

SPP waves in "dielectric-metal-dielectric" structures: influence of exchange correlations

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In this paper the results of investigation of the spectrum and the propagation length of SPP waves in the structures synthesized by the authors and the results of mathematical modeling of these characteristics are presented. It is shown that taking into account the simplest (interchange) interactions of metal layer electrons leads to considerable changes in the behaviour of the spectrum of SPP waves and their propagation length.

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1. Introduction

Surface plasmon-polariton waves (SPP waves) (i.e., electron density undulation) exist in the vicinity of the interface between a dielectric medium and a metal [1-3]. A free electron plasma provides the electromagnetic field confinement near the metal–dielectric interface of metals, making the real part of the metal dielectric permittivity negative in a wide range of frequencies up to the near-ultraviolet domain.

State-of-art approaches to mathematical modeling of propagation of SPP waves are based on the classical Drude theory [2, 3] which, as is known [4, 5], does not consider spatial dispersion of metal dielectric permittivity $\varepsilon(\mathbf{r}, t)$ and effects caused by the interaction of electrons in a metal layer. Those effects become especially significant (and in the same time complicated for modeling) in a low-scale (nanoscale) metallic systems which are used in "dielectric–metal–dielectric" structures (DMD structures) synthesis [6, 7].

An attempt of investigation of an electronic correlations influence on SPP waves characteristics for "dielectric–graphene–dielectric" structures was made in [8], where it was shown that the consideration of electronic correlations for 2D-dimensional graphene layers leads to the change of $\omega(\mathbf{Q})$ spectrum behaviour of SPP waves even in the case of a small ($|\mathbf{Q}| \ll k_F^{-1}$) wavevector. This confirms (domain $\mathbf{Q} \to 0$ is the domain of the Drude model correctness) the necessity of mathematical modeling describing SPP waves that would take into account anisotropy and electronic correlations effects.

In the paper there is considered the problem of mathematical modeling of SPP waves propagation in a layered DMD structure synthesized by the authors (Pavlysh, Nevinskyi) in the case of modeling the metal layer by two-dimensional electronic plasma [9]. It is shown that taking into account only the exchange correlations leads to considerable changes in the spectrum $\omega(\mathbf{Q})$ as well as in the propagation length of SPP wave.

2. Experiment

Onto the previously cleared glass (SiO₂) substrate in a deep vacuum (10^{-8} Torr) there was deposited a golden film of the controlled thickness 50 nm. The obtained structure "dielectric–metal" was covered with a polymer (layer thickness 1550 nm) and was illuminated (for excitation of SPP wave) by a 350

300

250 200

150

100

50

0

Intensity [arb. units]

laser with a wavelength of 632.5 nm and a pulse frequency of 250 fs. Schematic representation of the experiment is shown in Fig. 1.



Fig. 1. Schematic representation of SPP excitation.



Real Image

 $exp(-x/L_{SPP})$, $L_{SPP} = 20 \mu m$

20

Fig. 3. The intensity of SPP propagation along the waveguide. The solid curve corresponds to the experimental data, the dashed curve - to

the theoretical calculation of SPP attenuation.

30

x-direction [µm]



Fig. 2 shows the image of SPP wave displayed on LR microscope.

For 150 nm width of the waveguide, the length of SPP propagation was $10\,\mu m$ and for $200 \,\mathrm{nm} - 20 \,\mu\mathrm{m}$. Fig. 3 presents the result of SPP wave measurement depending on the distance to the excitation point.

Such experiments allow us to state that propagation length of SPP wave is $\sim 20 \,\mu \text{m}$.



