$$\hat{\sigma}_t=\begin{bmatrix} \frac{3}{2}(\sigma_{11}+\sigma_{22}) & \frac{3}{2}(\sigma_{12}-\sigma_{21}) & 0 \\ -\frac{3}{2}(\sigma_{12}-\sigma_{21}) & \frac{3}{2}(\sigma_{11}+\sigma_{22}) & 0 \\ 0 & 0 & 3\sigma_{33} \end{bmatrix}. \tag{47b}$$

Then the conductivity related to experiment is

$$\hat{\sigma}_t=Nec\begin{bmatrix} a_iH_iL & -HL & 0 \\ HL & a_iH_iL & 0 \\ 0 & 0 & b_iH_iL+c_i \end{bmatrix}, \tag{48a}$$

where

$$a_i\equiv\tfrac{1}{2}[(\alpha_{11}/\alpha_{22})^{1/2}+(\alpha_{22}/\alpha_{11})^{1/2}], \tag{48b}$$

$$b_i\equiv(\alpha_{23})^2(\alpha_{11})^{-1/2}(\alpha_{22})^{-3/2}, \tag{48c}$$

$$c_i\equiv\alpha_2\alpha_3(\alpha_{22})^{-1}e\tau/c, \tag{48d}$$

$$N\equiv 3n. \tag{48e}$$

SUMMARY

The foregoing example illustrates the ease with which one may write the total conductivity tensor for a Fermi surface consisting of a set of ellipsoids symmetrically placed about the direction of the magnetic field. It should be clear, however, that even for a set of ellipsoids having no symmetry with respect to the direction of the magnetic field one may follow the above recipe to determine the total conductivity. One need only write the $\hat{\alpha}$ tensors describing the various ellipsoids in a common coordinate system chosen such that the magnetic field is along the 3 axis. The rest of the problem is simply to perform the indicated tensor multiplications and then to add up the individual conductivities. Corresponding expressions for $\hat{\epsilon}''$, $\hat{\lambda}''$, and $\hat{\pi}''$ are obtained in an obvious manner [see Eq. (26)]. One may choose to represent the spherical geometry solutions in an approximation different from that of the Sondheimer-Wilson theory,[2] but the method of obtaining the ellipsoidal geometry solutions remains the same.

PHYSICAL REVIEW VOLUME 172, NUMBER 3 15 AUGUST 1968

Anomalous Skin Effect for Specular Electron Scattering and Optical Experiments at Non-Normal Angles of Incidence*

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Received 14 March 1968

The anomalous skin effect for specular electron scattering at the metal surface is studied, permitting the impinging plane wave to have an arbitrary angle of incidence. It is shown that the expressions for the surface impedance for a non-normal angle of incidence obtained by Reuter and Sondheimer as a generalization from their work at normal incidence are correct for S polarization but incorrect for P polarization. The correct surface impedance for P polarization leads to an additional absorption peak in the frequency range $10^{-2}\omega_p\lesssim\omega\lesssim\omega_p$, where ω_p is the free-electron plasma frequency. This additional absorption, particularly pronounced for long electron lifetimes, is investigated in detail. One important conclusion drawn from this work is that, in general, optical experiments performed at non-normal angles of incidence cannot be analyzed in terms of a single complex frequency-dependent dielectric function. In the frequency range of the additional P absorption, two such dielectric functions are needed, one function for describing P polarization and a different function for describing S polarization.

I. INTRODUCTION

The theory by which the anomalous skin effect was incorporated into the general theory of the optical properties of metals was developed in detail by Reuter and Sondheimer[1] and Dingle[2] for both specular and diffuse electron scattering at the metal surface. This work, utilizing the Boltzmann equation, treated the case of a plane wave incident normally on the metal surface, although conclusions were drawn concerning also the effect of non-normal incidence. A quantum-mechanical treatment of the anomalous skin effect at normal incidence with specular reflection was given by Mattis and Bardeen[3]; their result for the surface impedance was in agreement with that of Reuter and Sondheimer.

A recent study of the classical optical properties of an electron gas by the present authors[4] indicated that interesting absorption structure can occur at non-normal angles of incidence, structure that possesses no counterpart at normal incidence. This fact, together with the appearance of the striking results of optical studies performed by Mayer and his co-workers[5] on the alkali metals at large angles of incidence, suggested to us that a reexamination of the theory of the anomalous

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* Work performed in the Ames Laboratory of the U. S. Atomic Energy Commission. Contribution No. 2269.

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

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

[3] D. C. Mattis and J. Bardeen, Phys. Rev. **111**, 412 (1958).

[4] K. L. Kliewer and R. Fuchs, Phys. Rev. **153**, 498 (1967).

[5] H. Mayer and B. Hietel, in *Optical Properties and Electronic Structure of Metals and Alloys*, edited by F. Abeles (North-Holland Publishing Co., Amsterdam, 1966), p. 47, and references cited therein.

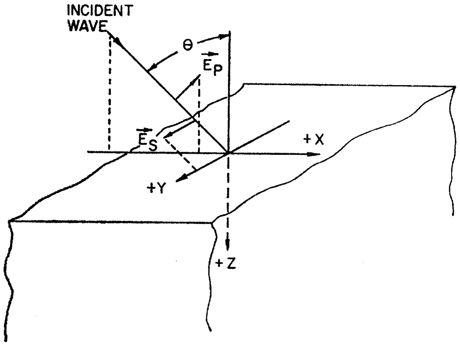

FIG. 1. Orientation of the semi-infinite metal showing the electric field of the incident wave for P polarization, $\mathbf{E}_P$, and for S polarization, $\mathbf{E}_S$. The angle of incidence is given by θ.

skin effect at non-normal incidence was warranted. This reexamination begins in the present paper with a discussion, based upon the Boltzmann equation, of the case of specular electron reflection.

A careful study of Refs. 1 and 2 suggests that there remain no unanswered questions for the case of specular reflection; that is, that the conclusions drawn for non-normal incidence from the detailed calculation at normal incidence are valid and complete. However, there are two points that indicate that this may not be the case. The first of these concerns the technique used by Reuter and Sondheimer to obtain the surface impedance for normal incidence. At normal incidence, $\nabla\cdot\mathbf{E}=0$ within the metal, where $\mathbf{E}$ is the electric field. Thus there occur no charge fluctuations within the metal, since the electric field is always parallel to the surface. If, however, one permits $\nabla\cdot\mathbf{E}\neq 0$, then, when there exists an electric-field component perpendicular to the surface, as will be the case for non-normal incidence with P-polarized fields, there can occur a charge imbalance penetrating the metal. The interaction of the electrons with this charge fluctuation would then be an additional absorption mechanism. As is true for similar situations where moving charged particles interact with self-generated longitudinal fields, we expect that the energy absorption in this case would be characterized by the imaginary part of $\epsilon_l(\mathbf{q},\omega)^{-1}$, where $\epsilon_l(\mathbf{q},\omega)$ is the nonlocal longitudinal dielectric constant of the system. The first point we wish to make here is that, when generalizing normal incidence results to non-normal incidence, one is going from a case where charge fluctuations do not exist to a case where they do. Since the mathematical description of these distinctly different physical situations must reflect the difference, it would appear that a possibly important absorption mechanism could be overlooked or treated incorrectly when making arguments based upon normal incidence conclusions.

Now, a reader familiar with the effects of an electric field normal to the surface of a metal might at this point counter with the argument that the screening distance for a charge imbalance of the sort discussed above should be the Fermi-Thomas screening distance λ_{FT} given by $\lambda_{\mathrm{FT}}=v_F/(\sqrt{3}\omega_p)$, where v_F is the Fermi velocity and ω_p the plasma frequency. Since $\lambda_{\mathrm{FT}}\lesssim 1$ Å, this would lead one to conclude that the screening is so effective that only very minor effects could be associated with the presence of the charge fluctuations. However, the utilization of λ_{FT} as a measure of the screening length is valid only for frequencies such that $\omega\tau\ll 1$, where τ is the effective electron lifetime. For higher frequencies, $\omega\tau\gg 1$, the longitudinal dielectric constant $\epsilon_l(\mathbf{q},\omega)$ is much more involved than it is in the low-frequency limit [see Eq. (2.47) below], and the simple screening concept breaks down. The above argument does indicate, though, that any important effects associated with the presence of the charge fluctuations will occur for frequencies where $\omega\tau\gg 1$.

The second, though related, point concerns the structure of the scattering term in the Boltzmann equation. When charge fluctuations are present, the relaxation of the perturbed distribution toward "equilibrium" will be to the local state of charge imbalance and not to the uniform distribution.[6] Thus a simple relaxation time approximation for the scattering term will not be adequate. These effects will be incorporated into the theory developed below. It should be emphasized that these points affect the calculation only when non-normal incidence is involved, and thus our results will reduce to those of Reuter and Sondheimer[1] for normal incidence.

The geometry used for the calculation is shown in Fig. 1. The metal is considered to be semi-infinite with the surface an x-y plane at $z=0$; the positive z direction is into the metal. When discussing $P(S)$ polarization the incident electric field will be that denoted $\mathbf{E}_P(\mathbf{E}_S)$ in Fig. 1.

A detailed solution, using the appropriate Boltzmann equation, for the surface impedance in the case of P polarization is given in Sec. II. This derivation includes a calculation of the longitudinal and transverse dielectric functions valid for the problem at hand. An alternative treatment of the problem based upon a general dielectric-constant tensor is given in Sec. III for both S and P polarization. The results are discussed in Sec. IV.

Although the emphasis in the present paper is on the basically free-electron properties of a metal, interband effects can be included in a phenomenological fashion. These interband effects will be considered in detail in a following paper.[7]

II. THEORY: BOLTZMANN-EQUATION APPROACH

Considering the magnetic permeability to be unity, Maxwell's equation for the electric field $\mathbf{E}(\mathbf{r})$ can here

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[6] J. L. Warren and R. A. Ferrell, Phys. Rev. 117, 1252 (1960).

[7] R. Fuchs and K. L. Kliewer (to be published).

be written as[2]

$$\nabla^2\mathbf{E}(\mathbf{r})-\nabla(\nabla\cdot\mathbf{E}(\mathbf{r}))+(\omega^2/c^2)(1+\epsilon_{\mathrm{IB}})\mathbf{E}(\mathbf{r})=-(4\pi i\omega/c^2)\mathbf{J}(\mathbf{r})\,, \quad (2.1)$$

where it has been assumed that the time dependence of all field and current components is $\exp(-i\omega t)$. $\mathbf{J}(\mathbf{r})$ is the current density, and c is the velocity of light. The quantity ϵ_{IB} represents the phenomenological inclusion of interband effects in the theory; that is, ϵ_{IB} is the (generally frequency-dependent) interband contribution to the dielectric response function. For simplicity, we take ϵ_{IB} to be zero here but will indicate below where it would reappear.

Since we are now interested in P polarization, with the electric field in the x-z plane as shown in Fig. 1, we can write[8]

$$\nabla\cdot\mathbf{E}(\mathbf{r})=\left(iq_xE_x(z)+\frac{\partial E_z(z)}{\partial z}\right)e^{iq_xx}\,, \quad (2.2)$$

where use has been made of the assumption that all fields and currents have a spatial dependence of the form

$$\mathbf{F}(\mathbf{r})=\mathbf{F}(z)e^{iq_xx}.$$

All field and current components appearing below without argument represent the z-dependent part of these quantities. Using (2.2), Eq. (2.1) can be written in component from as

$$\frac{d^2E_x}{dz^2}+\frac{\omega^2}{c^2}E_x-iq_x\frac{dE_z}{dz}=-\frac{4\pi i\omega}{c^2}J_x \quad (2.3\text{a})$$

and

$$\left(\frac{\omega^2}{c^2}-q_x^2\right)E_z-iq_x\frac{dE_x}{dz}=-\frac{4\pi i\omega}{c^2}J_z\,. \quad (2.3\text{b})$$

To proceed further with these equations requires expressions for J_x and J_z, which we now obtain from the Boltzmann equation.

For free electrons and field wavelengths long compared to the wavelength of an electron at the Fermi surface, the Boltzmann equation can here be written[9]:

$$\frac{\partial f_{\mathbf{k}}}{\partial t}-\frac{e}{\hbar}\mathbf{E}(\mathbf{r},t)\cdot\nabla_{\mathbf{k}}f_{\mathbf{k}}+\mathbf{v}\cdot\nabla_{\mathbf{r}}f_{\mathbf{k}}=-\sum_{\mathbf{k}'}W_{\mathbf{k}\mathbf{k}'}(f_{\mathbf{k}}-f_{\mathbf{k}'})\,, \quad (2.4)$$

where $f_{\mathbf{k}}$ is the one-particle distribution function, $\mathbf{v}=\hbar\mathbf{k}/m$ is the electron velocity, m is the electron mass, and e is the magnitude of the electronic charge. In addition it has been assumed that the scattering processes are elastic so that the scattering transition rates obey

$$W_{\mathbf{k}\mathbf{k}'}=W_{\mathbf{k}'\mathbf{k}}.$$

It is obvious that we are not making the simple relaxation approximation in the scattering term on the right-hand side of Eq. (2.4). This approximation is not legitimate in the problem under consideration here for the reasons discussed in the introduction. Relaxation to the local distribution is included by writing the Boltzmann equation in the form (2.4).

