LINEAR AND NONLINEAR MAGNETO-OPTICAL EFFECTS IN MATERIALS WITH SPATIAL DISPERSION AND MAGNETIC ORDERING*

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A theory of magneto-optical processes in aribtrary systems is proposed, comprising electric and magnetic multipole transitions. General formulae are derived for the light refractive index at Faraday's and Voigt's configurations. Gyrotropic rotation is discussed in detail for non-magnetic crystals, acted on by a dc magnetic field, as well as for magnetically ordered ones belonging to classes of the tetragonal, hexagonal, trigonal and cubic systems.

1. Introduction

It has been shown repeatedly that, in certain systems, spatial dispersion leads to new optical effects [1], and is particularly important in nonlinear phenomena induced by intense laser light [2–4].

Portigal and Burnstein [5], by a simplified approach, have considered the influence of spatial dispersion on magneto-optical effects. In this paper, we give a general theory of magneto-optical processes in arbitrary systems taking spatial dispersion and frequency dispersion into account. We proceed by multipolar theory of electromagnetic fields and quantum-mechanical perturbation calculus.

2. Fundamentals of the theory

The electric and magnetic permittivity tensors of an anisotropic system are:

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

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

δ_{ij} being elements of the symmetric unit Kronecker tensor.

The vector of total electric polarisation $\boldsymbol{P}_e(\boldsymbol{r}, t)$, induced in the point of space $\boldsymbol{r}$ and moment of time t by the electromagnetic field vectors $\boldsymbol{E}(\boldsymbol{r}, t)$ and $\boldsymbol{H}(\boldsymbol{r}, t)$ is given generally by the multipole expansion [2]:

$$\boldsymbol{P}_e(\boldsymbol{r}, t) = \sum_{n=1}^{\infty} \frac{(-1)^{n+1}}{(2n-1)!!} \nabla^{n-1}[n-1]\boldsymbol{P}_e^{(n)}(\boldsymbol{r}, t), \tag{3}$$

with $\boldsymbol{P}_e^{(n)}(\boldsymbol{r}, t)$ denoting 2^n-pole electric polarisation, and ∇ the spatial derivation operator. A similar equation holds for $\boldsymbol{P}_m(\boldsymbol{r}, t)$, the total magnetisation polarisation vector.

We assume the field vectors as;

$$\boldsymbol{E}(\boldsymbol{r}, t) = \boldsymbol{E}(\omega, \boldsymbol{k}) \exp\{i(\boldsymbol{k} \cdot \boldsymbol{r} - \omega t)\}, \tag{4}$$

with $\boldsymbol{k}$ the propagation vector of the electromagnetic wave, vibrating with the circular frequency ω. In a medium with the index n we have:

$$\boldsymbol{k} = n(\omega/c)\boldsymbol{s}, \tag{4a}$$

$\boldsymbol{s}$ being the unit vector in the direction of $\boldsymbol{k}$.

For moderate intensities of the wave, the multipolar polarisations depend linearly on $\boldsymbol{E}$ and $\boldsymbol{H}$, in a first approximation [2]:

$$\boldsymbol{P}_e^{(n)}(\boldsymbol{r}, t) = \sum_{s=1}^{\infty} \frac{i^{n-1}}{(2s-1)!!} \{{}_e^{(n)}\chi_e^{(s)}(\omega)[s]\boldsymbol{k}^{s-1}\boldsymbol{E}(\boldsymbol{r}, t) + {}_e^{(n)}\chi_m^{(s)}(\omega)[s]\boldsymbol{k}^{s-1}\boldsymbol{H}(\boldsymbol{r}, t)\}, \tag{5}$$

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where the $n+s$-rank tensor, defining the 2^n-pole electric susceptibility due to 2^s-pole magnetic transitions is of the quantum-mechanical form:

$$ {}^{(n)}_{\mathrm{e}}\chi^{(s)}_{\mathrm{m}}(\omega) = \frac{\rho}{\hbar}\sum_{abc}\rho_{ab}\left\{\frac{\langle a|M^{(n)}_{\mathrm{e}}|c\rangle\langle c|M^{(s)}_{\mathrm{m}}|b\rangle}{\omega_{cb}-\omega-\mathrm{i}\Gamma_{cb}}+\frac{\langle a|M^{(s)}_{\mathrm{m}}|c\rangle\langle c|M^{(n)}_{\mathrm{e}}|b\rangle}{\omega_{ca}+\omega+\mathrm{i}\Gamma_{ca}}\right\}. \tag{6}$$

Above, ρ_{ab} is the density matrix for a quantum transition with the frequency ω_{ba} and relaxation time Γ^{-1}_{ba}, whereas M^n_{e}, M^n_{m} are 2^n-pole electric (magnetic) moment operators of the microsystem (atom, molecule . . . .).

