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Nonlinear Magnetooptics of Colloids

STANISŁAW KIELICH

Department of Molecular Physics, A. Mickiewicz University, Poznań, Grunwaldzka 6, Poland

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Various nonlinearities due to the action of a strong d-c magnetic field and of the electric field of an intense laser beam on the optical or magnetic properties of isotropic bodies are discussed. The treatment is by classical theory along both phenomenological and statistical-microscopic lines. Three new magnetooptical effects are suggested for quantitative experimental study: (1) an inverse Cotton-Mouton, where calculations show that strong laser light induces magnetic anisotropy in a medium; (2) nonlinear optical variations in Faraday effect under the influence of laser light; and (3) magnetooptical cross birefringence, which is shown to result even if both the probe and laser beams propagate parallel to the magnetic field, as in linear Faraday. The laser and high magnetic field techniques now available are shown to make feasible the measurement of these effects, preferably in macromolecular and colloidal substances. Since such substances moreover easily permit total alignment of their microsystems in a d-c magnetic field or in the electric field of a laser beam, measurements of the anticipated effects at optical, magnetic, or magnetooptical saturation are suggested as a direct method of determining the sign and value of the optical and magnetic anisotropies of macromolecules and colloid particles.

INTRODUCTION

Magnetooptics takes existence from the fundamental work of Faraday, Kerr, Majorana, Cotton, and Mouton, and has since been the subject of extensive studies presented and reviewed in various monographs (1–4) and articles (5–8). Faraday (1845) showed plane polarized light, propagating in a medium in the direction of an externally applied magnetic field H, to undergo a rotation of the plane of polarization. With n_- and n_+ denoting the refractive indices at frequency $\omega = 2\pi c/\lambda$ for left and right circularly polarized light, respectively, the magnetooptical rotation is (2):

$$n_- - n_+ = \frac{\lambda}{\pi} V_\lambda H, \qquad [1]$$

where V_λ is the Verdet constant, specific to the medium and a function of the light wavelength λ.

If the light propagates at right angles to the magnetic field, the (naturally nonbirefringent) medium—as first shown by Majorana (1902) in colloidal solutions and subsequently by Cotton and Mouton (1905) in pure liquids—becomes optically birefringent with optical axis parallel to the direction of the magnetic field. The amount of this birefringence is given by the difference between the refractive indices for components of the light vibration parallel and perpendicular to the direction of the field:

$$n_\parallel - n_\perp = \lambda C_\lambda H^2, \qquad [2]$$

where C_λ is the Cotton-Mouton constant.

One of the earliest theoretical explanations of magnetooptical phenomena was proposed by Voigt (1) on the basis of Lorentz's (9) electron theory of matter. Voigt interpreted magnetically induced birefringence as due to the direct action of the applied magnetic field on the electrons

of atoms and molecules, conceived as isotropic oscillators, and undergoing an anisotropic deformation proportional to the square of the field strength H in accordance with Eq. [2]. By Voigt's theory, the absolute retardation ratio should be:

$$\frac{n_\parallel - n}{n_\perp - n} = 3, \qquad [3]$$

whereas the majority of experiments led to Havelock's (5) relation:

$$\frac{n_\parallel - n}{n_\perp - n} = -2, \qquad [4]$$

n being the refractive index in the absence of an applied magnetic field.

Langevin (10), by statistical mechanics, proved Havelock's relation for substances consisting of anisotropic molecules. According to Langevin, although molecules are generally anisotropic, their distribution in a fluid is entirely random, all orientations being equally probable, and the fluid as a whole remaining isotropic. When a strong magnetic field is applied, a new state of thermodynamical equilibrium sets in, favoring orientation in the magnetic field direction in accordance with the distribution function of Maxwell-Boltzmann statistics (10):

$$f(\mathbf{H}) = f(0)\left\{1 + \frac{a_\parallel{}^m - a_\perp{}^m}{6kT} \cdot \left(3\cos^2\vartheta - 1\right)H^2 + \cdots\right\}, \qquad [5]$$

where $f(0)$ is a distribution function defining the molecular orientation for equal probability in the absence of the field, and $a_\parallel{}^m$, $a_\perp{}^m$ are magnetic polarizabilities of the molecule parallel and perpendicular to its symmetry axis, the latter forming the angle ϑ with the field vector $\mathbf{H}$. From Eq. [5], the magnetic birefringence should increase as the square of the field strength H and decrease with the absolute temperature T; experiments have indeed shown this to be the case in most liquids.

The Cotton-Mouton constant resulting from Voigt's effect is of the form (2, 11)

$$C_\lambda^{NL} = \frac{\pi\rho}{n\lambda}(\eta_\parallel^{\omega m} - \eta_\perp^{\omega m}), \qquad [6]$$

where ρ stands for the number density of molecules, which undergo two different nonlinear changes in optical polarizability at frequency ω, $\eta_\parallel^{\omega m}$ and $\eta_\perp^{\omega m}$, in directions respectively parallel and perpendicular to the magnetic field $\mathbf{H}$. Voigt's effect plays the predominant role in atomic gases only, in which it is the sole process giving rise to birefringence (12).

Let us denote by $a_\parallel{}^\omega$ and $a_\perp{}^\omega$ the optical polarizabilities at frequency ω, respectively parallel and perpendicular to the axis of symmetry of the molecule. Langevin's effect of molecular reorientation, with the distribution function of Eq. [5], now leads to the following expression (10):

$$C_\lambda^{RO} = \frac{2\pi\rho}{15n\lambda kT}(a_\parallel{}^\omega - a_\perp{}^\omega)(a_\parallel{}^m - a_\perp{}^m), \qquad [7]$$

which permits determinations of the magnetic polarizabilities $a_\parallel{}^m$ and $a_\perp{}^m$ from measurements of magnetic birefringence in gases (12) and very dilute solutions (13). Studies of the Cotton-Mouton effect in liquids and solutions provide information regarding the nature and kind of intermolecular forces as well as the microscopic structure of the fluid (11, 14–16). Also, the magnetooptics of polymers and colloids is developing (3, 5, 6, 17–20); indeed, in such systems, total orientation of the particles is easily achieved in the magnetic field—a state referred to as magnetic birefringence saturation (18).

In recent years, a technique of obtaining very strong magnetic fields by pulse methods (16) has been evolved. Fields of the order of as much as 10^6 oe are achieved in measurable volumes. In such strong magnetic fields, the laws given by formulas [1] and [2] are invalidated even in gases (21); nonlinear changes of order higher than the second are produced in the refractive index,

and magnetic saturation is achieved even in substances consisting of simple molecules.

Moreover, laboratories can now use gigantic ruby lasers for the production of intense electric fields of the order of 10^5 v/cm in nonfocused and 10^9 v/cm in focused beams. This has led to the detection of a variety of nonlinear optical effects in various substances (22, 23). Among others, a strong electric field can cause the Verdet constant of Eq. [1] to become a nonlinear function of the light intensity (24). The Cotton-Mouton constant of Eq. [2] is affected similarly, as we shall show in the course of this paper. Obviously not only the optical but also the electric and magnetic properties of a body undergo variations under the influence of intense light (14, 25).

Debye (26) and subsequently Peterlin and Stuart (27) investigated the orientation of polar molecules (having an electric dipole moment μ_e) in an alternating electric field $\mathbf{E}^\omega = \mathbf{E}^0 e^{i\omega t}$ oscillating at frequency ω; here, orientation is given by the distribution function (28):

$$f(\mathbf{E}) = f(0)\left\{1 + \frac{\mu_e E_0 e^{i\omega t}\cos\vartheta}{kT(1 + i\omega\tau_D)} + \frac{E_0^2}{12kT}\left[a_\parallel^\omega - a_\perp^\omega + \frac{\mu_e^2}{kT(1 + i\omega\tau_D)}\right](3\cos^2\vartheta - 1)\cdot\left(1 + \frac{e^{i2\omega t}}{1 + i2\omega\tau_B}\right)\right\}, \quad [8]$$

where

$$\tau_D = \frac{4\pi r^3\eta}{kT} \quad [9]$$

is the Debye relaxation time of a molecule of radius r subject to a reorientation process in a medium of viscosity η, whereas $\tau_B = \tau_D/3$ is the relaxation time of birefringence (27).

In particular, at $\omega = 0$, the distribution function [8] takes the form used by Langevin and Born in their theory of Kerr's effect. In the other limiting case, when simultaneously $\omega\tau_D \to \infty$ and $\omega\tau_B \to \infty$, as occurs at optical frequencies, we have:

$$f(\mathbf{E}) = f(0)\left\{1 + \frac{a_\parallel^\omega - a_\perp^\omega}{12kT}\cdot\left(3\cos^2\vartheta - 1\right)E_0^2\right\}. \quad [10]$$

Thus one sees that although the permanent electric dipoles are unable to keep pace with the oscillations of the electric field, the electron polarizability ellipsoid of the molecule nevertheless does undergo an orientation. In this way, as shown by Buckingham (29), intense light can induce birefringence in a medium; this indeed has been observed to take place in liquids when laser technique is applied (30). Also, circular birefringence induced by a circularly polarized laser beam (31) has been detected.

It is well known that in molecular liquids the relaxation times range (32) from 10^{-10} to 10^{-13} sec and are thus sufficiently short for reorientation to take place during a single laser pulse, usually lasting from 10^{-6} to 10^{-12} sec. It can be asked whether these conditions admit of the reorientation of macromolecules or colloid particles. Now, relaxation times for aqueous solutions of protein molecules (33) amount to 10^{-6}–10^{-8} sec, so that reorientation can still occur in the electric fields of the light of certain lasers. Recently, optical orientation of gold particles in water has been observed at this Laboratory by Kaczmarek (34), who used a ruby laser of a pulse duration of 10^{-6} sec and an electric field strength at the focus of the order of 3×10^5 v/cm. The pulse duration of ruby lasers can be extended to 10^{-3}–10^{-4} sec, and on applying a two-step amplifying cascade one gets a beam with electric field strength about 10^4 v/cm. In the case of the larger macromolecules and particles, optical orientation can be achieved with the electric fields of focused beams from continuously operating gas or ion lasers.

