ОБМІН ПРАКТИЧНИМ ДОСВІДОМ ТА ТЕХНОЛОГІЯМИ

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ON BASIC FEATURES OF EM SIGNAL TRANSPORTING ALONG LINEAR WAVEGUIDES COMPOSED OF NOBEL METAL NANOSPHERES

Abstract – Propagation of surface dipole oscillations along a linear chain composed of spherical nanoparticles of noble metals is considered. Within the RPA method applied to the "jelly model" it was calculated the EM signal longitudinal group velocity $v_{gr,z}$ and the effective length L_z of the signal "run" along waveguides composed of Au (Ag, Cu) nanospheres at room temperature and the most frequently used parameters of such waveguides (nanoparticle radius a = 25 nm, with an interparticle spacing of 75 nm, merged into vacuum): the values are in good agreement with experimental data. The fact can be used to transfer information over relatively large distances: L_z > 1 μm, i.e. over the distances which exceed 200 periods of the nanoscale linear structure.

Keywords: surface dipole oscillations, metal nanoparticles, nanoscale waveguide.

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ОБ ОСОБЕННОСТЯХ РАСПРОСТРАНЕНИЯ ЭМ СИГНАЛА ВДОЛЬ ВОЛНОВОДА, СОСТАВЛЕННОГО ИЗ НАНОСФЕР БЛАГОРОДНЫХ МЕТАЛЛОВ

Рассмотрено распространение поверхностных дипольных колебаний вдоль линейной цепочки, составленной из сферических наночастиц благородных металлов. В рамках метода RPA применительно к «модели желе» вычислены продольная скорость прохождения ЭМ сигнала и эффективная длина «пробега» сигнала вдоль наноразмерного волновода при комнатной температуре и наиболее часто используемых параметрах такого рода волноводов (радиус наносфер 25 нм, расположенных в вакууме эквидистантно с расстоянием 75 нм): полученные расчеты находятся в хорошем согласии с экспериментом. Этот факт может быть использован для передачи информации на сравнительно большие расстояния: L_z > 1 мкм, т.е. на расстояния, превышающие 200 периодов такой линейной структуры.

Ключевые слова: поверхностные дипольные колебания, металлические наночастицы, наноразмерный волновод.

Introduction

Investigations of plasma oscillations in metallic nanoparticles has been discussed for a relatively long time and are of great practical value (nanophotonics and optical telecommunications). An important role in such systems assigned to nanoscale waveguides which are possible basis for development of new generation of optical communication systems [1-3]. One-dimensional ordered arrays of noble metal (Au, Ag, Cu) spherical nanoparticles[1] are increasingly seen as this kind of waveguides. It is well established that the nanoparticles of noble metals support plasmon resonances within a wide frequency range (UV → visible → near IR) [1-3].

When studying plasma oscillations in nanoparticles as small as 1 nm or less, methods of local density functional theory (LDA) and "a time-dependent density functional" (TDLDA) are generally used. When studying plasma oscillations in metallic nanoparticles of several tens of nanometers "random phase approximation" (method RPA) can be successfully used [4-6].

Most often, both theoretically and experimentally someone investigates the properties of plasma waveguides composed of noble metal nanoparticles of spherical shape (for example, silver and gold nanosphere of radius 25 nm and 50 nm) and of cylindrical form ("nanorods") of similar size [7].

Rather recently there were several experimental studies of plasma waveguides [8]. Within these studies there were published micrographs obtained by means of optical near-field microscopy, which clearly show the passage of an electromagnetic signal along nanochains of gold spheres (of the radius a = 50 nm): the signal passes along the waveguide with almost no damping, though the total length of nanoscale waveguides were more than $5 \cdot 10^{-6}$ m with following parameters: a = 50 nm (the radius of the nanospheres), d = 200 nm (d stands for the distance between the centers of metal nanospheres), ratio d/a = 4 [8].

The experimenters associate this observation with a point dipole model that can be applied when describing the transport process of the electromagnetic signal by a plasma waveguide, and they give their estimate of the group velocity and attenuation coefficients for one-dimensional arrays of nanoparticles. Within this model of a point dipole some characteristics of such waveguides were calculated using FDTD method. But the authors themselves recognize that there is a significant divergence between the calculations of linear damping coefficient for the waveguide (~3dB/15nm), presented in the above-mentioned papers, and the experimentally observed results (see [9]).

