[7]B. W. Batterman, Phys. Rev. Letters 22, 703 (1969).
[8]For a discussion, see J. W. Corbett, in *Solid State Physics*, edited by F. Seitz and D. Turnbull (Academic, New York, 1966), Suppl. 7.
[9]R. A. Johnson and E. Brown, Phys. Rev. 127, 446 (1962).
[10]A. Seeger, E. Mann, and R. J. van Jan, Phys. Chem. Solids 23, 639 (1962).
[11]M. Doyama and R. M. J. Cotterill, in *Lattice Defects and Their Interactions*, edited by R. R. Hasiguti (Gordon and Breach, New York, 1968).
[12]L. Tewordt, Phys. Rev. 109, 61 (1958).
[13]K. H. Bennemann, Z. Physik 165, 445 (1961).
[14]R. A. Johnson, Phys. Rev. 145, 423 (1966).
[15]G. Borrman, Z. Physik 127, 297 (1950).
[16]B. W. Batterman and H. Cole, Rev. Mod. Phys. 36, 681 (1964).
[17]R. W. James, in *Solid State Physics*, edited by F. Seitz and D. Turnbull (Academic, New York, 1963), Vol. 15, p. 53.
[18]P. H. Dederichs, Physik Kondensierten Materie 5, 347 (1966).
[19]Merlini and E. van der Voort, Euratom Report No. 1643, 1965 (unpublished).
[20]H. Wagenfeld, Phys. Rev. 144, 216 (1966).
[21]Y. Shimomura, Phil. Mag. 19, 773 (1969).
[22]Y. Shimomura, J. Appl. Phys. 41, 749 (1970).
[23]F. Haussermann, M. Ruhle, G. Roth, and G. P. Scheidler. Phys. Status Solidi 32, K 103 (1969).
[24]P. H. Dederichs, Phys. Status Solidi 23, 377 (1967).
[25]P. H. Dederichs (unpublished).
[26]F. W. Young, Jr., T. O. Baldwin, and P. H. Dederichs, in *Vacancies and Interstitials in Metals*, edited by A. Seeger, D. Schumacher, W. Schilling, and J. Diehl (North-Holland, Amsterdam, 1970), p. 619.
[27]F. W. Young, Jr., and J. R. Savage, J. Appl. Phys. 35, 1917 (1964).
[28]F. W. Young, Jr., and T. R. Wilson, Rev. Sci. Instr. 32, 559 (1961).
[29]R. M. Nicklow, F. A. Sherrill, and F. W. Young, Jr., Phys. Rev. 137, A1417 (1965).
[30]B. Okkerse and P. Penning, Phillips Res. Rept. 18, 82 (1963).
[31]P. Penning and D. Polder, Phillips Res. Rept. 16, 419 (1961).
[32]L. P. Hunter, J. Appl. Phys. 30, 874 (1958).
[33]L. S. Edelheit, Ph. D. thesis, University of Illinois, 1970 (unpublished).
[34]O. S. Oen, ORNL Report No. ORNL-3813, 1965 (unpublished).
[35]R. A. Johnson, J. Phys. Chem. Solids 28, 275 (1967).
[36]A. C. Damask and G. J. Dienes, *Point Defects in Metals* (Gordon and Breach, New York, 1963).
[37]K. C. Lie and J. S. Koehler, Advan. Phys. 17, 421 (1968).
[38]R. A. Johnson, J. Phys. Chem. Solids 26, 75 (1965).
[39]T. O. Baldwin, F. W. Young, Jr., and A. Merlini, Phys. Rev. 163, 591 (1967).
[40]T. O. Baldwin, Phys. Status Solidi 25, 71 (1968).
[41]F. Seitz and J. S. Koehler, in *Solid State Physics*, edited by F. Seitz and D. Turnbull (Academic, New York, 1956), Vol. 2.
[42]J. W. Corbett, R. B. Smith, and R. M. Walker, Phys. Rev. 114, 1452 (1959).
[43]R. O. Simmons, J. S. Koehler, and R. W. Balluffi, *Radiation Damage in Solids* (International Atomic Energy Agency, Vienna, 1962).

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s-Polarized Optical Properties of Metals*

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A theory for the *s*-polarized optical properties of a metallic slab of arbitrary thickness is presented. The light can be incident upon the slab at an arbitrary angle of incidence, and it is assumed that the surface electron scattering is diffuse. Interesting oscillatory structure appears in the absorptance for a thin film when the frequency is in the range $0.01\omega_p$–$0.1\omega_p$, where ω_p is the plasma frequency. The origin of this structure is discussed. An expression is derived for the absorptance of a thick sample in the infrared.

I. INTRODUCTION

Significant progress has been made of late in incorporating nonlocality into the theory of the optical properties of metals.[1–6] These studies, in which the electron scattering at the surface has either implicitly or explicitly been taken to be specular, have, in general, involved extensions of the pioneering work of Reuter and Sondheimer,[7] Dingle,[8] and Mattis and Dresselhaus[9] to include arbitrary incident angles[1,3,5] and/or finite thickness samples.[2–4,6]

There is now considerable evidence that a more generally valid description of the optical properties of metals is afforded by considering the surface electron scattering to be diffuse rather than specular. That is not to say that specular scattering does not occur. Indeed, for some materials, surfaces can be prepared in such a way that the desired degree of specular reflection can be obtained.[10] However, a theory for the case of diffuse scattering would be highly desirable. For normally incident light, such a theory already exists for thick crystals[7,8] and also for thin films.[11] The ease with which these theories can be generalized to include light incident at non-normal angles depends in large measure on the type of polarization involved. For p polarization, where the light generates a longitudinal field, and effects such as electron-hole pair production and plasmon excitation result,[1-5] the problem is very difficult. However, for s polarization, where no longitudinal effects occur, the problem is somewhat easier and it is this case we consider here. Clearly, p polarization involves more processes of physical interest than does s polarization. However, s polarization is not without interesting aspects and, in addition, the mathematical techniques developed in the solution of the s-polarization problem are applicable also to the case of p polarization and are now being so used.

The basic relations between the optical properties and the surface impedance are developed in Sec. II. Section III contains some brief remarks concerning the surface impedance for a semi-infinite metal. Calculations of the surface impedances for thin films are given in Sec. IV, and in Sec. V these expressions are used to obtain an approximation for the absorptance of a semi-infinite crystal in the infrared. The results of the optical properties calculations are presented and discussed in Sec. VI.

II. BASIC RELATIONS

The coordinate system to be used in the analysis and the orientation of the slab are shown in Fig. 1. A plane wave of angular frequency ω with a velocity component in the $+z$ direction is incident from vacuum at an angle θ upon that surface of a nonpermeable metallic slab located at $z=0$. The slab is of thickness d and is backed by vacuum. In this s-polarized arrangement, the electric field points in the y direction. The amplitudes of the incident, reflected, and transmitted electric fields are, respectively, E_i, E_r, and E_t. With a phase factor $\exp[i(q_x x-\omega t)]$ understood, we can write

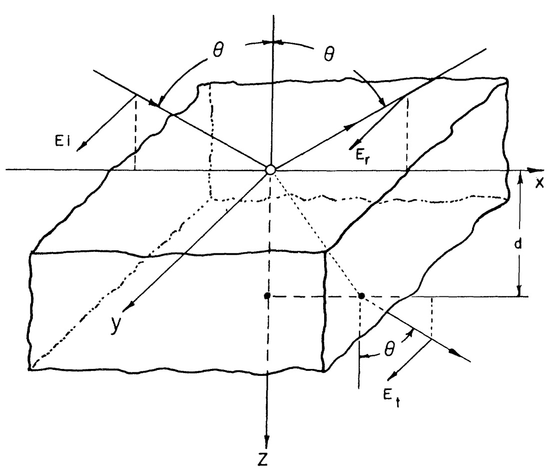

FIG. 1. Slab orientation and field geometry used in the calculation. The incident, reflected, and transmitted electric fields are denoted E_i, E_r, and E_t, and the incident angle is θ.

$$E_y(z)=\begin{cases} E_i \exp(iq_z z)+E_r\exp(-iq_z z), & z<0 \\ E_m(z)], & 0<z<d \\ E_t\exp[iq_z(z-d)], & d<z \end{cases} \tag{2.1}$$

where $q_x=\omega\sin\theta/c$, $q_z=\omega\cos\theta/c$, c is the velocity of light, and $E_m(z)$ is the electric field within the slab. From Maxwell's equations, we then find for the magnetic field component B_x

$$B_x(z)=\begin{cases} -E_i\cos\theta\exp(iq_z z)+E_r\cos\theta\exp(-iq_z z), & z<0 \\ \dfrac{ic}{\omega}\dfrac{dE_m(z)}{dz}, & 0<z<d \\ & d<z\ . \end{cases} \tag{2.2}$$

The continuity of the tangential field components E_y and B_x yields

$$E_i+E_r=E_m(0)\ , \tag{2.3}$$

$$E_t=E_m(d)\ , \tag{2.4}$$

$$-(E_i-E_r)\cos\theta=\frac{ic}{\omega}\frac{dE_m(0)}{dz}\ , \tag{2.5}$$

and

$$-E_t\cos\theta=\frac{ic}{\omega}\frac{dE_m(d)}{dz}\ . \tag{2.6}$$

Defining the surface impedance Z_s by[12]

$$Z_s=-E_y/B_x, \tag{2.7}$$

we have, from Eqs. (2.2), (2.4), and (2.6),

$$Z_s(d)=1/\cos\theta\ . \tag{2.8}$$

Equations (2.2), (2.3), and (2.5) lead to

$$\frac{E_i+E_r}{E_i-E_r}=Z_s(0)\cos\theta\ , \tag{2.9}$$

and thus,

$$\frac{E_r}{E_i}=\frac{Z_s(0)\cos\theta-1}{Z_s(0)\cos\theta+1} \quad . \tag{2.10}$$

Subtracting (2.5) divided by $\cos\theta$ from (2.3) yields

$$E_i=\tfrac{1}{2}E_m(0)\{1+[Z_s(0)\cos\theta]^{-1}\}, \tag{2.11}$$

and hence

$$\frac{E_t}{E_i}=\frac{2E_m(d)}{E_m(0)}\left(\frac{Z_s(0)\cos\theta}{Z_s(0)\cos\theta+1}\right) \quad . \tag{2.12}$$

The above equations represent a complete description of the optical properties since the reflectance is given by

$$R=\left|E_r/E_i\right|^2 , \tag{2.13}$$

the transmittance by

$$T=\left|E_t/E_i\right|^2 , \tag{2.14}$$

and the absorptance by

$$A=1-R-T . \tag{2.15}$$

Thus, the electric field at the surfaces and the surface impedances are needed to specify the optical parameters.

