Optical transmittance and reflectance and dynamic current density for thin metallic films

J. Szczyrbowski*

Department of Solid State Physics, Institute of Metallurgy, Academy of Mining and Metallurgy, al Mickiewicza 30, Cracow, Poland

K. Schmalzbauer and H. Hoffmann

Institut für Angewandte Physik der Universität Regensburg, Universitätsstr. 31, D-8400 Regensburg, Federal Republic of Germany (Received 26 November 1984)

Formulas for reflection and transmission coefficients R and T for electromagnetic radiation in thin metallic films are given, where the anomalous skin effect and electron scattering at the film boundaries are taken into account. The equations are valid in the near-infrared, visible, and ultraviolet spectral range, and at low frequencies where the skin effect is nearly classical. The classical size effect in the optical absorption for different values of the specularity parameters is considered. The observed oscillatory nature of the absorptance provides a promising method for verifying the boundary conditions for the free electrons at the film surfaces. Finally, a generalized Fuchs formula for the dynamic current density is developed, which is valid in the same frequency ranges as R and T.

I. INTRODUCTION

In this paper the influence of the skin effect on the optical transmission and reflection of thin films will be considered. This problem was theoretically investigated by Reuter and Sondheimer¹ for the case of bulk material. They gave an exact relation for the electric field in the metal, assuming the free electrons to be either totally specularly or diffusely reflected at the sample boundary (specularity parameter p = 1 or 0). Dingle² has given approximate relations for the transmission and reflection amplitudes t and r for thin films with p = 0 at both boundaries. Hutchison and Hansen³ have tried to extend the Dingle consideration for all values of the p parameter. The same was recently done by Szczyrbowski *et al.*⁴

Unfortunately, it turned out that the relation for the current density $J(\omega,z)$ given by Hutchison and Hansen [relation (9) in that paper] is not complete. We shall see that it represents only the special case when the specularity parameter at one of the boundaries equals zero. One of the consequences of this limitation was the theoretically obtained weak influence of the *p* parameter on the optical properties of thin metallic films in the near infrared (NIR).

The aim of this paper is to give approximate relations for r and t for thin homogeneous metallic films with values of the specularity parameters at both film boundaries in the range 0–1. In our solution of the wave equation we use the method developed by Dingle.^{2,5–7}

II. SOLVING THE WAVE EQUATION

We consider a system given in Fig. 1. The sample is limited by the surfaces z=0 and z=d. A plane electromagnetic wave is incident from the z direction (normal incidence). Omitting the time factor throughout, one can

write the electric field as

$$E_1 = \iota e^{-i(\omega/c)n_1 z} + \rho e^{i(\omega/c)n_1 z} \text{ on the left-hand side },$$

$$E_3 = \gamma e^{-i(\omega/c)n_3 z} \text{ on the right-hand side}$$
(1)

in Fig. 1, where n_1, n_3 are the refractive indices of the surrounding media 1,3 and ι , ρ , and γ are the amplitudes of the incident, reflected, and transmitted field, respectively.

Explicit relations for the complex coefficients of reflection, $r = \rho/\iota$, and transmission, $t = \gamma/\iota$, may be obtained using the boundary conditions for Maxwell's equations at each interface. They are

$$\iota + \rho = E(0), \quad \gamma = E(d) ,$$

$$\iota - \rho = \frac{ic}{\omega\mu} E'(0), \quad \gamma = \frac{ic}{\omega\mu} E'(d) ,$$
(2)

where E and E' are the electric field and its derivative in the sample, μ is the relative magnetic permeability, and ϵ_0 the permittivity of free space. Thus to obtain the expres-

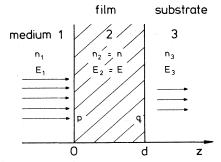


FIG. 1. Schematic diagram for the propagation of an electromagnetic wave through a film-substrate system.

