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Frequency Doubling and Second Order Nonlinear Optics

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Outline of the talk

- The first observation of second harmonic generation
- Nonlinear optical effects in solids
- The nonlinear wave equation (formulation of the interaction in nonlinear materials)
- Second order nonlinear optics
- Contracted notation for the second order susceptibility
- Three wave mixing
- Frequency doubling
- Phase matching in materials with birefringence

The first observation of second harmonic generation

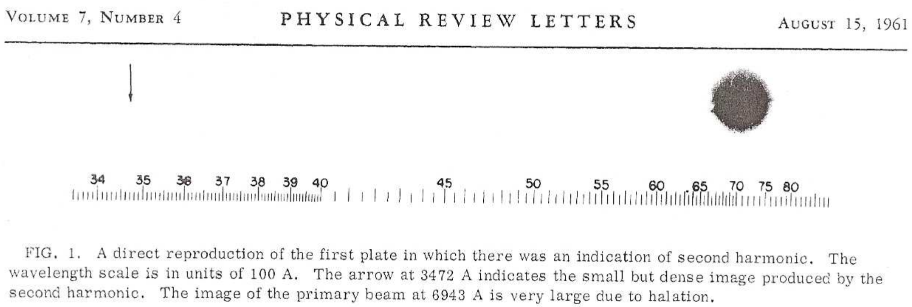
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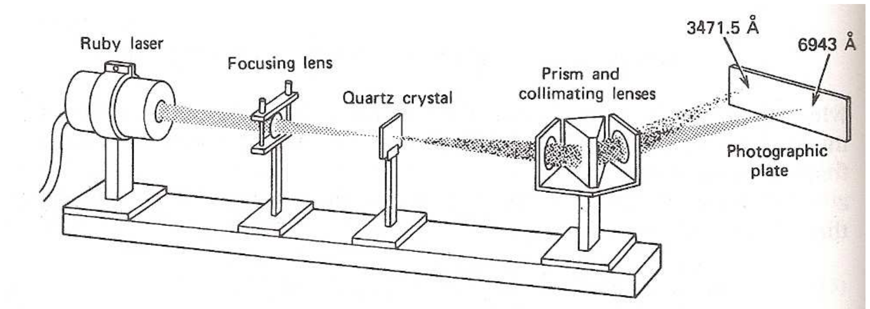
FIG. 1. A direct reproduction of the first plate in which there was an indication of second harmonic. The wavelength scale is in units of 100 A. The arrow at 3472 A indicates the small but dense image produced by the second harmonic. The image of the primary beam at 6943 A is very large due to halation.



Second harmonic generation (SHG) was first demonstrated by P. A. Franken et al. at the University of Michigan in 1961. They focused a ruby laser with a wavelength of 694 nm into a quartz sample. They sent the output light through a spectrometer and recording the spectrum on photographic paper, which indicated the production of light at 347 nm. (Note that the SHG spot does not appear in the publication!)

