

PACS 78.67.-n;78.40.-q

INTERACTION OF OPTICAL RADIATION WITH METAL NANOPARTICLES IN DIELECTRIC MATRIX

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Using effective medium approximation absorption of electromagnetic radiation by the composite thin film representing a transparent dielectric matrix included metal nanoparticles with size much smaller than the wavelength of the incident light are considered. Absorption cross-section of metal nanoparticles is treated in the quasistatic approximation of classical electrodynamics. By means of size dependent effective optical parameters for refraction index and extinction coefficient optical resonance is evaluated. Dependencies of spectral characteristics on parameters of the system are analysed in detail.

Key words: Drude dispersion, metal nanoparticles, size dependent plasmon resonances, nanocomposite thin film.

1. Introduction

Optical properties of metal nanoparticles, essentially differ both from characteristics of massive samples, and from characteristics of separate atoms. These properties are connected with collective oscillations of conductivity electrons near to particle surface under the influence of electromagnetic radiation. Apparently, wide applications of metal nanoparticles are connected with their special optical properties, such as the absorption of optical radiation, selective character of this absorption cross-section and high concentration of electromagnetic energy in an area close to nanoparticle, much smaller than the wavelength of radiation [1-7].

Systems containing metal nanoparticles or nanostructured metal films are photonic and plasmonic systems of much current interest. In these systems can be excited special electronic excitations near the metallic surfaces. Surface plasmons can be intense and localized, and correctly describing their behavior in complex systems can require numerically rigorous modeling techniques. This contribution concerns interaction of local surface plasmon excitations of metal nanoparticles in host thin film material with optical radiation. Study of the phenomena, connected with oscillations of conductivity electrons in metal

nanostructures and nanoparticles (plasmon oscillations) and interaction of these oscillations with light represents great scientific and practical interest. Plasmon oscillations are spatially localized and occur with high frequency (from ultra-violet to infra-red).

In this work using the Maxwell Garnett model, absorption of electromagnetic radiation by the composite thin film consisting of transparent dielectric matrix with metal nanoparticles inclusions are investigated. By means of nanoparticle size dependent effective optical parameters for refraction index and extinction coefficient the analysis of absorption processes of optical radiation in such dielectric composite film is carried out. In so doing, it is assumed that the characteristic size of inclusions and their volume concentration satisfy the conditions of applicability of the effective medium Maxwell Garnett model [1].

For the coordination of experimental results with theoretical data the formula for complex dielectric permeability of the metal particles, including dependence on radius of particle has been offered. In this work the analysis of considered optical processes is realized taking into account the sizes of metal particles.

The account of nanoparticle size effects in expression of complex dielectric function of metal particles $\varepsilon_2(\omega, r) = \varepsilon_2' - i\varepsilon_2''$, is carried out by means of the following expression [7]:

$$\varepsilon_2(\omega, r) = \varepsilon_b(\omega) + \frac{\omega_p^2}{\omega(\omega + i\gamma_0)} - \frac{\omega_p^2}{\omega(\omega + i\gamma)} \quad (1)$$

where ω is the frequency of the incident field, r is the characteristic particle radius along the electric field direction of external electromagnetic wave, ω_p is the plasma frequency, $\varepsilon(r \rightarrow \infty) = \varepsilon_b$ is dielectric permeability of bulk metal, γ_0 is the electron relaxation rate in the metal, γ is size-dependent damping constant which equals to a inverse average transit time of electron in nanoparticle:

$$\gamma = \gamma_0 + v_F/r \quad (2)$$

v_F is the mean electron velocity at the Fermi surface.