We now linearize Eq. (2.4) in a fashion consistent with the $\exp(iq_xx-i\omega t)$ dependence of the electric field by writing

$$f_{\mathbf{k}}=f_0+f_1(\mathbf{v},z)e^{iq_xx}e^{-i\omega t}\,, \quad (2.5)$$

where

$$f_0=[e^{\beta(\epsilon_k-\mu)}+1]^{-1}\,,$$
$$\beta=(k_BT)^{-1}\,,$$

μ is the electronic chemical potential, and the as yet undetermined function f_1 is proportional to the field. If only the lowest-order terms in the field are retained, (2.4) can be written

$$\frac{\partial f_1(\mathbf{v},z)}{\partial z}+\xi f_1(\mathbf{v},z)=e\left(\frac{v_x}{v_z}E_x+E_z\right)\left(\frac{\partial f_0}{\partial\epsilon_k}\right)+\frac{1}{v_z}\sum_{\mathbf{k}'}W_{\mathbf{k}\mathbf{k}'}f_1(\mathbf{v}',z)\,, \quad (2.6)$$

where

$$\xi=\frac{1}{v_z}\left[\frac{1}{\tau}+i(q_xv_x-\omega)\right],$$

and the relaxation time, dependent only upon the energy ϵ_k, is defined by

$$\frac{1}{\tau(\epsilon_k)}=\sum_{\mathbf{k}'}W_{\mathbf{k}\mathbf{k}'}\,.$$

A formal solution of Eq. (2.6) is

$$f_1(\mathbf{v},z)=e^{-\xi z}\left\{F(\mathbf{v})+e\left(\frac{\partial f_0}{\partial\epsilon_k}\right)\int_0^z\left(\frac{v_x}{v_z}E_x(s)+E_z(s)\right)e^{\xi s}ds+\frac{1}{v_z}\left(\frac{\partial f_0}{\partial\epsilon_k}\right)\int_0^zG(s)e^{\xi s}ds\right\}, \quad (2.7)$$

where $F(\mathbf{v})$ is an arbitrary function of $\mathbf{v}$ and $G(z)$ is introduced because of the self-consistency requirement resulting from the presence of the last term of (2.6); that is, $G(z)$ must be so determined that

$$\left(\frac{\partial f_0}{\partial\epsilon_k}\right)G(z)=\sum_{\mathbf{k}'}W_{\mathbf{k}'\mathbf{k}}f_1(\mathbf{v}',z)\,. \quad (2.8)$$

Consider now the electrons moving toward the surface, those electrons which have $v_z<0$. The distribution function f_1' for such electrons must approach zero as $z\to\infty$ and thus the corresponding value of $F(\mathbf{v})$,

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[8] Note that at this point the fact that $\nabla\cdot\mathbf{E}\neq0$ is used explicitly.

[9] G. V. Chester, Proc. Phys. Soc. (London) 81, 938 (1963).

$F'(\mathbf{v})$, is given by

$$F'(\mathbf{v})=-e\left(\frac{\partial f_0}{\partial\epsilon_k}\right)\int_0^\infty\left(\frac{v_x}{v_z}E_x(s)+E_z(s)\right)e^{\xi s}ds-\frac{1}{v_z}\left(\frac{\partial f_0}{\partial\epsilon_k}\right)\int_0^\infty G(s)e^{\xi s}ds\,,$$

from which follows

$$f_1'(\mathbf{v},z)=e^{-\xi z}\left(\frac{\partial f_0}{\partial\epsilon_k}\right)\left\{-e\int_z^\infty ds\left(\frac{v_x}{v_z}E_x+E_z\right)e^{\xi s}-\frac{1}{v_z}\int_z^\infty G(s)e^{\xi s}ds\right\}.\quad(2.9)$$

The distribution function $f_1''(\mathbf{v},z)$ for electrons moving away from the surface ($v_z>0$) depends upon the nature of the surface scattering. Since we are herein considering specular reflection, we have the condition

$$f_1''(v_x,v_y,v_z,z=0)=f_1'(v_x,v_y,-v_z,z=0)\,,$$

and so

$$f_1''(\mathbf{v},z=0)=\left(\frac{\partial f_0}{\partial\epsilon_k}\right)\left\{-e\int_0^\infty ds\left(-\frac{v_x}{v_z}E_x+E_z\right)e^{-\xi s}+\frac{1}{v_z}\int_0^\infty G(s)e^{-\xi s}ds\right\}.\quad(2.10)$$

From (2.7), this can also be written

$$f_1''(\mathbf{v},z=0)=F''(\mathbf{v})\,,\quad(2.11)$$

where $F''(\mathbf{v})$ is the value of $F(\mathbf{v})$ associated with electrons having $v_z>0$. We therefore find

$$f_1''(\mathbf{v},z)=e^{-\xi z}\left(\frac{\partial f_0}{\partial\epsilon_k}\right)\left[-e\int_0^\infty ds\left(-\frac{v_x}{v_z}E_x+E_z\right)e^{-\xi s}+e\int_0^z ds\left(\frac{v_x}{v_z}E_x+E_z\right)e^{\xi s}+\frac{1}{v_z}\int_0^\infty ds\,G(s)e^{-\xi s}+\frac{1}{v_z}\int_0^z ds\,G(s)e^{\xi s}\right].\quad(2.12)$$

Defining now

$$E_x(z)=E_x(-z)\,,\quad(2.13a)$$

$$E_z(z)=-E_z(-z)\,,\quad(2.13b)$$

$$G(z)=G(-z)\,,\quad(2.13c)$$

Eq. (2.12) can be written

$$f_1''(\mathbf{v},z)=e^{-\xi z}\left(\frac{\partial f_0}{\partial\epsilon_k}\right)\left[e\int_{-\infty}^z ds\left(\frac{v_x}{v_z}E_x+E_z\right)e^{\xi s}+\frac{1}{v_z}\int_{-\infty}^z ds\,G(s)e^{\xi s}\right].\quad(2.14)$$

The symmetry requirements (2.13a) and (2.13b) are suggested by Maxwell's equations, whereas (2.13c) will be seen below to be a consequence of the prior two.

Having now equations for the distributions, we can obtain the current

$$\mathbf{J}(z)=-2e\left(\frac{m}{h}\right)^3\int\mathbf{v}f_1(\mathbf{v},z)d^3v\,.\quad(2.15)$$

If one writes the volume element in velocity space in spherical coordinates, the azimuthal integration can be performed immediately using the fact that[10]

$$\int_0^{2\pi}\cos^n\varphi e^{iA\cos\varphi}d\varphi=\frac{2\pi}{i^n}J_0^{(n)}(A)\,,$$

where $J_0^{(n)}(A)$ is the nth derivative of the zero-order Bessel function with respect to its argument. Recalling now that

$$\left(\frac{\partial f_0}{\partial\epsilon_k}\right)\simeq-\delta(\epsilon-\epsilon_F)\,,\quad(2.16)$$

where ϵ_F is the Fermi energy, permits immediate evaluation of the integrals over the magnitude of the velocity. The integrals over the polar angle must be done separately for $v_z>0$ (where f_1'' must be used) and $v_z<0$ (where f_1' is needed). These latter integrals cannot, at this point, be evaluated in closed form, so our expressions for the current components are

$$J_x(z)=\left(\frac{4\pi m^2e^2v^2}{h^3}\right)\left\{-\int_{-\infty}^\infty ds\,E_x(s)K''(z-s)+i\int_{-\infty}^\infty ds\,E_z(s)K'(z-s)+\frac{i}{ev}\int_{-\infty}^\infty ds\,G(s)\bar{K}'(z-s)\right\}\quad(2.17a)$$

and

$$J_z(z)=\left(\frac{4\pi m^2e^2v^2}{h^3}\right)\left\{\int_{-\infty}^\infty ds\,E_z(s)\tilde{K}(z-s)+i\int_{-\infty}^\infty ds\,E_x(s)K'(z-s)-\frac{1}{ev}\int_{-\infty}^\infty ds\,G(s)\bar{K}(z-s)\right\},\quad(2.17b)$$

where now v is the magnitude of the Fermi velocity. The kernels in Eqs. (2.17) and an additional kernel

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[10] E. Jahnke and F. Emde, *Tables of Functions* (Dover Publications, Inc., New York, 1945), 4th ed., p. 149.

needed below are given by

$$K''(z-s)=\int_0^{\pi/2} d\theta \sin^2\theta \tan\theta J_0''\{q_x \tan\theta|z-s|\} \times e^{-\eta|z-s|}, \quad (2.18a)$$

$$K'(z-s)=\int_0^{\pi/2} d\theta \sin^2\theta J_0'\{q_x \tan\theta(z-s)\}e^{-\eta(z-s)}, \quad z>s$$

$$=-\int_0^{\pi/2} d\theta \sin^2\theta J_0'\{q_x \tan\theta(s-z)\}e^{-\eta(s-z)}, \quad s>z \quad (2.18b)$$

$$\bar{K}'(z-s)=\int_0^{\pi/2} d\theta \sin\theta \tan\theta J_0'\{q_x \tan\theta|z-s|\} \times e^{-\eta|z-s|}, \quad (2.18c)$$

$$\tilde{K}(z-s)=\int_0^{\pi/2} d\theta \sin\theta \cos\theta J_0\{q_x \tan\theta|z-s|\} \times e^{-\eta|z-s|}, \quad (2.18d)$$

$$\bar{K}(z-s)=-\int_0^{\pi/2} d\theta \sin\theta J_0\{q_x \tan\theta(z-s)\}e^{-\eta(z-s)}, \quad z>s$$

$$=\int_0^{\pi/2} d\theta \sin\theta J_0\{q_x \tan\theta(s-z)\}e^{-\eta(s-z)}, \quad s>z \quad (2.18e)$$

$$K(z-s)=\int_0^{\pi/2} d\theta \tan\theta J_0\{q_x \tan\theta|z-s|\} \times e^{-\eta|z-s|}, \quad (2.18f)$$

where the primes on the Bessel functions denote derivatives with respect to the arguments, and

$$\eta=\frac{\sec\theta}{v}\left(\frac{1}{\tau_0}-i\omega\right),$$

with τ_0 the relaxation time for electrons at the Fermi surface. The integration angle in these kernels should not be confused with the incident angle θ defined in Fig. 1.

If we now assume that the transition probabilities $W_{\mathbf{kk}'}$ are independent of the angle between $\mathbf{k}$ and $\mathbf{k}'$,[11] and use the integration techniques discussed above, the self-consistency equation (2.8) becomes

$$G(z)=\frac{ie}{2\tau_0}\int_{-\infty}^{\infty} ds\, E_x(s)\bar{K}'(z-s) -\frac{e}{2\tau_0}\int_{-\infty}^{\infty} ds\, E_z(s)\bar{K}(z-s) +\frac{1}{2\tau_0 v}\int_{-\infty}^{\infty} ds\, G(s)K(z-s). \quad (2.19)$$

The kernels $\bar{K}'$, $\bar{K}$, and K are given in Eqs. (2.18c), (2.18e), and (2.18f). Equations (2.3) and (2.17)-(2.19) are clearly consistent with the symmetry requirements (2.13).

