Size and Surface Effects on the Magneto-Optical Properties of Metallic Thin Films

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The variational principle of Jones and Sondheimer is applied to a thin film to discuss its reflective and transmissive properties in a constant magnetic wave at optical frequencies. First, generalized admittance tensors are defined via the variational integral, and the reflection and transmission tensors found in terms of these. Then the reflectivity, transmittivity, absorptivity, and Voigt effect are treated for a simple case with \mathbf{B}_0 parallel to the surfaces of the film, and the effect of surface collisions on these is discussed.

1. INTRODUCTION

IN a pure metal at low temperatures, collisions of electrons in the bulk of the metal are infrequent and the absorption of energy from electromagnetic waves at optical frequencies due to such collisions is consequently small. Under these conditions the absorption due to surface collisions becomes important and even dominant. Holstein¹ showed that this effect is considerably greater for diffuse than for specular surface scattering of electrons, and both he and Dingle² obtained expressions for the absorptivity of a semi-infinite medium in the absence of a magnetic field. (Recently, Fedders³ has obtained more general results for a rough surface.) Later, Jones and Sondheimer⁴ obtained expressions for the absorptivity of a semi-infinite medium, with a magnetic field parallel to the surface, for both diffuse and specular scattering. In order to do this they used a variational principle, in which a variational integral I_{ω} is made stationary with respect to small changes in the frequency component of field \mathbf{E}_{ω} to give the surface admittance Y_{ω} . Here, their method will be extended to the case of a slab of thickness d, which will later be taken to be small compared with other parameters which appear. A preliminary discussion of this problem has been published,⁵ but for completeness some of the results are repeated here.

Jones and Sondheimer¹ reformulated Maxwell's equations for a finite medium in which linear but nonlocal current-field and polarization-field relations obtain as the vanishing of the first variation of I_{ω} , given by

$$I_{\omega} = \int_{S} \mathbf{E}_{\omega}^{\dagger} \times (\nabla \times \mathbf{E}_{\omega}) \cdot \mathbf{dS}, \qquad (1.1)$$

where $\mathbf{E}_{\omega}(\mathbf{r})e^{i\omega t}$ is the electric field, the conjugate field $\mathbf{E}_{\omega}^{\dagger}$ is that obtained by reversing the magnetic field, and the integral is over the surface of the medium. Their

- ¹ T. Holstein, Phys. Rev. **88**, 1427 (1952). ² R. B. Dingle, Physica **19**, 311 (1953); **19**, 348 (1953); **19**, 729 (1953); **19**, 1187 (1953).
- ⁵ P. A. Fedders, Phys. Rev. 181, 1053 (1969).
 ⁴ M. C. Jones and E. H. Sondheimer, Proc. Roy. Soc. (London) A278, 256 (1964). ⁵ L. E. G. Ah-Sam and M. C. Jones, Alta Frequenza 38, 20
- (1969).

result is obtained by writing

$$I_{\omega} = \int_{V} \nabla \cdot \left[\mathbf{E}_{\omega}^{\dagger} \times (\nabla \times \mathbf{E}_{\omega}) \right] d\mathbf{x}, \qquad (1.2)$$

the integral being taken over the volume of the medium, and then

$$I_{\omega} = \int_{V} (\nabla \times \mathbf{E}_{\omega}^{\dagger} \cdot \nabla \times \mathbf{E}_{\omega} + i\omega\mu_{0}\mathbf{E}_{\omega}^{\dagger} \cdot \mathbf{J}_{\omega} - \omega^{2}\mu_{0}\mathbf{E}_{\omega}^{\dagger} \cdot \mathbf{D}_{\omega})d\mathbf{x}, \quad (1.3)$$

where

$$\nabla \times (\nabla \times \mathbf{E}_{\omega}) = -i\omega\mu_0 \mathbf{J}_{\omega} + \omega^2 \mu_0 \mathbf{D}_{\omega}$$
(1.4)

has been used. Varying (1.3),

$$\delta I_{\omega} = \int_{V} \left[\nabla \times \mathbf{\delta} \mathbf{E}_{\omega}^{\dagger} \cdot \nabla \times \mathbf{E}_{\omega} + \nabla \times \mathbf{E}_{\omega}^{\dagger} \cdot \nabla \times \mathbf{\delta} \mathbf{E}_{\omega} + i\omega\mu (\mathbf{\delta} \mathbf{E}_{\omega}^{\dagger} \cdot \mathbf{J}_{\omega} + \mathbf{E}_{\omega}^{\dagger} \cdot \mathbf{\delta} \mathbf{J}_{\omega}) - \omega^{2}\mu_{0} (\mathbf{\delta} \mathbf{E}_{\omega}^{\dagger} \cdot \mathbf{D}_{\omega} + \mathbf{E}_{\omega}^{\dagger} \cdot \mathbf{\delta} \mathbf{D}_{\omega}) \right] d\mathbf{x}, \quad (1.5)$$

and, so long as \mathbf{J}_{ω} , \mathbf{D}_{ω} satisfy linear relations of the form

$$\mathbf{J}_{\omega} = \int_{V} \mathbf{L}_{J}(\mathbf{x}, \mathbf{x}') \cdot \mathbf{E}_{\omega}(\mathbf{x}') d\mathbf{x}', \qquad (1.6)$$
$$\mathbf{D}_{\omega} = \int_{V} \mathbf{L}_{D}(\mathbf{x}, \mathbf{x}') \cdot \mathbf{E}_{\omega}(\mathbf{x}') d\mathbf{x}',$$

in which $L_{J,D}$ satisfies the generalized Onsager relation

$$\mathbf{L}_{J,D}{}^{T}(\mathbf{x},\mathbf{x}') = \mathbf{L}_{J,D}{}^{\dagger}(\mathbf{x}',\mathbf{x}), \qquad (1.7)$$

$$\int_{V} \mathbf{E}_{\omega}^{\dagger} \cdot \mathbf{\delta} \mathbf{J}_{\omega} d\mathbf{x} = \int_{V} \mathbf{\delta} \mathbf{E}_{\omega}^{\dagger} \cdot \mathbf{J}_{\omega}, \, d\mathbf{x} \,, \qquad (1.8)$$

