OPTICAL NONLINEAR PHENOMENA IN MAGNETIZED CRYSTALS AND ISOTROPIC BODIES

By S. Kielich and R. Zawodny

Institute of Physics, A. Mickiewicz University, Poznań*

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A theory of nonlinear optical effects taking into account frequency and spatial dispersion in linearly magnetized matter is formulated. The effects are described by axial magneto-optical susceptibility tensors χ_{ijk}^{eem}, χ_{ijkl}^{eeem}, χ_{ijkln}^{eeeem}, whose nonzero and independent elements are calculated by group theory for all crystallographical classes and the isotropic case. Beside frequency mixing of laser beams and harmonics generation, the feasibility and conditions of observation of nonlinear changes in magneto-optical birefringence and rotation due to intense light or a DC electric field at both Faraday and Voigt configurations are discussed closely.

1. Introduction

The properties of matter in external electric and magnetic fields are highly relevant to science and technology. Basic treatments of the subject are available in Refs [1–6]. For over 10 years, owing to the steady perfection of laser techniques, studies on the nonlinear optical properties of crystals and various other materials have been rapidly developing by methods of multi-photon absorption [7, 8], optical harmonics generation [7–11], laser light frequency mixing [8, 10], and other higher multi-harmonic processes [12–14]. Of late, studies have begun on a variety of new nonlinear magneto-optical processes in numerous materials [15–27].

In the present paper, we propose a theory of nonlinear magneto-optical effects taking into account frequency dispersion and space dispersion described by electric and magnetic multipole transitions of arbitrary order. We restrict ourselves to optical processes due to linear magnetization, in the configurations of Faraday as well as of Voigt. The effects under consideration (thus, frequency mixing, harmonics generation, and magneto-optical rotation) are described by axial tensors of magneto-optical susceptibility χ_{ijkl}^{eeem} and χ_{ijkln}^{eeeem}, the nonzero and mutually independent elements of which are calculated by methods of group theory [3, 5] for all crystallographical classes. A discussion is given of the conditions

* Address: Instytut Fizyki, Uniwersytet im. A. Mickiewicza, Grunwaldzka 6, 60–780 Poznań, Poland.

in which some of these new nonlinear magneto-optical effects can be made accessible to observation, and of the information to be gleaned from them regarding the electro-magnetic macroscopic properties of crystals and isotropic bodies, as well as their microstructure.

2. General phenomenological fundamentals

In the case of an arbitrary continuous medium, Maxwell's macroscopic electromagnetic field equations [28]:

$$\nabla \times \boldsymbol{E}(\boldsymbol{r}, t) = -\frac{1}{c}\frac{\partial \boldsymbol{B}(\boldsymbol{r}, t)}{\partial t}, \quad \nabla \cdot \boldsymbol{B}(\boldsymbol{r}, t) = 0, \tag{1}$$

$$\nabla \times \boldsymbol{H}(\boldsymbol{r}, t) = \frac{1}{c}\frac{\partial \boldsymbol{D}(\boldsymbol{r}, t)}{\partial t} + \frac{4\pi}{c}\boldsymbol{J}(\boldsymbol{r}, t), \quad \nabla \cdot \boldsymbol{D}(\boldsymbol{r}, t) = 4\pi\varrho_e(\boldsymbol{r}, t) \tag{2}$$

where $\varrho_e(\boldsymbol{r}, t)$ and $\boldsymbol{J}(\boldsymbol{r}, t)$ are the electric charge and current densities, can be derived from the well-known Lorentz microscopic field equations by applying a suitable statistical averaging procedure [29].

The electric and magnetic displacement vectors at position $\boldsymbol{r}$ and time t are:

$$\boldsymbol{D}(\boldsymbol{r}, t) = \boldsymbol{E}(\boldsymbol{r}, t) + 4\pi\boldsymbol{P}_e(\boldsymbol{r}, t), \tag{3}$$

$$\boldsymbol{B}(\boldsymbol{r}, t) = \boldsymbol{H}(\boldsymbol{r}, t) + 4\pi\boldsymbol{P}_m(\boldsymbol{r}, t). \tag{4}$$

In the general case considered here, the electric and magnetic polarization vectors at the space-time point $(\boldsymbol{r}, t)$ can be obtained in the form of the following multipole expansion [10]:

$$\boldsymbol{P}_e(\boldsymbol{r}, t) = \sum_{n=1}^{\infty} (-1)^{n+1} \frac{2^n n!}{2n!} \nabla^{n-1}[n-1]\boldsymbol{P}_e^{(n)}(\boldsymbol{r}, t), \tag{5}$$

$$\boldsymbol{P}_m(\boldsymbol{r}, t) = \sum_{n=1}^{\infty} (-1)^{n+1} \frac{2^n n!}{2n!} \nabla^{n-1}[n-1]\boldsymbol{P}_m^{(n)}(\boldsymbol{r}, t), \tag{6}$$

where the multipole electric and magnetic polarizations (or moment densities) of order n are defined by:

$$\boldsymbol{P}_e^{(n)}(\boldsymbol{r}, t) = \langle \sum_{p=1}^{N} \boldsymbol{M}_{ep}^{(n)}\delta(\boldsymbol{r}_p - \boldsymbol{r})\rangle, \tag{7}$$

$$\boldsymbol{P}_m^{(n)}(\boldsymbol{r}, t) = \tfrac{1}{2}\langle \sum_{p=1}^{N} \{\boldsymbol{M}_{mp}^{(n)}\delta(\boldsymbol{r}_p - \boldsymbol{r}) + \delta(\boldsymbol{r}_p - \boldsymbol{r})\boldsymbol{M}_{mp}^{(n)}\}\rangle. \tag{8}$$

Above, summation extends over all N microsystems (atoms, molecules, or their ions) at positions $\boldsymbol{r}_p$, brackets $\langle\ \rangle$ symbolize a suitable statistical averaging procedure (classical or quantal), and $\delta(\boldsymbol{r}_p-\boldsymbol{r})$ is the three-dimensional Dirac δ-function. In Eqs (5) and (6), ∇ is the spatial differential operator, and the symbol $[n-1]$ denotes $(n-1)$-fold contraction of the tensor operators ∇^{n-1} and $\boldsymbol{P}^{(n)}$.

Let the p-th microsystem consist of ν_p point particles (nuclei and electrons) with electric charges e_{ps} $(s = 1, 2, \dots \nu_p)$ and positional vectors $\boldsymbol{r}_{ps}$. The 2^n-pole electric and magnetic moment operators of a p-th microsystem are defined, respectively, as [29]:

$$\boldsymbol{M}_{ep}^{(n)} = \sum_{s=1}^{\nu_p} e_{ps} r_{ps} \boldsymbol{Y}_{ps}^{(n)}, \tag{9}$$

$$\boldsymbol{M}_{mp}^{(n)} = \frac{n}{(n+1)c} \sum_{s=1}^{\nu_p} e_{ps} r_{ps}^n \boldsymbol{Y}_{ps}^{(n)} \times \dot{\boldsymbol{r}}_{ps}, \tag{10}$$

where

$$\boldsymbol{Y}_{ps}^{(n)} = [(-1)^n/n!] r_{ps}^{n+1} \nabla^n (1/r_{ps})$$

is an operator of order n having the properties of spherical harmonics.

In the well-known manner we obtain from Eqs (1)–(4) the following electromagnetic wave equations [10]:

$$\Box \boldsymbol{E}(\boldsymbol{r}, t) + 4\pi \hat{\Box} \cdot \boldsymbol{P}_e(\boldsymbol{r}, t) = 4\pi \left\{ \nabla \varrho_e(\boldsymbol{r}, t) + \frac{1}{c} \frac{\partial \boldsymbol{J}(\boldsymbol{r}, t)}{\partial t} + \frac{1}{c} \nabla \times \frac{\partial \boldsymbol{P}_m(\boldsymbol{r}, t)}{\partial t} \right\}, \tag{11}$$

$$\Box \boldsymbol{H}(\boldsymbol{r}, t) + 4\pi \hat{\Box} \cdot \boldsymbol{P}_m(\boldsymbol{r}, t) = -\frac{4\pi}{c} \nabla \times \left\{ \boldsymbol{J}(\boldsymbol{r}, t) + \frac{\partial \boldsymbol{P}_e(\boldsymbol{r}, t)}{\partial t} \right\} \tag{12}$$

which, through the polarization vectors (5) and (6), contain all multipole contributions. Above, we have introduced the scalar Dalambertian operator $\Box = \nabla^2 - c^{-2}\partial^2/\partial t^2$ and an analogous tensorial operator $\hat{\Box} = \nabla\nabla - c^2 \boldsymbol{U} \partial^2/\partial t^2$ with $\boldsymbol{U}$ denoting the second-rank unit tensor.

The general wave equations (11) and (12) reduce to various particular cases for which solutions can be obtained in both linear and nonlinear approximation [7, 30]. This requires the explicit prescription of the dependence of the polarization vectors $\boldsymbol{P}_e$ and $\boldsymbol{P}_m$ on the electric and magnetic field strengths $\boldsymbol{E}$ and $\boldsymbol{H}$.

3. *Linear processes*

An electric (and likewise a magnetic) field varying with the time t and spatial coordinates $\boldsymbol{r}$ can be represented in the form of Fourier components:

$$\boldsymbol{E}(\boldsymbol{r}, t) = \sum_a \boldsymbol{E}(\omega_a, \boldsymbol{k}_a) \exp\{i(\boldsymbol{k}_a \cdot \boldsymbol{r} - \omega_a t)\}, \tag{13}$$

where ω_a is the circular oscillation frequency and $\mathbf{k}_a$ the wave vector of the a-th mode. With the aim of obtaining real quantities, summation in (13) is extended over all frequencies and wave vectors, both positive and negative, with $\omega_{-a} = -\omega_a$, $\mathbf{k}_{-a} = -\mathbf{k}_a$ and, for the Fourier transform (field amplitude), $\mathbf{E}^*(\omega_a, \mathbf{k}_a) = \mathbf{E}(-\omega_a, -\mathbf{k}_a)$.

