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QUASISTATIC (LOCALIZED) PLASMONS: FROM LANGMUIR TO FERRELL

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Abstract

We review main results in electrostatic aspects of plasmonics. Although many applications of plasmonics require full-wave approach, plasmon resonance has an electrostatic nature. In this paper we focused on fundamentals of plasmonics, which are easier understood in the electrostatic approximation. We also touch upon a history of first insights in resonances in subwavelength electromagnetic systems.

Keywords: plasmonics, localized plasmons, plasmon resonance, electrostatic approximation

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1. Introduction

Recently electrodynamics of media with negative values of permittivity and/or permeability has attracted huge interest in the literature [1–13]. Basic phenomena in this field, such as, e.g., SERS [6, 14–20], SPASER, STM [21], and numerous effects observed in metamaterials [1–13], are related to plasmon resonances. All these phenomena can be united by a single term—plasmonics.

A characteristic feature of plasmonics, which singles it out from ordinary optics and electrodynamics, is that main phenomena in plasmonics occur on scales that are much smaller than the wavelength of light in vacuum. This endows plasmonics with many features of near-field optics and makes it to be in great demand for modern technologies.

Since the “stage”, on which events in plasmonics take place, is small, plasmonics is akin to physics of magnetostatic waves [14, 15, 22] with the only difference that magnetostatic phenomena occur in the microwave range, while plasmonic events are observed in optics. As well as in the description of magnetostatic waves, the majority of phenomena of plasmonics can be treated in terms of the quasistatic approach. This is related to the fact that the spatial derivatives in Maxwell’s equations greatly exceed the time derivatives, which, therefore, can be neglected.

In this paper, we review plasmonic systems, which may be considered in quasistatic approximation.

2. The frequency of plasmon resonance

The term “plasmon” has appeared in plasma physics to describe longitudinal collective oscillations of electrons (Langmuir waves[1]) in plasmas. On average, plasma is quasineutral, this means that the mean local charge equals zero. Assume that, at a certain moment of time, a fluctuation in the charge distribution arose; namely, all particles of like charges in a plane layer with cross section $ABCD$ (see Fig. 1), e.g., electrons, are spontaneously displaced in the same direction by distance x. As a result of this spontaneous charge separation, a plane capacitor is formed, inside of which homogeneous electric field $E = enx$, appeared, where e is the electron charge, and n is the electron density. This field will act on a single uncompensated electron

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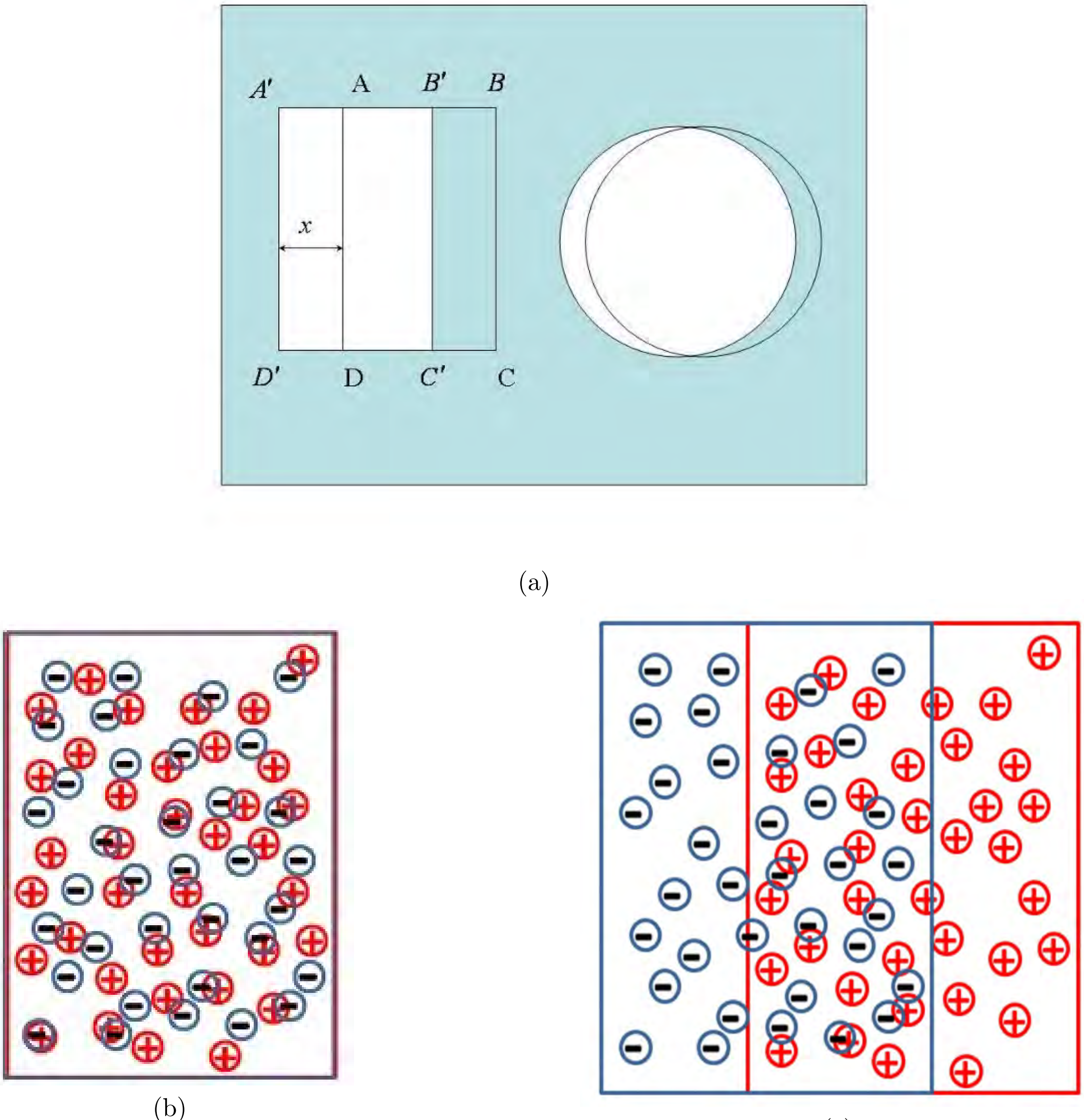
[1] The term “plasma” itself has been introduced by Irving Langmuir [23]

