Optical properties of metallic multilayer films

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Optical properties of multilayer films consisting of alternating layers of two different metals are studied on the basis of the Maxwell equations and the Boltzmann transport theory. The influence of free-electron scattering at the film external surface and at the interfaces is taken into account and considered as a function of the electromagnetic-field frequency and the structure modulation wavelength. Derived formulas for optical coefficients are valid at low frequencies, where the skin effect is nearly classical, as well as in the near-infrared, visible, and ultraviolet spectral ranges, where the skin effect has an anomalous nature. It is shown that the obtained results are apparently dependent on the values of scattering parameters. Also, the oscillatory nature of analyzed spectra is observed, where the two oscillation periods may appear on certain conditions. The oscillations result from the electron surface and interface scattering and their amplitudes and periods depend on the boundary conditions for free-electron scattering. Finally, an application of the interference phenomenon in dielectric layers is proposed to enhance nondistinct features, which can appear in optical spectra of metallic films.

I. INTRODUCTION

Recently, the advance in epitaxial growth techniques has made the production of high-quality compositionally modulated structures or superlattices possible. These structures are studied intensively because of their physical properties. The interest is concentrated in the structural analysis, mechanical, magnetic, and transport properties,[1] and in investigations of the multilayer films for extended ultraviolet and x-ray optics.[2,3] Only a few works[4–6] have considered the optical properties of metallic compositionally modulated structures. The work by Dimmich[6] gave tentative information about the oscillatory nature of optical spectra of these samples.

An approach presented in this paper is based on the results derived for skin-effect theories given by Reuter and Sondheimer,[7] Dingle,[8] and Manz and co-workers[9,10] for the case of a semi-infinite sample, and their later developments obtained for single- and double-layer films by Hutchison and Hansen,[11] Dimmich and Warkusz,[12] Szczyrbowski, Schmalzbauer, and Hoffmann,[13,14] Dryzek and Dimmich,[15,16] and Dudek.[17,18]

It is the aim of this paper to formulate a theoretical description of the optical properties of films which consist of alternating layers of two different metals and have a constant modulation wavelength. A detailed set of expressions for optical coefficients of multilayer films is presented, which was derived for the case of normal electromagnetic wave incidence on a film.

II. WAVE EQUATION

We consider the metallic multilayer film which is presented in Fig. 1. A plane electromagnetic wave is incident from the z direction on a film, where the electric field $E(z)e^{i\omega t}$ is taken in the x direction and magnetic field $H(z)e^{i\omega t}$ in the y direction. The film consists of n repeating double-layer systems of thicknesses $d = b + a$, so the film thickness is $d_f = nd$, where we are interested in the case of $n \gg 1$. For such geometry Maxwell's equations reduce to the following wave equation:

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

where $J(\omega,z)$ is the density of free-electron current generated by the electric field $E(z)$ of the incident radiation, $1+S$ takes care of the displacement current, atomic polarization, etc. The current density may be obtained from the linearized Boltzmann transport equation for the electron distribution function $f(\mathbf{v},z)$ of the conduction electrons

$$\frac{\partial f^n_{1\,[2]}}{\partial z} + \frac{(1+i\omega\tau_{1\,[2]})f^n_{1\,[2]}}{\tau_{1\,[2]}v_z} = \frac{eE(z)}{m_{1\,[2]}v_z}\frac{\partial f_0}{\partial v_x}\ , \qquad (2)$$

where the notation with optional lower indices in the

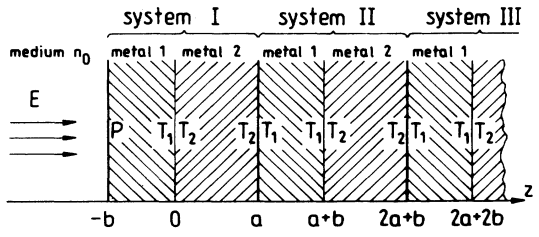

FIG. 1. Metallic multilayer film in the field of the electromagnetic wave.

square brackets is used, i.e., an equation or a quantity can be rewritten with the index 2 to describe the layer of metal 2. It is assumed that the distribution function is in the form $f_{1\,[2]}(\mathbf{v},z) = f_0 + f^n_{1\,[2]}(\mathbf{v},z)$, where f_0 is the Fermi-Dirac equilibrium distribution function, $f^n_1(\mathbf{v},z)$ and $f^n_2(\mathbf{v},z)$ are the deviations from this function induced by the electric field in layers of metal 1 and metal 2 in the nth layer system, respectively (see Fig. 1), $\mathbf{v} = (v_x, v_y, v_z)$ is the electron velocity, and $\tau_{1\,[2]}$ and $m_{1\,[2]}$ are the electron relaxation times and the effective masses. The general solution of Eq. (2) for the considered geometry has the form

$$f^{n+}_1(\mathbf{v},z) = e^{-A_1[z+b-(n-1)d]}\left(F^{n+}_1 + B_1\int_{-b+(n-1)d}^{z} E(t)e^{A_1[t+b-(n-1)d]}dt\right) , \tag{3}$$

$$f^{n-}_1(\mathbf{v},z) = e^{-A_1[z-(n-1)d]}\left(G^{n-}_1 - B_1\int_{z}^{(n-1)d} E(t)e^{A_1[t-(n-1)d]}dt\right) , \tag{4}$$

