Investigation of some electromagnetic modes in the metal-dielectric system by the absorption spectrum

Gang Sun*, Junjuan Zheng

Institute of Physics, Chinese Academy of Science, Beijing 10080, China

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Abstract

The surface plasmon resonance and the spherical cavity resonance in the system of a layer of periodically arranged dielectric spheres laid on outside of a metal slab or embedded in the metal substrate are investigated by observing the absorption spectrum. The absorption spectra for these systems can be accurately calculated by multiple scattering approaches. The calculated results show that there are some absorption peaks, which can be identified to the resonances of surface plasmon or cavity modes. The surface plasmon resonance occurs when the metal-dielectric interface is periodically modulated, as in both of the systems we considered. The frequency of the surface plasmon resonance is mainly determined by the periodicity of the modulated surface, and can be obtained by an analytic theory. Our calculated results are consistent very well with the analytic ones for both thick and thin metal slab. The cavity resonances only occur for the system of dielectric spheres embedded in a metal substrate. In this system, the resonances of both of the cavity mode and the surface plasmon coexist. In general, the absorption peaks for both of them are relatively weak, because their resonance frequency is far from each other. However, we can design the system to be that the resonances of the cavity mode and the surface plasmon are in almost the same frequency. In this case, a sympathetic resonance will occur, where the absorbance will be dramatically enhanced at the frequency.

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1. Introduction

Recent experimental discovery of the enhanced optical transmission through metal films with periodic subwavelength holes has given rise to a considerable interest in the optical properties of such structures due to their possible numerous applications in optics and optoelectronics as well as rich physics behind the phenomenon of the transmission enhancement [1–4]. The different, sometimes contradictory mechanisms of the transmission enhancement have been proposed and are being debated invoking surface-plasmon polaritons, different types of waveguiding modes in holes or slits, localized plasmons, and various combinations of the above [4–12]. The aim of all these research focuses on the understanding of electromagnetic mode in a system composed of metal and dielectric material and controlling the useable properties of them in applications.

This paper will introduce our recent research on the system of a layer of dielectric (glass) micro-spheres laid on outside of a metal (Ag) slab (Fig. 1(a)) or embedded in the metal substrate (Fig. 1(b)). The glass micro-spheres are arranged in a periodic triangular lattice with lattice constant a, which is the nearest neighbor distance between the spheres. The thickness of the metal slab and the distance between the center of the glass sphere and the upper surface of the metal in Fig. 1(a) will be denoted as t and h, respectively. The thickness of the cover layer in Fig. 1(b) is denoted as dc, which is chosen to be smaller than the skin depth so that external field can excite the electromagnetic modes in the cavity. In our calculation, the permittivity of the glass is set to be a constant with no absorption, $\varepsilon_{\text{sph}} = 1.96$, and that of the silver $\varepsilon_{\text{Ag}}$ is taken to be a Drude metal with plasma frequency $\omega_p h = 9.2$ eV and $(\omega_p \tau)^{-1} = 0.02$, as generally used in the literature. In this paper, the radius of the glass sphere and the thickness of the cover layer are fixed at $S = 0.1 \mu m$ and $d_c = 5$ nm, respectively.

* Corresponding author
E-mail address: gsun@aphy.iphy.ac.cn (G. Sun).

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Surface plasmon and cavity modes

For a perfect interface composed of semi-infinite air and metal Ag, surface plasmon can exist and it has following dispersion relation [16,17],

\[ k_{sp}^2 = \frac{\omega^2}{c^2} \left( \frac{\varepsilon_{Ag}}{1 + \varepsilon_{Ag}} \right), \]  

(2)

where \( k_{sp} \) is the in-plan wave vector of the surface plasmon. The surface plasmon can not be excited directly by a plane incident wave if the interface is perfect, but can be excited by evanescent waves or by scattering centers near the surface, such as the dielectric spheres in our system. The scattered wave from each sphere is strongly coherent, because they come from a same incident plane wave. If the spheres are arranged periodically, according to Brag scattering principle, the scattered wave can only exist in some particular direction, in which the scattered wave from every spheres has the same phase, so they will form a strong scattered wave on the whole system, otherwise they will cancel each other because their phase is different. This criterion is also correct for the component of surface plasmon in the scattered wave. So for a plane incident wave with wave vector \( k_0 \) in air, the surface plasmon on a periodically modulated surface can only occur at the frequencies that satisfy,

\[ k_{sp} = k_0 \sin \theta_0 \pm p \frac{2\pi}{\alpha} u_1 \pm q \frac{2\pi}{\alpha} u_2, \]  

(3)

where \( \theta_0 \) is the angle of incidence, \( u_1 \) and \( u_2 \) are the unit reciprocal lattice vectors of the periodic surface, and \( p \) and \( q \) are integer numbers. In another words, a periodically modulated surface has a surface plasmon resonance at the frequencies determined by Eq. (3), because the surface plasmon would be excited by the incident wave at these frequencies. For normal incident, the lowest resonance frequency corresponds to the in-plan wave vector being equal to one of the reciprocal vector in Eq. (3). It follows that,

\[ \frac{2\pi}{\alpha} = \frac{\omega}{c} \frac{\sqrt{3}}{2} \frac{\varepsilon_{Ag}}{\sqrt{1 + \varepsilon_{Ag}}}, \]  

(4)

which is only a function of the periodicity of the metal surface.

