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Plasmonic extraordinary transmittance in array of metal nanorods

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Abstract The optical response of an array of metal nanorods is studied in the case when the cylinders almost touch by their generatrices. As the cylinders approach each other, a series of surface plasmon resonances are excited. The first longitudinal mode is different from the higher-order lateral modes. The lateral resonances occur near the frequency where the real part of the metal permittivity changes sign. The plasmon resonances result in maxima and minima in the reflectance and transmittance. The resonances also result in a huge enhancement of the local electric field in the gap between cylinders.

1 Introduction

The fundamental optical properties of nanometer-size metal particles have been intensively studied for the last hundred years [1, 2]. Until recently, most effort focused on statistically large numbers of particles in disordered arrays. Modern technology has allowed research and design of regular chains and arrays of metal nanoparticles. The investigation of the periodic chains started in the seminal works [3, 4]. The surface plasmon (SP) excitations in the ordered one-dimensional arrays of nanoparticles have attracted significant attention in recent years due to numerous potential applications in nanoplasmonics [5–7]. The chains of nanoparticles can be used for transmitting and processing optical signals on a scale much smaller than the wavelength λ. The plasmon modes propagating in the chain, where the radius a of the particle is on the order of the distance δ between particles, are investigated in most detail (see e.g., [8]). In this mode the dipole excitation jumps between particles due to the near field interaction. Electromagnetic (em) energy concentrates in the vicinity a of the chain. Guided modes, where the em field propagates in the region $\sim\lambda \gg a$ around the chain, were investigated in [9–11]. Scattering and diffraction of the electromagnetic wave on a periodical array of separated metal nanorods was experimentally investigated, for example, in the papers [12, 13]. Recently much attention attracts problem of the wave propagation along metal nanorods since the optical negative refraction was obtained in these systems [14–16]. Stacked nanorods are also proposed for the superlensing, i.e., imaging with subwavelength resolution [17–23]. The field distribution in the close-packed array of nanoparticles and nanoshells were considered in the works [24] and [25], respectively. Our results are in qualitative agreement with these computer simulations.

2 Local electric field between nanorods

We investigate various SP modes propagating in the gap between closely packed metal nanorods provided the distance δ between them is much smaller than the radius a, as is shown in Fig. 1. The rods are infinitely long. The lateral

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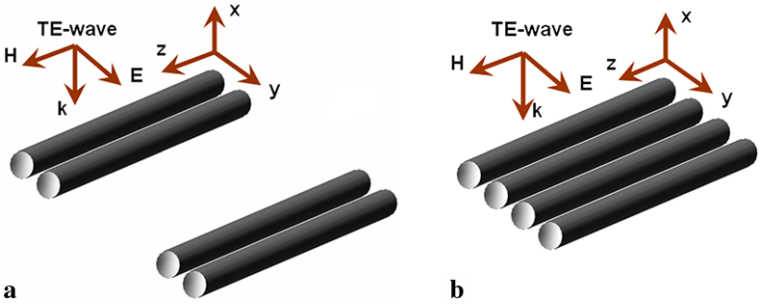

Fig. 1 (**a**) TE-wave propagation through array of pairs (dimers) of nanocylinders. (**b**) TE-wave propagation through array of closely packed nanocylinders

mode propagates in the direction perpendicular to the axis connecting the centers of the cylinders. The electric field of the SP concentrates in the gaps between cylinders. Thus the local electric field can be controllably concentrated on the sub nanometer scale, which is much smaller than the radius a. The excitation of the SP results in a series of resonances in the optical transmittance of the periodic planar ($2d$) array of metal cylinders whose period is much smaller than wavelength λ of the incident light (Fig. 1b). The enhancement of the local electric field and corresponding plasmonic extra ordinary transmittance (EOT) depends on the ratio δ/a, while the period $2a+\delta$ of the nanostructure can be arbitrary small. The modes propagating around the metal nanoparticles have been observed in the computer simulations [26, 27]. SP oscillations in a cluster of two nearly touching metallic nanospheres, resonant optical antennas, and crescent shaped cylinders, were discussed in the works [28–30], [31, 32], and [33] correspondingly.

