Optical Scattering Properties of Bimetallic Nanodisk Dimers

Zou Weibo Zhou Jun Zhang Haopeng Ma Yanan Shu Lei

(*Department of Microelectronic Science and Engineering, Faculty of Science, Ningbo University, Ningbo, Zhejiang* 315211, *China*)

Abstract According to the dipole-dipole approximation of plasmon hybridization theory, the localized surface plasmon resonance (LSPR) spectra of a pair of closely spaced gold and silver nanodisks, i.e. the plasmonic scattering characteristics of the bimetallic nanodisk dimer, have been studied by using the finite element method (FEM). The results show that, the plasmonic scattering intensities of the bimetallic nanodisk dimer are affected by the polarization of incident electromagnetic field and its structure parameters, such as the disk diameter, thickness and gap distance. Moreover, the LSPR spectra of the bimetallic nanodisk dimer have considerably red-shift as the increasing of diameters, the decreasing of thicknesses and gap distance. It is significant to apply the heterodimer plasmonic systems on biological sensors and photosensitive detectors.

Key words materials; bimetallic dimer; localized surface plasmon; finite element method; scattering cross section

OCIS codes 290.5850; 290.5838; 160.3900; 160.4760

复合金属纳米盘二聚物的散射特性

邹伟博 周 骏 张昊鹏 马亚楠 束 磊

(宁波大学理学院微电子科学与工程系，浙江 宁波 315211)

摘要 采用有限元方法(FEM)研究了一组相邻金纳米盘和银纳米盘的表面等离激元共振(LSPR)特性，并根据等离激元杂化理论的偶极-偶极近似对复合金属纳米盘二聚物的散射光谱进行分析。结果表明，金-银纳米盘的等离激元散射强度受到入射电磁场偏振方向以及金属纳米盘二聚物的结构参数(如纳米盘直径、厚度和纳米盘间间隙)变化的影响。而且，随着纳米盘直径的增加，纳米盘厚度以及盘间间隙减小，金-银纳米盘二聚物等离激元共振散射谱共振峰位置会发生相应的红移。研究结果对于纳米金属异质二聚物等离激元物系在生物传感和光敏探测器件等方面的应用有重要意义。

关键词 材料；复合金属二聚物；局域表面等离激元；有限元方法；散射截面

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1 Introduction

It is well known that the localized surface plasmon resonance (LSPR) properties of noble metal nanostructure are originated from the oscillations of the free electrons on the metal surface of the metallic nanostructure under the excitation of the external electromagnetic field[1-2]. LSPR effect can result in a strong enhancement of the optical absorption, scattering, and electric near-field intensity of metal nanoparticles, and the wavelength and intensity of LSPR peaks are sensitively dependent on the geometric parameters and the dielectric materials of the nanoparticles[3-4]. LSPR properties have a wide range of applications such as nonlinear optics[5], surface enhanced Raman scattering (SERS)[6], waveguide sensor[7] and chemical and biological sensors[8].

During the past decade, much of metallic nanostructures have received great attention, especially

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作者简介：邹伟博(1988—)，男，硕士研究生，主要从事微纳金属介质结构方面的研究。E-mail：wbzou1988@126.com

导师简介：周 骏(1958—)，男，教授，博士生导师，主要从事光学与光电子学方面的研究。E-mail：zhoujun@nbu.edu.cn (通信联系人)

