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Nanofilm thickness measurement by resonant frequencies

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Abstract. **We report a theoretical investigation of monochromatic laser light–thin metal film interaction. The dependences of transmission, reflection and absorption coefficients of an electromagnetic wave on the incidence angle, layer thickness and effective electron collision frequency are obtained. The above coefficients are analysed in the region of resonant frequencies. The resulting formula for the transmission, reflection and absorption coefficients are found to be valid for any angles of incidence. The case of mirror boundary conditions is considered. A formula is derived for contactless measurement of the film thickness by the observed resonant frequencies.**

Keywords: *nanofilms, resonant frequencies, transmission, reflection and absorption coefficients of an electromagnetic waves, film thickness.*

1. Introduction

The problem of interaction of an electromagnetic wave with a spatially confined substance has long attracted the attention of researchers [1–9], which is due to both theoretical interest in this issue and many practical applications (see [8, 9]).

Lesskis et al. [10, 11] developed a theory of interaction of electromagnetic radiation with a spherical particle, and the authors of [12, 13] generalised this theory to the case of nonspherical particles. Petrov [14] showed that the electromagnetic properties of small particles may differ significantly from the properties of larger particles.

In papers [15, 16] we considered, respectively, the interaction of H- and E-waves with a thin metal film. We showed that the problem can be solved analytically for films whose thickness is less than the thickness of the skin layer. In addition, we analysed transmission, reflection and absorption coefficients of an electromagnetic wave as functions of the angle of incidence, film thickness, coefficient of specular reflection and field-oscillation frequency.

In paper [17] we considered surface plasmon oscillations in a thin metal film, and in [18, 19] we studied the interaction of electromagnetic H- and E-waves, respectively, with a metal film located between two dielectric media.

To date, there are a considerable number of experimental studies of different kinds of variants of electromagnetic radiation with matter. Let us mention typical papers [20, 21]. Pokrowsky [20] proposed two different experimental method for measuring the thickness and dielectric constants of thin films on a glass substrate. Xu and Tang [21] performed experiments to determine the optical constants and optical absorbance for films of different thicknesses. The results of these experiments agree well with theoretical predictions.

This paper is a continuation of our work [15] and presents an analysis of resonances, which are observed in the transmission, reflection and absorption coefficients during the interaction of an electromagnetic wave with a thin metal film. These resonances are very sensitive to the film thickness and are sufficiently narrow. They can be experimentally observed only if use is made of monochromatic UV laser radiation.

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2. Statement of the problem

Consider a thin layer of a conductive material onto which an electromagnetic wave is incident at angle θ. The magnetic field vector of the electromagnetic wave is assumed parallel to the layer surface and use is made of the Cartesian coordinate system with the origin at one of the layer surfaces and the x axis directed deep into the layer. The y axis is directed parallel to the magnetic field vector of the electromagnetic wave.

The vectors of the electric and magnetic field strengths have the structure: $\boldsymbol{E} = \{E_x, 0, E_z\}$ and $\boldsymbol{H} = \{0, H_y, 0\}$. The components of these vectors are as follows:

$$E_x = E_x(x)\exp(-\mathrm{i}\omega t + \mathrm{i}k\sin\theta z),$$

$$E_z = E_z(x)\exp(-\mathrm{i}\omega t + \mathrm{i}k\sin\theta z),$$

$$H_y = H_y(x)\exp(-\mathrm{i}\omega t + \mathrm{i}k\sin\theta z).$$

The behaviour of the electric and magnetic fields of the wave inside the layer is described by the system of equations [3]

$$\frac{\mathrm{d}E_x}{\mathrm{d}x} - \mathrm{i}kE_x\sin\theta + \mathrm{i}kH_y = 0, \quad \mathrm{i}kE_x - \mathrm{i}kH_y\sin\theta = \frac{4\pi}{c}j_x,$$

$$\frac{\mathrm{d}H_y}{\mathrm{d}x} + \mathrm{i}kE_z = \frac{4\pi}{c}j_z,$$

where c is the velocity of light; j is the current density; and k is the wavenumber.