When determining the frequency dispersion and absorption of the multipole susceptibility tensor, it is convenient to resolve (6) into symmetric and antisymmetric parts:

$$ {}^{(n)}_{\mathrm{e}}S^{(s)}_{\mathrm{m}}(\omega) = (\rho/\hbar)\sum_{ab}\rho_{aa}\omega_{ba}\{\langle a|M^{(n)}_{\mathrm{e}}|b\rangle\langle b|M^{(s)}_{\mathrm{m}}|a\rangle + \langle a|M^{(s)}_{\mathrm{m}}|b\rangle\langle b|M^{(n)}_{\mathrm{e}}|a\rangle\}F_{ba}(\omega), \tag{7}$$

$$ {}^{(n)}_{\mathrm{e}}A^{(s)}_{\mathrm{m}}(\omega) = (\rho/\hbar)\sum_{ab}\rho_{aa}(\omega+\mathrm{i}\Gamma_{ba})\{\langle a|M^{(n)}_{\mathrm{e}}|b\rangle\langle b|M^{(s)}_{\mathrm{m}}|a\rangle - \langle a|M^{(s)}_{\mathrm{m}}|b\rangle\langle b|M^{(n)}_{\mathrm{e}}|a\rangle\}F_{ba}(\omega). \tag{8}$$

where we have introduced the complex function of frequency $F_{ba}(\omega) = F'_{ba}(\omega) + \mathrm{i}F''_{ba}(\omega)$, with:

$$ F'_{ba}(\omega) = \frac{\omega^2_{ba}-\omega^2+\Gamma^2_{ba}}{(\omega^2_{ba}-\omega^2)^2+2(\omega^2_{ba}+\omega^2)\Gamma^2_{ba}+\Gamma^4_{ba}}, \tag{9}$$

$$ F''_{ba}(\omega) = \frac{2\omega\Gamma_{ba}}{(\omega^2_{ba}-\omega^2)^2+2(\omega^2_{ba}+\omega^2)\Gamma^2_{ba}+\Gamma^4_{ba}}. \tag{10}$$

Since in tensorial notation the Maxwell equations of the field vectors (4) are:

$$ n\delta_{ijk}s_jE_k(\boldsymbol{r},t) = \mu_{ij}H_j(\boldsymbol{r},t), \tag{11}$$

$$ -n\delta_{ijk}s_jH_k(\boldsymbol{r},t) = \epsilon_{ij}E_j(\boldsymbol{r},t), \tag{12}$$

we obtain by (1), (3) and (5) for the electric permittivity tensor:

$$ \epsilon_{ij}(\omega,\boldsymbol{k}) = \epsilon^{(0)}_{ij}(\omega,\boldsymbol{k}) + 4\pi\,\mathrm{i}\delta_{ijk}\{G^{\mathrm{ee}(0)}_{kl}(\omega,\boldsymbol{k}) + G^{\mathrm{em}(0)}_{kl}(\omega,\boldsymbol{k})\}s_l, \tag{13}$$

δ_{ijk} being the antisymmetric Levi–Cività tensor.

In (13), $\epsilon^{(0)}_{ij}(\omega,\boldsymbol{k})$ describes the symmetric part of the electric permittivity in the absence of natural or magnetic gyration. The remaining part of (13) describes natural optical activity, defined by the tensors of electric gyration due to electric multipole transitions, $G^{\mathrm{ee}(0)}_{kl}(\omega,\boldsymbol{k})$, and to magnetic multipole transitions, $G^{\mathrm{em}(0)}_{kl}(\omega,\boldsymbol{k})$. The gyration tensors are expressed in terms of the appropriate tensors of multipole susceptibilities of eqs. (5) and (6).

Similarly to (13), we write the magnetic permittivity tensor:

$$ \mu_{ij}(\omega,\boldsymbol{k}) = \mu^{(0)}_{ij}(\omega,\boldsymbol{k}) + 4\pi\,\mathrm{i}\delta_{ijk}\{G^{\mathrm{mm}(0)}_{kl}(\omega,\boldsymbol{k}) + G^{\mathrm{me}(0)}_{kl}(\omega,\boldsymbol{k})\}s_l, \tag{14}$$

where $G^{\mathrm{mm}(0)}_{kl}(\omega,\boldsymbol{k})$ and $G^{\mathrm{me}(0)}_{kl}(\omega,\boldsymbol{k})$ are tensors of magnetic gyration, induced by magnetic and electric multipole transitions.

