Three-Dimensional Analysis of Scattering Field Interactions and Surface Plasmon Resonance in Coupled Silver Nanospheres

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Abstract Scattering field interactions and surface plasmon resonance (SPR) in coupled silver nanospheres are simulated by using the finite-element method, which includes the influences of near-field enhancements of electric field by the particle sizes, separation distances, propagation directions, as well as the polarizations of the incident wave. The proposed structures exhibit a red- and blue-shifted that can be tuned by varying the particle sizes and the separation distances, respectively. Implications for surface-enhance Raman scattering and nano-optics are discussed in three-dimensional models. The evolution of SPR and nanophotonic device with the structural variations can be designed in a controlled manner.

Keywords Silver nanospheres · Finite-element method · Surface plasmon resonances

Introduction

It is well known that the individual metallic nanoparticles can support surface plasmon resonance (SPR) in the optical wavelength range. The collective oscillations of electric charges excited and confined at the metal boundary which cause large enhancements of local fields near the particles. Such enhancements play an important role in surface-enhanced Raman scattering (SERS) [1]. Recently, several

works have attracted considerable interest as an alternative to conventional optics in manipulating the properties of light and applications, such as subwavelength optics nanophotonics [2], magneto-optic data storage [3], chemical and biological sensors [4], super-resolution near-field structure [5, 6], waveguides for propagating light in nanoscale structures [7], and acting as a key role in the amplification of the Raman signal of molecules adsorbed on metal particles [8].

Recently, Rechberger et al. have prepared two-identical gold nanoparticles [9] in the range of 150–450 nm and found a strong anisotropy in the scattering spectra for different polarization directions. As the electromagnetic field enhancements of scatterers with spherical shape which cannot explain the experimentally observed enhancement factors in SERS, especially the particle size less than 100 nm, a few extensions of these numerical studies have been performed on different particle shapes such as ellipsoids [10], cylinders [11], triangular [12], cubes [13], and particles surrounded by shells [14]. However, the mechanism is not clearly discussed in three-dimensional (3D) model. The resolution provided by the two-dimensional (2D) model is lower than that expected on the basis of experiments. Of the several possible reasons, the soft decay of the fields in 2D ($r^{1/2}$ instead of r^1) might be the most fundamental reason. This shows the need for 3D calculations, which, however, require much computation times.

A 3D finite-element method (FEM) model has been used to compute the optical fields that occur at near-field interactions in coupled silver nanospheres. The goal of this work is to find the influences of various factors, such as the influences of near field enhancements of electric field by the particle sizes, separation distances, propagation directions, as well as the polarizations of the incident wave,

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which produce an optical field on the gap and the edge surface of couple Ag nanospheres that is significantly smaller in size than the incident focused spot and which has appreciable intensity.

In this paper, the plasmon resonances of 3D Ag nanospheres are studied in the range of 20–100 nm. Special attention on this range is based on three reasons, i.e., the analytic solution that the field enhancement at resonance is maximized for spherical particles with diameters in this range, the experimentally observed enhancement in SERS is the largest for nanoparticle dimensions of this scale, and the technology is now reaching the state where particles of this size can be produced in a controlled manner. If such a spatial distribution of materials within 3D Ag nanosphere models that produces the desired results are able to find, it would enable us to model the Super-RENS [5, 6] writing and reading process and to better understand the Super-RENS phenomenon. In addition, by manipulating the localized SPR, local field enhancement, and near-field

coupling of nanoparticles, it is promising to be able to focus, to transport, and to interact with light in nanoscale region and are being explored for various applications in chemical and biological sensors [15], nano-photonic devices [16], plasmonics [17, 18], etc.

