

## Theory of the Faraday and Kerr Effects in Ferromagnetics\*

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Both the Faraday and (magneto-optic, polar) Kerr effects in ferromagnetics are treated on the basis of the band theory of metals. The spin-orbit interaction gives the electron wave functions such left-right asymmetry that the "magnetic" electrons, under the action of a plane polarized light wave, produce an average current perpendicular to the plane of polarization. The polarizability and conductivity tensors are evaluated. The model is capable of describing the rotation of the plane of polarization of the light and the elliptical polarization resulting from transmission or reflection on the ferromagnetic medium. Order of magnitude estimates of the tensor components, based on plausible assumptions on the nature of the electronic wave functions and the energy bands in ferromagnetics, give values that agree reasonably well with experimental results. The temperature and frequency dependence of these effects as given by the formulas is also in agreement with experiment.

### 1. INTRODUCTION

IT was observed in the last century that the transmitted and reflected beams, into which a plane polarized beam of light splits upon incidence on a ferromagnetic body magnetized in a direction parallel to the beam, become elliptically polarized with their major axes rotated with respect to the plane of polarization of the incident beam. This phenomenon, when it refers to the transmitted beam, is designated as the Faraday effect; it is known as the (magneto-optic, polar) Kerr effect when it refers to the reflected beam. Experiments have shown that these effects are all proportional to the net magnetization of the sample and not to the external magnetic field, as is the case with the nonferromagnetic bodies. The difference between ferromagnetic and nonferromagnetic materials extends to the order of magnitude of the effects. For example, in quartz the Faraday rotation is of the order of  $2^\circ$  per centimeter in an external magnetic field of the order of  $10^4$  gauss, whereas for iron it is of the order of  $380\,000^\circ$  per centimeter under the same conditions. It has been established that these effects are connected with the ferromagnetic properties of the specimen, since for temperatures higher than the Curie temperature of the material these effects disappear along with the ferromagnetic properties.

Macroscopically all these effects can be described, as we shall see below, by assigning to the medium a "refractive" tensor which takes the place of the ordinary complex index of refraction, or, equivalently, by two different complex indices of refraction for light of right- and left-handed circular polarization.

Early attempts to explain these phenomena on a microscopic theory of matter<sup>1</sup> consisted in using the Becquerel formula for the difference of the indices of refraction for right- and left-circularly polarized light

under the action of an external magnetic field, with the assumption that in ferromagnetics there is an effective magnetic field active of considerably higher order of magnitude than that of the external field. In fact, Voigt found that the effective field necessary to produce the observed effects is of the order of  $10^6$ - $10^7$  oersteds, i.e., of the order of magnitude of the so-called Weiss field, which was postulated to account for the existence of the ferromagnetic properties of matter.

It is impossible to explain the origin of such a strong magnetic field. The effective magnetic field for a charge inside a magnetized medium is, according to Wannier,<sup>2</sup> equal to  $\mathbf{H} + 2\pi(1+p)\mathbf{M}$ , where  $p$  is a parameter depending on the motion of the electron, varying between 0 and 1. The nature of the Weiss field, responsible for ferromagnetism, was explained by Heisenberg as a result of exchange interactions among the electrons. Although such an interaction energy can be thought of as an equivalent effective magnetic field as far as the alignment of the elementary magnets is concerned, it cannot affect the motion of the electrons as an equivalent magnetic field. It is the motion of the charges that is of importance here, since this motion gives rise to the electric current and thus affects the optical properties of the specimen.

The answer to this problem was provided by Hulme,<sup>3</sup> who introduced into the picture the spin-orbit interaction. This is the energy of interaction of the magnetic moment of an electron,  $\mathbf{y}$ , with the magnetic field it "sees" as it moves through the electric field,  $-\nabla V$ , inside the medium with momentum,  $\mathbf{p}$ , and it has the form,  $\sim \mathbf{y} \times \nabla V \cdot \mathbf{p}$ . We see immediately that such an interaction provides a relation between the motion ( $\mathbf{p}$ ) and the magnetic moment ( $\mathbf{y}$ ) of the electron, and thus it is plausible that it may be responsible for the connection between the optical and ferromagnetic properties that the Faraday and Kerr effects indicate. Indeed, the spin-orbit interaction can, in a certain approximation, be thought of as an effective magnetic

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<sup>1</sup> For an account of these attempts see: Handbuch der Experimental-Physik, XVI, 1. Teil.

<sup>2</sup> G. Wannier, Phys. Rev. 72, 304 (1947).

<sup>3</sup> H. R. Hulme, Proc. Roy. Soc. (London) A135, 237 (1932); see also O. Halpern, Ann Physik [5] 12, 181 (1932).

field, of vector potential  $\sim \mathbf{y} \times \nabla V$ , on the motion of the electrons. This also indicates why the spin-orbit interaction, although present in all matter, is operative in this manner only in ferromagnetics. In nonferromagnetics the electrons are in pairs with opposite  $\mathbf{y}$ , and thus half of the electrons find themselves in one magnetic field and the other half in an opposite field, the net result being no effect at all.

Hulme calculated the two indices of refraction, using the Heisenberg model of a ferromagnetic and the Kramers-Heisenberg dispersion formula, which gives the index of refraction in terms of the energy eigenvalues and the matrix elements of the appropriate electric dipole moment operator with respect to the eigenfunctions of the system. He accounted for the difference between the indices of refraction of the right- and left-circularly polarized beams by considering the splitting of the energy eigenvalues of the system due to the spin-orbit interaction and the fact that now states with different energy differences are combined under the two appropriate dipole moment operators. He neglected, however, the effect of the spin-orbit interaction on the wave functions. Kittel<sup>4</sup> showed, by an order of magnitude argument on a simple atomic model, that this change of the wave functions can give rise, on the basis again of the Kramers-Heisenberg dispersion formula, to a difference of the two indices of refraction of the desired order of magnitude. This is important, since several experiments have shown that the orbital angular momentum in ferromagnetic materials is quenched and thus there is no shifting of the energy eigenvalues resulting from spin-orbit interaction. Also, on account of the adopted model, Hulme found only the real indices of refraction, thus neglecting absorption. However, consideration of absorption is essential in discussing all the effects under study here, as can be seen from Eqs. (38), (39), (40), and (41). In fact, if, e.g., we assume the extinction coefficient for iron equal to zero, we can see that the Faraday rotation changes sign. Similarly, the Faraday elliptical polarization and the Kerr rotation vanish unless there is a difference between the coefficients of extinction of right- and left-circularly polarized light.

In order to describe both dispersion and absorption in a direct manner, the band theory of metals is used, on the basis of which the phenomenon of ferromagnetism has been discussed by Slater,<sup>5</sup> Stoner,<sup>6</sup> and others.<sup>7</sup> The main point here is that the exchange forces effectively displace the energy bands of electrons with spin "up" with respect to those of electrons with spin "down," and thus in equilibrium there are more electrons with one spin than with the opposite spin.

<sup>4</sup> C. Kittel, *Phys. Rev.* **83**, 208 (A) (1951); also (private communication).

<sup>5</sup> J. C. Slater, *Phys. Rev.* **49**, 537, 931 (1936); **52**, 198 (1937); see also J. C. Slater, *Solid State and Molecular Theory Group, Technical Report No. 6* (unpublished).

<sup>6</sup> E. C. Stoner, *Proc. Roy. Soc. (London)* **A165**, 372 (1938); **A169**, 339 (1939).

<sup>7</sup> See several articles in *Revs. Modern Phys.* **25**, No. 1 (1953).

This is the distribution of electrons among the possible Bloch states in a single ferromagnetic domain. It should be noted that the actual ferromagnetic samples consist of many domains with their net spins oriented in different directions and that, in addition, the samples are in general polycrystalline. This structure of the specimen has been taken into account in the calculations below.

Since we are interested in computing only the coherent radiation scattered by the system, which alone determines the refraction and extinction of the incident electromagnetic wave, the problem can be treated on the basis of the semiclassical theory of radiation. According to this theory, the current induced in the system by the incident radiation is considered as giving rise to the coherent scattered radiation according to the laws of classical electrodynamics. Thus, the main task is to calculate the current density induced in the system by an electromagnetic wave of certain frequency and, hence to find the conductivity and polarizability tensors of the system under consideration.

The calculation of the induced current had presented some difficulties which have been resolved by Wilson.<sup>8</sup> He showed that if all frequencies are kept in the expression for the induced current, both the polarization and conduction currents can be obtained directly, and there are no infinities to be explained away. The calculations below are carried out in a manner analogous to that of Wilson.

In the following the first-order effect of the spin-orbit interaction on the optical properties of ferromagnetics is examined on the basis of the band theory of metals, and it is shown that such an effect can account for the order of magnitude, dispersion, and temperature dependence of the Faraday and (magneto-optic, polar) Kerr phenomena in ferromagnetics.

## 2. CALCULATION OF THE TENSORS

The first step is to take as a system a single crystal of a ferromagnetic substance magnetized spontaneously in a given direction, i.e., to consider a single ferromagnetic domain. It is assumed that the lattice vibrations are of negligible importance in the optical phenomena that are considered. This is justified as long as the period of the incident electromagnetic wave is much smaller than the electron-lattice relaxation time. If  $\tau$  is this relaxation time and  $\omega$  the angular frequency of radiation, this condition gives  $(1/\omega) \ll \tau$ , or  $\omega \gg (1/\tau) \sim 10^{13}$  per second. Thus, the following considerations apply to optical, ultraviolet, and higher frequencies. It will be seen later that other approximations in the calculations will render the theory inapplicable to frequencies much higher than ultraviolet. Also the influence of the external magnetic field is neglected, since, as mentioned in the introduction, it is not sufficient to produce the observed effects. In the following, the effect on the magneto-optic phenomena of the spin-orbit interaction

<sup>8</sup> A. H. Wilson, *Proc. Roy. Soc. (London)* **A151**, 274 (1935).

only is considered. Thus, the results will be strictly valid in the case of a ferromagnetic specimen magnetized in the absence of a magnetic field. Actually, however, they will be very good approximations even in the case of a ferromagnetic under saturation conditions, since magnetic fields of the order of  $10^4$  gauss produce magneto-optic effects entirely negligible in comparison to those observed. Thus, the role of the external magnetic field reduces to merely magnetizing the sample in a certain direction.