Transmission, reflection and absorption coefficients of an electromagnetic wave by a layer are described by the expressions [1, 22]:

$$T = \frac{1}{4}|P_1 - P_2|^2, \quad R = \frac{1}{4}|P_1 + P_2|^2, \quad A = 1 - T - R,$$

where

$$P_j = \frac{\cos\theta + Z_j}{\cos\theta - Z_j};\ \text{and}\ j = 1, 2.$$

The quantity Z_1 corresponds to the impedance on the lower surface of the layer for the external field configuration which is symmetric with respect to the magnetic field. This is case 1, for which

$$H_y(0) = H_y(d),\quad E_x(0) = E_x(d),\quad E_z(0) = -E_z(d),$$

where d is the layer thickness. The quantity Z_2 corresponds to the impedance on the lower surface of the layer for the external field configuration which is anti-symmetric with respect to the magnetic field. This is case 2, for which

$$H_z(0) = -H_z(d),\quad E_x(0) = -E_x(d),\quad E_z(0) = E_z(d).$$

In this case, the impedance has the form

$$Z_j = \frac{E_z(-0)}{H_y(-0)}.$$

Consider the case when the layer thickness d is smaller than the skin-layer depth δ. Note that the skin-layer depth essentially depends on the radiation frequency, monotonically decreasing with the growth of the latter. The δ takes the smallest value in a so-called infrared case [23], and $\delta_0 = c/\omega_p$, where ω_p is the plasma frequency. For typical metals [23], $\delta_0 \sim 100$ nm. Thus, for films with $d < \delta_0$ our assumption is valid at any frequencies. From below the skin-layer thickness is limited by atomic dimensions, i.e., the film thickness should be substantially greater than the thickness of a monoatomic layer, and therefore the film should consist of a sufficient number of atomic layers. It follows from the experiment [24] that when the film thickness is less than a few nanometres, the character of kinetic processes in it varies considerably; in particular, its continuity can be broken. In this case, our theory becomes inapplicable. Thus, the films in question have a thickness from several nanometres to 100 nm.

3. Transmission, reflection and absorption coefficients

Let us assume that the wavelength of incident radiation substantially exceeds the layer thickness. Then, the expressions for transmission (T) and reflection (R) coefficients have the form [16]:

$$T = \cos^2\theta \left| \frac{1 - \mathrm{i}k(d/2)G\sin^2\theta(2\pi d\sigma_d/c)}{[\cos\theta + \mathrm{i}k(d/2)G\sin^2\theta][1 + (2\pi d\sigma_d/c)\cos\theta]} \right|^2, \quad (1)$$

$$R = \left| \frac{\mathrm{i}k(d/2)G\sin^2\theta - (2\pi d\sigma_d/c)\cos^2\theta}{[\cos\theta + \mathrm{i}k(d/2)G\sin^2\theta][1 + (2\pi d\sigma_d/c)\cos\theta]} \right|^2. \quad (2)$$

For the case $kl \ll 1$ (l is the electron mean free path), the quantity G can be calculated from the problem of the behaviour of the plasma layer in an alternating electric field that is perpendicular to the layer surface [25, 26]:

$$G = \frac{1}{2}\int_0^d e(x)\,\mathrm{d}x,$$

where $e(x)$ is the electric field. In the case of a nearly grazing incidence, when $\theta \to \pi/2$, we obtain $T \to 0$, $R \to 1$ and $A \to 0$.

Let the relation $kl \ll 1$ be fulfilled. Then, at arbitrary frequencies, $l = v_F\tau/(1 - \mathrm{i}\omega\tau)$, where τ is the electron mean free time; v_F is the Fermi velocity; $\sigma_d = \sigma_0/(1 - \mathrm{i}\omega\tau)$; and $\sigma_0 = \omega_p^2\tau/(4\pi)$ is the static conductivity of a bulk sample. The plasma frequency ω_p of potassium, gold, aluminium and silver are respectively 6.5×10^{15}, 1.37×10^{16}, 3.82×10^{15} and 0.96×10^{15} s^{-1}.