The electric and magnetic permittivity tensors (13) and (14) provide a complete description of frequency dispersion for arbitrary anisotropic bodies.

3. Magneto-optical processes

Consider a crystal with electric permittivity tensor of the form:

$$ (\epsilon_{ij}) = \begin{pmatrix} \epsilon_{xx} & \epsilon_{xy} & 0 \\ \epsilon_{yx} & \epsilon_{yy} & 0 \\ 0 & 0 & \epsilon_{zz} \end{pmatrix} \tag{15}$$

and similar magnetic permittivity tensor.

Assuming the light wave to propagate along the z-axis, the Maxwell equations (11) and (12) yield for the refractive index:

$$n_\pm^2 = \tfrac{1}{2}\{\epsilon_{xx}\mu_{yy} + \epsilon_{yy}\mu_{xx} - \epsilon_{xy}\mu_{xy} - \epsilon_{yx}\mu_{yx} \pm [(\epsilon_{xx}\mu_{yy} - \epsilon_{yy}\mu_{xx} - \epsilon_{yx}\mu_{yx} + \epsilon_{xy}\mu_{xy})^2 + 4(\epsilon_{xx}\mu_{xy} - \mu_{xx}\epsilon_{yx})(\epsilon_{yy}\mu_{yx} - \mu_{yy}\epsilon_{xy})]^{1/2}\}. \tag{16}$$

For particular cases, (16) reduces to well known results [6].

If the wave propagates along the x-axis, we obtain for the indices in the two directions perpendicular to x:

$$n_y^2 = \epsilon_{yy}\mu_{zz} - (\mu_{zz}/\epsilon_{xx})\epsilon_{xy}\epsilon_{yx}, \quad n_z^2 = \epsilon_{zz}\mu_{yy} - (\epsilon_{zz}/\mu_{xx})\mu_{yx}\mu_{xy}. \tag{17}$$

If the system is acted on by a dc magnetic field $\boldsymbol{H}^0$, the electric (13) and magnetic (14) permittivity tensors are functions of $\boldsymbol{H}^0$:

$$\epsilon_{ij}(\omega, \boldsymbol{k}, \boldsymbol{H}^0) = \epsilon_{ij}^s(\omega, \boldsymbol{k}, \boldsymbol{H}^0) + 4\pi\, \mathrm{i}\delta_{ijk}\{F_k^{ee}(\omega, \boldsymbol{k}, \boldsymbol{H}^0) + [G_{kl}^{ee}(\omega, \boldsymbol{k}, \boldsymbol{H}^0) + G_{kl}^{em}(\omega, \boldsymbol{k}, \boldsymbol{H}^0)]s_l\}, \tag{18}$$

$F_k^{ee}(\omega, \boldsymbol{k}, \boldsymbol{H}^0)$ being the magneto-gyration vector. The magnetic permittivity can be written similarly.

Assuming $\boldsymbol{H}^0$ as not excessively strong though sufficient for modifying the electric and magnetic properties of the system appreciably we have, in a quadratic approximation:

$$\epsilon_{ij}^s(\omega, \boldsymbol{k}, \boldsymbol{H}^0) = \epsilon_{ij}^{(0)}(\omega, \boldsymbol{k}) + \epsilon_{ijm}^{(1)}(\omega, \boldsymbol{k})H_m + \epsilon_{ijmn}^{(2)}(\omega, \boldsymbol{k})H_m^0H_n^0 + \cdots, \tag{19}$$

$$F_k^{ee}(\omega, \boldsymbol{k}, \boldsymbol{H}^0) = F_{km}^{ee}(\omega, \boldsymbol{k})H_m^0 + \cdots, \tag{20}$$

$$G_{kl}^{ee}(\omega, \boldsymbol{k}, \boldsymbol{H}^0) = G_{kl}^{ee(0)}(\omega, \boldsymbol{k}) + G_{klm}^{ee(1)}(\omega, \boldsymbol{k})H_m^0 + G_{klmn}^{ee(2)}(\omega, \boldsymbol{k})H_m^0H_n^0 + \cdots, \tag{21}$$

and similar expressions for the other gyration tensors.

