

1.7. NONLINEAR OPTICAL PROPERTIES

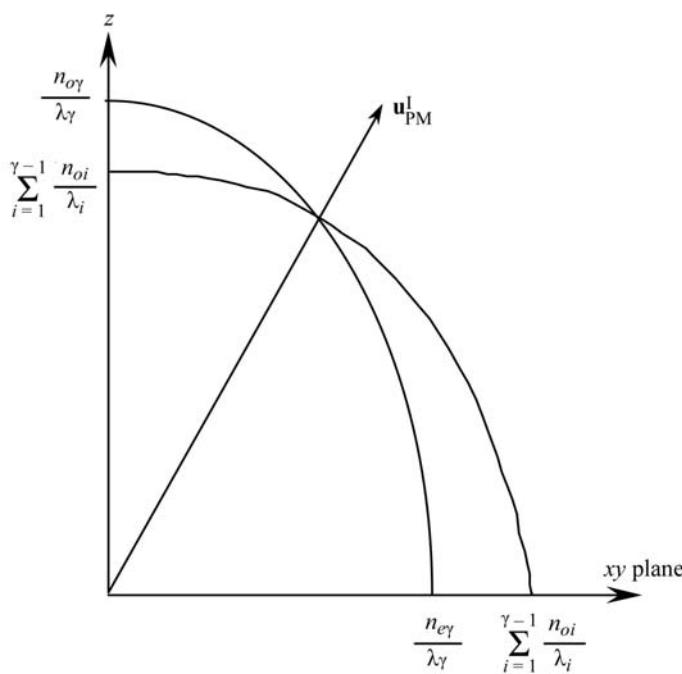


Fig. 1.7.3.4. Index surface sections in a plane containing the optic axis z of a negative uniaxial crystal allowing collinear type-I phase matching for SFG ($\omega_3 = \omega_1 + \omega_2$), $\gamma = 3$, or for SFG ($\omega_4 = \omega_1 + \omega_2 + \omega_3$), $\gamma = 4$. \mathbf{u}_{PM}^I is the corresponding phase-matching direction.

three-wave interaction is allowed for three configurations of polarization given in Table 1.7.3.1.

The designation of the type of phase matching, I, II or III, is defined according to the polarization states at the frequencies which are added or subtracted. Type I characterizes interactions for which these two waves are identically polarized; the two corresponding polarizations are different for types II and III. Note that each phase-matching relation corresponds to one sum-frequency generation SFG ($\omega_3 = \omega_1 + \omega_2$) and two difference-frequency generation processes, DFG ($\omega_1 = \omega_3 - \omega_2$) and DFG ($\omega_2 = \omega_3 - \omega_1$). Types II and III are equivalent for SHG because $\omega_1 = \omega_2$.

For a four-wave process, only seven combinations of refractive indices allow phase matching in the case of normal dispersion; they are given in Table 1.7.3.2 with the corresponding configurations of polarization and types of SFG and DFG.

The convention of designation of the types is the same as for three-wave interactions for the situations where one polarization state is different from the three others, leading to the types I, II, III and IV. The criterion corresponding to type I cannot be applied to the three other phase-matching relations where two waves have the same polarization state, different from the two others. In this case, it is convenient to refer to each phase-matching relation by the same roman numeral, but with a different index: V^i , VI^i and VII^i , with the index $i = 1, 2, 3, 4$ corresponding to the index of the frequency generated by the SFG or DFG. For THG ($\omega_1 = \omega_2 = \omega_3$), types II, III and IV are equivalent, and so are types V^4 , VI^4 and VII^4 .

Table 1.7.3.2. Correspondence between the phase-matching relations, the configurations of polarization and the types according to SFG ($\omega_4 = \omega_1 + \omega_2 + \omega_3$), DFG ($\omega_1 = \omega_4 - \omega_2 - \omega_3$), DFG ($\omega_2 = \omega_4 - \omega_1 - \omega_3$) and DFG ($\omega_3 = \omega_4 - \omega_1 - \omega_2$) (Boulanger et al., 1993)