It is thus obvious that present laboratory equipment is well adapted to the study of the various new magnetooptical effects which result specifically when a medium is acted on simultaneously by a strong magnetic field and by the electric field of an intense laser beam.

In particular, apart from the previously considered effects, we propose here for experimental investigation the magnetic anisotropy induced in media by intense light—an effect that may justly be referred to as inverse Cotton-Mouton (35), since the agent inducing magnetic anisotropy in a naturally isotropic medium is now laser light. Terminologically, this is an opticomagnetic effect. It is noteworthy that an inverse Faraday effect has recently been discovered by Pershan *et al.* (36) by laser technique. We moreover intend to consider changes in the optical and magnetic properties of bodies due to optical (37) and magnetic (18) as well as to magnetooptical saturation. However, we shall refrain from discussing variations due to strong d-c electric fields as belonging to the domain of electrooptics (5, 38, 39).

Obviously, besides the classical theory touched on above, quantum-mechanical theories of magnetooptical phenomena (40–42) have been evolved. The considerations of this paper, however, will be on a classical level and based on phenomenological-statistical methods (22, 43–45), which will enable us to derive results valid for arbitrary systems regardless of their microstructure and state of condensation. These results can be particularized to molecular, macromolecular, and dispersed systems.

THEORETICAL BACKGROUND

Consider incident upon an arbitrary medium an electromagnetic wave (probe light) with electric vector $\mathbf{E}^{\omega} = \mathbf{E}^{0}e^{i\omega t}$ oscillating at frequency ω, inducing in it an electric polarization vector $\mathbf{P}_e^{\omega}$ at the same frequency.

If the medium is isotropic in the absence of externally applied fields, one has the relation:

$$(\epsilon^{\omega} - 1)\mathbf{E}^{\omega} = 4\pi\mathbf{P}_e^{\omega}, \qquad [11]$$

where ϵ^{ω} is the scalar electric permittivity of the medium at frequency ω.

In the case of an anisotropic medium, the electric permittivity is a second rank tensor, $\epsilon_{\sigma\tau}$, the σ, τ-components of which if referred to a Cartesian coordinate system can be arrayed in the shape of a 3×3 matrix (43):

$$(\epsilon_{\sigma\tau}) = \begin{pmatrix} \epsilon_{xx} & \epsilon_{xy} & \epsilon_{xz} \\ \epsilon_{yx} & \epsilon_{yy} & \epsilon_{yz} \\ \epsilon_{zx} & \epsilon_{zy} & \epsilon_{zz} \end{pmatrix}. \qquad [12]$$

The number of its nonvanishing components and the mutual relationships between these components will in each case depend on the symmetry elements presented by the medium. Thus, in the simplest case of an isotropic body, the six nondiagonal components of the tensor matrix [12] will vanish, and the three remaining (diagonal) ones will be equal to one another, so that we can write

$$\epsilon_{\sigma\tau} = \epsilon\delta_{\sigma\tau}, \qquad [13]$$

$\delta_{\sigma\tau}$ being the unit Kronecker tensor with components equaling unity for $\sigma = \tau$ and vanishing for $\sigma \neq \tau$.

For an anisotropic body, the vectorial relation [11] has to be replaced by the following, fundamental relation in tensor notation:

$$(\epsilon_{\sigma\tau}^{\omega} - \delta_{\sigma\tau})E_{\tau}^{\omega} = 4\pi P_{e\sigma}^{\omega}, \qquad [14]$$

where we have applied (and shall henceforth apply) Einstein's summation convention, stating that an index appearing twice (here τ) indicates a summation. Obviously, with regard to [13], Eq. [14] immediately goes over into Eq. [11].

If, as was the case of classical light sources, the electric field strength $\mathbf{E}^{\omega}$ is not very large, the medium undergoes a linear polari-

zation (depending on the first power of the field strength only):

$$P_{e\sigma}^{\omega} = \chi_{\sigma\tau}^{e\omega} E_{\tau}^{\omega}, \qquad [15]$$

where $\chi_{\sigma\tau}^{e\omega}$ is the tensor of linear electric susceptibility at frequency ω, which, being a second-rank tensor, can be put in matrix form like [12]. With regard to Eqs. [14] and [15], we have the following relation between the two tensors:

$$\epsilon_{\sigma\tau}^{\omega} - \delta_{\sigma\tau} = 4\pi\chi_{\sigma\tau}^{e\omega}.$$

The linear relation [15] holds as long as the probe light is weak and the medium is not acted on by external forces. Now, if the medium is subjected to an external, e.g., magnetic field $\mathbf{H}$ of sufficiently high intensity, the polarization vector will in general become a nonlinear function $\mathbf{P}_e^{\omega}(\mathbf{H})$ of $\mathbf{H}$. It has to be stated clearly that if, as we shall see further on, the medium undergoing nonlinear polarization is a naturally isotropic one, it becomes anisotropic and its electric properties have to be expressed by a tensor equation of the form [14].

In considering a naturally isotropic medium of volume V we are interested in properties that are averaged microscopic quantities. Thus, in a dipole approximation, the electric polarization vector can be written in the form:

$$V\mathbf{P}_e^{\omega} = \int \mathbf{M}_e^{\omega}(\tau, \mathbf{E}^{\omega}, \mathbf{F}) f(\tau, \mathbf{F})\, d\tau, \qquad [16]$$

$\mathbf{M}_e^{\omega}(\tau, \mathbf{E}^{\omega}, \mathbf{F})$ being the electric dipole moment induced in the medium by the electric field $\mathbf{E}^{\omega}$ in the presence of an external field $\mathbf{F}$ when the microsystems (atoms, molecules, macromolecules, or particles) are at configuration τ and their statistical distribution at thermodynamical equilibrium of the system as a whole is defined by the function

$$f(\tau, \mathbf{F}) = \frac{\exp\{-\beta \cup (\tau, \mathbf{F})\}}{\int \exp\{-\beta \cup (\tau, \mathbf{F})\}\, d\tau}. \qquad [17]$$

Here $\beta = 1/kT$, whereas integration in [16] and [17] extends over all possible configurations of the microsystems.

The total potential energy of the system $U(\tau, \mathbf{F})$ at τ and $\mathbf{F}$ can be resolved into a nonperturbated part $U(\tau, 0)$ in the absence of the external field ($\mathbf{F} = 0$) and a perturbated part $W(\tau, \mathbf{F})$ due to the action of the field $\mathbf{F}$ on the system. In a great number of problems, the perturbation energy W is very small as compared to $U(\tau, 0)$ and kT, so that one is justified in expanding the Boltzmann factor in [17] in a power series and writing in a second approximation

$$f(\tau, \mathbf{F}) = f(\tau, 0)\{1 - \beta(W - \langle W\rangle) + \tfrac{1}{2}\beta^2[W^2 - \langle W^2\rangle - 2(W - \langle W\rangle)\langle W\rangle] + \cdots\}, \qquad [18]$$

where

$$\langle W^n\rangle = \int W^n(\tau, \mathbf{F}) f(\tau, 0)\, d\tau \qquad [19]$$

is the statistical average calculated with the function $f(\tau, 0)$ of the nonperturbated state by Eq. [17] for $\mathbf{F} = 0$.

In general, the magnetic vector $\mathbf{H}^{\omega} = \mathbf{H}^0 e^{i\omega t}$ of the light wave will also cause magnetic polarization of the medium given by a vector $\mathbf{P}_m^{\omega}$, and in analogy to Eq. [14] we write for the tensor of magnetic permittivity:

$$(\mu_{\sigma\tau}^{\omega} - \delta_{\sigma\tau}) H_{\tau}^{\omega} = 4\pi P_{m\sigma}^{\omega}. \qquad [20]$$

On going over to the microscopic interpretation we average this polarization as follows:

$$V\mathbf{P}_m^{\omega} = \int \mathbf{M}_m^{\omega}(\tau, \mathbf{H}^{\omega}, \mathbf{F}) f(\tau, \mathbf{F})\, d\tau, \qquad [21]$$

with $\mathbf{M}_m^{\omega}(\tau, \mathbf{H}, \mathbf{F})$ denoting the magnetic dipole moment induced in the medium by the field $\mathbf{H}^{\omega}$ in the presence of the field $\mathbf{F}$.

FARADAY EFFECT

Consider the medium to be in a d-c homogeneous magnetic field $\mathbf{H}$ of strength so low as to cause only linear polarization.

In addition to the electric linear polarization given by Eq. [15], we shall now have moreover a mixed (second-order) polarization:

$$P^{\omega}_{e\sigma} = \chi^{e\omega m}_{\sigma\tau\nu} E^{\omega}_{\tau} H_{\nu}, \qquad [22]$$

where the electric susceptibility pseudotensor $\chi^{e\omega m}_{\sigma\tau\nu}$ of third rank defines the linear changes in electric polarizability of the medium induced by the magnetic field.