4. Electric field

In expanding the solution of the initial boundary value problem, we constructed [26] a dimensionless electric field in a metal layer:

$$e(x) = \frac{\lambda_1}{\lambda_\infty} + \frac{2\lambda_1\eta_0\cosh[z_0(2x-d)/\eta_0]}{(ac - \eta_0^2)\lambda'(\eta_0)\cosh(z_0/\eta_0)} + \frac{\lambda_1}{2}\int_{-1}^{1} \frac{\eta^2\cosh[z_0(2x-d)/\eta]}{\lambda^+(\eta)\lambda^-(\eta)\cosh(z_0/\eta)}\,\mathrm{d}\eta, \quad (3)$$

where

$$\lambda(z) = c^2 + \frac{z^2}{2}\int_{-1}^{1}\frac{\eta_1^2 - \tau^2}{\tau^2 - z^2}\,\mathrm{d}\tau$$

is the dispersion function; η_0 is its zero; $\eta_1^2 = ac$; $a = dv/(2v_F\kappa)$; $\kappa^2 = 9a_0^2/r_D^2$; $r_D^2 = 3v_F^2/\omega_p^2$; $c = z_0/\kappa$; $z_0 = d(\nu - \mathrm{i}\omega)/2v_F$; r_D is the Debye radius; and $\nu = 1/\tau$ is the electron collision frequency. The dimensionless electric field $e(x)$ is related to the dimensional field $E(x)$ by the expression $e(x) = E(x)/E_0$ (E_0 is the value of the field at the boundary $x = 0$). Then, the value of G has the form (see also [26]):

$$G = \frac{\lambda_1}{\lambda_\infty} + \frac{2\lambda_1\eta_0^2\tanh(z_0/\eta_0)}{z_0(ac - \eta_0^2)\lambda'(\eta_0)} + \frac{\lambda_1}{2z_0}\int_{-1}^{1}\frac{\tanh(z_0/\eta)\eta^3}{\lambda^+(\eta)\lambda^-(\eta)}\,\mathrm{d}\eta. \quad (4)$$

Here, $\lambda_1 = \lambda(\eta_1) = c^2 - ac$ and $\lambda^\pm(\eta) = \lambda(\eta) \pm \mathrm{i}(\pi/2)\eta(\eta_1^2 - \eta^2)$.

The value of G is well approximated by the first two terms of expansion (4):

$$G \approx G_2 = \frac{\lambda_1}{\lambda_\infty} + \frac{2\lambda_1\eta_0^2\tanh(z_0/\eta_0)}{z_0(ac - \eta_0^2)\lambda'(\eta_0)}, \quad (5)$$

i.e., we have replaced the electric field by the first two Drude and Debye terms corresponding to a discrete spectrum.

To calculate G_2 we need an explicit expression for the dispersion-function zero $\eta_0 = \eta_0(\Omega, \varepsilon)$, where $\Omega = \omega/\omega_p$.

We will present without proof a formula (see [26]) of the dispersion-function factorization

$$\lambda(z) = \lambda_\infty(\eta_0^2 - z^2)X(z)X(-z). \quad (6)$$

Here,

$$\lambda_\infty = \lambda(\infty) = \frac{1}{3} + ac - c^2 = \frac{1}{3}(1 - \Omega^2 - \mathrm{i}\varepsilon\Omega);$$

$$X(z) = \frac{1}{z}\exp V(z);\quad V(z) = \frac{1}{2\pi\mathrm{i}}\int_0^1 \frac{\ln G(\tau) - 2\pi\mathrm{i}}{\tau - z}\,\mathrm{d}\tau;$$

$$G(\tau) = \frac{\lambda^+(\tau)}{\lambda^-(\tau)};\quad \lambda^\pm(\tau) = c^2 - ac - (\tau^2 - ac)\lambda_0^\pm(\tau);$$

$$\lambda_0^{\pm}(\tau) = \lambda_0(\tau) \pm \frac{\pi}{2}\tau\mathrm{i};$$

$$\lambda_0(\tau) = 1 + \frac{\tau}{2}\int_{-1}^{1}\frac{\mathrm{d}\tau'}{\tau'-\tau} = 1 + \frac{\tau}{2}\ln\frac{1-\tau}{1+\tau}.$$

If we calculate the values of the left and right sides of equation (6) at point $z = \mathrm{i}$, then for the square of the dispersion-function zero after some transformations we obtain the expression:

$$\eta_0^2 = -1 + \frac{\lambda(\mathrm{i})}{\lambda_\infty X(\mathrm{i})X(-\mathrm{i})} = -1 + \frac{\lambda(\mathrm{i})}{\lambda_\infty}\exp[-V(\mathrm{i}) - V(-\mathrm{i})].$$

