BULLETIN DE LA SOCIÉTÉ DES AMIS
DES SCIENCES ET DES LETTRES DE POZNAŃ

Série B — Livraison XXI — 1968/69

Nadbitka

ON THE INFLUENCE OF STRONG LASER LIGHT ON FARADAY'S EFFECT

by

S. KIELICH

(*Présenté à la séance du 19 décembre 1968*)

Summary. A semi-macroscopic theory of Faraday's effect in the presence of an intense laser beam is proposed. Relevant molecular mechanisms are discussed, including the rôle of molecular symmetry and mutual correlations. In molecular gases and liquids, the light intensity-dependent variations in Faraday constant are slight, though accessible to observation if using the focussed beam of a giant ruby laser. In solutions of macromolecules or colloidal systems, the variations are by several orders of magnitude larger, and can be observed when using non-focussed laser beams.

1. INTRODUCTION

Born [1] proposed a formal molecular theory of magneto-optical phenomena in gases, on the assumption that the optical polarizability tensor of the molecules $a^e_{\alpha\beta}$ quite generally undergoes the following variation when they are acted on by a *DC* magnetic field:

$$a^e_{\alpha\beta}(\mathbf{H}) = a^e_{\alpha\beta}(0) + b^{em}_{\alpha\beta\gamma} H_\gamma + \frac{1}{2} c^{em}_{\alpha\beta\gamma\delta} H_\gamma H_\delta + \ldots \tag{1}$$

In this expansion series, the pseudotensor $b^{em}_{\alpha\beta\gamma}$ characterizes the linear change in electric polarizability tensor due to the magnetic field observable in experiments on the usual Faraday effect [1], whereas the tensor $c^{em}_{\alpha\beta\gamma\delta}$ defines the nonlinear change in polarizability of the molecule caused by the square of a strong magnetic field as apparent in the Cotton-Mouton effect [1, 2].

At present, with regard to existing laser techniques, we have the possibility of studying a large variety of nonlinear optical phenomena [3, 4], due i. a. to the circumstance that light of high intensity $I = \mathbf{E}\cdot\mathbf{E}/2$ causes yet other changes in the polarizability tensor (1) according to the expansion

$$a^e_{\alpha\beta}(\mathbf{E}, \mathbf{H}) = \frac{1}{4} c^{ee}_{\alpha\beta\gamma\delta} E_\gamma E^*_\delta + \frac{1}{4} d^{em}_{\alpha\beta\gamma\delta\varepsilon} E_\gamma E^*_\delta H_\varepsilon, \tag{2}$$

where the tensor $c^{ee}_{\alpha\beta\gamma\delta}$ defines the changes in polarizability caused by an intense optical field and investigated recently in work on light intensity-dependent refractive index [5].

The pseudotensor $d^{em}_{\alpha\beta\gamma\delta\varepsilon}$ results by concomitant action on the molecule of an intense laser beam and a *DC* magnetic field, and describes the light intensity-dependent change in magneto-optical rotation [6]:

$$n_L - n_R = \left(\frac{n^2+2}{3n}\right)(F_0 + F_1 I + F_2 I^2 + \ldots)H, \tag{3}$$

where the constants $F_1, F_2, \ldots$ define departure from the usual Faraday constant F_0.

This paper contains an extension of Born's "gas" theory to the case of an arbitrarily dense isotropic medium in a *DC* magnetic field parallel to which, in addition to the probe beam, a laser beam of high intensity propagates. At first, the theory will be formulated in a semi-macroscopic approach. Then, its general results will be particularized for several simple cases, rendering apparent the part played by the symmetry properties of molecules and their mutual correlations in the Faraday effect.

2. GENERAL SEMI-MACROSCOPIC THEORY

Let us consider a dense, homogeneous medium of volume V, isotropic in the absence of externally applied fields and presenting scalar electric (ε) and magnetic (μ) permittivity. Under the influence of an intense external electromagnetic field the medium becomes nonlinear and anisotropic with electric permittivity tensor

$$(\varepsilon_{\sigma\tau} - \delta_{\sigma\tau}) E^M_\tau(\mathbf{r}, t) = 4\pi P_\sigma(\mathbf{r}, t), \tag{4}$$

$\mathbf{P}(\mathbf{r}, t)$ being the electric polarization vector at the space-time point $(\mathbf{r}, t)$.