We now have three simultaneous equations to solve for the field components: Eq. (2.3a) with J_x given by (2.17a), Eq. (2.3b) with J_z given by (2.17b), and Eq. (2.19). The solution can be most easily obtained utilizing Fourier transforms. Thus we define the Fourier transform for the field components

$$\mathcal{E}(q_z)=\int_{-\infty}^{\infty} dz\, E(z)e^{-iq_z z}, \quad (2.20a)$$

with the inverse transform given by

$$E(z)=\frac{1}{2\pi}\int_{-\infty}^{\infty} dq_z \mathcal{E}(q_z)e^{iq_z z}. \quad (2.20b)$$

Similarly, we write

$$g(q_z)=\int_{-\infty}^{\infty} dz\, G(z)e^{-iq_z z}, \quad (2.21)$$

$$j(q_z)=\int_{-\infty}^{\infty} dz\, J(z)e^{-iq_z z}, \quad (2.22)$$

and, for the kernels,

$$k(q_z)=\int_{-\infty}^{\infty} dz\, K(z)e^{-iq_z z}. \quad (2.23)$$

Because of the symmetry requirement (2.13a), there is a discontinuity in the derivative dE_x/dz at $z=0$, and the Fourier transform of d^2E_x/d_z^2 is given by

$$\int_{-\infty}^{\infty} dz\frac{d^2E_x}{dz^2}e^{-iq_z z}=-q_z^2\mathcal{E}_x-2\frac{dE_x(0+)}{dz}. \quad (2.24a)$$

The symmetry requirement (2.13b) indicates the existence of a discontinuity in E_z at $z=0$, so the Fourier transform of dE_z/dz is given by

$$\int_{-\infty}^{\infty} dz\frac{dE_z}{dz}e^{-iq_z z}=-2E_z(0+)+iq_z\mathcal{E}_z. \quad (2.24b)$$

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[11] The self-consistency correction will be seen below to be relatively small, and hence this approximation is indeed valid.

Using Eqs. (2.24), the Fourier transform of Eq. (2.3a) becomes

$$\mathcal{E}_x\left(-q_z^2+\frac{\omega^2}{c^2}\right)+q_zq_x\mathcal{E}_z+\left\{-2\frac{dE_x(0+)}{dz}+2iq_xE_z(0+)\right\}=-\frac{4\pi i\omega}{c^2}j_x. \quad (2.25)$$

Since

$$\nabla\times\mathbf{E}=-\frac{1}{c}\frac{\partial\mathbf{H}}{\partial t}, \quad (2.26)$$

we have

$$-2\frac{dE_x(0+)}{dz}+2iq_xE_z(0+)=-\frac{2i\omega}{c}H_y(0+), \quad (2.27)$$

and Eq. (2.25) can be written

$$\mathcal{E}_x\left(-q_z^2+\frac{\omega^2}{c^2}\right)+q_zq_x\mathcal{E}_z-\frac{2i\omega}{c}H_y(0+)=-\frac{4\pi i\omega}{c^2}j_x. \quad (2.28)$$

Writing the Fourier transform j_x using Eq. (2.17a) we have finally, for the Fourier transform of (2.3a),

$$\mathcal{E}_x\left(-q_z^2+\frac{\omega^2}{c^2}\right)+q_zq_x\mathcal{E}_z-\frac{2i\omega}{c}H_y(0+)=\kappa\left\{-\mathcal{E}_xk''+i\mathcal{E}_zk'+\frac{i}{ev}g\bar{k}'\right\}, \quad (2.29)$$

with

$$\kappa=-16\pi^2im^2e^2v^2\omega/c^2h^3. \quad (2.30)$$

In like manner, the Fourier transform of Eq. (2.3b) is found to be

$$\mathcal{E}_z\left(-q_x^2+\frac{\omega^2}{c^2}\right)+q_zq_x\mathcal{E}_x=\kappa\left\{\mathcal{E}_z\tilde{k}+i\mathcal{E}_xk'-\frac{1}{ev}g\bar{k}\right\}, \quad (2.31)$$

while the Fourier transform of the self-consistency condition, Eq. (2.19), is given by

$$g=\frac{ie}{2\tau_0}\mathcal{E}_x\bar{k}'-\frac{e}{2\tau_0}\mathcal{E}_z\bar{k}+\frac{1}{2\tau_0v}gk. \quad (2.32)$$

The Fourier-transformed kernels are evaluated in Appendix A.

Equation (2.32) can be solved directly for g, the result being

$$g=\frac{e}{2\tau_0}\{i\mathcal{E}_x\bar{k}'-\mathcal{E}_z\bar{k}\}\Big/\left\{1-\frac{1}{2l}k\right\}, \quad (2.33)$$

where the mean free path l is defined by $l\equiv v\tau_0$. Using this expression for g, our equations for $\mathcal{E}_x$ and $\mathcal{E}_z$ become

$$\mathcal{E}_x\left[-q_z^2+\frac{\omega^2}{c^2}+\kappa\left\{k''+\frac{\bar{k}'^2}{2l[1-(k/2l)]}\right\}\right]+\mathcal{E}_z\left[q_zq_x+i\kappa\left\{-k'+\frac{\bar{k}\bar{k}'}{2l[1-(k/2l)]}\right\}\right]=\frac{2i\omega}{c}H_y(0+), \quad (2.34a)$$

and

$$\mathcal{E}_x\left[q_zq_x+i\kappa\left\{-k'+\frac{\bar{k}\bar{k}'}{2l[1-(k/2l)]}\right\}\right]+\mathcal{E}_z\left[-q_x^2+\frac{\omega^2}{c^2}+\kappa\left\{-\tilde{k}-\frac{\bar{k}^2}{2l[1-(k/2l)]}\right\}\right]=0. \quad (2.34b)$$

Writing Eq. (2.34a) as

$$\mathcal{E}_xT_{xx}+\mathcal{E}_zT_{xz}=\frac{2i\omega}{c}H_y(0+), \quad (2.35a)$$

and Eq. (2.34b) as

$$\mathcal{E}_xT_{zx}+\mathcal{E}_zT_{zz}=0, \quad (2.35b)$$

we find

$$\frac{\mathcal{E}_x}{H_y(0+)}=\frac{2i\omega}{c}\left\{\frac{T_{zz}}{T_{xx}T_{zz}-T_{xz}T_{zx}}\right\}. \quad (2.36)$$

To facilitate comparison with the derivation of Sec. III, we write

$$T_{xx}=-q_z^2+(\omega^2/c^2)\epsilon_{xx}, \quad (2.37a)$$

$$T_{zz}=-q_x^2+(\omega^2/c^2)\epsilon_{zz}, \quad (2.37b)$$

and

$$T_{xz}=T_{zx}=q_xq_z+(\omega^2/c^2)\epsilon_{xz}. \quad (2.37c)$$

Using Eqs. (A1) of Appendix A, one can show that

$$\epsilon_{xx}=(1/q^2)\{\epsilon_tq_z^2+\epsilon_lq_x^2\}, \quad (2.38a)$$

$$\epsilon_{zz}=(1/q^2)\{\epsilon_tq_x^2+\epsilon_lq_z^2\}, \quad (2.38b)$$

and

$$\epsilon_{xz}=(\epsilon_l-\epsilon_t)q_xq_z/q^2, \quad (2.38c)$$

where q is defined by Eq. (A2). The transverse and longitudinal dielectric constants, ϵ_t and ϵ_l, are given by

$$\epsilon_t=1+\frac{ic^2\kappa}{\omega^2q}\left[-\frac{i}{ql'}+\frac{1}{2}\left(1+\frac{1}{(ql')^2}\right)\ln\left\{\frac{1+iql'}{1-iql'}\right\}\right] \quad (2.39)$$

and

$$\epsilon_l = 1 + \frac{2i\tau_0 c^2 \kappa l'}{\omega(ql')^2}\left[\left(ql' + \tfrac{1}{2}i\ln\left\{\frac{1+iql'}{1-iql'}\right\}\right)\Big/ \left(ql + \tfrac{1}{2}i\ln\left\{\frac{1+iql'}{1-iql'}\right\}\right)\right], \quad (2.40)$$

with l' defined by Eq. (A3).

Using Eqs. (2.37) and (2.38) in Eq. (2.36), we obtain the result

$$\frac{\mathcal{E}_x}{H_y(0+)} = \frac{2i\omega}{cq^2}\left[\frac{q_x^2}{(\omega^2/c^2)\epsilon_l} + \frac{q_z^2}{(\omega^2/c^2)\epsilon_t - q^2}\right]. \quad (2.41)$$

The surface impedance[12] for P polarization, Z_P', defined by

$$Z_P' = E_x(0+)/H_y(0+),$$

is then given by the inverse Fourier transform of (2.41) evaluated at $z=0$, or

$$Z_P' = \frac{1}{2\pi}\left(\frac{2i\omega}{c}\right)\int_{-\infty}^{\infty}\frac{dq_z}{q^2}\left[\frac{q_x^2}{(\omega^2/c^2)\epsilon_l} + \frac{q_z^2}{(\omega^2/c^2)\epsilon_t - q^2}\right]. \quad (2.42)$$

Transforming to a set of dimensionless variables defined by

$$\Omega = \omega/\omega_p, \quad (2.43a)$$

$$Q_x = q_x c/\omega_p, \quad (2.43b)$$

$$Q_z = q_z c/\omega_p, \quad (2.43c)$$

and

$$\gamma = 1/\omega_p \tau_0, \quad (2.43d)$$

$$b = (v/c)(1/\gamma), \quad (2.43e)$$

$$b' = b/(1 - i\Omega/\gamma), \quad (2.43f)$$

where $\omega_p = (4\pi Ne^2/m)^{1/2}$ is the free-electron plasma frequency (N is the electron density), Eq. (2.42) becomes

$$Z_P' = \frac{2i\Omega}{\pi}\int_0^{\infty}\frac{dQ_z}{Q^2}\left[\frac{Q_x^2}{\Omega^2\epsilon_l} + \frac{Q_z^2}{\Omega^2\epsilon_t - Q^2}\right], \quad (2.44)$$

where

$$Q^2 = Q_x^2 + Q_z^2, \quad (2.45)$$

and we have used the fact that the integrand is even in Q_z. In terms of the variables of Eqs. (2.43), the dielectric constants are

$$\epsilon_t = 1 - \frac{1}{\Omega(\Omega+i\gamma)}\frac{3}{2}\frac{1}{(b'Q)^3} \times\left[\frac{\{(b'Q)^2+1\}}{2i}\ln\left\{\frac{1+ib'Q}{1-ib'Q}\right\} - b'Q\right] \quad (2.46)$$

and

$$\epsilon_l = 1 - \frac{1}{\Omega(\Omega+i\gamma)}\left(-\frac{3i\Omega}{\gamma}\right)\frac{1}{(b'Q)^2} \times\left[\left(b'Q - \frac{1}{2i}\ln\left\{\frac{1+ib'Q}{1-ib'Q}\right\}\right)\Big/ \left(bQ - \frac{1}{2i}\ln\left\{\frac{1+ib'Q}{1-ib'Q}\right\}\right)\right]. \quad (2.47)$$

Several comments about these nonlocal dielectric functions are in order. In the limit $Q \to 0$, both reduce to the "ordinary" dielectric constant

$$\epsilon(\omega) = 1 - 1/\Omega(\Omega + i\gamma). \quad (2.48)$$

ϵ_t is the same function obtained by Reuter and Sondheimer and, indeed, if we consider normal incidence, for which $Q_x = 0$, our Eq. (2.44) for the surface impedance is the same as that obtained by Reuter and Sondheimer for the case of specular reflection. The presence of the longitudinal dielectric function is a manifestation of the fact that a P-polarized wave incident non-normally on the metal has a field component perpendicular to the surface, giving rise to charge fluctuations to which the system responds via the longitudinal dielectric function. These effects, of course, vanish for normal incidence. If we had made the relaxation-time approximation in the Boltzmann equation or, equivalently considered $G(z)$ in Eq. (2.7) to be zero, the result (2.44) would have the same structure but with ϵ_l replaced by ϵ_w, where

$$\epsilon_w = 1 - \frac{1}{\Omega(\Omega+i\gamma)}\frac{3}{(b'Q)^3}\left[b'Q - \frac{1}{2i}\ln\left\{\frac{1+ib'Q}{1-ib'Q}\right\}\right]. \quad (2.49)$$

Finally, it should be noted that had we retained the phenomenological term ϵ_{IB} in Eq. (2.1) representing interband effects, it would simply be added to the expressions for the dielectric function; that is, the leading "1" in Eqs. (2.39), (2.40), and (2.46)-(2.49) would be replaced by $(1+\epsilon_{\mathrm{IB}})$.

An expression for Z_S', the surface impedance for S polarization could clearly be determined in much the same manner as that used above to calculate Z_P'. However, in Sec. III a more adroit scheme is presented for the determination of the surface impedance and a discussion of Z_S' will be deferred to that point.

III. THEORY: DIELECTRIC-CONSTANT APPROACH

In the calculation of the surface impedance at non-normal incidence presented in Sec. II, the Boltzmann equation was used to solve for the current **J** in terms of the field **E** in the presence of a boundary. An alternative procedure will be used in this section to derive the sur-

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[12] The surface impedance is defined by $Z_P = (4\pi/c)(E_x/H_y)$. However, in the case of optical properties the important quantity is $(c/4\pi)Z_P$. Thus, to avoid useless repetition of factors $(c/4\pi)$, we call E_x/H_y the surface impedance but denote it by Z_P' to remind the reader that the definition is not the normal one. A similar convention will be used below for S polarization with $Z_S' \equiv -E_y(0+)/H_x(0+)$.

