ELECTRIC AND MAGNETIC ANISOTROPY INDUCED IN NONABSORBING ISOTROPIC MEDIA BY AN INTENSE LASER BEAM

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By a semi-macroscopic theory, intense light is shown to induce electric and magnetic anisotropy in isotropic media. In diamagnetics, the magnetic effect is small as compared to the optically-induced electric anisotropy, which *e.g.* in carbon disulphide or nitrobenzene should be accessible to observation by using a giant pulsed ruby laser. Investigation of the two effects in gases will yield new information on the electric and optical anisotropies, both linear and non-linear, of various molecules; in condensed media it can moreover provide data on the molecular correlations. The variations in electric and magnetic permeability due to optico-striction and the optico-caloric effect are calculated thermodynamically.

1. Introduction

Recently, some papers [1—2] appeared on the theory and measurement of the inversed Faraday effect. We wish to draw attention to a similar effect, consisting in the induction of magnetic anisotropy in isotropic media by an intense (*e. g.* laser) beam. This, indeed, is an inversed Cotton-Mouton effect, related with the previously considered non-linear variations of magnetic permeability which can be induced in gases or liquids by a strong optical field [3].

Similarly, an optical field of very high intensity can affect the electric properties of isotropic systems [3, 4]. We have in mind nonlinear variations of the electric permittivity and the optically induced electric anisotropy, which is indeed the inverse of the well-known effect of DC field-induced birefringence *i. e.* of Kerr's effect. At present the detection of the electric anisotropy induced in an isotropic medium by an intense light beam is only a matter of evolving appropriate ingenious measuring laser techniques.

From a phenomenological point of view, the above-mentioned effects are closely related with the circumstance that a medium will become nonlinear when acted on by light of high

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intensity. In this case the electric (ε) as well as the magnetic permittivity (μ) become non linear functions of the intensity I, their nonlinear variations being given as

$$\varepsilon-\varepsilon_0 = Q_1^{eo}I+Q_2^{eo}I^2+Q_3^{eo}I^3+\ldots, \tag{1}$$

$$\mu-\mu_0 = Q_1^{mo}I+Q_2^{mo}I^2+Q_3^{mo}I^3+\ldots. \tag{2}$$

Here, the expansion coefficients $Q_1^{eo}, \ldots Q_1^{mo}, \ldots$ describe the electro-optical and magneto--optical properties of the medium as well as its structure and thermodynamical state.

In the present paper, it is our aim to show that under the action of intense light the medium becomes not only nonlinear but moreover anisotropic in its electric and magnetic properties. In order to obtain results of sufficient generality, we proceed by the well-known semi-macroscopic approach to linear [5] and nonlinear [6—8] phenomena in isotropic dielectrics. The results thus derived are further subjected to a microscopic interpretation in a statistical-molecular approach. Here, however, our considerations will be restricted to variations in ε and μ linearly dependent on I, and thus to a discussion of the coefficients Q_1^{eo} and Q_1^{mo} in the expansions of Eqs (1) and (2).

2. *Optically induced magnetic anisotropy*

We consider a spherical, macroscopic sample of volume V within a dense isotropic medium in which the weak (measuring) magnetic field $\boldsymbol{H}$ induces magnetization given by the vector $\boldsymbol{P}_m$. Under the effect of the oscillating electric field $\boldsymbol{E}$ associated with an intense light beam, the medium becomes anisotropic, with magnetic properties given by the magnetic permeability tensor

$$\mu_{\sigma\tau} - \delta_{\sigma\tau} = 4\pi \frac{\partial P_{m\sigma}}{\partial H_\tau}. \tag{3}$$

where the indices σ and τ relate to the axes X, Y, Z of the laboratory reference frame.

Since the magnetization vector $\boldsymbol{P}_m$ is defined by the ratio of the mean magnetic dipole moment $\langle \boldsymbol{M}^m \rangle$ and the volume V of the sphere, we have by (3) for a diamagnetic medium

$$\mu_{\sigma\tau}-\delta_{\sigma\tau} = \frac{4\pi}{V_I}\left\langle \frac{\partial M_\sigma^m}{\partial H_\tau}\right\rangle_I, \tag{4}$$

where, in the classical case, the mean statistical value $\langle\ \rangle_I$ in the presence of light intensity I is defined as follows:

$$\left\langle \frac{\partial M_\sigma^m}{\partial H_\tau}\right\rangle_I = \frac{\int \frac{\partial M_\sigma^m}{\partial H_\tau} \exp\left\{-\frac{U(\tau, I)}{kT}\right\} d\tau}{\int \exp\left\{-\frac{U(\tau, I)}{kT}\right\} d\tau}. \tag{5}$$

Here, the total potential energy of the system at configuration τ may by expanded in a power series in I and we obtain in a linear approximation (see Appendix A)

$$U(\tau, I) = U(\tau, 0)-\frac{1}{2}A_{\sigma\tau}^o I_{\sigma\tau}-\ldots \tag{6}$$

with $U(\tau, 0)$ — the potential energy at zero light intensity I, which is defined as the trace of the incident light intensity tensor $I_{\sigma\tau} = \frac{1}{2} E_\sigma E_\tau^*$. In (6), $A^o_{\sigma\tau}$ denote the $\sigma\tau$ components of the optical polarizability tensor of the medium.

Similarly, the magnetic dipole moment in (4) is a function linear in $\boldsymbol{H}$ and nonlinear in $\boldsymbol{E}$, and we can write in the same approximation as (6)

$$M^m_\sigma = M^m_{0\sigma} + \frac{1}{2} B^{mo}_{\sigma\tau\nu} I_{\tau\nu} + \ldots + A^m_{\sigma\tau} H_\tau + \frac{1}{2} C^{mo}_{\sigma\tau\nu\varrho} H_\tau I_{\nu\varrho} + \ldots, \tag{7}$$

where $\boldsymbol{M}^m_0$ is the magnetic dipole moment of the sphere in the absence of external fields (which vanishes in the case of diamagnetic media), and the tensor $B^{mo}_{\sigma\tau\nu}$ describes the magnetization induced in the medium by light of intensity tensor $I_{\tau\nu}$. Thus, the second term in (7) corresponds to the inverse Faraday effect, as introduced and discussed by Pershan *et al.* [1, 2]. $A^m_{\sigma\tau}$ is the magnetic polarizability tensor of the medium whereas the tensor $C^{mo}_{\sigma\tau\nu\varrho}$ accounts for the change in magnetic polarizability due to the light intensity tensor $I_{\nu\varrho}$.

