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Influence of the shape of the particles covering the metal surface on the dispersion relations of surface plasmons

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Abstract

The Green function method was used for calculation of dispersion relations of surface plasmon generated at the metal surface covered by ellipsoidal particles. The influence of the shape of the particles on the surface plasmon dispersion relations was studied. It was established that both s- and p-polarized surface plasmons can be excited at the surface covered by the particles. It was shown that the shape of the particles covering the surface strongly influences on the surface plasmons dispersion. © 2008 Elsevier B.V. All rights reserved.

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1. Introduction

The study of surface plasmon-polaritons is in interest as well as from fundamental and applications points of view [1–4]. By altering the structure of a metal surface, the properties of surface plasmons can be tailored, which offers the potential for developing the new types of photonic devices. Surface plasmons are studied for their using in sub-wavelength optics [5,6], data storage, light generation [7,8], microscopy and bio-photonics [3,9–11]. Therefore, study of the surface plasmon propagation along the surfaces covered by meso-particles, bio- and organic molecules, ultrathin organic and biopolymer films is in great interest this time [12,13]. The effects of the local dielectric environment on the surface plasmon resonances of annealed gold-island films were studied in [14,15]. The depolarizing effect of the surrounding medium was clearly demonstrated by shifts in the frequencies of the resonance peaks. The usage of the surface plasmons in microscopy is developed this time (see, for example, Ref. [9]). A far-field optical microscopy

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technique capable of reaching nanometre-scale resolution has been developed using the in-plane image magnification by surface plasmons. This microscopy is based on the optical properties of a metal-dielectric interface that may, in principle, provide extremely large values of the effective refractive index $n_{\rm eff}$ up to $10^2 - 10^3$ as 'seen' by the surface plasmons. Thus, the theoretical diffraction limit on the resolution becomes $\lambda/2n_{\text{eff}}$, and falls into the nanometre-scale range [9]. Furthermore, the surface-sensitive optical technique of surface plasmon resonance (SPR) imaging is used to characterize ultrathin organic and biopolymer films at metal interfaces [10]. Because of its high surface sensitivity and its ability to measure in real time the interaction of unlabeled biological molecules with arrays of surfacebound species, SPR imaging has a potential to become a powerful tool in bimolecular investigations. Recently, SPR imaging has been successfully implemented in the characterization of supported lipid bilayer films, the monitoring of antibody-antigen interactions at surfaces, and the study of DNA hybridization adsorption. At the same time, the intensity studies of surface plasmon resonance are performed for developing of SPR sensor method [13,16,17]. In this connection, one should point the works [17-22] in

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which the SPR method for sensors of organic (bio-) molecules and problems of surface wave propagation along the surface covered by molecular layer were performed. All mentioned above studies touch upon the surface plasmon excitation. Then, one should know the dispersion relations of surface waves. It is clear that dispersion relations are depended on the state of the surface at which the surface plasmon is excited. For example, SPR method is based on this fact (see, for example Refs. [13,16,17]). Indeed the shift of the resonance curve shows that the conditions of the exciting of the surface wave are changed. This means that the knowledge of the influence of surface cover on the surface plasmon dispersion curves is very important. Then, the main purpose of the work is study of influence of the particles shape covering the metal surface on the surface waves dispersion. As it was mentioned above, this problem is very important for nano-optics, sensorics and plasmonics.

It should be noted there is a general approach to determine the optical properties of the metal inclusions of ellipsoidal shape in dielectric matrix. This is effective medium approximation [23–26]. It is used when dimensions of the particles are smaller than average distances between particles and wavelength of the probing field. Other words, the effective medium method consists in replacing the complex inhomogeneous system under consideration by any other system which optical properties are similar to initial system. The new system is homogeneous one which properties are described by some effective parameters. For example, it could be the effective permittivity. When the particles form sub-monolayer cover this approximation is not valid because, for example, it is difficult to say about interfaces of such film. That's why in the problem under consideration it is reasonably to use more adequate method to find the response of the system on the external field which was used in the present work.