for $(n-1)d - b < z < (n-1)d$ and

$$f^{n+}_2(\mathbf{v},z) = e^{-A_2[z-(n-1)d]}\left(F^{n+}_2 + B_2\int_{(n-1)d}^{z} E(t)e^{A_2[t-(n-1)d]}dt\right) , \tag{5}$$

$$f^{n-}_2(\mathbf{v},z) = e^{-A_2[z+b-(n-1)d]}\left(G^{n-}_2 - B_2\int_{z}^{nd-b} E(t)e^{A_2(t+b-nd)}dt\right) , \tag{6}$$

for $(n-1)d < z < nd - b$ and where

$$A_{1\,[2]} = \frac{1 + i\omega\tau_{1\,[2]}}{v_z\tau_{1\,[2]}} ,$$

$$B_{1\,[2]} = \frac{e}{m_{1\,[2]}v_z}\frac{\partial f_0}{\partial v_x} ,$$

the functions $f^{n+}_{1\,[2]}$ and $f^{n-}_{1\,[2]}$ are $f^n_{1\,[2]}$ for $v_z > 0$ and $v_z < 0$, respectively. $F^n_{1\,[2]}$ and $G^n_{1\,[2]}$ are the arbitrary velocity functions determined by the appropriate boundary conditions at the film surface and interfaces. These conditions should serve as a description of the electron scattering processes. Therefore, we introduce the Fuchs-Sondheimer scattering parameters and we assume for the simplification of the boundary condition system that $n = \infty$. According to the previous works[6,19] we consider the following equations as the boundary condition system:

$$f^{\mathrm{I}+}_1(v_z, z=-b) = Pf^{\mathrm{I}-}_1(-v_z, z=-b) ,$$

$$f^{\mathrm{I}-}_1(v_z, z=0) = T_2 f^{\mathrm{I}-}_2(v_z, z=0) ,$$

$$f^{\mathrm{I}+}_2(v_z, z=0) = T_1 f^{\mathrm{I}+}_1(v_z, z=0) , \tag{7}$$

$$f^{\mathrm{I}-}_2(v_z, z=a) = T_1 f^{\mathrm{II}-}_1(v_z, z=a) ,$$

$$f^{\mathrm{II}+}_1(v_z, z=a) = T_2 f^{\mathrm{I}+}_2(v_z, z=a) , \ \ldots ,$$

where P is the specularity parameter describing the electron scattering at the film surface $z = -b$, and T_1 and T_2 are the probabilities of coherent passage of an electron across an interface from a layer of metal 1 (or from a layer of metal 2) to a layer of metal 2 (or to a layer of metal 1), and $0 \le P, T_1, T_2 \le 1$. The above description of the surface and interface scattering is quantitative only. There is no information about the nature of scattering mechanisms. Nevertheless, our approach can be developed in certain cases and the actual scattering mechanisms can be taken into account using boundary conditions, for example, derived by Soffer[20] for scattering from surface and interface roughnesses or derived by Greene and O'Donnell[21] for scattering from charge centers randomly located on surfaces or interfaces. It means, in fact, that the expressions appropriate for a considered scattering mechanism should be applied in the final formulas instead of the parameters used in Eqs. (7).[22,23]

Using the boundary conditions, Eqs. (7), for the electron distribution functions, Eqs. (3)–(6), and introducing the polar coordinates one may obtain the equation for the current density $J(\omega,z)$ dependence on the electric field $E(z)$ in the following form:

$$J^n_1(z) = \frac{2\pi e^2 m_1^2 v_{F1}^2}{h^3}\int_1^\infty ds\left(\frac{1}{s} - \frac{1}{s^3}\right)\left[e^{-sw_1(z_n+b)}\left(\frac{F^{n+}_1}{B_1} + \int_{(n-1)d-b}^{z} dt\, E(t)e^{sw_1[t+b-(n-1)d]}\right) - e^{sw_1 z_n}\right.$$
$$\left.\times\left(\frac{G^{n+}_1}{B_1} - \int_z^{(n-1)d} dt\, E(t)e^{-sw_1[t-(n-1)d]}\right)\right] \quad \text{for } -b \le z_n \le 0 , \tag{8}$$

$$J_2^n(z) = \frac{2\pi e^2 m_2^2 v_{F2}^2}{h^3} \int_1^\infty ds \left(\frac{1}{s} - \frac{1}{s^3}\right) \left[e^{-sw_2 z_n} \left(\frac{F_2^{n+}}{B_2} + \int_{(n-1)d}^{z} dt\, E(t) e^{sw_2[t-(n-1)d]} \right) - e^{sw_2(z_n-a)} \times \left(\frac{G_2^{n+}}{B_2} - \int_z^{(n-1)d+a} dt E(t) e^{-sw_2[t-a-(n-1)d]} \right) \right] \quad \text{for } 0 \le z_n \le a \ , \tag{9}$$

where $w_{1\,[2]} = (1 + i\omega\tau_{1\,[2]})/(v_{F1\,[F2]}\tau_{1\,[2]})$, $z_n = z-(n-1)d$, and v_F is the Fermi velocity of electrons. Consequently, having $J(\omega, z)$ as a function $E(z)$ it is apparent that the wave equation, Eq. (1), becomes the equation for the electric field $E_1^n(z)$ and $E_2^n(z)$ within particular layers. This equation cannot be solved in a compact form. However, according to the Dingle method[8] the approximate solution may be found. In order to obtain this solution (it means the electric field) we consider in the first step the possible field contribution in the form

$$E_1^{n(1)}(z) = e^{-u_1 w_1 (z_n + b)} \quad \text{for } -b \le z_n \le 0 \ , \tag{10}$$

$$E_2^{n(1)}(z) = e^{-u_2 w_2 z_n} \quad \text{for } 0 \le z \le a \ . \tag{11}$$

