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LOCALIZED SURFACE PLASMONS OF GOLD NANOPARTICLES

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Abstract. In this paper, gold nanoparticle far-field and near-field optical responses are studied and simulated numerically. The electromagnetic field was excited by an electric dipole near one end of the nanorod, which is used to model the emission of a quantum dot. Another excitation method was also simulated in which an incident plane wave was used. The excitation of dark plasmon modes of the gold nanorod is presented. The Poynting equation was solved numerically to study the influence of the gold nanorod on the dipole radiative power. In addition, the extinction cross section of the gold nanoparticle illuminated by the incident plane wave was calculated to estimate the amount of the scattered and absorbed light.

Keywords: biosensing, extinction cross section, nanoparticle, plasmon, Raman scattering

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ЛОКАЛИЗОВАННЫЕ ПОВЕРХНОСТНЫЕ ПЛАЗМОНЫ НАНОЧАСТИЦ ЗОЛОТА

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Аннотация. В статье изучены и численно промоделированы оптические характеристики наночастиц золота в специально возбужденных электромагнитных полях (дальнем и ближнем). Электромагнитное поле возбуждали электрическим диполем вблизи одного из концов наностержня, который служил для моделирования излучения квантовой точки. Для моделирования другого метода возбуждения была использована падающая плоская волна. Представлено возбуждение темных плазмонных мод золотого наностержня. Чтобы установить влияние такого наностержня на мощность дипольного излучения, было численно решено уравнение Пойнтинга. Кроме того, с целью оценки количества рассеянного и поглощенного света было рассчитано поперечное сечение экстинкции наночастицы золота, освещенной падающей плоской волной.

Ключевые слова: биочувствительность, поперечное сечение экстинкции, наночастица, плазмон, комбинационное рассеяние

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1. Introduction

Surface plasmons were first observed by Wood in 1902 [1] as unexplained features in the reflection spectra of metallic particles; later, Mie [2] proposed the theory of light scattering by nanoparticles. The theoretical description of the energy loss encountered by electrons traveling in metals, given by Pines [3], was considered the first explanation of the surface plasmons. Pines introduced the term plasmons to describe the free electron oscillations in metals responsible for the energy loss. After that, Ritchie [4] theoretically investigated the electron energy loss in thin metal layers, and the term 'surface plasmons' was introduced. Since then, surface plasmons have found enormous applications in different fields such as biosensors, solar cells, and subwave length imaging [5].

Plasmons in metals are described as coherent oscillations of the electrons in the conduction bands [6]. Plasmons either propagate along the surface of the metal (surface plasmons) or inside the metal (bulk plasmons). Illuminating metals allows for coupling photon energy to the oscillated electrons and exciting surface plasmons polaritons (SPPs) [7]. SPPs can be either propagating waves, which occur at the interface between metal and dielectric, or a localized SPP in small-sized particles (smaller or comparable to the photon wavelengths). The propagating SPPs are confined at the metal – dielectric interface and decays exponentially based on the dielectric and metal refractive indices. The propagating SPPs can be excited by illuminating the metal – dielectric interface by free-space light wave only if the SPPs and light wave's propagation constants are coinciding. Different arrangements were proposed to create coupling between light and SP to excite the propagating SPPs [8].

Localized surface plasmons (LSPs) are the most outstanding optical property of small-sized metallic particles. The electromagnetic (EM) field of light will excite the conducting electrons to produce collective oscillations, which are the origin of these optical properties. EM field results in the displacing of conducting electrons from the positive metal lattice. However, the attraction force will act as a restoring force to return the cloud of electrons to its original levels. A resonance behavior will lead to the excitation of LSP, as shown in Figs. 1 and 2. A comparison between the propagating and localized SPP shows that the EM wave of the propagating SPP is decaying slower than that of the localized one. In addition, the propagating SPPs (PSPPs) have a frequency range, while the localized SPPs (LSPPs) are discrete modes. LSPPs differ from the PSPPs in the excitation process. While the PSPPs require a special arrangement for light coupling, LSPPs are excited directly by illuminating nanoparticles with free-space light. The similarity between LSPPs and PSPPs is that both are lossy modes. The frequency of LSPPs modes depends on the geometry of the particle and metal type, while the shape and size of the particle are of great importance to determine its resonance frequency. The frequency band for spherical nanoparticles (NPs) is located in the visible band. At the same time, that of cylindrical NPs is near red shifted. In addition, the quality factor of modes for gold nanorods is higher than that of spherical counter part due to the lower ohmic loss in the near red band compared to the visible region.

