Field-factor formalism for the study of the tensorial symmetry of four-wave nonlinear optical parametric interactions in uniaxial and biaxial crystals

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We establish a complete treatment of the tensorial symmetry properties of the four-wave sum and difference frequency mixing, phase matched in uniaxial and biaxial crystals. This study is based on the formalism of the "field factor" which we have previously introduced [B. Boulanger and G. Marnier, Opt. Commun. **79**, 102 (1990)]. The 14 configurations of polarization allowing phase matching are considered and the corresponding effective coefficient is calculated for the 19 uniaxial and 8 biaxial classes. The effective coefficient is nil in a few cases. The inequalities between refractive indices, which determine the collinear phase-matching directions, are given according to the optical sign. We calculate the field factors for three real nonlinear crystals: BaB_2O_4, $KTiOPO_4$, and thiosemicarbazide cadmium chloride monohydrate.

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I. INTRODUCTION

Maker [1] considered the irreducible tensorial decomposition of the product of the coupled electric fields in order to study the time dependence of the orientation-dependent molecular-pair distribution function of liquids by "quasielastic" second-harmonic light scattering. Later, we took an interest in the study of the tensorial product of the coupled electric fields, which we called the "field factor," for the determination of the independent elements of the second- and third-order electric susceptibility tensors $\chi^{(2)}$ and $\chi^{(3)}$ of crystals from phase-matched second- and third-harmonic generation experiments [2,3].

We developed the formalism of the field factor for the complete study of the three-wave nonlinear optical interactions phase matched in uniaxial and biaxial crystals, which allows a unified description of the different types of interactions [4]. Zyss used this formalism and demonstrated that the most efficient quadratic nonlinear mixing in an octupolar medium (D_{3h}) is obtained with circularly polarized waves [5].

This paper deals with the complete study of the collinear phase-matched four-wave nonlinear optical mixing in uniaxial and biaxial crystals. We show how the field-factor formalism allows one to precisely obtain the real tensorial contribution of the linear optical properties to the symmetry of the third-order nonlinear optical properties. In fact, the refractive indices and their dispersion in frequency determine the existence and the loci of the phase-matching directions which impose the directions of the electric-field vectors of the interacting waves. Then, we describe a four-wave parametric interaction in a crystal by two four-rank tensors: the third-order electric susceptibility tensor $\chi^{(3)}$ and the field tensor $\mathbf{F}^{(3)}$, which is equal to the tensorial product of the electric-field vectors. Each element F_{ijkl}, called the field factor, is a trigonometric function of the direction of propagation. The beam interacting with a third-order nonlinear crystal is then described by its pulsations $\omega_1,\omega_2,\omega_3,\omega_4$ ($\omega_4=\omega_1+\omega_2+\omega_3$) and its field tensor. The effective coefficient χ_{eff}, which depends on the efficiency of the interaction, is equal to the tensorial contraction of $\mathbf{F}^{(3)}$ and $\chi^{(3)}$. The symmetry of $\chi^{(3)}$ is imposed by the orientation symmetry of the crystal and the symmetry of $\mathbf{F}^{(3)}$ is governed by the vectorial properties of the electric fields characteristic of the optical class, uniaxial or biaxial. In this paper, we systematically study the symmetry of $\mathbf{F}^{(3)}$ according to the 14 configurations of polarization which allow phase matching. We also contract $\mathbf{F}^{(3)}$ and $\chi^{(3)}$ for the 19 uniaxial and the 8 biaxial crystal classes for each phase-matched configuration of polarization. The effective coefficient is nil in a few cases and we find the same forbidden crystal classes as those determined by Midwinter and Warner for the particular case of the third-harmonic generation assuming Kleinman's conjecture and without consideration of the field factor [6].

We take the example of three real nonlinear crystals, BaB_2O_4 (BBO), $KTiOPO_4$ (KTP), and thiosemicarbazide cadmium chloride monohydrate (TSCCC), for the calculation of the field factors. We show with BBO how the study of the field-factor functions simply allows the judicious choice of the configurations of polarization and phase-matching directions for the determination of the independent coefficients of $\chi^{(3)}$ by third-harmonic-generation (THG) efficiency measurements.

II. DEFINITIONS

A. Four-rank electric susceptibility and field tensors

The efficiency of a nonlinear optical parametric interaction depends on the effective coefficient $\chi_{\text{eff}}(\theta,\phi)$, defined by the tensorial contraction of the nonlinear polarization vector $\mathbf{P}^{\mathrm{NL}}(\omega,\theta,\phi)$ with the unit electric-field vector $\mathbf{e}(\omega,\theta,\phi)$. ω is the circular frequency of the considered wave in the direction of propagation, with the spherical coordinates (θ,ϕ) [7]:

$$\chi_{\mathrm{eff}}(\theta,\phi)=\mathbf{e}(\omega,\theta,\phi)\cdot\mathbf{P}^{\mathrm{NL}}(\omega,\theta,\phi)\ . \tag{1}$$

θ and ϕ refer to the optical frame (x,y,z).

$\mathbf{P}^{\mathrm{NL}}(\omega,\theta,\phi)$ is also linked to the nonlinear electric susceptibility tensor by a tensorial contraction. For the four-wave sum-frequency mixing (SFM) and difference-frequency mixing (DFM) with the circular frequencies ω_a, ω_b, ω_c, and ω_d, we have

$$\mathbf{P}^{\mathrm{NL}}(\omega_a,\theta,\phi)=\chi^{(3)}(\omega_a)\vdots\mathbf{e}(\omega_b,\theta,\phi)\mathbf{e}(\omega_c,\theta,\phi)\mathbf{e}(\omega_d,\theta,\phi)\ . \tag{2}$$

$(\omega_a,\omega_b,\omega_c,\omega_d)$ correspond to $(\omega_4,\omega_1,\omega_2,\omega_3)$ for the SFM $(\omega_4=\omega_1+\omega_2+\omega_3)$, to $(\omega_1,\omega_4,\omega_2,\omega_3)$ for the DFM $(\omega_1=\omega_4-\omega_2-\omega_3)$, to $(\omega_2,\omega_4,\omega_1,\omega_3)$ for the DFM $(\omega_2=\omega_4-\omega_1-\omega_3)$, and to $(\omega_3,\omega_4,\omega_1,\omega_2)$ for the DFM $(\omega_3=\omega_4-\omega_1-\omega_2)$.

Thus the effective coefficient is the tensorial concentration of two four-rank tensors:

$$\chi_{\mathrm{eff}}(\omega_a,\omega_b,\omega_c,\omega_d,\theta,\phi)=\chi^{(3)}\cdot\mathbf{F}^{(3)}(\theta,\phi)$$
$$=\sum_{i,j,k,l}\chi_{ijkl}(\omega_a)F_{ijkl}(\omega_a,\omega_b,\omega_c,\omega_d,\theta,\phi)\ . \tag{3}$$

The indices i, j, k, and l refer to the optical frame.

$\mathbf{F}^{(3)}(\omega_a,\omega_b,\omega_c,\omega_d,\theta,\phi)$ is the field tensor given by the tensorial product of the unit electric-field vectors of the four interacting waves:

$$\mathbf{F}^{(3)}(\omega_a,\omega_b,\omega_c,\omega_d,\theta,\phi)$$
$$=\mathbf{e}(\omega_a,\theta,\phi)\mathbf{e}(\omega_b,\theta,\phi)\mathbf{e}(\omega_c,\theta,\phi)\mathbf{e}(\omega_d,\theta,\phi)\ . \tag{4}$$

The correspondence between $\omega_a,\omega_b,\omega_c,\omega_d$ and $\omega_1,\omega_2,\omega_3,\omega_4$, according to SFM and DFM, is the same as that for relation (2). Each element F_{ijkl} is called a field factor and is a trigonometric function of the spherical coordinates (θ,ϕ) and only depends on refractive indices.

Thus $\mathbf{F}^{(3)}(\theta,\phi)$ is a tensor characteristic of the configuration of polarization of the beams interacting with the nonlinear crystal. Note that $\mathbf{F}(\theta,\phi)$ must not be confused with $F(\theta,\phi,\mathbf{d})$ which is the designation given by Midwinter and Warner [6] for the effective coefficient, $\mathbf{d}$ being the nonlinear polarization tensor.

From (4), it is obvious that the field factor remains unchanged by concomitant permutations of the electric-field vectors and the corresponding Cartesian indices. Thus there exist particular relationships between field factors of SFM and DFM, i.e., for all directions of propagation:

$$F_{ijkl}^{e_4e_1e_2e_3}(\omega_4=\omega_1+\omega_2+\omega_3)$$
$$=F_{jikl}^{e_1e_4e_2e_3}(\omega_1=\omega_4-\omega_2-\omega_3)$$
$$=F_{kijl}^{e_2e_4e_1e_3}(\omega_2=\omega_4-\omega_1-\omega_3)$$
$$=F_{lijk}^{e_3e_4e_1e_2}(\omega_3=\omega_4-\omega_1-\omega_2)\ . \tag{5}$$

e_i is the electric-field vector of the wave at ω_i $(i=1,2,3,4)$. The symmetry of tensor $\mathbf{F}^{(3)}(\theta,\phi)$ is governed by the vectorial properties of the interacting electric fields which impose restrictions and relations between F_{ijkl} elements and so reduce the number of independent elements. This will be studied in Secs. III and IV with the symmetry introduced by equalities between frequencies according to the configuration of polarization.

B. Refractive indices in a direction of propagation

We consider four collinear wave vectors $\mathbf{k}(\omega_i,\theta,\phi)$:

$$\mathbf{k}(\omega_i,\theta,\phi)=[\omega_i/c]n(\omega_i,\theta,\phi)\mathbf{u}(\theta,\phi)\quad(i=1,2,3,4) \tag{6}$$

with $\omega_i=2\pi c/\lambda_i$, where λ_i is the wavelength of the ith wave. $\mathbf{u}(\theta,\phi)$ is the unit vector of the direction of propagation with the Cartesian coordinates (u_x,u_y,u_z) given by

$$u_x=\cos\phi\sin\theta\ ,\quad u_y=\sin\phi\sin\theta\ ,\quad u_z=\cos\theta\ . \tag{7}$$

x,y,z refer to the orthonormal optical frame which corresponds to the principal axes of the index ellipsoid.

$n(\omega_i,\theta,\phi)$ is the refractive index, at the circular frequency ω_i, given by the Fresnel equation which admits two solutions [7]:

$$n^+(\omega_i)=\left[\frac{2}{-B_i-(B_i^2-4C_i)^{1/2}}\right]^{1/2} \tag{8}$$

and

$$n^-(\omega_i)=\left[\frac{2}{-B_i+(B_i^2-4C_i)^{1/2}}\right]^{1/2} \tag{9}$$

$[n^+(\omega_i)>n^-(\omega_i)]$ with

$$B_i=-u_x^2(b_i+c_i)-u_y^2(a_i+c_i)-u_z^2(a_i+b_i)\ , \tag{10}$$

$$C_i=u_x^2b_ic_i+u_y^2a_ic_i+u_z^2a_ib_i\ , \tag{11}$$

with

$$a_i=n_x^{-2}(\omega_i)\ ,\quad b_i=n_y^{-2}(\omega_i)\ ,\quad c_i=n_z^{-2}(\omega_i)\ . \tag{12}$$

$n_x(\omega_i)$, $n_y(\omega_i)$, and $n_z(\omega_i)$ are the principal refractive indices of the index ellipsoid at the circular frequency ω_i.

The biaxial class corresponds to the case where n_x, n_y, and n_z are different. The equality between two principal refractive indices defines the uniaxial class. The anaxial class, which corresponds to the equality of all refractive indices at a given circular frequency, is not studied in the present work. The calculation of the electric-field vectors $\mathbf{e}^+$ and $\mathbf{e}^-$, the two eigenmodes associated with n^+ and n^-, will be specified in Sec. III for the uniaxial class and in Sec. IV for the biaxial class.

C. Conservation of momentum and configuration of polarization

The conservation of momentum of the nonlinear interaction in the direction $\mathbf{u}(\theta,\phi)$ is satisfied when the wave vectors of the four interacting waves verify the relation

$$\mathbf{k}(\omega_1,\theta,\phi)+\mathbf{k}(\omega_2,\theta,\phi)+\mathbf{k}(\omega_3,\theta,\phi)=\mathbf{k}(\omega_4,\theta,\phi) . \quad (13)$$

Such a direction is called a phase-matching direction. According to (6), Eq. (13) becomes

$$\omega_1 n(\omega_1,\theta,\phi)+\omega_2 n(\omega_2,\theta,\phi)+\omega_3 n(\omega_3,\theta,\phi) = \omega_4 n(\omega_4,\theta,\phi) . \quad (14)$$

The birefringence $[n^-(\omega_i)\neq n^+(\omega_i),\ i=1,2,3,4]$ and the dispersion in frequency of the refractive indices $[n^{+,-}(\omega_1)<n^{+,-}(\omega_2)<n^{+,-}(\omega_3)<n^{+,-}(\omega_4)$ when $\omega_1<\omega_2<\omega_3<\omega_4]$ condition the possibility and loci of collinear phase-matching directions and thus the electric-field vectors of the interacting waves.

There are two possible values, n^+ and n^-, given by (8) and (9), for each of the four refractive indices, that is, 2^4 possible combinations. Among these combinations, only seven are compatible with the dispersion in frequency and with the conservations of energy and momentum. Thus the phase matching of four-wave interaction is allowed for seven configurations of polarization, given in Table I.

The designation of the seven corresponding phase-matching relations according to the four SFM and DFM interactions is of the same kind as for three-wave interactions [4], for the configurations $(+++-)$, $(--+-)$, $(-+--)$, and $(+---)$: type I corresponds to the case where the three waves whose frequencies are added or subtracted have the same polarization state. The designation of types II, III, and IV is then arbitrary.

The criteria corresponding to type I cannot be applied to the three other configurations $(-++-)$, $(+-+-)$, and $(++--)$. We choose to designate each of the corresponding phase-matching relations by the same number V^i, VI^i, and VII^i, respectively, with the superscript $(i=1,2,3,4)$ corresponding to the number of the frequency generated by the sum or difference. Table I gives the correspondence between phase-matching relations, configurations of polarization, and types according to SFM and DFM.

III. UNIAXIAL CLASSES

The uniaxial class is characterized by the equality of two principal indices, called ordinary indices $(n_x=n_y=n_o)$; the other index is called the extraordinary index $(n_z=n_e)$. The indices' surface, whose external and internal sheets are given by Eqs. (8) and (9), respectively, has one ombilic along the z axis, called the optical axis. The ordinary sheet is spherical and the extraordinary one is ellipsoidal with z as the revolution axis [8]. The sign of the class is defined by the sign of the birefringence n_e-n_o. Thus, according to these definitions, (n_e,n_o) corresponds to (n^+,n^-) for a positive class and to (n^-,n^+) for a negative class.

The phase-matching directions of the seven phase-matching relations are given by the intersection of the internal sheet at ω_4 and a combination of the internal and external sheets at ω_1, ω_2, and ω_3 according to Table I. For the positive uniaxial class, the principal refractive indices must verify

$$\omega_4 n^o(\omega_4)<\omega_1 n^a(\omega_1)+\omega_2 n^b(\omega_2)+\omega_3 n^c(\omega_3) . \quad (15)$$

For the SFM(ω_4), (n^a,n^b,n^c) correspond to

(n^e,n^e,n^e) for type I ,

(n^o,n^o,n^e) for type II ,

(n^o,n^e,n^o) for type III ,

(n^e,n^o,n^o) for type IV ,

(n^o,n^e,n^e) for type V^4 ,

(n^e,n^o,n^e) for type VI^4 ,

(n^e,n^e,n^o) for type VII^4 .

The correspondence between DFM and SFM is given in Table I. The inequalities for the negative uniaxial class are

$$\omega_4 n^e(\omega_4)<\omega_1 n^a(\omega_1)+\omega_2 n^b(\omega_2)+\omega_3 n^c(\omega_3) . \quad (16)$$

For the SFM(ω_4), (n^a,n^b,n^c) correspond to

(n^o,n^o,n^o) for type I ,

(n^e,n^e,n^o) for type II ,

(n^e,n^o,n^e) for type III ,

(n^o,n^e,n^e) for type IV ,

TABLE I. Definition of the types of interactions according to the phase-matching relations and the configurations of polarization. $\mathbf{e}^{+,-}$ are the electric-field vectors associated with the refractive indices $n^{+,-}$. $(\omega_1,\omega_2,\omega_3,\omega_4)$ are the pulsations of the four interacting waves.

Phase-matching relations	Configurations of polarization				Types of interaction			
	ω_4	ω_1	ω_2	ω_3	SFM(ω_4)	DFM(ω_1)	DFM(ω_2)	DFM(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	III
$\omega_4 n_4^- =\omega_1 n_1^- +\omega_2 n_2^+ +\omega_3 n_3^+$	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

(n^e,n^o,n^o) for type V^4 ,

(n^o,n^e,n^o) for type VI^4 ,

(n^o,n^o,n^e) for type VII^4 .

It is obvious that any phase matching is possible along the optical axis ($n^o=n^e$) of the nondispersive crystal.

Table II gives the configuration of ordinary and extraordinary polarizations for the negative and positive uniaxial classes corresponding to the seven phase-matching relations. The 14 possible configurations of polarization can be divided into three groups on the basis of the number of ordinary (o) and extraordinary (e) waves: the four interactions of three ordinary and one extraordinary waves, which we call 3*oe*, and the four other, coupling three extraordinary and one ordinary waves, 3*eo*; these two groups correspond to types I, II, III, and IV. The six configurations of two ordinary and two extraordinary waves, 2*o*2*e*, are related to types V^a, VI^a, VII^a ($a=1,2,3,4$). The components, in the optical frame (x,y,z), of the ordinary and extraordinary unit electric-field vectors $\mathbf{e}^o$ and $\mathbf{e}^e$ at the circular frequency ω are

$$e_x^o=-\sin\phi\ ,\quad e_y^o=+\cos\phi\ ,\quad e_z^o=0\ , \tag{17}$$

$$\begin{aligned} e_x^e&=-\cos[\theta\pm\rho(\theta,\omega)]\cos\phi\ ,\\ e_y^e&=-\cos[\theta\pm\rho(\theta,\omega)]\sin\phi\ ,\\ e_z^e&=\sin[\theta\pm\rho(\theta,\omega)]\ , \end{aligned} \tag{18}$$

with $-$ for the positive class and $+$ for the negative class.

