Effect of surface scattering on optical properties of metallic double-layer films

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The effects of free-electron scattering at film surfaces and at the interface upon the reflection, transmission, and absorption coefficients of metallic double-layer films are studied on the basis of the Maxwell equations and the Boltzmann transport theory. In order to calculate the surface electric current generated by the electric field of an incident electromagnetic wave, the microscopic transport parameters analogous to those introduced in the existing models of dc conductivity are used. The anomalous nature of the skin effect is taken into account. Formulas found for the optical coefficients are valid in the near-infrared, visible, and ultraviolet spectral ranges as well as at low frequencies where the skin effect is nearly classical. It is also shown that the model developed may be used to find the optical coefficients for systems which consist of a bulk metallic base and metallic covering. The results obtained for both kinds of structures are apparently dependent on the values of scattering parameters of electrons at the surfaces and the interface. Among others, the oscillatory nature of the absorption spectrum is observed. The amplitude and period of the oscillations change with the boundary conditions for free-electron scattering. This enables the verification of the values of the scattering parameters found.

I. INTRODUCTION

Interest in optical properties of thin metallic films has been substantial over recent years. Reuter and Sondheimer[1] were the first to theoretically study the effect of the surface scattering of electrons on these properties of bulk metals. They consider a semi-infinite metallic sample and derived equations for the electric field in metal for two limiting cases when electrons are exclusively specularly or diffusely scattered at the sample surface, i.e., the specularity parameter $p=1$ or 0, respectively. Dingle[2-4] gave a very detailed analysis of the optical properties of bulk metals within the Reuter and Sondheimer theory. Moreover, he developed an approximate method of solution of the integro-differential equation obeyed by the electric field in metal and he applied this method to films of arbitrary thicknesses with totally diffuse electron scattering at the surfaces[5] ($p=0$). Hutchison and Hansen[6] tried to improve upon the solution given by Dingle.[5] They took into consideration a case with arbitrary values of the specularity parameter p, but a boundary condition describing the electron scattering was formulated for the external surface of the film, only, i.e., for the surface upon which the electromagnetic wave is incident. The other surface was treated, without employing an appropriate boundary condition, as the one scattering the electrons diffusely. This limitation resulted from the fact that they used the electron distribution function in the form derived by Reuter and Sondheimer[1] for bulk metal, where the electric current generated by the electromagnetic wave becomes negligible sufficiently far from the surface. Thus the formulation of the wave equation given by Hutchinson and Hansen[6] is valid when the specularity parameter at the other film boundary equal zero and, consequently, their model is valid with the same restriction. The facts mentioned above cause the Hutchinson and Hansen theory[6] to show a weaker influence of free-electron surface scattering on optical properties of thin metallic films than can be seen in the case of the general approach. It was not until Dimmich and Warkusz[7] raised the arguments used for deriving the second-order integro-differential wave equation that the general form of this equation was obtained. Based upon the work by Reuter and Sondheimer[1] the authors[7] developed the model for the case of thin metallic films. It has been formulated by use of the Lucas boundary conditions[8] and by means of the Mayadas-Shatzkes method.[9] Thus the electron scattering at both the film surfaces and at the grain boundaries was taken into account. However, using the general form of the wave equation Dimmich and Warkusz[7] derived the exact expression for the optical reflectance of a polycrystalline bulk metal with specularity parameter $p=1$, only. The solutions of this newly formulated wave equation for single-crystalline and polycrystalline metallic films were obtained by Szczyrbowski *et al.*[10,11] They used the Dingle method[5] for this purpose. On the basis of the solutions found, Szczyrbowski *et al.*[10,11] derived formulas for reflection and transmission coefficients in thin metallic films. This set of expressions and the model of dc conductivity by Mayadas and Shatzkes[9] represent a unified physical description of the optical and electrical properties of metallic thin films. It permits the optical characteristics to be related to the dc conductivity in terms of the microscopic parameters when both the optical and electri-

cal characteristics are measured simultaneously. Such examinations have been performed for Au, Ag, and Cu films,[6,12-14] where reasonable agreement between the theoretical and experimental values for both kinds of analyzed quantities has been obtained for the same set of free-electron parameters.

In this paper, the theory of optical properties of metallic double-layer films is formulated. Approximate relations for the reflection, transmission, and absorption coefficients are obtained where the previously developed calculation method[1,5,7,10] is used. The resulting model provides optical characteristics, both in the case of thin double-layer films and bulk metals with thin metallic coverings.

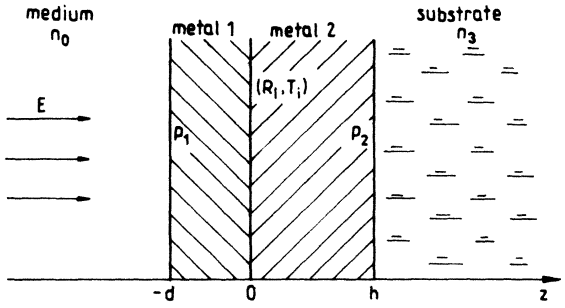

FIG. 1. Metallic double-layer film in the field of the electromagnetic wave.

II. WAVE EQUATION

The two-layer thin-film configuration considered in this analysis is shown in Fig. 1. The overlayer with surfaces at $z=-d$ and $z=0$ is made of metal 1 and the base layer with surfaces at $z=0$ and $z=h$ of metal 2. It is assumed that the conduction electrons in both the layers (metal 1 and metal 2) have an identical Fermi momentum, i.e., no contact potential difference arises at the interface $z=0$. The electric field $E(z)e^{i\omega t}$ is taken to be in the x direction and the magnetic field $H(z)e^{i\omega t}$ in the y direction. In further considerations, the time-dependent factor $e^{i\omega t}$ will be omitted. On eliminating H from Maxwell equations, these are reduced to the following wave equation:

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

where $J(\omega,z)$ is the free-electron current density generated by the electric field $E(z)$ of the incident radiation, $\omega/2\pi$ is the field frequency, μ is the relative magnetic permeability, and ϵ_0 is the permittivity of free space. The quantity $1+S$ arises from the displacement current, internal photoelectric absorption, and atomic polarization.

