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Resonant surface plasmons of a metal nanosphere treated as propagating surface plasmons*

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On the assumption that the resonant surface plasmons on a spherical nanoparticle are formed by standing waves of two counter-propagating surface plasmon waves along the surface, by using Mie theory simulation, we find that the dispersions of surface plasmon resonant modes supported by silver nanospheres match with those of the surface plasmons on a semi-infinite medium-silver interface very well. This suggests that the resonant surface plasmons of a metal nanosphere can be treated as a propagating surface plasmon wave.

Keywords: surface plasmons, surface waves, scattering of particles, dispersion relations

PACS: 73.20.Mf, 91.30.Fn, 52.25.Tx, 11.55.Fv **DOI:** 10.1088/1674-1056/27/6/067302

1. Introduction

Surface plasmon polaritons (SPPs) are electromagnetic (EM) excitations propagating at the interface between a conductor and a dielectric material, evanescently confined in the perpendicular direction. These EM surface waves arise via the coupling of the EM fields to oscillations of the conductor electron plasma. SPPs are usually divided into two different kinds: one is localized surface plasmons and the other is propagating surface plasmons, depending on whether the wavevector has a real part. The localized multiple surface plasmon resonances in one-dimensional metallic nanostructures such as nanowires, nano rods, and nano rice, can be considered as Fabry–Pérot resonance of the propagating SPPs, which has been studied intensively.[1–7] Several studies focused on the surface plasmon dispersion relation of spherical metal nanoparticles or a curved metal–dielectric interface. In 1978, Ogale *et al.* studied the surface plasmon resonant frequencies versus the radius of spherical metal nanoparticles from 0.5 nm to 6 nm.[8] However, their results only showed the quantum effect in very small particles (clusters), which was confirmed by recent studies.[9] A quasi-analytical method has been proposed to study the waves propagating along generally curved smooth interfaces of metal and dielectric medium.[10] Liaw *et al.* recently studied the surface plasmon waves propagating along a curved metal–dielectric interface and on big metallic nanoparticles (larger than 400 nm in size), and obtained the dispersion relations of the surface plasmon waves creeping along a curved interface.[11–13] However, as usually considered, big nanoparticles originally support propagating surface plasmon waves. Later, Guasoni's excellent study extended the propagating surface plasmon waves to a 200-nm diameter nanoparticle which is usually considered to only support localized surface plasmons.[14] Nordlander *et al.* also obtained the plasmon dispersion relation of a planar thin metal film from the plasmon resonance of a metallic nanoshell with a limit of infinite radius.[15] In this paper, we extend this further. With Mie theory simulation of silver spherical nanoparticles, the dispersion relation of the SPPs is obtained and analyzed. It is found that the dispersion curve of localized SPPs on a spherical nanoparticle excellently matches with that of propagating SPPs on the plane interface of metal and dielectric medium, where the radius of the nanoparticle is finite, ranging from several hundreds of nanometers to 10 nm.

2. Assumptions

First we define that when a spherical metal nanoparticle with radius r, embedded in a homogeneous medium, is resonant at certain wavelengths, the orders (dipole or multiples) of different resonant modes are expressed with $m = 1$ (first order, dipole), 2 (second order), 3 (third order), ..., as shown in Figs. 1(a) and 1(b). Then we assume that for each order, when the surface plasmon resonance happens, a standing wave is formed by two SPP waves propagating clockwise and counterclockwise along the surface. So the wavelength of the wave will be

$$\lambda_m = \frac{2\pi r}{m}. \quad (1)$$

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When $m = 1$ (dipole resonance), the wavelength of the surface plasmons equals the perimeter of the sphere, i.e., $2\pi r$. We also assume that the resonant surface plasmons are equivalent to those with the same wavelength propagating on the semi-infinity silver-air interface (Fig. 1(c)). Thus the wave vector of the SPP on the sphere can be defined as

$$k_m = \frac{2\pi}{\lambda_m} = \frac{m}{r}. \tag{2}$$

From the resonant peak and the wave vector, the dispersion relation of the spherical particles can be deduced.