Using Maxwell Garnett model for volume concentration $f \ll 1$ one can find real $\varepsilon' = n^2 - \chi^2$ and imaginary $\varepsilon'' = 2n\chi$ parts of dielectric constant of nanocomposite thin film containing metal nanoparticles [2]:

$$\varepsilon' = \varepsilon_1 \left[1 + 3f \cdot \frac{(\varepsilon_2' - \varepsilon_1)(\varepsilon_2' + 2\varepsilon_1) + \varepsilon_2''^2}{(\varepsilon_2' + 2\varepsilon_1)^2 + \varepsilon_2''^2} \right] \quad (3)$$

$$\varepsilon'' = 9f \cdot \frac{\varepsilon_1^2 \varepsilon_2''}{(\varepsilon_2' + 2\varepsilon_1)^2 + \varepsilon_2''^2} \quad (4)$$

where $\varepsilon_2' = n_2^2 - \chi_2^2$; $\varepsilon_2'' = 2n_2 \chi_2$ and for transparent matrix material $\varepsilon_1 = n_1^2$.

It is well known next expressions to find effective values for effective refraction index and extinction coefficient of nanocomposite thin film:

$$n^2 = \frac{1}{2}(\varepsilon' + \sqrt{\varepsilon'^2 + \varepsilon''^2}) \quad (5)$$

$$\chi^2 = \frac{1}{2}(-\varepsilon' + \sqrt{\varepsilon'^2 + \varepsilon''^2}) \quad (6)$$

From equation (4) we can obtain the plasmon resonance condition $\varepsilon_2'(\omega_{SP}) = -2\varepsilon_1$ for small values of ε_2'' . It is seen from (1) that plasmon resonance frequency depends on real parts of metal particle dielectric constant ε_2 , matrix material dielectric constant ε_1 , the electron relaxation rate in the metal γ_0 , filling factor f and the characteristic particle radius r .

Let us find frequency interval with negative real part of effective dielectric constant of thin nanocomposite film. In a points ω_1 and ω_2 real part of effective dielectric constant addresses in zero:

$$\omega_1 = \omega_p \sqrt{\frac{1-f}{3\varepsilon_1 + (1-f)(\varepsilon_b - \varepsilon_1)}} \quad (7)$$

$$\omega_2 = \omega_p \sqrt{\frac{1+2f}{2\varepsilon_1 + \varepsilon_b + 2f(\varepsilon_b - \varepsilon_1)}} \quad (8)$$

On an interval $[\omega_1, \omega_2]$ function $\varepsilon'(\omega)$ accepts negative values $\varepsilon'(\omega) < 0$, that is in this area of frequencies nanocomposite is similar to metal. One can see from formulae (7) and (8) that it is possible to operate width of this interval $\omega_2 - \omega_1$ by changing of metal nanoparticles filling factor f . Plots of the real (solid line) and the imaginary (dots) parts of the complex dielectric function of a composite medium consisting of Ag nanoparticles dispersed in a medium with $\varepsilon = 2.5$ with volume fraction $f = 0.01$ and $f = 0.1$ are presented in Figs.1, and 2. The silver parameters are: $\varepsilon_b = 5,00$, $\omega_p = 9$ eV, $\gamma_0 = 0,02$ eV [1].

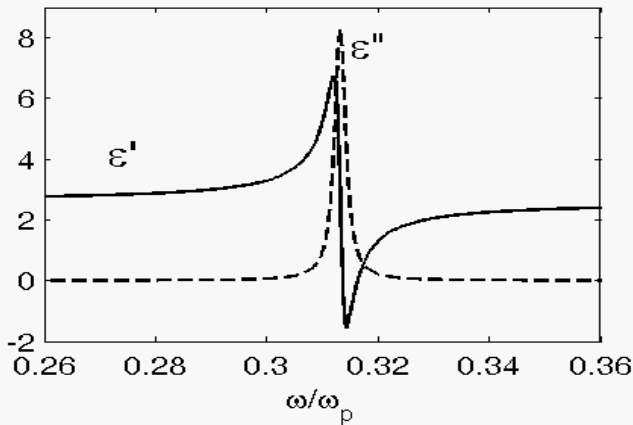


Fig.1. The real (solid line) and the imaginary (dots) parts of the complex dielectric function of a composite medium consisting of Ag nanoparticles dispersed in a medium with $\varepsilon = 2.5$ with volume fraction $f = 0.01$.