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several of nanoparticle homodimers and heterodimers structures. The plasmon coupling responses of these nanodimers exhibit the distinct shift of LSPR wavelength and the enormous enhancement of electric near-field. Bala Krishna Juluri *et al*.[9] have found that the tunability of the optical scattering spectra of the gold-gold nanodisk dimers can be realized by controlling the various structure parameters of the nanoparticle dimers. Atay et al.[10] have observed a splitting of the surface plasmon polariton energy by varying the gap distance of particle-pair in the periodic array which is composed of gold-gold nanodisks homodimers. Tanaka et al.[11] have proposed a new concept of light-matter interactions, which shows the nanoscale spatial profiles of gap-mode LSPR fields within gap-region of the gold nanodisk dimers. Zhang Zhenming et al.[12] have derived the influence of LSPR formed on silver nanodisk array on the LED performance. Furthermore, the far-field optical properties of silver-gold heterodimer systems have also been explored by Encina et al.[13] and an additional Fano profile peak appears in the absorption spectrum of the Au nanosphere, which is in agreement with the result in Ref.[14]. The plasmon coupling of the silver-gold and the silver-copper heterodimers has been theoretically studied and the collective plasmon mode has been predicted[15]. Recently, a prominent directional scattering phenomenon arising from coupling plasmon modes in the material-asymmetrical gold-silver nanodisk heterodimers is experimentally observed[16]. It is worthy to deeply explore the underline feature of optical scattering properties of gold and silver bimetallic nanodimers owing the coupling plasmon modes.

In the present work, the optical scattering spectra of the gold-silver nanodisk heterodimers are theoretically researched by the commercial finite element method (FEM) software (COMSOL). It has proved that FEM is an effective method for solving electromagnetic problems with complex geometries such as metal nanostructures[17]. The optical scattering spectra of the bimetallic nanodisks are simulated and the shift of LSPR peaks is clearly related with the diameter, thickness and gap distance of the bimetallic nanodisks. The physical mechanism of the plasmon coupling in the bimetallic nanodisks is discussed in detail by a simple dipole-dipole coupling model.

2　Model and theory

The optical scattering properties of bimetallic nanodimers are studied in the framework of the scattered field formulation by using a commercially available FEM package (COMSOL Multiphysics 4.1 with the radio frequency module). The scattering spectrum of the bimetallic nanodimer C_{sca} is integrated by the normalized electric field around a far-field transform boundary enclosing the bimetallic nanodimer[18]. The formula is

$$C_{sca}=\frac{1}{E_{inc}^2}\int|\boldsymbol{E}_{far}|^2\mathrm{d}S, \tag{1}$$

where $\boldsymbol{E}_{far}$ is the far-field electric vector of the scattering field and can be calculated by Stratton-Chu formula[19], E_{inc} is electric amplitude of the incident field, S is the cross-sectional area of calculating boundary.

The geometry model of the bimetallic nanodimer is depicted in Fig. 1. It is consists of a gold nanodisk and a silver nanodisk with a gap distance d and the nanodisks have the same shape and size, i.e., the diameter D and the thickness H are the same. In the calculation, the medium surrounding the model is air with the refractive index of 1.0, and the tabulated values in Ref.[20] are employed as the dielectric constants of gold and silver.

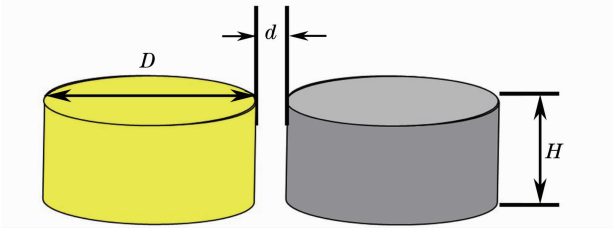

Fig. 1　Schematic diagram of the bimetallic nanodimer

When the incident light excites the bimetallic nanodimer, as shown in Fig. 2, the coulombic interactions in and between the two closely spaced nanodisks are produced due to the different distributions of the surface charges on the nanodisks. The features of scattering spectra of the bimetallic nanodimer can be explained by the dipole-dipole coupling model[21-22], which is similar to the analysis of the single metal nanodisk. For single gold or silver nanodisk, each of the electrons oscillates back and forth under the applied electromagnetic field, which is expected to dominantly contribute to the LSPR, and the coulombic attractions between electrons and cations in the surface of the nanodisks serve as the restoring force in the nanodisks. While the two nanodisks are close to each other, as sketched in Fig. 2, the additional polarization force appears and acts on the nanodisks because of the induced dipoles in the neighboring nanodisks. In brief, if the electromagnetic field is parallel to the long axis of the bimetallic nanodimer seen in Fig. 2(a). The positive charges in the left nanodisk face and the corresponding negative charges in the right nanodisk. The additional force will occur in the gap region and weaken the