In this paper, LSPPs are studied and modelled numerically to estimate the effect of the excitation of the LSP on the far and near field optical response of spherical and nanorod particles. Spherical NPs are studied first, and the absorption cross section is calculated. Excitation of LSPs in small-sized spherical particles is of great importance in fabricating low-cost solar cells. The effect of particle shape was also studied by simulating nanorods. Excitation of the LSPs in nanorods leads to an enhancement of the electric field results from a dipole in close proximity of nanorods, which can be used in many applications based on the enhanced Raman scattering.

The rest of this paper is organized as follows: Section 2 is devoted to introducing the theoretical modelling of the localized surface plasmons. In Section 3, the numerical results were introduced and discussed. Finally, the conclusions are in Section 4.

2. Theoretical modelling of LSPPs

Spherical NPs. Surface plasmons are excited by the attractive force resulting from the interaction of light and free electrons of metallic nanoparticles. To analyze SPP modes, Maxwell's equations need to be solved using the appropriate boundaries. However, the resulted mathematical equations will not describe what the SPPs are. To understand the physical implication of SPPs, let us consider the particle illuminated by the electromagnetic field of light (see Fig. 1).

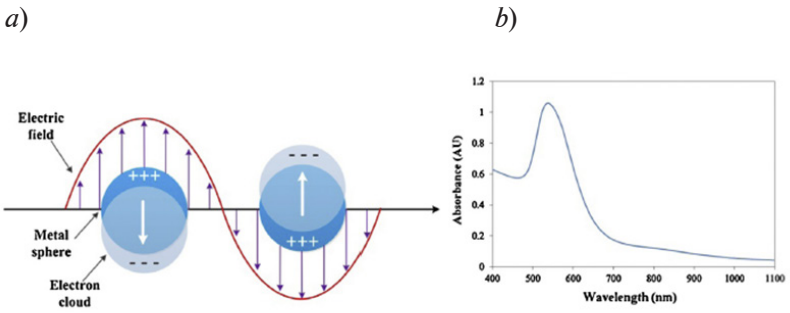

Fig. 1 [9]. The electrons' oscillations due to the plane EM wave: schematic representation (*a*) and the absorption spectrum (*b*)

This field will lead to accumulating electrons on one side, leaving the positive charge on the other side in a similar manner of an electric dipole. The electric field inside the particle will be generated due to the dipole effect opposite to that of the light to restore the electrons to its equilibrium position. For the oscillated electrons, the kinetic and electrostatic energies result from the incident light, therefore the excitement of SPP inside the nanoparticle leads to the partial extinguishing of the light due to the conservation of energy. Calculating the optical absorption spectrum provides a good method to notice the excitation of the SPPs. Absorption cross section of a particle is a measure of the absorption efficiency; if a given nanoparticle absorbs half of the photons hitting its surface, then the absorbing cross section is the half of its geometrical sector. In addition to the absorbing, light also can be scattered in different directions and the scattering cross section can be calculated. The sum of scattering and absorption cross sections is called the extinction cross section, which is of great importance for nanoparticles SPPs calculations.

To obtain the scattering cross section of spherical metallic NPs, the ratio of the total radiated power of the dipole to the intensity of the exciting wave was calculated in Ref. [9] as below.

The scattering cross section is

$$\sigma_{scatter} = \frac{8\pi}{3} k^4 a^6 \left| \frac{\varepsilon_m - \varepsilon_d}{\varepsilon_m + 2\varepsilon_d} \right|^2, \quad (1)$$

where k is the magnitude of the wave vector of the incident light, ε_m and ε_d are the permittivity of the metal and its surrounding respectively, and a is the radius of the spherical NP.

Using the Poynting's theorem [10], the absorption cross section is

$$\sigma_{abs} = 4\pi k a^3 I_m \left(\frac{\varepsilon_m - \varepsilon_d}{\varepsilon_m + 2\varepsilon_d} \right). \quad (2)$$

Finally, the extinction cross section is defined as the sum of the absorption and scattering cross sections, as below:

$$\sigma_{ext} = \sigma_{abs} + \sigma_{sca}. \quad (3)$$

Exploiting SPPs in metallic nanoparticles based solar cells can increase the efficiency of solar panels, the large scattering cross section of NPs allows for high scattering of light at the surface

of the panel. As a result, the absorption layer will decrease, leading to a big reduction in the fabrication cost [11].

Nanorods. The surface plasmon is strongly affected by the shape of nanoparticle since the restoring force of the accumulated electrons depends mainly on the particle geometry as shown in Fig. 2. In this figure, the electrons accumulated along the rod axis generate different plasmons (longitudinal plasmon) from that of perpendicular direction (transversal plasmons) [12].