For describing processes of SPP waves emergence and propagation on the surface of nanoscale film covered with dielectrics on top and on bottom ("dielectric-metal-dielectric" structure), we will use the model shown in Fig.4; $\Omega = \Omega_1 \cup \Omega_2 \cup \Omega_3$, where $\Omega_1 = \mathbb{R}^2 \times (+\delta, +\infty)$, $\Omega_2 = \mathbb{R}^2 \times [-\delta, +\delta]$, $\Omega_3 = \mathbb{R}^2 \times (-\delta, -\infty).$

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As initial relationships for the construction of mathematical model, we will use the system of Maxwell's equations [4, 5]

$$\operatorname{rot} \boldsymbol{H}(\boldsymbol{r},t) = \frac{\partial \boldsymbol{D}(\boldsymbol{r},t)}{\partial t} + \boldsymbol{j}(\boldsymbol{r},t), \quad \operatorname{rot} \boldsymbol{E}(\boldsymbol{r},t) = \frac{\partial \boldsymbol{B}(\boldsymbol{r},t)}{\partial t}, \quad (1)$$
$$\operatorname{div} \boldsymbol{D}(\boldsymbol{r},t) = \rho(\boldsymbol{r},t), \quad \operatorname{div} \boldsymbol{B}(\boldsymbol{r},t) = 0,$$

where $D(\mathbf{r},t)$ is electric flux density, $E(\mathbf{r},t)$ is electric field, $B(\mathbf{r},t)$ is magnetic flux density, $H(\mathbf{r},t)$ is magnetic field strength, $\rho(\mathbf{r},t)$ is electric charge density, $j(\mathbf{r},t)$ is electric current density, $(x, y, z) \in \mathbb{R}^3$, $t \in [0,\infty)$ is time.

We assume that interconnection between the vectors of strength Hand E and between B and D electric flux density vectors of electromagnetic field is non-local [4,5], namely

$$D(\mathbf{r},t) = \int_{\Omega} d\mathbf{r}' \int_{t} dt' \varepsilon(\mathbf{r},\mathbf{r}',t-t') E(\mathbf{r}',t'),$$

$$B(\mathbf{r},t) = \mu_{0} \mu H(\mathbf{r},t), \quad \mu = 1,$$
(2)

 μ_0 is magnetic permeability of vacuum, $\varepsilon(\mathbf{r}, \mathbf{r}', t - t')$ is the response function (relative permittivity).

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50

40

2D Metal σ(q,ω) Fig. 4. Schematic representation of "dielectric-metal-di-

electric" structure.

The dependence between current density $\boldsymbol{j}(\boldsymbol{r},t)$ and electric field vector $\boldsymbol{E}(\boldsymbol{r},t)$ is similar to (2)

$$\boldsymbol{j}(\boldsymbol{r},t) = \int_{\Omega} d\boldsymbol{r}' \int_{t} dt' \sigma(\boldsymbol{r},\boldsymbol{r}',t-t') E(\boldsymbol{r}',t'), \qquad (3)$$

 $\sigma(\mathbf{r}, \mathbf{r}', t - t')$ is dynamic conductivity.

We will search for a solution of the system (1) for so called TM-waves [2,3], i.e. we assume that

$$H = (0, H_y, 0), \quad E = (E_x, 0, E_z)$$

For TM-waves $\frac{\partial H_y}{\partial y} \equiv 0$ [2,3], therefore $H_y = H(x, z, t)$. This means that for our model (2) (it is easy to show) $E_x = E_x(x, z, t)$, $E_z = E_z(x, z, t)$, thus the electric field is uniform along OY.

Let us introduce into the model the assumptions about the structure of the functions $\varepsilon(\mathbf{r}, \mathbf{r}', t-t')$ and $\sigma(\mathbf{r}, \mathbf{r}', t-t')$. We assume that in domains Ω_1 and Ω_3 (dielectrics)

$$\varepsilon_i(\boldsymbol{r}, \boldsymbol{r}', t - t') \cong \varepsilon_0 \,\varepsilon_i(t - t') \,\delta(\boldsymbol{r} - \boldsymbol{r}'), \quad i = 1, 3, \tag{4}$$

where ε_0 is the vacuum permittivity. This means that in the dielectric environment we will neglect the spatial dispersion and will take into account only the frequency dispersion [4,5]. Since in dielectrics free carriers of charge are absent we will assume that in this domains $\sigma(\mathbf{r}, \mathbf{r}', t - t') \equiv 0$.

