Effect of a Static Magnetic Field on Optical Rotatory Dispersion

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(Received May 28, 1984)

The expression for the optical rotatory dispersion of optically active isotropic systems in the presence of a static magnetic field is studied on the basis of the classical theory of harmonic oscillators. A magnetic optical rotatory dispersion term and a product of dispersion terms for the magnetic optical activity (the Faraday effect) and the natural optical activity are obtained as the effects of the static magnetic field. An equation similar to the Becquerel formula also is derived by use of the oscillator model.

1. Introduction

A number of theoretical investigations of natural optical activity[1] and the Faraday effect[2,3] have been made by using various methods. Since these two phenomena are similar, they can be discussed from the same theoretical point of view.[3]

In classical treatments of the harmonic oscillator model, the index of refraction and the Faraday rotation for optically inactive substances are explained in terms of familiar expressions for optical dispersion.[4,5] For optically active substances one needs to introduce a perturbing force giving rise to the natural optical activity into the equation of motion for the oscillator in the classical theory.[5,6] The force is induced by the electromagnetic field of incident light and is defined with a rotatory parameter.

In the present paper, the optical rotatory dispersion of optically active substances in the presence of a static magnetic field is studied on the basis of the classical theory of dispersion using a non-interacting oscillator model. The effect of the external magnetic field on the natural optical rotation is derived and the relation[7] between the optical rotation and the index of refraction in the absence of external magnetic field is discussed.

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2. Formulation

A system consisting of isotropically-bound charged particles is under consideration. As a model of an optically active molecule we adopt a charged harmonic oscillator, which receives the force that gives rise to the natural optical activity in the presence of electromagnetic field of incident light. The calculation is made in a similar fashion to the classical theory of dispersion for the index of refraction and the Faraday effect.[4-6]

In order to study the effect of a static uniform magnetic field on the natural optical rotatory dispersion, we assume a perturbing force which gives rise to the natural optical rotation in the system and is linearly proportional to rot$\boldsymbol{E}$ where $\boldsymbol{E}$ is the oscillating electric field strength of the electromagnetic field.[5,6] We take the origin of our frame of reference at the equilibrium position of the charged particle. The equation of motion of the charged particle with mass m and electric charge e in the presence of a static uniform magnetic field of strength $\boldsymbol{H}_0$ and an electromagnetic field of strength $\boldsymbol{E}$ is of the following form:

$$m\frac{\mathrm{d}^2\boldsymbol{x}}{\mathrm{d}t^2} = -m\omega_0^2\boldsymbol{x} - m\gamma\frac{\mathrm{d}\boldsymbol{x}}{\mathrm{d}t} + 2eg_0\,\mathrm{rot}\,\boldsymbol{E} + \frac{e}{c}\frac{\mathrm{d}\boldsymbol{x}}{\mathrm{d}t}\times\boldsymbol{H}_0 + e\boldsymbol{E}, \tag{1}$$

where $\boldsymbol{x}$ is the displacement from the equilibrium position, ω_0 the natural vibrational angular frequency of the oscillator, g_0 a rotatory parameter for the natural optical activity and c is the light velocity in the vacuum, and here we introduce a damping force with damping constant γ linearly proportional to the velocity of the particle. The fourth term on the right-hand side of Eq. (1) gives the Lorentz force acting on the particle in the static magnetic field of strength $\boldsymbol{H}_0$, where the magnitude of $\boldsymbol{H}_0$ is much greater than the amplitude of the magnetic field strength $\boldsymbol{H}$ of the electromagnetic field. The last term of Eq. (1) represents the force due to the electromagnetic field. The force due to the magnetic field of strength $\boldsymbol{H}$ is here neglected because of the magnitude of the order of $\frac{e}{c}\left|\frac{\mathrm{d}\boldsymbol{x}}{\mathrm{d}t}\right||\boldsymbol{E}|\left(\left|\frac{\mathrm{d}\boldsymbol{x}}{\mathrm{d}t}\right| \ll c\right)$, where $\boldsymbol{E}$ is the electric field strength of the electromagnetic field. The electric field due to the polarization of the other particles in the system is also neglected, that is, the non-interacting particles are to be under consideration.