with force $F = eE = e^2nx$, so that the equation of motion of such an electron has the form

$$\ddot{x} = -\frac{e}{m}E = -\frac{e^2n}{m}x. \tag{1}$$

The solution to this equation is a harmonic oscillation with a frequency $\omega_p = \sqrt{e^2n/m}$, which is referred to as the plasma frequency.



(a)

(b)

(c)

Fig. 1 – Charge separation as a result of plasmon oscillations

If the fluctuation has the shape of a sphere rather than of a layer, the field inside of the fluctuation region will also be homogeneous. Indeed, uncompensated negative charges in the examined fluctuation will shift outside from the surface of the initial volume by the distance $\delta x = x\cos\varphi$ (R and φ are the spherical coordinates fixed to the center of the fluctuation); as a result, on the surface of the surface charge $\delta\sigma = nex\cos\varphi$ appears. Mathematically, this problem is equivalent to the problem on the field inside a dielectric sphere with the polarization $P = nex$. As is well known [24], the field E inside of this sphere is given by $4\pi P/3$. Substituting this field $E = 4\pi enx/3$ into Eq. (1), we arrive at the equation governing motion of an oscillator with the frequency equal to

$$\omega_{sp} = \sqrt{\frac{e^2n}{3m}} = \omega_p/\sqrt{3}.$$

For the fluctuation having the shape of an ellipsoid, the field in it as a result of the charge separation will be n_iP, where $i = x, y, z$ coincides with one of the principal axes of the ellipsoid, while n_i is the depolarization factor [25]. Correspondingly, the resonant frequency will be given by $\omega_{ep\alpha} = \omega_p\sqrt{n_\alpha}$. Note that $n_x + n_y + n_z = 1$ [25]. For a circular cylinder whose axis is directed along the x coordinate, we have

$$n_x = 0,\ n_y = n_z = 1/2, \text{ and } \omega_{cp} = \omega_p/\sqrt{2}.$$

3. Description of the plasmon resonance in terms of the permittivity

In order to describe plasmon oscillations, microscopic description methods have been developed, which take into account quantum effects, and so on [18, 19]. However, in view of the collective character of oscillations, the description of plasmons in terms of the permittivity is often employed. In this chapter, we restrict ourselves precisely to this approach. To estimate the permittivity, we consider plasma in the free-electron approximation [1, 6, 21]. In other words, we will assume that the field generates the current $\mathbf{J} = ne\boldsymbol{\nu}$, where $\boldsymbol{\nu}$ is the average velocity of electrons. Microscopic Maxwell's equations are reduced to the wave equation

$$\nabla \times \nabla \times\ \mathbf{E} = -\frac{\varepsilon\mu}{c^2}\frac{\partial^2\mathbf{E}}{\partial t^2} - \frac{4\pi\mu}{c^2}ne\frac{\partial}{\partial t}\boldsymbol{\nu}. \tag{2}$$

According to Newton's second law, $m\dot{\boldsymbol{\nu}} = -e\mathbf{E}$. This finally yields the following closed equation for the electric field:

$$\nabla \times \nabla \times \mathbf{E} = -\frac{1}{c^2}\frac{\partial^2\mathbf{E}}{\partial t^2} + \frac{4\pi\mu}{c^2}\frac{ne^2}{m}\mathbf{E}. \tag{3}$$

For its solution in the form of plane wave $\exp\left(-i\omega t + i\mathbf{kr}\right)$, we obtain the dispersion equation

$$k^2 = \frac{\omega^2}{c^2}\left(1 - \frac{\omega_p^2}{\omega^2}\right), \tag{4}$$

which yields the following expression for the permittivity of the plasma:

$$\varepsilon\left(\omega\right) = 1 - \frac{\omega_p^2}{\omega^2}. \tag{5}$$

The condition $\omega = \omega_p$ of excitation of the volume Langmuir oscillations (see Eq. (1)) looks now as $\varepsilon_{res} = \varepsilon\left(\omega\right) = 0$. We note that this condition is unambiguously related to the geometry of the problem defined by plane wave. In the case of a spherical geometry, plasmon oscillations are excited at the frequency $\omega = \omega_p/3$, i.e., at resonant value of permittivity $\varepsilon_{res} = -2$. In the case of an ellipsoid,

$$\varepsilon_{res} = -(1/n_\alpha + 1);$$

in particular, for a cylinder, $\varepsilon_{res} = -1$. Note that, in all these cases, the dipole moment d induced by external field E as well as polarizability $\alpha = P/\left(EV\right)$ become infinite. Here V is the particle volume. In particular, for a sphere that consists of a material with ε_{int} and is placed into a medium with ε_{ext}, we have

$$\alpha = \frac{3}{4\pi}V\frac{\varepsilon_{\text{int}} - \varepsilon_{ext}}{\varepsilon_{\text{int}} + 2\varepsilon_{ext}}. \tag{6}$$

In other words, at resonant values of permittivity a zero field can cause a finite response of a plasmonic particle. This means that plasmon oscillations are nonzero eigensolutions of the Maxwell equations—plasmons that exist in the absence of an external field.