When the metal is not semi-infinite but with a finite thickness, there are two air–metal interfaces. Surface plasmons can exist on both of them, and they will weakly couple each other through the field in the metal slab. As a result, they form two altered surface modes, correspond to symmetric and antisymmetric surface plasmons, if the absorption in the metal is negligible. These surface modes have following dispersion relations [18], if the metal slab is relatively thick (\( \exp(-tk_{sp}\sqrt{-\varepsilon_{Ag}}) \ll 1 \)),

\[ k_{1,2} = k_{sp} \left[ 1 \pm \frac{2\varepsilon_{Ag}}{1 - \varepsilon_{Ag}} \exp(-tk_{sp}\sqrt{-\varepsilon_{Ag}} \right], \]  

(5)

where the in-plan wave vectors \( k_1 \) and \( k_2 \) correspond to symmetric and antisymmetric surface plasmons, respectively. By the same reason that we mentioned previously for the case of semi-infinite metal system, the surface plasmon can only be excited by the incident plane wave with
frequencies that satisfy,
\[ \mathbf{k}_{1,2} = \mathbf{k}_0 \sin \theta_0 \pm p \frac{2\pi}{\lambda} \mathbf{u}_1 \pm q \frac{2\pi}{\lambda} \mathbf{u}_2. \]  
(6)

We notice that there exist two resonance frequencies for each frequency of Eq. (3).

Another analytic soluble electromagnetic mode is the cavity mode for dielectric spheres embedded in an infinite large metal [19]. The frequencies for these spherical harmonic electric modes can be obtained from the peaks of coefficients \( T^F_0(\omega) \) [13], which can be written as,
\[ T^F_0(\omega) = \frac{j_l(k_{qph}) - j_l(k_{Ag})}{h_l(k_{Ag})} \frac{(r_1(j_l(k_{qph})) r_{Ag} - j_l(k_{qph}) r_{qph}) (r_1 j_l(k_{Ag})) r_{qph}}{h_l(j_l(k_{Ag})) r_{qph}} |_{l=5}, \]
(7)

where \( k_{qph} = \sqrt{\varepsilon_{qph} \mu_{qph}} \) and \( k_{Ag} = \sqrt{\varepsilon_{Ag} \mu_{Ag}} \) with \( c \) being the velocity of light in vacuum. When the cover layer \( d_c \) is not infinite, these spherical harmonic electric modes exist approximately, and they will couple with the incident plane wave through the little leakage of the mode into the vacuum. So we can see a cavity resonance at these frequencies, because external field will easily excite them. In this paper, the cavity properties of the glass sphere is not changed, since the permittivity of the glass spheres and that of the silver substrate, and the radius of the glass sphere are all fixed. The lowest cavity resonance frequency corresponds to that with \( l = 1 \), and its free-space wavelength is at about \( \lambda = 425 \) nm, which is in the range of visible light. In this paper, we use free-space wavelength \( \lambda = 2\pi c/\omega \) to denote the frequency.

3. Surface plasmon induced by periodically arranged glass spheres

We firstly study the system of a metal slab with a layer of dielectric (glass) micro-spheres laid on outside of its surface as shown in Fig. 1(a). The multiple scattering approach is used to calculate the absorption spectrum of the system. In the method, the maximum value of the angular moment index \( L_{max} \) in the spherical wave expansion and the number of plane waves \( R_{max} \) in plane wave expansion based on reciprocal lattice vectors control the convergence of the calculation. The convergence criterion depends on the details of the system to be calculated, and we determine them by numerical convergence tests, which increase the values of \( L_{max} \) and \( R_{max} \) gradually until the results do not change. Our numerical convergence tests show that \( L_{max} = 19 \) and \( R_{max} = 36 \) are enough for the calculation in this system.