However, the medium is acted on by the intense field $\boldsymbol{E}$ too, and quite generally this can affect also the volume V. If, for simplicity, we assume that the sphere changes its volume isotropically (without undergoing a change in shape) under the effect of the field $\boldsymbol{E}$, and that this change is a quadratic function of E, we can write (see Appendix B)

$$V_I - V = -\frac{V}{8\pi}\left\{\left(\frac{\partial n^2}{\partial p}\right)_T - (n^2-1)\varkappa_T\right\} I \tag{8}$$

for the change in volume due to the light intensity I (opticostriction); p denotes the pressure, n — the refractivity and $\varkappa_T$ — the isothermal compressibility of the medium.

By Eqs (5)—(8), we obtain from Eq. (4) for the change in magnetic permeability tensor due to light of intensity I

$$\mu_{\sigma\tau} - \mu_0\,\delta_{\sigma\tau} = A_{mo} I \delta_{\sigma\tau} + B_{mo}(3\,I_{\sigma\tau} - I\delta_{\sigma\tau}), \tag{9}$$

where μ_0 is the scalar permeability in the absence of light ($I = 0$).

The constant B_{mo} in Eq. (9) defines the magnetic anisotropy induced optically in the medium, and is in general given as (see Eq. A12)

$$B_{mo} = \frac{\pi}{45V}\left\langle 3C^{mo}_{\alpha\beta\alpha\beta} + 3C^{mo}_{\alpha\beta\beta\alpha} - 2C^{mo}_{\alpha\alpha\beta\beta} + \right.$$

$$\left. + \frac{1}{kT}\left(3A^m_{\alpha\beta}A^o_{\alpha\beta} + 3A^m_{\alpha\beta}A^o_{\beta\alpha} - 2A^m_{\alpha\alpha}A^o_{\beta\beta}\right)\right\rangle, \tag{10}$$

where the brackets $\langle\ \rangle$ denote statistical averaging (5) at zero light intensity.

The constant A_{mo} describing the isotropic changes in $\mu_{\sigma\tau}$ consists of two parts: firstly

$$A^P_{mo} = \frac{2\pi}{9V}\left\{\langle C^{mo}_{\alpha\alpha\beta\beta}\rangle + \frac{1}{kT}\left(\langle A^m_{\alpha\alpha}A^o_{\beta\beta}\rangle - \langle A^m_{\alpha\alpha}\rangle\langle A^o_{\beta\beta}\rangle\right)\right\} \tag{11}$$

related with variations in magnetic polarizability and fluctuations in linear magnetic and optical polarizability, and secondly

$$A_{mo}^{V}=\frac{\mu_0-1}{8\pi}\left\{\left(\frac{\partial n^2}{\partial p}\right)_T-(n^2-1)\,\varkappa_T\right\} \tag{12}$$

resulting from opticostriction (electrostriction at optical frequencies given by Eq. (8)).

For the case of light incident in the direction of the Z-axis of the laboratory system, Eq. (9) yields the relative difference in diagonal components of the magnetic permeability tensor in the form

$$\mu_{xx}-\mu_{yy}=3B_{mo}(I_{xx}-I_{yy}), \tag{13}$$

which defines only the magnetic anisotropy induced in the isotropic medium by intense light. For natural light $\bar{I}_{xx}=\bar{I}_{yy}=\frac{1}{2}I$ and the magnetic anisotropy (13) vanishes.

In the case when the incident beam is elliptically polarized and propagates again along the Z-axis, equation (9) leads formally to the difference between the nondiagonal components of the magnetic permeability tensor

$$\mu_{xy}-\mu_{yx}=3iB_{mo}(I_+-I_-), \tag{14}$$

where I_+ and I_- are the intensities of right and left circularly polarized light with electric amplitudes $E_\pm=\frac{1}{\sqrt{2}}(E_x\pm iE_y)$.

The magnetic anisotropy constant in the general form (10) can be written in molecular form by recurring to classical statistics. Namely, for a medium of density $\varrho=N/V$ of magnetically and electrically anisotropic molecules, one obtains

$$B_{mo}=\varrho B_{mo}^{(1)}+\varrho^2 B_{mo}^{(2)}+\ldots, \tag{15}$$

where

$$B_{mo}^{(1)}=\frac{2\pi}{45}\left\{3c_{\alpha\beta\alpha\beta}^{mo}-c_{\alpha\alpha\beta\beta}^{mo}+\frac{1}{kT}\left(3a_{\alpha\beta}^{m}a_{\alpha\beta}^{o}-a_{\alpha\alpha}^{m}a_{\beta\beta}^{o}\right)\right\} \tag{16}$$

is a constant determining the magneto-optical properties of the medium in the absence of molecular interactions. The tensors $a_{\alpha\beta}^{o}$ and $a_{\alpha\beta}^{m}$ define the optical and magnetic linear polarizabilities of the isolated molecule, and $c_{\alpha\beta\gamma\delta}^{mo}$ the — change in its magnetic polarizability under the effect of the light intensity I.

The other constants in (15) account for the influence of molecular correlations on the magnetic anisotropy in a dense medium; in a first approximation [9, 10],

$$B_{mo}^{(2)}=\frac{2\pi}{45VkT}\int\int\left\{3a_{\alpha\beta}^{m(p)}a_{\alpha\beta}^{o(q)}-a_{\alpha\alpha}^{m(p)}a_{\beta\beta}^{o(q)}\right\}g^2(\tau_p,\tau_q)\,d\tau_p d\tau_q, \tag{17}$$

where $g^{(2)}(\tau_p,\tau_q)$ is the binary correlation function for molecules p and q at configurations τ_p and τ_q.