Phase-matching relations	Configurations of polarization				Types of interaction			
	ω_4	ω_1	ω_2	ω_3	SFG (ω_4)	DFG (ω_1)	DFG (ω_2)	DFG (ω_3)
$\omega_4 n_4^- = \omega_1 n_1^+ + \omega_2 n_2^+ + \omega_3 n_3^+$	e^-	e^+	e^+	e^+	I	II	III	IV
$\omega_4 n_4^- = \omega_1 n_1^- + \omega_2 n_2^+ + \omega_3 n_3^+$	e^-	e^-	e^-	e^+	II	III	IV	I
$\omega_4 n_4^- = \omega_1 n_1^- + \omega_2 n_2^+ + \omega_3 n_3^-$	e^-	e^-	e^+	e^-	III	IV	I	II
$\omega_4 n_4^- = \omega_1 n_1^+ + \omega_2 n_2^- + \omega_3 n_3^-$	e^-	e^+	e^-	e^-	IV	I	II	IV
$\omega_4 n_4^- = \omega_1 n_1^- + \omega_2 n_2^+ + \omega_3 n_3^+$	e^-	e^-	e^+	e^+	V^4	V^1	V^2	V^3
$\omega_4 n_4^- = \omega_1 n_1^+ + \omega_2 n_2^- + \omega_3 n_3^+$	e^-	e^+	e^-	e^+	VI^4	VI^1	VI^2	VI^3
$\omega_4 n_4^- = \omega_1 n_1^+ + \omega_2 n_2^+ + \omega_3 n_3^-$	e^-	e^+	e^+	e^-	VII^4	VII^1	VII^2	VII^3

The index surface allows the geometrical determination of the phase-matching directions, which depend on the relative ellipticity of the internal (–) and external (+) sheets divided by the corresponding wavelengths: according to Tables 1.7.3.1 and 1.7.3.2 the directions are given by the intersection of the internal sheet of the lowest wavelength [$n^-(\lambda_\gamma, \theta, \varphi)]/(\lambda_\gamma)$ with a linear combination of the internal and external sheets at the other frequencies $\sum_{i=1}^{\gamma-1} [n^\pm(\lambda_i, \theta, \varphi)]/(\lambda_i)$. The existence and loci of these intersections depend on specific inequalities between the principal refractive indices at the different wavelengths. Note that independently of phase-matching considerations, normal dispersion and energy conservation impose $\sum_{i=1}^{\gamma-1} [n_a(\lambda_i)]/(\lambda_i) < [n_a(\lambda_\gamma)]/(\lambda_\gamma)$ with $a = x, y, z$.

1.7.3.2.2.1. Cubic crystals

There is no possibility of collinear phase matching in a dispersive cubic crystal because of the absence of birefringence. In a hypothetical non-dispersive anaxial crystal, the 2^3 three-wave and 2^4 four-wave phase-matching configurations would be allowed in any direction of propagation.

1.7.3.2.2.2. Uniaxial crystals

The configurations of polarization in terms of ordinary and extraordinary waves depend on the optic sign of the phase-matching direction with the convention given in Section 1.7.3.1: Tables 1.7.3.1 and 1.7.3.2 must be read by substituting (+, –) by (e, o) for a positive crystal and by (o, e) for a negative one.

Because of the symmetry of the index surface, all the phase-matching directions for a given type describe a cone with the optic axis as a revolution axis. Note that the previous comment on the anaxial class is valid for a propagation along the optic axis ($n_o = n_e$).

Fig. 1.7.3.4 shows the example of negative uniaxial crystals ($n_o > n_e$) like $\beta\text{-BaB}_2\text{O}_4$ (BBO) and KH_2PO_4 (KDP).

From Fig. 1.7.3.4, it clearly appears that the intersection of the sheets is possible only if $(n_{e_\gamma})/(\lambda_\gamma) < \sum_{i=1}^{\gamma-1} (n_{o_i})/(\lambda_i)$ [$< (n_{o_\gamma})/(\lambda_\gamma)$] with $\gamma = 3$ for a three-wave process and $\gamma = 4$ for a four-wave one. The same considerations can be made for the positive sign and for all the other types of phase matching. There are different situations of inequalities allowing zero, one or several types: Table 1.7.3.3 gives the five possible situations for the three-wave interactions and Table 1.7.3.4 the 19 situations for the four-wave processes.

1.7.3.2.2.3. Biaxial crystals

The situation of biaxial crystals is more complicated, because the two sheets that must intersect are both elliptical in several cases. For a given interaction, all the phase-matching directions generate a complicated cone which joins two directions in the principal planes; the possible loci a, b, c, d are shown on the stereographic projection given in Fig. 1.7.3.5.

The basic inequalities of normal dispersion (1.7.3.7) forbid collinear phase matching for all the directions of propagation located between two optic axes at the two frequencies concerned.