$\rho(\theta,\omega)$ is the walkoff angle, given by

$$\rho(\theta,\omega)=\arccos\left[\left[\frac{\cos^2\theta}{n_a^2(\omega)}+\frac{\sin^2\theta}{n_b^2(\omega)}\right]\times\left[\frac{\cos^2\theta}{n_a^4(\omega)}+\frac{\sin^2\theta}{n_b^4(\omega)}\right]^{1/2}\right]\ . \tag{19}$$

For a uniaxial crystal, $n_a=n_o$ and $n_b=n_e$.

Note that $\rho(\theta,\omega)=0$ for a propagation along one of the three principal axes. For each direction of propagation (θ,ϕ), allowing phase matching or not, the ordinary electric-field vector is orthogonal to the extraordinary one:

$$\mathbf{e}^o(\omega_i,\theta,\phi)\cdot\mathbf{e}^e(\omega_j,\theta,\phi)=0\ . \tag{20}$$

This relation is satisfied when ω_i and ω_j are equal or different.

A. Interactions between three ordinary waves and one extraordinary wave (3*oe*)

Four 3*oe* configurations of polarization are possible according to Table II: (*eooo*), (*oooe*), (*ooeo*), and (*oeoo*).

(a) The number of nonzero elements of the field tensors varies with the direction of propagation (θ,ϕ).

(i) Out of the principal planes ($\theta\neq0°,90°$ and $\phi\neq0°,90°$) only the z components of the ordinary waves, e_z^o, are nil by definition, which leads to the following nil field factors:

$$\begin{aligned} &F_{izkl}=F_{ijzl}=F_{ijkz}=0 \quad \text{for } (eooo),\\ &F_{zjkl}=F_{izkl}=F_{ijzl}=0 \quad \text{for } (oooe)\ ,\\ &F_{zjkl}=F_{izkl}=F_{ijkz}=0 \quad \text{for } (ooeo),\\ &F_{zjkl}=F_{ijzl}=F_{ijkz}=0 \quad \text{for } (oeoo)\ . \end{aligned} \tag{21}$$

(ii) In the x-y plane ($\theta=90°$, any ϕ), the three zero components are e_z^o, e_x^e, and e_y^e, which leads to relations (21) and

$$\begin{aligned} &F_{xjkl}=0 \text{ and } F_{yjkl}=0 \quad \text{for } (eooo)\ ,\\ &F_{ijkx}=0 \text{ and } F_{ijky}=0 \quad \text{for } (oooe)\ ,\\ &F_{ijxl}=0 \text{ and } F_{ijyl}=0 \quad \text{for } (ooeo)\ ,\\ &F_{ixkl}=0 \text{ and } F_{iykl}=0 \quad \text{for } (oeoo)\ . \end{aligned} \tag{22}$$

(iii) In the x-z plane ($\phi=0°$, any θ), the three zero components are e_z^o, e_x^o, and e_y^e, and in the y-z plane ($\phi=90°$, any θ), they are e_z^0, e_y^o, and e_x^e, which leads, for the two planes, to relations (21) and the following:

TABLE II. Correspondence between the types of interactions and the configurations of polarization in term of ordinary (o) and extraordinary (e) waves according to the optical sign of the direction of propagation.

Types of interaction				Configurations of polarization							
				Negative sign				Positive sign			
SFM(ω_4)	DFM(ω_1)	DFM(ω_2)	DFM(ω_3)	ω_4	ω_1	ω_2	ω_3	ω_4	ω_1	ω_2	ω_3
I	II	III	IV	*e*	*o*	*o*	*o*	*o*	*e*	*e*	*e*
II	III	IV	I	*e*	*e*	*e*	*o*	*o*	*o*	*o*	*e*
III	IV	I	II	*e*	*e*	*o*	*e*	*o*	*o*	*e*	*o*
IV	I	II	III	*e*	*o*	*e*	*e*	*o*	*e*	*o*	*o*
V^4	V^1	V^2	V^3	*e*	*e*	*o*	*o*	*o*	*o*	*e*	*e*
VI^4	VI^1	VI^2	VI^3	*e*	*o*	*e*	*o*	*o*	*e*	*o*	*e*
VII^4	VII^1	VII^2	VII^3	*e*	*o*	*o*	*e*	*o*	*e*	*e*	*o*

$$F_{iakl}=F_{ijal}=F_{ijka}=0 \text{ and } F_{bjkl}=0 \text{ for } (eooo),$$
$$F_{ajkl}=F_{iakl}=F_{ijal}=0 \text{ and } F_{ijkb}=0 \text{ for } (oooe),$$
$$F_{ajkl}=F_{iakl}=F_{ijka}=0 \text{ and } F_{ijbl}=0 \text{ for } (ooeo),$$
$$F_{ajkl}=F_{ijal}=F_{ijka}=0 \text{ and } F_{ibkl}=0 \text{ for } (oeoo). \quad (23)$$

$(a,b)=(x,y)$ for the x-z plane and $(a,b)=(y,x)$ for the y-z plane. Hence, according to (21)–(23), the $3oe$ field tensors have 24 $(=2^3\times3^1)$ nonzero elements for the phase-matching directions out of the principal planes, 8 $(=2^3\times1^1)$ in the x-y plane, and 2 $(=1^3\times2^1)$ in the x-z and y-z planes. The only nonzero element along the x axis and the y axis are F_{zaaa} for $(eooo)$, F_{aaaz} for $(oooe)$, F_{aaza} for $(ooeo)$, and F_{azaa} for $(oeoo)$ with $a=y$ along the x axis and $a=x$ along the y axis.

(b) The number of relations between field factors which are due to the orthogonality property (20) is given by the number of possible choices without repetition of two orthogonal polarizations among the four polarizations, i.e., 6 $(=4!/[2!(4-2)!])$:

$$F_{xxij}+F_{yyij}\ (+F_{zzij}=0)=0, \quad (24)$$
$$F_{xixj}+F_{yiyj}\ (+F_{zizj}=0)=0, \quad (25)$$
$$F_{ixxj}+F_{iyyj}\ (+F_{izzj}=0)=0, \quad (26)$$
$$F_{ixjx}+F_{iyjy}\ (+F_{izjz}=0)=0, \quad (27)$$
$$F_{xijx}+F_{yijy}\ (+F_{zijz}=0)=0, \quad (28)$$
$$F_{ijxx}+F_{ijyy}\ (+F_{ijzz}=0)=0. \quad (29)$$

i and j are equal to x or y (the field factors with i or j equal to z are nil). Each tensor obeys three of the previous equalities:

$(eooo)$ (24), (25), and (28),

$(oooe)$ (27), (28), and (29),

$(ooeo)$ (25), (26), and (29),

$(oeoo)$ (24), (26), and (27).

The combination of the three relations of orthogonality for each configuration of polarization leads to specific equalities. For example, the combination of (24), (25), and (28) for $(eooo)$ leads to

$$F_{xxxx}=-F_{yyxx}=-F_{yxyx}=-F_{yxxy},$$
$$F_{yyyy}=-F_{xxyy}=-F_{xyxy}=-F_{xyyx}, \quad (30)$$
$$F_{yxyy}=F_{yyxy}=F_{yyyx}=-F_{xyxx}=-F_{xxyx}=-F_{xxxy}.$$

The four $3oe$ field tensors are symmetric in the three Cartesian indices relative to the ordinary waves for the field factors with x or y as the Cartesian index relative to the extraordinary wave.

(c) The nondispersion in frequency of the direction of the ordinary electric-field vectors leads to the same symmetry as the previous ones but also to symmetry in the three ordinary Cartesian indices of the field factors with z as an extraordinary Cartesian index:

$$F_{ijkl}^{eooo}(\theta,\phi,\omega_a,\omega_b,\omega_c,\omega_d)[j\text{-}k,k\text{-}l], \quad (31)$$
$$F_{ijkl}^{oooe}(\theta,\phi,\omega_a,\omega_b,\omega_c,\omega_d)[i\text{-}j,j\text{-}k], \quad (32)$$
$$F_{ijkl}^{ooeo}(\theta,\phi,\omega_a,\omega_b,\omega_c,\omega_d)[i\text{-}j,j\text{-}l], \quad (33)$$
$$F_{ijkl}^{oeoo}(\theta,\phi,\omega_a,\omega_b,\omega_c,\omega_d)[i\text{-}k,k\text{-}l]. \quad (34)$$

$T_{abkl}[a\text{-}b]$ signifies that the tensor $\mathbf{T}$ is symmetric in the two indices a and b, i.e. [9],

$$T_{abkl}=T_{bakl}. \quad (35)$$

These equations are valid for any value of ω_a, ω_b, ω_c, and ω_d contained in the transparency range of the crystal. Equalities between frequencies do not create any new symmetry.

(d) The four $3oe$ $\mathbf{F}^{(3)}$ tensors have nine independent elements according to orthogonality relations (24)–(30) and to equalities (31)–(34) due to the nondispersion in frequency of the ordinary electric-field vectors.

The matrix representation of the $(eooo)$ field tensor for phase-matching directions out of the principal planes is given in Table III, taking into account the previous relations. The three other field tensors are deduced from the previous one by associated permutations of the Cartesian indices and the corresponding polarizations. According to relations (5), the magnitudes of two permutated elements are equal if the permutation of polarizations are associated with the corresponding frequencies. Thus, according to the definition of the types given in Table II, it is the case for permutations between the following interactions.

(i) $(eooo)$ SFM(ω_4) type I<0 and the three $(oeoo)$ interactions, DFM(ω_1) type II<0, DFM(ω_2) type III<0, DFM(ω_3) type IV<0.

(ii) The three $(oooe)$ interactions, SFM(ω_4) type II>0, DFM(ω_1) type III>0, DFM(ω_2) type IV>0, and $(eooo)$ DFM(ω_3) type I>0.

(iii) The two $(ooeo)$ interactions, SFM(ω_4) type III>0, DFM(ω_1) type IV>0, $(eooo)$ DFM(ω_2) type I>0, and $(oooe)$ DFM(ω_3) type II>0.

(iv) $(oeoo)$ SFM(ω_4) type IV>0, $(eooo)$ DFM(ω_1) type I>0, and the two interactions $(ooeo)$, DFM(ω_2) type II>0, DFM(ω_3) type III>0.

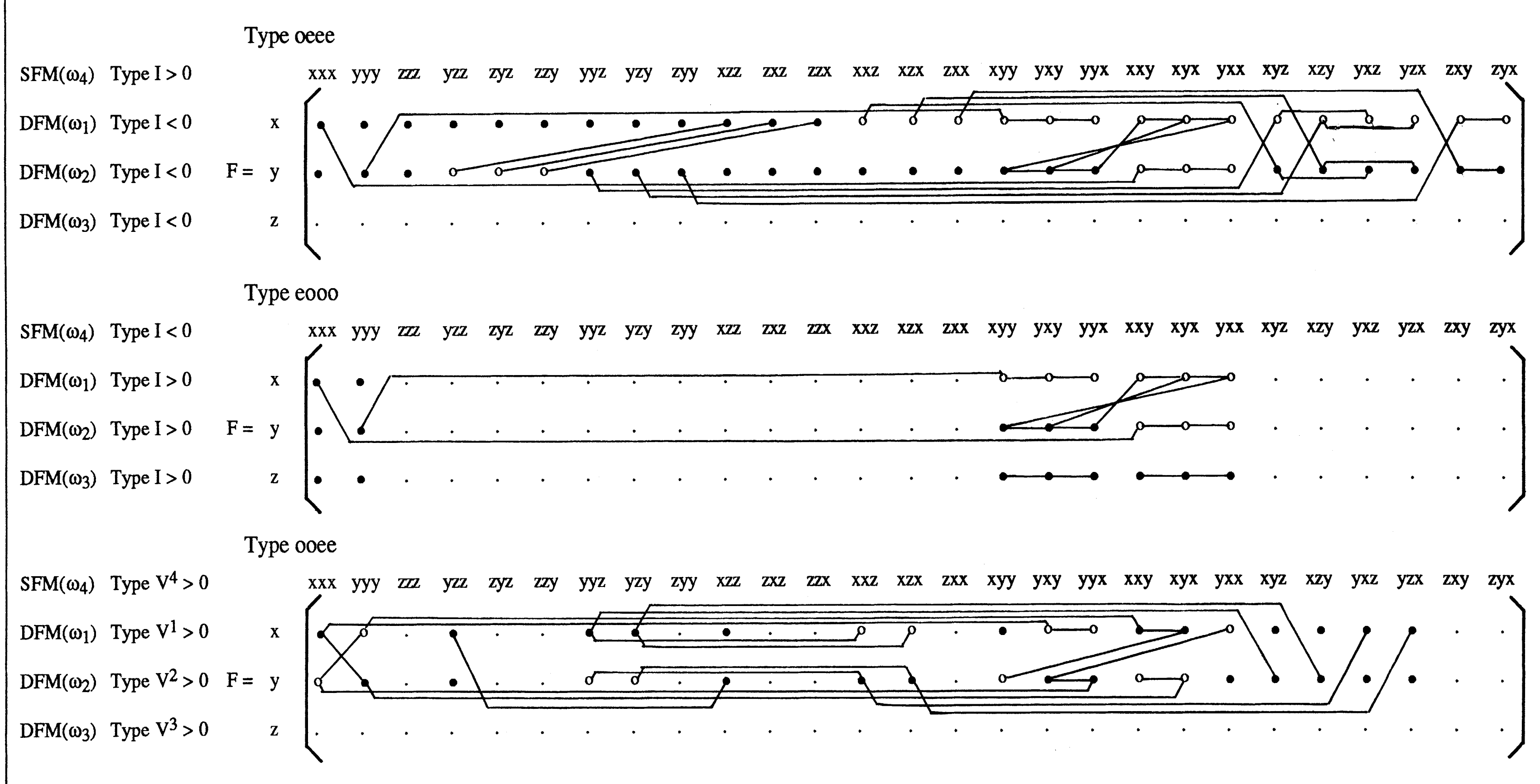
Equalities between frequencies do not create any new symmetry.

(e) According to (4), (17), (18), and (19), the trigonometric functions of the nine independent elements of the $3oe$ field tensors are

$$(1)=f_{\varepsilon}^{(\phi)}g_{\Sigma}^{(\theta)},$$
$$(\bar{1})=f_{\xi}^{(\phi)}g_{\Sigma}^{(\theta)},$$
$$(2)=f_{\eta}^{(\phi)}g_{\zeta}^{(\theta)},$$
$$(\bar{2})=f_{\kappa}^{(\phi)}g_{\zeta}^{(\theta)},$$
$$(3)=(3')=(3'')=f_{\rho}^{(\phi)}g_{\Sigma}^{(\theta)}, \quad (36)$$
$$(\bar{3})=(\bar{3}')=(\bar{3}'')=f_{\nu}^{(\phi)}g_{\Sigma}^{(\theta)},$$
$$(4)=(4')=(4'')=-(5)=f_{\tau}^{(\phi)}g_{\zeta}^{(\theta)},$$
$$(\bar{4})=(\bar{4}')=(\bar{4}'')=-(\bar{5})=f_{\omega}^{(\phi)}g_{\zeta}^{(\theta)},$$
$$(6)=(6')=(6'')=-(\bar{6})=-(\bar{6}')=-(\bar{6}'')=f_{\sigma}^{(\phi)}g_{\zeta}^{(\theta)},$$

TABLE III. Matrix representations of the (*eooo*), (*oeee*), and (*ooee*) field tensors of the uniaxial class for a propagation out of the principal planes. The relations not indicated in the matrices, for more clarity, can be deduced from the present ones.

Interactions	4-rank $\mathbf{F}_{ijkl}(\theta,\phi)$ field tensors for uniaxial class



. $F_{ijkl} = 0$ ●—●, o—o $F_{ijkl} = F_{mnop}$ ●—o $F_{ijkl} = - F_{mnop}$

with

$$f_{\sigma}^{(\phi)}=\cos^2\phi\sin^2\phi\ ,\quad f_{\varepsilon}^{(\phi)}=\cos^3\phi\ ,$$
$$f_{\zeta}^{(\phi)}=-\sin^3\phi\ ,\quad f_{\eta}^{(\phi)}=-\cos^4\phi\ ,$$
$$f_{\kappa}^{(\phi)}=\sin^4\phi\ ,\quad f_{\rho}^{(\phi)}=-\cos^2\phi\sin\phi\ , \tag{37}$$
$$f_{\nu}^{(\phi)}=\sin^2\phi\cos\phi\ ,\quad f_{\tau}^{(\phi)}=\sin\phi\cos^3\phi\ ,$$
$$f_{\omega}^{(\phi)}=-\sin^3\phi\cos\phi\ ,$$

and

$$g_{\Sigma}^{(\theta)}=\sin[\theta\pm\rho(\omega q,\theta)]\ ,$$
$$g_{\zeta}^{(\theta)}=\cos[\theta\pm\rho(\omega q,\theta)] \tag{38}$$

with $-$ for the positive class and $+$ for the negative class. $\rho(\omega q,\theta)$ is the walkoff angle, given by (19). ω_q is the pulsation of the extraordinary wave. ω_q is equal to ω_4 for SFM type I<0, to ω_3 for SFM type II>0; to ω_2 for SFM type III>0, and to ω_1 for SFM type IV>0. The relationship with DFM can be done according to Table II.

The correspondence between functions (36) and field factors F_{ijkl} of the different configurations of polarization is given in Table IV for SFM. The correspondence for DFM can be done from Table IV, relations (5), and Table II.

Each trigonometric function is written as the product of a ϕ angular contribution, $f(\phi)$, with a θ angular contribution, $g(\theta)$. This writing is justified because many functions $f(\phi)$ are common to the three groups of configuration of polarizations 3*oe*, 3*eo*, and 2*o*2*e*; the functions $g(\theta)$ are specific to each group but are common to several trigonometric functions of a given group.

