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SMR 1302 - 20

WINTER SCHOOL ON LASER SPECTROSCOPY AND APPLICATIONS

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Introduction to Non-Linear Optics for lecture on Novel Laser Sources for Applied Spectroscopy

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These are preliminary lecture notes, intended only for distribution to participants.

Introduction to Non-linear Optics

- The binding energy of an external e^- in atomic or molecular systems is:

  $E \sim 10\, eV$ ($1 eV \rightarrow \sim 1\, \mu m$ wavelength)

- Average distance from nucleus: $1 Å = 10^{-8} cm$

$$\Downarrow$$

The electric field is: $E_a \sim 10^9 V/cm$

- In most dielectrics (insulators), "breakdown" occurs for external electric fields $E \sim 10^6 V/cm$
- Therefore, in many cases, the maximum value of the ratio $\boxed{E/E_a \sim 10^{-3}}$

If $\Delta K \neq 0$ the typical length for the non-linear process that maintains Coherent emission is $l_c \approx 1/|\Delta K|$

Within l_c there is constructive interference that increases the, e.g. SHG, field, outside l_c energy is reconverted to fundamental radiation frequency and, periodically, to SHG again.

How to achieve Phase Matching?

Since:

$K(\omega) = \frac{\omega}{c} n(\omega)$ n = real part of refractive index

$\Delta K = 0$ for SHG if $\cancel{2} n_{2\omega} \cancel{\omega_1} = \cancel{2} n_{\omega} \cancel{\omega_1}$

$$\downarrow$$

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

But $n(\omega)$ monotonically changes with frequency, therefore: → Anisotropic crystals

→ Birefringent Phase Matching (first trick)

→ "Quasi" Phase Matching (second trick)

As a consequence ($\frac{E}{E_d} \ll 1$) it is possible to expand the electric dipole moment per unit volume, called $\vec{P}$, in a power series of the externally applied electric field $\vec{E}$:

$$\bar{P}(\bar{E}) = \epsilon_0 \chi(\bar{E}) \bar{E} = \epsilon_0 [\chi^{(1)} \bar{E} + \chi^{(2)} \bar{E}^2 + \chi^{(3)} \bar{E}^3 + \dots]$$

$\bar{P}$ is a function of $\bar{E}$ that describes how the medium behaves when the e.m. wave, with field $\bar{E}$, passes through it.

All phenomena, both in linear and non-linear optics, can be described by using Maxwell equations:

$$\bar{\nabla} \times \bar{E} = -\frac{1}{c} \frac{\partial}{\partial t} \bar{B} \qquad \bar{\nabla} \cdot (\bar{E} + 4\pi \bar{P}) = 0$$

$$\bar{\nabla} \times \bar{B} = \frac{1}{c} \frac{\partial}{\partial t} (\bar{E} + 4\pi \bar{P}) \qquad \bar{\nabla} \cdot \bar{B} = 0$$

Writing Maxwell eqs., we assumed no charges nor external currents are present:

$$\boxed{\rho = \bar{J} = 0}$$

We have assumed $\bar{B} = \bar{H}$, that means the magnetic dipole moment per unit volume $\bar{M}$ is zero $\rightarrow$ $\boxed{\bar{M} = 0}$.

$\bar{J} = 0$ only if the special case of e.m wave propagation through conductors is excluded.

If propagation through <u>transparent</u> <u>media</u> (where "<u>long</u>" optical paths are possible) is considered, it is possible to put $\underline{\bar{J} = 0}$, because we are using <u>insulators</u>.

By combining Maxwell eqs., it is possible to write:

$$\bar{\nabla}^2 \bar{E} - \bar{\nabla}(\bar{\nabla} \cdot \bar{E}) - \frac{1}{c^2} \frac{\partial^2 \bar{E}}{\partial t^2} - \frac{4\pi}{c^2} \frac{\partial^2 \bar{P}}{\partial t^2} = 0$$

To solve this equation, a relation between $\bar{P}$ and $\bar{E}$ is needed → A microscopic knowledge of propagation medium is required, i.e.

$\chi^{(i)}$ → (i-th order susceptibility)

is required to describe i-th order effects.

$\chi^{(i)}$ is a tensor and can be very difficult to have a detailed knowledge of it.

We restrict to second order phenomena, i.e. those that are described by $\chi^{(2)}$. We neglect, in the following, linear effects ($\chi^{(1)}$ dependant), as well as non-linear terms of order >2.

$\chi^{(1)}$ → is a tensor of rank 2

$\chi^{(2)}$ → is a tensor of rank 3

$$\downarrow$$

$$\chi^{(2)}_{ijk}$$

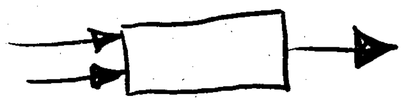
$\chi^{(2)} \neq 0$ only in media that do not possess a center of symmetry

whereas

$\chi^{(3)} \neq 0$ in isotropic media

$\chi^{(2)}$ processes

$\omega_3 = \omega_1 + \omega_2$
parametric luminescence
(or oscillation)



DFG $\omega_1 - \omega_2 = \omega_3$
SFG $\omega_1 + \omega_2 = \omega_3$
SHG $2\omega_1 = \omega_3$

To observe non-linear effects strong fields are needed ($\gtrsim 1\,kV/cm$)

$\Downarrow$

- First Second Harmonic Generation by Franken et al. (1961) $\rightarrow$ ruby laser ($\lambda = 6942$ Å) was doubled in a quartz crystal (at $\lambda = 3471$ Å)

$\downarrow$

Laser sources are needed!