It is worth emphasizing that the values of permittivity at which the plasmon resonance is observed are independent of the particular form of the frequency dispersion. Thus the resonance can be observed at resonant values of permittivity even in materials with a permittivity dispersion different from (5). The frequency of the plasmon resonance, more exactly, the negative value of the permittivity at which it is observed, is determined by the geometry of the problem. Indeed, the equation $\operatorname{div}\left(\varepsilon\mathbf{E}\right) = 0$ for plane waves $E \sim \exp\left(-i\omega t + i\mathbf{kr}\right)$, which define the planar geometry, is reduced to the following equation

$$\varepsilon\left(\omega\right)\mathbf{kE} = 0. \tag{7}$$

At $\varepsilon\left(\omega\right) \neq 0$ the solution is a transverse travelling wave, while, at $\varepsilon = 0$, the localized longitudinal oscillations of the electric field may appear. It is the case of the Langmuir resonance. It is evident that the curl of this field is zero and, therefore, the magnetic field is also zero. The latter fact means that the vector potential can be neglected and the electric field can be considered to be the gradient of the scalar potential.

4. Multipole resonances of a plasmonic particle

As we have seen, if the symmetry of the particle changes, the resonant value of the permittivity also changes. The same effect may be achieved by changing the symmetry of the field distribution. Even if the shape of the plasmonic particle is specified, the resonance can be observed at several values of the permittivity, corresponding to excitation of multipole plasmons, such as quadrupole, octupole, etc.

For simplicity, let us consider a spherical plasmonic particle. As is well known, in a homogeneous electric field only an electric dipole moment may be induced in the sphere. In order to excite a higher-order (multipole) moment the sphere should be placed in an inhomogeneous field. The inhomogeneity of the field can be caused by different reasons. For example, it can be created by a periodic inhomogeneity in the space of an incident plane wave, or by an inhomogeneity formed by neighboring inclusions in a composite material, or by some other inhomogeneity of the system, such as, e.g., corners, sharp edges, tips, and so on. In any case, the field of a plasmon is the solution of the Laplace equation that vanishes at infinity.

The problem on excitation of a small sphere by an inhomogeneous field of the form of $\varphi_0 = r^l Y_{l,m}(\theta,\varphi)$ is reduced to solving the Laplace equation [2]

$$\frac{1}{r^2}\frac{\partial}{\partial r}\left(r^2\frac{\partial\varphi}{\partial r}\right)+\frac{1}{r^2\sin\vartheta}\frac{\partial}{\partial\vartheta}\left(\sin\vartheta\frac{\partial\varphi}{\partial\vartheta}\right)+\frac{1}{r^2\sin^2\vartheta}\frac{\partial^2\varphi}{\partial\varphi^2}=0. \tag{8}$$

In order to find its solution, it is convenient to use the method of separation of variables. By substituting the solution in the form of $\varphi = R(r)\,\Theta(\vartheta)\,\Phi(\varphi)$ into (8), we arrive at the systems of equations

$$\begin{aligned}&\tfrac{d}{dr}\left(r^2\tfrac{d}{dr}R\right)-l(l+1)R=0,\\&\tfrac{1}{\sin\theta}\tfrac{d}{d\theta}\left(\sin^2\theta\tfrac{d\Theta}{d\theta}\right)-\tfrac{m^2}{\sin^2\theta}\Theta+l(l+1)\Theta=0,\\&\tfrac{d^2\Phi}{d\varphi^2}+m\Phi=0,\end{aligned} \tag{9}$$

the solution of the first of them is given by

$$R(r)=Ar^l+\frac{B}{r^{l+1}},$$

while the solutions of the other two equations are Legendre polynomials $P_l^m(\cos\vartheta)$ and exponentials $\exp(\pm im\varphi)$. At $\theta=\pm\pi$, the second equation has finite solutions only at $l \geq |m|$ [3]. For the potentials inside and outside of the sphere, φ_{int} and φ_{ext}, respectively, the well-known expressions are obtained

$$\begin{aligned}&\varphi_{int}=ar^lY_{l,m}(\theta,\phi), \qquad |\varphi_{int}(r=0)|<\infty\\&\varphi_{ext}=r^lY_{l,m}(\theta,\varphi)+br^{-(l+1)}Y_{l,m}(\theta,\phi), \qquad |\varphi_{ext}(r=\infty)|<\infty,\end{aligned} \tag{10}$$

where $Y_{l,m}(\theta,\varphi)$ are the spherical functions:

$$Y_{l,m}(\theta,\varphi)=(-1)^{(m+|m|)/2}i^l\left[\frac{2l+1}{4\pi}\frac{(l-|m|)!}{(l+|m|)!}\right]^{1/2}P_l^m(\cos\vartheta)\,e^{im\varphi}.$$

In a sphere that is placed in an inhomogeneous field of the form of

$$\varphi_0 \sim r^lY_{l,m}(\theta,\phi),$$

multipole moment a arises, which has a pole at

$$\varepsilon_{\text{int}}^{(l)}=-\frac{l+1}{l}\varepsilon_{ext} \tag{11}$$

(problem 2.1). At $l=1$, a dipole resonance occurs, and the field inside of the sphere is homogeneous (see [13, 26]). At $l>1$, the field inside of the sphere is inhomogeneous. With an increase in the order of multipole l, the field inside of the sphere concentrates near the surface,

$$\varphi_{int}=ar^lY_{l,m}(\theta,\phi),$$

and, in this respect, the solution can be called the surface plasmon. At $l\to\infty$, the spacing between poles $\varepsilon_{\text{int}}^{(l)}$ decreases. Moreover, this leads to the appearance of a point of condensation of poles at $\varepsilon_{\text{int}}^{(\infty)} = -\varepsilon_{\text{ext}}$ [3, 6, 11, 12, 14, 24–33].