It should be noted that the present system for specifying the optical parameters results in a reflectance which is identical in form for a slab and a semi-infinite medium. This is in contrast to the usual system[13] for slabs in which two fields of specific symmetry about the slab center are utilized. The reason the system used here was selected is the form which results for the surface impedance at the slab back, Eq. (2.8). This expression can very easily be modified to allow for the presence of something other than vacuum backing the metallic film.

For convenience, we will develop our results below in terms of a set of dimensionless parameters. The electron gas with which we shall be concerned is comprised of electrons of charge $-e$, mass m, and mean density n_0. In addition, the electronic system is characterized by a Fermi velocity v, a Fermi energy ϵ_F, and a relaxation time[14] τ. In terms of the plasma frequency ω_p,

$$\omega_p=(4\pi n_0 e^2/m)^{1/2} ,$$

we define

$$\Omega=\omega/\omega_p , \qquad \gamma=(\omega_p\tau)^{-1} , \quad W=\omega_p d/c ,$$
$$Q_x=\Omega\sin\theta , \quad \Omega'=\Omega+i\gamma . \tag{2.16}$$

Ω is the dimensionless frequency, γ a measure of damping effects, W the dimensionless slab thickness, and Q_x the wave-vector component of the incident wave parallel to the slab surface in dimensionless form.

III. SEMI-INFINITE MEDIUM

It is a straightforward exercise to generalize the results of Reuter and Sondheimer[7] and Dingle[8] to obtain the surface impedance for diffuse scattering when the light is nonnormally incident. The result is

$$Z_s=-i\Omega\pi\Big/\int_0^\infty dQ_z \ln\left(\frac{Q^2-\Omega^2\epsilon_t(Q,\Omega)}{Q_z^2}\right) , \tag{3.1}$$

where

$$Q^2=Q_x^2+Q_z^2 , \tag{3.2}$$

and $\epsilon_t(Q,\Omega)$ is the transverse dielectric constant of the electron gas which we take here to be[15]

$$\epsilon_t(Q,\Omega)=1-f_t/(\Omega\Omega') , \tag{3.3}$$

with

$$f_t=\tfrac{3}{8}(z^2+u'^2+1)-\frac{3}{32z}\left[[1-(z-u')^2]^2\ln\left(\frac{z-u'+1}{z-u'-1}\right)\right.$$
$$\left.+[1-(z+u')^2]^2\ln\left(\frac{z+u'+1}{z+u'-1}\right)\right] , \tag{3.4}$$

where

$$z=Q\omega_p/2mvc, \quad u'=\Omega'[Q(v/c)]^{-1} . \tag{3.5}$$

This dielectric function is an extension of the transverse dielectric function of Lindhard[16] in that it includes a finite electron lifetime. Its use here represents a generalization over the use of the transverse dielectric function obtained from the Boltzmann equation.[17]

For comparison with Eq. (3.1), we give here the surface impedance for s polarization and specular scattering[1]

$$Z_s\big|_{\text{spec.}}=\frac{2i\Omega}{\pi}\int_0^\infty\frac{dQ_z}{\Omega^2\epsilon_t(Q,\Omega)-Q^2} \quad . \tag{3.6}$$

Results for the specular case have previously been presented.[1,3] Equation (3.1) for diffuse scattering can be integrated numerically to obtain values of the surface impedance. However, an accurate integration requires a significant amount of computer time. Since the general result for a finite thickness sample yields the same information more easily, a discussion of the results for a semi-infinite medium will be deferred until Sec. VI.

IV. FINITE THICKNESS SAMPLE

Consider now the slab of Fig. 1. We write Ampere's law as

$$\nabla\times\vec{B}=\frac{4\pi}{c}\vec{J}+\frac{\epsilon}{c}\frac{\partial\vec{E}}{\partial t}, \tag{4.1}$$

where ϵ, a function of ω in general, includes effects of core polarization and interband transitions when present, and reduces to 1 when such effects are ignored or unimportant.[18] With the electric field of Eq. (2.1), we obtain from Maxwell's equations

$$\frac{d^2E_m(z)}{dz^2}+\left(\epsilon\frac{\omega^2}{c^2}-q_x^2\right)E_m(z)=-\frac{4\pi i\omega}{c^2}J_y(z), \tag{4.2}$$

since $\nabla\cdot\vec{E}=0$ for this s-polarized situation.

To obtain $J_y(z)$ we use the Boltzmann equation,

$$\frac{\partial f_{\vec{k}}}{\partial t}-\frac{e}{\hbar}\vec{E}\cdot\nabla_{\vec{k}}f_{\vec{k}}+\vec{v}_{\vec{k}}\cdot\nabla_{\vec{r}}f_{\vec{k}}=\left(\frac{\partial f_{\vec{k}}}{\partial t}\right)_{\text{collisions}}, \tag{4.3}$$

where $f_{\vec{k}}$ is the one-particle distribution function and $\vec{v}_{\vec{k}}=\hbar\vec{k}/m$. We assume

$$f_{\vec{k}}=f_0+f_1(\vec{v}_{\vec{k}},z)\exp[i(q_xx-\omega t)], \tag{4.4}$$

where the equilibrium distribution f_0 for the electrons is the Fermi distribution

$$f_0=\{\exp[\beta(\epsilon_{\vec{k}}-\mu)]+1\}^{-1}$$

with $\beta=(k_BT)^{-1}$, and μ the electronic chemical potential. The function f_1 is to be proportional to the electric field. Making the relaxation time approximation[14]

$$\left(\frac{\partial f_{\vec{k}}}{\partial t}\right)_{\text{collisions}}=-\frac{f_{\vec{k}}-f_0}{\tau}, \tag{4.5}$$

we have

$$\frac{\partial f_1}{\partial z}+\xi f_1=eE_m\left(\frac{v_y}{v_z}\right)\left(\frac{\partial f_0}{\partial\epsilon_{\vec{k}}}\right), \tag{4.6}$$

where

$$\xi=iq_x(v_x/v_z)-i\omega'/v_z,\qquad \omega'=\omega+i/\tau, \tag{4.7}$$

and v_x, v_y, and v_z are components of $\vec{v}_{\vec{k}}$.

The general solution of Eq. (4.6) is

$$f_1=e^{-\xi z}\left[F(\vec{v}_{\vec{k}})+e\left(\frac{\partial f_0}{\partial\epsilon_{\vec{k}}}\right)\left(\frac{v_y}{v_z}\right)\int_0^z E_m(s)e^{\xi s}\,ds\right], \tag{4.8}$$

where $F(\vec{v}_{\vec{k}})$ is an arbitrary function of $\vec{v}_{\vec{k}}$. For electrons moving with $v_z<0$,

$$f_1^-=e^{-\xi z}\left[F^-(\vec{v}_{\vec{k}})+e\left(\frac{\partial f_0}{\partial\epsilon_{\vec{k}}}\right)\left(\frac{v_y}{v_z}\right)\int_0^z E_m(s)e^{\xi s}\,ds\right], \tag{4.9a}$$

and for electrons moving with $v_z>0$,

$$f_1^+=e^{-\xi z}\left[F^+(\vec{v}_{\vec{k}})+e\left(\frac{\partial f_0}{\partial\epsilon_{\vec{k}}}\right)\left(\frac{v_y}{v_z}\right)\int_0^z E_m(s)e^{\xi s}\,ds\right]. \tag{4.9b}$$

The diffuse scattering boundary conditions will be used to determine $F^\pm$. These boundary conditions require (i) $f_1^+=0$ at $z=0$, (ii) $f_1^-=0$ at $z=d$. Thus,

$$F^+(\vec{v}_{\vec{k}})=0 \tag{4.10a}$$

and

$$F^-(\vec{v}_{\vec{k}})=-e\left(\frac{\partial f_0}{\partial\epsilon_{\vec{k}}}\right)\left(\frac{v_y}{v_z}\right)\int_0^d E_m(s)e^{\xi s}\,ds. \tag{4.10b}$$

Equations (4.10) together with Eqs. (4.9) lead to

$$f_1^-=e^{-\xi z}\left[-e\left(\frac{\partial f_0}{\partial\epsilon_{\vec{k}}}\right)\left(\frac{v_y}{v_z}\right)\int_z^d E_m(s)e^{\xi s}\,ds\right] \tag{4.11a}$$

and

$$f_1^+=e^{-\xi z}\left[e\left(\frac{\partial f_0}{\partial\epsilon_{\vec{k}}}\right)\left(\frac{v_y}{v_z}\right)\int_0^z E_m(s)e^{\xi s}\,ds\right]. \tag{4.11b}$$