In fact:

I (incoherent sources) $\propto$ photon number (N)

$$\left(\left|\bar{E}_1+\bar{E}_2+\ldots\right|^2=\left|\bar{E}_1\right|^2+\left|\bar{E}_2\right|^2+\ldots\right.$$

whereas

I (coherent fields) $\propto$ $\underline{\underline{N^2}}$

In the non-linear process, first the e.m. wave generates a non-linear response of the medium and this acts modifying the fields (in a non-linear way $\rightarrow$ generation of waves at new frequencies).

In principle, all media can give non-linear effects $\longrightarrow$ Also VACUUM (Photons can interact through vacuum polarization)

Of course, for practical purposes, vacuum is, to a good approximation, a linear medium

To fix the ideas, let us consider Second Harmonic Generation (SHG)

Energy Conservation states that:

$$\hbar\omega_1 + \hbar\omega_1 = \hbar\omega_2 \qquad \omega_2 = 2\omega_1$$

How can conversion from $\omega_1 \rightarrow \omega_2$ radiation be maximized?

$\Downarrow$

"Phase Matching Condition"

Can be seen from a Quantum Mechanical point of view or classically:

In Quantum Mechanics $\rightarrow$ we consider Photons $\left(\begin{array}{l} E = h\nu \\ \bar{p} = \hbar\bar{k} \end{array}\right.$

When $\chi^{(2)}_{\alpha\beta\gamma} \neq 0$ photons may interact with each other

$\hookrightarrow$ Two photons may "fuse" to form a third

Momentum is conserved if $\hbar\bar{k}_2 = 2\hbar\bar{k}_1$

Classical explanation of Phase Matching:

$\Delta \bar{k} = 0$ means that all "second harmonic" dipoles created by the e.m. wave emit radiation <u>exactly</u> <u>in</u> <u>phase</u> with all the other set of dipoles in the medium

$\Downarrow$

<u>Constructive Interference</u>

If $\Delta \bar{k} \neq 0 \longrightarrow$ <u>Destructive Interference</u>

How can the Interference term be written?

From Maxwell eqs. it is obtained:

$$E_3(\ell) = -\frac{i\omega_3 d_3}{c n_3} E_1 E_2 \frac{e^{i\Delta k \ell} - 1}{i \Delta k}$$

$$I_3 = P_3 / A = \frac{1}{2} \varepsilon_0 c n_3 |E_3|^2$$

$$\eta(A, \ell) \equiv \frac{P_3}{P_1 P_2} = \frac{2}{\varepsilon_0 c^3} \frac{\omega_3^2 d_3^2 \ell^2}{A n_1 n_2 n_3} \operatorname{sinc}^2\left(\frac{\Delta k \ell}{2}\right)$$

(For plane waves) $\longrightarrow \operatorname{sinc}(x) = \frac{\sin(x)}{x}$

crystal with phase matching is the necessary and sufficient condition for an effective three-wave interaction.

2.3 Optics of Uniaxial Crystals

In *uniaxial* crystals a special direction exists called the *optic axis* (Z axis). The plane containing the Z axis and the wave vector $\boldsymbol{k}$ of the light wave is termed the *principal plane*. The light beam whose polarization (i.e., the direction of the vector $\boldsymbol{E}$ oscillations) is normal to the principal plane is called an *ordinary beam* or an *o-beam* (Fig. 2.2). The beam polarized in the principal plane is known as the *extraordinary beam* or *e-beam* (Fig. 2.3). The refractive index of the o-beam does not depend on the propagation direction, whereas for the e-beam it does. Thus, the refractive index in anisotropic crystals generally depends both on light polarization and propagation direction.

