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MAJORANA EFFECT IN THE PRESENCE OF AN INTENSE LASER BEAM

by

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Summary. The classical theory of Majorana's effect is extended to the case when a colloidal system is immersed in a strong magnetic field and, beside the measuring light wave, laser light of high intensity is incident on the system. It is shown that, with the intense electric fields of laser beams and the strong pulsed magnetic fields now available in laboratories, measurements of magnetic or optical or magneto-optical saturation have become feasible. In particular, the author suggests that certain new, magneto-optical cross-effects be measured in colloidal systems or polymers in skilfully contrived experiments, since the laser beam, polarized or not, can be applied perpendicularly to the magnetic field lines, or parallel as in the Faraday effect. A study of these effects can be predicted to yield abundant data on the linear and nonlinear electromagnetic properties of colloid particles, as well as on their anisotropy and shape.

1. INTRODUCTION

The optical birefringence induced in gases and liquids by a strong *DC* magnetic field, and generally referred to as the Cotton-Mouton effect, is relatively weak and not easily measurable [1 - 3]. On the other hand, the magneto-optical birefringence of colloidal systems —— Majorana's effect —— is in general 10^6 times larger than in liquids [2], and thus represents a quite considerable effect. Although 65 years have elapsed since Majorana's discovery, the magneto-optics of colloids has hardly been given the attention it deserves. Neither has it been sufficiently widely recurred to for obtaining information on the magnetic properties of colloidal particles, apart from some papers [4, 5]. Recently, advances have been achieved in the magneto-optics of polymers [6 - 8], diamagnetic gases [9], as well as dilute [10] and concentrated solutions of liquids [11, 12].

Laser techniques now available for producing very intense electric fields (with E of order 10^5 V/cm in a non-focussed beam, or 10^9 V/cm in a focussed beam) in conjunction with existing methods of producing strong pulsed magnetic fields with H of order 10^6 Oe suggest new possibilities in the domain of magneto-optical investigations of colloidal systems. Thus, an intense laser beam can induce measurable variations in magneto-optical rotation [13] as well as in magneto-optical birefringence. It is found that these variations can be particularly large in solutions of macromolecules and colloidal systems.

In the present paper, the Majorana effect will be discussed for the case when a colloidal system is acted on by a strong magnetic field and simultaneously by the electric field of an intense laser beam. Especial stress will be laid on magneto-optical cross-effects and on the possibility of their being made apparent in several new, experimental variants.

We shall show e. g. that magneto-optical birefringence can arise when the probe and intense laser beams simultaneously propagate along the magnetic field lines, by analogy to Faraday's effect [1, 2, 14]. Obviously, Majorana's normal effect does not appear in these conditions, since it requires the probe beam to propagate perpendicularly to the magnetic field [2].

With regard to the fact that colloid particles are strongly anisotropic both geometrically and optically [15], the basic mechanism of the above-mentioned effects will obviously consist in the orientation (alignment) of particles in the *DC* magnetic field [2] or in the electric field of the laser beam [16]. In the case when the two fields are very strong, we shall consider an effect of magneto-optical saturation by analogy to magnetic saturation [4] and electric saturation [17]. We shall moreover take into consideration magneto-optical deformation of the particles — an effect hitherto studied only with regard to molecules [1, 9, 11, 14]. However, for the sake of simplicity, we shall abstain here from considering mutual interactions of the particles and their interaction with the surrounding medium.

It may well be worth reminding that orientation of particles in strong optical fields is very considerable, and that this is the reason of nonlinear light scattering [16], optically induced birefringence, as well as self-focussing of laser beams [18]. Indeed, it would be most interesting to proceed to a study of self-focussing and self-trapping of laser beams in colloidal systems placed in a strong *DC* magnetic field, when conditions of observation can be chosen so that the nonlinear changes in refractive index will increase or decrease according to the direction in which the magnetic field is applied. But we shall not discuss these effects here, it being only our wish to indicate their feasibility.

2. MAJORANA EFFECT IN A STRONG DC MAGNETIC FIELD

Consider a medium of scalar refractive index n_0 containing in suspension ellipsoidal particles having anisotropic optical properties given by the principal values n_1, n_2, n_3 of their refractive index. In the absence of solvation and external fields such a medium is optically isotropic, but under the influence of a strong magnetic field $\mathbf{H}$ it becomes optically anisotropic with electric permittivity tensor $\varepsilon_{\sigma\tau}^{\omega}$ when the frequency of the electric field $\mathbf{E}^{\omega}$ of the measuring light beam is ω. In the case of a dilute solution, when the number density ρ of particles is not excessive, the electric permittivity tensor is given by the fundamental formula [14]

$$\varepsilon_{\sigma\tau}^{\omega}-n_0^2\,\delta_{\sigma\tau}=4\pi\rho\int\frac{\partial M_{\sigma}^{\omega}}{\partial E_{\tau}^{\omega}}f(\Omega\,,\mathbf{H})\,d\Omega\,. \tag{1}$$