Provided the field strengths $\mathbf{E}$ and $\mathbf{H}$ of the electromagnetic wave are small, the electric polarization induced in the medium is linear and additive as the vector (13):

$$\mathbf{P}_e^{(1)}(\mathbf{r}, t) = \sum_a \mathbf{P}_e^{(1)}(\omega_a, \mathbf{k}_a) \exp\{i(\mathbf{k}_a \cdot \mathbf{r} - \omega_a t)\} \tag{14}$$

where the component of dipolar polarization induced at frequency ω_a is:

$$\mathbf{P}_e^{(1)}(\omega_a, \mathbf{k}_a) = \chi_{ee}^{(1)}(\omega_a; \mathbf{k}_a) \cdot \mathbf{E}(\omega_a, \mathbf{k}_a) + \chi_{em}^{(1)}(\omega_a; \mathbf{k}_a) \cdot \mathbf{H}(\omega_a, \mathbf{k}_a). \tag{15}$$

The second-rank polar tensor $\chi_{ee}^{(1)}(\omega_a, \mathbf{k}_a)$ defines linear (*i.e.* first-order) electric susceptibility, the dependence on ω_a defining frequency dispersion and the dependence on the wave vector $\mathbf{k}_a$ spatial dispersion [4]. The (polar-axial) pseudo-tensor $\chi_{em}^{(1)}(\omega_a, \mathbf{k}_a)$ defines linear magneto-electric susceptibility. Explicitly, the dependence of the tensors on ω_a and $\mathbf{k}_a$ are found by methods of quantum mechanics [8] on resorting to a Hamiltonian of the multipole form [10]:

$$H = H_0 - \sum_{n=1}^{\infty} \frac{2^n n!}{2n!} \{\mathbf{M}_e^{(n)}[n]\mathbf{E}^{(n)}(\mathbf{r}, t) + \mathbf{M}_m^{(n)}[n]\mathbf{H}^{(n)}(\mathbf{r}, t)\} - \ldots, \tag{16}$$

where

$$\mathbf{E}^{(n)}(\mathbf{r}, t) = \nabla^{n-1}\mathbf{E}(\mathbf{r}, t) \text{ and } \mathbf{H}^{(n)}(\mathbf{r}, t) = \nabla^{n-1}\mathbf{H}(\mathbf{r}, t).$$

With regard to Eqs (13)–(16), we derive the following tensor of linear magneto-electric susceptibility with multipole spatial dispersion taken into account:

$$\chi_{em}^{(1)}(\omega_a; \mathbf{k}_a) = \sum_{n_a=1}^{\infty} i^{n_a-1} \frac{2^{n_a} n_a!}{2n_a!} \chi_{em}^{(n_a)}(\omega_a)\,[n_a-1]\mathbf{k}_a^{n_a-1}, \tag{17}$$

where we have the component

$$\chi_{em}^{(n_a)}(\omega_a) = \frac{\varrho}{\hbar} \sum_{klr} \varrho_{kl} \left\{ \frac{\langle k|\mathbf{M}_e^{(1)}|r\rangle \langle r|\mathbf{M}_m^{(n_a)}|l\rangle}{\omega_{rl}+\omega_a+i\Gamma_{rl}} + \frac{\langle k|\mathbf{M}_m^{(n_a)}|r\rangle \langle r|\mathbf{M}_e^{(1)}|l\rangle}{\omega_{rk}-\omega_a-i\Gamma_{rk}} \right\} \tag{18}$$

dependent on spatial dispersion only. Above, ϱ is the number density of the medium and ϱ_{kl} the statistical matrix for the transition from quantum state $|k\rangle$ to state $|l\rangle$ with Bohr frequency ω_{kl} and relaxation time Γ_{kl}^{-1}.

For the case of weak spatial dispersion, we write by (17) in satisfactory approximation [4]:

$$\chi_{em}^{(1)}(\omega_a; \boldsymbol{k}_a) = \chi_{em}^{(1)}(\omega_a) + i\chi_{em}^{(2)}(\omega_a) \cdot \boldsymbol{k}_a - \chi_{em}^{(3)}(\omega_a) : \boldsymbol{k}_a\boldsymbol{k}_a + \dots \tag{17a}$$

where the second-rank tensor $\chi_{em}^{(1)}$ defines the linear electric dipolar susceptibility in magnetic dipole approximation (in the absence of spatial dispersion), the third-rank tensor $\chi_{em}^{(2)}$ describes the magnetic quadrupole contribution, the fourth-rank tensor $\chi_{em}^{(3)}$ — the magnetic octupole contribution, and so forth.

4. Magneto-optical processes of order 2

At sufficiently high electromagnetic field strengths, second-order polarization for which we have:

$$\boldsymbol{P}_e^{(2)}(\boldsymbol{r}, t) = \tfrac{1}{2}\sum_{ab} \boldsymbol{P}_e^{(2)}(\omega_a+\omega_b, \boldsymbol{k}_a+\boldsymbol{k}_b) \exp\{i[(\boldsymbol{k}_a+\boldsymbol{k}_b)\cdot \boldsymbol{r} - (\omega_a+\omega_b)t]\} \tag{19}$$

has to be taken into account. Here, the magneto-optical polarization component at frequency $\omega_a+\omega_b$ of interest to us is:

$$\boldsymbol{P}_e^{(2)}(\omega_a+\omega_b, k_a+k_b) = \chi_{eem}^{(2)}(\omega_a, \omega_b; \boldsymbol{k}_a, \boldsymbol{k}_b) : \boldsymbol{E}(\omega_a, \boldsymbol{k}_a)\boldsymbol{H}(\omega_a, \boldsymbol{k}_b). \tag{20}$$

Above, the third-rank pseudo-tensor $\chi_{eem}^{(2)}(\omega_a, \omega_b; \boldsymbol{k}_a, \boldsymbol{k}_b)$ describes the (second-order) nonlinear magneto-optical susceptibility, given by the expression:

$$\chi_{eem}^{(2)}(\omega_a, \omega_b; \boldsymbol{k}_a, \boldsymbol{k}_b) = \sum_{n_a=1}^{\infty}\sum_{n_b=1}^{\infty} i^{n_a+n_b-2} \frac{2^{n_a+n_b} n_a! n_b!}{2n_a! 2n_b!} \times$$
$$\times \chi_{eem}^{(n_a+n_b)}(\omega_a, \omega_b)\, [n_a+n_b-2] \boldsymbol{k}_a^{n_a-1} \boldsymbol{k}_b^{n_b-1}, \tag{21}$$

where the quantum-mechanical form of the component accounting for spatial dispersion is:

$$\chi_{eem}^{(n_a+n_b)}(\omega_a, \omega_b) = \frac{\varrho}{\hbar^2} S(a, b) \sum_{klrs} \varrho_{kl} \left\{ \frac{\langle k|\boldsymbol{M}_e^{(1)}|r\rangle \langle r|\boldsymbol{M}_e^{(n_a)}|s\rangle \langle s|\boldsymbol{M}_m^{(n_b)}|l\rangle}{(\omega_{rl}+\omega_a+\omega_b+i\Gamma_{rl})(\omega_{sl}+\omega_b+i\Gamma_{sl})} + \right.$$
$$+ \frac{\langle k|\boldsymbol{M}_e^{(n_a)}|r\rangle \langle r|\boldsymbol{M}_e^{(1)}|s\rangle \langle s|\boldsymbol{M}_m^{(n_b)}|l\rangle}{(\omega_{rk}-\omega_a-i\Gamma_{rk})(\omega_{sl}+\omega_b+i\Gamma_{sl})} +$$
$$\left. + \frac{\langle k|\boldsymbol{M}_e^{(n_a)}|r\rangle \langle r|\boldsymbol{M}_m^{(n_b)}|s\rangle \langle s|\boldsymbol{M}_e^{(1)}|l\rangle}{(\omega_{rk}-\omega_a-i\Gamma_{rk})(\omega_{sk}-\omega_a-\omega_b-i\Gamma_{sk})} \right\}; \tag{22}$$

$S(a, b, \dots)$ denoting a symmetrizing operator, consisting in summation over all permutations of $a, b \dots$.

The polarization (20) describes the magneto-optical contribution to bulk second--harmonic generation (especially at metal surfaces) discussed by Jha [11]. In the DC magnetic field case ($\omega_b = 0$, $\boldsymbol{k}_b = 0$), Eq. (20) describes magneto-optical rotation *i. e.* linear Faraday effect.

Likewise to (19), one can write the following second-order magnetic polarization, where the component at frequency $\omega_a+\omega_b$ of interest to us is:

$$\boldsymbol{P}_m^{(2)}(\omega_a+\omega_b;\boldsymbol{k}_a+\boldsymbol{k}_b) = \chi_{mee}^{(2)}(\omega_a+\omega_b;\boldsymbol{k}_a+\boldsymbol{k}_b):\boldsymbol{E}(\omega_a,\boldsymbol{k}_a)\boldsymbol{E}(\omega_b,\boldsymbol{k}_b), \tag{23}$$

with the nonlinear electro-magnetic susceptibility pseudo-tensor $\chi_{mee}^{(2)}(\omega_a+\omega_b;\boldsymbol{k}_a+\boldsymbol{k}_b)$ defined similarly to (21) in conjunction with (22), where $\boldsymbol{M}_e^{(1)}$ has to be replaced by $\boldsymbol{M}_m^{(1)}$ and $\boldsymbol{M}_m^{(n_b)}$ by $\boldsymbol{M}_e^{(n_b)}$.

In the particular case when $\omega_a = -\omega_b$ and $\boldsymbol{k}_a = -\boldsymbol{k}_b$, Eq. (23) describes the magnetization induced by intense light observed by Pershan *et al.* [16] as inverse Faraday effect.