The current density $J(\omega,z)$ can be obtained from the linearized Boltzmann transport equation for the distribution function $f(\mathbf{v},z)$ of the conduction electrons. For the sample in the electric field $f(\mathbf{v},z)=f_0+f_{1,2}(\mathbf{v},z)$, where f_0 is the Fermi-Dirac equilibrium distribution function and $f_1(\mathbf{v},z)$ and $f_2(\mathbf{v},z)$ are the deviations from this function induced by the electric field for $-d\le z\le 0$ and $0\le z\le h$, respectively. Then the notation that index 1 refers to metal 1 and index 2 to metal 2 will be made. The Boltzmann equation for each layer takes the form

$$\frac{\partial f_{1,2}}{\partial z}+\frac{w_{1,2}f_{1,2}}{\tau_{1,2}v_z}=\frac{e}{m_{1,2}v_z}\frac{\partial f_0}{\partial v_x}E(z)\ , \tag{2}$$

where $\mathbf{v}=(v_x,v_y,v_z)$ is the electron velocity, $w_{1,2}=1+i\omega\tau_{1,2}$, $\tau_{1,2}$ and $m_{1,2}$ are the relaxation times and the effective masses of the electrons. The general solution of Eq. (2) for a plane electromagnetic wave incident along the z direction (Fig. 1.) has the form

$$f_{1,2}(\mathbf{v},z)=\exp\left[-\frac{w_{1,2}(z+d)}{\tau_{1,2}v_z}\right]\left[F_{1,2}(\mathbf{v})+\frac{e}{m_{1,2}v_z}\frac{\partial f_0}{\partial v_x}\int_{-d}^{z}dt\,E(t)\exp\left[\frac{w_{1,2}(t+d)}{\tau_{1,2}v_z}\right]\right]\ , \tag{3}$$

where $F_{1,2}(\mathbf{v})$ are the arbitrary velocity functions determined by the appropriate boundary conditions at the film surfaces and the interface. These conditions should serve as a description of the electron scattering processes. We use the boundary conditions and the microscopic parameters for the surface and interface scattering given by Dimmich and Warkusz,[15]

$$f_0+f_1^+(v_z,z=-d)=p_1[f_0+f_1^-(-v_z,z=-d)]+H_1\ , \tag{4}$$

$$f_0+f_2^-(v_z,z=h)=p_2[f_0+f_2^+(-v_z,z=h)]+H_2\ , \tag{5}$$

$$f_0+f_1^-(v_z,z=0)=R_i[f_0+f_1^+(-v_z,z=0)]+T_i[f_0+f_2^-(v_z,z=0)]+I_1\ , \tag{6}$$

$$f_0+f_2^+(v_z,z=0)=R_i[f_0+f_2^-(-v_z,z=0)]+T_i[f_0+f_1^+(v_z,z=0)]+I_2\ , \tag{7}$$

where $f_{1,2}^+(\mathbf{v},z)=f_{1,2}(\mathbf{v},z)$ for $v_z>0$, $f_{1,2}^-(\mathbf{v},z)=f_{1,2}(\mathbf{v},z)$ for $v_z<0$, and H_1, H_2, I_1, and I_2 account for the diffusely scattered current carriers at the respective surfaces. R_i, T_i, p_1, and p_2 are the phenomenological parameters which can be interpreted as the probabilities that an electron is specularly reflected at the interface $z=0$ (R_i), crosses it without diffuse scattering (T_i), and is specularly reflected at the external surfaces $z=-d$ (p_1) and $z=h$ (p_2) of the double-layer film, respectively. The parameters range between the assumption of complete specularity, $R_i=1$, $T_i=1$, $p_1=1$, $p_2=1$, and complete diffuseness, $R_i=0$, $T_i=0$, $p_1=0$, $p_2=0$, where the restriction that $R_i+T_i\le 1$ must be fulfilled. This is because R_i and T_i describe the additional electron scattering occurring at the interface between the two layers where an electron can undergo two processes, a reflection and a transmission through the interface.

The above treatment of the surfaces is quantitative and

there is no information in our boundary conditions [Eqs. (4)—(7)] about the nature of scattering mechanisms. Various mechanisms have been proposed[16] for the origin of surface scattering. For example, scattering from charge centers located randomly on a crystal surface was considered by Greene and O'Donnell.[17] Scattering from neutral atoms chemisorbed on a film was derived by the use of quantum-mechanical theories by Watanabe and Hiratuka.[18,19] Scattering from a rough surface in the geometrical-optics limit was studied by Ziman[20] and Soffer.[21] One of the consequences of the above-mentioned works is a conclusion that the scattering parameters must at least be dependent on an angle of incidence upon the surface. Thus the boundary conditions in Eqs. (4)—(7) are too simple to be realistic. It should be emphasized, however, that there will be no problem with the generalization of our results by means of the substitution of an expression appropriate for the considered mechanism of scattering into the final formulas (cf. next section) instead of the parameters introduced in Eqs. (4)—(7).