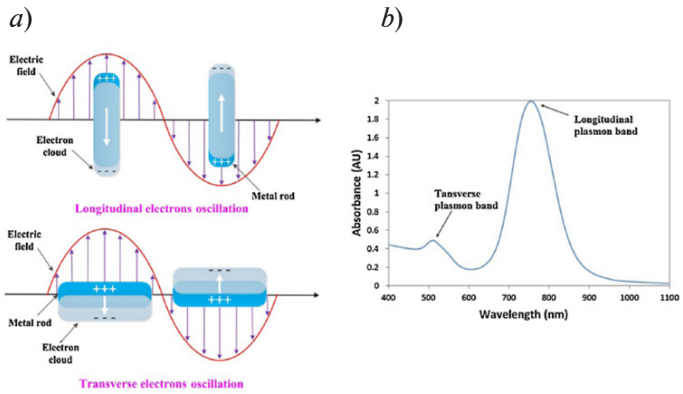

Fig. 2 [12]. The electrons' longitudinal and transverse oscillations due to the plane EM wave: schematic representation (*a*) and the absorption cross section (*b*)

For transversal plasmons, the resonant frequency is slightly higher than that for spherical NPs and it is insensitive to the change of aspect ratio, while that of the longitudinal plasmons is lower when the aspect ratio of the nanorod increases. The longitudinal plasmon bands are highly affected by the dielectric properties of the metallic NP as well as the surrounding medium, and the excitation of longitudinal plasmon results in a high absorption of light. The extinction cross section can be calculated based on the Gan's theory [11] as below:

$$\sigma_{ext} = \frac{2\pi V N \varepsilon_m^{3/2}}{3\lambda} \sum_j \frac{\left(\frac{1}{P_j^2}\right)\varepsilon_i}{\left(\varepsilon_r + \left(\frac{1-P_j}{P_j}\right)\varepsilon_m\right)^2 + \varepsilon_i^2}, \tag{4}$$

where P_j is the depolarization factor and V is the particle volume.

The depolarization factor for nanorods is described as below:

$$P_{length} = \frac{1-e^2}{e^2}\left[\frac{1}{2e}\ln\left(\frac{1+e}{1-e}\right) - 1\right], \tag{5}$$

$$P_{width} = \frac{1-P_{length}}{2}, \tag{6}$$

where e is given by

$$e^2 = 1 - (\text{aspect ratio})^{-2}, \tag{7}$$

and the aspect ratio is the length/width ratio of the nanorod.

Eq. (7) shows that any change in the aspect ratio will lead to a high change in the plasmon band as shown in Fig. 2.

An important application of the longitudinal plasmons is the enhancement of emitters scattering near the nanorods. It is described as the surface-enhanced Raman scattering (SERS) which is called the lightning rod effect. The nanoantenna is another application for optical nanorods where the fluorescence of an emitter placed close to the nanorod can be enhanced. Finally, due to the high sensitivity of the nanorods optical response to the refractive index of the surrounding medium, nanorods are used in biosensing applications.

Section 3 will present a numerical modelling of both a spherical NP and a nanorod made of gold. The scattering and absorption cross sections for the spherical NP will be calculated numerically and the SERS in the gold nanorod is modelled.

3. CST simulation of nanoparticles

A 3D simulation software is used to numerically calculate the optical properties of a nanoparticle exposed to an EM field. CST is a 3D EM solver that solves Maxwell's equation in both the time domain with Finite integration method (FIT) and frequency domain with Finite element method (FEM). A spherical nanoparticle and cylindrical nanorod were simulated using frequency domain solver as below.

Spherical NPs simulation. A spherical gold nanoparticle is modelled first using CST with a plane wave source to model the electromagnetic field of light. The radius of nanoparticle was taken as 20 nm to calculate the absorption cross section numerically. For plasmonic applications, two quantities should be calculated. First, the electric field distribution on the outer surface of the spherical NP and the other one is the absorption cross section. Fig. 3 shows the electric field distribution due to the excitation of LSP, while Fig. 4 shows the absorption cross section of the gold nanoparticle. As shown in Fig. 3, the excitation of the LSP results in an increase in field strength at the NP surface. This plasmon is called bright plasmon due to the scattering of light. In the other hand, Fig. 4 shows clearly an increase in light absorption due to the light coupling with the surface plasmon. Comparison of Figs. 3 and 4 with Fig. 1 shows a good agreement with the published results and allows for using the CST in the modelling of gold nanorods as presented in the next section.