32

763

©1985 The American Physical Society

sions for r and t one has to know the field E in the sample area which obeys the wave equation (SI units)

$$\frac{d^2 E(z)}{dz^2} + \frac{\omega^2}{c^2} \mu (1+S) E(z) = \frac{i\omega\mu}{c^2\epsilon_0} J(\omega,z) .$$
(3)

The quantity $(1+S) = \epsilon_l$ is the residual dielectric constant which takes care of the displacement current, atomic polarization, etc. J(z) is the current density of the free electrons in the sample and is given by

$$J(\omega,z) = -2e \left[\frac{m}{h}\right]^{3} \int \int \int v_{x} f_{1} dv_{x} dv_{y} dv_{z} , \qquad (4)$$

where m is the effective mass of an electron.

 f_1 may be obtained from the linearized Boltzmann transport equation

$$\frac{\partial f_1}{\partial z} + \frac{wf_1}{\tau v_z} = \frac{e}{mv_z} \frac{\partial f_0}{\partial v_x} E(z) , \qquad (5)$$

where it is assumed that the distribution function f may be written as $f = f_0 + f_1$ ($f_1 \ll f_0$, f_0 is the Fermi function, $w = 1 + i\omega\tau$, and τ is the relaxation time). The general solution of Eq. (5) is given by Reuter and Sondheimer¹ and f_1 has the form

$$f_{1} = \exp\left[-\frac{wz}{\tau v_{z}}\right] \left[C(v) + \frac{e}{mv_{z}}\frac{\partial f_{0}}{\partial v_{x}} \times \int_{0}^{z} E(y) \exp\left[\frac{wy}{\tau v_{z}}\right] dy\right], \quad (6)$$

where C(v) is an arbitrary function of the velocity which is determined by the appropriate conditions at the film boundaries. We use the Lucas⁸ boundary conditions

$$f_{0}+f_{1}^{+}(v_{z},z=0)=p[f_{0}+f_{1}^{-}(-v_{z},z=0)]+G_{1},$$

$$f_{0}+f_{1}^{-}(v_{z},z=d)=q[f_{0}+f_{1}^{+}(-v_{z},z=d)]+G_{2},$$
(7)

where $f_1^+ = f_1$ for $v_z > 0$, $f_1^- = f_1$ for $v_z < 0$ and p and q are the specularity parameters related to the surfaces z = 0 and z = d, respectively. The magnitudes G_1 and G_2 are connected with the diffuse scattering of electrons at the film boundaries. Using (6) and (7), from (4) one obtains

$$J(\omega,z) = \frac{2\pi (emv_F)^2}{h^3} \int_1^\infty F \, ds \, \int_0^d E(y) \, dy \left[p e^{-w(y+z)s/l} + pq e^{w(|y-z|-2d)s/l} + q e^{w(y+z-2d)s/l} + e^{-w|y-z|s/l} \right], \tag{8}$$

where

$$F = \left(\frac{1}{s} - \frac{1}{s^3}\right) \frac{1}{1 - pqe^{-2wds/l}} ,$$
(9)

 v_F is the Fermi velocity and *l* the mean-free path of an electron. Relation (8) is the same as given by Dimmich and Warkusz⁹ for polycristalline films assuming the electron transmission through the grain boundaries to be unity. One can see now that at q = 0 Eq. (8) becomes the expression for the current density given by Hutchison and Hansen.³

It is convenient for mathematical analysis to introduce dimensionless coordinates z = z/l and y = y/l. Emphasizing that below also the film thickness d is measured in units of the mean-free path l, Eq. (3) then takes the form

$$\frac{d^{2}E(z)}{dz^{2}} + \frac{\omega^{2}l^{2}}{c^{2}}\mu(1+S)E(z) = \frac{i\omega l^{2}\mu}{c^{2}\epsilon_{0}}J(\omega,z) .$$
(10)

To obtain the electric field from (10) with (8) we consider first the possible solution $E^{(1)} = e^{-uwz}$ which results in

$$(u^2 - \eta)e^{-uwz} = \xi[K(u)e^{-uwz} + \Lambda], \qquad (11)$$

where

$$\eta = -\left[\frac{\omega l}{cw}\right]^{2} (S+1)\mu ,$$

$$K(u) = \int_{1}^{\infty} F\left[\frac{1}{s-u} + \frac{1}{s+u}\right] (1-pqe^{-2swd}) ds = \int_{1}^{\infty} \left[\frac{1}{s} - \frac{1}{s^{3}}\right] \left[\frac{1}{s-u} + \frac{1}{s+u}\right] ds$$

