# Optical Absorption of a Composite Based on Two-Layer Nanowires

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The optical properties of two-layer nanowires are analyzed within the classical Drude theory. The expressions and frequency dependences of the real and imaginary parts of the dielectric function for metal oxide nanowires and nanoshells are calculated. The optical absorption of a composite on the basis of both approaches is investigated. It is shown that the variation of the thickness of both the core and the shell leads to a change in the magnitude and position of the optical characteristics maxima of specified systems. The presence of two maxima of the absorption coefficient of the composite based on metal nanoshells is demonstrated. Splitting into two resonant peaks is due to the hybridization of surface plasmons, which depends on the inner and outer radii of the shell. The influence of the metal layer final thickness on the frequency dependences of the absorption coefficient is analyzed. Calculations are performed for metals Au, Ag and Al, which are in different dielectric matrices. It is established that the behavior of the optical absorption maxima of the composite is determined by the dielectric constant of the matrix.

**Keywords**: Metal oxide nanowire, Nanoshell, Composite, Dielectric function, Frequency dependence, Absorption coefficient.

## 1. INTRODUCTION

One-dimensional metal nanostructures attract considerable attention due to the possibility of their application as active components in the manufacture of nanoscale electronic, biosensor and optical devices [1-4]. Additionally, such systems are used as models in physical phenomena studies such as Raman scattering [5], surface-enhanced fluorescence [6] and resonant light scattering [7], which are based on the surface plasmon resonance phenomenon. The spectral position of plasmon resonance directly depends on the size, shape, degree of interaction of particles and the dielectric properties of the local environment in which the particles are [8, 9]. By applying a dielectric coating, the properties of the local environment around the nanostructures are changed. As a result, one of the areas of one-dimensional systems study is composite structures such as core-shell, which are synthesized using semiconductors, metals and dielectrics [10]. By changing the material of the shell or core without changing the total radius of the nanowires, it is possible to control the electrical and optical properties from the visible to the infrared regions of the electromagnetic spectrum.

The optical properties of the core-shell type structure are studied by researchers experimentally [11] and theoretically [12, 13] due to their unique physicochemical properties. Technologies for surface coating and control of the thickness of the oxide layer on the surface of one-dimensional structures have been developed [14, 15]. The light scattering on finite arrays of metal nanowires with a dielectric coating has been numerically investigated by the method of multiple scattering [12]. An experimental study of the polarization light scattering on homogeneous and multiseg-mental Ag, Au, and Ni nanowires with a diameter of ~ 30 nm and a length of ~ 7 μm is presented in [11]. In [13], the interaction of light with single and multiple silver nanowires of nanometer size, covered with a dielectric shell, was studied by the method finite-differences time-domain. The presence of the oxide layer shifts the position of the plasmon resonance to the red with increase of dielectric thickness.

On the other hand, structures which consist of a dielectric or hollow core surrounded by a metal wall, called nanotubes, are studied. Hybridization of surface plasmon resonances due to excitation on the inner and outer walls of the nanotube gives the possibility of more flexible regulation of optical characteristics by changing the thickness of the metal shell [16, 17]. The effect of changing the radius of a metal nanocylinder with a dielectric core on the dispersion of surface plasmons is considered in [18]. In [19], the formation of nanowires of $SiO_2$ and $TiO_2$ with a length of ~ 10 μm and a diameter of 90-200 nm, covered with a thin shell of Au, is discussed. It is shown that by varying the diameter of the oxide core and the thickness of the Au coating, it is possible to adjust the optical properties of such structures [19]. An approach for obtaining, synthesizing and controlling the walls of hollow nanostructures Au, Pt and Pd is given in [20].

In this article, on the basis of simple relations, the influence of various factors on the frequency dependences of the real and imaginary parts of the dielectric function of two-layer metal oxide nanowires and nanoshells is considered, which are further used to study optical light absorption by composites based on them. The approach adapted in [21] for composites based on two-layer spherical nanoparticles is used to calculate the absorption coefficient.