For describing the electromagnetic field behavior in the domain Ω_2 (metal nanoscale film), the functions $\varepsilon(\mathbf{r}, \mathbf{r}', t - t')$ and $\sigma(\mathbf{r}, \mathbf{r}', t - t')$ we will model by functions of 2D-electron liquid [8, 9], namely, we will assume that

$$\varepsilon_{2}(\boldsymbol{r},\boldsymbol{r}',t-t') \cong \varepsilon_{0}\varepsilon_{2}(\boldsymbol{r}_{||}-\boldsymbol{r}_{||}',t-t')\delta(z-z'),$$

$$\sigma_{2}(\boldsymbol{r},\boldsymbol{r}',t-t') \cong \sigma_{0}\sigma_{2}(\boldsymbol{r}_{||}-\boldsymbol{r}_{||}',t-t')\delta(z-z'),$$
(5)

 $r_{||}$ is a component of the radius vector r that lies in XOY, i.e. $r = (r_{||}, z)$.

Under such assumptions we are neglecting the dispersion ε_2 and σ_2 along OZ, which is, generally speaking, true for metal films with the thickness 2δ that satisfies the condition $2\delta k_F \sim 1 \div 10$, where $k_F = (\frac{3}{4\pi})^{\frac{2}{3}} r_s a_0^{-1}$, r_s is Gell-Mann-Brakner parameter, a_0 is Bohr raduis [6,7] and the influence of fringe effects in the XOY plane (this is true when geometric dimensions of a metal film in XOY are considerably larger than its thickness along OZ, $2\delta(\sqrt{S})^{-1} \ll 1$, where S is an area of the metal film surface).

For the further analysis of the system of Maxwell's equations it is convenient to write it down for Fourier components of H, E, B, D vectors (all the characteristics of the model is homogeneous with respect to time), by defining the Fourier transform with respect to time as [10]

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

we have:

• domain Ω_1 :

$$\operatorname{rot} \boldsymbol{H}(\boldsymbol{r},\omega) = i\omega\varepsilon_0\tilde{\varepsilon}_1(\omega)\boldsymbol{E}(\boldsymbol{r},\omega),\tag{7}$$

• domain Ω_3 :

$$\operatorname{rot} \boldsymbol{H}(\boldsymbol{r},\omega) = i\omega\varepsilon_0\tilde{\varepsilon}_3(\omega)\boldsymbol{E}(\boldsymbol{r},\omega),\tag{8}$$

• domain Ω_2 :

$$\operatorname{rot} \boldsymbol{H}(\boldsymbol{r},\omega) = i\omega\varepsilon_0 \int_{\Omega_2} d\boldsymbol{r}_{||}'\tilde{\varepsilon}_2(\boldsymbol{r}_{||} - \boldsymbol{r}_{||}',\omega) \boldsymbol{E}(\boldsymbol{r}_{||}',\omega,z) + \sigma_0 \int_{\Omega_2} d\boldsymbol{r}_{||}'\tilde{\sigma}_2(\boldsymbol{r}_{||} - \boldsymbol{r}_{||}',\omega) \boldsymbol{E}(\boldsymbol{r}_{||}',\omega,z).$$
(9)

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For the domain Ω_2 (9) it is also convenient to define the Fourier transform with respect to $r_{||}$ that allows us to write (9) as follows:

$$\operatorname{rot} \boldsymbol{H}(\boldsymbol{r}_{||},\omega) = \frac{i\omega\varepsilon_0}{(2\pi)^2} \int_{\Omega_2} d\boldsymbol{q}\tilde{\varepsilon}_2(\boldsymbol{q},\omega) e^{i(\boldsymbol{q},\boldsymbol{r}_{||}')} \boldsymbol{E}(\boldsymbol{q},\omega,z) + \frac{\sigma_0}{(2\pi)^2} \int_{\Omega_2} d\boldsymbol{q}\tilde{\sigma}_2(\boldsymbol{q},\omega) e^{i(\boldsymbol{q},\boldsymbol{r}_{||}')} \boldsymbol{E}(\boldsymbol{q},\omega,z), \quad (10)$$

where $\boldsymbol{q} = (q_x, q_y), \, d\boldsymbol{q} = (dq_x, dq_y), \, (\boldsymbol{q}, \boldsymbol{r}_{||}) \equiv q_x x + q_y y.$

For describing of SPP waves propagation we will search solutions of the system (7)-(10) in the form of the following "ansatz"

$$H_y(x,\omega,z) = H_y(z)e^{iQx}.$$
(11)