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

$$\Lambda = \int_{1}^{\infty} F\frac{1}{s+u} ds (pe^{-swz} - pe^{-uwd}e^{-sw(d+z)} + pqe^{-sw(2d-z)} - e^{-uwd}e^{-sw(d-z)})$$

$$+ \int_{1}^{\infty} F\frac{1}{s-u} ds (-e^{-swz} + qe^{-uwd}e^{-sw(d-z)} + pqe^{-sw(d+z)} - qe^{-sw(2d-z)}) ,$$
(12)

and

$$\xi = i \frac{\omega}{c^2 \epsilon_0} \frac{2\pi (emv_F)^2 l^3 \mu}{h^3 w^3}$$

$$\xi = i\frac{3}{4}\omega_p^2 \frac{\omega \tau l^2 \mu}{c^2 w^3} = i\frac{3}{2} \frac{l^2}{w^3 \delta^2}$$

where $\delta = [2\epsilon_0 c^2 / \mu \omega \sigma(0)]^{1/2}$ is the classical penetration depth.

The next contribution to the electric field may be taken as $E^{(2)} = \xi \Lambda$. Setting $E = E^{(1)} + E^{(2)}$ into the wave equation (10), one obtains:

$$(u^{2} - \eta)e^{-uwz} = \xi K(u)e^{-uwz} + O(\xi^{2}), \qquad (13)$$

where $O(\xi^2)$ is a term of order ξ^2 . This process can be repeated to form a series in ξ which converges for $|\xi| < 1$. Thus, generally, one can write

$$E^{+} = E^{(1)} + E^{(2)} + \cdots + O(\xi^{n}) .$$
(14)

Since by (12) K(u) is an even function of u, there is a further solution E^- obtained from (14) simply by replacing u by -u. Thus the general solution of (10) with (8) may be written in the form

$$E(z) = E_{0+}E^{+} + E_{0-}E^{-}$$
(15)

where E_{0+} and E_{0-} are constants.

....

III. RELATIONS FOR r AND t

In this section we assume additionally that $|\xi| \ll 1$ and $|\eta| \ll 1$. One consequence of this is that $|u| \ll 1$. This restricts the validity of the further considerations to either low frequencies, where the skin effect is nearly classical, or very high frequencies from the near infrared up to the ultraviolet (UV). A more detailed discussion of these assumptions is given in the next chapter. With sufficient accuracy one may then take

$$E(z) = E_{0+}e^{-uwz} + E_{0-}e^{uwz}, \qquad (16a)$$

$$\frac{ic}{\omega l\mu} E'(z) = \frac{ic}{\omega l\mu} \left[E_{0+}(E^+)' + E_{0-}(E^-)' \right] = E_{0+} n(z) e^{-u\omega z} - E_{0-} n'(z) e^{u\omega z} , \qquad (16b)$$

where

$$n(z) = \frac{n_b}{\mu} - \frac{3}{4} \left(\frac{\omega_p \tau}{w}\right)^2 \frac{v_F}{c} [(1-p)e^{uwz}I_0e^{-swz} + p(1-q)e^{-uw(d-z)}I_1e^{-swz}]$$

$$-q(1-p)e^{uwz}I_2e^{swz}-(1-q)e^{-uw(d-z)}I_1e^{swz}];$$
(17)

$$n'(z) = \frac{n_b}{\mu} + \frac{3}{4} \left[\frac{\omega_p \tau}{w} \right]^2 \frac{v_F}{c} [(1-p)e^{-uwz}I_0e^{-swz} + p(1-q)e^{uw(d-z)}I_1e^{-swz}]$$

$$-q(1-p)e^{-uwz}I_2e^{swz}-(1-q)e^{uw(d-z)}I_1e^{swz}];$$

and

$$I_{K} = \int_{1}^{\infty} \left[\frac{1}{s^{3}} - \frac{1}{s^{5}} \right] \frac{e^{-Kswd}}{1 - pqe^{-2swd}} ds \text{ with } K = 0, 1, 2.$$

The expressions $I_K e^{\pm suz}$ in Eqs. (17) represent an abbreviated notation and it means that the integrand of I_K is multiplied by the appropriate exponential function. n_b is the complex refractive index of the considered medium where the surface contribution is neglected. The appearance of l in (16b) is a consequence of the dimensionless notation of the wave equation. From (12) one obtains $K(u) \simeq 4/3$ and from (13):

$$uw = \frac{i\omega l}{c} \left[\left(1 + S - i\frac{\omega_p^2 \tau}{\omega w} \right) \mu \right]^{1/2} = \frac{i\omega l}{c} n_b , \qquad (18)$$

where $\omega_p^2 = Ne^2/(\epsilon_0 m)$ is the plasma frequency. The relation (18) is the familiar expression for the dielectric constant of a free-electron gas with correction for residual polarization.