If the electric field strength $\boldsymbol{E}$ of an electromagnetic wave is periodic and given by

$$\boldsymbol{E} = \boldsymbol{E}_0\exp\left[i\omega\left(t - \frac{n}{c}\boldsymbol{s}\cdot\boldsymbol{r}\right)\right], \tag{2}$$

the steady state solution to Eq. (1) has the following form with the same fre-

quency ω:

$$\boldsymbol{x} = \boldsymbol{x}_0 e^{i\omega t}, \tag{3}$$

where ω is the angular frequency of the incident light, s the unit vector in the direction of propagation of the light and n is the index of refraction for the system of non-interacting isotropic oscillators. Substitution of the forms given in Eqs. (2) and (3) into Eq. (1) yields

$$-m\omega^2\boldsymbol{x} = -m\omega_0{}^2\boldsymbol{x} - m\gamma i\omega\boldsymbol{x} + 2eg_0\left(-i\omega\frac{n}{c}\,\boldsymbol{s}\times\boldsymbol{E}\right) + \frac{e}{c}\,i\omega\boldsymbol{x}\times\boldsymbol{H}_0 + e\boldsymbol{E}. \tag{4}$$

Let us consider a linearly polarized light propagating in the z direction through the system and a static magnetic field of strength $\boldsymbol{H}_0$ pointing in the direction of z axis; that is, both the directions of the propagating light and the static magnetic field are the same along the z axis. Then, Eq. (4) becomes the set of simultaneous equations

$$\left.\begin{aligned}(\omega_0{}^2 - \omega^2 + i\gamma\omega)x - \frac{e}{mc}i\omega y H_0 &= \frac{e}{m}\left(E_x + i\frac{2g_0\omega}{c}n E_y\right)\\ (\omega_0{}^2 - \omega^2 + i\gamma\omega)y + \frac{e}{mc}i\omega x H_0 &= \frac{e}{m}\left(E_y - i\frac{2g_0\omega}{c}n E_x\right)\\ (\omega_0{}^2 - \omega^2 + i\gamma\omega)z &= \frac{e}{m}E_z\end{aligned}\right\}, \tag{5}$$

where x, y, z and E_x, E_y, E_z are the orthogonal components of $\boldsymbol{x}$ and $\boldsymbol{E}$, respectively.

Equation (5) can be solved by making use of the complex combinations of these components, which are defined by

$$\xi_\pm = x \pm iy \tag{6}$$

and

$$E_\pm = E_x \pm iE_y, \tag{7}$$

where the upper (lower) sign corresponds to the propagation along z of the left (right) circularly polarized light. One may obtain new equations for $\xi_\pm$ by use of Eq. (5) and find out their solutions given by

$$\left.\xi_+ = \frac{e}{m}\frac{1 + \dfrac{2g_0\omega}{c}n}{\omega_0{}^2 - \omega^2 + \omega(-2\omega_L + i\gamma)}E_+\right.$$

$$\left.\begin{aligned}\xi_- &= \frac{e}{m}\frac{1-\dfrac{2g_0\omega}{c}n}{\omega_0^2-\omega^2+\omega(2\omega_L+i\gamma)}E_- \\ z &= \frac{e}{m}\frac{1}{\omega_0^2-\omega^2+\omega(i\gamma)}E_z\end{aligned}\right\}, \tag{8}$$

where ω_L is the Larmor frequency, i.e.,

$$\omega_L = \frac{eH_0}{2mc}. \tag{9}$$

If there are non-interacting localized particles in the system, one obtains easily the electric polarization and the tensor of complex dielectric constant. Let N be the number of particles per unit volume. If there is no polarization in the absence of the electromagnetic wave, the polarization $\boldsymbol{P}$ is related to ξ_+, ξ_-, z and the orthogonal components of $\boldsymbol{P}$ are expressed as

$$\left.\begin{aligned}P_x &= Nex = \frac{1}{2}Ne(\xi_+ + \xi_-) \\ P_y &= Ney = -\frac{i}{2}Ne(\xi_+ - \xi_-) \\ P_z &= Nez\end{aligned}\right\}. \tag{10}$$

Let

$$P_i = \sum_j \alpha_{ij} E_j \quad (i,\ j = x,\ y,\ z), \tag{11}$$

where (α_{ij}) is the polarizability tensor. Then, the dielectric constant tensor is given by

$$\varepsilon_{ij} = \delta_{ij} + 4\pi\alpha_{ij} \quad (i,\ j = x,\ y,\ z), \tag{12}$$

where δ_{ij} is the Kronecker δ symbol. Thus, the dielectric constant tensor is found to be

$$(\varepsilon_{ij}) = \begin{bmatrix} \varepsilon_{xx} & \varepsilon_{xy} & 0 \\ -\varepsilon_{xy} & \varepsilon_{xx} & 0 \\ 0 & 0 & \varepsilon_{zz} \end{bmatrix}, \tag{13a}$$