To obtain the current density $J_y(z)$ given by

$$J_y(z)=-2e(m/h)^3\int v_yf_1(\vec{v}_{\vec{k}},z)\,dv_x\,dv_y\,dv_z, \tag{4.12}$$

we use Eqs. (4.11) and proceed as in Ref. 1. We find

$$J_y(z)=(4\pi e^2/h^3)(mv)^2\int_0^d E_m(s)\,ds$$
$$\times\int_0^{\pi/2}\sin^2\alpha\tan\alpha\{J_0[q_x\tan\alpha(z-s)]$$
$$+J_0''[q_x\tan\alpha(z-s)]\}\exp(i\omega'\sec\alpha\,|z-s|/v)\,d\alpha. \tag{4.13}$$

Making the variable change $t=\sec\alpha$, J_y can be written

$$J_y(z)=\frac{2\pi e^2}{h^3}(mv)^2\int_0^d E_m(s)\,ds\int_1^\infty dt\left(\frac{1}{t}-\frac{1}{t^3}\right)$$
$$\times\{J_0[q_x(z-s)(t^2-1)^{1/2}]$$
$$+J_2[q_x(z-s)(t^2-1)^{1/2}]\}\exp(i\omega't\,|z-s|/v), \tag{4.14}$$

and so, from Eqs. (4.2) and (4.14), the integro-differential equation we must solve is

$$\frac{d^2E_m(z)}{dz^2}+\frac{\omega^2}{c^2}(\epsilon-\sin^2\theta)E_m(z)=\frac{\kappa}{2}\int_0^d E_m(s)\,ds$$
$$\times\int_1^\infty dt\left(\frac{1}{t}-\frac{1}{t^3}\right)\{J_0[q_x(z-s)(t^2-1)^{1/2}]$$

$$+ J_2[q_x(z-s)(t^2-1)^{1/2}]\} \exp \frac{i\omega' t|z-s|}{v} \ , \tag{4.15}$$

where

$$\kappa = -(i\omega/h^3)(4\pi emv/c)^2 \ . \tag{4.16}$$

The imaginary part of the exponent appearing in the expression for $J_y(z)$ is

$$\omega t\,|z-s|/v \ , \tag{4.17}$$

while the magnitude of the argument of the Bessel functions appearing in $J_y(z)$ is

$$\omega \sin\theta\,|z-s|\,(t^2-1)^{1/2}/c \ . \tag{4.18}$$

Since $t \geq 1$, (4.18) is much less than (4.17)[19] and the integral on t in J_y is dominated by the oscillatory behavior of the exponential. Indeed, essentially all of the contribution to the integral on t will occur for (4.17) $\lesssim 1$ which means (4.18) $\ll 1$. Hence, we can expand the Bessel functions,

$$\begin{aligned} J_0[q_x(z-s)(t^2-1)^{1/2}] &+ J_2[q_x(z-s)(t^2-1)^{1/2}] \\ &= 1 - \tfrac{1}{8} q_x^2 (z-s)^2 (t^2-1) \\ &\quad + \tfrac{1}{192} q_x^4 (z-s)^4 (t^2-1)^2 - \cdots \ , \end{aligned} \tag{4.19}$$

integrate term by term, and expect a rapidly converging series.

The above procedure also reflects an important physical point. Rapid convergence of the series resulting from the expansion (4.19) indicates that the kernel in the current density is nearly independent of the incident angle as (4.19) is also an expansion in $\sin^2\theta$.

Using (4.19), the equation we must solve becomes

$$\begin{aligned} \frac{d^2E_m(z)}{dz^2} + \frac{\omega^2}{c^2}(\epsilon - \sin^2\theta)\,E_m(z) &\simeq \frac{\kappa}{2}\int_0^d E_m(s)\,ds \\ &\times \int_1^\infty dt \left(\frac{1}{t} - \frac{1}{t^3}\right)[1 - \tfrac{1}{8} q_x^2 (z-s)^2 (t^2-1) \\ &+ \tfrac{1}{192} q_x^4 (z-s)^4 (t^2-1)^2] \exp(i\omega' t|z-s|/v) \ . \end{aligned} \tag{4.20}$$

Assume now

$$E_m^{(0)}(z) = e^{i\omega'\alpha z/v} \ , \tag{4.21}$$

where α is an undetermined parameter. The use of (4.21) in the left-hand side of (4.20) yields

$$[-(\omega'\alpha/v)^2 + \omega^2(\epsilon - \sin^2\theta)/c^2]\,e^{i\omega'\alpha z/v}, \tag{4.22}$$

while from the right-hand side we get

$$\begin{aligned} &\frac{\kappa v}{2i\omega'}\left[-e^{i\omega'\alpha z/v}K_1(\alpha) + \int_1^\infty dt\,F_1(t,\alpha)e^{i\omega' tz/v} + e^{i\omega'\alpha d/v}\int_1^\infty dt\,F_1(t,-\alpha)\,e^{i\omega' t(d-z)/v}\right] - \tfrac{1}{16}\kappa q_x^2\,(v/i\omega')^3 \\ &\times\left[-2e^{i\omega'\alpha z/v}K_3(\alpha) + \int_1^\infty dt\,F_3(t,\alpha)e^{i\omega' tz/v}G(z,\alpha,t) + e^{i\omega'\alpha d/v}\int_1^\infty dt\,F_3(t,-\alpha)e^{i\omega' t(d-z)/v}G(d-z,-\alpha,t)\right] \\ &+ \tfrac{1}{384}\kappa q_x^4 (v/i\omega')^5[-24\,e^{i\omega'\alpha z/v}K_5(\alpha) + \int_1^\infty dt\;F_5(t,\alpha)e^{i\omega' tz/v}H(z,\alpha,t) \\ &+ e^{i\omega'\alpha d/v}\int_1^\infty dt\,F_5(t,-\alpha)\,e^{i\omega' t(d-z)/v}H(d-z,-\alpha,t)] \ , \end{aligned} \tag{4.23}$$

where

$$K_n(\alpha) = \int_1^\infty dt\,\frac{(t^2-1)^{(n+1)/2}}{t^3}\left(\frac{1}{(t-\alpha)^n} + \frac{1}{(t+\alpha)^n}\right) \ , \tag{4.24a}$$

$$F_n(t,\alpha) = \frac{(t^2-1)^{(n+1)/2}}{t^3}\,\frac{1}{(t-\alpha)^n} \ , \tag{4.24b}$$

$$G(z,\alpha,t) = (Az)^2 - 2Az + 2 \ , \tag{4.24c}$$

$$H(z,\alpha,t) = (Az)^4 - 4(Az)^3 + 12(Az)^2 - 24Az + 24 \ , \tag{4.24d}$$

and

$$A = i\omega'(t-\alpha)/v \ . \tag{4.24e}$$

Now choose α such that

$$-\left(\frac{\omega'}{v}\alpha\right)^2+\frac{\omega^2}{c^2}(\epsilon-\sin^2\theta)=-\frac{\kappa v}{2i\omega'}K_1(\alpha)+\frac{\kappa q_x^2}{8}\left(\frac{v}{i\omega'}\right)^3K_3(\alpha)-\frac{\kappa q_x^4}{16}\left(\frac{v}{i\omega'}\right)^5K_5(\alpha)\ , \tag{4.25}$$

which leaves terms in (4.23) proportional to $\kappa v/2i\omega'$ and involving the functions F_n. If we then add to $E_m^{(0)}(z)$ the field term $E_m^{(1)}(z)$,

$$E_m^{(1)}(z)=\tfrac{1}{2}\kappa(v/i\omega')^3\left[\int_1^\infty dt\, e^{i\omega' tz/v}D(z,\alpha,t)+e^{i\omega'\alpha d/v}\int_1^\infty dt\, e^{i\omega' t(d-z)/v}D(d-z,-\alpha,t)\right]\ , \tag{4.26}$$

where

$$D(z,\alpha,t)=\frac{F_1(t,\alpha)}{(t^2+\Gamma)}-\frac{1}{8}\left(\frac{q_xv}{i\omega'}\right)^2F_3(t,\alpha)\left(\frac{G(z,\alpha,t)}{(t^2+\Gamma)}-\frac{[4Az(t^2-\alpha t)-2(t^2-\alpha^2)]}{(t^2+\Gamma)^2}+\frac{8t^2(t-\alpha)^2}{(t^2+\Gamma)^3}\right)$$
$$+\frac{1}{192}\left(\frac{q_xv}{i\omega'}\right)^4F_5(t,\alpha)\left(\frac{H(z,\alpha,t)}{(t^2+\Gamma)}+\frac{[-8(Az)^3(t^2-\alpha t)+12(Az)^2(t^2-\alpha^2)-24Az(t^2-\alpha^2)+24(t^2-\alpha^2)]}{(t^2+\Gamma)^2}\right.$$
$$+\frac{[48(Az)^2t^2(t-\alpha)^2-96Azt\alpha(t-\alpha)^2+24(t^2-\alpha^2)^2]}{(t^2+\Gamma)^3}$$
$$\left.+\frac{[-192Azt^3(t-\alpha)^3+96t^2(t-\alpha)^3(-t+3\alpha)]}{(t^2+\Gamma)^4}+\frac{384t^4(t-\alpha)^4}{(t^2+\Gamma)^5}\right) \tag{4.27a}$$

and

$$\Gamma=-(\omega/\omega')^2(v/c)^2(\epsilon-\sin^2\theta)\ , \tag{4.27b}$$

then operating on $E_m^{(1)}$ with the operator on the left-hand side of Eq. (4.20)

$$\frac{d^2}{dz^2}+\frac{\omega^2}{c^2}(\epsilon-\sin^2\theta) \tag{4.28}$$

yields terms which precisely cancel those terms in (4.23) which involve the functions F_n, that is, those terms in (4.23) not included in the determining equation for α, Eq. (4.25). However, there then arise terms proportional to δ^2, where δ is defined by

$$\delta=\tfrac{1}{2}\kappa(v/i\omega')^3\ , \tag{4.29}$$

when $E_m^{(1)}$ is used in the right-hand side of Eq. (4.20). These terms would in turn be cancelled by adding a field term $E_m^{(2)}$, proportional to δ^2, which would yield these terms when operated on by the left-hand side operator (4.28). Clearly, this procedure yields a solution for the electric field E_m which is a power series in δ. Each term of this power series, except the first, is also a power series, but in the parameter $[(v\sin\theta)/c]^2$. Thus, the solution represented by

$$E_m=E_m^{(0)}+E_m^{(1)} \tag{4.30}$$

includes terms up to order δ^2 and, within the terms in the δ series, terms up to order $[(v\sin\theta/c]^6$ have been retained.[20]