5. *Third-order magneto-optical processes*

In third-order approximation, the electric dipole polarization assumes the form:

$$\boldsymbol{P}_e^{(3)}(\boldsymbol{r},t) = \frac{1}{6}\sum_{abc}\boldsymbol{P}_e^{(3)}(\omega_a+\omega_b+\omega_c;\boldsymbol{k}_a+\boldsymbol{k}_b+\boldsymbol{k}_c)\exp\{i[(\boldsymbol{k}_a+\boldsymbol{k}_b+\boldsymbol{k}_c)\cdot\boldsymbol{r}- -(\omega_a+\omega_b+\omega_c)t]\}. \tag{24}$$

$\boldsymbol{P}_e^{(3)}$ consists in general of 8 components. Here, however, we shall restrict ourselves to writing out the only one of relevance to our considerations, namely:

$$\boldsymbol{P}_e^{(3)}(\omega_a+\omega_b+\omega_c;\boldsymbol{k}_a+\boldsymbol{k}_b+\boldsymbol{k}_c) = = \chi_{eeem}^{(3)}(\omega_a,\omega_b,\omega_c,\boldsymbol{k}_a,\boldsymbol{k}_b,\boldsymbol{k}_c)\vdots\boldsymbol{E}(\omega_a,\boldsymbol{k}_a)\boldsymbol{E}(\omega_b,\boldsymbol{k}_b)\boldsymbol{H}(\omega_c,\boldsymbol{k}_c) \tag{25}$$

where the fourth-rank axial tensor defining the third-order magneto-optical suscepti bility is, in the presence of spatial dispersion, of the form:

$$\chi_{eeem}^{(3)}(\omega_a,\omega_b,\omega_c;\boldsymbol{k}_a,\boldsymbol{k}_b,\boldsymbol{k}_c) = = \sum_{n_a=1}^{\infty}\sum_{n_b=1}^{\infty}\sum_{n_c=1}^{\infty} i^{n_a+n_b+n_c-3}\frac{2^{n_a+n_b+n_c}n_a!n_b!n_c!}{2n_a!2n_b!2n_c!}\chi_{eeem}^{(n_a+n_b+n_c)}(\omega_a,\omega_b,\omega_c)\,[n_a+ +n_b+n_c-3]\boldsymbol{k}_a^{n_a-1}\boldsymbol{k}_b^{n_b-1}\boldsymbol{k}_c^{n_c-1}. \tag{26}$$

The quantum mechanical form of the third-order magneto-electric multipole susceptibility tensor is:

$$\chi_{eeem}^{(n_a+n_b+n_c)}(\omega_a, \omega_b, \omega_c) =$$

$$= \frac{\varrho}{\hbar^3} S(a, b, c) \sum_{klrst} \varrho_{kl} \left\{ \frac{\langle k|M_e^{(1)}|r\rangle \langle r|M_e^{(n_a)}|s\rangle \langle s|M_e^{(n_b)}|t\rangle \langle t|M_m^{(n_c)}|l\rangle}{(\omega_{rl}+\omega_a+\omega_b+\omega_c+i\Gamma_{rl})(\omega_{sl}+\omega_b+\omega_c+i\Gamma_{sl})(\omega_{tl}+\omega_c+i\Gamma_{tl})} + \right.$$

$$+ \frac{\langle k|M_e^{(n_a)}|r\rangle \langle r|M_e^{(1)}|s\rangle \langle s|M_e^{(n_b)}|t\rangle \langle t|M_m^{(n_c)}|l\rangle}{(\omega_{rk}-\omega_a-i\Gamma_{rk})(\omega_{sl}+\omega_b+\omega_c+i\Gamma_{sl})(\omega_{tl}+\omega_c+i\Gamma_{tl})} +$$

$$+ \frac{\langle k|M_e^{(n_a)}|r\rangle \langle r|M_e^{(n_b)}|s\rangle \langle s|M_e^{(1)}|t\rangle \langle t|M_m^{(n_c)}|l\rangle}{(\omega_{rk}-\omega_a-i\Gamma_{rk})(\omega_{sk}-\omega_a-\omega_b-i\Gamma_{sk})(\omega_{tl}+\omega_c+i\Gamma_{tl})} +$$

$$\left. + \frac{\langle k|M_e^{(n_a)}|r\rangle \langle r|M_e^{(n_b)}|s\rangle \langle s|M_m^{(n_c)}|t\rangle \langle t|M_e^{(1)}|l\rangle}{(\omega_{rk}-\omega_a-i\Gamma_{rk})(\omega_{sk}-\omega_a-\omega_b-i\Gamma_{sk})(\omega_{tk}-\omega_a-\omega_b-\omega_c-i\Gamma_{tk})} \right\}. \tag{27}$$

Nonlinear magneto-electric polarization, as given by Eq. (25), in general describes a process of interaction of three waves in a medium leading to the emergence of a fourth wave, of the frequency $\omega_a+\omega_b+\omega_c$. At $\omega_c = \boldsymbol{k}_c = 0$, one has mixing of two waves in the presence of a DC magnetic field, or, in particular, DC magnetic field-induced SHG, if $\omega_a = \omega_b = \omega$ and $\omega_c = 0$ [12, 27, 31]. On putting $\omega_a = \omega$ and $\omega_b = \omega_c = \boldsymbol{k}_a = \boldsymbol{k}_c = 0$ in (25), one comes upon a new effect, consisting in optical rotation in crossed DC electric $\boldsymbol{E}(0)$ and magnetic $\boldsymbol{H}(0)$ fields [32].

6. *Fourth-order magneto-optical processes*

In centro-symmetric bodies, the third-order magneto-optical polarization (25) vanishes, and we have to proceed to the next, higher approximation of order 4:

$$\boldsymbol{P}_e^{(4)}(\boldsymbol{r}, t) = \frac{1}{24} \sum_{abcd} \boldsymbol{P}_e^{(3)}(\omega_a+\omega_b+\omega_c+\omega_d; \boldsymbol{k}_a+\boldsymbol{k}_b+\boldsymbol{k}_c+\boldsymbol{k}_d) \exp\{i[(\boldsymbol{k}_a+ \\ +\boldsymbol{k}_b+\boldsymbol{k}_c+\boldsymbol{k}_d)\cdot\boldsymbol{r}-(\omega_a+\omega_b+\omega_c+\omega_d)t]\}. \tag{28}$$

On restricting ourselves to the magneto-optical component linear in the magnetic field strength, we have, at the frequency $\omega_a+\omega_b+\omega_c+\omega_d$:

$$\boldsymbol{P}_e^{(4)}(\omega_a+\omega_b+\omega_c+\omega_d; \boldsymbol{k}_a+\boldsymbol{k}_b+\boldsymbol{k}_c+\boldsymbol{k}_d) =$$

$$= \chi_{eeeem}^{(4)}(\omega_a, \omega_b, \omega_c, \omega_d; \boldsymbol{k}_a, \boldsymbol{k}_b, \boldsymbol{k}_c, \boldsymbol{k}_d) \vdots\vdots \boldsymbol{E}(\omega_a, \boldsymbol{k}_a)\boldsymbol{E}(\omega_b, \boldsymbol{k}_b)\boldsymbol{E}(\omega_c, \boldsymbol{k}_c)\boldsymbol{H}(\omega_d, \boldsymbol{k}_d), \tag{29}$$

with the fifth-rank axial tensor, defining fourth-order magneto-electric susceptibility, in the form:

$$\chi_{eeeem}^{(4)}(\omega_a, \omega_b, \omega_c, \omega_d; \boldsymbol{k}_a, \boldsymbol{k}_b, \boldsymbol{k}_c, \boldsymbol{k}_d) = \sum_{n_a=1}^{\infty} \sum_{n_b=1}^{\infty} \sum_{n_c=1}^{\infty} \sum_{n_d=1}^{\infty} i^{n_a+n_b+n_c+n_d-4} \times$$

$$\times \frac{2^{n_a+n_b+n_c+n_d} n_a! n_b! n_c! n_d!}{2n_a! 2n_b! 2n_c! 2n_d!} \chi_{eeeem}^{(n_a+n_b+n_c+n_d)}(\omega_a, \omega_b, \omega_c, \omega_d) \times$$

$$\times [n_a+n_b+n_c+n_d-4] \boldsymbol{k}_a^{n_a-1} \boldsymbol{k}_b^{n_b-1} \boldsymbol{k}_c^{n_c-1} \boldsymbol{k}_d^{n_d-1}. \tag{30}$$

Fourth-order quantum mechanical calculation, taking into account all multipole contributions, yields:

$$\chi_{eeeem}^{(n_a+n_b+n_c+n_d)}(\omega_a, \omega_b, \omega_c, \omega_d) = \frac{\varrho}{\hbar^4} S(a, b, c, d) \sum_{klrstu} \varrho_{kl} \times$$

$$\times \left\{ \frac{\langle k|M_e^{(1)}|r\rangle \langle r|M_e^{(n_a)}|s\rangle \langle s|M_e^{(n_b)}|t\rangle \langle t|M_e^{(n_c)}|u\rangle \langle u|M_m^{(n_d)}|l\rangle}{(\omega_{rl}+\omega_a+\omega_b+\omega_c+\omega_d+i\Gamma_{rl})(\omega_{sl}+\omega_b+\omega_c+\omega_d+i\Gamma_{sl})(\omega_{tl}+\omega_c+\omega_d+i\Gamma_{tl})(\omega_{ul}+\omega_d+i\Gamma_{ul})} + \right.$$

$$+ \frac{\langle k|M_e^{(n_a)}|r\rangle \langle r|M_e^{(1)}|s\rangle \langle s|M_e^{(n_b)}|t\rangle \langle t|M_e^{(n_c)}|u\rangle \langle u|M_m^{(n_d)}|l\rangle}{(\omega_{rk}-\omega_a-i\Gamma_{rk})(\omega_{sl}+\omega_b+\omega_c+\omega_d+i\Gamma_{sl})(\omega_{tl}+\omega_c+\omega_d+i\Gamma_{tl})(\omega_{ul}+\omega_d+i\Gamma_{ul})} +$$

$$+ \frac{\langle k|M_e^{(n_a)}|r\rangle \langle r|M_e^{(n_b)}|s\rangle \langle s|M_e^{(1)}|t\rangle \langle t|M_e^{(n_c)}|u\rangle \langle u|M_m^{(n_d)}|l\rangle}{(\omega_{rk}-\omega_a-i\Gamma_{rk})(\omega_{sk}-\omega_a-\omega_b-i\Gamma_{sk})(\omega_{tl}+\omega_c+\omega_d+i\Gamma_{tl})(\omega_{ul}+\omega_d+i\Gamma_{ul})} +$$

$$+ \frac{\langle k|M_e^{(n_a)}|r\rangle \langle r|M_e^{(n_b)}|s\rangle \langle s|M_e^{(n_c)}|t\rangle \langle t|M_e^{(1)}|u\rangle \langle u|M_m^{(n_d)}|l\rangle}{(\omega_{rk}-\omega_a-i\Gamma_{rk})(\omega_{sk}-\omega_a-\omega_b-i\Gamma_{sk})(\omega_{tk}-\omega_a-\omega_b-\omega_c-i\Gamma_{tk})(\omega_{ul}+\omega_d+i\Gamma_{ul})} +$$

$$\left. + \frac{\langle k|M_e^{(n_a)}|r\rangle \langle r|M_e^{(n_b)}|s\rangle \langle s|M_e^{(n_c)}|t\rangle \langle t|M_m^{(n_d)}|u\rangle \langle u|M_e^{(1)}|l\rangle}{(\omega_{rk}-\omega_a-i\Gamma_{rk})(\omega_{sk}-\omega_a-\omega_b-i\Gamma_{sk})(\omega_{tk}-\omega_a-\omega_b-\omega_c-i\Gamma_{tk})(\omega_{ul}-\omega_a-\omega_b-\omega_c-\omega_d-i\Gamma_{uk})} \right\}. \tag{31}$$