Given that $\lambda_0(\mathrm{i}) = 1 - \pi/4$, we have

$$\lambda(\mathrm{i}) = c^2 - ac + \left(1 - \frac{\pi}{4}\right)(1 + ac)$$

$$= -\frac{1}{3}(\Omega^2 + \mathrm{i}\varepsilon\Omega) + \left(1 - \frac{\pi}{4}\right)\left[1 + \frac{1}{3}(\varepsilon^2 - \mathrm{i}\varepsilon\Omega)\right].$$

The function $X(z) = (1/z)\exp V(z)$ can be written in the form

$$X(z) = \frac{1}{z-1}\exp V_0(z),$$

where

$$V_0(z) = \frac{1}{2\pi\mathrm{i}}\int_0^1 \frac{\ln G(\tau)}{\tau - z}\,\mathrm{d}\tau.$$

Let us find the sum

$$V_0(\mathrm{i}) + V_0(-\mathrm{i}) = \frac{1}{2\pi\mathrm{i}}\int_{-1}^{1}\frac{\ln G(\tau)\mathrm{d}\tau}{\tau - \mathrm{i}} + \frac{1}{2\pi\mathrm{i}}\int_0^1\frac{\ln G(\tau)\mathrm{d}\tau}{\tau + \mathrm{i}}$$

$$= \frac{1}{2\pi\mathrm{i}}\int_{-1}^{1}\frac{\ln G(\tau)\mathrm{d}\tau}{\tau - \mathrm{i}} = \frac{1}{2\pi\mathrm{i}}\int_{-1}^{1}\frac{\tau\ln G(\tau)\mathrm{d}\tau}{\tau^2 + 1}.$$

With these formulas taken into account we transform the formula for the square of the dispersion-function zero to the form

$$\eta_0^2 = -1 + \frac{2\lambda(\mathrm{i})}{\lambda_\infty}\exp\left[-\frac{1}{2\pi\mathrm{i}}\int_{-1}^{1}\frac{\tau\ln G_l(\tau)\mathrm{d}\tau}{\tau^2+1}\right]$$

or

$$\eta_0^2 = -1 + \frac{2\lambda(\mathrm{i})}{\lambda_\infty}\exp\left[\frac{\mathrm{i}}{\pi}\int_{-1}^{1}\frac{\tau\ln G_l(\tau)\mathrm{d}\tau}{\tau^2+1}\right],$$

where

$$G_l(\tau) = \ln\frac{(3\tau^2 - \varepsilon^2 + \mathrm{i}\varepsilon\Omega)(\lambda_0(\tau) + (\pi/2)\tau\mathrm{i}) + \Omega^2 + \mathrm{i}\varepsilon\Omega}{(3\tau^2 - \varepsilon^2 + \mathrm{i}\varepsilon\Omega)(\lambda_0(\tau) - (\pi/2)\tau\mathrm{i}) + \Omega^2 + \mathrm{i}\varepsilon\Omega}.$$

Now the relative error

$$O_2(\Omega, \varepsilon, d) = \left|\frac{G - G_2}{G}\right| \times 100\%$$

for 5- and 10-nm-thick films made of potassium at $\omega = \omega_\mathrm{p}$ and $\nu = 10^{-3}\omega_\mathrm{p}$, respectively, is equal to 0.003% and 0.0004%.

Numerical calculations show that in the $\omega > \omega_\mathrm{p}$ region, the contribution to the electric field, corresponding to a continuous spectrum, is insignificant and can be neglected. Thus, the function G can be approximated by two Drude and Debye terms corresponding according to (5) to a discrete spectrum.

5. Behaviour of the coefficients and discussion of the results

Using (1) and (2) and expression (5) for the function G, we will study transmission, reflection and absorption coefficients in graphic form.

Consider the case of a thin film made of potassium. We construct the dependences of the transmission coefficient on the value of $\Omega = \omega/\omega_\mathrm{p}$ at an incidence angle $\theta = 75°$ (Fig. 1). This value of the angle is used in [1, 2].