The electric field $\mathbf{E}(t)$ in the absence of the medium (in vacuo) is related with the mean macroscopic field $\mathbf{E}^M(\mathbf{r}, t)$ existing in a given point of the medium, considered as a macroscopic sphere, as follows:

$$3E_\sigma = (\varepsilon_{\sigma\tau} + 2\delta_{\sigma\tau}) E^M_\tau. \tag{5}$$

For the case of an isotropic body in a weak electric field, we have $\varepsilon_{\sigma\tau} = \varepsilon\delta_{\sigma\tau}$ and Eq. (5) reduces to the well-known form

$$3\mathbf{E} = (\varepsilon + 2)\mathbf{E}^M. \tag{5a}$$

We assume the (probe) field measuring the polarization of the medium to be the optical electric field $\mathbf{E}(\omega_p)$ of an incident light beam oscillating at frequency ω_p. Let us consider the case when the medium is placed in a *DC* uniform magnetic field $\mathbf{H}$ and is also illuminated with an intense laser beam (oscillating at a frequency $\omega_L \neq \omega_p$ and having the intensity $I_L = \mathbf{E}(\omega_L)\cdot\mathbf{E}(-\omega_L)/2$). Since the field $\mathbf{E}(\omega_p)$ inducing polarization in the medium at frequency ω_p is weak, the polarization vector $\mathbf{P}(\omega_p)$ will depend linearly on the field strength of $\mathbf{E}(\omega_p)$ but moreover has to depend on I_L and $\mathbf{H}$ (for the sake of simplicity, the polarization component induced by the magnetic vector of the probe light wave is neglected here):

$$P_\sigma(\omega_p, I_L, H) = \frac{1}{V}\langle A^e_{\sigma\tau}(\omega_p)\rangle_{I_L,H} E_\tau(\omega_p). \tag{6}$$

Above, $A^e_{\sigma\tau}(\omega_p)$ is the polarizability tensor of the medium and can be expanded thus:

$$A^e_{\sigma\tau}(\omega_p, I_L, H) = A^e_{\sigma\tau}(-\omega_p, \omega_p) + B^{em}_{\sigma\tau\nu}(-\omega_p, \omega_p) H_\nu + \\ + \tfrac{1}{4} C^{ee}_{\sigma\tau\nu\rho}(-\omega_p, \omega_p, \omega_L, -\omega_L) E_\nu(\omega_L) E_\rho(-\omega_L) + \\ + \tfrac{1}{4} D^{em}_{\sigma\tau\nu\rho\lambda}(-\omega_p, \omega_p, \omega_L, -\omega_L) E_\nu(\omega_L) E_\rho(-\omega_L) H_\lambda + \ldots \quad (7)$$

with $A^e_{\sigma\tau}(-\omega_p, \omega_p)$ denoting the polarizability tensor of the medium at $I_L = H = 0$, and the pseudotensor $B^{em}_{\sigma\tau\nu}(-\omega_p, \omega_p)$ defining the linear change in polarizability due to the *DC* magnetic field alone. The tensor $C^{ee}_{\sigma\tau\nu\rho}(-\omega_p, \omega_p, \omega_L, -\omega_L)$ describes the nonlinear change in polarizability of the medium caused by the laser beam of intensity $E_\nu(\omega_L)\, E_\rho(-\omega_L)/2$. The pseudotensor $D^{em}_{\sigma\tau\nu\rho\lambda}(-\omega_p, \omega_p, \omega_L, -\omega_L)$ expresses the nonlinear change in polarizability caused jointly by the laser and *DC* magnetic fields.

By classical statistical mechanics, for a system at thermodynamical equilibrium (at temperature T and configuration Γ) in the presence of I_L and H, one can write

$$\langle A^e_{\sigma\tau}(\omega_p)\rangle_{I_L,H} = \frac{\int A^e_{\sigma\tau}(\omega_p, I_L, H) \exp\left\{-\dfrac{U(I_L, H)}{kT}\right\} d\Gamma}{\int \exp\left\{-\dfrac{U(I_L, H)}{kT}\right\} d\Gamma}, \quad (8)$$

where the total potential energy of the system, in the same approximation as that of the expansion (7), is:

$$U(I_L, H) = U(0) - M^m_\sigma H_\sigma - \tfrac{1}{4} A^e_{\sigma\tau}(\omega_L, -\omega_L) E_\sigma(\omega_L) E_\tau(-\omega_L) - \\ - \tfrac{1}{4} B^{em}_{\sigma\tau\nu}(\omega_L, -\omega_L) E_\sigma(\omega_L) E_\tau(-\omega_L) H_\nu - \ldots, \quad (9)$$

$\mathbf{M}^m$ being the magnetic dipole moment of the medium.

