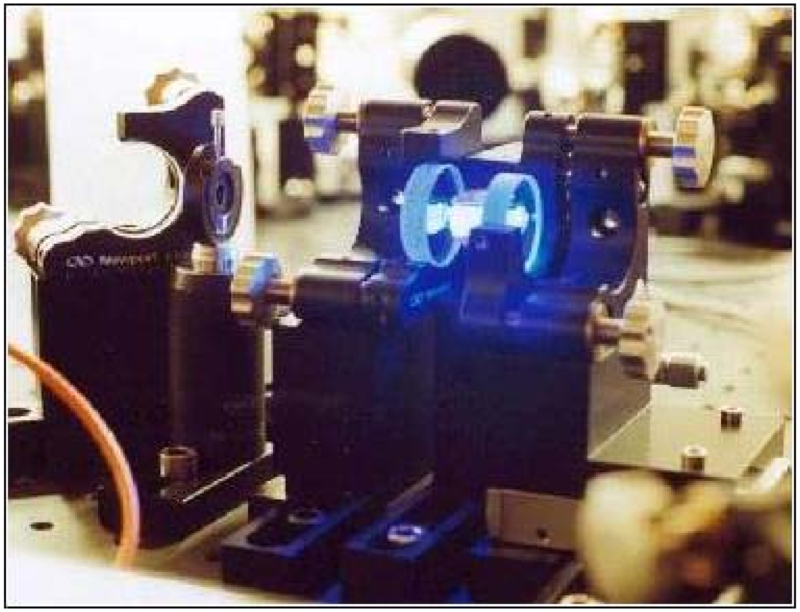
This curve may be interpreted as SHG as a function of angle

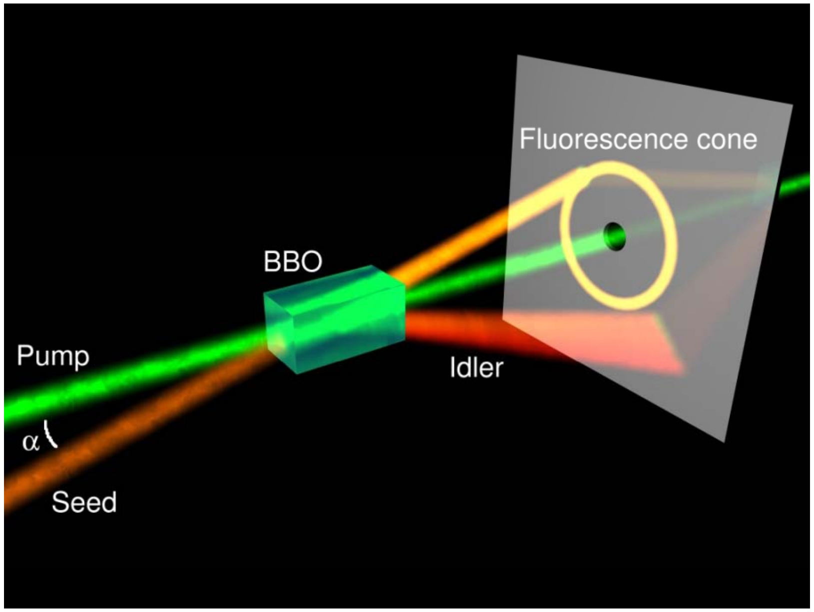


Electronic scheme
Diodes in differential measurement



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 ω_1 (signal) and a photon at ω_2 (idler).

Start again from coupled wave equations:
- no absorption
- phase-matched $\Delta k=0$
- k defined

$$\kappa = d\frac{1}{2}\sqrt{\frac{\mu_0}{\varepsilon_0}}\sqrt{\frac{\omega_1\omega_2\omega_3}{n_1 n_2 n_3}}$$

This leads to coupled amplitudes

$$\frac{dA_1}{dz} = -\frac{1}{2} i\kappa A_3 A_2^* e^{-i\Delta kz}$$

$$\frac{dA_2^*}{dz} = \frac{1}{2} i\kappa A_1 A_3^* e^{i\Delta kz}$$

Assume no depletion of the pump:

$$A_3(z) = A_3(0)$$

Define: $g = \kappa A_3(0)$

Coupled amplitudes:

$$\frac{dA_1}{dz} = -\frac{1}{2} igA_2^* \qquad \frac{dA_2^*}{dz} = \frac{1}{2} igA_1$$

Boundary conditions: $A_2(0)=0$ and $A_1(0)=small$

$$A_1(z) = A_1(0)\cosh\left(\frac{gz}{2}\right)$$

$$A_2^*(z) = A_1(0)\sinh\left(\frac{gz}{2}\right)$$

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.