For a plasmonic sphere with permittivity (11), the excitation frequencies of corresponding resonances may be easily estimated as

$$\varepsilon_{int} = 1 - \frac{\omega_p^2}{\omega^2} = -\frac{l+1}{l}\varepsilon_{ext} \ \Rightarrow \omega^2 = \frac{\omega_p^2}{\left[1 + (l+1)\,\varepsilon_{ext}/l\right]} \ \underset{l\to\infty}{\to} \ \omega_{surf}^2 = \frac{\omega_p^2}{1+\varepsilon_{ext}}. \tag{12}$$

To solve the problem for dielectric cylinder we should consider cylindrical coordinates. The Laplace equation takes the form [15]

$$\frac{1}{r}\frac{\partial}{\partial r}\left(r\frac{\partial \varphi}{\partial r}\right) + \frac{1}{r^2}\frac{\partial^2 \varphi}{\partial \varphi^2} + \frac{\partial^2 \varphi}{\partial z^2} = 0. \tag{13}$$

Assuming that the field is perpendicular to the cylinder axis we can search for solution independent of z. Substituting into the Laplace equation the sought solution in the form of $\varphi = R\,(r)\,\Phi\,(\varphi)$, we arrive at the systems of equations

$$\begin{aligned} &\tfrac{d}{d\,r}\left(r\tfrac{dR}{dr}\right) - \tfrac{m}{r}R = 0, \\ &\tfrac{d^2\Phi}{d\,\varphi^2} + m\Phi = 0. \end{aligned} \tag{14}$$

The first of them has the solution $R\,(r) = Ar^m + \frac{B}{r^m}$, while the solution of the second equation is $\exp\left(\pm m\varphi\right)$. This yields the following well-known expressions for the potentials inside and outside of the sphere, φ_{int} and $\varphi_{ext} + \varphi_0$, respectively:

$$\begin{aligned} &\varphi_{int} = ar^m\cos(m\varphi), \qquad |\,\varphi_{int}\,(r=0)| < \infty, \\ &\varphi_{ext} = br^{-m}\cos(m\varphi), \quad |\,\varphi_{ext}\,(r=\infty)| < \infty. \end{aligned} \tag{15}$$

In a cylinder placed in an external homogeneous field that is described by potential $\varphi_{ext} = \cos\left(m\varphi\right)$, a multipole moment appears, which has a pole at

$$\varepsilon_{int} = -\varepsilon_{ext}, \tag{16}$$

(problem 2.2).

In other words, all the multipoles of the cylinder have resonance at one and the same value of the permittivity and one and the same frequency.

5. Plasmon resonance in a system of particles (plasmonic nanolens)

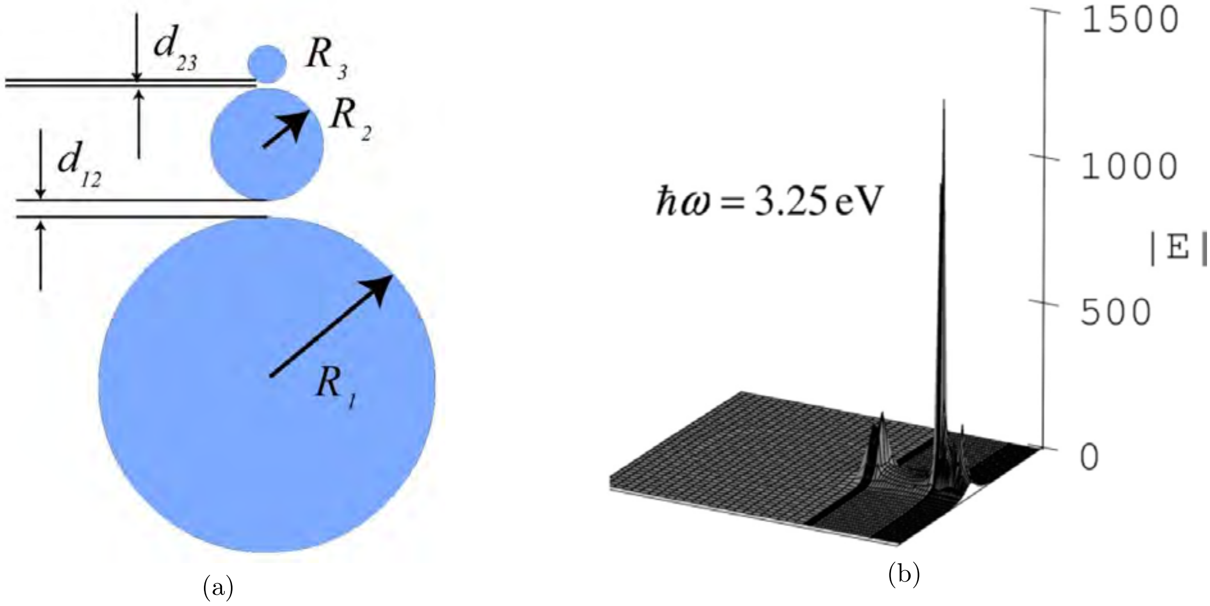
The plasmon resonance can be used to enhance the local field strength [17, 34]. Let us consider a scheme that has been proposed in [34]. In this work, a finite chain of metal nanospheres was examined. Let R_i denote the radius of the ith nanosphere and $d_{i,i+1}$ denote the spacing between the surfaces of the ith and $(i+1)$-th nanospheres. Then, the system is constructed implying that the self-similarity takes place, i.e.,

$$R_{i+1} = \kappa R_i, \qquad d_{i,i+1} = \kappa d_{i+1,i+2} \quad i = 1, 2, ...N,$$