Combining Eqs (6) - (9) and on averageing over all possible directions of the fields acting on the medium, we obtain (see Appendix):

$$4\pi P_\sigma(\omega_p, I_L, H) = \{3R_{LL}\delta_{\sigma\tau} + F_0 \varepsilon_{\sigma\tau\nu} H_\nu + \tfrac{1}{2}(D^*_1 \delta_{\sigma\tau} \varepsilon_{\nu\rho\lambda} + \\ + D^*_2 \varepsilon_{\sigma\tau\lambda} \delta_{\nu\rho} + D_3 \varepsilon_{\sigma\nu\lambda} \delta_{\tau\rho} + D_4 \varepsilon_{\sigma\rho\lambda} \delta_{\tau\nu} + D_5 \delta_{\sigma\nu} \varepsilon_{\tau\rho\lambda} + \\ + D_6 \delta_{\sigma\rho} \varepsilon_{\tau\nu\lambda}) E_\nu(\omega_L) E_\rho(-\omega_L) H_\lambda + \ldots\} E_\tau(\omega_p), \quad (10)$$

where

$$R_{LL} = \frac{4\pi}{9V} \langle A^e_{\alpha\alpha}(-\omega_p, \omega_p)\rangle \quad (11)$$

is the well-known Lorentz-Lorenz function, and [7]

$$F_0 = \frac{2\pi}{3V} \langle B^{em}_{\alpha\beta\gamma}(\omega_p, -\omega_p) + \frac{1}{kT} A^e_{\alpha\beta}(-\omega_p, \omega_p) M^m_\gamma \rangle \varepsilon_{\alpha\beta\gamma} \quad (12)$$

is a constant characterizing the Faraday effect in the absence of the intense laser beam. The symbol $\langle\ \rangle$ stands for statistical averageing at zero external fields, and $\varepsilon_{\alpha\beta\gamma}$ is the Levi-Cività extensor.

The remaining constants $D_i (i = 1, \ldots 6)$ in (10) define the change in Faraday effect due to the intense laser light beam, and are of the form:

$$D_1^* = D_1 - \frac{\pi}{9VkT} \langle A^e_{\alpha\beta}(-\omega_p, \omega_p)\rangle \langle B^{em}_{\gamma\delta\varepsilon}(\omega_L, -\omega_L) + \\ + \frac{1}{kT} A^e_{\gamma\delta}(\omega_L, -\omega_L) M^m_\varepsilon\rangle \delta_{\alpha\beta}\varepsilon_{\gamma\delta\varepsilon},$$

$$D_2^* = D_2 - \frac{\pi}{9VkT} \langle B^{em}_{\alpha\beta\varepsilon}(-\omega_p, \omega_p)\rangle \langle A^e_{\gamma\delta}(-\omega_L, \omega_L)\rangle \varepsilon_{\alpha\beta\varepsilon}\delta_{\gamma\delta},$$

$$D_i = \frac{\pi}{15V}\Big\langle D^{em}_{\alpha\beta\gamma\delta\varepsilon}(-\omega_p, \omega_p, \omega_L, -\omega_L) + \frac{1}{kT}[A^e_{\alpha\beta}(-\omega_p, \omega_p)\cdot \\ \cdot B^{em}_{\gamma\delta\varepsilon}(\omega_L, -\omega_L) + B^{em}_{\alpha\beta\varepsilon}(-\omega_p, \omega_p) A^e_{\gamma\delta}(\omega_L, -\omega_L)] + \\ + \frac{1}{kT} C^{ee}_{\alpha\beta\gamma\delta}(-\omega_p, \omega_p, \omega_L, -\omega_L) M^m_\varepsilon + \\ + \frac{1}{k^2T^2} A^e_{\alpha\beta}(-\omega_p, \omega_p) A^e_{\gamma\delta}(\omega_L, -\omega_L) M^m_\varepsilon\Big\rangle \varepsilon^{(i)}_{\alpha\beta\gamma\delta\varepsilon}. \tag{13}$$

The tensors $\varepsilon^{(i)}_{\alpha\beta\gamma\delta\varepsilon}$ are explained in the Appendix.

Assuming the *DC* magnetic field to be applied along the z-axis of the laboratory reference system i. e. in the direction in which the probe light wave propagates, one obtains by Eqs (4), (5) and (10) the following difference between nondiagonal electric permittivity tensor components (putting $\varepsilon_{xx} \simeq \varepsilon_{yy} = n^2$):

$$\varepsilon_{xy} - \varepsilon_{yx} = 2\left(\frac{n^2+2}{3}\right)(F_0 + F_1 I_L + \ldots) H_z, \tag{14}$$

where the Faraday constant F_0 is of the form (12) and its variation under the influence of laser light of intensity $I_L = E_x(\omega_L)\, E_x(-\omega_L)/2 + E_y(\omega_L)\, E_y(-\omega_L)/2$ is given by the constant:

$$F_1 = \tfrac{1}{2}(2D_2^* + D_3 + D_4 - D_5 - D_6). \tag{15}$$

From Eq. (14), the change in magneto-optical rotation is seen to be independent of how the laser light inducing it is polarized.