The relations for r and t may be obtained using the interference matrix M defined as^{9,10}

$$\begin{pmatrix} E(0) \\ H(0) \end{pmatrix} = M \begin{pmatrix} E(d) \\ H(d) \end{pmatrix}$$

with

$$\boldsymbol{M} = \begin{bmatrix} \boldsymbol{m}_{11} & \boldsymbol{m}_{12} \\ \boldsymbol{m}_{21} & \boldsymbol{m}_{22} \end{bmatrix}$$

and

$$r = \frac{(m_{11} + m_{12}n_3)n_1 - (m_{12} + m_{22}n_3)}{(m_{11} + m_{12}n_3)n_1 + (m_{12} + m_{22}n_3)},$$

$$t = \frac{2n_1}{(m_{11} + m_{12}n_3)n_1 + (m_{12} + m_{22}n_3)}.$$
(19)

The matrix elements can be found from (2) and (16). After some transformations one obtains

$$m_{11} = \frac{n'(d)e^{uwd} + n(d)e^{-uwd}}{n'(d) + n(d)},$$

$$m_{12} = \frac{e^{uwd} - e^{-uwd}}{n'(d) + n(d)},$$

$$m_{21} = \frac{n(0)n'(d)e^{uwd} - n'(0)n(d)e^{-uwd}}{n'(d) + n(d)},$$

$$m_{22} = \frac{n'(0)e^{-uwd} + n(0)e^{uwd}}{n'(d) + n(d)}.$$
(20)

The explicit expressions for the optical constants are

$$n(0) = \frac{n_b}{\mu} - \frac{3}{4} \left[\frac{\omega_p \tau}{w} \right]^2 \frac{v_F}{c} (1-p) \\ \times [I_0 - (1-q)e^{-uwd}I_1 - qI_2] ,$$

$$n(d) = \frac{n_b}{\mu} + \frac{3}{4} \left[\frac{\omega_p \tau}{w} \right]^2 \frac{v_F}{c} (1-q) \\ \times [I_0 - (1-p)e^{uwd}I_1 - pI_2] ,$$

$$n'(0) = \frac{n_b}{\mu} + \frac{3}{4} \left[\frac{\omega_p \tau}{w} \right]^2 \frac{v_F}{c} (1-p) \\ \times [I_0 - (1-q)e^{uwd}I_1 - qI_2] ,$$

$$n'(d) = \frac{n_b}{\mu} - \frac{3}{4} \left[\frac{\omega_p \tau}{w} \right]^2 \frac{v_F}{c} (1-q) \\ \times [I_0 - (1-p)e^{-uwd}I_1 - qI_2] .$$
(21)

Therefore Eqs. (19) reduce to the familiar expressions

$$r = \frac{r_{12} + r_{23} U e^{-2uwd}}{1 + r'_{12} r_{23} e^{-2uwd}} ,$$

$$t = \frac{t_{12}t_{23}e^{-uwd}}{1 + r'_{12}r_{23}e^{-2uwd}} ,$$

where the Fresnel amplitudes are defined as follows:

$$r_{12} = \frac{n_1 - n(0)}{n_1 + n(0)}, \quad r'_{12} = \frac{n_1 - n'(0)}{n_1 + n(0)}$$
$$r_{23} = \frac{n(d) - n_3}{n'(d) + n_3}, \quad t_{ij} = 1 + r_{ij}$$

and

$$U = \frac{n_1 + n'(0)}{n_1 + n(0)} \; .$$

The measured intensity coefficients R and T for reflection and transmission of a film on a transparent substrate are given by

$$R = R' + T'^{2} \frac{R_{31}}{1 - R_{31}R''} ,$$

$$T = \frac{(1 - R_{31})T'}{1 - R_{31}R''} ,$$
(23)

where

$$T' = \frac{n_3}{n_1} |t|^2, \ R' = |r|^2, \ R_{31} = \left(\frac{n_3 - n_1}{n_3 + n_1}\right)^2.$$