Micrograms obtained with a scanning tunneling microscope clearly show that the electromagnetic signal passes through the Au nanochain a considerable distance (the length of about 6.75×10^3 nm) without any significant

[1] so-called *nanochains*

attenuation. Very similar result to this experimental one was obtained when passing an electromagnetic signal along nanochain of gold spheres with a diameter of 100 nm [9]: it is evident that the electromagnetic signal while passing along such nanochains slightly damped over a length of at least of 5×10^3 nm.

The author investigates plasma (dipole) waves, which arise within a linear array made of metallic spherical nanoparticles. It is shown that the dipole eigenwaves generated in the linear array, decay with time exponentially. The waves which arise in the chain due to the external electromagnetic wave source (generator), do not decay eventually, the frequencies of these plasma waves equal to the frequencies of the external alternating field, and their amplitudes depend on these frequencies. The greatest amplitudes of the plasma waves appear at the resonance frequencies.

The aim of this work is the calculation of certain characteristics of nanoscale waveguides in the form of a chain composed of nanospheres of noble metals with the following parameters: the radius of a nanosphere is $a = 25$ nm; the nanochain length is L_{chain} ($L_{chain} >> a$). It was assumed that metal nanospheres are arranged equidistantly with the distance d between the centers of the nearest particles, and that nanochain merged into a medium with dielectric constant of ε_h .

Theory

Consider an infinite chain composed of noble metal nanoparticles of spherical shape of radius a and merged in a dielectric medium with the permittivity ε_h . We suppose that the nanospheres are arranged along an axis Z at an equal distance of $d > 2a$ from each other (i.e., their centers are spaced at equal distances from each other). Let the origin of coordinates is located at the center of one of the particles, for example at the nanosphere with index $l = 0$. Further suppose that there is an external source of electric field, which is located on one of the particles of the nanochain, for example, on the particle with index $l = 0$. At the center of the particle with $l = 0$ this electric field produces a point dipole moment which, in its turn, will also radiate a certain electric field.

Let the intensity of the radiated electric field at any point of the axis Z at any given time t is equal to $\vec{E}_0(0;z;t)$, in this case $\vec{E}_0(0;z;t) = \vec{E}_0(0;-z;t)$.

Then projection of the considered electric field intensity onto the coordinate axe Z at each l -th node of the chain, where are located metal nanoparticles, will be determined by the following relations: $\vec{E}_{0z}(0;ld;t) = \lim\limits_{z\to ld} E_{0z}(0;z;t)$.

It means that at the center of each metal nanoparticles (see [5, p. 124322, Eq. (27)]) it will arise additional dipole moments, committing with time compelled transverse and longitudinal vibrations.

According to the general theory (see [9]) dipole moment $\vec{D}(\vec{R},t)$ located at the point $\vec{R}$ radiates an electric field intensity at a point $(\vec{R}+\vec{R}_0)$ is equal to:

$$\vec{E}(\vec{R};\vec{R}_0;t) = \frac{1}{\varepsilon_h}\left(-\frac{1}{R_0^3} - \frac{1}{R_0^2}\frac{1}{v_h}\frac{\partial}{\partial t} - \frac{1}{R_0}\frac{1}{v_h^2}\frac{\partial^2}{\partial t^2}\right)\times$$

$$\times\,\vec{D}\left(\vec{R};t-R_0/v_h\right) + \frac{1}{\varepsilon_h}\left(\frac{3}{R_0^3} + \frac{3}{R_0^2}\frac{1}{v_h}\frac{\partial}{\partial t} + \frac{1}{R_0}\frac{1}{v_h^2}\frac{\partial^2}{\partial t^2}\right)\vec{n}_0\cdot\left(\vec{n}_0\cdot\vec{D}\left(\vec{R};t-R_0/v_h\right)\right),$$

where $\vec{n}_0 = \vec{R}_0/R_0$, $v_h = c/\sqrt{\varepsilon_h}$.