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## 2. BASIC RELATIONS

Consider a system in the form of a two-layer nanowire with a radius $R = R_c + t$ (where $R_c$ is the core radius; $t$ is the shell thickness), located in a medium with dielectric constant $\varepsilon_m$. The polarizability of such a system is found by solving the Laplace equation in cylindrical coordinate system

$$\frac{\partial^2 \varphi_i}{\partial r^2} + \frac{1}{r}\frac{\partial \varphi_i}{\partial r} = 0 \quad (i = 1, 2, 3), \tag{1}$$

taking into account the boundary conditions at the interface

$$\varphi_1\big|_{r=R_c} = \varphi_2\big|_{r=R_c}; \qquad \varphi_2\big|_{r=R} = \varphi_3\big|_{r=R};$$

$$\varepsilon_c \frac{\partial \varphi_1}{\partial r}\bigg|_{r=R_c} = \varepsilon_{sh}\frac{\partial \varphi_2}{\partial r}\bigg|_{r=R_c}; \qquad \varepsilon_{sh}\frac{\partial \varphi_2}{\partial r}\bigg|_{r=R} = \varepsilon_m\frac{\partial \varphi_3}{\partial r}\bigg|_{r=R},$$

where $\varepsilon_c$ and $\varepsilon_{sh}$ are the core and shell dielectric functions, respectively.

The solution of the Laplace equation has the form:

$$\varphi(r) = \begin{cases} Ar, & r < R_c; \\ Br + Cr^{-1}, & R_c \le r \le R; \\ -\mathscr{E}_0 r + Dr^{-1}, & r > R, \end{cases}$$

where $\mathscr{E}_0$ is the external electric field strength.

After substitution of the solutions of the Laplace equation into boundary conditions, we have a system of equations from which we obtain the following relation:

$$\frac{\varepsilon_c - \varepsilon_{sh}}{\varepsilon_c + \varepsilon_{sh}} = \frac{1}{\beta_c^2}\frac{(\varepsilon_m - \varepsilon_{sh}) + (\varepsilon_m + \varepsilon_{sh})\bar{\alpha}}{(\varepsilon_m + \varepsilon_{sh}) + (\varepsilon_m - \varepsilon_{sh})\bar{\alpha}},$$

whence for polarizability we will have:

$$\bar{\alpha} = \frac{(\varepsilon_c + \varepsilon_{sh})(\varepsilon_{sh} - \varepsilon_m) + \beta_c^2(\varepsilon_c - \varepsilon_{sh})(\varepsilon_{sh} + \varepsilon_m)}{(\varepsilon_c + \varepsilon_{sh})(\varepsilon_{sh} + \varepsilon_m) + \beta_c^2(\varepsilon_c - \varepsilon_{sh})(\varepsilon_{sh} - \varepsilon_m)}, \tag{2}$$

where $\beta_c = R_c/R = 1 - t/R \ (t/R \ll 1)$ is the volume content of the core material.

On the other hand, the polarizability of the 1D system is

$$\bar{\alpha} = \frac{\varepsilon_{c-sh} - \varepsilon_m}{\varepsilon_{c-sh} + \varepsilon_m}, \tag{3}$$

where $\varepsilon_{c-sh}$ is the dielectric constant of the two-layer nanowire.

Equating (2) and (3) we obtain:

$$\varepsilon_{c-sh} = \varepsilon_{sh}\frac{(\varepsilon_c + \varepsilon_{sh}) + \beta_c^2(\varepsilon_c - \varepsilon_{sh})}{(\varepsilon_c + \varepsilon_{sh}) - \beta_c^2(\varepsilon_c - \varepsilon_{sh})}. \tag{4}$$

The absorption coefficient of composite is determined by the ratio [22]:

$$\eta(\omega) = \frac{\omega}{3c}\gamma\varepsilon_m^{3/2}\sum_j \frac{1}{P_j^2}\frac{\operatorname{Im}\varepsilon_{c-sh}}{\left(\operatorname{Re}\varepsilon_{c-sh} + \dfrac{1-P_j}{P_j}\varepsilon_m\right)^2 + \operatorname{Im}\varepsilon_{c-sh}^2},$$

where $\gamma$ is the volume content of 1D systems; $c$ is the speed of light; $\omega$ is the frequency of the incident electromagnetic wave; $P_j$ is the depolarization factor; $j = x, y, z$.