Assuming for simplicity that the particles are linearly polarizable, one can write for the induced electric dipole moment component

$$M_\sigma^\omega = A_{\sigma\tau}^\omega E_\tau^\omega \tag{2}$$

with $A_{\sigma\tau}^\omega$ – the polarizability tensor at frequency ω. Greek indices σ and τ denote tensor components, and repeated indices in (2) imply, by the Einstein convention, a summation over the three Cartesian components x, y, z of the laboratory reference frame; $\delta_{\sigma\tau}$ is the unit Kronecker tensor.

In the presence of a DC magnetic field the particles tend to align themselves, but this is opposed by the Brownian motion in the solution. By classical Maxwell-Boltzmann statistics, at thermodynamical equilibrium of the system when the temperature is T, the probability for the particle to have its axis in the elementary solid angle $d\Omega$ is determined by the distribution function:

$$f(\Omega, \mathbf{H}) = \frac{\exp\{-\beta U(\Omega, \mathbf{H})\}}{\int \exp\{-\beta U(\Omega, \mathbf{H})\}\, d\Omega}, \tag{3}$$

where the integrations are over all particle directions in $d\Omega$; $\beta = 1/kT$.

The potential energy $U(\Omega, \mathbf{H})$ possessed by a diamagnetic particle of the colloid in the magnetic field is, to within the second power of $\mathbf{H}$:

$$U(\Omega, \mathbf{H}) = -\tfrac{1}{2} A_{\sigma\tau}^m H_\sigma H_\tau, \tag{4}$$

where $A_{\sigma\tau}^m$ is the $\sigma\tau$–component of the diamagnetic polarizability tensor of the particle.

The principal values of $A_{\sigma\tau}^\omega$ are given by [3, 15]

$$A_i^\omega = \frac{v(\varepsilon_i^\omega - n_0^2)\, n_0^2}{4\pi\left[n_0^2 + (\varepsilon_i^\omega - n_0^2) L_i\right]}, \tag{5}$$

where the L_i's $(L_1 + L_2 + L_3 = 1)$ are shape parameters of the particle of volume v. For magnetic principal polarizabilities, one need only replace the electrical parameters in (5) by the corresponding magnetic ones.

We shall now consider the simple and particularly interesting case of particles possessing an axis of symmetry, e. g. along the principal 3-axis defined by a unit vector $\mathbf{k}$. For such particles, we obtain

$$A_{\sigma\tau}^\omega = A_1^\omega \delta_{\sigma\tau} + (A_3 - A_1) k_\sigma k_\tau,$$

and by (2) we obtain from (1)

$$\varepsilon_{zz}^\omega - \varepsilon_{xx}^\omega = 4\pi\rho (A_3^\omega - A_1^\omega) \int (z_\sigma z_\tau - x_\sigma x_\tau) k_\sigma k_\tau f(\Omega, \mathbf{H})\, d\Omega \tag{6}$$

for the difference in refractive indices for oscillations along the z-axis and x-axis of the incident light wave propagating in the direction of the y-axis.

Similarly, the energy (3) can be rewritten as

$$U(\Omega, \mathbf{H}) = -\tfrac{1}{2}\{A_1^m \delta_{\sigma\tau} + (A_3^m - A_1^m) k_\sigma k_\tau\} H_\sigma H_\tau. \tag{7}$$

In the case of a not very high magnetic field, we can expand the right hand side of (3) in a power series in the temperature parameter β, obtaining by (7) from eq. (6) up to the third-order approximation:

$$\varepsilon_{zz}^{\omega}-\varepsilon_{xx}^{\omega}=\frac{2\pi}{15}\,\rho\beta(A_3^{\omega}-A_1^{\omega})(A_3^{m}-A_1^{m})\left\{1+\frac{\beta}{21}(A_3^{m}-A_1^{m})H^2-\right.$$
$$\left.-\frac{\beta^2}{315}(A_3^{m}-A_1^{m})^2H^4-\ldots\right\}(H_z^2-H_x^2). \tag{8}$$

The preceding expression is applicable to the case when $\beta(A_3^m-A_1^m)\,H^2\ll 2$ i. e. for colloidal systems with particles of a size of 100 Å acted on by a magnetic field of considerably less than 10^5 Oe. The first term of (8) describes the well-known Langevin formula [1].