In this calculation the usual one-electron approximation is used in describing the ground and excited states of the system. The one-electron Hamiltonian

$$\mathcal{H} = \mathcal{H}_0 + \mathcal{H}' + \mathcal{H}'' ,$$

where

$$\mathcal{H}_0 = (1/2m)\mathbf{p}^2 + V(\mathbf{r}) ,$$

$$\mathcal{H}' = (1/2m^2c^2)[\nabla V(\mathbf{r}) \times \mathbf{p}] \cdot \mathbf{s} ,$$

$$\mathcal{H}'' = (e/mc)\mathbf{A}(\mathbf{r}, t) \cdot \mathbf{p} .$$

In these expressions  $V(\mathbf{r})$  is the potential energy of an electron in the crystal in the absence of radiation, and represents the averaged influence of the nuclei and all the other electrons on the electron under consideration.  $\mathbf{A}(\mathbf{r}, t)$  is the vector potential of the electromagnetic field inside the material. Such a potential of a monochromatic light wave of angular frequency  $\omega (= 2\pi\nu)$  is

$$\mathbf{A}(\mathbf{r}, t) = \mathbf{a}(\mathbf{r})e^{-i\omega t} + \mathbf{a}^*(\mathbf{r})e^{i\omega t} . \quad (1)$$

$\mathbf{p}$  is the momentum operator  $(\hbar/i)\nabla$ , and  $\mathbf{s} = (\hbar/2)\boldsymbol{\sigma}$  is the electron spin operator. Finally,  $m$  is the mass of the electron and  $(-e)$  its charge.  $\mathcal{H}'$  is the spin-orbit interaction energy of the electron, of which the significance in these problems was discussed in the introduction.  $\mathcal{H}''$  is the interaction of the electron with the electromagnetic field in the material, only the largest term being kept.

The wave equation to be solved is

$$\mathcal{H}\Psi = i\hbar(\partial\Psi/\partial t) .$$

The solution is obtained by the method of the variation of constants,  $\mathcal{H}''$  being treated as a perturbation. Let the orthonormalized eigenfunctions of  $(\mathcal{H}_0 + \mathcal{H}')$  be  $\phi_{\lambda, s}(\mathbf{r}) \cdot \exp(-i\omega_{\lambda} t)$ , i.e.,

$$(\mathcal{H}_0 + \mathcal{H}')\phi_{\lambda, s}(\mathbf{r}) = \epsilon_{\lambda}^s \cdot \phi_{\lambda, s}(\mathbf{r}) , \quad (2)$$

$$\hbar\omega_{\lambda}^s = \epsilon_{\lambda}^s . \quad (2a)$$

Here  $\lambda$  denotes all the quantum numbers necessary for the space part of the wave function and  $s (= \pm 1)$  stands for the spin quantum number. If the electron is in the state  $\phi_{\lambda, s}(\mathbf{r})$  at  $t=0$ , i.e., before the application of the light wave, its wave function at time  $t$  is, to the first order in the amplitude of the light wave,

$$\Psi_{\lambda, s}(\mathbf{r}, t) = \phi_{\lambda, s}(\mathbf{r}) \exp(-i\omega_{\lambda} t) + \sum_{\lambda', \sigma} a_{\lambda\lambda', \sigma\sigma}(t) \phi_{\lambda', \sigma}(\mathbf{r}) \exp(-i\omega_{\lambda'} t) , \quad (3)$$

where

$$a_{\lambda\lambda', \sigma\sigma}(t) = \frac{ie}{mc} \left[ \frac{\exp[i(\omega_{\lambda'} - \omega)t] - 1}{\omega_{\lambda'} - \omega} \times \int \phi_{\lambda', \sigma}^*(\mathbf{r}) \mathbf{a}(\mathbf{r}) \cdot \nabla \phi_{\lambda, s}(\mathbf{r}) d\tau + \frac{\exp[i(\omega_{\lambda'} + \omega)t] - 1}{\omega_{\lambda'} + \omega} \times \int \phi_{\lambda', \sigma}^*(\mathbf{r}) \mathbf{a}^*(\mathbf{r}) \cdot \nabla \phi_{\lambda, s}(\mathbf{r}) d\tau \right] , \quad (3a)$$

with the notation

$$\omega_{\lambda'} = \omega_{\lambda'}^{\sigma} - \omega_{\lambda'}^s = (\epsilon_{\lambda'}^{\sigma} - \epsilon_{\lambda'}^s)/\hbar ; \quad \sigma = \pm 1 ,$$

( $d\tau$  will always denote integration over the whole crystal). The summation over  $\lambda', \sigma$  in this formula extends over all possible states; it will be seen later how other considerations change this.

The current density of an electron in a state  $\Psi_{\lambda, s}(\mathbf{r}, t)$  and in the presence of a vector potential  $\mathbf{A}(\mathbf{r}, t)$  is given by the formula (part of the current operator that would give terms proportional to magnetization square has been dropped)

$$\mathbf{j}_{\lambda, s}(\mathbf{r}, t) = ie\hbar/2m(\Psi_{\lambda, s}^* \nabla \Psi_{\lambda, s} - \Psi_{\lambda, s} \nabla \Psi_{\lambda, s}^*) - (e^2/mc)\mathbf{A}\Psi_{\lambda, s}^* \Psi_{\lambda, s} . \quad (4)$$

Using Eqs. (3) and (3a), the easily verified relations [see Eq. (1)]

$$\mathbf{a}e^{-i\omega t} = \frac{1}{2}\mathbf{A} + \frac{i}{2\omega} \frac{\partial \mathbf{A}}{\partial t} ,$$

$$\mathbf{a}^*e^{i\omega t} = \frac{1}{2}\mathbf{A} - \frac{i}{2\omega} \frac{\partial \mathbf{A}}{\partial t} ,$$

and neglecting terms of second or higher order in the amplitude of the light wave, after some simplification, we find that Eq. (4) takes the form,

$$\mathbf{j}_{\lambda, s}(\mathbf{r}, t) = ie\hbar/2m(\phi_{\lambda, s}^* \nabla \phi_{\lambda, s} - c.c.) - \frac{e^2\hbar}{4m^2c} \sum_{\lambda', \sigma} \left\{ f_{\lambda', \lambda}^{\sigma\sigma} (\phi_{\lambda, s}^* \nabla \phi_{\lambda', \sigma} - \phi_{\lambda', \sigma} \nabla \phi_{\lambda, s}^*) \int \phi_{\lambda', \sigma}^* \mathbf{A} \cdot \nabla \phi_{\lambda, s} d\tau + c.c. \right\} - \frac{ie^2\hbar}{4m^2c} \frac{1}{\omega} \sum_{\lambda', \sigma} \left\{ f_{\lambda', \lambda}^{\sigma\sigma} (\phi_{\lambda, s}^* \nabla \phi_{\lambda', \sigma} - \phi_{\lambda', \sigma} \nabla \phi_{\lambda, s}^*) \int \phi_{\lambda', \sigma}^* \frac{\partial \mathbf{A}}{\partial t} \cdot \nabla \phi_{\lambda, s} d\tau - c.c. \right\} - \frac{e^2}{mc} \mathbf{A} \phi_{\lambda, s}^* \phi_{\lambda, s} , \quad (5)$$

where

$$f_{\lambda,\lambda'}^{\sigma_s \pm} = \frac{1 - \exp[-i(\omega_{\lambda',\lambda}^{\sigma_s} - \omega)t]}{\omega_{\lambda',\lambda}^{\sigma_s} - \omega} \pm \frac{1 - \exp[-i(\omega_{\lambda',\lambda}^{\sigma_s} + \omega)t]}{\omega_{\lambda',\lambda}^{\sigma_s} + \omega}. \quad (5a)$$

$\phi_{\lambda,s}$  can be found by perturbation theory. Let  $\psi_n(\mathbf{k}, \mathbf{r})$  be the orthonormalized eigenfunctions of  $\mathcal{H}_0$  and  $E_n(\mathbf{k})$  their eigenvalues, i.e.,

$$\mathcal{H}_0 \psi_n(\mathbf{k}, \mathbf{r}) = E_n(\mathbf{k}) \psi_n(\mathbf{k}, \mathbf{r}), \quad (6)$$

$$\int \psi_n^*(\mathbf{k}', \mathbf{r}) \psi_m(\mathbf{k}, \mathbf{r}) d\tau = \delta_{nm} \delta(\mathbf{k}' - \mathbf{k}). \quad (6a)$$

Here the reduced Brillouin zone scheme is used in enumerating the Bloch functions  $\psi_n(\mathbf{k}, \mathbf{r})$ ;  $n$  denotes the energy band, and  $\mathbf{k}$  is the wave vector.  $\psi_n(\mathbf{k}, \mathbf{r})\alpha(s)$  can be taken as unperturbed eigenfunctions, where  $\alpha(s)$ ,  $s = \pm 1$ , are the spin eigenfunctions of the Pauli spin operator  $\sigma_{z_0}$ , i.e.,

$$\sigma_{z_0} \alpha(\pm 1) = \pm \alpha(\pm 1). \quad (7)$$

Here  $z_0$  denotes an arbitrary direction of quantization of the spin. In the "molecular field" approximation of the exchange forces the quantities  $E_n(\mathbf{k}) \pm \delta$  are taken as the energy eigenvalues of the eigenfunctions  $\psi_n(\mathbf{k}, \mathbf{r})\alpha(\pm 1)$ . Now, since the perturbation  $\mathcal{H}'$  has the periodicity of the lattice, it connects only Bloch states of different bands with the same  $\mathbf{k}$ . This also proves that the Bloch functions are the correct zero-order wave functions for the perturbation  $\mathcal{H}'$  for all  $\mathbf{k}$ , except possibly for few  $\mathbf{k}$ 's for which there might be degeneracy of different bands. Such cases can be disregarded, as their effect is certainly negligible.