It is obviuos that in this case

$$\boldsymbol{E}(\boldsymbol{r},\omega) = \boldsymbol{E}(x,\omega,z) = \boldsymbol{E}(\omega,z)e^{i\boldsymbol{Q}x}.$$
(12)

Since for such "ansatz"

$$\boldsymbol{E}(\boldsymbol{q},\omega,z) = (2\pi)^2 \boldsymbol{E}(\omega,z) \delta(Q-q_x) \delta(-q_y),$$

 $(\delta(\ldots))$ is the Dirac delta function [10]), then the system of equations (7)–(10) takes the form

$$\operatorname{rot} \boldsymbol{H}(Q,\omega,z) = i\omega\varepsilon_0\tilde{\varepsilon}_2(\omega)\boldsymbol{E}(Q,\omega,z), \quad j = 1,3,$$
(13)

$$\operatorname{rot} \boldsymbol{H}(Q,\omega,z) = i\omega\varepsilon_0\tilde{\varepsilon}_2(Q,\omega)\boldsymbol{E}(Q,\omega,z) + \tilde{\sigma}(Q,\omega)\boldsymbol{E}(Q,\omega,z).$$
(14)

Complementing the system (13), (14) with the Helmholtz equations [5] for $H(\omega, z)$ (which are easy to obtain for such a layered structure) we have a mathematical model of SPP waves propagation:

• domain Ω_1 :

$$\frac{d^{2}H_{y}(\omega,z)}{dz^{2}} - \left(Q^{2} - \tilde{\varepsilon}_{1}(\omega)\frac{\omega^{2}}{c^{2}}\right)H_{y}(\omega,z) = 0,$$

$$E_{x}(\omega,z) = -\frac{i}{\omega\varepsilon_{0}\tilde{\varepsilon}_{1}(\omega)}\frac{\partial H_{y}(\omega,z)}{\partial z},$$

$$E_{z}(\omega,z) = \frac{Q}{\omega\varepsilon_{0}\tilde{\varepsilon}_{1}(\omega)}H_{y}(\omega,z);$$
(15)

• domain Ω_3 :

$$\frac{d^{2}H_{y}(\omega,z)}{dz^{2}} - \left(Q^{2} - \tilde{\varepsilon}_{3}(\omega)\frac{\omega^{2}}{c^{2}}\right)H_{y}(\omega,z) = 0,$$

$$E_{x}(\omega,z) = -\frac{i}{\omega\varepsilon_{0}\tilde{\varepsilon}_{3}(\omega)}\frac{\partial H_{y}(\omega,z)}{\partial z},$$

$$E_{z}(\omega,z) = \frac{Q}{\omega\varepsilon_{0}\tilde{\varepsilon}_{3}(\omega)}H_{y}(\omega,z);$$
(16)

• domain Ω_2 :

$$\frac{d^{2}H_{y}(\omega,z)}{dz^{2}} - \left(Q^{2} - \gamma(Q,\omega,0)\frac{\omega^{2}}{c^{2}}\right)H_{y}(\omega,z) = 0,$$

$$-\frac{\partial H_{y}(\omega,z)}{\partial z} = E_{x}(\omega,z)\left(i\omega\varepsilon_{0}\tilde{\varepsilon}_{2}(Q,\omega,0) - \sigma_{0}\tilde{\sigma}_{2}(Q,\omega,0)\right),$$

$$H_{y}(\omega,z) = E_{z}(\omega,z)\left(i\omega\varepsilon_{0}\tilde{\varepsilon}_{2}(Q,\omega,0) - \sigma_{0}\tilde{\sigma}_{2}(Q,\omega,0)\right).$$
(17)

The system of equations (15)-(17) should be solved using the corresponding electrodynamic joining conditions at the boundary of environments: continuity of the corresponding components of H and D