The above nested-series solution is useful only if the expansion parameters are such that the series converge quickly. Since $v/c<10^{-2}$, the series involving $(v/c)^2$ converges quickly as was already noted above. In terms of the dimensionless parameters of Eqs. (2.16), we have

$$\delta=\tfrac{3}{4}(v/c)^2\Omega/\Omega'^3\ . \tag{4.31}$$

With damping such that $\gamma\lesssim10^{-3}$, the solution (4.30) will be valid for

$$\left(\frac{v}{c}\frac{1}{\Omega}\right)^2\ll1\quad\text{when }\Omega\gg\gamma$$

and

$$(v/c)^2\frac{\Omega}{\gamma^3}\ll1\quad\text{when }\Omega\ll\gamma\ .$$

Potassium, for example, has[1] $v=0.85\times10^8$ cm/sec, and thus our solution is valid for $\Omega\gtrsim10^{-2}$ or $\Omega\lesssim10^{-4}$. If damping is severe, $\gamma\gtrsim10^{-2}$, then $|\delta|\ll1$ for all Ω, and thus our solution is valid for all Ω. In general, then, we have a solution valid everywhere but in the frequency region of the extreme anomalous skin effect. In particular, we have a solution valid in the optical region $\Omega\gtrsim10^{-2}$.

Equation (4.25) which determines α is even in α. Thus, a general solution for the electric field is $E_m^T(z)$ given by

$$E_m^T(z)=E_m(z)+PE_m^-(z)\ , \tag{4.32}$$

where $E_m(z)$ is given by (4.30), $E_m^-(z)$ is $E_m(z)$ with $\alpha\to-\alpha$, and P is a parameter to be determined.[21] We can now obtain the necessary component of the magnetic field from Eqs. (2.2) and determine P by using (2.8). We then have available all the quantities needed to calculate the optical properties. However, they are not yet in useful form. To begin to remedy this defect, let us make an estimate of the size of α. This turns out to be very useful since the result leads to marked simplifications in evaluating integrals such as those in Eq. (4.26).

Expression $K_n(\alpha)$, Eq. (4.24a), can be evaluated exactly. The results for those values of n needed here are

$$K_1(\alpha)=\frac{1}{\alpha^3}\left[2\alpha+(\alpha^2-1)\ln\left(\frac{1+\alpha}{1-\alpha}\right)\right] ,$$

$$K_3(\alpha)=\frac{2}{\alpha^5}\left[-6\alpha+(3-\alpha^2)\ln\left(\frac{1+\alpha}{1-\alpha}\right)\right], \tag{4.33}$$

$$K_5(\alpha)=\frac{1}{\alpha^7}\left[26\alpha+\frac{4\alpha}{1-\alpha^2}+(3\alpha^2-15)\ln\left(\frac{1+\alpha}{1-\alpha}\right)\right].$$

Consider then Eq. (4.25) for the electron gas, i.e., $\epsilon=1$. The terms involving K_3 and K_5 should involve small corrections so ignore them for the moment. Then,

$$\alpha^2\simeq\left(\frac{v}{c}\frac{\Omega}{\Omega'}\right)^2[\epsilon_t(\alpha)-\sin^2\theta] , \tag{4.34}$$

where

$$\epsilon_t(\alpha)=1-\frac{3}{2}\frac{1}{\Omega\Omega'}\left[\frac{1}{\alpha^2}+\frac{\alpha^2-1}{2\alpha^3}\ln\left(\frac{1+\alpha}{1-\alpha}\right)\right] . \tag{4.35}$$

$\epsilon_t(\alpha)$ is the transverse dielectric function for the electron gas as obtained from the Boltzmann equation.[22] The presence of the factor $(v/c)^2$ outside the bracket in (4.34) suggests that α is of order (v/c), and thus small. α_0, the lowest-order approximation for α, results from taking $\alpha=0$ on the right-hand side of (4.34). Thus,

$$\alpha_0=\frac{v}{c}\frac{\Omega}{\Omega'}\left(\cos^2\theta-\frac{1}{\Omega\Omega'}\right)^{1/2} . \tag{4.36}$$

Keeping terms of order α^2 on the right-hand side of (4.25) leads to

$$\alpha\cong\alpha_0[1-\tfrac{1}{10}(v/c)^2\Omega/\Omega'^3] \tag{4.37}$$

through terms of order $(v/c)^2$. Hence, $\alpha\sim(v/c)\times(1/\Omega)$ for $\Omega\gg\gamma$, so $|\alpha|\ll1$ in the frequency range $\Omega\gtrsim10^{-2}$. Since this is the frequency range of interest here, we can exploit this fact in evaluating the electric and magnetic fields. This field evaluation is tedious and only the results will be given. Defining

$$I_n(s)=\int_1^\infty e^{sx}x^{-n}\,dx, \tag{4.38}$$

we find

$$E_m(0)=1+\delta\left[\frac{2}{15}+\frac{\alpha}{12}+\frac{2}{35}(\alpha^2+\Gamma)-\frac{12}{105}\left(\frac{v}{c}\frac{\Omega}{\Omega'}\frac{\sin\theta}{i}\right)^2\right]+\delta\, e^{i\Omega'W\alpha/(v/c)}\left\{(I_4-I_6)-\alpha(I_5-I_7)+(\alpha^2+\Gamma)(I_6-I_8)\right.$$
$$\left.-\frac{1}{8}\left(\frac{v}{c}\frac{\Omega}{\Omega'}\frac{\sin\theta}{i}\right)^2\left[\left(\frac{i\Omega'W}{v/c}\right)^2(I_2-2I_4+I_6)-6\left(\frac{i\Omega'W}{v/c}\right)(I_3-2I_5+I_7)+12(I_4-2I_6+I_8)\right]\right\} , \tag{4.39}$$

where we have assumed α is small and terms through order $(v/c)^2$ have been retained. The argument of all the I_n in Eq. (4.39) and the equations below is $[i\Omega'W/(v/c)]$. Similarly

$$E_m(d)=\exp\left(\frac{i\Omega'\alpha W}{v/c}\right)+\delta\left\{(I_4-I_6)+\alpha(I_5-I_7)+(\alpha^2+\Gamma)(I_6-I_8)-\frac{1}{8}\left(\frac{v}{c}\frac{\Omega}{\Omega'}\frac{\sin\theta}{i}\right)^2\left[\left(\frac{i\Omega'W}{v/c}\right)^2(I_2-2I_4+I_6)\right.\right.$$
$$\left.\left.-6\left(\frac{i\Omega'W}{v/c}\right)(I_3-2I_5+I_7)+12(I_4-2I_6+I_8)\right]\right\}+\delta\exp\left(\frac{i\Omega'\alpha W}{v/c}\right)\left[\frac{2}{15}-\frac{\alpha}{12}+\frac{2}{35}(\alpha^2+\Gamma)-\frac{12}{105}\left(\frac{v}{c}\frac{\Omega}{\Omega'}\frac{\sin\theta}{i}\right)^2\right] , \tag{4.40}$$

$$B_x^+(0)=-\frac{c}{v}\frac{\Omega'}{\Omega}\alpha-\frac{c}{v}\frac{\Omega'}{\Omega}\delta\left[\frac{1}{4}+\frac{2}{15}\alpha+\frac{1}{12}(\alpha^2+\Gamma)-\frac{1}{8}\left(\frac{v}{c}\frac{\Omega}{\Omega'}\frac{\sin\theta}{i}\right)^2\right]-\frac{c}{v}\frac{\Omega'}{\Omega}\delta\exp\left(\frac{i\Omega'\alpha W}{v/c}\right)\left\{-(I_3-I_5)+\alpha(I_4-I_6)\right.$$
$$\left.-(\alpha^2+\Gamma)(I_5-I_7)+\frac{1}{8}\left(\frac{v}{c}\frac{\Omega}{\Omega'}\frac{\sin\theta}{i}\right)^2\left[\left(\frac{i\Omega'W}{v/c}\right)^2(I_1-2I_3+I_5)-4\left(\frac{i\Omega'W}{v/c}\right)(I_2-2I_4+I_6)+6(I_3-2I_5+I_7)\right]\right\} , \tag{4.41}$$

and

$$B_x^+(d)=-\frac{c}{v}\frac{\Omega'}{\Omega}\alpha\exp\left(\frac{i\Omega'\alpha W}{v/c}\right)-\frac{c}{v}\frac{\Omega'}{\Omega}\delta\exp\left(\frac{i\Omega'W}{v/c}\right)\left[-\frac{1}{4}+\frac{2}{15}\alpha-\frac{1}{12}(\alpha^2+\Gamma)+\frac{1}{8}\left(\frac{v}{c}\frac{\Omega}{\Omega'}\frac{\sin\theta}{i}\right)^2\right]$$

$$-\frac{c}{v}\frac{\Omega'}{\Omega}\delta\Big\{(I_3-I_5)+\alpha(I_4-I_6)+(\alpha^2+\Gamma)(I_5-I_7)$$

$$-\frac{1}{8}\left(\frac{v}{c}\frac{\Omega}{\Omega'}\frac{\sin\theta}{i}\right)^2\left[\left(\frac{i\Omega' W}{v/c}\right)^2(I_1-2I_3+I_5)-4\left(\frac{i\Omega' W}{v/c}\right)(I_2-2I_4+I_6)+6\quad_3-2I_5+I_7)\right]\Big\} \quad . \tag{4.42}$$

The properties of the exponential-type integrals I_n are discussed in the Appendix.