R'' represents the reflection coefficient of the film for light incident from the semi-infinite substrate. Because R'' is multiplied by R_{31} , which is much less than 1 in the expression for it, one can practically neglect the considered skin effect. Thus in the numerical calculation we can use the expression

$$R'' = \left| \frac{r_{32} + r_{21}e^{-uwd}}{1 + r_{12}r_{23}e^{-2uwd}} \right|^2,$$
(24)

where r_{ij} are the Fresnel coefficients defined as

$$r_{32} = \frac{n_3 - n'(d)}{n_3 + n(d)}, \quad r_{21} = \frac{n'(0) - n_1}{n'(0) + n_1}$$

IV. DISCUSSION AND CONCLUSIONS

Our discussion will be limited to the case of nonmagnetic materials, i.e., we assume $\mu = 1$.

The expressions (22) are similar to those in the literature^{10,11} for metallic or dielectric films, where the skin effect is neglected. The influence of the skin effect and the interaction of the electrons with the film boundaries are described by U and the new definitions of r_{12} and r_{23} .

The validity of (22) is conditioned by two assumptions: $|\xi| \ll 1$ and $|\eta| \ll 1$. One can consider three cases:

(i) For $\omega \tau \ll 1$ one has

$$|\xi| \simeq \frac{3}{4}\omega_p^2 \tau^2 \left(\frac{v_F}{c}\right)^2 \omega \tau$$

and

(22)

$$\mid \eta \mid \simeq \omega^2 \tau^2 \left(rac{v_F}{c}
ight)^2 (S+1) \; .$$

The maximum value of $(v_F/c)^2$ for any metal is of the order of 10^{-5} . Thus the second inequality $|\eta| \ll 1$ is always fulfilled when $\omega \tau \ll 1$, but $|\xi| \ll 1$ is true when $\omega_p \tau \leq c/v_F$. If we take the typical parameter values for gold $(\hbar \omega_p \simeq 9 \text{ eV}, \tau \simeq 2 \times 10^{-14} \text{ sec})$ we obtain $\omega_p \tau \simeq c/v_F$, which means that for this material $|\xi| \ll 1$ is obeyed.

(ii) For $\omega \tau \gg 1$ the discussed $|\xi|$ and $|\eta|$ magnitudes may be written as $|\xi| \simeq \frac{3}{4} (\omega_p / \omega)^2 (v_F / c)^2$ and $|\eta| \simeq (v_F / c)^2 (S + 1)$. Thus again the second inequality $(|\eta| \ll 1)$ is fulfilled, while the first one is obeyed only then when $(\omega_p / \omega)^2 \ll (c / v_F)^2$. For materials such as Au, Ag, and Cu both inequalities are fulfilled when $\hbar \omega > 0.2$ eV.

(iii) For $\omega \tau \simeq 1$ the discussed assumptions are fulfilled when $(\omega_p \tau)^2 \ll (c/v_F)^2$.

The optical constants n and n' [Eq. (21)] depend on both the film thickness d and the specularity parameters pand q. For bulk material or thick films ($d \gg 1$, in practice $d \ge 2$) the dependence on d disappears, and in this case only n(0) is of importance. It becomes

$$n(0) = n_b - \frac{3}{16} \frac{(\omega_p \tau)^2 v_F}{w^2 c} (1 - p) = n_b + n_s .$$
 (25)

The second term n_s in (25) represents the surface contribution resulting from the interaction of free electrons with the sample boundary and it disappears when p = 1.