Plasma oscillations arising on the particle with index l also emit electromagnetic field, which is accompanied by power emitted by the influence of the field on the conduction electrons of the particle with index l (Lorentz force). The presence of this force is equivalent to the presence of an external electric field effective, the source of which is located in the center of the particle with index l , and the intensity of this field is given by the expression (see [9]):

$$\vec{E}_L(ld,0,t) = \frac{2e\sqrt{\varepsilon_h}}{3c^3\varepsilon_h}\frac{\partial^3\vec{D}(ld,t)}{\partial t^3} = \frac{2}{3d^3\varepsilon_h}\left(\frac{d}{v_h}\right)^3\frac{\partial^3\vec{D}(ld,t)}{\partial t^3}.$$

Thus, the equations describing the behavior of the dipole moment $\vec{D}_z(ld,t)$ along the longitudinal direction (Z axis) of the particle with index l are of the form (see [5, p. 124322, see Eq. (28-29)]):

$$\frac{\partial^2 D_z(ld,t)}{\partial t^2} + \frac{2}{\tau_0}\frac{\partial D_z(ld,t)}{\partial t} + \omega_1^2 D_z(ld,t) = \varepsilon_h a^3\omega_1^2\left(\sum_{\substack{m=-\infty\\(m\neq l)}}^{+\infty} E_z(R_m,R_{ml},t) + E_{Lz}(ld,0,t) + E_{0z}(0,ld,t)\right) \quad (1)$$

,

where $\tau_0 = \left(\frac{v_F}{2\lambda_0} + \frac{v_F}{2a}\right)^{-1}$ is the decay time of plasma oscillations due to their interaction with phonons of bulk metal and surface of the nanoparticles.

For the particle with index m ($m \neq l$), equations (1) can be rewritten as

$$\frac{\partial^2}{\partial t^2} D_z(ld,t) + \frac{2}{\tau_0}\frac{\partial}{\partial t} D_z(ld,t) + \omega_1^2 D_z(ld,t) =$$

$$= 2\omega_1^2 \frac{a^3}{d^3}\left[\sum_{\substack{m=-\infty \\ (m\neq 0)}}^{+\infty}\left(\frac{1}{|m|^3} + \frac{d}{v_h |m|^2}\frac{\partial}{\partial t}\right)\cdot D_z\left(md + ld; t - \frac{|m|}{v_h}d\right) + \frac{d^3}{3v_h^3}\frac{\partial^3}{\partial t^3} D_z(ld;t)\right] + \varepsilon_h a^3 \omega_1^2 E_{0z}(0;ld;t)\,.$$

Here $l \cdot d$ is the discrete variable. To solve these equations, assume that $D_z(ld,t) = \lim\limits_{z\to ld} D_z(z,t)$, where z is the continuous independent variable, and functions $D_z(z,t)$ satisfy the following equations:

$$\frac{\partial^2}{\partial t^2} D_z(z,t) + \frac{2}{\tau_0}\frac{\partial}{\partial t} D_z(z,t) + \omega_1^2 D_z(z,t) =$$

$$= 2\omega_1^2 \frac{a^3}{d^3}\left[\sum_{\substack{m=-\infty \\ (m\neq 0)}}^{+\infty}\left(\frac{1}{|m|^3} + \frac{d}{v_h |m|^2}\frac{\partial}{\partial t}\right)\cdot D_z\left(md + z; t - \frac{|m|}{v_h}d\right) + \frac{d^3}{3v_h^3}\frac{\partial^3}{\partial t^3} D_z(z;t)\right] + \varepsilon_h a^3 \omega_1^2 E_{0z}(0;z;t)\,.$$

Here $\omega_1^2 = \frac{\omega_p^2}{3\varepsilon_h}$ is the frequency of plasma oscillations in a medium where the nanoparticles are placed;

ω_p is the eigenfrequency of plasma oscillations of the electron gas of the nanoparticles.

If the condition

$$\frac{1}{\omega_1 \tau_0} \sim \left(\frac{\omega_p a}{c\sqrt{3}}\right)^2 \leq \left(\frac{a}{d}\right)^3 << 1\,,$$

is satisfied, these equations become much simpler and for functions $\widetilde{D}_z(k,t)$ in a first approximation they take the form:

$$\left(\frac{\partial^2}{\partial t^2} + \frac{2}{\tau_0}\frac{\partial}{\partial t} + \widetilde{\omega}_z^2(kd)\right)\widetilde{D}_z(k,t) = \varepsilon_h a^3 \omega_1^2 \widetilde{E}_{0z}(k,t)\,. \qquad (2)$$

The frequencies of $\widetilde{\omega}_z(kd)$ have the following properties: $\widetilde{\omega}_z^2(kd) = \widetilde{\omega}_z^2(-kd) = \widetilde{\omega}_z^2(kd + 2\pi)$.