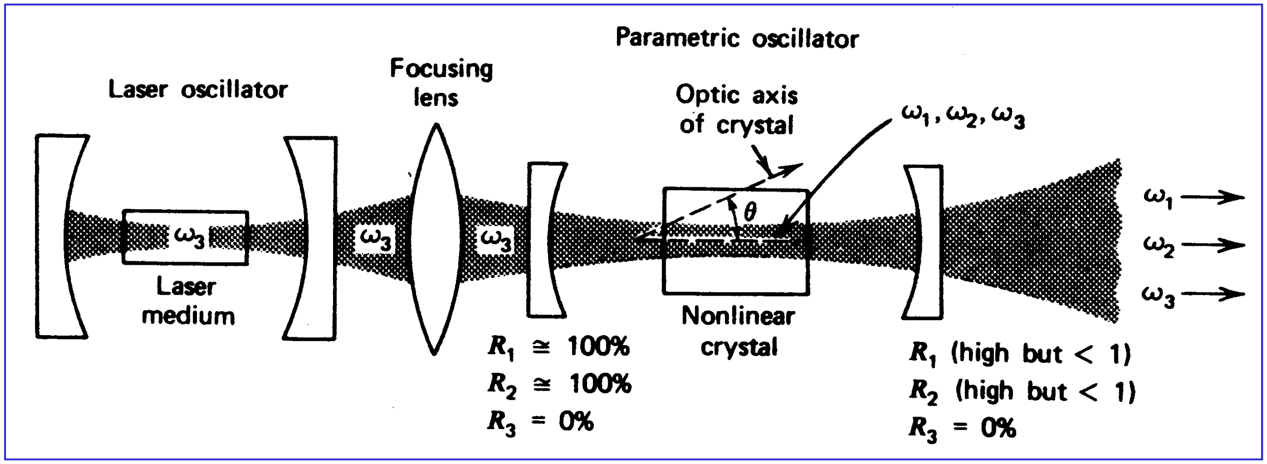
Verify that: $-\frac{dA_3A_3^*}{dz} = \frac{dA_1A_1^*}{dz} = \frac{dA_2A_2^*}{dz}$

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)

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

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

Nontrivial solution at threshold if:

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

Above threshold if

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

Gain > losses

Tuning of an OPO

Parameter is the phase-matching condition:

$$\Delta\vec{k} = 0 \qquad \vec{k}_3 = \vec{k}_1 + \vec{k}_2$$

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 θ_m the OPO will produce a combination of two frequencies ω_1 and ω_2

Rotation of angle near θ_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

$$\omega_1 \rightarrow \omega_1 + \Delta\omega_1 \qquad \omega_2 \rightarrow \omega_2 + \Delta\omega_2$$

Energy conservation; $\Delta\omega_2 = -\Delta\omega_1$

Index n_3 changes if it is extra-ordinary

$$\Delta n_3 = \left|\frac{\partial n_3}{\partial\theta}\right|_{\theta_m} \Delta\theta$$ angle dependence

$$\Delta n_1 = \left|\frac{\partial n_1}{\partial\omega_1}\right|_{\omega_1} \Delta\omega_1$$ dispersion

$$\Delta n_2 = \left|\frac{\partial n_2}{\partial\omega_2}\right|_{\omega_2} \Delta\omega_2$$ 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: $\Delta\omega_2 = -\Delta\omega_1$ and solve:

$$\Delta\omega_1 = \frac{\omega_3\Delta n_3 - \omega_1\Delta n_1 - \omega_2\Delta n_2}{n_1 - n_2}$$

Tuning of an OPO -2

Then:

$$\Delta\omega_1 = \frac{\omega_3\left|\frac{\partial n_3}{\partial\theta}\right|\Delta\theta - \omega_1\left|\frac{\partial n_1}{\partial\omega_1}\right|\Delta\omega_1 + \omega_2\left|\frac{\partial n_2}{\partial\omega_2}\right|\Delta\omega_1}{n_1 - n_2}$$

Solve for: $\Delta\omega_1$

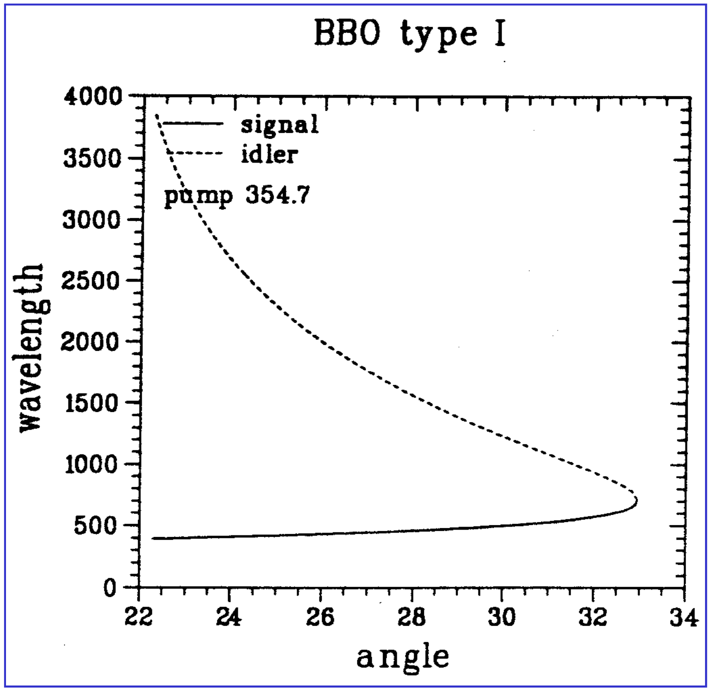
$$\frac{\Delta\omega_1}{\Delta\theta} = \frac{\omega_3\left|\frac{\partial n_3}{\partial\theta}\right|}{(n_1 - n_2) + \left[\omega_1\left|\frac{\partial n_1}{\partial\omega_1}\right| - \omega_2\left|\frac{\partial n_2}{\partial\omega_2}\right|\right]}$$