Keep in mind that the above field expressions (4.39)–(4.42) are the $+\alpha$ contributions to the fields, that is, the total electric field E^T and the total x component of the magnetic field B_x^T are given by

$$E_m^T = E_m + P E_m^- \tag{4.43}$$

and

$$B_x^T = B_x^+ + P B_x^- \ , \tag{4.44}$$

where E_m^- is obtained from E_m by letting $\alpha \to -\alpha$, and B_x^- is obtained in like fashion from B_x^+.

V. THICK CRYSTALS AND INFRARED FREQUENCIES

A technique which has been found useful[23,24] for testing our understanding of optical properties of metals is to measure at normal incidence the optical parameters of thick crystals in the infrared where the absorptivity is nearly independent of frequency and then, assuming diffuse scattering, compare the absorptivity with the expression

$$A_{uc} = 2\gamma + \tfrac{3}{4}v/c \ , \tag{5.1}$$

which results from a simple extension of the work of Holstein[25] and also appears in papers of Dingle.[26] The limits of validity of Eq. (5.1) have not been examined. We do so now.

Letting $W\to\infty$ in Eqs. (4.39) and (4.41) permits us to write the surface impedance Z_s for the semi-infinite crystal as

$$Z_s = Z_{c1} - \frac{3}{16}\frac{v}{c}\frac{Z_{c1}^2}{\Omega'^2} + \left(\frac{3}{16}\right)^2\left(\frac{v}{c}\right)^2\frac{Z_{cl}^3}{\Omega'^4} + \frac{1}{10}\left(\frac{v}{c}\right)^2\frac{\Omega}{\Omega'^3}Z_{c1} \ , \tag{5.2}$$

retaining terms through $(v/c)^2$. Z_{c1}, the classical (or local) expression for the surface impedance for a semi-infinite metal is

$$Z_{c1} = [\epsilon - \sin^2\theta - (\Omega\Omega')^{-1}]^{-1/2} \ . \tag{5.3}$$

Since we are here interested in the infrared, let us assume $\Omega \ll 1$, $\Omega^2|\epsilon| \ll 1$, and $\gamma \ll \Omega$. We then find

$$Z_s = \frac{\gamma}{2}\left[1-\frac{1}{8}\left(\frac{\gamma}{\Omega}\right)^2+\frac{3}{2}\Omega^2(\epsilon-\sin^2\theta)\right]+\frac{3}{16}\left(\frac{v}{c}\right)\left[1\left(-\frac{\gamma}{\Omega}\right)^2+\Omega^2(\epsilon-\sin^2\theta)\right]+\left(\frac{v}{c}\right)^2\frac{1}{\Omega}\left(-\frac{65}{512}\frac{\gamma}{\Omega}\right)$$

$$-i\left\{\Omega\left[1+\frac{1}{8}\left(\frac{\gamma}{\Omega}\right)^2+\frac{\Omega^2}{2}(\epsilon-\sin^2\theta)\right]+\frac{3}{16}\frac{v}{c}\frac{\gamma}{\Omega}\left[1-\left(\frac{\gamma}{\Omega}\right)^2\right]+\frac{83}{1280}\left(\frac{V}{C}\right)^2\frac{1}{\Omega}\right]\right\} \quad . \tag{5.4}$$

Note that Eq. (5.4) is *not*, in general, separated into real (Re) and imaginary (Im) parts since ϵ is complex.

The above expression together with the absorptance expression for an infinite crystal,

$$A_\infty = \frac{4\mathrm{Re}Z_s\cos\theta}{|Z_s\cos\theta+1|^2}\ , \tag{5.5}$$

give the desired approximation to the absorptance. If we now consider frequencies below which interband effects occur, so that $\epsilon = 1$, or, alternatively, frequencies such that the important damping effects can be taken into account by considering γ to be an effective γ, Eqs. (5.4) and (5.5) yield

$$A_\infty = 2\gamma\cos\theta\left[1-\frac{1}{8}\left(\frac{\gamma}{\Omega}\right)^2+\tfrac{1}{2}\Omega^2\cos^2\theta-\gamma\cos\theta\right]$$

$$+\frac{3}{4}\left(\frac{v}{c}\right)\cos\theta\left[1-\left(\frac{\gamma}{\Omega}\right)^2-2\gamma\cos\theta\right]$$

$$+\left(\frac{v}{c}\right)^2\cos\theta\left[-\frac{65}{128}\frac{\gamma}{\Omega^2}-\frac{9}{32}\cos\theta\right] \quad . \tag{5.6}$$

The correction terms appearing in (5.6) but not in (5.1) will be seen in Sec. VI to be important in some cases.

VI. RESULTS AND DISCUSSION

Calculations of the optical properties have been made for $v = 0.85\times10^8$ cm/sec representing potassium and $v = 1.34\times10^8$ cm/sec representing aluminum. We have also considered $\epsilon = 1$, thereby eliminating interband effects. However, some comments on the effect of interband transitions appear below.

The absorptance for $W = 0.3$[27] is shown in Fig. 2 as a function of frequency. The most striking feature of these curves is the oscillatory structure for $10^{-2} \lesssim \Omega \lesssim 10^{-1}$, the origin of which we will discuss below. This structure appears for $0.1 \lesssim W \lesssim 1.0$, being more prominent at large incident angles for small thickness because the marked decrease in absorptivity with increasing frequency for small incident angles tends to deemphasize the oscillations. The curves in Fig. 2 were obtained for $\gamma = 10^{-3}$ representing moderate damping. If the damping is reduced such that $\gamma = 10^{-4}$, these absorptivity curves drop only ~ 10%. This points up the insensitivity of the optical properties to moderate damping when the surface scattering is diffuse. Increasing γ to 10^{-2} roughly doubles the absorptivities shown in Fig. 2 and the structure is nearly obscured. The sharp absorptivity drop for $\Omega \sim 3\times10^{-3}$ is probably due, in part, to a breakdown of the theory.[28]

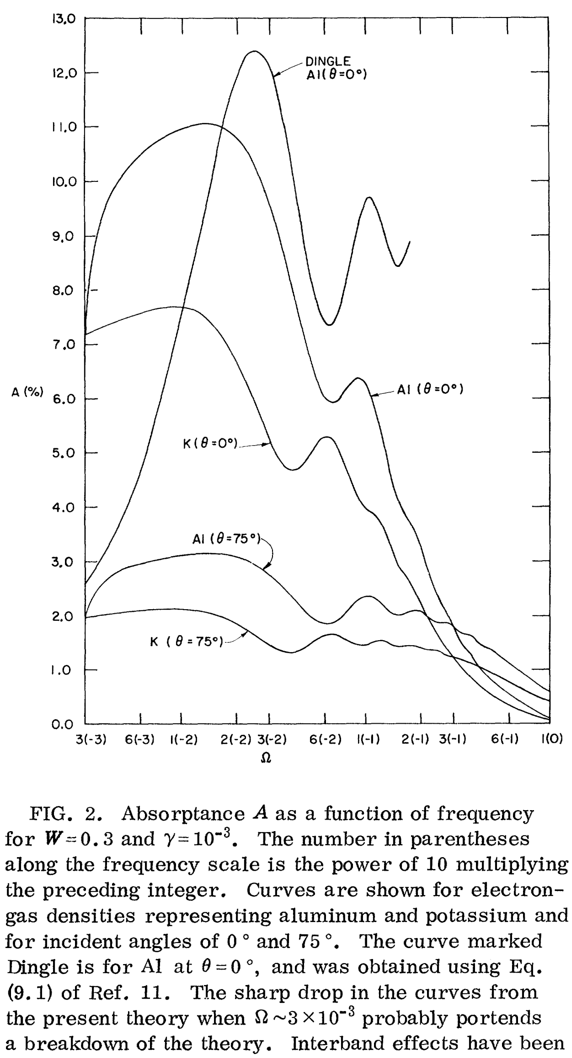

FIG. 2. Absorptance A as a function of frequency for $W = 0.3$ and $\gamma = 10^{-3}$. The number in parentheses along the frequency scale is the power of 10 multiplying the preceding integer. Curves are shown for electron-gas densities representing aluminum and potassium and for incident angles of 0° and 75°. The curve marked Dingle is for Al at $\theta = 0°$, and was obtained using Eq. (9.1) of Ref. 11. The sharp drop in the curves from the present theory when $\Omega \sim 3\times10^{-3}$ probably portends a breakdown of the theory. Interband effects have been neglected.