On inserting [15] and [22] into the fundamental equation [14], we obtain for an arbitrary body:

$$\epsilon^{\omega}_{\sigma\tau} - \delta_{\sigma\tau} = 4\pi(\chi^{e\omega}_{\sigma\tau} + \chi^{e\omega m}_{\sigma\tau\nu} H_{\nu}). \qquad [23]$$

For an isotropic body, we obtain:

$$\chi^{e\omega}_{\sigma\tau} = \chi_e{}^{\omega}\delta_{\sigma\tau}, \qquad \chi^{e\omega m}_{\sigma\tau\nu} = \chi^{\omega}_{e\,m}\epsilon_{\sigma\tau\nu}, \qquad [24]$$

where the scalar susceptibilities are:

$$\chi_e{}^{\omega} = \tfrac{1}{3}(\chi^{e\omega}_{xx} + \chi^{e\omega}_{yy} + \chi^{e\omega}_{zz}), \qquad [24a]$$

$$\chi^{\omega}_{em} = \tfrac{1}{6}(\chi^{e\omega m}_{xyz} - \chi^{e\omega m}_{yxz} + \chi^{e\omega m}_{yzx} - \chi^{e\omega m}_{zyx} + \chi^{e\omega m}_{zxy} - \chi^{e\omega m}_{xzy}), \qquad [24b]$$

$\epsilon_{\sigma\tau\nu}$ being the Levi-Cività extensor with components equal to 1 or -1 according to whether $\sigma\tau\nu$ is an even or odd permutation of xyz, and to zero if any two indices are the same.

With regard to [24], we can rewrite [23] as follows:

$$\epsilon^{\omega}_{\sigma\tau} - \epsilon_0{}^{\omega}\delta_{\sigma\tau} = 4\pi\chi^{\omega}_{em}\epsilon_{\sigma\tau\nu}H_{\nu} \qquad [25]$$

with $\epsilon_0{}^{\omega} - 1 = 4\pi\chi_e{}^{\omega}$ denoting the electric permittivity in the absence of a magnetic field.

Once we assume the magnetic field as directed along the z-axis (which is the propagation direction of the probe light), only the nondiagonal components of [25] will be nonzero:

$$\epsilon^{\omega}_{xy} = -\epsilon^{\omega}_{yx} = \frac{n\lambda}{\pi i} V_{\lambda} H_z, \qquad [26]$$

Verdet's constant now being of the form:

$$V_{\lambda} = \frac{4\pi^2 i}{n\lambda}\chi^{\omega}_{em}. \qquad [27]$$

If now we take into account that the indices for right and left rotating oscillations are (2):

$$n_{\pm} = n \mp \frac{i\epsilon^{\omega}_{xy}}{2n}$$

we go over directly from [26] to Eq. [1].

Similar calculations yield contributions to Verdet's constant from the magnetic polarization induced by the magnetic vector $\mathbf{H}^{\omega}$ of the light wave in the presence of a d-c magnetic field $\mathbf{H}$ (8, 41); these, however, are insignificant in the case of diamagnetic substances, and we shall refrain from considering them here.

The preceding, purely phenomenological considerations are valid for diamagnetic (and not excessively condensed) substances, and have to be extended if the microsystems constituting the medium possess permanent magnetic moments, in Langevin's classical meaning (46), undergoing orientation in the applied magnetic field. The potential energy, which in this case is that of a paramagnetic material immersed in a magnetic field, is, in a linear approximation,

$$W(\tau, \mathbf{H}) = -M^{0}_{m\sigma}H_{\sigma}, \qquad [28]$$

whence the distribution function [18] assumes (in the same approximation) the form:

$$f(\tau, \mathbf{H}) = f(\tau, 0)(1 + \beta M^{0}_{m\sigma}H_{\sigma}), \qquad [29]$$

$\mathbf{M}_m{}^0$ denoting the permanent magnetic dipole moment of the medium in the absence of externally applied fields.

On the other hand, the electric dipole moment induced in the medium by the oscillating electric vector $\mathbf{E}^{\omega}$ in the presence of a magnetic field $\mathbf{H}$ is:

$$M^{\omega}_{e\sigma} = (A^{e\omega}_{\sigma\tau} + B^{e\omega m}_{\sigma\tau\nu}H_{\nu})E^{\omega}_{\tau}, \qquad [30]$$

where $A^{e\omega}_{\sigma\tau}$ is the tensor of linear electric polarizability of the medium at frequency ω, and its linear variation induced by the magnetic field is defined by the polarizability pseudotensor $B^{e\omega m}_{\sigma\tau\nu}$.

On inserting the polarization vector

component [16], calculated by means of [29] and [30], into Eq. [14], we finally get (see Appendix A):

$$\epsilon^{\omega}_{\sigma\tau} - \epsilon_0{}^{\omega}\delta_{\sigma\tau} = \frac{n\lambda}{\pi i} V_\lambda \epsilon_{\sigma\tau\nu} H_\nu , \qquad [31]$$

where the Faraday constant is now of the following general form derived by statistical thermodynamics:

$$V_\lambda = \frac{2\pi^2 i}{3n\lambda V}\left(B^{e\omega m}_{\alpha\beta\gamma} + \beta A^{e\omega}_{\alpha\beta} M^0_{m\gamma}\right)\epsilon_{\alpha\beta\gamma} . \qquad [32]$$

This expression is correct for isotropic media of arbitrary structure and degree of condensation. Particularized for a medium of mutually noninteracting microsystems, it takes a form analogous to the well-known result of Born (2):

$$V_\lambda = \frac{2\pi^2 i\rho}{3n\lambda}\left(b^{e\omega m}_{\alpha\beta\gamma} + \beta a^{e\omega}_{\alpha\beta}\mu^m_\gamma\right)\epsilon_{\alpha\beta\gamma} , \qquad [33]$$

$\rho = N/V$ being the number density of microsystems presenting magnetic moments $\mu_\alpha{}^m$ and polarizability tensors $a^{e\omega}_{\alpha\beta}$ and $b^{e\omega m}_{\alpha\beta\gamma}$.

The first terms of Eqs. [32] and [33], which do not depend directly on temperature, define a diamagnetic Faraday effect, consisting in changes in the optical properties of a body or of its microsystems induced directly by an applied magnetic field. The second term in Eq. [33], which depends directly on the temperature by way of β, defines the paramagnetic part of Faraday's constant related to reorientation of the magnetic dipoles in the applied magnetic field (46).

COTTON-MOUTON EFFECT

From the phenomenological viewpoint, the Cotton-Mouton effect stems from the nonlinear electric polarization induced by the square of an applied d-c magnetic field; this polarization, when inserted into Eq. [14], leads to the following variation of the electric permittivity tensor:

$$\epsilon^{\omega}_{\sigma\tau} - \epsilon_0{}^{\omega}\delta_{\sigma\tau} = 4\pi\chi^{e\omega m}_{\sigma\tau\nu\rho} H_\nu H_\rho , \qquad [34]$$

where the electromagnetic susceptibility tensor $\chi^{e\omega m}_{\sigma\tau\nu\rho}$ of fourth rank has nonzero components in the case of an isotropic medium also. The phenomenological equation [34], applicable though it be to arbitrary bodies, fails to permit any statement whatsoever as to the microscopic mechanism of the effect. In order to bridge the gap, we have to develop a statistical theory, as has already been done with regard to the Faraday effect.

To begin with, let us restrict ourselves to the case when no magnetic moment exists in the absence of externally applied fields ($\mathbf{M}_m{}^0 = 0$) and when no electric polarization, linear in the magnetic field strength, as described by [22], occurs. With these restrictions, the electric dipole moment induced in the medium can be written as follows:

$$M^{\omega}_{e\sigma} = \left(A^{e\omega}_{\sigma\tau} + \tfrac{1}{2}C^{e\omega m}_{\sigma\tau\nu\rho} H_\nu H_\rho + \cdots\right) E^{\omega}_\tau . \qquad [35]$$

Above, a new fourth-rank tensor $C^{e\omega m}_{\sigma\tau\nu\rho}$ appears. It describes the change in electric polarizability of the medium due to the square of the applied magnetic field.

In the case now under consideration the potential energy [28] of the first approximation is equal to zero, and we have to proceed to the second approximation:

$$W(\tau, \mathbf{H}) = -\tfrac{1}{2}A^m_{\sigma\tau} H_\sigma H_\tau , \qquad [36]$$

where $A^m_{\sigma\tau}$ is the tensor of linear magnetic polarizability of the medium. The distribution function [18] is now of the form:

$$f(\tau, \mathbf{H}) = f(\tau, 0)\{1 + \tfrac{1}{2}\beta(A^m_{\sigma\tau} - \langle A^m_{\sigma\tau}\rangle) H_\sigma H_\tau\} . \qquad [37]$$

Returning to Eqs. [35] and [39], we now easily calculate the polarization vector component [16], which on insertion into Eq. [14] leads to the following result for isotropic bodies (see Appendix A):

$$\epsilon^{\omega}_{\sigma\tau} - \epsilon_0{}^{\omega}\delta_{\sigma\tau} = 2n\lambda\{Q^{em}_\lambda \delta_{\sigma\tau} H^2 + C_\lambda(H_\sigma H_\tau - \tfrac{1}{3}\delta_{\sigma\tau} H^2)\} , \qquad [38]$$

wherein the constant

$$Q_\lambda^{em} = \frac{\pi}{3n\lambda}\{\langle C_{\alpha\alpha\beta\beta}^{e\omega m}\rangle + \beta\langle \Delta A_{\alpha\alpha}^{e\omega}\Delta A_{\beta\beta}^{m}\rangle\} \quad [39]$$

defines isotropic variations of the tensor $\epsilon_{\sigma\tau}$ due to the nonlinear polarizability of the medium (first term) and to fluctuations of the linear electric and magnetic polarizabilities (the second term of Eq. [39]—the counterpart of magnetostriction). The constant $C_\lambda^{\,d}$ defines the diamagnetic part of the Cotton-Mouton constant:

$$C_\lambda^{\,d} = \frac{\pi}{15n\lambda V}\{\langle 3C_{\alpha\beta\alpha\beta}^{e\omega m} - C_{\alpha\alpha\beta\beta}^{e\omega m}\rangle + \beta\langle 3A_{\alpha\beta}^{e\omega}A_{\alpha\beta}^{m} - A_{\alpha\alpha}^{e\omega}A_{\beta\beta}^{m}\rangle\}. \quad [40]$$