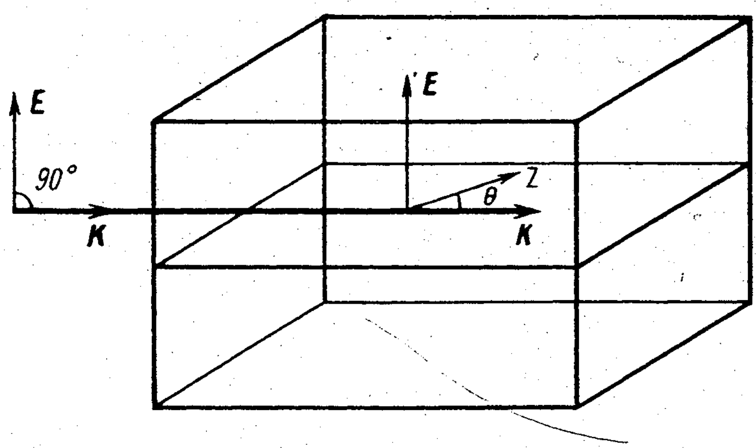


Fig. 2.2. Principal plane of the crystal (kZ) and ordinary beam

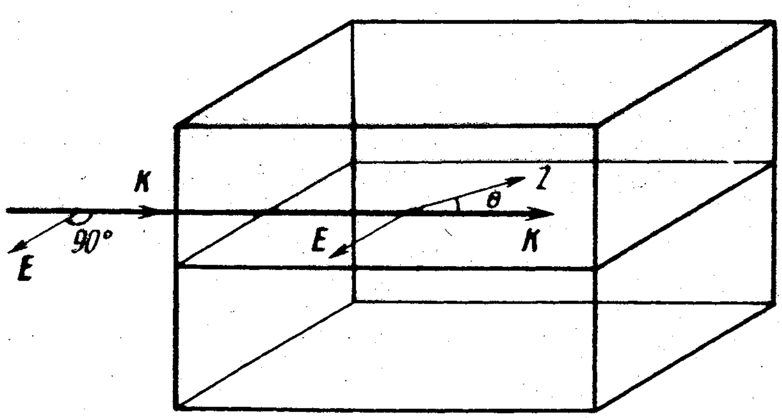


Fig. 2.3. Principal plane of the crystal (kZ) and extraordinary beam

from "Handbook of Nonlinear Crystals" by Dmitriev, Gurzadyan & Nikogosyan (Springer-Verlag)

Types of Phase Matching

Type I: $\bar{k}_{o1} + \bar{k}_{o2} = \bar{k}_3^e$ $\boxed{(o,o,e)}$

in negative crystals ($n_o > n_e$)

$\bar{k}_1^e(\theta) + \bar{k}_2^e(\theta) = \bar{k}_{o3}$ $\boxed{(e,e,o)}$

in positive crystals ($n_o < n_e$)

Type II: $\bar{k}_{o1} + \bar{k}_2^e(\theta) = \bar{k}_3^e(\theta)$ $\boxed{(o,e,e)}$

in negative crystals

$\bar{k}_1^e(\theta) + \bar{k}_{o2} = \bar{k}_3^e(\theta)$ $\boxed{(e,o,e)}$

in positive crystals

At $\theta = 90° \rightarrow$ non-critical phase matching

Therefore, the three waves never have the same polarization for Birefringent Phase Matching

Note that conversion efficiency depends on d_{ie}^2

For $LiNbO_3$ (one of the most efficient crystals)
is: $d_{33} \simeq 30\ pm/V$ at $\lambda = 1.06\ \mu m$
$d_{31} \simeq 4\ pm/V$
$d_{22} \simeq 2\ pm/V$

but $d_{ooe} = d_{31} \sin\vartheta - d_{22} \cos\vartheta \sin 3\vartheta$
if $\vartheta = 90°$ for BPM $\longrightarrow d_{eff} = d_{ooe} = d_{31}$

For QPM, instead, is $\boxed{d_{eff} = d_{33}}$

Hence, $$\frac{\eta(QPM)}{\eta(BPM)} \rightarrow \frac{\left[(d_{eff})^2\right]_{QPM}}{\left[(d_{eff})^2\right]_{BPM}} \xrightarrow[(m=1)]{} \frac{\left(\frac{2}{\pi}\right)^2 d_{33}^2}{d_{31}^2} \simeq$$

$\simeq \underline{\underline{20}}$ times more efficient QPM than BPM

Notation:

Instead of $\chi^{(2)}$, d is generally tabulated, where:

$$\chi^{(2)}_{ijk} \equiv 2\, d_{ijk}$$

But it is easier to use a bi-dimensional notation for d: $(i,j,k) \rightarrow (i,l)$ $\quad d_{ijk} \rightarrow d_{il}$

Therefore
$i=1 \rightarrow X$
$i=2 \rightarrow Y$
$i=3 \rightarrow Z$

$l =$	1	2	3	4	5	6
	xx	yy	zz	$yz=zy$	$xz=zx$	$xy=yx$

Hence d_{il} has 18 components

GENERALITÀ DI OTTICA NON LINEARE AL 2° ORDINE

Equazione costituente per la polarizzazione:

$$\mathbf{P}(\mathbf{r},t) = \mathbf{P}^{(L)}(\mathbf{r},t) + \mathbf{P}^{(NL)}(\mathbf{r},t)$$

$$= \chi^{(1)} \cdot \mathbf{E}(\mathbf{r},t) + \chi^{(2)} \cdot \mathbf{E}(\mathbf{r},t) \cdot \mathbf{E}(\mathbf{r},t)$$

Equazione del moto per il campo elettrico:

$$\left(\nabla \times \nabla \times + \frac{1}{c^2} \frac{\partial^2}{\partial t^2} \right) \mathbf{E}(\mathbf{r},t) =$$

$$= -\frac{4\pi}{c^2} \frac{\partial^2}{\partial t^2} \left[\chi^{(1)} \cdot \mathbf{E}(\mathbf{r},t) + \chi^{(2)} \cdot \mathbf{E}(\mathbf{r},t) \cdot \mathbf{E}(\mathbf{r},t) \right]$$

