Second order nonlinear processes in optical crystals

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Part I. (6.12.2000)

the effective nonlinearity: symmetries, crystal classes and conventions

Part II. (13.12.2000)

- the phase-matching (collinear, non-collinear and quasi-phase matching)

- the bandwidths (acceptances): spectral, angular, temperature

- frequency conversion with ultrashort pulses

Literature:

1. R. W. Boyd, Nonlinear Optics, 1992
2. W. Kleber, Einführung in die Kristallographie, 1985
3. V. G. Dmitriev, G. G. Gurzadyan, D. N. Nikogosyan, Handbook of nonlinear optical crystals, 1999
4. R. L. Sutherland, Handbook of nonlinear optics, 1996

Δk: phase-mismatch

For collinear interaction: $\Delta k = k_1 + k_2 - k_3$.

$\Delta k = 0$ means that the generated wave (i. e. in SFG) remains always in phase with the nonlinear polarisation. The amplitude A_3 (or the intensity) increase linearly (quadratic) with z under the condition of low conversion efficiency (constant fields A_1 und A_2).

This is the *„fixed-field"-approximation.*

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In the stationary (without the term $\partial/\partial t \cdots$) „fixed-field"-case we can integrate the SFG equations to get at z=L:

$$A_3(z=L) = i\frac{2\omega_3 d_{eff} A_1(z=0)A_2(z=0)}{n_3 c}\int_0^L e^{i\Delta kz}dz = \frac{2\omega_3 d_{eff} A_1(0)A_2(0)}{n_3 c}\left(\frac{e^{i\Delta kL}-1}{\Delta k}\right)$$

Introducing the intensities: $I_n(z) = 2\varepsilon_0 n_n c\left|A_n(z)\right|^2$ we get:

$$I_3(L) = \frac{8\varepsilon_0\omega_3^2 d_{eff}^2 \left|A_1(0)\right|^2 \left|A_2(0)\right|^2}{n_3 c}\left|\frac{e^{i\Delta kL}-1}{\Delta k}\right|^2$$

with $$\left|\frac{e^{i\Delta kL}-1}{\Delta k}\right|^2 = L^2 \sin c^2\left(\frac{\Delta kL}{2}\right)$$

The conversion efficiency assuming $I_1(0) \ll I_2(0)$ is:

$$\eta_e = \frac{I_3(L)}{I_1(0)} = \frac{8\pi^2 d_{eff}^2 L^2 I_2(0)}{\varepsilon_0 c n_1 n_2 n_3 \lambda_3^2} \operatorname{sinc}^2\left(\frac{\Delta kL}{2}\right)$$

The bandwidth of the phase-matching (FWHM) is defined by the points where $\operatorname{sinc}^2(...)=0.5$: $\Delta k = 0.886\pi L^{-1}$

Coherence length $L_c = \pi / \Delta k$:

half the distance at which $\operatorname{sinc}^2(\Delta kL/2)$ drops to 0.

It determines the energy flow direction.

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If $\Delta k=0$ is realized by phase-matching then the conversion efficiency can be considerable and the „fixed-field"-approximation is violated. Nevertheless, for plane waves analytical formula for the conversion efficiency can be found in the literature [Sutherland].

In the simplest case of SHG ($\omega_1=\omega_2$) we have:

$$\eta_e = \frac{I_3(L)}{I_1(0)} = \tanh^2\left(\frac{L}{L_{NL}}\right) \quad \text{with} \quad L_{NL} = \frac{1}{4\pi d_{eff}} \sqrt{\frac{2\varepsilon_0 n_1^2 n_3 c \lambda_1^2}{I_1(0)}}$$

as a nonlinear length. With perfectly phase-matched SHG we get $\eta_e(L=L_{NL})=58\ \%$.

OPA: strong pump at ω_3 + weak seed (signal) at ω_2 propagated

In the stationary „fixed-field"-approximation [$I_3(z)=I_3(0)$] we obtain exponential gain for the signal and idler waves:

$$I_1(z) = I_2(0)\frac{\omega_1}{\omega_2}\sinh^2(gz)$$

$$I_2(z) = I_2(0)\cosh^2(gz)$$

Exponential parametric gain: $g = [g_0^{\,2} - (\Delta k/2)^2]^{1/2}$

where $g_0 = 2\sqrt{2}\pi d_{eff}[I_3(0)/\varepsilon_0 c n_1 n_2 n_3 \lambda_1 \lambda_2]^{1/2} = (L_{NL})^{-1}$.

For $gz>>1$ we obtain after expansion of the hyperbolic function:

$G_0 = I_2(z)/I_2(0) = 0.25\exp(2gz)$

Gain bandwidth:

$$\frac{1}{2} = \frac{\exp(2[g_0^2 - (\Delta k/2)^2]^{1/2} L)}{\exp(2g_0 L)} \quad \text{or} \quad \Delta k \approx 2\left(\frac{\ln 2g_0}{L}\right)^{1/2}$$

For $L>L_{NL}$: strong interaction and violated „fixed field" approximation. Approximate analytical solutions of the coupled equations with $\Delta k=0$ and $I_1(0)=0$ are known in terms of the Jacobi elliptic function (sn).

Phase-matching in birefringent nonlinear crystals

Collinear (scalar) phase-matching:

$$k_3 = k_2 + k_1 \qquad \text{or} \qquad \omega_3 n_3 = \omega_2 n_2 + \omega_1 n_1$$

This condition cannot be satisfied with waves having equal polarisation neither in isotropic nor in anisotropic crystals because of the dispersion.

In anisotropic crystals we can use, however, different polarisations!

a) We choose the propagation direction z with respect to the dielectric XYZ frame.

b) At a fixed propagation direction the phase-matching can be realized by temperature change.

Uniaxial crystals

Z: optical axis, Z and $\vec{k}$//z define the principal plane.

o-beam (ordinary) is polarized perpendicularly to this plane

e-beam (extraordinary) is polarized in this plane

n_o is independent of the propagation direction

$n^e(\theta)$ depends on the polar angle θ between Z and z.

$\Delta n = n_o - n_e$: birefringence.

$\Delta n = 0$ along Z and Δn=max for z in the XY plane.

In general:

$$n^e(\theta) = n_o \sqrt{\frac{1+\tan^2\theta}{1+\left(\frac{n_o}{n_e}\right)^2 \tan^2\theta}}$$

where n_o and $n_e=n^e(90°)$ denote the principal values.

Optical negative (positive) crystals are defined by $n_o>n_e$ ($n_o<n_e$).

Birefringence „walk-off" ange ρ for an e-beam: angle between the Poynting vector $\vec{V}$ (or **S**) and the $\vec{k}$-vector:

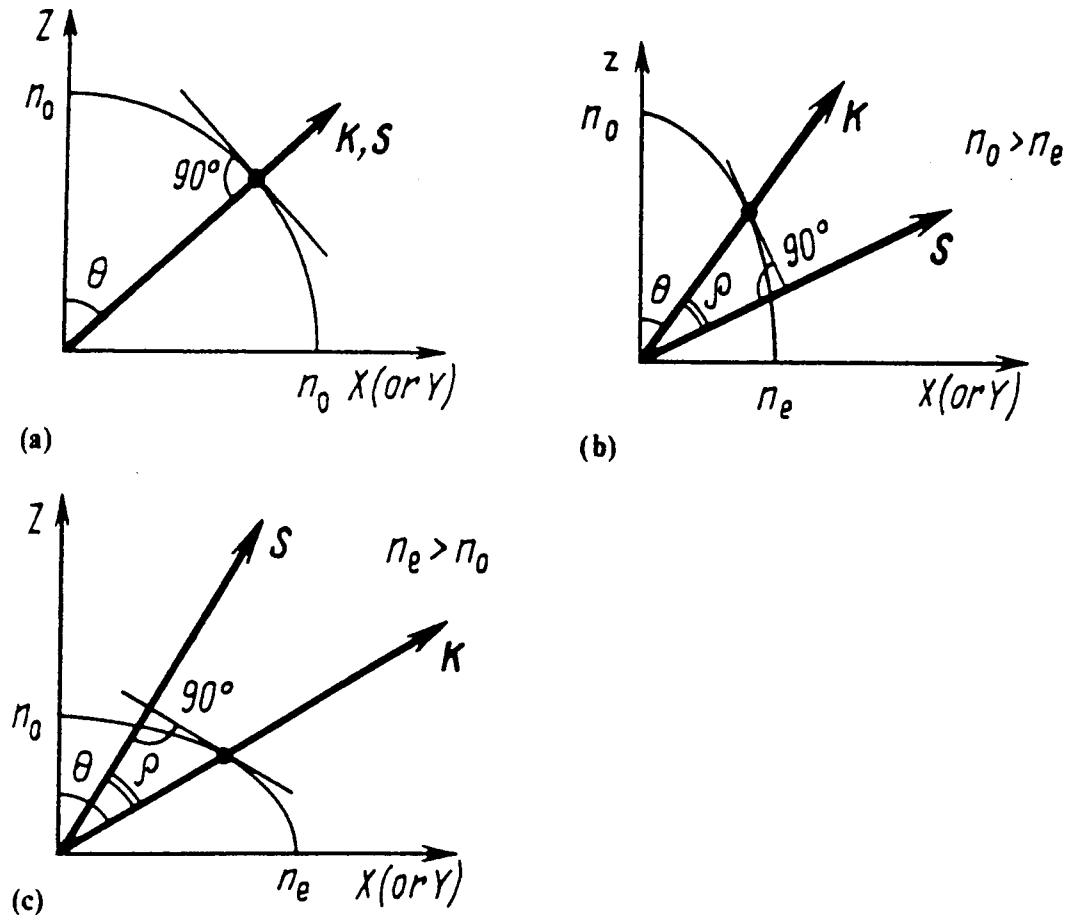
$$\rho(\theta) = \pm \arctan\left[\left(\frac{n_o}{n_e}\right)^2 \tan\theta\right] \mp \theta$$

[Upper (lower) sign for negative (positive) crystals].