In expression (5) for the current density, terms up to the order of (spin-orbit energy)  $\times$  (amplitude of light wave) are kept, and thus, effectively,

$$\phi_{\lambda, \pm 1}(\mathbf{r}) = [\psi_n(\mathbf{k}, \mathbf{r}) \pm \chi_n(\mathbf{k}, \mathbf{r})] \alpha(\pm 1), \quad (8)$$

where

$$\epsilon_{\lambda}^{\pm 1} = E_n(\mathbf{k}) \pm \delta, \quad (8a)$$

$$\chi_n(\mathbf{k}, \mathbf{r}) = \sum_{m \neq n} b_{nm}(\mathbf{k}) \psi_m(\mathbf{k}, \mathbf{r}), \quad (8b)$$

with

$$b_{nm}(\mathbf{k}) = \frac{-i\hbar^2/4m^2c^2}{E_n(\mathbf{k}) - E_m(\mathbf{k})} \times \int \psi_m^*(\mathbf{k}) (\nabla V \times \nabla)_{z_0} \psi_n(\mathbf{k}) d\tau. \quad (8c)$$

The other parts of  $\phi_{\lambda, \pm 1}$  give terms of order higher than that worked with. The diagonal matrix element of  $\mathcal{H}'$  is zero, as can be seen directly by making use of the time and space inversion symmetry properties of the Bloch functions, thus exhibiting the quenching of orbital angular momentum.

To this approximation, Eq. (5) takes the form,

$$\begin{aligned} \mathbf{j}_{\lambda, \pm 1}(\mathbf{r}, t) = & \frac{ie\hbar}{2m} (\phi_{\lambda, \pm 1}^* \nabla \phi_{\lambda, \pm 1} - \text{c.c.}) \\ & - \frac{e^2\hbar}{4m^2c} \sum_{\lambda'} \{ f_{\lambda', \lambda}^+ [\mathbf{K}_{\lambda', \lambda}(\mathbf{A}) \pm \mathbf{L}_{\lambda', \lambda}(\mathbf{A})] + \text{c.c.} \} \\ & - \frac{ie^2\hbar}{4m^2c} \frac{1}{\omega} \sum_{\lambda'} \{ f_{\lambda', \lambda}^- [\mathbf{K}_{\lambda', \lambda}(\partial\mathbf{A}/\partial t) \\ & \pm \mathbf{L}_{\lambda', \lambda}(\partial\mathbf{A}/\partial t)] - \text{c.c.} \} \\ & - \frac{e^2}{mc} \mathbf{A} \{ \psi_{\lambda}^* \psi_{\lambda} \pm [\psi_{\lambda\chi}^* + \text{c.c.}] \}, \quad (9) \end{aligned}$$

where

$$\mathbf{K}_{\lambda', \lambda}(\mathbf{A}) = (\psi_{\lambda}^* \nabla \psi_{\lambda'} - \psi_{\lambda'} \nabla \psi_{\lambda}^*) \int \psi_{\lambda'}^* \mathbf{A} \cdot \nabla \psi_{\lambda} d\tau,$$

$$\begin{aligned} \mathbf{L}_{\lambda', \lambda}(\mathbf{A}) = & (\psi_{\lambda}^* \nabla \psi_{\lambda'} - \psi_{\lambda'} \nabla \psi_{\lambda}^*) \int (\psi_{\lambda'}^* \mathbf{A} \cdot \nabla \chi_{\lambda} \\ & + \chi_{\lambda'}^* \mathbf{A} \cdot \nabla \psi_{\lambda}) d\tau + (\psi_{\lambda}^* \nabla \chi_{\lambda'} + \chi_{\lambda'}^* \nabla \psi_{\lambda} \\ & - \psi_{\lambda'} \nabla \chi_{\lambda}^* - \chi_{\lambda} \nabla \psi_{\lambda'}^*) \int \psi_{\lambda'}^* \mathbf{A} \cdot \nabla \psi_{\lambda} d\tau, \end{aligned}$$

$$f_{\lambda', \lambda}^{\pm} = \frac{1 - \exp[-i(\omega_{\lambda', \lambda} - \omega)t]}{\omega_{\lambda', \lambda} - \omega} \pm \frac{1 - \exp[-i(\omega_{\lambda', \lambda} + \omega)t]}{\omega_{\lambda', \lambda} + \omega},$$

$$\omega_{\lambda', \lambda} = \omega_{\lambda'} - \omega_{\lambda} = (E_{\lambda'} - E_{\lambda})/\hbar,$$

$\lambda$  denotes quantum numbers ( $n, \mathbf{k}$ ),  $\lambda'$  denotes quantum numbers ( $m, \mathbf{k}'$ ), and

$$\sum_{\lambda'} \rightarrow \frac{\Omega}{(2\pi)^3} \sum_m \int_{\text{first B.Z.}} d\mathbf{k}'. \quad (\Omega = \text{volume of specimen}).$$

The next step is to find the total current density. In this Hartree approximation,

$$\mathbf{j} = \mathbf{j}_{+1} + \mathbf{j}_{-1}, \quad (10)$$

where

$$\mathbf{j}_{+1} = \sum_{\lambda} \mathbf{j}_{\lambda, +1}, \quad \mathbf{j}_{-1} = \sum_{\lambda} \mathbf{j}_{\lambda, -1}, \quad (10a)$$

the summations extending over states occupied by electrons with the corresponding spin. This is equivalent to assuming a completely degenerate electron distribution. For a nondegenerate distribution,  $\mathbf{j}_{\pm 1}$  are obtained from  $\mathbf{j}_{\lambda, \pm 1}$  through Eqs. (10a) by multiplication with the Fermi distribution function.  $\mathbf{j}_{\lambda, \pm 1}$  are given by Eq. (9) where, according to the time-dependent perturbation theory,  $\lambda'$  in the summation should go over all possible one-electron states, whether occupied or unoccupied. This is a result of the Hartree approximation, where each electron is assumed to be in a definite state and the Pauli principle is taken into account by having not more than two electrons (and in the case of two, with opposite spins) in the same Bloch state. It

was observed<sup>9</sup> in connection with the optical properties of atoms that agreement with experiment obtains, if only the nonoccupied states in Eq. (9) are summed over. This can be explained theoretically on the basis of the Fock approximation, which takes the exclusion principle into account directly by describing the states of the whole system by determinantal wave functions constructed out of the one-electron wave functions used.<sup>10</sup> Thus,  $\mathbf{j}$  is given by Eqs. (10) and (10a), where  $\mathbf{j}_{\lambda, \pm 1}$  are given by Eq. (9) with the understanding that now  $\lambda'$  stands for any one of the nonoccupied states, which is denoted symbolically by  $\lambda' > \lambda$ .

Making use of the relations

$$\psi_n(-\mathbf{k}, \mathbf{r}) = \psi_n^*(\mathbf{k}, \mathbf{r}), \quad E_n(-\mathbf{k}) = E_n(\mathbf{k}),$$

$$\chi_n(-\mathbf{k}, \mathbf{r}) = -\chi_n^*(\mathbf{k}, \mathbf{r}),$$

which give

$$f_{mn}^\pm(-\mathbf{k}', -\mathbf{k}) = f_{mn}^\pm(\mathbf{k}', \mathbf{k}), \quad \mathbf{K}_{mn}(-\mathbf{k}', -\mathbf{k})$$

$$= \mathbf{K}_{mn}^*(\mathbf{k}', \mathbf{k}), \quad \mathbf{L}_{mn}(-\mathbf{k}', -\mathbf{k}) = -\mathbf{L}_{mn}^*(\mathbf{k}', \mathbf{k}),$$

and noting that if state  $(n, \mathbf{k})$  is occupied so is  $(n, -\mathbf{k})$ , the total current density

$$\mathbf{j} = \mathbf{j}^{(0)} + \mathbf{j}^{(1)}, \quad (11)$$

where

$$\mathbf{j}^{(0)} = -\frac{e^2 \hbar}{2m^2 c} \sum_{\lambda(+)} \sum_{\lambda' > \lambda} (\Re f_{\lambda', \lambda^+}) \mathbf{K}_{\lambda', \lambda}(\mathbf{A})$$

$$+ \frac{e^2 \hbar}{2m^2 c} \frac{1}{\omega} \sum_{\lambda(+)} \sum_{\lambda' > \lambda} (\Im f_{\lambda', \lambda^-}) \mathbf{K}_{\lambda', \lambda}(\partial \mathbf{A} / \partial t)$$

$$- \frac{e^2}{mc} \mathbf{A} \sum_{\lambda(+)} \psi_\lambda^* \psi_\lambda, \quad (11a)$$

and

$$\mathbf{j}^{(1)} = -\frac{ie^2 \hbar}{2m^2 c} \sum_{\lambda(-)} \sum_{\lambda' > \lambda} (\Im f_{\lambda', \lambda^+}) \mathbf{L}_{\lambda', \lambda}(\mathbf{A})$$

$$- \frac{ie^2 \hbar}{2m^2 c} \frac{1}{\omega} \sum_{\lambda(-)} \sum_{\lambda' > \lambda} (\Re f_{\lambda', \lambda^-}) \mathbf{L}_{\lambda', \lambda}(\partial \mathbf{A} / \partial t)$$

$$- \frac{e^2}{mc} \mathbf{A} \sum_{\lambda(-)} (\psi_\lambda \chi_\lambda^* + \text{c.c.}). \quad (11b)$$

$\sum_{\lambda(-)}$  indicates that the summation is to be taken over the states that are occupied *only* by electrons with spin  $\alpha(+1)$ , i.e., the electrons responsible for the magnetic properties of the sample, and which shall be called magnetic electrons.  $\sum_{\lambda(+)}$  denotes a summation over all states occupied by electrons with spin  $\alpha(+1)$  and all states occupied by electrons with spin  $\alpha(-1)$ .  $\Re$  and  $\Im$  denote the real and imaginary parts of a complex quantity, respectively. The term  $\sum_{\lambda} (\phi_{\lambda, \pm 1}^* \nabla \phi_{\lambda, \pm 1} - \text{c.c.})$

<sup>9</sup> H. Kramers and R. de L. Krönig, Z. Physik 48, 174 (1938).