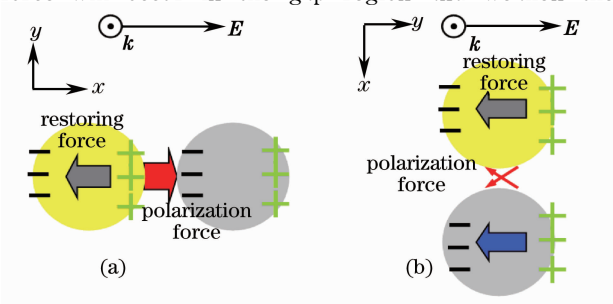

Fig. 2　Schematic diagram of the coulombic interactions of two closely placed nanodisks. Polarization directions of the incident light are (a) parallel and (b) perpendicular to the long axis of the bimetallic nanodimer, respectively

restoring force of the surface charges in both nanodisks. In contrast, when the electromagnetic field is perpendicular to the long axis of the bimetallic nanodimer, the surface charges in both nanodisks are distributed from negative to positive along the polarization direction as shown in Fig. 2(b), similar to the charge distributions in Fig. 2(a).

3 Results and discussion

3.1 Effect of the polarization angle

The scattering spectra of the bimetallic nanodimer (D = 100 nm, H = 30 nm and d = 10 nm) with different polarization angles are shown in Fig. 3. As shown in Fig. 3(a), the scattering spectrum (black line) displays two distinct peaks at 466 nm and 616 nm respectively. When the polarization direction of incident light is parallel to the long axis of the bimetallic nanodimer, only the longitudinal resonance mode (L-mode) is induced due to the longitudinal LSPR coupling between the two nanodisks and the two peaks corresponding to the higher energy L-mode and the lower energy L-mode respectively. When the polarization direction of incident light is perpendicular to the long axis of the bimetallic nanodimer, the transverse resonance mode (T-mode) presents in the scattering spectrum due to the transverse LSPR coupling between the two nanodisks. As shown in Fig. 3(b), two distinct peaks are displayed in their scattering spectrum (pink line), which are located at 473 nm and 582 nm, corresponding to the higher energy T-mode and the lower energy T-mode respectively. As expected, the lower energy L- and T-modes (616 nm, 582 nm) and the high energy L- and T- modes (466 nm, 473 nm) can be assigned as the dipole-dipole pattern and the quadrupole-quadrupole pattern respectively from the dipole-dipole approximation of the plasmon hybridization theory. Moreover, when the incident polarization angle ranges from 0° to 90°, the scattering spectra are resulted from the interaction of L- and T- orthogonal modes. The dominant modes are changed from L- to T-mode. Thus, the peak intensity of lower energy plasmon mode decreases, whereas the peak intensity of higher energy plasmon mode increases, and an isosbestic point is clearly observed at 540 nm. Moreover, for the case of the bimetallic nanodimer, the peaks of lower energy plasmon modes have slightly blue-shifts with the increase of the incident polarization angles, which agrees with the case of Ag-Au nanosphere dimer, whereas the position of the peaks of higher energy plasmon modes do not change comparing with that of Ag-Au nanosphere dimer[15]. In Fig. 3(b), the dipole resonance peaks of the isolated silver nanodisk and gold nanodisk are 466 nm and 573 nm respectively. By comparing with the case of the isolated nanodisks, the lower energy L- and T-modes of the bimetallic nanodimer have larger red-shifts, while the higher energy T-mode has only a small red-shift. It can be known from Fig. 3 that the lower energy and higher energy resonance peaks correspond to the dipole LSPR of Au nanodisk and Ag nanodisk respectively, which demonstrates the bimetallic nanodimer can scatter light preferably in the LSPR regions of Au and Ag[16].