At the beginning we consider periodic array of cylinder dimers shown in Fig. 1a. The lateral size l of the region between two cylinder, where the SP is localized, estimates as $l=\sqrt{\delta(4a+\delta)}/2\cong\sqrt{a\delta}$ so that $\delta\ll l\ll a$. This estimate follows from the conformal map

$$w=u+iv=\ln\big[\big(x+i(y-l)\big)\big/\big(x+i(y+l)\big)\big], \tag{1}$$

which gives the potential u of two perfect conductors axially aligned cylinders, provided the centers of the cylinders are placed at the coordinates $x=0, y=\pm(a+\delta/2)$ and the cylinders have charges $1/2$ and $-1/2$ per unit length. The conformal map (1) transforms the plane $\{x, y\}$ into the strip $0<v<2\pi$. The left end of the strip ($u<-u_0, u_0=\operatorname{arcsinh} l$) represents upper cylinder. The right dead end ($u>u_0$) represents the lower cylinder. Then the capacitance between cylinders equals to $C=1/(4\operatorname{arcsinh} l)$per unit length [34].

The map (1) solves the problem of the field distribution between two metal or dielectric cylinders excited by the external electric field $\mathbf{E}_0=\{0,E_0,0\}$. The centers of the cylinders are still placed at $y_{1,2}=\pm(a+\delta/2)$ and we assume that the $y_{1,2}$ cylinders have permittivity ε_m while the space between cylinders is filled by dielectric with permittivity ε_d. To find the electric field inside and between the cylinders it is enough to express the potential $\Phi_0=izE_0=E_0l\coth(w/2)$ of the external field in terms of the harmonics $\exp(\pm kw)$, namely $\Phi_0=E_0l[1+2\sum_{k=1}^{\infty}\exp(-kw)]$ for $0<u<u_0$, and $\Phi_0=-E_0l[1+2\sum_{k=1}^{\infty}\exp(kw)]$ for $-u_0<u<0$. Then the complex potential Φ takes the following form: $\Phi=\sum A_k\exp(kw)$ for $u<-u_0$, $\Phi=\Phi_0+\sum[B_k\exp(kw)+C_k\exp(-kw)]$ for $-u_0<u<u_0$, and $\Phi=\sum H_k\exp(-kw)$ for $u>u_0$. The coefficients A_k, B_k, C_k, and H_k, we find by matching the real part of the potential $\varphi=\mathrm{Re}[\Phi(w)]$ at $u=\pm u_0$ and matching normal components of the electric displacement $D_n=\varepsilon\mathrm{Re}[d\Phi(w)/dw]|dw/dz|$ at $u=\pm u_0$. It follows from the above equations that the length l is the characteristic length for the field distribution and that the electric field oscillates as $E\propto B_k\exp(ikv)-C_k\exp(-ikv)$ along the x axis ($u=0$) between the cylinders. Thus we obtain equation for the resonance frequencies ω_n:

$$\mathrm{Re}\big[\varepsilon_m(\omega_n)\big]=-\varepsilon_d\big(\gamma^{2n}+1\big)\big/\big(\gamma^{2n}-1\big), \tag{2}$$

where $\gamma=(l/a)[1+\sqrt{1+(a/l)^2}]\sim 1+l/a$. Let us consider, for example, the resonance in the dimer of two silver nanorods in the vacuum ($\varepsilon_d=1$). We approximate the metal optical permittivity ε_m in (2) by the Drude formula $\varepsilon_m=\varepsilon_b-(\omega_p/\omega)^2/(1+i\omega_\tau/\omega)$, where the optical constants for silver are taken from experimental results [35, 36]: $\varepsilon_b=5, \omega_p=9.1$ eV, $\omega_\tau=0.02$ eV. The cylinders in the dimer are separated by the distance $\delta=0.2a$. Then the resonance frequencies equal to 3.35, 3.6, 3.67, ... eV (cf. Fig. 3).

We use the COMSOL Multiphysics package to calculate the electric field for the dimers of almost touching nanorods shown in Fig. 1a. In Fig. 2 we compare the analytical solution for the enhancement of the electric field in the gap between the nanorods with results of electrodynamics simulations. In the quasistatic limit $D=2a\ll\lambda$ numerical and analytical results are in a good agreement. We observe huge enhancement of the local field. The frequency dependence of the enhancement is shown in Fig. 3, where we see the series of the resonances. Note that positions of the resonances can be tuned by variation of the ratio δ/a.

