

Surface plasmon polaritons in dielectric/metal/dielectric structures: metal layer thickness influence

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A model is proposed and studied that makes it possible to explain experimental data on a metal layer thickness influence on the spectrum of SPPs waves in heterogeneous dielectric/metal/dielectric structures.

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1. Introduction

Surface plasmon polaritons (SPPs) are collective excitations of electrons, propagating at the interface between a metal and a dielectric [1, 2]. They are used to manipulate electromagnetic energy at the subwavelength scales, which necessitates the study of their characteristics.

A considerable amount of research on the study of SPPs spectrum in heterogeneous dielectric/metal/dielectric structures has been published for today (see bibliography in [1, 3]), in which a metal layer is mainly considered as 2D metal or metallic graphene using the corresponding characteristic expressions for the dielectric function $\varepsilon(\mathbf{q}, \omega)$ ($\mathbf{q} = (k_x, k_y)$ is 2D vector, ω is frequency) of a metal.

In the case when a metal layer is a 3D structure, the Drude model is widely used to describe SPPs [1, 4] in which the dielectric permittivity of a metal is expressed by the formula:

$$\begin{aligned} \varepsilon(\mathbf{q}, z, z', \omega) &= \varepsilon^D(\omega) \delta(z - z'), \\ \varepsilon^D(\omega) &= 1 - \frac{\omega_p^2}{\omega^2 + i\gamma\omega}. \end{aligned} \quad (1)$$

Unfortunately, this approach to the description of SPPs does not allow taking into account the influence of the thickness of a metal layer and size effects on their properties.

Recently, in the paper [5] there has been presented the results of experimental studies on the influence of a thickness of a metal film on the spectrum $\hbar\omega$ (\hbar is the reduced Planck constant [6]) of SPPs, where it has been demonstrated that such a dependence is significant in the area of small ($\sim 1 - 5$ nm) thicknesses. In this paper, a mathematical model for $\varepsilon(\mathbf{q}, \omega, z, z')$ is proposed and studied and it is shown that taking into account the thickness of a metal layer can be described by such a model; and the obtained results qualitatively coincide with the experimental results.

2. Problem formulation

Consider a heterogeneous structure (whose geometry is depicted in Fig.1) formed by two non-conducting media with dielectric permittivities ε_1 and ε_3 between which a metal nanofilm with thickness L is sandwiched. We assume that dielectric permittivities ε_1 and ε_3 are functions of the time variable, i.e.

$$\varepsilon_1 = \varepsilon_1(t), \quad \varepsilon_3 = \varepsilon_3(t). \quad (2)$$

Dielectric permittivity of metal is $\varepsilon_2(\mathbf{r}, \mathbf{r}', t)$ and for the geometry of a dielectric/metal/dielectric heterostructure has the form

$$\varepsilon_2 = \varepsilon_2(\mathbf{r}_{\parallel} - \mathbf{r}'_{\parallel}, z, z', t), \quad \mathbf{r}_{\parallel} = (x, y). \tag{3}$$

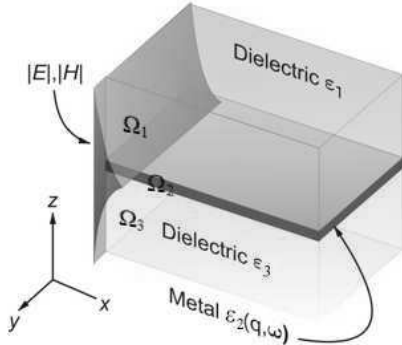


Fig. 1. Schematic representation of dielectric/metal/dielectric structure.

Let us consider a problem of describing the propagation of electromagnetic waves, which are localized at the interface between a dielectric ($z \geq L$) and a metal ($0 < z < L$). These waves are called surface plasmons [1]. A mathematical model describing the propagation of surface plasmons is based on the Maxwell’s equations system [1, 2]:

$$\begin{aligned} \nabla \cdot \mathbf{D} &= \rho, & \nabla \cdot \mathbf{B} &= 0, \\ \nabla \times \mathbf{E} &= -\dot{\mathbf{B}}, & \nabla \times \mathbf{H} &= \mathbf{J} + \dot{\mathbf{D}}, \end{aligned} \tag{4}$$

where \mathbf{D} is electric flux density, \mathbf{B} is magnetic flux density, \mathbf{E} is electric field strength, \mathbf{H} is magnetic field strength, ρ is electric charge density and \mathbf{J} is electric current density. We assume that external charges ρ in the area of contact between dielectrics and metal are absent, namely $\nabla \cdot \mathbf{D} = 0$. Here “ \cdot ” is the dot product, “ \times ” is the cross product.