The silver parameters: $\varepsilon_b = 5,00$, $\omega_p = 9$ eV, $\gamma_0 = 0,02$ eV .

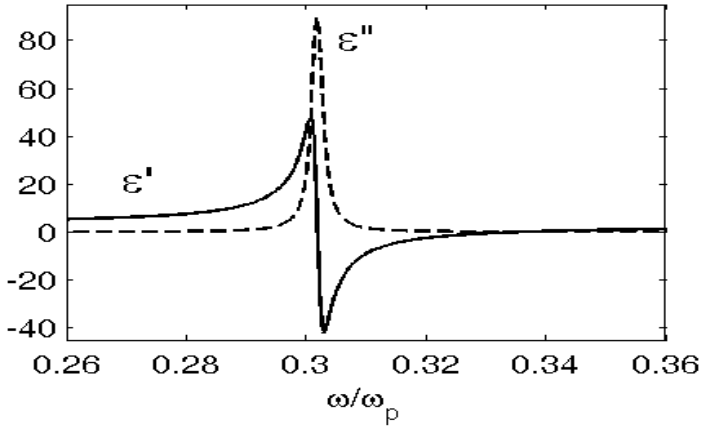


Fig.2. The real (solid line) and the imaginary (dots) parts of the complex dielectric function of a composite medium consisting of Ag nanoparticles dispersed in a medium with $\epsilon = 2.5$ with volume fraction $f = 0.1$. The silver parameters: $\epsilon_b = 5,00$, $\omega_p = 9\text{eV}$, $\gamma_0 = 0,02 \text{ eV}$.

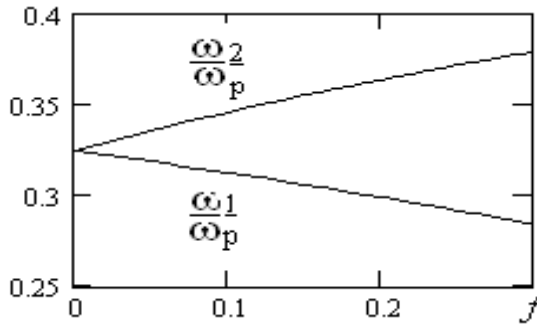


Fig.3. Dependences of ω_1/ω_p and ω_2/ω_p on filling factor f .

One can see that the imaginary part of the dielectric function basically resembles the typical behavior inherent in elementary absorption bands, for example for a gas of atoms. The imaginary part will accordingly give rise to the absorptive band whereas the real part will define the refraction with abnormal dispersion inside that band. The spectral position of this absorptive-like behavior is determined by the metal parameters and by its volume fraction. For the specific case of Figs. 1 and 2 it corresponds to approximately 440 nm wavelength. Similar to atomic absorption lines, the real part of the dielectric function at first grows with growing frequency (normal dispersion), then reaches its maximal value near a frequency corresponding to the half-maximum of the imaginary part, falls down (anomalous dispersion), crosses the zero axis near the frequency ω_1 corresponding to the maximum in the imaginary part, and acquires the negative sign onwards. For the case under consideration, the zero value of the real part of dielectric function occurs at the two frequencies

determined by the metal parameters, the ambient medium parameter ϵ_1 , and the composite medium under consideration parameter f .

Dependences of ω_1/ω_p and ω_2/ω_p on filling factor f are presented in Fig.3. One can see dependence of width of this interval $\omega_2 - \omega_1$ by changing of metal nanoparticles filling factor f when function $\epsilon'(\omega)$ accepts negative values. For this interval of frequencies $\epsilon'(\omega) < 0$, and nanocomposite is similar to metal [2,9].