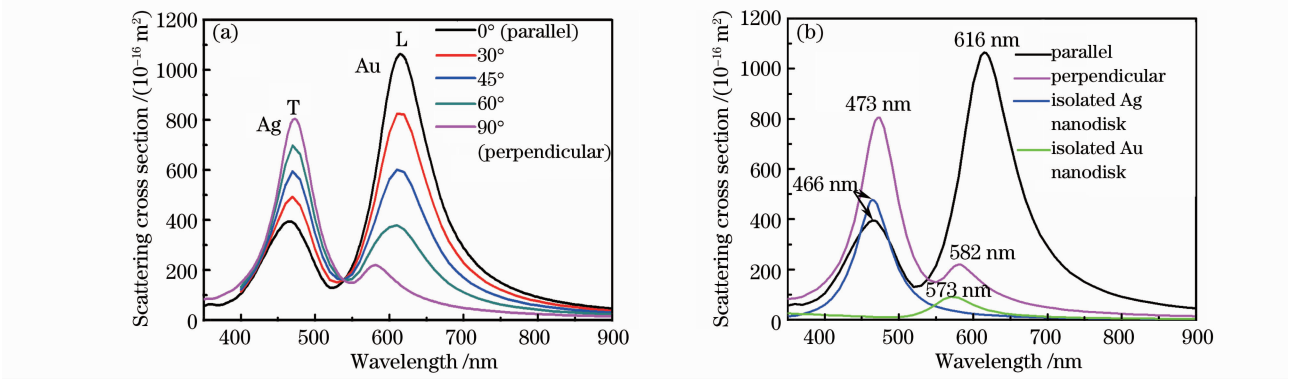

Fig. 3 (a) Scattering spectra with different incidenct polarization angles for the bimetallic nanodimer; (b) scattering spectra of the isolated silver and gold nanodisks

3.2 Effect of the diameter

The scattering spectra of the bimetallic nanodimer with different diameters are shown in Fig. 4. In Fig. 4 (a), the polarization direction of the incident light is parallel to the long axis of the bimetallic nanodimer. The peaks corresponding to the higher energy L-mode and the lower energy L-mode are located at 440 nm and 550 nm for D = 80 nm, 466 nm and 615 nm for D = 100 nm, 580 nm and 660 nm for D = 120 nm, respectively. When the polarization direction of the incident light is perpendicular to the long axis of the bimetallic nanodimer, as shown in Fig. 4(b), the peaks are located at 440 nm and 560 nm for D = 80 nm, 473 nm and 582 nm for D = 100 nm, 510 nm and 600 nm for D = 120 nm, respectively. It is clearly found that these peaks evidently shift toward long wavelengths with the increasing of nanodisk diameters owing to the coupling interaction of the LSPRs in the nanodisks. In fact, as shown in Fig. 2, the negative and positive charges mainly distribut at the end sides of nanodisks along the polarization direction of the applied electromagnetic field. With the increasing of the nanodisk diameters, the coulombic attractions of the surface charges are weakened due to the larger distance between positive and negative charges in both nanodisks, which weakens the restoring force and

results in the lower plasmon energy and the red-shifts of LSPR peaks. It is similar to the diameter-dependent LSPR shifts for single gold or silver nanodisk[23-24]. In addition, comparing Fig. 4(a) with Fig. 4(b), the resonance intensities of the LSPR peaks of the gold and silver nanodisks are distinctly different for the two polarization direction, which is originated from the same mechanism discussed in section 3.1.