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vectors. For the considered in this paper an ultrathin metal film $2\delta k_F \sim 1 \div 10$, we obtain that:

$$H_{y}^{(1)}(\omega, z) = A_{1}e^{-\lambda z}, \quad E_{x}^{(1)}(\omega, z) = \frac{\lambda}{\omega\varepsilon_{0}\tilde{\varepsilon}_{1}(\omega)}A_{1}e^{-\lambda z}, \quad E_{z}^{(1)}(\omega, z) = \frac{Q}{\omega\varepsilon_{0}\tilde{\varepsilon}_{1}(\omega)}A_{1}e^{-\lambda z}, \quad \lambda = \sqrt{Q^{2} + \frac{\omega^{2}}{c^{2}}\tilde{\varepsilon}_{1}(\omega)}, \quad (18)$$

$$H_{y}^{(3)}(\omega, z) = A_{1}e^{\xi z}, \quad E_{x}^{(3)}(\omega, z) = \frac{\lambda}{\omega\varepsilon_{0}\tilde{\varepsilon}_{3}(\omega)}B_{3}e^{\xi z}, \quad E_{z}^{(3)}(\omega, z) = \frac{Q}{\omega\varepsilon_{0}\tilde{\varepsilon}_{3}(\omega)}B_{3}e^{\xi z}, \quad \xi = \sqrt{Q^{2} + \frac{\omega^{2}}{c^{2}}\tilde{\varepsilon}_{3}(\omega)}, \quad (19)$$

The constants A_1 and B_3 we will obtain from the joining conditions of the solutions (18), (19) at the boundary z = 0, taking into account that at this boundary there exists the surface current $j_x = \sigma_0 \sigma(Q, \omega) \tilde{E}_x(Q, \omega, 0)$, where \tilde{E}_x is electric field in the plane z = 0. From this condition it follows that

 $c^2 = \varepsilon_0 \mu_0.$

$$H_{y}^{(1)}(\omega, +0) - H_{y}^{(3)}(\omega, -0) = j_{x},$$

$$\varepsilon_{0}\tilde{\varepsilon}_{1}(\omega)E_{y}^{(1)}(\omega, +0) = \varepsilon_{0}\tilde{\varepsilon}_{3}(\omega)E_{y}^{(3)}(\omega, -0) = \rho_{x},$$

$$E_{z}^{(1)}(\omega, +0) = E_{z}^{(3)}(\omega, -0).$$
(20)

Non-zero solution of the equation system (20) exists only if the following condition is fulfilled

$$\frac{\tilde{\varepsilon}_1(\omega)}{\lambda} + \frac{\tilde{\varepsilon}_3(\omega)}{\xi} = -\frac{1}{\omega\varepsilon_0}\sigma(Q,\omega),\tag{21}$$

which is dispersion relation for finding $\omega(Q)$ of SPP wave in this model. Notice that the dispersion equation (21) coincides with the dispersion equation of the SPP wave spectrum obtained in [8] for the case of "dielectric–graphene–dielectric" model.

4. Solution of dispersion equation

To solve the equation (21), a two-dimensional dynamic conductivity of an electron liquid $\sigma(Q, \omega)$ should be set. In the paper, solutions of (21) for two well-known in literature models of $\sigma(Q, \omega)$ are investigated:

• the Drude model [2]:

$$\sigma_0 \sigma(Q, \omega) = \frac{e^2 \varepsilon_F}{\pi \hbar^2} \frac{i}{\omega + i\tau^{-1}};$$
(22)

• the RPA model [9]:

$$\sigma(Q,\omega) = -i\omega\chi(Q,\omega), \quad \chi(Q,\omega) = \chi_1(Q,\omega) + i\chi_2(Q,\omega), \tag{23}$$

$$\chi_{1} = G\left(\frac{Q}{k_{F}} - C_{-}\left(\left(\frac{Q}{2k_{F}} - \frac{\omega}{Qv_{F}}\right)^{2} - 1\right)^{\frac{1}{2}} - C_{+}\left(\left(\frac{Q}{2k_{F}} + \frac{\omega}{Qv_{F}}\right)^{2} - 1\right)^{\frac{1}{2}}\right),$$

$$\chi_{2} = G\left(D_{-}\left(1 - \left(\frac{Q}{2k_{F}} - \frac{\omega}{Qv_{F}}\right)^{2}\right)^{\frac{1}{2}} - D_{+}\left(1 - \left(\frac{Q}{2k_{F}} + \frac{\omega}{Qv_{F}}\right)^{2}\right)^{\frac{1}{2}}\right),$$

$$G = \frac{Ne^{2}}{\varepsilon_{0}mq^{2}v_{F}^{2}}, \quad v_{F} = \frac{\hbar k_{F}}{m},$$
(24)