<sup>10</sup> F. Seitz, *Modern Theory of Solids* (McGraw-Hill Book Company, Inc., New York, 1940), Chap. XVII.

has been dropped, since its macroscopic value is zero.  $\mathbf{j}^{(0)}$  is the induced current density in the absence of any spin-orbit interaction.  $\mathbf{j}^{(1)}$  arises from spin-orbit interaction; only the magnetic electrons contribute to  $\mathbf{j}^{(1)}$ , and this points out the connection of the magnetic and optical properties of metals in so far as the spin-orbit interaction is responsible for it.

In this semiclassical theory of radiation, variations in a region small compared to the wavelength of the wave are unimportant, and thus the calculations can be simplified by finding the average current density over a unit cell. This restricts the applicability of the results to light waves of frequencies  $\omega$  such that  $(2\pi c/\omega) \gg a$ , where  $a$  is the lattice constant. Since  $a \sim 10^{-8}$  cm,  $\omega \ll 10^{18}$  per second; hence, the results do not apply to x-ray or higher frequencies. This and the previous approximation of disregarding collisions of electrons with lattice vibrations limit the range of validity of the formulas pretty well to optical and ultraviolet frequencies. However, as will be shown below, this averaging will give a simplified expression for the total current density, part of which will be proportional to the electric field  $\mathbf{E}$  of the light wave (conduction current) and the rest proportional to  $\partial \mathbf{E} / \partial t$  (polarization current). This was first demonstrated by Wilson<sup>8</sup> in connection with the optical properties of solids.

A coordinate system  $x, y, z$  is taken so that the wave propagates parallel to the  $z$ -axis. It is assumed that  $\mathbf{A}(\mathbf{r}, t)$  depends only on  $z$ , i.e.,  $\mathbf{A}(\mathbf{r}, t) = \mathbf{A}(z, t)$ , and it will be proved later that this is a self-consistent assumption, i.e., that this wave gives rise to such a current density that Maxwell's equations for this density admit of a solution  $\mathbf{A}(z, t)$  depending only on  $z$ . The averaging calculations are straightforward generalizations of those by Wilson, and as such they will not be reported here. However, this averaging process neglects the momentum of the light wave in comparison to that of the electrons (which is reasonable in the frequencies used), and gives essentially the selection rule  $\mathbf{k}' = \mathbf{k}$  for the nonvanishing of the integral  $\int \psi_\lambda^* \mathbf{A} \cdot \nabla \psi_\lambda d\tau = \int \psi_m^*(\mathbf{k}', \mathbf{r}) \mathbf{A} \cdot \nabla \psi_n(\mathbf{k}, \mathbf{r}) d\tau$ . The Bloch character of the wave functions is used here, namely,

$$\psi_n(\mathbf{k}, \mathbf{r}) = (1/N^{1/2}) e^{i\mathbf{k} \cdot \mathbf{r}} u_n(\mathbf{k}, \mathbf{r}), \quad (12)$$

and

$$\chi_n(\mathbf{k}, \mathbf{r}) = (1/N^{1/2}) e^{i\mathbf{k} \cdot \mathbf{r}} w_n(\mathbf{k}, \mathbf{r}), \quad (13)$$

where, as can be seen from Eqs. (8b) and (12),

$$w_n(\mathbf{k}, \mathbf{r}) = \sum_{m \neq n} b_{nm}(\mathbf{k}) u_m(\mathbf{k}, \mathbf{r}), \quad (14)$$

$b_{nm}(\mathbf{k})$  being given by Eq. (8c). Here  $u_n(\mathbf{k}, \mathbf{r})$ , and consequently  $w_n(\mathbf{k}, \mathbf{r})$  also, has the periodicity of the lattice and is normalized over a unit cell, i.e.,

$$\int u_n^*(\mathbf{k}, \mathbf{r}) u_n(\mathbf{k}, \mathbf{r}) d\tau_0 = 1.$$

$d\tau_0$  will denote integration over a unit cell, in contradistinction to  $d\tau$  that denotes integration over the whole

crystal. Thus,  $\psi_n(\mathbf{k}, \mathbf{r})$  is normalized over the whole crystal according to Eq. (6a);  $N$  is the number of unit cells in the crystal.

Using the relations giving the vector potential in terms of the electric field  $\mathbf{E}(z, t)$ , i.e.,

$$\mathbf{E} = -\frac{1}{c} \frac{\partial \mathbf{A}}{\partial t}, \quad \frac{\partial \mathbf{E}}{\partial t} = -\frac{\omega^2}{c} \mathbf{A},$$

the result of this calculation, i.e., the average values of  $\mathbf{j}$ ,  $\mathbf{j}^{(0)}$ , and  $\mathbf{j}^{(1)}$ , given by Eqs. (11), (11a), and (11b), respectively, is

$$\mathbf{J}(\mathbf{r}, t) = \mathbf{J}^{(0)}(\mathbf{r}, t) + \mathbf{J}^{(1)}(\mathbf{r}, t), \quad (15)$$

where  $\mathbf{J}$ , the total macroscopic current density (which, by the nature of the calculation, includes both the conduction and polarization current), and  $\mathbf{J}^{(0)}$ ,  $\mathbf{J}^{(1)}$  are given by the relations,

$$\mathbf{J}^{(0)} = \boldsymbol{\sigma}^{(0)} \cdot \mathbf{E} + \boldsymbol{\alpha}^{(0)} \cdot (\partial \mathbf{E} / \partial t), \quad (15a)$$

$$\mathbf{J}^{(1)} = \boldsymbol{\sigma}^{(1)} \cdot \mathbf{E} + \boldsymbol{\alpha}^{(1)} \cdot (\partial \mathbf{E} / \partial t). \quad (15b)$$

The  $\boldsymbol{\sigma}$ 's and  $\boldsymbol{\alpha}$ 's are conductivity and polarizability tensors, of which the components are:

$$\sigma_{ij}^{(0)} = \frac{e^2 \hbar}{8\pi^3 m^2} \frac{1}{\omega} \sum_{m>n} \int_V [\mathcal{G} f_{mn}^-(\mathbf{k})] P_{mn}^{ij}(\mathbf{k}) d\mathbf{k},$$

$$\alpha_{ij}^{(0)} = -\frac{e^2 \hbar}{8\pi^3 m^2} \frac{1}{\omega^2} \int_V \left\{ \frac{m}{\hbar} \delta_{ij} - \sum_{m>n} [\mathcal{R} f_{mn}^+(\mathbf{k})] P_{mn}^{ij}(\mathbf{k}) \right\} d\mathbf{k},$$

$$\sigma_{ij}^{(1)} = \frac{e^2 \hbar}{8\pi^3 m^2} \frac{1}{\omega} \sum_{m>n} \int_V [\mathcal{R} f_{mn}^-(\mathbf{k})] Q_{mn}^{ij}(\mathbf{k}) d\mathbf{k},$$

$$\alpha_{ij}^{(1)} = -\frac{e^2 \hbar}{8\pi^3 m^2} \frac{1}{\omega^2} \sum_{m>n} \int_V [\mathcal{G} f_{mn}^+(\mathbf{k})] Q_{mn}^{ij}(\mathbf{k}) d\mathbf{k},$$

with

$$P_{mn}^{ij}(\mathbf{k}) = \int u_m^*(\mathbf{k}) \frac{\partial u_n(\mathbf{k})}{\partial x_i} d\tau_0 \times \int u_m(\mathbf{k}) \frac{\partial u_n^*(\mathbf{k})}{\partial x_j} d\tau_0, \quad (16a)$$

$$Q_{mn}^{ij}(\mathbf{k}) = \frac{1}{i} \left[ \int u_m^* \frac{\partial u_n}{\partial x_i} d\tau_0 \times \int \left( u_m^* \frac{\partial w_n}{\partial x_j} + w_m^* \frac{\partial u_n}{\partial x_j} \right) d\tau_0 + \int u_m^* \frac{\partial u_n}{\partial x_j} d\tau_0 \times \int \left( u_m \frac{\partial w_n^*}{\partial x_i} + w_m \frac{\partial u_n^*}{\partial x_i} \right) d\tau_0 \right]. \quad (16b)$$

$$f_{mn}^{\pm}(\mathbf{k}) = \frac{1 - \exp[-i(\omega_{mn} - \omega)t]}{\omega_{mn} - \omega} \pm \frac{1 - \exp[-i(\omega_{mn} + \omega)t]}{\omega_{mn} + \omega},$$

$$\omega_{mn} = \frac{E_m(\mathbf{k}) - E_n(\mathbf{k})}{\hbar}.$$

$\sum_{m>n}$  extends over all nonoccupied bands.  $\int_V$  denotes integration over all occupied states, *once* for *each* spin orientation (this includes effectively a summation over  $n$ ).  $\int_v$  denotes integration over the states occupied by *magnetic* electrons only. Here, for convenience, the  $x$ ,  $y$ ,  $z$  components of a vector have been denoted by the subscripts  $i$ ,  $j$  ( $i, j = 1, 2, 3$ ).  $f^{\pm}$  is a function that often appears in collision and radiation problems and has the property,<sup>11</sup>

$$\lim_{t \rightarrow \infty} f_{mn}^{\pm}(\mathbf{k}) = \left[ \frac{\mathcal{P}}{\omega_{mn} - \omega} + i\pi\delta(\omega_{mn} - \omega) \right] \pm \left[ \frac{\mathcal{P}}{\omega_{mn} + \omega} + i\pi\delta(\omega_{mn} + \omega) \right],$$

where  $\mathcal{P}/x$  is called the principal value of  $x$  (it behaves like  $1/x$  for  $x \neq 0$ , but it is zero for  $x = 0$ ), and  $\delta(x)$  is the Dirac delta function. Thus, Eqs. (16), in the limit of large  $t$ , take on the form,