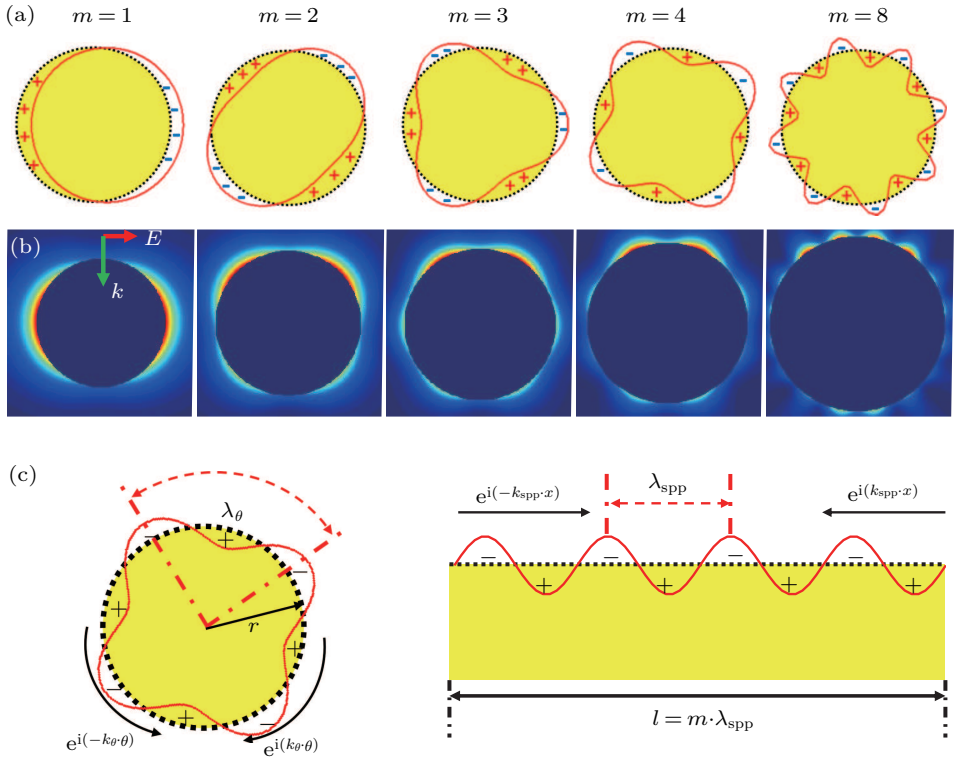


Fig. 1. (a) Illustration of charge distributions of different orders of surface plasmon resonant modes. (b) Corresponding electric field patterns of a silver nanosphere (Drude model, $\varepsilon = 3.7 - \omega_p^2/(\omega^2 + i \cdot \omega/\tau)$, $\omega_p = 9.149$ eV, $\tau = 3.1 \times 10^{-14}$ s) in air calculated by Mie theory (from left to right, $R = 20$ nm, $\lambda = 335$ nm; $R = 30$ nm, $\lambda = 315$ nm; $R = 55$ nm, $\lambda = 310$ nm; $R = 100$ nm, $\lambda = 315$ nm; $R = 300$ nm, $\lambda = 335$ nm). (c) Illustration of the assumption of a surface plasmon resonant mode of a spherical nanoparticle and surface plasmons on the interface between plane metal and dielectric medium. The surface charges of an $m = 4$ order surface plasmon resonance can be viewed as a standing wave of an SPP wavelength λ_θ, which is formed as two waves (clockwise and counterclockwise). It is equivalent to the surface charge distribution of a surface plasmon wave on a plane interface between the metal and dielectric medium.

3. Results and discussion

Based on the assumption above, silver (Johnson and Christy experiment[16]) spherical nanoparticles in air are calculated with Mie theory and verified with full wave calculation by using the FEM method.[17] Figures 2(a) and 2(b) show the extinction spectra of nanospheres in air, with diameters from 10 nm to 380 nm. The excitation light is a plane wave and the polarization is fixed in one direction. For metal particles larger than 10 nm, the quantum effect is negligible and the dielectric function is still usable without further modification (simulations with electron mean free path modified dielectric function show very little difference). Wavelengths and thus frequencies of surface plasmon resonant peaks of different orders ($m = 1$, 2, 3, ...) can be obtained from the spectra with $\omega_m = 2\pi c/\lambda_m$ where c is the light speed in a vacuum; wave vectors of spheres with different radii and multipole resonant order are deduced from formula (2). Then we plot all of the corresponding values of ω_m and k_m of particles with different sizes together in the same figure, which are the SPP dispersion relations of the nanospheres (Fig. 2(c)). In Fig. 2(c) the dispersion relations of the SPPs on the interface of a semi-infinite silver-air interface (TM waves) are plotted together (green circles). It can be seen that the dispersion relations of all of the orders of SPP resonant modes on the nanosphere match very well with those of the SPPs on a planar surface. This suggests that our assumption is correct, i.e., the localized SPPs on a spherical nanoparticle actually are a kind of propagating SPP wave. However, in the range of large wave vectors, there are still some deviations between the dispersion relation curves of the nanosphere and those of the planar film, and the deviations are different for different-order modes. All modes of the nanospheres have lower energies than those of the SPPs in the planar film, which is consistent with the electrostatic approximation of spherical SPPs.