Using the boundary conditions given by Eqs. (4)—(7) and introducing the polar coordinates (v,θ,ϕ) with $v_z = v\cos\theta$, we obtain the current densities $J_{1,2}(\omega,z)$ in the layers as

$$
\begin{aligned}
J_1(\omega,z) &= -2e\left[\frac{m_1}{h}\right]^3 \int\int\int v_x f_1\, dv_x\, dv_y\, dv_z \\
&= \frac{2\pi(em_1 v_F)^2}{h^3}\int_1^\infty ds\left[\frac{1}{s}-\frac{1}{s^3}\right]\left[\frac{(p_1+e^{2sw_1(z+d)/l_1})}{W_y(s)}\right. \\
&\quad \times\left[p_2T_i^2+R_i(e^{2sw_2h/l_2}-p_2R_i)\right]\int_{-d}^{0} dt\,E(t)e^{sw_1(t-z)/l_1} \\
&\quad +T_i e^{-sw_1z/l_1}\int_0^h dt\,E(t)e^{-sw_2(t-2h)/l_2} \\
&\quad \left.+p_2T_i e^{-sw_1z/l_1}\int_0^h dt\,E(t)e^{sw_2t/l_2}\right] \\
&\quad +\frac{p_1}{W_y(s)}\left[(e^{2sw_2h/l_2}-p_2R_i)(1+R_i^{2sw_1z/l_1})+p_2T_i^2e^{2sw_1z/l_1}\right] \\
&\quad \times\int_{-d}^{0} dt\,E(t)e^{-sw_1(t+z)/l_1}+\int_{-d}^{z} dt\,E(t)e^{sw_1(t-z)/l_1} \\
&\quad \left.+\int_z^0 dt\,E(t)e^{-sw_1(t-z)/l_1}\right]
\end{aligned}
\tag{8}
$$

and

$$
\begin{aligned}
J_2(\omega,z) &= -2e\left[\frac{m_2}{h}\right]^3 \int\int\int v_x f_2\, dv_x\, dv_y\, dv_z \\
&= \frac{2\pi(em_2 v_F)^2}{h^3}\int_1^\infty ds\left[\frac{1}{s}-\frac{1}{s^3}\right]\left[\frac{(p_2+e^{2sw_2(h-z)/l_2})e^{sw_2z/l_2}}{W_y(s)}\right. \\
&\quad \times\left[T_i\int_{-d}^{0} dt\,E(t)e^{sw_1(t+2d)/l_1}+p_1T_i\int_{-d}^{0} dt\,E(t)e^{-sw_1t/l_1}\right. \\
&\quad \left.+e^{sw_2z/l_2}[p_1T_i^2+R_i(e^{2sw_1d/l_1}-p_1R_i)]\int_0^h dt\,E(t)e^{-sw_2(t+z)/l_2}\right] \\
&\quad +p_2\frac{(e^{2sw_1d/l_1}-p_1R_i)(1+R_ie^{-2sw_2z/l_2})+p_1T_i^2e^{-2sw_2z/l_2}}{W_y(s)}\int_0^h dt\,E(t)e^{sw_2(t+z)/l_2} \\
&\quad \left.+\int_0^z dt\,E(t)e^{sw_2(t-z)/l_2}+\int_z^h dt\,E(t)e^{-sw_2(t-z)/l_2}\right],
\end{aligned}
\tag{9}
$$

$$
W_y(s)=(e^{2sw_1d/l_1}-p_1R_i)(e^{2sw_2h/l_2}-p_2R_i)-p_1p_2T_i^2\,,
$$

where v_F is the Fermi velocity and $l_{1,2}$ are the mean free paths of electrons. It is convenient to introduce dimensionless coordinates in the layers $z=z/l_1$, $t=t/l_1$ for $-d\le z\le 0$ and $z=z/l_2$, $t=t/l_2$ for $0\le z\le h$ and to use the reduced values of layer thicknesses $d=d/l_1$ and $h=h/l_2$. The wave equation for the electric field $E_1(z)$ and $E_2(z)$ within the layers takes the form

$$\frac{d^2E_{1,2}(z)}{dz^2}+\frac{\omega^2 l_{1,2}^2}{c^2}\mu_{1,2}(1+S_{1,2})E_{1,2}(z)=\frac{i\omega l_{1,2}^2}{c^2\epsilon_0}\mu_{1,2}J_{1,2}(\omega,z)\ . \quad (10)$$

Generally, this equation cannot be solved in compact form. However, the approximate solution may be found by use of the Dingle[5] method. In order to obtain the electric field, we first consider the possible field contribution in the external layer as

$$E_1^{(1)}(z)=e^{-u_1w_1(z+d)}\ , \quad (11)$$

which when substituted into Eqs. (8) and (10) and after the integration over t leads to

$$(u_1^2-\eta_1)e^{-u_1w_1(z+d)}=\xi_1[K(u_1)e^{-u_1w_1(z+d)}+\Lambda_1]\ , \quad (12)$$

where

$$\eta_1=-\left[\frac{\omega l_1}{cw_1}\right]^2(S_1+1)\mu_1\ ,$$

$$K(u_1)=\int_1^\infty ds\left[\frac{1}{s}-\frac{1}{s^3}\right]\left[\frac{1}{s-u_1}+\frac{1}{s+u_1}\right]\ ,$$

$$\Lambda_1=\int_1^\infty ds\left[\frac{1}{s}-\frac{1}{s^3}\right]\left[\frac{(p_1+e^{2sw_1(d+z)})}{W_y(s)}\left[[p_2T_i^2+R_i(e^{2sw_2h}-p_2R_i)](e^{(s-u_1)w_1d}-1)\frac{e^{-sw_1(z+d)}}{s-u_1}\right.\right.$$
$$+T_i\frac{e^{-sw_1z}e^{-u_1w_1d}}{u_1+sa}(e^{2sw_2h}-e^{-h(u_1w_1-w_2s)})$$
$$\left.+p_2T_i\frac{e^{-sw_1z}e^{-u_1w_1d}}{sa-u_1}(e^{h(sw_2-u_1w_1)}-1)\right]$$
$$+p_1\frac{(e^{2sw_2h}-p_2R_i)(1+R_ie^{2sw_1z})+p_2T_i^2e^{2sw_1z}}{(u_1+s)W_y(s)}(e^{2sw_1d}-e^{(s-u_1)w_1d})e^{-sw_1(d+z)}$$
$$\left.-\frac{e^{-sw_1(z+d)}}{s-u_1}-\frac{e^{sw_1z}e^{-u_1w_1d}}{s+u_1}\right]\ , \quad (13)$$