By (17), $B_{mo}^{(2)}$ is non-zero only if molecular correlations are present; if not, it vanishes, and the magnetic anisotropy is given by the constant (16), which consists of a temperature-independent part defining the effect of non-linear molecular deformation (a counterpart of Voigt's effect), and a temperature-dependent part related to the effect of molecular orientation in a strong optical field (a counterpart of the Langevin effect).

Since in the diamagnetic case the magnetic anisotropy constant B_{mo} can be replaced by the Cotton-Mouton constant, which for nitrobenzene is of the order of 10^{-16} esu, Eq. (13) yields $\mu_{xx}-\mu_{yy} = 3\times 10^{-16} I$. Hence, the optically induced magnetic anisotropy is seen to be very small in the case of diamagnetic substances and is accessible to measurement only by highly sensitive experimental methods and using a laser beam of intensity $I \simeq 10^8$ esu. Presumably, larger effects can be expected in paramagnetic substances *e. g.* in oxygen.

3. Optically induced electric anisotropy

In a similar way, the semi-macroscopic theory of optically induced electric anisotropy can be formulated. We shall restrict ourselves here to writing out the equation for the change in electric permittivity tensor due to light of intensity I:

$$\varepsilon_{\sigma\tau}-\varepsilon_0\delta_{\sigma\tau} = \left(\frac{\varepsilon_0+2}{3}\right)\{A_{eo}I\delta_{\sigma\tau}+B_{eo}(3I_{\sigma\tau}-I\delta_{\sigma\tau})\}, \tag{18}$$

where ε_0 is the scalar dielectric constant at $I=0$.

The quantity A_{eo} in Eq. (18) describes isotropic changes in ε related with the linear and nonlinear polarizabilities and opticostriction (8), whereas the constant B_{eo} defines the electric anisotropy induced in the medium by intense light and is given by

$$B_{eo} = \frac{\pi}{45V}(3\delta_{\alpha\gamma}\delta_{\beta\delta}+3\delta_{\alpha\delta}\delta_{\beta\gamma}-2\delta_{\alpha\beta}\delta_{\gamma\delta})\left\langle C_{\alpha\beta\gamma\delta}^{eo}+\right.$$
$$\left.+\frac{1}{kT}(2M_\alpha^e B_{\beta\gamma\delta}^{eo}+A_{\alpha\beta}^e A_{\gamma\delta}^o)+\frac{1}{k^2T^2}M_\alpha^e M_\beta^e A_{\gamma\delta}^o\right\rangle, \tag{19}$$

where M_α^e is the α-component of the electric dipole moment of the sphere and $A_{\alpha\beta}^e$ the components of its electric polarizability tensor. The tensors $B_{\alpha\gamma\delta}^{eo}$ and $C_{\alpha\beta\gamma\delta}^{eo}$ describe respectively the change in M_α^e and $A_{\alpha\beta}^e$ produced by the light intensity tensor $I_{\gamma\delta}$.

We now proceed from the foregoing semimacroscopic form of the electric anisotropy constant of (19) to a molecular form aimed at gaining insight into the microscopic mechanism of this phenomenon. Namely, by methods of statistical mechanics, the constant B_{eo} can be written formally as the following expansion in a power series in the number density ϱ:

$$B_{eo} = \varrho B_{eo}^{(1)}+\varrho^2 B_{eo}^{(2)}+\varrho^3 B_{eo}^{(3)}+\ldots, \tag{20}$$

where

$$B_{eo}^{(1)} = \frac{2\pi}{45}\left\{3c_{\alpha\beta\alpha\beta}^{eo}-c_{\alpha\alpha\beta\beta}^{eo}+\frac{2}{kT}(3\mu_\alpha^e b_{\beta\alpha\beta}^{eo}-\mu_\alpha^e b_{\alpha\beta\beta}^{eo})+\right.$$
$$\left.+\frac{1}{kT}(3a_{\alpha\beta}^e a_{\alpha\beta}^o-a_{\alpha\alpha}^e a_{\beta\beta}^o)+\frac{1}{k^2T^2}(3\mu_\alpha^e\mu_\beta^e a_{\alpha\beta}^o-\mu_\alpha^e\mu_\alpha^e a_{\beta\beta}^o)\right\} \tag{21}$$

is the electric anisotropy constant for an ideal system of noninteracting molecules having permanent electric dipole moments μ^e. On interchanging the indices e and o, Eq. (21) becomes analogous to the Kerr constant [3, 11—14].

The remaining constants in (20) are non-zero only for systems in which molecular interactions occur and are of the form (for comparison see Ref. [14])

$$B_{eo}^{(2)} = \frac{\pi}{45kT}(3\delta_{\alpha\gamma}\delta_{\beta\delta}+3\delta_{\alpha\delta}\delta_{\beta\gamma}-2\delta_{\alpha\beta}\delta_{\gamma\delta})\int\int\Big\{\mu_\alpha^{e(p)}b_{\beta\gamma\delta}^{eo(q)}+$$

$$+b_{\alpha\gamma\delta}^{eo}\mu_\beta^{e(q)}+a_{\alpha\beta}^{e(p)}a_{\gamma\delta}^{o(q)}+\frac{1}{kT}(\mu_\alpha^{e(p)}\mu_\beta^{e(p)}a_{\gamma\delta}^{o(q)}+\mu_\alpha^{e(p)}\mu_\beta^{e(q)}a_{\gamma\delta}^{o(p)}+$$

$$+\mu_\alpha^{e(p)}\mu_\beta^{e(q)}a_{\gamma\delta}^{o(q)})\Big\}g^{(2)}(\tau_p,\tau_q)\,d\tau_p d\tau_q, \tag{22}$$

$$B_{eo}^{(3)} = \frac{2\pi}{45k^2T^2}\int\int\int\{3\mu_\alpha^{e(p)}\mu_\beta^{e(q)}a_{\alpha\beta}^{o(r)}-\mu_\alpha^{e(p)}\mu_\alpha^{e(q)}a_{\beta\beta}^{o(r)}\}\,g^{(3)}(\tau_p,\tau_q,\tau_r)\,d\tau_p d\tau_q d\tau_r, \tag{23}$$

where $g^{(3)}(\tau_p, \tau_q, \tau_r)$ is the ternary correlation function for triples of molecules p, q and r,

The expressions (21)—(23) hold for systems of interacting molecules of arbitrary symmetry, and their further simplification can be achieved by assuming a particular type of molecular symmetry, *e. g.* spherical, tetrahedral, axial or other point group symmetries [8, 14].

