Energy and thickness dependences of optical coefficients for polycrystalline metallic double-layer films

R. Dimmich
Institute of Physics, Technical University of Wrocław, Wybrzeże Stanisława Wyspiańskiego 27, PL-50-370 Wrocław, Poland

J. Dryzek
Institute of Nuclear Physics, ul. Radzikowskiego 152, PL-31-342 Kraków, Poland

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Formulas for reflection, transmission, and absorption coefficients for electromagnetic radiation in polycrystalline metallic double-layer films are derived. On the basis of the presented model it is shown that the absorption of double-layer systems considered as a function of thickness may exhibit significant departures from the single-layer behavior. The influence of the electron scattering at the film surfaces and interface between layers as well as the scattering at the grain boundaries on the predicted effect is analyzed.

In this paper the influence of free-electron scattering at surfaces, interface, and grain boundaries on the optical coefficients of metallic double-layer films (MDLF's) is studied. In our previous work[1] (paper I) we have presented only the effect of surface and interface scattering on transmission, reflection, and absorption spectra. However, real films have polycrystalline structure, and it is necessary to consider this modification. First, Dimmich and Warkusz[2] and Szczyrbowski *et al.*[3] performed the appropriate calculations for metallic single films (MSF's). Szczyrbowski *et al.*[3,4] formulated a model which enables an analysis of optical-coefficient measurements for MSF's. This model is a development of the Reuter-Sondheimer theory.[5] Deriving new formulas Szczyrbowski *et al.* used the results of the works by Dingle,[6-9] by Hutchison and Hansen,[10] and by Dimmich and Warkusz.[2] Unfortunately, in the case of MDLF's the abovementioned models cannot be applied. Thus, we have elaborated a more general model for monocrystalline MDLFs.[1] The results for polycrystalline MDLF's presented in this paper are also based on those works.[1-10]

It is the aim of this paper to extend the previously developed model[1] taking into account electron–grain-boundary scattering. Moreover, in connection with results of intensive examinations of MDLF dc electrical properties,[11-16] where the MDLF resistance first increases, then passes through a maximum, and finally decreases with increasing overlayer thickness, we would like to show that the MDLF optical-absorption dependence on overlayer thickness may be analogous, and consequently, a significant departure from the behavior of metallic single films may be expected.

The two-layer thin film considered in this analysis consists of the overlayer with surfaces at $z=-d$ and $z=0$ which is made of metal 1 and the base layer with surfaces at $z=0$ and $z=h$ of metal 2. The grain sizes in both layers are D_1 and D_2, respectively. It is also assumed that the metals are isotropic so the background mechanisms of electron scattering can be described by the mean free paths l_1 and l_2, and the influence of contact-potential differences arising at the interface $z=0$ on the discussed effects can be neglected. An electromagnetic wave is incident from the z direction where the electric field $E(z)\exp(i\omega t)$ is taken to be in the x direction and the magnetic field $H(z)\exp(i\omega t)$ in the y direction. Omitting the time-dependent factor and introducing dimensionless coordinates in the layers (i.e., $z=z/l_1$ for $-d<z<0$ and $z=z/l_2$ for $0<z<h$) as well as the reduced layer thicknesses and grain sizes $d=d/l_1$, $h=h/l_2$, and $D_{1,2}=D_{1,2}/l_{1,2}$ one can show that Maxwell equations transform to the following wave equation for the electric fields $E_1(z)$ and $E_2(z)$ within the layers:

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

where $J_{1,2}(\omega,z)$ are the free-electron current densities generated by the electric fields $E_{1,2}(z)$, $\mu_{1,2}$ are the relative magnetic permeabilities, ϵ_0 is the permittivity of free space, and the quantities $(1+S_{1,2})$ are the residual dielectric constants. The notation that index 1 refers to metal 1 and index 2 to metal 2 is used. The current densities $J_{1,2}(\omega,z)$ can be obtained from the linearized Boltzmann transport equation solved with appropriate boundary conditions for surface and interface electron scattering [see Eqs. (4)–(7) in paper I]. The grain-boundary scattering can be taken into account by means of the Mayadas-Shatzkes method,[17] where the grain boundaries are represented as parallel, partially reflecting planes, perpendicular both to the field E and to the plane of the layers. Therefore, the surface scattering will be described by the specularity parameters p_1 and p_2 related to the surfaces $z=-d$ and $z=h$. The interaction between the electrons and the interface will be characterized by two parameters, the specular reflection R_i and the transmission without the diffuse scattering T_i. According to the Mayadas-Shatzkes model[17] the electron scattering at the grain boundaries in metals 1 and 2 will be considered by using the reflection coefficients R_{g1} and R_{g2}, respectively. The values of these parameters are limited by the conditions $0\le p_1,p_2,R_i,T_i,R_{g1},R_{g2}\le 1$ and

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$R_i + T_i \leq 1$.