$$a=\frac{w_2l_1m_2}{w_1l_2m_1}\ ,$$

$$\xi_1=i\frac{\omega}{c^2\epsilon_0}\frac{2\pi(em_1v_F)^2}{h^3w_1^3}l_1^3\mu_1=i\frac{3}{2}\frac{l_1^2}{w_1^3\delta_1^2}\ ,$$

$\delta_1=[2\epsilon_0c^2/\mu_1\omega\sigma_1(0)]^{1/2}$ is the classical penetration depth, and $\sigma_1(0)$ is the dc conductivity of bulk metal 1. The closest agreement between the left-hand and right-hand sides of the relation (12) may be attained by choosing u_1 such that

$$u_1=\xi_1K(u_1)+\eta_1\ . \quad (14)$$

Thus the contribution E_1 [Eq. (11)] will leave us with uncompensated terms $\xi_1\Lambda_1$. The next possible contribution to the electric field may be taken in the following form:

$$E_1^{(2)}(z)=\frac{\xi_1\Lambda_1}{s^2-\eta_1}\ . \quad (15)$$

Now, we have for the electric field $E_1=E_1^{(1)}+E_1^{(2)}$ from Eqs. (8) and (10) that

$$(u_1^2-\eta_1)e^{-u_1w_1(z+d)}=\xi_1K(u_1)e^{-u_1w_1(z+d)}+O(\xi_1^2)\ . \quad (16)$$

It is obvious that the two contributions applied here leave only terms of order ξ_1^2 so the process can be repeated to form a series in ξ_1 which converges for $|\xi_1|<1$. The solution is of the form

$$E_1^+(z)=E_1^{(1)}(z)+E_1^{(2)}(z)+O(\xi_1^2)\ . \quad (17)$$

Since $K(u_1)$ is an even function of u_1, $-u_1$ is also a root of the transcendental Eq. (14). Thus there is a further solution, $E_1^-(z)$, obtained from (17) simply by replacing u_1 by $-u_1$. Thus the electric field within the layer of metal 1 is a linear combination of these two solutions:

$$E_1(z)=A_1E_1^+(z)+B_1E_1^-(z)\ , \tag{18}$$

where A_1 and B_1 are constants.

The method of solving Eqs. (9) and (10), i.e., the wave equation for the layer of metal 2, follows essentially the same lines as those described above. The general solution may be written in the form

$$E_2(z)=A_2E_2^+(z)+B_2E_2^-(z)\ , \tag{19}$$

where

$$E_2^+(z)=E_2^{(1)}(z)+E_2^{(2)}(z)+O(\xi_2^2)\ , \tag{20}$$

$$E_2^{(1)}(z)=e^{-u_2w_2z}\ , \tag{21a}$$

$$E_2^{(2)}(z)=\frac{\xi_2\Lambda_2}{s^2-\eta_2}\ , \tag{21b}$$

$$u_2^2=\xi_2K(u_2)+\eta_2\ , \tag{22}$$

$$\Lambda_2=\int_1^\infty ds\left[\frac{1}{s}-\frac{1}{s^3}\right]\left[\frac{(p_2+e^{2sw_2(h-z)})e^{sw_2z}}{W_y(s)}\left[[p_1T_i^2+R_i(e^{2sw_1d}-p_1R_i)]\frac{(1-e^{-(s+u_2)w_2h})}{s+u_2}\right.\right.$$
$$\left.+T_i\frac{ae^{2sw_1d}}{s-au_2}(1-e^{-d(sw_1-u_2w_2)})+p_1T_ia\frac{(e^{d(u_2w_2+w_1s)}-1)}{s+au_2}\right]$$
$$+p_2\frac{(e^{2sw_1d}-p_1R_i)(1+R_ie^{-2sw_2z})+p_1T_i^2e^{-2sw_2z}}{(s-u_2)W_y(s)}(e^{(s-u_2)w_2h}-1)e^{sw_2z}$$
$$\left.-\frac{e^{-sw_2z}}{s-u_2}-\frac{e^{-(s+u_2)w_2h}e^{sw_2z}}{s+u_2}\right]\ , \tag{23}$$

$$\eta_2=-\left[\frac{\omega l_2}{cw_2}\right]^2(S_2+1)\mu_2,\quad \xi_2=l\frac{3}{2}\frac{l_2^2}{w_2^3\delta_2^2}\ ,$$

and $E_2^-(z)$ is found by using $-u_2$ instead of u_2 in $E_2^+(z)$.