Notazione per la suscettività al 2° ordine:

$$\chi_{ijk} = 2d_{ijk} = 2d_{il}$$

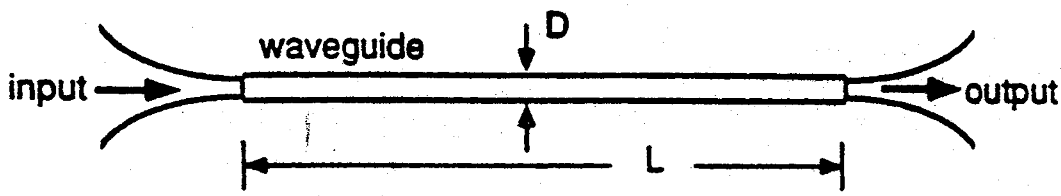
$$\begin{pmatrix} P_X \\ P_Y \\ P_Z \end{pmatrix} = \begin{pmatrix} d_{11} & d_{12} & d_{13} & d_{14} & d_{15} & d_{16} \\ d_{21} & d_{22} & d_{23} & d_{24} & d_{25} & d_{26} \\ d_{31} & d_{32} & d_{33} & d_{34} & d_{35} & d_{36} \end{pmatrix} \begin{pmatrix} E_X^2 \\ E_Y^2 \\ E_Z^2 \\ 2E_Y E_Z \\ 2E_X E_Z \\ 2E_X E_Y \end{pmatrix}$$

Quasi phasematching waveguide doubling

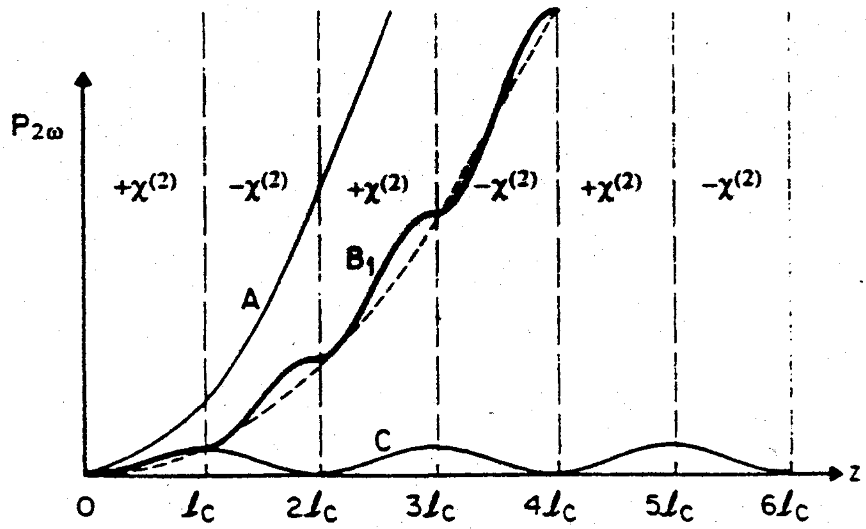
Waveguide confinement improves the conversion efficiency by a factor of

$$\lambda_\omega L / A_{eff}$$

with repect to bulk nonlinear conversion.
(λ_ω - pump wavelength, L - interaction length, A_{eff} - effective waveguide cross-sectional area).



Quasi phasematching - phase velocity matching by periodic modulation of the nonlinear coefficient:
Higher efficiency (~20 times higher) in $LiNbO_3$, by using the large d_{33} coefficient which is not accesible to birefringent phasematching.
Near room temperature operation.
Relaxed temperature and wavelength tolerances (both fundamental and SH are extraordinarily polarized).

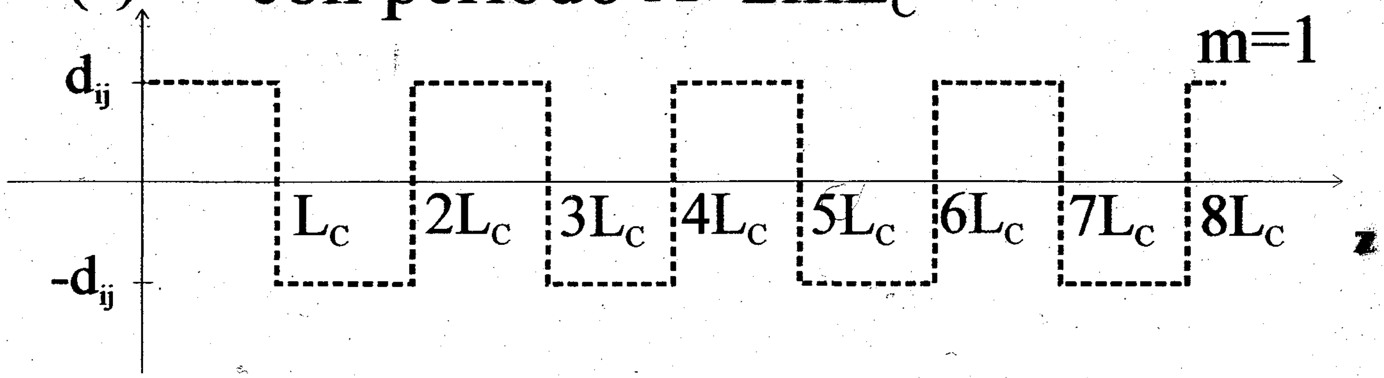


Quasi-Phase Matching (QPM)

Inversione della fase relativa ripetuta ad ogni multiplo della lunghezza di coerenza

modulazione periodica di d_{ij}

con periodo $\Lambda = 2mL_C$



Condizione di QPM

$$\boxed{\Delta k' = \Delta k - \frac{2\pi m}{\Lambda} = 0}$$

$$2\Lambda \cdot \left[n^{2\omega}\left(T_{QPM}\right) - n^{\omega}\left(T_{QPM}\right)\right] = m\lambda$$