We assume that interconnection between the vectors \mathbf{E} and \mathbf{D} [2], namely

$$\mathbf{D}(\mathbf{r}_{\parallel}, z, t) = \iiint d\mathbf{r}'_{\parallel} dz' dt' \varepsilon_i(\mathbf{r}_{\parallel} - \mathbf{r}'_{\parallel}, z, z', t - t') \mathbf{E}(\mathbf{r}'_{\parallel}, z', t'), \quad i = 1, 2, 3. \tag{5}$$

Let us write the system of Maxwell’s equations (4) in Fourier variables. We will define the Fourier transform with respect to time as

$$f(t) = \frac{1}{2\pi} \int_{-\infty}^{\infty} \tilde{f}(\omega) e^{i\omega t} d\omega, \quad \tilde{f}(\omega) = \int_{-\infty}^{\infty} f(t) e^{-i\omega t} dt. \tag{6}$$

And $\varepsilon_i(\mathbf{r}_{\parallel} - \mathbf{r}'_{\parallel}, z, z', t - t')$ is expressed by the following equation:

$$\varepsilon_i(\mathbf{r}_{\parallel} - \mathbf{r}'_{\parallel}, z, z', t - t') = \frac{\Omega}{(2\pi)^3} \int_{-\infty}^{\infty} d\omega \int_{\Omega} d\mathbf{q} \varepsilon_i(\mathbf{q}, z, z', \omega) e^{-i(\mathbf{q}, \mathbf{r}_{\parallel} - \mathbf{r}'_{\parallel}) - i\omega(t - t')}, \tag{7}$$

where Ω is the domain of the 2D vector $\mathbf{q} = (k_x, k_y)$.

We will assume that

$$\begin{aligned} \varepsilon_1(\mathbf{r}_{\parallel} - \mathbf{r}'_{\parallel}, z, z', t - t') &= \varepsilon_1(t - t') \delta(\mathbf{r}_{\parallel} - \mathbf{r}'_{\parallel}) \delta(z - z'), \\ \varepsilon_2(\mathbf{r}_{\parallel} - \mathbf{r}'_{\parallel}, z, z', t - t') &= \varepsilon_2(\mathbf{r}_{\parallel} - \mathbf{r}'_{\parallel}, z, z', t - t') \delta(z - z'), \\ \varepsilon_3(\mathbf{r}_{\parallel} - \mathbf{r}'_{\parallel}, z, z', t - t') &= \varepsilon_3(t - t') \delta(\mathbf{r}_{\parallel} - \mathbf{r}'_{\parallel}) \delta(z - z'), \end{aligned} \tag{8}$$

where $\delta(z - z')$ is the Dirac delta function [6].

The polarization of the waves corresponds to the transverse magnetic (TM) mode for the vectors \mathbf{E} and \mathbf{H} , that is

$$\mathbf{E} = (E_x, 0, E_y), \quad \mathbf{H} = (0, H_y, 0). \tag{9}$$

Consequently [1], the magnetic field propagates along the axis OX and is homogeneous along the axis OY

$$\mathbf{H}(\mathbf{r}, \omega) = \mathbf{H}(z, \omega) e^{ik_x x}, \tag{10}$$

k_x is a wave vector in the direction of propagation.

For \mathbf{H} we obtain a system of wave equations for all the domains of the heterogeneous structure [1]:

$$\frac{\partial^2 H_y(z, \omega)}{\partial z^2} + (k_0^2 \varepsilon_1(\omega) - k_x^2) H_y(z, \omega) = 0, \tag{11}$$

$$\frac{\partial^2 H_y(z, \omega)}{\partial z^2} + (k_0^2 \varepsilon_2(\mathbf{q}, z, z, \omega) - k_x^2) H_y(z, \omega) = 0, \tag{12}$$

$$\frac{\partial^2 H_y(z, \omega)}{\partial z^2} + (k_0^2 \varepsilon_3(\omega) - k_x^2) H_y(z, \omega) = 0, \tag{13}$$

where $k_0 = \omega/c$. In order to solve the system (11)–(13), we need to find out expressions for dielectric permittivities $\varepsilon_1(\omega)$, $\varepsilon_2(\mathbf{q}, z, z, \omega)$, and $\varepsilon_3(\omega)$.