Also shown in Fig. 2 is the absorptivity resulting from an approximate expression, valid when $W \gg \Omega$, obtained by Dingle.[29] Since this result is inapplicable when the frequency drops down toward the frequency range of the extreme anomalous skin effect, it is clear from Fig. 2 that this expression cannot be used in a quantitative manner for thicknesses such that $W \lesssim 0.3$. However, the oscillatory structure is very similar to that of the exact theory.

The absorptivity oscillations shown in Fig. 2 occur because the surface scattering is diffuse and can be understood in the following way. Consider a plane wave of angular frequency ω ($\omega \sim 0.1\omega_p$) incident normally upon a slab of electron gas of thickness d with the geometry as shown in Fig. 1. Assume for simplicity that the slab is thin ($d \ll c/\omega_p$ or $W \ll 1$) and that the absorptivity is small so we can take the electric field amplitude inside the slab to be constant and equal to that of the incident wave. We consider the relaxation time $\tau \to \infty$, thereby ignoring damping, assume the surface electron scattering is diffuse, and adopt the basic philosophy of Holstein.[25]

We express the y-directed electric field in the slab as

$$E_y = E_0 \cos(\omega t + \alpha), \tag{6.1}$$

where α is a phase angle to be subsequently averaged over. Consider an electron in the slab moving from the surface at $z = 0$ toward the surface at $z = d$ at an angle φ measured with respect to the slab normal. When the electron leaves the front surface it has a velocity $\vec{v}_f$. The velocity of this electron when it reaches the back surface is $\vec{v}_b$, where

$$\vec{v}_b = \vec{v}_f - (eE_0/m)\,\hat{j} \int_0^{d/(v_f \cos\varphi)} \cos(\omega t + \alpha)\, dt, \tag{6.2}$$

and $\hat{j}$ is a unit vector in the y direction. Equation (6.2) yields

$$\vec{v}_b = \vec{v}_f - \frac{eE_0}{m}\hat{j}\left[\sin\left(\frac{\omega d}{v_f \cos\varphi} + \alpha\right) - \sin\alpha\right]. \tag{6.3}$$

The energy change for this electron as it traverses the slab,

$$\Delta E = \tfrac{1}{2} m (\vec{v}_b^{\,2} - \vec{v}_f^{\,2}), \tag{6.4}$$

becomes, after averaging on α,

$$\Delta E = \tfrac{1}{2} m \left(\frac{eE_0}{m\omega}\right)^2 \left[1 - \cos\left(\frac{\omega d}{v_f \cos\varphi}\right)\right]. \tag{6.5}$$

Since we are here dealing with diffuse scattering, we can consider the effect of the surface scattering to be the restoration of the velocity magnitude to the value $|\vec{v}_f|$. Thus, ΔE is the energy extracted from the field and deposited within the slab for a single traversal of the slab by an electron of the type being considered. Since this same energy loss results for electrons leaving the back surface with an initial velocity $-\vec{v}_f$, the rate of energy loss for electrons which leave both surfaces with velocity magnitude $|\vec{v}_f|$ at angle φ is ($\varphi \leq \frac{1}{2}\pi$)

$$\frac{e^2 v_f \cos\varphi E_0^2}{m d \omega^2}\left[1-\cos\left(\frac{\omega d}{v_f \cos\varphi}\right)\right] . \tag{6.6}$$

The number of electrons per unit area with velocities such that $|\vec{v}_f|$ is between v_f and $v_f + dv_f$ and whose direction is within $d\varphi$ about φ is

$$\tfrac{3}{2}(n_0 d/v^3)v_f^2 \sin\varphi \, dv_f \, d\varphi , \tag{6.7}$$

where v is the Fermi velocity. Multiplying (6.6) by (6.7) and integrating over half the Fermi sphere (half because electrons moving both toward and away from a given surface have already been included) yields the total rate of energy loss per unit area

$$\frac{3}{2}\frac{n_0 e^2 E_0^2}{m v^3 \omega^2}\int_0^{\pi/2} d\varphi \int_0^v dv_f \, v_f^3 \sin\varphi\cos\varphi \times\left[1-\cos\left(\frac{\omega d}{v_f\cos\varphi}\right)\right] . \tag{6.8}$$

To further simplify the analysis, let us consider

$$\omega d/v \gg 1, \tag{6.9}$$

so that (6.8) becomes, after averaging over v_f,

$$\frac{3}{2}\frac{n_0 e^2 E_0^2}{m v^3 \omega^2}\left[\frac{v^4}{8}+\frac{v^5}{\omega d}\int_0^{\pi/2} d\varphi \sin\varphi\cos^2\varphi \sin\left(\frac{\omega d}{v\cos\varphi}\right)\right] . \tag{6.10}$$

Doing the final integral in (6.10) using (6.9) yields for the total rate of energy loss per unit area

$$\frac{3}{16}\frac{n_0 e^2 v}{m}\frac{E_0^2}{\omega^2}\left[1+8\left(\frac{v}{\omega d}\right)^2\cos\left(\frac{\omega d}{v}\right)\right] . \tag{6.11}$$

Dividing (6.11) by the incident flux $cE_0^2/8\pi$ yields the absorptance

$$A=\frac{3}{8}\frac{v}{c}\frac{1}{\Omega^2}\left[1+8\left(\frac{v}{c}\frac{1}{W\Omega}\right)^2\cos\left(\frac{W\Omega}{v/c}\right)\right] , \tag{6.12}$$

which is valid for

$$W \ll 1, \tag{6.13a}$$

$$\frac{W\Omega}{v/c} \gg 1, \tag{6.13b}$$

$$\frac{v/c}{\Omega^2} \ll 1. \tag{6.13c}$$

Equation (5.12) yields an oscillatory absorptivity with maxima when

$$\frac{W\Omega}{v/c} = 2\pi n, \tag{6.14}$$

where n is a positive integer,[30] which is consistent with the results shown in Fig. 2. In addition, Eq. (6.12) agrees well with the general theory when damping is ignored and the conditions (6.13) are satisfied.

Expression (6.14) indicates that the absorptivity maxima occur when the time for an electron to cross the slab in a normal direction d/v is an integer times the period T of the incident radiation, or

$$d/(vT) = n. \tag{6.15}$$

Now this is not the result which would be expected from a simple resonance-type idea. That is, absorptivity maxima might be expected to occur when

$$d/(vT) = \tfrac{1}{2}(2k-1) , \tag{6.16}$$

where k is a positive integer, since this condition corresponds to an electron with the Fermi velocity crossing the slab normally while the electric field goes through an odd number of half cycles of oscillation. Condition (6.16) does appear, in a sense, at an intermediate point in the derivation leading to Eq. (6.12). Consider the integrand in Eq. (6.10).[31] If we eliminate the factor associated with the number of electrons in $d\varphi$, $\sin\varphi \, d\varphi$, then what remains is

$$\cos^2\varphi \sin(\omega d/v\cos\varphi) . \tag{6.17}$$

This expression has maxima when $\varphi = 0$ and $\omega d/v = (2k-1)\pi$, or $(d/v)/T = \frac{1}{2}(2k-1)$, which is just condition (6.16). Thus, when considering one electron moving with the Fermi velocity, the rate of energy loss is maximized consistent with the simple resonance idea. However, when (6.17) is angular averaged, as in Eq. (6.10), rather than maximized in angle, the positions of the absorptivity maxima occur as given by (6.14) or (6.15). Thus, it is the angular average which finally determines the exact character of the oscillatory absorptance.

Condition (6.14) for the absorptance maxima is symmetric in W and Ω. However, the structure in A as a function of W for fixed Ω is less pronounced than that of Fig. 2, particularly for large incident angles. Absorptivity curves are shown in Fig. 3 for $\Omega = 0.05$ and in Fig. 4 for $\Omega = 0.15$; in both figures $\gamma = 10^{-3}$. For an incident angle of

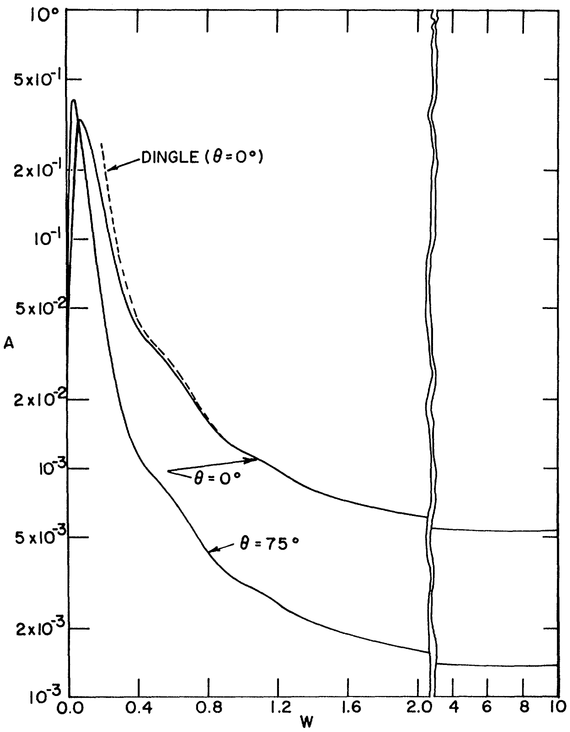

FIG. 3. Absorptance A of aluminum as a function of thickness for $\Omega = 0.05$ and $\gamma = 10^{-3}$. Curves for $\theta = 0°$ and $\theta = 75°$ are given. The curve marked Dingle was obtained using Eq. (9.1) of Ref. 11. Interband effects have been ignored.