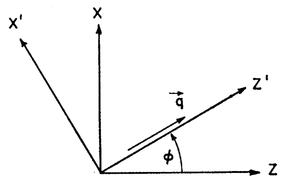

FIG. 2. The original and rotated coordinate systems for obtaining the dielectric tensor.

face impedance for both P and S polarizations. The relationship between $\mathbf{J}$ and $\mathbf{E}$ for the metal is expressed at the outset in terms of a nonlocal dielectric-response function, defined for an infinite medium. The effect of the metal surface is then taken into account by solving Maxwell's equations using appropriate boundary conditions. It is still necssary to find the dielectric function from the Boltzmann equation or by any other method desired; however, this can now be done for an infinite medium rather than a finite medium, significantly reducing the mathematical complexity.

P Polarization

We begin with Maxwell's equations in the form (2.3), and include the current density $\mathbf{J}$ in a displacement field $\mathbf{D}$ by the relation

$$\mathbf{D}=\mathbf{E}+(4\pi i/\omega)\mathbf{J}. \tag{3.1}$$

Equations (2.3) then become

$$\frac{d^2E_x}{dz^2}-iq_x\frac{dE_z}{dz}+\frac{\omega^2}{c^2}D_x=0 \tag{3.2a}$$

and

$$-q_x^2E_z-iq_x\frac{dE_x}{dz}+\frac{\omega^2}{c^2}D_z=0. \tag{3.2b}$$

These equations are valid for $z>0$, that is, inside the metal. We now imagine that the metal fills all space, extending the domain of z from $-\infty$ to $+\infty$. The fact that electrons are reflected specularly is taken into account by imposing the symmetry requirements $E_x(z)=E_x(-z)$, $E_z(z)=-E_z(-z)$, $D_x(z)=D_x(-z)$, and $D_z(z)=-D_z(-z)$, as in Eqs. (2.13). The Fourier transform of Eq. (3.2a) is

$$-q_z^2\mathcal{E}_x+q_zq_z\mathcal{E}_z+\frac{\omega^2}{c^2}\mathfrak{D}_x=2\frac{dE_x(0+)}{dz}-2iq_xE_z(0+)\,, \tag{3.3}$$

where we have used Eq. (2.20a) as the definition of the transform from $E(z)$ to $\mathcal{E}(q_z)$ and from $D(z)$ to $\mathfrak{D}(q_z)$. The terms on the right-hand side of Eq. (3.3) originate from discontinuities in dE_x/dz and E_z at $z=0$. Using Eq. (2.27), Eq. (3.3) can be written

$$-q_z^2\mathcal{E}_x+q_xq_z\mathcal{E}_z+(\omega^2/c^2)\mathfrak{D}_x=(2i\omega/c)H_y(0+). \tag{3.4}$$

Similarly, the Fourier transform of Eq. (3.2b) is

$$-q_x^2\mathcal{E}_z+q_zq_x\mathcal{E}_x+(\omega^2/c^2)\mathfrak{D}_z=0. \tag{3.5}$$

$\boldsymbol{\mathfrak{D}}$ and $\boldsymbol{\mathcal{E}}$ are related by the dielectric function $\epsilon_{ij}(\mathbf{q},\omega)$:

$$\mathfrak{D}_i=\sum_j \epsilon_{ij}\mathcal{E}_j. \tag{3.6}$$

To express ϵ_{ij} in terms of ϵ_t and ϵ_l, the transverse and longitudinal dielectric functions, we introduce a coordinate system x', z', rotated by the angle φ about the y axis as shown in Fig. 2. Placing the z' axis along the direction of $\mathbf{q}$, we have

$$\mathfrak{D}_{x'}=\epsilon_t\mathcal{E}_{x'} \tag{3.7a}$$

and

$$\mathfrak{D}_{z'}=\epsilon_l\mathcal{E}_{z'}. \tag{3.7b}$$

Thus the dielectric function is a diagonal tensor in the x', z' coordinate system:

$$\epsilon'=\begin{pmatrix}\epsilon_t & 0\\ 0 & \epsilon_l\end{pmatrix}. \tag{3.8}$$

ϵ' can be expressed in the x, z coordinate system by making the transformation

$$\epsilon=S^{-1}\epsilon' S\,, \tag{3.9}$$

where

$$S=\begin{pmatrix}\cos\varphi & -\sin\varphi\\ \sin\varphi & \cos\varphi\end{pmatrix}, \tag{3.10}$$

with $\sin\varphi=q_x/q$, $\cos\varphi=q_z/q$, and $q=(q_x^2+q_z^2)^{1/2}$. The result is that ϵ_{xx}, ϵ_{zz}, ϵ_{xz}, and ϵ_{zx} are as given by Eqs. (2.38).

Using the relations $\mathfrak{D}_x=\epsilon_{xx}\mathcal{E}_x+\epsilon_{xz}\mathcal{E}_z$ and $\mathfrak{D}_z=\epsilon_{zx}\mathcal{E}_x+\epsilon_{zz}\mathcal{E}_z$, we can solve Eqs. (3.4) and (3.5) for $\mathcal{E}_x/H_y(0+)$. We find

$$\frac{\mathcal{E}_x}{H_y(0+)}=\frac{2i\omega}{c}\left(\frac{T_{zz}}{T_{xx}T_{zz}-T_{zx}T_{xz}}\right), \tag{3.11}$$

where

$$T_{ij}=(\omega^2/c^2)\epsilon_{ij}-q^2\delta_{ij}+q_iq_j. \tag{3.12}$$

Equations (3.11) and (3.12) are identical to Eqs. (2.36) and (2.37). If the expressions (2.38) for ϵ_{ij} are inserted into (3.12), we find

$$T_{xx}=\left(\frac{\omega^2}{c^2}\frac{\epsilon_t}{q^2}-1\right)q_z^2+\frac{\omega^2}{c^2}\epsilon_l q_x^2\,, \tag{3.13a}$$

$$T_{zz}=\left(\frac{\omega^2}{c^2}\frac{\epsilon_t}{q^2}-1\right)q_x^2+\frac{\omega^2}{c^2}\epsilon_l q_z^2\,, \tag{3.13b}$$

$$T_{xz}=T_{zx}=\left[\frac{\omega^2}{c^2q^2}(\epsilon_l-\epsilon_t)+1\right]q_xq_z. \tag{3.13c}$$

Thus Eq. (3.11) finally becomes

$$\frac{\mathcal{E}_x}{H_y(0+)}=\frac{2i\omega}{cq^2}\left[\frac{q_x^2}{(\omega^2/c^2)\epsilon_l}+\frac{q_z^2}{(\omega^2/c^2)\epsilon_t-q^2}\right]. \tag{3.14}$$

The inverse Fourier transform of Eq. (3.14) evaluated at $z=0$ gives

$$Z_P'\equiv\frac{E_x(0+)}{H_y(0+)}=\frac{2i\Omega}{\pi}\int_0^\infty\frac{dQ_z}{Q^2}\left[\frac{Q_x^2}{\Omega^2\epsilon_l}+\frac{Q_z^2}{\Omega^2\epsilon_t-Q^2}\right] \tag{3.15}$$

in terms of the dimensionless variables defined in Eqs. (2.43). Equation (3.15) is the same as Eq. (2.44); however, ϵ_l and ϵ_t are as yet unspecified. This result therefore has a general validity independent of the particular model used for obtaining ϵ_l and ϵ_t. The only requirement is that $\epsilon_l(\mathbf{q},\omega)$ and $\epsilon_t(\mathbf{q},\omega)$ exist.[13] In the present context, the appropriate value of ϵ_t is that of Eq. (2.46), whereas ϵ_l is correctly given by (2.47). However, were the relaxation to the perturbed state not to be considered, ϵ_l would be given by ϵ_w, Eq. (2.49).[14]

S Polarization

The only nonvanishing fields are now E_y, H_x, and H_z. The y component of Eq. (3.2) is

$$\frac{d^2E_y}{dz^2}-q_x^2E_y+\frac{\omega^2}{c^2}D_y=0, \tag{3.16}$$

where Eq. (3.1) has again been used to introduce the displacement D. The Fourier transform of Eq. (3.16) is

$$-(q_z^2+q_x^2)\mathcal{E}_y+\frac{\omega^2}{c^2}\mathfrak{D}_y=2\frac{dE_y(0+)}{dz} \tag{3.17}$$

or

$$-q^2\mathcal{E}_y+\frac{\omega^2}{c^2}\mathfrak{D}_y=-\frac{2i\omega}{c}H_x(0+). \tag{3.18}$$

In arriving at Eq. (3.17), we have taken $E_y(z)=E_y(-z)$ so that the discontinuity in dE_y/dz gives the term on the right. From the Maxwell equation (2.26) or $dE_y/dz=-(i\omega/c)H_x$, Eq. (3.18) follows immediately. Since the electric field is perpendicular to $\mathbf{q}$ for S polarization,

$$\mathfrak{D}_y=\epsilon_t\mathcal{E}_y. \tag{3.19}$$

Using Eq. (3.19), we solve Eq. (3.18) for $\mathcal{E}_y/H_x(0+)$:

$$\frac{\mathcal{E}_y}{H_x(0+)}=-\frac{2i\omega}{c}\frac{1}{(\omega^2/c^2)\epsilon_t-q^2}. \tag{3.20}$$

The inverse Fourier transform of Eq. (3.20) evaluated at $z=0$ gives

$$Z_S'\equiv-\frac{E_y(0+)}{H_x(0+)}=\frac{2i\Omega}{\pi}\int_0^\infty\frac{dQ_z}{\Omega^2\epsilon_t-Q^2}, \tag{3.21}$$

where the dimensionless variables defined in Eqs. (2.43) have again been introduced. The appropriate expression for ϵ_t is that of Eq. (2.46).

Reflectance

For completeness, we shall present expressions for the reflectance in terms of the surface impedance. The fields associated with the incident and reflected waves for P polarization are of the following form:

$$E_z=a_Ie^{ik_zz}+a_Re^{-ik_zz}, \tag{3.22a}$$

$$E_x=(k_x/k_z)(-a_Ie^{ik_zz}+a_Re^{-ik_zz}), \tag{3.22b}$$

$$H_y=-(\omega/k_xc)(a_Ie^{ik_zz}+a_Re^{-ik_zz}), \tag{3.22c}$$

where a_I and a_R are unknown amplitudes for the incident and reflected waves, $k_z=(\omega/c)\cos\theta$ and $k_x=(\omega/c)\times\sin\theta$, θ being the angle of incidence as indicated in Fig. 1. These equations hold in the vacuum, for $z<0$. The Maxwell equations $ik_xE_x+dE_z/dz=0$ and $ik_xH_y=-i(\omega/c)E_z$ have been used to obtain Eqs. (3.22b) and (3.22c) from Eq. (3.22a). Solving Eqs. (3.22b) and (3.22c) for the amplitude ratio a_R/a_I in terms of $E_x(0-)/H_y(0-)$, we find the expression for the reflectance,

$$R_P=\left|\frac{a_R}{a_I}\right|^2=\left|\frac{\cos\theta-E_x(0-)/H_y(0-)}{\cos\theta+E_x(0-)/H_y(0-)}\right|^2. \tag{3.23}$$

From the continuity of E_x and H_y, $E_x(0-)/H_y(0-)=E_x(0+)/H_y(0+)=Z_P'$. Therefore,

$$R_P=\left|\frac{\cos\theta-Z_P'}{\cos\theta+Z_P'}\right|^2. \tag{3.24}$$

Since the x component of the wave vector is the same inside and outside the material, $q_x=k_x=(\omega/c)\sin\theta$ or $Q_x=\Omega\sin\theta$, where $Q_x=q_xc/\omega_p$ and $\Omega=\omega/\omega_p$ are the dimensionless quantities appearing in Eq. (3.15).

The fields associated with the incident and reflected waves for S polarization are

$$E_y=a_Ie^{ik_zz}+a_Re^{-ik_zz}, \tag{3.25a}$$

$$H_x=(ck_z/\omega)(-a_Ie^{ik_zz}+a_Re^{-ik_zz}), \tag{3.25b}$$

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[13] Equations (3.15) and (3.21) below are applicable rigorously only to a microscopically uniform system such as a free-electron gas. Actual metals and insulators are microscopically nonuniform due to the presence of a periodic potential and can be characterized only by more general dielectric functions of the form $\epsilon_{ij}(\mathbf{q}+\mathbf{G}, \mathbf{q}+\mathbf{G}', \omega)$, where $\mathbf{G}$ and $\mathbf{G}'$ are reciprocal lattice vectors. See L. J. Sham and J. M. Ziman, Solid State Phys. **15**, 221 (1963).