Consider now the planar array of closely packed nanorods (Fig. 1b). The presence of other cylinders cannot qualitatively change the field distribution between two neighboring

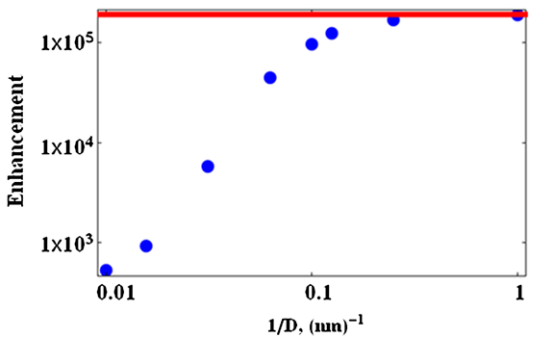

Fig. 2 Electric field enhancement $|E_m(\omega_1)/E_0|^2$ in the central point between nanocylinders as function of the cylinder diameter D: $\delta/D = 0.1$; E_0 is amplitude of the incident field; distance between dimers 300 nm; *red line* corresponds to analytical results: $\omega = 3.35$ eV; *blue disks* results of numerical simulation: for each D frequency ω_1 is fitted to maximum enhancement: $\omega_1 = 3.34$ eV for $D = 10$ nm, $\omega_1 = 2.85$ eV for $D = 100$ nm

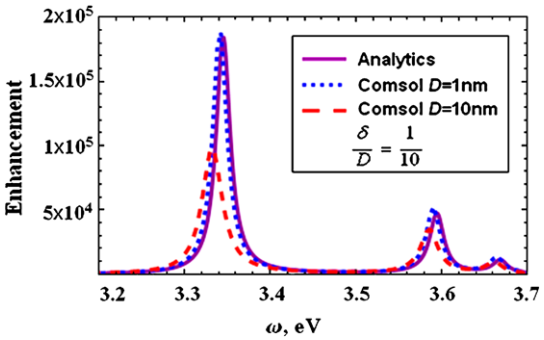

Fig. 3 Comparison of analytical (*purple line*) and numerical (*blue and red dashed lines*) enhancement $|E_m/E_0|^2$ of the electric field in the middle of dimer; $\delta/D = 0.1$, diameter of nanorods in COMSOL simulations is $D = 10$ nm, 1 nm; distance between dimers 300 nm; E_0 is amplitude of the incident field

cylinders since we consider the case when $l \ll a$. Therefore, l remains to be the lateral field characteristic length in the array of the metal nanorods. Since we consider the metal nanorods where $a \ll \lambda$ we use the Laplace equation $\nabla^2\varphi = 0$ to calculate the SP potential φ. Then the potential in the gap between the cylinders ($|x| < l$) approximately is

$$\varphi_{\text{out}} = \varphi_0 \exp(i\beta x)\sinh(\beta y)/\sinh(\beta\delta/2) \tag{3}$$

and inside the metal

$$\varphi_{\text{in}} = \varphi_0 \exp(i\beta x)\exp\big(-\beta|y|\big). \tag{4}$$

Matching the solutions (3) and (4) at $y = \pm\delta/2$ we obtain the dispersion equation $\coth(\beta\delta/2) \cong 2/\beta\delta = -\varepsilon_d/\varepsilon_m$, where we take into account that $\beta\delta \propto \delta/l \ll 1$. The resonance condition $2\beta l \cong n, n = 1, 2, \ldots$ gives the resonance values of the metal permittivity

$$\varepsilon_m(\omega_n) \cong -\varepsilon_d a/(nl), \tag{5}$$

which coincides with (2) in the limit $\delta/a \ll 1$. Thus we obtain the estimate of the resonance frequencies ω_n for the

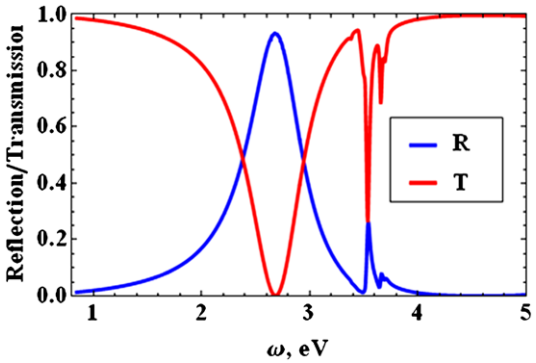

Fig. 4 Reflectance $R(\omega)$ (*blue line*) and transmittance $T(\omega)$ (*red line*) of array of silver cylinders with radius $a = 5$ nm and inter-rod spacing $\delta = 1$ nm