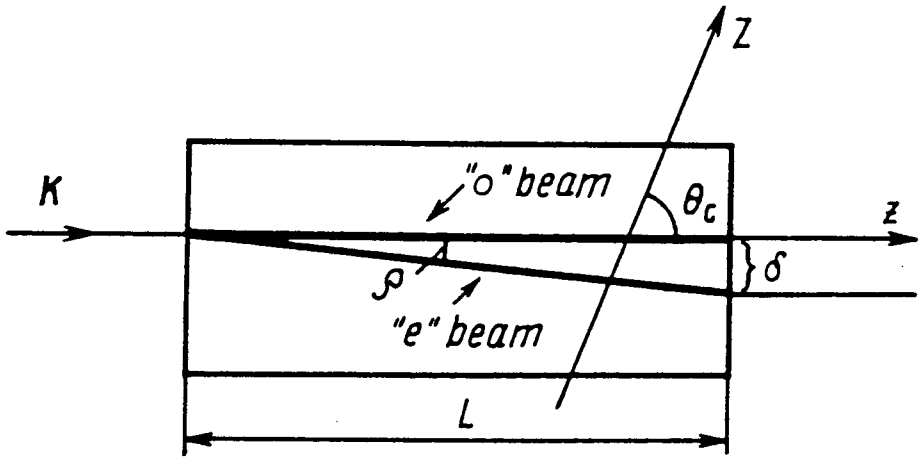
Real beams are Gaussian with a radial distribution of $w(z) = w_0[1+(z/z_R)^2]^{1/2}$ where w(z) is the radius (at $1/e^2$-level), $w_0 = w(0)$ is the waist and $z_R = n\pi w_0^2/\lambda$ denotes the Rayleigh-length. In this case for $\rho \neq 0$ the o- and e-beams walk-off after

$L_\rho = \sqrt{\pi} w_0 / \tan\rho$: spatial walk-off length or „aperture" length.

Birefringence effect: pronounced with focussed beams in long crystals.



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 angle)



Determination of the cut angle θ_c for the uniaxial crystal

Type I - phase-matching: the two waves at the lower frequencies ω_1 and ω_2 have equal polarisation.

Type II- phase-matching: the two waves at the lower frequencies ω_1 and ω_2 have perpendicular polarisation.

The 3 symbols „o" or „e" are always in the sequence λ_1, λ_2, λ_3.

The same convention is valid for DFG or OPA following always $\lambda_1 > \lambda_2 > \lambda_3$.

In negative crystals,

$$k_{o1} + k_{o2} = k_3^e(\theta)$$

(this is called "ooe" *phase matching* or "ooe" *interaction* or type $I^{(-)}$ *phase matching*). In positive crystals,

$$k_1^e(\theta) + k_2^e(\theta) = k_{o3}$$

("eeo" *phase matching* or "eeo" *interaction* or type- $I^{(+)}$ *phase matching*)

$$k_{o1} + k_2^e(\theta) = k_3^e(\theta)$$

("oee" *phase matching* or "oee" *interaction* or type $II^{(-)}$ *phase-matching*) or

$$k_1^e(\theta) + k_{o2} = k_3^e(\theta)$$

("eoe" *phase matching* or "eoe" interaction or type $II^{(-)}$ *phase-matching*); and to an ordinary wave in positive crystals:

$$k_{o1} + k_2^e(\theta) = k_{o3}$$

("oeo" *phase matching* or "oeo" *interaction* or type $II^{(+)}$ *phase-matching*), or

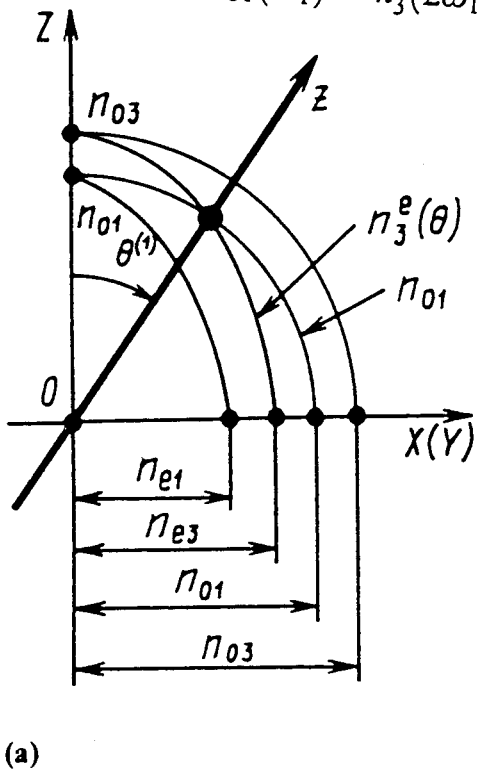
$$k_1^e(\theta) + k_{o2} = k_{o3}$$

("eoo" *phase matching* or "eoo" *interaction* or type $II^{(+)}$ *phase-matching*).

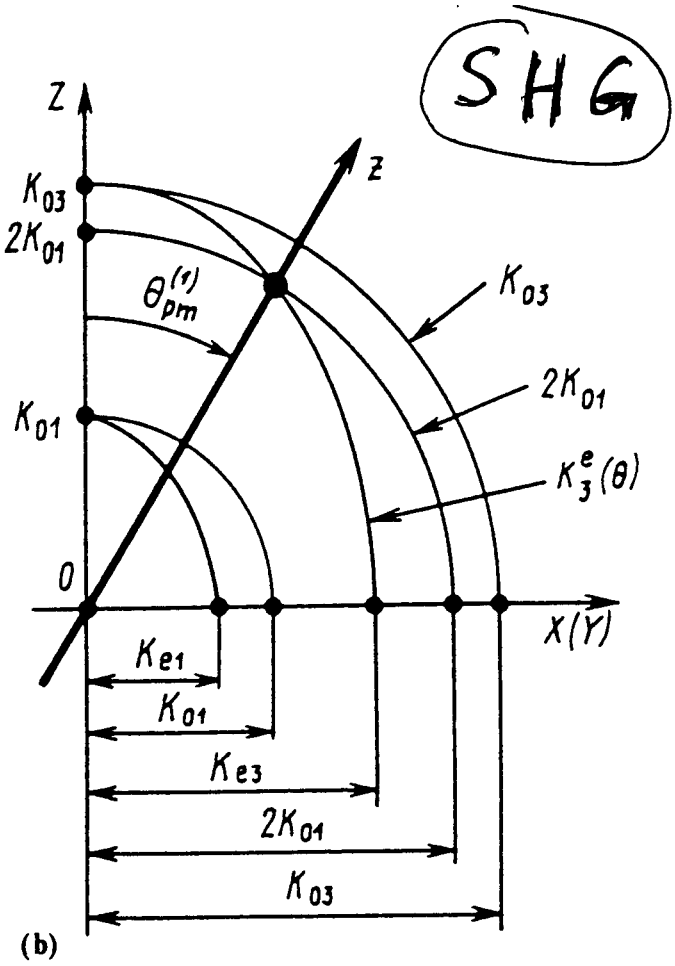
$$n_{o1}(\omega_1) = n_3^e(2\omega_1, \theta_{pm}^{(1)})$$

or

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

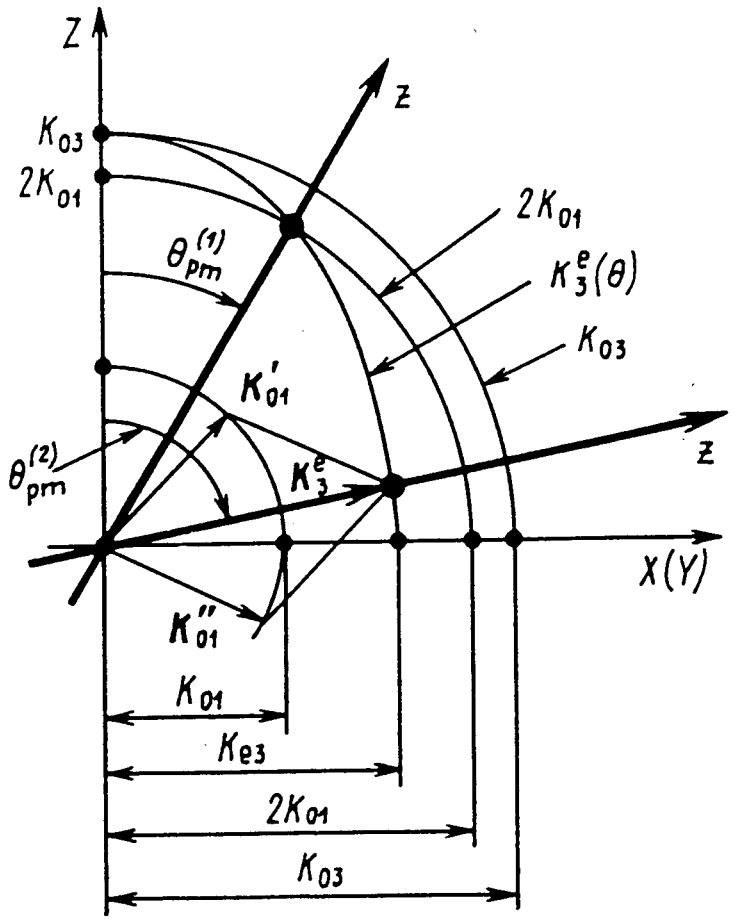


(a)



(b)

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



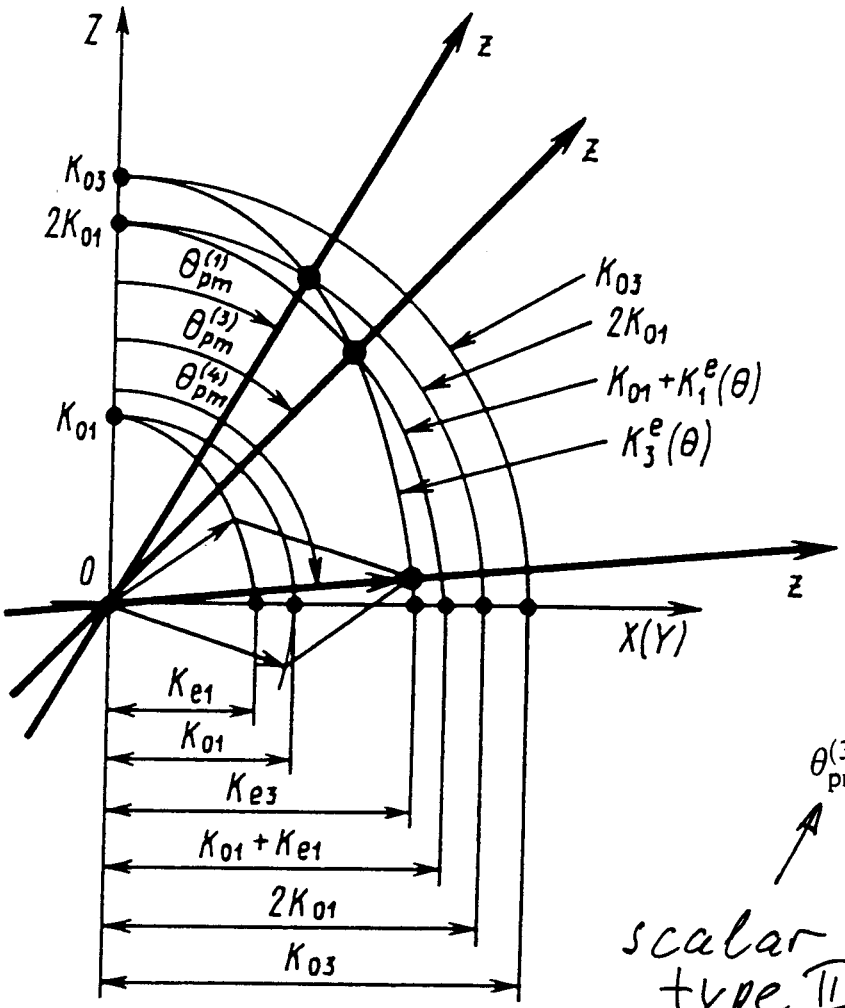
SHG

scalar

vector

$$\theta_{pm}^{(1)} \le \theta_{pm}^{(2)} \le \pi - \theta_{pm}^{(1)},$$

Scalar (collinear) and vector (noncollinear) phase matching of type I ("ooe") in a uniaxial negative crystal