Consider the case where the electromagnetic wave is incident on a cylindrical nanowire perpendicular to the wire axis. The wave vector lies in the $xy$ plane, as a result $P_z = 0, P_x = P_y = (1 - P_z)/2 = 1/2$. Then we finally have

$$\eta(\omega) = \frac{8\omega}{3c}\gamma\varepsilon_m^{3/2}\frac{\operatorname{Im}\varepsilon_{c-sh}}{(\operatorname{Re}\varepsilon_{c-sh} + \varepsilon_m)^2 + \operatorname{Im}\varepsilon_{c-sh}^2}. \tag{5}$$

**2.1** First consider a metal oxide nanowire which consists of a metal core covered with a thin dielectric layer. As a result, the expression (4) takes the form:

$$\varepsilon_{c-sh} = \varepsilon_{ox}\frac{(\varepsilon_{metal} + \varepsilon_{ox}) + \beta_c^2(\varepsilon_{metal} - \varepsilon_{ox})}{(\varepsilon_{metal} + \varepsilon_{ox}) - \beta_c^2(\varepsilon_{metal} - \varepsilon_{ox})},$$

where $\varepsilon_{ox}$ is the dielectric function of the oxide, $\varepsilon_{metal}$ is the Drude function:

$$\varepsilon_{metal}(\omega) = \varepsilon_1(\omega) + i\varepsilon_2(\omega) = \varepsilon_\infty - \frac{\omega_{pl}^2\tau^2}{1 + \omega^2\tau^2} + i\frac{\omega_{pl}^2\tau}{\omega(1 + \omega^2\tau^2)},$$

$\varepsilon_\infty$ is the component that describes the contribution of the ionic framework, $\omega_{pl}^2 = \dfrac{e^2 n_e}{\varepsilon_0 m^*}$ is the plasmon frequency, $\varepsilon_0$ is the electric constant, $n_e$ is the conductivity electron concentration, $m^*$ is the effective electron mass, $\tau$ is the relaxation time.

Substituting the Drude function and separating real and imaginary parts, we have

$$\operatorname{Re}\varepsilon_{c-sh} = \varepsilon_{ox}\frac{(\varepsilon_1^2 + \varepsilon_2^2 + \varepsilon_{ox}^2)(1 - \beta_c^4) + 2\varepsilon_1\varepsilon_{ox}(1 + \beta_c^4)}{\{\varepsilon_1(1 - \beta_c^2) + \varepsilon_{ox}(1 + \beta_c^2)\}^2 + \varepsilon_2^2(1 - \beta_c^2)^2};$$

$$\operatorname{Im}\varepsilon_{c-sh} = \varepsilon_{ox}^2\frac{4\beta_c^2\varepsilon_2}{\{\varepsilon_1(1 - \beta_c^2) + \varepsilon_{ox}(1 + \beta_c^2)\}^2 + \varepsilon_2^2(1 - \beta_c^2)^2}. \tag{6}$$

For the boundary case, when the thickness of the oxide layer $t \to 0$, we have $\beta_c = 1$ and, as a consequence, $\operatorname{Re}\varepsilon_{c-sh} = \varepsilon_1$ and $\operatorname{Im}\varepsilon_{c-sh} = \varepsilon_2$; when $t = R$ and $\beta_c = 0$ we obtain the dielectric constant of the oxide $\operatorname{Re}\varepsilon_{c-sh} = \varepsilon_{ox}$.

