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EFFECT OF ELECTRON PARAMAGNETIC RESONANCE ON THE OPTICAL FARADAY EFFECT AT LOW TEMPERATURES

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An expression is derived for the angle of rotation of the plane of polarization of light in a sample in a fixed magnetic field to which is applied an rf magnetic field in the region of a paramagnetic resonance. A comparison is made with the experimental data.

1. Daniels and Wesemeyer[1] have reported experimental results concerning the effect of a magnetic resonance on the optical Faraday effect. The measurements were made with a neodymium ethylsulfate single crystal at 1.5°K, using microwave power at 9060 Mcs and the green mercury line at 5461 A. With no microwave power the angle of rotation of the plane of polarization was a linear function of the strength of the fixed magnetic field. When the microwave power was switched on a noticeable reduction in the angle of rotation was observed near resonance. In microwave fields strong enough for saturation no rotation of the plane of polarization could be observed even at very strong fields ($H_0 \sim 1750$ oe).

The relation between the optical Faraday effect and paramagnetic resonance was first noted by Kastler[2] and the quantum-mechanical calculation was carried out by Opechowski.[3] In these papers it is pointed out that the effect of the radio-frequency field is to cause a redistribution in the electron populations in the energy levels which are split by the fixed field (corresponding to different values of the magnetic quantum number). This redistribution leads to a change in the intensity of the Zeeman lines and affects the polarization of the medium.

On the basis of a quantum-mechanical calculation of the magnetic and electric dipole transitions caused by the radio-frequency and optical radiation, Opechowski gives an expression for the polarization tensor which contains a correction for the magnetic dipole transitions. However, it is difficult to compare his results with experiment because of the dipole-transition matrix elements which appear in the expression; also, the population numbers for the initial and final energy levels, between which these transitions take place, are unknown. The analysis in reference 3 does not take account of the magnetic polarization due to the light and this effect is important at low temperatures.

Below we give a simple macroscopic analysis of the effect; this analysis yields an explicit expression for the angle rotation of the plane of polarization of the light near the paramagnetic resonance in a radio-frequency field, h, which is weak compared with the fixed magnetic field $H_0 = H_z$. The constants which appear in the final expressions can be determined from optical, magnetic, and radio-frequency measurements.

2. The effect of a paramagnetic resonance on optical phenomena is due primarily to spin-orbit interactions. The optical effects are characterized by the dielectric permittivity, which is determined by the orbital motion of the electrons in the atom. In the region of a paramagnetic resonance there is a marked change in the spin system. When spin-orbit coupling is taken into account the change in the state of the spin system results in a change in the dielectric permittivity. The latter effect is important in the low-temperature region, where the magnetization of a paramagnet is close to saturation even in a relatively weak magnetic field. The following system of equations[4] can be used for a macroscopic description of these effects:

$$\Delta \mathbf{A} = -\frac{4\pi}{c}\left\{c \operatorname{curl} \mathbf{M} + \dot{\mathbf{P}} - \frac{1}{4\pi c}\ddot{\mathbf{A}}\right\}; \tag{1}$$

$$\ddot{\mathbf{P}}_\alpha + \omega_\alpha^2 \mathbf{P}_\alpha = -(e^2/mc)\dot{\mathbf{A}} + (e/mc)\dot{\mathbf{P}}\times\mathbf{B}; \tag{2}$$

$$\dot{\mathbf{M}} + \mathbf{M}/\tau = (\chi_0/\tau)\mathbf{H} + \gamma\mathbf{M}\times\mathbf{B}; \tag{3}$$

$$\mathbf{P} = \sum_\alpha N_\alpha \mathbf{P}_\alpha;\ \ \mathbf{B} = \operatorname{curl}\mathbf{A} = \mathbf{H}_0 + \mathbf{H}_i + 4\pi\mathbf{M}, \tag{4}$$

where **P** and **M** are the densities of the electric and magnetic moments respectively. The vector potential **A** is given in the form

$$\mathbf{A} = \mathbf{A}_0 + \mathbf{A}(r, t), \text{ where } \operatorname{curl}\mathbf{A}_0 = \mathbf{B}_0 = \mathbf{H}_0 + 4\pi\mathbf{M}_0.$$

As has been shown in reference 4, using this system it is possible to introduce the spin-orbit interaction within the framework of a macroscopic theory. The self-consistent internal field $\mathbf{H}_i$ is due to the spin-spin (source of c curl $\mathbf{M}$) and spin-orbit (source of $\dot{\mathbf{P}}$) interactions. The $\dot{\mathbf{P}}$ term in Eq. (1) describes the polarization current associated with the change of electric dipole moment of the atom due to the rotational electric field. This rotational electric field produces oscillations in the magnetization. Equation (2) describes the change in polarization in the ensemble of atoms which interact with the internal electromagnetic field. Equation (2) contains no damping term; there is no significant change in the final results of the calculation when damping is introduced.

3. If a linearly polarized electromagnetic wave is propagated in the medium in the direction of the fixed magnetizing field $\mathbf{H}_0$, the plane of polarization is rotated. The specific angle of rotation[5]

$$\theta = (\omega / 4c)\,(n_-^2 - n_+^2) / n \tag{5}$$

is determined by the refractive index $n_\pm = ck/\omega$ for waves with right- and left-handed polarization and by the refractive index n if there is no magnetic field $\mathbf{H}_0$.