and similarly for \mathbf{D}_{ω} . Thus (1.5) can be written

$$\delta I_{\omega} = \int_{V} \{ \delta \mathbf{E}_{\omega}^{\dagger} \cdot [\nabla \times (\nabla \times \mathbf{E}_{\omega}) + i \omega \mu_{0} \mathbf{J}_{\omega} - \omega^{2} \mu_{0} \mathbf{D}_{\omega}] \\ + \delta \mathbf{E}_{\omega} \cdot [\nabla \times (\nabla \times \mathbf{E}_{\omega}^{\dagger}) + i \omega \mu_{0} \mathbf{J}_{\omega}^{\dagger} - \omega^{2} \mu_{0} \mathbf{D}_{\omega}^{\dagger}] \} d\mathbf{x} \\ + \int_{S} [\delta \mathbf{E}_{\omega}^{\dagger} \times (\nabla \times \mathbf{E}_{\omega}) + \delta \mathbf{E}_{\omega} \times (\nabla \times \mathbf{E}_{\omega}^{\dagger})] \cdot \mathbf{dS}. \quad (1.9)$$

1 3896 From (1.4) and $(1.4)^{\dagger}$ the volume integral in (1.9)vanishes, and, if the components of $\delta E_{\omega}^{\dagger}$ and δE_{ω} tangential to the surface are zero, (1.9) gives

$$\delta I_{\omega} = 0. \tag{1.10}$$

2. REFLECTION AND TRANSMISSION TENSORS IN TERMS OF GENERALIZED ADMITTANCE TENSORS

For the case where V is the region between the planes z=0 and z=d (x, y, z being rectangular Cartesian coordinates), (1.4) is slightly modified by carrying out a partial Fourier transformation $F(\mathbf{r}) \rightarrow \mathfrak{F}(\mathbf{s}, z)$, where $\mathbf{s} = (s_x, s_y)$, and

$$F(\mathbf{r}) = (1/2\pi) \int \mathfrak{F}(\boldsymbol{s}, z) e^{-i(s_x x + s_y y)} d\boldsymbol{s}. \qquad (2.1)$$

Equations (1.10) and (1.4) now give

$$\delta \mathcal{J}_{\omega} = 0, \qquad (2.2)$$

with

$$\mathfrak{s}_{\omega} = \{ \mathfrak{S}_{\omega}^{\dagger}(d) \times [\nabla_{0} \times \mathfrak{S}_{\omega}(d)] - \mathfrak{S}_{\omega}(0) \times [\nabla_{0} \times \mathfrak{S}_{\omega}(0)] \}_{z}, \tag{2.3}$$

$$= \int_{0}^{d} [\nabla_{0} \times \mathfrak{S}_{\omega}(z) \cdot \nabla_{0} \times \mathfrak{S}_{\omega}^{\dagger}(z) + i\omega\mu_{0} \mathfrak{S}_{\omega}^{\dagger}(z) \cdot \mathfrak{F}_{\omega}(z) - \omega^{2}\mu_{0} \mathfrak{S}_{\omega}^{\dagger}(z) \cdot \mathfrak{F}_{\omega}(z)] dz, \tag{2.4}$$

where

and

$$\mathfrak{G}_{\omega}(z) = \mathfrak{G}_{\omega}(\boldsymbol{s}, z), \quad \mathfrak{G}_{\omega}^{\dagger} = \mathfrak{G}_{\omega}^{\dagger}(-\boldsymbol{s}, z),$$
$$\nabla_{0} = (i\boldsymbol{s}, \partial/\partial z), \quad \nabla_{0}^{\dagger} = (-i\boldsymbol{s}, \partial/\partial z).$$

For the case where the field in the medium is produced by a monochromatic plane wave incident on the surface z=0, and with electric field $ie^{i(\omega t-q \cdot x)}$, then s=q, where $q = (q_x, q_y)$. The boundary condition on $\delta \mathbf{E}, \, \delta \mathbf{E}^{\dagger}$ requires

$$\delta \mathcal{E}_{\alpha}(0) = \delta \mathcal{E}_{\alpha}(d) - \delta \mathcal{E}_{\alpha}^{\dagger}(0) = \delta \mathcal{E}_{\alpha}^{\dagger}(d) = 0 \quad (\alpha = x, y). \quad (2.5)$$

Equation (2.5) indicates that \mathcal{I} , given by (2.3), (2.4), is a quadratic function of $\mathcal{E}(0)$ and $\mathcal{E}(d)$. This may be seen explicitly by defining tensors X, X', Y, Y' by the equations

$$[\nabla_0 \times \mathfrak{G}(0)]_x = \sum_{\beta = x, y} [Y_{y\beta} \mathcal{E}_{\beta}(0) + X_{y\beta} \mathcal{E}_{\beta}(d)], \qquad (2.6)$$

$$\left[\nabla_{0} \times \mathfrak{G}(0)\right]_{y} = -\sum_{\beta=x,y} \left[Y_{x\beta} \mathcal{E}_{\beta}(0) + X_{x\beta} \mathcal{E}_{\beta}(d)\right], \quad (2.7)$$

$$\left[\nabla_{0} \times \mathfrak{G}(d)\right]_{x} = -\sum_{\beta=x,y} \left[X_{y\beta}' \mathcal{E}_{\beta}(0) + Y_{y\beta}' \mathcal{E}_{\beta}(d)\right], \quad (2.8)$$

$$[\nabla_0 \times \mathfrak{G}(d)]_y = \sum_{\beta=x,y} [X_{x\beta}' \mathcal{E}_{\beta}(0) + Y_{x\beta}' \mathcal{E}_{\beta}(d)].$$
(2.9)

Using (2.3) \mathcal{I} may now be written

$$\begin{split} \mathcal{I} &= \sum_{\alpha,\beta=x,y} \mathcal{E}_{\alpha^{\dagger}}(d) Y_{\alpha\beta}' \mathcal{E}_{\beta}(d) + \mathcal{E}_{\alpha^{\dagger}}(d) X_{\alpha\beta}' \mathcal{E}_{\beta}(0) \\ &+ \mathcal{E}_{\alpha^{\dagger}}(0) X_{\alpha\beta} \mathcal{E}_{\beta}(d) + \mathcal{E}_{\alpha^{\dagger}}(0) Y_{\alpha\beta} \mathcal{E}_{\beta}(0) \,. \end{split}$$
(2.10)