**2.2** For the case of a nanoshell that consists of a dielectric core covered with a thin metal layer, the formula for the dielectric function is:

$$\varepsilon_{c-sh} = \varepsilon_{metal} \frac{\left(\varepsilon_{ox} + \varepsilon_{metal}\right) + \beta_c^2 \left(\varepsilon_{ox} - \varepsilon_{metal}\right)}{\left(\varepsilon_{ox} + \varepsilon_{metal}\right) - \beta_c^2 \left(\varepsilon_{ox} - \varepsilon_{metal}\right)},$$

or after the separation of real and imaginary parts

$$\operatorname{Re}\varepsilon_{c-sh} = \frac{\varepsilon_1\left(\varepsilon_1^2 + \varepsilon_2^2 + \varepsilon_{ox}^2\right)\left(1-\beta_c^4\right) + 2\varepsilon_1^2\varepsilon_{ox}\left(1+\beta_c^4\right) + 4\varepsilon_2^2\varepsilon_{ox}\beta_c^2}{\left\{\varepsilon_1\left(1+\beta_c^2\right) + \varepsilon_{ox}\left(1-\beta_c^2\right)\right\}^2 + \varepsilon_2^2\left(1+\beta_c^2\right)^2};$$

$$\operatorname{Im}\varepsilon_{c-sh} = \varepsilon_2 \frac{\left(\varepsilon_1^2 + \varepsilon_2^2 + \varepsilon_{ox}^2\right)\left(1-\beta_c^4\right) + 2\varepsilon_1\varepsilon_{ox}\left(1-\beta_c^2\right)^2}{\left\{\varepsilon_1\left(1+\beta_c^2\right) + \varepsilon_{ox}\left(1-\beta_c^2\right)\right\}^2 + \varepsilon_2^2\left(1+\beta_c^2\right)^2}.$$

(7)

For the boundary case, when the thickness of the metal layer $t \to 0$, we have $\operatorname{Re}\varepsilon_{c-sh} = \varepsilon_{ox}$; when $t = R$ and $\beta_c = 0$ we get $\operatorname{Re}\varepsilon_{c-sh} = \varepsilon_1$ and $\operatorname{Im}\varepsilon_{c-sh} = \varepsilon_2$.

When the particle size is less than the wavelength of the electron, it is necessary to take into account the scattering of electrons on the surface of the 1D system, as a result of which the relaxation time $\tau$ is recorded through $\tau_{eff}$:

$$\frac{1}{\tau_{eff}} = \frac{1}{\tau_{bulk}} + A\frac{\upsilon_F}{R},$$

(8)

where $\tau_{bulk}$ is the relaxation time in 3D metal; $A$ is the effective parameter determined by electron scattering processes; $\upsilon_F$ is the Fermi velocity; $R$ is the radius of the metal component.

Usually, the constant $A$ for the sphere is taken equal to $A = 3/4$, or it is considered a parameter that needs to be adjusted. In [23], within the kinetic approach in describing the processes of electron scattering in volume and on the surface, expressions were obtained for the effective parameter $A$ for both spherical nanoparticles and for the case of oblate and prolate spheroids. For the form of a two-layer nanowire we use the results of [24], where the parameter $A$ based on the kinetic approach is calculated as

$$A = \frac{27\pi}{128\left(\varepsilon_m + 1\right)}\left(\frac{\omega_{pl}}{\omega}\right)^2.$$

Further, formulas (6)-(7) are used to calculate the dielectric function, taking into account expression (8) for the effective relaxation time.

## 3. CALCULATION RESULTS AND DISCUSSION

The calculations were performed for metal oxide nanowires and nanoshells Au, Ag and Al. The parameters of the metals are given in Table 1.

**Table 1** – Metal parameters ($a_0$ is the Bohr radius)

| Parameter \ Metal | Au | Ag | Al |
|---|---|---|---|
| $r_s/a_0$ | 3.01 | 3.02 | 2.07 |
| $m^*/m_e$ | 0.99 | 0.96 | 1.48 |
| $\varepsilon_\infty$ | 9.84 | 3.7 | 1 |
| $\tau$, fs | 29 | 40 | 8 |