The magneto-electric mixing processes of four waves in a medium described by Eqs (28) and (29) lead in particular at $\omega_a = \omega_b = \omega_c$ and $\omega_d = \boldsymbol{k}_d = 0$ to the Third Harmonic Generation in the presence of a magnetic field observed in semiconductors [19] and discussed theoretically taking into account the nonparabolic energy band [17, 18]. At $\omega_c = \omega_d = \boldsymbol{k}_c = \boldsymbol{k}_d = 0$, Eq. (29) describes frequency mixing and, at $\omega_a = \omega_b$, SHG in crossed DC electric and magnetic fields [33]. In particular at $\omega_a = \omega$, $\omega_b = -\omega_c = \omega_L$ and $\omega_d = \boldsymbol{k}_d = 0$, Eq. (29) describes the influence of intense laser light on Faraday's effect [21] recently observed by Kubota [22] in semiconductor CdS and ZnS single crystals. At $\omega_a = \omega$ and $\omega_b = \omega_c = \omega_d = 0$, $\boldsymbol{k}_b = \boldsymbol{k}_c = \boldsymbol{k}_d = 0$, we have a new nonlinear magneto-optical effect, consisting in DC electric field-changes in magneto-optical rotation.

In the case when $\omega_d = \boldsymbol{k}_d = 0$, and $\omega_a = \omega_b = -\omega_c$, one has the two-photon absorption in a DC magnetic field observed in semiconductors [20, 22] and discussed theoretically for various models [17]. In general, Eqs (28)–(31) describe new nonlinear magneto-optical processes of the Raman type.

7. *Nonzero and mutually independent tensor elements of magneto-optical susceptibility*

The higher order magneto-optical processes discussed in the preceding Sections involve the presence of a body, possessing some kind of crystallographical symmetry. The processes (effects) will, consequently, run variously from one case to another depending on which of the nonlinear susceptibility tensor elements $\chi^{(2)}_{eem}$, $\chi^{(3)}_{eeem}$, and $\chi^{(4)}_{eeeem}$ do not vanish for the crystallographical class in occurrence. With the aim of calculating (determining) these tensor elements, we now go over from vectorial notation to indicial notation of the tensors. Thus,

in place of $\chi^{(2)}_{eem}$ we write $\chi^{eem}_{i,j,k}$,

in place of $\chi^{(3)}_{eeem}$ we write $\chi^{eeem}_{i,j,k,l}$, and

in place of $\chi^{(4)}_{eeeem}$ we write $\chi^{eeeem}_{i,j,k,l,n}$.

The commas between the indices i, j, k, l, n... (which run through the values x, y, z of laboratory axes) signify that the tensors possess no permutational symmetry with respect to the indices. If a tensor is symmetric in a pair of indices i, j, we write (i,j), whereas if it is antisymmetric, we write $[ij]$. By resorting to methods of group theory [34, 35], we derived relations between the nonzero axial tensor elements $\chi^{eem}_{i,j,k}$, $\chi^{eeem}_{i,j,k,l}$ and $\chi^{eeeem}_{i,j,k,l,n}$, not possessing permutational symmetry in the 32 crystallographical classes, 2 icosahedral classes (Y and Y_h), and isotropic medium (K and K_h). Moreover, we calculated the numbers of mutually independent and nonzero elements of these tensors as well as of other axial tensors of ranks 3, 4 and 5 describing the nonlinear magnetic susceptibilities and presenting the following permutational symmetries (Table I):

a) 3-rd rank axial tensors: $\chi^{eem}_{ij,k} = \chi^{eem}_{ji,k} \equiv \chi^{eem}_{(ij)k}$,

$$\chi^{eem}_{ij,k} = -\chi^{eem}_{ji,k} \equiv \chi^{eem}_{[ij]k} .$$

b) 4-th rank axial tensors: $\chi^{eeem}_{ij,k,l} = \chi^{eeem}_{ji,k,l} \equiv \chi^{eeem}_{(ij)k,l}$,

$$\chi^{eeem}_{ij,k,l} = -\chi^{eeem}_{ji,k,l} \equiv \chi^{eeem}_{[ij]k,l}, \quad \chi^{eeem}_{ijk,l} \equiv \chi^{eeem}_{(ijk)l} \text{ (symmetric in } i, j, k),$$

c) 5-th rank axial tensors: $\chi^{eeeem}_{ij,k,l,n} = \chi^{eeeem}_{ji,k,l,n} \equiv \chi^{eeeem}_{(ij)k,l,n}$,

$$\chi^{eeeem}_{ij,k,l,n} = -\chi^{eeeem}_{ji,k,l,n} \equiv \chi^{eeeem}_{[ij]k,l,n} ,$$

$$\chi^{eeeem}_{ij,kl,n} = \chi^{eeeem}_{ji,kl,n} = \chi^{eeeem}_{ij,lk,n} = \chi^{eeeem}_{ji,lk,n} = \chi^{eeeem}_{(ij)(kl)n} ,$$

$$\chi^{eeeem}_{ij,kl,n} = -\chi^{eeeem}_{ji,kl,n} = \chi^{eeeem}_{ij,lk,n} = -\chi^{eeeem}_{ji,lk,n} = \chi^{eeeem}_{[ij](kl)n} ,$$

$$\chi^{eeeem}_{ijk,l,n} \equiv \chi^{eeeem}_{(ijk)l,n} \text{ (symmetric in } i, j, k\text{), and}$$

$$\chi^{eeeem}_{ijkl,n} \equiv \chi^{eeeem}_{(ijkl)n} \text{ (symmetric in } i, j, k, l).$$

The number of independent elements of an axial tensor of arbitrary rank, for the group $G(g_1, g_2, ..., g_N)$, is given by the formula [37]:

$$\nu = \frac{1}{N} \sum_{g_r \in G} \chi^0(g_r)\chi_A(g_r), \tag{32}$$

The number of non-zero (N) and independent (I) elements of nonlinear

Class	$\chi^{eem}_{(ij)k}$		$\chi^{eem}_{[ij]k}$		$\chi^{eeem}_{(ij)k,l}$		$\chi^{eeem}_{[ij]k,l}$		$\chi^{eeem}_{(ijk)l}$	
	N	I	N	I	N	I	N	I	N	I
1 (C_1)	27	18	18	9	81	54	54	27	81	30
$\bar{1}$ (C_i)	27	18	18	9	0	0	0	0	0	0
m (C_s)	13	8	10	5	40	26	28	14	40	14
2 (C_2)	13	8	10	5	41	28	26	13	41	16
2/m (C_{2h})	13	8	10	5	0	0	0	0	0	0
222 (D_2)	6	3	6	3	21	15	12	6	21	9
mm2 (C_{2v})	6	3	6	3	20	13	14	7	20	7
mmm (D_{2h})	6	3	6	3	0	0	0	0	0	0
4 (C_4)	11	4	10	3	39	14	26	7	35	8
$\bar{4}$ (S_4)	11	4	10	3	40	14	24	6	40	8
4/m (C_{4h})	11	4	10	3	0	0	0	0	0	0
422 (D_4)	4	1	6	2	21	8	12	3	21	5
4mm (C_{4v})	4	1	6	2	18	6	14	4	14	3
$\bar{4}$2m (D_{2d})	4	1	6	2	20	7	12	3	20	4
4/mmm (D_{4h})	4	1	6	2	0	0	0	0	0	0
3 (C_3)	19	6	10	3	71	18	42	9	67	10
$\bar{3}$ (S_6)	19	6	10	3	0	0	0	0	0	0
32 (D_3)	8	2	6	2	37	10	20	4	37	6
3m (C_{3v})	8	2	6	2	34	8	22	5	30	4
3m (D_{3d})	8	2	6	2	0	0	0	0	0	0
6 (C_6)	11	4	10	3	39	12	26	7	35	6
$\bar{6}$ (C_{3h})	11	4	10	3	32	6	16	2	32	4
6/m (C_{6h})	11	4	10	3	0	0	0	0	0	0
622 (D_6)	4	1	6	2	21	7	12	3	21	4
6mm (C_{6v})	4	1	6	2	18	5	14	4	14	2
$\bar{6}$m2 (D_{3h})	4	1	6	2	16	3	8	1	16	2
6/mmm (D_{6h})	4	1	6	2	0	0	0	0	0	0
23 (T)	6	1	6	1	21	5	12	2	21	3
m3 (T_h)	6	1	6	1	0	0	0	0	0	0
432 (O)	0	0	6	1	21	3	12	1	21	2
$\bar{4}$3m (T_d)	0	0	6	1	18	2	12	1	18	1
m3m (O_h)	0	0	6	1	0	0	0	0	0	0
Y	0	0	6	1	21	2	12	1	21	1
Y_h	0	0	6	1	0	0	0	0	0	0
K	0	0	6	1	21	2	12	1	21	1
K_h	0	0	6	1	0	0	0	0	0	0

susceptibility axial tensors χ^{eem}_{ijk}, χ^{eeem}_{ijkl} and χ^{eeeem}_{ijklm} TABLE I