The principle of designation of the trigonometric functions is the following: (i) The Cartesian indices of the field factor named $\overline{N}$ are obtained from the field factor named N by substitution of x by y and y by x. Thus the two functions N and $\overline{N}$ are out of phase of 90°. (ii) Three functions are named N, N', and N'' when the associated field factors are equal because of relations (31), (32), (33), or (34).

(f) According to the nonzero elements of $\chi^{(3)}$ and to the field factors given in Table III, we give in Table V the trigonometric functions of the 3*oe* field factors intervening in the tensorial contraction of $\mathbf{F}^{(3)}$ and $\chi^{(3)}$ for all the uniaxial classes of orientation symmetry. Table V must be read with Table IV for the correspondence between the designation of the function and the field factor according to the configuration of polarization.

The effective coefficient χ_{eff} is nil for the classes $D_6(622)$, $D_{6h}(6/m\ m\ m)$, $D_{3h}(\overline{6}2m)$, and $C_{6v}(6mm)$. These four classes and the three other hexagonal classes $C_{3h}(\overline{6})$, $C_6(6)$, $C_{6h}(6/m)$, have a nil χ_{eff} under Kleinman's symmetry conjecture [10] (i.e., $\chi_{ijkl}[i\text{-}j,j\text{-}k,k\text{-}l]$). Note that the notation of Herman Maügin for the crystalline classes must not be confused with numerotation of field-factor functions.

As an example and according to Table V, we calculate the intervening field factors for each type-I collinear phase-matching direction of direct THG 1.064 μm$\rightarrow$0.355 μm in BaB_2O_4 (BBO), a negative uniaxial

TABLE IV. Correspondence between 3*oe* uniaxial functions and field factors according to the four corresponding types of phase-matched SFM(ω_4). The symbols = and ≅ mean that the functions (for example, 3, 3′, and 3″) are equal in the case of uniaxial crystals and almost equal in the case of biaxial crystals.

		3*oe* uniaxial field-factor functions																							
	Optical sign	1	$\overline{1}$	2	$\overline{2}$	3	3′	3″	$\overline{3}$	$\overline{3}'$	$\overline{3}''$	4	4′	4″	$\overline{4}$	$\overline{4}'$	$\overline{4}''$	5	$\overline{5}$	6	6′	6″	$\overline{6}$	$\overline{6}'$	$\overline{6}''$
	Uniaxial						=	=		=	=		=	=		=	=				=	=		=	=
Interactions	Biaxial						≅	≅		≅	≅		≅	≅		≅	≅				≅	≅		≅	≅
Type I (*eooo*)	<0	*zyyy*	*zxxx*	*xyyy*	*yxxx*	*zxyy*	*zyxy*	*zyyx*	*zyxx*	*zxyx*	*zxxy*	*xxyy*	*xyxy*	*xyyx*	*yyxx*	*yxyx*	*yxxy*	*yyyy*	*xxxx*	*yxyy*	*yyxy*	*yyyx*	*xyxx*	*xxyx*	*xxxy*
Type II (*oooe*)	>0	*yyyz*	*xxxz*	*yyyx*	*xxxy*	*yxyz*	*yyxz*	*xyyz*	*xyxz*	*xxyz*	*yxxz*	*yxyx*	*yyxx*	*xyyx*	*xyxy*	*xxyy*	*yxxy*	*yyyy*	*xxxx*	*yxyy*	*yyxy*	*xyyy*	*xyxx*	*xxyx*	*yxxx*
Type III (*ooeo*)	>0	*yyzy*	*xxzx*	*yyxy*	*xxyx*	*yxzy*	*yyzx*	*xyzy*	*xyzx*	*xxzy*	*yxzx*	*yxxy*	*yyxx*	*xyxy*	*xyyx*	*xxyy*	*yxyx*	*yyyy*	*xxxx*	*yxyy*	*yyyx*	*xyyy*	*xyxx*	*xxxy*	*yxxx*
Type IV (*oeoo*)	>0	*yzyy*	*xzxx*	*yxyy*	*xyxx*	*yzxy*	*yzyx*	*xzyy*	*xzyx*	*yzxy*	*yzxx*	*yxxy*	*yxyx*	*xxyy*	*xyyx*	*xyxy*	*yyxx*	*yyyy*	*xxxx*	*yyxy*	*yyyx*	*xyyy*	*xxyx*	*xxxy*	*yxxx*

TABLE V. $3oe$, $3eo$, and $2o2e$ uniaxial field-factor functions intervening in the calculation of the effective coefficient for the uniaxial crystal classes.

Uniaxial crystal classes	S_4, C_4, C_{4h}, C_{3h}, C_6, C_{6h}	C_{4v}, D_{2d}, D_4, D_{4h}, C_{6v}, D_{3h}, D_6, D_{6h}	C_3, C_{3i}	C_{3v}, D_3, D_{3d}
Intervening $3oe$ field-factor functions	$2,\bar{2}$ $4,4',4'',\bar{4},\bar{4}',\bar{4}''$ $5,\bar{5}$ $6,6',6'',\bar{6},\bar{6}',\bar{6}''$	$4,4',4'',\bar{4},\bar{4}',\bar{4}''$ $5,\bar{5}$	$1,\bar{1}$ $2,\bar{2}$ $3,3',3'',\bar{3},\bar{3}',\bar{3}''$ $4,4',4'',\bar{4},\bar{4}',\bar{4}''$ $5,\bar{5}$ $6,6',6'',\bar{6},\bar{6}',\bar{6}''$	1 $\bar{3},\bar{3}',\bar{3}''$ $4,4',4'',\bar{4},\bar{4}',\bar{4}''$ $5,\bar{5}$
Intervening $3eo$ field-factor functions	$2a,2b,2c,\bar{2}a,\bar{2}b,\bar{2}c$ $4a,4b,4c,\bar{4}a,\bar{4}b,\bar{4}c$ $7,\bar{7}$ $8,8',8'',\bar{8},\bar{8}',\bar{8}''$ $9,\bar{9}$ $10,10',10'',\overline{10},\overline{10}',\overline{10}''$	$4a,4b,4c,\bar{4}a,\bar{4}b,\bar{4}c$ $9,\bar{9}$ $10,10',10'',\overline{10},\overline{10}',\overline{10}''$	$2a,2b,2c,\bar{2}a,\bar{2}b,\bar{2}c$ $3a,3b,3c,\bar{3}a,\bar{3}b,\bar{3}c$ $4a,4b,4c,\bar{4}a,\bar{4}b,\bar{4}c$ $5a,5b,5c,\bar{5}a,\bar{5}b,\bar{5}c$ $5'a,5'b,5'c,\bar{5}'a,\bar{5}'b,\bar{5}'c$ $6a,6b,6c,\bar{6}a,\bar{6}b,\bar{6}c$ $7,\bar{7}$ $8,8',8'',\bar{8},\bar{8}',\bar{8}''$ $9,\bar{9}$ $10,10',10'',\overline{10},\overline{10}',\overline{10}''$	$3a,3b,3c$ $4a,4b,4c,\bar{4}a,\bar{4}b,\bar{4}c$ $\bar{5}a,\bar{5}b,\bar{5}c,\bar{5}'a,\bar{5}'b,\bar{5}'c$ $\bar{6}a,\bar{6}b,\bar{6}c$ $9,\bar{9}$ $10,10',10'',\overline{10},\overline{10}',\overline{10}''$
Intervening $2o2e$ field-factor functions	$1,1'$ $4,\bar{4}$ $6,6',\bar{6},\bar{6}'$ $7,7',\bar{7},\bar{7}'$ $8,\bar{8}$ $9,\bar{9}$ $10,10',\overline{10},\overline{10}'$	$4,\bar{4}$ $8,\bar{8}$ $9,\bar{9}$ $10,10',\overline{10},\overline{10}'$	all	$2a,2b,2'a,2'b$ $3a,3b$ $4,\bar{4}$ $\bar{5}a,\bar{5}b$ $8,\bar{8}$ $9,\bar{9}$ $10,10',\overline{10},\overline{10}'$

nonlinear crystal which belongs to the crystal class $C_{3v}(3m)$. The refractive indices at the interacting wavelengths are the following [11]:

$$n_o=1.6545\ ,\quad n_e=1.5339 \quad \text{at } \lambda_1=1.064\ \mu\text{m}\ ,$$
$$n_o=1.7055\ ,\quad n_e=1.5766 \quad \text{at } \lambda_4=0.355\ \mu\text{m}\ . \tag{39}$$

The phase-matching directions of SFM, calculated according to Table II and from (39), are located at $\theta=37.32°$ for any ϕ. The field factors are plotted in Fig. 1.

B. Interactions between three extraordinary waves and one ordinary wave ($3eo$)

$(oeee)$, $(eeeo)$, $(eeoe)$, and $(eoee)$ are the four $3eo$ configurations of polarization allowing phase-matching according to Table II.

(a) The counting of the nil field factors according to the localization of the phase-matching direction is done on the same basis than for the $3oe$ interactions. We have the following.

(i) Out of the principal planes:

$$F_{zjkl}=0 \ \text{ for } (oeee)\ ,\quad F_{ijkz}=0 \ \text{ for } (eeeo)\ ,$$
$$F_{ijzl}=0 \ \text{ for } (eeoe)\ ,\quad F_{izkl}=0 \ \text{ for } (eoee)\ . \tag{40}$$

(ii) In the x-y plane: Equations (40) and

$$F_{ixkl}=F_{ijxl}=F_{ijkx}=0 \ \text{ and } \ F_{iykl}=F_{ijyl}=F_{ijky}=0 \ \text{ for } (oeee)\ ,$$
$$F_{xjkl}=F_{ixkl}=F_{ijxl}=0 \ \text{ and } \ F_{yjkl}=F_{iykl}=F_{ijyl}=0 \ \text{ for } (eeeo)\ ,$$
$$F_{xjkl}=F_{ixkl}=F_{ijkx}=0 \ \text{ and } \ F_{yjkl}=F_{iykl}=F_{ijky}=0 \ \text{ for } (eeoe)\ ,$$
$$F_{xjkl}=F_{ijxl}=F_{ijkx}=0 \ \text{ and } \ F_{yjkl}=F_{ijyl}=F_{ijky}=0 \ \text{ for } (eoee)\ . \tag{41}$$

(iii) In the x-z and y-z planes: Equations (40) and the same relations (23) as for the $3oe$ interactions but with $(a,b)=(y,x)$ for the x-z plane and $(a,b)=(x,y)$ for the y-z plane.

Thus the $3eo$ field tensors have 54 ($=2^1\times3^3$) nonzero field factors out of the principal planes, 2 ($=2^1\times1^3$) in the x-y plane and 8 ($=1^1\times2^3$) in the x-z and y-z planes. There is only one nonzero element along the x axis and the y axis: F_{azzz} for $(oeee)$, F_{zzza} for $(eeeo)$, F_{zzaz} for $(eeoe)$, and F_{zazz} for $(eoee)$ with $a=y$ along the x axis and $a=x$ along the y axis.

(b) The relations between the $3eo$ field factors which

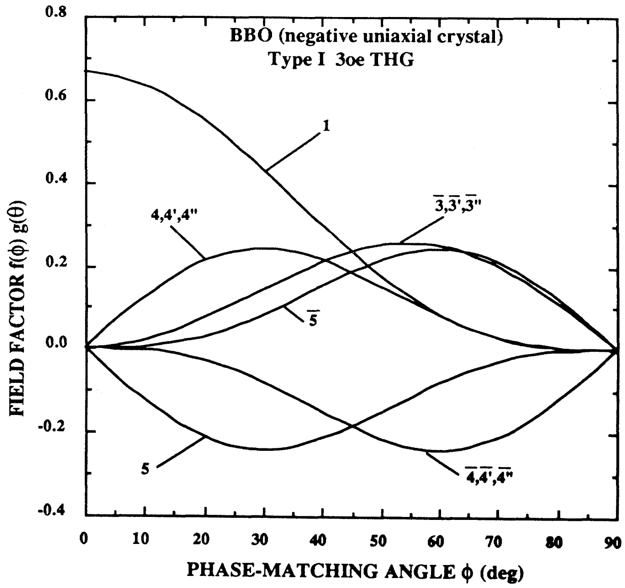


FIG. 1. Intervening $3oe$ uniaxial field-factor functions vs the phase-matching spherical coordinate ϕ, calculated for direct type-I THG $1.064 \rightarrow 0.355$ μm in BBO (crystal class C_{3v}).

are due to orthogonality of the ordinary and extraordinary electric-field vectors are the same as those of the $3oe$ field factors.

Each tensor obeys the following equalities:

$(oeee)$, (24), (25), and (28) ,

$(eeeo)$, (27), (28), and (29) ,

$(eeoe)$, (25), (26), and (29) ,

$(eoee)$, (24), (26), and (27) .

The Cartesian indices i and j can be equal to x, y, and z.

The combination of the three relations specific to each tensor lead to the same equalities of type (30) as for the $3oe$ field tensors. The $3eo$ field tensors are also symmetric in the Cartesian indices x and y relative to extraordinary waves.

(c) This symmetry can also be deduced from the nondispersion in frequency of the projection of the extraordinary electric-field vector in the x-y plane. This property does not create any new symmetry.

(d) Thus the $3eo$ field tensors are "less symmetric" than the $3oe$. They have 28 independent elements. The matrix representation of the $(oeee)$ field tensor is given in Table III for directions of propagation out of the principal planes. The three other $3eo$ field tensors are deduced from this one by associated permutation of the Cartesian indices and the corresponding polarizations.

According to relations (5) and Table II, two permutated field factors have the same magnitude for permutation between the following interactions:

(i) $(oeee)$ SFM(ω_4) type I>0 and the three $(eoee)$ interactions, DFM(ω_1) type II>0, DFM(ω_2) type III>0, DFM(ω_3) type IV>0.

(ii) The three $(eeeo)$ interactions, SFM(ω_4) type II<0, DFM(ω_1) type III<0, DFM(ω_2) type IV<0, and $(oeee)$ DFM(ω_3) type I<0.

(iii) The two $(eeoe)$ interactions, SFM(ω_4) type III<0, DFM(ω_1) type IV<0, $(oeee)$ DFM(ω_2) type I<0, and $(eeeo)$ DFM(ω_3) type II<0.

(iv) $(eoee)$ SFM(ω_4) type IV<0, $(oeee)$ DFM(ω_1) type I<0, and the two interactions $(eeoe)$, DFM(ω_2) type II<0, DFM(ω_3) type III<0.

(e) Equalities between circular frequencies create new symmetries which are not valid for all the SFM and DFM interactions of the same configuration of polarization. The field tensors are symmetric in the Cartesian indices relative to extraordinary waves at the same pulsation ω:

$$\begin{aligned}
&F_{ijkl}^{oeee}(\omega_4=\omega_1+\omega+\omega)[k\text{-}l] ,\\
&F_{ijkl}^{oeee}(\omega_4=\omega+\omega_2+\omega)[j\text{-}l] ,\\
&F_{ijkl}^{oeee}(\omega_4=\omega+\omega+\omega_3)[j\text{-}k] ,\\
&F_{ijkl}^{oeee}(3\omega=\omega+\omega+\omega)[j\text{-}k,k\text{-}l] ,\\
&F_{ijkl}^{eeeo}(\omega_4=\omega+\omega+\omega_3)[j\text{-}k] ,\\
&F_{ijkl}^{eeoe}(\omega_4=\omega+\omega_2+\omega)[j\text{-}l] ,\\
&F_{ijkl}^{eoee}(\omega_4=\omega_1+\omega+\omega)[k\text{-}l] .
\end{aligned} \tag{42}$$

The symmetry for DFM can be obtained from (42) and the permutation relations (5).

(f) The $3eo$ field tensors are symmetric in the three extraordinary Cartesian indices in the general case $(\omega_b \neq \omega_c \neq \omega_d)$ only if the dispersion in frequency of the walk-off angle can be neglected $(\partial\rho(\omega)/\partial\omega \cong 0)$ in the range containing the four frequencies.