Fig. 1 shows the frequency dependences of the real $\operatorname{Re}\varepsilon_{c-sh}$ and imaginary $\operatorname{Im}\varepsilon_{c-sh}$ parts of the dielectric function calculated by formula (6) for Au nanowires covered with the layer $Al_2O_3$ $\left(\varepsilon_{ox} = 3.13\right)$. Increasing the radius $R_c$ of the metal core Au (curves 1-3 of Fig. 1) at a fixed value of the thickness $t = 5$ nm of the $Al_2O_3$ shell leads to an increase in the absolute value of the maxima of the real and imaginary parts of the dielectric function and shift them to lower frequencies. Increasing the thickness $t$ of the $Al_2O_3$ dielectric shell (curves 1'-3' in Fig. 1) at a fixed value of the radius $R_c = 20$ nm of the Au core leads to a decrease in the magnitude of the maxima $\operatorname{Re}\varepsilon_{c-sh}$ and $\operatorname{Im}\varepsilon_{c-sh}$ and their shift to the high frequency range. Fig. 1 demonstrates an important fact that $\operatorname{Im}\varepsilon_{c-sh}$ is nonnegative in the entire investigated frequency range, while $\operatorname{Re}\varepsilon_{c-sh}$ is an alternating function of frequency.
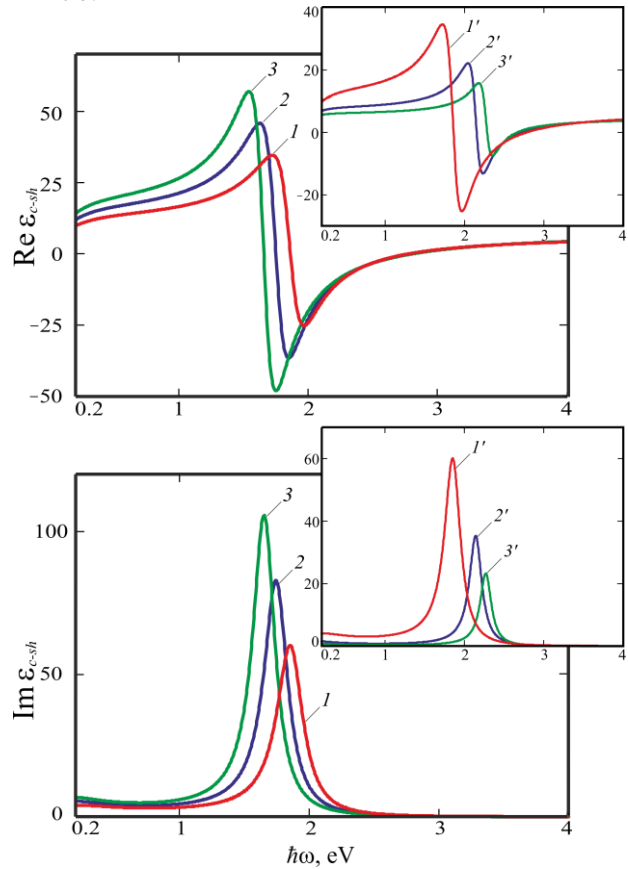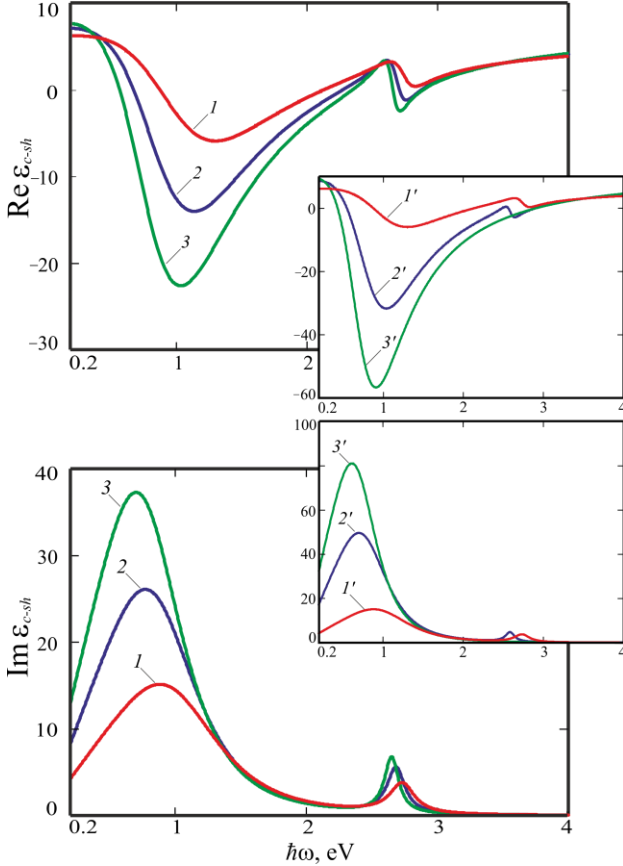