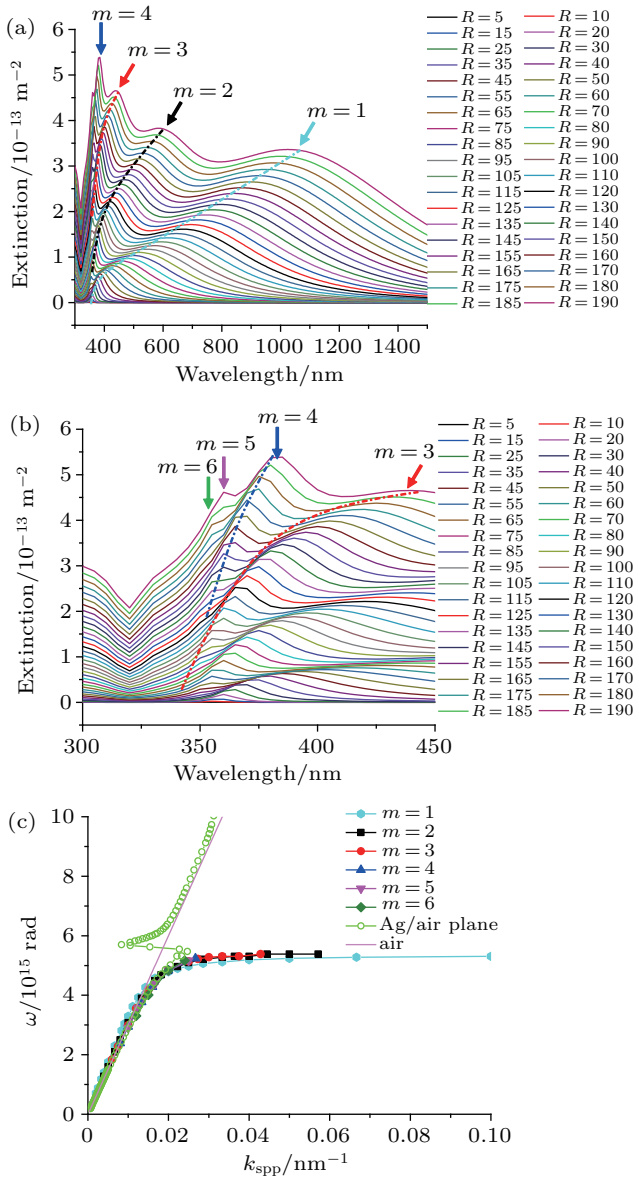


Fig. 2. (color online) (a) Mie theory calculated extinction spectra of silver nanosphere in the air, with radii ranging from 5 nm to 190 nm. (b) Zoomed plots in panel (a). (c) Dispersion relations of the SPPs of the spherical nanoparticles (the same as those in panel (a)) and silver planar surface. The different resonant orders ($m = 1, 2, 3, \ldots$) are indicated with the same colors as the arrows in panels (a) and (b).

To further check the relationship between the localized SPPs on spherical nanoparticles and the SPPs propagating on a planar surface, Mie theoretical simulations are performed under the Drude model. The dielectric function is set to be

$$\varepsilon = 3.7 - \frac{\omega_{\mathrm{p}}^2}{\omega^2 + \mathrm{i} \cdot \omega * (1/\tau)}, \tag{3}$$