Fig. 2(a)–(d) show the absorbance of the system with a thick enough metal slab as a function of the free-space wavelength for a series of lattice constant \( \alpha \). The distance between the center of the glass sphere and the metal surface is set to \( h = 100 \) nm, i.e. the spheres touch the metal surface. In each panel from 2(a) to (d), a distinct absorption peak is found, which can be traced to surface plasmon induced by the layer of periodically arranged glass spheres outside the metal surface. The position of the absorption peak moves to longer wavelength as lattice constant \( \alpha \) increases (from 2(a) to (d)), agreeing with the trend described by Eq. (4). To convince that the absorption peak is due to the surface plasmon resonance, the position of absorption peak as a function of \( \alpha \) is shown in Fig. 3. The lowest resonance frequency of surface plasmon obtained by Eq. (4) was also
plotted in the figure (solid lines). From Fig. 3, we can see that the analytic results and the calculated ones agree well, especially for larger lattice constant. While at small lattice constant, more deviation between them is found. This is quite understandable since the existence of the glass spheres in the air will affect the effective permittivity of dielectric media near the metal surface. This effect would become weaker when the glass spheres were more spare or the whole layer of the glass sphere was apart further away from the metal surface.

For the system with a thinner metal slab, according to Eq. (6) the resonance of surface plasmons will occur at two frequencies, correspond to symmetric and antisymmetric modes, respectively. In this case, the absorption peak occurred in very thick metal slab (as shown in Fig. 2) splits to double peak structure. Fig. 4(a)–(d) show the absorbance for the system with a thinner metal slab as a function of the free-space wavelength for a series of thickness of the metal slab \( t \). The lattice constant is fixed at \( a = 500 \) nm, and the distance between the center of the glass sphere and the metal surface is also set to \( h = 100 \) nm. From Fig. 4, we clearly see the double peak structure resulting from the limited thickness of metal slab. The splitting is larger for thinner metal slab. However, for this system the upper metal-dielectric interfaces and the lower one are not strictly equivalent, because of the existence of the glass spheres on the upper interface but without the spheres at lower one. Therefore, Eq. (5) is not applicable in this system, unless the layer of glass spheres is apart far away from the metal surface. In Fig. 4, instead of the two absorption peaks tend to a common frequency as the thickness of the metal increases as described by Eq. (5), they tend to two different frequencies at \( \lambda = 473 \) nm and \( \lambda = 455 \) nm, which correspond to the frequencies that consider the affection of glass spheres to the effective permittivity of the dielectric side (closed circle at \( \alpha = 500 \) nm in Fig. 3) and that do not consider this affect (solid line at \( \alpha = 500 \) nm in Fig. 3), respectively. These two surface plasmons are almost independent, and exist on the upper and lower metal surface, respectively.

When the layer of glass spheres is apart far away from the metal surface, both side of the metal tend to same, and Eq. (5) becomes a good approximation. Fig. 5 shows the position of absorption peaks as a function of the thickness of the metal slab \( t \) for a very long distance between the center of the glass sphere and the metal surface \( h = 500 \) nm. The resonance frequencies of the surface plasmon given by Eq. (6) were also plotted in the figure (solid and dashed lines). We found that they are in good agreement when the metal slab is thicker, which is the applicable condition of Eq. (6).

4. Surface plasmon, cavity modes, and its coupling

In this section, we study the system of a metal substrate with a layer of dielectric (glass) micro-spheres embedded inside as shown in Fig. 1(b). To use multiple scattering approach in this system, the Ewald sum used in the computation of the Z matrices [14] should be replaced by direct summation. The convergence requirement for this system is rather demanding, especially when the spheres become close each other or the cover layer becomes very thinner. For this reason, we restrict our calculations to
Fig. 6. $T_F^l(\omega)$ for several value of $l$ (a). The absorbance for various lattice constant $\alpha$ are shown in (b)–(e). The symbols $S$ and $C$ labels surface-plasmon-like and cavity-like resonances.

$\alpha > 3S$ and $d_h = 5$ nm. By numerical convergence tests, we found that we need to use $L_{\text{max}} = 19$ and $R_{\text{max}} = 48$ to obtain reliable results for the most demanding case of the convergence.

Fig. 6(a) shows the value of $T_F^l(\omega)$ for several spherical harmonic electric mode $l$. From Figure (a), we can see that the strongest resonance comes from $l = 1$, which located at the longest wavelength ($\lambda = 425$ nm). The resonance become weaker and shift to shorter wavelength as $l$ increases.

Fig. 6(b)–(e) shows the absorbance as a function of the free-space wavelength for a series of lattice constant $\alpha$. We focus our attention on the absorption peaks at several lowest frequencies, labeled by $S$ and $C$, respectively. Fig. 6(b) corresponds to the case in which the embedded glass spheres are at $\alpha = 0.6 \mu$m apart from each other. In this panel, the absorption peak labeled as $C(l = 1)$ can be traced to $l = 1$ cavity mode of the embedded dielectric sphere. The fact that this is a $l = 1$ mode can be identified by varying the maximum angular momentum index used in the multiple scattering calculations from one progressively to higher values. This $l = 1$ cavity mode has nearly the same frequency as the cavity resonance frequency of a sphere embedded in bulk Ag, shown by the solid line in Fig. 6(a).