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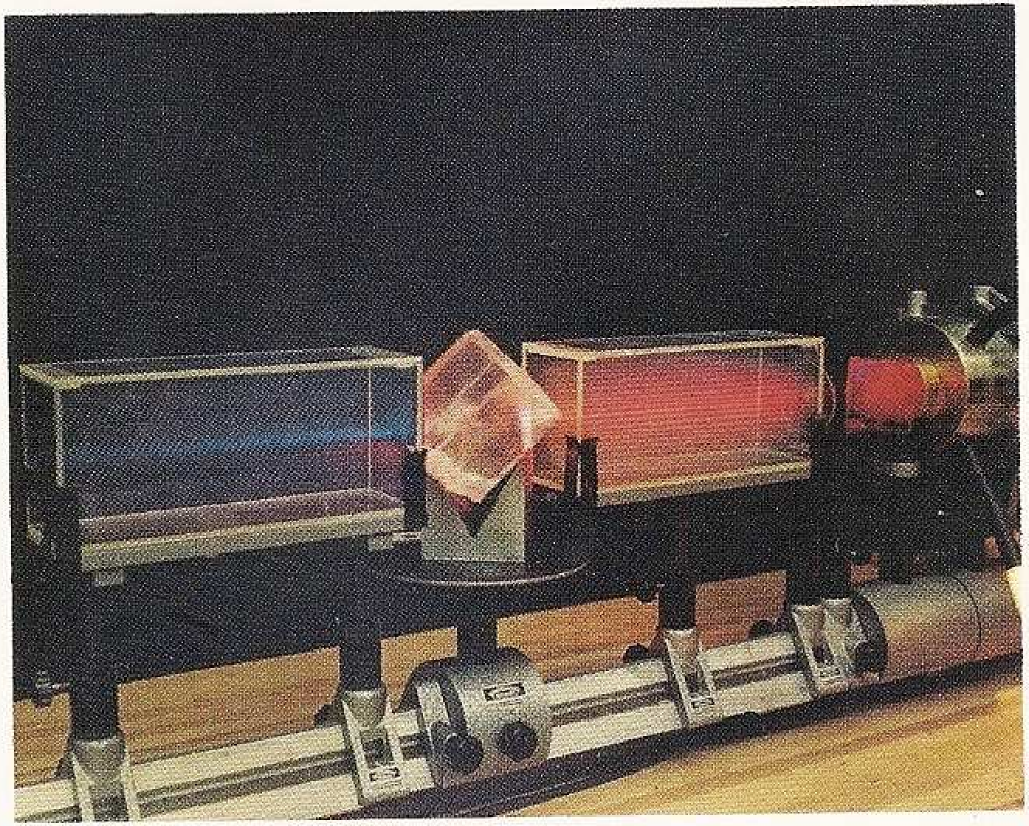
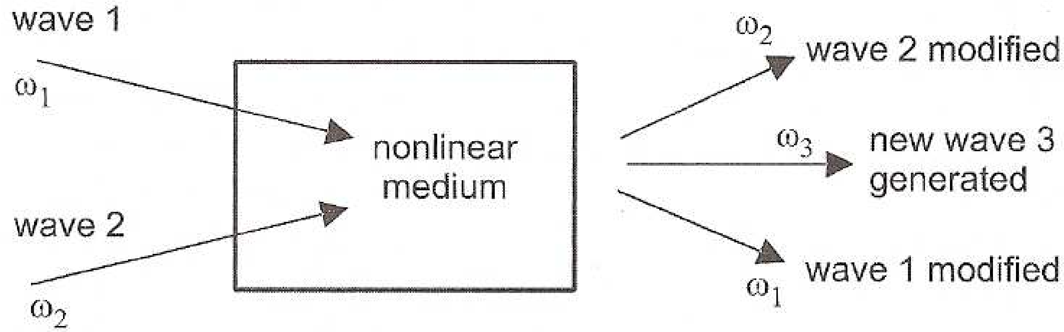
Conversion of red lasers into blue lasers



Laser beam enters a crystal of ammonium dihydrogen phosphate as red light and emerges as blue—the second harmonic. Courtesy of R. W. Terhune.

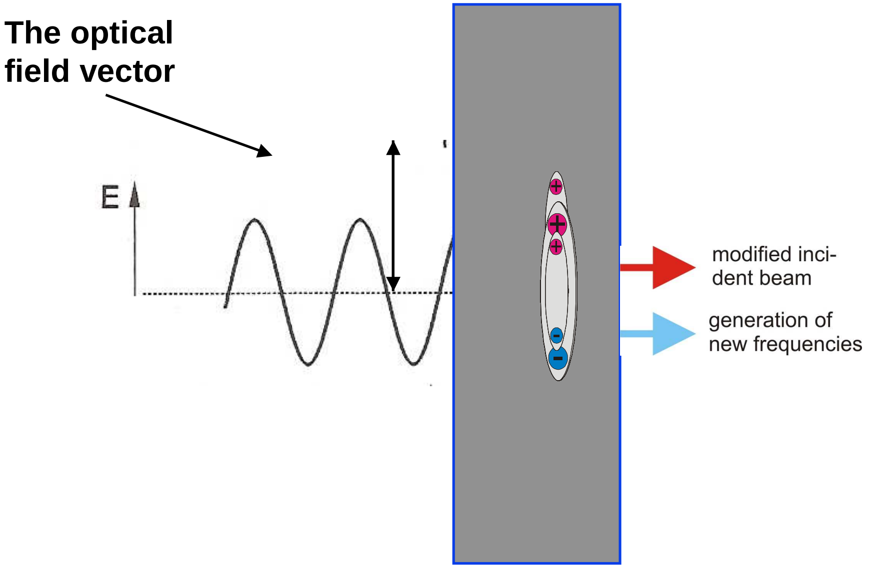
Generation of new waves in a nonlinear medium



Light interacts with the nonlinear medium. The incident beams are modified and new beams are generated.