It is well known, that the general solution of the inhomogeneous equation (2) with constant coefficients equals the sum of two functions (see [9]): $\widetilde{D}_z(k,t) = \widetilde{D}_{0z}(k,t) + \widetilde{D}_{1z}(k,t)$, where $\widetilde{D}_{0z}(k,t)$ is the general solution of the homogeneous equation as it follows:

$$\left(\frac{\partial^2}{\partial t^2} + \frac{2}{\tau_0}\frac{\partial}{\partial t} + \widetilde{\omega}_z^2(kd)\right)\widetilde{D}_{0z}(k,t) = 0\,, \qquad (3)$$

and the functions $\widetilde{D}_{1z}(k,t)$ are particular solutions of inhomogeneous equations (2).

Solutions of equations (3) have the form (see [9]):

$$\widetilde{D}_{0z}(k,t) = A_z(k)\cdot\exp\left(-i\widetilde{\omega}'_z t - t/\tau_0\right) + B_z(k)\cdot\exp\left(i\widetilde{\omega}'_z t - t/\tau_0\right),$$

where $\widetilde{\omega}'_z = \widetilde{\omega}'_z(kd) = \sqrt{\widetilde{\omega}_z^2(kd) - 1/\tau_0^2}$, and $A_z(k)$ and $B_z(k)$ are arbitrary functions of k determined from both the initial and physical conditions. Consider the case when an external electric field is turned on at the moment $t = 0$. Since before the event the system under consideration was in a state of equilibrium and it was free of any excitations, it is necessary to assume that $A_z(k) = B_z(k) = 0$. In this case $\widetilde{D}_{0z}(k, t=0) = 0$, and when $t > 0$ we obtain:

$$\widetilde{D}_z(k,t) = \widetilde{D}_{1z}(k,t) = \varepsilon_h \frac{a^3\omega_1^2}{\widetilde{\omega}'_z(kd)}\frac{1}{\exp(t/\tau_0)}\cdot\int_0^t \exp(t_1/\tau_0)\widetilde{E}_{0z}(k,t_1)\cdot\sin\left(\widetilde{\omega}'_z(kd)\cdot(t-t_1)\right)\cdot dt_1\,,$$

here t_1 is time, and besides $t_1 \neq t$.

The desired function $D_z(z,t)$, according to (2) is defined by the equation $D_z(z,t) = \int_{-\infty}^{+\infty} e^{ikz}\widetilde{D}_z(k,t)dk$.

In the linear array, we will primarily be interested in the electric dipole moment of the metal nanospheres located in the l-th node of the chain, and the dipole moment is defined by the following relationship: $D_z(ld,t) = \lim_{z\to ld} D_z(z,t) = \lim_{z\to ld} D_{1z}(z,t) = D_{1z}(ld,t)$.

Next we suppose that the external electric field is turned off at the moment of T. Under the conditions $z = ld$ and $t > T$ the mentioned above short-term effect of external electric field on the dipole moment of the l-th nanospheres of the nanochain takes the form (see [9]):

$$D_z(ld,t) = \frac{\varepsilon_h a^3 \omega_1^2}{\widetilde{\omega}'_z(\pi)} \frac{2\Delta k}{\exp(t/\tau_0)} \times$$

$$\times \mathrm{Im}\left[F_z(k_0,T) \frac{\sin\left(ld - t\Delta k \cdot \left.\frac{d\widetilde{\omega}'_z}{dk}\right|_{\pi/d} \right)}{ld - t\Delta k \cdot \left.\frac{d\widetilde{\omega}'_z}{dk}\right|_{\pi/d}} \right] \cdot \left[\exp\left(i\widetilde{\omega}'_z(\pi)\cdot t + ik_0 ld\right) + \exp\left(i\widetilde{\omega}'_z(\pi)\cdot t - ik_0 ld\right)\right], \quad (4)$$

here we have denoted: $k_0 = \Delta k = \pi/d$.

Results

From Eq. (4) we can see that the function is a wave packet: the last factor here is so-called a "rapidly-oscillating" function, and the factor in square brackets before is the amplitude of the changing with time dipole moment of the nanoparticle with index l.

This maximum of the amplitude can be achieved at the moment $t_l = ld/v_{gr,z}$, where $v_{gr,z} = \left(\frac{d\widetilde{\omega}'_z(kd)}{dk}\right)\Big|_{k=\pi/d} = \frac{\tau_0\omega'_z(kd)}{\sqrt{(\tau_0\cdot\omega'_z(kd))^2 - 1}} \cdot \left.\frac{d\omega'_z(kd)}{dk}\right|_{k=\pi/d}$ is the group velocity of the wave packet, at which it moves along the nanochain.