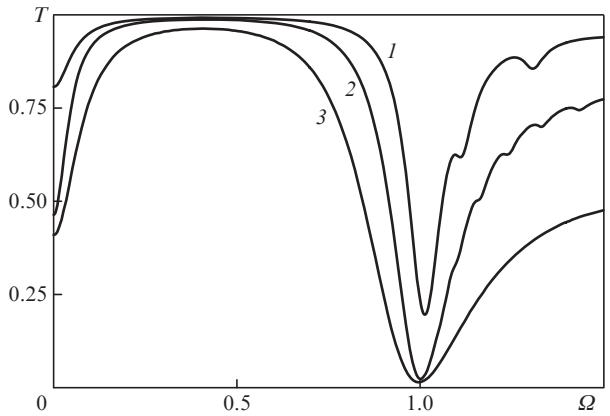

Figure 1. Dependences of the transmission coefficient T on Ω for (*1*) $d = 2$ nm, $\nu = 0.05\omega_\mathrm{p}$, (*2*) $d = 5$ nm, $\nu = 0.03\omega_\mathrm{p}$ and (*3*) $d = 10$ nm, $\nu = 0.05\omega_\mathrm{p}$ at $\theta = 75°$.

Note that near the plasma resonance ($\omega \sim \omega_\mathrm{p}$) the transmission coefficient has a minimum, whereas the reflection and absorption coefficients have a maximum. At a film thickness of 1.5 nm and $\nu = 0.05\omega_\mathrm{p}$, all the coefficients in the region of super-resonant frequencies ($\omega > \omega_\mathrm{p}$) have one more maximum. With increasing film thickness from 1.5 nm to 10 nm the second maximum disappears.

At a film thickness of 5 nm and $\nu = 0.02\omega_\mathrm{p}$, the behaviour of all the coefficients in the region of super-resonant frequencies exhibits a so-called comb-like structure ('stockade'). With a further increase in the film thickness the comb frequency increases, and we can see an increase in the amplitude of its teeth. If at a film thickness of 5 nm, the value of $\varepsilon = \nu/\omega_\mathrm{p}$ decreases, the amplitude of the comb teeth increases sharply.

A further increase in the film thickness leads to an increase in the frequency of the comb teeth (number of palings in the stockade). Figure 2 shows the behaviour of the reflection and absorption coefficients as a function of Ω for $d = 10$ nm, $\nu = 0.001\omega_\mathrm{p}$ and $\theta = 75°$. Note that Figs 2a and 2b virtually coincide with Figs 2 and 3, respectively, from [1].

Figure 3 shows the dependences of the transmission, reflection and absorption coefficients on the film thickness d at $\nu = 0.001\omega_\mathrm{p}$ and $\theta = 75°$. In this range of thicknesses in question the transmission coefficient has one minimum and the absorption coefficient has one maximum.

We proceed to the derivation of the formula for calculating the film thickness by the points Ω_n at which the coefficients of

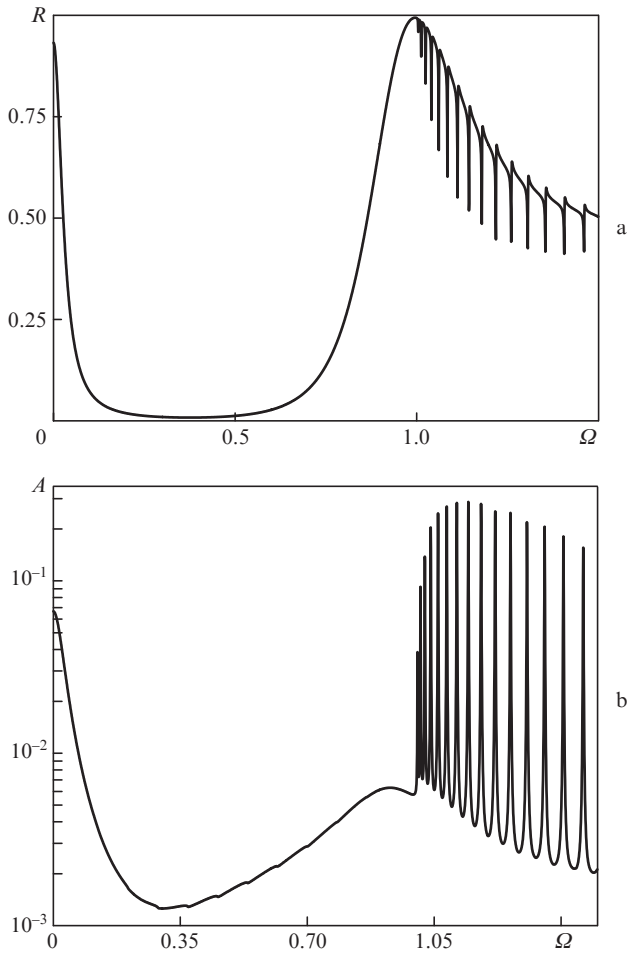

Figure 2. Dependences of the (a) reflection R and (b) absorption A coefficients on Ω for $d = 10$ nm, $\nu = 0.001\,\omega_p$ and $\theta = 75°$.