Efficienza:

$$\eta \equiv \frac{P^{2\omega}}{P^{\omega}} = 2\left(\frac{\mu_0}{\varepsilon_0}\right)^{\frac{3}{2}} \frac{\omega^2 d_Q^{\,2} L^2}{n_2 n_1^{\,2}} \left(\frac{P^{\omega}}{A}\right) sinc^2\left(\frac{\Delta k' L}{2}\right)$$

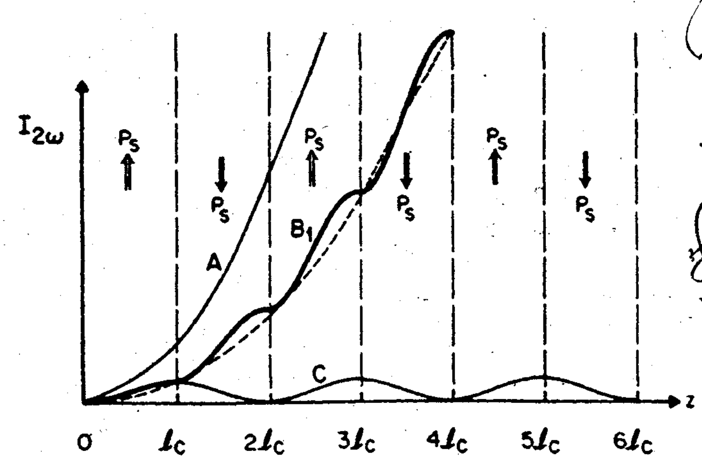
$$d_{ij} \longrightarrow d_Q = \frac{2}{m\pi} d_{ij} \qquad \Delta k \longrightarrow \Delta k'$$

QUASI-PHASE MATCHING

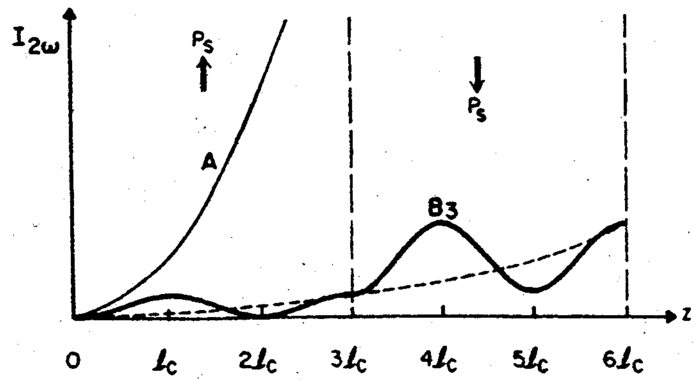
$$\begin{cases} \Delta k = k_3 - k_1 - k_2 - \dfrac{2\pi m}{\Lambda} \\ \Delta k L \approx 0 \end{cases}$$

$$\frac{d_{eff}^{QPM}}{d_{eff}^{B}} = \frac{2}{m\pi}\sin(m\pi D)$$

D = duty cycle (best efficiency is obtained with $D = \frac{1}{2}$ (50% duty cycle) for $m = 1$
$D = \frac{1}{2}$ if all domains have the same width)

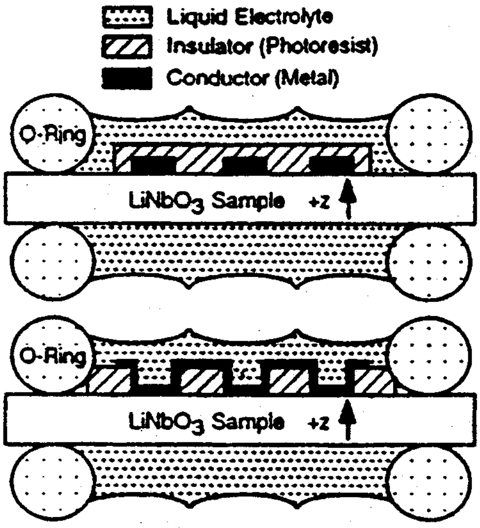


(a)

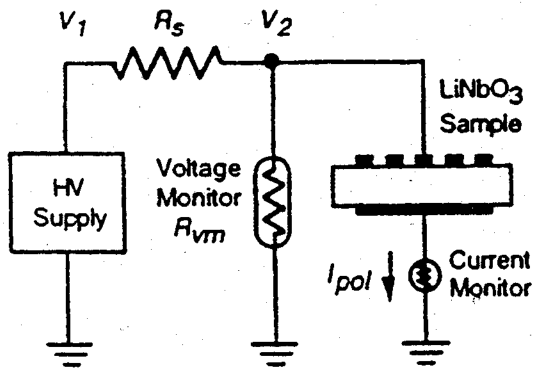


(b)

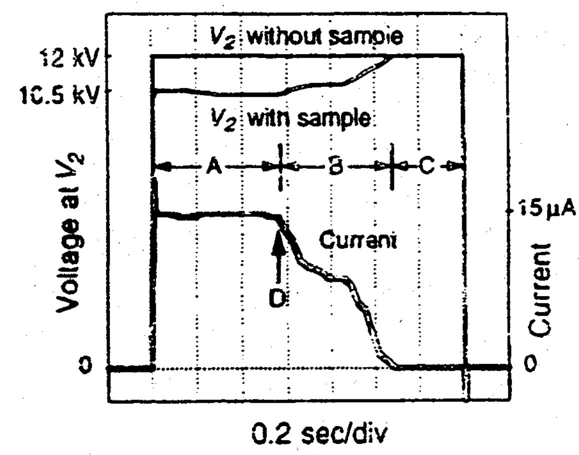
Fig. 1. Effect of phase matching on the growth of second harmonic intensity with distance in a nonlinear crystal. (a) A: perfect phase matching in a uniformly poled crystal; C: nonphase-matched interaction; B_1: first-order QPM by flipping the sign of the spontaneous polarization every coherence length of the interaction of curve C. (b) A: perfect phase matching; B_3: third-order QPM by flipping P_S every three coherence lengths.