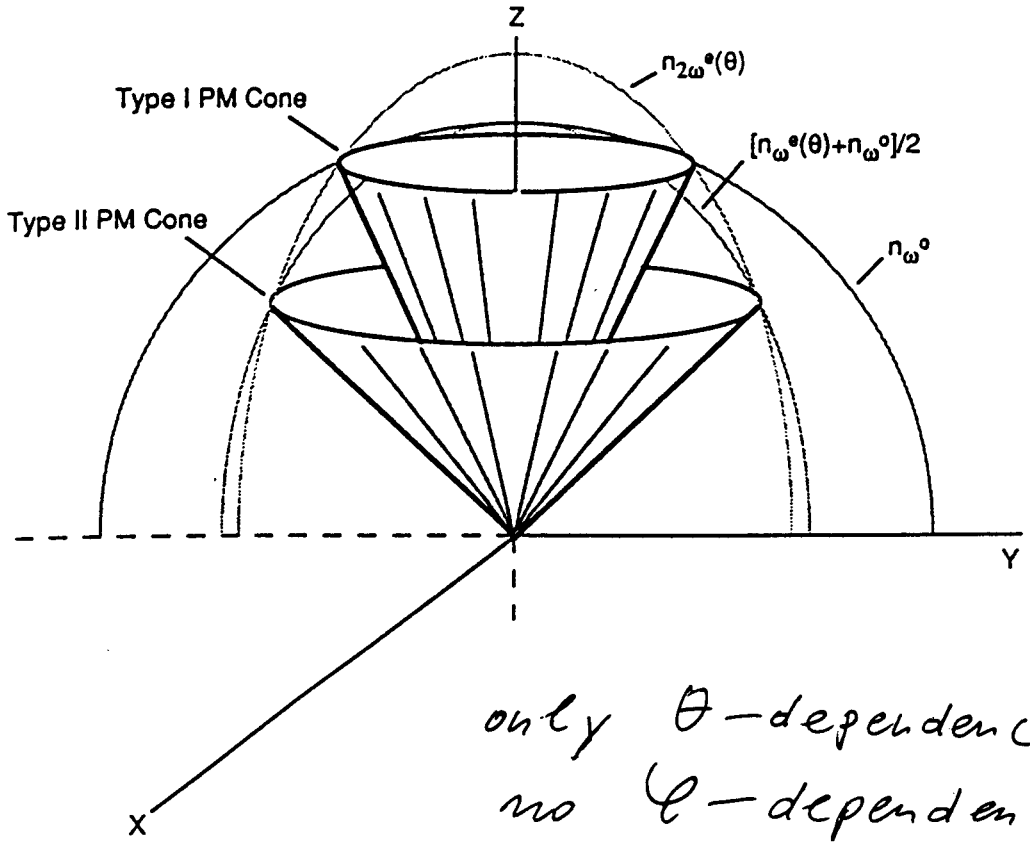


Scalar (collinear) phase matching of type I ("ooe") and type II ("eoe"), and vector (noncollinear) phase matching of type II ("eoe") in a uniaxial negative crystal

$$\theta_{pm}^{(3)} < \theta_{pm}^{(4)} < \pi - \theta_{pm}^{(3)}.$$

scalar type II

vector type II



only θ-dependence

no φ-dependence

Types I and II phase matching cones for SHG in a uniaxial crystal.

The phase-matching angle θ_{PM} is calculated by the Sellmeier-equations for $n_o(\lambda)$ and $n_e(\lambda)$.

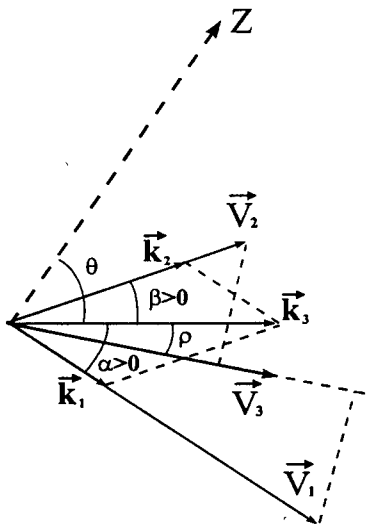
Temperature phase-matching: the angle θ is fixed but the temperature is varied.

Tuning at $\theta_{PM}=90°$ is possible in that case:

noncritical phase-matching with $\rho=0$.

(maximum angular acceptance)

Vector phase-matching:



Type-I (ooe) phase-matching in a negative uniaxial crystal.

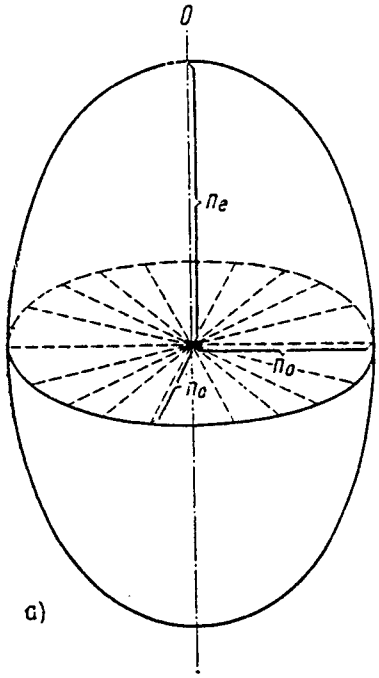
From $\vec{k}_3 = \vec{k}_2 + \vec{k}_1$ we calculate θ for a given β, and then α.

$$\left(k_{o1}\right)^2 = k_{o2}^2 + k_{e3}^2 - 2k_{o2}k_{e3}\cos\beta$$

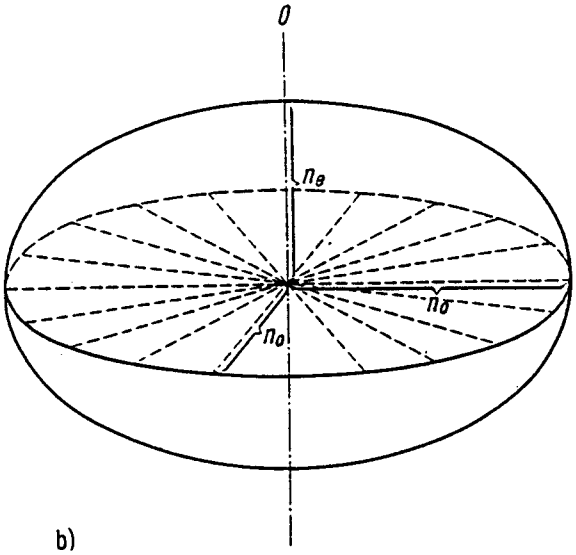
$$\left(k_{o2}\right)^2 = k_{o1}^2 + k_{e3}^2 - 2k_{o1}k_{e3}\cos\alpha$$

Optical Indicatrix

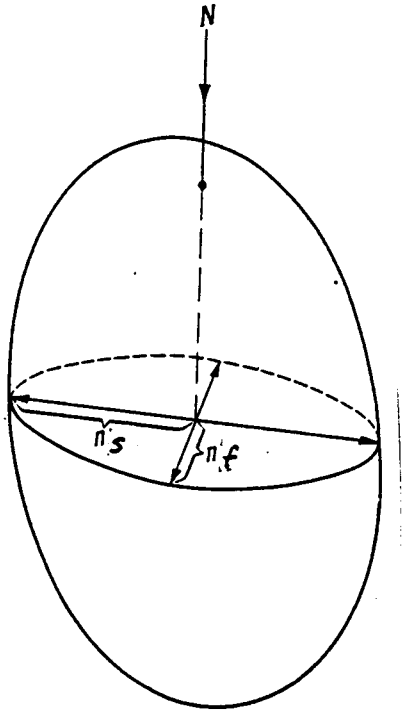
uniaxial-negative



uniaxial-positive



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



biaxial

n_s: slow wave

n_f: fast wave

$n_s \equiv n^{e_1}$

$n_f \equiv n^{e_2}$

roots of the Fresnel equation

In order to express this relation analytically, let angle $AZH = \phi$, angle $ZOH = \theta$, and δ be the angle between e_1 and ZH at the point H, which is measured clockwise for the positive value. Accordingly δ is expressed as a function of ϕ, θ, and Ω as follows[15]:

$$\cot 2\delta = \frac{\cot^2\Omega \sin^2\theta - \cos^2\theta \cos^2\phi + \sin^2\phi}{\cos\theta - \sin 2\phi},$$

where Ω is the angle between the Z axis and the optic axis.