(g) The expression of the trigonometric functions of the 28 independent elements of the $3eo$ field tensors are the following:

$$\begin{aligned}
&(1)=f_\alpha^{(\phi)}g_\Delta^{(\theta)} ,\\
&(\bar{1})=f_\beta^{(\phi)}g_\Delta^{(\theta)} ,\\
&(\bar{2}a)=f_\delta^{(\phi)}g_\Lambda^{(\theta)} , \quad (\bar{2}b)=f_\delta^{(\phi)}g_\Pi^{(\theta)} , \quad (\bar{2}c)=f_\delta^{(\phi)}g_\Psi^{(\theta)} ,\\
&(2a)=f_\gamma^{(\phi)}g_\Lambda^{(\theta)} , \quad (2b)=f_\gamma^{(\phi)}g_\Pi^{(\theta)} , \quad (2c)=f_\gamma^{(\phi)}g_\Psi^{(\theta)} ,\\
&(3a)=f_\varepsilon^{(\phi)}g_H^{(\theta)} , \quad (3b)=f_\varepsilon^{(\phi)}g_K^{(\theta)} , \quad (3c)=f_\varepsilon^{(\phi)}g_\Omega^{(\theta)} ,\\
&(\bar{3}a)=f_\zeta^{(\phi)}g_H^{(\theta)} , \quad (\bar{3}b)=f_\zeta^{(\phi)}g_K^{(\theta)} , \quad (\bar{3}c)=f_\zeta^{(\phi)}g_\Omega^{(\theta)} ,\\
&(\bar{4}a)=-(4a)=f_\mu^{(\phi)}g_\Lambda^{(\theta)} , \quad (\bar{4}b)=-(4b)=f_\mu^{(\phi)}g_\Pi^{(\theta)} ,\\
&(\bar{4}c)=-(4c)=f_\mu^{(\phi)}g_\Psi^{(\theta)} ,\\
&(6a)=-(5a)=-(5'a)=f_\rho^{(\phi)}g_H^{(\theta)} ,\\
&(6b)=-(5b)=-(5'b)=f_\rho^{(\phi)}g_K^{(\theta)} ,\\
&(6c)=-(5c)=-(5'c)=f_\rho^{(\phi)}g_\Omega^{(\theta)} ,\\
&(\bar{6}a)=-(\bar{5}a)=-(\overline{5'a})=f_\upsilon^{(\phi)}g_H^{(\theta)} ,\\
&(\bar{6}b)=-(\bar{5}b)=-(\overline{5'b})=f_\upsilon^{(\phi)}g_K^{(\theta)} ,\\
&(\bar{6}c)=-(\bar{5}c)=-(\overline{5'c})=f_\upsilon^{(\phi)}g_\Omega^{(\theta)} ,\\
&(7)=f_\eta^{(\phi)}g_\Gamma^{(\theta)} ,\\
&(\bar{7})=f_\kappa^{(\phi)}g_\Gamma^{(\theta)} ,\\
&(\bar{8})=-(8)=(\bar{8}')=-(8')=(\bar{8}'')=-(8'')=f_\sigma^{(\phi)}g_\Gamma^{(\theta)} ,\\
&(9)=-(10)=-(10')=-(10'')=f_\tau^{(\phi)}g_\Gamma^{(\theta)} ,\\
&(\bar{9})=-(\overline{10})=-(\overline{10'})=-(\overline{10''})=f_\omega^{(\phi)}g_\Gamma^{(\theta)} .
\end{aligned} \tag{43}$$

There are nine functions $f(\phi)$ which are equal to those of the $3oe$ field factors; their expressions are given in (37). The five others are

$$f_{\alpha}^{(\phi)}=\cos\phi \ , \quad f_{\beta}^{(\phi)}=-\sin\phi \ , \quad f_{\gamma}^{(\phi)}=-\cos^2\phi \ , \quad f_{\delta}^{(\phi)}=\sin^2\phi \ , \quad f_{\mu}^{(\phi)}=\sin\phi\cos\phi \ . \tag{44}$$

There are no common functions with the $3oe$ field factors among the eight $g(\theta)$ functions:

$$\begin{aligned}
g_{\Gamma}^{(\theta)}&=\cos[\theta\pm\rho(\omega_q,\theta)]\cos[\theta\pm\rho(\omega_r,\theta)]\cos[\theta\pm\rho(\omega_s,\theta)] \ ,\\
g_{\Delta}^{(\theta)}&=\sin[\theta\pm\rho(\omega_q,\theta)]\sin[\theta\pm\rho(\omega_r,\theta)]\sin[\theta\pm\rho(\omega_s,\theta)] \ ,\\
g_{H}^{(\theta)}&=\cos[\theta\pm\rho(\omega_q,\theta)]\cos[\theta\pm\rho(\omega_r,\theta)]\sin[\theta\pm\rho(\omega_s,\theta)] \ ,\\
g_{K}^{(\theta)}&=\cos[\theta\pm\rho(\omega_q,\theta)]\sin[\theta\pm\rho(\omega_r,\theta)]\cos[\theta\pm\rho(\omega_s,\theta)] \ ,\\
g_{\Lambda}^{(\theta)}&=\cos[\theta\pm\rho(\omega_q,\theta)]\sin[\theta\pm\rho(\omega_r,\theta)]\sin[\theta\pm\rho(\omega_s,\theta)] \ ,\\
g_{\Pi}^{(\theta)}&=\sin[\theta\pm\rho(\omega_q,\theta)]\cos[\theta\pm\rho(\omega_r,\theta)]\sin[\theta\pm\rho(\omega_s,\theta)] \ ,\\
g_{\Psi}^{(\theta)}&=\sin[\theta\pm\rho(\omega_q,\theta)]\sin[\theta\pm\rho(\omega_r,\theta)]\cos[\theta\pm\rho(\omega_s,\theta)] \ ,\\
g_{\Omega}^{(\theta)}&=\sin[\theta\pm\rho(\omega_q,\theta)]\cos[\theta\pm\rho(\omega_r,\theta)]\cos[\theta\pm\rho(\omega_s,\theta)] \ ,
\end{aligned} \tag{45}$$

with $-$ for the positive class and $+$ for the negative class. $\rho(\omega,\theta)$ is given by (19). ω_q, ω_r, and ω_s are relative to the extraordinary waves. $(\omega_q,\omega_r,\omega_s)$ are equal to $(\omega_1,\omega_2,\omega_3)$ for SFM type I>0, to $(\omega_4,\omega_1,\omega_2)$ for SFM type II<0, to $(\omega_4,\omega_1,\omega_3)$ for SFM type III<0, and to $(\omega_4,\omega_2,\omega_3)$ for SFM type IV<0. The relationship with DFM can be done according to Table II.

The correspondence between functions (43) and field factors is given in Table VI for SFM and from Table VI, relations (5) and Table II for DFM. Two functions N and $\bar{N}$ are related to field factors which correspond by substitution of x by y or y by x. Functions N, N', and N'' are equal and are relative to field factors symmetric in the Cartesian indices x and y relative to extraordinary waves. Functions named N_a, N_b, and N_c have the same contribution $f(\phi)$ and differ by $g(\theta)$; the associated field factors correspond by permutation of z by x and y.

(h) Table V gives the trigonometric functions of the $3eo$ field factors which intervene in the tensorial contraction of $\mathbf{F}^{(3)}$ and $\chi^{(3)}$. The effective coefficient is nil for the same crystal classes than those of the $3oe$ interactions.

We calculate the intervening field factors for each type-II collinear phase-matching direction of direct THG 1.064 μm$\rightarrow$0.355 μm in BBO.

The phase-matching directions calculated according to Table II and from the refractive indices (39) are located at $\theta=81.22°$ for any ϕ. The field factors are plotted in Fig. 2.

C. Interactions between two ordinary waves and two extraordinary waves ($2o2e$)

According to Table II, the six $2o2e$ possible phase-matched configurations of polarization are $(eeoo)$, $(eoeo)$, $(eooe)$, $(ooee)$, $(oeoe)$, and $(oeeo)$.

(a) The nil components of the $2o2e$ field tensors are the following according to the localization of the direction of propagation.

(i) Out of the principal planes:

$$\begin{aligned}
&F_{ijkz}=F_{ijzl}=0 \quad \text{for } (eeoo) \ ,\\
&F_{ijkz}=F_{izkl}=0 \quad \text{for } (eoeo) \ ,\\
&F_{ijzl}=F_{izkl}=0 \quad \text{for } (eooe) \ ,\\
&F_{izkl}=F_{zjkl}=0 \quad \text{for } (ooee) \ ,\\
&F_{ijzl}=F_{zjkl}=0 \quad \text{for } (oeoe) \ ,\\
&F_{ijkz}=F_{zjkl}=0 \quad \text{for } (oeeo) \ .
\end{aligned} \tag{46}$$

(ii) In the x-y plane: Equations (46) and

$$\begin{aligned}
&F_{ajkl}=F_{iakl}=0 \quad \text{for } (eeoo) \ ,\\
&F_{ajkl}=F_{ijal}=0 \quad \text{for } (eoeo) \ ,\\
&F_{ajkl}=F_{ijka}=0 \quad \text{for } (eooe) \ ,\\
&F_{ijal}=F_{ijka}=0 \quad \text{for } (ooee) \ ,\\
&F_{iakl}=F_{ijka}=0 \quad \text{for } (oeoe) \ ,\\
&F_{iakl}=F_{ijal}=0 \quad \text{for } (oeeo) \ .
\end{aligned} \tag{47}$$

a is equal to x or y.

(iii) In the x-z and y-z planes: Equations (46) and

$$\begin{aligned}
&F_{ijal}=F_{ijka}=F_{bjkl}=F_{ibkl}=0 \quad \text{for } (eeoo) \ ,\\
&F_{iakl}=F_{ijka}=F_{bjkl}=F_{ijbl}=0 \quad \text{for } (eoeo) \ ,\\
&F_{iakl}=F_{ijal}=F_{bjkl}=F_{ijkb}=0 \quad \text{for } (eooe) \ ,\\
&F_{ajkl}=F_{iakl}=F_{ijbl}=F_{ijkb}=0 \quad \text{for } (ooee) \ ,\\
&F_{ajkl}=F_{ijal}=F_{ibkl}=F_{ijkb}=0 \quad \text{for } (oeoe) \ ,\\
&F_{ajkl}=F_{ijka}=F_{ibkl}=F_{ijbl}=0 \quad \text{for } (oeeo) \ .
\end{aligned} \tag{48}$$

$(a,b)=(x,y)$ for the x-z plane and $(a,b)=(y,x)$ for the y-z plane. Hence, according to (46)–(48), the $2o2e$ field tensors have 36 $(=3^2\times2^2)$ nonzero elements out of the principal planes, 4 $(=2^2\times1^2)$ in the x-y plane, and 4 $(=1^2\times2^2)$ in the x-z and y-z planes. The only nonzero element along the x axis and the y axis are F_{zzaa} for $(eeoo)$, F_{zaza} for $(eoeo)$, F_{zaaz} for $(eooe)$, F_{aazz} for $(ooee)$, F_{azaz} for $(oeoe)$, and F_{azza} for $(oeeo)$, with $a=y$ along the x axis and $a=x$ along the y axis.

(b) Each $2o2e$ field tensor obeys four orthogonality relations instead of three for the $3oe$ and $3eo$ field factors:

$(eeoo)$ and $(ooee)$ (25), (26), (27), (28) ,

$(eoeo)$ and $(oeoe)$ (24), (26), (28), (29) ,

$(eooe)$ and $(oeeo)$ (24), (25), (27), (29) .

i and j are equal to x and y (the field factors with i or j equal to z are nil).

For each tensor, the combination of the four orthogonality relations leads to specific equalities. For example, we have the following equalities by combination of (25)–(28):

TABLE VI. Correspondence between $3eo$ uniaxial functions and field factors according to the four corresponding types of phase-matched SFM(ω_4).

		3eo uniaxial field-factor functions																			
Interactions	Optical sign	1	$\bar{1}$	2a	2b	2c	$\bar{2}a$	$\bar{2}b$	$\bar{2}c$	3a	3b	3c	$\bar{3}a$	$\bar{3}b$	$\bar{3}c$	4a	4b	4c	$\bar{4}a$	$\bar{4}b$	$\bar{4}c$
Type I	>0	yzzz	xzzz	yxzz	yzxz	yzzx	xyzz	xzyz	xzzy	yxxz	yxzx	yzxx	xyyz	xyzy	xzyy	yyzz	yzyz	yzzy	xxzz	xzxz	xzzx
Type II	<0	zzzy	zzzx	zxzy	zzxy	xzzy	zyzx	zzyx	yzzx	zxxy	xxzy	xzxy	zyyx	yyzx	yzyx	zyzy	zzyy	yzzy	zxzx	zzxx	xzzx
Type III	<0	zzyz	zzxz	zxyz	zzyx	xzyz	zyxz	zzxy	yzxz	zxyx	xxyz	xzyx	zyxy	yyxz	yzxy	zyyz	zzyy	yzyz	zxxz	zzxx	xzxz
Type IV	<0	zyzz	zxzz	zyxz	zyzx	xyzz	zxyz	zxzy	yxzz	zyxx	xyxz	xyzx	zxyy	yxyz	yxzy	zyyz	zyzy	yyzz	zxxz	zxzx	xxzz

Interactions	Optical sign	5a	5′a	5b	5′b	5c	5′c	$\bar{5}a$	$\bar{5}'a$	$\bar{5}b$	$\bar{5}'b$	$\bar{5}c$	$\bar{5}'c$	6a	6b	6c	$\bar{6}a$	$\bar{6}b$	$\bar{6}c$
	Uniaxial	=		=		=		=		=		=							
	Biaxial	≅		≅		≅		≅		≅		≅							
Type I	>0	yxyz	yyxz	yxzy	yyzx	yzxy	yzyx	xyxz	xxyz	xyzx	xxzy	xzyx	xzxy	xxxz	xxzx	xzxx	yyyz	yyzy	yzyy
Type II	<0	zxyy	zyxy	yxzy	xyzy	yzxy	xzyy	zyxx	zxyx	xyzx	yxzx	xzyx	yzxx	zxxx	xxzx	xzxx	zyyy	yyzy	yzyy
Type III	<0	zxyy	zyyx	yxyz	xyyz	yzyx	xzyy	zyxx	zxxy	xyxz	yxxz	xzxy	yzxx	zxxx	xxxz	xzxx	zyyy	yyyz	yzyy
Type IV	<0	zyxy	zyyx	yyxz	xyyz	yyzx	xyzy	zxyx	zxxy	xxyz	yxxz	xxzy	yxzx	zxxx	xxxz	xxzx	zyyy	yyyz	yyzy

Interactions	Optical sign	7	$\bar{7}$	8	8′	8″	$\bar{8}$	$\bar{8}'$	$\bar{8}''$	9	$\bar{9}$	10	10′	10″	$\overline{10}$	$\overline{10}'$	$\overline{10}''$
	Uniaxial			=	=		=	=				=	=		=	=	
	Biaxial			≅	≅		≅	≅				≅	≅		≅	≅	
Type I	>0	yxxx	xyyy	yxyy	yyxy	yyyx	xyxx	xxyx	xxxy	xxxx	yyyy	yyxx	yxyx	yxxy	xxyy	xyxy	xyyx
Type II	<0	xxxy	yyyx	yxyy	yyxy	xyyy	xyxx	xxyx	yxxx	xxxx	yyyy	xyxy	xxyy	yxxy	yxyx	yyxx	xyyx
Type III	<0	xxyx	yyxy	yxyy	yyyx	xyyy	xyxx	xxxy	yxxx	xxxx	yyyy	xyyx	xxyy	yxyx	yxxy	yyxx	xyxy
Type IV	<0	xyxx	yxyy	yyxy	yyyx	xyyy	xxyx	xxxy	yxxx	xxxx	yyyy	xyyx	xyxy	yyxx	yxxy	yxyx	xxyy

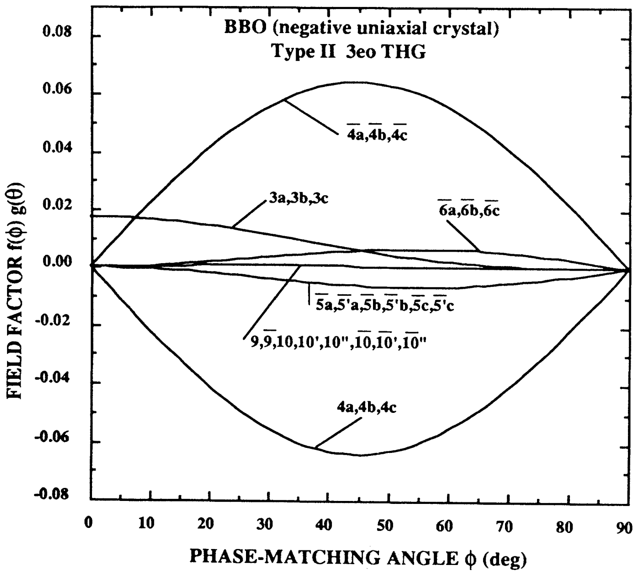


FIG. 2. Intervening $3eo$ uniaxial field-factor functions vs the phase-matching spherical coordinate ϕ, calculated for direct type-II THG $1.064 \rightarrow 0.355$ μm in BBO (crystal class C_{3v}).

$$
\begin{aligned}
&F_{xxxx}=F_{yyyy}=-F_{yxyx}=-F_{xyyx}=-F_{xyxy}=-F_{yxxy}\ ,\\
&F_{xxxy}=F_{xxyx}=-F_{xyyy}=-F_{yxyy}\ ,\\
&F_{yyyx}=F_{yyxy}=-F_{yxxx}=-F_{xyxx}\ .
\end{aligned}
\tag{49}
$$

(c) The nondispersion in frequency of the direction of the ordinary electric field vectors leads to symmetry of the field tensors in the two Cartesian indices relative to the ordinary waves:

$$
\begin{aligned}
&F_{ijkl}^{eeoo}(\theta,\phi,\omega_a,\omega_b,\omega_c,\omega_d)[k\text{-}l]\ ,\\
&F_{ijkl}^{ooee}(\theta,\phi,\omega_a,\omega_b,\omega_c,\omega_d)[i\text{-}j]\ ,\\
&F_{ijkl}^{eoeo}(\theta,\phi,\omega_a,\omega_b,\omega_c,\omega_d)[j\text{-}l]\ ,\\
&F_{ijkl}^{oeoe}(\theta,\phi,\omega_a,\omega_b,\omega_c,\omega_d)[i\text{-}k]\ ,\\
&F_{ijkl}^{eooe}(\theta,\phi,\omega_a,\omega_b,\omega_c,\omega_d)[j\text{-}k]\ ,\\
&F_{ijkl}^{oeeo}(\theta,\phi,\omega_a,\omega_b,\omega_c,\omega_d)[i\text{-}l]\ .
\end{aligned}
\tag{50}
$$

These symmetries are valid for all i, j, k, and l. Note that according to (49), the field tensors are symmetric in the Cartesian indices relative to extraordinary waves only if these indices are x or y; it is not the case with z.

(d) The six $2o2e$ field tensors have 16 independent elements in the general case ($\omega_b \neq \omega_c \neq \omega_d$) according to (49) and (50). The matrix representation of the ($ooee$) field tensor is given in Table III for phase-matching directions out of the principal planes. The five other $2o2e$ field tensors are deduced from Table III by associated permutation of the Cartesian indices and the corresponding polarizations.

According to relations (5) and Table II, two permutated field factors have the same magnitude for permutation between the following interactions:

(i) For type $\mathrm{V}^i<0$, the two ($eeoo$) interactions, SFM(ω_4), DFM(ω_1), and the two ($oeeo$) interactions, DFM(ω_2), DFM(ω_3).

(ii) For type $\mathrm{VI}^i<0$, ($eoeo$) SFM(ω_4), ($oeeo$) DFM(ω_1), ($eeoo$) DFM(ω_2), and ($oeoe$) DFM(ω_3).

(iii) For type $\mathrm{VII}^i<0$, ($eooe$) SFM(ω_4), the two ($oeoe$) interactions, DFM(ω_1), DFM(ω_2), and ($eeoo$) DFM(ω_3).

(iv) For type $\mathrm{V}^i>0$, the two ($ooee$) interactions, SFM(ω_4) and DFM(ω_1), and the two ($eooe$) interactions, DFM(ω_2), DFM(ω_3).

(v) For type $\mathrm{VI}^i>0$, ($oeoe$) SFM(ω_4), ($eooe$) DFM(ω_1), ($ooee$) DFM(ω_2), and ($eoeo$) DFM(ω_3).