After the substitution of Eqs. (10) and (11) into Eqs. (8), (9) and, in turn, (1) and then, after the integration over t one may derive the following system of equations:

$$(u_1^2 - \eta_1) E_1^{n(1)}(z) = \xi_1 \left[K(u_1) E_1^{n(1)}(z) + \int_1^\infty ds \left(\frac{1}{s} - \frac{1}{s^3}\right) \Lambda_1^n \right] , \tag{12}$$

$$(u_2^2 - \eta_2) E_2^{n(1)}(z) = \xi_2 \left[K(u_2) E_2^{n(1)}(z) + \int_1^\infty ds \left(\frac{1}{s} - \frac{1}{s^3}\right) \Lambda_2^n \right] , \tag{13}$$

where the dimensionless coordinates are introduced, i.e., $z_n = z_n/l_1$, $b = b/l_1$ for $-b < z_n < 0$ and $z_n = z_n/l_2$, $a = a/l_2$ for $0 < z_n < a$, l_1 and l_2 are the mean free paths of the electrons in metal 1 and metal 2, and where

$$\xi_{1\,[2]} = \frac{i\omega\mu_{1\,[2]}}{c^2\epsilon_0} \frac{2\pi(em_{1\,[2]}v_{F1\,[F2]})^2 l_{1\,[2]}^3}{h^3 w_{1\,[2]}^3} \ ,$$

$$\eta_{1\,[2]} = -\left(\frac{\omega l_{1\,[2]}}{c w_{1\,[2]}}\right)^2 (1 + S_{1\,[2]})\mu_{1\,[2]} \ ,$$

$$K(u_{1\,[2]}) = \int_1^\infty ds \left(\frac{1}{s} - \frac{1}{s^3}\right) \left(\frac{1}{s - u_{1\,[2]}} + \frac{1}{s + u_{1\,[2]}} \right) .$$

Λ_1^n and Λ_2^n are the functions of P, T_1, T_2, w_1, w_2, u_1, u_2, a, b, and z. It is apparent that taking u_1 and u_2 as follows,

$$u_{1\,[2]} = \xi_{1\,[2]} K(u_{1\,[2]}) + \eta_{1\,[2]} \ , \tag{14}$$

we leave with uncompensated terms

$$\xi_{1\,[2]} \int_1^\infty ds \left(\frac{1}{s} - \frac{1}{s^3}\right) \Lambda_{1\,[2]}^n$$

in the wave equation. Thus, the next possible contribution to the electric field may be chosen in the form

$$E_1^{n(2)}(z) = \xi_1 \int_1^\infty ds \left(\frac{1}{s} - \frac{1}{s^3}\right) \frac{\Lambda_1^n}{s^2 - \eta_1} \quad \text{for } -b \le z_n \le 0 \ , \tag{15}$$

$$E_2^{n(2)}(z) = \xi_2 \int_1^\infty ds \left(\frac{1}{s} - \frac{1}{s^3}\right) \frac{\Lambda_2^n}{s^2 - \eta_2} \quad \text{for } 0 \le z_n \le a. \tag{16}$$

The two contributions $E_{1\,[2]}^{n(1)}$ and $E_{1\,[2]}^{n(2)}$ applied to the wave equation leave, in turn, terms of order ξ_1^2 and ξ_2^2 only. Therefore, the process can be repeated to form a series in ξ_1 for layers of metal 1 and a series in ξ_2 for layers of metal 2. The series converge for $|\xi_1| < 1$ and $|\xi_2| < 1$. The solution is the following:

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

in the layers of metal 1,

$$E_2^{n(+)}(z) = E_2^{n(1)}(z) + E_2^{n(2)}(z) + O(\xi_2^2) \tag{18}$$

in the layers of metal 2. Taking into account the fact that $K(u_1)$ and $K(u_2)$ are even functions of u_1 and u_2 one may notice that $-u_1$ and $-u_2$ are also roots of the transcendental Eqs. (14), and consequently, the next solution of wave equation $E_1^{n(-)}(z)$ and $E_2^{n(-)}(z)$ may be obtained from Eqs. (17) and (18) simply by replacing u_1 by $-u_1$ and u_2 by $-u_2$. Thus, the electric field is a linear combination of these two solutions which in both kinds of layers takes the form

$$E_1^n(z) = A_1^n E_1^{n(+)}(z) + B_1^n E_1^{n(-)}(z) \ , \tag{19}$$

$$E_2^n(z) = A_2^n E_2^{n(+)}(z) + B_2^n E_2^{n(-)}(z) \ , \tag{20}$$

where A_1, A_2, B_1, and B_2 are constants.

