LASER-BEAM INTENSITY DEPENDENT, OPTICAL CIRCULAR BIREFRINGENCE IN CRYSTALS*

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Using a phenomenological approach, the optical circular birefringence of crystals in the presence of intense laser light is shown to be the sum of natural gyration, self-induced optical rotation (exhibited by optically inactive bodies), and a self-induced nonlinear variation in the natural gyration. These effects are discussed for all crystallographic classes and the conditions for the separate observation of each effect are specified.

1. Introduction

Intense laser light induces an optical Kerr effect, as observed by Mayer [1] in liquids and recently studied by Paillette [2] in glasses, by Lalanne [3] and Wong [4] in liquid crystals, and by Bischofberger [5] in molecular crystals. Maker et al. [6] showed that the light, if elliptically polarized, undergoes a rotation of its polarization plane when traversing the liquid. This self-induced rotation of the polarization ellipse has recently been observed by Owyoung [7] in glasses and cubic crystals, and by Wong and Shen [4] in liquid crystals. As shown by Kielich [8] and Atkins [9], besides these two effects a nonlinear change in the optical natural gyration can occur in optically active isotropic bodies. Such variations have indeed been observed by Vlasov and Zaitsev [10] in tinted quartz crystals and in optically active organic solutions.

In this communication, we give a complete analysis of the conditions for the self induction of optical circular birefringence (OCB) by an intense laser beam in istotropic and crystalline bodies of all kinds exhibiting in general spatial dispersion [11,12]. When calculating light refraction indices, we assume in accordance with an earlier theory of Piekara and Kielich [13] that intense light causes nonlinear changes in the electric permittivity as well as in the the magnetic permeability. We shall consider systematically, in addition to the electric and the magnetic multipolar susceptibilities, also the cross electro–magnetic and magneto–electric multipolar susceptibilities [12]. For the relevant 5th rank tensors of the nonlinear susceptibilities, the non-zero and mutually independent tensor elements have been found by methods of group theory [14].

2. Fundamentals of the phenomenological theory

The tensors of the dynamic electric and magnetic permittivity of a medium with natural or induced anisotropy are given by the following, well-known equations:

$$(\epsilon_{ij} - \delta_{ij}) E_j(\boldsymbol{r}, t) = 4\pi \{P_{ei}(\boldsymbol{r}, t) + P_{ei}^{m}(\boldsymbol{r}, t)\} , \tag{1}$$

$$(\mu_{ij} - \delta_{ij}) H_j(\boldsymbol{r}, t) = 4\pi \{P_{mi}(\boldsymbol{r}, t) + P_{mi}^{e}(\boldsymbol{r}, t)\} , \tag{2}$$

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where $\boldsymbol{E}(\boldsymbol{r}, t)$ and $\boldsymbol{H}(\boldsymbol{r}, t)$ are the electric and magnetic vectors of the electromagnetic field at the moment of time t in the space point $\boldsymbol{r}$.

In an appropriately strong electromagnetic field, the ith component of the total electric polarization at the fundamental frequency ω is in Fourier representation:

$$P_{ei}(\omega, \boldsymbol{k}) = \{\chi_{ij}^{ee}(\omega, \boldsymbol{k}) + \chi_{ijkl}^{eeee}(\omega, \boldsymbol{k})\, E_k(\omega, \boldsymbol{k})\, E_1^*(\omega, \boldsymbol{k}) + ...\}\, E_j(\omega, \boldsymbol{k}) \,, \tag{3}$$

χ_{ij}^{ee} being the tensor of the linear electric susceptibility and χ_{ijkl}^{eeee} that of the nonlinear third-order electric susceptibility.

The notation of the expansion (3) is such that the tensors $\chi_{ij}^{ee}(\omega, \boldsymbol{k})$ and $\chi_{ijkl}^{eeee}(\omega, \boldsymbol{k})$ comprise phenomenologically not only frequency dispersion but notably the spatial dispersion [11] resulting from electric multipole transitions [12]. Hence we have for the linear susceptibility at not excessively strong spatial dispersion [11]:

$$\chi_{ij}^{ee}(\omega, \boldsymbol{k}) = \chi_{ij}^{ee}(\omega) + \mathrm{i}\eta_{ijm}^{ee}(\omega)\, k_m + \eta_{ijmn}^{ee}(\omega)\, k_m k_n + ... \,, \tag{4}$$

where $\chi_{ij}^{ee}(\omega)$ is the tensor of the linear electric dipole susceptibility. The third-rank tensor $\eta_{ijm}^{ee}(\omega)$, in accordance with quantum-mechanical theory, comprises the electric dipole susceptibility with electric quadrupole transition and the electric quadrupole susceptibility with electric dipole transition. The fourth-rank tensor $\eta_{ijmn}^{ee}(\omega)$ results from electric dipole, quadrupole and octupole transitions.