3. Model of dielectric permittivity of a metal layer

Here and subsequently, we will use a high-frequency approximation for dielectric layers, this implies that in (8) the first and the last expressions can be rewritten as follows

$$\begin{aligned} \varepsilon_1(\omega) &= \varepsilon_1(\infty) = \varepsilon_1 = \text{const}, \\ \varepsilon_3(\omega) &= \varepsilon_3(-\infty) = \varepsilon_3 = \text{const}. \end{aligned} \tag{14}$$

As a model for the dielectric function $\varepsilon_2(\mathbf{q}, z, z', \omega)$ of the metal layer, we will use the diagonal component of the dielectric permittivity tensor of a metal film which is obtained in [7],

$$\varepsilon(\mathbf{r}, \mathbf{r}', \omega) = \left(1 - \frac{\omega_p^2}{n_e \omega^2} \sum_n f_n |\psi_n(\mathbf{r}')|^2 \right) \delta(\mathbf{r} - \mathbf{r}'). \tag{15}$$

Here $\omega_p = \sqrt{4\pi n_e e^2 / m_e}$ is the plasma frequency [1, 4], n_e is an electron density in a metal, $f_n = \Theta(\varepsilon_n - \varepsilon_F)$ is the Fermi–Dirac function [7, 8], $\Theta(x)$ is the Heaviside step function [8], ε_F is the Fermi energy [4, 8], $\mathbf{r} = (\mathbf{r}_{||}, z)$.

The function

$$\psi_n(x, y, z) = \sqrt{\frac{2}{S}} e^{i(\mathbf{q} \cdot \mathbf{r}_{||})} \phi_n(z) \tag{16}$$

is a wave function [6] of an electron in the metal layer and $\phi_n(z)$ is the solution of equation

$$-\frac{\hbar^2}{2m} \frac{d^2}{dz^2} \phi_n(z) + U(z) \phi_n(z) = W \phi_n(z), \tag{17}$$

which is the Schrödinger equation [3] that describes behaviour of an electron in a metal film [6, 9, 10]. Potential $U(z)$ that simulates surfaces bounding the film has the form

$$U(z) = \begin{cases} U_1 & \text{if } z < 0, \\ 0 & \text{if } 0 < z < L, \\ U_2 & \text{if } z > L, \end{cases} \tag{18}$$

where L is the film thickness. The solutions of the equation (17) that satisfies the conditions $\phi(z \rightarrow \pm\infty) \rightarrow 0$ can be presented as follows

$$\phi_n(z) = \begin{cases} A e^{\chi_1 z}, & \chi_1 = \sqrt{\frac{2m}{\hbar^2} (U_1 - W)} & \text{if } z < 0, \\ C_1 e^{ikz} + C_2 e^{-ikz}, & k = \sqrt{\frac{2m}{\hbar^2} W} & \text{if } 0 < z < L, \\ B e^{-\chi_1 z}, & \chi_2 = \sqrt{\frac{2m}{\hbar^2} (U_2 - W)} & \text{if } z > L. \end{cases} \tag{19}$$

Constants A , C_1 , C_2 , and B we will determine using continuity conditions for $\phi_n(z)$ and $\frac{d\phi_n(z)}{dz}$ on the boundaries $z = 0$ and $z = L$ and a normalization condition

$$\int_{-\infty}^{\infty} |\phi_n(z)|^2 = 1, \tag{20}$$

which, actually, yields the condition $\phi(z \rightarrow \pm\infty) \rightarrow 0$. Hence, the expressions for constants have the form

$$C_1 = A \left(\frac{1}{2} - \frac{i\chi_1}{2k} \right), \quad C_2 = A \left(\frac{1}{2} + \frac{i\chi_1}{2k} \right). \tag{21}$$

$$B = A \left(\cos(kL) + \frac{\chi_1}{k} \sin(kL) \right) e^{\chi_2 L}, \tag{22}$$

$$|A|^2 = \left(\frac{1}{2\chi_1} + \frac{1}{2\chi_2} \left(\cos(kL) + \frac{\chi_1}{k} \sin(kL) \right) \right)^2 + \frac{L}{2} \left(\frac{\chi_1^2}{k^2} + 1 \right) + \frac{\chi_1}{2k^2} \left(1 - \cos(2kL) \right) + \frac{1}{4} \left(\frac{1}{k} - \frac{\chi_1^2}{k^3} \right) \sin(2kL) \Big)^{-1}. \tag{23}$$

In order to find k , we need to solve the following equation [3, 9, 10]

$$kL = \pi n - \left(\arcsin \frac{k\hbar}{\sqrt{2mU_1}} + \arcsin \frac{k\hbar}{\sqrt{2mU_2}} \right), \tag{24}$$

the roots of (24) will determine a value of

$$W_n = \frac{\hbar^2 k_n^2}{2m} \tag{25}$$

which is discrete.