In the case of metal films, starting from the NIR region up to the UV, the inequalities $\omega \tau \gg 1$ and $|\xi| \ll 1$ are obeyed. Thus, especially from (25), the real value of n(0)is influenced. For gold the corresponding contributions to the refractive index are shown in Fig. 2. As we see, the surface contribution of n at p = q = 0 is of the same order

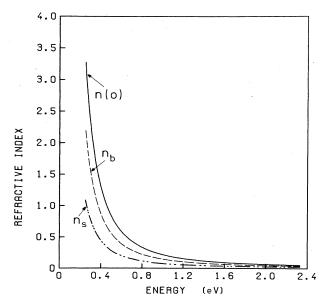


FIG. 2. Computed contributions to the real part of the refractive index $[n(z=0)=n_b+n_s]$ for a hypothetical bulk gold sample $[l=30 \text{ nm}, p=0, m^*=m_e, S=7, \hbar\omega_p=9.0 \text{ eV}$ $(N=5.9\times10^{28} \text{ m}^{-3})].$

as n_b . For metals with a high electron concentration N (e.g., for Al, $N = 18 \times 10^{28} \text{ m}^{-3}$) the surface contribution may be very large. The influence of n_s on the reflection spectrum of a hypothetical gold film (of thickness 500 nm) at different p values is shown in Fig. 3. As we see, the surface contribution may lower the measured reflection by about 0.5%. One can expect also that the observed strong dependence of optical properties of metals on the preparation conditions is at least in part connected with the value of the p parameter.

If d < 1 one has to use the complete relations for n and n' [Eqs. (17)]. The plots of R and T as a function of energy for two hypothetical gold films of different thickness at different p values are shown in Fig. 4. Firstly, one can see that the influence of the p parameter on the reflection spectrum is relatively larger in the low-energy region, where the reflectivity may be several percent lower for q = p = 0 than for q = p = 1. The value of this effect depends on d (the ratio of the thickness to the mean-free path) and increases with decreasing d value. Comparing our results with the conclusions made in Ref. 4 one must say that the influence of the specularity parameters on the reflection and transmission spectra is essentially greater than in Ref. 4. Concluding, one can expect that the value of p may now be determined from optical measurements.

Since w is a complex quantity, all the integrals I_K are periodic functions of $K\omega\tau d$. This periodicity was theoretically predicted by Dingle⁵ for p = q = 0. The oscillations should appear in the measured transmission and reflection spectra and especially in the absorption A = 1 - R - Tand its derivative. The plots of A and $dA/d(\hbar\omega)$ as a function of energy for different values of p = q are shown in Figs. 5(a) and 5(b). One can see that in the thickness range considered the absorptivity increases as the thickness is decreased and for low values of p the distinct oscillations of A appear. For p = q the phase of the oscillatory

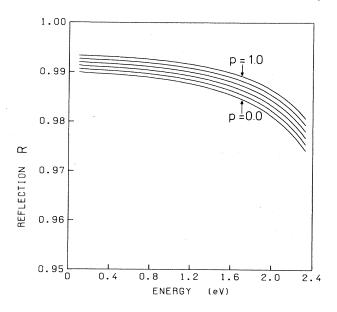


FIG. 3. Reflection spectra of a hypothetical bulk gold sample for different p values (0, 0.2, 0.4, 0.6, 0.8, 1.0). The other parameters are the same as in Fig. 2.

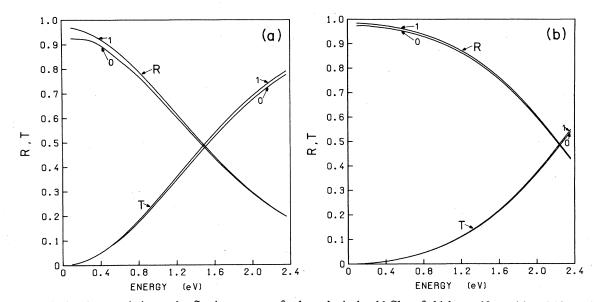


FIG. 4. Calculated transmission and reflection spectra of a hypothetical gold film of thickness 10 nm (a) and 20 nm (b) for two values of the p parameter (p = q = 0 and p = q = 1). The other parameters as in Fig. 2.