Thus, using the Mayadas-Shatzkes method[17] along with the previous approach[1] one can obtain for $J_{1,2}(\omega,z)$ the formulas analogous to those in paper I [see Eqs. (8) and (9)] with new relaxation times $\tau^*_{1,2}$ instead of $\tau_{1,2}$ only. After introducing the polar coordinates (v,θ,ϕ) with $v_z = v\cos\theta$ the relaxation times for bulk polycrystalline metals 1 and 2 can be written as

$$\frac{1}{\tau^*_{1,2}} = \frac{1}{\tau_{1,2}}\left[1+\frac{\beta_{1,2}}{\sin\theta\cos\phi}\right],$$

where

$$\beta_{1,2} = \frac{1}{D_{1,2}}\frac{R_{g1,2}}{1-R_{g1,2}} . \tag{2}$$

The method of further solving the wave equation [Eq. (1)] to find expressions for the electric field in each of the layers follows essentially the same lines as those described in paper I. The solutions form series in $\xi_{1,2} = il^2_{1,2}/(2\omega^3\delta^2_{1,2})$, where $\delta_{1,2}$ are the classical penetration depths in both layers. With the assumption that $|\xi_{1,2}| \ll 1$ they may be easily limited but this restricts the validity of our formulas to two ranges: low frequencies where the skin effect is nearly classical and high frequencies from the near-infrared up to the ultraviolet region. The approximate equations for complex refractive indices of the layers can be derived in the form

$$n_1(z) = \frac{n_{1b}}{\mu_1} - F_1(u_{1,2},z) , \tag{3a}$$

$$n'_1(z) = \frac{n_{1b}}{\mu_1} + F_1(-u_{1,2},z) , \tag{3b}$$

$$n_2(z) = \frac{n_{2b}}{\mu_2} - F_2(u_{2,1},z) , \tag{4a}$$

$$n'_2(z) = \frac{n_{2b}}{\mu_2} + F_2(-u_{2,1},z) , \tag{4b}$$

where

$$\begin{aligned}
F_1(u_{1,2},z) = {} & \frac{3}{\pi}\left[\frac{l_1\omega_{p1}}{v_F}\right]^2\frac{v_F}{c}\int_0^{\pi/2}d\phi\,(\cos^2\phi) \\
& \times\int_1^{\infty} ds\left[\frac{1}{s^3}-\frac{1}{s^5}\right]\frac{\exp[u_1w_1(z+d)]}{w'^2_1} \\
& \times\left[\frac{\exp[sw'_1(z+2d)]-p_1\exp(-sw'_1z)}{W'_Y(s)}\right. \\
& \times\left[\{p_2T_i^2+R_i[\exp(2sw'_2h)-p_2R_i]\}[\exp(-u_1w_1d)-\exp(-sw'_1d)]\right. \\
& \quad +\frac{T_i}{a}\{\exp(2sw'_2h)-\exp[-(u_2w_2-sw'_2)h]\} \\
& \quad \left.+p_2\frac{T_i}{a}\{\exp[(sw'_2-u_2w_2)h]-1\}\right] \\
& +p_1\frac{[\exp(2sw'_2h)-p_2R_i][R_i\exp(sw'_1z)-\exp(-sw'_1z)]+p_2T_i^2\exp(s\omega'_1z)}{W'_Y(s)} \\
& \times[\exp(sw'_1d)-\exp(-u_1w_1d)] \\
& \left.-\exp(sw'_1z-u_1w_1d)+\exp[-sw'_1(z+d)]\right] ,
\end{aligned} \tag{5}$$

$$\begin{aligned}
F_2(u_{2,1},z) = {} & \frac{3}{\pi}\left[\frac{l_2\omega_{p2}}{v_F}\right]^2\frac{v_F}{c}\int_0^{\pi/2}d\phi(\cos^2\phi)\int_1^{\infty}ds\left[\frac{1}{s^3}-\frac{1}{s^5}\right]\frac{\exp(u_2w_2z)}{w'^2_2} \\
& \times\left[\frac{p_2\exp(sw'_2z)-\exp[sw'_2(2h-z)]}{W'_Y(s)}\right. \\
& \times(\{p_1T_i^2+R_i[\exp(2sw'_1d)-p_1R_i]\}\{1-\exp[(sw'_2-u_2w_2)h]\} \\
& \left.+aT_i\exp(sw'_1d)\{\exp[(sw'_1-u_1w_1)d]-1\}+p_1aT_i[\exp(sw'_1d)-\exp(-u_1w_1d)]\right)
\end{aligned}$$