Similarly, one can calculate magnetic contributions to the tensor $\varepsilon_{\sigma\tau}$, as well as variations of the magnetic permittivity tensor $\mu_{\sigma\tau}$ [8 - 10].

3. APPLICATIONS AND DISCUSSION

We shall now apply the general relations (12) and (15) to a medium consisting of N identical molecules and presenting neither electronic dispersion nor absorption appreciably. Expressing all macroscopic quantities M^m_α, $A^a_{\alpha\beta}$ and so forth by their molecular counterparts m^m_α, $a^e_{\alpha\beta}$, ... :

$$M^m_\alpha = \sum_{p=1}^{N} m^{m(p)}_\alpha, \qquad A^e_{\alpha\beta} = \sum_{p=1}^{N} a^{e(p)}_{\alpha\beta}, \ldots \tag{16}$$

we can rewrite the constants F_0 and F_1 in the form:

$$F_0=\frac{2\pi}{3V}\left\langle \sum_{p=1}^{N} b_{\alpha\beta\gamma}^{em(p)}+\frac{1}{kT}\sum_{p=1}^{N}\sum_{q=1}^{N} a_{\alpha\beta}^{e(p)}\, m_{\gamma}^{m(q)}\right\rangle \varepsilon_{\alpha\beta\gamma}, \tag{17}$$

$$F_1=\frac{\pi}{30V}\left\langle \sum_{p=1}^{N} d_{\alpha\beta\gamma\delta\varepsilon}^{em(p)}+\frac{1}{kT}\sum_{p=1}^{N}\sum_{q=1}^{N}\left(a_{\alpha\beta}^{e(p)}\, b_{\gamma\delta\varepsilon}^{em(q)}+c_{\alpha\beta\gamma\delta}^{ee(p)}\, m_{\varepsilon}^{m(q)}+\right.\right.$$

$$\left.\left.+\, b_{\alpha\beta\varepsilon}^{em(p)}\, a_{\gamma\delta}^{e(q)}\right)+\frac{1}{k^2T^2}\sum_{p=1}^{N}\sum_{q=1}^{N}\sum_{r=1}^{N} a_{\alpha\beta}^{e(p)}\, a_{\gamma\delta}^{e(q)}\, m_{\varepsilon}^{m(r)}\right\rangle \chi_{\alpha\beta\gamma\delta\varepsilon}-$$

$$-\frac{\pi}{9VkT}\left\langle \sum_{p=1}^{N} b_{\alpha\beta\varepsilon}^{em(p)}\right\rangle\left\langle \sum_{q=1}^{N} a_{\gamma\delta}^{e(q)}\right\rangle \varepsilon_{\alpha\beta\varepsilon}\,\delta_{\gamma\delta}, \tag{18}$$

where we have introduced the notation

$$\chi_{\alpha\beta\gamma\delta\varepsilon}=2\varepsilon_{\alpha\beta\varepsilon}\,\delta_{\gamma\delta}+\varepsilon_{\alpha\gamma\varepsilon}\,\delta_{\beta\delta}+\varepsilon_{\alpha\delta\varepsilon}\,\delta_{\beta\gamma}-\delta_{\alpha\gamma}\,\varepsilon_{\beta\delta\varepsilon}-\delta_{\alpha\delta}\,\varepsilon_{\beta\gamma\varepsilon}.$$

3.1. PERFECT GASES

On particularizing the Faraday constant (17) to the case of a gas free of intermolecular interactions one gets Born's result [1]:

$$F_0=\frac{2\pi\rho}{3}\left(b_{\alpha\beta\gamma}^{em}+\frac{1}{kT}\, a_{\alpha\beta}^{e}\,\mu_{\gamma}^{m}\right)\varepsilon_{\alpha\beta\gamma} \tag{19}$$

($\rho=N/V$ being the number density of molecules). Here, the first, temperature-independent term describes the diamagnetic effect consisting in a direct influence of the magnetic field on the optical polarizability of the molecules. The second, temperature-dependent, paramagnetic term results in Langevin's [11] classical approach by orientation of the magnetic dipoles μ_m in the magnetic field.

As seen from Eq. (18), the mechanism of the nonlinear Faraday effect is more complicated than that of the usual Faraday effect given by Eq. (17). Indeed, the constant F_1 contains, beside the diamagnetic effect consisting in nonlinear polarization of the molecule described by the tensor $d_{\alpha\beta\gamma\delta\varepsilon}^{em}$ as caused by the intense laser beam and the *DC* magnetic field, yet other, temperature-dependent effects due to orientation of the polarizability ellipsoids of the molecules in the optical field and of the magnetic dipoles in the magnetic field.