[14] Derivations of expressions for $\epsilon_l(\mathbf{q},\omega)$, $\epsilon_t(\mathbf{q},\omega)$, and $\epsilon_w(\mathbf{q},\omega)$, using the Boltzmann equation for a free-electron gas of infinite extent, have been presented by several authors. For example, Warren and Ferrell (Ref. 6) give expressions for the complex conductivities σ_t and σ_l [their Eqs. (20) and (24)], from which the dielectric constants ϵ_t and ϵ_l can be obtained using $\epsilon=1+4\pi i\sigma/\omega$. In C. Kittel, *Quantum Theory of Solids* (John Wiley & Sons, Inc., New York, 1963), Chap. 17, σ_t is given by Eq. (23), σ_w by Eq. (22), while σ', defined by Eq. (33), is the longitudinal conductivity σ_l if c_s is written as ω/q. Therefore, the methods of Sec. II need not be employed to determine the dielectric constants.

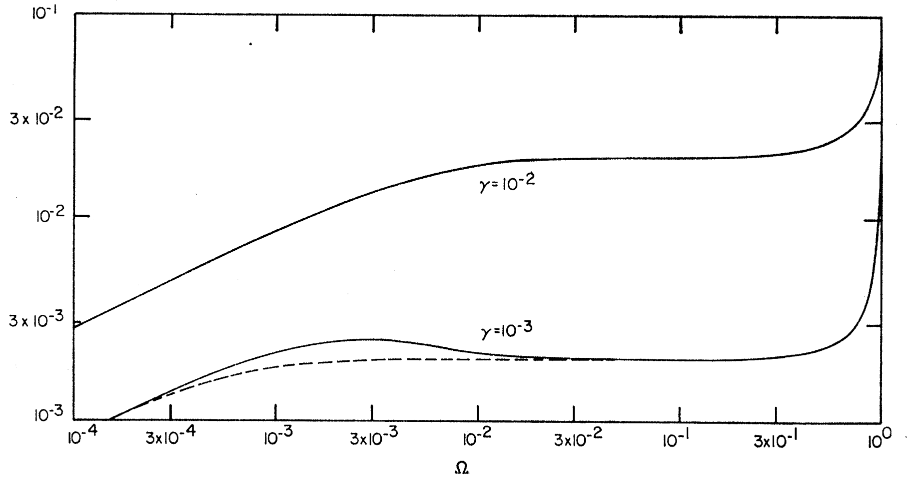

FIG. 3. Classical (dashed lines) and anomalous (solid lines) calculations of the frequency dependence of the absorptance for $\theta=0$, with $\gamma=10^{-2}$ and 10^{-3}. For $\gamma=10^{-2}$, the classical and anomalous values cannot be distinguished with this scale.

where the Maxwell equation $H_x=i(c/\omega)dE_y/dz$ has been used to obtain Eq. (3.25b) from (3.25a). H_z is also present, but need not be considered. The reflectance is

$$R_S=\left|\frac{a_R}{a_I}\right|^2=\left|\frac{1+[E_y(0-)/H_x(0-)]\cos\theta}{1-[E_y(0-)/H_x(0-)]\cos\theta}\right|^2 \tag{3.26}$$

or

$$R_S=\left|\frac{1-Z_S'\cos\theta}{1+Z_S'\cos\theta}\right|^2. \tag{3.27}$$

The general expressions (3.15) and (3.21) for the surface impedances Z_P' and Z_S' can be evaluated in the classical limit (that is, neglecting nonlocal effects) by setting $\epsilon_t=\epsilon_l=\epsilon(\omega)$, a function only of the frequency. The integration over Q_z can then be done exactly and we find

$$Z_P^{(\mathrm{cl})}=(\epsilon(\omega)-\sin^2\theta)^{1/2}/\epsilon(\omega) \tag{3.28}$$

and

$$Z_S'^{(\mathrm{cl})}=(\epsilon(\omega)-\sin^2\theta)^{-1/2}. \tag{3.29}$$

Therefore, the classical expressions for the reflectance are

$$R_P^{(\mathrm{cl})}=\left|\frac{\epsilon(\omega)\cos\theta-(\epsilon(\omega)-\sin^2\theta)^{1/2}}{\epsilon(\omega)\cos\theta+(\epsilon(\omega)-\sin^2\theta)^{1/2}}\right|^2 \tag{3.30}$$

and

$$R_S^{(\mathrm{cl})}=\left|\frac{(\epsilon(\omega)-\sin^2\theta)^{1/2}-\cos\theta}{(\epsilon(\omega)-\sin^2\theta)^{1/2}+\cos\theta}\right|^2. \tag{3.31}$$

Since we are here considering free-electron effects, the dielectric constant $\epsilon(\omega)$ for the classical calculation is that given by Eq. (2.48).

In all cases the absorptance A is given by $A=1-R$, where R is the appropriate reflectance.

IV. RESULTS AND DISCUSSION

Since we are herein investigating effects associated with the essentially free electrons, we consider ϵ_{IB} to be zero. Thus there are in the present formalism two quantities characteristic of the metal under consideration: one is the Fermi velocity and the other is the electron lifetime. Since the electron lifetime will depend on

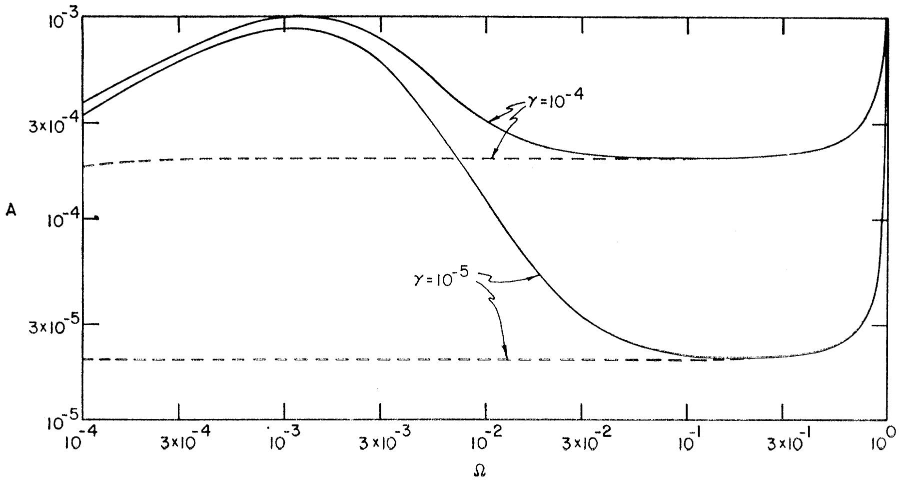

FIG. 4. Classical (dashed lines) and anomalous (solid lines) calculations of the frequency dependence of the absorptance for $\theta=0$, with $\gamma=10^{-4}$ and 10^{-5}.

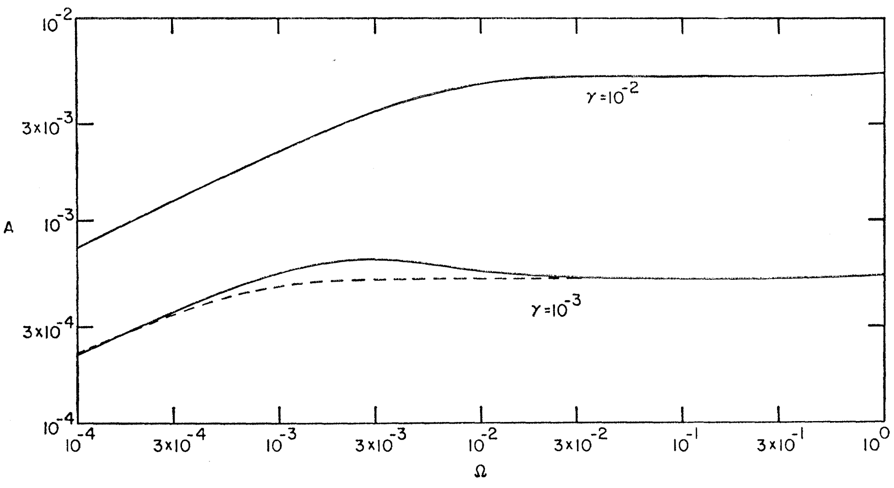

FIG. 5. Classical (dashed lines) and anomalous (solid lines) calculations of the frequency dependence of the absorptance for $\theta=75°$, S polarization, and $\gamma=10^{-2}$ and 10^{-3}. For $\gamma=10^{-2}$, the classical and anomalous values cannot be distinguished with this scale.

the specific conditions involved in a given experiment, we will illustrate its effect in the theory by considering a range of γ [15] from 10^{-2} (short lifetime) to 10^{-5} (long lifetime). We have selected for illustration the value $v_F=0.85\times10^8$ cm/sec characteristic of potassium.[16]

To indicate the type of absorptance structure associated with the anomalous skin effect at normal incidence, we show in Figs. 3 and 4 the absorptance as obtained from the normal-incidence limit of either Eq. (3.24) or (3.27). In this case Z_P' and Z_S' are both given by the normal-incidence limit, or

$$Z_N'=\frac{2i\Omega}{\pi}\int_0^\infty\frac{dQ_z}{\Omega^2\epsilon_t-Q_z^2}, \tag{4.1}$$

with ϵ_t given by (2.46) with $Q=Q_z$. Shown also are the classical values of the absorptance. Clearly evident in these figures is the peak due to the anomalous skin effect for $\Omega\sim10^{-3}$. The difference between the anomalous and the classical calculations is very small for $\gamma=10^{-2}$, but becomes large as γ decreases to 10^{-5}. It should be noted that essentially all of the anomalous effects occur for $\Omega<0.1$.

A comparison of the anomalous and classical calculations of the absorptance for an angle of incidence of 75° and S polarization is shown in Figs. 5 and 6. These curves are very similar to those for normal incidence, the anomalous skin effect giving rise to a marked peak for $\Omega\sim10^{-3}$ and low γ. The only significant difference for $\theta=75°$ is the absence of the abrupt rise in absorptance near $\Omega=1$. This abrupt rise occurs for $\Omega\sim1/\cos\theta$ or near $\Omega\sim1$ for $\theta=0$ and near $\Omega\sim3.9$ for $\theta=75°$. Thus

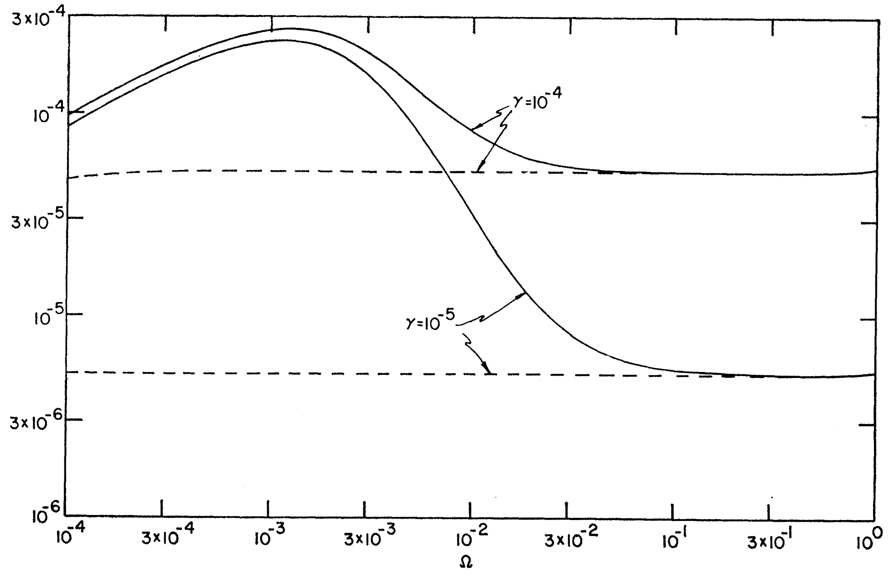

FIG. 6. Classical (dashed lines) and anomalous (solid lines) calculations of the frequency dependence of the absorptance for $\theta=75°$, S polarization, and $\gamma=10^{-4}$ and 10^{-5}.

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[15] Note that $\gamma=(\omega_p\tau_0)^{-1}$. See Eq. (2.43).

[16] C. Kittel, *Introduction to Solid State Physics* (John Wiley & Sons, Inc., New York, 1966), 3rd ed., p. 208. This free-electron value is valid here since the effective electron mass is equal to the actual electron mass for potassium. See Ref. 5.