This formula holds for arbitrarily condensed media. If particularized to the case of noninteracting microsystems, it becomes (2, 11):

$$C_\lambda^{\,d} = \frac{\pi\rho}{15n\lambda}\{3c_{\alpha\beta\alpha\beta}^{e\omega m} - c_{\alpha\alpha\beta\beta}^{e\omega m} + \beta(3a_{\alpha\beta}^{e\omega}a_{\alpha\beta}^{m} - a_{\alpha\alpha}^{e\omega}a_{\beta\beta}^{m})\}. \quad [41]$$

The first, not directly temperature-dependent term is related to the nonlinear change in optical polarizability of the microsystems induced directly by the magnetic field (Voigt's effect). The second, temperature-dependent term arises by orientation of the magnetic polarizability ellipsoids in the magnetic field (Langevin's effect). Since for isotropically polarizable microsystems (11) we have:

$$a_{\alpha\beta}^{e\omega} = a_e^{\,\omega}\delta_{\alpha\beta}\,, \qquad a_{\alpha\beta}^{m} = a_m\delta_{\alpha\beta}\,,$$
$$c_{\alpha\beta\gamma\delta}^{e\omega m} = \eta_\perp^{\omega m}\delta_{\alpha\beta}\delta_{\gamma\delta} + \tfrac{1}{2}(\eta_\parallel^{\omega m} - \eta_\perp^{\omega m})(\delta_{\alpha\gamma}\delta_{\beta\delta} + \delta_{\alpha\delta}\delta_{\beta\gamma}),$$

one notes that [41] reduces directly to Eq. [6], which describes the Voigt effect alone, since now the Langevin molecular orientation term vanishes.

Voigt's effect of nonlinear deformation plays an essential part in atomic substances whereas in substances consisting of molecules beyond absorption regions it plays a part subordinate to that of the second term of Eq. [41], since in such substances the effect of orientation of the anisotropic microsystems predominates. Accordingly, in such cases, we are justified in neglecting the very small Voigt effect (12) and in considering only the Langevin effect, which, in the case of linearly polarizable axially symmetric microsystems, is given by Eq. [7]. The latter formula results obviously from the general equation [41], since at symmetry with respect to the 3-axis given by the unit vector $\mathbf{k}$, the tensor of linear polarizability can be written in the form:

$$a_{\alpha\beta} = a_{11}\delta_{\alpha\beta} + (a_{33} - a_{11})k_\alpha k_\beta\,. \quad [42]$$

Had we included in our calculations (in addition to the diamagnetic energy [36]) a paramagnetic energy [28] as well as a moment linear in $\mathbf{H}$ in the expansion [30], we would have obtained in the general equation [38] (in addition to the diamagnetic part of the Cotton-Mouton constant [40]) a paramagnetic part of the form (15, 35):

$$C_\lambda^{\,p} = \frac{\pi\beta}{15Vn\lambda}\{2\langle 3B_{\alpha\beta\alpha}^{e\omega m}M_{m\beta}^{0} - B_{\alpha\alpha\beta}^{e\omega m}M_{m\beta}^{0}\rangle + \beta\langle 3A_{\alpha\beta}^{e\omega}M_{m\alpha}^{0}M_{m\beta}^{0} - A_{\alpha\alpha}^{e\omega}M_{m\beta}^{0}M_{m\beta}^{0}\rangle\}. \quad [43]$$

Cotton-Mouton constants of the forms [40] and [43] have found application to liquids if account is taken of various molecular correlations (11, 15) and to solutions (15, 35), as well as to macromolecular substances (19), and can readily be applied to suspensions of colloid particles.

In concluding this Section it may be worth noting that, in the case of probe light propagating along the y-axis perpendicularly to the xz-plane containing the magnetic field vector, the general equation [38] yields the following expression for the difference between mutually perpendicular components of the permittivity tensor:

$$\epsilon_{zz}^{\omega} - \epsilon_{xx}^{\omega} = 2n\lambda C_\lambda(H_z^{\,2} - H_x^{\,2}). \quad [44]$$

For a magnetic field acting along the z-axis we get, in approximation, Eq. [2] since $\epsilon^{\omega}_{zz} - \epsilon^{\omega}_{xx} \simeq 2n(n_{\parallel} - n_{\perp})$.

OPTICALLY INDUCED MAGNETIZATION

Before the coming of lasers, it had already been suggested (25) that strong light could nonlinearly affect the magnetic properties of matter. Recently, Pershan *et al.* (36) carried out an experiment in which they succeeded in magnetizing nonabsorbing bodies by means of laser light. In this case, laser light with electric vector $\mathbf{E}^{\omega_L} = \mathbf{E}^0 e^{i\omega_L t}$ oscillating at frequency ω_L induces in the medium a magnetic polarization vector of second rank whose component is:

$$P_{m\sigma} = \chi^{m\omega\omega_L}_{\sigma\tau\nu} E^{\omega_L}_{\tau} E^{-\omega_L}_{\nu}. \qquad [45]$$

Above, the pseudotensor $\chi^{m\omega\omega_L}_{\sigma\tau\nu}$ of third rank describes the nonlinear change in magnetic susceptibility induced by the laser beam of intensity $E^{\omega_L} E^{-\omega_L}$. In the absence of dispersion and absorption, the pseudotensor $\chi^{m\omega\omega_L}_{\sigma\tau\nu}$ is the inverse of the pseudotensor $\chi^{e\omega m}_{\sigma\tau\nu}$ appearing in the theory of Faraday's effect, and in the case of isotropic bodies is given by the expression [24]. We can accordingly bring [45] to the form:

$$4\pi P_{m\sigma} = \frac{n_L \lambda_L}{\pi i} V_{\lambda_L} \epsilon_{\sigma\tau\nu} E^{\omega_L}_{\tau} E^{-\omega_L}_{\nu}, \qquad [46]$$

where V_{λ_L} is a Verdet constant defined by analogy to [27] for the laser wavelength λ_L.

Since in the case of light circularly polarized and propagating along the z-axis the amplitudes of the electric vectors of right and left rotating oscillations are:

$$E_{\pm} = (E_x \pm iE_y)/\sqrt{2}, \qquad [47]$$

Eq. [46] reduces to the form (36):

$$P_{mz} = \frac{n_L \lambda_L}{2\pi^2} V_{\lambda_L} (I_+^{\ L} - I_-^{\ L}), \qquad [48]$$

where $I_{\pm}^{\ L} = E \pm E_{\pm}^{*}/2$ is the intensity of right and left circularly polarized laser light.

The formula [48] defines an inverse Faraday effect [36] consisting in the induction, in a nonabsorbing medium, of magnetic polarization by circularly polarized light.

We shall now show that intense light can induce magnetic anisotropy in a medium. This new effect originates in the circumstance that the electric vector $\mathbf{E}^{\omega_L}$ of the laser beam not only causes magnetic polarization of the second order as given by formula [45] but moreover modifies nonlinearly the magnetic polarization $\mathbf{P}_m^{\omega}$ induced by the probe light's magnetic vector $\mathbf{H}^{\omega}$. We are thus dealing here with a mixed polarization of third order:

$$P^{\omega}_{m\sigma} = \chi^{m\omega\omega_L}_{\sigma\tau\nu\rho} H^{\omega}_{\tau} E^{\omega_L}_{\nu} E^{-\omega_L}_{\rho}, \qquad [49]$$

wherein, in the absence of electron dispersion and absorption as well as dipole reorientation, the fourth-rank tensor $\chi^{m\omega\omega_L}_{\sigma\tau\nu\rho}$ possesses properties analogous to those of the tensor $\chi^{e\omega m}_{\sigma\tau\nu\rho}$ of Eq. [34] describing the Cotton-Mouton effect. By inserting [49] into Eq. [20] we obtain the following expression for the variation induced in the magnetic permittivity tensor by intense laser light:

$$\mu^{\omega}_{\sigma\tau} - \mu_0{}^{\omega}\delta_{\sigma\tau} = 4\pi\chi^{m\omega\omega_L}_{\sigma\tau\nu\rho} E^{\omega_L}_{\nu} E^{-\omega_L}_{\rho}. \qquad [50]$$

This expression is valid both for static magnetic permittivity ($\omega = 0$) and for magnetic permittivity dependent on the frequency ω; as will be shown further on, it describes an effect which is the inverse of the Cotton-Mouton effect and in which a body changes its magnetic properties under the influence of intense light. In particular, for isotropic bodies, Eq. [50] becomes:

$$\mu^{\omega}_{\sigma\tau} - \mu_0{}^{\omega}\delta_{\sigma\tau} = a\delta_{\sigma\tau} E^{\omega_L}_{\nu} E^{-\omega_L}_{\nu} + bE^{\omega_L}_{\sigma} E^{-\omega_L}_{\tau} + cE^{\omega_L}_{\tau} E^{-\omega_L}_{\sigma}, \qquad [51]$$

where a, b, c are constants involving components of the tensor $\chi^{m\omega\omega_L}_{\alpha\beta\gamma\delta}$.