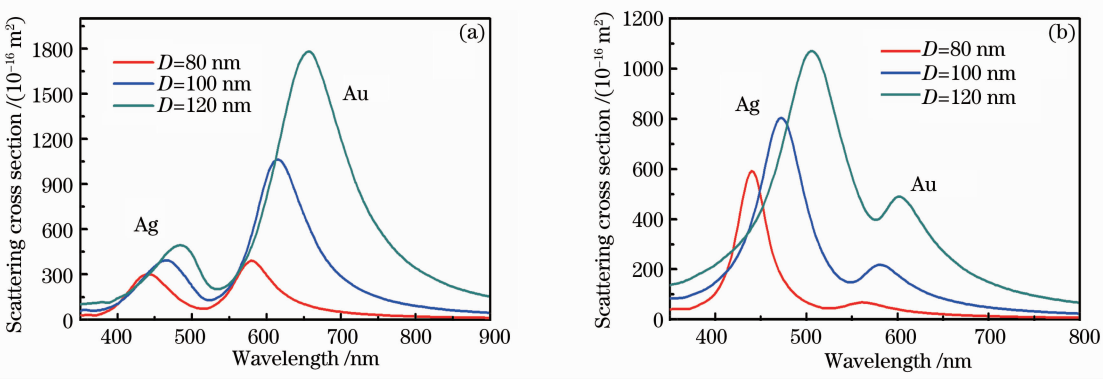

Fig. 4 Scattering spectra with different nanodisk diameters for the polarization directions of the incident light (a) parallel and (b) perpendicular to the long axis of the bimetallic nanodimer

3.3 Effect of the thickness

The scattering spectra of the bimetallic nanodimer with different thicknesses are shown in Fig. 5. In Fig. 5(a), the polarization direction of the incident light is parallel to the long axis of the bimetallic nanodimer. The dipole LSPR peaks of the silver nanodisk and the gold nanodisk are respectively located at the short wavelengths and the long wavelengths for different nanodisk thicknesses, such as 466 nm and 615 nm for H = 30 nm, 510 nm and 657 nm for H = 15 nm, 555 nm and 703 nm for H = 10 nm. It is found that the resonance peaks of the silver nanodisk and the gold nanodisk have the red-shifts of 44 nm and 42 nm for H from 30 nm to 15 nm, and the red-shifts of 45 nm and 56 nm for H from 15 nm to 10 nm, respectively. When the polarization direction of the incident light is perpendicular to the long axis of the bimetallic nanodimer, as shown in Fig. 5(b), the peaks are located at 473 nm and 582 nm for H = 30 nm, 520 nm and 620 nm for H = 15 nm, and 580 nm and 670 nm for H = 10 nm respectively. It can be seen from Fig. 5 that the red-shift trends of the silver nanodisk and the gold nanodsk are the same for both cases of the polarization direction of the incident light. Thus, the red-shift of resonance peaks can be obtained by decreasing the nanodisk thickness.

The plasmon resonance red-shift can be explained by the dipole-dipole coupling model while decreasing the thicknesses of nanodisks. With the decreasing of the nanodisk thickness, the side surfaces of two nanodisks are decreased so that the surface charge density of nanodisks and the coupled strength in coupling region are increased. We conjectured that the surface charge density waves propagate circularly around the edge of the nanodisk when an incident electric field couples with the electron oscillations in the gold or silver nanodisk. Thus, the coulombic interaction from top- and bottom-surface would strengthen the influence of the coulombic interaction from self-surface with the decreasing of thickness, resulting in the weakening of the restoring force on the surface of the nanodisks. In turn, with weakening the restoring force, the lower plasmon energy results in the larger red-shift of LSPR peaks of the bimetallic nanodimer. In addition, the intensities of the LSPR peaks of the gold nanodisks in Fig. 5(a) are much stronger than that of the silver nanodisks, but a reverse phenomenon is exhibited in Fig. 5(b), and the reason has been discussed in section 3.1.