The general Eq. (18) leads to the electric anisotropy

$$\varepsilon_{xx}-\varepsilon_{yy} = 3\left(\frac{\varepsilon_0+2}{3}\right)^2 B_{eo}(I_{xx}-I_{yy}), \tag{24}$$

and the difference between the nondiagonal components of the electric permittivity tensor

$$\varepsilon_{xy}-\varepsilon_{yx} = 3i\left(\frac{\varepsilon_0+2}{3}\right)B_{eo}(I_+-I_-). \tag{25}$$

Hence, the electric anisotropy induced optically in an isotropic medium is independent of the constant A_{eo} containing *i. a.* the opticostriction given by (8).

On certain assumptions, the electric anisotropy constant B_{eo} can be replaced by the Kerr constant. Since for nitrobenzene the latter is of the order of 10^{-11} esu, the electric anisotropy by (24) is of that of $10^{-9}\,I$ and is accessible to measurement at $I \simeq 10^{-4}$—10^5 esu. It is noteworthy that, recently, measurements of induced intensity-dependent rotation [15] and optical birefringence [16] in some organic liquids have been performed by laser techniques.

4. Magneto-optical phenomena

As stated at the outset, optically induced magnetic anisotropy is an inversed Cotton-Mouton effect, related with the Faraday effect by the equation [10]

$$n_{\sigma\tau}^2-n_0^2\delta_{\sigma\tau} = \left(\frac{n_0^2+2}{3}\right)\{F_{om}\varepsilon_{\sigma\tau\nu}H_\nu+A_{em}\delta_{\sigma\tau}H^2+B_{om}(3H_\sigma H_\tau-\delta_{\sigma\tau}H^2)\}, \tag{26}$$

which determines the linear and non-linear changes in optical permittivity tensor (electric permittivity tensor for optical frequencies) due to a strong, homogeneous magnetic field $\boldsymbol{H}$. $\varepsilon_{\sigma\tau\nu}$ is the Levi-Cività extensor.

Thus, Eq. (26) now describes both the linear effect of magnetic rotation of the polarization plane as given by the Faraday constant

$$F_{om}^{\blacksquare} = \frac{2\pi}{3V}\,\varepsilon_{\alpha\beta\gamma}\left\langle B_{\alpha\beta\gamma}^{om} + \frac{1}{kT}\,A_{\alpha\beta}^{o} M_{\gamma}^{m}\right\rangle \tag{27}$$

and the quadratic effect of magnetic birefringence given by the Cotton-Mouton constant B_{om}, whose shape in the diamagnetic case coincides with Eq. (10) provided we interchange therein the indices m and o. A detailed discussion of B_{om} is given elsewhere [10].

Also, the constant A_{om} consists of two parts analogous to (11) and (12); however, the change in volume is now related with magnetostriction (see Appendix B).

Applying in particular the Faraday constant (27) to a gas consisting of noninteracting molecules, one obtains directly Born's formula [11] (derived by formal molecular theory [12]):

$$F_{om} = \frac{2\pi}{3}\,\varrho\varepsilon_{\alpha\beta\gamma}\left(b_{\alpha\beta\gamma}^{om} + \frac{1}{kT}\,a_{\alpha\beta}^{o}\mu_{\gamma}^{m}\right), \tag{28}$$

where $\boldsymbol{\mu}^{m}$ is the permanent magnetic moment of the molecule, and $b_{\alpha\beta\gamma}^{om}$ — a tensor defining the change in optical polarizability tensor due to the linear magnetic field. The first term in (28) accounts for the diamagnetic effect, whereas the second term — for the paramagnetic effect directly dependent on the temperature.

From (26) we obtain for magneto-optical rotation:

$$n_{xy}^{2\,\mathrm{I}} - n_{yx}^{2\,\mathrm{II}} = 2\left(\frac{n_0^2+2}{3}\right) F_{om} H_z \tag{29}$$

if the incident light beam propagates parallel to the magnetic field acting along the Z-axis, whereas for magnetic birefringence we have

$$n_{xx}^2 - n_{yy}^2 = 3\left(\frac{n_0^2+2}{3}\right)^2 B_{om}(H_x^2 - H_y^2) \tag{30}$$

if propagation is parallel to the Z-axis and perpendicular to the XY-plane containing the strong magnetic field vector $\boldsymbol{H}$.

5. Application to special cases

Eqs (15)—(17), when applied to isotropically polarizable atoms or molecules, yield the simple formula

$$B_{mo} = \frac{2\pi}{3}\,\varrho(c_{3333}^{mo} - c_{1133}^{mo}) \tag{31}$$

meaning that in atomic substances magnetic anisotropy can arise solely as a result of the anisotropy induced in the atoms by the light beam of high intensity. The optical molecular orientation effect is in this case absent [3, 18].

For the case of axially symmetric molecules, the result on omitting the nonlinear deformation term is

$$B_{mo} = \frac{4\pi\varrho}{45kT}(a^m_{33}-a^m_{11})(a^o_{33}-a^o_{11})(1+J^{(2)}_A), \tag{32}$$

where [10]

$$J^{(2)}_A = \frac{1}{2}\frac{\varrho}{V}\int\int(3\cos^2\theta_{pq}-1)g^{(2)}(\tau_p, \tau_q)d\tau_p d\tau_q \tag{33}$$

is an integral parameter describing angular correlations between molecules p and q whose symmetry axes subtend the angle θ_{pq}.