III. OPTICAL PROPERTIES

The validity of the presented considerations is determined by the assumption $|\xi_{1\,[2]}| < 1$. Analyzing the dependence of $\xi_{1\,[2]}$ on ω for metal films it is evident that generally, this condition is fulfilled. Moreover, in two spectral ranges (for low frequencies, where the skin effect is nearly classical, and for high frequencies, from the near infrared to the ultraviolet) the extended assumption $|\xi_{1\,[2]}| \ll 1$ may be used. Thus, in the above frequency ranges our approach, i.e., the approach where the first two contributions in the series for the electric field are taken into account, is substantiated. Consequently, we can find that inequalities $|\eta_{1\,[2]}| \ll 1$ and $|u_{1\,[2]}| \ll 1$ are also fulfilled. Therefore, with sufficient accuracy we may take that

$$E_1^n(z) = A_1^n e^{-u_1 w_1 (z_n + b)} + B_1^n e^{u_1 w_1 (z_n + b)} ,$$
$$\frac{ic}{\omega l_1 \mu_1} \frac{dE_1^n(z)}{dz} = \frac{ic}{\omega l_1 \mu_1} \frac{d[A_1^n E_1^{n(+)}(z) + B_1^n E_1^{n(-)}(z)]}{dz} = A_1^n n_1^n(z) e^{-u_1 w_1 (z_n + b)} - B_1^n n_1^{n\prime}(z) e^{u_1 w_1 (z_n + b)} , \tag{21}$$

and

$$E_2^n(z) = A_2^n e^{-u_2 w_2 z_n} + B_2^n e^{u_2 w_2 z_n} ,$$
$$\frac{ic}{\omega l_2 \mu_2} \frac{dE_2^n(z)}{dz} = \frac{ic}{\omega l_2 \mu_2} \frac{d[A_2^n E_2^{n(+)}(z) + B_2^n E_2^{n(-)}(z)]}{dz} = A_2^n n_2^n(z) e^{-u_2 w_2 z_n} - B_2^n n_2^{n\prime}(z) e^{u_2 w_2 z_n} , \tag{22}$$

where the refractive indices have the forms

$$n_1^n(z) = \frac{n_{1b}}{\mu_1} - F_1^n(u_1, u_2, z_n) , \quad n_1^{n\prime}(z) = \frac{n_{1b}}{\mu_1} + F_1^n(-u_1, -u_2, z_n) ,$$

$$\begin{aligned} F_1^n(u_1, u_2, z_n) = \frac{3 v_{F1}}{4c} \left(\frac{l_1 \omega_{p1}}{w_1 v_{F1}} \right)^2 \int_1^\infty ds \left(\frac{1}{s^3} - \frac{1}{s^5} \right) \frac{1}{1 - T_1 T_2 H_1 H_2} \\ \times [(1 - T_1 T_2 G_1 H_2 + C(T_1 H_1 - T_2 G_2) \\ - \{P(1 - H_1 G_1) + T_1 T_2 (H_1 H_2 - G_1 H_2) + C T_2 [P H_1 (1 - H_2 G_2) + H_2 - G_2]\} \\ \times (T_1 T_2 H_1 H_2)^{n-1}) e^{-s w_1 (z_n + b)} \\ + [T_1 T_2 H_2 - G_1 + C T_2 (1 - H_2 G_2)] e^{s w_1 z_n}] e^{u_1 w_1 (z_n + b)} , \end{aligned} \tag{23}$$

and

$$n_2^n(z) = \frac{n_{2b}}{\mu_2} - F_2^n(u_2, u_1, z_n) , \quad n_2^{n\prime}(z) = \frac{n_{2b}}{\mu_2} + F_2^n(-u_2, -u_1, z_n) ,$$

$$\begin{aligned} F_2^n(u_2, u_1, z_n) = \frac{3 v_{F2}}{4c} \left(\frac{l_2 \omega_{p2}}{w_2 v_{F2}} \right)^2 \int_1^\infty ds \left(\frac{1}{s^3} - \frac{1}{s^5} \right) \frac{1}{1 - T_1 T_2 H_1 H_2} \\ \times [(1 - T_1 T_2 H_1 G_2 + C^{-1} T_1 (H_1 G_1) \\ - \{T_1 T_2 H_1 [P H_1 (1 - H_2 G_2) + H_2 - G_2] + C^{-1} T_1 \\ \times [P H_1 (1 - H_1 G_1) + H_1 - G_1]\} (T_1 T_2 H_1 H_2)^{n-1}) e^{-s w_2 z_n} \\ + [T_1 T_2 H_1 - G_2 + C^{-1} T_1 (1 - H_1 G_1)] e^{s w_2 (z_n - a)}] e^{u_2 w_2 z_n} , \end{aligned} \tag{24}$$

where

$$H_1 = e^{-s w_1 b} , \quad H_2 = e^{-s w_2 a} , \quad G_1 = e^{-u_1 w_1 b} , \quad G_2 = e^{-u_2 w_2 a} , \quad C = \frac{m_1 v_{F1} w_1 l_2}{m_2 v_{F2} w_2 l_1} ,$$

ω_{p1} and ω_{p2} are the plasma frequencies for metal 1 and metal 2. The quantities n_{1b} and n_{2b}, i.e., the complex refractive indices of the bulk metal 1 and the bulk metal 2, respectively, are introduced by means of the following relations:

$$u_{1\,[2]} w_{1\,[2]} = \frac{i \omega l_{1\,[2]}}{c} \left[\left((1 + S_{1\,[2]}) - \frac{i \omega_{p1\,[p2]}^2 \tau_{1\,[2]}}{\omega w_{1\,[2]}} \right) \mu_{1\,[2]} \right]^{1/2} = \frac{i \omega l_{1\,[2]}}{c} n_{1b\,[2b]} , \tag{25}$$

which are obtained from Eqs. (12) and (13) with neglected terms of order $\xi_{1\,[2]}^2$ and higher, and where for bulk metals $K(u_{1\,[2]}) \simeq \frac{4}{3}$.