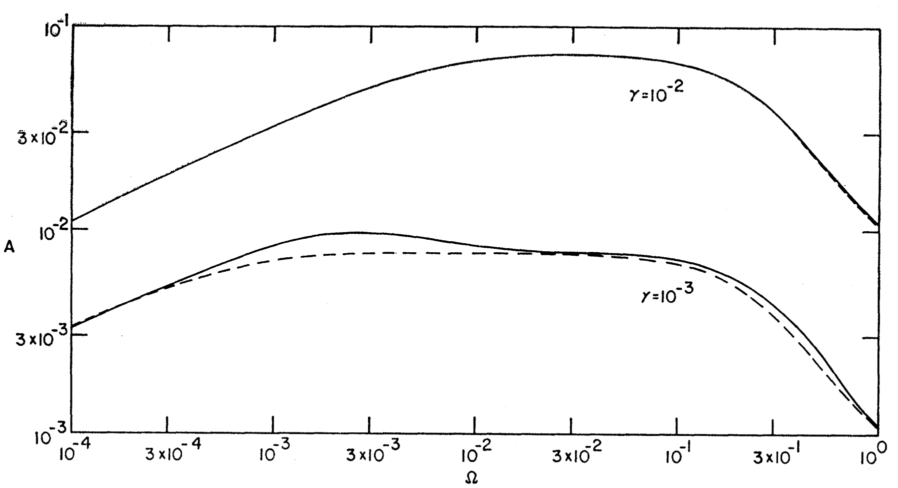

FIG. 7. Classical (dashed lines) and anomalous (solid lines) calculations of the frequency dependence of the absorptance for $\theta=75°$, P polarization, and $\gamma=10^{-2}$ and 10^{-3}.

in the curves for 75° this rise is removed to the right and off the scale. Note once again that essentially all anomalous effects occur for $\Omega<0.1$.

The anomalous and classical absorptance calculations for P polarization and $\theta=75°$ are shown in Figs. 7 and 8. Here, one sees the "usual" anomalous peak near $\Omega\sim10^{-3}$ but in addition there exists an important absorption peak in the range $0.1\lesssim\Omega\lesssim1.0$. The existence of this latter peak is, as shown in Fig. 8, not predicted by the non-normal-incidence generalization given by Reuter and Sondheimer, where, as discussed above, conclusions were drawn based upon a situation where $\nabla\cdot\mathbf{E}=0$.[17] This fact, together with the results shown in Figs. 5 and 6 indicating no analogous peak for S polarization, indicates that this additional absorption is due to the presence of charge fluctuations within the metal, an effect which we have allowed for in the present calculation.[18]

To further assess the nature and consequences of this additional absorption, we have done the following calculation. Much optical data in the "free-electron" regime is analysed using the classical dielectric constant of Eq. (2.48). Hence it would be of interest to know the effective value of γ needed in a classical calculation to

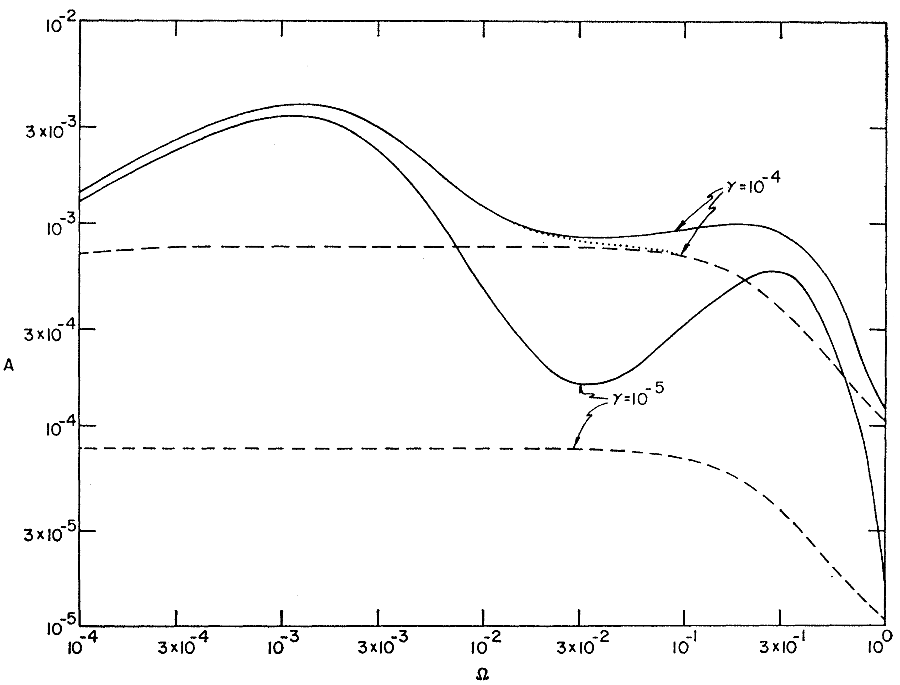

FIG. 8. Classical (dashed lines) and anomalous (solid lines) calculations of the frequency dependence of the absorptance for $\theta=75°$, P polarization, and $\gamma=10^{-4}$ and 10^{-5}. The dotted line shows the absorptance obtained for $\gamma=10^{-4}$ using the surface impedance given by Reuter and Sondheimer (see Appendix B). The dotted line coincides with the solid line (the anomalous absorptance) at low frequencies and with the dashed line (the classical absorptance) at high frequencies.

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[17] In Appendix B is presented a discussion of the Reuter and Sondheimer conclusions for non-normal incidence.

[18] If ϵ_w rather than ϵ_l is used in Eq. (3.15) when evaluating the surface impedance for P polarization, the absorption changes by $\lesssim0.1\%$. Hence no further mention of ϵ_w will be made.

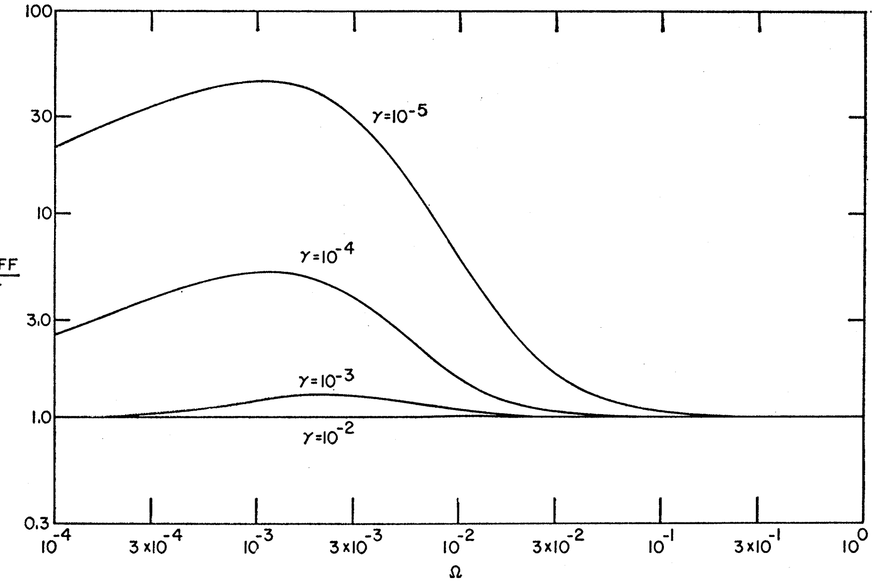

FIG. 9. The frequency dependence of γ_{eff}/γ for $\theta=0$.

reproduce the absorptance curves of Figs. 3–8. These effective values of γ were determined by using ϵ from Eq. (2.48) in Eqs. (3.30) and (3.31) and adjusting γ until the reflectance agreed with that obtained from the anomalous calculations.

The frequency dependence of γ_{eff} thereby determined divided by the fixed γ of the anomalous calculation is shown in Fig. 9 for $\theta=0$. If there were no anomalous effects, all curves would be independent of Ω, and γ_{eff}/γ would have the value 1.0. For $\gamma=10^{-2}$, there occurs only a small increase above 1.0 for $\Omega\sim10^{2}$. However, with decreasing γ the significant additional absorption associated with the anomalous skin effect produces a marked peak for $\Omega\sim10^{-3}$. For S polarization and an arbitrary value of the angle of incidence, the values of γ_{eff}/γ are identical to those for $\theta=0$ indicating that γ_{eff} is a meaningful quantity.

Using the absorptance values of Figs. 7 and 8, curves of γ_{eff}/γ for P polarization have been calculated and are shown in Fig. 10. When $\Omega\lesssim10^{-2}$, the values of γ_{eff}/γ are again identical to those for S polarization and arbitrary θ. However, the higher-frequency absorption structure for P polarization gives rise to significant peaks in γ_{eff} near $\Omega\sim0.4$ when $\gamma\lesssim10^{-3}$. For $\gamma=10^{-2}$, there is only a small increase above 1.0, the maximum value of γ_{eff}/γ being ~1.02 for $\Omega\sim0.40$. It is interesting to note that if we write $\gamma_{eff}=\gamma+\Delta\gamma$, the maximum value of $\Delta\gamma$ in the frequency region above 0.1 for $\theta=75°$ is roughly 2×10^{-4}, independent of the value of γ.

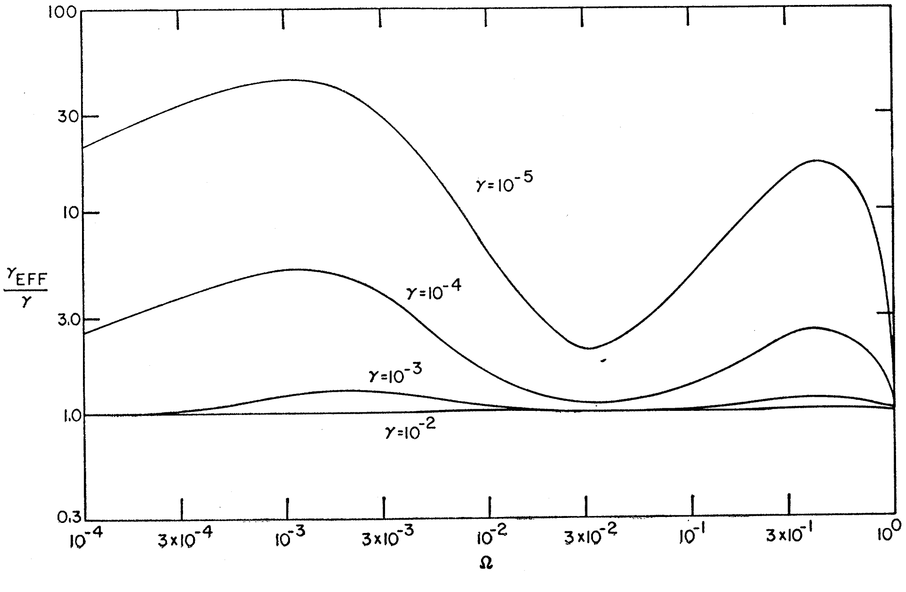

FIG. 10. The frequency dependence of γ_{eff}/γ for $\theta=75°$ and P polarization.

Once again we point out that the results given above are for potassium. Since potassium has a small Fermi velocity compared with most metals, all of the anomalous structure will be greater for most other materials than that discussed above. For example, the Fermi velocity for gold is 1.34×10^8cm/sec and since $\Delta\gamma$, as defined in the previous paragraph, scales approximately linearly with the Fermi velocity, $\Delta\gamma$ for gold has a maximum value of roughly 3.2×10^{-4} for $\theta=75°$, the maximum occuring when $\Omega\sim0.4$.

The most striking conclusion from the results discussed above is that, for the case of non-normal incidence,[19] *the assumption that there exists a single, complex, frequency-dependent dielectric function* $\epsilon(\omega)$ *which describes the metal classically is invalid.* The fact that γ_{eff} for P polarization differs from that for S polarization when $10^{-2}\lesssim\Omega\lesssim1$ means that two different frequency-dependent dielectric constants are needed to characterize the reflectance of the system, one for P and one for S polarization. Thus analyzing optical data in terms of a single dielectric constant in this frequency range gives results which have questionable meaning.