3.4 Effect of the gap distance

The gap distance of the bimetallic nanodimer is

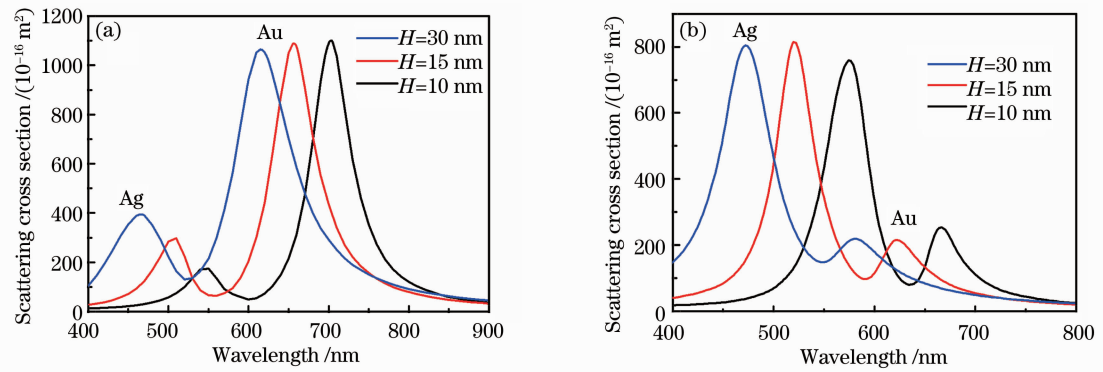

Fig. 5 Scattering spectra with different thicknesses of nanodisk for bimetallic nanodimer. The polarization directions of the incident light (a) parallel and (b) perpendicular to the long axis of bimetallic nanodimer, respectively

significant for tuning the LSPR peaks in the scattering spectra. Fig. 6 shows the scattering spectra of the bimetallic nanodimer for the gap distance d ranging from 60 nm to 3 nm. In Fig. 6(a), the polarization direction of the incident light is parallel to the long axis of the bimetallic nanodimer, and the peaks are corresponding to the dipole LSPRs of the silver nanodisk and the gold nanodisk. With the decreasing of gap distance, the intensity of LSPR peak of the silver nanodisk is stronger than that of the gold nanodisk until the gap distance decreases to a critical size of 27 nm, whereas the intensity of LSPR peak of the gold nanodisk is stronger than that of the silver nanodisk for $d<27$ nm. Meanwhile, it is found that the LSPR peaks of gold nanodisk red-shift from 581 nm to 700 nm and the LSPR peaks of silver nanodisk almost keep constant at $\lambda=466$ nm without flat shoulder[15] for gap d from 60 nm to 3 nm. This variation of scattering spectra of the bimetallic nanodimer with decreasing gap distance are appreciable contribution from both metal materials, which makes the higher order resonances remain unchanged and the dipole mode considerably red-shift. For the sake of contrast, the scattering spectra of the Ag and Au nanodisk homodimer with different gap distances for $d=20$, 10, 5 nm are investigated and shown in Fig. 6(b). The intensities of LSPR peaks become stronger with decreasing the gap by the plasmon hybridization of the two nanodisks, which agrees with the previous report[25]. The LSPR peaks of the Ag or Au nanodisk homodimers are located at 520 nm and 610 nm for $d=20$ nm, 560 nm and 630 nm for $d=10$ nm, 600 nm and 660 nm for $d=5$ nm respectively. It is clear that all of LSPR peaks of the homodimer red-shift with decreasing the gap, which can be explained on the basis of a simple dipole-dipole coupling model. For parallel polarization, the dipole-dipole interaction in the gap region of the homodimer will weaken the restoring force within each nanodisk and lead to the reduction of the plasmon energy, i.e. the red-shift of the LSPR peak, which agrees well with the results of Refs.[26-27]. Comparing Fig. 6(a) with Fig. 6(b). The scattering characteristic of bimetallic nanodimer is mainly caused by the gold nanodisk and attributed to the different properties of gold and silver materials, in which the imaginary part of permittivity of silver is larger than that of gold[16,28].