where

$$\left.\varepsilon_{xx} = 1 + \omega_p^2\frac{\omega_0^2-\omega^2+i\gamma\omega+\dfrac{2g_0\omega}{c}n\cdot 2\omega_L\omega}{(\omega_0^2-\omega^2+i\gamma\omega)^2-4\omega_L^2\omega^2}\right\}$$

$$\left.\begin{aligned}\varepsilon_{zz} &= 1+\omega_p{}^2\frac{1}{\omega_0{}^2-\omega^2+i\gamma\omega}\\ \varepsilon_{xy} &= i\omega_p{}^2\frac{\frac{2g_0\omega}{c}n\cdot(\omega_0{}^2-\omega^2+i\gamma\omega)+2\omega_L\omega}{(\omega_0{}^2-\omega^2+i\gamma\omega)^2-4\omega_L{}^2\omega^2}\end{aligned}\right\} \tag{13b}$$

with

$$\omega_p{}^2=\frac{4\pi Ne^2}{m}, \tag{14}$$

ω_p being plasma frequency.

Expressions for the relation between the index of refraction and the dielectric constant are given by Maxwell's equations expressed in terms of $\boldsymbol{E}$ as[4]

$$\left.\begin{aligned}&(\varepsilon_{xx}-n^2)E_x+\varepsilon_{xy}E_y=0\\ &-\varepsilon_{xy}E_x+(\varepsilon_{xx}-n^2)E_y=0\\ &\varepsilon_{zz}E_z=0\end{aligned}\right\}, \tag{15}$$

where use has been made of Eqs. (2) and (13a) and the linearly polarized light propagating in the z direction is under consideration. The set of homogeneous equations (15) has solutions only if

$$n_\pm{}^2=\varepsilon_{xx}\mp i\varepsilon_{xy}, \tag{16}$$

and the corresponding solutions to the equations are $E_y/E_x=\mp i$ and $E_z=0$. Equation (16) with the upper signs corresponds to the left and that with the lower to the right circularly polarized light.

At frequencies where there is little absorption, one may obtain equatins for $n_\pm$ by substituting Eq. (13b) into Eq. (16) as

$$n_\pm{}^2\mp 2n_\pm\frac{g_0\omega}{c}\frac{\omega_p{}^2}{\omega_0{}^2-\omega^2\mp 2\omega_L\omega}-\left(1+\frac{\omega_p{}^2}{\omega_0{}^2-\omega^2\mp 2\omega_L\omega}\right)=0, \tag{17}$$

where use has been made of $\gamma=0$. Then, the two indices of refraction become

$$n_\pm=\sqrt{1+\frac{\omega_p{}^2}{\omega_0{}^2-\omega^2\mp 2\omega_L\omega}+\left(\frac{\omega_p{}^2\frac{g_0\omega}{c}}{\omega_0{}^2-\omega^2\mp 2\omega_L\omega}\right)^2}\pm\frac{\omega_p{}^2\frac{g_0\omega}{c}}{\omega_0{}^2-\omega^2\mp 2\omega_L\omega}. \tag{18}$$

The rotational angle ϕ of the plane of polarization per unit path length is represented in terms of the difference n_--n_+ between the indices of refraction of

the right and the left circularly polarized light as

$$\phi = \frac{\omega}{c}\frac{n_- - n_+}{2}, \tag{19}$$

where the sense of the rotation is defined so that positive ϕ corresponds to counterclockwise rotation as seen by an observer against the z direction.

For the low magnetic intensity H_0, we consider the expression ϕ containing the terms up to the first order in H_0. With the assumption

$$\left|\frac{eH_0}{mc}\omega\right| \ll \left|\omega_0^2 - \omega^2\right|, \tag{20}$$

one obtains the relation

$$\frac{1}{\omega_0^2 - \omega^2 \mp 2\omega_L\omega} = \frac{1}{\omega_0^2 - \omega^2}\left(1 \pm \frac{2\omega_L\omega}{\omega_0^2 - \omega^2} + \cdots\right). \tag{21}$$

In order to have the expression for ϕ, it is convenient to define the following quantities:

$$\left.\begin{aligned} G &= \frac{g_0\omega}{c} \\ \Omega_\pm &= \Omega \mp 2L \end{aligned}\right\}, \tag{22}$$

where

$$\left.\begin{aligned} \Omega &= \omega_0^2 - \omega^2 \\ L &= \omega_L\omega \end{aligned}\right\}. \tag{23}$$