The index surface for a given frequency in biaxial crystals is determined analytically by the two real solutions, n^{e_1} and n^{e_2}, of the equation

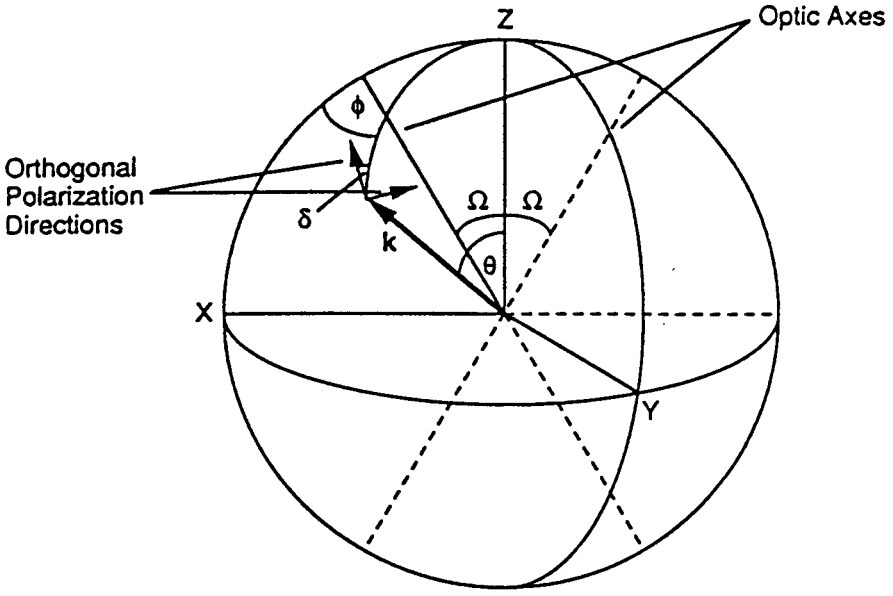
$$\frac{\sin^2\theta \cos^2\phi}{n^{-2} - (n^x)^{-2}} + \frac{\sin^2\theta \sin^2\phi}{n^{-2} - (n^y)^{-2}} + \frac{\cos^2\theta}{n^{-2} - (n^z)^{-2}} = 0,$$

where n^x, n^y, and n^z are principal refractive indices.

$0 < \delta < \pi/2$

$n_x < n_y < n_z$

Frequency Doubling and Mixing



Definitions of angles for propagation in a biaxial crystal used in the formulas

Biaxial crystals: *phase-matching determined both by θ and φ.*

The indices of refraction are defined in the dielectric frame XYZ (or xyz) for polarisation parallel to the corresponding axis:

we have 3 Sellmeier equations for the principal values n_X, n_Y und n_Z.

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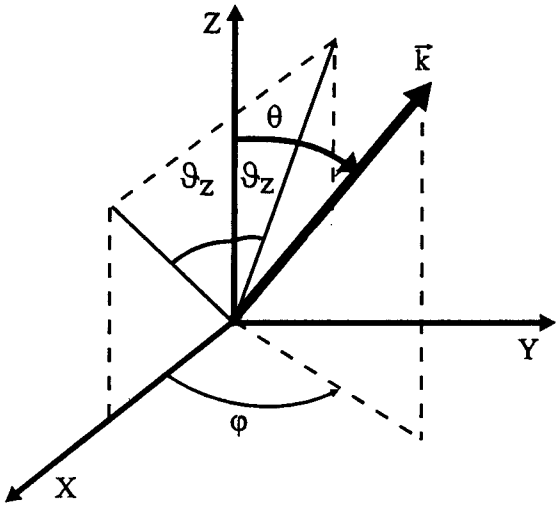
The 2 optical axes which lie per definition in the XZ plane which means that we assume either $n_X<n_Y<n_Z$ or $n_X>n_Y>n_Z$.

They intersect at $2\vartheta_Z=2\Omega$ and lie symmetrically with respect to Z:



In the principal planes XY, XZ or YZ biaxial crystals can be regarded as uniaxial and the terms „o“ and „e“ can be used, too. E.g. assuming $n_X<n_Y<n_Z$, in the XY plane we have an analogy with a negative uniaxial crystal defining $n_o=n_Z$ and $n^e(\varphi)$ by:

$$n^e(\varphi) = n_Y \frac{\left(1+\tan^2\varphi\right)^{1/2}}{\left[1+\left(\frac{n_Y}{n_X}\right)^2 \tan^2\varphi\right]^{1/2}}$$

Equations for calculating phase-matching angles in uniaxial crystals

Negative uniaxial crystals	Positive uniaxial crystals
$\tan^2\theta_{pm}^{ooe} = (1-U)/(W-1)$	$\tan^2\theta_{pm}^{eeo} \cong (1-U)/(U-S)$
$\tan^2\theta_{pm}^{eoe} \cong (1-U)/(W-R)$	$\tan^2\theta_{pm}^{oeo} = (1-V)/(V-Y)$
$\tan^2\theta_{pm}^{oee} \cong (1-U)/(W-Q)$	$\tan^2\theta_{pm}^{eoo} = (1-T)/(T-Z)$

Notations:
$U = (A+B)^2/C^2$; $W = (A+B)^2/F^2$; $R = (A+B)^2/(D+B)^2$;
$Q = (A+B)^2/(A+E)^2$; $S = (A+B)^2/(D+E)^2$; $V = B^2/(C-A)^2$;
$Y = B^2/E^2$; $T = A^2/(C-B)^2$; $Z = A^2/D^2$;
$A = n_{o1}/\lambda_1$; $B = n_{o2}/\lambda_2$; $C = n_{o3}/\lambda_3$;
$D = n_{e1}/\lambda_1$; $E = n_{e2}/\lambda_2$; $F = n_{e3}/\lambda_3$.

The expressions presented in Table 2.1 can be generalized to the noncollinear phase matching. In this case, for example, the phase-matching angle θ_{pm}^{ooe} is determined from the above presented equation using the new coefficients U and W:

$$U = (A^2 + B^2 + 2AB\cos\gamma)/C^2, \quad W = (A^2 + B^2 + 2AB\cos\gamma)/F^2$$

where γ is the angle between wave vectors $\boldsymbol{k}_1$ and $\boldsymbol{k}_2$.

Equations for calculating phase-matching angles in biaxial crystals upon light propagation in the principal planes

(a) $n_X < n_Y < n_Z$

Principal plane	Type of interaction	Equations	Notations
1	2	3	4
XY	ooe	$\tan^2\phi = \frac{1-U}{W-1}$	$U = \left(\frac{A+B}{C}\right)^2;\ W = \left(\frac{A+B}{F}\right)^2;\ A = \frac{n_{Z1}}{\lambda_1};\ B = \frac{n_{Z2}}{\lambda_2};\ C = \frac{n_{Y3}}{\lambda_3};\ F = \frac{n_{X3}}{\lambda_3}$
	eoe	$\tan^2\phi \cong \frac{1-U}{W-R}$	$U = \left(\frac{A+B}{C}\right)^2;\ W = \left(\frac{A+B}{F}\right)^2;\ R = \left(\frac{A+B}{D+B}\right)^2;\ A = \frac{n_{Y1}}{\lambda_1};\ B = \frac{n_{Z2}}{\lambda_2};\ C = \frac{n_{Y3}}{\lambda_3};\ D = \frac{n_{X1}}{\lambda_1};\ F = \frac{n_{X3}}{\lambda_3}$
	oee	$\tan^2\phi \cong \frac{1-U}{W-Q}$	$U = \left(\frac{A+B}{C}\right)^2;\ W = \left(\frac{A+B}{F}\right)^2;\ Q = \left(\frac{A+B}{A+E}\right)^2;\ A = \frac{n_{Z1}}{\lambda_1};\ B = \frac{n_{Y2}}{\lambda_2};\ C = \frac{n_{Y3}}{\lambda_3};\ E = \frac{n_{X2}}{\lambda_2};\ F = \frac{n_{X3}}{\lambda_3}$
YZ	eeo	$\tan^2\theta \cong \frac{1-U}{U-S}$	$U = \left(\frac{A+B}{C}\right)^2;\ S = \left(\frac{A+B}{D+E}\right)^2;\ A = \frac{n_{Y1}}{\lambda_1};\ B = \frac{n_{Y2}}{\lambda_2};\ C = \frac{n_{X3}}{\lambda_3};\ D = \frac{n_{Z1}}{\lambda_1};\ E = \frac{n_{Z2}}{\lambda_2}$
	oeo	$\tan^2\theta = \frac{1-V}{V-Y}$	$V = \left(\frac{B}{C-A}\right)^2;\ Y = \left(\frac{B}{E}\right)^2;\ A = \frac{n_{X1}}{\lambda_1};\ B = \frac{n_{Y2}}{\lambda_2};\ C = \frac{n_{X3}}{\lambda_3};\ E = \frac{n_{Z2}}{\lambda_2}$
	eoo	$\tan^2\theta = \frac{1-T}{T-Z}$	$T = \left(\frac{A}{C-B}\right)^2;\ Z = \left(\frac{A}{D}\right)^2;\ A = \frac{n_{Y1}}{\lambda_1};\ B = \frac{n_{X2}}{\lambda_2};\ C = \frac{n_{X3}}{\lambda_3};\ D = \frac{n_{Z1}}{\lambda_1}$
XZ	ooe	$\tan^2\theta = \frac{1-U}{W-1}$	$U = \left(\frac{A+B}{C}\right)^2;\ W = \left(\frac{A+B}{F}\right)^2;\ A = \frac{n_{Y1}}{\lambda_1};\ B = \frac{n_{Y2}}{\lambda_2};\ C = \frac{n_{X3}}{\lambda_3};\ F = \frac{n_{Z3}}{\lambda_3}$
$\theta < V_Z$	eoe	$\tan^2\theta \cong \frac{1-U}{W-R}$	$U = \left(\frac{A+B}{C}\right)^2;\ W = \left(\frac{A+B}{F}\right);\ R = \left(\frac{A+B}{D+B}\right)^2;\ A = \frac{n_{X1}}{\lambda_1};\ B = \frac{n_{Y2}}{\lambda_2};\ C = \frac{n_{X3}}{\lambda_3};\ D = \frac{n_{Z1}}{\lambda_1};\ F = \frac{n_{Z3}}{\lambda_3}$
	oee	$\tan^2\theta \cong \frac{1-U}{W-Q}$	$U = \left(\frac{A+B}{C}\right)^2;\ W = \left(\frac{A+B}{F}\right)^2;\ Q = \left(\frac{A+B}{A+E}\right)^2;\ A = \frac{n_{Y1}}{\lambda_1};\ B = \frac{n_{X2}}{\lambda_2};\ C = \frac{n_{X3}}{\lambda_3};\ E = \frac{n_{Z2}}{\lambda_2};\ F = \frac{n_{Z3}}{\lambda_3}$
XZ	eeo	$\tan^2\theta \cong \frac{1-U}{U-S}$	$U = \left(\frac{A+B}{C}\right)^2;\ S = \left(\frac{A+B}{D+E}\right)^2;\ A = \frac{n_{X1}}{\lambda_1};\ B = \frac{n_{X2}}{\lambda_2};\ C = \frac{n_{Y3}}{\lambda_3};\ D = \frac{n_{Z1}}{\lambda_1};\ E = \frac{n_{Z2}}{\lambda_2}$
$\theta > V_Z$	oeo	$\tan^2\theta = \frac{1-V}{V-Y}$	$V = \left(\frac{B}{C-A}\right)^2;\ Y = \left(\frac{B}{E}\right)^2;\ A = \frac{n_{Y1}}{\lambda_1};\ B = \frac{n_{X2}}{\lambda_2};\ C = \frac{n_{Y3}}{\lambda_3};\ E = \frac{n_{Z2}}{\lambda_2}$
	eoo	$\tan^2\theta = \frac{1-T}{T-Z}$	$T = \left(\frac{A}{C-B}\right)^2;\ Z = \left(\frac{A}{D}\right)^2;\ A = \frac{n_{X1}}{\lambda_1};\ B = \frac{n_{Y2}}{\lambda_2};\ C = \frac{n_{Y3}}{\lambda_3};\ D = \frac{n_{Z1}}{\lambda_1}$