As the optical polarizabilities of Eq. (5) are in general, mathematically, complex quantities, we can resolve the birefringence (8) in the well-known manner into a real part describing Majorana's effect and an imaginary part describing magneto-optical dichroism [2].

3. MAGNETO-OPTICAL CROSS-EFFECTS

We shall now consider the case when the medium immersed in the *DC* magnetic field is moreover acted on by a laser electric field $\mathbf{E}^{\omega_L}$ oscillating at frequency ω_L. This situation gives rise to novel, magneto-optical cross-effects.

The time-averaged potential energy over many cycles of the optical frequency is, in a quadratic approximation [16]:

$$U(\Omega,\mathbf{E}_L)=-\tfrac{1}{4}A_{\sigma\tau}^{\omega_L}E_{\sigma}^{\omega_L}E_{\tau}^{-\omega_L}, \tag{9}$$

or, for axially-symmetric particles:

$$U(\Omega,\mathbf{E}_L)=-\tfrac{1}{4}\{A_1^{\omega_L}\delta_{\sigma\tau}+(A_3^{\omega_L}-A_1^{\omega_L})k_{\sigma}k_{\tau}\}E_{\sigma}^{\omega_L}E_{\tau}^{-\omega_L}. \tag{10}$$

By the expressions (7) and (10), the distribution function is of the form

$$f(\Omega,\mathbf{H},\mathbf{E}_L)=\frac{\exp\{(y_m H_{\sigma}H_{\tau}+y_{\omega_L}I_{\sigma\tau})k_{\sigma}k_{\tau}\}}{\int\exp\{(y_m H_{\sigma}H_{\tau}+y_{\omega_L}I_{\sigma\tau})k_{\sigma}k_{\tau}\}\,d\Omega}, \tag{11}$$

where

$$y_m=\tfrac{1}{2}\beta(A_3^m-A_1^m)\quad\text{and}\quad y_{\omega_L}=\tfrac{1}{2}\beta(A_3^{\omega_L}-A_1^{\omega_L}) \tag{12}$$

are parameters of orientation of the particle in the magnetic and laser fields, respectively, and $I_{\sigma\tau}=E_{\sigma}^{\omega_L}E_{\tau}^{-\omega_L}/2$ is the intensity tensor of the laser light wave.

Inserting (11) in Eq. (6) we obtain for the birefringence due only to the magneto-optical cross-effects (see Appendix):

$$\varepsilon_{zz}^{\omega}-\varepsilon_{xx}^{\omega}=3C_{em}(z_{\sigma}z_{\tau}-x_{\sigma}x_{\tau})(3H_{\sigma}H_{\nu}I_{\nu\tau}+3H_{\tau}H_{\nu}I_{\nu\sigma}-$$
$$-2H_{\nu}H_{\nu}I_{\sigma\tau}-2H_{\sigma}H_{\tau}I_{\nu\nu}), \tag{13}$$

where we have used the notation:

$$C_{em}=\frac{4\pi\rho}{945}\,\beta^2(A_3^{\omega}-A_1^{\omega})\;(A_3^m-A_1^m)(A_3^{\omega_L}-A_1^{\omega_L}). \tag{14}$$

From (13), one obtains various magneto-optical cross-effects. For the present discussion, we select only the more important ones.

In the case when the direction of propagation of the laser beam is the same or opposite to that of the probe light wave (y-axis), Eq. (13) yields

$$\varepsilon_{zz}^{\omega}-\varepsilon_{xx}^{\omega}=6C_{em}\{3(H_z^2 I_{zz}-H_x^2 I_{xx})-H^2(I_{zz}-I_{xx})-(H_z^2-H_x^2)(I_{xx}+I_{zz})\}. \quad (15)$$

For a non-polarized laser beam $I_{xx}=I_{zz}=I_L/2$, and (15) reduces to

$$\varepsilon_{zz}^{\omega}-\varepsilon_{xx}^{\omega}=3C_{em}(H_z^2-H_x^2)I_L, \quad (16)$$

whereas for a polarized beam with oscillations along the z-axis

$$\varepsilon_{zz}^{\omega}-\varepsilon_{xx}^{\omega}=6C_{em}(H_z^2-H_y^2)I_{zz}. \quad (17)$$

In the case when the magnetic field is applied parallel to the propagation of the beams, Eq. (15) yields for the birefringence

$$\varepsilon_{zz}^{\omega}-\varepsilon_{xx}^{\omega}=-6C_{em}H_y^2(I_{zz}-I_{xx}), \quad (18)$$

which vanishes if the laser beam is unpolarized.