Simulation models and results

The 3D FEM used in this work as applied to metal nanoparticles was comprehensively described in [19]. Before investigating complex particles, it is educative to start with a simple structure. First, we investigate the case of two identical Ag nanospheres which embedded in vacuum with the same diameter d and a separation gap g illuminated with an electromagnetic plane wave. The arrangement of two identical Ag nanoparticles is along x -axis (long axis direction). Throughout this paper, the Ag permittivity data are obtained from [20–24] and corrected

Fig. 1 The SCS (a, c) and ACS (b, d) of Ag nanosphere pair with different Ag diameters d varied from 20 to 100 nm as a function of wavelengths (the d/g ratio remains constant: $d/g=5$, where g is gap spacing). In (a) and (b), the incident electric field E is along x -axis

(longitudinal mode, x -polarization), whereas in (c) and (d), it is along y -axis (transverse mode, y -polarization). Scattering near-field patterns is in the inset of a x -polarization ($\lambda=430$ nm), and c y -polarization ($\lambda=367$ nm)

by Drude model, which is including size effects [25]. Here, two different polarizations of incident wave are considered, i.e., the illumination is under normal incidence by the plane wave linearly polarized along the x and y axes, respectively. The distributions of the positive and negative polarization charges corresponding to two coupled modes are also shown in Fig. 1a. When the external field E is parallel to the y-axis (the wave-vector k is along negative z-axis), the charges oscillate in phase, and the positive and negative charges accumulate on the sides of the gap (see Fig. 1a and c); for the other illumination direction, another mode can be excited (see Fig. 1b).

The insets of Fig. 1a,c show the calculated results of the 3D near-field scattering patterns for Ag nanosphere pair with diameter $d=100$ nm, $d/g=5$ [11], and the polarization of incident plane wave is x-polarization ($\lambda=430$ nm, Fig. 1a) and y-polarization ($\lambda=367$ nm, Fig. 1c), respectively. The fields have a characteristic spherical wave appearance. The enhanced intensity in the gap between two Ag nanospheres is due to the SPR and the concentration of energy flow. In the sectional image plane shown in the inset of Fig. 1a, highly localized enhancements of field were produced between the gap of Ag nanospheres in the near-field zone only for x-polarization. The Ag nanosphere pair behave like a dipole and a very strong scattering center. The local fields from adjacent nanosphere are coupled together and produce highly enhanced evanescent fields. When free electrons of Ag nanosphere pair oscillate collectively in resonance with the incident light, the interaction may constitute a SPR. Since the boundary conditions of Maxwell's equations state that induced surface charge density is proportional to the discontinuity of the electric field component normal to the surface, only x-polarized light can excite SPR between the gap of Ag nanosphere pair. On the other hand, no enhancement appears between the gap shown in the inset of Fig. 1c, where the external field E is parallel to the y-axis since the boundary conditions now require this field to be continuous. These near-field optical signals of Ag nanospheres corresponded to the nonlinear near-field optical properties which were previously found in experiments by Nakano et al. [26] and by our research group [27, 28].

The scattering cross section (SCS) [18] indicates the amount of light scattered in the far field. In Fig. 1, we show the SCS (see Fig. 1a,c) and absorption cross section (ACS; see Fig. 1b,d) of Ag nanosphere pair with different Ag diameters d varied from 20 to 100 nm (the d/g ratio remains constant: $d/g=5$) as a function of wavelengths. In Fig. 1a,b, the incident electric field E is along x-axis (longitudinal mode), whereas in Fig. 1c,d, it is along y-axis (transverse mode). We find a distinct red shift of the surface plasmon extinction peak with increasing particle size as, shown in Fig. 1a, and a smaller but the same distinct red shift, as

shown in Fig. 1c. Note that the resonance peak shift toward larger wavelength and all the resonances broaden as the size of the d value increases [10, 11].