III. OPTICAL PROPERTIES

It is evident from the preceding section that the validity of the presented considerations is determined by the assumption $|\xi_{1,2}|<1$. We will extend this assumption taking $|\xi_{1,2}|\ll 1$. Analyzing the dependence of $\xi_{1,2}$ on ω we find that this restricts the validity of our formulas to two spectra ranges: low frequencies where the skin effect is nearly classical and high frequencies from the near infrared (NIR) and visible (VIS) up to the ultraviolet (uv).[5,10] In the above frequency ranges an inequality $|\eta_{1,2}|\ll 1$ is also fulfilled. Consequently, we have $|u_{1,2}|\ll 1$. Thus we may take for the external layer with a sufficient accuracy that

$$E_1(z)=A_1e^{-u_1w_1(z+d)}+B_1e^{u_1w_1(z+d)}\ . \tag{24}$$

Also, we can approximate $E_1'(z)$ by the expression

$$\frac{ic}{\omega l_1\mu_1}E_1'(z)=\frac{ic}{\omega l_1\mu_1}[A_1E_1^+(z)+B_1E_1^-(z)]'=A_1n_1(z)e^{-u_1w_1(z+d)}-B_1n_1'(z)e^{u_1w_1(z+d)}\ , \tag{25}$$

where

$$n_1(z)=\frac{n_{1b}}{\mu_1}-F_1(u_1,z)\ , \tag{26a}$$

$$n_1'(z)=\frac{n_{1b}}{\mu_1}+F_1(-u_1,z)\ , \tag{26b}$$

$$F_1(u_1,z)=\frac{3}{4}\left[\frac{l_1\omega_{p_1}}{w_1v_F}\right]^2\frac{v_F}{c}\int_1^\infty ds\left[\frac{1}{s^3}-\frac{1}{s^5}\right]$$
$$\times\left[\frac{(e^{sw_1(z+2d)}-p_1e^{-sw_1z})}{W_y(s)}\left[[p_2T_i^2+R_i(e^{2sw_2h}-p_2R_i)]\times(e^{-u_1w_1d}-e^{-sw_1d})+\frac{T_i}{a}e^{-u_1w_1d}\right.\right.$$
$$\left.\left.\times(e^{2sw_2h}-e^{-h(u_1w_1-sw_2)})+p_2\frac{T_i}{a}e^{-u_1w_1d}(e^{h(sw_2-u_1w_1)}-1)\right]\right.$$

$$+p_1\frac{(e^{2sw_2h}-p_2R_i)(R_ie^{sw_1z}-e^{-sw_1z})+p_2T_i^2e^{sw_1z}}{W_y(s)}(e^{sw_1d}-e^{-u_1w_1d})$$
$$-(e^{sw_1z}e^{-u_1w_1d}-e^{-sw_1(z+d)})\Bigg]e^{u_1w_1(z+d)}. \tag{27}$$

The quantity n_{1b}, i.e., the complex refractive index of the bulk metal, is introduced by means of the following relation:

$$u_1w_1=\frac{i\omega l_1}{c}\left[\left[(1+S_1)-i\frac{\omega_{p_1}^2\tau_1}{\omega w_1}\right]\mu_1\right]^{1/2}=\frac{i\omega l_1}{c}n_{1b}\,, \tag{28}$$

which is obtained from Eq. (16) taking into account that $K(u_1)\simeq\frac{4}{3}$ for the neglected surfaces contribution [Eq. (13)]. ω_{p_1} is the plasma frequency.

The considerations for the layer of metal 2, similar to those carried out above, yield the optical constants $n_2(z)$ and $n_2'(z)$ as follows:

$$n_2(z)=\frac{n_{2b}}{\mu_2}-F_2(u_2,z)\,, \tag{29a}$$

$$n_2'(z)=\frac{n_{2b}}{\mu_2}+F_2(-u_2,z)\,, \tag{29b}$$

$$F_2(u_2,z)=\frac{3}{4}\left[\frac{l_2\omega_{p_2}}{w_2v_F}\right]^2\frac{v_F}{c}\int_1^\infty ds\left[\frac{1}{s^3}-\frac{1}{s^5}\right]$$
$$\times\Bigg[\frac{(p_2e^{sw_2z}-e^{sw_2(2h-z)})}{W_y(s)}\{[p_1T_i^2+R_i(e^{2sw_1d}-p_1R_i)](1-e^{(s-u_2)w_2h})$$
$$+T_ia(e^{2sw_1d}-e^{(u_2w_2+sw_1)d})+p_1aT_i(e^{(u_2w_2+sw_1)d}-1)\}$$
$$+p_2\frac{(e^{2sw_1d}-p_1R_i)(e^{sw_2z}-R_ie^{-sw_2z})-p_1T_i^2e^{-sw_2z}}{W_y(s)}$$
$$\times(e^{(s-u_2)w_2h}-1)-(e^{sw_2z}e^{-(s+u_2)w_2h}-e^{-sw_2z})\Bigg]e^{u_2w_2z}. \tag{30}$$

The definition of n_{2b} is analogous to that of n_{1b}. The expressions for the reflected (r) and transmitted (t) amplitudes of the electric field may be obtained using the interference matrix $\underline{M}$ defined as[22,23]

$$\begin{bmatrix}E_1(-d)\\H_1(-d)\end{bmatrix}=\underline{M}\begin{bmatrix}E_2(h)\\H_2(h)\end{bmatrix},$$

with

$$\underline{M}=\begin{bmatrix}m_{11}&m_{12}\\m_{21}&m_{22}\end{bmatrix}=\underline{M}_1\underline{M}_2=\begin{bmatrix}m_{11}^1&m_{12}^1\\m_{21}^1&m_{22}^1\end{bmatrix}\begin{bmatrix}m_{11}^2&m_{12}^2\\m_{21}^2&m_{22}^2\end{bmatrix}$$

and

$$r=\frac{(m_{11}+m_{12}n_3)n_0-(m_{21}+m_{22}n_3)}{m_{11}+m_{12}n_3)n_0+(m_{21}+m_{22}n_3)}\,,$$
$$t=\frac{2n_0}{(m_{11}+m_{21}n_3)n_0+(m_{21}+m_{22}n_3)}\,. \tag{31}$$