(vi) For type $\mathrm{VII}^i>0$, ($oeeo$) SFM(ω_4), the two ($eoeo$) interactions, DFM(ω_1), DFM(ω_2), and ($ooee$) DFM(ω_3).

i refers to ω_i.

(e) Equalities between frequencies add symmetry in the Cartesian indices relative to the extraordinary waves for particular interactions. Then, according to (50), we have

$$
\begin{aligned}
&F_{ijkl}^{oeoe}(\omega_4=\omega+\omega_2+\omega)[i\text{-}k,j\text{-}l]\ ,\\
&F_{ijkl}^{oeoe}(3\omega=\omega+\omega+\omega)[i\text{-}k,j\text{-}l]\ ,\\
&F_{ijkl}^{oeeo}(\omega_4=\omega+\omega+\omega_3)[i\text{-}l,j\text{-}k]\ ,\\
&F_{ijkl}^{oeeo}(3\omega=\omega+\omega+\omega)[i\text{-}l,j\text{-}k]\ ,\\
&F_{ijkl}^{ooee}(\omega_4=\omega_1+\omega+\omega)[i\text{-}j,k\text{-}l]\ ,\\
&F_{ijkl}^{ooee}(3\omega=\omega+\omega+\omega)[i\text{-}j,k\text{-}l]\ .
\end{aligned}
\tag{51}
$$

The symmetries for DFM can be obtained from (51) and (5).

(f) In the general case ($\omega_b \neq \omega_c \neq \omega_d$), the six $2o2e$ field factors are symmetric in the two Cartesian indices relative to extraordinary wave only if the dispersion in frequency of the walkoff angle can be neglected.

(g) The trigonometric functions of the 16 independent $2o2e$ field factors are the following:

$$
\begin{aligned}
&(1)=(1')=-f_{\mu}^{(\phi)}g_{\Theta}^{(\theta)}\ ,\\
&(2a)=(2'a)=-(3a)=-f_{\rho}^{(\phi)}g_{X}^{(\theta)}\ ,\\
&(2b)=(2'b)=-(3b)=-f_{\rho}^{(\phi)}g_{\Xi}^{(\theta)}\ ,\\
&(\bar{2}a)=(\bar{2}'a)=-(\bar{3}a)=f_{\upsilon}^{(\phi)}g_{X}^{(\theta)}\ ,\\
&(\bar{2}b)=(\bar{2}'b)=-(\bar{3}b)=f_{\upsilon}^{(\phi)}g_{\Xi}^{(\theta)}\ ,\\
&(4)=f_{\delta}^{(\phi)}g_{\Theta}^{(\theta)}\ ,\\
&(\bar{4})=-f_{\gamma}^{(\phi)}g_{\Theta}^{(\theta)}\ ,\\
&(5a)=-f_{\varepsilon}^{(\phi)}g_{X}^{(\theta)}\ ,\quad (5b)=-f_{\varepsilon}^{(\phi)}g_{\Xi}^{(\theta)}\ ,\\
&(\bar{5}a)=f_{\zeta}^{(\phi)}g_{X}^{(\theta)}\ ,\quad (\bar{5}b)=f_{\zeta}^{(\phi)}g_{\Xi}^{(\theta)}\ ,\\
&(6)=(6')=-(7)=-(7')=-f_{\omega}^{(\phi)}g_{M}^{(\theta)}\ ,\\
&(\bar{6})=(\bar{6}')=-(\bar{7})=-(\bar{7}')=f_{\tau}^{(\phi)}g_{M}^{(\theta)}\ ,\\
&(8)=f_{\kappa}^{(\phi)}g_{M}^{(\theta)}\ ,\\
&(\bar{8})=-f_{\eta}^{(\phi)}g_{M}^{(\theta)}\ ,\\
&(9)=-(10)=-(10')=(\bar{9})=-(\overline{10})=-(\overline{10}')=f_{\sigma}^{(\phi)}g_{M}^{(\theta)}\ .
\end{aligned}
\tag{52}
$$

The 12 functions $f(\phi)$ are common to those of $3oe$ or $3eo$ interactions. The functions $g(\theta)$ are specific:

$$\begin{aligned} g_{\Theta}^{(\theta)} &= \sin[\theta \pm \rho(\omega_r,\theta)]\sin[\theta \pm \rho(\omega_s,\theta)] \ , \\ g_{M}^{(\theta)} &= \cos[\theta \pm \rho(\omega_r,\theta)]\cos[\theta \pm \rho(\omega_s,\theta)] \ , \\ g_{\Xi}^{(\theta)} &= \sin[\theta \pm \rho(\omega_r,\theta)]\cos[\theta \pm \rho(\omega_s,\theta)] \ , \\ g_{X}^{(\theta)} &= \cos[\theta \pm \rho(\omega_r,\theta)]\sin[\theta \pm \rho(\omega_s,\theta)] \ , \end{aligned} \tag{53}$$

with $-$ for the positive class and $+$ for the negative class. ω_r and ω_s are the pulsations of the two extraordinary waves. (ω_r,ω_s) is equal to (ω_4,ω_1) for SFM type $\mathrm{V}^4<0$, to (ω_4,ω_2) for SFM type $\mathrm{VI}^4<0$, to (ω_4,ω_3) for SFM type $\mathrm{VII}^4<0$, to (ω_2,ω_3) for SFM type $\mathrm{V}^4>0$, to (ω_1,ω_3) for the SFM type $\mathrm{VI}^4>0$, and to (ω_1,ω_2) for SFM type $\mathrm{VII}^4>0$.

The relationship with DFM can be done according to Table II. The correspondence between functions (52) and field factors is given in Table VII for SFM and from this table, relation (5) and Table II for the correspondence with DFM.

The principle of designation of the trigonometric functions is the same as for $3oe$ and $3eo$ interactions. Thus, for $2o2e$ interactions, two functions N and N' are equal and correspond to field factors which are symmetric in the Cartesian indices x and y relative to the two ordinary waves or the two extraordinary waves.

(h) The $2o2e$ trigonometric functions involved in the tensorial contraction of $\mathbf{F}^{(3)}$ and $\chi^{(3)}$ are given in Table V. At the opposite of $3oe$ and $3eo$ interactions, all the crystal classes allow $2o2e$ interactions.

We give the example of collinear type V^4 THG 1.064 μm$\rightarrow$0.355 μm in BBO. The phase-matching directions are located at $\theta=46.91°$ for any ϕ, according to the refractive indices (39). The intervening field factors are plotted in Fig. 3.

The particular interactions which we have chosen for BBO show the interest of $\mathbf{F}^{(3)}$ for the study of $\chi^{(3)}$. For the crystal class $3m$ and under Kleinman's symmetry conjecture, the relations between the χ_{ijkl} coefficients are

TABLE VII. Correspondence between $2o2e$ uniaxial functions and field factors according to the six corresponding types of phase-matched SFM(ω_4).

Interactions	Optical sign Uniaxial Biaxial	1	1′	2a	2′a	2b	2′b	$\bar{2}a$	$\bar{2}'a$	$\bar{2}b$	$\bar{2}'b$	3a	3b	$\bar{3}a$	$\bar{3}b$
		= ≅		= ≅		= ≅		= ≅		= ≅					
Type V^4	>0	*xyzz*	*yxzz*	*yxxz*	*xyxz*	*yxzx*	*xyzx*	*xyyz*	*yxyz*	*xyzy*	*yxzy*	*yyyz*	*yyzy*	*xxxz*	*xxzx*
Type VI^4	>0	*xzyz*	*yzxz*	*yxxz*	*xxyz*	*yzxx*	*xzyx*	*xyyz*	*yyxz*	*xzyy*	*yzxy*	*yyyz*	*yzyy*	*xxxz*	*xzxx*
Type VII^4	>0	*xzzy*	*yzzx*	*yxzx*	*xxzy*	*yzxx*	*xzxy*	*xyzy*	*yyzx*	*xzyy*	*yzyx*	*yyzy*	*yzyy*	*xxzx*	*xzxx*
Type V^4	<0	*zzxy*	*zzyx*	*zxyx*	*zxxy*	*xzyx*	*xzxy*	*zyxy*	*zyyx*	*yzxy*	*yzyx*	*zyyy*	*yzyy*	*zxxx*	*xzxx*
Type VI^4	<0	*zxzy*	*zyzx*	*zyxx*	*zxxy*	*xyzx*	*xxzy*	*zxyy*	*zyyx*	*yxzy*	*yyzx*	*zyyy*	*yyzy*	*zxxx*	*xxzx*
Type VII^4	<0	*zxyz*	*zyxz*	*zyxx*	*zxyx*	*xyxz*	*xxyz*	*zxyy*	*zyxy*	*yxyz*	*yyxz*	*zyyy*	*yyyz*	*zxxx*	*xxxz*

Interactions	Optical sign Uniaxial Biaxial	4	$\bar{4}$	5a	$\bar{5}a$	5b	$\bar{5}b$	6	6′	$\bar{6}$	$\bar{6}'$	7	7′	$\bar{7}$	$\bar{7}'$
								= ≅		= ≅		= ≅		= ≅	
Type V^4	>0	*xxzz*	*yyzz*	*yyxz*	*xxyz*	*yyzx*	*xxzy*	*xxxy*	*xxyx*	*yyyx*	*yyxy*	*xyyy*	*yxyy*	*yxxx*	*xyxx*
Type VI^4	>0	*xzxz*	*yzyz*	*yxyz*	*xyxz*	*yzyx*	*xzxy*	*xxxy*	*xyxx*	*yyyx*	*yxyy*	*xyyy*	*yyxy*	*yxxx*	*xxyx*
Type VII^4	>0	*xzzx*	*yzzy*	*yxzy*	*xyzx*	*yzxy*	*xzyx*	*xxyx*	*xyxx*	*yyxy*	*yxyy*	*xyyy*	*yyyx*	*yxxx*	*xxxy*
Type V^4	<0	*zzxx*	*zzyy*	*zxyy*	*zyxx*	*xzyy*	*yzxx*	*yxxx*	*xyxx*	*xyyy*	*yxyy*	*yyxy*	*yyyx*	*xxyx*	*xxxy*
Type VI^4	<0	*zxzx*	*zyzy*	*zyxy*	*zxyx*	*xyzy*	*yxzx*	*yxxx*	*xxyx*	*xyyy*	*yyxy*	*yxyy*	*yyyx*	*xyxx*	*xxxy*
Type VII^4	<0	*zxxz*	*zyyz*	*zyyx*	*zxxy*	*xyyz*	*yxxz*	*yxxx*	*xxxy*	*xyyy*	*yyyx*	*yxyy*	*yyxy*	*xyxx*	*xxyx*

Interactions	Optical sign Uniaxial Biaxial	8	$\bar{8}$	9	$\bar{9}$	10	10′	$\overline{10}$	$\overline{10}'$
				= ≅		= ≅		= ≅	
Type V^4	>0	*xxyy*	*yyxx*	*xxxx*	*yyyy*	*xyxy*	*xyyx*	*yxyx*	*yxxy*
Type VI^4	>0	*xyxy*	*yxyx*	*xxxx*	*yyyy*	*xxyy*	*xyyx*	*yyxx*	*yxxy*
Type VII^4	>0	*xyyx*	*yxxy*	*xxxx*	*yyyy*	*xxyy*	*xyxy*	*yyxx*	*yxyx*
Type V^4	<0	*yyxx*	*xxyy*	*xxxx*	*yyyy*	*yxxy*	*xyxy*	*xyyx*	*yxyx*
Type VI^4	<0	*yxyx*	*xyxy*	*xxxx*	*yyyy*	*yxxy*	*xxyy*	*xyyx*	*yyxx*
Type VII^4	<0	*yxxy*	*xyyx*	*xxxx*	*yyyy*	*yxyx*	*xxyy*	*xyxy*	*yyxx*

$$
\begin{aligned}
&xxxx=yyyy=xxyy+xyxy+xyyx, xxzz=xzxz=xzzx=yyzz=yzyz=yzzy=zyyz=zyzy=zzyy\\
&=zxxz=zxzx=zzxx, xxyy=xyxy=xyyx=yxxy=yxyx=yyxx, xxyz=xxzy=xyxz\\
&=xyzx=xzxy=xzyx=-yyyz=-yyzy=-yzyy=yxxz=yxzx=yzxx\\
&=-zyyy=zxxy=zxyx=zyxx, zzzz\ .
\end{aligned} \tag{54}
$$

There are five independent coefficients according to (54). The curves of Figs. 1–3 allow the judicious choice of phase-matching directions for the magnitude determination of the four useful coefficients at 0.355 μm by THG efficiency measurements (χ_{zzzz} is never solicited because $F_{zzzz}=0$ in all cases). All the measurements can be done in the x-z and y-z principal planes. In the undepleted pump approximation, we have:

$$\chi_{zyyy}=\left[\eta_{x\text{-}z}^{\mathrm{I},3oe}\right]^{1/2}\Big/B_{x\text{-}z}^{\mathrm{I},3oe}F_{zyyy}\ , \tag{55}$$

$$\chi_{xxzy}=\left[\eta_{x\text{-}z}^{\mathrm{II},3eo}\right]^{1/2}\Big/B_{x\text{-}z}^{\mathrm{II},3eo}(F_{zxxy}+F_{xxzy}+F_{xzxy})\ . \tag{56}$$

From (55) and (56), it is possible to solve the following system for the determination of χ_{yyxx} and χ_{zzxx}:

$$
\begin{aligned}
\left[\eta_{y\text{-}z}^{\mathrm{V}^4,2o2e}\right]^{1/2}&=B_{y\text{-}z}^{\mathrm{V}^4,2o2e}[\chi_{xxzy}(F_{zyxx}+F_{yzxx})\\
&\qquad+\chi_{yyxx}F_{yyxx}+\chi_{zzxx}F_{zzxx}]\ ,\\
\left[\eta_{x\text{-}z}^{\mathrm{V}^4,2o2e}\right]^{1/2}&=B_{x\text{-}z}^{\mathrm{V}^4,2oe}(\chi_{yyxx}F_{xxyy}+\chi_{zzxx}F_{zzyy})\ .
\end{aligned} \tag{57}
$$

η is the THG efficiency and B is a factor which depends on the refractive indices and on the fundamental beam parameters [11].

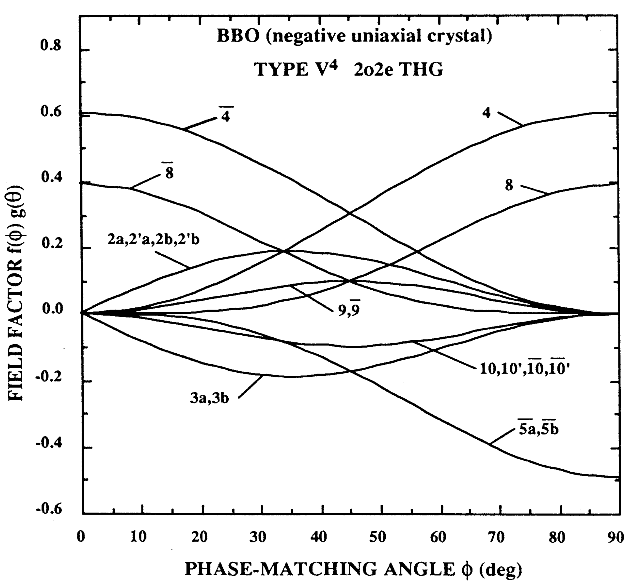


FIG. 3. Intervening $2o2e$ uniaxial field-factor functions vs the phase-matching spherical coordinate ϕ, calculated for direct type-V^4 THG 1.064→0.355 μm in BBO (crystal class C_{3v}).

IV. BIAXIAL CLASSES

In a biaxial crystal, the three principal refractive indices n_x, n_y, and n_z are different. The equations of the two sheets of the indices surface $n^+(\theta,\phi)$ and $n^-(\theta,\phi)$ are given by Eqs. (8) and (9), respectively, as a function of the direction of propagation (θ,ϕ). The graphical representations of the indices surfaces are given in Fig. 4 for the positive biaxial class ($n_x<n_y<n_z$) and for the negative one ($n_x>n_y>n_z$) [4]. These conventional cases are representative of all the possible situations with the appropriate permutation of the principal refractive indices. There are two directions, contained in the x-z plane, for which the two refractive indices n^+ and n^- are equal; this defines the two optical axes.

As for uniaxial classes, the phase-matching directions of the seven phase-matching relations are calculated from (8), (9), and (14) and correspond to the intersection of the internal sheet at ω_4 and a combination of the internal and external sheets at ω_1, ω_2, and ω_3 according to Table I.

We give in Table VIII the inequalities between refractive indices which determine collinear phase matching in the principal planes of biaxial crystals according to the optical sign of the class and according to the different situations of birefringence. These conditions are established on the same bases we used in a previous paper devoted to the complete study of phase-matching conditions of three-wave collinear SFM and DFM [12]. The inequalities written a, b, c, and d in Table VIII correspond to phase-matching directions in specific areas of the principal planes according to the principal axes and to the optical axes. The areas are defined in Table VIII. The existence of a phase-matching cone joining two of these areas depends on the type of interaction and on

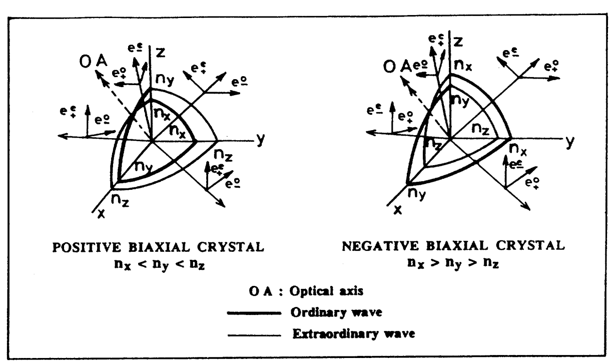


FIG. 4. Index surfaces of the positive and negative biaxial classes. $\mathbf{e}^{o,e}_{+,-}$ are the ordinary (o) and extraordinary (e) electric-field vectors relative to the external (+) or internal (−) sheets for propagation in the principal planes.