In particular, for diamagnetic isotropically polarizable molecules, we can write:

$$a_{\alpha\beta}^{e}=a_e\,\delta_{\alpha\beta}, \qquad c_{\alpha\beta\gamma\delta}^{ee}=\tfrac{1}{3}c_e(\delta_{\alpha\beta}\,\delta_{\gamma\delta}+\delta_{\alpha\gamma}\,\delta_{\beta\delta}+\delta_{\alpha\delta}\,\delta_{\beta\gamma}),$$

$$b_{\alpha\beta\gamma}^{em}=b_{em}\,\varepsilon_{\alpha\beta\gamma}, \qquad d_{\alpha\beta\gamma\delta\varepsilon}^{em}=d_{em}(\varepsilon_{\alpha\beta\varepsilon}\,\delta_{\gamma\delta}+\varepsilon_{\alpha\beta\delta}\,\delta_{\varepsilon\gamma}+\varepsilon_{\alpha\beta\gamma}\,\delta_{\varepsilon\delta}), \tag{20}$$

whence the constants (17) and (18) reduce to:

$$F_0=4\pi\rho b_{em}, \qquad F_1=2\pi\rho d_{em}. \tag{21}$$

Evaluations of nonlinear electronic polarizabilities b_{em} and d_{em} yield a ratio $F_1/F_0=d_{em}/b_{em}$ of the order of 10^{-14} or at the most 10^{-12}. Consequently, to detect variations in Faraday constant resulting from distortion electronic processes, one would have to

use a laser field of the strength of 10^5 - 10^6 esu, which can only be achieved with the focussed beam of a giant ruby laser.

In the case of a diamagnetic gas in the absence of intermolecular correlations, the constant (18) becomes

$$F_1 = \frac{\pi\rho}{15}\left\{d^{em}_{\alpha\gamma\beta\gamma\delta} + d^{em}_{\alpha\gamma\gamma\beta\delta} + d^{em}_{\alpha\beta\gamma\gamma\delta} + \frac{1}{3kT}(3b^{em}_{\alpha\gamma\delta}a^{e}_{\beta\gamma} + 3b^{em}_{\alpha\gamma\delta}a^{e}_{\gamma\beta} - 2b^{em}_{\alpha\beta\delta}a^{e}_{\gamma\gamma})\right\}\varepsilon_{\alpha\beta\delta}. \tag{22}$$

Hence, the change in Faraday constant in a diamagnetic gas is seen to be due to the nonlinear distortion effect as well as to the effect of molecular reorientation in the laser field. In the case of strongly anisotropic molecules this latter effect predominates so that we can assess the order of magnitude of the ratio F_1/F_0, resulting in $a_e/5kT \simeq 10^{-10}$. This leads to a change in Faraday constant of $10^{-10}\, I_L$ observable if using a laser field with strength of order $10^3 - 10^4$ esu.

3.2. COMPRESSED GASES AND LIQUIDS

In compressed gases and in molecular liquids, the effects under consideration must surely be larger. However, as can be seen from the expressions (17) and (18), numerical evaluations taking into account molecular correlations involve difficulties, and we shall refrain here from adducing a full derivation of the results. Instead, we prefer to draw attention to a fact of interest pointing to an essential rôle of molecular correlations in Faraday's effect. Thus, in dense bodies, even in the absence of external fields, there exist relatively intense electric molecular fields, which contribute supplementarily to polarize the molecules, lowering their symmetry (e. g. by causing a shift of the symmetry centre). Taking this into account, the total magnetic moment of the medium in the absence of external fields can be expressed by the expansion:

$$M^m_\alpha = \sum_{p=1}^{N}\{\mu^{m(p)}_\alpha + a^{me(p)}_{\alpha\beta}F^{(p)}_\beta + \tfrac{1}{2}b^{me(p)}_{\alpha\beta\gamma}F^{(p)}_\beta F^{(p)}_\gamma + \ldots\} \tag{23}$$

where the second term describes the linear magnetic moment induced in the p-th molecule by the field [12]

$$F^{(p)}_\alpha = -\sum_{q=1}^{N}\{T^{(pq)}_{\alpha\beta}\mu^{e(q)}_\beta - \tfrac{1}{3}T^{(pq)}_{\alpha\beta\gamma}\Theta^{(q)}_{\beta\gamma} + \tfrac{1}{15}T^{(pq)}_{\alpha\beta\gamma\delta}\Omega^{(q)}_{\beta\gamma\delta} - \ldots\} \tag{24}$$

of the electric multipoles of neighbouring molecules. The tensors $T^{(pq)}_{\alpha\beta}$, $T^{(pq)}_{\alpha\beta\gamma}$, $T^{(pq)}_{\alpha\beta\gamma\delta}$, ... characterize interaction, successively, between the dipole $\mu^{e(q)}_\alpha$, quadrupole $\Theta^{(q)}_{\alpha\beta}$, octupole $\Omega^{(q)}_{\alpha\beta\gamma}$ of the q-th molecule and the dipole induced in the p-th molecule. Similarly, the third term in Eq. (23) defines the nonlinear magnetic moment induced by the square of the electric molecular field (24).