The dipole moment $D_z(ld,t)$ of the particle with index l after termination of external influence, i.e. at $t > T$, has the following form:

$$D_z(ld,t) = \varepsilon_h \frac{a^3\omega_1^2}{\widetilde{\omega}'_z(\pi)}(-1)^l \cdot \frac{\sin\left(\widetilde{\omega}'_z(\pi)\cdot t\right)}{\exp(t/\tau_0)} \cdot \frac{\sin\left(\pi l - \pi\cdot v_{gr,z}t/d\right)}{\pi\cdot\left(l - v_{gr,z}t/d\right)} \cdot \int_0^T \exp(t_1/\tau_0) E_{0z}(0;0;t_1)dt_1, \quad (5)$$

From the eq. (5) one can see that the dipole moment of the l-th nanoparticle reaches its maximum value at $t = t_l = ld/v_{gr,z}$, and the maximum value at this moment is directly proportional to the value of $\exp(-t_l/\tau_0)$. Hence the distance $L_z = v_{gr,z}\cdot\tau_0$, on which a signal can be transmitted along a linear array of nanoparticles, is about of $L_z \sim l\cdot d$.

Calculations of some characteristics of nanoscale waveguides were carried out *ab initio* within the RPA method and theoretical positions, published in [6, 10]. After Fourier transformation of Eq. (5) one can get the following relation for the longitudinal group velocity $v_{gr,z}$:

$$v_{gr,z} = \frac{2d\cdot\omega_1^2\cdot\sum_{m=1}^{\infty}\frac{\sin(m\cdot kd)}{m}\cdot\left(\frac{\cos(m\cdot\omega_1 d/v_h)}{m} + \omega_1 d/v_h\cdot\sin(m\cdot\omega_1 d/v_h)\right)}{\left(\frac{d}{a}\right)^3\cdot\sqrt{\omega_1^2\cdot\left[1 - 4\cdot\left(\frac{a}{d}\right)^3\cdot\sum_{m=1}^{\infty}\frac{\cos(m\cdot kd)}{m^2}\cdot\left(\frac{\cos(m\cdot\omega_1 d/v_h)}{m} + \omega_1 d/v_h\cdot\sin(m\cdot\omega_1 d/v_h)\right)\right] - \frac{1}{\tau_0^2}}}.$$

The results of calculations are presented in the Table 1.

Table 1

The results of calculations of certain parameters of nanoscale waveguides (T = 300 K):

nanoparticle material	the signal group velocity $v_{gr,Z}$	the lifetime of the wave packet τ_0, sec	the length of the signal propagation L_z, μm
Ag	(0,1600±0,0002)×$\boldsymbol{c}$	$2{,}42\times10^{-14}$	1,16
Au	(0,1580±0,0002)× $\boldsymbol{c}$	$2{,}14\times10^{-14}$	1,01
Cu	(0,0480±0,0001)× $\boldsymbol{c}$	$1{,}91\times10^{-14}$	0,27

Note #1: here $\boldsymbol{c}$ – the speed of light in vacuum. These characteristics of such excitations are in good agreement with the experimental data [11-13].

Note #2: for presented in the Table #1 calculations it was taken the following basic conditions: a = 25 nm; d/a = 3; temperature = 27 °C; dielectric medium is vacuum.

Summary

In this paper, the subject of the study was the spread of EM signal along a one-dimensional array composed of noble metal spherical nanoparticles. If external electric field source will be placed amongst such ensemble of nanoparticles, and one of the particles would be affected by a short pulse, then in this linear array the pulse will cause additionally arising of dipole waves with a frequency lower than the plasma one. As it was shown, under certain parameters of described nanoscale waveguides undamped dipole oscillations may occur in the form of a wave packet. The fact was confirmed experimentally [3, 6] and it can be used to transfer information over relatively large distances: L_{chain} > 1 μm, i.e. over the distances which exceed 200 periods of the nanoscale linear structure.

It was calculated the signal longitudinal group velocity $v_{gr,z}$ and the characteristic length of the signal propagation along the linear array of nanoparticles L_z, and at the same time good agreement between calculations and experimental results was achieved. The presented results clearly show that the longitudinal group velocity of EM signal propagation along Ag or Au nanochains much more (in ≈3.3 times more) than along Cu ones of the same parameters (see Table 1).

One can see the perspective use of the described nanoscale waveguides: 1) light energy conversion inside semiconductor photodiode systems with active nano-modified surface; and/or 2) transportation of EM energy and information within respective nano-optical devices.

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