Since, as is apparent from (1.6)-(1.8) and (2.4), \mathcal{G} is symmetric in \mathcal{E}^{\dagger} and \mathcal{E} , it follows that

$$\mathbf{X}^{\prime T} = \mathbf{X}^{\dagger}, \quad \mathbf{Y}^{T} = \mathbf{Y}^{\dagger}, \quad \mathbf{Y}^{\prime T} = \mathbf{Y}^{\prime \dagger}.$$
 (2.11)

For the case of a semi-infinite medium $\mathcal{E}^{\dagger}(d), \mathcal{E}(d) \to 0$, and then \mathbf{Y} is just the surface admittance tensor; (2.10) provides a generalization of that case in a form convenient for obtaining the reflection and transmission tensors **R** and **T** defined by

$$\boldsymbol{r} = \boldsymbol{\mathsf{R}} \cdot \boldsymbol{i}, \qquad (2.12)$$

$$t = \mathbf{T} \cdot \mathbf{i}, \qquad (2.13)$$

where $\mathbf{r}e^{i(\omega t-\mathbf{q'}\cdot\mathbf{x})}$ and $\mathbf{t}e^{i(\omega t-\mathbf{q}\cdot\mathbf{x})}$, with $\mathbf{q'}=(q_x, q_y-q_z)$ are, respectively, the reflected and transmitted electric fields. Since the boundary conditions at the surfaces are

$$i_{\alpha} + r_{\alpha} = \mathcal{E}_{\alpha}(0), \qquad (2.14)$$

$$t_{\alpha} = \mathcal{E}_{\alpha}(d) , \qquad (2.15)$$

$$-i[\mathbf{q}\times\mathbf{i}+\mathbf{q}'\times\mathbf{r}]_{\alpha}=[\nabla_{0}\times\mathfrak{G}(0)]_{\alpha},\qquad(2.16)$$

$$-i[\mathbf{q} \times \mathbf{t}]_{\alpha} = [\nabla_0 \times \mathfrak{G}(d)]_{\alpha}, \quad (\alpha = x, y) \quad (2.17)$$

and also

or

$$\mathbf{r} = \mathbf{a} \cdot \mathbf{r} = \mathbf{a} \cdot \mathbf{f} + \mathbf{a} \cdot \mathbf{f} = 0 \tag{2.10}$$

(2.18)

$$\boldsymbol{q} \cdot \boldsymbol{r} - \boldsymbol{q}_z \boldsymbol{r}_z = \boldsymbol{q} \cdot \boldsymbol{\iota} + \boldsymbol{q}_z \boldsymbol{\iota}_z = 0. \tag{2.19}$$

Equations (2.6)–(2.9) together with (2.12), (2.13) give

 $\mathbf{q} \cdot \mathbf{i} = \mathbf{q}' \cdot \mathbf{r} = \mathbf{q} \cdot \mathbf{t} = 0$,

 $(\mathbf{I} + \mathbf{Q}) \cdot \mathbf{T} = (i/q_z) [\mathbf{X}' \cdot (\mathbf{I} + \mathbf{R}) + \mathbf{Y}' \cdot \mathbf{T}]$ (2.20)

$$(\mathbf{I}+\mathbf{Q})\cdot(\mathbf{I}-\mathbf{R}) = (-i/q_z)[\mathbf{Y}\cdot(\mathbf{I}+\mathbf{R})+\mathbf{X}\cdot\mathbf{T}], \quad (2.21)$$

where $Q_{\alpha\beta} = q_{\alpha}q_{\beta}/q_{z}^{2}$. Solving (2.20) and (2.21) for **T** and **R**.

$$\mathbf{T} = \left[\left(\frac{i}{q_z} \mathbf{X}' \right)^{-1} \cdot \left(\mathbf{I} + \mathbf{Q} - \frac{i}{q_z} \mathbf{Y}' \right) - \left(\mathbf{I} + \mathbf{Q} - \frac{i}{q_z} \mathbf{Y} \right)^{-1} \cdot \left(\frac{i}{q_z} \mathbf{X} \right) \right]^{-1} \left[\mathbf{I} + \left(\mathbf{I} + \mathbf{Q} - \frac{i}{q_z} \mathbf{Y} \right)^{-1} \cdot \left(\mathbf{I} + \mathbf{Q} + \frac{i}{q_z} \mathbf{Y} \right) \right]$$
(2.22)
and

$$\mathbf{R} = \left[\left(\frac{i}{q_z} \mathbf{X} \right)^{-1} \cdot \left(\mathbf{I} + \mathbf{Q} - \frac{i}{q_z} \mathbf{Y} \right) - \left(\mathbf{I} + \mathbf{Q} - \frac{i}{q_z} \mathbf{Y}' \right)^{-1} \cdot \left(\frac{i}{q_z} \mathbf{X}' \right) \right]^{-1} \\ \times \left[\left(\mathbf{I} + \mathbf{Q} - \frac{i}{q_z} \mathbf{Y}' \right)^{-1} \cdot \left(\frac{i}{q_z} \mathbf{X}' \right) + \left(\frac{i}{q_z} \mathbf{X} \right)^{-1} \cdot \left(\mathbf{I} + \mathbf{Q} + \frac{i}{q_z} \mathbf{Y} \right) \right]. \quad (2.23)$$

Once X, X', Y, Y' are known, (2.22) and (2.23) allow all the properties of the reflected and transmitted waves to be found. In the following sections it is shown how the variational principle may be used to obtain approximate expressions for these.

3. CURRENT-FIELD RELATION FROM BOLTZMANN EQUATION

In order to apply the variational principle, specific forms for the nonlocal tensor $L(\mathbf{x},\mathbf{x}')$ appearing in (1.6) are required. In the simple case of a degenerate quasifree-electron gas with a constant lattice permittivity \mathscr{E} is considered, the current density can be found by solving the Boltzmann equation. Jones and Sondheimer⁴ found a general solution to this for an arbitrarily shaped medium and their solution gives the following results for a film of thickness d with a constant magnetic field \mathbf{B}_0 parallel to its surfaces.