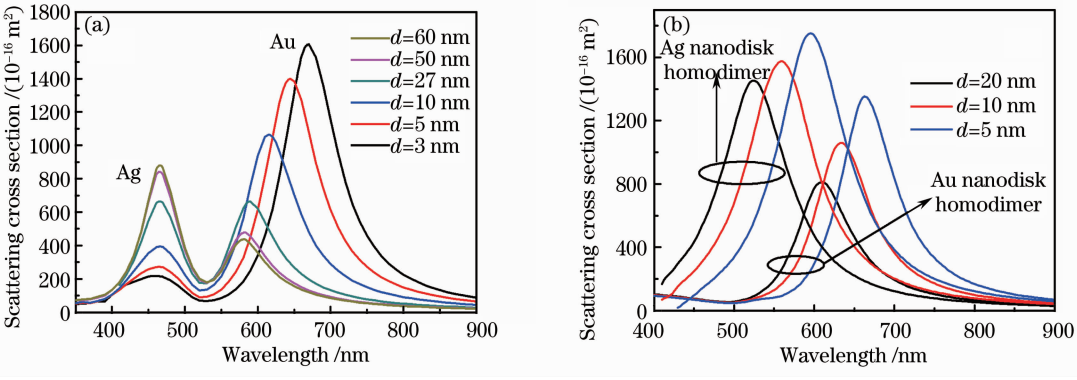

Fig. 6 Scattering spectra of (a) bimetallic nanodisk heterodimer and (b) silver and gold nanodisks homodimer with different gap distances for the polarization direction of the incident light parallel to the long axis of the nanodimers

When the polarization direction of the incident light is perpendicular to the long axis of the bimetallic nanodimer, the scattering spectra with different gap distances are shown in Fig. 7, and the details of the LSPR peaks are displayed in the insets of Fig. 7. It is found that, by varying the gap distance from 60 nm to 5 nm, the LSPR peaks of silver nanodisk red-shift slightly change from 470 nm to 475 nm and the LSPR peaks of gold nanodisk red-shift change from 575 nm to 583 nm, which is different from the blue-shifts of the scattering spectra of gold particle pairs[10,26]. The intensities of LSPR peaks have slight declines with decreasing the gap distance. Here, from Fig. 2(b), the slight change of the scattering spectra is attributed to the smaller polarization force in gap region between two nanodisks, which agrees well with the results of Refs.[13, 29]. Similarly, the scattering spectra are also strongly affected by the properties of materials in the nanodimer for the perpendicular polarization of the incident light.

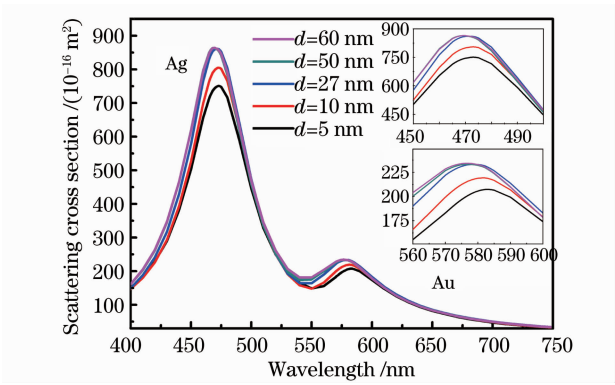

Fig. 7 Scattering spectra as a function of the gap distance with the polarization direction of the incident light perpendicular to the long axis of the bimetallic nanodimer (details of LSPR peaks of silver and gold nanodisks are shown in the upper inset and the lower inset, respectively)

4 Conclusion

The scattering characteristics of the gold and silver

nanodisk dimer have been systematically studied by using FEM. The effects of the polarization angle, the diameter, thickness and gap distance of nanodisks on the scattering spectra of bimetallic nanodimer are analyzed. The results show that, there are two kinds of LSPR peaks in the scattering spectra of bimetallic nanodimer, which are associated to the dipole LSPR of the silver and gold nanodisks respectively. The intensity of LSPR peak of silver nanodisk decreases with the polarization angle increasing from 0° to 90°, however, that of the gold nanodisk increases. The LSPR peaks of red-shift change with the increase of the diameter. The decrease of the thickness and the gap distance for the incident polarization parallel and perpendicular to long axis of the bimetallic nanodimer. Especially, the positions of the LSPR peaks of silver nanodisk are almost unchanged with the change of gap distance for the parallel polarization. In addition, the red-shifts of LSPR peaks in scattering spectra can be attributed to the interaction between the dipole LSPRs of the gold and silver nanodisk, while a simple dipole-dipole coupling model is adopted to qualitatively explain the physical mechanism of the scattering of the bimetallic nanodimer. It exhibits that the scattering characteristics of heterodimer plasmonic systems could be useful for designing the plasmonic devices in the practical application, such as a biological sensor and photosensitive detector.

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