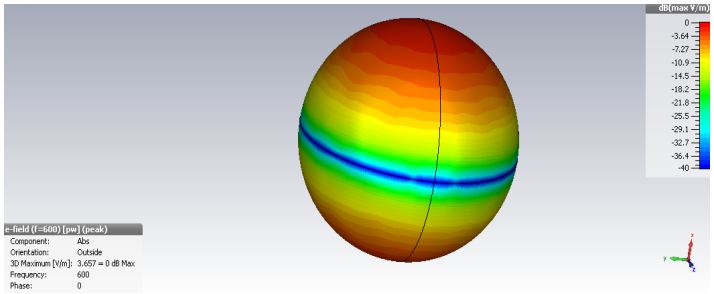

Fig. 3. The electric field distribution on the surface of a spherical nanoparticle

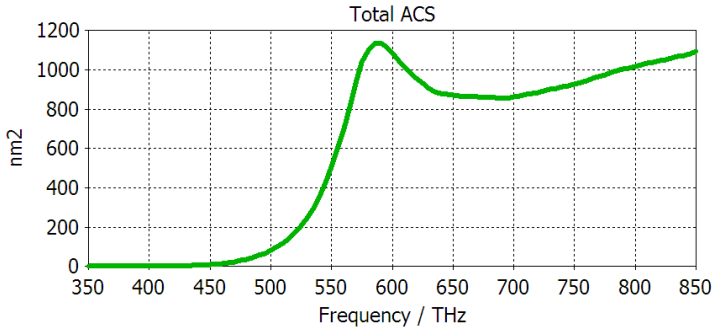

Fig. 4. The absorption cross section of a spherical nanoparticle

Nanorods simulation. In Section 2, a localized surface plasmon has been shown to be excited in the spherical NP by a plane wave. Illuminating a nanorod by a plane wave will result in the excitation of the bright plasmons, which is recognized by the light scattering. Other plasmons, which will not be coupled to the radiating field and being called 'dark plasmons', are simulated in this section. To excite dark plasmons, an emitting source was placed near the nanorod. A gold nanorod was modelled as a circular cylinder, then the upper and lower edges were selected to perform a blend function in the CST to obtain the two hemispherical shape caps. The CST model consisted of a gold nanorod with a length of 100 nm and a radius of 5 nm. This nanorod was excited by a small dipole of 3 nm in length located at a distance of 0.25 nm from the apex

of the nanorod. A frequency range from 200 to 650 THz was used for simulation with a normal background and open boundaries.

The first estimation of the nanorod effect on the electric field distribution is shown in Fig. 5, *a*, where the field pattern was taken at a distance of 100.25 nm from the dipole without the presence of the nanorod while Fig. 5, *b* shows the enhancement of the electric field at the same point with the nanorod. It can be seen that the excitation of the LSP improves the level of the electric field at the same probe point.

In the presence of the gold NP, two types of power of the dipole can be calculated, one is the radiative (*r*) power that is modelled theoretically by the Poynting vector [12] (see Eq. (8) below). The other type of power is the non-radiative (*nr*) one, which is also mathematically represented by the Poynting vector as in Eq. (9) below:

$$P_r = \frac{1}{2}\mathrm{Re}\iint(\mathbf{E}\times\mathbf{H})\,ds, \tag{8}$$

$$P_{nr} = \frac{1}{2}\mathrm{Re}\iint(\mathbf{E}\times\mathbf{H})\,ds. \tag{9}$$

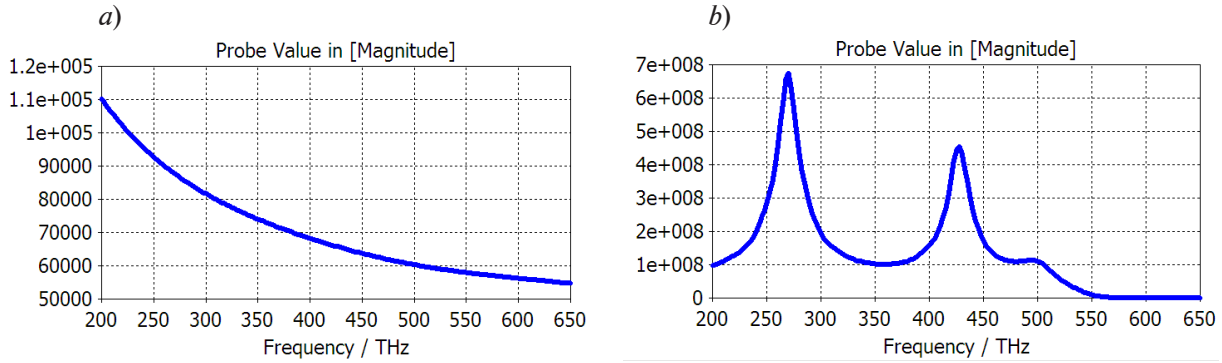

Fig. 5. The electric field patterns without nanorod (*a*) and with its presence (*b*) at a distance of 100.25 nm from the dipole