where κ is a certain constant that is smaller than unity (see Fig. 2). At $\kappa << 1$, the local field of the ith nanosphere is only insignificantly perturbed by the field of the $(i+1)$-th nanosphere. Due to the plasmon resonance, the local field near the largest nanosphere is enhanced compared to exciting field E_0 by a factor of Q, where $Q \sim \mathrm{Re}\varepsilon\,(\omega)\,/\mathrm{Im}\varepsilon\,(\omega)$ is the quality factor of the resonance, and $\varepsilon\,(\omega)$ is the relative permittivity of the metal of which nanospheres are made of, and ω is the frequency of the exciting field. The local field of the first, largest nanosphere can be considered to be homogeneous on the scale of the second nanosphere, and it can be treated as an external exciting field. Therefore, near the second nanosphere, the field will be enhanced Q^2 times. By continuing this construction, we find that, near the n-th nanosphere, the local field will be equal to $Q^n E_0 >> E_0$. For example, for a really achievable value $Q \sim 10$ and n equal to three[2], we find that the local field near the smallest nanosphere is $E_n = 10^3 E_0$. If this nanolens is used in Raman spectroscopy, then, in accordance with the presented estimate, the Raman scattering will be enhanced by a factor of $(E/E_0)^4 \sim 10^{12}$ [18].

Really, it is necessary to take into account the mutual influence of nanospheres. This mutual influence manifests itself in that all the nanospheres are involved in the resultant oscillation, so that its frequency becomes different from the resonant frequency of an individual sphere. The main consequence is that the stronger enhancement is obtained for not-too-small , but, when a smaller nanosphere can rather strongly affect the field of a larger nanosphere. In this case, the greatest enhancement is achieved in between the smallest spheres.

[2]The size of the smallest nanosphere is restricted by the electron free path length in the metal; i.e., at optical frequencies, this radius should be chosen to be not smaller than 5 nm.



(a) (b)

Fig. 2 – (a) Geometry of a nanolens consisting of three metal spheres with the radii R_1, R_2, and R_3 being equal to 45, 15, and 5 nm, respectively; the spacings d_{12} and d_{23} between the surfaces of the corresponding spheres are equal to 4.5 and 1.5 nm, respectively. (b) Enhancement coefficient of a local field in relation to the coordinate in the gap between the spheres. The figures were taken from [6, 19]

The efficiency of the proposed scheme was verified in the course of the numerical simulation using the multipole spectral expansion method [21], which is based on the spectral method in the differential form [1].

6. Spatial distribution of the field energy under plasmon resonance conditions

When studying complex systems that consist of several plasmonic particles, it is also necessary to take into account that, at plasmon resonance, the fraction of energy that is contained outside of a plasmonic particle, namely, in the dielectric, is always smaller than the fraction of energy that is concentrated inside of plasmonic particles [2].

Let us consider a limiting losseless case. In accordance with the Kramers–Kronig relations, the permittivity of a metal has the form [25]: $\varepsilon_M = \varepsilon_\infty - \omega_p^2/\omega^2$, where ε_∞ is the constant that does not depend on the frequency. Depending on the shape of a plasmonic particle or on the space distribution of a set of such particles, there is a frequency or the negative value of the permittivity of the metal, at which the plasmon resonance is observed; i.e., there is an eigensolution that differs from zero at a zero external field. A particular feature of this eigensolution is that the field tends to zero at infinity.

The following relation can be written:

$$\begin{aligned} &\int_\Omega \varepsilon \mathbf{E}\cdot\mathbf{E}dV = \int_\Omega \mathbf{D}\cdot\mathbf{E}dV = -\int_\Omega \mathbf{D}\cdot\vec{\nabla}\varphi dV \\ &= -\int_\Omega \left[\nabla\cdot(\varphi\mathbf{D}) - \varphi\vec{\nabla}\cdot\mathbf{D}\right]dV = \oint_{S_\Omega}\varphi\,(\mathbf{Dn})ds = 0 \end{aligned}. \quad (17)$$

Then, by dividing the whole volume into a part that is occupied by the dielectric and a part that contains plasmonic particles and assuming that the permittivity distributions inside of these volumes are homogeneous, we obtain

$$\int_{\Omega_D} \varepsilon_D \mathbf{E}\cdot\mathbf{E}dV = -\int_{\Omega_M} \varepsilon_M \mathbf{E}\cdot\mathbf{E}dV. \quad (18)$$

The energy stored in the plasmonic particle has the form

$$U_M = \frac{1}{8\pi}\int_{\Omega_M} \frac{d\omega\varepsilon_M}{d\omega}\mathbf{E}\cdot\mathbf{E}dV. \quad (19)$$

where

$$\frac{d\omega\varepsilon_M}{d\omega} = \frac{d\left[\omega\left(\varepsilon_\infty - \frac{\omega_p^2}{\omega^2}\right)\right]}{d\omega} = 2\varepsilon_\infty - \varepsilon_M > -\varepsilon_M, \quad (20)$$

that yields

$$U_M = \frac{2\varepsilon_\infty - \varepsilon_M}{8\pi} \int_{\Omega_M} \mathbf{E} \cdot \mathbf{E} dV. \tag{21}$$

The energy stored inside of the dielectric is given by

$$U_D = \frac{1}{8\pi} \int_{\Omega_D} \varepsilon_D \mathbf{E} \cdot \mathbf{E} dV = - \int_{\Omega_M} \varepsilon_M \mathbf{E} \cdot \mathbf{E} dV. \tag{22}$$

For the ratio of these energies, we have

$$\frac{U_M}{U_D} = \frac{(2\varepsilon_\infty - \varepsilon_M) \int_{\Omega_M} \mathbf{E} \cdot \mathbf{E} dV}{-\varepsilon_M \int_{\Omega_M} \mathbf{E} \cdot \mathbf{E} dV} = \frac{(2\varepsilon_\infty - \varepsilon_M)}{-\varepsilon_M} > 1. \tag{23}$$