The maximum number of energy levels n_{\max} we determine from the condition

$$n_{\max} = \left\lceil \frac{1}{\pi} \left(L \min(S_1, S_2) + \arcsin \frac{\min(S_1, S_2)}{S_1} + \arcsin \frac{\min(S_1, S_2)}{S_2} \right) \right\rceil, \tag{26}$$

$$S_i = \sqrt{\frac{2m}{\hbar^2} U_i}, \quad i = 1, 2.$$

Square brackets indicate taking the integer part.

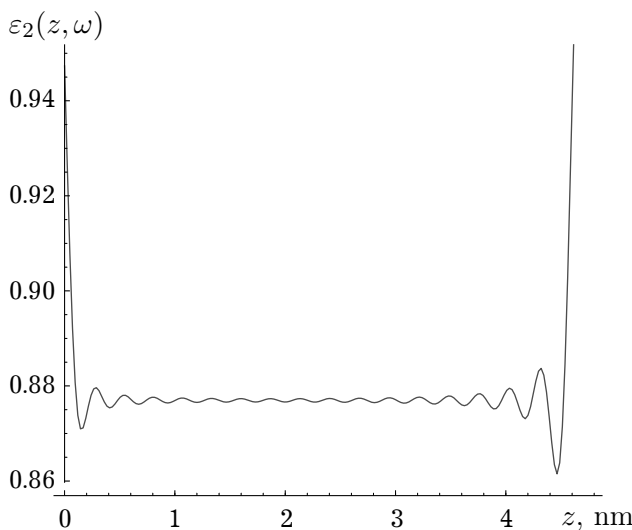


Fig. 2. The dielectric permittivity of the metal layer ($L = 5$ nm) when $\frac{\omega}{\omega_p} = 4$.

Expression for $\varepsilon(\mathbf{q}, z, z', \omega)$ (7) for the model (15) has the form (for details see [7])

$$\begin{aligned} \varepsilon_2(0, z, z', \omega) &= \varepsilon_2(z, \omega) \delta(z - z') \\ &= \left(1 - \frac{\omega_p^2}{\pi n_e \omega^2} \sum_{n=1}^{n_{\max}} (k_F^2 - k_n^2) |\phi_n(z)|^2 \right) \times \delta(z - z'). \end{aligned} \tag{27}$$

The results of numerical calculations of $\varepsilon_2(z, \omega)$ for specific values $U_1 = 4.2$ eV and $U_2 = 5$ eV, which correspond to the dielectrics (1 – polyethylene, 2 – SiO₂), are shown in Fig. 2.

The results obtained have shown that the dielectric function $\varepsilon_2(z, \omega)$ is different from constant only near the contact areas ($z = L$ and $z = 0$) (Fig. 2). This allows making some simplifications when studying the system (11)–(13).

4. Investigation of the influence of the thickness of metal film on the wave spectrum

To solve the system of wave equations (11)–(13), Eq. (27) will be assumed that

$$\varepsilon_2(z, z', \omega) = (\varepsilon_2(L, \omega) + \alpha \Delta \varepsilon_2(z, \omega)) \delta(z - z'), \tag{28}$$

where

$$\varepsilon_2(L, \omega) = \frac{1}{L} \int_0^L \varepsilon_2(z, \omega) dz = 1 - \frac{\omega_p^2}{2\pi n_e \omega^2} \sum_{n=1}^{n_{\max}} (k_F^2 - k_n^2) \overline{|\phi_n(z)|^2}, \tag{29}$$

$$\begin{aligned} \overline{|\phi_n(z)|^2} &= \frac{1}{L} \int_0^L |\phi_n(z)|^2 dz \\ &= |A|^2 \left(\frac{1}{2} \left(\frac{\chi_1^2}{k^2} + 1 \right) + \frac{\chi_1}{2k^2 L} (1 - \cos(2kL)) + \frac{1}{4L} \left(\frac{1}{k} - \frac{\chi_1^2}{k^3} \right) \sin(2kL) \right). \end{aligned} \tag{30}$$