(a)



(b)



(c)

Fig. 9. (a) Schematic of the electrode configuration for electric field poling of a ferroelectric crystal. The ferroelectric domains can be reversed by the application of a sufficient electric field. (b) Electric field poling circuit. Typically $R_x = 100M\Omega$, $R_{vm} = 1G\Omega$, and V_2 is set at 12 kV for a 0.5 mm thick sample of $LiNbO_3$. During poling V_2 clamps at the coercive voltage V_c. (c) Voltage and current waveforms for poling a 3 mm diameter 0.5 mm thick $LiNbO_3$ sample. Section A is poling under the metal electrode or liquid contact, Sec. B is poling under the photoresist, and Sec. C is after completion of poling. For a patterned device the voltage would be reduced to zero at point D. [after Ref. 58]

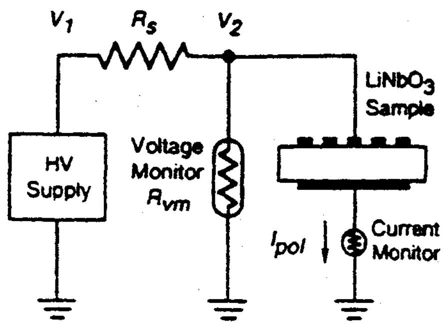


Fig. 5. Electric-field poling circuit. Typically $R_s = 100\ M\Omega$, $R_{vm} = 1\ G\Omega$, and V_2 is set at 12 kV with no sample in the circuit. During poling, V_2 clamps at the coercive voltage $V_c \approx 10.5$ kV for a 0.5-mm-thick $LiNbO_3$ sample.

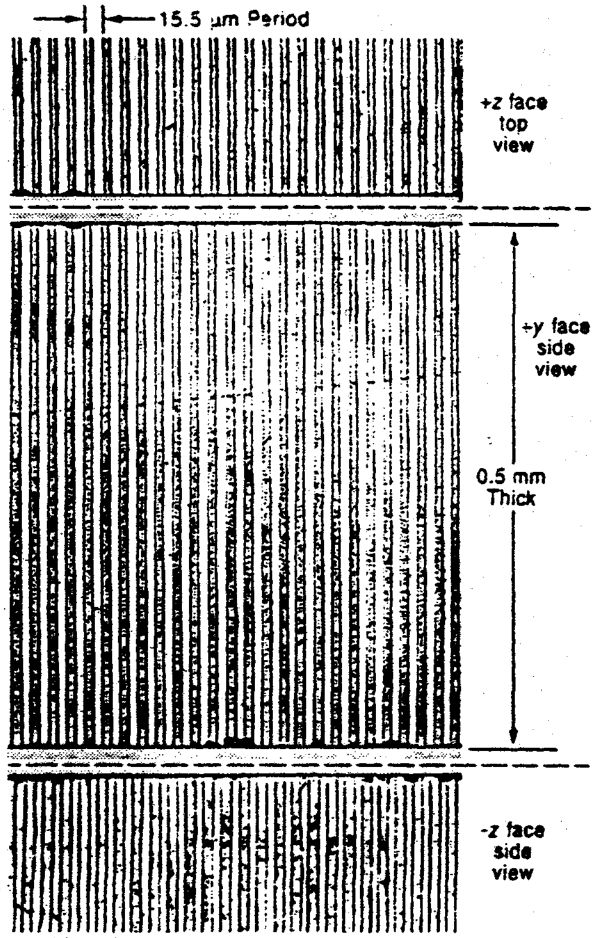


Fig. 7. Orthographic view of 0.5-mm-thick PPLN with a 15.5-μm period, after etching in HF acid to reveal the domain structure. The three panels are top, side, and bottom views taken at the same location in the crystal by cutting and polishing into the grating region. The top panel is the $+z$ face upon which the lithographic electrode was applied. The middle panel is a cross-sectional view of the $+y$ face. The bottom panel is the $-z$ face, which had the unpatterned ground electrode.