**Fig. 1** – Frequency dependences of the real and imaginary parts of the Au nanowires dielectric function of radius $R_c = 20(1,1',2',3')$, 25(2), 30(3) nm, covered with a layer of $Al_2O_3$ with a thickness of $t = 5(1,2,3,1')$, 10(2'), 15(3') nm

Fig. 2 shows the frequency dependences of the real and imaginary parts of the dielectric function, calculated by formula (7), for Au nanoshells with an $Al_2O_3$ core. An increase in the thickness $t$ of the metal shell (curves 1-3 of Fig. 2) at a fixed value of the radius $R_c = 20$ nm of the dielectric core leads to an increase in the maxima of the real and imaginary parts of the dielectric function and shift them to lower frequencies. Decreasing the content of the dielectric core at a fixed value of the radius $R = R_c + t = 30$ nm of the two-layer cylindrical sys-

tem leads to similar results (curves 1′-3′ of Fig. 2). In this case, formula (7) passes to the Drude function.

Let us estimate the order of magnitude $\mathrm{Im}\,\varepsilon_{c-sh}$ for $\hbar\omega = 0.6$ eV ($\omega = 9.12 \cdot 10^{14}$ s$^{-1}$). Taking into account that at $t = R = 30$ nm we have

$$\mathrm{Im}\,\varepsilon_{c-sh} = \frac{\omega_{pl}^2 \tau_{eff}}{\omega\left(1 + \omega^2 \tau_{eff}^2\right)}.$$



**Fig. 2** – Frequency dependences of the real and imaginary parts of the Au nanoshells dielectric function with thickness $t = 10(1, 1')$, $15(2)$, $20(3, 2')$, $30(3')$ nm with Al$_2$O$_3$ core with radius $R_c = 20(1, 2, 3, 1')$, $10(2')$, $0(3')$ nm
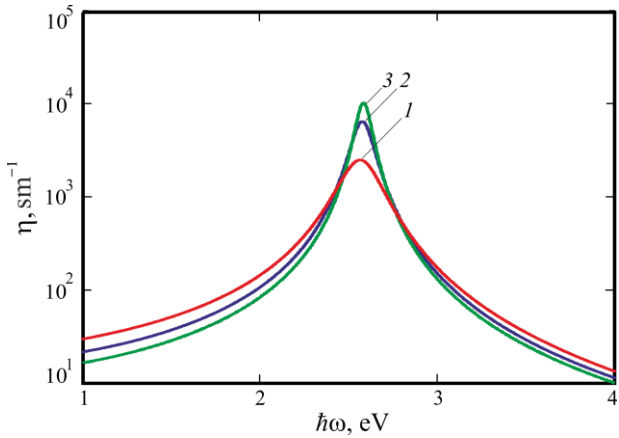


**Fig. 3** – Frequency dependences of the absorption coefficient of a composite based on Au nanowires with radius $R_c = 10(1)$, $20(2)$, $30(3)$ nm covered with a layer of Al$_2$O$_3$ with a thickness of $t = 5$ nm
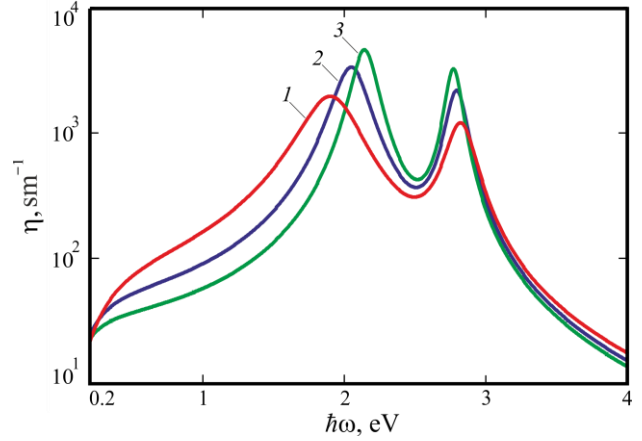