$$\sigma_{ij}^{(0)} = \frac{e^2 \hbar}{8\pi^3 m^2} \sum_{m>n} \int_V \frac{\delta(\omega_{mn} - \omega)}{\omega} P_{mn}^{ij}(\mathbf{k}) d\mathbf{k}, \quad (17)$$

$$\alpha_{ij}^{(0)} = -\frac{e^2 \hbar}{4\pi^3 m^2} \int_V \left[ \frac{m}{2\hbar\omega^2} \delta_{ij} - \sum_{m>n} \frac{\omega_{mn}(\mathbf{k})}{\omega^2(\omega_{mn}^2 - \omega^2)} P_{mn}^{ij}(\mathbf{k}) \right] d\mathbf{k},$$

$$\sigma_{ij}^{(1)} = \frac{e^2 \hbar}{4\pi^3 m^2} \sum_{m>n} \int_V \frac{1}{\omega_{mn}^2 - \omega^2} Q_{mn}^{ij}(\mathbf{k}) d\mathbf{k}, \quad (18)$$

$$\alpha_{ij}^{(1)} = -\frac{e^2 \hbar}{8\pi^3 m^2} \sum_{m>n} \int_V \frac{\delta(\omega_{mn} - \omega)}{\omega^2} Q_{mn}^{ij}(\mathbf{k}) d\mathbf{k},$$

where now the integral sign means the principal value of the integral. Thus, the theory is free of infinities. Here the term containing  $\delta(\omega_{mn} + \omega)$  has been dropped, as it contributes nothing to the integral. Taking  $t$  very large does not invalidate the use of time-dependent perturbation theory, since this can always be done for a small enough radiation field.

The tensors now can be simplified by making use of the symmetry properties of the wave functions. The

<sup>11</sup> W. Heitler, *The Quantum Theory of Radiation* (Clarendon Press, London, 1954), third edition.

calculations are not carried out here,<sup>12</sup> but only the general scheme of simplification is indicated. The tensor components of  $\sigma^{(0)}$  and  $\alpha^{(0)}$  are of the general form,

$$p_{ij} = \int_V g[\omega_{mn}(\mathbf{k})] P_{mn}^{ij}(\mathbf{k}) d\mathbf{k}, \quad (19)$$

and those of  $\sigma^{(1)}$  and  $\alpha^{(1)}$  are of the form,

$$q_{ij} = \int_v g[\omega_{mn}(\mathbf{k})] Q_{mn}^{ij}(\mathbf{k}) d\mathbf{k}, \quad (20)$$

where  $g(\omega_{mn})$  is some function of  $\omega_{mn}(\mathbf{k})$ , and  $P_{mn}^{ij}$ ,  $Q_{mn}^{ij}$  are given by Eqs. (16a) and (16b). The region  $V$  (or  $v$ ) in  $\mathbf{k}$ -space over which we integrate can be divided into a number of equal regions, all of which can be obtained from a "fundamental" region  $V_f$  (or  $v_f$ ) by application of the symmetry operations,  $R$ , of the point symmetry group of the lattice.<sup>13</sup> It is known<sup>14</sup> that for a general  $\mathbf{k}$ , with a nondegenerate star, the following equations are true:

$$E_m(R\mathbf{k}) = E_m(\mathbf{k}); \quad \omega_{mn}(R\mathbf{k}) = \omega_{mn}(\mathbf{k}); \quad (21)$$

and

$$\psi_m(R\mathbf{k}, \mathbf{r}) = \psi_m(\mathbf{k}, R^{-1}\mathbf{r}); \quad u_m(R\mathbf{k}, \mathbf{r}) = u_m(\mathbf{k}, R^{-1}\mathbf{r}). \quad (22)$$

Equations (19) and (20) can then be rewritten in the following form:

$$\begin{aligned} p_{ij} &= \int_V g[\omega_{mn}(\mathbf{k})] P_{mn}^{ij}(\mathbf{k}) d\mathbf{k} \\ &= \sum_R \int_{(RV_f)} g[\omega_{mn}(\mathbf{k})] P_{mn}^{ij}(\mathbf{k}) d\mathbf{k} \\ &= \int_{V_f} \sum_R g[\omega_{mn}(R\mathbf{k})] P_{mn}^{ij}(R\mathbf{k}) d\mathbf{k} \\ &= \int_{V_f} g[\omega_{mn}(\mathbf{k})] \sum_R P_{mn}^{ij}(R\mathbf{k}) d\mathbf{k}, \\ q_{ij} &= \int_{v_f} g[\omega_{mn}(\mathbf{k})] \sum_R Q_{mn}^{ij}(R\mathbf{k}) d\mathbf{k}. \end{aligned}$$

Within the fundamental regions  $V_f$  and  $v_f$  the functions  $\omega_{mn}(\mathbf{k})$ ,  $P_{mn}^{ij}(\mathbf{k})$ , and  $Q_{mn}^{ij}(\mathbf{k})$  depend on the detailed nature of the crystal potential  $V(\mathbf{r})$ , and no general statement can be made about them; we can hope, then, to detect the vanishing or nonvanishing of  $p_{ij}$  and  $q_{ij}$ , not through the integration over  $\mathbf{k}$ , but rather through the vanishing or nonvanishing of the factors  $\sum_R P^{ij}(R\mathbf{k})$  and  $\sum_R Q^{ij}(R\mathbf{k})$  in the integrand.

<sup>12</sup> For more details at this and other points see author's thesis, University of California, Berkeley, 1954 (unpublished).

<sup>13</sup> H. Brooks, thesis, Harvard University, 1940 (unpublished).

<sup>14</sup> F. Seitz, *Ann. Math.* **37**, 17 (1936); see also Bouckaert, Smoluchowski, and Wigner, *Phys. Rev.* **50**, 58 (1936).

Using Eq. (22), it can be proved that

$$\int u_m^*(R\mathbf{k}) \nabla u_n(R\mathbf{k}) d\tau_0 = \int u_m^*(\mathbf{k}) (R\nabla) u_n(\mathbf{k}) d\tau_0,$$

and thus,

$$\begin{aligned} \sum_R P_{mn}^{ij}(R\mathbf{k}) &= \sum_R \int u_m^*(\mathbf{k}) (R\nabla) u_n(\mathbf{k}) d\tau_0 \\ &\quad \times \int u_m^*(\mathbf{k}) (R\nabla)_j u_n(\mathbf{k}) d\tau_0. \end{aligned}$$

An analogous equation can be written for  $\sum_R Q_{mn}^{ij}(R\mathbf{k})$ . We are interested in the case of the cubic group  $O_h$ ; this is the point symmetry group for cubic lattices, and thus the following apply to the ferromagnetic Fe and Ni crystals. In the special case of the beam propagating along one of the cubic axes of the crystal,  $(R\nabla)_i$  can be found easily, and thus some tensor components are proved to be zero. By a tensor transformation the tensors for a general arrangement of the direction of propagation of the beam, the cubic axes, and the spin direction can then be found. Then the average tensors over the different ferromagnetic domains and the crystal orientations can be found, and these are

$$\begin{aligned} \sigma_{ij} &= \sigma_0 \delta_{ij} + \sigma^{(1)} \beta_{ij}, \\ \alpha_{ij} &= \alpha_0 \delta_{ij} + \alpha^{(1)} \beta_{ij}, \end{aligned} \quad (23)$$

where

$$\delta_{ij} = \begin{pmatrix} 1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & 1 \end{pmatrix}, \quad \beta_{ij} = \begin{pmatrix} 0 & -\beta_3 & \beta_2 \\ \beta_3 & 0 & -\beta_1 \\ -\beta_2 & \beta_1 & 0 \end{pmatrix},$$

$[\beta_i (i=1,2,3)]$  are the direction cosines of the net spin direction of the specimen with respect to coordinate system attached to the beam propagating along the 3-direction].

$$\begin{aligned} \sigma_0 &= \frac{e^2 \hbar}{8\pi^2 m^2} \frac{1}{3} \sum_{m>n} \int_V \frac{\delta(\omega_{mn} - \omega)}{\omega} \left| \int u_m^* \nabla u_n d\tau_0 \right|^2 d\mathbf{k}, \\ \alpha_0 &= \frac{e^2 \hbar}{4\pi^3 m^2} \frac{1}{3} \sum_{m>n} \int_V \frac{\omega_{mn}}{\omega^2 (\omega_{mn}^2 - \omega^2)} \\ &\quad \times \left| \int u_m^* \nabla u_n d\tau_0 \right|^2 d\mathbf{k} - \frac{e^2 n}{m \omega^2}, \end{aligned} \quad (24)$$

where  $n$  = total number of electrons per unit volume.

$$\begin{aligned} \sigma^{(1)} &= \frac{e^2 \hbar}{4\pi^3 m^2} \sum_{m>n} \int_V \frac{1}{\omega_{mn}^2 - \omega^2} Q_{mn}(\mathbf{k}) d\mathbf{k}, \\ \alpha^{(1)} &= -\frac{e^2 \hbar}{8\pi^2 m^2} \sum_{m>n} \int_V \frac{\delta(\omega_{mn} - \omega)}{\omega^2} Q_{mn}(\mathbf{k}) d\mathbf{k}, \end{aligned} \quad (25)$$

with

$$Q_{mn}(\mathbf{k}) = 2\Re \left[ \frac{1}{i} \int u_m \frac{\partial u_n^*}{\partial y} d\tau_0 \right. \\ \left. \times \int \left( u_m^* \frac{\partial w_n}{\partial x} + w_m^* \frac{\partial u_n}{\partial x} \right) d\tau_0 \right]. \quad (26)$$

$w_n$  is given by Eq. (14), where now

$$b_{nm}(\mathbf{k}) = -\frac{i\hbar/4m^2c^2}{\omega_{mn}(\mathbf{k})} \int \psi_m^* (\nabla V \times \nabla)_z \psi_n d\tau.$$

Thus, the average macroscopic total current density

$$\mathbf{J}_{\text{total}} = \boldsymbol{\sigma} \cdot \mathbf{E} + \boldsymbol{\alpha} \cdot \partial \mathbf{E} / \partial t, \quad (27)$$

where  $\boldsymbol{\sigma}$  and  $\boldsymbol{\alpha}$  are given by Eqs. (23). This is the basic equation which will give all the optical phenomena when used in conjunction with Maxwell's equations for the electromagnetic field.