Substitution of (29) into (12) yields

$$\frac{\partial^2 H_y(z, \omega)}{\partial z^2} + (k_0^2 (\varepsilon_2(L, \omega) + \alpha \Delta \varepsilon_2(z, \omega)) - k_x^2) H_y(z, \omega) = 0. \tag{31}$$

The solution of the equation (31) for $H_y(z, \omega)$ can be found as expansion into a series in increasing powers of α

$$H_y(z, \omega) = \sum_{m=0}^{\infty} \alpha^m H_{y,m}(z, \omega). \tag{32}$$

In particular, the first two equations $H_{y,0}(z, \omega)$ and $H_{y,1}(z, \omega)$ are following:

$$\frac{\partial^2 H_{y,0}(z, \omega)}{\partial z^2} + (k_0^2 \varepsilon(L, \omega) - k_x^2) H_{y,0}(z, \omega) = 0, \tag{33}$$

$$\frac{\partial^2 H_{y,1}(z, \omega)}{\partial z^2} + (k_0^2 \varepsilon(L, \omega) - k_x^2) H_{y,1}(z, \omega) = -k_0^2 \alpha \Delta \varepsilon_2(z, \omega) H_{y,0}(z, \omega). \tag{34}$$

When modeling the influence of the thickness of a metal film L on SPPs spectrum, we will limit ourselves to the case $H_y(z, \omega) \approx H_{y,0}(z, \omega)$. In this case, the dispersion relation has the form

$$e^{-4k_1 \frac{L}{2}} = \frac{k_1/\varepsilon_1 + k_2/\varepsilon_2}{k_1/\varepsilon_1 + k_2/\varepsilon_2} \frac{k_3/\varepsilon_1 + k_3/\varepsilon_2}{k_3/\varepsilon_3 + k_2/\varepsilon_2}, \tag{35}$$

$$k_i^2 = k_x^2 - k_0^2 \varepsilon_i, \quad i = 1, 2, 3; \quad k_0 = \frac{\omega}{c}, \tag{36}$$

which coincides with the results obtained in Ref. [1]. Here $\varepsilon_1 = \varepsilon(\omega)$, $\varepsilon_2 = \varepsilon(L, \omega)$, and $\varepsilon_3 = \varepsilon(\omega)$.

Similarly as in our previous work [11], for the upper layer we took a polyethylene with a permittivity constant $\varepsilon_1 = 2.3$ and the electron work function $U_1 = 4.24$ eV; SiO₂ for the lower dielectric substrate with a permittivity constant $\varepsilon_1 = 4$ and the electron work function $U_2 = 5$ eV.

The dielectric function of the metal layer (gold) is described by the function (16)

$$\varepsilon_2(L, \omega) = 1 - \frac{\omega_p^2}{2\pi n_e \omega^2} \sum_{n=1}^{n_{\max}} (k_F^2 - k_n^2) \overline{|\phi_n(z)|^2}. \tag{37}$$

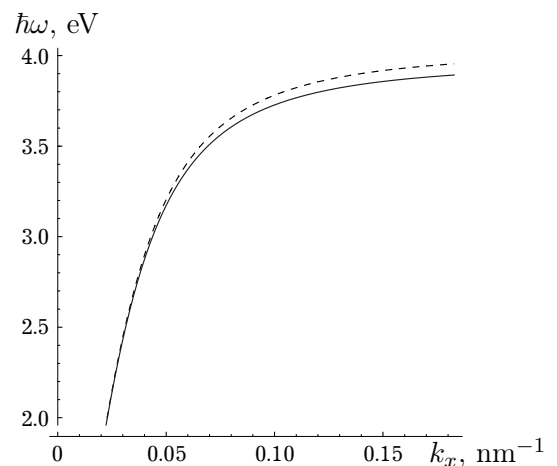


Fig. 3. SPPs spectrum for the Drude model $\varepsilon^D(\omega)$ (dotted line) and $\varepsilon_2(L, \omega)$ (solid line).

Fig. 3 shows the result of spectrum calculations obtained from (35) using (29), (30). These results we compared to the data obtained for the Drude model with negligible damping [1]

$$\varepsilon(L, \omega) = \varepsilon^D(\omega) = 1 - \frac{\omega_p^2}{\omega} \quad (38)$$

in which the plasmon frequency spectrum does not depend on the thickness L .