$\chi^{eeeem}_{(ij)k,l,n}$		$\chi^{eeeem}_{[ij]k,l,n}$		$\chi^{eeeem}_{(ij)(kl)n}$		$\chi^{eeeem}_{[ij](kl)n}$		$\chi^{eeeem}_{(ijk)l,n}$		$\chi^{eeeem}_{(ijkl)n}$	
N	I	N	I	N	I	N	I	N	I	N	I
243	162	162	81	243	108	162	54	243	90	243	45
243	162	162	81	243	108	162	54	243	90	243	45
121	80	82	41	121	52	82	28	121	44	121	21
121	80	82	41	121	52	82	28	121	44	121	21
121	80	82	41	121	52	82	28	121	44	121	21
60	39	42	21	60	24	42	15	60	21	60	9
60	39	42	21	60	24	42	15	60	21	60	9
60	39	42	21	60	24	42	15	60	21	60	9
119	40	82	21	117	26	78	14	115	22	109	11
119	40	82	21	117	26	78	14	115	22	109	11
119	40	82	21	117	26	78	14	115	22	109	11
58	19	42	11	56	11	42	8	54	10	48	4
58	19	42	11	56	11	42	8	54	10	48	4
58	19	42	11	56	11	42	8	54	10	48	4
58	19	42	11	56	11	42	8	54	10	48	4
231	54	146	27	229	36	142	18	227	30	213	15
231	54	146	27	229	36	142	18	227	30	213	15
114	26	74	14	112	16	74	10	110	14	96	6
114	26	74	14	112	16	74	10	110	14	96	6
114	26	74	14	112	16	74	10	110	14	96	6
119	32	82	19	117	20	78	12	115	16	101	7
119	32	82	19	117	20	78	12	115	16	101	7
119	32	82	19	117	20	78	12	115	16	101	7
58	15	42	10	56	8	42	7	54	7	40	2
58	15	42	10	56	8	42	7	54	7	40	2
58	15	42	10	56	8	42	7	54	7	40	2
58	15	42	10	56	8	42	7	54	7	40	2
60	13	42	7	60	8	42	5	60	7	60	3
60	13	42	7	60	8	42	5	60	7	60	3
54	6	42	4	48	3	42	3	42	3	24	1
54	6	42	4	48	3	42	3	42	3	24	1
54	6	42	4	48	3	42	3	42	3	24	1
54	3	42	3	48	1	42	2	42	1	0	0
54	3	42	3	48	1	42	2	42	1	0	0
54	3	42	3	48	1	42	2	42	1	0	0
54	3	42	3	48	1	42	2	42	1	0	0

where χ_A as well as

$$\chi^0(g_r) = \frac{1}{N_P} \sum_{p\in P} (-1)^q \chi(g_r^{l_1})\chi(g_r^{l_2})\dots\chi(g_r^{l_\sigma}) \tag{33}$$

and

$$\chi(g_r) = \pm 1 + 2\cos\varphi_r \tag{34}$$

denote characters of the element g_r in the irreducible antisymmetric vector representation of the group G, reducible tensorial representation taking into account the permutational symmetry group of indices $P(p_1, p_2, \dots p_{N_P})$, and reducible vectorial representation of the group G. Summation in (32) runs through all elements of the group G (of which there are N in all), and in (33) — through all N_P elements of the permutation group of the tensor indices. By $l_1, l_2, \dots, l_\sigma$, we denote the lengths of the cycles into which the permutation of tensor indices $p \in P$, being the element of the group P, decomposes, whereas q is a factor, taking the following 2 values: if the permutation $p \in P$ leaves the sign of the tensor element unchanged, then $q = 2$; if it does not, then $q = 1$. The signs "+" and "−" in (34) are, respectively, for proper and improper rotations by the angle φ_r.

By using Wigner's projection operator [38]:

$$\hat{O}_u = \frac{f_\alpha}{N} \sum_{g_r\in G} \chi^\alpha_{uu}(g_r)\hat{g}_r \tag{35}$$

we find the tensor elements or linear combinations of elements transforming according to the various irreducible representations of the group G, or in other words, the transformational properties of the tensor. This operator, acting on a linear combination of tensor elements, sifts out therefrom the part belonging to the u-th column of the representation α. The irreducible representation is of dimension f_α, whereas $\chi^\alpha_{uu}(g_r)$ denotes the character of the element g_r of G in the irreducible representation α.

By resorting to (35), we previously determined [36, 39] the transformational properties of the tensors $\chi^{eem}_{i,j,k}$, $\chi^{eeem}_{i,j,k,l}$ and $\chi^{eeeem}_{i,j,k,l,n}$, which served for finding the relations existing between their nonzero elements and for determining the numbers of their nonzero elements. The results thus obtained are listed in Tables II–IV. By taking into account the relevant permutational symmetry in the results of Tables II–IV, one quite easily obtains relations between nonzero elements for the other axial tensors of ranks 3, 4 and 5 describing the nonlinear magneto-optical susceptibilities. In Table I, we list the numbers of independent and nonzero elements of the axial tensors of ranks 3, 4 and 5 describing those susceptibilities.

8. *Nonlinear magneto-optical birefringence and rotation*

For an anisotropic medium the electric and magnetic properties of which are described, respectively, by the second-rank electric permittivity tensor $\boldsymbol{\varepsilon}$ (element ε_{ij}) and magnetic permittivity tensor $\boldsymbol{\mu}$ (element μ_{ij}), we have the following fundamental relations:

$$\boldsymbol{D}(\boldsymbol{r}, t) = \boldsymbol{\varepsilon} \cdot \boldsymbol{E}(\boldsymbol{r}, t), \qquad \boldsymbol{B}(\boldsymbol{r}, t) = \boldsymbol{\mu} \cdot \boldsymbol{H}(\boldsymbol{r}, t), \tag{36}$$

TABLE II

Non-zero and independent elements of the axial tensor $\chi^{eem}_{i,j,k}$ for all crystallographical classes. The tensor elements are denoted by their subscripts x, y, z only

Class	Number of non-zero elements	Number of independent elements	Elements
1 (C_1) $\bar{1}$ (C_i)	27	27	zzz, zxx, xxz, xzx, zyy, yyz, yzy, xxx, xxy, yxx, xyx, xyy, yyx, yxy, yyy, xzz, zzx, zxz, yzz, zzy, zyz, xyz, xzy, yxz, yzx, zxy, zyx,
m (C_s) 2 (C_2) 2/m (C_{2h})	13	13	zzz, zxx, xxz, xzx, zyy, yyz, yzy, xyz, xzy, yxz, yzx, zxy, zyx
222 (D_2) mm2 (C_{2v}) mmm (D_{2h})	6	6	xyz, xzy, yxz, yzx, zxy, zyx
4 (C_4) $\bar{4}$ (S_4) 4/m (C_{4h}) 6 (C_6) $\bar{6}$ (C_{3h}) 6/m (C_{6h})	13	7	zzz, zxx = zyy, xxz = yyz, xzx = yzy, xyz = yxz, yzx =-xzy, zxy =-zyx
422 (D_4) 4mm (C_{4v}) $\bar{4}$2m (D_{2d}) 4/mmm (D_{4h}) 622 (D_6) 6mm (C_{6v}) $\bar{6}$m2 (D_{3h}) 6/mmm (D_{6h})	6	3	xyz =-yxz, yzx =-xzy, zxy =-zyx,
3 (C_3) $\bar{3}$ (S_6)	21	9	zzz, zxx = zyy, xxz = yyz, xzx = yzy, xyz =-yxz, yzx =-xzy, zxy =-zyx, xxx =-xyy =-yxy =-yyx, yyy =-yxx =-xyx =-xxy,
32 (D_3) 3m (C_{3v}) $\bar{3}$m (D_{3d})	10	4	xxx =-xyy =-yxy =-yyx, xyz =-yxz, yzx =-xzy, zxy =-zyx,
23 (T) m3 (T_h)	6	2	xyz = yzx = zxy, xzy = zyx = yxz,
m3m (O_h) 432 (O) $\bar{4}$3m (T_d) Y Y_h K K_h	6	1	xyz = yzx = zxy =-yxz =-xzy =-zyx.

TABLE III

Non-zero and independent elements of the axial tensor $\chi^{eem}_{i,j,k,l}$ for all crystallographical classes. The tensor elements are denoted by their subscripts x, y, z only. In order to reduce space, elements shared by various classes have been assembled under one capital letter

Class	Number of non-zero elements	Number of independent elements	Elements
1 (C_1)	81	81	A ≡ xxxx, yyyy, zzzz, xxyy, yyxx, xyxy, yxyx, xyyx, yxxy, xxzz, zzxx, xzxz, zxzx, xzzx, zxxz, yyzz, zzyy, yzyz, zyzy, yzzy, zyyz; B ≡ xxxy, xxyx, xyxx, yxxx, yyyx, yyxy, yxyy, xyyy, xzzy, xzyz, xyzz, yzzx, yzxz, yxzz, zzxy, zxzy, zxyz, zzyx, zyzx, zyxz; D ≡ xxxz, xxzx, xzxx, zxxx, zzzx, zzxz, zxzz, xzzz, xyyz, xyzy, xzyy, zyyx, zyxy, zxyy, yyxz, yxyz, yxzy, yyzx, yzyx, yzxy; E ≡ yyyz, yyzy, yzyy, zyyy, zzzy, zzyz, zyzz, yzzz, xxyz, xyxz, xyzx, xxzy, xzxy, xzyx, yzxx, yxzx, yxxz, zyxx, zxyx, zxxy,
$\bar{1}$ (C_i)	0	0	
m (C_s)	40	40	D and E
2 (C_2)	41	41	A and B
2/m (C_{2h})	0	0	
222 (D_2)	21	21	A
mm2 (C_{2v})	20	20	B
mmm (D_{2h})	0	0	
4 (C_4)	41	21	F ≡ xxxx = yyyy, zzzz, xxyy = yyxx, xyxy = yxyx, xyyx = yxxy , xxzz = yyzz, xzxz = yzyz, xzzx = yzzy, zzxx = zzyy, zxzx = zyzy, zxxz = zyyz; G ≡ zzxy =-zzyx, zxyz =-zyxz, xzzy =-yzzx, xyzz =-yxzz, zxzy =-zyzx, xzyz =-yzxz, xxxy =-yyyx, xxyx =-yyxy, xyxx =-yxyy, yxxx =-xyyy,
$\bar{4}$ (S_4)	40	20	H ≡ xxxx =-yyyy, xxyy =-yyxx, xyxy =-yxyx, xyyx =-yxxy, xxzz =-yyzz, xzxz =-yzyz, xzzx =-yzzy, zzxx =-zzyy, zxzx =-zyzy, zxxz =-zyyz, J ≡ zzxy = zzyx, zxyz = zyxz, xzzy = yzzx, xyzz = yxzz, zxzy = zyzx, xzyz = yzxz, xxxy = yyyx, xxyx = yyxy, xyxx = yxyy, yxxx = xyyy,
4/m (C_{4h})	0	0	
422 (D_4)	21	11	F
4mm (C_{4v})	20	10	G
$\bar{4}$2m (D_{2d})	20	10	H
4/mmm (D_{4h})	0	0	

TABLE III (continued)