75°, the oscillations have been reduced to gentle undulations. As the frequency increases, the structure moves toward smaller thicknesses as is clear from (6.14). Note that the absorptivity as a function of thickness has an absolute maximum at a very small thickness.

Also shown in Figs. 3 and 4 are curves obtained using Dingle's approximate expression,[29] a result which clearly becomes more accurate as the thickness increases. It should be noted, however, that damping is included in Dingle's expression only in the fashion that it appears in Eq. (5.1). In fact, the limit of Dingle's expression as $W \to \infty$ is just Eq. (5.1) as noted above. This means that the expression of Dingle can be valid only under conditions where (5.6) reduces to (5.1) when $\theta = 0$, in addition to the requirement that $W \gg \Omega$.

Reflectance and transmittance curves as a function of thickness for the conditions corresponding to the absorptances of Figs. 3 and 4 are given in Figs. 5 and 6. In general, structure in A does not result in apparent structure in either T or R. Figure 6 does indicate that this is not always the case.

For thick crystals and infrared frequencies, Eq. (5.6) contains correction terms to Eq. (5.1) which can be important. As an illustration consider the case of aluminum for which the poorest agreement between theory and experiment was reported by Biondi and Goubadia.[24] Recent measurements by Bos and Lynch[32] have shown: (i) There is an interband peak with a threshold somewhat above 0.26 eV although interband effects may extend to lower frequencies. (ii) For an energy of 0.26 eV, or $\Omega = 0.020$, $A = 0.90\%$ for normal incidence. (iii) The effective value of γ for $\Omega \sim 0.020$ is 3.7×10^{-3}. Since the room-temperature to liquid-helium-temperature resistance ratio was 3000, this suggests strongly that interband effects slowly varying with frequency are occurring for these energies in addition to the peak noted in (i) above. For the above conditions, Eq. (5.1) yields $A_{uc} = 1.08\%$. From Eq. (5.6), we obtain $A = 1.04\%$ for $\Omega = 0.02$ and $A = 0.97\%$ for $\Omega = 0.01$. These latter results differ significantly from A_{uc} and point up the inadequacy of A_{uc} for this frequency range. The fact that the calculated A is larger than the experimental A could result

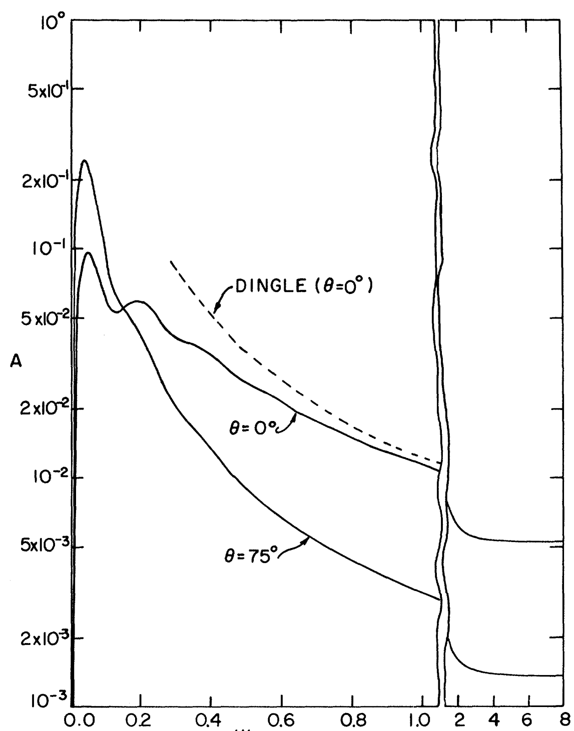

FIG. 4. Absorptance A of aluminum as a function of thickness for $\Omega = 0.15$ and $\gamma = 10^{-3}$. Curves for $\theta = 0°$ and $\theta = 75°$ are given. The curve marked Dingle was obtained using Eq. (9.1) of Ref. 11. Interband effects have been ignored.

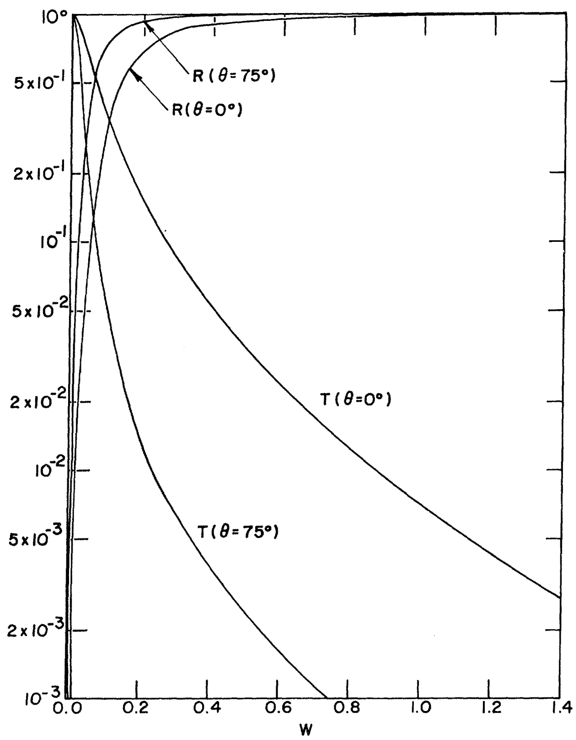

FIG. 5. Reflectance R and transmittance T for aluminum as a function of thickness for $\Omega = 0.05$ and $\gamma = 10^{-3}$. Interband effects have been ignored.

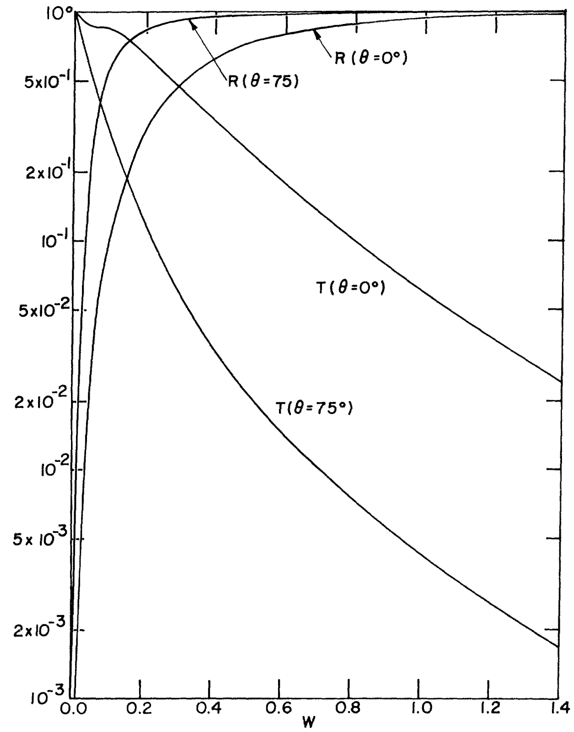

FIG. 6. Reflectance R and transmittance T for aluminum as a function of thickness for $\Omega = 0.15$ and $\gamma = 10^{-3}$. Interband effects have been ignored.

from γ_{eff} being smaller than 3.7×10^{-3} or the surface electron scattering being partially specular rather than completely diffuse, as assumed.

The absorptance for a semi-infinite sample is shown in Fig. 7. These curves were obtained using Eq. (3.1) [33] for $\Omega < 10^{-2}$ and the above developed theory[34] with $W \to \infty$ for $\Omega > 10^{-2}$. A comparison of these curves with those for specular scattering given in Refs. 1 and 3 indicates that the absorptance dip occurring for specular scattering as the frequency moves above that of the extreme anomalous skin effect does not occur for diffuse

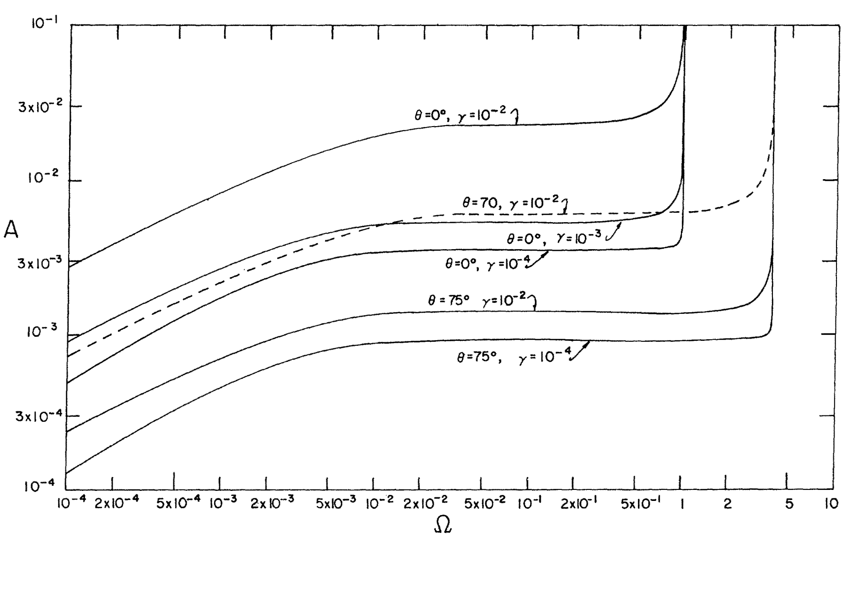

FIG. 7. The absorptance for a semi-infinite aluminum sample as a function of frequency. Curves for $\theta = 0°$ and $\theta = 75°$ are given for $\gamma = 10^{-4}$, 10^{-3}, and 10^{-2}. The sharp absorptance rise occurs near $\Omega = (\cos\theta)^{-1}$ or $\Omega = 1$ for $\theta = 0°$, and $\Omega = 3.86$ for $\theta = 75°$. Interband effects have been ignored.

scattering. For the semi-infinite sample, the absorptance is somewhat more sensitive to moderate damping than it is for a thin film.