Above, $\epsilon_{ij}^{(0)}(\omega, \boldsymbol{k})$ is a second-rank polar tensor, symmetric with respect to the operation of time-inversion [7] (polar i-tensor). The third-rank axial tensor $\epsilon_{ijm}^{(1)}(\omega, \boldsymbol{k})$, antisymmetric with respect to time inversion (axial c-tensor), is a function of odd powers of the wave vector [1] and defines magneto-spatial dispersion [5]. The polar i-tensor $\epsilon_{ijkl}^{(2)}(\omega, \boldsymbol{k})$ describes the electric contribution to the quadratic Voigt effect, Cotton–Mouton effect, and magnetostriction; like $\epsilon_{ij}^{(0)}(\omega, \boldsymbol{k})$, it can but be an even function $\boldsymbol{k}$. The polar second-rank i-tensor $F_{km}^{ee}(\omega, \boldsymbol{k})$ describes the electric contribution to the linear Faraday effect, and is an even function of $\boldsymbol{k}$. The polar i-tensors of the type of $G_{ijm}^{(1)}(\omega, \boldsymbol{k})$ are odd functions of $\boldsymbol{k}$, and describe magneto-spatial rotation. The gyration axial c-tensors of the type $G_{ij}^{(0)}(\omega, \boldsymbol{k})$ and $G_{ijmn}^{(2)}(\omega, \boldsymbol{k})$ are even functions of $\boldsymbol{k}$ and describe, respectively, natural optical activity and its variation proportional to $(H^0)^2$; the latter can occur as well in isotropic, naturally optically active media.

4. Results

By having recourse to tabulated [3, 7] nonzero independent elements of the tensors of (19), (20) and (21), we find for all crystals having symmetry of the classes of the tetragonal, hexagonal, trigonal and cubic systems except $\underline{4}$, $\underline{\bar{4}}$, $\underline{4}/\mathrm{m}$, $\underline{4}2\underline{2}$, $\underline{4}\mathrm{m}\underline{\mathrm{m}}$, $\underline{4}2\underline{\mathrm{m}}$, $\underline{\bar{4}}\mathrm{m}\underline{2}$, $\underline{4}/\mathrm{mm}\underline{\mathrm{m}}$, 23, m3, $\underline{\mathrm{m}}3$, $\underline{4}3\underline{2}$ and $\underline{\bar{4}}3\underline{\mathrm{m}}$:

$$n_+ - n_- = (4\pi/n)\{\epsilon_{xx}^s(\omega, k_z, H_z^0)[F_{zz}^{mm}(\omega, k_z)H_z^0 + (G_{zz}^{mm}(\omega, k_z, H_z^0) + G_{zz}^{me}(\omega, k_z, H_z^0))s_z] + \mu_{xx}^s(\omega, k_z, H_z^0)[F_{zz}^{ee}(\omega, k_z)H_z^0 + (G_{zz}^{ee}(\omega, k_z, H_z^0) + G_{zz}^{em}(\omega, k_z, H_z^0))s_z]\}, \tag{22}$$

with $n = \tfrac{1}{2}(n_+ + n_-)$, and:

$$\epsilon_{xx}^s(\omega, k_z, H_z^0) = \epsilon_{xx}^{(0)}(\omega, k_z) + \epsilon_{xxz}^{(1)}(\omega, k_z)H_z^0 + \epsilon_{xxzz}^{(2)}(\omega, k_z)H_z^{0^2} + \cdots, \tag{23}$$