The parameter (33) occurs also in the theory of molecular light scattering, thus in the depolarization ratio [19]

$$D = \frac{6(a^o_{33}-a^o_{11})^2(1+J^{(2)}_A)}{45\bar{a}^2_o\varrho kT\varkappa_T+7(a^o_{33}-a^o_{11})^2(1+J^{(2)}_A)}, \tag{34}$$

and can be calculated numerically from the experimental data [20] (a_o is the mean optical polarizability of the molecule).

Similarly, Eqs (20)—(23) yield for atoms

$$B_{eo} = \frac{2\pi}{3}\varrho(c^{eo}_{3333}-c^{eo}_{1133}), \tag{35}$$

whereas for dipolar axially-symmetric molecules (on omitting nonlinear deformation terms)

$$B^{\text{el}}_{eo} = \frac{4\pi\varrho}{45kT}\left\{(a^e_{33}-a^e_{11})(a^o_{33}-a^o_{11})(1+J^{(2)}_A)+\right.$$
$$\left.+\frac{\mu^2_e}{kT}(a^o_{33}-a^o_{11})(1+2J^{(1)}_A+J^{(2)}_A+J^{(3)}_A)\right\}, \tag{36}$$

where, in addition to (33), we have the correlation parameter

$$J^{(1)}_A = \frac{\varrho}{V}\int\int\cos\theta_{pq}\, g^{(2)}(\vec{\tau_p}, \tau_q)d\vec{\tau_p}d\tau_q \tag{37}$$

occurring in Kirkwood's theory of linear dielectrics [5], as well as the triplets angular correlation parameter

$$J^{(3)}_A = \frac{\varrho^2}{2V}\int\int\int(3\cos\theta_{pr}\cos\theta_{qr}-\cos\theta_{pq})g^{(3)}(\tau_p, \tau_q, \tau_r)\,d\tau_p d\tau_q d\tau_r \tag{38}$$

appearing in that of the Kerr effect [14].

Applying Eq. (36) to non-dipolar substances ($\mu_e = 0$) and recurring to Eq. (34), one obtains a relationship between the electric anisotropy constant and the depolarization ratio:

$$B_{eo} = 4\pi\varrho^2 a^2_o\frac{(a^e_{33}-a^e_{11})}{(a^o_{33}-a^o_{11})}\frac{\varkappa_T D}{6-7D}, \tag{39}$$

which can be checked experimentally.

Considering the relation

$$I^M = \left(\frac{3}{n^2+2}\right)^2 I \tag{40}$$

between the light intensity within the medium (I^M) and in vacuum (I), Eq. (24) can be rewritten thus:

$$\varepsilon_{xx}-\varepsilon_{yy} = 3\left(\frac{\varepsilon_0+2}{3}\right)^2\left(\frac{n^2+2}{3}\right)^2 B_{eo}(I^M_{xx}-I^M_{yy}). \tag{41}$$

For CS_2 we have [19] at $t = 20°C$: $n = 1.636$, $\varepsilon = 2.26$, $\varrho = 9.99\times10^{21}$ cm^{-3}, $\varkappa_T = 92\times$ $\times10^{-12}$ cgs, $D = 0.62$, $a_e = a_o = 8.77\times10^{-24}$ cm^3, whence Eq. (39) yields $B_{eo} = 3.3\times10^{-12}$, and in accordance with Eq. (41)

$$\varepsilon_{xx}-\varepsilon_{yy} = 55\times10^{-12}(I_{xx}-I_{yy}). \tag{42}$$

Using a laser beam of intensity 10^6 esu, one has an electric anisotropy of 5×10^{-5}, which is accessible to measurement by present techniques.

6. *Conclusions*

By semi-macroscopic theory, an isotropic medium is shown to become anisotropic in the presence of intense light, the variations of its electric and magnetic properties being described by the tensor equations (9) and (18), which contain *i.a.* the constants of optically--induced electric and magnetic anisotropy B_{eo} and B_{mo}. Eqs (10) and (19), which define these constants, are of sufficient generality for application to various special cases, such as rarefied or dense one-component systems (gases or liquids) or multi-component systems (gas mixtures, or solutions) consisting of mutually interacting or non-interacting molecules of arbitrary symmetry. The evaluated order of magnitude of the electric anisotropy in the case of carbon disulphide or nitrobenzene lies within the range of measurement by existing laser techniques which, ingeniously applied, have already permitted to measure certain other, equally subtle nonlinear effects [1, 2, 15—17]. Investigation of the phenomena considered above, in conjunction with work on the Cotton-Mouton and Kerr effects which has continued successfully over a period of many years, can provide information much fuller than has hitherto been possible on the linear and nonlinear electric, magnetic and optical properties of atoms and molecules subjected to intenese optical fields.

A detailed discussion of these effects, on a quantum-mechanical level and based on the foregoing general nonlinear formalism [21] will be given in a separate paper.

APPENDIX A

Derivation of Eqs (9), (18) and (26) from the free energy in external fields

All nonlinear effects discussed in this paper can be derived from the free energy

$$F(\boldsymbol{E}, \boldsymbol{H}) = -\beta^{-1}\ln Z(\boldsymbol{E}, \boldsymbol{H}), \tag{A1}$$

related with the partition function of the system

$$Z(\boldsymbol{E}, \boldsymbol{H}) = \Omega \int \exp\{-\beta U(\tau, \boldsymbol{E}, \boldsymbol{H})\}\, d\tau, \tag{A2}$$

which involves the total potential energy of the latter $U(\tau, \boldsymbol{E}, \boldsymbol{H})$ when its molecules are at configuration τ in the presence of external fields $\boldsymbol{E}$ and $\boldsymbol{H}$; $\beta = 1/kT$, and Ω is a normalizing factor such that $\Omega \int d\tau = 1$.