The distribution function $f = f_0 + g(\mathbf{k}, z)$, where f_0 is the equilibrium (Fermi) distribution function. Coordinates $\mathcal{E}_{\mathbf{k}}$, k_h , and ϕ are used in \mathbf{k} space where $\mathcal{E}_{\mathbf{k}} = (\hbar^2/2m^*)\mathbf{k}^2$, k_h is the component of \mathbf{k} parallel to the magnetic field, and ϕ is the azimuthal angle about the field direction. The cyclotron frequency eB_0/m^* is denoted by ω_0 , and a constant time of relaxation τ is assumed. For $(2v/\omega_0) \sin\theta < d$, and

$$-1 + \frac{\omega_0(d-z)}{v\sin\theta} > \cos\phi > 1 - \frac{\omega_0 z}{v\sin\theta},$$
$$g(z,\phi) = \frac{e}{\omega_0} \frac{1}{e^{2\pi\gamma} - 1} \int_{\phi}^{\phi+2\pi} e^{\gamma(\phi'-\phi)} \times \left[z + \frac{v}{\omega_0} \sin\theta(\cos\phi - \cos\phi') \right] \cdot v(\phi') d\phi'; \quad (3.1)$$

otherwise,

for

$$g(z,\phi) = \frac{e}{\omega_0} \int_{\phi_0}^{\phi} e^{\gamma (\phi' - \phi)} \\ \times \mathfrak{G}\left[z + \frac{v}{\omega_0} \sin\theta (\cos\phi - \cos\phi')\right] \cdot \mathbf{v}(\phi') d\phi', \quad (3.2)$$

where ϕ_0 is the greatest value of $\psi(\langle \phi \rangle)$ satisfying

$$z + (v/\omega_0)\sin\theta(\cos\phi - \cos\psi) = z_s \qquad (3.3)$$

(3.4)

with $z_s=0$ or d. In these expressions **v** is the electron velocity on the Fermi surface which is given by $k^2 = k_0^2$; $k_h = k_0 \cos\theta$; $\gamma = (1 + i\omega\tau)/(\omega_0\tau)$; in deriving (3.2), diffuse surface scattering has been assumed. Equation (3.3) gives

$$\phi_0 = \cos^{-1}(\cos\phi + \omega_0 z/v \sin\theta)$$
,

 $(2v/\omega_0)\sin\phi < d$ and $\cos\phi < 1 - \omega_0 z/v \sin\theta$, or for

$$(2v/\omega_0)\sin\theta < d$$

and

$$\cos^{-1}(1-\omega_0 z/v \sin\theta) < \phi < 2\pi$$
$$-\cos^{-1}[-1+\omega_0(d-z)/v \sin\theta];$$
$$\phi_0 = -\cos^{-1}[\cos\phi - \omega_0(d-z)/v \sin\theta], \text{ if } \phi < \pi$$
$$\phi_0 = 2\pi - \cos^{-1}[\cos\phi - \omega_0(d-z)/v \sin\theta], \text{ if } \phi > \pi \quad (3.5)$$
for

$$(2v/\omega_0)\sin\theta < d$$
 and $\cos\phi > -1 + \omega_0(d-z)/v\sin\theta$,

or for

$$\begin{array}{l} (2v/\omega_0)\sin\theta > d \quad \text{and} \quad 0 < \phi < \cos^{-1}(1-\omega_0 z/v\sin\theta) ,\\ 2\pi - \cos^{-1}[-1+\omega_0(d-z)/v\sin\theta] < \phi < 2\pi . \end{array}$$

It can be seen from these expressions that there are two distinct cases: (a) $d>2v/\omega_0$ and (b) $d<2v/\omega_0$. In case (a) complete electron orbits are possible in the bulk of the medium, giving rise to a solution of the form (3.1). Although case (b) can be treated by the same methods used here for case (a), it is even more laborious and only the latter case will be considered here.

For $d > 2v/\omega_0$ then, the current density is given by

$$\mathfrak{F}(z) = \frac{em^2v^2}{4\pi^3\hbar^3} \int_0^\pi \sin\theta d\theta \int_0^{2\pi} \mathbf{n}(\theta, \phi) g(z, \theta, \phi) d\phi \,, \quad (3.6)$$

g being given by (3.1) or (3.2) in the appropriate region and **n** being a unit vector normal to **v**. Since $\Im(z)$ appears only in a scalar product with $\mathfrak{G}^{\dagger}(z)$ which is integrated [Eq. (2.4)], there is no need to display \mathfrak{L} explicitly; it can, however, be shown to satisfy conditions corresponding to (1.7).

4. EVALUATION OF VARIATION INTEGRAL WITH EXPONENTIALLY VARYING FIELD

Before proceeding to evaluate the variational integral or a specific form of trial field, it may be noted that, if the z axis is a twofold axis of symmetry (a condition certainly satisfied for quasifree electrons), the variational integral can be simplified for the case where \mathbf{B}_0 is parallel to the surfaces of the slab. In this case

$$\mathcal{E}_{\alpha}^{\dagger} = \mathcal{E}_{\alpha}(\alpha = x, y), \quad \mathcal{E}_{z}^{\dagger} = -\mathcal{E}_{z},$$

and consequently $\mathfrak{G}^{\dagger} \cdot \mathfrak{J} = \mathcal{E} \cdot \mathfrak{g} - \mathcal{E}_z \mathfrak{g}_z$. This is valid for arbitrary angles of incidence, and allows \mathfrak{I} to be varied by varying \mathfrak{G} . The fact that variations in \mathfrak{G}^{\dagger} and \mathfrak{G} are no longer independent does not affect the validity of (2.2) as may easily be seen from (1.9).