Similarly to the linear susceptibility (4), we write the following expansion for the nonlinear susceptibility:

$$\chi_{ijkl}^{eeee}(\omega, \boldsymbol{k}) = \chi_{ijkl}^{eeee}(\omega) + \mathrm{i}\eta_{ijklm}^{ee}(\omega)\, k_m + \eta_{ijklmn}^{ee}(\omega)\, k_m k_n + ... \,, \tag{5}$$

where the tensor $\chi_{ijkl}^{eeee}(\omega)$ is taken in the electric dipole approximation, whereas $\eta_{ijklm}^{ee}(\omega)$ and higher tensors arise in the higher electric multipolar transitions [12].

The magnetic polarization of eq. (2) too comprises all electric and magnetic contributions [12]; in the present approximation, it can be written as follows:

$$P_{mi}(\omega, \boldsymbol{k}) = \{\chi_{ij}^{mm}(\omega, \boldsymbol{k}) + \chi_{ijkl}^{mmee}(\omega, \boldsymbol{k})\, E_k(\omega, \boldsymbol{k})\, E_1^*(\omega, \boldsymbol{k}) + ...\}\, H_j(\omega, \boldsymbol{k}) \,, \tag{6}$$

χ_{ij}^{mm} being the tensor of the linear magnetic dipole susceptibility, and χ_{ijkl}^{mmee} that of the nonlinear magneto-electric susceptibility in the presence of spatial dispersion as given by expansions similar to (4) and (5).

In the general case of electrically and magnetically gyrotropic media one has to take into account, besides expansions (3) and (6), the cross electro–magnetic polarization

$$P_{ei}^{m}(\omega, \boldsymbol{k}) = \{\chi_{ij}^{em}(\omega, \boldsymbol{k}) + \chi_{ijkl}^{emee}(\omega, \boldsymbol{k})\, E_k(\omega, \boldsymbol{k})\, E_1^*(\omega, \boldsymbol{k}) + ...\}\, H_j(\omega, \boldsymbol{k}) \,, \tag{7}$$

and the magneto–electric polarization

$$P_{mi}^{e}(\omega, \boldsymbol{k}) = \{\chi_{ij}^{me}(\omega, \boldsymbol{k}) + \chi_{ijkl}^{meee}(\omega, \boldsymbol{k})\, E_k(\omega, \boldsymbol{k})\, E_1^*(\omega, \boldsymbol{k}) + ...\}\, E_j(\omega, \boldsymbol{k}) \,. \tag{8}$$

The tensors of the linear and nonlinear electro–magnetic and magneto–electric susceptibilities occurring in these expansions depend in general on higher, and higher electric and magnetic multipole transitions [12].

Table 1 specifies the numbers of the non-zero and independent elements of the tensors occurring in eqs. (3)–(8) as found by methods of group theory [14].

3. Optical circular birefringence (OCB)

On transforming the polarizations (3) and (6)–(8) to a circular representation $E_\pm = (2)^{-1/2}(E_x \pm \mathrm{i}E_y)$ and having recourse to eqs. (1) and (2) and Maxwell's equations, we find for the difference in the refractive indices of light propagating along the Z-axis for the two cases of circular polarization:

$$n_+ - n_- = G_\mathrm{N} + \tfrac{1}{2}R_\mathrm{s}\{|E_-(\omega)|^2 - |E_+(\omega)|^2\} + \tfrac{1}{2}G_\mathrm{s}\{|E_+(\omega)|^2 + |E_-(\omega)|^2\} + ... \,. \tag{9}$$

Table 1
The number of non-zero (N) and independent (I) elements of the nonlinear susceptibility tensors occurring in eqs. (3)–(6). Parentheses ( ) contain those indices in which the tensors are symmetric. The number of non-zero and independent elements of the nonlinear susceptibility tensors occurring in eqs. (7); (8) are given in ref. [14].