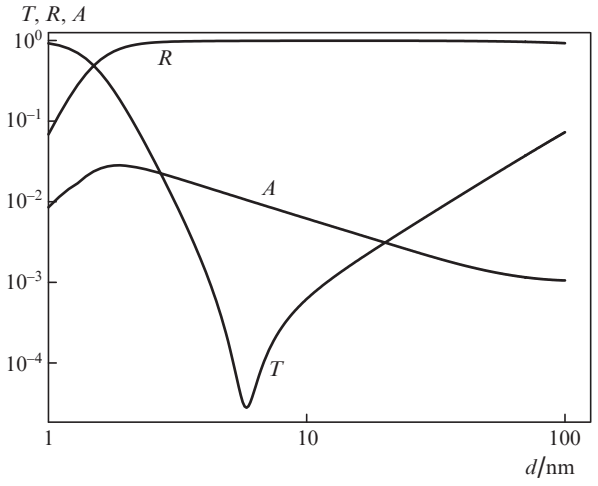

Figure 3. Dependences of the transmission, reflection and absorption coefficients on the film thickness d at $\omega = \omega_p$, $\nu = 0.001\,\omega_p$ and $\theta = 75°$.

transmission, reflection and absorption have extremes. Consider the reflection coefficient.

Figure 4 shows the dependences of the reflection coefficient on Ω for 10-nm-thick films made of potassium, gold and silver at $\nu = 0.001\omega_p$. Thus, Fig 4 presents the first teeth of the comb shown earlier in Fig. 2.

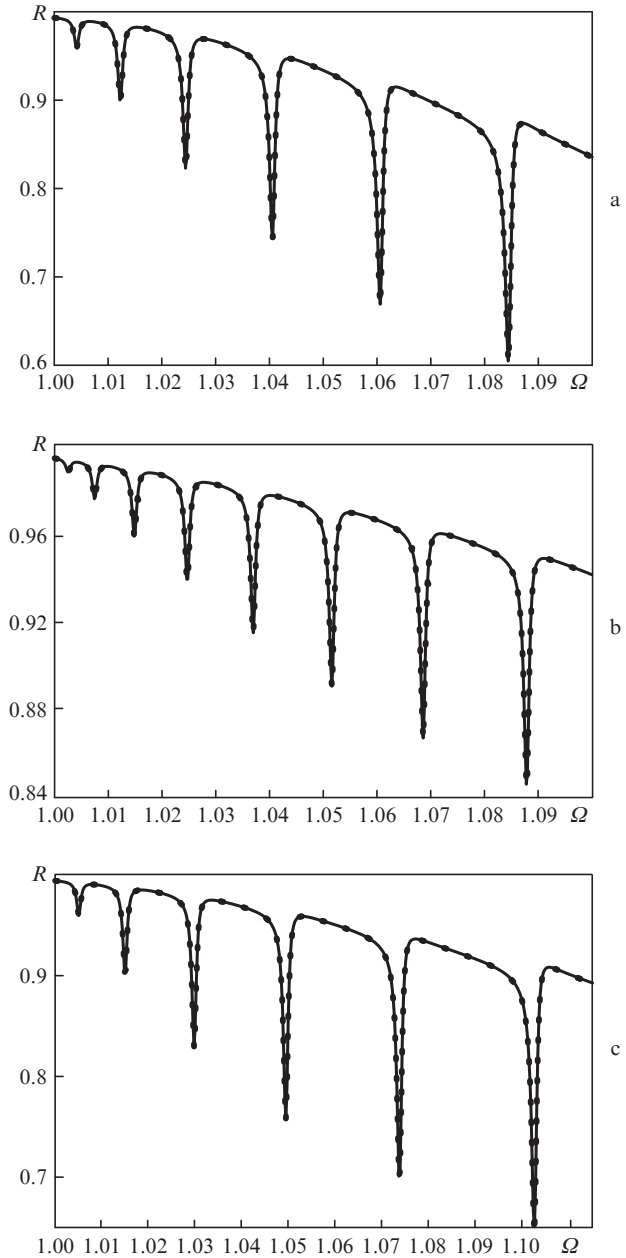

Figure 4. Dependences of the reflection coefficient R on Ω for a film made of (a) potassium, (b) gold and (c) silver at $d = 10$ nm, $\nu = 0.001\,\omega_p$ and $\theta = 75°$.