Optical Polarisation induced in Solids

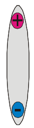
The optical electrical field induces electrical dipole moments in the nonlinear material. The induced dipoles modify the incident beam and for strong optical fields leads to the generation of new k-vectors and new frequencies.

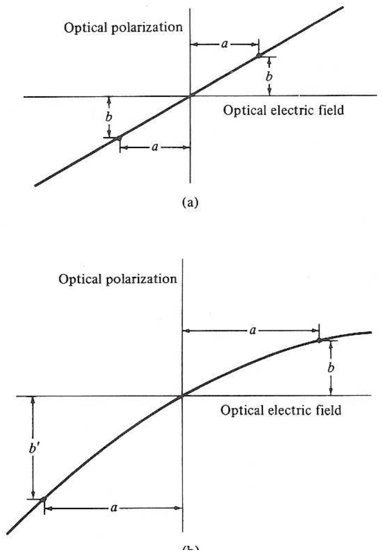


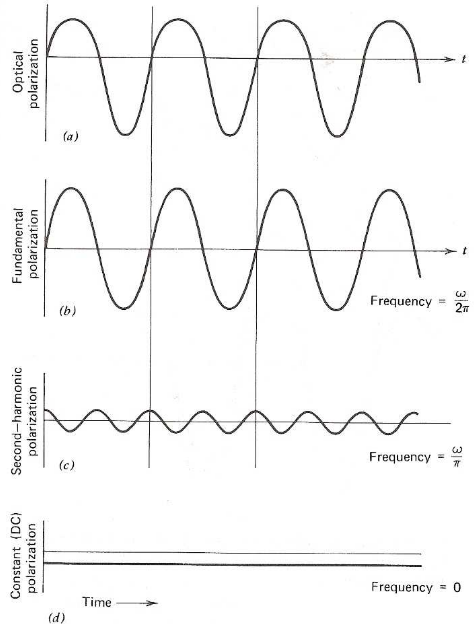
P is the dipole moment per unit volume.

The induced dipoles represents accelerating charged particles that radiates electromagnetic radiation perpendicular to the acceleration vector.

Second Order Nonlinear Polarization







The relation between the induced optical polarisation in the material and the electric field. a) In a linear material b) In a crystal without inversion symmetry.

The optical polarisation induced by an applied sinussoidal electric field with frequency ω. The polarisation consists of three contributions with frequencies ω , 2ω and 0 (dc-term)

Nonlinear Optical Effects in Solids

The nonlinear optical effects are described by the polarization:

$$\underline{D} = \varepsilon_0 \underline{E} + \underline{P} \quad , \qquad P_i(E) = (\varepsilon_0 \chi_{ij}^{(1)} E_j + 2\chi_{ijk}^{(2)} E_j E_k + 4\chi_{ijkl}^{(3)} E_j E_k E_l +)$$

<u>Linear optics is described by $\chi_{ij}^{(1)}$:</u>

The optical properties are independent of the light intensity

The frequency of light does not change due to interaction with light

The principle of superposion is valid

<u>Nonlinear optics is described by $\chi_{ijk}^{(2)}$ and $\chi_{ijkl}^{(3)}$</u>

Light can interact with light

The principle of superposion is not valid

New waves with new k-vectors and new frequencies are generated

Nonlinear Optical Effects in Solids

The nonlinear optical effects are described by the polarization:

$$\underline{D} = \varepsilon_0 \underline{E} + \underline{P} \quad , \qquad P_i(E) = (\varepsilon_0 \chi_{ij}^{(1)} E_j + 2\chi_{ijk}^{(2)} E_j E_k + 4\chi_{ijkl}^{(3)} E_j E_k E_l +)$$

$\chi_{ij}^{(1)}$ describes the linear 1. order optical properties:

absorption (α) and refraction (n)

$\chi_{ijk}^{(2)}$ describes the 2. order nonlinearities:

frequency doubling, electro-optic effect, and

parametric oscillation, etc.