Class	$\eta^{ee}_{i(jm)}(\omega)$		$\chi^{eeee}_{ijkl}(\omega)$		$\eta^{ee}_{i(jmnu)}(\omega)$		$\eta^{ee}_{(im)(jnu)}(\omega)$		$\eta^{eeee}_{i(jm)kl}(\omega)$		$\eta^{ee}_{ijklm}(\omega)$		$\eta^{eeee}_{i(jm)(kn)l}(\omega)$	
	N	I	N	I	N	I	N	I	N	I	N	I	N	I
$1(C_1)$	27	18	81	81	243	45	243	60	243	162	243	243	729	324
$\bar{1}(C_i)$	0	0	81	81	0	0	0	0	0	0	0	0	729	324
$m(C_s)$	14	10	41	41	122	24	122	32	122	82	122	122	365	164
$2(C_2)$	13	8	41	41	121	21	121	28	121	80	121	121	365	164
$2/m(C_{2h})$	0	0	41	41	0	0	0	0	0	0	0	0	365	164
$222(D_2)$	6	3	21	21	60	9	60	12	60	39	60	60	183	84
$mm2(C_{2v})$	7	5	21	21	61	12	61	16	61	41	61	61	183	84
$mmm(D_{2h})$	0	0	21	21	0	0	0	0	0	0	0	0	183	84
$4(C_4)$	11	4	41	21	109	11	113	14	119	40	121	61	361	82
$\bar{4}(S_4)$	12	4	41	21	114	10	108	14	120	40	120	60	361	82
$4/m(C_{4h})$	0	0	41	21	0	0	0	0	0	0	0	0	361	82
$422(D_4)$	4	1	21	11	48	4	52	5	58	19	60	30	183	43
$4mm(C_{4v})$	7	3	21	11	61	7	61	9	61	21	61	31	183	43
$\bar{4}2m(D_{2d})$	6	2	21	11	60	5	60	7	60	20	60	30	183	43
$4/mmm(D_{4h})$	0	0	21	11	0	0	0	0	0	0	0	0	183	43
$3(C_3)$	19	6	73	27	213	15	225	20	231	54	233	81	713	108
$\bar{3}(S_6)$	0	0	73	27	0	0	0	0	0	0	0	0	713	108
$32(D_3)$	8	2	37	14	96	6	108	8	114	26	116	40	359	56
$3m(C_{3v})$	11	4	37	14	117	9	117	12	117	28	117	41	359	56
$\bar{3}m(D_{3d})$	0	0	37	14	0	0	0	0	0	0	0	0	359	56
$6(C_6)$	11	4	41	19	101	7	113	10	119	32	121	51	361	60
$\bar{6}(C_{3h})$	8	2	41	18	112	8	112	10	112	22	112	30	361	60
$6/m(C_{6h})$	0	0	41	19	0	0	0	0	0	0	0	0	361	60
$622(D_6)$	4	1	21	10	40	2	52	3	58	15	60	25	183	32
$6mm(C_{6v})$	7	3	21	10	61	5	61	7	61	17	61	26	183	32
$\bar{6}m2(D_{3h})$	4	1	21	10	56	4	56	5	56	11	56	15	183	32
$6/mmm(D_{6h})$	0	0	21	10	0	0	0	0	0	0	0	0	183	32
$23(T)$	6	1	21	7	60	3	60	4	60	13	60	20	183	28
$m3(T_h)$	0	0	21	7	0	0	0	0	0	0	0	0	183	28
$432(O)$	0	0	21	4	24	1	36	1	54	6	60	10	183	15
$\bar{4}3m(T_d)$	6	1	21	4	60	2	60	3	60	7	60	10	183	15
$m3m(O_h)$	0	0	21	4	0	0	0	0	0	0	0	0	183	15
Y	0	0	21	3	0	0	0	0	54	3	60	6	183	7
Y_h	0	0	21	3	0	0	0	0	0	0	0	0	183	7
K	0	0	21	3	0	0	0	0	54	3	60	6	183	7
K_h	0	0	21	3	0	0	0	0	0	0	0	0	183	7