(b) $n_X > n_Y > n_Z$

XY	eeo	$\tan^2\phi \cong \frac{1-U}{U-S}$	$U = \left(\frac{A+B}{C}\right)^2;\ S = \left(\frac{A+B}{D+E}\right)^2;\ A = \frac{n_{Y1}}{\lambda_1};\ B = \frac{n_{Y2}}{\lambda_2};\ C = \frac{n_{Z3}}{\lambda_3};\ D = \frac{n_{X1}}{\lambda_1};\ E = \frac{n_{X2}}{\lambda_2}$
	oeo	$\tan^2\phi = \frac{1-V}{V-Y}$	$V = \left(\frac{B}{C-A}\right)^2;\ Y = \left(\frac{B}{E}\right)^2;\ A = \frac{n_{Z1}}{\lambda_1};\ B = \frac{n_{Y2}}{\lambda_2};\ C = \frac{n_{Z3}}{\lambda_3};\ E = \frac{n_{X2}}{\lambda_2}$
	eoo	$\tan^2\phi = \frac{1-T}{T-Z}$	$T = \left(\frac{A}{C-B}\right)^2;\ Z = \left(\frac{A}{D}\right)^2;\ A = \frac{n_{Y1}}{\lambda_1};\ B = \frac{n_{Z2}}{\lambda_2};\ C = \frac{n_{Z3}}{\lambda_3};\ D = \frac{n_{X1}}{\lambda_1}$
YZ	ooe	$\tan^2\theta = \frac{1-U}{W-1}$	$U = \left(\frac{A+B}{C}\right)^2;\ W = \left(\frac{A+B}{F}\right)^2;\ A = \frac{n_{X1}}{\lambda_1};\ B = \frac{n_{X2}}{\lambda_2};\ C = \frac{n_{Y3}}{\lambda_3};\ F = \frac{n_{Z3}}{\lambda_3}$
	eoe	$\tan^2\theta \cong \frac{1-U}{W-R}$	$U = \left(\frac{A+B}{C}\right)^2;\ W = \left(\frac{A+B}{F}\right)^2;\ R = \left(\frac{A+B}{D+B}\right)^2;\ A = \frac{n_{Y1}}{\lambda_1};\ B = \frac{n_{X2}}{\lambda_2};\ C = \frac{n_{Y3}}{\lambda_3};\ D = \frac{n_{Z1}}{\lambda_1};\ F = \frac{n_{Z3}}{\lambda_3}$
	oee	$\tan^2\theta \cong \frac{1-U}{W-Q}$	$U = \left(\frac{A+B}{C}\right)^2;\ W = \left(\frac{A+B}{F}\right)^2;\ Q = \left(\frac{A+B}{A+E}\right)^2;\ A = \frac{n_{X1}}{\lambda_1};\ B = \frac{n_{Y2}}{\lambda_2};\ C = \frac{n_{Y3}}{\lambda_3};\ E = \frac{n_{Z2}}{\lambda_2};\ F = \frac{n_{Z3}}{\lambda_3}$
XZ	eeo	$\tan^2\theta \cong \frac{1-U}{U-S}$	$U = \left(\frac{A+B}{C}\right)^2;\ S = \left(\frac{A+B}{D+E}\right)^2;\ A = \frac{n_{X1}}{\lambda_1};\ B = \frac{n_{X2}}{\lambda_2};\ C = \frac{n_{Y3}}{\lambda_3};\ D = \frac{n_{Z1}}{\lambda_1};\ E = \frac{n_{Z2}}{\lambda_2}$
$\theta < V_Z$	oeo	$\tan^2\theta = \frac{1-V}{V-Y}$	$V = \left(\frac{B}{C-A}\right)^2;\ Y = \left(\frac{B}{E}\right)^2;\ A = \frac{n_{Y1}}{\lambda_1};\ B = \frac{n_{X2}}{\lambda_2};\ C = \frac{n_{Y3}}{\lambda_3};\ E = \frac{n_{Z2}}{\lambda_2}$
	eoo	$\tan^2\theta = \frac{1-T}{T-Z}$	$T = \left(\frac{A}{C-B}\right)^2;\ Z = \left(\frac{A}{D}\right)^2;\ A = \frac{n_{X1}}{\lambda_1};\ B = \frac{n_{Y2}}{\lambda_2};\ C = \frac{n_{Y3}}{\lambda_3};\ D = \frac{n_{Z1}}{\lambda_1}$
XZ	ooe	$\tan^2\theta = \frac{1-U}{W-1}$	$U = \left(\frac{A+B}{C}\right)^2;\ W = \left(\frac{A+B}{F}\right)^2;\ A = \frac{n_{Y1}}{\lambda_1};\ B = \frac{n_{Y2}}{\lambda_2};\ C = \frac{n_{X3}}{\lambda_3};\ F = \frac{n_{Z3}}{\lambda_3}$
$\theta > V_Z$	eoe	$\tan^2\theta \cong \frac{1-U}{W-R}$	$U = \left(\frac{A+B}{C}\right)^2;\ W = \left(\frac{A+B}{F}\right)^2;\ R = \left(\frac{A+B}{D+B}\right)^2;\ A = \frac{n_{X1}}{\lambda_1};\ B = \frac{n_{Y2}}{\lambda_2};\ C = \frac{n_{X3}}{\lambda_3};\ D = \frac{n_{Z1}}{\lambda_1};\ F = \frac{n_{Z3}}{\lambda_3}$
	oee	$\tan^2\theta \cong \frac{1-U}{W-Q}$	$U = \left(\frac{A+B}{C}\right)^2;\ W = \left(\frac{A+B}{F}\right)^2;\ Q = \left(\frac{A+B}{A+E}\right)^2;\ A = \frac{n_{Y1}}{\lambda_1};\ B = \frac{n_{X2}}{\lambda_2};\ C = \frac{n_{X3}}{\lambda_3};\ E = \frac{n_{Z2}}{\lambda_2};\ F = \frac{n_{Z3}}{\lambda_3}$

Periodically poled crystals and quasi-phase-matching

QPM

With QPM one can achieve tunable noncritical (90°) phase-matching with larger d_{eff} because of the free polarisation choice (utilize d_{33}).

QPM is possible in crystals with insufficient or vanishing birefringence.

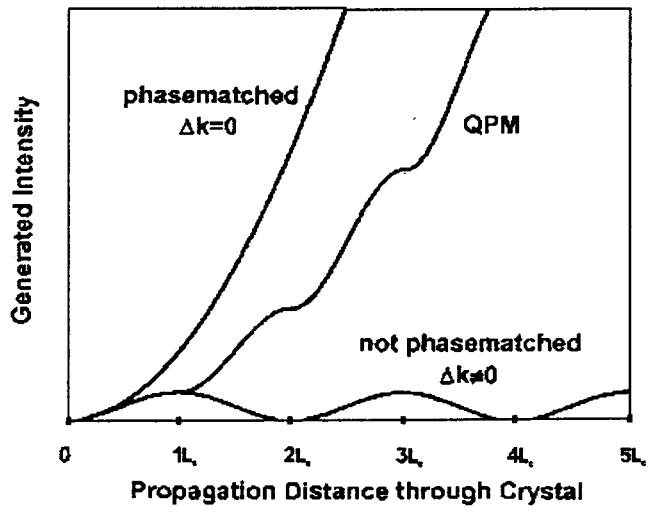
In general it corrects too small Δn but also too large Δn.

The idea: the nonlinear coefficient is modulated with a period that compensates the accumulated phase-mismatch, e.g. stationary SFG:

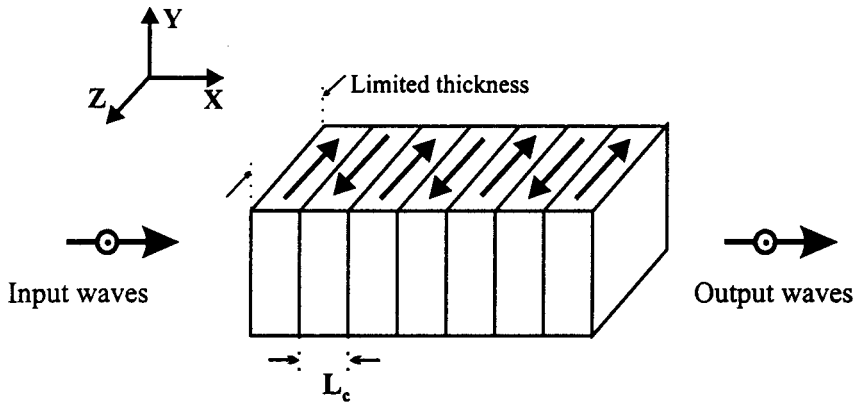
$$\frac{\partial A_3(z)}{\partial z} \propto d_{eff} A_1(z) A_2(z) e^{i\Delta kz}$$

180° Poling of the structure e.g. ferroelectric materials means:

$-d_{eff}=d_{eff}\exp(i\pi)$ instead of d_{eff} or addition of π to the phase Δkz.



QPM is not perfect and is less efficient than birefringent phase-matching but one can utilize larger nonlinearity:



Effective (1-st order QPM) nonlinearity:

$$d_Q = \frac{2}{\pi} d_{eff}$$

Example:

$LiNbO_3$-crystal, the largest nonlinearity d_{33}, (6 times d_{31}) can be utilized for all polarisations parallel to the Z-axis.

$(d_{33}/d_{31})^2(2/\pi)^2 \approx 15$: expected increase in the conversion efficiency

Phase-mismatch in QPM, Δk_Q:

$$\Delta k_Q = 2\pi\left(\frac{n_1}{\lambda_1} + \frac{n_2}{\lambda_2} - \frac{n_3}{\lambda_3} + \frac{1}{\Lambda_g}\right) = k_1 + k_2 - k_3 + \frac{2\pi}{\Lambda_g}$$

with grating period $\Lambda_g = 2L_c = \dfrac{2\pi}{|k_1 + k_2 - k_3|}$

The first three terms can be influnced by temperature: fine tuning.