Considering, then, the case in which \mathbf{B}_0 is parallel to the surfaces of the slab,

$$\vartheta = \int_{0}^{d} \left\{ (\mathscr{E}_{x}') + (\mathscr{E}_{y}')^{2} - 2i(q_{x}\mathscr{E}_{x}' + q_{y}\mathscr{E}_{y}')\mathscr{E}_{z} - (q_{x}^{2} + q_{y}^{2})\mathscr{E}_{z}^{2} + (q_{x}\mathscr{E}_{y} - q_{y}\mathscr{E}_{x})^{2} + i\omega\mu_{0}[\mathscr{E}_{x}\mathscr{G}_{x} + \mathscr{E}_{y}\mathscr{G}_{y} - \mathscr{E}_{z}\mathscr{G}_{z}] - \omega^{2}\mu_{0}\mathscr{E}[\mathscr{E}_{x}^{2} + \mathscr{E}_{y}^{2} - \mathscr{E}_{z}^{2}] \right\} dz. \quad (4.1)$$

For simplicity, only the case of normal incidence will be considered here, and then the x and y axes will be principal axes for the generalized admittance tensors introduced in Sec. 2, and the two cases, where the electric field is parallel to \mathbf{B}_0 and where it is perpendicular to \mathbf{B}_0 , can be considered separately. These two cases will be referred to as longitudinal and transverse, respectively, and subscripts L and T will be used to distinguish quantities where necessary.

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The variational method consists of taking a trial function for $\mathfrak{G}(z)$ which is linear in $\mathscr{E}(0)$ and $\mathscr{E}(d)$, and involves a number of parameters, say, $\lambda_1, \ldots, \lambda_N$. On substitution into (4.1), \mathscr{I} becomes quadratic in $\mathscr{E}(0)$ and $\mathscr{E}(d)$, and depends on $\lambda_1, \ldots, \lambda_N$. Equation (2.2) now gives the best values of $\lambda_1, \ldots, \lambda_N$ as the solutions to

$$\frac{\partial \mathcal{I}}{\partial \lambda_i} = 0 (i = 1, \dots, N). \tag{4.2}$$

These values are then replaced in (4.1) and approximations for $\mathbf{X}, \mathbf{X}', \mathbf{Y}, \mathbf{Y}'$ obtained by comparing coefficients between the resulting expression and (2.10). The classical (constant local conductivity) theory has exact solutions in which the fields vary exponentially with z. Jones and Sondheimer⁴ found that an exponential trial function was adequate for the semi-infinite case, and it seems reasonable to use similar trial functions here also. However, the results thus obtained in this section and Sec. 5 suggest that these trial functions may not be good enough when the film thickness is of the same order as the cyclotron diameter. This will be discussed further in Sec. 5.

For the longitudinal case the trial function is

$$\mathcal{E}_x(z) = \alpha e^{-\sigma z} + \beta e^{-\sigma (d-z)}, \qquad (4.3)$$

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where the boundary condition gives, in a self-explanatory notation,

$$\alpha = \frac{\mathcal{E}_0 - e^{-\sigma d} \mathcal{E}_d}{1 - e^{-2\sigma d}}, \quad \beta = \frac{\mathcal{E}_d - \mathcal{E}_0 e^{-\sigma d}}{1 - e^{-2\sigma d}}.$$
 (4.4)

For the transverse case, $\mathcal{E}_y(z)$ is represented in the form found in (4.3) and the Hall field $\mathcal{E}_z(z)$ by

$$\mathcal{E}_{z}(z) = (\chi_{1}\mathcal{E}_{0} + \chi_{2}\mathcal{E}_{d})e^{-\sigma z} + (\chi_{3}\mathcal{E}_{0} + \chi_{4}\mathcal{E}_{d})e^{-\sigma (d-z)}. \quad (4.5)$$

In these expressions σ and χ_1, \ldots, χ_4 are to be varied. For the longitudinal case immediately, and for the transverse case on eliminating χ_1, \ldots, χ_4 and using (4.2) and (4.1), gives

$$\vartheta = \frac{1}{2} \sigma(\alpha^2 + \beta^2) (1 - e^{-2\sigma d}) - 2\alpha \beta \sigma^2 d e^{-\sigma d} + c [(\alpha^2 + \beta^2) A(\sigma) + 2\alpha \beta B(\sigma)], \quad (4.6)$$

where $c = Ne^2 \mu_0 / m$ and

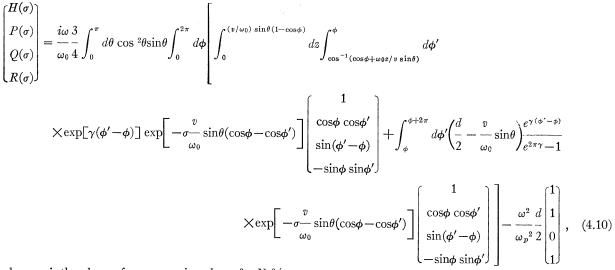
$$A^{L}(\sigma) = G(\sigma) + e^{-2\sigma d}G(-\sigma), \quad B^{L}(\sigma) = e^{-\sigma d}[H(\sigma) + H(-\sigma)], \tag{4.7}$$

$$A^{T}(\sigma) = L(\sigma) + e^{-2\sigma d}L(-\sigma) - \frac{1}{4} \{ [N(\sigma) + e^{-2\sigma d}N(-\sigma)]^{2} - e^{-2\sigma d}[R(\sigma) + R(-\sigma)]^{2} \}^{-1} \\ \times \{ [N(\sigma) + e^{-2\sigma d}N(-\sigma)][(M(\sigma) + e^{-2\sigma d}M(-\sigma))^{2} + e^{-2\sigma d}(Q(\sigma) + Q(-\sigma))^{2}] \\ -2e^{-2\sigma d}[M(\sigma) + e^{-2\sigma d}M(-\sigma)][Q(\sigma) + Q(-\sigma)][R(\sigma) + R(-\sigma)] \}, \qquad (4.8)$$

$$B^{T}(\sigma) = e^{-\sigma d}[P(\sigma) + e^{-2\sigma d}P(-\sigma)] - \frac{1}{4} \{ e^{-2\sigma d}[R(\sigma) + R(-\sigma)]^{2} - [N(\sigma) + e^{-2\sigma d}N(-\sigma)]^{2} \}^{-1} \\ \times \{ e^{-\sigma d}[R(\sigma) + R(-\sigma)][(M(\sigma) + e^{-2\sigma d}M(-\sigma))^{2} + e^{-2\sigma d}(Q(\sigma) + Q(-\sigma))^{2}] \\ -2e^{-\sigma d}[M(\sigma) + e^{-2\sigma d}M(-\sigma)][Q(\sigma) + Q(-\sigma)][N(\sigma) + e^{-2\sigma d}N(-\sigma)] \}. \tag{4.8}$$