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$$C_{\pm} = \frac{\left(\frac{Q}{2k_F} \pm \frac{\omega}{Qv_F}\right)}{\left|\frac{Q}{2k_F} + \frac{\omega}{Qv_F}\right|}, \quad D_{\pm} = 0 \quad \text{for} \quad \left|\frac{Q}{2k_F} + \frac{\omega}{Qv_F}\right| > 1$$
$$C_{\pm} = 0, \quad D_{\pm} = 1 \quad \text{for} \quad \left|\frac{Q}{2k_F} + \frac{\omega}{Qv_F}\right| < 1.$$

In the given expressions (22)–(24) for dynamic conductivity k_F is the Fermi momentum, $\varepsilon_F = \frac{\hbar^2 k_F^2}{2m}$ is the Fermi energy, v_F is the Fermi velocity, \hbar is reduced Plank constant, m, e are the mass and the charge of electron respectively (see [11]), τ^{-1} is the reverse dumping time [8], $\tau = 1.3 \cdot 10^{-13}$ [3].

Numerical analysis of the equation was carried out using "MATLAB" for layered structure "polyethylene–gold–SiO₂" with parameters:

$$\varepsilon_1(\omega) = \tilde{\varepsilon}_1(+\infty) = 2.3, \quad \varepsilon_3(\omega) = \tilde{\varepsilon}_3(-\infty) = 4, \quad k_F a_0 = \left(\frac{3}{4\pi}\right)^{\frac{2}{3}} r_s, \quad r_s = 2.9.$$
(25)

The results of numerical calculation of $\omega = \omega(Q)$ are presented in Figs. 5–7.



Fig. 5. Graphics of dispersion $\omega(Q)$ for the Drude model (dashed curves) and for the RPA model (solid curves).



Fig. 6. Imaginary part $Im(\omega(Q))$ of disperse relation solution for the Drude model.

Fig. 7. Imaginary part $\text{Im}(\omega(Q))$ of disperse relation solution for the RPA model.

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As we can see in Fig. 5, the difference between the Drude and the RPA models becomes considerable when the wavevector Q in the domain is $\frac{|Q|}{k_F} \sim 0.05$. This means that for describing $\varepsilon(\mathbf{r}, t)$ and $\sigma(\mathbf{r}, t)$ other models should be used instead of the Drude model. A similar result was obtained for "dielectric–graphene–dielectric" structures [8].

Figs. 6, 7 present the dependence of the imaginary part of wavevector $\frac{\text{Im}(Q)}{k_F}$ on $\frac{Q}{k_F}$. This characteristic can be interpreted as inverse propagation length of SPP (L_{SPP}^{-1}) . For the Drude model $L_{SPP} \sim 0.2 \,\mu\text{m}$ and for the RPA model $L_{SPP} \sim 0.003 \,\mu\text{m}$. Decreasing of propagation length for the RPA model is understandable because this model takes into account the plasmon scattering effects connected with interchange interactions (Landau damping) [8]. The propagation length $L_{SPP} \sim 20 \,\mu\text{m}$ is obtained experimentally due to a laser illumination.

5. Conclusions

Being obtained as a result of mathematical modeling the dependences of the spectrum $\omega(Q)$ and the propagation length L_{SPP} of the plasmon-polariton wave show that for theoretical investigation of these characteristics we should take into account both interaction effects of electrons in a metal layer and anisotropy effects of a metal film in DMD structures. This will be a subject of the future researches.

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SPP-хвилі в структурах «діелетрик-метал-діелектрик»: вплив обмінних кореляцій

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Подано результати дослідження спектра та довжини поширення SPP-хвиль в синтезованих авторами структурах та результати математичного моделювання цих характеристик. Показано, що врахування найпростіших (обмінних) взаємодій електронів металевого прошарку приводить до значних змін у поведінці спектра SPP-хвиль та довжини їхнього поширення.

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