Let us now assume the laser beam to propagate in the direction of the x-axis perpendicularly to propagation of the probe light wave; computation from (13) leads to the expressions:

$$\varepsilon_{zz}^{\omega}-\varepsilon_{xx}^{\omega}=6C_{em}H_x^2 I_{yy}, \quad (19)$$

$$\varepsilon_{zz}^{\omega}-\varepsilon_{xx}^{\omega}=6C_{em}H_z^2(I_{zz}-I_{yy}), \quad (20)$$

for magnetic field applied parallel or perpendicularly to the propagation direction of the laser beam, respectively.

The preceding considerations permit to state that various technically feasible experiments still await performing. These are expected to make apparent the effects on a colloidal system of the concomitant action of a *DC* magnetic field and electric field of a laser beam, with the latter propagating perpendicularly to **H** (as in Majorana's effect) or parallel to **H** (as in Faraday's effect).

4. MAGNETO-OPTICAL SATURATION

When immersed in a very strong magnetic field or in the electric field of a laser beam, colloid particles tend to total alignment, leading in well-defined conditions to magneto--optical saturation. In the case now under consideration, we have to ask whether we are still justified in expanding the Boltzmann factor of the distribution function (11) in a series in powers of β, as done previously. For colloid particles of dimensions of order 100 Å, the anisotropy of optical polarizability is of order 10^{-18} whereas the anisotropy of magnetic polarizability is about 10^{-24}, whence the orientational parameters (12) are of the orders $y_m \simeq 10^{-10}$ and $y_{\omega L} \simeq 10^{-5}$ at room temperature. As a consequence, expansion of the function (11) in a power series is justifiable only as long as the magnetic field strength is maintained below 10^5 Oe and the laser field below 10^3 esu. One sees that with the now generally available strong magnetic and laser fields, calculations of the birefringence (6) have to be performed with the full distribution function in the form (11).

However, so as to be able to perform the calculations throughout, let us assume as simplification that the DC magnetic field is imposed along the z-axis and that the laser electric field oscillates also parallel to the z-axis. We now have:

$$x_\sigma k_\sigma = \sin\vartheta\cos\varphi, \quad z_\sigma k_\sigma = \cos\vartheta,$$

where ϑ is the angle between the symmetry axis of the particle and the z-axis of the laboratory reference system. With regard to Eqs (6) and (11), we can now write:

$$\varepsilon_{zz}^{\omega} - \varepsilon_{xx}^{\omega} = 4\pi\rho(A_3^{\omega} - A_1^{\omega})\Phi(\gamma), \tag{21}$$

where we have used the function due to O'Konski et al [17]:

$$\Phi(\gamma) = \tfrac{1}{2}\int_0^{\pi}(3\cos^2\vartheta - 1)f(\vartheta,\gamma)\,2\pi\sin\vartheta\,d\vartheta \tag{22}$$

with

$$f(\vartheta,\gamma) = \frac{\exp(\gamma\cos^2\vartheta)}{\int_0^{\pi}\exp(\gamma\cos^2\vartheta)\,2\pi\sin\vartheta\,d\vartheta}, \tag{23}$$

where, in our case,

$$\gamma = y_m H_z^2 + y_{\omega L} I_{zz}. \tag{24}$$

For not too strong fields, one can expand (23) in a power series, as a result of which the function (22) takes the form

$$\Phi(\gamma) = 2\sum_{n=0}^{\infty} c_n \gamma^n, \tag{25}$$

wherein $c_0 = 0$, and for $n \geqslant 1$ we have [18]:

$$c_n = \frac{n}{n!(2n+1)(2n+3)} - \sum_{k=1}^{n}\frac{c_{n-k}}{k!(2k+1)}. \tag{26}$$

For the case of weak fields ($\gamma \ll 1$), one can represent (25) by the following approximation:

$$\Phi(\gamma) = \frac{2}{15}\gamma + \frac{4}{315}\gamma^2 - \frac{8}{4725}\gamma^3 - \frac{16}{31185}\gamma^4 + \ldots \tag{27}$$

For the case of very strong fields and positive anisotropy one has to recur to the function (22) in the form calculated, tabulated numerically and plotted by O'Konski et al [17]:

$$\Phi(\gamma) = \frac{3}{4}\left\{\frac{e^{\gamma}}{\sqrt{\gamma}\int_0^{\sqrt{\gamma}} e^{t^2}dt} - \frac{1}{\gamma}\right\} - \frac{1}{2}. \tag{28}$$

The limiting form of (28) for very strong fields is [17]:

$$\Phi(\gamma)=1-\frac{3}{2\gamma-1}, \tag{29}$$

and for $\gamma\to\infty$ one has $\Phi\to 1$.