It seems to be reasonable to take into account the case that the considered metallic multilayer film, as a sample of finite thickness, may be partly transparent. Thus, the expressions for the reflected (r) and transmitted (t) amplitudes of the electric field may be obtained using the interference matrix method.[24,25] For n repeated double-layer systems in the film the interference matrix $\mathbf{M}$ is defined as

$$\begin{pmatrix} E(-b) \\ H(-b) \end{pmatrix} = \mathbf{M} \begin{pmatrix} E(nd-b) \\ H(nd-b) \end{pmatrix}$$

$$\text{with } \mathbf{M} = \begin{pmatrix} m_{11} & m_{12} \\ m_{21} & m_{22} \end{pmatrix} , \quad (26)$$

where $\mathbf{M} = M_1^{\mathrm{I}} M_2^{\mathrm{I}} M_1^{\mathrm{II}} M_2^{\mathrm{II}} \cdots M_1^n M_2^n$ is the product of the matrices for the particular layers. In this formalism the coefficients r and t are given by the following expressions:

$$r = \frac{(m_{11} + m_{12} n_s) n_0 - (m_{21} + m_{22} n_s)}{(m_{11} + m_{12} n_s) n_0 + (m_{21} + m_{22} n_s)} , \quad (27)$$

$$t = \frac{2 n_0}{(m_{11} + m_{12} n_s) n_0 + (m_{21} + m_{22} n_s)} , \quad (28)$$

where n_s is the substrate refractive index and the elements of matrices $M_{1\,[2]}^n$ may be found from boundary conditions[24,25] describing the fact that the tangential components of the electromagnetic field must be equal across the surface and across the interfaces. The elements of the matrix M_1^{I} for the external layer of metal 1 are given in the form

$$m_{11(1)}^{\mathrm{I}} = \frac{n_1^{\mathrm{I}}(0) e^{-u_1 w_1 b} + n_1^{\mathrm{I}\prime}(0) e^{u_1 w_1 b}}{n_1^{\mathrm{I}}(0) + n_1^{\mathrm{I}\prime}(0)} ,$$

$$m_{12(1)}^{\mathrm{I}} = \frac{e^{u_1 w_1 b} - e^{-u_1 w_1 b}}{n_1^{\mathrm{I}}(0) + n_1^{\mathrm{I}\prime}(0)} ,$$

$$m_{21(1)}^{\mathrm{I}} = \frac{n_1^{\mathrm{I}}(-b) n_1^{\mathrm{I}\prime}(0) e^{u_1 w_1 b} - n_1^{\mathrm{I}}(0) n_1^{\mathrm{I}\prime}(-b) e^{-u_1 w_1 b}}{n_1^{\mathrm{I}}(0) + n_1^{\mathrm{I}\prime}(0)} ,$$

$$m_{22(1)}^{\mathrm{I}} = \frac{n_1^{\mathrm{I}}(-b) e^{u_1 w_1 b} + n_1^{\mathrm{I}\prime}(-b) e^{-u_1 w_1 b}}{n_1^{\mathrm{I}}(0) + n_1^{\mathrm{I}\prime}(0)} ,$$

and the others can be calculated by analogy.

The reflection (R) and transmission (T) intensity coefficients of a multilayer film on a transparent substrate are as follows:

$$R = |r|^2 + |t|^2 \frac{n_s R_{s0}}{n_0 (1 - R_{s0} R')} , \quad (29)$$

$$T = |t|^2 \frac{n_s (1 - R_{s0})}{n_0 (1 - R_{s0} R')} , \quad (30)$$

where r and t are given by Eqs. (27) and (28), and the reflection coefficient of the medium-substrate interface

$$R_{s0} = \left(\frac{n_s - n_0}{n_s + n_0} \right)^2$$

and R' is the reflection coefficient of the multilayer film for the electromagnetic wave incident through the substrate. In calculations of R' we may neglect the surface contribution to the skin effect because in Eqs. (29) and (30) the term $R_{s0} R'$ is much less than unity.

IV. DISCUSSION

The metallic multilayer structure considered in this paper needs the approach taking into account the complexity of the problems which would reflect in the properties of optical reflectance and absorptance spectra. Such an approach was developed in the previous sections and the obtained formulas [Eqs. (23) and (24)] reveal that the optical coefficients depend on the field frequency, the modulation wavelength of the structure $d = b + a$, and scattering parameters P, T_1, T_2. Thus, the influence of the skin effect on the optical film coefficients is modified by the size effects. Moreover, the feasibility of coherent passages of conduction electrons across interfaces (for $T_1, T_2 > 0$) and the nonlocal character of the relationship between the electric field and the surface current in the wave equation lead to a mutual influence of transport parameters characterizing one of the metallic layers on the optical coefficients of the others.