One sees that, in general, the magnetic moment (23) can be non-zero for diamagnetic molecules having $\mu^m_a = 0$. Extending the procedure, one can write the following expansions in polarizability tensors of higher orders:

$$A^e_{\alpha\beta} = \sum_{p=1}^{N}\{a^{e(p)}_{\alpha\beta} + b^{ee(p)}_{\alpha\beta\gamma}F^{(p)}_\gamma + \tfrac{1}{2}c^{ee(p)}_{\alpha\beta\gamma\delta}F^{(p)}_\gamma F^{(p)}_\delta + \ldots\}, \tag{25}$$

$$B^{em}_{\alpha\beta\gamma} = \sum_{p=1}^{N}\{b^{em(p)}_{\alpha\beta\gamma} + c^{em(p)}_{\alpha\beta\gamma\delta}F^{(p)}_\delta + \tfrac{1}{2}d^{em(p)}_{\alpha\beta\gamma\delta\varepsilon}F^{(p)}_\delta F^{(p)}_\varepsilon + \ldots\}. \tag{26}$$

If now, instead of (16), one substitutes the expansions (23) - (26) into the Faraday constant (12) and carries out statistical averageing with appropriately chosen molecular distribution functions, it becomes apparent that the diamagnetic part, which in gases was temperature-independent, can result in the form of a function of higher powers of the density ρ as well as of the temperature. The second part (the one in T^{-1}), which was zero in diamagnetic gases, can now in general be non-zero owing to molecular redistribution and statistical fluctuations of the molecular electric multipoles.

Not aiming at a full discussion of the constant F_1 with molecular correlations taken into account (which would lead to highly involved results), we nevertheless wish to point to an interesting property of the diamagnetic (temperature-dependent) part, which by (13) and (15) can be put in the general form:

$$F_1^d = \frac{\pi}{45VkT}\{5\langle \Delta B^{em}_{\alpha\beta\delta}\Delta A^{e}_{\gamma\gamma}\rangle + \langle 3B^{em}_{\alpha\gamma\delta}A^{e}_{\beta\gamma} + 3B^{em}_{\alpha\gamma\delta}A^{e}_{\gamma\beta} - 2B^{em}_{\alpha\beta\delta}A^{e}_{\gamma\gamma}\rangle\}\,\varepsilon_{\alpha\beta\delta}\,. \tag{27}$$

We hence note that F_1 consists of a part presenting an isotropic nature (the term with fluctuations of polarizability $\Delta A^{e}_{\alpha\beta}$ and $\Delta B^{em}_{\alpha\beta\gamma}$) and of a part characterizing anisotropic changes in Faraday constant. The occurrence of the isotropic component is due chiefly to statistical fluctuations in the medium (in particular – fluctuations in number density of molecules $\Delta\rho$), as is the case in molecular light scattering or electrostriction in the molecular approach. The second term of Eq. (27) differs from zero in diamagnetic gases only if their molecules are optically anisotropic. In condensed bodies, where the expansions (25) and (26) are valid, the anisotropic part of Eq. (27) can differ from zero even if the molecules are optically isotropic, as a result of translational fluctuations of the induced electric dipoles [5, 13].

Similarly, from the expansions (23) - (25), it can be shown that also the most strongly temperature-dependent part of F_1, namely:

$$F_1^p = \frac{\pi}{15Vk^2T^2}\langle (A^{e}_{\alpha\gamma}A^{e}_{\beta\gamma} + A^{e}_{\alpha\gamma}A^{e}_{\gamma\beta} + A^{e}_{\alpha\beta}A^{e}_{\gamma\gamma})\,M^{m}_{\delta}\,\varepsilon_{\alpha\beta\delta}\rangle \tag{28}$$

can differ from zero in diamagnetics whose molecular electric multipoles interact mutually. In this context, it would be of interest to investigate the Faraday effect in its dependence on the temperature, as such measurements would make apparent the rôles of the various components of the Faraday constants:

$$F_0 = \rho\left(a_0 + \frac{b_0}{T}\right), \tag{29}$$

$$F_1 = \rho\left(a_1 + \frac{b_1}{T} + \frac{c_1}{T^2}\right), \tag{30}$$

where in general the constants $a_0, \ldots, c_1$ can be implicite functions of the density and temperature of the system.