Class	Number of non-zero elements	Number of independent elements	Elements
$3\ (C_3)$	73	27	L ≡ zzzz, xxxx = yyyy = xxyy + xyxy + xyyx, xxyy = yyxx, xyxy = yxyx, xyyx = yxxy, xxzz = yyzz, xzxz = yzyz, xzzx = yzzy, zzxx = zzyy, zxzx = zyzy, zxxz = zyyz, M ≡ xxxy =-yyyx =-/xxyx + xyxx + yxxx/, xxyx =-yyxy, xyxx =-yxyy, yxxx =-xyyy, zzxy =-zzyx, zxyz =-zyxz, xzzy =-yzzx, xyzz =-yxzz, zxzy =-zyzx, xzyz =-yzxz, P ≡ xxxz =-xyyz =-yxyz =-yyxz, xxzx =-xyzy =-yxzy =-yyzx, xzxx =-xzyy =-yzxy =-yzyx, zxxx =-zxyy =-zyxy =-zyyx, Q ≡ yyyz =-yxxz =-xyxz =-xxyz, yyzy =-yxzx =-xyzx =-xxzy, yzyy =-yzxx =-xzyx =-xzxy, zyyy =-zyxx =-zxyx =-zxxy,
$\bar{3}\ (S_6)$	0	0	
$32\ (D_3)$	37	14	L and Q
$3m\ (C_{3v})$	36	13	M abd P
$\bar{3}m\ (D_{3d})$	0	0	
$6\ (C_6)$	41	19	L and M
$\bar{6}\ (C_{3h})$	32	8	P and Q
$6/m\ (C_{6h})$	0	0	
$622\ (D_6)$	21	10	L
$6mm\ (C_{6v})$	20	9	M
$\bar{6}m2\ (D_{3h})$	16	4	Q
$6/mmm\ (D_{6h})$	0	0	
$23\ (T)$	21	7	xxxx = yyyy = zzzz, xxyy = yyzz = zzxx, yyxx = xxzz = zzyy, xyxy = yzyz = zxzx, yxyx = xzxz = zyzy, xyyx = yzzy = zxxz, yxxy = xzzx = zyyz,
$m3\ (T_h)$	0	0	
$432\ O$	21	4	xxxx = yyyy = zzzz, xxyy = yyzz = zzxx = yyxx = xxzz = zzyy, xyxy = yzyz = zxzx = yxyx = xzxz = zyzy, xyyx = yzzy = zxxz = yxxy = xzzx = zyyz,
$m3m\ (O_h)$	0	0	
$\bar{4}3m\ (T_d)$	18	3	xxyy = yyzz = zzxx =-yyxx =-xxzz =-zzyy, xyxy = yzyz = zxzx =-yxyx =-xzxz =-zyzy, xyyx = yzzy = zxxz =-yxxy =-xzzx =-zyyz,
Y K	21	3	xxxx = yyyy = zzzz = xxyy + xyxy + xyyx, xxyy = yyxx = xxzz = zzxx = yyzz = zzyy, xyxy = yxyx = xzxz = zxzx = yzyz = zyzy, xyyx = yxxy = xzzx = zxxz = yzzy = zyyz,
Y_h K_h	0	0	

TABLE IV

Non-zero and independent elements of the axial tensor $\chi^{eeeem}_{i,j,k,l,n}$ for all crystallographical classes. The tensor elements are denoted by their subscripts x, y, z only. In order to reduce space, elements shared by various classes have been assembled under one capital letter

Class	Number of non-zero elements	Number of independent elements	Elements
1 (C_1) $\bar{1}$ (C_i)	243	243	A_1= zzzzz, xxxxz, xxxzx, xxzxx, xzxxx, zxxxx, yyyyz, yyyzy, yyzyy, yzyyy, zyyyy, zzzxx, zzxzx, zzxxz, zxzxz, zxzzx, zxxzz, xzzzx, xzzxz, xzxzz, xxzzz, zzzyy, zzyzy, zzyyz, zyzyz, zyzzy, zyyzz, yzzzy, yzzyz, yzyzz, yyzzz; B_1= xxxyz, xxxzy, xxyxz, xxyzx, xxzxy, xxzyx, xyxxz, xyxzx, xyzxx, xzxxy, xzxyx, xzyxx, yxxxz, yxxzx, yxzxx, yzxxx, zxxxy, zxxyx, zxyxx, zyxxx, yyyxz, yyyzx, yyxyz, yyxzy, yyzyx, yyzxy, yxyyz, yxyzy, yxzyy, yzyyx, yzyxy, yzxyy, xyyyz, xyyzy, xyzyy, xzyyy, zyyyx, zyyxy, zyxyy, zxyyy; zzzxy, zzzyx, zzxzy, zzxyz, zzyzx, zzyxz, zxzzy, zxzyz, zxyzz, zyzzx, zyzxz, zyxzz, xzzzy, xzzyz, xzyzz, xyzzz, yzzzx, yzzxz, yzxzz, yxzzz; D_1= xxyyz, xxyzy, xxzyy, xyxyz, xyxzy, xyyzx, xyyxz, xyzxy, xyzyx, xzxyy, xzyxy, xzyyx, yxxyz, yxxzy, yxyzx, yxyxz, yxzyx, yxzxy, yyxxz, yyxzx, yyzxx, yzxxy, yzxyx, yzyxx, zxxyy, zxyxy, zxyyx, zyyxx, zyxyx, zyxxy; E_1= xxxxx, yyyyy, xxxxy, xxxyx, xxyxx, xyxxx, yxxxx, yyyyx, yyyxy, yyxyy, yxyyy, xyyyy, zzzzx, zzzxz, zzxzz, zxzzz, xzzzz, zzzzy, zzzyz, zzyzz, zyzzz, yzzzz, xxxyy, xxyxy, xxyyx, xyxyx, xyxxy, xyyxx, yxxxy, yxxyx, yxyxx, yyxxx, yyyxx, yyxyx, yyxxy, yxyxy, yxyyx, yxxyy, xyyyx, xyyxy, xyxyy, xxyyy, xxxzz, xxzxz, xxzzx, xzxzx, xzxxz, xzzxx, zxxxz, zxxzx, zxzxx, zzxxx, yyyzz, yyzyz, yyzzy, yzyzy, yzyyz, yzzyy, zyyyz, zyyzy, zyzyy, zzyyy, xxzzy, xxzyz, xxyzz, xzxzy, xzxyz, xzzyx, xzzxy, xzyxz, xzyzx, xyxzz, xyzxz, xyzzx, zxxzy, zxxyz, zxzyx, zxzxy, zxyzx, zxyxz, zzxxy, zzxyx, zzyxx, zyxxz, zyxzx, zyzxx, yxxzz, yxzxz, yxzzx, yzzxx, yzxzx, yzxxz, yyzzx, yyzxz, yyxzz, yzyzx, yzyxz, yzzxy, yzzyx, yzxyz, yzxzy, yxyzz, yxzyz, yxzzy, zyyzx, zyyxz, zyzxy, zyzyx, zyxzy, zyxyz, zzyyx, zzyxy, zzxyy, zxyyz, zxyzy, zxzyy, xyyzz, xyzyz, xyzzy, xzzyy, xzyyz, xzyzy;
m (C_s) 2 (C_2) 2/m (C_{2h})	121	121	A_1, B_1 and D_1
222 (D_2) mm2 (C_{2v}) mmm (D_{2h})	60	60	B_1

TABLE IV (continued)

Class	Number of non-zero elements	Number of independent elements	Elements

4 (C_4)
$\bar{4}$ (S_4)
4/m (C_{4h})
	121	61	zzzzz,
			xxxxz = yyyyz, xxxzx = yyyzy, xxzxx = yyzyy, xzxxx = yzyyy,
			zxxxx = zyyyy, zzzxx = zzzyy, zzxxz = zzyyz, zzxzx = zzyzy,
			zxzzx = zyzzy, zxxzz = zyyzz, zxzxz = zyzyz, xxzzz = yyzzz,
			xzxzz = yzyzz, xzzxz = yzzyz, xzzzx = yzzzy, xxyyz = yyxz,
			xxyzy = yyxzx, xxzyy = yyzxx, xyxyz = yxyxz, xyxzy = yxyzx,
			xyyxz = yxxyz, xyyzx = yxxzy, xyzyx = yxzxy, xyzxy = yxzyx,
			xzyyx = yzxxy, xzxyy = yzyxx, xzyxy = yzxyx, zxxyy = zyyxx,
			zxyxy = zyxyx, zxyyx = zyxxy;
			
			$F_1 \equiv$ xxxyz =-yyyxz, xxxzy=-yyyzx, xxzxy =-yyzyx, xxyxz =-yyxyz,
			xxyzx =-yyxzy, xxzyx =-yyzxy, xzxyx =-yzyxy, xzyxx =-yzxyy,
			xzxxy =-yzyyx, xyzxx =-yxzyy, xyxzx =-yxyzy, xyxxz =-yxyyz,
			yxxxz =-xyyyz, yxxzx =-xyyzy, yxzxx =-xyzyy, yzxxx =-xzyyy,
			zyxxx =-zxyyy, zxxyx =-zyyxy, zxyxx =-zyxyy, zxxxy =-zyyyx,
			zzzxy =-zzzyx, zzyzx =-zzxzy, zzxyz =-zzyxz, zyzxz =-zxzyz,
			zyxzz =-zxyzz, zyzzx =-zxzzy, xzzzy =-yzzzx, xzzyz =-yzzxz,
			xzyzz =-yzxzz, xyzzz =-yxzzz;