$\chi_{ijkl}^{(3)}$ describes 3. order nonlinearities:

quadratic kerr effect, intensity-dependent refractive

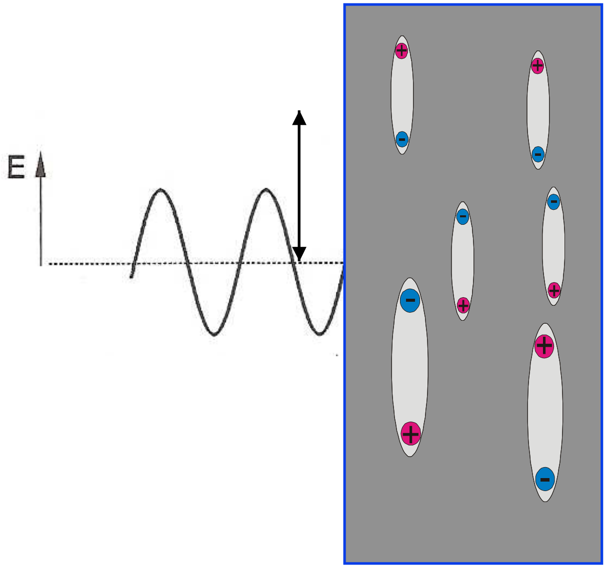
index, four-wave mixing, self-focusing, etc.

Quantum mechanical determination of the susceptibility χ

The material response is given by the polarization:

$$P(t) = \tfrac{1}{2}\varepsilon_0 E\{\chi(\omega)e^{-i\omega t} + \chi(-\omega)e^{i\omega t}\} \qquad (1)$$

The susceptibility $\chi(\omega)$ may be determined by calculating P(t) quantum mechanically and then determine $\chi(\omega)$ by comparison with Eq.(1).



The macroscopic polarization with N atoms in a volume V is:

$$P(t) = Nd(t)/V$$

Where the electric dipole moment is

$$d(t) = -\int \Psi(t) eX \Psi(t) d^3r \quad , \quad X = \sum_{j=1}^{z} x_j$$

Formulation of the interaction in nonlinear materials

The Maxwell equations:

$$\nabla \times \underline{H} = \underline{J} + \frac{\partial \underline{D}}{\partial t} \ , \quad \nabla \times \underline{E} = -\mu_0 \frac{\partial \underline{H}}{\partial t} \ ,$$

$$\underline{D} = \varepsilon_0 \underline{E} + \underline{P} \ , \quad \underline{J} = \sigma \underline{E}$$

lead to:

$$\nabla^2 \underline{E} = \mu_0 \sigma \frac{\partial \underline{E}}{\partial t} + \mu_0 \varepsilon_0 \varepsilon \frac{\partial^2 \underline{E}}{\partial t^2} + \mu_0 \frac{\partial^2 \underline{P}^{NL}}{\partial t^2}$$ "the nonlinear wave equation"

The nonlinear polarization term P^{NL} leads to the generation of new waves with new k-vectors, polarization, and frequencies:

For a new wave: $E_i(z,t) = ½ A_i e^{i(\omega_i t - k_i z)} + c.c.$

the nonlinear wave equation reduces (SVEA-approximation) to:

$$\frac{dA_i}{dz} e^{i(\omega_i t - k_i z)} + c.c. = -\frac{1}{2}\sqrt{\frac{\mu_0}{\varepsilon_0 \varepsilon_i}} \sigma_i A_i e^{i(\omega_i t - k_i z)} + c.c. + i\sqrt{\frac{\mu_0}{\varepsilon_0 \varepsilon_i}} \frac{1}{\omega_i} \frac{\partial^2 P^{NL}}{\partial t^2}$$

The nonlinear wave equation

$$\frac{dA_i}{dz} e^{i(\omega_i t - k_i z)} + c.c. = -\frac{1}{2}\sqrt{\frac{\mu_0}{\varepsilon_0 \varepsilon_i}}\sigma_i A_i e^{i(\omega_i t - k_i z)} + c.c. + i\sqrt{\frac{\mu_0}{\varepsilon_0 \varepsilon_i}} \frac{1}{\omega_i} \frac{\partial^2 P^{NL}}{\partial t^2}$$

This is the fundamental equation that governs the generation of a new wave $E_i(z,t) = ½ A_i e^{i(\omega_i t - k_i z)} + c.c.$

Second Order Nonlinear Optics

The interaction between light and matter is given by:

$$P_i(E) = (\varepsilon_0 \chi_{ij}^{(1)} E_j + 2\chi_{ijk}^{(2)} E_j E_k + 4\chi_{ijkl}^{(3)} E_j E_k E_l +)$$

where

$$P_{i,NL2}(E) = 2\chi_{ijk}^{(2)} E_j E_k$$

describes the second order nonlinearities. The notation

$$\chi_{ijk}^{(2)} = d_{ijk}$$

is often used in the literature where d_{ijk} is a third order tensor. with 27 elements.