For the case of laser light propagating along the z-axis, Eq. [51] leads to the following expressions for the differences between the diagonal and the nondiagonal components of the magnetic permittivity

tensor:

$$\mu_{xx}^{\omega} - \mu_{yy}^{\omega} = 2(b + c)(I_{xx}^{L} - I_{yy}^{L}); \quad [52]$$

$$\mu_{xy}^{\omega} - \mu_{yx}^{\omega} = 2i(b - c)(I_{+}^{L} - I_{-}^{L}). \quad [53]$$

One thus sees that magnetic anisotropy of the kind given by Eq. [52] can be induced only by light that is linearly polarized in the direction of the x- or y-axis, since in the case of natural light $I_{xx}^{L} = I_{yy}^{L} = I/2$ and the anisotropy $\mu_{xx}^{\omega} - \mu_{yy}^{\omega}$ vanishes. On the other hand, circularly polarized light can induce magnetic anisotropy according to Eq. [53] only in a medium with dispersion and absorption, since in an optically transparent and magnetically lossless medium we have:

$$b = c = \frac{2\pi}{15}(3\chi_{\alpha\beta\alpha\beta}^{m\omega\omega_L} - \chi_{\alpha\alpha\beta\beta}^{m\omega\omega_L}). \quad [54]$$

We now proceed to a statistical-microscopic interpretation of the results in the special case of mutually noninteracting microsystems; however, we assume that these microsystems (molecules, particles) interact with the electric field of laser light according to the following formula of their potential energy in the field:

$$W(\tau, \mathbf{E}_L) = -\tfrac{1}{4}a_{\sigma\tau}^{\omega_L}E_{\sigma}^{\omega_L}E_{\tau}^{-\omega_L}, \quad [55]$$

where $a_{\sigma\tau}^{\omega_L}$ is the linear electric polarizability tensor at frequency ω_L.

On inserting this energy into the distribution function [18] we obtain, in an approximation sufficient for our aims,

$$f(\tau, \mathbf{E}_L) = f(\tau, 0)\left\{1 + \frac{\beta}{4}(a_{\sigma\tau}^{\omega_L} - \langle a_{\sigma\tau}^{\omega_L}\rangle)E_{\sigma}^{\omega_L}E_{\tau}^{-\omega_L}\right\}. \quad [56]$$

Assuming moreover that the tensor of magnetic polarizability of a microsystem $a_{\sigma\tau}^{m}$ undergoes no nonlinear optical variation, we get with regard to Eqs. [20] and [21] for diamagnetic substances:

$$\mu_{\sigma\tau} - \delta_{\sigma\tau} = 4\pi\rho\int a_{\sigma\tau}^{m} f(\tau, \mathbf{E}_L)\, d\tau. \quad [57]$$

On substituting herein the distribution function [56] and on performing an averaging procedure one comes to an equation of the form of Eq. [51], where the constants a, b, and c in the absence of dispersion take the form:

$$2b = 2c = -3a = n_L\lambda_L C_{\lambda_L}^{RO}, \quad [58]$$

where

$$C_{\lambda_L}^{RO} = \frac{\pi\rho\beta}{15n_L\lambda_L}(3a_{\alpha\beta}^{m}a_{\alpha\beta}^{\omega_L} - a_{\alpha\alpha}^{m}a_{\beta\beta}^{\omega_L}) \quad [59]$$

is the counterpart of the Cotton-Mouton constant [41], arising by reorientation of the microsystems in the laser field of frequency ω_L.

Recurring to the relations [58], we can now write the magnetic anisotropy [52] in the form:

$$\mu_{xx} - \mu_{yy} = 2n_L\lambda_L C_{\lambda_L}^{RO}(I_{xx}^{L} - I_{yy}^{L}). \quad [60]$$

This formula, patently, describes an inverse Cotton-Mouton effect.

NONLINEAR FARADAY EFFECT

Let us now consider magnetooptical rotation in the presence of intense laser light —a situation involving in addition to the polarization [22] the following nonlinear contribution:

$$P_{e\sigma}^{\omega} = \chi_{\sigma\tau\nu\rho\lambda}^{e\omega\omega_L m}E_{\tau}^{\omega}E_{\nu}^{\omega_L}E_{\rho}^{-\omega_L}H_{\lambda} \quad [61]$$

leading to the following variation of the electric permittivity tensor:

$$\epsilon_{\sigma\tau}^{\omega} - \epsilon_0^{\omega}\delta_{\sigma\tau} = 4\pi\{\chi_{\sigma\tau\lambda}^{e\omega m} + \chi_{\sigma\tau\nu\rho\lambda}^{e\omega\omega_L m}E_{\nu}^{\omega_L}E_{\rho}^{-\omega_L}\}H_{\lambda}. \quad [62]$$

One notes that this equation contains (in addition to the earlier term describing the usual, linear Faraday effect) a new term the experimental meaning of which is that of a nonlinear variation in Faraday effect due to intense light. Hence, this term leads us to anticipate a new experiment in which the measured, optical properties of matter vary under the simultaneously exercised influence of a d-c magnetic field $\mathbf{H}$ and the electric field $\mathbf{E}^{\omega_L}$ of laser light.

Let us now consider this nonlinear Faraday effect more closely in a statistical approach, starting from the following relation:

$$\epsilon^{\omega}_{\sigma\tau} - \epsilon^{\omega}_{0}\delta_{\sigma\tau} = 4\pi\rho \int b^{e\omega m}_{\sigma\tau\nu} H_\nu f(\tau, \mathbf{E}_L)\, d\tau, \quad [63]$$

where for the sake of simplicity we have omitted the direct nonlinear optical variation of the pseudotensor $b^{e\omega m}_{\sigma\tau\nu}$.

We now introduce, by analogy to the gyration tensor $g_{\sigma\tau}$ of optical activity theory (2, 4), a tensor $g^{em}_{\sigma\tau}$ defined thus:

$$b^{e\omega m}_{\sigma\tau\rho} = -i\epsilon_{\sigma\tau\nu} g^{em}_{\nu\rho}. \quad [64]$$

Invoking moreover the distribution function [56], we obtain (see Appendix A):

$$\begin{aligned}\epsilon^{\omega}_{\sigma\tau} - \epsilon^{\omega}_{0}\delta_{\sigma\tau} = \frac{n\lambda}{\pi i}\{&V^{(0)}_{\lambda}\,\delta_{\nu\rho} \\ &+ \tfrac{1}{4}\, V^{(2)}_{\lambda}(3E^{\omega_L}_{\nu}E^{-\omega_L}_{\rho} \\ &+ 3E^{\omega_L}_{\rho}E^{-\omega_L}_{\nu} \\ &- 2\delta_{\nu\rho}E^{\omega_L}_{\lambda}E^{-\omega_L}_{\lambda})\}\epsilon_{\sigma\tau\nu}H_\rho,\end{aligned} \quad [65]$$

where

$$V^{(0)}_{\lambda} = \frac{4\pi^2\rho}{3n\lambda} g^{em}_{\alpha\alpha} = \frac{4\pi^2\rho}{n\lambda} g_{em} \quad [66]$$

is the usual Verdet constant (its diamagnetic part) in the absence of the laser beam, whereas

$$V^{(2)}_{\lambda} = \frac{2\pi^2\rho\beta}{45n\lambda}(3g^{em}_{\alpha\beta}a^{\omega_L}_{\alpha\beta} - g^{em}_{\alpha\alpha}a^{\omega_L}_{\beta\beta}) \quad [67]$$

is a Verdet constant from induction by light and is due to the effect of optical reorientation of the microsystems alone.

If the tensors $g^{em}_{\alpha\beta}$ and $a^{\omega_L}_{\alpha\beta}$ are referred to principal axes 1, 2, 3 of the microsystem, Eq. [67] can be rewritten in expanded form:

$$\begin{aligned}V^{(2)}_{\lambda} = \frac{2\pi^2\rho\beta}{45n\lambda}\{&(g^{em}_{11} - g^{em}_{22})(a^{\omega_L}_{11} - a^{\omega_L}_{22}) \\ &+ (g^{em}_{22} - g^{em}_{33})(a^{\omega_L}_{22} - a^{\omega_L}_{33}) \\ &+ (g^{em}_{33} - g^{em}_{11})(a^{\omega_L}_{33} - a^{\omega_L}_{11})\}.\end{aligned} \quad [68]$$

Assuming as previously the light to propagate along the z-axis, in whose direction the d-c magnetic field is applied, one obtains from Eq. [65]:

$$\begin{aligned}\epsilon^{\omega}_{xy} - \epsilon^{\omega}_{yx} = \frac{2n\lambda}{\pi i}\{&V^{(0)}_{\lambda} \\ &+ \tfrac{1}{2}\, V^{(2)}_{\lambda}(3E^{\omega_L}_{z}E^{-\omega_L}_{z} \\ &- E^{\omega_L}_{\sigma}E^{-\omega_L}_{\sigma})\}H_z.\end{aligned} \quad [69]$$

The nonlinear variation in Verdet constant is thus seen to be positive if the electric vector of the laser beam oscillates along the z-axis, and negative if the laser beam propagates along the z-axis and its electric vector has components $E^{\omega_L}_{x}$ and $E^{\omega_L}_{y}$. This statement is expressed by writing:

$$V^{E_L}_{\lambda} - V^{(0)}_{\lambda} = \begin{cases} 2V^{(2)}_{\lambda} I^{L}_{zz}, \\ -V^{(2)}_{\lambda}(I^{L}_{xx} + I^{L}_{yy}). \end{cases} \quad [70]$$

OPTICAL SATURATION

We hitherto assumed that the potential energy [55] of the system in the electric field of the laser beam was small as compared to kT, and recurred to the distribution function in its approximate form given by Eq. [56]. In very strong optical fields, the polarizability ellipsoids of the microsystems can undergo complete alignment, so that optical saturation takes place in the whole volume of the system. We can no longer use the approximate expansion [56] in calculating optical reorientation effects, but have instead to recur to the distribution function [17], which in the present case with regard to [55] takes the form:

$$f(\tau, \mathbf{E}_L) = \frac{\exp\left(\frac{\beta}{4} a^{\omega_L}_{\sigma\tau} E^{\omega_L}_{\sigma} E^{-\omega_L}_{\tau}\right)}{\int \exp\left(\frac{\beta}{4} a^{\omega_L}_{\sigma\tau} E^{\omega_L}_{\sigma} E^{-\omega_L}_{\tau}\right) d\tau}. \quad [71]$$

With the aim of simplifying further calculations, let us assume that the microsystems present the axial symmetry; expression [42] is the one to be used now, reducing the function [71] to:

$$f(\tau, \mathbf{E}_L) = \frac{\exp\{y_{\omega_L}(\mathbf{k}\cdot\mathbf{e})^2\}}{\int \exp\{y_{\omega_L}(\mathbf{k}\cdot\mathbf{e})^2\}\, d\tau}, \quad [72]$$

where

$$y_{\omega_L} = \frac{\beta}{4}(a_{33}^{\omega_L} - a_{11}^{\omega_L})E^{\omega_L}E^{-\omega_L} \quad [73]$$

is a parameter of reorientation of the microsystems in the electric field $\mathbf{E}^{\omega_L}$, the direction of which is defined by the unit vector $\mathbf{e}$.