In (4.7) and (4.8) G, H, L, M, N, P, Q, and R are given by

$$\begin{bmatrix} G(\sigma) \\ L(\sigma) \\ M(\sigma) \\ N(\sigma) \end{bmatrix} = \frac{i\omega}{\omega_0} \frac{3}{4} \int_0^{\pi} d\theta \cos^2\theta \sin\theta \int_0^{2\pi} d\phi \Biggl\{ \int_0^{(v/\omega_0) \sin\theta(1-\cos\phi)} dz \ e^{-2\sigma z} \int_{\cos^{-1}(\cos\phi+\omega_0 z/v\sin\theta)}^{\phi} d\phi' \exp[\gamma(\phi'-\phi)] \\ \times \exp\left[-\sigma \frac{v}{\omega_0} \sin\theta(\cos\phi-\cos\phi') \right] \Biggl[\frac{1}{\cos\phi\cos\phi'} \frac{1}{\sin(\phi'-\phi)} \\ -\sin\phi\sin\phi' \right] + \frac{1}{2\sigma} \int_{\phi}^{\phi+2\pi} d\phi' \frac{e^{\gamma(\phi'-\phi)}}{e^{2\pi\gamma}-1} \\ \times \exp\left[-\sigma \frac{v}{\omega_0} \sin\theta(2-\cos\phi-\cos\phi') \right] \Biggl[\frac{1}{\cos\phi\cos\phi'} \frac{1}{\sin(\phi'-\phi)} \\ -\sin\phi\sin\phi' \right] \Biggr\} - \frac{\omega^2}{\omega_p^2} \frac{1}{2\sigma} \begin{bmatrix} 1 \\ 0 \\ 1 \end{bmatrix}, \quad (4.9)$$



where ω_p is the plasma frequency given by $\omega_p^2 = N e^2 / m \epsilon$.

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5. EVALUATION OF GENERALIZED ADMITTANCES AT OPTICAL FREQUENCIES

At sufficiently high frequencies the solution for σ obtained by making (4.6) stationary is such that the exponentials in (4.9) and (4.10) can be expanded in powers of $\sigma(v/\omega_0)$; typically, this requires frequencies in the infrared or higher. In carrying out such an expansion there are two cases to be distinguished—(a) $d \gg r_0$, (b) $d \sim r_0$, where $r_0 = v/\omega_0$. In the first case the exponentials $e^{-\sigma d}$ are not expanded, while in case (b) a consistent treatment requires them to be expanded also. For case (a) the absorption is much the same as that for the semi-infinite case considered by Jones and Sondheimer^{4,6,7} but slightly modified by factors arising from terms involving $e^{-\sigma d}$; in particular, the zeroth-order approximation for σ is just the classical value in both cases. For case (b), however, the lowest-order terms in the expansion of \mathcal{G} are independent of σ and the nexthigher-order terms give rise to an approximation for σ which is not independent of \mathcal{E}_0 and \mathcal{E}_d . This latter effect is presumably due to the inadequacy of the exponential (4.3) and (4.5) and suggests that some improvement of the results obtained here could be obtained by use of an improved trial function. Nevertheless, as the results shown here are derived from the zeroth-order term and so do not involve σ , it is reasonable to suppose that even if their exact form is incorrect their order of magnitude is probably right, and they are used to illustrate the method which, clearly, can be equally well applied to improved results.

For the case
$$d \gg r_0$$
, it may first be noted that if $G(\sigma)$ is expanded in the form

$$G(\sigma) = g_0/2\sigma + g_1r_0 + g_2\sigma r_0^2 + \cdots, \qquad (5.1)$$

and $H(\sigma)$ in the form

$$H(\sigma) = h_0 + h_1 \sigma r_0 + h_2 (\sigma r_0)^2 + \cdots, \qquad (5.2)$$

then

(

$$h_0 = \frac{1}{2} dg_0 + g_1 r_0, \qquad (5.3)$$

with similar relations between corresponding terms for P, L; Q, M; R, N. Expanding the terms in the square bracket in (4.6) in powers of σr_0 , and treating σd as of order $(\sigma r_0)^0$, keeping terms up to order $(\sigma r_0)^1$ gives

$$\begin{aligned} \alpha^{2} + \alpha^{2} A\left(\sigma\right) + 2\alpha\beta B(\sigma) \\ &= \frac{1}{2}\sigma(\alpha^{2} + \beta^{2})(1 - e^{-2\sigma d}) - 2\alpha\beta\sigma^{2}de^{-\sigma d} \\ &+ c\left[(\alpha^{2} + \beta^{2})(g_{0}/2\sigma)(1 - e^{-2\sigma d}) + 2\alpha\beta dg_{0}e^{-\sigma d} \\ &+ (\alpha^{2} + \beta^{2})g_{1}r_{0}(1 + e^{-2\sigma d}) + 2\alpha\beta \cdot 2g_{1}r_{0}e^{-\sigma d}\right]. \end{aligned}$$
(5.4)

In the transverse case g_0 and g_1 are replaced by $l_0 - \frac{1}{4}m_0^2/n_0$ and $l_1 - \frac{1}{2}m_1/(m_0/n_0) + \frac{1}{4}n_1(m_0/n_0)^2$, respectively. Differentiating (5.4) gives, in the lowest order,

$$\sigma^2 = cg_0. \tag{5.5}$$

On evaluating g_0, l_0, m_0, n_0 , Eq. (5.5) is found to be just the classical result,

$$\sigma^2 = \frac{N e^2 \mu_0}{m} \left(\frac{i\omega\tau}{1 + i\omega\tau} - \frac{\omega^2}{\omega_p^2} \right), \tag{5.6}$$

for the longitudinal case, and

$$\sigma^{2} = \frac{Ne^{2}\mu_{0}}{m} \left[\frac{i\omega\tau}{1+i\omega\tau} \frac{\gamma^{2}}{1+\gamma^{2}} - \frac{\omega^{2}}{\omega_{p}^{2}} + \left(\frac{i\omega\tau}{1+i\omega\tau} \frac{\gamma}{1+\gamma^{2}} \right)^{2} / \left(\frac{i\omega\tau}{1+i\omega\tau} \frac{\gamma^{2}}{1+\gamma^{2}} - \frac{\omega^{2}}{\omega_{p}^{2}} \right) \right]$$
(5.7)

⁶ M. C. Jones and E. H. Sondheimer, Phys. Rev. Letters 14, 643 (1965). ⁷ M. C. Jones and E. H. Sondheimer, in *Proceedings of the International Colloquium on Optical Properties and Electronic Structure of Metals and Alloys, Paris, 1965* (North-Holland Publishing Co., Amsterdam, 1966).