### 3. FARADAY AND KERR EFFECTS

Consider now the case of a ferromagnetic sample (such as iron or nickel) magnetized in the  $+z$ -direction. In such instance,

$$\beta_1 = \beta_2 = 0, \quad \beta_3 \neq 0,$$

and the conductivity and polarizability tensors are

$$\boldsymbol{\sigma} = \begin{bmatrix} \sigma_0 & -\sigma_1 & 0 \\ \sigma_1 & \sigma_0 & 0 \\ 0 & 0 & \sigma_0 \end{bmatrix}, \quad \boldsymbol{\alpha} = \begin{bmatrix} \alpha_0 & -\alpha_1 & 0 \\ \alpha_1 & \alpha_0 & 0 \\ 0 & 0 & \alpha_0 \end{bmatrix}, \quad (28)$$

with

$$\sigma_1 = \sigma^{(1)}\beta_3; \quad \alpha_1 = \alpha^{(1)}\beta_3. \quad (28a)$$

The tensors have the form required by general symmetry arguments except for the last diagonal element, which does not necessarily have to be equal to the other two. This may arise from magnetostriction or other purely optical effects which have been neglected. However, it will be seen that the magnitude of the third diagonal element does not enter the discussion of the Faraday and Kerr effects.

The relevant Maxwell equations are

$$\nabla \times \mathbf{E} = -\frac{1}{c} \frac{\partial \mathbf{H}}{\partial t}, \\ \nabla \times \mathbf{H} = -\frac{1}{c} \frac{\partial \mathbf{E}}{\partial t} + \frac{4\pi}{c} \mathbf{J}_{\text{total}}.$$

It should be noted here that both the so-called conduction and polarization currents have been included in  $\mathbf{J}_{\text{total}}$ . Also, it has been assumed that  $(\partial \mathbf{M} / \partial t) = 0$ , where  $\mathbf{M}$  is the average magnetization of the sample. This assumption is justified, since the induced magnetization by the  $\mathbf{H}$  field of the light wave is zero in our frequency range.

Using expression (27) for  $\mathbf{J}_{\text{total}}$ ,

$$\nabla \times \mathbf{E} = -\frac{1}{c} \frac{\partial \mathbf{H}}{\partial t}, \quad (29)$$

$$\nabla \times \mathbf{H} = -\frac{1}{c} \frac{\partial \mathbf{E}}{\partial t} + \frac{4\pi}{c} \boldsymbol{\alpha} \cdot \frac{\partial \mathbf{E}}{\partial t} + \frac{4\pi}{c} \boldsymbol{\sigma} \cdot \mathbf{E}. \quad (30)$$

Looking for solutions of the form of a monochromatic plane wave of angular frequency  $\omega$  propagating along the  $z$ -axis, we have

$$\mathbf{E} = \mathbf{E}_0 e^{i\omega(t - Nz/c)} \\ \mathbf{H} = \mathbf{H}_0 e^{i\omega(t - Nz/c)}, \quad (31)$$

where  $\mathbf{E}_0$ ,  $\mathbf{H}_0$  are constant vectors and  $N$  is the complex index of refraction. It is known from the electromagnetic theory of optical phenomena that such solutions exist for suitable  $N$ . This proves the self-consistency of the assumption made before, namely, that  $\mathbf{E}$  is a function of  $z$  and  $t$  only. Equations (29) and (30) become, for such time and space dependence of  $\mathbf{E}$  and  $\mathbf{H}$ ,

$$N(\mathbf{E} \times \mathbf{k}) = -\mathbf{H}, \quad (32)$$

$$N(\mathbf{H} \times \mathbf{k}) = \mathfrak{M} \cdot \mathbf{E}, \quad (33)$$

where

$$\mathfrak{M} = \mathbf{1} + 4\pi\boldsymbol{\alpha} + \frac{4\pi}{i\omega}\boldsymbol{\sigma} = \begin{bmatrix} A_0 & -A_1 & 0 \\ A_1 & A_0 & 0 \\ 0 & 0 & A_0 \end{bmatrix}, \\ A_0 = 1 + 4\pi\alpha_0 + \frac{4\pi}{i\omega}\sigma_0, \\ A_1 = 4\pi\alpha_1 + \frac{4\pi}{i\omega}\sigma_1, \quad (33a)$$

and  $\mathbf{k}$  = unit vector in the  $z$ -direction. Substituting Eq. (32) for  $\mathbf{H}$  in Eq. (33), the fundamental equation for the optics of anisotropic bodies is obtained, namely,

$$\mathfrak{M} \cdot \mathbf{E} = N^2 [\mathbf{E} - \mathbf{k}(\mathbf{E} \cdot \mathbf{k})], \quad (34)$$

or, in component form,

$$(A_0 - N^2)E_x - A_1E_y = 0, \\ A_1E_x + (A_0 - N^2)E_y = 0, \quad (35) \\ A_0E_z = 0.$$

The first two equations for  $E_x$ ,  $E_y$  have a nontrivial solution only if the determinant of their coefficients vanishes, i.e.,  $(A_0 - N^2)^2 + A_1^2 = 0$ , and thus  $N^2$  is equal to either

$$N_+^2 = A_0 - iA_1, \quad (36)$$

or

$$N_-^2 = A_0 + iA_1. \quad (37)$$

The solution corresponding to  $N_+^2 = A_0 - iA_1$  is  $E_y^{(+)} = +iE_x^{(+)}$ , and the one corresponding to  $N_-^2 = A_0 + iA_1$



is  $E_y^{(-)} = -iE_z^{(-)}$ . Thus, there are two possible ways for a wave to propagate along the  $z$ -axis: either as  $E_0^{(+)} \exp\{i\omega[t - (N_+/c)z]\}$ , which denotes a right-handed circularly polarized wave traveling with the complex velocity  $c/N_+$ ; or as  $E_0^{(-)} \exp\{i\omega[t - (N_-/c)z]\}$ , which is a left-handed circularly polarized wave traveling with complex velocity  $c/N_-$ . Any linear superposition of the two is also a possible wave.

The constants pertaining to the Faraday and (magneto-optic, polar) Kerr effects can now be found. A plane polarized beam, upon incidence on the surface of a sample, assuming normal incidence, splits into the refracted and reflected beams. The refracted beam becomes elliptically polarized with the major axis rotated by an angle

$$\phi_F = \frac{1}{2}(\omega z/c) \mathfrak{R}(N_+ - N_-), \quad (38)$$

at a distance  $z$  from the boundary (a positive sign of  $\phi_F$  denotes a rotation of the major axis from  $x$ - to  $y$ -axis;  $xyz$  is a right-handed triad), and with an ellipticity (ratio of the minor to the major axis) given by

$$\epsilon_F = -\tanh\left[\frac{1}{2}(\omega z/c) \mathfrak{I}(N_+ - N_-)\right]. \quad (39)$$

Equations (38) and (39) are the constants of the Faraday effect. The reflected beam also becomes elliptically polarized with the major axis rotated by an angle

$$\phi_K = -\mathfrak{I}[(N_+ - N_-)/(N_+ N_- - 1)], \quad (40)$$

with an ellipticity equal to

$$\epsilon_K = -\mathfrak{R}[(N_+ - N_-)/(N_+ N_- - 1)]. \quad (41)$$

Equations (40) and (41) are the constants of the (magneto-optic, polar) Kerr effect, in the special case of normal incidence.  $\phi_K$ , too, counts as positive for rotation from  $x$ - to  $y$ -axis, but it must be kept in mind that the beam to which it refers is traveling along the  $-z$ -direction. These formulas are valid for  $(N_+ - N_-) \ll N_+$  or  $N_-$ .

It is interesting to note that all four phenomena can be observed only in absorbing media. If the medium is transparent, i.e., with real  $N_+$  and  $N_-$ , then the ellipticity of the refracted beam is zero, i.e., it is a plane polarized beam, and the rotation of the polarization of the reflected beam also vanishes.

The phenomena connected with the case of a ferromagnetic magnetized in a direction perpendicular to the propagation of the beam, such as the Cotton-Moutton effect, etc., cannot be discussed, since a consistent treatment of this problem requires the calculation of the tensor components to the second order in spin-orbit interaction.

$N_+$ ,  $N_-$  are now calculated for our case. From Eqs. (33a) it can be seen that

$$A_0 = 1 + 4\pi(\alpha_0 - i\sigma_0/\omega), \quad (42)$$

$$iA_1 = 4\pi[(\sigma_1/\omega) + i\alpha_1]. \quad (43)$$

From the definitions of  $\sigma_0$ ,  $\alpha_0$ ,  $\sigma_1$ ,  $\alpha_1$ , it is noted that  $(\sigma_1/\omega) \ll \alpha_0$  and  $\alpha_1 \ll (\sigma_0/\omega)$ , since only the magnetic electrons contribute to  $\sigma_1$ ,  $\alpha_1$ , whereas all of them contribute to  $\sigma_0$ ,  $\alpha_0$ . This is especially so near the Curie point, since at the Curie temperature  $\sigma_1 = \alpha_1 = 0$ . Experimental values, on the other hand, indicate that at room temperatures  $(\sigma_1/\omega)$  and  $\alpha_1$  are about 30–50 times smaller than  $\alpha_0$  and  $(\sigma_0/\omega)$ , respectively. Using this approximation,  $iA_1 \ll A_0$ , and Eqs. (36) and (37),

$$N_+ = (A_0 - iA_1)^{1/2} \cong \sqrt{A_0} - \frac{1}{2}iA_1/\sqrt{A_0},$$

$$N_- = (A_0 + iA_1)^{1/2} \cong \sqrt{A_0} + \frac{1}{2}iA_1/\sqrt{A_0}.$$

Thus, in this approximation,

$$N_+ - N_- = -iA_1/\sqrt{A_0}, \quad (44)$$

$$\frac{N_+ - N_-}{N_+ N_- - 1} = \frac{-iA_1}{(A_0 - 1)\sqrt{A_0}}. \quad (45)$$

It should be observed that  $\sqrt{A_0}$  is the complex index of refraction,  $N = n - ik$  ( $n$  = real index of refraction;  $k$  = extinction coefficient), of the medium in the absence of any spin-orbit interaction. This is approximately equal to the complex index of refraction of the ferromagnetic above the Curie point, as can be seen with the help of Eqs. (24) and verified experimentally by reflectivity vs temperature experiments.<sup>15</sup>

Thus, from Eqs. (42), (43), (44), and (45),

$$N_+ - N_- = (-4\pi) \frac{(\sigma_1/\omega) + i\alpha_1}{n - ik}, \quad (46)$$

$$\frac{N_+ - N_-}{N_+ N_- - 1} = (-4\pi) \frac{(\sigma_1/\omega) + i\alpha_1}{(n - ik)[(n - ik)^2 - 1]}. \quad (47)$$

These are to be used to estimate the Faraday and Kerr constants given by Eqs. (38), (39), (40), (41).