If one applies Eq. [57] to the case of axially symmetric microsystems, the magnetic anisotropy results in the form

$$\mu_{xx} - \mu_{yy} = 4\pi\rho(a_{33}^m - a_{11}^m) \cdot \int \{(\mathbf{k}\cdot\mathbf{x})^2 - (\mathbf{k}\cdot\mathbf{y})^2\} f(\tau, \mathbf{E}_L)\, d\tau \quad [74]$$

with $\mathbf{x}$, $\mathbf{y}$, $\mathbf{z}$ denoting unit vectors parallel to the axes of laboratory coordinates.

On assuming that the laser light is polarized with electric vector oscillating parallel to the x-axis, and on denoting by ϑ the angle between the latter and the symmetry axis $\mathbf{k}$ of the microsystem, we can write Eq. [74] as follows:

$$\mu_{xx} - \mu_{yy} = 4\pi\rho(a_{33}^m - a_{11}^m)\Phi(y_{\omega_L}), \quad [75]$$

where

$$\Phi(y_{\omega_L}) = \frac{1}{2} \cdot \int_0^\pi (3\cos^2\vartheta - 1) f(\vartheta, E_L) 2\pi \sin\vartheta\, d\vartheta \quad [76]$$

is a function introduced by O'Konski *et al.* (38) in the description of electric saturation of optical birefringence. The distribution function [72] is now of the form:

$$f(\vartheta, E_L) = \frac{\exp(y_{\omega_L}\cos^2\vartheta)}{2\pi\int_0^\pi \exp(y_{\omega_L}\cos^2\vartheta)\sin\vartheta\, d\vartheta}. \quad [77]$$

Keeping in mind, in calculating the latter, that the optical anisotropy of the microsystems can be positive (as is the case of CS_2, where $a_{33} > a_{11}$) or negative (e.g., for C_6H_6, where $a_{33} < a_{11}$), we obtain the function [76] in the form:

$$\Phi(\pm y) = \pm\frac{3}{4}\left\{\frac{1}{\sqrt{|y|}\, I(\pm y)} - \frac{1}{|y|}\right\} - \frac{1}{2}, \quad [78]$$

involving integrals of the form:

$$I(\pm y) = \frac{1}{2}e^{\mp|y|}\int_{-\sqrt{|y|}}^{\sqrt{|y|}} e^{\pm t^2}\, dt, \quad [79]$$

the upper and lower signs referring to positive and negative anisotropy, respectively.

The function [78] has been tabulated and plotted for both positive (37, 38) and negative (47) anisotropy. For $y = 0$ we obviously have $\Phi(0) = 0$, whereas in the limiting case as $y \to \infty$ the orientation function [78] tends to unity in the case of positive anisotropy and to $-\frac{1}{2}$ in that of negative anisotropy.

The procedure to be followed is similar if we wish to calculate the influence of optical saturation on the Faraday effect from Eq. [63] and the relation [64]. Thus, denoting by ϑ the angle between the symmetry axis of the microsystem and the z-axis, parallel to which the magnetic field $\mathbf{H}$ lies and the laser vector $\mathbf{E}^{\omega_L}$ oscillates, one obtains the following nonlinear change in Verdet constant:

$$V_\lambda^{E_L} - V_\lambda^{(0)} = \frac{4\pi^2\rho}{3n\lambda}(2g_{33}^{em} - g_{11}^{em} - g_{22}^{em})\Phi(\pm y_{\omega_L}). \quad [80]$$

A similar result is obtained if one supposes that the laser beam propagates along the z-axis; one has only to replace the reorientation function by $\Phi(\mp y)$.

For not too strong reorientation ($y < 1$), the function [76] can be represented in the form of the series (48):

$$\Phi(y) = \frac{2y}{15} + \frac{4y^2}{315} - \frac{8y^3}{4725} - \frac{16y^4}{31185} + \cdots. \quad [81]$$

One sees that in a first approximation of the expansion [81] the result [80] reduces to the variation [70], with Verdet constant of the form:

$$V_\lambda^{(2)} = \frac{2\pi^2\rho\beta}{45n\lambda}(2g_{33}^{em} - g_{11}^{em} - g_{22}^{em}) \cdot (a_{33}^{\omega_L} - a_{11}^{\omega_L}) \qquad [82]$$

being a particular case of the constant [68], on the assumption that $a_{11} = a_{22} \neq a_{33}$.

MAGNETOOPTICAL SATURATION OF BIREFRINGENCE

Finally, let us consider the case when axially symmetric microsystems undergo a reorientation induced concomitantly by a d-c magnetic field $\mathbf{H}$ and a laser electric field $\mathbf{E}_L$ given by the distribution function

$$f(\tau, \mathbf{E}_L, \mathbf{H}) = \frac{\exp\{y_{\omega_L}(\mathbf{k}\cdot\mathbf{e})^2 + y_m(\mathbf{k}\cdot\mathbf{h})^2\}}{\int \exp\{y_{\omega_L}(\mathbf{k}\cdot\mathbf{e})^2 + y_m(\mathbf{k}\cdot\mathbf{h})^2\}\, d\tau}, \qquad [83]$$

where y_{ω_L} is defined by Eq. [73], whereas

$$y_m = \frac{\beta}{2}(a_{33}^m - a_{11}^m)H^2 \qquad [84]$$

is a reorientation parameter of the microsystem in the magnetic field $\mathbf{H}$, the latter being directed along the unit vector $\mathbf{h}$.

Let the probe light beam propagate parallel to the y-axis. The birefringence is now: (Fig. 1):

$$\epsilon_{zz}^\omega - \epsilon_{xx}^\omega = 4\pi\rho(a_{33}^\omega - a_{11}^\omega) \cdot \int \{(\mathbf{k}\cdot\mathbf{z})^2 - (\mathbf{k}\cdot\mathbf{x})^2\} f(\tau, \mathbf{E}_L, \mathbf{H})\, d\tau. \qquad [85]$$

Insertion of the first-order expansion of the function [83] into Eq. [85] yields the usual Cotton-Mouton effect and the optical Kerr effect (30).

However, if one recurs to the second-order approximation of [83], one obtains (in addition to the birefringence [44]) the following magnetooptical cross birefringence:

$$\epsilon_{zz}^\omega - \epsilon_{xx}^\omega = 2n\lambda C_\lambda^{(2)}(z_\sigma z_\tau - x_\sigma x_\tau)\{3(e_\sigma h_\tau + e_\tau h_\sigma)(\mathbf{e}\cdot\mathbf{h}) - 2(e_\sigma e_\tau + h_\sigma h_\tau)\}H^2 I^L \qquad [86]$$

involving the constant

$$C_\lambda^{(2)} = \frac{2\pi\rho\beta^2}{315n\lambda} \cdot (a_{33}^\omega - a_{11}^\omega)(a_{33}^m - a_{11}^m)(a_{33}^{\omega_L} - a_{11}^{\omega_L}), \qquad [87]$$

the value of which is dependent simultaneously on three polarizability anisotropies, two of them electric for frequencies ω and ω_L, and one magnetic. As to the birefringence of Eq. [86], it depends on the square of the magnetic field strength H and on the intensity $I^L = E^{\omega_L}E^{-\omega_L}/2$ of the strong laser beam.

An analysis of the expression [86] suggests several experimental variants for the

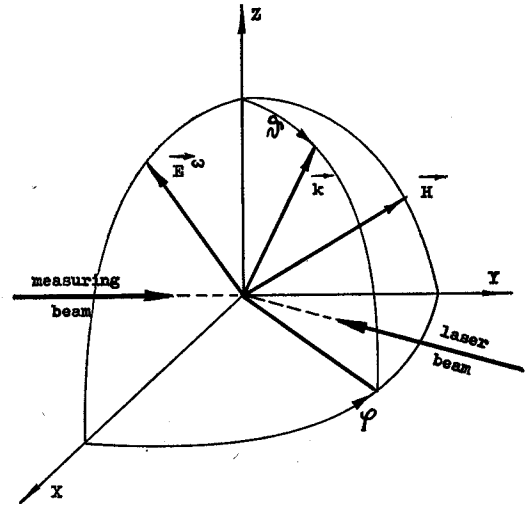

FIG. 1. Diagram showing setup of proposed experiments. The measuring (probe) light beam with electric vector $\mathbf{E}^\omega$ is in all cases chosen to propagate along the y-axis. The propagation direction of the laser beam with electric vector $\mathbf{E}_L$ can be chosen arbitrarily; however, the most advantageous direction of propagation for it would be along the y-axis. The d-c magnetic field $\mathbf{H}$ can be applied along one of the reference axes x, y, or z. The symmetry vector $\mathbf{k}$ of the particle subtends the angle ϑ with the z-axis, its azimuth being φ.