If one puts $\gamma=y_m H^2=10^{-10}\vartheta H^2$ in the function (28), magnetic saturation can appear at a magnetic field strength of $3\cdot 10^5$ Oe, which is accessible in laboratories [12]. For $\gamma=y_{\omega L}I=10^{-5}\ I$, optical saturation can occur already at electric field strengths of 10^3 esu available in non-focussed laser beams. With γ defined by Eq. (24), one has the case of magneto-optical saturation caused by simultaneous action on the colloid of a strong magnetic field and an intense laser field.

Calculations of the function (22) can be extended to cases when the particles possess a magnetic moment, or when they do not present axial symmetry, as done for the case of saturation of electric birefringence in solutions of macromolecules [17, 19].

5. GENERAL THEORY OF MAJORANA EFFECT

In the general case, similarly as for molecules [1, 2], the polarizability tensor of the particles $A^{\omega}_{\sigma\tau}$ undergoes the following variation when they are acted on by the *DC* magnetic field $\mathbf{H}$:

$$A^{\omega}_{\sigma\tau}(\mathbf{H})=A^{\omega}_{\sigma\tau}+B^{\omega m}_{\sigma\tau\nu}H_\nu+\frac{1}{2}C^{\omega m}_{\sigma\tau\nu\rho}H_\nu H_\rho+\dots. \tag{30}$$

Above, the third-rank pseudotensor $B^{\omega m}_{\sigma\tau\nu}$ characterizes the linear change in electric polarizability tensor due to $\mathbf{H}$, whereas the fourth-rank tensor $C^{\omega m}_{\sigma\tau\nu\rho}$ defines the nonlinear change of $A^{\omega}_{\sigma\tau}$ caused by the square of the strong magnetic field.

If the particles have a magnetic dipole moment $\mathbf{M}$, the potential energy (4) has to be replaced by the following expansion [11]:

$$U(\Omega,\mathbf{H})=-M_\sigma H_\sigma-\frac{1}{2}A^{m}_{\sigma\tau}H_\sigma H_\tau-\dots, \tag{31}$$

and we obtain from (1) by (2) with (30) and (3) with (31) the general expression

$$\varepsilon^{\omega}_{\sigma\tau}-n_0^2\delta_{\sigma\tau}=\{3R_{LL}\delta_{\sigma\tau}+F_0\varepsilon_{\sigma\tau\nu}H_\nu+QH^2\delta_{\sigma\tau}+C_M(3H_\sigma H_\tau-H^2\delta_{\sigma\tau})\}, \tag{32}$$

where

$$R_{LL}=\frac{4\pi\rho}{3}A_e \tag{33}$$

is the well-known Lorentz-Lorenz function, with $A_e=(A^{\omega}_{11}+A^{\omega}_{22}+A^{\omega}_{33})/3$ denoting the mean polarizability of the particle. Mathematically, (Eq. 32) recalls a formula derived by this author [14] for molecular liquids.

The term of (32) linear in H determines the well-known Faraday effect with the constant

$$F_0=\frac{2\pi}{3}\rho\left(B^{\omega m}_{\alpha\beta\gamma}+\beta A^{\omega}_{\alpha\beta}M_\gamma\right)\varepsilon_{\alpha\beta\gamma}, \tag{34}$$

where $\varepsilon_{\alpha\beta\gamma}$ is the Levi-Cività extensor with components equal to 1 or $-$ 1 according to whether $\alpha\beta\gamma$ is an even or odd permutation of xyz, and zero if any two indices are the same.

The constant in Eq. (32):

$$Q = \frac{2\pi\rho}{9} C^{\omega m}_{\alpha\alpha\beta\beta} \tag{35}$$

describes isotropic changes in the tensor $\varepsilon^{\omega}_{\sigma\tau}$ due to the square of H, whereas

$$C_M = \frac{\pi\rho}{45}\left(C^{\omega m}_{\alpha\beta\gamma\delta} + 2\beta B^{\omega m}_{\alpha\beta\gamma} M_\delta + \beta A^{\omega}_{\alpha\beta} A^{m}_{\gamma\delta} + \beta^2 A^{\omega}_{\alpha\beta} M_\gamma M_\delta\right)\chi_{\alpha\beta\gamma\delta} \tag{36}$$

accounts for the optical anisotropy induced in the medium by a DC magnetic field, with the notation

$$\chi_{\alpha\beta\gamma\delta} = 3\delta_{\alpha\gamma}\delta_{\beta\delta} + 3\delta_{\alpha\delta}\delta_{\beta\gamma} - 2\delta_{\alpha\beta}\delta_{\gamma\delta}.$$

When light propagates within the medium parallel to the magnetic field lines along the y-axis, Eq. (32) yields

$$n_L - n_R = \frac{\varepsilon^{\omega}_{xz} - \varepsilon^{\omega}_{zx}}{2n} = \frac{F_0}{n} H_y \tag{37}$$

for the difference in refractive indices n_L and n_R for left and right circularly polarized light, respectively.