It should, however, be noted that significant differences in the two dielectric functions occur only when $\gamma\lesssim10^{-3}$. When $\gamma=10^{-3}$, the differences in the imaginary parts of the dielectric functions as determined by the γ_{eff} analysis can be as large as 20%, while the difference for $\gamma=10^{-2}$ is less than 2%, these numbers being for the case of potassium with $\theta=75°$. Also we saw above that for $\Omega\lesssim10^{-2}$ the values of γ_{eff} were independent of angle or polarization. Thus if the frequency is sufficiently low or γ is sufficiently large, there does exist a unique frequency-dependent dielectric function by which the reflectance of the system can be characterized.[20]

One might object that analyzing the absorption data in terms of γ_{eff} is too rigid a requirement; that is, that there may yet be an alternative dielectric function $\epsilon(\omega)$, valid for the system, having a form differing significantly from that of (2.48). That such is not, in general, the case will be demonstrated now. If there exists a dielectric function $\epsilon(\omega)$, the classical values of the surface impedance $Z_P'^{(\text{cl})}$ and $Z_S'^{(\text{cl})}$ are given by Eqs. (3.28) and (3.29). These equations imply a particular relationship between the surface impedances,

$$Z_P'^{(\text{cl})}=\frac{Z_S'^{(\text{cl})}}{1+\sin^2\theta[Z_S'^{(\text{cl})}]^2}, \tag{4.2}$$

and indicate that $\epsilon(\omega)$ is given by

$$\epsilon(\omega)=[Z_P'^{(\text{cl})}Z_S'^{(\text{cl})}]^{-1}. \tag{4.3}$$

If, conversely, the true surface impedances Z_P' and Z_S', as calculated by Eqs. (3.15) and (3.21), were to obey the relationship

$$Z_P'=\frac{Z_S'}{1+\sin^2\theta[Z_S']^2}, \tag{4.4}$$

in analogy to (4.2), a unique dielectric function could be found from

$$\epsilon(\omega)=[Z_P'Z_S']^{-1}, \tag{4.5}$$

as in Eq. (4.3). This dielectric function would then characterize the metal completely, giving not only the correct reflectances for both S and P polarization but also the correct phases of the reflected waves. It will be shown in Appendix B that Z_P' and Z_S' do not, in general, obey Eq. (4.4); this means that a unique $\epsilon(\omega)$ does not exist. It is important to note that the relationship (4.4) becomes invalid precisely where the new absorption peak for P polarization associated with charge fluctuations appears. Two different dielectric functions $\epsilon_P(\omega)$ and $\epsilon_S(\omega)$, related to the actual surface impedances Z_P' and Z_S' by equations of the form (3.28) and (3.29), would therefore be needed to characterize the metal. However, if $\gamma\gtrsim10^{-2}$ or Ω is in the usual anomalous region ($\Omega\lesssim10^{-2}$), Eq. (4.4) is satisfied and a unique $\epsilon(\omega)$ exists.

It is also of interest to examine one of the standard methods of analysis of optical data at non-normal angles of incidence, the method used by Mayer and coworkers.[5] In this method, a plane polarized beam of light is incident at some angle θ (75° is much used) with the beam polarized at an angle of 45° with respect to the plane defined by the incident direction and the normal to the metal surface, i.e., midway between P and S polarization. Using a system of polarizers the complex reflectance amplitudes are then measured for both S and P polarization. Assuming that a single complex dielectric function then exists, it is obtained from the expression[21]

$$\epsilon^{SP}(\Omega)=\epsilon_1^{SP}+i\epsilon_2^{SP}=\left(\frac{1-s}{1+s}\right)^2\frac{\sin^4\theta}{\cos^2\theta}+\sin^2\theta, \tag{4.6}$$

where $s=r_P/r_S$, $r_P(r_S)$ being the complex reflection amplitude for $P(S)$ polarization. There is clearly no particular form implied for the dielectric constant in this type of analysis.

We shall show below that if the present theory is used to calculate the ratio $s=r_P/r_S$, which is then inserted into Eq. (4.6) to obtain ϵ^{SP}, the value of ϵ^{SP} found in this way does not yield the actual reflectance (or absorptance) for either S or P polarization. That is, if ϵ^{SP} is used in Eqs. (3.30) and (3.31) to recalculate the absorptance, the result in the range $10^{-2}\lesssim\Omega\lesssim1$ is too large for P polarization and much too large for S polarization when $\gamma\lesssim10^{-3}$. Thus, even though ϵ^{SP} has

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[19] We have only considered $\theta=75°$ above. Other angles are mentioned below.

[20] This statement does not imply that such a dielectric constant determined by the γ_{eff} analysis can characterize the system completely. For when there exists an anomalous skin effect, even if γ_{eff} can be chosen so as to reproduce the correct reflectance, the phase of the reflected wave would not, in general, be given correctly by the classical analysis.

[21] D. J. Price, Proc. Phys. Soc. (London) 58, 704 (1946).

been constructed so as to give the correct *ratio* r_P/r_S or R_P/R_S, the separate values of R_P and R_S (or A_P and A_S) are incorrect. Only when $\gamma \gtrsim 10^{-2}$ or $1 \lesssim \Omega \lesssim 10^{-2}$ does the use of Eq. (4.6) become reasonable.

The conclusions of the preceding paragraph can be arrived at by considering the imaginary part of the dielectric constant, ϵ_2^{SP}, or the associated conductivity, σ_{SP}, related to ϵ_2^{SP} by

$$\sigma_{SP} = (\omega/4\pi)\epsilon_2^{SP}. \tag{4.7}$$

The conductivity σ_{SP} can be compared with σ_{eff}, which is found in the following manner: Taking $\epsilon(\omega)$ to be given by (2.48), γ is replaced by γ_{eff}. The associated dielectric constant is

$$\epsilon^{\mathrm{eff}}(\Omega) = \epsilon_1^{\mathrm{eff}}(\Omega) + i\epsilon_2^{\mathrm{eff}}(\Omega) = 1 - 1/\Omega(\Omega + i\gamma_{\mathrm{eff}}), \tag{4.8}$$

and the conductivity is

$$\sigma_{\mathrm{eff}} = \frac{\omega}{4\pi}\epsilon_2^{\mathrm{eff}} = \frac{\omega_p}{4\pi}\frac{\gamma_{\mathrm{eff}}}{\Omega^2 + \gamma_{\mathrm{eff}}^2}, \tag{4.9}$$

with ω_p the plasma frequency defined in (2.43).[22] The two values of σ_{eff} calculated in this way, one for S and the other the P polarization, are those values which will yield the correct absorption classically. In comparing σ_{SP} and σ_{eff}, it is convenient to plot the quantities $(\sigma_{SP} - \sigma_{\mathrm{cl}})/\sigma_{\mathrm{cl}}$ and $(\sigma_{\mathrm{eff}} - \sigma_{\mathrm{cl}})/\sigma_{\mathrm{cl}}$, where

$$\sigma_{\mathrm{cl}} = \frac{\omega_p}{4\pi}\frac{\gamma}{\Omega^2 + \gamma^2}.$$

The solid lines in Fig. 11 are the values of $(\sigma_{\mathrm{eff}} - \sigma_{\mathrm{cl}})/\sigma_{\mathrm{cl}}$ for P polarization. The only curve shown for S polarization is the dotted line for $\gamma = 10^{-5}$. For higher values of γ, the values of $(\sigma_{\mathrm{eff}} - \sigma_{\mathrm{cl}})/\sigma_{\mathrm{cl}}$ for S polarization are approximately two orders of magnitude below the values for P polarization at $\Omega \sim 0.1$ and are decreasing rapidly as Ω increases. The values of $(\sigma_{SP} - \sigma_{\mathrm{cl}})/\sigma_{\mathrm{cl}}$ are given by the dashed lines in Fig. 11. This figure shows that the value of ϵ^{SP} or the conductivity σ_{SP} derived from Eqs. (4.6) and (4.7) cannot characterize the system completely. While this type of analysis yields values of the conductivity which have the right form but the wrong magnitude for P polarization, the resultant values of the conductivity are meaningless for S polarization. Thus we would conclude that since σ_{SP} is somewhat greater than σ_{eff} for P polarization and much greater than σ_{eff} for S polarization, the absorptance calculated from ϵ^{SP} or σ_{SP} would be too large for P polarization and much too large for S polarization. This indicates again that two distinct frequency-dielectric functions are needed in general, one each for P and S polarization.

Nothing has been said above concerning the values for ϵ_1^{SP}. In the frequency range where two dielectric functions are needed the values of ϵ_1^{SP} and $\epsilon_1^{\mathrm{eff}}$ are essentially the same. Thus the comparison of σ_{SP} and ϵ_{eff} in Fig. 11, involving only the imaginary parts of the dielectric constants, is justified.

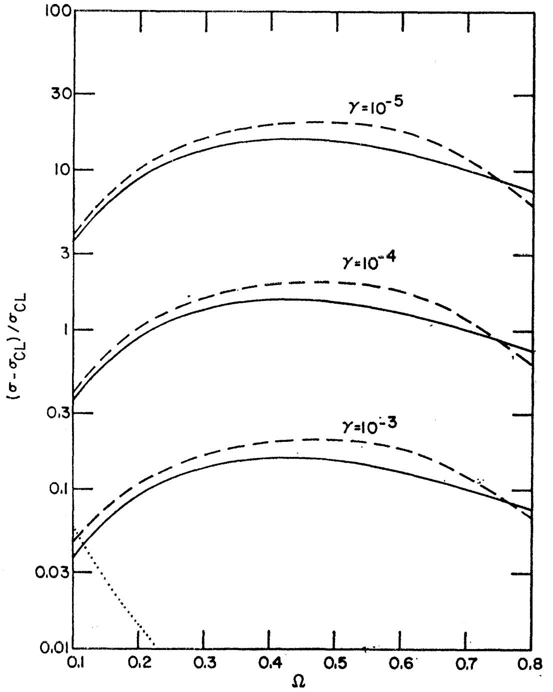

FIG. 11. Values of $(\sigma_{\mathrm{eff}} - \sigma_{\mathrm{cl}})/\sigma_{\mathrm{cl}}$ for P polarization are given by the solid lines. Values of $(\sigma_{SP} - \sigma_{\mathrm{cl}})/\sigma_{\mathrm{cl}}$ are given by the dashed lines. $(\sigma_{\mathrm{eff}} - \sigma_{\mathrm{cl}})/\sigma_{\mathrm{cl}}$ for S polarization and $\gamma = 10^{-5}$ is given by the dotted line.

To this point, we have discussed only the cases of $\theta = 0$ and $\theta = 75°$. To indicate the angular dependence of the anomalous P absorption in the range $0.1 \leq \Omega \leq 1$, we have shown in Fig. 12 curves of the P absorptance minus the classical absorptance for $\gamma = 10^{-4}$ and various values of θ. From these curves it is clear that, as expected, the anomalous effect decreases as θ decreases, but important effects occur still for $\theta = 30°$. The decrease from $\theta = 75°$ to $\theta = 85°$ is really a consequence of the quantity plotted. For if $(A - A_{\mathrm{cl}})/A_{\mathrm{cl}}$ were plotted, the 85° curve would peak above the others.

Much has been said above concerning the point that, in the frequency range $10^{-2} \lesssim \Omega \lesssim 1$, two dielectric functions are needed to characterize the system for non-normal angles of incidence. Yet we have only considered the case of specular reflection of the electrons at the surface. Is it not possible that if the electron scattering is diffuse rather than specular these particular anomalous effects will vanish? Possibly, yes, but, in our estimation, highly unlikely. Indeed, an unpublished calculation by the present authors suggests that these effects might well be considerably enhanced. This calculation involved a generalization of the Holstein model[23] to include a finite lifetime and an arbitrary angle of in-

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[22] For potassium, $\omega_p = 6.61 \times 10^{15}$ sec^{-1}. See Ref. 5.

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

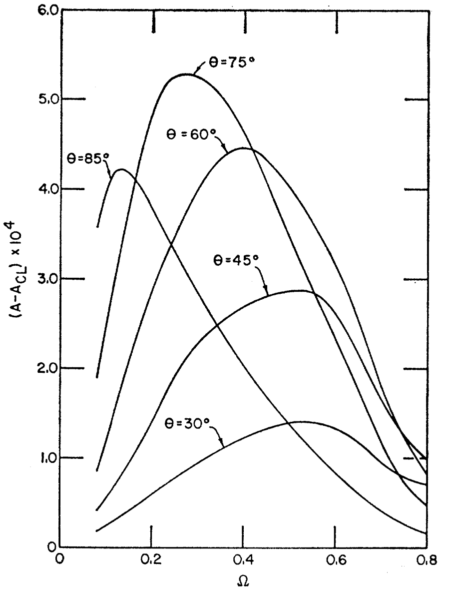

FIG. 12. The angular dependence of the anomalous absorptance A minus the classical absorptance A_{cl} for $\gamma=10^{-4}$.

cidence. Now the principle assumption of Holstein was that in the high-frequency range of interest here ($\Omega\sim0.1$), the electric field inside the metal can be approximated by its classical value. This assumption eliminates any charge fluctuations in the metal since the classical field has $\nabla\cdot\mathbf{E}=0$. Thus we expect, and find, no absorption peaks in this frequency range for either polarization. However, making an analysis as discussed above, we do obtain a value of $\gamma_{\text{eff}}=\gamma+1.1\times10^{-3}$ for both P and S polarization at $\theta=75°$ when the metal under consideration is potassium. This, then, is the background value from which any anomalous peaks associated with the charge fluctuation must emerge. Since the background (the value of γ_{eff} for S polarization) in the case of specular reflection corresponds to $\gamma_{\text{eff}}\approx\gamma$, we see immediately that the absorptance will be significantly higher for the case of diffuse scattering. If the peaks associated with the charge fluctuation scale even roughly as for specular reflection, the anomalous effect in this frequency range for diffuse reflection would be considerably larger than for specular reflection. A calculation for the case of diffuse reflection has been completed; the results will be reported elsewhere.