The last term is an additional parameter of choice: „engineerable“.

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Ferroelectric materials:

$LiNbO_3$, $KNbO_3$, $KTiOPO_4$ or $LiTaO_3$.

Photolithography + electric field poling

(21 kV/mm along Z for $LiNbO_3$).

GaAs und ZnSe: slicing (difusion bonding) of ≈100 μm thick plates with opposite orientation possible because the coherence length is large in the MIR.

Bandwidths:

The angular ($\Delta\theta$), spectral ($\Delta\nu$) and temperature acceptance (ΔT) correspond to the maximum allowed divergence and spectral width of the radiation and the temperature stability.

First order expansion of Δk gives:

$$\Delta k(T,\theta,\nu) \cong \frac{\partial(\Delta k)}{\partial T}\Delta T + \frac{\partial(\Delta k)}{\partial \theta}\Delta\theta + \frac{\partial(\Delta k)}{\partial \nu}\Delta\nu$$

For SFG and DFG in the „fixed-field"-approximation we arrive at:

$$\Delta\nu = 1.772\frac{\pi}{L}\left(\frac{\partial(\Delta k)}{\partial \nu}\right)^{-1}_{\nu_{PM}}$$

$$\Delta T = 1.772\frac{\pi}{L}\left(\frac{\partial(\Delta k)}{\partial T}\right)^{-1}_{T_{PM}}$$

$$\Delta\theta = 1.772\frac{\pi}{L}\left(\frac{\partial(\Delta k)}{\partial \theta}\right)^{-1}_{\theta_{PM}}$$

For $\theta_{pm} = 90°$ (90° *phase matching*) the first derivative $\partial(\Delta k)/\partial(\delta\theta)$ becomes equal to zero and the corresponding second derivative becomes important. Hence, the 90° phase-matching internal angular bandwidth is

$$\Delta\theta|_{\theta_{pm}=90°} \simeq 2\left[0.886\frac{\pi}{L}\left(\frac{\partial^2(\Delta k)}{\partial(\delta\theta)^2}\right)^{-1}\right]^{1/2}.$$

Equations for calculating the SHG internal angular bandwidth for the different types of interaction

Type of interaction	Internal angular bandwidth for SHG ($\omega_1 + \omega_1 = \omega_2$)
ooe	$\Delta\theta = \dfrac{0.443\lambda_1\left[1 + (n_{o2}/n_{e2})^2 \tan^2\theta\right]}{L\tan\theta\left\lvert 1 - (n_{o2}/n_{e2})^2\right\rvert n_2^e(\theta)}$
eoe, oee	$\Delta\theta = \dfrac{0.886}{L\tan\theta}\left\lvert \dfrac{n_1^e(\theta)\left[1 - (n_{o1}/n_{e1})^2\right]}{\lambda_1\left[1 + (n_{o1}/n_{e1})^2\tan^2\theta\right]} - \dfrac{n_2^e(\theta)\left[1 - (n_{o2}/n_{e2})^2\right]}{\lambda_2\left[1 + (n_{o2}/n_{e2})^2\tan^2\theta\right]}\right\rvert^{-1}$
eeo	$\Delta\theta = \dfrac{0.443\lambda_1\left[1 + (n_{o1}/n_{e1})^2\tan^2\theta\right]}{L\tan\theta\left[1 - (n_{o1}/n_{e1})^2\right]n_1^e(\theta)}$
eoo, oeo	$\Delta\theta = \dfrac{0.886\lambda_1\left[1 + (n_{o1}/n_{e1})^2\tan^2\theta\right]}{L\tan\theta\left[1 - (n_{o1}/n_{e1})^2\right]n_1^e(\theta)}$

Equations for calculating the SFG internal angular bandwidth for the different types of interaction

Type of interaction	Internal angular bandwidth for SFG ($\omega_1 + \omega_2 = \omega_3$)
ooe	$\Delta\theta = \dfrac{0.886\lambda_3\left[1 + (n_{o3}/n_{e3})^2\tan^2\theta\right]}{L\tan\theta\left\lvert 1 - (n_{o3}/n_{e3})^2\right\rvert n_3^e(\theta)}$
eoe	$\Delta\theta = \dfrac{0.886}{L\tan\theta}\left\lvert \dfrac{n_1^e(\theta)\left[1 - (n_{o1}/n_{e1})^2\right]}{\lambda_1\left[1 + (n_{o1}/n_{e1})^2\tan^2\theta\right]} - \dfrac{n_3^e(\theta)\left[1 - (n_{o3}/n_{e3})^2\right]}{\lambda_3\left[1 + (n_{o3}/n_{e3})^2\tan^2\theta\right]}\right\rvert^{-1}$
oee	$\Delta\theta = \dfrac{0.886}{L\tan\theta}\left\lvert \dfrac{n_2^e(\theta)\left[1 - (n_{o2}/n_{e2})^2\right]}{\lambda_2\left[1 + (n_{o2}/n_{e2})^2\tan^2\theta\right]} - \dfrac{n_3^e(\theta)\left[1 - (n_{o3}/n_{e3})^2\right]}{\lambda_3\left[1 + (n_{o3}/n_{e3})^2\tan^2\theta\right]}\right\rvert^{-1}$
eeo	$\Delta\theta = \dfrac{0.886}{L\tan\theta}\left\{\dfrac{n_1^e(\theta)\left[1 - (n_{o1}/n_{e1})^2\right]}{\lambda_1\left[1 + (n_{o1}/n_{e1})^2\tan^2\theta\right]} + \dfrac{n_2^e(\theta)\left[1 - (n_{o2}/n_{e2})^2\right]}{\lambda_2\left[1 + (n_{o2}/n_{e2})^2\tan^2\theta\right]}\right\}^{-1}$
eoo	$\Delta\theta = \dfrac{0.886\lambda_1\left[1 + (n_{o1}/n_{e1})^2\tan^2\theta\right]}{L\tan\theta\left[1 - (n_{o1}/n_{e1})^2\right]n_1^e(\theta)}$
oeo	$\Delta\theta = \dfrac{0.886\lambda_2\left[1 + (n_{o2}/n_{e2})^2\tan^2\theta\right]}{L\tan\theta\left[1 - (n_{o2}/n_{e2})^2\right]n_2^e(\theta)}$

Equations for calculating the SHG internal angular 90° phase-matching bandwidth for all types of interaction

Types of Interaction	Internal angular 90° phase-matching bandwidth for SHG ($\omega_1 + \omega_1 = \omega_2$)
ooe	$\Delta\theta = 2\left(\dfrac{0.443\lambda_1}{Ln_{e2}[1-(n_{e2}/n_{o2})^2]}\right)^{1/2}$
eoe oee	$\Delta\theta = 2\left(\dfrac{0.886}{L}\left\lvert\dfrac{n_{e1}}{\lambda_1}\left[1-\left(\dfrac{n_{e1}}{n_{o1}}\right)^2\right]-\dfrac{n_{e2}}{\lambda_2}\left[1-\left(\dfrac{n_{e2}}{n_{o2}}\right)^2\right]\right\rvert^{-1}\right)^{1/2}$
eeo	$\Delta\theta = 2\left(\dfrac{0.443\lambda_1}{Ln_{e1}\lvert 1-(n_{e1}/n_{o1})^2\rvert}\right)^{1/2}$
eoo oeo	$\Delta\theta = 2\left(\dfrac{0.886\lambda_1}{Ln_{e1}\lvert 1-(n_{e1}/n_{o1})^2\rvert}\right)^{1/2}$

Equations for calculating the SFG internal angular 90° phase-matching bandwidth for all types of interaction

Types of interaction	Internal angular 90° phase-matching bandwidth for SFG ($\omega_1 + \omega_2 = \omega_3$)
ooe	$\Delta\theta = 2\left(\dfrac{0.886\lambda_3}{Ln_{e3}\left[1-(n_{e3}/n_{o3})^2\right]}\right)^{1/2}$
eoe	$\Delta\theta = 2\left(\dfrac{0.886}{L}\left\lvert\dfrac{n_{e1}}{\lambda_1}\left[1-\left(\dfrac{n_{e1}}{n_{o1}}\right)^2\right]-\dfrac{n_{e3}}{\lambda_3}\left[1-\left(\dfrac{n_{e3}}{n_{o3}}\right)^2\right]\right\rvert^{-1}\right)^{1/2}$
oee	$\Delta\theta = 2\left(\dfrac{0.886}{L}\left\lvert\dfrac{n_{e2}}{\lambda_2}\left[1-\left(\dfrac{n_{e2}}{n_{o2}}\right)^2\right]-\dfrac{n_{e3}}{\lambda_3}\left[1-\left(\dfrac{n_{e3}}{n_{o3}}\right)^2\right]\right\rvert^{-1}\right)^{1/2}$
eeo	$\Delta\theta = 2\left(\dfrac{0.886}{L}\left\lvert\dfrac{n_{e1}}{\lambda_1}\left[1-\left(\dfrac{n_{e1}}{n_{o1}}\right)^2\right]+\dfrac{n_{e2}}{\lambda_2}\left[1-\left(\dfrac{n_{e2}}{n_{o2}}\right)^2\right]\right\rvert^{-1}\right)^{1/2}$
eoo	$\Delta\theta = 2\left(\dfrac{0.886\lambda_1}{Ln_{e1}\lvert 1-(n_{e1}/n_{o1})^2\rvert}\right)^{1/2}$
oeo	$\Delta\theta = 2\left(\dfrac{0.886\lambda_2}{Ln_{e2}\lvert 1-(n_{e2}/n_{o2})^2\rvert}\right)^{1/2}$

Equations for calculating the SHG temperature bandwith for the different types of interaction

Type of interaction	Temperature bandwidth for SHG $\omega_1 + \omega_1 = \omega_2$
ooe	$\Delta T = \frac{0.443\lambda_1}{L}\left\lvert\frac{\partial n_{o1}}{\partial T} - \frac{\partial n_2^e(\theta)}{\partial T}\right\rvert^{-1}$
eoe, oee	$\Delta T = \frac{0.886\lambda_1}{L}\left\lvert\frac{\partial n_1^e(\theta)}{\partial T} + \frac{\partial n_{o1}}{\partial T} - \frac{2\partial n_2^e(\theta)}{\partial T}\right\rvert^{-1}$
eeo	$\Delta T = \frac{0.443\lambda_1}{L}\left\lvert\frac{\partial n_1^e(\theta)}{\partial T} - \frac{\partial n_{o2}}{\partial T}\right\rvert^{-1}$
eoo, oeo	$\Delta T = \frac{0.886\lambda_1}{L}\left\lvert\frac{\partial n_1^e(\theta)}{\partial T} + \frac{\partial n_{o1}}{\partial T} - \frac{2\partial n_{o2}}{\partial T}\right\rvert^{-1}$