(5.8)

for the transverse case. Resubstituting into (5.4) from (5.5) gives additional nonclassical terms arising from surface collisions. In order to calculate the effect of these on the reflection and transmission of the slab, the components of the (diagonal) generalized admittances are found by comparing coefficients in (5.4) and (2.10). Thus,

 $X = X' = -2\sigma e^{-\sigma d} / (1 - e^{-2\sigma d})$

and

$$Y = Y' = \sigma(1 + e^{-2\sigma d}) / (1 - e^{-2\sigma d}) + cg_1 r_0, \quad (5.9)$$

where X, Y are either X_{xx} , Y_{xx} or X_{yy} , Y_{yy} , and g_1 is taken accordingly. In the limit $d \rightarrow \infty$ Eqs. (5.8), and (2.23) just give the result of Jones and Sondheimer.^{4,6} Further discussion of the effects of the additional non-classical term in (5.9) will be left until Sec. 6.

In the case $d \sim r_0$, it is no longer possible to treat σd as of order $(\sigma r_0)^0$, and so terms involving $e^{-\sigma d}$ in (5.4) must also be expanded as power series. Equation (4.6) now gives

$$\begin{split} \mathcal{I} &= \frac{1}{d} \bigg[\left(\mathcal{S}_{0}^{2} + \mathcal{S}_{d}^{2}\right) \bigg(1 + \frac{(\sigma d)^{4}}{45} + \cdots \bigg) + 2\mathcal{S}_{0}\mathcal{S}_{d} \bigg(-1 + \frac{7}{360} (\sigma d)^{4} + \cdots \bigg) \bigg] + c \bigg\{ \left(\mathcal{S}_{0}^{2} + \mathcal{S}_{d}^{2}\right) \bigg[\frac{1}{3} g_{0} d + g_{1} r_{0} \\ &+ g_{s} \frac{r_{0}^{2}}{d} + g_{s} \frac{r_{0}^{3}}{d^{2}} - h_{2} \frac{r_{0}^{2}}{d^{2}} + \bigg(-\frac{2}{45} g_{0} d + \frac{1}{3} g_{1} r_{0} + \frac{2}{3} g_{3} \frac{r_{0}^{3}}{d^{2}} + h_{2} \frac{r_{0}^{2}}{d^{2}} + g_{4} \frac{r_{0}^{4}}{d^{3}} + g_{5} \frac{r_{0}^{5}}{d^{4}} - h_{4} \frac{r_{0}^{4}}{d^{2}} \bigg) (\sigma d)^{2} + \cdots \bigg] \\ &+ 2\mathcal{S}_{0} \mathcal{S}_{d} \bigg[\frac{1}{6} g_{0} d - g_{2} \frac{r_{0}^{2}}{d} - g_{3} \frac{r_{0}^{3}}{d^{3}} + h_{2} \frac{r_{0}^{2}}{d^{2}} + \bigg(\frac{-7}{180} g_{0} d - \frac{1}{6} \frac{r_{0}^{2}}{d} + \frac{1}{6} \frac{r_{0}^{3}}{3} - \frac{1}{6} h_{2} \frac{r_{0}^{2}}{d^{2}} \\ &+ g_{4} \frac{r_{0}^{4}}{d^{3}} + g_{5} \frac{r_{0}^{5}}{d^{4}} - h_{4} \frac{r_{0}^{4}}{d^{4}} \bigg) (\sigma d)^{2} + \cdots \bigg] \bigg\}, \quad (5.10) \end{split}$$

where again, as in (5.4), this expression is valid for the transverse case as well as the longitudinal, provided that g_0 and g_1 , etc., are suitably redefined. Differentiating (5.10) with respect to σ and setting the resulting expression equal to zero gives σ —but this is found to depend on \mathcal{E}_0 and \mathcal{E}_d . This indicates that the exponential form for the fields is inadequate when $d \sim r_0$, but, nevertheless, because the leading terms are independent of σ , Eq. (5.10) can be used to give approximations for X and Y which go beyond the classical results. To fully assess the reliability of these requires finding

a better trial function for the field, but this is not attempted here. The functions g_0 and g_1 , etc., appearing in (5.10), are of considerable length and will not be displayed here explicitly; they can be obtained from (4.9) and (4.10).

6. RESULTS

To illustrate the effect of size and surfaces the transmittivity $(|T|^2)$, reflectivity $(|R|^2)$, and absorptivity $(1-|R|^2-|T|^2)$ have been computed for a model of a degenerate semiconductor. This has been chosen so that it is possible to satisfy $\omega_0 = \omega_p$ at values of the magnetic

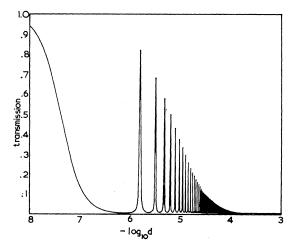


FIG. 1. The variation of classical longitudinal transmittivity with film thickness d (measured in meters). In this figure and Figs. 2 and 3 $\omega = 1.5\omega_p$ and the other parameters have the values given in the text.

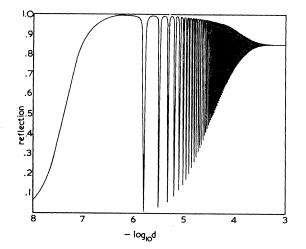


FIG. 2. The variation of classical longitudinal reflectivity with film thickness d.