Eq. (9) describes the intensity-dependent OCB, consisting of

(i) natural optical gyration:

$$G_{\mathrm{N}} = -2\pi \mathrm{i}\{(\mu/\epsilon)^{1/2}[\chi^{\mathrm{ee}}_{xy}(\omega,\boldsymbol{k}) - \chi^{\mathrm{ee}}_{yx}(\omega,\boldsymbol{k})] - \chi^{\mathrm{em}}_{xx}(\omega,\boldsymbol{k}) - \chi^{\mathrm{em}}_{yy}(\omega,\boldsymbol{k})$$
$$+ \chi^{\mathrm{me}}_{xx}(\omega,\boldsymbol{k}) + \chi^{\mathrm{me}}_{yy}(\omega,\boldsymbol{k}) + (\epsilon/\mu)^{1/2}[\chi^{\mathrm{mm}}_{xy}(\omega,\boldsymbol{k}) - \chi^{\mathrm{mm}}_{yx}(\omega,\boldsymbol{k})]\}\,, \tag{10}$$

(ii) self-induced optical rotation:

$$R_{\mathrm{s}} = \pi\{(\mu/\epsilon)^{1/2}\, r^{\mathrm{ee}}_{\mathrm{s}} + r^{\mathrm{em}}_{\mathrm{s}} - r^{\mathrm{me}}_{\mathrm{s}} + (\epsilon/\mu)^{1/2}\, r^{\mathrm{mm}}_{\mathrm{s}}\}\,, \tag{11}$$

(iii) a self-induced nonlinear variation in natural optical gyration:

$$G_{\mathrm{s}} = \pi\mathrm{i}\{(\mu/\epsilon)^{1/2}\, g^{\mathrm{ee}}_{\mathrm{s}} + g^{\mathrm{em}}_{\mathrm{s}} - g^{\mathrm{me}}_{\mathrm{s}} + (\epsilon/\mu)^{1/2}\, g^{\mathrm{mm}}_{\mathrm{s}}\}\,. \tag{12}$$

In (11), we have used the notation:

$$r^{\mathrm{ee}}_{\mathrm{s}} = -\chi^{\mathrm{eeee}}_{xxyy}(\omega,\boldsymbol{k}) - \chi^{\mathrm{eeee}}_{yyxx}(\omega,\boldsymbol{k}) - \chi^{\mathrm{eeee}}_{xyxy}(\omega,\boldsymbol{k}) - \chi^{\mathrm{eeee}}_{yxyx}(\omega,\boldsymbol{k})$$
$$+ \chi^{\mathrm{eeee}}_{xxxx}(\omega,\boldsymbol{k}) + \chi^{\mathrm{eeee}}_{yyyy}(\omega,\boldsymbol{k}) + 3\{\chi^{\mathrm{eeee}}_{xyyx}(\omega,\boldsymbol{k}) + \chi^{\mathrm{eeee}}_{yxxy}(\omega,\boldsymbol{k})\}\,. \tag{13}$$

$$r^{\mathrm{em}}_{\mathrm{s}} = 3\{\chi^{\mathrm{emee}}_{yyxy}(\omega,\boldsymbol{k}) - \chi^{\mathrm{emee}}_{xxyx}(\omega,\boldsymbol{k})\} + \chi^{\mathrm{emee}}_{xyxx}(\omega,\boldsymbol{k}) - \chi^{\mathrm{emee}}_{yxyy}(\omega,\boldsymbol{k})$$
$$+ \chi^{\mathrm{emee}}_{yxxx}(\omega,\boldsymbol{k}) - \chi^{\mathrm{emee}}_{xyyy}(\omega,\boldsymbol{k}) + \chi^{\mathrm{emee}}_{xxxy}(\omega,\boldsymbol{k}) - \chi^{\mathrm{emee}}_{yyyx}(\omega,\boldsymbol{k})\,. \tag{14}$$

The other quantities $r^{\mathrm{mm}}_{\mathrm{s}}$ and $r^{\mathrm{me}}_{\mathrm{s}}$ can be expressed similarly in terms of the appropriate tensor elements $\chi^{\mathrm{mmee}}_{ijkl}$ and $\chi^{\mathrm{meee}}_{ijkl}$.