measurement of this magnetooptical cross effect. We shall now consider some of these variants. To begin with, let us consider the case when the direction of propagation of the laser beam is the same as that of the probe light, i.e., parallel to the y-axis. Equation [86] now yields for an unpolarized laser beam:

$$\epsilon_{zz}^{\omega} - \epsilon_{xx}^{\omega} = n\lambda C_{\lambda}^{(2)}(H_z^2 - H_x^2)I^L \quad [88]$$

and for a laser beam which is polarized with oscillations of the electric vector parallel to the z-axis:

$$\epsilon_{zz}^{\omega} - \epsilon_{xx}^{\omega} = 2n\lambda C_{\lambda}^{(2)}(H_z^2 - H_y^2)I_{zz}^L. \quad [89]$$

It is thus apparent that Eq. [88] describes a nonlinear contribution, due specifically to the influence of the strong laser light of intensity I^L, to the Cotton-Mouton effect of Eq. [44]. Obviously, the Cotton-Mouton effect [44] and its nonlinear variation [88] can take place only if the magnetic field **H** is applied along the z-axis or x-axis. However, Eq. [89] shows that a birefringence can also appear in the case when the magnetic field acts along the propagation direction of the two light beams (y-axis), as in Faraday's effect. This "parallel" magnetooptical birefringence is given in general form by the expression:

$$\epsilon_{zz}^{\omega} - \epsilon_{xx}^{\omega} = -2n\lambda C_{\lambda}^{(2)} H_y^2 (I_{zz}^L - I_{xx}^L) \quad [90]$$

and should be accessible to measurement independently of the "perpendicular" Cotton-Mouton effect [44]. Clearly, the birefringence of Eq. [90] can occur only if the electric vector of the laser beam oscillates perpendicularly to the direction in which the magnetic field is applied, e.g., along the x- or z-axis; an unpolarized laser beam is unable to produce such birefringence, since in this case $I_{xx}^L = I_{zz}^L = I^L/2$ and the birefringence [90] vanishes. Equation [86] suggests yet other experiments; however, we refrain from discussing them here.

When reorientation of the microsystems is considerable, one is no longer justified in expanding the distribution function [83]. For a magnetic field **H** and electric vector $\mathbf{E}_L$ acting along the z-axis, we can now write:

$$f(\vartheta, \mathbf{E}_L, \mathbf{H}) = \frac{\exp\{(y_{\omega_L} + y_m)\cos^2\vartheta\}}{2\pi\int_0^{\pi}\exp\{(y_{\omega_L} + y_m)\cos^2\vartheta\}\sin\vartheta\, d\vartheta}, \quad [91]$$

where ϑ is the angle subtended by the axis **k** of the microsystem and the unit vector **z** of laboratory coordinates (Fig. 1). Since in this case $\mathbf{k}\cdot\mathbf{z} = \cos\vartheta$ and $\mathbf{k}\cdot\mathbf{x} = \sin\vartheta\cos\varphi$, Eqs. [85] and [91] yield:

$$\epsilon_{zz}^{\omega} - \epsilon_{xx}^{\omega} = 4\pi\rho(a_{33}^{\omega} - a_{11}^{\omega})\Phi(\pm y_{\omega_L} \pm y_m). \quad [92]$$

This formula defines optical birefringence in the presence of magnetooptical saturation. It reduces to the formula for optical saturation of birefringence (37) if $y_{\omega_L} \neq 0$, $y_m = 0$, and to magnetic saturation of birefringence (18) if $y_{\omega_L} = 0$, $y_m \neq 0$.

DISCUSSION AND CONCLUSIONS

In molecules like CS_2 or C_6H_6, the reorientation parameters [73] and [84] are of the order of $y_{\omega_L} \simeq 10^{-10} E_L^2$ and $y_m \simeq 10^{-15}H^2$ at 300°K. With such low values, it would be necessary to apply a laser beam with electric field E_L of the order of 10^4 esu or a magnetic field H of as much as 3×10^6 oe in order to bring the reorientation function $\Phi(y)$ of Eq. [78] to a value of 10^{-3}, corresponding to a value of $y = 10^{-2}$. Hence, in order to be able to observe in molecular liquids the nonlinear effects anticipated by our calculations, one would have to apply fields of the utmost intensity, thus incurring experimental difficulties (optical breakdown due to laser light).

The situation changes spectacularly for the better if one considers using macromolecular or colloidal substances. Thus, large molecules and colloid particles of about 100 Å present reorientation parameters of the order of $y_{\omega_L} \simeq 10^{-5}E_L^2$ and $y_m \simeq 10^{-10}H^2$, so that fields of less than

$E_L \simeq 10^2$ esu and $H \simeq 10^4$ oe are now sufficient for raising the value of the function $\Phi(y)$ to 10^{-3}. Such fields are readily available in laboratories at present. Optical saturation would thus demand a field E_L of order 10^3 esu, involving the focused beam of a ruby laser with a pulse duration of at least 10^{-6} sec so that the macromolecules can keep pace with the field in the process of reorientation.

For the case of optical saturation, Eqs. [75] and [92] lead to the following expressions:

$$\mu_{xx} - \mu_{yy} = \begin{cases} + 4\pi\rho(a^m_{33} - a^m_{11}), \\ - 2\pi\rho(a^m_{33} - a^m_{11}), \end{cases} \quad [93]$$

$$\epsilon^{\omega}_{zz} - \epsilon^{\omega}_{xx} = \begin{cases} + 4\pi\rho(a^{\omega}_{33} - a^{\omega}_{11}), \\ - 2\pi\rho(a^{\omega}_{33} - a^{\omega}_{11}), \end{cases} \quad [94]$$

permitting direct determinations not only of the optical and magnetic anisotropy of macromolecules and colloid particles but moreover of its sign (in Eqs. [93] and [94], the signs "−" and "+" refer to negative and positive anisotropy, respectively).

From Eq. [80], in the case of optical saturation, investigation of the Verdet constant, which is now of the form:

$$V^{E_L}_{\lambda} - V^{(0)}_{\lambda} = \begin{cases} + \dfrac{4\pi^2\rho}{3n\lambda}(2g^{em}_{33} - g^{em}_{11} - g^{em}_{22}), \\ - \dfrac{2\pi^2\rho}{3n\lambda}(2g^{em}_{33} - g^{em}_{11} - g^{em}_{22}), \end{cases} \quad [95]$$

will permit determinations of the sign and value of the magnetooptical anisotropy induced in the macromolecules or colloid particles.

Colloid systems (e.g., anisaldazine, see reference 18) with particles of the order of 10^3 A and a high magnetic anisotropy amounting to about 10^{-20} are quite easy to obtain, leading to a magnetic reorientation parameter [84] of order $y_m \simeq 10^{-7} H^2$. In such systems, magnetic saturation can be achieved even at magnetic field strengths H of less than 10^4 oe. Hence, the properties given by Eqs. [93]–[95] should prove accessible to measurement in the presence of magnetic saturation, induced by a d-c magnetic field of intensity easily achievable in laboratories with electromagnets. Similarly, the study of the influence of magnetooptical saturation suggests itself with the system acted on simultaneously by a strong d-c magnetic field and the electric field of intense laser light.

From the preceding considerations, it can be said that real possibilities are now available for the standardization of new and simple methods of determining the magnetooptical properties of macromolecules and colloid particles as well as their anisotropy and geometrical shape (see Appendix B). The new nonlinear magnetooptical effects calculated in this paper should also be easily apparent in liquid crystals (49) and other substances the microsystems of which can undergo reorientation.

Historically, the linear magnetooptical effects were first found in colloidal systems. It is only natural that these systems should still be the most appropriate for the detection of the nonlinear effects anticipated here.

The classical theory presented above can be easily extended to multicomponent systems involving various molecular correlations of both the radial and angular kinds (15, 35). Also, one can evolve a coherent, quantum-mechanical theory of these nonlinear magnetooptical effects, as has been done for the case of other, simpler phenomena (40–42, 50). This will provide new information regarding the electronic structure of atomic, molecular, macromolecular, and colloid systems.

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APPENDIX A

UNWEIGHTED AVERAGING METHOD

Tensor components referred to laboratory coordinates with unit vector basis **x**, **y**, **z** will be labeled σ, τ, ν, ρ, $\cdots$; referred to the axes of the molecular coordinate system, with unit vectors **i**, **j**, **k**, they will be labeled α, β, γ, δ, $\cdots$. Let us accordingly transform the tensors $a_{\sigma\tau}$, $b_{\sigma\tau\nu}$, and so forth, from laboratory to molecular coordinates:

$$\begin{aligned} a_{\sigma\tau} &= c_{\sigma\alpha}c_{\tau\beta}a_{\alpha\beta} , \\ b_{\sigma\tau\nu} &= c_{\sigma\alpha}c_{\tau\beta}c_{\nu\gamma}b_{\alpha\beta\gamma} , \cdots , \end{aligned} \qquad [A1]$$

where, if all reference systems are Cartesian, the transformation coefficients $c_{\sigma\alpha}$ have the meaning of cosines of the angles between axes σ and α. The directional cosines $c_{\sigma\alpha}$ and $c_{\tau\beta}$ fulfill the orthonormality relations:

$$c_{\sigma\alpha}c_{\tau\beta}\delta_{\sigma\tau} = \delta_{\alpha\beta} , \qquad c_{\sigma\alpha}c_{\tau\beta}\delta_{\alpha\beta} = \delta_{\sigma\tau} . \qquad [A2]$$