In order to see the above-mentioned effects the reflection R and the absorption A are numerically evaluated for a hypothetical Nb-Cu system. Artificial Nb-Cu superstructures were the object of studies of several researchers.[26–31] It is assumed that $l_{\mathrm{Nb}} = 5.8$ nm, $l_{\mathrm{Cu}} = 42$ nm, $m_{\mathrm{Nb}} = m_{\mathrm{Cu}} = m_e, S_{\mathrm{Nb}} = S_{\mathrm{Cu}} = 0$, the electron concentrations are $N_{\mathrm{Nb}} = 5.6 \times 10^{22}$ cm^{-3}, $N_{\mathrm{Cu}} = 8.5 \times 10^{22}$ cm^{-3}. Values $n_0 = 1$ and $n_s = 1.5$ are taken as the optical constants of the medium and substrate, respectively. The theoretical results are presented in the standard form versus the electromagnetic field frequency.

The basis for all our calculations are the expressions for the refractive indices of the whole stack of layers given by Eqs. (23)–(25). However, the refractive index of the external surface of the multilayer structure is of prime importance for the optical spectra of semi-infinite samples. One can expect that the influence of the electron interaction with the external surface would change in a significant way the real part of $n_1^{\mathrm{I}}(-b)$. It is shown in Fig. 2. As we may see comparing the curves for $P = 1$ and for $P = 0$, the contribution of the electron surface scattering leads to an increase in the $\mathrm{Re}[n_1^{\mathrm{I}}(-b)]$ value. Moreover, the effect increases with decreasing value of the reduced thickness of the external layer ($b_{\mathrm{Cu}}/l_{\mathrm{Cu}} \ll b_{\mathrm{Nb}}/l_{\mathrm{Nb}}$). It means, in turn, that the surface scattering is more effective in the case of metal 1 being a good conductor, i.e., for the metals with longer electron mean free path. Generally, it is apparent that the surface contribution to the refractive index value may be of the same order as the refractive index value for an ideal surface. This multilayer film behavior is analogous to that found for the single- and double-layer structures.[8,11–16]

The optical reflection of the multilayer films is illustrated in Fig. 3. The plots demonstrate the reflection coefficient dependence on the modulation wavelength d for Nb-Cu systems with constant ratio a/b. It is shown

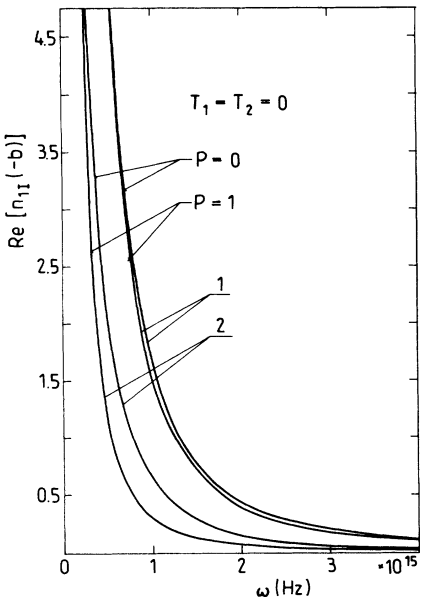

FIG. 2. Frequency dependence of the real part of the refractive index at the external film surface for hypothetical Nb-Cu (1, b_{Nb}=2 nm, a_{Cu}=2.3 nm) and Cu-Nb (2, b_{Cu}=2.3 nm, a_{Nb}=2 nm) bimetallic systems.

for effectively operating mechanisms of electron scattering at the surface and at the interfaces that the reflection decreases with decreasing modulation wavelength and the reflection oscillations appear for $b_{Nb} < l_{Nb}$ and $a_{Cu} < l_{Cu}$. These oscillations were predicted by Dingle[8] for single films. Their amplitudes depend on the efficiency of the surface and interface scattering, i.e., they

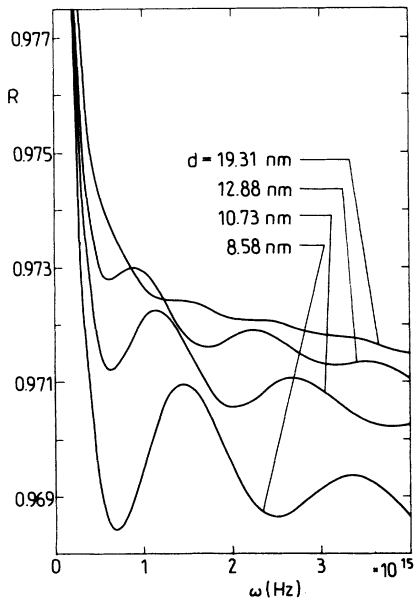

FIG. 3. Reflection spectra of a hypothetical multilayer film consisting of Nb-Cu bimetallic systems for the case of diffuse electron scattering at the surface and at the interfaces ($P = T_1 = T_2 = 0$). The Nb layer thicknesses are 9, 6, 5, and 4 nm for the curves from the top to the bottom, respectively.