We consider the system to be in the DC magnetic field $\boldsymbol{H}$ and in the optical field $\boldsymbol{E}$ of the incident light wave of intensity I. In the case of a non-dissipative medium, the following expansion can be written, to within terms quadratic in H and linear in I (see Ref. [1]):

$$U(\tau, \boldsymbol{H}, \boldsymbol{I}) = U(\tau, 0) - M^m_{0\sigma} H_\sigma - \frac{1}{2} A^m_{\sigma\tau} H_\sigma H_\tau - \frac{1}{2} A^o_{\sigma\tau} I_{\sigma\tau} -$$

$$- \frac{1}{2} B^{mo}_{\sigma\tau\nu} H_\sigma I_{\tau\nu} - \frac{1}{4} C^{mo}_{\sigma\tau\nu\varrho} H_\sigma H_\tau I_{\nu\varrho} - \ldots, \tag{A3}$$

where the part of the mechanical energy has been omitted.

Since the energy (A3) does not depend explicitly on the time (the rapidly-variable terms vanish on time-averageing), it can be inserted into the partition function (A2) which, obviously, holds for systems at thermal equilibrium.

Dealing with the H- and I-dependent part of the energy (A3) as a perturbation to the energy $U(\tau, 0)$ of the non-perturbated system, we obtain from (A2) the consecutive approximations

$$Z_0 = \Omega \int \exp\{-\beta U(\tau, 0)\} d\tau,$$

$$Z_1 = \beta Z_0 \left\langle M^m_{0\sigma} H_\sigma + \frac{1}{2} A^m_{\sigma\tau} H_\sigma H_\tau + \frac{1}{2} A^o_{\sigma\tau} I_{\sigma\tau} + \right.$$

$$\left. + \frac{1}{2} B^{mo}_{\sigma\tau\nu} H_\sigma I_{\tau\nu} + \frac{1}{4} C^{mo}_{\sigma\tau\nu\varrho} H_\sigma H_\tau I_{\nu\varrho} + \ldots \right\rangle,$$

$$Z_2 = \frac{1}{2} \beta^2 Z_0 \left\langle M^m_{0\sigma} M^m_{0\tau} H_\sigma H_\tau + M^m_{0\sigma} A^o_{\nu\varrho} H_\sigma I_{\nu\varrho} + M^m_{0\sigma} B^{mo}_{\tau\nu\varrho} H_\sigma H_\tau I_{\nu\varrho} + \frac{1}{2} A^m_{\sigma\tau} A^o_{\nu\varrho} H_\sigma H_\tau I_{\nu\varrho} + \ldots \right\rangle,$$

$$Z_3 = \frac{1}{6} \beta^3 Z_0 \langle 3 M^m_{0\sigma} M^m_{0\tau} A^o_{\nu\varrho} H_\sigma H_\tau I_{\nu\varrho} + \ldots \rangle, \ldots, \tag{A4}$$

neglecting approximations of higher orders. The symbol $\langle\ \rangle$ stands for the statistical average calculated in the absence of external fields:

$$\langle G \rangle = Z_0^{-1} \Omega \int G \exp\{-\beta U(\tau, 0)\} d\tau. \tag{A5}$$

We assume the summation indices $\sigma, \tau, \nu, \varrho$ in (A3) and (A4) to refer to the laboratory coordinates in which the direction of the magnetic fields is given. For further calculations, it is convenient to transform the tensors $M_{0\sigma}$, $A_{\sigma\tau}$, $B_{\sigma\tau\nu}$, ... from the laboratory reference

system to one attached to the medium (or to any single molecule) with axes labelled by the indices $\alpha, \beta, \gamma, \delta$. The transformation formulas are of the form

$$M_\sigma = \omega_{\sigma\alpha} M_\alpha, \qquad A_{\sigma\tau} = \omega_{\sigma\alpha}\omega_{\tau\beta} A_{\alpha\beta}, \ldots, \tag{A6}$$

where, if the axes of either reference system are rectangular, the transformation coefficients $\omega_{\sigma\alpha}$, $\omega_{\tau\beta}$, ... have the meaning of directional cosines of the angles between the respective axes.

For the case of an isotropic medium in the absence of external fields all orientations of the axes $\alpha, \beta, \ldots$ with regard to the axes $\sigma, \tau, \ldots$ are equally probable, and averageing yields [22]

$$\langle \omega_{\sigma\alpha} \rangle = 0, \langle \omega_{\sigma\alpha}\omega_{\tau\beta} \rangle = \frac{1}{3}\,\delta_{\alpha\beta}\delta_{\sigma\tau}, \langle \omega_{\sigma\alpha}\omega_{\tau\beta}\omega_{\nu\gamma} \rangle = \frac{1}{6}\,\varepsilon_{\alpha\beta\gamma}\varepsilon_{\sigma\tau\nu},$$

$$\langle \omega_{\sigma\alpha}\omega_{\tau\beta}\omega_{\nu\gamma}\omega_{\varrho\delta} \rangle = \frac{1}{30}\,\{(4\delta_{\alpha\beta}\delta_{\gamma\delta} - \delta_{\alpha\gamma}\delta_{\beta\delta} - \delta_{\alpha\delta}\delta_{\beta\gamma})\delta_{\sigma\tau}\delta_{\nu\varrho} + (4\delta_{\alpha\gamma}\delta_{\beta\delta} -$$

$$- \delta_{\alpha\delta}\delta_{\beta\gamma} - \delta_{\alpha\beta}\delta_{\gamma\delta})\delta_{\sigma\nu}\delta_{\tau\varrho} + (4\delta_{\alpha\delta}\delta_{\beta\gamma} - \delta_{\alpha\beta}\delta_{\gamma\delta} - \delta_{\alpha\gamma}\delta_{\beta\delta})\delta_{\sigma\varrho}\delta_{\tau\nu}\}. \tag{A7}$$

Here, $\delta_{\alpha\beta}$ is the Kronecker unit tensor, and $\varepsilon_{\alpha\beta\gamma}$ — the Levi-Cività pseudo-tensor.