lateral SP resonances in the array of the metal nanocylinders. In the case of the metal sphere the radial distribution is given by the Bessel function $\varphi = \varphi_0 J_n(\beta r)\exp(in\varphi)$ and the resonance condition is $\beta l \propto \mu_{np}$, where μ_{pn} is a pth zero of the Bessel of the order $n = 1, 2, \ldots$. Therefore the resonance frequencies ω_n for the chain or array of the touching metal nano spheres can be estimated from the condition $\varepsilon_m(\omega) \cong -\mu_{pn}^{-1}(a/l)$. Note that the potential distribution between two spheres can be found from Laplace equation, which can be solved by separation of variables in the bispherical system of coordinates [37, 38].

3 Reflection and transmission in array of nanorods

The discussed resonances result in anomalous optical behavior of the array of metal nanorods shown in Fig. 1b. The centers of the cylinders are placed at coordinates $x = 0$, $y = \delta/2 + a \pm n(\delta + 2a)$, $\delta \ll a$. The light impinges normal to the film. The surface conductivity is defined as $\langle\mathbf{j}\rangle = \sigma\langle\mathbf{E}\rangle$, where current density $\mathbf{j}$ and local electric field $\mathbf{E}$ are averaged over the layer $-a < y < a$. The transmittance and reflectance estimate as $T \cong |1/(1 + 2\pi\sigma/c)|^2$ and $R \cong |(2\pi\sigma/c)/(1 + 2\pi\sigma/c)|^2$, respectively, where c is the speed of light. At a resonance the current density and, therefore, the surface conductivity is much enhanced. The absolute value of the conductivity σ reaches a maximum. Then the reflectance also has a maximum while the transmittance goes down. The full scale electrodynamics simulations reveal maxima and minima in reflectance and transmittance as is shown in Fig. 4. The reflectance R as a function of the frequency has three distinguishable peaks. The first reflection maximum corresponds to the longitudinal SP, where current flows through the cylinders (see Figs. 5a and 6a) while the next two maxima are due to the excitation of the lateral SP (see Figs. 5b, c and 6b, c). The film becomes almost opaque at the resonance frequencies. Out of a resonance the surface conductivity σ decreases. Correspondingly the