The area here is the closed surface that encloses both the nanorod and the dipole. To solve Eqs. (8) and (9) numerically using CST, a closed surface was modelled and the radiative and non-radiative powers were calculated. The radiative power of the dipole was simulated numerically by creating a surface containing both the NP and dipole, while the non-radiative power due to the ohmic loss in the gold NP was calculated on the surface of NP only.

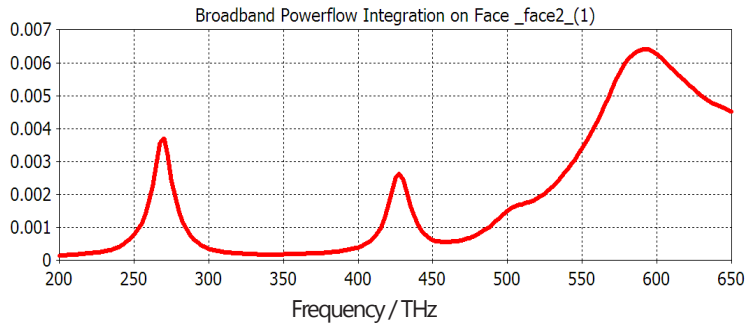

Fig. 6. The power leaving the port

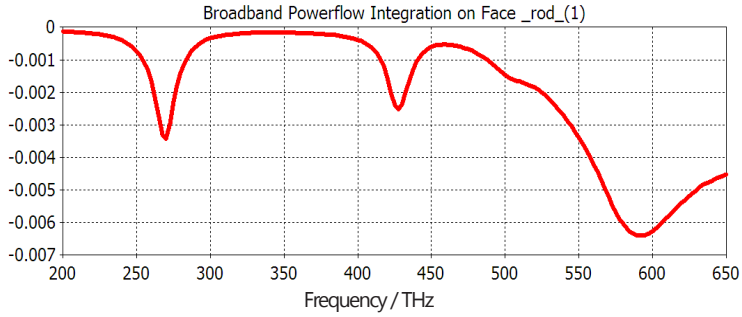

Fig. 7. The non-radiative power which represents the power absorbed by the nanorod

Fig. 6 shows the numerical solution of Eq. (8) where the power leaving the port was calculated on the face surface containing the emitter and the nanorod, while Fig. 7 shows the non-radiative power due to the nanorod absorption. Fig. 7 shows a negative power since the power was calculated as a power flow out to the outer surface. Finally, the radiative power is shown in Fig. 8 where the far field displays one plasmon that decays due to radiation.

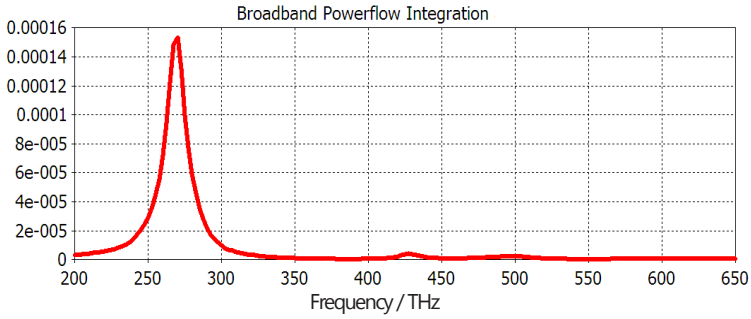

Fig. 8. The radiative power from the power flow

4. Conclusion

Surface plasmons are of great interest in many fields of technology ranging from solar cell to optical biosensing. In this work, localized surface plasmon has been studied and simulated numerically. Spherical nanoparticles and nanorods (both objects were made of gold) were considered, and theoretical and numerical modelling of localized surface plasmons in the gold nanoparticles were presented. An absorption cross section of these nanoparticles illuminated by a plane wave were calculated. Exciting of the LSP was shown by observing the electric field on the surface of the nanoparticle. The nanorods were also modelled and simulated to estimate the enhancement of near and far fields induced by a small dipole close to the nanorod. The enhancement of the electric field was shown to be due to the excitation of the LSPs.

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