By the transformation formulas (A6) and mean values (A7), and on adding up the consecutive approximations of the partition function (A4) and ordering according to powers of the field strengths, we have

$$Z(\boldsymbol{H}, \boldsymbol{I}) = Z_0 \Big\{ 1 + \frac{1}{6}\,\beta\,\langle A^m_{\alpha\alpha} + \beta M^m_{0\alpha} M^m_{0\alpha} \rangle H^2 + \frac{1}{6}\,\beta\,\langle A^o_{\alpha\alpha} \rangle I +$$

$$+ \frac{1}{12}\,\beta\varepsilon_{\alpha\beta\gamma}\langle B^{mo}_{\alpha\beta\gamma} + \beta M^m_{0\alpha} A^o_{\beta\gamma} \rangle\, \varepsilon_{\sigma\tau\nu} H_\sigma I_{\tau\nu} +$$

$$+ \frac{1}{360}\,\beta\,[10\delta_{\alpha\beta}\delta_{\gamma\delta} I\delta_{\sigma\tau} + (3\delta_{\alpha\gamma}\delta_{\beta\delta} + 3\delta_{\alpha\delta}\delta_{\beta\gamma} - 2\delta_{\alpha\beta}\delta_{\gamma\delta}) \times$$

$$\times (3I_{\sigma\tau} - I\delta_{\sigma\tau})]\,\langle C^{ma}_{\alpha\beta\gamma\delta} + 2\beta M^m_{0\alpha} B^{mo}_{\beta\gamma\delta} + \beta A^m_{\alpha\beta} A^o_{\gamma\delta} + \beta^2 M^m_{0\alpha} M^m_{0\beta} A^o_{\gamma\delta} \rangle\, H_\sigma H_\tau + \ldots \Big\}. \tag{A8}$$

Now, since the magnetic polarization vector component is

$$P_{m\sigma} = -\,\frac{1}{V}\left(\frac{\partial F}{\partial H_\sigma}\right)_{T,V}, \tag{A9}$$

we have by (A8) in the present approximation

$$P_{m\sigma} = \frac{1}{3V}\,\langle A^m_{\alpha\alpha} + \beta M^m_{0\alpha} M^m_{0\alpha} \rangle\, H_\sigma +$$

$$+ \frac{1}{12V}\,\varepsilon_{\alpha\beta\gamma}\langle B^{mo}_{\alpha\beta\gamma} + \beta M^m_{0\alpha} A^o_{\beta\gamma} \rangle\, \varepsilon_{\sigma\tau\nu} I_{\tau\nu} +$$

$$+ \frac{1}{180V}\,[10\delta_{\alpha\beta}\delta_{\gamma\delta} I\delta_{\sigma\tau} + (3\delta_{\alpha\gamma}\delta_{\beta\delta} + 3\delta_{\alpha\delta}\delta_{\beta\gamma} - 2\delta_{\alpha\beta}\delta_{\gamma\delta}) \times$$

$$\times (3I_{\sigma\tau} - I\delta_{\sigma\tau})]\,\langle C^{mo}_{\alpha\beta\gamma\delta} + 2\beta M^m_{0\alpha} B^{mo}_{\beta\gamma\delta} + \beta A^m_{\alpha\beta} A^o_{\gamma\delta} + \beta^2 M^m_{0\alpha} M^m_{0\beta} A^o_{\gamma\delta} \rangle H_\tau + \ldots. \tag{A10}$$

In (A10), the first term defines linear magnetisation, the second — magnetisation due to light alone [1, 2] and the third — magnetisation due to the simultaneous action of the magnetic field and the intense light beam.

Inserting the magnetisation (A10) in the fundamental equation (3) one obtains immediately Eq. (9) for the permeability tensor; however, the constants A_{mo} and B_{mo} are now of more generality, and formally applicable to paramagnetics as well:

$$A^P_{mo} = \frac{2\pi}{9V}\{\langle C^{mo}_{\alpha\alpha\beta\beta}\rangle + 2\beta\langle M^m_{o\alpha} B^{mo}_{\alpha\beta\beta}\rangle + \\ + \beta\langle (A^m_{\alpha\alpha} + \beta M^m_{o\alpha} M^m_{o\alpha})(A^o_{\beta\beta} - \langle A^o_{\beta\beta}\rangle)\}, \tag{A11}$$

$$B_{mo} = \frac{\pi}{45V}(3\delta_{\alpha\gamma}\delta_{\beta\delta} + 3\delta^{\alpha\gamma}_{\alpha\delta}\delta^{\beta\gamma}_{\beta\gamma} - 2\delta^{\alpha\beta}_{\alpha\beta}\delta^{\gamma\delta}_{\gamma\delta})\langle C^{mo}_{\alpha\beta\gamma\delta} + \\ + 2\beta M^m_{0\alpha} B^{mo}_{\beta\gamma\delta} + \beta A^m_{\alpha\beta} A^o_{\gamma\delta} + \beta^2 M^m_{0\alpha} M^m_{0\beta} A^o_{\gamma\delta}\rangle. \tag{A12}$$

If in particular $M^m_0 = 0$, as is the case of diamagetics, (A11) and (A12) go over directly into Eqs (10) and (11).

With regard to (3) and (A9), the permeability tensor can be linked directly to the free energy:

$$\mu_{\sigma\tau} - \delta^{\sigma\tau}_{\sigma\tau} = -\frac{4\pi}{V}\left(\frac{\partial^2 F}{\partial H_\sigma \partial H_\tau}\right)_{T,V} \tag{A13}$$

if V is assumed independent of H.

On replacing magnetic by electrical quantities in (A3) et seqq., one comes to Eqs (18) and (19) for the variations of the electrical properties of the medium under the effect of intense light. Similarly, the method leads to the fundamental equation (26) for magneto-optical phenomena.

APPENDIX B

The optico-strictional and optico-caloric effects

When the medium is acted on by light of high intensity I, the variations of its electric (1) and magnetic (2) permeabilities are given as [3]

$$\Delta\varepsilon = \Delta\varepsilon_I + \Delta\varepsilon_V + \Delta\varepsilon_T \tag{B1}$$

$$\Delta\mu = \Delta\mu_I + \Delta\mu_V + \Delta\mu_T. \tag{B2}$$

The variations $\Delta\varepsilon_I$ and $\Delta\mu_I$ due to the optical field have been calculated in Appendix A; here, we shall derive those related with optico-striction ($\Delta\varepsilon_V$, $\Delta\mu_V$) and with the optico-caloric effect ($\Delta\varepsilon_T$, $\Delta\mu_T$). We begin by introducing the total thermodynamical potential Φ related with the free energy (A1) as follows [23]:

$$d\Phi = -SdT + Vdp + dF, \tag{B3}$$

S denoting the entropy, T — the temperature and p — the pressure.