Assuming that the losses in the plasmonic particle are small and using (18), we can obtain the following estimate for the quality factor of the plasmon resonance, $Q = \frac{\omega(U_M+U_D)}{d(U_M+U_D)/dt}$:

$$Q = \omega \frac{\frac{d[\omega \mathrm{Re}\varepsilon_M(\omega)]}{d\omega} \int_{\Omega_M} \mathbf{E} \cdot \mathbf{E} dV + \varepsilon_D(\omega) \int_{\Omega_D} \mathbf{E} \cdot \mathbf{E} dV}{2\mathrm{Im}\varepsilon_M(\omega) \int_{\Omega_M} \mathbf{E} \cdot \mathbf{E} dV} = \omega \frac{d\mathrm{Re}\varepsilon_M(\omega)/d\omega}{2\mathrm{Im}\varepsilon_M(\omega)}, \tag{24}$$

An impression can be formed that, for the restricted system, the quality factor of the resonance does not depend on the particle shape or on the mutual arrangement of particles. However, the geometrical factor determines the frequency of the resonance, which, in turn, determines the value of ε_M in expression (24) for the quality factor.

To conclude we should emphasize that appearing singularities are related to overidealization of the problem. In the reality, losses are always present in the system, which shifts the frequency of the resonance to the complex domain. At real-valued frequencies, all quantities prove to be finite. Moreover, at low Joule losses, it is necessary to take into account losses for the emission of radiation. In this case, the quasistatic approximation is inapplicable (see [13, 26]), and it is necessary to solve the exact problem [11, 12].

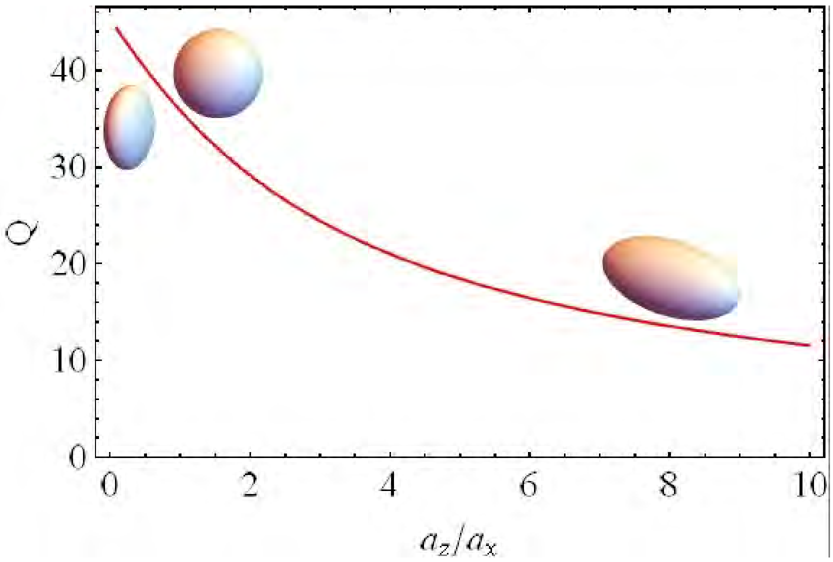


Fig. 3 – Quality factor of the plasmon resonance of an ellipsoid of revolution in relation to the ratio of its axes a_z/a_x (a unit value of this ratio corresponds to a sphere). The material of the ellipsoid is silver

7. The Ferrell solution for a plasmon on a thin film

We note that the above-described enhancement of the field in the nanolens is a collective plasmon resonance in which the electromagnetic oscillation is localized not on a single inclusion [35, 36] but, rather, on a system of noncontacting particles. The manifestation of this phenomenon is most clearly pronounced upon consideration of a plasmonic composite—a dielectric matrix filled by inclusions with a negative permittivity. At certain concentrations, which depend on the magnitude of the negative permittivity, the effective permittivity of the composite has a pole [34] that is related to the excitation of a collective plasmon, which involves all particles and which is not restricted in the volume of the composite. It is clear that the external field at the resonant frequency should lead to the excitation of an infinitely large polarization, i.e., to the appearance of strong local fields.

Thus, we logically pass to the consideration of quasistatic problems for boundaryless objects. In essence, these solutions are transitional between localized and propagating plasmons.

In the late 1950s, works by Ritchie [37] and, later, by Ferrell [38] have been published in which energy losses of an electron beam in a metal foil were considered. It was shown that a part of losses is associated with the excitation of collective oscillations of electrons on the boundary surface of the metal film (see the discussion in [34]).

It turned out that, even in the planar geometry, the equation $\mathrm{div}\,(\varepsilon\mathbf{E}) = 0$ can have solutions that are more complicated than the Langmuir plasmon solution (1). Following [38, 39] consider a metallic layer of thickness 2τ to be situated in the (x, y) plane. Then, it follows from the symmetry of the problem that two solutions should exist at a given value of τ, one of which is symmetric with respect to the plane $z = 0$, while the other solution is antisymmetric. Correspondingly, we will seek a solution of the equation $div\ (\varepsilon\,\mathrm{grad}\,\varphi) = 0$ inside of the layer in the form

$$\varphi_k = \cos{(kx)}\,(e^{kz} \pm e^{-kz}).$$

Using the condition that potential φ_k turns to zero at $|z| \to \infty$ and the continuity condition of the potential on the plate surface, we obtain the following expressions for the value of the potential outside of the plate:

$$\varphi_k^{z>\tau} = \cos{(kx)}\,e^{-kz}(e^{k\tau} \pm e^{-k\tau})e^{k\tau} \text{ at } z > \tau,$$
$$\varphi_k^{z<-\tau} = \cos{(kx)}\,e^{kz}(e^{-k\tau} \pm e^{k\tau})e^{k\tau} \text{ at } z < -\tau.$$