**Fig. 4** – Frequency dependences of the absorption coefficient of a composite based on Au nanoshells with a thickness of $t = 10(1)$, $15(2)$, $20(3)$ nm with an Al$_2$O$_3$ core with a radius of $R_c = 20$ nm
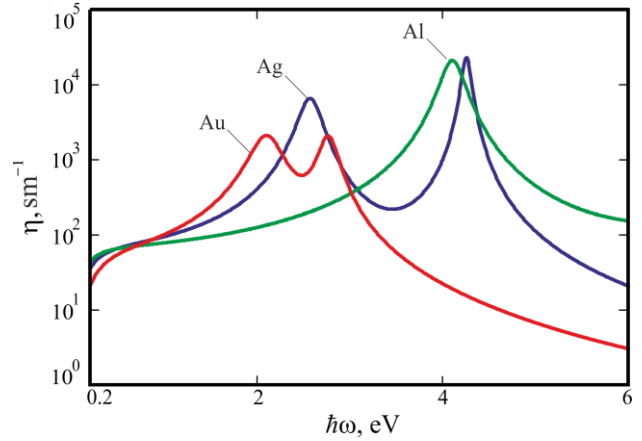


**Fig. 5** – Frequency dependences of the absorption coefficient of the composite based on nanoshells of different metals with a thickness of $t = 10$ nm, covered with a layer of Cu$_2$O with a radius of $R_c = 10$ nm.

Given for Au $n_e = 5.9 \cdot 10^{28}$ m$^{-3}$ and $m^* = 0.99 m_e$, we obtain $\omega_{pl} = 1.38 \cdot 10^{16}$ s$^{-1}$. Substituting in (8) $\upsilon_F = 1.4 \cdot 10^6$ m/s, $\tau_{bulk} = 29 \cdot 10^{15}$ s, $A = 45.9$, we have $\tau_{eff} = 4.6 \cdot 10^{-16}$ s. Then $\mathrm{Im}\,\varepsilon_{c-sh} \approx 80$, which agrees well with the calculated data presented in Fig. 2 (curve 3').

The frequency dependences of the absorption coefficient of the composite based on Au nanowires covered with a layer of Al$_2$O$_3$ with a thickness of $t = 5$ nm are shown in Fig. 3. Teflon $\left(\varepsilon_m = 2.3\right)$ is used as a matrix. Increasing the absolute value of the maxima of the absorption coefficient and shifting them to the high frequency range is directly related to the increase in the content of the metal core.

Fig. 4 shows similar dependences for composite based on Au nanoshells with an Al$_2$O$_3$ dielectric core. The main difference between the nanoshell and the case of a nanowire is the presence of two maxima in the frequency dependences of the absorption coefficient of the composite due to hybridization of plasmons [25]. Electromagnetic excitations of plasmons induce surface charges on the inner and outer surfaces of the metal shell. The interaction of plasmons with each other is

due to the final thickness of the metal layer. The strength of the plasmon interaction is determined by controlling the thickness of the nanoshell. As a result of the interaction, the plasmon resonance splits into a low-energy symmetric mode and a high-energy anti-symmetric mode.
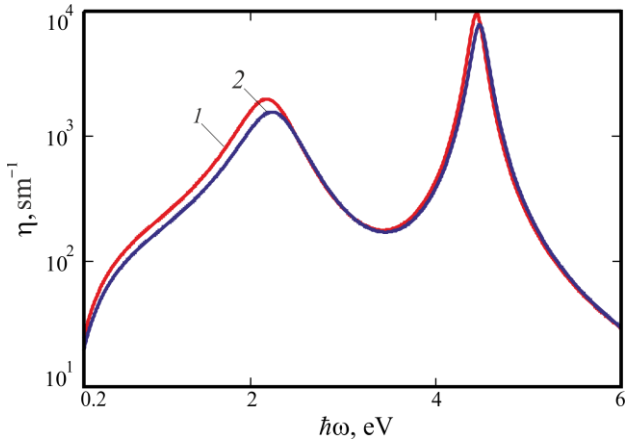