The elements of matrices $\underline{M}_1$ and $\underline{M}_2$ can be found from the boundary conditions[23] describing the fact that the tangential components of the electromagnetic field must be equal across the surfaces and across the interface. It is then calculated that these elements are

$$m_{11}^1=\frac{(n_1'(-d)e^{u_1w_1d}+n_1(-d)e^{-u_1w_1d}}{n_1'(-d)+n_1(-d)}\,,$$
$$m_{12}^1=\frac{e^{u_1w_1d}-e^{-u_1w_1d}}{n_1'(-d)+n_1(-d)}\,,$$
$$m_{21}^1=\frac{n_1(0)n_1'(-d)e^{u_1w_1d}-n_1'(0)n_1(-d)e^{-u_1w_1d}}{n_1'(-d)+n_1(-d)}\,,$$
$$m_{22}^1=\frac{n_1'(0)e^{-u_1w_1d}+n_1(0)e^{u_1w_1d}}{n_1'(-d)+n_1(-d)}\,, \tag{32}$$
$$m_{11}^2=\frac{n_2'(h)e^{u_2w_2h}+n_2(h)e^{-u_2w_2h}}{n_2'(h)+n_2(h)}\,,$$

$$m_{12}^2=\frac{e^{u_2w_2h}-e^{-u_2w_2h}}{n_2'(h)+n_2(h)},$$

$$m_{21}^1=\frac{n_2(0)n_2'(h)e^{u_2w_2h}-n_2'(0)n_2(h)e^{-u_2w_2h}}{n_2'(h)+n_2(h)},$$

$$m_{22}^2=\frac{n_2'(0)e^{-u_2w_2h}+n_2(0)e^{u_2w_2h}}{n_2'(h)+n_2(h)}.$$

The intensity coefficients R and T for reflection and transmission of the double-layer film on a transparent substrate are given by

$$R=R'+T'^2\frac{R_{30}}{1-R_{30}R''},$$
$$T=\frac{(1-R_{30})T'}{1-R_{30}R''}, \tag{33}$$

where $T'=(n_3/n_0)|t|^2$, $R'=|r|^2$, $R_{30}=[(n_3-n_0)/(n_3+n_0)]^2$, and R'' is the reflection coefficient of the film for the electromagnetic wave incident through the semi-infinite substrate. In Eqs. (33) for R and T the term $R_{30}R''$ is much less than unity so we can neglect surface contributions to the skin effect in the calculations of R''.

IV. DISCUSSION

The relations obtained in this paper for the reflection and transmission coefficients of the double-layer film depend on both the layers thicknesses d and h and the scattering parameters p_1, p_2, R_i, and T_i. Moreover, it is apparent from Eqs. (26), (27), (29), and (30) that there exists a mutual influence of the transport parameters characterizing one of the metallic layers on the optical constants of the other one. This results from both the feasibility of coherent passages of conduction electrons across the interface (for $T_i>0$) and the nonlocal character of the relationship between the electric field and the surface current in the wave equation. The above-mentioned facts reveal the complexity of the problem and indicate the necessity for numerical evaluations for further discussion.

In order to see the effects of surface and interface scattering, the reflection R, transmission T, and absorption $A=1-R-T$ are calculated for a hypothetical double-layer system with a base layer of Ag (metal 2) and an overlayer of Au (metal 1). It is assumed that $l_1=30$ nm, $l_2=53$ nm, $m_1=m_e$, $m_2=0.85m_e$, $S_1=7$, $S_2=2.55$, and the electron concentrations $N_1=N_2=5.9\times10^{22}$ cm^{-3}.[14] Values $n_0=1$ and $n_3=1.5$ are taken as the optical constants of the medium and the substrate, respectively. The theoretical results are presented in the standard form versus energy $\hbar\omega$.

The optical reflection and transmission of the double-layer films are illustrated in Fig. 2. The plots demonstrate both the limiting cases in which the surfaces do not scatter the electrons diffusely ($p_1=p_2=1$) and the surface scattering is exclusively diffuse ($p_1=p_2=0$) as well as limiting cases for the interface scattering, i.e., exclusively diffuse ($R_i=T_i=0$) and the coherent transmission of the carriers across the interface ($R_i=0$, $T_i=1$). These curves