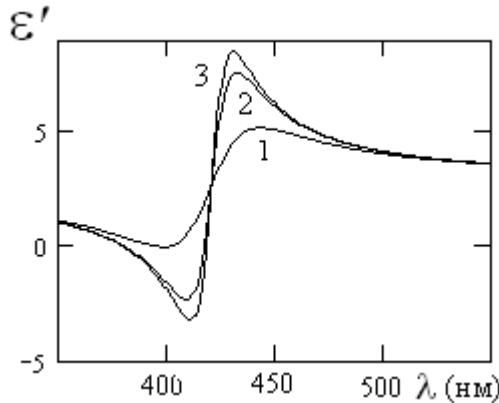


Fig.4. Real part of the complex dielectric function of a composite medium consisting of Ag nanoparticles with radii $r=5$ (1), $r=20$ (2) and $r=40$ (3) nm dispersed in a medium with $\epsilon = 2.25$ with volume fraction $f = 0.1$. The silver parameters: $\epsilon_b = 5,00$, $\omega_p = 9\text{eV}$, $\gamma_0 = 0,02$ eV.

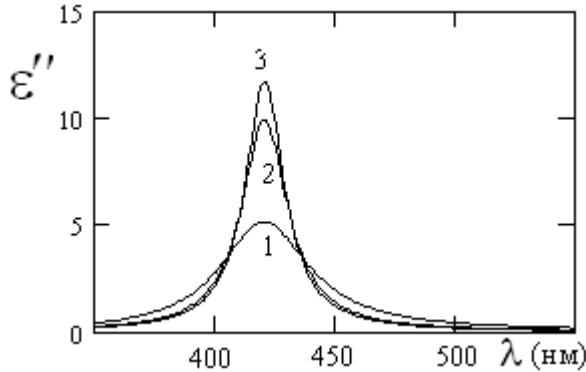


Fig.5. Imaginary part of the complex dielectric function of a composite medium consisting of Ag nanoparticles with radii $r=5$ (1), $r=20$ (2) and $r=40$ (3) nm dispersed in a medium with $\epsilon = 2.25$ volume fraction $f = 0.1$. The silver parameters: $\epsilon_b = 5,00$, $\omega_p = 9\text{eV}$, $\gamma_0 = 0,02$ eV.

Plots of the real ϵ' and the imaginary ϵ'' parts of the complex dielectric function of a composite medium consisting of Ag nanoparticles dispersed in a medium with $\epsilon = 2.25$ with radii $r=5$ (1), $r=20$ (2) and $r=40$ (3) nm for volume fraction $f = 0.1$ are presented in Figs.4, and 5. It is seen that real ϵ' and the imaginary ϵ'' parts of the complex dielectric function of a nanocomposite medium depend on size of metal nanoparticles. That is why interaction of electromagnetic radiation with plasmonic oscillations of metallic nanoparticles depends on size of metal nanoparticles. And plasmon resonance frequency also

depends on particle radius.

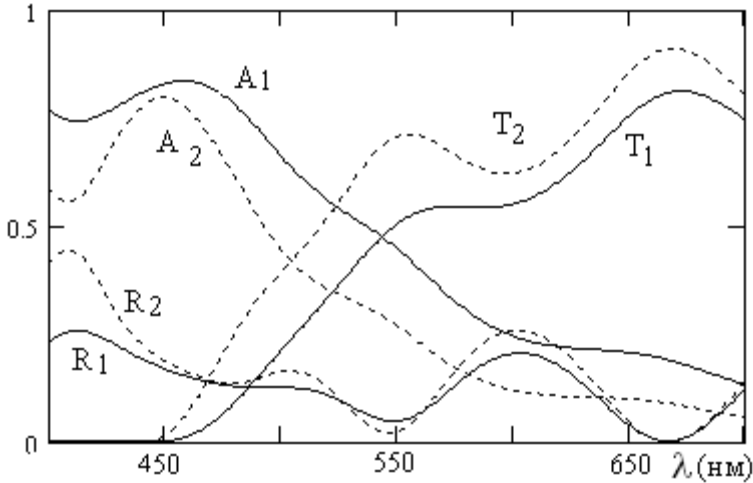


Fig.6. Spectral dependences of the reflection R, transmission T and absorption A coefficients of nanocomposite thin film with silver particles of radii $r = 5$ (R_1, A_1, T_1), 20 nanometers (R_2, A_2, T_2). The filling factor $f=0.1$. Matrix with refraction index $n_1=1.5$; a substrate - transparent dielectric with refraction index $n_3=3.6$.