$$+p_2\frac{[\exp(2sw_1'd)-p_1R_i][\exp(sw_2'z)-R_i\exp(-sw_2'z)]-p_1T_i^2\exp(-sw_2'z)}{W_Y'(s)}\{\exp[(sw_2'-u_2w_2)h]-1\}$$

$$-\exp[sw_2'z-(sw_2'+u_2w_2)h]+\exp(-sw_2'z)\Big], \tag{6}$$

$$W_Y'(s)=[\exp(2sw_1'd)-p_1R_i][\exp(2sw_2'h)-p_2R_i]-p_1p_2T_i^2,$$

$$a=\frac{w_2'l_1m_2}{w_1'l_2m_1},\quad w_{1,2}=1+i\omega\tau_{1,2},\quad w_{1,2}'=w_{1,2}+\frac{\beta_{1,2}}{(\cos\phi)(1-1/s^2)^{1/2}},$$

$$u_{1,2}w_{1,2}=\frac{i\omega l_{1,2}}{c}\left[\left[1+S_{1,2}-i\frac{\omega_{p1,2}^2\tau_{1,2}}{\omega w_{1,2}}f(\beta_{1,2},w_{1,2})\right]\mu_{1,2}\right]^{1/2}=\frac{i\omega l_{1,2}}{c}n_{1b,2b}, \tag{7}$$

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

is the Mayadas-Shatzkes function, $\omega_{p1,2}$ are the plasma frequencies, $m_{1,2}$ are the effective electron masses, and $n_{1b,2b}$ are the refractive indices of the bulk metals.

The expressions for reflected and transmitted amplitudes of the electric field can be obtained using the interference matrix M defined in paper I [see Eqs. (31)–(33)] on the basis of the Knittl approach.[18] Therefore, the intensity coefficients R, T, and A for optical reflection, transmission, and absorption of MDLF are given by the set of Eqs. (3)–(7) from this work and Eqs. (31)–(33) from paper I.

In order to see the effect of grain-boundary scattering along with surface and interface scattering on MDLF optical spectra and to analyze their thickness and grain-size dependences, the absorption A is calculated for a hypothetical MDLF with a base layer of Ag and an overlayer of Au. It is assumed that the grain-boundary reflection coefficients and specularity parameters are equal in both layers, i.e., $R_{g1}=R_{g2}=R_g$ and $p_1=p_2=p$, as well as that the grain diameters are equal to the layer thicknesses, i.e., $D_1=d$ and $D_2=h$. The other system parameters are taken as in paper I, i.e., $l_1=30$ nm, $l_2=53$ nm, $m_1=m_e$, $m_2=0.85m_e$, $S_1=7$, $S_2=2.55$, the electron concentrations $N_1=N_2=5.9\times10^{22}$ cm^{-3}, and values $n_0=1$ and $n_3=1.5$ for the optical constants of the medium and substrate, respectively.

The theoretical results for optical absorption of the MDLF are presented in Fig. 1 versus energy $\hbar\omega$ and in Fig. 2 versus overlayer thickness d for fixed wavelength of the electromagnetic radiation $\lambda=1$ μm. In Fig. 1 the plots demonstrate the modifications arising from the grain-boundary scattering against a background of the changes evoked by the surface [Fig. 1(a)], and surface and interface [Fig. 1(b)] scattering. It is apparent that in general, the absorption increases with increasing value of the grain-boundary reflection coefficient ($R_g=0,0.2,0.4$) and at the same time the relative absorption enhancement resulting from surface and interface scattering (related to the case without them) decreases. Nevertheless, comparing Figs. 1(a) and 1(b) one may notice that all three scattering mechanisms can be significant in the polycrys-