Assuming the incident light to propagate along the y-axis perpendicularly to the xz-plane containing the DC magnetic field vector, Eq. (32) yields

$$n_z - n_x = \frac{\varepsilon^{\omega}_{zz} - \varepsilon^{\omega}_{xx}}{2n} = \frac{3C_M}{2n}(H_z^2 - H_x^2) \tag{38}$$

for the difference between the refractive indices in the directions of z and x.

The general expressions (34) and (36) reduce easily to the form obtained by Born [1, 11] for the Faraday and Cotton-Mouton constants of atomic and molecular gases. The first two terms of (36) are related with the change in polarizability of the particles due to direct action of the magnetic field; in the case of strongly anisotropic particles, they can be neglected with regard to the other two terms, which result by orientation of the particles in the magnetic field. If moreover the tensors $A^{\omega}_{\alpha\beta}$ and $A^{m}_{\alpha\beta}$ are referred to principal axes, then by (36) we get for diamagnetic linearly polarizable particles:

$$C^d_M = \frac{2\pi\rho}{45kT}\{(A^{\omega}_1 - A^{\omega}_2)(A^m_1 - A^m_2) + (A^{\omega}_2 - A^{\omega}_3)(A^m_2 - A^m_3) + (A^{\omega}_3 - A^{\omega}_1)(A^m_3 - A^m_1)\}. \tag{39}$$

If the particles possess a magnetic moment, Eq. (36) yields the paramagnetic part of the Cotton-Mouton constant as follows:

$$C^p_M = \frac{2\pi\rho}{45k^2T^2}\{(A^{\omega}_1 - A^{\omega}_2)(M_1^2 - M_2^2) + (A^{\omega}_2 - A^{\omega}_3)(M_2^2 - M_3^2) + (A^{\omega}_3 - A^{\omega}_1)(M_3^2 - M_1^2)\}. \tag{40}$$

If in particular one assumes the particles as spherical ($L_1 = L_2 = L_3 = 1/3$) and optically isotropic, all tensors become isotropic [11]:

$$A^{\omega}_{\alpha\beta} = A^{\omega}\delta_{\alpha\beta}, \qquad A^{m}_{\alpha\beta} = A^{m}\delta_{\alpha\beta}, \tag{41}$$

$$C^{\omega m}_{\alpha\beta\gamma\delta} = C^{\omega m}_{\perp}\delta_{\alpha\beta}\,\delta_{\gamma\delta} + \frac{1}{2}(C^{\omega m}_{||} - C^{\omega m}_{\perp})(\delta_{\alpha\gamma}\,\delta_{\beta\delta} + \delta_{\alpha\delta}\,\delta_{\beta\gamma}).$$

$C^{\omega m}_{||}$ and $C^{\omega m}_{\perp}$ are the nonlinear changes in polarizability of the particles in the directions parallel and, respectively, perpendicular to the vector **H**.

Obviously, with regard to Eq. (41), the contributions (39) and (40) arising by orientation of particles vanish, and the only source of birefringence in the case now under consideration resides in the nonlinear distortional effect which, with regard to Eqs (36) and (41), is given by the simple formula:

$$C_M = \frac{2\pi\rho}{3}(C^{\omega m}_{||} - C^{\omega m}_{\perp}) \tag{42}$$

similar to the one valid for a diamagnetic gas consisting of spherical molecules [9].

In a similar manner, a discussion of the general formulas (34) and (36) can be carried out for particles of yet other symmetries. In particular, when considering rotational ellipsoid symmetry, one comes to the results of refs [2] and [3].

The theory thus generally formulated can be extended to comprise the presence of a laser field, but the final results are of a rather complicated form, and we refrain from adducing them here.

6. CONCLUDING REMARKS

High magnetic field techniques now generally available permit to go beyond the quadratic Majorana effect and to detect and measure variations depending on higher powers of the magnetic field as given by Eq. (8). In very strong magnetic fields when $\gamma = \gamma_m H^2$ in the O'Konski function (28), one can investigate saturation of magnetic birefringence, easily achievable in appropriate solutions of macromolecules or systems of colloidal particles. Electro-magnetic saturation can easily be achieved in systems containing large particles. E. g., for particles with a volume of order 10^{-12} cm^3 the magnetic anisotropy is of order 10^{-19} [4], so that magnetic saturation results already at a magnetic field of the order of $3 \cdot 10^3$ Oe.