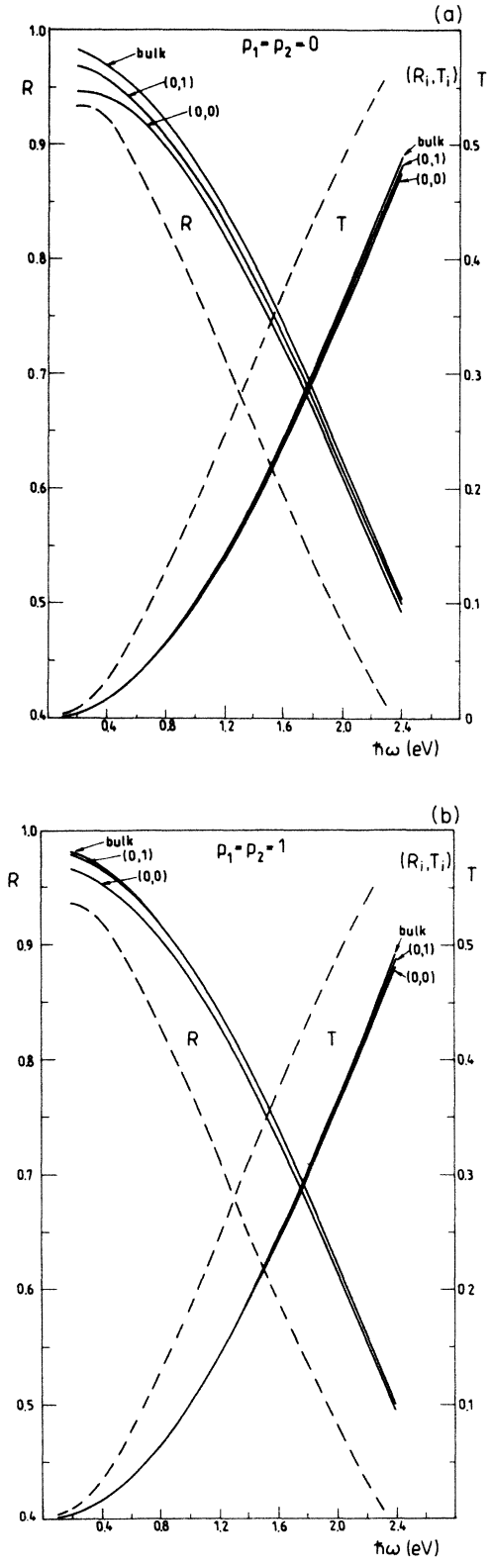

FIG. 2. Reflection and transmission spectra of a hypothetical double-layer film with layer thicknesses $d=5$ nm (Au) and $h=10$ nm (Ag) (solid curves) compared with the spectra of a single layer with thickness $h=10$ nm (Ag) (dashed curves) for (a) the diffuse and (b) the specular scattering of electrons at the surfaces.

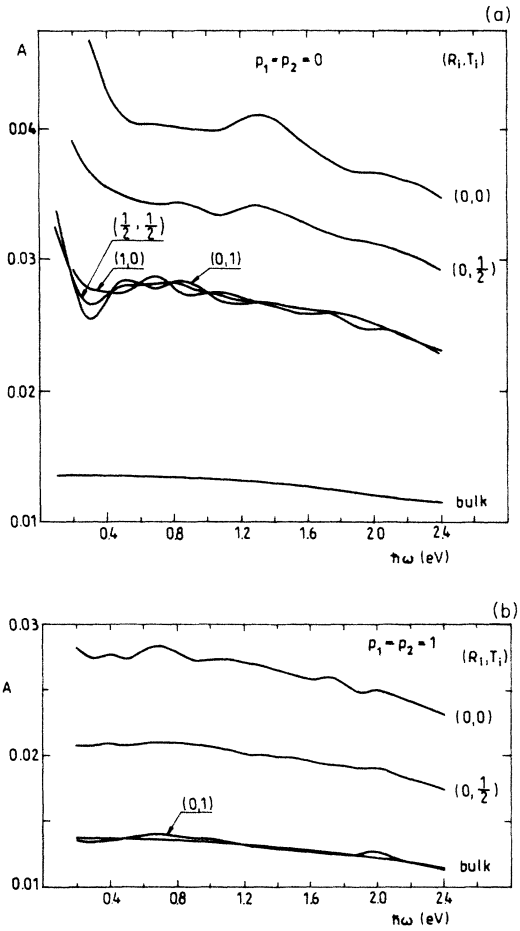

FIG. 3. Absorption spectrum of a hypothetical double-layer film (Au: $d=5$ nm, Ag: $h=10$ nm) for (a) the diffuse and (b) the specular scattering of electrons at the surfaces.

are compared with those for the double-layer films of Ag and Au with the neglected surface and interface contributions (labeled bulk) and with the curves for the single layer of Ag. The results of the absorption for the analogous films are illustrated in Fig. 3, where additionally the cases of the partially coherent transmission of the electrons across the interface have been included.

The theoretical plots show that the influence of the parameters p_1, p_2, R_i, and T_i on the reflection spectrum is relatively large in the low-energy region and slightly decreases with the increasing energy of the electromagnetic wave. As one can expect, the energy dependence of transmission is reversed. However, for both the spectra this influence remains significant in the NIR, VIS, and uv ranges. The changes of the absorption spectrum evoked by the surface and interface scattering are more essential when compared to the bulk. The additional contributions may enhance the absorption calculated for our hypothetical example by about 2.7%. It should be stressed here that in general the values of the described effects depend on d and h, and increase with decreasing thicknesses.

The curves in Fig. 3 also demonstrate absorption oscillations. They were predicted by Dingle[5] for single films. The oscillations appear because $w_{1,2}$ are complex quantities and, consequently, the integrals derived in our model are periodic functions of $\omega\tau_1 d$ and $\omega\tau_2 h$. One can see that their amplitudes depend on the efficiency of the surface and interface scattering where the most distinct oscillations of A appear for low values of the parameters p_1, p_2, R_i, and T_i. It is obvious that such effects should also be revealed in the reflection and transmission spectra, but on the other hand they are more pronounced in the absorption.

The obtained oscillations result from the nature of the electron movement in the film in the field of electromagnetic wave. On its way between the film surfaces the electron collects the energy from the field and loses this energy, being scattered diffusely at the surfaces and interfaces. The maximum of dissipated energy is attained when the electron approaching the surface has the maximal energy, i.e., duration, of its movement between the scattering surfaces is equal to the period of the electric field or a multiplicity of this period. On the other hand, the process of the energy dissipation should be most efficient for the electrons moving perpendicularly to the plane of the film. Therefore, the periods of the oscillations depend on the distances between the surfaces and interface which scatter the current carriers diffusely or partly diffusely. The described influence of the scattering at the particular surfaces on the oscillations of A is illustrated in Fig. 4, where two fragments of the plots from Fig. 3 are enlarged.