are the most distinct for low values of the parameters P, T_1, T_2 and they increase with decreasing modulation wavelength. The obtained oscillations result from the nature of the electron movement in the film layers in the field of electromagnetic waves. Generally, they are conditioned by the surface and interface electron scattering and they disappear when those scattering mechanisms are not operating ($P = T_1 = T_2 = 1$). Our understanding of this phenomenon was given in detailed form in the previous work.[15]

According to the above ideas one may expect for a bimetallic structure that the oscillations would have two different periods corresponding to two different kinds of metallic layers. In the case of Fig. 3 the values of parameters (a/b) are chosen in such a way that oscillation periods for both structure components are the same. Nevertheless, the problem of influence of the transport parameters characterizing particular metallic layers on the optical absorption of the multilayer structure is presented in Figs. 4 and 5. In Fig. 4 the two cases in which the surface does not scatter the electrons diffusely ($P = 1$) and the surface scattering is exclusively diffuse ($P = 0$) are demonstrated for curves labeled as 1. The Cu layer thicknesses are taken much greater than the Nb ones, and, simultaneously thin enough to obtain the influence of the bimetallic systems farther from the external surface evident. The oscillation periods appearing due to the existence of two types of layers should differ several times. However, it is obvious that Cu, as a very good conductor, can dominate the oscillatory behavior of the whole film, especially when Cu is the external layer metal. It should be emphasized that for the considered structure the elimination of the external surface scattering ($P = 1$) does not change the oscillation period of the multilayer film though the amplitudes may change. It is contrary to the case of a single-layer film where for one of the surfaces

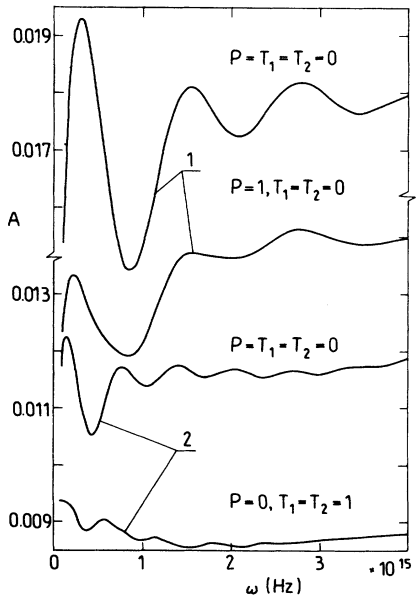

FIG. 4. Absorption spectra of a hypothetical multilayer film consisting of Cu-Nb bimetallic systems for 1, b_{Cu}=7.4 nm, a_{Nb}=1 nm; and 2, b_{Cu}=14.8 nm, a_{Nb}=2 nm.

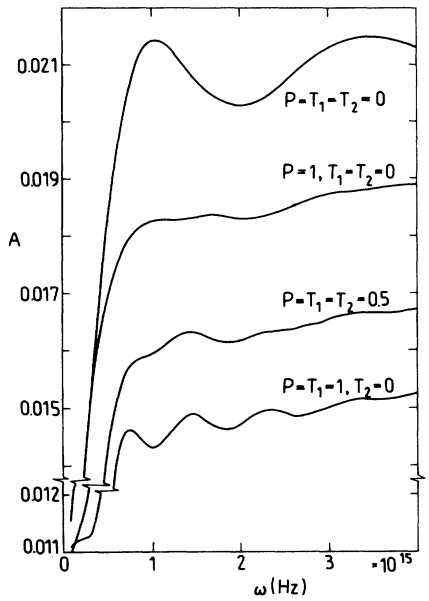

FIG. 5. Influence of the surface and interface scattering parameters on the oscillations in the absorption spectra of Nb-Cu multilayer films, where b_{Nb}=3 nm and a_{Cu}=11 nm.

assumed to reflect electrons specularly ($P = 1$) the oscillation period increases twice. Therefore, from our curves one may easily draw a conclusion that the influence of the whole stack of systems appears in the spectra.

The interpretation is not so obvious for the second choice of the parameters made for the curves labeled as 2. The thickness ratio a/b is the same as in the first case, but the modulation wavelength is much greater. The plots demonstrate the difference between the absorption spectrum with the interface scattering included ($T_1 = T_2 = 0$) and the spectrum for the case without the interface scattering contribution ($T_1 = T_2 = 1$). For the parameters $P = T_1 = T_2 = 0$ the oscillations are still distinct with the period determined by the Cu layers. It is two times shorter than for the curves labeled 1 because the modulation wavelength is two times longer. For $P = 0$ and $T_1 = T_2 = 1$, when the electrons can pass across the interfaces coherently, the mutual influence of the parameters characterizing particular layers on the transport parameters characterizing the others leads to the spectrum where the oscillation period created originally in the Cu layers is modified by the Nb layer transport parameters.

The curve behavior quite opposite to that described above is presented in Fig. 5. The optical absorption spectra of Nb-Cu multilayer structures show that for a certain choice of layer thicknesses one can observe the domination of the oscillations created in the Nb layers or the oscillations created in the Cu layers depending on the scattering parameter values only. Thus, in two limiting cases, for $P = T_1 = T_2 = 0$ (i.e., for the most efficient surface and interface electron scattering) and for $P = T_1 = 1$ and $T_2 = 0$ (i.e., when the interface scattering of electrons in the Cu layers is efficient) the two different oscillation periods are apparent. In the intermediate cases the oscillations are not distinct enough, i.e., the oscillations created in both kinds of layers damp mutually. Moreover, it is illustrated in Fig. 5 that the presence of surface and interface scattering of the electrons may enhance the absorption of the multilayer films significantly.