where $\omega_{\mathrm{p}} = 9.149$ eV, $\tau = 3.1 \times 10^{14}$ s ($\varepsilon_\infty = 3.7$ is used as the permittivity to make the value closer to the permittivity value of the real metal). With the same method stated above, the dispersion relations are plotted in Fig. 3. Figure 3(a) shows the dispersion relations of nanospheres with the different radii, and the dispersion relations of SPPs on a semi-infinite silver-air interface (dark cyan) and the light line in the air. Different numbers refer to different resonant orders ($m = 1, 2, 3, \ldots$) of plasmonic modes. It can be seen that the dispersion relation curve of each order mode matches very well with its corresponding one of SPPs on the silver-air interface. In the range of large wave vectors, the dipole modes have the lowest energy. Higher order modes have gradually increasing energies, which is consistent with the resonant scenario of spherical nanoparticles under the electrostatic approximation ($\omega_{\mathrm{sp,l}} = \omega_{\mathrm{p}}/\sqrt{l/(2l+1)}$). As the wave vector becomes smaller, the dispersion relation curves of all modes converge together with the one of SPPs on the silver-air interface. The deviations also show that the surface plasmon waves propagating on the spherical nanoparticles have a smaller phase velocity, which was confirmed in Liaw's work.[12] When the radius of curvature becomes larger, the phase velocity is closer to the one of the planar surface. Figure 3(b) shows the scenario of big nanoparticles, in which excellent matching is obtained.

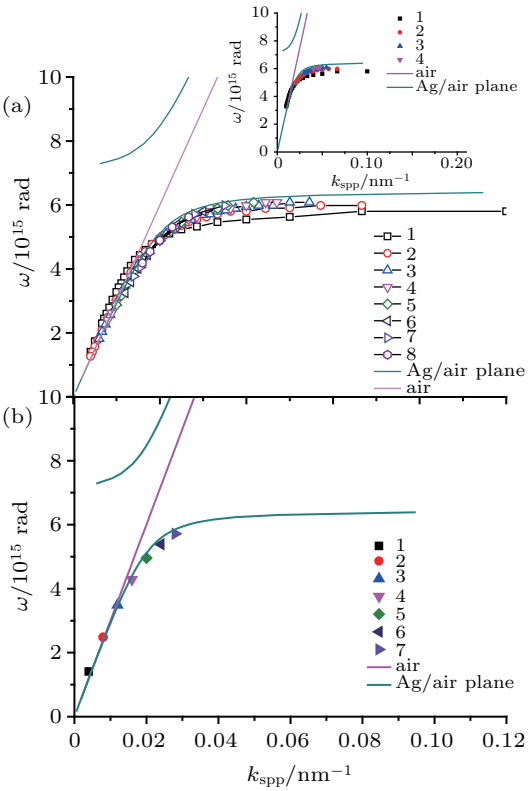


Fig. 3. (color online) Dispersion relations of SPPs on silver spherical nanoparticles in air and those of SPPs on the interface between air and silver planar surface, obtained from the Drude mode, at the same parameters as those in Fig. 1. In panel (a) nanoparticle radii range from 5 nm to 500 nm and the inset shows the dispersion relations of nanoparticles with radii ranging from 5 nm to 100 nm. In panel (b) only the dispersion relation of the nanoparticle with $R = 250$ nm is plotted.

As analyzed above, the localized surface plasmon resonance can be considered as two counter-propagating surface plasmon waves which form a standing wave, which is applicable not only to the high order modes of big nanoparticles, but also to small particles and dipole modes. However, the conclusion does not hold to the case of an infinite metal cylinder. The reason may be that the cylinder and sphere belong to different classes of structures in plasmonics as indicated in transform

optics.[18,19] The results from both Schmidt and Chang also show that the SPPs on a metal cylinder have a mixed wave vectors both in the angular direction in the cross section and along the cylinder direction.[20,21] It should be noted that even in the range of small wave vectors, the dispersion relation curve of the dipole mode (but not for the higher modes) still deviates a little bit from the one of SPPs on the silver-air interface, though the deviation is so small and not obvious. It may be because for big particles, the retardation effect cannot be ignored and thus the resonant frequency is blue-shift.

4. Conclusions

In this work, under the assumption of surface plasmon waves on a spherical nanoparticle analogous to the propagating ones on a planar surface, we have plotted the dispersion relations of surface plasmon resonances of spherical nanoparticles. The results show that the dispersion relations of SPPs on a spherical particle excellently match with those of the surface plasmons on a planar surface. This means that the localized SPPs can be considered as two anti-propagating surface plasmon waves propagating on the surface particles and forming a standing wave. The conclusion still holds even for small nanoparticles with radius smaller than 200 nm to 5 nm, and for dipole resonance as well. This gives us another perspective on localized surface plasmon polaritons.

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