Finally let us discuss the optical properties of metallic samples which consist of a thin covering and a thick base. In order to obtain the reflection spectrum of such a system, the expressions for the refractive indices given by Eqs. (26), (27), (29), and (30) are calculated in the limit $h\rightarrow\infty$. The explicit relations for the optical constants are

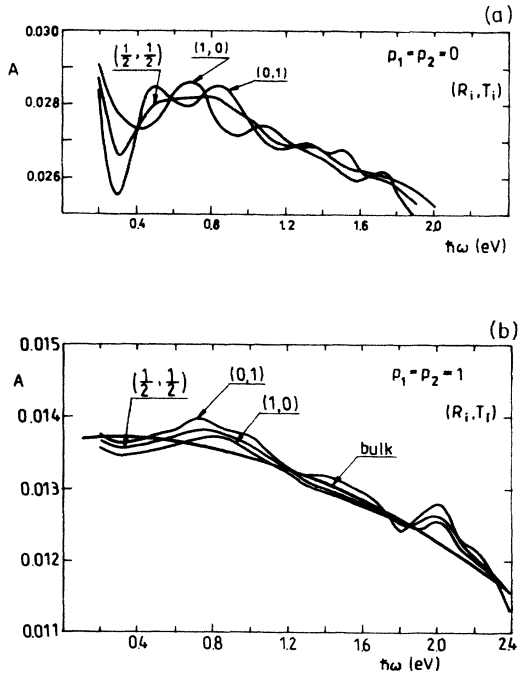

FIG. 4. Influence of the parameters of the interface scattering on the oscillations in the absorptions spectrum for (a) the diffuse and (b) the specular surface scattering of electrons.

$$n_1(-d)=n_{1b}+\frac{3}{4}\left[\frac{l_1\omega_{p_1}}{w_1 v_F}\right]^2\frac{v_F}{c}\int_1^\infty ds\left[\frac{1}{s^3}-\frac{1}{s^5}\right]\frac{(1-p_1)}{(1-p_1R_ie^{-2sw_1d})}$$

$$\times\left[R_ie^{-2sw_1d}+\left[1-R_i-T_i\frac{1}{a}\right]e^{-sw_1d}e^{-u_1w_1d}-1\right],$$

$$n_1(0)=n_{1b}+\frac{3}{4}\left[\frac{l_1\omega_{p_1}}{w_1 v_F}\right]^2\frac{v_F}{c}\int_1^\infty ds\left[\frac{1}{s^3}-\frac{1}{s^5}\right]\frac{1}{(1-p_1R_ie^{-2sw_1d})}\left[\left[1-R_i-T_i\frac{1}{a}\right]-p_1\left[1-R_i-T_i\frac{1}{a}\right]e^{-2sw_1d}\right.$$

$$\left.-(1-p_1)(1-R_i)e^{-sw_1d}e^{u_1w_1d}\right],$$

$$n_2(0)=n_{2b}-\frac{3}{4}\left[\frac{\omega_{p_2}l_2}{w_2 v_F}\right]^2\frac{v_F}{c}\left[\int_1^\infty ds\left[\frac{1}{s^3}-\frac{1}{s^5}\right]\frac{1}{(1-p_1R_ie^{-2sw_1d})}[aT_i(1-p_1)e^{-sw_1d}e^{u_2w_2d}\right.$$

$$\left.+p_1T_i(a-T_i)e^{-2sw_1d}-aT_i]+\tfrac{1}{4}(1-R_i)\right].$$

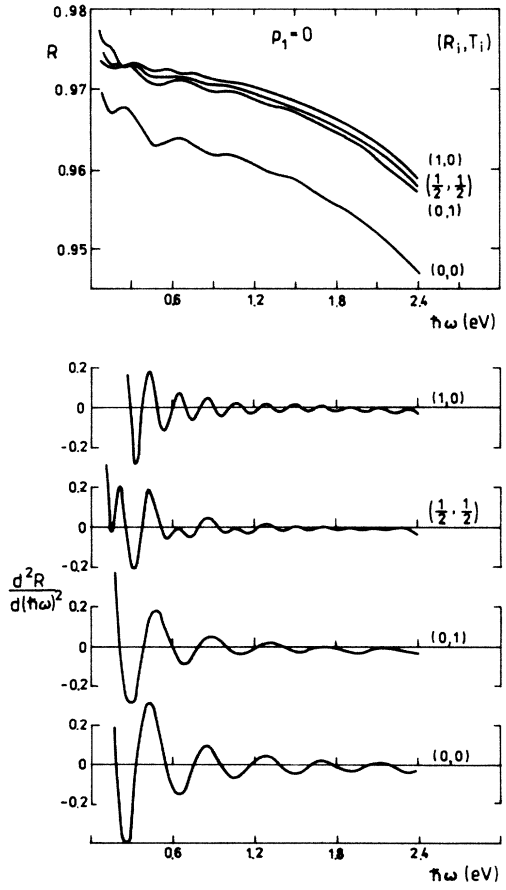

FIG. 5. Plots of the reflection coefficient and the second derivative $d^2R/d(\hbar\omega)^2$ for a hypothetical semi-infinite base with the covering [thicknesses: $d=16$ nm (Ag), $h=\infty$ (Au)].

In this case only the contributions of the external covering surface and of the interface between the two metals are significant. The theoretical results obtained for the Au base and the Ag covering are demonstrated in Fig. 5. One can see the influence of the surface and interface scattering on both the value of the reflection coefficient and the appearance of the oscillations. The second derivative of R reveals differences in periods of these oscillations for some cases where the previous elucidations are adequate. It is apparent that the oscillations may also be observed in the case of thick metallic samples on condition that two surface scattering electrons are operative.

V. CONCLUSION

It is worth noticing that the values of the scattering parameters may be determined from optical measurements. Furthermore, the detailed analysis of the oscillations should enable a distinction between the surface and interface scattering. It seems that a combined analysis of the optical and electrical data should be the most effective tool for investigating the microscopic parameters of free electrons in double-layer films. The first successful attempts of such studies were made for single films.[6,12–14] In the case of double-layer films, the theoretical models for dc conductivity (Ref. 15) and optical properties (the present paper) create the analogous feasibilities.

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