On averaging these products of directional cosines over all possible orientations at equal probability, we get (35):

$$\begin{aligned} \langle c_{\sigma\alpha}c_{\tau\beta}\rangle &= \tfrac{1}{3}\delta_{\alpha\beta}\delta_{\sigma\tau} , \\ \langle c_{\sigma\alpha}c_{\tau\beta}c_{\nu\gamma}\rangle &= \tfrac{1}{6}\epsilon_{\alpha\beta\gamma}\epsilon_{\sigma\tau\nu} , \end{aligned}$$

$$\begin{aligned} &\langle c_{\sigma\alpha}c_{\tau\beta}c_{\nu\gamma}c_{\rho\delta}\rangle \\ &= \tfrac{1}{30}\{(4\delta_{\alpha\beta}\delta_{\gamma\delta} - \delta_{\alpha\gamma}\delta_{\beta\delta} - \delta_{\alpha\delta}\delta_{\beta\gamma})\delta_{\sigma\tau}\delta_{\nu\rho} \\ &\quad + (4\delta_{\alpha\gamma}\delta_{\beta\delta} - \delta_{\alpha\delta}\delta_{\beta\gamma} - \delta_{\alpha\beta}\delta_{\gamma\delta})\delta_{\sigma\nu}\delta_{\tau\rho} \\ &\quad + (4\delta_{\alpha\delta}\delta_{\beta\gamma} - \delta_{\alpha\beta}\delta_{\gamma\delta} - \delta_{\alpha\gamma}\delta_{\beta\delta})\delta_{\sigma\rho}\delta_{\tau\nu}\} . \end{aligned} \qquad [A3]$$

As an example, let us calculate the nonlinear variation of the Verdet constant as given by the following equation:

$$\begin{aligned} \epsilon^{\omega}_{\sigma\tau} - \epsilon_0{}^{\omega}\delta_{\sigma\tau} &= -4\pi\rho i\epsilon_{\sigma\tau\nu} \\ &\quad \cdot \int g^{em}_{\nu\rho}H_{\rho}f(\boldsymbol{\tau}, \mathbf{E}_L)\, d\boldsymbol{\tau}, \end{aligned} \qquad [A4]$$

which results from Eq. [63] on insertion of [64]. Returning to the distribution function in the form [56], we obtain:

$$\begin{aligned} \epsilon^{\omega}_{\sigma\tau} - \epsilon_0{}^{\omega}\delta_{\sigma\tau} &= -4\pi\rho i\epsilon_{\sigma\tau\nu}H_{\rho}\{\langle g^{em}_{\nu\rho}\rangle \\ &\quad + \frac{\beta}{4}(\langle g^{em}_{\nu\rho}a^{\omega L}_{\lambda\mu}\rangle - \langle g^{em}_{\nu\rho}\rangle\langle a^{\omega L}_{\lambda\mu}\rangle)E^{\omega L}_{\lambda}E^{-\omega L}_{\mu}\}, \end{aligned} \qquad [A5]$$

or, using the transformation formulas [A1]:

$$\begin{aligned} \epsilon^{\omega}_{\sigma\tau} - \epsilon_0{}^{\omega}\delta_{\sigma\tau} &= -4\pi\rho i\epsilon_{\sigma\tau\nu}H_{\rho}\{g^{em}_{\alpha\beta}\langle c_{\nu\alpha}c_{\rho\beta}\rangle \\ &\quad + \frac{\beta}{4}g^{em}_{\alpha\beta}a^{\omega L}_{\gamma\delta}(\langle c_{\nu\alpha}c_{\rho\beta}c_{\lambda\gamma}c_{\mu\delta}\rangle \\ &\quad - \langle c_{\nu\alpha}c_{\rho\beta}\rangle\langle c_{\lambda\gamma}c_{\mu\delta}\rangle)E^{\omega L}_{\lambda}E^{-\omega L}_{\mu}\}. \end{aligned} \qquad [A6]$$

On inserting herein the mean values [A3] of the directional cosines, we find:

$$\begin{aligned} \epsilon^{\omega}_{\sigma\tau} - \epsilon_0{}^{\omega}\delta_{\sigma\tau} &= -\frac{4\pi}{3}\rho i\epsilon_{\sigma\tau\nu}H_{\rho}\Big\{g^{em}_{\alpha\alpha}\delta_{\nu\rho} \\ &\quad - \frac{\beta}{120}(3g^{em}_{\alpha\beta}a^{\omega L}_{\alpha\beta} + 3g^{em}_{\alpha\beta}a^{\omega L}_{\beta\alpha} \\ &\quad - 2g^{em}_{\alpha\alpha}a^{\omega L}_{\beta\beta})\delta_{\nu\rho}E^{\omega L}_{\lambda}E^{-\omega L}_{\lambda} \\ &\quad + \frac{\beta}{40}(4g^{em}_{\alpha\beta}a^{\omega L}_{\alpha\beta} - g^{em}_{\alpha\beta}a^{\omega L}_{\beta\alpha} \\ &\quad - g^{em}_{\alpha\alpha}a^{\omega L}_{\beta\beta})E^{\omega L}_{\nu}E^{-\omega L}_{\rho} \\ &\quad + \frac{\beta}{40}(4g^{em}_{\alpha\beta}a^{\omega L}_{\beta\alpha} - g^{em}_{\alpha\beta}a^{\omega L}_{\alpha\beta} \\ &\quad - g^{em}_{\alpha\alpha}a^{\omega L}_{\beta\beta})E^{\omega L}_{\rho}E^{-\omega L}_{\nu}\Big\} . \end{aligned} \qquad [A7]$$

If, above, one assumes the tensor $a^{\omega L}_{\alpha\beta}$ to be symmetrical as is the case in the absence of dispersion and absorption for optically nonactive molecules, the formula [65] results immediately. The other expressions, [38] and [59], are similarly derived.

APPENDIX B

POLARIZABILITY OF THE ANISOTROPY ELLIPSOID

Let us consider a medium of electric permittivity ϵ^e within which an externally applied uniform electric field $\mathbf{E}^e$ extends. We consider an ellipsoid, of electric permittivity ϵ^i, immersed in the medium. The electric field $\mathbf{E}^i$ existing within this ellipsoid is related to the field $\mathbf{E}^e$ by the following

equation, which results by electrostatics of anisotropic bodies (51, 52):

$$\epsilon^e_{\sigma\tau} E_\tau{}^e = \{(\delta_{\sigma\nu} - L_{\sigma\nu})\epsilon^e_{\nu\tau} + L_{\sigma\nu}\epsilon^i_{\nu\tau}\} E_\tau{}^i. \quad [B1]$$

The symmetric tensor $L_{\sigma\tau} = L_{\tau\sigma}$ describes the geometrical shape of the ellipsoid and has the following properties: its trace equals unity,

$$L_{\sigma\tau}\delta_{\sigma\tau} = L_{\sigma\sigma} = L_{xx} + L_{yy} + L_{zz} = 1, \quad [B2]$$

and its principal values are given in the well-known manner:

$$L_\sigma = \tfrac{1}{2} r_x r_y r_z \int_0^\infty \frac{dS}{(r_\sigma{}^2 + S)R_S}, \quad [B3]$$

where

$$R_S{}^2 = (r_x{}^2 + S)(r_y{}^2 + S)(r_z{}^2 + S),$$

with r_x, r_y, r_z denoting the lengths of the semiaxes of the ellipsoid.

In the particular case of a sphere ($r_x = r_y = r_z = r$), the shape tensor becomes isotropic:

$$L_{\sigma\tau} = \tfrac{1}{3}\delta_{\sigma\tau} \quad [B4]$$

and [B1] reduces to the simpler form:

$$3\epsilon^e_{\sigma\tau} E_\tau{}^e = (\epsilon^i_{\sigma\tau} + 2\epsilon^e_{\sigma\tau}) E_\tau{}^i. \quad [B5]$$

If, on the other hand, the permittivity of the medium in which the ellipsoid is immersed is isotropic, i.e, if

$$\epsilon^e_{\sigma\tau} = \epsilon_e \delta_{\sigma\tau}, \quad [B6]$$

Eqs. [B1] and [B2] become for an ellipsoid:

$$\epsilon_e E_\sigma{}^e = \{(\delta_{\sigma\tau} - L_{\sigma\tau})\epsilon_e + L_{\sigma\nu}\epsilon^i_{\nu\tau}\} E_\tau{}^i \quad [B7]$$

and for a sphere:

$$3\epsilon_e E_\sigma{}^e = (\epsilon^i_{\sigma\tau} + 2\epsilon_e\delta_{\sigma\tau}) E_\tau{}^i. \quad [B8]$$

By referring the tensor components $\epsilon_{\sigma\tau}$ and $L_{\sigma\tau}$ to the principal axes, we derive relations for the electric field within the ellipsoid:

$$E_\sigma{}^i = \frac{\epsilon_e E_\sigma{}^e}{\epsilon_e + (\epsilon_\sigma{}^i - \epsilon_e)L_\sigma}, \quad [B9]$$

or the sphere:

$$E_\sigma{}^i = \frac{3\epsilon_e}{\epsilon_\sigma{}^i + 2\epsilon_e} E_\sigma{}^e. \quad [B10]$$

For the case of isotropic bodies, these relations lead to certain well-known expressions (51, 52).

For a cylinder about the x-axis, we have $L_{xx} = 0$ and $L_{yy} = L_{zz} = \frac{1}{2}$. In the case of a circular disc, $L_{xx} = L_{yy} = 0$ and $L_{zz} = 1$.

By Eq. [14], one gets for an anisotropic ellipsoid of volume v immersed in an isotropic medium of electric permittivity ϵ^e:

$$(\epsilon^i_{\sigma\tau} - \epsilon_e\delta_{\sigma\tau}) E_\tau{}^i = \frac{4\pi}{v} a_{\sigma\tau} E_\tau{}^e. \quad [B11]$$

With regard to the relation [B7], we hence derive the tensor $a_{\sigma\tau}$ of electric polarizability of the ellipsoid; in particular, its principal values are given by the well-known expression (17):

$$a_\sigma = \frac{v(\epsilon_\sigma{}^i - \epsilon_e)\epsilon_e}{4\pi[\epsilon_e + (\epsilon_\sigma{}^i - \epsilon_e)L_\sigma]}, \quad [B12]$$

Similarly, expressions are obtained for the tensor of magnetic polarizability of the anisotropic ellipsoid under consideration. The present discussion can be extended to the case of dispersive and absorbing media.

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