2. Effective susceptibility and dispersion relations

A collective oscillation of electron plasma near the surface of a metal is known as a surface plasma wave or surface plasmon. The speed of such surface wave is slower then the speed of light in the medium adjacent to the metal surface, then the electromagnetic wave is evanescent. Surface plasmon localized on the flat interface is longitudinal mode and correspond to oscillations of the near-surface electron density. Electric field decays exponentially as an evanescent wave in the direction normal to the interface. Solving Maxwell's equations with boundary conditions (or studying the pole part of the Green function of the system [31,32]) one can find the condition when surface plasmon exists

$$\kappa_{\rm m}\varepsilon_{\rm d}(\omega) + \kappa_{\rm d}\varepsilon_{\rm m}(\omega) = 0, \tag{1}$$

where $\varepsilon_{\rm d}(\omega)$ and $\varepsilon_{\rm m}(\omega)$ are dielectric constants of the adjacent mediums (dielectric and metal), $\kappa_{\rm d,m} = \sqrt{k^2 - \varepsilon_{\rm d,m}(\omega/c)^2}$ are the propagation constants in the media "d" (dielectric) and "m" (metal), respectively. One can see that surface plasmons can be excited only in the case when permittivities of the adjacent mediums have opposite signs. Since the surface wave is associated with an evanescent field it does not couple to any freely propagating electromagnetic mode (this means that it cannot be excited by light impinging on the interface). Excitation of the surface plasmon can be performed with the evanescent field generated by either total reflection method, a fine grating, or any other sub-wavelength structure. One should note that the surface plasmon at the perfect flat interface can be excited by only p-polarized (or TH) external radiation [27].

Dispersion relations connect frequency with wave vector of the waves and show under which conditions such waves can be excited. Using Eq. (1) one can find that the wave vector k of the surface plasmon is given by

$$k_{\rm sp}(\omega) = \frac{\omega}{c} \sqrt{\frac{\varepsilon_{\rm d}(\omega)\varepsilon_{\rm m}(\omega)}{\varepsilon_{\rm d}(\omega) + \varepsilon_{\rm m}(\omega)}},\tag{2}$$

where ω is a frequency of the surface plasmon. Fig. 1 is plot of the $\omega - k$ relationship. It is clear that when the state of the surface is changed the conditions of the surface plasmon exciting are violated. This leads to the changing of the dispersion relations. Thus dispersion relations are depended on the state of the surface at which the surface plasmon is excited. In the case when the interface is not ideal (covered, for example, by sub-monolayer of nanoparticles or by dielectric ultrathin film) it is possible to excite s-polarized or TE surface plasmon [18,19].

Let us consider the molecular cover of the metal surface. Molecules are presented as ellipse-like homogeneous particles uniformly distributed along the plane of metal surface. To calculate the effective susceptibility one can use the approach developed in Refs. [18,28]. As a result, one obtains the effective susceptibility of the molecular layer in the form

$$X_{ij}(\mathbf{k},\omega) = [\chi_{ij}^{-1}(\omega) - nG_{ji}(\mathbf{k},l,l,\omega)]^{-1},$$
(3)

with $G_{ij}(\mathbf{k}, l, l, \omega)$ is electrodynamical Green function of the medium in which molecular layer is embedded, *n* is a concentration of particles at the surface and $\chi_{ij}(\omega)$ is a susceptibility of single molecule at the surface. Obviously that in the case under consideration one should use the Green function of two semi-spaces with flat interface. Because



Fig. 1. Dispersion relation of surface plasmon.

we assume that the molecules can be presented as ellipsoidal homogeneous particles one can use for molecular susceptibility the polarizability of ellipsoid-like particle at the surface which has a form

$$\alpha_{ij} = \begin{pmatrix} \alpha_{\parallel} & 0 & 0\\ 0 & \alpha_{\parallel} & 0\\ 0 & 0 & \alpha_{\perp} \end{pmatrix}, \tag{4}$$

where the components of polarizability are written as [29]

$$\alpha_{\parallel,\perp} = \varepsilon_{\rm m} V_{\rm p} \frac{(\varepsilon_{\rm p} - \varepsilon_{\rm m})}{\varepsilon_{\rm m} + (\varepsilon_{\rm p} - \varepsilon_{\rm m}) m_{\parallel,\perp}} L_{\parallel,\perp}.$$
(5)

In this expression the next designations are used: $\varepsilon_{\rm p}$ is a dielectric constant of the particle, $\varepsilon_{\rm d}$ is a dielectric constant of external medium, $\varepsilon_{\rm m}$ is a dielectric constant of a substrate (metal), and

$$L_{\parallel,\perp} = \left[1 + \frac{(\varepsilon_{\rm m} - \varepsilon_{\rm d})(\varepsilon_{\rm p} - \varepsilon_{\rm m})}{3(\varepsilon_{\rm m} + \varepsilon_{\rm d})(\varepsilon_{\rm m} + (\varepsilon_{\rm p} - \varepsilon_{\rm m})m_{\parallel,\perp})}U_{\parallel,\perp}\right]^{-1} \tag{6}$$

with $m_{\parallel,\perp}$ are depolarization factors of the particle [30], and $U_{\parallel,\perp} = \vartheta, 2\vartheta$, where parameter $\vartheta = h_x h_y h_z (2z_p)^{-3}$ is determined by linear dimensions of the particle (See, Fig. 2). The semi-axes of the ellipsoids are denoted as h_i , i = x, y, z. Prolate and oblate ellipsoids are characterized by different depolarization factors. According to [29] and [30] these depolarization factors can be written in the form:

- for prolate ellipsoidal particle, where $h_z > h_x = h_y$ depolarizing factor describing the polarization along to the surface plane

$$m_{\parallel} = \frac{1}{2}(1 - m_{\perp}),$$
 (7)

and along the normal to surface plane

1

$$m_{\perp} = \frac{1 - \zeta^2}{\zeta^3} \left(\frac{1}{2} \ln \frac{1 + \zeta}{1 - \zeta} - \zeta \right), \tag{8}$$

where parameter describing the shape of the particle

$$\zeta = \sqrt{1 - h_x^2/h_z^2}.\tag{9}$$

– for oblate ellipsoidal particle, where $h_z < h_x = h_y$ depolarizing factor describing the polarization along to the surface plane

$$m_{\perp} = \frac{1+\zeta^2}{\zeta^3} (\zeta - \arctan \zeta), \qquad (10)$$

$$m_{\parallel} = \frac{1}{2}(1 - m_{\perp}),$$
 (11)

$$\zeta = \sqrt{h_x^2 / h_z^2 - 1}.$$
 (12)

Then, the effective susceptibility can be considered as defined.

As it is well known [31,32], the dispersion relations can be obtained as zeros of pole part of susceptibility of the system under consideration. In the case considered in this work, this condition is reduced to fulfilling the requirements of

$$\det[\alpha_{ij}^{-1}(\omega) - nG_{ji}(\mathbf{k}, l, l, \omega)] = 0.$$
(13)

When one uses the electrodynamical Green function of two semi-spaces with flat perfect interface (see, for example, Refs. [18,33]), and, supposing that surface wave propagates along OX axes, one can see that Eq. (13) brakes apart into two equations – the first of them describes the TE-

$$\alpha_{\parallel}^{-1}(\omega) - nG_{yy}(k,\omega) = 0 \tag{14}$$

and the second describes TH-

$$[\alpha_{\parallel}^{-1}(\omega) - nG_{xx}(k,\omega)] \cdot [\alpha_{\perp}^{-1}(\omega) - nG_{zz}(k,\omega)] - n^2 G_{zx}(k,\omega) G_{zx}(k,\omega) = 0$$
(15)

branches of dispersion relations.

3. Numerical calculations

To demonstrate the influence of shape of the particles covering the surface on the dispersion relations of surface waves one chooses the metal characterized by dielectric function

$$\varepsilon_m(\omega) = \varepsilon_\infty (1 - \omega_{\rm pl}^2 / \omega^2), \tag{16}$$

with plasma frequency ω_{pl} . This parameter was chosen as typical plasma frequency of the metal

$$\omega_{\rm pl} = 2 \times 10^5 \,\rm cm^{-1} \tag{17}$$

(for example, $\omega_{\rm pl} = 1.2 \times 10^5 \, {\rm cm}^{-1}$ for Al [34], and $\omega_{\rm pl} = 4 \times 10^5 \, {\rm cm}^{-1}$ for Ag [35]). The high-frequency dielectric



Fig. 2. Ellipsoidal particles at the surface: (a) as prolate ellipsoid; (b) as oblate ellipsoid.

constant was supposed as $\varepsilon_{\infty} = 4$. Then, using the normalized variables $x = \omega/\omega_{\rm pl}$, one can rewrite the dielectric function of the substrate in the form

$$\varepsilon_{\rm m}(x) = \varepsilon_{\infty} \left(1 - \frac{1}{x^2} \right). \tag{18}$$

As covering particles (molecules) one supposes two kinds of the ellipsoids

- prolate ellipsoids with parameters

$$h_x = h_y = 1 \text{ nm}, \quad h_z = 2 \text{ nm},$$
 (19)

- oblate ellipsoids with semi-axes

$$h_x = h_y = 2 \text{ nm}, \quad h_z = 0.5 \text{ nm}.$$
 (20)