TABLE VIII. Inequalities between refractive indices determining the collinear phase-matching directions in the principal planes of biaxial crystals according to the seven types of phase-matched SFM(ω_4). (n_{xi}, n_{yi}, n_{zi}) are the principal refractive indices at the wavelength λ_i $(i=1,2,3,4)$. The areas *a*, *b*, *c*, and *d* are defined as following: *a*, between the *z* axis and the optical axis of smallest angle θ; *b*, between the optical axis of greatest angle θ and the *x* axis; *c*, between the *x* axis and the *y* axis; *d*, between the *y* axis and the *z* axis.

Phase-matching directions in the principal planes	Inequalities determining four-wave collinear phase matching in biaxial crystals: Positive sign	Negative sign
	SFM Type I	
a	$\frac{n_{x4}}{\lambda_4} < \frac{n_{y1}}{\lambda_1} + \frac{n_{y2}}{\lambda_2} + \frac{n_{y3}}{\lambda_3} < \frac{n_{z4}}{\lambda_4}$	$\frac{n_{z1}}{\lambda_1} + \frac{n_{z2}}{\lambda_2} + \frac{n_{z3}}{\lambda_3} < \frac{n_{y4}}{\lambda_4} < \frac{n_{x1}}{\lambda_1} + \frac{n_{x2}}{\lambda_2} + \frac{n_{x3}}{\lambda_3}$
b	$\frac{n_{x1}}{\lambda_1} + \frac{n_{x2}}{\lambda_2} + \frac{n_{x3}}{\lambda_3} < \frac{n_{y4}}{\lambda_4} < \frac{n_{z1}}{\lambda_1} + \frac{n_{z2}}{\lambda_2} + \frac{n_{z3}}{\lambda_3}$	$\frac{n_{z4}}{\lambda_4} < \frac{n_{y1}}{\lambda_1} + \frac{n_{y2}}{\lambda_2} + \frac{n_{y3}}{\lambda_3} < \frac{n_{x4}}{\lambda_4}$
c	$\frac{n_{x4}}{\lambda_4} < \frac{n_{z1}}{\lambda_1} + \frac{n_{z2}}{\lambda_2} + \frac{n_{z3}}{\lambda_3} < \frac{n_{y4}}{\lambda_4}$	$\frac{n_{y1}}{\lambda_1} + \frac{n_{y2}}{\lambda_2} + \frac{n_{y3}}{\lambda_3} < \frac{n_{z4}}{\lambda_4} < \frac{n_{x1}}{\lambda_1} + \frac{n_{x2}}{\lambda_2} + \frac{n_{x3}}{\lambda_3}$
d	$\frac{n_{y1}}{\lambda_1} + \frac{n_{y2}}{\lambda_2} + \frac{n_{y3}}{\lambda_3} < \frac{n_{x4}}{\lambda_4} < \frac{n_{z1}}{\lambda_1} + \frac{n_{z2}}{\lambda_2} + \frac{n_{z3}}{\lambda_3}$	$\frac{n_{z4}}{\lambda_4} < \frac{n_{x1}}{\lambda_1} + \frac{n_{x2}}{\lambda_2} + \frac{n_{x3}}{\lambda_3} < \frac{n_{y4}}{\lambda_4}$
	SFM Type II $(i=1,j=2,k=3)$, SFM Type III $(i=3,j=1,k=2)$, and SFM Type IV $(i=2,j=3,k=1)$	
a	$\frac{n_{x4}}{\lambda_4} < \frac{n_{xi}}{\lambda_i} + \frac{n_{xi}}{\lambda_j} + \frac{n_{yk}}{\lambda_k}\ ;\ \frac{n_{zi}}{\lambda_i} + \frac{n_{zj}}{\lambda_j} + \frac{n_{yk}}{\lambda_k} < \frac{n_{z4}}{\lambda_4}$	$\frac{n_{yi}}{\lambda_i} + \frac{n_{yj}}{\lambda_j} + \frac{n_{zk}}{\lambda_k} < \frac{n_{y4}}{\lambda_4} < \frac{n_{yi}}{\lambda_i} + \frac{n_{yj}}{\lambda_j} + \frac{n_{xk}}{\lambda_k}$
b	$\frac{n_{yi}}{\lambda_i} + \frac{n_{yj}}{\lambda_j} + \frac{n_{xk}}{\lambda_k} < \frac{n_{y4}}{\lambda_4} < \frac{n_{yi}}{\lambda_i} + \frac{n_{yj}}{\lambda_j} + \frac{n_{zk}}{\lambda_k}$	$\frac{n_{z4}}{\lambda_4} < \frac{n_{zi}}{\lambda_i} + \frac{n_{zj}}{\lambda_j} + \frac{n_{yk}}{\lambda_k}\ ;\ \frac{n_{xi}}{\lambda_i} + \frac{n_{xj}}{\lambda_j} + \frac{n_{yk}}{\lambda_k} < \frac{n_{x4}}{\lambda_4}$
c	$\frac{n_{x4}}{\lambda_4} < \frac{n_{xi}}{\lambda_i} + \frac{n_{xj}}{\lambda_j} + \frac{n_{zk}}{\lambda_k}\ ;\ \frac{n_{yi}}{\lambda_i} + \frac{n_{yj}}{\lambda_j} + \frac{n_{zk}}{\lambda_k} < \frac{n_{y4}}{\lambda_4}$	$\frac{n_{zi}}{\lambda_i} + \frac{n_{zj}}{\lambda_j} + \frac{n_{yk}}{\lambda_k} < \frac{n_{z4}}{\lambda_4} < \frac{n_{zi}}{\lambda_i} + \frac{n_{zj}}{\lambda_j} + \frac{n_{xk}}{\lambda_k}$
*c**	$\frac{n_{xi}}{\lambda_i} + \frac{n_{xj}}{\lambda_j} + \frac{n_{zk}}{\lambda_k} < \frac{n_{x4}}{\lambda_4};\ \frac{n_{y4}}{\lambda_4} < \frac{n_{yi}}{\lambda_i} + \frac{n_{yj}}{\lambda_j} + \frac{n_{zk}}{\lambda_k}$	$\frac{n_{zi}}{\lambda_i} + \frac{n_{zj}}{\lambda_j} + \frac{n_{yk}}{\lambda_k} < \frac{n_{z4}}{\lambda_4} < \frac{n_{zi}}{\lambda_i} + \frac{n_{zj}}{\lambda_j} + \frac{n_{xk}}{\lambda_k}$
d	$\frac{n_{xi}}{\lambda_i} + \frac{n_{xj}}{\lambda_j} + \frac{n_{yk}}{\lambda_k} < \frac{n_{x4}}{\lambda_4} < \frac{n_{xi}}{\lambda_i} + \frac{n_{xj}}{\lambda_j} + \frac{n_{zk}}{\lambda_k}$	$\frac{n_{z4}}{\lambda_4} < \frac{n_{zi}}{\lambda_i} + \frac{n_{zj}}{\lambda_j} + \frac{n_{xk}}{\lambda_k}\ ;\ \frac{n_{yi}}{\lambda_i} + \frac{n_{yj}}{\lambda_j} + \frac{n_{xk}}{\lambda_k} < \frac{n_{y4}}{\lambda_4}$
*d**	$\frac{n_{xi}}{\lambda_i} + \frac{n_{xj}}{\lambda_j} + \frac{n_{yk}}{\lambda_k} < \frac{n_{x4}}{\lambda_4} < \frac{n_{xi}}{\lambda_i} + \frac{n_{xj}}{\lambda_j} + \frac{n_{zk}}{\lambda_k}$	$\frac{n_{zi}}{\lambda_i} + \frac{n_{zj}}{\lambda_j} + \frac{n_{xk}}{\lambda_k} < \frac{n_{z4}}{\lambda_4}\ ;\ \frac{n_{y4}}{\lambda_4} < \frac{n_{yi}}{\lambda_i} + \frac{n_{yj}}{\lambda_j} + \frac{n_{xk}}{\lambda_k}$
	SFM Type V[4] $(i=1,j=2,k=3)$, SFM Type VI[4] $(i=2,j=3,k=1)$, and SFM Type VII[4] $(i=3,j=1,k=2)$	
a	$\frac{n_{x4}}{\lambda_4} < \frac{n_{xi}}{\lambda_i} + \frac{n_{yj}}{\lambda_j} + \frac{n_{yk}}{\lambda_k}\ ;\ \frac{n_{zi}}{\lambda_i} + \frac{n_{yj}}{\lambda_j} + \frac{n_{yk}}{\lambda_k} < \frac{n_{z4}}{\lambda_4}$	$\frac{n_{yi}}{\lambda_i} + \frac{n_{zj}}{\lambda_j} + \frac{n_{zk}}{\lambda_k} < \frac{n_{y4}}{\lambda_4} < \frac{n_{yi}}{\lambda_i} + \frac{n_{xj}}{\lambda_j} + \frac{n_{xk}}{\lambda_k}$
b	$\frac{n_{yi}}{\lambda_i} + \frac{n_{xj}}{\lambda_j} + \frac{n_{xk}}{\lambda_k} < \frac{n_{y4}}{\lambda_4} < \frac{n_{yi}}{\lambda_i} + \frac{n_{zj}}{\lambda_j} + \frac{n_{zk}}{\lambda_k}$	$\frac{n_{z4}}{\lambda_4} < \frac{n_{zi}}{\lambda_i} + \frac{n_{yj}}{\lambda_j} + \frac{n_{yk}}{\lambda_k}\ ;\ \frac{n_{xi}}{\lambda_i} + \frac{n_{yj}}{\lambda_j} + \frac{n_{yk}}{\lambda_k} < \frac{n_{x4}}{\lambda_4}$
c'	$\frac{n_{x4}}{\lambda_4} < \frac{n_{xi}}{\lambda_i} + \frac{n_{zj}}{\lambda_j} + \frac{n_{zk}}{\lambda_k}\ ;\ \frac{n_{yi}}{\lambda_i} + \frac{n_{zj}}{\lambda_j} + \frac{n_{zk}}{\lambda_k} < \frac{n_{y4}}{\lambda_4}$	$\frac{n_{zi}}{\lambda_i} + \frac{n_{yj}}{\lambda_j} + \frac{n_{yk}}{\lambda_k} < \frac{n_{z4}}{\lambda_4} < \frac{n_{zi}}{\lambda_i} + \frac{n_{xj}}{\lambda_j} + \frac{n_{xk}}{\lambda_k}$
*c***	$\frac{n_{xi}}{\lambda_i} + \frac{n_{zj}}{\lambda_j} + \frac{n_{zk}}{\lambda_k} < \frac{n_{x4}}{\lambda_4}\ ;\ \frac{n_{y4}}{\lambda_4} < \frac{n_{yi}}{\lambda_i} + \frac{n_{zj}}{\lambda_j} + \frac{n_{zk}}{\lambda_k}$	$\frac{n_{zi}}{\lambda_i} + \frac{n_{yj}}{\lambda_j} + \frac{n_{yk}}{\lambda_k} < \frac{n_{z4}}{\lambda_4} < \frac{n_{zi}}{\lambda_i} + \frac{n_{xj}}{\lambda_j} + \frac{n_{xk}}{\lambda_k}$

TABLE VIII. (*Continued*).

Phase-matching directions in the principal planes	Inequalities determining four-wave collinear phase matching in biaxial crystals	
	Positive sign	Negative sign
d'	$\frac{n_{xi}}{\lambda_i}+\frac{n_{yj}}{\lambda_j}+\frac{n_{yk}}{\lambda_k}<\frac{n_{x4}}{\lambda_4}<\frac{n_{xi}}{\lambda_i}+\frac{n_{zj}}{\lambda_j}+\frac{n_{zk}}{\lambda_k}$	$\frac{n_{z4}}{\lambda_4}<\frac{n_{zi}}{\lambda_i}+\frac{n_{xj}}{\lambda_j}+\frac{n_{xk}}{\lambda_k}$; $\frac{n_{yi}}{\lambda_i}+\frac{n_{xj}}{\lambda_j}+\frac{n_{xk}}{\lambda_k}<\frac{n_{y4}}{\lambda_4}$
d^{**}	$\frac{n_{xi}}{\lambda_i}+\frac{n_{yj}}{\lambda_j}+\frac{n_{yk}}{\lambda_k}<\frac{n_{x4}}{\lambda_4}<\frac{n_{xi}}{\lambda_i}+\frac{n_{zj}}{\lambda_j}+\frac{n_{zk}}{\lambda_k}$	$\frac{n_{zi}}{\lambda_i}+\frac{n_{xj}}{\lambda_j}+\frac{n_{xk}}{\lambda_k}<\frac{n_{z4}}{\lambda_4}$; $\frac{n_{y4}}{\lambda_4}<\frac{n_{yi}}{\lambda_i}+\frac{n_{xj}}{\lambda_j}+\frac{n_{xk}}{\lambda_k}$
	SFM Type II $(i,j)=(1,2)$, SFM Type III $(i,j)=(1,3)$, and SFM Type IV $(i,j)=(2,3)$	
Conditions c,d are applied if	$\frac{n_{yi}}{\lambda_i}+\frac{n_{yj}}{\lambda_j}-\frac{n_{xi}}{\lambda_i}-\frac{n_{xj}}{\lambda_j}<\frac{n_{y4}}{\lambda_4}-\frac{n_{x4}}{\lambda_4}$	$\frac{n_{yi}}{\lambda_i}+\frac{n_{yj}}{\lambda_j}-\frac{n_{zi}}{\lambda_i}-\frac{n_{zj}}{\lambda_j}<\frac{n_{y4}}{\lambda_4}-\frac{n_{z4}}{\lambda_4}$
Conditions c^*,d^* are applied if	$\frac{n_{y4}}{\lambda_4}-\frac{n_{x4}}{\lambda_4}<\frac{n_{yi}}{\lambda_i}+\frac{n_{yj}}{\lambda_j}-\frac{n_{xi}}{\lambda_i}-\frac{n_{xj}}{\lambda_j}$	$\frac{n_{y4}}{\lambda_4}-\frac{n_{z4}}{\lambda_4}<\frac{n_{yi}}{\lambda_i}+\frac{n_{yj}}{\lambda_j}-\frac{n_{zi}}{\lambda_i}-\frac{n_{zj}}{\lambda_j}$
	SFM Type V^4 $(i=1)$, SFM Type VI^4 $(i=2)$, and SFM Type VII^4 $(i=3)$	
Conditions c',d' are applied if	$\frac{n_{yi}}{\lambda_i}-\frac{n_{xi}}{\lambda_i}<\frac{n_{y4}}{\lambda_4}-\frac{n_{x4}}{\lambda_4}$	$\frac{n_{yi}}{\lambda_i}-\frac{n_{zi}}{\lambda_i}<\frac{n_{y4}}{\lambda_4}-\frac{n_{z4}}{\lambda_4}$
Conditions c^{**},d^{**} are applied if	$\frac{n_{y4}}{\lambda_4}-\frac{n_{x4}}{\lambda_4}<\frac{n_{yi}}{\lambda_i}-\frac{n_{xi}}{\lambda_i}$	$\frac{n_{y4}}{\lambda_4}-\frac{n_{z4}}{\lambda_4}<\frac{n_{yi}}{\lambda_i}-\frac{n_{zi}}{\lambda_i}$

birefringence. The cones joining a and b, a and c, b and d, and c and d (inequalities c and d for SFM types I, II, III, and IV, inequalities c' and d' for SFM types V^4, VI^4, and VII^4) are possible for the seven types of interaction. Except for type I, cones which join b and c for the positive class and a and d for the negative class are also allowed (inequalities c^* and d^* for SFM types II, III, and IV, inequalities c^{**} and d^{**} for SFM types V^4, VI^4, and VII^4). The dispersion in frequency of the refractive indices forbid collinear phase matching for all directions located between the four optical axes even if their dispersion in frequency is high. The counting of the possible cases of coexistence of the different cones is not done in this paper.

It is impossible to define ordinary and extraordinary waves out of the principal planes of a biaxial crystal. The two electric-field vectors $\mathbf{e}^+$ and $\mathbf{e}^-$ have a nonzero component along the z axis. They are calculated from the equation of propagation projected on the three axes x, y, and z of the optical frame [7,8]:

$$\frac{e_p^{+,-}}{(n^{+,-})^2}=\frac{u_p(u_x e_x^{+,-}+u_y e_y^{+,-}+u_z e_z^{+,-})}{(n^{+,-})^2-(n_p)^2}, \quad (58)$$

with $p=x$, y, and z and $\|\mathbf{e}^{+,-}\|=1$.

According to (58), $\mathbf{e}^+$ and $\mathbf{e}^-$ are not perpendicular: the walkoff angles of the two waves are nonzero and different. The field tensor relations of orthogonality (20) which exist for uniaxial crystals are not valid for biaxial ones. Furthermore, according to (58), there is a rotation of 90° of the electric-field vectors $\mathbf{e}^+$ and $\mathbf{e}^-$ from the directions b or d to a for phase-matching cones a-b and a-d; thus an ordinary wave becomes an extraordinary one and vice versa.

As for uniaxial crystals, it is possible to define ordinary and extraordinary waves in the principal planes of a biaxial crystal: the ordinary electric-field vector is perpendicular to the z axis and to the extraordinary one; relation (20) is satisfied. The electric fields are represented in Fig. 4 for a propagation in the principal planes.

For a propagation in the x-y plane (area c), the ordinary electric-field vector has a nonzero walkoff angle and the extraordinary walkoff angle is nil:

$$e_x^o=-\sin[\phi\pm\rho(\phi,\omega)]\ ,$$
$$e_y^o=\cos[\phi\pm\rho(\phi,\omega)]\ ,\quad e_z^o=0\ , \quad (59)$$
$$e_x^e=0\ ,\quad e_y^e=0\ ,\quad e_z^e=1\ , \quad (60)$$

with + for the positive class and − for the negative class. $\rho(\phi,\omega)$ is the walkoff angle of the ordinary wave, given by (19) with $n_a=n_y$ and $n_b=n_x$.

Note that in the x-y plane of an uniaxial crystal, the extraordinary and ordinary waves have a nil walkoff angle for all direction of propagation according to (17)–(19). The optical sign in the x-y plane is defined by the sign of the birefringence n_z-$n_{ba}(\phi)$, where $n_{ba}(\phi)$ is given by

$$n_{ba}(\phi)=[\cos^2(\phi)/n_a^2+\sin^2(\phi)/n_b^2]^{-1/2}\ . \quad (61)$$

$n_{ba}=n_{yx}$, $n_a=n_y$, and $n_b=n_x$ in the x-y plane.