Likewise, in (12) we use the notation:

$$g^{\mathrm{ee}}_{\mathrm{s}} = -3\{\chi^{\mathrm{eeee}}_{xyyy}(\omega,\boldsymbol{k}) - \chi^{\mathrm{eeee}}_{yxxx}(\omega,\boldsymbol{k})\} - \chi^{\mathrm{eeee}}_{xxxy}(\omega,\boldsymbol{k}) + \chi^{\mathrm{eeee}}_{yyyx}(\omega,\boldsymbol{k})$$
$$- \chi^{\mathrm{eeee}}_{xxyx}(\omega,\boldsymbol{k}) + \chi^{\mathrm{eeee}}_{yyxy}(\omega,\boldsymbol{k}) - \chi^{\mathrm{eeee}}_{xyxx}(\omega,\boldsymbol{k}) + \chi^{\mathrm{eeee}}_{yxyy}(\omega,\boldsymbol{k})\,, \tag{15}$$

$$g^{\mathrm{em}}_{\mathrm{s}} = -\chi^{\mathrm{emee}}_{xyxy}(\omega,\boldsymbol{k}) - \chi^{\mathrm{emee}}_{yxyx}(\omega,\boldsymbol{k}) - \chi^{\mathrm{emee}}_{xyyx}(\omega,\boldsymbol{k}) - \chi^{\mathrm{emee}}_{yxxy}(\omega,\boldsymbol{k})$$
$$+ \chi^{\mathrm{emee}}_{xxxx}(\omega,\boldsymbol{k}) + \chi^{\mathrm{emee}}_{yyyy}(\omega,\boldsymbol{k}) + 3\{\chi^{\mathrm{emee}}_{xxyy}(\omega,\boldsymbol{k}) + \chi^{\mathrm{emee}}_{yyxx}(\omega,\boldsymbol{k})\}\,. \tag{16}$$

The other quantities, $g^{\mathrm{mm}}_{\mathrm{s}}$ and $g^{\mathrm{me}}_{\mathrm{s}}$, can be expressed similarly in terms of the appropriate tensor elements $\chi^{\mathrm{mmee}}_{ijkl}$ and $\chi^{\mathrm{meee}}_{ijkl}$.

The general formulae (9)–(16) hold for optically active as well as optically inactive bodies both isotropic and crystalline.

4. Applications and discussion

We begin with optically inactive crystals. For the classes 4mm and 43m, we have by (11) and (13) in the absence of spatial dispersion:

$$R = 2\pi(\mu/\epsilon)^{1/2}\,\{\chi^{\mathrm{eeee}}_{xxxx}(\omega) + 3\chi^{\mathrm{eeee}}_{xyyx}(\omega) - \chi^{\mathrm{eeee}}_{xxyy}(\omega) - \chi^{\mathrm{eeee}}_{xyxy}(\omega)\} \tag{17}$$

In particular, for the classes 3m, 6mm, $\bar{6}$ and $\bar{6}$m2 and an isotropic medium we have additionally the symmetry relation:

$$\chi_{xxxx} = \chi_{xxyy} + \chi_{xyxy} + \chi_{xyyx}\,,$$

Table 2
Crystallographical classes in which optical rotations G_N, R_s and G_s can occur in the presence of spatial linear ($k_z \neq 0$), quadratic ($k_z^2 \neq 0$) or cubic ($k_z^3 \neq 0$) dispersion and its absence ($k_z = 0$) .