These parameters provide us for the particles with the same volume $V_p = 4/3\pi h_x h_y h_z$ is equal to $V_p \simeq 8.38 \times 10^{-21} \text{ cm}^3$. Such dimensions of the particles were chosen because it is characteristic sizes of many biomolecules and colloidal gold particles. One supposed during the calculations that monolayer cover corresponds to concentration $n_0 = 0.25 \times 10^{13}$ particles/cm² and dielectric constant of ellipsoids is $\varepsilon_n = 5$. In some cases, when we in more detail studied the influence of particles shape on the dispersion of surface plasmon we supposed the particles with different semi-axes but with the same volume $V_{\rm p}$. Since Eqs. (14) and (15) are soluble when particle concentration $n \neq 0$, one can see that analogously to previous study [18] both p- and s-polarized surface waves can be excited in the system under consideration. Moreover the surface waves in this case can be excited in the frequency range where dielectric function of a substrate is positive (one should remember that standard surface plasmon can be excited in the frequency range where dielectric function of a substrate is negative). Here for definiteness one will consider only surface waves which can be excited in the frequency range, where $\varepsilon_m < 0$, i.e., in the range of surface plasmon existence. In the terms of normalized frequency x the range is 0.001 < x < 0.886.

4. Results and discussion

The purpose of the present calculations was a demonstration of the influence of covering particles shape on the dispersion curves of surface plasmon. To demonstrate this influence the dispersion curves of surface plasmon were calculated when the oblate and prolate ellipsoidal particles (with different relationships between its semi-axes) cover the surface. The dispersion curves of s-polarized surface plasmon at the surface covered by oblate ellipsoids are represented in Fig. 3. As one can see, more broadening of dispersion (the more wide frequency range the dispersion curve occupies) when the particles covering the surface are more oblate. This behavior of surface plasmon dispersion of s-polarization can, obviously, be explained by strong polarizability of oblate ellipsoids along the direction parallel to the surface plane. Indeed, the s-polarized wave



Fig. 3. The influence of the shape of the particles as oblate ellipsoids covering the surface on the dispersion of surface plasmon of s-polarization (concentration of the particles $n = n_0$). (1) $h_x = 2 \text{ nm}$, $h_z = 0.5 \text{ nm}$; (2) $h_x = 1.8 \text{ nm}$, $h_z = 0.62 \text{ nm}$; (3) $h_x = 1.26 \text{ nm}$, $h_z = 1.26 \text{ nm}$.

has only *y*-component of electric field (parallel to surface plane). This component effectively interacts with polarization of the particle parallel to the surface of a substrate, which are stronger for more oblate particles. This point is the additional confirmation of the main role of cover of the particles in formation of s-polarized surface wave. In contrast to the case of oblate ellipsoids, the influence of the shape of prolate ellipsoids on the dispersion of s-polarized surface plasmon is not so essential. As one can see from Fig. 4 the extremely small dispersion – the relative changes of dispersion is not more than 0.3% is observed in this case. This fact can be easily understood within previous speculations, because the polarizability of prolate ellipsoids along the direction of its short semi-axis (parallel to the substrate surface) is rather small.



Fig. 4. The influence of the shape of the particles as prolate ellipsoid covering the surface on the dispersion of surface plasmon of s-polarization (concentration of the particles $n = n_0$). (1) $h_z = 2 \text{ nm}$, $h_x = 1 \text{ nm}$; (2) $h_z = 1.8 \text{ nm}$, $h_x = 1.05 \text{ nm}$; (3) $h_z = 1.5 \text{ nm}$, $h_x = 1.15 \text{ nm}$; (4) $h_z = 1.26 \text{ nm}$, $h_x = 1.26 \text{ nm}$.



Fig. 5. Dispersion curves of p-polarized surface plasmon when the oblate ellipsoidal and spherical particles (marked by "Sph") cover the surface of concentration n_0 .



Fig. 6. Dispersion curves of p-polarized surface plasmon when the oblate ellipsoidal particles cover the surface of concentration 0.1 n_0 . Dashed curve corresponds to SP at free surface.