In order to find $n_\pm$ and n we start from Eqs. (1) to (4), taking the solution in the form

$$\mathbf{A} = {}^1/_2\, \mathbf{r}_\cdot \times \mathbf{B}_0 + \sum_{\mathbf{k}\neq 0} \mathbf{A}_\mathbf{k} \exp\{-i\omega t + i\mathbf{k}\cdot\mathbf{r}\}; \tag{6}$$

$$\mathbf{P}_\alpha = \sum_\mathbf{k} \mathbf{P}_{\alpha\mathbf{k}} \exp\{-i\omega t + i\mathbf{k}\cdot\mathbf{r}\}; \tag{7}$$

$$\mathbf{M} = \mathbf{M}_0 + \sum_{\mathbf{k}\neq 0} \mathbf{m}_\mathbf{k} \exp\{-i\omega t + i\mathbf{k}\cdot\mathbf{r}\}. \tag{8}$$

Substituting Eq. (7) in Eq. (2), including higher-order terms we have

$$\mathbf{P}_{\alpha k} = i\frac{e^2}{mc}\,\frac{\omega}{\omega_\alpha^2 - \omega^2}\,\mathbf{A}_k. \tag{9}$$

Assuming further that $\mathbf{M} = \mathbf{M}_0 + \mathbf{m}$, in the same approximation we have

$$\operatorname{curl} \mathbf{m}_k = i\frac{\gamma}{\omega}\,\mathbf{k}\cdot\mathbf{M}_0(\mathbf{k}\times\mathbf{A}_k). \tag{10}$$

Now, from Eqs. (6) and (1)

$$\left\{\left[k^2 - \frac{\omega^2}{c^2} - \sum_\alpha \frac{4\pi N_\alpha e^2}{mc^2}\,\frac{\omega^2}{\omega_\alpha^2 - \omega^2}\right]^2 - \left(\frac{4\pi\gamma}{\omega}\right)^2 (\mathbf{k}\cdot\mathbf{M})^2 k^2\right\} \mathbf{A}_\mathbf{k} = 0. \tag{11}$$

Thus, at optical frequencies the refractive index is

$$n_\pm^2 = (kc/\omega)^2 \equiv n^2\{1 \mp (4\pi\gamma/\omega)\,M_{0z}\}, \tag{12}$$

where

$$n = 1 + \sum_\alpha (4\pi N_\alpha e^2/m)/(\omega_\alpha^2 - \omega^2). \tag{13}$$

Hence, the specific angle of rotation of the plane of polarization

$$\theta = (2\pi\gamma/c)\,nM_{0z} \tag{14}$$

is determined by the dependence of the magnetization component M_{0Z} on the fixed magnetizing field H_0 and the amplitude of the radio-frequency field. At saturation, where $M_Z = 0$, both values of the index of refraction $n_\pm = n$ and $\theta = 0$.

4. If the radio-frequency field h_0 is circularly polarized

$$h_x = h_0 \cos\omega_0 t;\; h_y = h_0 \sin\omega_0 t, \tag{15}$$

the expression for M_{0Z} is found easily, writing Eq. (3) in a coordinate system which rotates about the z-axis at the frequency ω_0.

Under steady-state conditions

$$\boldsymbol{\omega}_0 \times \mathbf{M}_0 = \gamma\mathbf{M}\times\mathbf{H}_0 + (\chi_0\mathbf{H} - \mathbf{M}_0)/\tau.$$

Solving the last equation for M_Z, we have

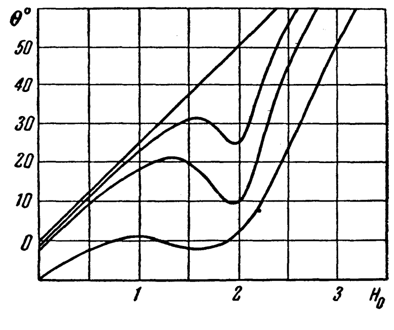
$$M_{0z} = \chi_0 H_0 \left\{1 - \gamma^2\,\frac{1 - \Delta\omega/\gamma H_0}{\Delta\omega^2 + \tau^{-2} + \gamma^2 h_0^2}\,h_0^2\right\}, \tag{16}$$

where

$$\Delta\omega = \omega_0 + \gamma H_0.$$

Equations (14) and (16) determine the explicit dependence of the angle of rotation of the plane of polarization in a paramagnetic medium as a function of H_0 and h_0.

It follows from Eq. (16) that when $h_0 = 0$ the angle of rotation is proportional to the magnetizing field H_0. If the radio-frequency field is weak $|\gamma|\tau h \ll 1$ the function $\theta(H_0)$ has a valley in the resonance region $\omega_0 = |\gamma| H_0$; the depth of the valley increases with h_0. At stronger radio-frequency fields, where $|\gamma|\tau h < 1$, θ starts to increase rapidly when $|\gamma| H_0 > \omega_0$. For value of H_0 smaller than $\omega/|\gamma|$ there is a change in the sign of the angle, as is apparent from the figure. Because of the approximations made in the derivation Eq. (14), in strong radio-frequency fields we can expect only qualitative agreement with experiment.



Theoretical dependence of the angle θ on the intensity of the magnetizing field for different values of the radio-frequency field.

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