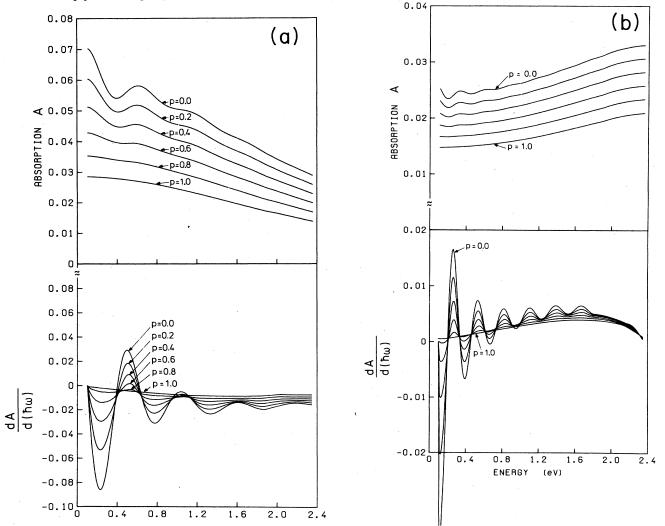


FIG. 5. Computed values of the absorption A = 1 - T - R and the derivative $dA/d(\hbar\omega)$ vs energy for two gold films [thickness 10 nm (a), 20 nm (b)] for different values of p = q (0, 0.2, 0.4, 0.6, 0.8, 1.0) and the values of the other parameters as in Fig. 2.

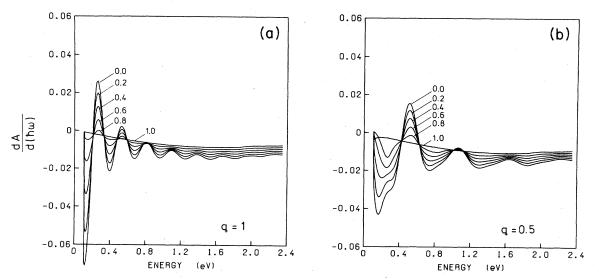


FIG. 6. Plots of the derivative $dA/d(\hbar\omega)$ vs the energy of a 10-nm-thick gold film (the parameters are given in Fig. 2) for q = 1 (a) and q = 0.5 (b) with p ranging from 0 to 1.

function in A is given by $\omega \tau d$ and at the maxima $\omega \tau d = K 2\pi t/T$, where t is the minimum time for an electron to cross the film, T is the period of radiation and $K = 1, 2, \ldots$. This oscillatory absorption is called the classical size effect in the optical absorption. The period length may be understood by analyzing the relations (21) for n and n'. For $|uwd| \ll 1$ and p = q the expression in brackets in (21) may be well approximated by the Fuchs integral [see relation (28)]. This gives

$$n(0) \simeq n_b - \frac{3}{4} \frac{(\omega_p \tau)^2 v_F}{w^2 c} (1-p) \\ \times \int_1^\infty \left[\frac{1}{s^3} - \frac{1}{s^5} \right] \frac{1-e^{-swd}}{1-pe^{-swd}} ds .$$

The relations for the other *n* and *n'* may be written in an analogous form. Therefore, the periodicity in *A* is connected only with the function e^{-swd} in the integrand. On the other hand for p = 1 or q = 1, instead of e^{-swd} , the magnitude e^{-2swd} appears in the Fuchs integrand. Thus, the period length of the oscillatory function in *A* [and $dA/d(\hbar\omega)$] is half that at p = q [compare Figs. 5(a) and 6(a)]. For $p \neq 1$, $q \neq 1$, and $p \neq q$ both e^{-swd} and e^{-2swd} remain in the expressions for *n* and *n'*. Thus two components of the frequency appear in the absorptivity spectrum [Fig. 6(b)].

Concluding, one can say that study of these oscillations may lead to valuable information concerning the transport parameters in thin metallic film as follows: (i) The appearance of oscillations in the absorption spectrum means that at least one of the specularity parameters is different from unity, (ii) the appearance of the second component of the frequency tells us that p is different from q. It is worth noting here that this fact cannot be simply deduced from measurements of the dc conductivity σ only. For instance, using the Lucas⁸ formula one can show that for three different sets of parameters (l,p=q),(l'=2l, p=1, q), and (l'=2l, p, q=1) the ratio σ/σ_b is exactly the same, thus, these three cases cannot be distinguished. From the experimental point of view this conclusions also remain true, if the corresponding p and qvalues differ strongly, (iii) since the magnitude $\omega \tau d = \omega l v_F d$ does not depend on the mean-free path of an electron (d is measured in units of l), from the length of the period in A, the Fermi velocity v_F may be immediately deduced.

From an experimental point of view, care must be taken when measuring A. Figure 5 shows that the oscillation amplitude is about 0.001, i.e., the measurement accuracy must be much better than 0.1%. The film surfaces should be sufficiently smooth, otherwise the oscillations would be smeared out. It is worth noting, however, that the surface roughness only lowers the amplitude of the oscillations, but does not change the length of the period.