422 (D_4) 4mm (C_{4v}) $\bar{4}$2m (D_{2d}) 4/mmm (D_{4h})	60	30	F_1
3 (C_3) $\bar{3}$ (S_6)	233	81	$G_1 \equiv$ -xxxyz = yyyxz = xxyxz + xyxxz + yxxxz, -xxxzy = yyyzx = xxyzx + xyxzx + yxxzx, -xxzxy = yyzyx = xxzyx + xyzxx + yxzxx, -xzxxy = yzyyx = xzxyx + xzyxx + yzxxx, -zxxxy = zyyyx = zxxyx + zxyxx + zyxxx, xxyxz =-yyxyz, xxyzx =-yyxzy, xxzyx =-yyzxy, xzxyx =-yzyxy, zxxyx =-zyyxy, yxxxz =-xyyyz, yxxzx =-xyyzy, yxzxx =-xyzyy, yzxxx =-xzyyy, zyxxx =-zxyyy, xyxxz =-yxyyz, xyxzx =-yxyzy, xyzxx =-yxzyy, xzyxx =-yzxyy, zxyxx =-zyxyy, zzzxy =-zzzyx, zzyzx =-zzxzy, zzxyz =-zzyxz, zyzxz =-zxzyz, zyxzz =-zxyzz, zyzzx =-zxzzy, xzzzy =-yzzzx, xzzyz =-yzzxz, xzyzz =-yzxzz, xyzzz =-yxzzz; $H_1 \equiv$ xxxzz =-xyyzz =-yxyzz =-yyxzz, xzzxx =-xzzyy =-yzzxy =-yzzyx, xxzxz =-xyzyz =-yxzyz =-yyzxz, zxxzx =-zxyzy =-zyxzy =-zyyzx, xxzzx =-xyzzy =-yxzzy =-yyzzx, zxxxz =-zxyyz =-zyxyz =-zyyxz, xzxxz =-xzyyz =-yzxyz =-yzyxz, zxzxx =-zxzyy =-zyzxy =-zyzyx, xzxzx =-xzyzy =-yzxzy =-yzyzx, zzxxx =-zzxyy =-zzyxy =-zzyyx, xxxxx = $-\frac{1}{3}$ /yyyyx + yyyxy + yyxyy + yxyyy + xyyyy/ , xxxyy = $\frac{1}{3}$ /2yyyyx + 2yyyxy - yyxyy - yxyyy - xyyyy/, xxyxy = $\frac{1}{3}$ /2yyyyx - yyyxy + 2yyxyy - yxyyy - xyyyy /, xxyyx = $\frac{1}{3}$ /-yyyyx + 2yyyxy + 2yyxyy - yxyyy - xyyyy/, xyxxy = $\frac{1}{3}$ /2yyyyx - yyyxy - yyxyy + 2yxyyy - xyyyy /, xyyxx = $\frac{1}{3}$ /-yyyyx - yyyxy + 2yyxyy + 2yxyyy - xyyyy/, xyxyx = $\frac{1}{3}$ /-yyyyx + 2yyyxy - yyxyy + 2yxyyy - xyyyy/, yxxxy = $\frac{1}{3}$ /2yyyyx - yyyxy - yyxyy - yxyyy + 2xyyyy /, yxxyx = $\frac{1}{3}$ /-yyyyx + 2yyyxy - yyxyy - yxyyy + 2xyyyy/, yxyxx = $\frac{1}{3}$ /-yyyyx - yyyxy + 2yyxyy - yxyyy + 2xyyyy/, yyxxx = $\frac{1}{3}$ /-yyyyx - yyyxy - yyxyy + 2yxyyy + 2xyyyy/, yyyyx, yyyxy, yyxyy, yxyyy, xyyyy ;

Class	Number of non-zero elements	Number of independent elements	Elements
			$J_1 \equiv$ zzzzz, zzzxx = zzzyy, zzxxz = zzyyz, zzxzx = zzyzy, zxzzx = zyzzy, zxxzz = zyyzz, zxzxz = zyzyz, xxzzz = yyzzz, xzxzz = yzyzz, xzzxz = yzzyz, xzzzx = yzzzy, xxxxz = yyyyz = xxyyz + xyxyz + xyyxz, xxxzx = yyyzy = xxyzy + xyxzy + xyyzx, xxzxx = yyzyy = xxzyy + xyzxy + xyzyx, xzxxx = yzyyy = xzxyy + xzyxy + xzyyx, zxxxx = zyyyy = zxxyy + zxyxy + zxyyx, xxyyz = yyxxz, xxyzy = yyxzx, xxzyy = yyzxx, xyxyz = yxyxz, xyxzy = yxyzx, xyyxz = yxxyz, xyyzx = yxxzy, xyzyx = yxzxy, xyzxy = yxzyx, xzyyx = yzxxy, xzxyy = yzyxx, xzyxy = yzxyx, zxxyy = zyyxx, zxyxy = zyxyx, zxyyx = zyxxy, $L_1 \equiv$ yyyzz =-yxxzz =-xyxzz =-xxyzz, yzzyy =-yzzxx =-xzzyx =-xzzxy, yyzyz =-yxzxz =-xyzxz =-xxzyz, zyyzy =-zyxzx =-zxyzx =-zxxzy, yyzzy =-yxzzx =-xyzzx =-xxzzy, zyyyz =-zyxxz =-zxyxz =-zxxyz, yzyyz =-yzxxz =-xzyxz =-xzxyz, zyzyy =-zyzxx =-zxzyx =-zxzxy, yzyzy =-yzxzx =-xzyzx =-xzxzy, zzyyy =-zzyxx =-zzxyx =-zzxxy, yyyyy =- $\frac{1}{3}$ /xxxxy + xxxyx + xxyxx + xyxxx + yxxxx/, . yyyxx = $\frac{1}{3}$ /2xxxxy + 2xxxyx - xxyxx - xyxxx - yxxxx/, yyxyx = $\frac{1}{3}$ /2xxxxy - xxxyx + 2xxyxx - xyxxx - yxxxx/, yyxxy = $\frac{1}{3}$ /-xxxxy + 2xxxyx + 2xxyxx - xyxxx - yxxxx/, yxyyx = $\frac{1}{3}$ /2xxxxy - xxxyx - xxyxx + 2xyxxx - yxxxx/, yxxyy = $\frac{1}{3}$ /-xxxxy - xxxyx + 2xxyxx + 2xyxxx - yxxxx/, yxyxy = $\frac{1}{3}$ /-xxxxy + 2xxxyx - xxyxx + 2xyxxx - yxxxx/, xyyyx = $\frac{1}{3}$ /2xxxxy - xxxyx - xxyxx - xyxxx + 2yxxxx /, xyyxy = $\frac{1}{3}$ /-xxxxy + 2xxxyx - xxyxx - xyxxx + 2yxxxx/, xyxyy = $\frac{1}{3}$ /-xxxxy - xxxyx + 2xxyxx - xyxxx + 2yxxxx/, xxyyy = $\frac{1}{3}$ /-xxxxy - xxxyx - xxyxx + 2xyxxx + 2yxxxx/, xxxxy, xxxyx, xxyxx, xyxxx, yxxxx ;
32 (D_3) 3m (C_{3v}) $\bar{3}$m (D_{3d})	116	40	G_1 and H_1
6 (C_6) $\bar{6}$ (C_{3h}) 6/m (C_{6h})	121	51	G_1 and J_1
622 (D_6) 6mm (C_{6v}) $\bar{6}$m2 (D_{3h}) 6/mmm (D_{6h})	60	25	G_1

TABLE IV (continued)

Class	Number of non-zero elements	Number of in-dependent elements	Elements
23 (T) m3 (T_h)	60	20	xxxyz = yyyzx = zzzxy, xxxzy = yyyxz = zzzyx, xxzxy =yyxyz=zzyzx, xxyxz = yyzyx = zzxzy, xxyzx = yyzxy = zzxyz, xzxyx =yxyzy=zyzxz, xzxxy = yxyyz = zyzzx, xzyxx = yxzyy = zyxzz, xyzxx =yzxyy=zxyzz, xxzyx = yyxzy = zzyxz, xyxzx = yzyxy = zxzyz, xyxxz = yzyyx=zxzzy, yxxxz = zyyyx = xzzzy, yxxzx = zyyxy = xzzyz, yxzxx =zyxyy=xzyzz, yzxxx = zxyyy = xyzzz, zyxxx = xzyyy = yxzzz, zxyxx =xyzyy=yzxzz, zxxyx = xyyzy = yzzxz, zxxxy = xyyyz = yzzzx.
432 (O) $\bar{4}3m$ (T_d) m3m (O_h)	60	10	xxxyz = yyyzx = zzzxy =-xxxzy =-yyyxz =-zzzyx, xxyxz = yyzyx = zzxzy =-xxzxy =-yyxyz =-zzyzx, xyxxz = yzyyx = zxzzy =-xzxxy =-yxyyz =-zyzzx, yxxxz = zyyyx = xzzzy =-zxxxy =-xyyyz =-yzzzx, xxyzx = yyzxy = zzxyz =-xxzyx =-yyxzy =-zzyxz, xyxzx = yzyxy =zxzyz =-xzxyx =-yxyzy =-zyzxz, yxxzx = zyyxy = xzzyz =-zxxyx =-xyyzy =-yzzxz, xyzxx = yzxyy = zxyzz =-xzyxx =-yxzyy =-zyxzz, yxzxx = zyxyy = xzyzz =-zxyxx =-xyzyy =-yzxzz, yzxxx = zxyyy = xyzzz =-zyxxx =-xzyyy =-yxzzz,
Y Y_h K K_h	60	6	xxxyz = yyyzx = zzzxy =-xxxzy =-yyyxz =-zzzyx = =- xxyzx + xyxzx + yxxzx , xxyxz = yyzyx = zzxzy =-xxzxy =-yyxyz =-zzyzx = =- xxyzx + xyzxx + yxzxx , xyxxz = yzyyx = zxzzy =-xzxxy =-yxyyz =-zyzzx = =- xyxzx - xyzxx + yzxxx , yxxxz = zyyyx = xzzzy =-zxxxy =-xyyyz =-yzzzx = =-/yxxzx + yxzxx + yzxxx/, xxyzx = yyzxy = zzxyz =-yyxzy =-zzyxz =-xxzyx , xyxzx = yzyxy = zxzyz =-xzxyx =-yxyzy =-zyzxz, yxxzx = zyyxy = xzzyz =-zxxyx =-xyyzy =-yzzxz, xyzxx = yzxyy = zxyzz =-xzyxx =-yxzyy =-zyxzz, yxzxx = zyxyy = xzyzz =-zxyxx =-xyzyy =-yzxzz, yzxxx = zxyyy = xyzzz =-zyxxx =-xzyyy =-yxzzz.

which, together with the relations (3) and (4), permit the determination of the light refractive indices $n \approx (\varepsilon\mu)^{1/2}$.

For the sake of simplicity, we shall restrict our considerations to changes in the electric permittivity tensor due to a strong electric, optical or magnetic field in the presence of a DC magnetic field.