Contracted Notation for the Second Order Susceptibility

$$P_{i,NL2}(E) = 2d_{ijk}E_jE_k \quad \text{where} \quad \chi^{(2)}_{ijk} = d_{ijk}$$

since

$$d_{ijk} = d_{ikj}$$

The third order tensor may be represented by a (3x6) matrix:

$$d_{il} = \begin{pmatrix} d_{11} & \mathrm{K} & d_{16} \\ \mathrm{M} & \mathrm{O} & \mathrm{M} \\ d_{31} & \mathrm{L} & a_{36} \end{pmatrix}$$

with 18 elements

Generation of new waves using second order nonlinearities

Sum frequency generation

$\omega_1\ ,\ \omega_2 \rightarrow$ Nonlinear crystal $\rightarrow \omega_1 + \omega_2$

Second harmonic generation

$\omega\ ,\ \omega \rightarrow$ Nonlinear crystal $\rightarrow 2\omega$

Difference frequency generation

$\omega_1\ ,\ \omega_2 \rightarrow$ Nonlinear crystal $\rightarrow \omega_1 - \omega_2$

Optical parametric oscillator (OPO)

$\omega_{pump} \rightarrow$ M | Nonlinear crystal | M $\rightarrow \omega_{signal}\ ,\ \omega_{idler}$

Three Wave Mixing

Two waves E(ω_1) and E(ω_2) may due to the interaction with the second order nonlinear polarization generate a third beam E(ω_3)

ω_1 , ω_2 → Nonlinear crystal → ω_1, ω_2, ω_3

The three waves are given by:

$$E_1(\omega_1) = \tfrac{1}{2} A_1 e^{i(\omega_1 t + k_1 z)} + c.c.\,, \quad E_2(\omega_2) = \tfrac{1}{2} A_2 e^{i(\omega_2 t + k_2 z)} + c.c.\,, \quad E_3(\omega_3) = \tfrac{1}{2} A_3 e^{i(\omega_3 t + k_3 z)} + c.c.$$

where we have assumed all waves to be polarized in the same direction.
For the three waves the nonlinear wave Equation reduces to:

$$\frac{dA_1}{dz} = -\frac{\mu_0 \sigma_1 c}{2n_1} A_1 - \frac{i\omega_1 \mu_0 \alpha d c}{n_1} A_3 A_2^{\,*} e^{-i\Delta kz}$$

$$\frac{dA_2}{dz} = -\frac{\mu_0 \sigma_2 c}{2n_2} A_2 - \frac{i\omega_2 \mu_0 \alpha d c}{n_2} A_3 A_1^{\,*} e^{-i\Delta kz}\,,$$

$$\frac{dA_3}{dz} = -\frac{\mu_0 \sigma_3 c}{2n_3} A_3 - \frac{i\omega_3 \mu_0 \alpha d c}{n_3} A_1 A_2 e^{i\Delta kz}$$

The fundamental equations that describes second order nonlinear interaction.

where $\Delta k = k_3 - k_1 - k_2$, $\alpha = (2 - \delta_{\omega 1\, \omega 2})/2$, and where we have used $\sqrt{\frac{\mu_0}{\varepsilon_0 \varepsilon_i}} = \frac{\mu_0 c}{n_3}$
($\delta_{\omega 1\, \omega 2} = 1$ for $\omega_1 = \omega_2$ and $\delta_{\omega 1\, \omega 2} = 0$ for ω_1 # ω_2)

Frequency doubling

$\omega_1 = \omega$, $\omega_2 = \omega$ → Nonlinear crystal → ω, $\omega_3 = 2\omega$

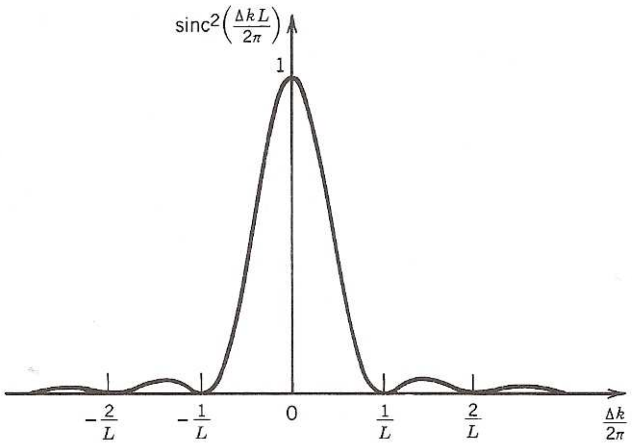
The fundamental equation $\frac{dA_3}{dz} = -\frac{\mu_0 \sigma_3 c}{2n_3} A_3 - \frac{i\omega_3 \mu_0 \alpha d c}{n_3} A_1 A_2 e^{i\Delta k z}$ reduces to:

$$\frac{dA(2\omega)}{dz} = -\frac{i\omega\mu_0 dc}{n(2\omega)} A^2(\omega) e^{i\Delta k z}$$

where absorption is neglected. If we assume that $A_1 = A_2 = A(\omega)$ is undepleted we obtain the second harmonic intensity:

$$I(2\omega) = \frac{2\mu_0^3 c^3 \omega^2 d^2}{n^2(\omega) n(2\omega)} L^2 I(\omega)^2 \left(\frac{\sin(\Delta k L/2)}{\Delta k L/2} \right)^2$$

where L is the length of the nonlinear crystal.

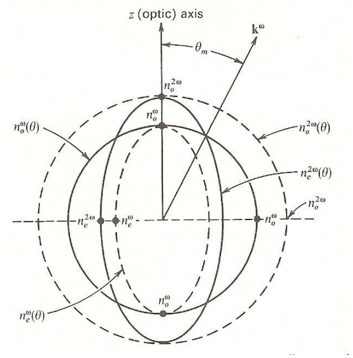


Phase matching in materials with birefringence

Strong SHG signals are obtained when :

$$\Delta k = k_3 - k_1 - k_2 = k_3 - 2k_1 = \frac{2\omega}{c}\left(n(2\omega) - n(\omega)\right) = 0$$

Phase matching condition



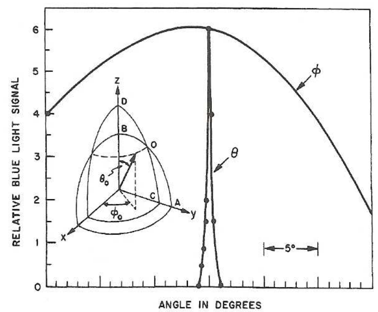


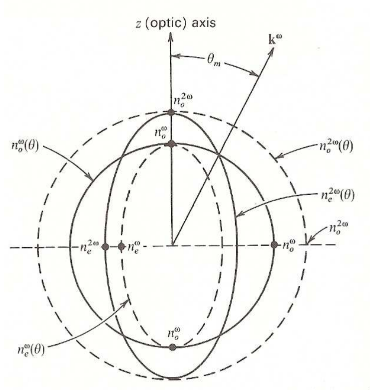
FIG. 2. Blue light intensity as a function of crystal orientation for KDP. Maximum output occurs at $\theta_0 = 52° \pm 2°$, $\phi_0 = 45°$. Laser beam collimated to within $\pm\frac{1}{4}°$. *AOB* is an arc on the index of refraction surface for red ordinary rays, *COD* for blue extraordinary rays.

It was suggested independently by Giordmaine(ref.1) and Maker et al(ref.2)) that index matching may be obtained in birefractive crystals by correct choice of polarisation and angle θ of propagation. The intersection defines the propagation direction.

In Ref. 2 phase matching in KDP with an 300-fold increase of blue light intensity was observed in 1962

Ref.1: J.A.Giordmaine, Phys. Rev. Letters, 8, 19(1962); Ref2: P. Maker et. al, Phys. Rev. Letters, 8, 21(1962),

Type I and II phase matching



Type I phase matching:

The refractive index of an extraordinary beam is given by: $n_e(\theta) = \dfrac{n_o n_e}{\sqrt{n_0{}^2 \sin^2\theta + n_e{}^2 \cos^2\theta}}$

Phase matching is obtained in negative uniaxial crystals (n_e<n_0) when $n_e(2\omega, \theta_m) = n_0$

Type II phase matching:

In this case the fundamental beam ω consist of both an ordinary beam and an extraordinary beam and in this configuration the beam at 2ω fulfils the condition:

$$n_e(\theta_m, 2\omega) = ½(n_e(\theta_m, \omega) + n_0(\omega))$$

and the frequency doubled beam will be an **extraordinary** beam

Suggested further reading:

- *Nonlinear Optics* by R. W. Boyd
  Academic Press
- *Optical Electronics in Modern Communications*
  by A. Yariv, Oxford University Press
- *The Principles of Nonlinear Optics* by Y. R. Shen,
  Wiley