Optical properties calculations were also performed incorporating the experimentally determined interband dielectric function for Al. These effects do not obscure the oscillatory absorptance structure if the resulting absorptance is less than it would be for $\epsilon = 1$ and $\gamma \sim 10^{-2}$.

Finally, we wish to comment on the fact that non-normal angles of incidence manifest themselves in the absorptance essentially through a multiplicative factor $\cos\theta$. This is the fashion in which the approximate expression (6.12) changes for $\theta \neq 0$, which indicates that the oscillatory structure is nearly independent of the angle of incidence. Crudely, this occurs since the dielectric function of the metal is high for $\Omega \lesssim 10^{-1}$, and thus, through a somewhat questionable use of Snell's law, one can conclude that the "propagation direction" of the fields within the metal is roughly normal to the surface, independent of the incident angle. Hence the $\cos\theta$ factor, arising from the incident flux, is essentially the only source of θ dependence.

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APPENDIX

The exponential integrals $I_n(x)$ defined by Eq. (4.38) must be evaluated with some care since the argument

$$x = \frac{iW(\Omega + i\gamma)}{v/c} \tag{A1}$$

has an imaginary part which is much larger than the magnitude of the real part for most cases of interest. It is only necessary to evaluate $I_1(x)$ numerically since the recurrence relation

$$I_n(x) = (n-1)^{-1}\left[e^x + xI_{n-1}(x)\right] \tag{A2}$$

can be used to obtain I_n for $n > 1$.

It is not difficult to show that $I_1(x)$ is given, in general, by

$$I_1(x) = -C - \ln(-x) - \sum_{j=1}^{\infty} \frac{x^j}{j!\,j} \quad , \tag{A3}$$

where $C = 0.5772157\ldots$ is Euler's constant. However, in the present application the imaginary part of x can be as large as 10^3. Thus, the series (A3) is of questionable utility is some cases. The asymptotic series

$$I_1(x) = -\frac{e^x}{x}\left(1 + \frac{1!}{x} + \frac{2!}{x^2} + \frac{3!}{x^3} + \cdots + \frac{k!}{x^k} + \cdots\right) \tag{A4}$$

can then be used. For the parameters of interest here, $W \lesssim 10$, $10^{-3} \lesssim \Omega \lesssim 10$, $\gamma \lesssim 10^{-1}$, $v/c \sim 10^{-2}$, we have determined empirically that this asymptotic series yields accurate results when $|x| > 14$.

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[1]K. L. Kliewer and R. Fuchs, Phys. Rev. 172, 607 (1968).

[2]W. E. Jones, K. L. Kliewer, and R. Fuchs, Phys. Rev. 178, 1201 (1969).

[3]R. Fuchs and K. L. Kliewer, Phys. Rev. 185, 905 (1969).

[4]A. R. Melnyk and M. J. Harrison, Phys. Rev. Letters 21, 85 (1968).

[5]F. Forstmann, Z. Physik 203, 495 (1967).

[6]R. J. Esposito, P. E. Bloomfield, and L. Muldawer, Phys. Rev. 176, 886 (1968).

[7]G. E. H. Reuter and E. H. Sondheimer, Proc. Roy. Soc. (London) A195, 336 (1948).

[8]R. B. Dingle, Physica 19, 311 (1953).

[9]D. C. Mattis and G. Dresselhaus, Phys. Rev. 111, 403 (1958).

[10]H. E. Bennett, J. M. Bennett, E. J. Ashley, and R. J. Motyka, Phys. Rev. 165, 755 (1968).

[11]R. B. Dingle, Physica 19, 1187 (1953).

[12]This definition is unconventional but convenient for optical problems. See footnote 12 of Ref. 1.

[13]R. Fuchs, K. L. Kliewer, and W. J. Pardee, Phys. Rev. 150, 589 (1966).

[14]The single relaxation-time approximation can be shown to be good when the electron scattering is from impurities. See Ref. 15 and S. H. Liu, Ann. Phys. (N. Y.) 59, 165 (1970).

[15]K. L. Kliewer and R. Fuchs, Phys. Rev. 181, 552 (1969).

[16]J. Lindhard, Kgl. Danske Videnskab. Selskab, Mat.-Fys. Medd. 28, No. 8 (1954).

[17]The difference between the results using the above transverse dielectric function and the results using the transverse dielectric function obtained from the Boltzmann equation is, however, very small. See Ref. 3.

[18]Treating core polarization and interband effects via a local dielectric function is a reasonable approximation for s polarization but not for p polarization (see Refs. 1–3). Since we are here interested in s polarization, we shall so proceed.

[19]$v/c \sim 10^{-2}$.

[20]The determining equation for α, Eq (4.25), also involves a power series in $(v \sin\theta/c)^2$.

[21]The fact that field terms having both $\pm\alpha$ appear is the analog of the usual local result wherein terms appear corresponding to plane waves moving in both directions within a film. Thus, P is a measure of the

extent to which the wave moving in the film is reflected at the back surface.

[22]The transverse dielectric function as obtained from the Boltzmann equation is given, for example, by Eq. (2.46) of Ref. 1. To obtain Eq. (2.46) of Ref. 1, one need only replace α in Eq. (4.35) above by $ib'Q$, where Q is the magnitude of the wave vector and $b' = (v/\gamma c)/(1 - i\Omega/\gamma)$.

[23]J. N. Hodgson, J. Phys. Chem. Solids 24, 1213 (1963).

[24]M. A. Biondi and A. I. Guobadia, Phys. Rev. 166, 667 (1968).

[25]T. Holstein, Phys. Rev. 88, 1427 (1952).

[26]Equation (5.1) results if the $\vec{d} \rightarrow \infty$ limit is taken in Eq. (9.1) of Ref. 11.

[27]$W = 0.3$ corresponds to a film thickness of 136 Å for K, and 47 Å for Al.

[28]Both δ and α are of order 1 for $\Omega \sim 10^{-3}$ and $\gamma \sim 10^{-3}$.

[29]The expression in Eq. (9.1) of Ref. 11.

[30]Note that this is consistent with (6.13b) and (6.9).

[31]This expression, having already been averaged over v_f, involves only the Fermi velocity, and, thus, corresponds to the general theory wherein physical effects occur only for electrons with the Fermi velocity because of the factor $\partial f_0/\partial \epsilon_{\vec{k}}^*$ appearing, for example, in Eqs. (4.11).

[32]L. Bos and D. W. Lynch, Phys. Rev. Letters 25, 156 (1970); D. W. Lynch (private communication).

[33]Equation (3.1) for the surface impedance can be integrated exactly in the region of the extreme anomalous skin effect. See L. E. Hartmann and J. M. Luttinger, Phys. Rev. 151, 430 (1966); 156, 1038 (1967).

[34]Obviously, Eq. (3.1) can be used for all frequencies. However, numerical integration of Eq. (3.1) becomes particularly time consuming when $\Omega > 10^{-1}$. In this frequency range the present theory with $W \rightarrow \infty$ is a much more efficient way to obtain results for thick samples.

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Effect of Hydrostatic Pressure on the Fermi Surface of Bi†

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We present data on the pressure dependence of the smallest extremal cross-sectional areas of both the hole and electron sheets of the Fermi surface of Bi. The cross sections were determined from measurements of the frequency of de Haas–van Alphen oscillations detected with the field modulation technique. Pressure derivatives obtained using solid He and fluid He were in excellent agreement, thus removing any doubts about the hydrostatic nature of the stress in the solid-He portions of the work. Values for the pressure derivatives of the effective masses associated with these cross sections were obtained from magnetization-amplitude-versus-temperature measurements in solid He to ~4 kbar. Our values for the pressure derivatives of the cross sections and masses differ in detail from and have somewhat less uncertainty than the most recent literature values. No change in the initial phase of the de Haas–van Alphen oscillations associated with the smallest extremal cross-sectional area of the hole sheet was detected in contrast to the findings of Overton and Berlincourt.

I. INTRODUCTION

Bi was the first metal in which studies of the size and shape of the Fermi surface as a function of pressure were attempted.[1–3] Although much of the earlier work suffered from the inadequacies of the pressure techniques in achieving hydrostatic compression on nonisotropic crystals, it was established that large changes in the electronic energy spectrum were occurring with pressure. These large changes are not surprising in view of the anisotropy of the structure and elastic properties of Bi.

Bi crystallizes in the arsenic structure, which can be thought of as two face-centered-cubic sublattices displaced from each other along a body diagonal with a small rhombohedral distortion along this diagonal. The effect of pressure on the Bi lattice[4] is to change the dimension in the trigonal direction at the fractional rate of -18.1×10^{-4} kbar^{-1} and the dimension perpendicular to the trigonal axis at -6.4×10^{-4} kbar^{-1}. The distance between the sublattices or the atomic positional parameter increases[5] at the fractional rate of $\sim 11 \times 10^{-4}$ kbar^{-1}.

The Fermi surface of Bi is quite simple, consisting of nearly ellipsoidal hole and electron sheets.[6] The hole sheets are located near the point T in the Brillouin zone with their long axes parallel to the trigonal direction. The electron sheets are in the trigonal-bisectrix plane with their long axes tipped $\sim 96°$ from the trigonal direction in the sense T toward X.[7]