The increase in electric permeability related with a change in volume $\Delta V = V_I - V$ is

$$\Delta\varepsilon_V = \left(\frac{\partial\varepsilon}{\partial V}\right)_T \Delta V, \tag{B4}$$

where, by (A1), (A8) and (B3), and on neglecting higher order terms, we have

$$V_I = \left(\frac{\partial\Phi}{\partial p}\right)_T = V - \frac{1}{6}\left\{\frac{\partial}{\partial p}\langle A^o_{\alpha\alpha}\rangle\right\}_T I - \\ - \frac{1}{6}\left\{\frac{\partial}{\partial p}\langle A^m_{\alpha\alpha} + \beta M^m_{0\alpha} M^m_{0\alpha}\rangle\right\}_T H^2 - \ldots \tag{B5}$$

Since in the absence of external fields (A11) and (A13) yield

$$n^2 - 1 = \frac{4\pi}{3V}\langle A^o_{\alpha\alpha}\rangle \quad \text{and} \quad \mu - 1 = \frac{4\pi}{3V}\langle A^m_{\alpha\alpha} + \beta M^m_{0\alpha} M^m_{0\alpha}\rangle, \tag{B6}$$

the change of volume can be expressed in the form of

$$\Delta V = -\frac{1}{8\pi}\left\{\frac{\partial}{\partial p}[(n^2-1)V]\right\}_T I - \frac{1}{8\pi}\left\{\frac{\partial}{\partial p}[(\mu-1)V]\right\}_T H^2, \tag{B7}$$

where the term proportional to the light intensity I defines the effect of optico-striction (electro-striction [23] at optical frequencies), whereas the one proportional to H^2 defines magneto-striction [6, 10].

Restricting further considerations to optico-striction, we have by (B7)

$$\Delta V = -\frac{V}{8\pi}\left\{\left(\frac{\partial n^2}{\partial p}\right)_T - (n^2-1)\varkappa_T\right\} I, \tag{B8}$$

$\varkappa_T = -\frac{1}{V}\left(\frac{\partial V}{\partial p}\right)_T$ being the isothermal compressibility.

Since by analogy to (B6) we have

$$\varepsilon - 1 = \frac{4\pi}{3V}\langle A^e_{\alpha\alpha} + \beta M^e_{0\alpha} M^e_{0\alpha}\rangle, \tag{B9}$$

$\left(\frac{\partial\varepsilon}{\partial V}\right)_T = -\frac{\varepsilon-1}{V}$ and Eqs (B4), (B8) finally yield

$$\Delta\varepsilon_V = -\frac{V}{8\pi}\left\{\left(\frac{\partial n^2}{\partial p}\right)_T - (n^2-1)\varkappa_T\right\}\left(\frac{\partial\varepsilon}{\partial V}\right)_T I \\ = \frac{\varepsilon-1}{8\pi}\left\{\left(\frac{\partial n^2}{\partial p}\right)_T - (n^2-1)\varkappa_T\right\}\left(\frac{n^2+2}{3}\right)^2 I_M, \tag{B10}$$

where I_M is the light intensity within the medium given by Eq. (40).

Similarly,

$$\Delta\varepsilon_T = \left(\frac{\partial\varepsilon}{\partial T}\right)_V \Delta T, \tag{B11}$$

where the rise in temperature due to absorption of heat by the system when the external fields are switched on adiabatically is

$$\Delta T = \frac{T}{C_p}\left(\frac{\partial}{\partial T}\,\Delta\Phi\right)_p = -\frac{T}{8\pi C_p}\left\{\frac{\partial}{\partial T}\,[(n^2-1)V]\right\}_p I - \\ -\frac{T}{8\pi C_p}\left\{\frac{\partial}{\partial T}\,[(\mu-1)V]\right\}_p H^2. \tag{B12}$$

Here, the first term defines the optico-caloric and the second — the magneto-caloric effect. $C_p = T\left(\frac{\partial S}{\partial T}\right)_p$ is the specific heat at constant pressure.

Considering solely the rise in temperature due to the optico-caloric effect we have

$$\Delta T = -\frac{TV}{8\pi C_p}\left\{\left(\frac{\partial n^2}{\partial T}\right)_p + (n^2-1)\alpha_p\right\} I, \tag{B13}$$

where $\alpha_p = \frac{1}{V}\left(\frac{\partial V}{\partial T}\right)_p$ is the volume oeficient of expansion.

The expressions (B8) and (B13) are counterparts of electro-striction and the electro--caloric effect [23].

From (B9), (B11), (B13) the variation in electric permeability resulting from the optico--caloric effect is

$$\Delta\varepsilon_T = -\frac{VT}{8\pi C_p}\left(\frac{\partial\varepsilon}{\partial T}\right)_V\left\{\left(\frac{\partial n^2}{\partial T}\right)_p + (n^2-1)\alpha_p\right\} I. \tag{B14}$$

Quite similarly, one calculates the changes in magnetic permeability and refractive index due to optico-striction and the optico-caloric effect. In particular, the change in refractive index from optico-striction, by (A8), is

$$\Delta n^2_V = \left(\frac{\partial n^2}{\partial V}\right)_T \Delta V = \frac{n^2-1}{8\pi}\left\{\left(\frac{\partial n^2}{\partial p}\right)_T - (n^2-1)\varkappa_T\right\}\left(\frac{n^2+2}{3}\right)^2 I_M. \tag{B15}$$

Numerical evaluations for optico-striction [24—26] show that in liquids its contribution is of an order comparable to that of optical molecular orientation [3, 18], whereas in isotropic solids it is predominant [24].

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