Finally, it would seem worth noting that, since the polarizabilities of macromolecules and colloidal particles are by 2 to 10 orders of magnitude larger than those of molecules in gases (thus, the polarizability of colloid gold particles of diameter 300 Å is of the order

10^{-18} cm^3 at $\lambda = 6500$ Å whereas that of tobacco mosaic virus is of order 10^{-14} cm^3), the variations in Faraday constant in macromolecular and colloidal solutions represent quantities ranging from 10^{-2} I to 10^{-8} I. Such considerable variations ΔF are accessible to observation already if using a non-focussed laser beam with electric field strength 100 esu.

4. CONCLUDING REMARKS

We have proposed a semi-macroscopic theory of non-linear changes in magneto-optical rotation induced in arbitrary isotropic media by an optical laser field of high intensity. These changes are shown to be described in a quadratic approximation by the new Faraday constant F_1, which expresses the molecular structure and thermodynamical state of the medium as well as the changes it undergoes in the presence of the strong optical field. Numerical evaluations of F_1/F_0 show that in some substances the light intensity-dependent changes in magneto-optical rotation should be accessible to observation by appropriately applied, available laser techniques.

On the other hand, a quantum-mechanical treatment [9, 14] of the nonlinear Faraday effect can yield new information on the strongly nonlinear electromagnetic properties of atoms, molecules and macromolecules as well as on the structure of substances. It is hoped that experiments at this Department on intense laser light propagation in media immersed in a strong *DC* magnetic field will help to further clarify various nonlinear optical processes, including self-trapping [3, 4].

The author is indebted to K. Flatau ,M. Sc., for conversations and the English translation of this paper.

Appendix

UNWEIGHTED AVERAGING

Let us transform the tensors M^m_σ, $A^e_{\sigma\tau}$, $B^{em}_{\sigma\tau\upsilon}$.... from the laboratory reference system to another system as follows:

$$M^m_\sigma = h_{\sigma\alpha} M^m_\alpha, \qquad A^e_{\sigma\tau} = e_{\sigma\alpha} e_{\tau\beta} A^e_{\alpha\beta}, \tag{A.1}$$

$$B^{em}_{\sigma\tau\nu} = e_{\sigma\alpha} e_{\tau\beta} h_{\nu\gamma} B^{em}_{\alpha\beta\gamma}, \ldots,$$

where $e_{\sigma\alpha}$ and $h_{\sigma\alpha}$ are transformation coefficients related with the directions of the electric and magnetic vectors **E** and **H**.

On averageing with equal probability over all possible directions of the fields **E** and **H**, we obtain:

$$\langle e_{\sigma\alpha} e_{\tau\beta} \rangle = \tfrac{1}{3} \delta_{\alpha\beta} \delta_{\sigma\tau}, \qquad \langle e_{\sigma\alpha} e_{\tau\beta} h_{\nu\gamma} \rangle = \tfrac{1}{6} \varepsilon_{\alpha\beta\gamma} \varepsilon_{\sigma\tau\nu},$$

$$\langle e_{\sigma\alpha} e_{\tau\beta} e_{\nu\gamma} e_{\rho\delta} h_{\lambda\varepsilon} \rangle = \frac{1}{30} \{ \varepsilon^{(1)}_{\alpha\beta\gamma\delta\varepsilon} \delta_{\sigma\tau} \varepsilon_{\nu\rho\lambda} + \varepsilon^{(2)}_{\alpha\beta\gamma\delta\varepsilon} \varepsilon_{\sigma\tau\lambda} \delta_{\nu\rho} +$$

$$+ \varepsilon^{(3)}_{\alpha\beta\gamma\delta\varepsilon} \varepsilon_{\sigma\nu\lambda} \delta_{\tau\rho} + \varepsilon^{(4)}_{\alpha\beta\gamma\delta\varepsilon} \varepsilon_{\sigma\rho\lambda} \delta_{\tau\nu} + \varepsilon^{(5)}_{\alpha\beta\gamma\delta\varepsilon} \delta_{\sigma\nu} \varepsilon_{\tau\rho\lambda} + \varepsilon^{(6)}_{\alpha\beta\gamma\delta\varepsilon} \delta_{\sigma\rho} \varepsilon_{\tau\nu\lambda} \}, \tag{A.2}$$

with the notation:

$$\varepsilon^{(1)}_{\alpha\beta\gamma\delta\varepsilon} = 3\delta_{\alpha\beta} \varepsilon_{\gamma\delta\varepsilon} + \varepsilon_{\alpha\gamma\varepsilon} \delta_{\beta\delta} - \varepsilon_{\alpha\delta\varepsilon} \delta_{\beta\gamma} - \delta_{\alpha\gamma} \varepsilon_{\beta\delta\varepsilon} + \delta_{\alpha\delta} \varepsilon_{\beta\gamma\varepsilon},$$