in the classes 4, $\bar{4}$, 4/m, $4\underline{22}$, $4\underline{\mathrm{mm}}$, $\bar{4}\underline{2\mathrm{m}}$, $4/\mathrm{m}\underline{\mathrm{mm}}$, 3, $\bar{3}$, $3\underline{2}$, $3\underline{\mathrm{m}}$, $\bar{3}\underline{\mathrm{m}}$, 6, $\bar{6}$, 6/m, $6\underline{22}$, $6\underline{\mathrm{mm}}$, $\bar{6}\underline{\mathrm{m}2}$, $6/\mathrm{m}\underline{\mathrm{mm}}$, but $\epsilon_{xxz}^{(1)}(\omega, k_z) = 0$ in the others: $4/\underline{\mathrm{m}}$, $\underline{4}/\underline{\mathrm{m}}$, 422, 4mm, $\bar{4}$2m, 4/mmm, $4/\underline{\mathrm{mmm}}$, $4/\underline{\mathrm{m}}\mathrm{mm}$, $\underline{4}/\underline{\mathrm{mm}}\mathrm{m}$, $\underline{3}$, 32, 3m, $\bar{3}$m, $\underline{\bar{3}}$m, $\underline{\bar{3}\mathrm{m}}$, $\underline{6}$, $\underline{\bar{6}}$, $\underline{6}/\underline{\mathrm{m}}$, $\underline{6}$/m, $6/\underline{\mathrm{m}}$, $\underline{6}2\underline{2}$, 622, 6mm, $\underline{6}\mathrm{m}\underline{\mathrm{m}}$, $\bar{6}$m2, $\underline{\bar{6}}2\underline{\mathrm{m}}$, $\underline{\bar{6}}\mathrm{m}\underline{2}$, 6/mmm, $\underline{6}/\mathrm{m}\underline{\mathrm{m}}\mathrm{m}$, $\underline{6}/\underline{\mathrm{mm}}\mathrm{m}$, $6/\underline{\mathrm{mmm}}$, $6/\underline{\mathrm{m}}\mathrm{mm}$, 432, $\bar{4}$3m, m3m, $\underline{\mathrm{m}}3\underline{\mathrm{m}}$, $\underline{\mathrm{m}}$3m, Y, $\mathrm{Y_h}$, K, $\mathrm{K_h}$. The component $\mu_{xx}^s(\omega, k_z, H_z^0)$ is similar for these classes. The polar i-tensor elements $F_{zz}^{ee}(\omega, k_z)$ and $F_{zz}^{mm}(\omega, k_z)$ occur in all classes. The remaining gyration tensor elements amount to:

$$G_{zz}^{ee}(\omega, k_z, H_z^0) = G_{zz}^{ee(0)}(\omega, k_z) + G_{zzz}^{ee(1)}(\omega, k_z)H_z^0 + G_{zzzz}^{ee(2)}(\omega, k_z)H_z^{0^2}, \tag{24}$$

$$G_{zz}^{em}(\omega, k_z, H_z^0) = G_{zz}^{em(0)}(\omega, k_z) + G_{zzz}^{em(1)}(\omega, k_z)H_z^0 + G_{zzzz}^{em(2)}(\omega, k_z)H_z^{0^2} \tag{25}$$

with similar expansions for $G_{zz}^{mm}(\omega, k_z, H_z^0)$ and $G_{zz}^{me}(\omega, k_z, H_z^0)$, but $G_{zz}^{ee(0)}(\omega, k_z)$, $G_{zzzz}^{ee(2)}(\omega, k_z)$, $G_{zz}^{em(0)}(\omega, k_z)$, $G_{zzzz}^{em(2)}(\omega, k_z)$, $G_{zz}^{mm(0)}(\omega, k_z)$, $G_{zzzz}^{mm(2)}(\omega, k_z)$, $G_{zz}^{me(0)}(\omega, k_z)$ and $G_{zzzz}^{me(2)}(\omega, k_z)$ will occur for the classes 4, $4/\underline{m}$, 422, $4\underline{mm}$, $4/\underline{mmm}$, 3, $\underline{3}$, 32, $3\underline{m}$, $\bar{3}\underline{m}$, 6, $\bar{6}$, $6/\underline{m}$, 622, $6\underline{mm}$, $\bar{6}2\underline{m}$, $6/\underline{mmm}$, 432, $\underline{m}3\underline{m}$, Y and K whereas $G_{zzz}^{ee(1)}(\omega, k_z)$, $G_{zzz}^{em(1)}(\omega, k_z)$, $G_{zzz}^{mm(1)}(\omega, k_z)$) and $G_{zzz}^{me(1)}(\omega, k_z)$ only for 4, 4mm, $4\underline{mm}$, 3, 3m, $3\underline{m}$, 6, $\underline{6}$, 6mm, $\underline{6}m\underline{m}$ and $6\underline{mm}$.

For isotropic optically inactive bodies of symmetry $\bar{4}3m$, m3m, Y_h and K_h we obtain the well known results for Faraday's and Voigt's configurations.

Magnetically ordered crystals with nonzero spontaneous magnetisation M_z in the z-axis direction (parallel to the symmetry axis of the highest multiplicity) exhibit, even without an external dc magnetic field, nonzero differences $n_+ - n_-$ and $n_y - n_z$ given by (16), (17) and (22) (with, now, M_z instead of H_z^0) [8, 9]. Whereas an external dc magnetic field will give rise therein to new non-additive terms, proportional to $H_z^0 M_z$.

The above results should prove very helpful when choosing the best crystal for experimental observations of birefringence and optical rotation, arising from the various terms of eqs. (19), (20) and (21).

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