For a propagation in the y-z plane, the components of the electric-field vectors are the same as for the uniaxial class; they are given by (17) and (18) with $\phi=90°$. The ordinary walkoff angle is nil and the extraordinary one is given by (19) with $n_a=n_y$ and $n_b=n_z$.

Thus the y-z plane of a biaxial crystal has exactly the same characteristics for the optical propagation than any plane containing the optical axis (z axis) of an uniaxial crystal.

The optical sign is defined by the sign of the birefringence $n_{ba}(\theta)-n_x$, where $n_{ba}(\theta)$ is given by (61), with $n_{ba}(\theta)=n_{zy}(\theta)$, $n_a=n_y$, and $n_b=n_z$.

In the x-z plane, the optical axis creates discontinuity of the optical sign and discontinuity of the ellipticity of the external and internal sheets of the indices surface according to Fig. 4. The optical sign is defined by the sign of the birefringence $n_{ba}(\theta)$-n_y, where $n_{ba}(\theta)$ is given by (61) with $n_{ba}(\theta)=n_{zx}(\theta)$, $n_a=n_x$, and $n_b=n_z$.

The birefringence is nil along the optical axis and its sign changes on either side. The optical sign is so the same for all directions of propagation contained is the y-z plane, the x-y plane, and in the x-z plane from the x axis to the optical axis. Thus a positive biaxial crystal is negative in the x-z plane from the optical axis to the z axis; the situation is inverted for a negative biaxial crystal.

According to Fig. 4, the phase-matching directions a have an optical sign different from all the others. Then we call cones of type B the phase-matching cones a-b, a-d and a-c. The other cones b-c, b-d, and c-d are of type A [4].

For a phase-matching direction contained from the x axis to the optical axis (area b), the electric-field components are given by (17) and (18) with $\phi=0°$. The extraordinary walkoff angle is given by (19), where $n_a=n_x$ and $n_b=n_z$.

According to (58), the electric-field vectors for a direction of propagation contained from the optical axis to the z axis (area a) can be obtained by a rotation of 90° of $\mathbf{e}^o$ and $\mathbf{e}^e$ associated to a propagation in area b or area d. Then, according to (17), the extraordinary electric-field vector is given by (18) with $\phi=0°$ and the ordinary one is out of phase by 180° in relation to (17) according to (18), that is,

$$e_x^o=0, \quad e_y^o=-1, \quad e_z^o=0 \ . \tag{62}$$

The extraordinary walkoff angle is given by (19), with $n_a=n_x$ and $n_b=n_z$. Thus the symmetry of the field tensor of all phase-matching directions in the principal planes of a biaxial crystal is exactly the same as for any phase-matching direction in an uniaxial crystal. All the considerations developed in Sec. III are also suitable to the study of the principal planes of biaxial crystals. Out of the principal planes, the field tensors are less symmetric than for a propagation in the principal planes because of the nonperpendicularity of the two eigenmodes $\mathbf{e}^+$ and $\mathbf{e}^-$; thus the field tensors have 81 nonzero and independent elements in the general case. Out of the principal planes, the only possible symmetries are due to equalities between frequencies: the field tensors are symmetric in the Cartesian indices relative to electric-field vectors of same eigenmode at the same pulsation or at different pulsations if the dispersion in frequency of the walkoff angle is negligible.

We keep the designation of $3oe$, $3eo$, and $2o2e$ for the phase-matching cone of type A. We denote by $3oe$-$3eo$, $3eo$-$3oe$, and $2o2e$-$2e2o$ the configurations of polarization for the phase-matching cone of type B in order to show the change of polarization on either side of the optical axis; for example, a $3oe$ interaction for a propagation in area b, c, or d is $3eo$ in area a.

A. Cone of type A. Comparison with the uniaxial class

We take the example of $KTiOPO_4$ (KTP), which is a positive biaxial crystal belonging to the crystal class $C_{2v}(mm2)$. We consider the collinear phase-matched SFM (1/0.61 μm=1/3.0 μm+1/2.73 μm+1/1.064 μm). The corresponding refractive indices are the following [13]:

$$\begin{aligned} &n_x=1.7004, \quad n_y=1.7062, \quad n_z=1.7800 \quad \text{at } \lambda_1=3\ \mu\text{m}\ , \\ &n_x=1.7067, \quad n_y=1.7129, \quad n_z=1.7876 \\ &\qquad\qquad\qquad\qquad \text{at } \lambda_2=2.73\ \mu\text{m}\ , \\ &n_x=1.7399, \quad n_y=1.7480, \quad n_z=1.8296 \\ &\qquad\qquad\qquad\qquad \text{at } \lambda_3=1.064\ \mu\text{m}\ , \\ &n_x=1.7663, \quad n_y=1.7763, \quad n_z=1.8675 \\ &\qquad\qquad\qquad\qquad \text{at } \lambda_4=0.61\ \mu\text{m}\ . \end{aligned} \tag{63}$$

KTP is a quasiuniaxial crystal since n_x is close to n_y compared to n_z. We consider the SFM type II $3oe$, type I $3eo$, and type V[4] $2o2e$. The corresponding phase-matching directions calculated according to Table II and from (63) are located from ($\theta=78.71°,\phi=0°$) to

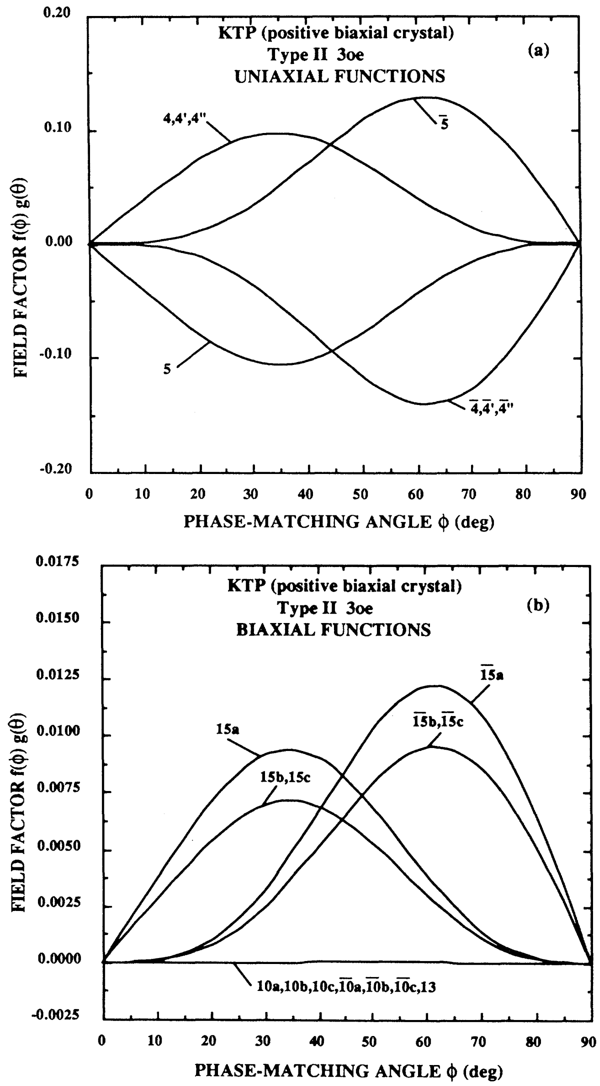


FIG. 5. Intervening $3oe$ uniaxial (a) and biaxial (b) field-factor functions vs the phase-matching spherical coordinate ϕ, calculated for type-II SFM $(1/0.61=1/3.0+1/2.73 +1/1.064\ \mu\text{m})$ in KTP (crystal class C_{2v}).

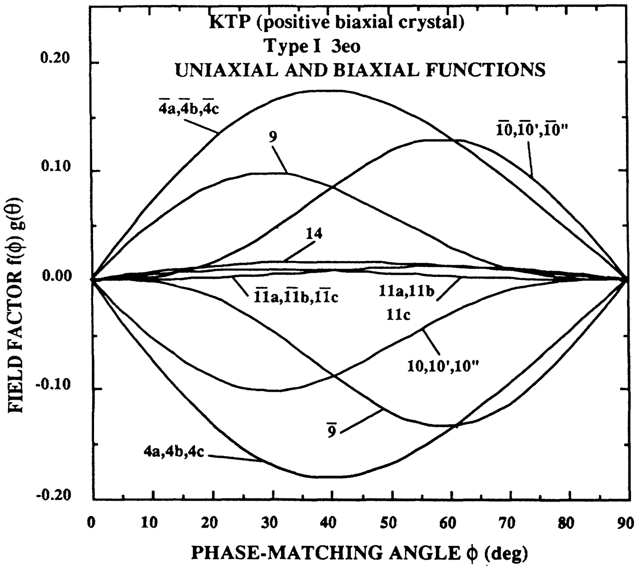


FIG. 6. Intervening 3*eo* uniaxial and biaxial field-factor functions vs the phase-matching spherical coordinate ϕ, calculated for type-I SFM $(1/0.61=1/3.0+1/2.73+1/1.064\ \mu m)$ in KTP (crystal class C_{2v}).

$(\theta=63.94°,\phi=90°)$, from $(\theta=52.09°,\phi=0°)$ to $(\theta=42.61°,\phi=90°)$, and from $(\theta=59.57°,\phi=0°)$ to $(\theta=49.68°,\phi=90°)$, respectively. The calculated intervening field factors are plotted as a function of the spherical coordinate ϕ of each phase-matching direction in Figs. 5(a) and 5(b) for 3*oe*, Fig. 6 for 3*eo*, and Fig. 7(a) and 7(b) for 2*o*2*e*. The correspondences between trigonometric functions and field factors are given in Tables IV and IX for 3*oe*, Tables VI and X for 3*eo*, and Tables VII and XI for 2*o*2*e*.

The functions of Figs. 5(a), 6, and 7(a) concern the same field factors as the nonzero elements of the corresponding interactions in uniaxial crystals and are the so-called uniaxial functions. The functions N, N', and N'' are now different.

The uniaxial functions of the biaxial class are all the more similar to those of the uniaxial class (Figs. 1–3) since n_x approaches n_y. The functions of Figs. 5(b), 6, and 7(b) are specific to the biaxial class and are called biaxial functions. They are nil in the principal planes. Out of the principal planes, these functions are all the smaller since the biaxial crystal "tends" to a uniaxial one, that is, n_x approaches n_y. Thus the elements of the tensor $\chi^{(3)}$ of a quasiuniaxial biaxial crystal are weakly involved by the specific biaxial field factors. Nevertheless, their contribution can be non-negligible in comparison with those solicited by the uniaxial field factors, according to their relative sign.

B. Comparison between cones of types *A* and *B*

We consider the thiosemicarbazide cadmium chloride monohydrate (TSCCC), a new positive biaxial crystal which belongs to the crystal class $C_s(m)$. We show the difference between area *a* and area *d* by comparison between two type-I collinear phase-matched THG ($1.15\rightarrow0.38\ \mu m$) which allows a cone of type *A* and ($1.32\rightarrow0.44\ \mu m$) which allows a cone of type *B*. The intervening refractive indices are the following [14]:

$$\begin{aligned}
&n_x=1.6458,\quad n_y=1.7088,\quad n_z=1.7337, &&\text{at } \lambda_1=1.32\ \mu m\ ,\\
&n_x=1.6978,\quad n_y=1.7757,\quad n_z=1.8033 &&\text{at } \lambda_4=0.44\ \mu m\ ,\\
&n_x=1.6481,\quad n_y=1.7127,\quad n_z=1.7366 &&\text{at } \lambda_1=1.15\ \mu m\ ,\\
&n_x=1.7196,\quad n_y=1.8031,\quad n_z=1.8339 &&\text{at } \lambda_4=0.38\ \mu m\ .
\end{aligned}\tag{64}$$

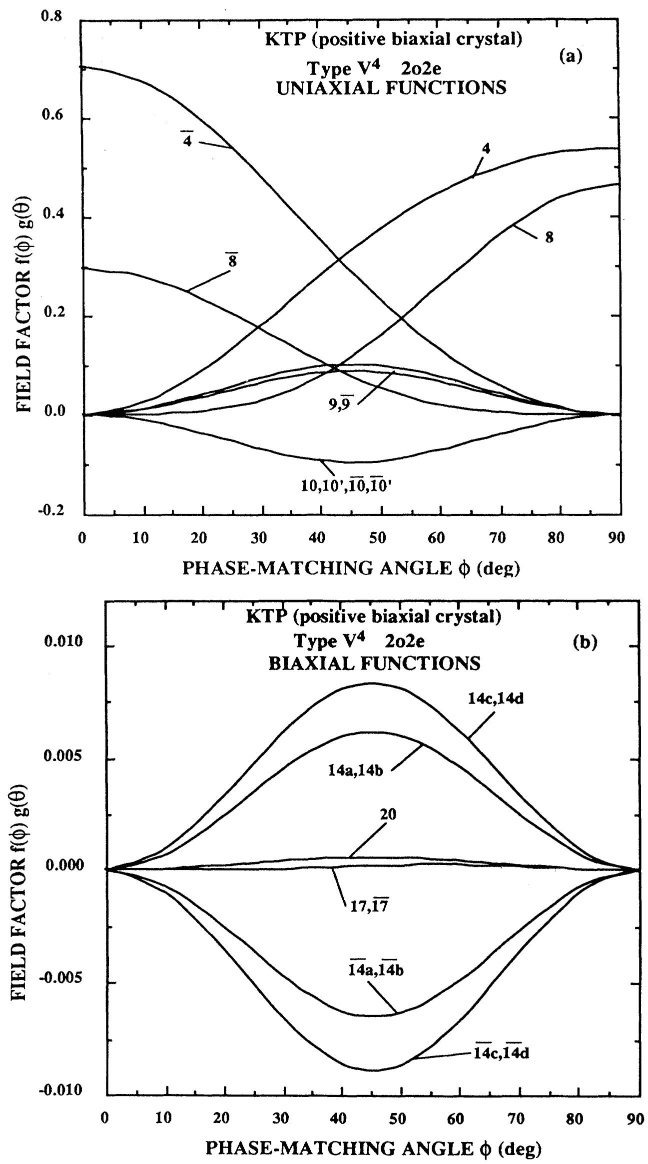


FIG. 7. Intervening 2*o*2*e* uniaxial (a) and biaxial (b) field-factor functions vs the phase-matching spherical coordinate ϕ, calculated for type-V[4] SFM $(1/0.61=1/3.0+1/2.73+1/1.064\ \mu m)$ in KTP (crystal class C_{2v}).

TABLE IX. Correspondence between 3*oe*, 3*oe*-3*eo* biaxial functions and field factors according to the four corresponding types of phase-matched SFM(ω_4).

Interactions	Optical sign	3*oe* and 3*oe*-3*eo* biaxial field-factor functions: 7	$\overline{7}$	8*a*	8*b*	8*c*	$\overline{8}a$	$\overline{8}b$	$\overline{8}c$	9*a*	9*b*	9*c*	$\overline{9}a$	$\overline{9}b$	$\overline{9}c$	10*a*	10*b*	10*c*	$\overline{10}a$	$\overline{10}b$	$\overline{10}c$
Type I	<0	yzzz	xzzz	xyzz	xzyz	xzzy	yxzz	yzxz	yzzx	xyyz	xyzy	xzyy	yxxz	yxzx	yzxx	yyzz	yzyz	yzzy	xxzz	xzxz	xzzx
Type II	>0	zzzy	zzzx	zyzx	zzyx	yzzx	zxzy	zzxy	xzzy	zyyx	yyzx	yzyx	zxxy	xxzy	xzxy	zyzy	zzyy	yzzy	zxzx	zzxx	xzzx
Type III	>0	zzyz	zzxz	zyxz	zzxy	yzxz	zxyz	zzyx	xzyz	zyxy	yyxz	yzxy	zxyx	xxyz	xzyx	zyyz	zzyy	yzyz	zxxz	zzxx	xzxz
Type IV	>0	zyzz	zxzz	zxyz	zxzy	yxzz	zyxz	zyzx	xyzz	zxyy	yxyz	yxzy	zyxx	xyxz	xyzx	zyyz	zyzy	yyzz	zxxz	zxzx	xxzz

Interactions	Optical sign	11*a*	11′*a*	11*b*	11′*b*	11*c*	11′*c*	$\overline{11}a$	$\overline{11}'a$	$\overline{11}b$	$\overline{11}'b$	$\overline{11}c$	$\overline{11}'c$	12*a*	12*b*	12*c*	$\overline{12}a$	$\overline{12}b$	$\overline{12}c$
Type I	<0	xyxz	xxyz	xyzx	xxzy	xzyx	xzxy	yxyz	yyxz	yxzy	yyzx	yzxy	yzyx	yyyz	yyzy	yzyy	xxxz	xxzx	xzxx
Type II	>0	zyxx	zxyx	xyzx	yxzx	xzyx	yzxx	zxyy	zyxy	yxzy	xyzy	yzxy	xzyy	zyyy	yyzy	yzyy	zxxx	xxzx	xzxx
Type III	>0	zyxx	zxxy	xyxz	yxxz	xzxy	yzxx	zxyy	zyyx	yxyz	xyyz	yzyx	xzyy	zyyy	yyyz	yzyy	zxxx	xxxz	xzxx
Type IV	>0	zxyx	zxxy	xxyz	yxxz	xxzy	yxzx	zyxy	zyyx	yyxz	xyyz	yyzx	xyzy	zyyy	yyyz	yyzy	zxxx	xxxz	xxzx

Interactions	Optical sign	13	14*a*	14*b*	14*c*	15*a*	15*b*	15*c*	$\overline{15}a$	$\overline{15}b$	$\overline{15}c$	16*a*	16*b*	16*c*	17*a*	17′*a*	17*b*	17′*b*	17*c*	17′*c*
Type I	<0	zzzz	zyzz	zzyz	zzzy	zyyz	zyzy	zzyy	zxxz	zxzx	zzxx	zxzz	zzxz	zzzx	zxyz	zyxz	zxzy	zyzx	zzxy	zzyx
Type II	>0	zzzz	zyzz	zzyz	yzzz	zyyz	yyzz	yzyz	zxxz	xxzz	xzxz	zxzz	zzxz	xzzz	zxyz	zyxz	yxzz	xyzz	yzxz	xzyz
Type III	>0	zzzz	zyzz	zzzy	yzzz	zyzy	yyzz	yzzy	zxzx	xxzz	xzzx	zxzz	zzzx	xzzz	zxzy	zyzx	yxzz	xyzz	yzzx	xzzy
Type IV	>0	zzzz	zzyz	zzzy	yzzz	zzyy	yzyz	yzzy	zzxx	xzxz	xzzx	zzxz	zzzx	xzzz	zzxy	zzyx	yzxz	xzyz	yzzx	xzzy

TABLE X. Correspondence between 3*eo*, 3*eo*-3*oe* biaxial functions and field factors according to the four corresponding types of phase-matched SFM(ω_4).