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

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

This results in the angle tuning fucntion:

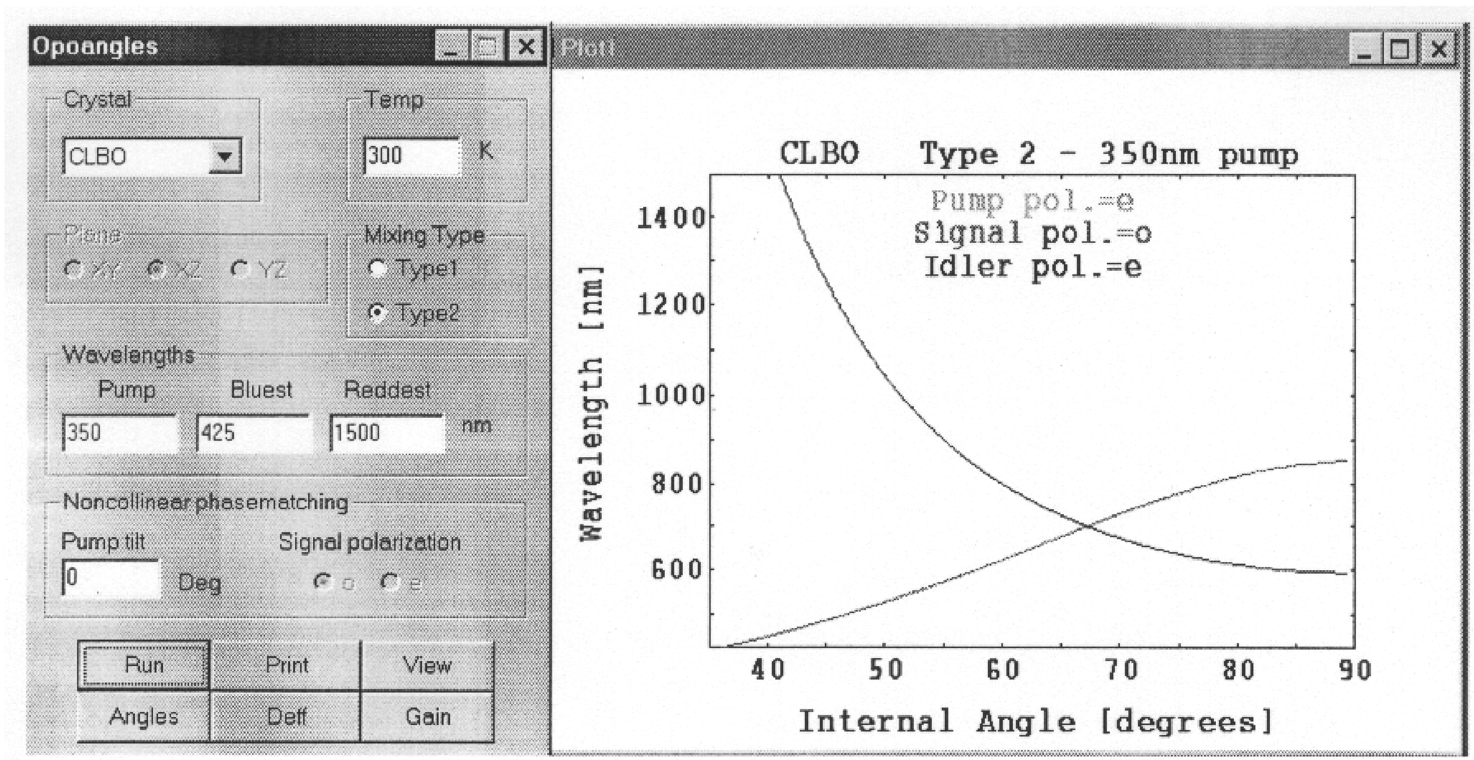
$$\frac{\partial\omega_1}{\partial\theta} = \frac{-\frac{1}{2}n_o^3\left[n_e^{-2}(\omega_3) - n_o^{-2}(\omega_3)\right]\omega_3\sin 2\theta_m}{(n_1 - n_2) + \left[\omega_1\frac{\partial n_1}{\partial\omega_1} - \omega_2\frac{\partial n_2}{\partial\omega_2}\right]}$$



SNLO – Public Domain Software for non-linear optics

SNLO.lnk

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



Practical problems

1) The $EF^1\Sigma_g^+$, v=0 state in the 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 $EF^1\Sigma_g^+$, v=6 state in the 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.