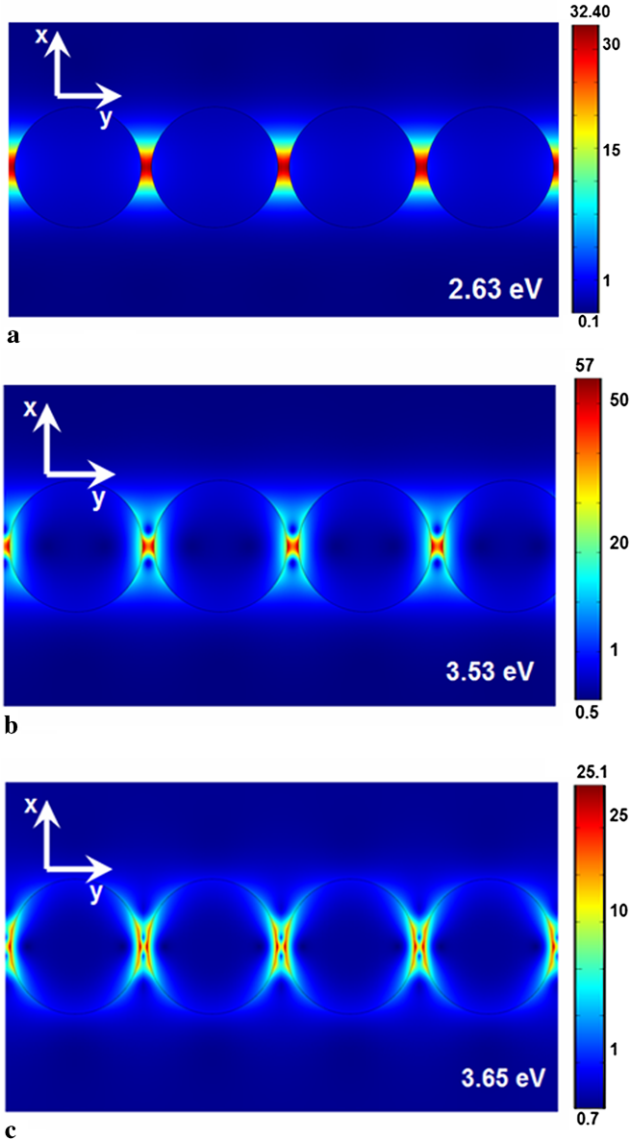

Fig. 5 Electric field distribution $|E(x, y)/E_0|$. Visualization of multiple surface plasmon resonances in array of silver cylinders (normalized to the amplitude of the incident field E_0): (**a**) $\omega = 2.63$ eV, $a = 5$ nm, $\delta = 1$ nm; (**b**) $\omega = 3.53$ eV, $a = 5$ nm, $\delta = 1$ nm; (**c**) $\omega = 3.65$ eV, $a = 5$ nm, $\delta = 1$ nm

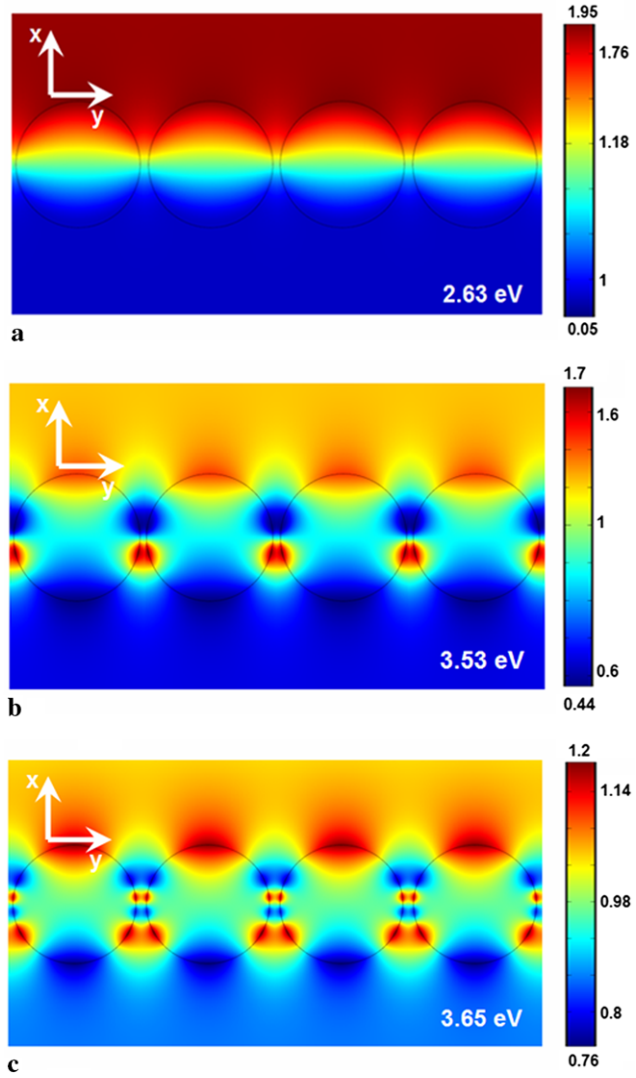

Fig. 6 Magnetic field distribution $|H(x, y)/H_0|$. Visualization of multiple surface plasmon resonances in array of silver cylinders (normalized to the amplitude of the incident field H_0): (**a**) $\omega = 2.63$ eV, $a = 5$ nm, $\delta = 1$ nm; (**b**) $\omega = 3.53$ eV, $a = 5$ nm, $\delta = 1$ nm; (**c**) $\omega = 3.65$ eV, $a = 5$ nm, $\delta = 1$ nm

transmittance $T(\omega)$ has maxima between the resonance frequencies (Fig. 4).

The resonance enhancement of the electric and magnetic fields between the nanorods can been seen from Figs. 5 and 6. The visualization of the electric field in Fig. 5 reveals excitation of multiple plasmon resonances. The red color corresponds to the maximum amplitude of electric component of TE-wave while the blue color corresponds to the minimum amplitude. System of nanorods exhibits SP modes which are strongly localized around the rods. The collective SP resonance is similar to the "whispering gallery" modes [38].

4 Conclusions

We propose that the array of metal nanorods has anomalous optical properties corresponding to the excitation various SP resonances. The computer simulations as well as the analytical theory are presented. The resonance electric field is much enhanced in the gaps between metal nanorods. The field concentrates at the scale much smaller than the diameter of a rod. We speculate that the resonance frequencies and field enhancement can be tuned by variation of the shape and arrangement of the metal nanoparticles.

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