		3*eo* and 3*eo*-3*oe* biaxial field-factor functions																	
Interactions	Optical sign	11*a*	11*b*	11*c*	$\overline{11}a$	$\overline{11}b$	$\overline{11}c$	12*a*	12*b*	12*c*	12*a*′	12*b*′	12*c*′	13*a*	13*b*	13*c*	$\overline{13}a$	$\overline{13}b$	$\overline{13}c$	14
Type I	>0	*zxxz*	*zxzx*	*zzxx*	*zyyz*	*zyzy*	*zzyy*	*zxyz*	*zxzy*	*zzxy*	*zyxz*	*zyzx*	*zzyx*	*zxzz*	*zzxz*	*zzzx*	*zyzz*	*zzyz*	*zzzy*	*zzzz*
Type II	<0	*zxxz*	*xxzz*	*xzxz*	*zyyz*	*yyzz*	*yzyz*	*zxyz*	*yxzz*	*yzxz*	*zyxz*	*xyzz*	*xzyz*	*zxzz*	*zzxz*	*xzzz*	*zyzz*	*zzyz*	*yzzz*	*zzzz*
Type III	<0	*zxzx*	*xxzz*	*xzzx*	*zyzy*	*yyzz*	*yzzy*	*zxzy*	*yxzz*	*yzzx*	*zyzx*	*xyzz*	*xzzy*	*zxzz*	*zzzx*	*xzzz*	*zyzz*	*zzzy*	*yzzz*	*zzzz*
Type IV	<0	*zzxx*	*xzxz*	*xzzx*	*zzyy*	*yzyz*	*yzzy*	*zzxy*	*yzxz*	*yzzx*	*zzyx*	*xzyz*	*xzzy*	*zzxz*	*zzzx*	*xzzz*	*zzyz*	*zzzy*	*yzzz*	*zzzz*

Interactions	Optical sign	15	$\overline{15}$	16*a*	16*b*	16*c*	$\overline{16}a$	$\overline{16}b$	$\overline{16}c$
Type I	>0	*zxxx*	*zyyy*	*zyxx*	*zxyx*	*zxxy*	*zxyy*	*zyxy*	*zyyx*
Type II	<0	*xxxz*	*yyyz*	*xyxz*	*xxyz*	*yxxz*	*yxyz*	*yyxz*	*xyyz*
Type III	<0	*xxzx*	*yyzy*	*xyzx*	*xxzy*	*yxzx*	*yxzy*	*yyzx*	*xyzy*
Type IV	<0	*xzxx*	*yzyy*	*xzyx*	*xzxy*	*yzxx*	*yzxy*	*yzyx*	*xzyy*

In Fig. 8(a), we give the angle of polarization α of each electric field as a function of the coordinate θ along the cones of types A and B. The angle α is defined in relation to an orthornormal frame (I,J,K) with K collinear to the wave vector $\mathbf{k}$:

$$\alpha=\arctan(e_J/e_I)\ . \tag{65}$$

e_I and e_J are the projections of each electric-field vector $\mathbf{e}^+$ or $\mathbf{e}^-$, calculated by (58), on the plane I-J, defined as the following in the optical frame (x,y,z):

$$\begin{aligned} &x_I=-\cos\phi\cos\theta,\quad y_I=-\sin\phi\cos\theta,\quad z_I=\sin\theta\ ,\\ &x_J=\sin\phi,\quad y_J=-\cos\phi,\quad z_J=0\ ,\\ &x_K=\sin\theta\cos\phi,\quad y_K=\sin\theta\sin\phi,\quad z_K=\cos\theta\ . \end{aligned} \tag{66}$$

TABLE XI. Correspondence between 2*o*2*e*, 2*o*2*e*-2*e*2*o* biaxial functions and field factors according to the six corresponding types of phase-matched SFM(ω_4).

		2*o*2*e* and 2*o*2*e*-2*e*2*o* biaxial field-factor functions													
Interactions	Optical sign	11*a*	11*b*	$\overline{11}a$	$\overline{11}b$	12*a*	12*b*	12*c*	12*d*	$\overline{12}a$	$\overline{12}b$	$\overline{12}c$	$\overline{12}d$	13*a*	13*b*
Type V^4	>0	*yzzz*	*zyzz*	*xzzz*	*zxzz*	*xzyz*	*xzzy*	*zxyz*	*zxzy*	*yzxz*	*yzzx*	*zyxz*	*zyzx*	*yzxx*	*zyxx*
Type VI^4	>0	*yzzz*	*zzyz*	*xzzz*	*zzxz*	*xyzz*	*xzzy*	*zyxz*	*zzxy*	*yxzz*	*yzzx*	*zxyz*	*zzyx*	*yxzx*	*zxyx*
Type VII^4	>0	*yzzz*	*zzzy*	*xzzz*	*zzzx*	*xyzz*	*xzyz*	*zyzx*	*zzyx*	*yxzz*	*yzxz*	*zxzy*	*zzxy*	*yxxz*	*zxxy*
Type V^4	<0	*zzyz*	*zzzy*	*zzxz*	*zzzx*	*zyxz*	*yzxz*	*zyzx*	*yzzx*	*zxyz*	*xzyz*	*zxzy*	*xzzy*	*xxyz*	*xxzy*
Type VI^4	<0	*zyzz*	*zzzy*	*zxzz*	*zzzx*	*zxyz*	*yxzz*	*zzyx*	*yzzx*	*zyxz*	*xyzz*	*zzxy*	*xzzy*	*xyxz*	*xzxy*
Type VII^4	<0	*zyzz*	*zzyz*	*zxzz*	*zzxz*	*zxzy*	*yxzz*	*zzxy*	*yzxz*	*zyzx*	*xyzz*	*zzyx*	*xzyz*	*xyzx*	*xzyx*

Interactions	Optical sign	$\overline{13}a$	$\overline{13}b$	14*a*	14*b*	14*c*	14*d*	$\overline{14}a$	$\overline{14}b$	$\overline{14}c$	$\overline{14}d$	15*a*	15′*a*	15*b*	15′*b*	$\overline{15}a$	$\overline{15}'a$
Type V^4	>0	*xzyy*	*zxyy*	*xzxz*	*xzzx*	*zxxz*	*zxzx*	*yzyz*	*yzzy*	*zyyz*	*zyzy*	*yzxy*	*yzyx*	*zyxy*	*zyyx*	*xzyx*	*xzxy*
Type VI^4	>0	*xyzy*	*zyxy*	*xxzz*	*xzzx*	*zxxz*	*zzxx*	*yyzz*	*yzzy*	*zyyz*	*zzyy*	*yxzy*	*yyzx*	*zxyy*	*zyyx*	*xyzx*	*xxzy*
Type VII^4	>0	*xyyz*	*zyyx*	*xxzz*	*xzxz*	*zxzx*	*zzxx*	*yyzz*	*yzyz*	*zyzy*	*zzyy*	*yxyz*	*yyxz*	*zxyy*	*zyxy*	*xyxz*	*xxyz*
Type V^4	<0	*yyxz*	*yyzx*	*zxxz*	*xzxz*	*zxzx*	*xzzx*	*zyyz*	*yzyz*	*zyzy*	*yzzy*	*yxyz*	*xyyz*	*yxzy*	*xyzy*	*xyxz*	*yxxz*
Type VI^4	<0	*yxyz*	*yzyx*	*zxxz*	*xxzz*	*zzxx*	*xzzx*	*zyyz*	*yyzz*	*zzyy*	*yzzy*	*yyxz*	*xyyz*	*yzxy*	*xzyy*	*xxyz*	*yxxz*
Type VII^4	<0	*yxzy*	*yzxy*	*zxzx*	*xxzz*	*zzxx*	*xzxz*	*zyzy*	*yyzz*	*zzyy*	*yzyz*	*yyzx*	*xyzy*	*yzyx*	*xzyy*	*xxzy*	*yxzx*

Interactions	Optical sign	$\overline{15}b$	$\overline{15}'b$	16*a*	16*b*	$\overline{16}a$	$\overline{16}b$	17	$\overline{17}$	18	18′	19*a*	19*b*	$\overline{19}a$	$\overline{19}b$	20
Type V^4	>0	*zxyx*	*zxxy*	*yzyy*	*zyyy*	*xzxx*	*zxxx*	*zzyy*	*zzxx*	*zzxy*	*zzyx*	*zzyz*	*zzzy*	*zzxz*	*zzzx*	*zzzz*
Type VI^4	>0	*zyxx*	*zxxy*	*yyzy*	*zyyy*	*xxzx*	*zxxx*	*zyzy*	*zxzx*	*zxzy*	*zyzx*	*zyzz*	*zzzy*	*zxzz*	*zzzx*	*zzzz*
Type VII^4	>0	*zyxx*	*zxyx*	*yyyz*	*zyyy*	*xxxz*	*zxxx*	*zyyz*	*zxxz*	*zxyz*	*zyxz*	*zyzz*	*zzyz*	*zxzz*	*zzxz*	*zzzz*
Type V^4	<0	*xyzx*	*yxzx*	*yyyz*	*yyzy*	*xxxz*	*xxzx*	*yyzz*	*xxzz*	*yxzz*	*xyzz*	*zyzz*	*yzzz*	*zxzz*	*xzzz*	*zzzz*
Type VI^4	<0	*xzyx*	*yzxx*	*yyyz*	*yzyy*	*xxxz*	*xzxx*	*yzyz*	*xzxz*	*yzxz*	*xzyz*	*zzyz*	*yzzz*	*zzxz*	*xzzz*	*zzzz*
Type VII^4	<0	*xzxy*	*yzxx*	*yyzy*	*yzyy*	*xxzx*	*xzxx*	*yzzy*	*xzzx*	*yzzx*	*xzzy*	*zzzy*	*yzzz*	*zzzx*	*xzzz*	*zzzz*

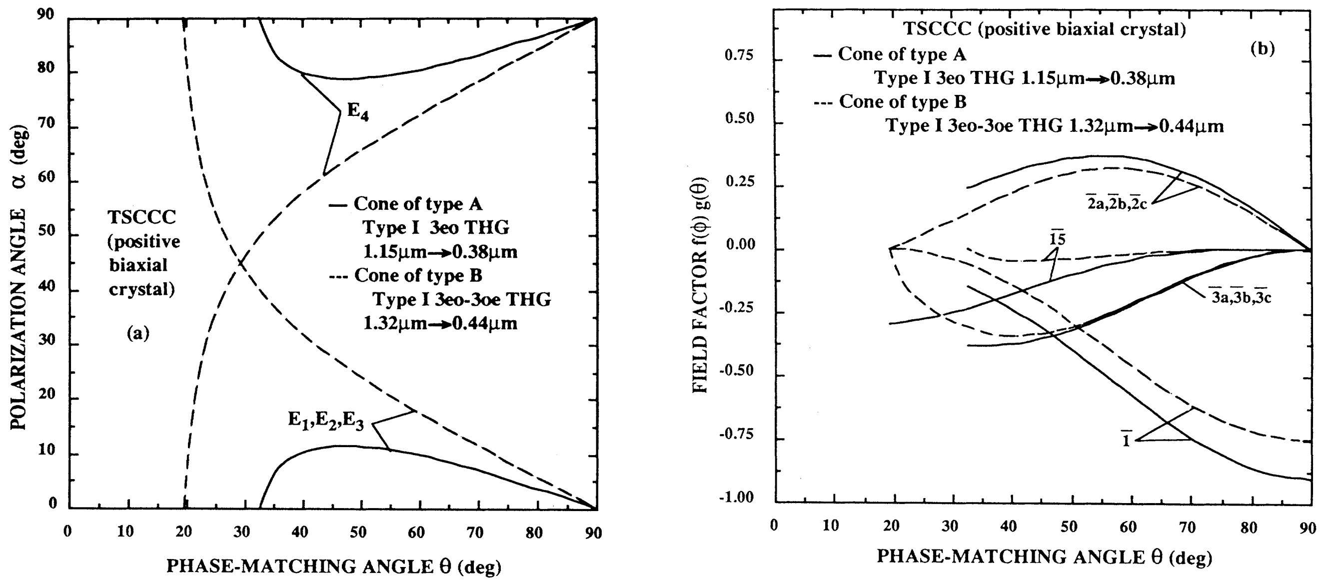


FIG. 8. (a) Polarization angles α of the interacting waves (E_i, $i=1,2,3,4$), and (b) field-factor functions which differ in areas a and d vs the phase-matching spherical coordinate θ, calculated for the positive biaxial TSCCC crystal (crystal class C_s).

TABLE XII. $3oe$, $3oe$-$3eo$, $3eo$, $3eo$-$3oe$, $2o2e$, $2o2e$-$2e2o$ field-factor functions intervening in the calculation of the effective coefficient χ_{eff} for the biaxial crystal classes.

Biaxial crystal classes	C_1,C_i	C_s,C_2,C_{2h}	C_{2v},D_2,D_{2h}
Intervening $3oe$ and $3oe$-$3eo$ field-factor functions	all	$2,\bar{2}$ $4,4',4'',\bar{4},\bar{4}',\bar{4}''$ $5,\bar{5}$ $6,6',6'',\bar{6},\bar{6}',\bar{6}''$ $8a,8b,8c,\bar{8}a,\bar{8}b,\bar{8}c$ $10a,10b,10c,\overline{10}a,\overline{10}b,\overline{10}c$ 13 $15a,15b,15c,\overline{15}a,\overline{15}b,\overline{15}c$ $17a,17'a,17b,17'b,17c,17'c$	$4,4',4'',\bar{4},\bar{4}',\bar{4}''$ $5,\bar{5}$ $10a,10b,10c,\overline{10}a,\overline{10}b,\overline{10}c$ 13 $15a,15b,15c,\overline{15}a,\overline{15}b,\overline{15}c$
Intervening $3eo$ and $3eo$-$3oe$ field-factor functions	all	$2a,2b,2c,\bar{2}a,\bar{2}b,\bar{2}c$ $4a,4b,4c,\bar{4}a,\bar{4}b,\bar{4}c$ $7,\bar{7}$ $8,8',8'',\bar{8},\bar{8}',\bar{8}''$ $9,\bar{9}$ $10,10',10'',\overline{10},\overline{10}',\overline{10}''$ $11a,11b,11c,\overline{11}a,\overline{11}b,\overline{11}c$ $12a,12b,12c,12'a,12'b,12'c$ 14	$4a,4b,4c,\bar{4}a,\bar{4}b,\bar{4}c$ $9,\bar{9}$ $10,10',10'',\overline{10},\overline{10}',\overline{10}''$ $11a,11b,11c,\overline{11}a,\overline{11}b,\overline{11}c$ 14
Intervening $2o2e$ and $2o2e$-$2e2o$ field-factor functions	all	$1,1',4,\bar{4}$ $6,6',\bar{6},\bar{6}'$ $7,7',\bar{7},\bar{7}'$ $8,\bar{8},9,\bar{9}$ $10,10',\overline{10},\overline{10}'$ $12a,12b,12c,12d$ $\overline{12}a,\overline{12}b,\overline{12}c,\overline{12}d$ $14a,14b,14c,14d$ $\overline{14}a,\overline{14}b,\overline{14}c,\overline{14}d$ $17,\overline{17},18,18'$ 20	$4,\bar{4}$ $8,\bar{8}$ $9,\bar{9}$ $10,10',\overline{10},\overline{10}'$ $14a,14b,14c,14d$ $\overline{14}a,\overline{14}b,\overline{14}c,\overline{14}d$ $17,\overline{17}$ 20

$\alpha=0°$ corresponds to an extraordinary wave and $\alpha=90°$ to an ordinary wave. According to Fig. 8(a), the four electric-field vectors turn 90° for the cone of type B; the configuration of polarization is $3eo$ in area c and $3oe$ in area a. For the cone of type A, the configuraiton is $3eo$ in area c and in area d. We give in Fig. 8(b) the field factors of types A and B cones, calculated from (4) and (64), which differ in area a.

According to the nonzero elements of $\chi^{(3)}$, we give in Table XII the trigonometric functions of the field factors intervening in the calculation of the effective coefficient for the eight biaxial crystal classes. Table XII must be read with Tables IV, VI, VII, IX, X, and XI. All the biaxial crystal classes allow the four-wave nonlinear optical parametric interactions for all types of collinear phase matching.

V. CONCLUSION

The use of the field-factor formalism allows an unified description of the phase-matched four-wave SFM and DFM. Even if the third-order nonlinearity of the crystal is high and even if phase-matching directions exist, the efficiency of the interaction can be nil because of symmetry of the $\chi^{(3)}$ and $\mathbf{F}^{(3)}$ tensors: the effective coefficient is nil for $3oe$ and $3eo$ configurations of polarization in the uniaxial crystal classes $D_6(622)$, $D_{6h}(6/m\ m\ m)$, $D_{3h}(\bar{6}2m)$, and $C_{6v}(6mm)$. It is also the case under Kleinman's conjecture for the four previous classes and the three other hexagonal classes $C_{3h}(\bar{6})$, $C_6(6)$, and $C_{6h}(6/m)$. Thus this study completes the calculation of the third-order electric susceptibility tensor elements from crystallographical and chemical criteria within the context of the study and optimization *a priori* of a crystal for a given nonlinear interaction [15,16]. The study of the angular variation of field factors for the 14 possible phase-matched configurations of polarization is a guide for the judicious choice of interaction and phase-matching direction in order to perform the best determination of all the usefull $\chi^{(3)}$ elements by phase-matching experiments.

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