Absolutely another behavior of dispersion demonstrates p-polarized surface plasmon. In this case very complicate spectra can be seen from Figs. 5, and 6. Four branches of the dispersion curves can be observed for the both types of particles oblate and prolate ellipsoids. The four modes appear as the result of interaction of plasmon wave with the transversal and longitudinal resonance frequencies of the ellipsoidal particles. For the spherical-like particles on the contrary (see Fig. 5) only two branches of the dispersion curves can be seen. This fact, obviously, can be explained by two components of the polarization of the particles which interact with two components of the electric field (directed along OX and OZ axes of Cartesian coordinates, respectively). Because polarizability along OX and OZ axes are equal one to other for the sphere, one can observe only two branches in dispersion curves. This fact is not universal, but caused by very weak influence of interaction between particles and surface at used in this work parameters. Then the difference between values of L_{\parallel} and L_{\perp} (see, Eq. (6) in which $m_{\parallel} = m_{\perp} = 1/3$ should be putted) is insufficient for forming sufficiently different in-plane and normal polarizabilities for sphere-like particles. One should note that because configuration resonances of prolate ellipsoid are situated very close over frequency, the upper two branches of the dispersion curves (see Fig. 7) are almost nondispersive. As well as oblate ellipsoidal particles, the influence of the particle shape on the dispersion of surface plasmon is obtained for prolate ellipsoids. This influence is demonstrated in Fig. 8 for low branches of dispersion of p-polarized surface plasmon. In Figs. 5-8 one can see dispersion curves bend to a lower frequency side at a higher k-region. This means that such waves have negative dispersion. It means that the direction of wave front propagation is opposite to the direction of the energy propagation. These waves can exist in solid states [36].

One needs to note that interpartical interactions (lateral interactions) contribution to surface waves dispersion.



Fig. 7. Dispersion curves of p-polarized surface plasmon for surface covered by prolate ellipsoids of concentration n_0 . Insert shows the two high frequency branches of the dispersion low.



Fig. 8. The influence of prolate ellipsoidal particles shape on the low branches of dispersion curves. Curves 1 correspond to particles with $(h_z = 4 \text{ nm}, h_x = h_y = 0.71 \text{ nm})$; curves 2 correspond to particles with $(h_z = 2 \text{ nm}, h_x = h_y = 1 \text{ nm})$; curves 3 correspond to spherical particles with the same volume $(h_z = h_x = h_y = 1.26 \text{ nm})$.

Moreover, s-polarized surface waves are formed mostly because of particle cover at the surface. Indeed, s-polarized surface wave does not exist for free (when the surface cover is absent) surface. Fig. 9 demonstrates the influence of lateral interactions (which become apparent via concentration of particles on the surface) on dispersion of s-polarized surface plasmon. As one can see, the weakening of lateral interactions leads to narrowing of dispersion of surface plasmon. This is rather transparent result because of arising of s-polarized surface wave in this case is caused by cover at the surface. It means that the limit curve for which the dispersion curves have to tend asymptotically to the line corresponding to the configuration resonance frequency of the single particle at the surface. Similar behavior of dispersion curves of s-polarized surface waves demonstrates when the prolate particles cover the surface.



Fig. 9. The dispersion curves of s-polarized surface plasmon when the surface is covered by oblate ellipsoidal particles. Curve 1 corresponds to concentration $n = n_0$, curve $2 - 0.5n_0$, curve $3 - 0.1n_0$.

Calculations for p-polarized waves show that decreasing of the concentration of particles at the surface lead to grouping of the dispersion curves along the dispersion curve of surface plasmon of free surface. At the same time the upper-frequency branches of ellipsoidal particles degenerate to the nondispersive lines which correspond to eigenmodes of configuration resonances of the ellepsiod-like particles at the surface.

5. Conclusions

In the frame of the Green function method using the effective susceptibility concept one developed the approach for calculations of surface electromagnetic waves dispersion. It was supposed that the metal surface is covered by the ellipsoidal particles. It was shown that the shape of the covering particles influences on the surface plasmons dispersion. Because of covering particles have an ellipsoidal shape, two configurational resonances characterize the response of the particle on the external field. The interference of the surface plasmon and these resonances leads to that new surface waves occur. These waves can be excited by sand p-polarized light. As far as spherical particles have only one resonance, then the number of dispersion curves in this case has to be less than for ellipsoidal particles. The shape of the particles strongly influences on the forming of surface electromagnetic waves. This fact is very important in SPR sensor method. When organic (bio-) molecules adsorb on the passivated metal surface they do not denaturate but take the ellipsoidal form. Consequently, using SPR sensor method one can measure dispersion curves and determine the shape of the molecules and, in that way, their type. The fact of influence of the shape of covering particles on the dispersion of surface plasmon is very important in plasmonics. Indeed, the dispersion relations point to the conditions under which the surface plasmon can be excited. Then the properties of the surface cover will define the conditions of the surface plasmon exciting.

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