8.1. Light intensity-dependent magneto-optical phenomena

From Eqs (15), (20), (29) and (3), (36), we obtain the change in electric permittivity tensor analyzed by means of light of the frequency ω_A, but induced by intense laser light of the frequency ω_I (for simplicity, we omit the dependences on the wave vectors), in the form:

$$\Delta\varepsilon_{ij}(\omega_A) = 4\pi\{\chi_{ijn}^{eem}(\omega_A, 0)+ \\ +\chi_{ijkln}^{eeeem}(\omega_A, \omega_I, -\omega_I, 0)E_k(\omega_I)E_l(-\omega_I)+\ldots\}H_n(0). \tag{37}$$

If the analyzing beam (frequency ω_A) is assumed to propagate along the z-axis, Eq. (37) yields for the difference between diagonal components:

$$\Delta\varepsilon_{xx}(\omega_A)-\Delta\varepsilon_{yy}(\omega_A) = 4\pi\{\chi_{xxn}^{eem}(\omega_A, 0)-\chi_{yyn}^{eem}(\omega_A, 0)+ \\ +2[\chi_{xxkln}^{eeeem}(\omega_A, \omega_I, -\omega_I, 0)-\chi_{yykln}^{eeeem}(\omega_A, \omega_I, -\omega_I, 0)]I_{kl}+\ldots\}H_n(0) \tag{38}$$

and for the difference between nondiagonal components (assuming in (37) antisymmetricity in the indices i, j):

$$\Delta\varepsilon_{xy}(\omega_A)-\Delta\varepsilon_{yx}(\omega_A) = 8\pi\{\chi_{xyn}^{eem}(\omega_A, 0)+ \\ +2\chi_{xykln}^{eeeem}(\omega_A, \omega_I, -\omega_I, 0)I_{kl}+\ldots\}H_n(0), \tag{39}$$

where we have introduced the intensity tensor of light inducing the optical nonlinearity as:

$$I_{kl} = E_k(\omega_I)E_l(-\omega_I)/2. \tag{40}$$

Hence, observations of the changes (38) and (39) are found to depend, on the one hand, on the crystallographical symmetry of the body and, on the other, on the mutual configuration at which we experimentally dispose the vector $\boldsymbol{H}(0)$ of the DC magnetic field, those of the analyzing wave (frequency ω_A), and those of the nonlinearity-inducing wave (frequency ω_I), as well as on the states of polarization of the two waves. The variety of these configurations admits of a classification of all nonlinear magneto-optical phenomena. Two fundamental, limiting configurations can be distinguished:

(*i*) Faraday's configuration, where the analyzing wave propagates parallel to the magnetic field $\boldsymbol{H}(0)$ (z-axis), and

(*ii*) Voigt's configuration, where it propagates perpendicularly to $\boldsymbol{H}(0)$ (along the x or y-axis).

Tables I, II and IV immediately enable us to predict what kind of magneto-optical phenomenon can or cannot occur in a given crystallographical class (see, Tables V and VI).

Obviously, the simplest result is obtained from Eq. (39) for the isotropic body (classes K and Y) at Faraday configuration, namely:

$$\Delta\varepsilon_{xy}(\omega_A)-\Delta\varepsilon_{yx}(\omega_A) = 8\pi\{\chi_{xyz}^{eem}(\omega_A, 0)+ \\ +2\chi_{xyxxz}^{eeeem}(\omega_A, \omega_I, -\omega_I, 0)\,(I_{xx}+I_{yy})+\ldots\}H_z(0). \tag{39a}$$

TABLE V

Situations admitting of the observation of third-order nonlinear magneto-optical birefringence, induced by intense laser light

Magnetic field at:	Propagation direction of inducing light	Tensor elements $\chi^{eeem}_{(ij)kln}$ relevant to the birefringence effect	Crystallographic classes admitting of the effect
Voigt configuration H_y	x	$xxyyy - yyyyy$, $xxyzy - yyyzy$ $xxzyy - yyzyy$, $xxzzy - yyzyy$	1, $\bar{1}$, m, 2, $2/m$, 4, $\bar{4}$, $4/m$, 3, $\bar{3}$, 6, $\bar{6}$, $6/m$
	y	$xxzzy - yyzzy$, $xxzxy - yyzxy$ $xxxzy - yyxzy$, $xxxxy - yyxxy$,	all crystallographical classes
	z	$xxxxy - yyxxy$, $xxxyy - yyxyy$, $xxyxy - yyyxy$, $xxyyy - yyyyy$,	1, $\bar{1}$, 3, $\bar{3}$, 32, $3m$, $\bar{3}m$
Faraday configuration H_z	x	$xxyyz - yyyyz$, $xxyzz - yyyzz$, $xxzyz - yyzyz$, $xxzzz - yyzzz$,	1, $\bar{1}$, m, 2, $2/m$, 4, $\bar{4}$, $4/m$, 3, $\bar{3}$, 6, $\bar{6}$, $6/m$
	y	$xxzzz - yyzzz$, $xxzxz - yyzxz$ $xxxzz - yyxzz$, $xxxxz - yyxxz$	1, $\bar{1}$, m, 2, $2/m$, 4, $\bar{4}$, $4/m$, 3, $\bar{3}$, 32, $3m$, $\bar{3}m$, 6, $\bar{6}$, $6/m$
	z	$xxxxz - yyxxz$, $xxxyz - yyxyz$, $xxyxz - yyyxz$, $xxyyz - yyyyz$	all crystallographical classes

TABLE VI

Situations admitting of the observation of thitd-order nonlinear magneto-optical rotation, induced by intense laser light

Magnetic field at:	Propagation direction of inducing light	Tensor elements $\chi^{eeem}_{[ij]kln}$ relevant to the rotation effect	Crystallographic classes admitting of the effect
Voigt configuration H_y	x	$xyyyy$, $xyyzy$, $xyzyy$, $xyzzy$	all crystallographical classes
	y	$xyzzy$, $xyzxy$ $xyxzy$, $xyxxy$	1, $\bar{1}$, 2, m, $2/m$, 4, $\bar{4}$, $4/m$, 3, $\bar{3}$, 6, $\bar{6}$, $6/m$, 32, $3m$, $\bar{3}m$
	z	$xyxxy$, $xyxyy$ $xyyxy$, $xyyyy$	1, $\bar{1}$, 3, $\bar{3}$, 32, $3m$, $\bar{3}m$
Faraday configuration H_z	x	$xyyyz$, $xyyzz$ $xyzyz$, $xyzzz$	all crystallographical classes
	y	$xyzzz$, $xyzxz$ $xyxzz$, $xyxxz$	all crystallographical classes
	z	$xyxxz$, $xyxyz$ $xyyxz$, $xyyyz$	all crystallographical classes

Here, the first term describes the usual linear Faraday effect whereas the next term describes its nonlinear variation due to intense light. A similar result was obtained previously by the semi-macroscopic method [21, 40] for liquids where, beside the effect of nonlinear distortion of the molecules, there occurs an effect of reorientation of the molecules by the electric field of the intense light beam and the DC magnetic field concomitantly.

8.2. DC electric field-induced magneto-optical phenomena

When studying magneto-optical phenomena in the presence of a DC electric field, the change in electric permittivity tensor with regard to Eqs (15), (20), (25) and (29) is:

$$\Delta\varepsilon_{ij}(\omega_A) = 4\pi\{\chi^{eem}_{ijn}(\omega_A, 0)+ + \chi^{eeem}_{ijkl}(\omega_A, 0, 0)E_k(0)+ + \chi^{eeeem}_{ijkln}(\omega_A, 0, 0, 0)E_k(0)E_l(0)+\dots\}H_n(0). \tag{41}$$

On comparison with (37), the variation (41) is found to contain a new mixed term, linear in the DC electric field, expressing a new phenomenon of optical rotation in crossed DC electric nad DC magnetic fields [32]. From Table III, this effect can occur only in classes without a centre of symmetry; consequently, in studying centro-symmetric classes, the next term of the expansion (41) quadratic in $E(0)$ has to be taken into account. Tables VII and VIII give the feasible variants for the observation of magneto-optical effects of the second order.

A detailed discussion of the experimental conditions for the observation of these new nonlinear magneto-optical phenomena indicating the materials in which they can

TABLE VII

Situations admitting of the observation of second-order magneto-optical birefringence, induced by a DC electric field

Magnetic field at:	Direction of electric field	Tensor elements $\chi^{eeem}_{(ij)kl}$ relevant to the birefringence effect	Crystallographic classes admitting of the effect
Voigt configuration H_y	x	$xxxy - yyxy$	1, 2, $mm2$, 4, $\bar{4}$, $4mm$, 3, $3m$, 6, $6mm$
	y	$xxyy - yyyy$	1, 2, 222, 4, $\bar{4}$, 422, $\bar{4}2m$, 3, 32 6, 622, 23, 432, $\bar{4}3m$, Y, K
	z	$xxzy - yyzy$	1, 3, 32, $\bar{6}$, $\bar{6}m2$
Faraday configuration H_z	x	$xxxz - yyxz$	1, m, 3, $3m$, $\bar{6}$
	y	$xxyz - yyyz$	1, m, 3, 32, $\bar{6}$, $\bar{6}m2$
	z	$xxzz - yyzz$	1, 2, 222, $\bar{4}$, $\bar{4}2m$, $\bar{4}3m$, 23

TABLE VIII

Situations admitting of the observation of second-order magneto-optical rotation, induced by a DC electric field

Magnetic field at:	Direction of electric field	Tensor elements $\chi^{eeem}_{[ij]kl}$ relevant to the rotation effect	Crystallographic classes admitting of the effect
Voigt configuration H_y	x	$xyxy = -yxxy$	1, 2, 222, 4, $\bar{4}$, 422, $\bar{4}2m$, 3, 32, 6, 622, 23, 432, $\bar{4}3m$, Y, K
	y	$xyyy = -yxyy$	1, 2, $mm2$, 4, $\bar{4}$, $4mm$, 3, $3m$, 6, $6m$
	z	$xyzy = -yxzy$	1, m
Faraday configuration H_z	x	$xyxz = -yxxz$	1, m
	y	$xyyz = -yxyz$	1, m
	z	$xyzz = -yxzz$	1, 2, $mm2$, 4, $4mm$, 3, $3m$, 6, $6mm$

take place will be given in a separate paper. The question is answered by calculating from Fresnel's equation the respective light refraction indices, as expressed by the changes in electric permittivity tensor (37) and (41) and analogous changes in magnetic permittivity tensor. This represents a quite highly involved problem of Nonlinear Crystallo-Magneto--Optics.

In concluding, it may well be worth stressing that the magneto-optical birefringence phenomena discussed in Tables V and VII primarily hinge on the presence of spatial dispersion. On the other hand, magneto-optical rotation of the second order (Table VIII) can be induced in crystals without natural optical activity belonging to the classes $4\,mm$, $3\,m$, $6\,mm$, $\bar{4}3\,m$, Y and K. Light intensity-dependent magneto-optical rotation can occur in all classes, as well as in isotropic bodies.

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