CONDITIONS FOR QPM (SHG)

$$\Delta k = k(2\omega) - 2k(\omega) = \frac{4\pi\,(n_{2\omega} - n_\omega)}{L}$$

coherence length: $L_c = \frac{\pi}{\Delta k}$

grating period: $\Lambda = 2 m L_c \quad m = 1, 3, 5 \ldots$

$$L_c = \frac{\lambda}{4(n_{2\omega} - n_\omega)}$$

QPM condition: $\bar{k}_{2\omega} - 2\bar{k}_\omega - \bar{K} = 0$

$$|\bar{K}| = \frac{2 m \pi}{\Lambda}$$

effective nonlinear coefficient for QPM:

$$d_{eff} = \left(\frac{2}{m\pi}\right) d_{ij} \operatorname{sen}(m\pi D)$$

D = duty cycle

Fejer et al., IEEE J. Q. E, QE-28, 2631 (1992)

INDICI DI RIFRAZIONE

$$\begin{cases} n_o(\theta) \equiv n_o \\ n_e(\theta) = \left(\dfrac{\cos^2\theta}{n_o^2} + \dfrac{\sin^2\theta}{n_e^2} \right)^{-1} \end{cases}$$

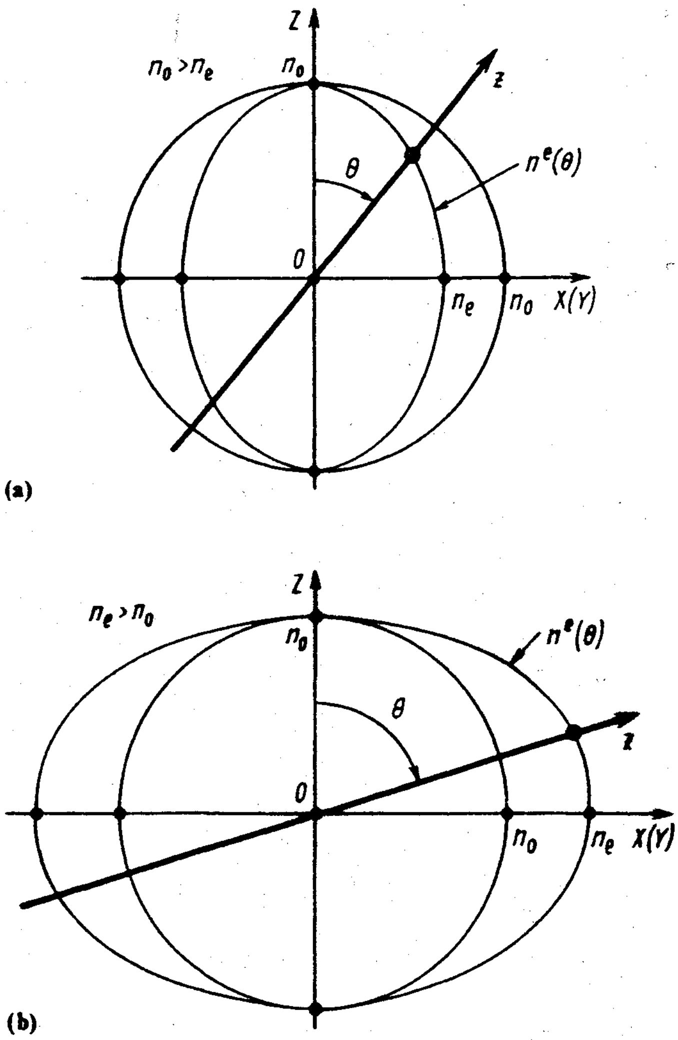


Fig. 2.5. Indicatrices of the refractive indices for ordinary and extraordinary waves in negative **(a)** and positive **(b)** uniaxial crystals