Another absorption peak at lower frequency, labeled as $S$, is due to the excitation of the surface plasmon on a periodically modulated metal-dielectric interface, which has frequency given approximately by Eq. (4). In this panel, the two resonance frequencies are far from each other, and both peaks are low and narrow, and their origins can be easily identified.

Fig. 6(c) shows the situation when the spheres are closer together, at $\alpha = 0.55 \mu$m. The cavity resonance remains at about the same frequency, as expected, while the plasmon absorption peak moves to shorter wavelength. From panels (b) to (d) we see that the absorption peak of the cavity mode remains more or less in the same position, while the plasmon absorption peak moves to shorter wavelength as the lattice constant decreases. The resonance frequency of surface plasmon is lower than that of the $l = 1$ cavity mode as the lattice constant being large, and they will cross at certain lattice constant. When the cavity resonance and the surface plasmon resonance are close in frequency, the coupling increases the absorbance of both resonances, as the absorption peaks become higher and wider.

When the two resonance frequencies are about the same, at $\alpha = 0.47 \mu$m, both peaks reaches the highest value. At that particular configuration, the assignment to cavity-like or plasmon-like resonance is somewhat arbitrary. If we further decrease the lattice constant, the resonance frequency of the surface mode become higher than that of the cavity. At $\alpha = 0.40 \mu$m (Fig. 6(e)), the surface plasmon resonance is between $l = 1$ and $l = 2$ cavity mode and the coupling leads to the enhancement of the absorption of both.

At higher frequencies, some small peaks can be traced to higher order cavity resonance ($l = 2$ or $l = 3$) and higher order surface plasmon mode. However, these modes are closely spaced in frequency and coupling between the cavity mode and the surface plasmon mode make the assignment to plasmon-like or cavity-like somewhat difficult.

In Fig. 7, we show the wavelengths of the lowest and second-lowest absorption peaks as a function of $\alpha$. The resonance frequency of $l = 1$ cavity mode obtained by Eq. (7) and the lowest resonance frequency of the surface plasmon obtained by Eq. (4) were also plotted as dashed and solid lines, respectively, in Fig. 7. The solid and dashed lines cross at about $\alpha = 0.46 \mu$m. Far away from the crossing-point, the assignment of the resonance to plasmon-like or cavity-like is visually obvious. When the lattice constant $\alpha$ near the crossing-point, the coupling will splits the resonance frequencies. As a results, the two absorption peak would never coincide, and an interchanging of the plasmon-like...
resonance and the cavity-like resonance is completed in the area adjacent to the crossing-point.

We also see that the position of the absorption peak corresponding to the cavity mode deviate from the analytic result somewhat. This is quite understandable since the cavity resonance frequency depends on the boundary condition, and in this case, it depends on the leakage of the mode into the vacuum. By the same reason that mentioned in Section 3, the position of the absorption peaks of the surface plasmon also approach the analytic value at larger separations of the spheres, but here the existence of the glass spheres affect the effective permittivity in the metal side.

5. Conclusions and discussions

The surface plasmon resonance for a periodically structured metal-dielectric interface and a spherical cavity resonance for a dielectric sphere mounted in a metal are investigated by observing the absorption spectrum. Two systems were selected to investigate the resonances. One is that a layer of periodically arranged dielectric spheres lay on outside of a metal slab, another is that the layer of dielectric spheres is embedded in a metal substrate. The absorption spectra for these systems can be accurately calculated by multiple scattering approaches.

The absorption spectrum for the system of periodically arranged dielectric spheres on the upper surface of the metal shows some peaks, which can be identified to surface plasmon resonances. For a very thick metal slab, the strongest resonance of the surface plasmon corresponds to that with lowest frequency, which is a single sharp peak in the absorption spectrum. The position of the absorption peak is mainly determined by the periodicity of dielectric spheres, and is consistent with the analytic theory very well.

For a thin metal slab, the single absorption peak occurred in thick metal slab splits to double peak structure, which correspond to resonances of symmetric and antisymmetric modes, respectively. The width of the splitting is related the thickness of the metal slab, which can also be explained by an analytic theory.

The absorption spectrum for the system of dielectric spheres periodically embedded in a metal substrate also shows some peaks. Besides that can be identified to the surface plasmon resonance, there are some other peaks that can be identified to the spherical cavity resonances, especially the $l=1$ cavity resonance has the strongest absorbance. In general, the absorption peaks for both of the cavity and the surface mode are relatively weak, because their resonance frequency is far from each other. However, if we design the system to be that the cavity mode and the surface mode in almost the same frequency, a sympathetic resonance will occurs. In this case, the absorbance will be dramatically enhanced at the frequency.

For the systems we considered in this paper, the physics of the electromagnetic modes are clear, and the resonance frequencies can be controlled easily by corresponding parameters. Therefore, we can design some useful system, for which the sharp absorption peaks are located at desired frequencies.

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