Concluding, one can say that the optical properties of the metallic multilayer films are determined by the transport parameters of metals being components of the alternating systems. It should be emphasized that the optical coefficient behavior results from the properties of the whole stack of repeated systems. The surface and interface scattering of electrons may lead in the case of multilayer structures to the oscillation effects which can appear in the optical spectra. The oscillation periods and amplitudes can be a source of valuable information about the transport parameters of the studied structures as well as about phenomena occurring at the interfaces and at the surface. The amplitude of these oscillations may be greater than that expected for single films. However, from the experimental point of view it seems that the oscillations are still too weak to be observable. Therefore, we would like to propose the application of the interference phenomena to enhance the above-mentioned oscillations. The idea and the results are presented in Fig. 6 for Cu-Nb systems. In the upper part of this figure the metallic multilayer film with a dielectric coating is shown. According to the approach developed in this paper we can obtain the modification resulting from the presence of the dielectric coverage by means of the inter-

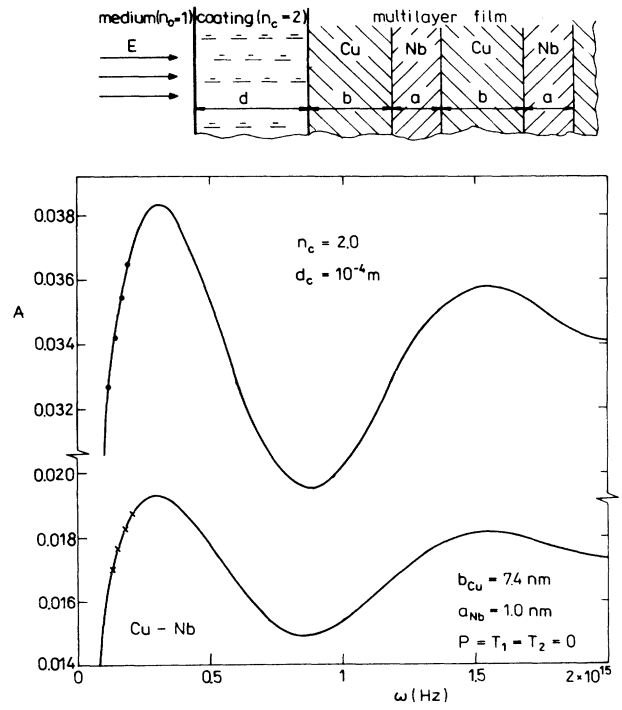

FIG. 6. Absorption spectrum of the uncovered Cu-Nb multilayer film (the bottom curve) compared with the envelopes for quickly varying with frequency absorption spectrum of the film with dielectric coverage (both the top and the bottom curves). Crosses and full circles show the minima and the maxima of the periodically varying absorption of the covered metallic structure.

ference matrix method. The interference matrix $\mathbf{M}_{d-m}$ for the metallic multilayer film with the dielectric coating having thickness d_c and refractive index n_c should be defined as

$$\mathbf{M}_{d-m} = M^c\,\mathbf{M}\ , \tag{31}$$

where the matrix M^c elements are given in the form

$$m^c_{11} = \cos\left(\frac{\omega}{c}n_c d_c\right)\ ,\quad m^c_{12} = \frac{i}{n_c}\sin\left(\frac{\omega}{c}n_c d_c\right)\ ,$$

$$m^c_{21} = i n_c \sin\left(\frac{\omega}{c}n_c d_c\right)\ ,\quad m^c_{22} = \cos\left(\frac{\omega}{c}n_c d_c\right)\ ,$$

and the method of calculation of the optical coefficients follows essentially the same lines as those described in the previous section [Eqs. (26)–(30)]. The obtained results are presented in Fig. 6 for the Cu-Nb absorption spectra. The absorption of the metallic film with the dielectric coverage varies periodically and very quickly with changing frequency. It exhibits a whole series of maxima and minima (marked in the left part of Fig. 6 as full circles and crosses, respectively). Thus, in Fig. 6 only the two curves are presented which are the envelopes for the interference oscillations of absorption of the structure under consideration. It should be emphasized that the bottom curve is also the absorption spectrum of an uncovered multilayer film whereas the top one (the second envelope) is the projection of that absorption spectrum of uncovered film with a significant enhancement. The enhancement depends on both coverage thickness and refractive index value. The changes of these parameters can change also the period of interference oscillations, so the choice needs some care about the measurement possibilities.

V. CONCLUSIONS

The optical properties of metallic multilayer films can be predicted theoretically, and the appropriate model is formulated in this paper. The influence of size effects and film structure (the modulation wavelength and the microscopic transport parameters of the layers) on the optical coefficients cannot be neglected. The presence of the surface and interface scattering of the electrons leads to the oscillation effects in the optical spectra of multilayer films. These effects are weak but, generally, it seems that the application of dielectric coatings with the aim to enhance the interesting details of spectra would be useful in studies of optical effects in metallic films.

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