In order to obtain the solution in the whole space, it is necessary to join the normal components of the electric induction, $\varepsilon\partial\varphi/\partial z$, at the boundaries. As a result, we obtain the eigenvalue and eigenfunction problem, which lies in finding a value of the permittivity at which a symmetric plasmon,

$$\varepsilon\,(\omega) = -\frac{(e^{k\tau} + e^{-k\tau})}{(e^{k\tau} - e^{-k\tau})}, \tag{25}$$

and an antisymmetric plasmon,

$$\varepsilon\,(\omega) = -\frac{(e^{k\tau} - e^{-k\tau})}{(e^{k\tau} + e^{-k\tau})}, \tag{26}$$

(see [12]) propagate (see problem 2.3). For the permittivity of the plasma $\varepsilon = 1 - \omega_p^2/\omega^2$, the corresponding frequencies are given by

$$\omega = \omega_p\sqrt{\frac{1 \mp e^{-k\tau/2}}{2}}. \tag{27}$$

8. Field enhancement in an apertureless SNOM

The use of near fields in optical instruments makes it possible to overcome the Rayleigh resolution limit [40, 41]. In particular, near fields that arise upon propagation of waves through small (subwavelength) holes are used in aperture schemes of scanning near-field optical microscopy (SNOM) [25, 42]. In the majority of the schemes of this type, the light enters the system via a tapered optical fiber with a metal-sprayed coating. The resolution of the instrument is determined by the cross-section of the tapered end of the fiber. The subwavelength resolution can be achieved if the fiber end is an evanescent waveguide. In this case, the intensity of near fields in the SNOM aperture configuration is very low, which lowers the sensitivity of the method.

This drawback is eliminated in apertureless methods of SNOM, in which the incident electromagnetic wave excites a plasmon on the metal tip. The nanofocusing of plasmons on the tip end [**?**, 40] makes it possible to create fields of high intensities in a small (subwavelength) region of space [43]. At a specially chosen geometry, the tip ensures the enhancement of the field intensity by a factor of up to 10^4 [44]. As a result of the scattering of the electric field of the plasmon by a sample under study, a far field arises the intensity of which (in some schemes, its phase as well) is registered and is used to retrieve the image of the sample. SNOM systems will make it possible to obtain images with a spatial resolution of about 20 nm [45].

The field on the tip end can be found analytically if the shape of the tip is approximated by a certain simple surface, e.g., by a paraboloid of revolution (a similar solution for a parabolic cylinder was presented in [46]). In this case, it is convenient to pass to the parabolic coordinate system [47]:

$$\begin{aligned} x &= \sigma\tau\cos\varphi, \\ y &= \sigma\tau\sin\varphi, \\ z &= \left(\tau^2 - \sigma^2\right)/2. \end{aligned} \tag{28}$$

The surfaces of constant values of coordinates and are paraboloids of revolution, which are defined, respectively, as

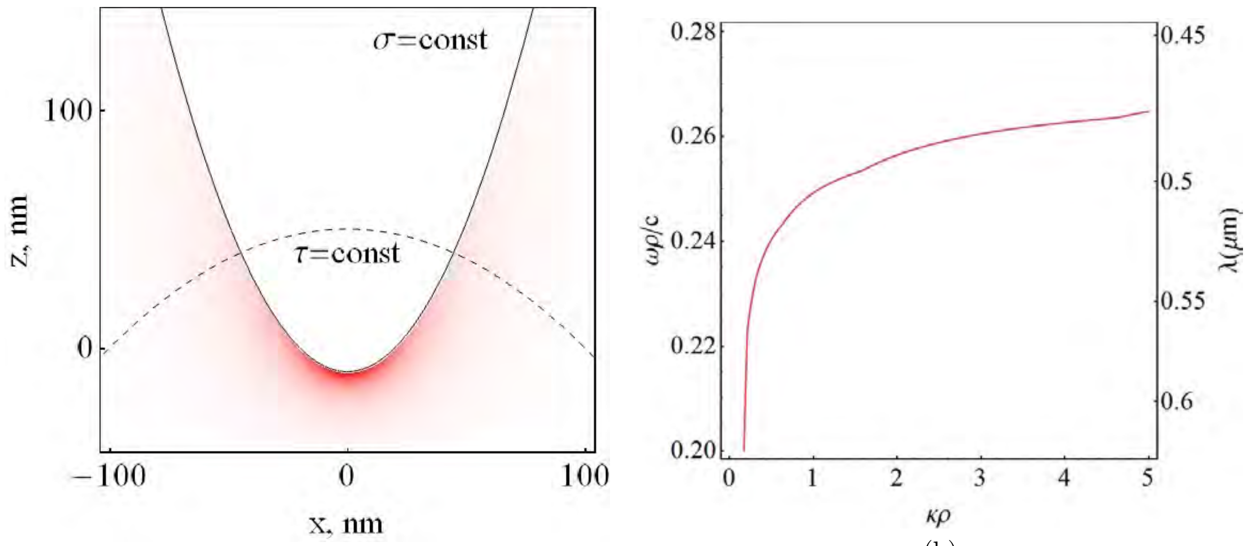
$$z = \left(x^2 + y^2\right)/2\sigma^2 - \sigma^2/2 \tag{29}$$

and

$$z = -\left(x^2 + y^2\right)/2\tau^2 + \tau^2/2 \tag{30}$$

(see Fig. 4a).

The surface of the metal tip is one of paraboloids with a constant value of σ. It follows from (29) that the value of σ is connected with the radius of curvature of the tip end ρ_0 by a simple relationship $\sigma = \sqrt{\rho_0}$.



(a) (b)

Fig. 4 – (a) Distribution of squared field modulus $|\nabla\Phi|^2$ in a plasmon wave on the tip end; (b) dispersion curve of a plasmon on a gold tip with a radius of the tip end of $\rho = 20$ nm

If the radius of curvature of the paraboloid is assumed to be much smaller than the wavelength, $k_0\rho_0 << 1$, the problem can be solved in the electrostatic approximation; i.e., the electric potential can be found from the Laplace equation $\Delta\Phi = 0$ taking into account that Φ and $\varepsilon\partial\Phi/\partial n$ are continuous on the tip surface and that Φ turns to zero at infinity.