#### 4. COMPARISON WITH EXPERIMENT

In comparing the results of this theory with experiment,  $n$  and  $k$  are assumed to be given by independent experiments on the optical constants of the specimen, although they are given in principle through Eqs. (24). We shall attempt to estimate the fundamental quantities of our theory,  $\sigma_1$  and  $\alpha_1$ , in terms of which the Faraday and Kerr effects are given.

The expressions for  $\sigma_1$  and  $\alpha_1$  [Eqs. (25)] can be approximated as follows:

$$\sigma_1 = \frac{e^2 \hbar}{4\pi^2 m^2} \beta_3 \sum_{m>n} \langle Q_{mn} / (\omega_{mn}^2 - \omega^2) \rangle_{\nu} \int_{\nu} d\mathbf{k},$$

$$\alpha_1 = -\frac{e^2 \hbar}{8\pi^2 m^2} \frac{1}{\omega^2} \sum_{m>n} \langle \delta(\omega_{mn} - \omega) Q_{mn} \rangle_{\nu} \int_{\nu} d\mathbf{k},$$

<sup>15</sup> L. Ornstein and J. H. van der Veen, *Physica* 3, 289 (1936); L. Ornstein and O. Koefoed, *Physica* 5, 175 (1938); J. H. van der Veen and L. Ornstein, *Physica* 6, 439 (1939).

where the angular parentheses indicate an obvious average. But now

$$\int_{\nu} d\mathbf{k} = (2\pi)^3 \times \left( \frac{\text{no. of magnetic electrons}}{\text{unit volume}} \right) = (2\pi)^3 \frac{M_s}{\frac{1}{2}g\mu_B},$$

where  $M_s$  is the saturation magnetization of a domain,  $g$  is the spectroscopic splitting factor, and  $\mu_B = (e\hbar/2mc)$  is the Bohr magneton. On the other hand,  $\beta_3$  is the average value of the direction cosine of the net spin direction with respect to the  $z$ -axis, and thus, since the magnetic moment of the electron is opposite to its spin,  $M$ , the  $z$ -component of the net magnetization, is given by  $M = -M_s\beta_3$ . Combining the last four equations and taking  $g \approx 2$ ,

$$\sigma_1 = -\frac{4ec}{m} \left\{ \sum_{m>n} \langle Q_{mn}/(\omega_{mn}^2 - \omega^2) \rangle_{Av} \right\} M, \quad (48)$$

$$\alpha_1 = \frac{2\pi ec}{m} \left\{ \frac{1}{\omega^2} \sum_{m>n} \langle \delta(\omega_{mn} - \omega) Q_{mn} \rangle_{Av} \right\} M. \quad (49)$$

Since  $n, k$  depend only slightly on magnetization, as explained above, it can be seen immediately that the Faraday and Kerr constants are proportional to the net magnetization  $M$  of the specimen.  $\epsilon_F$  is always observed for so small a film width,  $z$ , that it is always proportional to  $\mathcal{G}(N_+ - N_-)$  [see Eq. (39)]. This is what is observed experimentally, as was mentioned in the introduction. (See also footnote 1.)

The proportionality constant depends on frequency, but it is temperature-independent. All temperature dependence of the Faraday and Kerr constants is thus given by  $M(T)$  according to our theory. It should be noted that this is so mainly because, in the calculations for optical and ultraviolet frequencies, the effect of electron-phonon collisions was neglected. The effect of temperature on the electron distribution is certainly negligible. The available experimental evidence<sup>1</sup> indicates that this is indeed the case at these frequencies, the magneto-optic effects under consideration here decreasing slowly at first with rising temperature, much faster as the Curie point is approached, and finally vanishing at the Curie point, much in the same way the  $M$  vs  $T$  curve behaves.

The dispersion of the effects is also given in principle by our formulas, but it is rather difficult to state it explicitly, owing to the complexity of the integrals involved and the lack of accurate knowledge of  $\omega_{mn}(\mathbf{k})$ ,  $Q_{mn}(\mathbf{k})$ . A similar situation exists with the optical constants  $n$  and  $k$ . However, the fact that  $\sigma_1$  depends on  $\omega$  through the expression  $(\omega_{mn}^2 - \omega^2)$  gives rise to the possibility for the Faraday rotation to change sign (an experimentally observed fact) below a certain frequency; whereas an effective magnetic field approach

to the problem gives a dependence like  $(\omega_{mn}^2 - \omega^2)^2$ , which excludes such a possibility.

In order to get a numerical estimate of the order of magnitude of these effects, it is necessary to make further approximations as to the structure of the wave functions and the energy bands. We first approximate the integrals in the expression (26) for  $Q_{mn}(\mathbf{k})$ , the average value of which appears in Eqs. (48) and (49). Introducing the abbreviations

$$\int u_m^* \frac{\partial}{\partial x} u_n d\tau_0 = \langle m | \partial_x | n \rangle, \\ -\frac{i\hbar^2}{4m^2c^2} \int \psi_i^* \left( \frac{\partial V}{\partial x} \frac{\partial}{\partial y} - \frac{\partial V}{\partial y} \frac{\partial}{\partial x} \right) \psi_n d\tau = \langle l | \partial | n \rangle,$$

we have

$$Q_{mn} = 2 \frac{1}{i} \langle m | \partial_y | n \rangle^* \left[ \sum_{l \neq n} \frac{\langle l | \partial | n \rangle}{E_n - E_l} \langle m | \partial_x | l \rangle + \sum_{l \neq m} \frac{\langle l | \partial | m \rangle^*}{E_m - E_l} \langle l | \partial_x | n \rangle \right].$$

Here  $\mathcal{R}$  has been dropped, since the matrix elements of  $\partial_x$ ,  $\partial_y$ , and  $\partial$  are purely imaginary numbers, and thus the whole expression is real. As will be seen below, the most important  $m$ -band is the 4s-band, whereas  $n$  stands for one of the 3d-bands, namely that occupied by most of the magnetic electrons. Thus, in this crude estimate, the second sum compared to the first in the previous expression for  $Q_{mn}$  may be neglected, since  $E_n - E_l$  can become quite small with  $l$  any one of the other overlapping 3d-bands. Furthermore, the terms making the major contribution to the first sum are the ones with  $l$  one of the 3d-bands. Therefore, we shall take

$$Q_{mn} = \frac{2}{\Delta E} \frac{1}{i} \langle m | \partial_y | n \rangle^* \sum_l \langle l | \partial | n \rangle \langle m | \partial_x | l \rangle, \quad (50)$$

where

$$\Delta E = \langle E_n(\mathbf{k}) - E_l(\mathbf{k}) \rangle_{Av}, \quad (51)$$

and  $l$  denotes any one of the 3d-bands not occupied by most of the magnetic electrons (this is the  $n$ -band). The Bloch functions for the 3d-bands have been calculated by Fletcher<sup>16</sup> for nickel on the basis of the tight-binding approximation according to which Bloch functions are constructed as linear combinations of atomic wave functions centered about different lattice points. Since the five 3d-bands overlap, a linear combination of all five 3d-atomic wave functions should be taken for the construction of the corresponding Bloch functions with different coefficients for different values of the wave vector  $\mathbf{k}$ . Even this complicated procedure neglects the overlapping 4s-band. The resulting Bloch wave functions and their  $E$  vs  $\mathbf{k}$  curves are quite complicated. In the order of magnitude estimates for the

<sup>16</sup> G. C. Fletcher, Proc. Phys. Soc. (London) A65, 192 (1952).

matrix elements,  $(l|\Theta|n)$ , simpler expressions for the 3d-band Bloch wave functions will be taken, namely,

$$\psi_n(\mathbf{k}, \mathbf{r}) = N^{-\frac{1}{2}} \sum_{\mathbf{R}} e^{i\mathbf{k} \cdot \mathbf{R}} \phi_n(\mathbf{r} - \mathbf{R}), \quad (52)$$

where  $\phi_n(\mathbf{r})$  is the corresponding atomic wave function and  $\mathbf{R}$  denotes the position of the lattice points. Such an approximation is not likely to introduce an order of magnitude error. Brooks<sup>17</sup> has shown that in such a case

$$(l|\Theta|n) \approx A(l|L_z|n),$$

where  $A$  is the ordinary one-electron spin-orbit interaction parameter for free atoms, and  $(l|L_z|n)$  is the matrix element of  $L_z = (1/i)(x\partial/\partial y - y\partial/\partial x) = (1/i)\partial/\partial\phi$ , with respect to the angular parts of the atomic wave functions  $\phi_l$  and  $\phi_n$ . Most of the magnetic electrons, as is shown by Fletcher,<sup>16</sup> occupy the most energetic states of one of the 3d-bands. It is assumed that all of them are in the  $n$ th band, and that its Bloch wave functions are given by Eq. (52), with  $\phi_n(\mathbf{r}) = yzf(r)$ . This particular choice does not affect the order of magnitude, although it can alter the sign of the matrix element; thus, it is seen that a definite statement cannot be made as to the correct sign of the matrix elements, and, hence, of the final estimates of  $\sigma_1$  and  $\alpha_1$ . Bloch wave functions for the other four 3d-bands are constructed in an analogous manner from the other 3d-atomic wave functions,  $\phi_l(\mathbf{r}) : xyf(r), zxf(r), \frac{1}{2}(x^2 - y^2)f(r), (1/2\sqrt{3}) \times (x^2 + y^2 - 2z^2)f(r)$ .