Detailed comments concerning experimental results will be deferred until the analysis for diffuse reflection is presented. However, several comments will be made concerning the experiments of Mayer and co-workers[5] on K and Na and those of Hodgson[24] on Na, work done at $\theta=75°$ and analyzed via Eq. (4.6). The excess absorption (absorption above the classical or Drude value) obtained in the present calculation is lower than that found by these workers at frequencies below the interband edge. Based strictly upon the results for specular reflection we would then say that the results of Hodgson and of Mayer and co-workers for Na at temperatures of 100° and 20°C and for K at 85° and 20°C, even though based upon an equation which in principle is invalid, should be approximately valid since they correspond to $\gamma\sim3\times10^{-3}$. However, the results of Mayer and co-workers for K and Na at a temperature of −183°C are suspect since for this case $\gamma\sim10^{-3}$. In particular, the apparent dip of the conductivity below the classical value, about which much has been said,[25] cannot be considered significant at this point. *It is quite possible that if these experiments were redone at another angle of incidence, the results would be significantly different.*

We would like to stress that if optical experiments in the frequency range $10^{-2}\lesssim\Omega\lesssim1$ are to be done at non-normal angles of incidence, the experimenter should present the reflectance data for S and P polarization separately. In particular, analyses based upon equations like (4.6) should be avoided for pure metals except perhaps in the region of interband absorption, where the effective values of γ are sufficiently high that no inconsistencies can occur.

Finally, we wish to make several comments concerning the validity of the present calculation. The work of Chester[9] indicates that the Boltzmann equation used in Sec. II is valid (i) when the wave vector k is small compared to the Fermi wave vector k_F and (ii) when the frequency ω is small compared to $\epsilon_F/\hbar$, ϵ_F being the Fermi energy. The first condition is apparently well-satisfied in the present calculation since, although the surface impedance integrals over Q_z run from zero to infinity, less than 0.2% of the contribution to these integrals comes from $Q_z>k_F/k_p$, where k_p is the plasma wave vector, and less than 0.1% from $Q_z>2k_F/k_p$. Thus, any changes in the dielectric functions necessitated by the fact that the wave vector is not small compared to k_F should produce only a minor effect. However, if there occurs very pronounced high-wave-vector structure in the "correct" dielectric functions which our present expressions do not possess, the surface impedances calculated herein could be in error. Such a possibility is being investigated.

Much of the "new" absorptance for P polarization occurs in a frequency range where condition (ii) above is not satisfied, i.e., $\omega\sim\epsilon_F/\hbar$. This does not appear to be a problem, though, as can be seen from the following argument. The derivation of Sec. III, which is not dependent upon the Boltzmann equation, indicates that the form of the surface impedances as obtained in Sec. II is valid, and thus any difficulty with the present calculation must be associated with the expressions for ϵ_t and ϵ_l (or ϵ_w). But, when $\tau\rightarrow\infty$, our expressions for the dielectric functions are the long-wavelength limit of the Lindhard dielectric functions, and thus involve a wave-vector limitation but not a frequency

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

[25] J. C. Phillips, Solid State Phys. **18**, 56 (1966). See p. 157.

limitation. The wave-vector limitation has been discussed above and does not appear to be a problem. Thus we conclude that the present results are valid.

A final bit of evidence concerning the validity of the use of the present dielectric functions is the insensitivity of the results to the replacement of ϵ_l by ϵ_w. These functions are quite different in structure but the replacement of one by the other produces a negligible difference in the surface impedances (see Ref. 18).

Note added in proof. It has been pointed out to us by Dr. R. H. Ritchie that a terse derivation of the expressions (3.15) and (3.21) for the surface impedances at non-normal angles of incidence appears in V. P. Silin and E. P. Fetisov, Zh. Eksperim. i Teor. Fiz. **41**, 159 (1961) [English transl.: Soviet Phys.—JETP **14**, 115 (1962)]. These authors, however, were concerned primarily with the application of these expressions to classical and relativistic plasmas and do not indicate that they imply the existence of an additional absorption peak.

It should be noted, in addition, that J. G. Collins [Appl. Sci. Res. Sec. B **7**, 1 (1958)] showed that two frequency-dependent dielectric functions are needed to characterize optical experiments at non-normal angles of incidence.

ACKNOWLEDGMENTS

We are indebted to Dr. S. H. Liu and Dr. D. W. Lynch for many helpful discussions. We also thank T. E. Anderson and G. W. Hedrick for their assistance with the numerical computations.

APPENDIX A

We are here interested in evaluating the Fourier transforms of the kernels of Eq. (2.18). Consider, as an example, the transform of $\bar{K}(z-s)$ given by Eq. (2.18e). We have

$$\bar{k}(q_z)=\int_{-\infty}^{\infty} dz\, e^{-iq_z z}\bar{K}(z)$$

$$=\int_{-\infty}^{0} e^{-iq_z z}dz\int_0^{\pi/2} d\theta \sin\theta\, J_0\{q_x z\tan\theta\}e^{\eta z}$$

$$-\int_0^{\infty} e^{-iq_z z}dz\int_0^{\pi/2} d\theta\sin\theta\, J_0\{q_x z\tan\theta\}e^{-\eta z}$$

$$=2i\int_0^{\pi/2}\sin\theta d\theta\int_0^{\infty} dz\sin q_z z\, J_0\{q_x z\tan\theta\}e^{-\eta z}.$$

The integral on z can be done using a table of Laplace transforms[26] from which we obtain

$$\int_0^{\infty} dz \sin q_z z\, J_0\{q_x z\tan\theta\}e^{-\eta z}$$

$$=(2i)^{-1}[\{(\eta-iq_z)^2+(q_x\tan\theta)^2\}^{-1/2} -\{(\eta+iq_z)^2+(q_x\tan\theta)^2\}^{-1/2}].$$

Keeping in mind the fact that η is dependent upon θ, the θ integration can then be done with the result that

$$\bar{k}=\frac{q_z}{q^2}\left[2i-\frac{1}{ql'}\ln\left\{\frac{1+iql'}{1-iql'}\right\}\right], \tag{A1e}$$

with

$$q=(q_x^2+q_z^2)^{1/2} \tag{A2}$$

and

$$l'=\frac{l}{1-i\omega\tau_0}. \tag{A3}$$

In the same fashion, we find

$$k''=-\frac{1}{q^3}\left[\frac{(2q_x^2-q_z^2)}{ql'}+i\left\{\frac{q_x^2}{(ql')^2}-\tfrac{1}{2}q_z^2\left(1+\frac{1}{(ql')^2}\right)\right\}\times\ln\left\{\frac{1+iql'}{1-iql'}\right\}\right], \tag{A1a}$$

$$k'=-\frac{q_xq_z}{q^3}\left[\frac{3i}{ql'}-\left\{\frac{1}{(ql')^2}+\frac{1}{2}\left(1+\frac{1}{(ql')^2}\right)\right\}\times\ln\left\{\frac{1+iql'}{1-iql'}\right\}\right], \tag{A1b}$$

$$k'=-\frac{q_x}{q^2}\left[2+\frac{i}{ql'}\ln\left\{\frac{1+iql'}{1-iql'}\right\}\right], \tag{A1c}$$

$$\tilde{k}=\frac{1}{q^3}\left[\frac{(2q_z^2-q_x^2)}{ql'}+i\left\{\frac{q_z^2}{(ql')^2}-\tfrac{1}{2}q_x^2\left(1+\frac{1}{(ql')^2}\right)\right\}\times\ln\left\{\frac{1+iql'}{1-iql'}\right\}\right], \tag{A1d}$$

$$k=-\frac{i}{q}\ln\left\{\frac{1+iql'}{1-iql'}\right\}. \tag{A1f}$$

APPENDIX B

Reuter and Sondheimer (RS) have generalized their calculation of the surface impedance at normal incidence for specular reflection to the case of non-normal incidence without allowing explicitly for charge fluctua-

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[26] See, for example, G. E. Robert and H. Kaufman, *Table of Laplace Transforms* (W. B. Saunders Co., Philadelphia, 1966).

tions inside the metal. It is therefore of interest to compare our exact results with those of RS.[1]

If we translate Eq. (60) in RS into our notation and use the relationship

$$\bar{Z}' \equiv \frac{c}{4\pi}\bar{Z} = -\frac{4\pi i\omega l}{c^2}\frac{f(0)}{f'(0)}, \qquad \text{(B1)}$$

for the surface impedance $\bar{Z}$ as calculated by RS, we find

$$\bar{Z}' = \frac{2i\Omega}{\pi}\int_0^\infty \frac{dQ_z}{\Omega^2\epsilon_t - Q^2} = Z_S', \qquad \text{(B2)}$$

where Z_S' is the same quantity defined by Eq. (3.21). The prescription of RS to calculate the reflectance for non-normal incidence is to replace the dielectric constant $\epsilon(\omega)$ in any classical equation by the quantity $\sin^2\theta + (\bar{Z}')^{-2}$ (or $\sin^2\theta + (Z_S')^{-2}$). Therefore, the classical equation (3.28) for $Z_P'^{(\mathrm{cl})}$ becomes

$$\begin{aligned}\bar{Z}_P' &= \frac{(Z_S')^{-1}}{\sin^2\theta + (Z_S')^{-2}} \\ &= \frac{Z_S'}{1+\sin^2\theta (Z_S')^2} &\text{(B3)} \\ &= \frac{2i\Omega}{\pi}\int_0^\infty \frac{dQ_z}{\Omega^2\epsilon_t - Q^2}\Bigg/ \\ &\qquad \left[1+\sin^2\theta\left(\frac{2i\Omega}{\pi}\int_0^\infty \frac{dQ_z}{\Omega^2\epsilon_t - Q^2}\right)^2\right]. &\text{(B4)}\end{aligned}$$

The classical surface impedance $Z_P'^{(\mathrm{cl})}$ on the left-hand side of Eq. (3.28) is now denoted by $\bar{Z}_P'$, a quantity which, according to RS, is supposed to be the true surface impedance for P polarization. Similarly, Eq. (3.29) becomes

$$\bar{Z}_S' = Z_S' = \frac{2i\Omega}{\pi}\int_0^\infty \frac{dQ_z}{\Omega^2\epsilon_t - Q^2}. \qquad \text{(B5)}$$

We can now insert $\bar{Z}_P'$ and $\bar{Z}_S'$ into the generally valid equations (3.24) and (3.27) (in place of the correct Z_P' and Z_S') to find the reflectances R_P and R_S and absorptances A_P and A_S.

Equation (B5) shows that the surface impedance $\bar{Z}_S'$ for S polarization, as found by RS, is correct; the same is true for the absorptance A_S. However, Eq. (B4), which gives the surface impedance for P polarization, is clearly different from the correct equation (3.15). Equations (B4) and (3.24) have been used to calculate the absorptance A_P for potassium, taking $\theta = 75°$ and $\gamma = 10^{-4}$. The result is shown by the dotted line in Fig. 8. The absorptance agrees with the exact result (solid line) at low frequencies in the "usual" anomalous region, but at higher frequencies it drops down to the classical value (dashed line) and completely fails to reproduce the "new" peak centered at $\Omega \sim 0.4$; for this reason, we can attribute this peak to the presence of charge fluctuations. At higher frequencies ($\Omega \gtrsim 1$) the classical, exact, and RS absorptances all agree.

Combining Eqs. (B4) and (B5), we find that $\bar{Z}_S'$ and $\bar{Z}_P'$ are related by the expression

$$\bar{Z}_P' = \frac{\bar{Z}_S'}{1+\sin^2\theta(\bar{Z}_S')^2}, \qquad \text{(B6)}$$

which, according to Eq. (4.4), is the condition for the existence of a unique $\epsilon(\omega)$. In the region of the "new" peak, where the RS and exact absorptances are different, $Z_P' \neq \bar{Z}_P'$. Therefore, the exact surface impedances Z_P' and Z_S' do not satisfy Eq. (4.4), and a unique value of $\epsilon(\omega)$ does not exist.