$$\varepsilon^{(2)}_{\alpha\beta\gamma\delta\varepsilon} = 3\varepsilon_{\alpha\beta\varepsilon} \delta_{\gamma\delta} - \varepsilon_{\alpha\gamma\varepsilon} \delta_{\beta\delta} - \varepsilon_{\alpha\delta\varepsilon} \delta_{\beta\gamma} + \delta_{\alpha\gamma} \varepsilon_{\beta\delta\varepsilon} + \delta_{\alpha\delta} \varepsilon_{\beta\gamma\varepsilon},$$

$$\varepsilon^{(3)}_{\alpha\beta\gamma\delta\varepsilon} = \delta_{\alpha\beta} \varepsilon_{\gamma\delta\varepsilon} - \varepsilon_{\alpha\beta\varepsilon} \delta_{\gamma\delta} + 3\varepsilon_{\alpha\gamma\varepsilon} \delta_{\beta\delta} - \varepsilon_{\alpha\delta\varepsilon} \delta_{\beta\gamma} - \delta_{\alpha\delta} \varepsilon_{\beta\gamma\varepsilon},$$

$$\varepsilon^{(4)}_{\alpha\beta\gamma\delta\varepsilon} = -\delta_{\alpha\beta} \varepsilon_{\gamma\delta\varepsilon} - \varepsilon_{\alpha\beta\varepsilon} \delta_{\gamma\delta} - \varepsilon_{\alpha\gamma\varepsilon} \delta_{\beta\delta} + 3\varepsilon_{\alpha\delta\varepsilon} \delta_{\beta\gamma} - \delta_{\alpha\gamma} \varepsilon_{\beta\delta\varepsilon},$$

$$\varepsilon^{(5)}_{\alpha\beta\gamma\delta\varepsilon} = -\delta_{\alpha\beta} \varepsilon_{\gamma\delta\varepsilon} + \varepsilon_{\alpha\beta\varepsilon} \delta_{\gamma\delta} - \varepsilon_{\alpha\delta\varepsilon} \delta_{\beta\gamma} + 3\delta_{\alpha\gamma} \varepsilon_{\beta\delta\varepsilon} - \delta_{\alpha\delta} \varepsilon_{\beta\gamma\varepsilon},$$

$$\varepsilon^{(6)}_{\alpha\beta\gamma\delta\varepsilon} = \delta_{\alpha\beta} \varepsilon_{\gamma\delta\varepsilon} + \varepsilon_{\alpha\beta\varepsilon} \delta_{\gamma\delta} - \varepsilon_{\alpha\gamma\varepsilon} \delta_{\beta\delta} - \delta_{\alpha\gamma} \varepsilon_{\beta\delta\varepsilon} + 3\delta_{\alpha\delta} \varepsilon_{\beta\gamma\varepsilon}. \tag{A.3}$$

On expanding the Boltzmann-Maxwell factor in the definition of the statistical average (8) as follows:

$$\exp\left(-\frac{U}{kT}\right)=\exp\left(-\frac{U_0}{kT}\right)\left(1-\frac{U_p}{kT}+\frac{U_p^2}{2k^2T^2}-\ldots\right) \tag{A.4}$$

(with U_p denoting a perturbation to the energy U_0 in the presence of I_L and H) and applying the transformations (A.1) to the expansions (7) and (9), the mean values (A.2) will lead to Eq. (10).

REFERENCES

1. Born M.: Optik (J. Springer, Berlin 1933). — 2. Buckingham A. D., Pople, J. A.: Proc. Phys. Soc., B **69**, 1113 (1956). — 3. Bloembergen N.: American J. Phys. **35**, 989 (1967). — 4. Akhmanov S. A., Sukhorukov, A. P., Khokhlov, R. W.: Uspekhi Fiz. Nauk, **93**, 19 (1967); Akhmanov S. A., Khokhlov R. V.: Uspekhi Fiz. Nauk, **95**, 231 (1968). — 5. Kielich S.: Proc. Phys. Soc., **90**, 847 (1967). — 6. Kielich S.: Physics Letters, **25A**, 517 (1967). — 7. Kielich S., Acta Phys. Polonica, **31**, 929 (1967). — 8. Pershan P. S.: Phys. Rev., **130**, 919 (1963). — 9. Kielich S.: Proc. Phys. Soc., **86**, 709 (1965). — 10. Graham C., Raab R. E.: Proc. Phys. Soc., **90**, 417 (1967). — 11. Langevin P.: J. Physique, **4**, 678 (1905). — 12. Kielich, S.: Molec. Phys. **9**, 549 (1965). — 13. Kirkwood J. G.: J. Chem. Phys., **4**, 592 (1936). — 14. Daudel R., Galais F., Smet P.: Int. Quantum Chem. **1**, 873 (1967).

DOC. DR Hab. STANISŁAW KIELICH
Department of Molecular Physics,
Adam Mickiewicz University, Poznań