PHASE MATCHING BIRIFRANGENTE

$$n_{o1}(\omega_1) = n_3^e(2\omega_1, \theta_{pm}^{(1)}) \tag{2.30}$$

or

$$2k_{o1}(\omega_1) = k_3^e(2\omega_1, \theta_{pm}^{(1)}) \,. \tag{2.31}$$

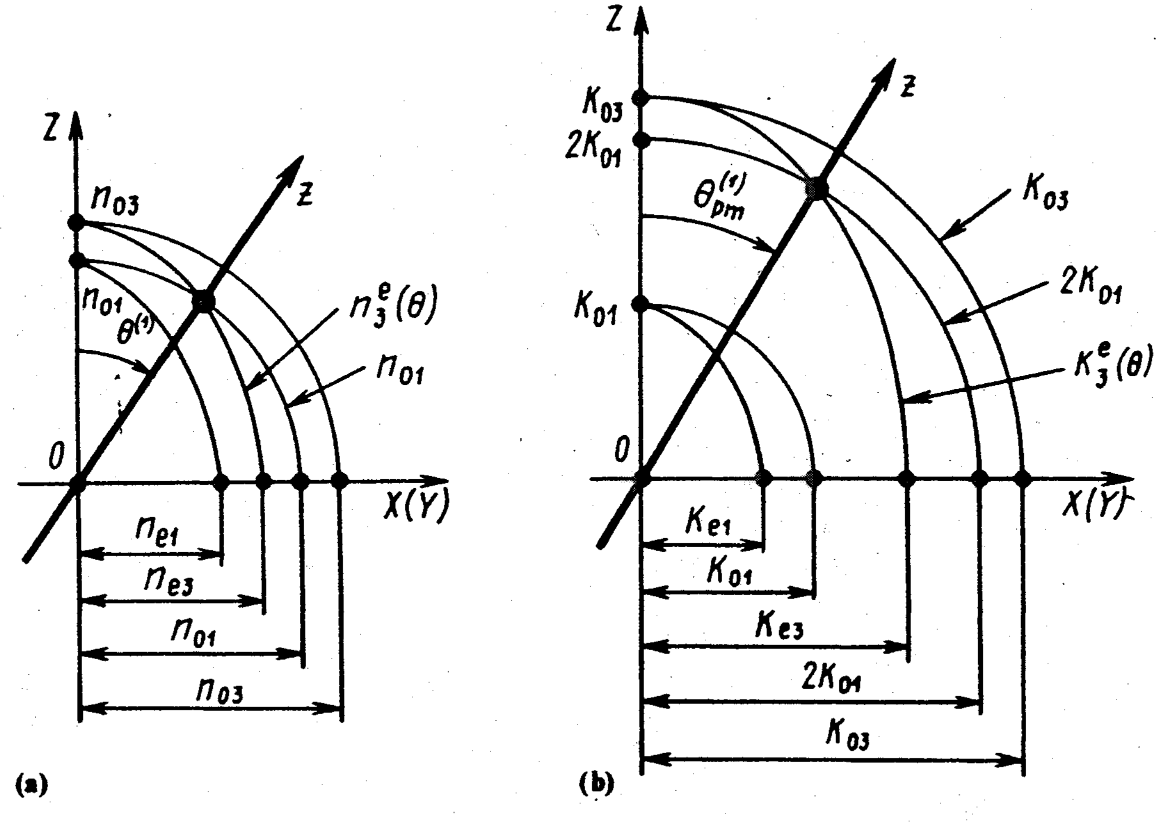
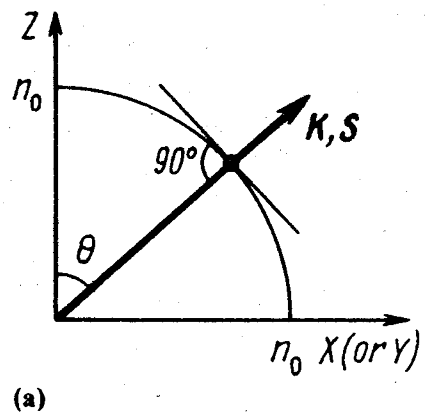


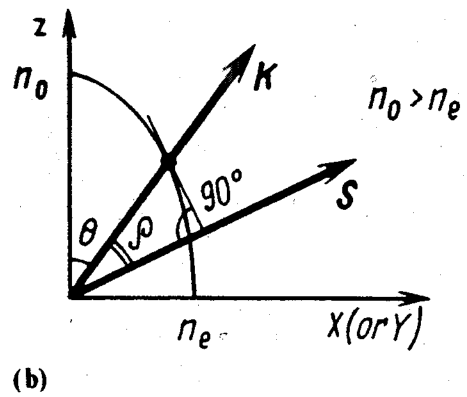
Fig. 2.8. Scalar (collinear) phase matching of type I ("ooe") in a uniaxial negative crystal in coordinates of refractive indices (a) and wave vectors (b) in the first quadrant of the XZ (YZ) plane

BIRIFRANGENZA

Angolo di birifrangenza:

$$\rho(\theta) = \pm \operatorname{arctg}\left[\left(\frac{n_o}{n_e}\right)^2 \operatorname{tg}(\theta)\right] \mp \theta$$





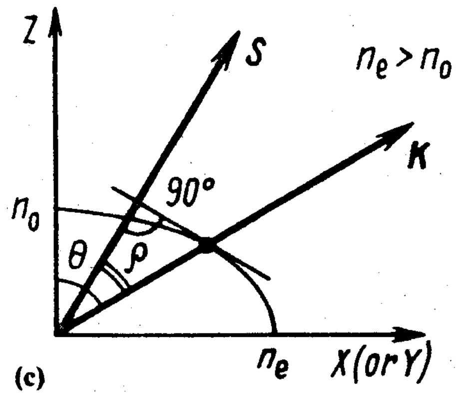


Fig. 2.6. Disposition of the wave (*k*) and **beam** (*s*) vectors in an isotropic medium (**a**) and anisotropic negative (**b**) and positive (**c**) uniaxial crystals (ρ is the birefringence or anisotropy angle)

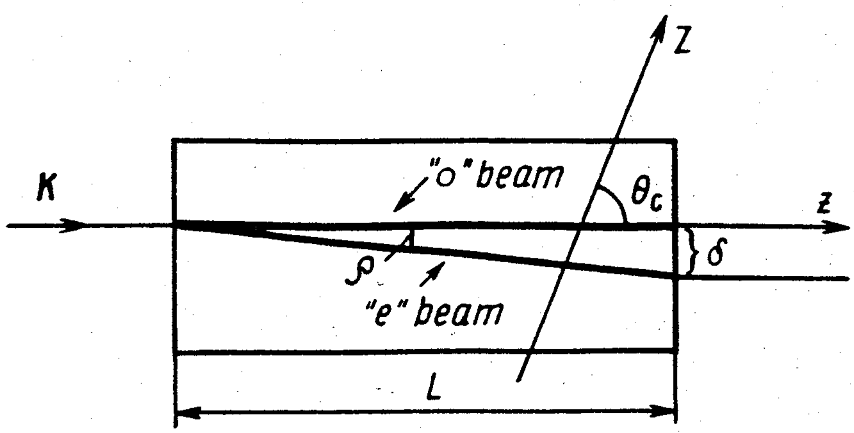


Fig. 2.7. Calculation of the cut angle θ_c in a uniaxial crystal