From Eqs. (18) one can obtain the quantity

$$\frac{n_- - n_+}{2} = \frac{1}{2}\frac{\omega_p^2 \cdot \frac{1}{2}\left(\frac{1}{\Omega_-} - \frac{1}{\Omega_+}\right) + (\omega_p^2 G)^2 \cdot \frac{1}{2}\left(\frac{1}{\Omega_-^2} - \frac{1}{\Omega_+^2}\right)}{\frac{1}{2}\left[\sqrt{1 + \frac{\omega_p^2}{\Omega_-} + \left(\frac{\omega_p^2 G}{\Omega_-}\right)^2} + \sqrt{1 + \frac{\omega_p^2}{\Omega_+} + \left(\frac{\omega_p^2 G}{\Omega_+}\right)^2}\right]} - \omega_p^2 G \cdot \frac{1}{2}\left(\frac{1}{\Omega_-} + \frac{1}{\Omega_+}\right), \tag{24}$$

and the mean n of n_+ and n_-

$$n = \frac{n_- + n_+}{2} = \frac{1}{2}\left[\sqrt{1 + \frac{\omega_p^2}{\Omega_-} + \left(\frac{\omega_p^2 G}{\Omega_-}\right)^2} + \sqrt{1 + \frac{\omega_p^2}{\Omega_+} + \left(\frac{\omega_p^2 G}{\Omega_+}\right)^2}\right] - \omega_p^2 G \cdot \frac{1}{2}\left(\frac{1}{\Omega_-} - \frac{1}{\Omega_+}\right). \tag{25}$$

As far as one investigates the external magnetic field effect up to the first order in H_0 on ϕ, the following approximation may be admitted:

$$\left.\begin{aligned}\frac{1}{2}\left(\frac{1}{\Omega_-}+\frac{1}{\Omega_+}\right)&\approx\frac{1}{\Omega}\\ \frac{1}{2}\left(\frac{1}{\Omega_-}-\frac{1}{\Omega_+}\right)&\approx-\frac{2L}{\Omega^2}\\ \frac{1}{2}\left(\frac{1}{\Omega_-{}^2}-\frac{1}{\Omega_+{}^2}\right)&\approx-\frac{4L}{\Omega^3}\end{aligned}\right\}. \tag{26}$$

This means that the numerator of the first term on the right-hand side of Eq. (24) is linearly proportional to H_0, so that its denominator can be replaced by n in the absence of H_0, leading to the following equation:

$$n_0(\omega)\equiv n(H_0=0)=\sqrt{1+\frac{\omega_p{}^2}{\Omega}+\left(\frac{\omega_p{}^2G}{\Omega}\right)^2}. \tag{27}$$

By taking into account Eqs. (19), (24), (26) and (27) one can write the expression for ϕ as follows:

$$\phi=-\frac{eH_0}{2mc}\frac{\omega_p{}^2}{cn_0(\omega)}\frac{\omega^2}{(\omega_0{}^2-\omega^2)^2}-g_0\frac{\omega_p{}^2}{c^2}\frac{\omega^2}{\omega_0{}^2-\omega^2}$$
$$-\frac{eH_0}{2mc}g_0{}^2\frac{2\omega_p{}^4}{c^3n_0(\omega)}\frac{\omega^4}{(\omega_0{}^2-\omega^2)^3}, \tag{28}$$

where use has been made of Eqs. (22) and (23). The first and the second term on the right-hand side of Eq. (28) show the magnetic and the natural optical rotatory dispersion, respectively, and the parameter g_0 corresponds to the rotational strength. The third term gives rise to the effect of interference between the magnetic and the natural optical activity, and this term may be rewritten in the form

$$\phi'=-\frac{eH_0}{2mc}\frac{\omega_p{}^2}{cn_0(\omega)}\frac{\omega^2}{(\omega_0{}^2-\omega^2)^2}\cdot g_0\frac{\omega_p{}^2}{c^2}\frac{\omega^2}{\omega_0{}^2-\omega^2}\cdot 2g_0. \tag{29}$$

This expression is composed of a product of dispersion terms due to the magnetic and the natural optical activity.

Since the first term on the right-hand side of Eq. (28) gives the dispersion of the Faraday effect, one can study the Faraday effect in optically active substances and the relation between the magnetic and the natural optical activity.