It is of particular interest to consider the case of Majorana's effect in the presence of intense laser light, rendering observable various magneto-optical cross-effects as given by formulas (13) - (20). Indeed, the ratio of the constant (14) accounting for these cross--effects and the constant (39) accounting for the quadratic Majorana effect, in the case of axially-symmetric particles, amounts to

$$\frac{C_{em}}{C_M} = \frac{\beta}{21}(A^{\omega L}_3 - A^{\omega L}_1) \simeq 10^{-6}.$$

This means that, in colloidal systems, variations in Majorana effect due to the cross-effects are of order $10^{-6}\, I$, thus being accessible to experimental observation recurring to usual laser techniques.

Noteworthy, too, is the case of light propagating as in Faraday's effect along the magnetic field applied in the y-direction; now $H_x = H_z = 0$ and in accordance with the expression (8) the usual Majorana effect does not appear. In this case however, according to Eq. (17), birefringence appears to the amount of

$$\varepsilon_{zz}^{\omega}-\varepsilon_{xx}^{\omega}=-6C_{em}H_y^2 I_{zz} \tag{43}$$

defining a new, magneto-optical cross-effect. In the experimental set-up under consideration, the cross-effect of Eq. (43) occurs simultaneously with laser-induced birefringence [16, 18], compared to which it increases with the magnetic field strength H as $\beta/21\ (A_3^m - A_1^m)\ H^2$. For particles of dimensions of the order of 10^3 Å, this ratio is of the order of $10^{-9}H^2$, whence the effect given by Eq.(43) should be quite easily detectable by means of the strong magnetic fields now generally in use.

These effects, when subjected to experimental investigation, can be predicted to constitute a direct source of data regarding the values and signs of the magnetic anisotropy of axially-symmetric particles. Similarly, formula (42) will permit to gain information on the nonlinear magneto-optical deformation of spherical particles.

In addition to the above-considered magneto-optical effects, one can investigate other, novel effects, such as light intensity-dependent changes in magneto-optical rotation [13]:

$$n_L-n_R=\frac{1}{n}(F_0+F_1 I+F_2 I^2+\ldots)H, \tag{44}$$

where F_1 and F_2 describe departures from the usual Faraday effect given by Eq. (37). Pershan et al [20] recently investigated an inverse Faraday effect, consisting in the induction of magnetic polarization in the medium by circularly polarized light of intensities I_R and I_L:

$$P_m=(G_0+G_1 I+G_2 I^2+\ldots)(I_R-I_L) \tag{45}$$

Also, magnetic anisotropy induced in the medium by intense laser light can be investigated [21]. The effects described by formulas (44) and (45), easily detectable in colloids, are discussed in detail in a separate paper [22].

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Appendix

UNWEIGHTED AVERAGING

As long as the magnetic $\mathbf{H}$ and laser $\mathbf{E}_L$ field strengths are not too large, Boltzmann's factor in the distribution function (11) can be expanded in a power series with the following accuracy:

$$f(\Omega, \mathbf{H}, \mathbf{E}_L) = f(\Omega, 0)\{1 + \beta(k_\sigma k_\tau - \langle k_\sigma k_\tau \rangle)(y_m H_\sigma H_\tau + y_{\omega L} I_{\sigma\tau}) + \tfrac{1}{2}\beta^2 [k_\sigma k_\tau k_\nu k_\rho - \langle k_\sigma k_\tau k_\nu k_\rho \rangle - \langle k_\sigma k_\tau \rangle k_\nu k_\rho - k_\sigma k_\tau \langle k_\nu k_\rho \rangle + 2\langle k_\sigma k_\tau \rangle \langle k_\nu k_\rho \rangle](y_m^2 H_\sigma H_\tau H_\nu H_\rho + 2 y_m y_{\omega L} H_\sigma H_\tau I_{\nu\rho} + y_{\omega L}^2 I_{\sigma\tau} I_{\nu\rho}) + \ldots\}, \quad (A.1)$$

where

$$\langle k_\sigma k_\tau k_\nu \ldots \rangle = \int k_\sigma k_\tau k_\nu \ldots f(\Omega, 0)\, d\Omega \quad (A.2)$$

denotes averageing over all possible orientations of particles with the distribution function $f(\Omega, 0)$ valid at zero external fields, i.e. averageing with equal probability. The definition (A.2) now yields:

$$\langle k_\sigma k_\tau \rangle = \tfrac{1}{3}\delta_{\sigma\tau},$$

$$\langle k_\sigma k_\tau k_\nu k_\rho \rangle = \tfrac{1}{15}(\delta_{\sigma\tau}\delta_{\nu\rho} + \delta_{\sigma\nu}\delta_{\tau\rho} + \delta_{\sigma\rho}\delta_{\tau\nu}),$$

$$\langle k_\sigma k_\tau k_\nu k_\rho k_\lambda k_\mu \rangle = \tfrac{1}{105}\{\delta_{\sigma\tau}(\delta_{\nu\rho}\delta_{\lambda\mu} + \delta_{\nu\lambda}\delta_{\rho\mu} + \delta_{\nu\mu}\delta_{\lambda\rho}) + \delta_{\sigma\nu}(\delta_{\tau\rho}\delta_{\lambda\mu} + \delta_{\tau\lambda}\delta_{\rho\mu} + \delta_{\tau\mu}\delta_{\lambda\rho}) + \delta_{\sigma\rho}(\delta_{\tau\nu}\delta_{\lambda\mu} + \delta_{\tau\lambda}\delta_{\nu\mu} + \delta_{\tau\mu}\delta_{\nu\lambda}) + \delta_{\sigma\lambda}(\delta_{\tau\nu}\delta_{\rho\mu} + \delta_{\tau\rho}\delta_{\nu\mu} + \delta_{\tau\mu}\delta_{\nu\rho}) + \delta_{\sigma\mu}(\delta_{\tau\nu}\delta_{\rho\lambda} + \delta_{\tau\rho}\delta_{\nu\lambda} + \delta_{\tau\lambda}\delta_{\nu\rho})\}. \quad (A.3)$$

On substituting the distribution function (A.1) into Eq. (6) and taking the mean values (A.3), one obtains formulas (8) or (13).

REFERENCES

1. Born M.: Optik (Julius Springer-Verlag, Berlin 1933); Volkenshteyn M. V.: Molekularnaya Optika (Molecular Optics, Gostekhizdat, Moskva 1951). — 2. Beams J. W.: Rev. Mod. Phys., **4**, 133 (1932); Schütz W.: Handbuch der Experimental Physik, Magnetooptik, Vol. 16, Akad. Verlagsges. Leipzig 1936, and references therein. — 3. Peterlin A. and Stuart H. A.: Z. Physik, **112**, 129 (1939). — 4. Tsvetkov V. N. and Sosinskii M.: Zh. Eksperim. i Teor. Fiz., **19**, 543 (1949). — 5. Prémilat S. and Horn P.: C. R. Acad. Sc. Paris, **257**, 3848 (1963); **258**, 6366 (1964); J. Chim. Physique, **62**, 395 (1965). — 6. Tsvetkov V. N.: Zh. Eksperim. i Teor. Fiz. **14**, 35 (1945); Miekshenkov M. J.: Biofizika, **12**, 30 (1967). — 7. Tinoco I. Jr. and Bush C. A.: Biopolymers Symposia **1**, 235 (1964). — 8. Harris R. A.: J. Chem. Phys., **46**, 3398 (1967); **47**, 4481 (1967). — 9. Buckingham A. D., Prichard W. H., and Whiffen D. H.: Trans. Faraday Soc. **63**, 1057 (1967). — 10. Le Fevre R. J. W. and Murthy D. S. N.: Australian J. Chem. **19**, 179, 1321 (1966). — 11. Kielich S.: Acta Phys. Polonica **22**, 299 (1962). — 12. Surma M.: Acta Phys. Polonica **25**, 485 (1964). — 13. Kielich S.: Physics Letters, **25A**, 517 (1967). — 14. Kielich S.: Acta Phys. Polonica, **31**,929 (1967). — 15. Gans R.: Ann. Physik **37**, 881 (1912). — 16. Kielich S.: J. Colloid Interf. Sci., **27**,432 (1968). — 17. O'Konski Ch. T,, Yoshioka K. and Ortung W. H.: J. Phys. Chem. **63**, 1558 (1959); Yoshioka K. and O'Konski C. T.: Biopolymers, **4**, 499 (1966). — 18. Kielich S.: Chem. Phys. Letters **1**, 675 (1968). — 19. Shah M. J.: Phys. Chem. **67**, 2215 (1963); Holcomb D. N. and Tinoco I. Jr.: J. Phys. Chem. **67**, 2691 (1963). — 20. Pershan P. S., Van der Ziel J. P. and Malmstrom L. D.: Phys. Rev. **143**, 574 (1966). — 21. Kielich S.: Acta Phys. Polonica, **32**, 405 (1967). — 22. Kielich S.: J. Colloid Interf. Sci., **30**, 159 (1969).

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