**Fig. 6** – Frequency dependences of the absorption coefficient of the composite based on Ag nanoshells with a thickness of $t = 5$ nm, covered with a layer of $Cu_2O$ with a radius of $R_c = 10$ nm, in different dielectric matrices: 1 – Teflon ($\varepsilon_m = 2.3$); 2 – Ar ($\varepsilon_m = 1.7$)

Hybridization of plasmons and splitting of resonant levels are quite significant for a small value of the shell thickness (curve 1, Fig. 4). Increasing the thickness of the metal shell brings the resonant peaks together due to the weakening of the interaction between plasmons.

The frequency dependences of the absorption coefficient for composite based on nanoshells of different metals at a fixed value of the shell thickness $t = 10$ nm and the radius $R_c = 10$ nm of the $Cu_2O$ ($\varepsilon_{ox} = 3.7$) core are shown in Fig. 5. The results of the calculations show qualitatively and quantitatively different type in

the position and magnitude of the peaks of the absorption coefficient of the composite of Au, Ag and Al nanoshells. This behavior is due to the different values of the contribution of the ionic core $\varepsilon_\infty$ and plasma frequency $\omega_{pl}$.

The influence of the dielectric matrix $\varepsilon_m$ on the absorption coefficient of the composite of Ag nanoshells with a $Cu_2O$ dielectric core is shown in Fig. 6. Increasing the dielectric constant of the matrix $\varepsilon_m$ leads to an increase in the maxima of the absorption coefficient and the shift of the maxima in the region of lower frequencies. As a result, the choice of dielectric matrix also makes it possible to control the optical properties of 1D two-layer systems.

## 4. CONCLUSIONS

The expressions for the real and imaginary parts of the dielectric function and the absorption coefficient of composite with two-layer nanowires are obtained. The evolution of frequency dependences of real and imaginary parts of dielectric function with radius variation of both core and shell for cases of metal oxide nanowires and nanoshells is analyzed. It is established that increasing the radius of the metal core of the metal oxide nanowire leads to an increase in the absolute value of the maximum absorption coefficient and its displacement in the high frequency range. The presence of two maxima of the metal nanoshell absorption coefficient due to hybridization of surface plasmons is shown. The magnitude and position of the resonant peaks directly depend on the metal shell thickness.

Calculations of the absorption coefficient were performed for the composite based on Au, Ag and Al nanowires. The differences in character of the frequency dependences of nanowires of various metals are due to different values of the contribution of the ionic core and plasma frequency.

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# Оптичне поглинання композиту на основі двошарових нанодротів

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В рамках класичної теорії Друде проаналізовано оптичні властивості двошарових нанодротів. Отримано вирази та розраховано частотні залежності дійсної та уявної частин діелектричної функції для метал-оксидних нанодротів та нанооболонок. Досліджено оптичне поглинання композиту на основі обох підходів. Показано, що варіювання розмірів товщини як ядра, так і оболонки, призводить до зміни величини та положення максимумів оптичних характеристик вказаних систем. Продемонстрована наявність двох максимумів коефіцієнту поглинання композиту на основі металевих нанооболонок. Розщеплення на два резонансних піки обумовлене гібридизацією поверхневих плазмонів, що залежать від внутрішнього та зовнішнього радіусу оболонки. Проаналізовано вплив кінцевої товщини металевого шару на частотні залежності коефіцієнту поглинання. Розрахунки проведені для металів Au, Ag та Al, що знаходяться у різних діелектричних матрицях. Встановлено, що поведінка максимумів оптичного поглинання композиту визначається діелектричною сталою матриці.

**Ключові слова:** Метал-оксидний нанодріт, Нанооболонка, Композит, Діелектрична функція, Частотна залежність, Коефіцієнт поглинання.