In parabolic coordinates, the Laplace equation for axially symmetric solutions, which do not depend on azimuthal angle φ of the sought solution, takes the form

$$\frac{1}{\sigma}\frac{\partial}{\partial\sigma}\left(\sigma\frac{\partial\Phi}{\partial\sigma}\right) + \frac{1}{\tau}\frac{\partial}{\partial t}\left(\tau\frac{\partial\Phi}{\partial\tau}\right) = 0. \tag{31}$$

Then, we will separate variables $\Phi(\sigma,\tau) = S(\sigma)\,T(\tau)$ and denote the separation constant by κ ($\frac{1}{S}\frac{1}{\sigma}\frac{\partial}{\partial\sigma}\left(\sigma\frac{\partial S}{\partial\sigma}\right) = \frac{1}{T}\frac{1}{\tau}\frac{\partial}{\partial\tau}\left(\tau\frac{\partial T}{\partial\tau}\right) = -\kappa$). As will be shown below, the choice $\kappa > 0$ ensures an exponential decrease of the solution along the line $\tau = \text{const}$, i.e., with increasing distance from the tip.

Function $T(\tau)$ is the solution of the zero-order Bessel equation. The solution that is bounded on the tip end has the form $T = J_0\left(\sqrt{\kappa}\tau\right)$. Function $S(\sigma)$ is the solution of the modified zero-order Bessel equation. Inside of the tip, the solution that is bounded at zero has the form $S\left(\sigma < \sqrt{\rho_0}\right) = I_0\left(\sqrt{\kappa}\sigma\right)$, whereas, outside of the tip, one should choose the solution that decreases at infinity, $S\left(\sigma > \sqrt{\rho_0}\right) = \alpha K_0\left(\sqrt{\kappa}\sigma\right)$. The unknown coefficient is determined from joining the conditions $S\left(\sqrt{\rho} - 0\right) = S\left(\sqrt{\rho} + 0\right)$ and $\varepsilon S'\left(\sqrt{\rho} - 0\right) = S'\left(\sqrt{\rho} + 0\right)$, which lead to the dispersion equation for $\kappa(\omega)$:

$$\varepsilon(\omega)\frac{I_0{}'\left(\sqrt{\kappa\rho}\right)}{I_0\left(\sqrt{\kappa\rho}\right)} = \frac{K_0{}'\left(\sqrt{\kappa\rho}\right)}{K_0\left(\sqrt{\kappa\rho}\right)}. \tag{32}$$

The corresponding dispersion curve is shown in Fig. 4b, while the field distribution is presented in Fig. 4a.

We note that, on the surface of the metal,

$$\sigma = \sqrt{\rho_0},$$

the solution has an oscillating character, as in the Ferrell problem. With increasing distance from the tip in the direction along the tip axis,

$$\tau = \sqrt{2z + \rho} \propto \sqrt{2z},$$

the potential takes the form

$$\Phi = I_0\left(\sqrt{\kappa\rho}\right) J_0\left(\sqrt{2\kappa\left(\rho + z\right)}\right) \propto sin\left(\sqrt{2\kappa z}\right) / (2\kappa z)^{1/4}.$$

Therefore, the behavior of the field has an oscillating character. However, unlike the exponentially decreasing Ferrell solution, this solution decreases at infinity according to a power law, which is related to the change in the radius of curvature of the tip along the z axis. The concentrating (focusing) of the field on the tip end is called the nanofocusing effect.

The oscillating character of the solution can be described by introducing the local wavenumber $k = \partial\sqrt{2\kappa z}/\partial z = \sqrt{\kappa/2z}$. The electrostatic solution presented above is valid only in the range in which the wavenumber $k >> \omega/c$. Therefore, the electrostatic approach is applicable in the neighborhood of the tip: $k_0 z << \kappa/2k_0$.

In the literature, other shapes of the tip are also considered, which refer to various schemes of scanning tunneling microscopy (STM). Thus, in [48], the dispersion of plasmons excited between the tip in the shape of a hyperboloid and a metal plane was determined. As in the case considered above, plasmons have a continuous spectrum and the field concentration due to the tip effect.

STM schemes that operate with localized plasmons were also realized. Thus, in [38], the sample was “probed” by the field of a plasmon localized on a nanoparticle that is placed on the tip of an optical fiber. The plasmon is excited by a far field. The sensitivity and the resolution of this scheme are on the same order as those discussed above.

5. Conclusions

Thus, plasmonics has passed a long way from first notes of longitudinal plasma resonances to contemporary applications in microscopy and sensing. The main feature of plasmonics, resonance in subwavelength structures, is manifested in enhancement of Raman scattering (SERS), perspective plasmonic lines and interconnects. We hope that simplified electrostatic approach adopted here allowed deep understanding of plasmonics.

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КВАЗИСТАТИЧЕСКИЕ (ЛОКАЛИЗОВАННЫЕ) ПЛАЗМОНЫ: ОТ ЛЭНГМЮРА ДО ФЕРРЕЛЯ

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Аннотация

Проведен обзор основных результатов по электростатическим аспектам плазмоники. Хотя многие применения плазмоники требуют электродинамического подхода, плазмонный резонанс имеет электростатическую природу. В этой статье мы сосредоточились на основах плазмоники, которые легче понять в электростатическом приближении. Мы также затрагиваем историю первых представлений в области резонансов в субволновых электромагнитных системах.

Ключевые слова: плазмоника, локализованные плазмоны, плазмонный резонанс, электростатическое приближение