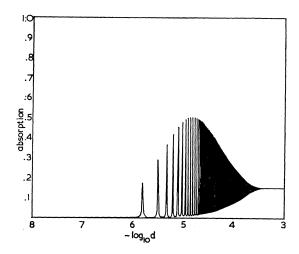


FIG. 3. The variation of classical longitudinal absorptivity with film thickness d.

field which are attainable in practice. In addition to the quantities mentioned above, the Voigt angle—the angle between the incident direction of polarization and the major axis of the elliptically polarized transmitted wave—has been computed.

In the model used, $N=10^{24}m^{-3}$, $m^*=0.1m$, and $\mathcal{E}=16\mathcal{E}_0$. Since $r_0=l/\omega_0\tau$, where *l* is the mean free path, the exact classical results are obtained from (5.8) and (5.9) by putting l=0. In order to check the validity of the expansion used in (5.10), values for l=0 are compared with the corresponding exact classical values.

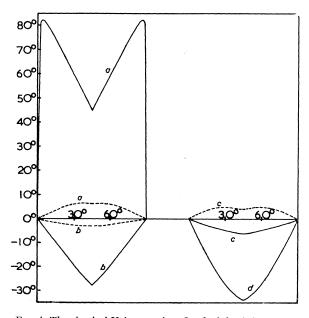


FIG. 4. The classical Voigt rotation. On the left of the figure ω_0 is fixed $(=0.1 \ \omega_p)$ and on the right is fixed $(=\omega_p)$. The broken curves are for $d=10^{-7}$ m the solid ones for $d=10^{-5}$ m. In curves (a) $\omega=\omega_p$ and in curves (b) $\omega=0.2 \ \omega_p$; in curves (c) $\omega_0=0.3 \ \omega_p$, and in curve (d) $\omega_0=\omega_p$.

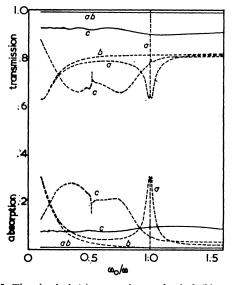


FIG. 5. The classical (a), approximate classical (b), and nonclassical (c) absorptivity and transmittivity in both the longitudinal (solid) and transverse (dashed) directions plotted against field for $\omega/\omega_p=1$, $d=10^{-7}$ m.

The figures show that there are pronounced size effects even in the classical case (Figs. 1–3) and that the surface effects can produce changes of the order of 10% or more (Figs. 5 and 6). The effects of a magnetic field are also considerable and, as Fig. 4 indicates, quite complicated behavior can result from varying, say, both frequency and film thickness even in the classical case. Figures 5–7 indicate that, apart from fields close to the bulk resonance at $\omega_0/\omega=1$, the expansion in (5.10) is valid, though for $\omega \ll_p$ "close to" covers a considerably wider range of fields than for larger values of ω . An exact physical description of the processes which give rise to

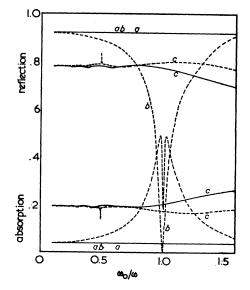


FIG. 6. The same curves as Fig. 6 for the absorptivity and reflectivity for $\omega/\omega_p = 0.1$, $d = 10^{-7}$ m.

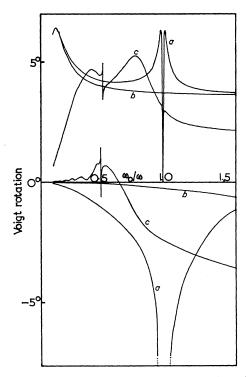


FIG. 7. Variation of Voigt rotation with field for $\omega/\omega_p=1$, $d=10^{-7}$ m (upper), and $\omega/\omega_p=0.1$ (lower). The labeling of the curves corresponds to that of Figs. 5 and 6.

the departures from classical behavior, because of the complicated expressions involved, offers some difficulty. A simple method of deriving the results of Jones and Sondheimer⁴ has been given by D'Haennens and Carter⁸ but this method cannot be used to give the results obtained in Sec. 5, although it throws some light on the processes involved.

⁸ J. P. D'Haennens and D. L. Carter, Phys. Rev. 140, A1992 (1965).

The reflectivity and absorptivity do not—except in a manner to be expected—differ appreciably qualitatively from the results of Jones and Sondheimer,⁴ but as they considered the semi-infinite case they were unable to consider the transmissive Voigt effect. As remarked above, in the absence of an improved solution for the field, the results probably only indicate the order of magnitude of the effects and are not to be taken as exact. The formalism introduced in Sec. 2, however, can be useful even in the classical theory. Donovan and Medcalf⁹ have considered size effects in the classical case, as have Ramey *et al.*¹⁰ and others, but such treatments are not always easy to compare. Because the results of Sec. 2 can be easily extended to a series of parallel slabs by using the relations

$$\begin{split} \mathbf{Y}_{1+2} &= \mathbf{Y}_1 - \mathbf{X}_1 (\mathbf{Y}_1' + \mathbf{Y}_2)^{-1} \mathbf{X}_1', \\ \mathbf{X}_{1+2} &= -\mathbf{X}_1 (\mathbf{Y}_1' + \mathbf{Y}_2)^{-1} \mathbf{X}_2, \\ \mathbf{Y}_{1+2}' &= \mathbf{Y}_2' - \mathbf{X}_2' (\mathbf{Y}_1' + \mathbf{Y}_2)^{-1} \mathbf{X}_2, \\ \mathbf{X}_{1+2}' &= -\mathbf{X}_2' (\mathbf{Y}_1' + \mathbf{Y}_2)^{-1} \mathbf{X}_1', \end{split}$$

in a self-explanatory notation, the results obtained here can be extended to an arbitrary arrangement of such slabs. Further effort will be directed towards the evaluation of the generalized admittances for other systems, for example, magnetic materials, and to more exact treatment of the surface terms.

ACKNOWLEDGMENTS

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⁹ B. Donovan and T. Medcalf, Phys. Letters 7, 304 (1963). ¹⁰ R. L. Ramey, W. H. Pitchen, Jr., J. M. Lloyd, and H. S. Landes, J. Appl. Phys. 39, 3883 (1968).