$n_+ - n_-$		Tensor elements relevant to $n_+ - n_- \neq 0$	Crystallographical classes admitting $n_+ - n_- \neq 0$
G_N	$k_z \neq 0$	$\eta^{ee}_{xyz}(\omega) - \eta^{ee}_{yxz}(\omega)$	1, 2, 222, 4, 422, 3, 32, 6, 622,
		$\chi^{em}_{xx}(\omega) + \chi^{em}_{yy}(\omega)$	1, 2, 222, 4, 422, 3, 32, 6, 622, 23, 432, Y, K,
	$k_z^3 \neq 0$	$\eta^{ee}_{xyzzz}(\omega) - \eta^{ee}_{yxzzz}(\omega)$	1, 2, 222, 4, 422, 3, 32, 6, 622, 23, 432,
		$\eta^{em}_{xxzz}(\omega) + \eta^{em}_{yyzz}(\omega)$	1, 2, 222, 4, 422, 3, 32. 6, 622, 23, 432, Y, K,
		$\eta^{mm}_{xyz}(\omega) - \eta^{mm}_{yxz}(\omega)$	1, 2, 222, 4, 422, 3, 32, 6, 622,
R_s	$k_z = 0$	$r^{ee}_{s(0)}$	all classes
	$k_z \neq 0$	$r^{ee}_{s(1)}$	1, 2, mm2, 4, 4mm, 3, 3m, 6, 6mm,
		$r^{em}_{s(0)}$	1, 2, mm2, 4, 4mm, 3, 3m, 6, 6mm,
	$k_z^2 \neq 0$	$r^{ee}_{s(2)}$	all classes
		$r^{em}_{s(1)}$	all classes
		$r^{mm}_{s(0)}$	all classes
G_s	$k_z = 0$	$g^{ee}_{s(0)}$	1, $\bar{1}$, m, 2, 2/m, 4, $\bar{4}$. 4/m, 3, $\bar{3}$, 6, $\bar{6}$, 6/m,
	$k_z \neq 0$	$g^{ee}_{s(1)}$	1, 2, 222, 4, 422, 3, 32, 6, 622, 23, 432, Y, K,
		$g^{em}_{s(0)}$	1, 2, 222, 4, 422, 3, 32, 6, 622, 23, 432, Y, K,
	$k_z^2 \neq 0$	$g^{ee}_{s(2)}$	1, $\bar{1}$, m, 2, 2/m, 4, $\bar{4}$, 4/m, 3, $\bar{3}$, 6, $\bar{6}$, 6/m,
		$g^{em}_{s(1)}$	1, $\bar{1}$, m, 2, 2/m, 4, $\bar{4}$, 4/m, 3, $\bar{3}$, 6, $\bar{6}$, 6/m,
		$g^{mm}_{s(0)}$	1, $\bar{1}$, m, 2, 2/m, 4, $\bar{4}$, 4/m, 3, $\bar{3}$, 6, $\bar{6}$, 6/m,

reducing eq. (17) to the following, simpler form:

$$R = 8\pi(\mu/\epsilon)^{1/2}\ \chi^{eeee}_{xyyx}(\omega)\ , \tag{17a}$$

which becomes identical with Maker's result [6] for the isotropic medium.

Obviously, self-induced rotation (11) occurs also in optically active classes (see table 2). If spatial dispersion is considered, further contributions (14) emerge.

We now apply eq. (12) to uniaxial crystals of the tetragonal system, obtaining in the absence of spatial dispersion [for brevity, we write only the electric part (15)] :

$$G = -2\pi i(\mu/\epsilon)^{1/2}\ \{3\chi^{eeee}_{xyyy}(\omega) + \chi^{eeee}_{xxyx}(\omega) + \chi^{eeee}_{xyxx}(\omega) + \chi^{eeee}_{xxxy}(\omega)\}\ . \tag{18}$$

In particular, for the classes 3, $\bar{3}$, 6 and 6/m we have moreover the relation: $\chi_{xyyy} = \chi_{xxxy} + \chi_{xxyx} + \chi_{xyxx}$, reducing (18) to the form:

$$G = -8\pi i(\mu/\epsilon)^{1/2}\ \chi^{eeee}_{xyyy}(\omega)\ . \tag{18a}$$

It is highly interesting that this result also holds for the class $\bar{6}$ although this class is not optically active, but is found to be susceptible to the induction of nonlinear optical activity under the influence of intense light.

If linear dispersion is present and the tensor $\eta^{eeee}_{[ij]klm}$ is assumed to be antisymmetric in the indices i, j we get by eqs. (12) and (15):

$$G = -4\pi i(\mu/\epsilon)^{1/2} \{\eta^{eeee}_{xyxxz}(\omega) + \eta^{eeee}_{xyyyz}(\omega)\} k_z \,. \tag{19}$$

This formula holds for the classes 222, 422, 32, 622 and 23. In particular, for the classes 432, Y and K we have $\eta^{eeee}_{xyxxz} = \eta^{eeee}_{xyyyz}$, corresponding to an earlier result [8], derived for isotropic optically active bodies by molecular-statistical methods.

The presence, or absence, of the effects (9)–(16) in each crystallographic class is indicated in table 2.

Thus, for self-induced rotation and nonlinear optical gyration one has available a method for the direct determination of the value and the sign of the individual elements of the 3rd order susceptibility tensor $\chi_{ijkl}(-\omega, \omega, \omega, -\omega, \boldsymbol{k})$. This is surely important, as methods of harmonic generation are essentially restricted to its modulus.

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