Finally let us discuss the dynamic current density $J(\omega,z)$. In our approximation $(|\xi| \ll 1)$ for p=q it is given by

$$J(\omega,z) = \sigma_b(\omega) \left\{ e^{-u\omega z} - \frac{3}{4}(1-p) \int_1^\infty \hat{F} \, ds (e^{-s\omega z} + pe^{-u\omega d} e^{-s\omega(d+z)} + pe^{-s\omega(2d-z)} + e^{-u\omega d} e^{-s\omega(d-z)}) \right\} E_0 \tag{26}$$

with $\sigma_b(\omega) = \sigma_b(0)/w$, where $\sigma_b(0)$ is the bulk dc conductivity, and

$$\widehat{F} = \left\lfloor \frac{1}{s^2} - \frac{1}{s^4} \right\rfloor \frac{1}{1 - p^2 e^{-2swd}}$$

The first term in (26) gives the familiar expression from the literature for the current density in bulk material. The second one is connected with electron scattering at the boundaries of the sample and is equal to zero for p = 1. The expression for J(z=0) using the assumption $d \gg 1$ may be written as

$$J(\omega,0) = \sigma_b(\omega) [1 - \frac{1}{2}(1-p)] E_0 .$$
(27)

For p=0 we obtain only half the conductivity compared to that in the interior of the sample. Comparing (25) with (27) one can conclude that the influence of the p parameter on the optical constants is smaller than on the electrical conductivity. Additionally one can see from (25) and (27) that the known simple relationship between the dynamic conductivity and the complex refractive index is not valid here.

To compare our results for dynamic current density with those given by Fuchs¹² one has to find the mean value of $J(\omega,z)$, i.e.,

$$J(\omega) = \langle J(\omega, z) \rangle = \frac{1}{d} \int_0^d J(\omega, z) dz$$

After integration one obtains

$$J(\omega) = \sigma_b(\omega) \left[\frac{1 - e^{-uwd}}{uwd} - \frac{3}{4} \frac{1 - p}{wd} (1 + e^{-uwd}) \times \int_1^\infty \left[\frac{1}{s^3} - \frac{1}{s^5} \right] \frac{1 - e^{-swd}}{1 - pe^{-swd}} ds \left] E_0 .$$

$$(28)$$

Equation (28) is the generalized Fuchs formula, valid for alternate current (ac) also. In the limit $\omega \rightarrow 0$ ($u \rightarrow 0$), expression (28) becomes exactly the Fuchs expression.

ACKNOWLEDGMENTS

One of the authors (J.S.) would like to thank the Alexander von Humboldt Stiftung and the Deutsche Forschungsgemeinschaft for supporting this work.

- *Present address: Institut für Angewandte Physik der Universität Regensburg, Universitätsstr. 31, D-8400 Regensburg, Federal Republic of Germany.
- ¹G. E. H. Reuter and E. H. Sondheimer, Proc. R. Soc. London, Ser. A **195**, 336 (1948).
- ²R. B. Dingle, Physica XIX, 1187 (1953).
- ³F. E. Hutchison and W. N. Hansen, Phys. Rev. B 20, 4069 (1979).
- 4J. Szczyrbowski, J. Dryzek, and A. Czapla, Thin Solid Films 112, 175 (1984).
- ⁵R. B. Dingle, Physica XIX, 311 (1953).

- ⁶R. B. Dingle, Physica XIX, 348 (1953).
- ⁷R. B. Dingle, Physica XIX, 729 (1953).
- ⁸M. S. P. Lucas, J. Appl. Phys. 36, 1632 (1965).
- ⁹R. Dimmich and F. Warkusz, Phys. Status Solidi A 72, 117 (1982).
- ¹⁰M. Born and E. Wolf, *Principles of Optics*, 5th ed. (Pergamon, Oxford, 1975), Sec. 1.6.
- ¹¹Z. Knittl, *Optics of Thin Films* (Wiley, London, 1976), Secs. 2.1–2.5 and 10.3.
- ¹²K. Fuchs, Proc. Cambridge Philos. Soc. 34, 100 (1938).