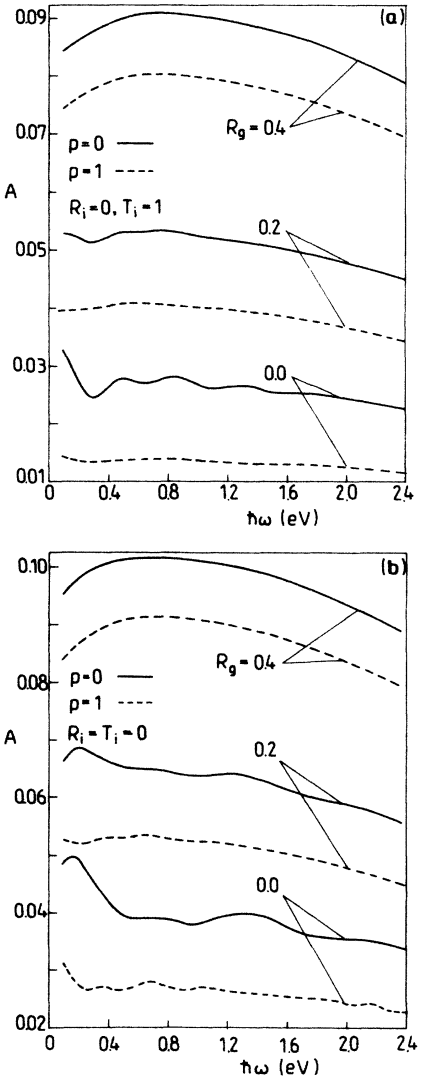

FIG. 1. Absorption spectra of MDLF with layer thicknesses $h=10$ nm (Ag) and $d=5$ nm (Au) for (a) the exclusively coherent passage of electrons across the interface and (b) the diffuse electron scattering at the interface.

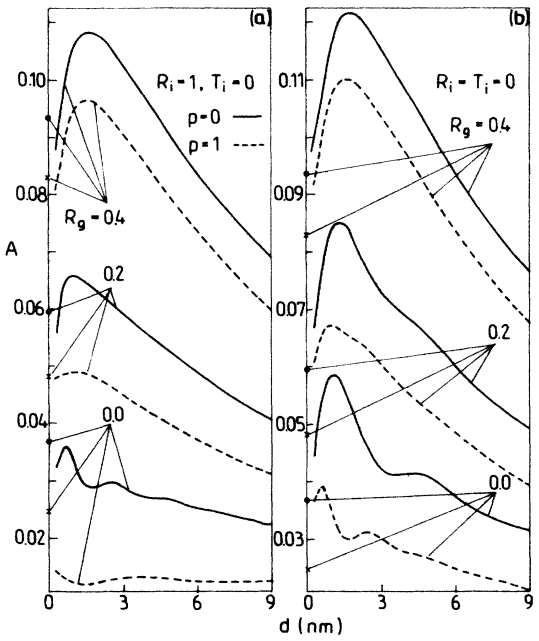

FIG. 2. MDLF absorption dependence on the Au overlayer thickness for fixed base layer thickness $h=10$ nm (Ag), the wavelength $\lambda=1$ μm, and for (a) the exclusively specular electron reflection and (b) the diffuse electron scattering at the interface. The points and crosses at the frames indicate the absorption values of the uncovered base layer with the surface specularity parameters $p=0$ and $p=1$, respectively.

talline MDLF. It is also obvious that the presence of the grain-boundary scattering causes a rapid decay of the absorption oscillations (discussed in paper I) with increasing R_g value.

Figure 2 illustrates the effect of overlayer deposition and its growth on the sample absorption. It is shown that the presence of surface or interface scattering in monocrystalline MDLF leads to the oscillations of absorption as a function of the overlayer thickness. The nature of this effect is the same as in the case of absorption oscillations with energy and analogously to that the polycrystallinity damps the above-mentioned oscillations. It is also worth noticing that, in the case of monocrystalline MDLF where only the surface scattering is operative and the interface reflects electrons specularly [Fig. 2(a)], the absorption of MDLF decreases with increasing overlayer thickness (on the condition that the oscillations are not taken into account). This is absorption behavior analogous to the case of MSF. An opposite effect arises when the interface scattering becomes operative and the electron scattering at the surface system comprising the interface and the external surface of MDLF is enhanced in comparison with scattering at the external surface of an uncovered base layer. In such a film, as is shown in Fig. 2(b), the MDLF absorption first increases, then passes through a maximum, and finally decreases with increasing overlayer thickness. This significant departure from the MSF behavior may be augmented when the grain-boundary scattering is operative in the overlayer. Generally, this scattering may evoke the considered effect even in samples where electrons reflect at the interface exclusively specularly, but only on the condition that the grain-boundary scattering is stronger in the overlayer than in the base layer [Fig. 2(a)].

Concluding, it should be emphasized that all three electron scattering mechanisms under question modify the optical spectra of MDLF in different ways, so it seems possible to discern them. Moreover, the detailed analysis of MDLF optical absorption and dc resistance dependences on the overlayer thickness should be a valuable source of information about the electron-interface interaction.

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