In these figures, the dotted curve corresponding to discrete and continuous spectra coincides with the solid curve corresponding only to the discrete spectrum, which is consistent with the above estimates. In constructing the dotted curve use is made of formula (4), while the solid curve makes use of formula (5).

The analysis shows that the coefficients of transmission, reflection and absorption have extremes at the same points Ω_n, regardless of the angle of incidence of the electromagnetic wave. These considerations allow us to find the film thickness by those points $\Omega_n = \omega_n/\omega_p$, at which the coefficients T, R and A have an extremum.

The second term (Debye mode) in formulas (4) and (5) includes the function $\mathrm{th}(z_0/\eta_0) = -\mathrm{i}\sin(\mathrm{i}z_0/\eta_0)/\cos\mathrm{i}(z_0/\eta_0)$, responsible for the quasi-periodic comb-like behaviour of the dependences. Points Ω_n, at which the reflection coefficient has

a minimum, exactly coincide with the points at which the function $\cos \mathrm{Re}(\mathrm{i}z_0/\eta_0)$ vanishes. From the equation $\cos \mathrm{Re}(\mathrm{i}z_0/\eta_0)$ we find

$$\mathrm{Re}\left(\mathrm{i}\frac{z_0(\Omega_n,\varepsilon,d)}{\eta_0(\Omega_n,\varepsilon)}\right)+\frac{\pi}{2}+\pi n,$$

or in the explicit form,

$$\mathrm{Re}\left(\mathrm{i}\,\frac{\omega_\mathrm{p}10^{-7}(\varepsilon-\mathrm{i}\Omega_n)}{2v_\mathrm{F}\eta_0(\Omega_n,\varepsilon)}\,\mathrm{d}\right)=\frac{\pi}{2}+\pi n, \tag{7}$$

where $n = 1, 2, 3, \ldots$.

In (7) d is measured in nanometres. It follows from this formula that the reflection coefficient has local minima at

$$d_n=\frac{10^7\pi v_\mathrm{F}(1+2n)}{\omega_\mathrm{p}\mathrm{Re}[(\Omega_n+\mathrm{i}\varepsilon)/\eta_0(\Omega_n\varepsilon)]}\,. \tag{8}$$

Table 1 shows the first six resonant frequencies at which the reflection coefficient has local minima. The results are shown for ∼10-nm-thick films made of different materials. The data presented in Table 1 suggest that the error in measuring the film thickness by the observed frequencies does not exceed 1%.

Table 1. Measurement results of the film thickness.

Film material	Extremum number	Frequency/ s^{-1}	Film thickness/ nm	Relative error (%)
Potassium	1	1.0046	9.959	–0.4
	2	1.0127	9.997	–0.03
	3	1.0250	9.968	–0.3
	4	1.0406	10.046	0.04
	5	1.0608	10.017	0.17
	6	1.0847	10.008	0.08
Gold	1	1.0028	9.928	–0.7
	2	1.0077	10.009	–0.09
	3	1.0149	10.073	–0.73
	4	1.0249	10.012	0.12
	5	1.0370	10.003	0.03
	6	1.0518	10.006	0.06
Silver	1	1.0056	9.975	–0.3
	2	1.0152	10.093	0.9
	3	1.0298	10.082	0.8
	4	1.0490	10.094	0.9
	5	1.0730	10.032	0.3
	6	1.1029	10.012	0.1

6. Conclusions

Thus, we have considered the films whose thicknesses are equal to tens of nanometres and do not exceed the thickness of the skin layer. We have derived formulas for calculating transmission, reflection and absorption coefficients of these films and have analysed these coefficients. We have noted that there exist resonant frequencies related to resonances in longitudinal plasmon oscillations. We have found that these frequencies exhibit a greater sensitivity to the film thickness, which in principle allows one to measure the film thickness by using the values of these frequencies. We have derived a formula for determining the film thickness by the position of the resonant frequencies.

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