In Table 1 the results of numerical calculations of the dependence of frequency $\omega^* = \frac{\omega}{\omega_p}$, and a wave vector $k_x^* = \frac{k_x c}{\omega_p}$ on the number of levels of dimensional quantization n_{\max} are provided [12].

Table 1.

L (nm)	$\varepsilon_2(L, \omega^*)$	$\varepsilon^D(\omega^*)$	n_{\max}
$k_x^* = 1$			
100	0.33992328	0.34237082	335
1000	0.34016832	0.34237082	3357
5000	0.34019295	0.34237082	16789
10000	0.34019603	0.34237082	33579
30000	0.34019749	0.34237082	100737
$k_x^* = 2$			
100	0.40974355	0.41421356	335
1000	0.41021317	0.41421356	3357
5000	0.41025793	0.41421356	16789
10000	0.41026353	0.41421356	33579
30000	0.41026618	0.41421356	100737
$k_x^* = 3$			
100	0.42679347	0.43187178	335
1000	0.42732626	0.43187178	3357
5000	0.42737706	0.43187178	16789
10000	0.42738341	0.43187178	33579
30000	0.42738642	0.43187178	100737
$k_x^* = 4$			
100	0.43313236	0.43844718	335
1000	0.43368968	0.43844718	3357
5000	0.43374282	0.43844718	16789
10000	0.43374947	0.43844718	33579
30000	0.43375261	0.43844718	100737

The data in the table show that difference between spectra $\omega(k_x)$ for both models increases with increasing of the wave vector k_x . Also, it should be noted that with increasing of L , $\omega(k_x)$ is steadily approaching from below the values obtained for the Drude model.

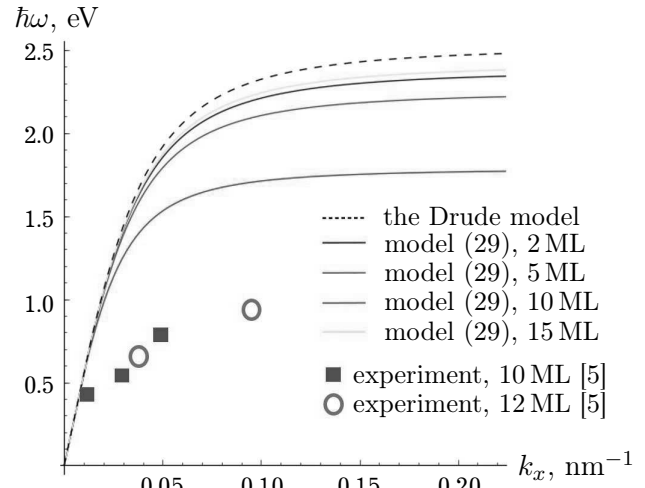


Fig. 4. SPPs spectrum for the Drude model $\varepsilon^D(\omega)$ (dotted line) and $\varepsilon_2(L, \omega)$ for the different thicknesses of a metal layer (solid lines), 1 ML ~ 0.24 nm.

In Fig. 4, the results of calculations for the structure “Si–silver–SiO₂” are shown. The dielectrics were simulated for the following parameters: $U_1 = 5$ eV, $\varepsilon_1 = 2.4$ and $U_1 = 4.8$ eV, $\varepsilon_1 = 11.7$ for Si and SiO₂ correspondingly [1].

The same figure shows the experimental results for the structure “Si–silver–SiO₂” published in [5]. These results demonstrate that the spectrum of plasmons strongly depends on the thickness of a metal layer when $L \sim 50$ ML. As can be seen from Fig. 4, the proposed approach gives a qualitative explanation of the influence of a metal film on the SPPs spectrum.

Note that the results obtained in [5] for the SPPs spectrum were obtained for structures that consist of 2–15 monolayers, thus simulation of $\varepsilon_2(\mathbf{q}, z, z', \omega)$ should be carried out for such thicknesses. In such metal structures, quantum effects become significant [9, 10, 13] and they should be taken into account.

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Поверхневі плазмон–поляритони в структурах “діелектрик–метал–діелектрик”: вплив товщини металевого прошарку

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Запропоновано та досліджено модель, яка дає змогу пояснити експериментальні дані щодо впливу товщини металевого прошарку на спектр SPP хвиль у гетерогенних структурах “діелектрик–метал–діелектрик”.

Ключові слова: *поверхневі плазмони, спектр плазмона, товщина металевого шару, діелектрична проникність.*

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