It is readily found that the only  $\phi_l$  which has a non-vanishing matrix element of  $L_z$  with  $\phi_n$  is the  $zxf(r)$ , and for this case  $(l|L_z|n) = 1/i$ . Thus, neglecting a minus sign, since no faith can be placed in it, Eq. (50) may be written,

$$Q_{mn} = (2/\Delta E)A \langle m|\partial_y|n \rangle^* \langle m|\partial_x|l \rangle.$$

Now, in order to get a rough estimate of the order of magnitude of the matrix elements of  $\partial/\partial x$ ,  $\partial/\partial y$ , they are transformed in the usual manner to matrix elements of  $x$  and  $y$ .

$$\begin{aligned} \langle m|\partial_x|n \rangle &= (m/\hbar^2)(E_n - E_m) \int u_m^* x u_n d\tau_0 \\ &= (m/\hbar^2)(E_n - E_m) \langle m|x|n \rangle, \end{aligned}$$

and similarly for  $\langle m|\partial_y|n \rangle$ . It should be noticed that the free electron approximation of the  $m$ th (4s-band) and higher bands gives zero matrix elements, since  $\langle m|\partial_x|n \rangle = \langle n|\partial_x|m \rangle = 0$  ( $u_m = \text{constant}$  for plane waves). The deviations of the wave functions from plane waves are thus essential for a correct calculation; the almost-free electron approximation usually gives fair estimates. However, in view of the dimensions of the unit cell and the relation  $\int u_m^* u_n d\tau_0 = \delta_{mn}$ , it may be assumed  $\langle m|x|n \rangle$  and  $\langle m|y|n \rangle$  are of the order of  $\sim 10^{-9}$  cm.

<sup>17</sup> H. Brooks, Phys. Rev. 58, 909 (1940).

Thus, Eqs. (48) and (49) may be written as

$$|\sigma_1| \sim 1.0 \times 10^{11} \frac{A(E_n - E_m)(E_l - E_m)}{\Delta E[(E_m - E_n)^2 - \hbar^2 \omega^2]} M,$$

$$|\alpha_1| \sim |\sigma_1/\omega|.$$

(A factor of 10 can easily enter the last expression.) Here all bands higher than the 4s-band are neglected, since for our frequencies they contribute much less on account of the resonant form of the denominator.

From Slater's diagram<sup>5</sup> the energy difference near the top of the magnetic 3d- and 4s-bands is estimated as

$$(E_m - E_n) \approx 4 \text{ ev} = 6.4 \times 10^{-12} \text{ erg.}$$

Also,  $\Delta E \approx 1 \text{ ev} = 1.6 \times 10^{-12} \text{ erg}$ , and therefore  $(E_m - E_l) \approx 5 \text{ ev} = 8.0 \times 10^{-12} \text{ erg}$ . For a light wave of wavelength, say,  $\lambda = 6000 \text{ \AA}$ ,  $\hbar\omega = 3.0 \times 10^{-12} \text{ erg}$ , and

$$|\sigma_1| \sim 1.0 \times 10^{23} AM.$$

Taking, with Brooks, the spin-orbit parameter to be  $A \approx 10^{-13} \text{ erg}$  for nickel and  $A \approx 0.7 \times 10^{-13} \text{ erg}$  for iron, and  $M \approx 500$  gauss for nickel, under saturation conditions at room temperature, and  $M \approx 1700$  gauss for iron, under the same conditions, we finally obtain:

$$\left. \begin{aligned} |\sigma_1| &\sim 5.2 \times 10^{12} \text{ sec}^{-1} \\ |\alpha_1| &\sim 1.6 \times 10^{-3} \end{aligned} \right\} \text{for Ni,} \quad (53)$$

and

$$\left. \begin{aligned} |\sigma_1| &\sim 12.4 \times 10^{12} \text{ sec}^{-1} \\ |\alpha_1| &\sim 4.0 \times 10^{-3} \end{aligned} \right\} \text{for Fe.} \quad (54)$$

These values, along with the remark about the approximation of  $\alpha_1$ , give, e.g., for the Faraday rotation in iron under saturation conditions at room temperature, an angle of  $\sim 280\,000^\circ$  per centimeter.

The experimental work on these effects is not satisfactory, mainly on account of the usual difficulties that beset all optical measurements of solids. The results depend markedly on the method of preparation of the films used. All experimental work up to 1936 has been summarized by Schültz.<sup>1</sup> The most recent measurement of the Faraday rotation in iron is that of König,<sup>18</sup> who measured  $\phi_F$  in saturated iron at room temperature for  $\lambda \sim 6000 \text{ \AA}$  and found it to be ( $\phi_F = 380\,000^\circ$  per centimeter) approximately twice as big as previous measurements had indicated. He was able to show that the oxidation of the films can account for such differences. An analysis of the work of Foote<sup>19</sup> on the Kerr constants and that of Skinner and Tool<sup>20</sup> on the Faraday constants for  $\lambda \sim 6000 \text{ \AA}$  gives the following values for the quantities  $\sigma_1$ ,  $\alpha_1$  within a factor of 2, taking  $n = 2.0$ ,  $k = 3.9$  for nickel, and  $n = 2.4$ ,  $k = 3.3$  for iron (see also

<sup>18</sup> H. König, J. Optik 3, 101 (1948).

<sup>19</sup> P. Foote, Phys. Rev. 34, 96 (1912).

<sup>20</sup> C. A. Skinner and A. Q. Tool, Phil. Mag. 16, 833 (1908).

Darwin<sup>21</sup>)

$$\left. \begin{aligned} \sigma_1 &\cong 3.1 \times 10^{12} \text{ sec}^{-1} \\ \alpha_1 &\cong 1.7 \times 10^{-2} \end{aligned} \right\} \text{for Ni,}$$

and

$$\left. \begin{aligned} \sigma_1 &\cong 30.0 \times 10^{12} \text{ sec}^{-1} \\ \alpha_1 &\cong 4.3 \times 10^{-2} \end{aligned} \right\} \text{for Fe.}$$

Values for different wavelengths in the visible region are not very much different. In view of the approximations

<sup>21</sup> C. G. Darwin, Proc. Roy. Soc. (London) A151, 512 (1935).

necessary to get the theoretical estimates, Eqs. (53) and (54), the comparison is satisfactory.

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Neutron Damage to the Structure of Vitreous Silica

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The x-ray diffraction pattern of vitreous silica has been investigated before and after exposure to neutrons. Small, but significant, changes are observed. A relation between these changes and those caused by neutron damage to crystalline forms of silica is suggested.

INTRODUCTION

SEVERAL observations have been reported on radiation damage in silica, both crystalline and vitreous. Wittels and Sherrill,<sup>1</sup> for instance, found that fast-neutron irradiation of a total of  $\sim 2 \times 10^{20}$  neutrons/cm<sup>2</sup> caused all crystalline forms to become apparently glassy with a density of 2.26. Vitreous silica also reached this density. Primak, Fuchs, and Day<sup>2</sup> report an increase in density and refractive index of vitreous silica. No extended study of the x-ray diffraction pattern of irradiated vitreous silica has been published. In view of the tendency of all crystalline silica materials to become glassy with a common density and of vitreous silica to increase in density to the same value, it is of interest to examine the diffraction patterns of these damaged materials in some detail. In this report, the effect of neutron irradiation on the x-ray intensity curve of vitreous silica is discussed.

EXPERIMENTAL

Vitreous silica of high purity<sup>3</sup> was irradiated by neutrons at the Materials Testing Reactor to an exposure of  $nvt \sim 2 \times 10^{20}$ , at a temperature of about 50°C. The specimen showed no visual change other than a slight violet discoloration. The density was found to have increased from 2.21 to 2.25, in excellent agreement with previous work. Also confirming earlier

observations, the refractive index (sodium D line) increased from  $1.45706 \pm 0.00004$  to  $1.46687 \pm 0.00010$ .

The x-ray intensity curves for both unirradiated and irradiated material were obtained using a spectrometer and Geiger counter. Values were measured at intervals of one degree  $2\theta$  or less, except at high angles where larger intervals were used. Readings were taken to  $\sin\theta/\lambda = 0.700$ , above which point all scattering is essentially incoherent. Filtered copper and molybdenum radiations were used. Since the radiation was not strictly monochromatic, no Fourier analysis has been made of the data. It is hoped that in the future, data suitable for such analysis can be obtained. The intensities were measured by using the fixed count method in which the time to register a fixed number of counts is recorded. The probable error is less than one percent.

Spectroscopic analysis of the unirradiated material is given in Table I. The results may be in error by a factor of two or three.

TABLE I. Spectroscopic analysis of vitreous silica.

Constituent	Abundance <sup>a</sup> (parts per million)
V	400
Cr	200
Ti	120
Mn	60
Cu	16
Mg	6
Al	6
B	<<200

\* May be in error by a factor of two or three.

\* The Knolls Atomic Power Laboratory is operated by the General Electric Company under contract with the United States Atomic Energy Commission.

<sup>1</sup> M. Wittels and F. A. Sherrill, Phys. Rev. 93, 1117 (1953).

<sup>2</sup> Primak, Fuchs, and Day, Phys. Rev. 92, 1064 (1953).

<sup>3</sup> Provided by Corning Glass Works, Corning, New York.