Equations for calculating the SFG temperature bandwidth for the different types of interaction

Type of interaction	Temperature bandwidth for SFG $\omega_1 + \omega_2 = \omega_3$
ooe	$\Delta T = \frac{0.886}{L}\left\lvert\frac{1}{\lambda_1}\frac{\partial n_{o1}}{\partial T} + \frac{1}{\lambda_2}\frac{\partial n_{o2}}{\partial T} - \frac{1}{\lambda_3}\frac{\partial n_3^e(\theta)}{\partial T}\right\rvert^{-1}$
eoe	$\Delta T = \frac{0.886}{L}\left\lvert\frac{1}{\lambda_1}\frac{\partial n_1^e(\theta)}{\partial T} + \frac{1}{\lambda_2}\frac{\partial n_{o2}}{\partial T} - \frac{1}{\lambda_3}\frac{\partial n_3^e(\theta)}{\partial T}\right\rvert^{-1}$
oee	$\Delta T = \frac{0.886}{L}\left\lvert\frac{1}{\lambda_1}\frac{\partial n_{o1}}{\partial T} + \frac{1}{\lambda_2}\frac{\partial n_2^e(\theta)}{\partial T} - \frac{1}{\lambda_3}\frac{\partial n_3^e(\theta)}{\partial T}\right\rvert^{-1}$
eeo	$\Delta T = \frac{0.886}{L}\left\lvert\frac{1}{\lambda_1}\frac{\partial n_1^e(\theta)}{\partial T} + \frac{1}{\lambda_2}\frac{\partial n_2^e(\theta)}{\partial T} - \frac{1}{\lambda_3}\frac{\partial n_{o3}}{\partial T}\right\rvert^{-1}$
eoo	$\Delta T = \frac{0.886}{L}\left\lvert\frac{1}{\lambda_1}\frac{\partial n_1^e(\theta)}{\partial T} + \frac{1}{\lambda_2}\frac{\partial n_{o2}}{\partial T} - \frac{1}{\lambda_3}\frac{\partial n_{o3}}{\partial T}\right\rvert^{-1}$
oeo	$\Delta T = \frac{0.886}{L}\left\lvert\frac{1}{\lambda_1}\frac{\partial n_{o1}}{\partial T} + \frac{1}{\lambda_2}\frac{\partial n_2^e(\theta)}{\partial T} - \frac{1}{\lambda_3}\frac{\partial n_{o3}}{\partial T}\right\rvert^{-1}$

Equations for calculating the SHG spectral bandwidth for the different types of interaction

Type of interaction	Spectral bandwidth for SHG ($\omega_1 + \omega_1 = \omega_2$)
ooe	$\Delta\nu_1 = \frac{0.443}{\lambda_1 L}\left\lvert\frac{\partial n_{o1}}{\partial\lambda_1} - \frac{\partial n_2^e(\theta)}{\partial\lambda_2}\right\rvert^{-1}$
eoe, oee	$\Delta\nu_1 = \frac{0.886}{\lambda_1 L}\left\lvert\frac{\partial n_{o1}}{\partial\lambda_1} + \frac{\partial n_1^e(\theta)}{\partial\lambda_1} - 2\frac{\partial n_2^e(\theta)}{\partial\lambda_2}\right\rvert^{-1}$
eeo	$\Delta\nu_1 = \frac{0.443}{\lambda_1 L}\left\lvert\frac{\partial n_1^e(\theta)}{\partial\lambda_1} - \frac{\partial n_{o2}}{\partial\lambda_2}\right\rvert^{-1}$
eoo, oeo	$\Delta\nu_1 = \frac{0.886}{\lambda_1 L}\left\lvert\frac{\partial n_{o1}}{\partial\lambda_1} + \frac{\partial n_1^e(\theta)}{\partial\lambda_1} - 2\frac{\partial n_{o2}}{\partial\lambda_2}\right\rvert^{-1}$

Equations for calculating the SFG spectral bandwidth when the lower-frequency interacting wave has a wide-band spectrum

Type of interaction	Spectral bandwidth for SFG ($\omega_1 + \omega_2 = \omega_3$) λ_1: wide-band spectrum; λ_2: fixed wavelength
ooe	$\Delta\nu_1 = \frac{0.886}{L}\left\lvert n_{o1} - n_3^e(\theta) - \lambda_1\frac{\partial n_{o1}}{\partial\lambda_1} + \lambda_3\frac{\partial n_3^e(\theta)}{\partial\lambda_3}\right\rvert^{-1}$
eoe	$\Delta\nu_1 = \frac{0.886}{L}\left\lvert n_1^e(\theta) - n_3^e(\theta) - \lambda_1\frac{\partial n_1^e(\theta)}{\partial\lambda_1} + \lambda_3\frac{\partial n_3^e(\theta)}{\partial\lambda_3}\right\rvert^{-1}$
oee	$\Delta\nu_1 = \frac{0.886}{L}\left\lvert n_{o1} - n_3^e(\theta) - \lambda_1\frac{\partial n_{o1}}{\partial\lambda_1} + \lambda_3\frac{\partial n_3^e(\theta)}{\partial\lambda_3}\right\rvert^{-1}$
eeo	$\Delta\nu_1 = \frac{0.886}{L}\left\lvert n_1^e(\theta) - n_{o3} - \lambda_1\frac{\partial n_1^e(\theta)}{\partial\lambda_1} + \lambda_3\frac{\partial n_{o3}}{\partial\lambda_3}\right\rvert^{-1}$
eoo	$\Delta\nu_1 = \frac{0.886}{L}\left\lvert n_1^e(\theta) - n_{o3} - \lambda_1\frac{\partial n_1^e(\theta)}{\partial\lambda_1} + \lambda_3\frac{\partial n_{o3}}{\partial\lambda_3}\right\rvert^{-1}$
oeo	$\Delta\nu_1 = \frac{0.886}{L}\left\lvert n_{o1} - n_{o3} - \lambda_1\frac{\partial n_{o1}}{\partial\lambda_1} + \lambda_3\frac{\partial n_{o3}}{\partial\lambda_3}\right\rvert^{-1}$

Equations for calculating the SFG spectral bandwidth when the higher-frequency interacting wave has a wide-band spectrum

Type of interaction	Spectral bandwidth for SFG ($\omega_1 + \omega_2 = \omega_3$) λ_1: fixed wavelength; λ_2: wide-band spectrum
ooe	$\Delta\nu_2 = \frac{0.886}{L}\left\lvert n_{o2} - n_3^e(\theta) - \lambda_2\frac{\partial n_{o2}}{\partial\lambda_2} + \lambda_3\frac{\partial n_3^e(\theta)}{\partial\lambda_3}\right\rvert^{-1}$
eoe	$\Delta\nu_2 = \frac{0.886}{L}\left\lvert n_{o2} - n_3^e(\theta) - \lambda_2\frac{\partial n_{o2}}{\partial\lambda_2} + \lambda_3\frac{\partial n_3^e(\theta)}{\partial\lambda_3}\right\rvert^{-1}$
oee	$\Delta\nu_2 = \frac{0.886}{L}\left\lvert n_2^e(\theta) - n_3^e(\theta) - \lambda_2\frac{\partial n_2^e(\theta)}{\partial\lambda_2} + \lambda_3\frac{\partial n_3^e(\theta)}{\partial\lambda_3}\right\rvert^{-1}$
eeo	$\Delta\nu_2 = \frac{0.886}{L}\left\lvert n_2^e(\theta) - n_{o3} - \lambda_2\frac{\partial n_2^e(\theta)}{\partial\lambda_2} + \lambda_3\frac{\partial n_{o3}}{\partial\lambda_3}\right\rvert^{-1}$
eoo	$\Delta\nu_2 = \frac{0.886}{L}\left\lvert n_{o2} - n_{o3} - \lambda_2\frac{\partial n_{o2}}{\partial\lambda_2} + \lambda_3\frac{\partial n_{o3}}{\partial\lambda_3}\right\rvert^{-1}$
oeo	$\Delta\nu_2 = \frac{0.886}{L}\left\lvert n_2^e(\theta) - n_{o3} - \lambda_2\frac{\partial n_2^e(\theta)}{\partial\lambda_2} + \lambda_3\frac{\partial n_{o3}}{\partial\lambda_3}\right\rvert^{-1}$

OPA bandwidths: they refer to the amplification and not to the conversion efficiency.

1. In the „fixed-field"-approximation we obtain a spectral bandwidth of:

$$\Delta\nu \approx 4\sqrt{\frac{\ln 2\ g}{L}}\left(\frac{\partial(\Delta k)}{\partial\nu}\right)^{-1}_{\nu_{PM}}$$

Monochromatic pump is assumed here and the result concerns the signal and idler bandwidths determined by the dispersive properties of the crystal i.e. $\chi^{(1)}$.

Other contributions to the OPA/OPG bandwidth:

2. the pump bandwidth.

$$G_0(\kappa I_3) = 0.5 G_0(I_3)$$

$\Delta\nu_3$ is determined by the values $\nu_3-\Delta\nu_3/2$ and $\nu_3+\Delta\nu_3/2$ corresponding to κI_3 in the frequency domain.

3. Pump divergence: for a cone angle Ω of the pump beam one obtains:

$$\Delta\nu_\Omega \approx \left(\frac{d\nu}{d\theta}\right)\frac{2\Omega}{n_3} \approx \frac{2\Omega\rho}{\lambda_3}\left(\frac{\partial(\Delta k)}{\partial\nu}\right)^{-1}_{\nu_{PM}}$$

4. The off-axis interaction, vector phase-matching (unwanted).

Multi-stage OPA schemes

Three-wave interactions with broadband (ps or fs) pulses:

Pulse duration T (FWHM) of $I(z,t)$ and the spectral bandwidth $\Delta\nu_L$(FWHM) of $|A(z,\nu)|^2$ are related through:

$\Delta\nu_L T \geq C$ (with C=0.441 for Gaussian and 0.316 for $sech^2$- pulses).

$\Delta\nu_L T = C$: Fourier-limited (bandwidth-limited) pulses.

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Advantages when operating with short pulses:

-High peak at low average intensities.

-Bulk damage threshold is higher.

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Besides saturation (deviation from the fixed-field approximation) we have to consider:

Dispersion, spectral broadening, higher order nonlinearities.