By means of the received formulae dependencies of spectral characteristics on parameters of the system are analysed in detail : dielectric permeability of a transparent matrix material of a film, complex dielectric permeability of silver nanoparticles, radius and the filling factor, and also film thickness. It is shown, that such nanocomposite films depending on its parameters selectively are reflected optical radiation falling on it.

In Fig.6 are presented spectral dependences of the reflection R, transmission T and absorption A coefficients of nanocomposite thin film with silver particles of radii $r = 5$ (R_1, A_1, T_1) and 20 nanometers (R_2, A_2, T_2) for filling factor $f=0.1$. Matrix material is considered to be transparent with refraction index $n_1=1.5$; a substrate is transparent dielectric with refraction index $n_3=3.6$ [8]. It is necessary to notice, that all coefficients depend on size of silver nanoparticles. It is seen from Fig.6 that near to resonant frequency transmission coefficient equals zero. It means that for these frequencies $\epsilon'(\omega) < 0$ and nanocomposite material likes metal. Near to resonant frequency occurs absorption and reflection which depends on size of silver particle. With removal from resonant frequency of silver nanoparticles absorption coefficient decreases and transmission coefficient increases. For some frequencies reflection coefficient equals zero.

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OPTİK ŞÜALANMANIN METAL NANOZƏRRƏCİKLƏRLƏ DİELEKTRİK MATRİSDƏ QARŞILIQLI TƏSİRİ

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XÜLASƏ

Effektiv mühit yaxınlaşmasında elektromaqnit şüalanmasının ölçüləri düşən işıq dalğası uzunluğundan çox kiçik olan metal nanozərrəcikləri şəffaf dielektrik kompozit nazik təbəqədə udulması tədqiq edilmişdir. Metal nanozərrəciklərin udulma en kəsiyi klassik elektrodinamikanın kvazistatik yaxınlaşmasında təhlil olunmuşdur. Optik rezonans ölçüdən asılı effektiv optik parametrlər olan sındırma əmsalı və ekstinksiya əmsalı ilə nəzərə alınmışdır. Spektral xarakteristikaların sistemin parametrlərindən asılılıqları ətraflı analiz olunmuşdur.

Açar sözlər: Drude dispersiyası, metal nanozərrəciklər, ölçüdən asılı plazmon rezonanslar, nanokompozit nazik təbəqə.

ВЗАИМОДЕЙСТВИЕ ОПТИЧЕСКОГО ИЗЛУЧЕНИЯ С МЕТАЛЛИЧЕСКИМИ НАНОЧАСТИЦАМИ В ДИЭЛЕКТРИЧЕСКОЙ МАТРИЦЕ

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РЕЗЮМЕ

В приближении эффективной среды рассмотрено поглощение электромагнитного излучения в прозрачной диэлектрической композитной тонкой пленке с наночастицами металла, размеры которых намного меньше по сравнению с длиной волны падающего света. Сечение поглощения наночастиц металла рассмотрено в квазистатическом приближении классической электродинамики. Оптический резонанс оценивался с помощью размерно-зависящих эффективных оптических параметров для показателя преломления и коэффициента экстинкции. Детально проанализированы зависимости спектральных характеристик от параметров системы.

Ключевые слова: дисперсия Друде, металлические наночастицы, размерно-зависимые плазмон-резонансы, нанокomпозитная тонкая пленка.

Поступила в редакцию: 23.09.2011 г.

Принята к печати: 05.03.2012 г.