Let us consider a system which is weakly optically active, namely for which $g_0\approx 0$. In the low frequency region $\omega_0{}^2-\omega^2\gg\frac{4\pi Ne^2}{m}\frac{\omega}{c}g_0$, the index $n_0(\omega)$ of refraction in the absence of $\boldsymbol{H}_0$ in Eq. (27) may be approximated to

$$n_0(\omega)=\sqrt{1+\frac{\omega_p^2}{\Omega}}\left[1+\frac{1}{2}\frac{\left(\frac{\omega_p^2}{\Omega}\right)^2}{1+\frac{\omega_p^2}{\Omega}}G^2\right]. \tag{30}$$

This formula contains the zeroth and the second order term in g_0. Furthermore, outside the absorption region, that is, if $\omega_0^2-\omega^2\gg\frac{4\pi Ne^2}{m}$, the mean index $n_0(\omega)$ of refraction reduces to

$$n_0(\omega)=1+\frac{2\pi Ne^2}{m}\frac{1}{\omega_0^2-\omega^2}, \tag{31}$$

and is independent of g_0. As for the rotational angle under these conditions, the rotational angle ϕ will be given by a linear combination of the magnetic and the natural optical rotatory power as

$$\phi_F=-\frac{eH_0}{2mc}\frac{1}{cn_0(\omega)}\frac{4\pi Ne^2}{m}\frac{\omega^2}{(\omega_0^2-\omega^2)^2} \tag{32}$$

and

$$\phi_N=-g_0\frac{1}{c^2}\frac{4\pi Ne^2}{m}\frac{\omega^2}{\omega_0^2-\omega^2}, \tag{33}$$

because ϕ_F is due to the zeroth order in g_0 and ϕ_N due to the first order. Then, let us consider only the terms up to the first order in g_0, and with the assumption that $n_0(\omega)$ is nearly unity, combination of Eqs. (31), (32) and (33) leads to

$$\phi=\phi_F+\phi_N=-\frac{eH_0}{2mc^2}\omega\frac{\partial n_0(\omega)}{\partial\omega}-g_0\frac{2\omega^2}{c^2}[n_0(\omega)-1], \tag{34}$$

where $n_0(\omega)=1$ in Eq. (32) has been used. We have here obtained an expression similar to the one originated by Becquerel,[7] the latter of which gives the relation between the optical rotatory power of weakly optically active substances in the presence of an external static magnetic field and the index of refraction in the absence of external magnetic field.

3. Summary and Discussion

We have studied the optical rotatory dispersion of optically active substances in the presence of an external static magnetic field on the basis of the classical theory on the Faraday effect by use of the non-interacting harmonic oscillator model. As the effect of the static magnetic field on the optical rotatory power, we have obtained an expression containing a term for the magnetic optical rota-

tory dispersion (i.e., the dispersion due to the Faraday effect) and a dispersion term of product of the magnetic and the natural rotatory dispersion. The former term specified by Eq. (32) is nearly independent of the natural optical activity in substances, whereas the latter given by Eq. (29) is related closely to it, and is derived from the third term in the square root in Eq. (18).

Under the condition $\omega_0^2 - \omega^2 \gg \frac{4\pi Ne^2}{m}\frac{\omega}{c}g_0$, the second term in the [ ] brackets in Eq. (30) may be neglected, so that $n_0(\omega)$'s in the first and the third term on the right-hand side of Eq. (28) have no g_0. Therefore, one may have the rotational angle ϕ consisting of the first term without g_0, the second term with g_0 and the third term with g_0^2 in Eq. (28). Consequently, the rotational angle due to the interference of the magnetic and the natural optical activity has been given only by the last term in Eq. (28).

This term, as shown by Eq. (29), is expressed in terms of a product of the dispersion formulae for the magnetic and the natural optical activity, but is of the second order in g_0. The magnetic optical rotation is governed only by properties of substances, whereas the natural optical rotation is governed by both properties of substances and the electromagnetic field of the light applied to substances. For these reasons the rotational angle ϕ' given by Eq. (29) appears. Since the natural optical rotation is caused by the first order spatial dispersion in the wave number q of light and the magnetic optical rotation by the zeroth order in q,[3] one may have a conclusion that the rotation ϕ' specified by Eq. (29) is caused by the second order in q of light.

Furthermore, under the condition $\omega_0^2 - \omega^2 \gg \frac{4\pi Ne^2}{m}$ and with up to the first order term of g_0 under consideration, the relation between the rotational angle ϕ and the index $n_0(\omega)$ of refraction in the absence of static magnetic field has been derived in the form of Eq. (34). This equation is in close connection with the Becquerel formula. The last term ϕ_N in Eq. (34), however, retains the parameter g_0 for natural optical activity, since $n_0(\omega)$ has not the first order term of g_0 but only the zeroth order one.

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