Since $|\vec{P}_{NL}| << |\vec{P}_L|$ holds, normally linear dispersion is condidered only.

$$k(\omega) = k_n + \left(\frac{dk}{d\omega}\right)_{\omega_n} (\omega - \omega_n) + \frac{1}{2}\left(\frac{d^2k}{d\omega^2}\right)_{\omega_n} (\omega - \omega_n)^2 + \cdots$$

Group-velocity v_n:

$$v_n = \left(\frac{\partial\omega}{\partial k}\right)_{\omega_n} = c\left(\frac{\partial\omega}{\partial(n\omega)}\right)_{\omega_n} = c\left[n_n + \omega_n\left(\frac{\partial n}{\partial\omega}\right)_{\omega_n}\right]^{-1}$$

Group-velocity dispersion (GVD):

$$k_n'' \equiv \left(\frac{\partial^2 k}{\partial\omega^2}\right)_{\omega_n} = \frac{2}{c}\left(\frac{\partial n}{\partial\omega}\right)_{\omega_n} + \frac{\omega_n}{c}\left(\frac{\partial^2 n}{\partial\omega^2}\right)_{\omega_n} = \frac{\lambda_n^3}{2\pi c^2}\left(\frac{\partial^2 n}{\partial\lambda^2}\right)_{\lambda_n}$$

Interaction length (between two pulses!) L_{GVM}:

$$L_{GVM} = \frac{T}{|1/v_n - 1/v_m|} = \frac{T}{|\Delta_{nm}|}$$

$\Delta_{nm}=1/v_n-1/v_m$: group-velicty mismatch (GVM).

Only in type I-SHG or degenerate OPG and OPA we have one GVM-parameter

A method to influence the GVM: noncollinear (vector) phase-matching.

Group velocities in the direction of the Poynting vector of the ω_3 wave (ooe interaction):

$$v_3=(d\omega_3/dk_3)\cos\rho\ ,$$

$$v_2=(d\omega_2/dk_2)\cos(\beta+\rho)\ ,$$

$$v_1=(d\omega_1/dk_1)\cos(\alpha-\rho)$$

The dispersion of $\chi^{(1)}$ is accounted for in the frequency domain were we have products insted of convolution integrals. Taylor expansion for $\chi^{(1)}(\omega)$:

$$\chi^{(1)}(\omega) = \chi_n^{(1)} + \left[\frac{d\chi^{(1)}}{d\omega}\right]_{\omega=\omega_n}(\omega-\omega_n) + \frac{1}{2}\left[\frac{d^2\chi^{(1)}}{d\omega^2}\right]_{\omega=\omega_n}(\omega-\omega_n)^2 + \ldots..$$

The multiplication by $(\omega-\omega_n)$ corresponds in the time domain to d/dt, d^2/dt^2 etc. for the slowly varying amplitudes. In the moving frame $\xi=z$ and $\eta=t-z/v_n$ and with the SVA approximation, we simply have to substitude $\frac{n_n}{c}\frac{\partial}{\partial t}$ by $\frac{1}{v_n}\frac{\partial}{\partial \eta} + \frac{i}{2}k_n''\frac{\partial^2}{\partial \eta^2}$ in the coupled equations. Choosing the moving frame to be related to the ω_3 wave (pump pulse in OPA or OPG) we obtain:

$$\left(\frac{\partial}{\partial\xi} + \Delta_{13}\frac{\partial}{\partial\eta} + i\frac{1}{2}k_1''\frac{\partial^2}{\partial\eta^2}\right)A_1(\xi,\eta) = i\frac{2\omega_1 d_{eff}}{n_1 c}A_3(\xi,\eta)A_2^*(\xi,\eta)e^{-i\Delta k\xi}$$

$$\left(\frac{\partial}{\partial\xi} + \Delta_{23}\frac{\partial}{\partial\eta} + i\frac{1}{2}k_2''\frac{\partial^2}{\partial\eta^2}\right)A_2(\xi,\eta) = i\frac{2\omega_2 d_{eff}}{n_2 c}A_3(\xi,\eta)A_1^*(\xi,\eta)e^{-i\Delta k\xi}$$

$$\left(\frac{\partial}{\partial\xi} + i\frac{1}{2}k_3''\frac{\partial^2}{\partial\eta^2}\right)A_3(\xi,\eta) = i\frac{2\omega_3 d_{eff}}{n_3 c}A_1(\xi,\eta)A_2(\xi,\eta)e^{i\Delta k\xi}$$

Gaussian pulse $A(\xi=0,\eta) = A_0\exp(-2\ln 2\eta^2/T_0^2)$,

with T_0 as FWHM of the intensity $I(\xi=0,\eta) \propto |A(\xi=0,\eta)|^2$.

Neglecting the nonlinearity we see that the pulse duration is doubled after $(\sqrt{3}/4\ln 2)L_{GVD} \approx 0.6L_{GVD}$, where $L_{GVD} = T_0^2/k_n''$

In general $L<<L_{GVD}$ holds for Fourier-limited pulses i.e. GVD is negligible.

Bandwidth control: In the „fixed-field"-approximation we can express the spectral acceptance for SFG and DFG as:

$$\Delta\nu = \frac{0.886}{L|\Delta_{nm}|}$$

and the OPA / OPG bandwidths as,

$$\Delta\nu = \frac{0.53(g/L)^{1/2}}{|\Delta_{12}|} \quad \text{and} \quad \Delta\nu_{\Omega} = 2\Omega\rho / \lambda_3|\Delta_{12}|$$

The spectral width of the signal and idler in a OPG is influenced also by the pump bandwidth, noncollinear interaction, beam divergence.

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Whereas in SFG and DFG the spectral acceptance is a limitation in an OPG this can be a (unwanted) spectral broadening mechanism.

Seeding of an OPA can control here the bandwidths of signal and idler.

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In an OPA also „temporal gain narrowing" is possible.

At degeneracy we have for $L_{NL} \ll L$ and $L_{GVM} > L$ a shortening factor of

$$T/T_0 \approx 1.4(gL)^{-1/2} \qquad \text{for Gaussian pulses}$$

limited by the GVM to a minimum duration of $T \approx \sqrt{T_0 L_{NL}|\Delta_{13}|}$.

The shortening mechanism is similar to the well known $\sqrt{2}$ *-shortening in SHG*

In general it is better to use shorter crystals ($L<L_{GVM}$) and higher intensity.

However, (mostly unwanted)

higher order nonlinearities as $\chi^{(3)}$-processes come into play:

Two-photon absorption,

self- and cross-phase modulation,

induced focussing, defocussing etc.

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Example:

coupled equations for SHG without GVM and GVD and for $\Delta k=0$:

$$\frac{\partial A_1}{\partial \xi} = i\frac{2\omega_1 d_{eff}}{n_1 c} A_1^* A_3 + i\frac{3\omega_1 \chi^{(3)}}{2n_1 c} A_1\left(|A_1|^2 + 2|A_3|^2\right)$$

$$\frac{\partial A_3}{\partial \xi} = i\frac{\omega_3 d_{eff}}{n_3 c} A_1^2 + i\frac{3\omega_3 \chi^{(3)}}{2n_3 c} A_3\left(|A_3|^2 + 2|A_1|^2\right)$$

$$\chi^{(3)} = \chi^{(3)}_{Re} + i\chi^{(3)}_{Im}.$$

(Re describes influence on the phase and Im - two-photon absorption),

For plane waves: $\chi^{(3)}_{Re}$: self-phase modulation (single pulse)

or cross-phase modulation (induced phase-modulation).

For propagation in a Kerr medium with Kerr coefficient n_I:

$$n(\eta) = n_0 + n_I I(\eta)$$

holds with $n_I = 3\chi^{(3)}_{Re}(-\omega;\omega,\omega,-\omega)/4\varepsilon_0 c n_0^2$.

n_0 is the linear refractive index (up to now n or n_n).

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n_I results in a phase shift $\Delta\varphi(\eta)$

$$\Delta\varphi(\eta) = \frac{\omega_0}{c} n_I I(\eta) L$$

Then $\omega(\eta)=\omega_0+\Delta\omega(\eta)$ with $\Delta\omega(\eta)=-(d/d\eta)\Delta\varphi(\eta)$:

$$\Delta\omega(\eta) = -\frac{\omega_0 n_I L}{c}\frac{dI(\eta)}{d\eta}$$

Leading pulse edge [$dI(\eta)/d\eta>0$]: red shift for $n_I > 0$,

Trailing pulse edge [$dI(\eta)/d\eta<0$]: blue shifted.

$\Rightarrow$

Chirped (not Fourier-limited) pulses. **Consequences:**

Stronger influence of GVD and the spectral acceptance =

lower conversion efficiency.

In the extreme case: continuum and white light generation

Two-photon absorption

Single pulse (e.g. the pump pulse for an OPA) with intensity I:

$$\frac{dI}{d\xi} = \alpha I - \beta_{TP} I^2$$

where $\beta_{TP} = 3\pi\chi^{(3)}_{Im}(-\omega;\omega,\omega,-\omega)/\varepsilon_0 c n_0^2 \lambda$.

Spatial effects (plane wave approximation violated):

$\chi^{(3)}_{Re}$ and n_I lead to modification of the transversal beam profile:

Self-focussing, defocussing, channelling.

Critical power for self-trapping (n_I>0), Gaussian cw beam:

$$P_{cr} = \frac{\lambda^2}{8\pi n_0 n_I}$$

If the above power is exceeded self-focussing takes place at:

$$L_{SF} = \frac{\pi w_0^2 / \lambda}{(P/P_{cr} - 1)^{1/2}}$$

Criterion to avoid all n_I -effects (the B-integral):

$$B = \frac{2\pi}{\lambda} n_I \int_0^L I_0(\xi,\eta) d\xi \ ,$$

B≤3–5 should hold for the axial peak intensity $I_0(\eta = 0) \equiv I_m$.

What we expect from second order nonlinear materials:

- phase-matching,
- high effective nonlinearity,
- high damage threshold, non-hygroscopic,
- high and broad transmission,
- small spatial walk-off due to the birefringence,
- no two-photon absorption and small Kerr constant
- large angular acceptance and possibly temperature tunable
- advantageous (low) GVM.

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Real beams are Gaussian: effective focal length $L_F=\pi z_R$

Real crystals have absorption losses.

These two and the birefringence length L_ρ can be treated theoretically and formulae exist in the fixed-field approximation.

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Noncritical phase-matching:	always desirable since the angular acceptance is maximized and birefringence walk-off is absent.
Temperature tuning:	always convenient: no beam walking when tuning.