In Fig. 1a, for small Ag nanosphere pair, $d=20, 35$, and 50 nm, only one peak in the spectra of SCS around $\lambda=375$ nm associated with the individual nanosphere pair is visible. It is only when the nanospheres reach a diameter of $d=75$ nm that a second peak in the spectrum starts to appear. These peaks in the spectra should be associated to the plasmonic resonance modes of the 3D Ag nanosphere

Fig. 2 Spectra of SCS of single nanosphere (multiplied by factor of two) and nanosphere pair with $d=50$ nm of different separation distance g varied from 5 to 25 nm as a function of wavelengths for different polarization; **a** x-polarization (E along x axis, k along negative z axis), **b** y-polarization (E along y axis, k along negative z axis), **c** transverse-polarization (E along y axis, k along x axis)

pair system. In comparison with the results of 2D calculation system of two interacting Ag cylinders found by Kottmann et al. [11], the qualitative spectral evolution of the red-shifted plasmon peak fits quite similar. However, a second peak reported in [11] is found at any particle size is different from the particle size larger than $d=75$ nm in our case. We assign this result to a different calculation dimensions (in our case is a 3D Ag nanosphere with finite diameter, in [11] is a 2D cylinder with infinite rod length) and quantitatively difference in retardation and in differently strong contribution of higher multipoles in both systems. In addition, we can also observe in Fig. 1b,d that the more ACS is presented when the wavelength of incident is slightly smaller than $\lambda=450$ nm.

To illustrate the role plays by the separation distance between Ag nanosphere pair, Fig. 2a–c show the spectra of SCS of single Ag nanosphere and nanosphere pair with $d=50$ nm of different gap distance g varied from 5 to 25 nm as a function of wavelengths for different polarization. The gap distance g and its relative peak resonance wavelengths λ are also shown in the inset of the figure. For the case of x-polarization (E along x axis, $\mathbf{k}$ along negative z axis) shown in Fig. 2a, the peak of the SCS (resonant SP modes) splits into two different modes ($\lambda=351$ and 391 nm) as the gap distance is small enough (for example $g=5$ nm). Figure 3a shows the x–y sectional plane of scattered field patterns at light wavelengths $\lambda=351$ and 391 nm, respectively, which are corresponding to the two peaks in SCS of Ag nanosphere pair with diameter $d=50$ nm and $g=5$ nm shown in the Fig. 2a. The electric fields are relatively large in the gap and side surface, and low frequency mode

(longer wavelength, right side of Fig. 3a) is larger than that of high frequency mode (shorter wavelength, left side of Fig. 3a). It can be also observed in the right side of Fig. 3a that the field in the side surface is larger than that in the left side of Fig. 3a. These two localized plasmonic resonance modes are resulted from the splitting of single particle plasmonic resonance mode due to the effect of coupling between the Ag nanosphere pair. The resonance peak behavior of a simple dipole interaction model is well known from the molecular systems [29, 30]. These results for interacting particles, together with recent calculations for 2D analysis of elliptic and triangular [10, 12] and cylinder-shaped [4] particles, should develop our insight into the plasmon resonances in complex systems. They could help us design specific nanoparticle configurations in which extremely intense and localized electromagnetic fields are established in a controlled manner.

It can be also seen that in Fig. 2a, when the gap distance g increases, the SCS peaks in visible light region show blue-shifted, and the intensity of SCS decreases as the gap distance increases. The intensity of SCS for two particle pairs is much larger than that obtained from single particles (multiplied by a factor of two [31]). A very different behavior is observed for the other polarization direction, as illustrated in Fig. 2b. For the case of y-polarization (E along y axis, $\mathbf{k}$ along negative z axis), the SCS of Ag nanosphere pair with different gap distance is almost the same as the single particle case. Figure 3b shows the x–y sectional plane of scattered field patterns at light wavelengths $\lambda=359$ nm, which is corresponding to the peaks of SCS shown in Fig. 2b. The electric fields in Fig. 3b are relatively larger

Fig. 3 The cross-sectional plane of scattered field patterns at excited peak wavelengths, which are corresponding to the peaks in SCS of Ag nanosphere pair with diameter $d=50$ nm and $g=5$ nm shown in a Fig. 2a, b Fig. 2b and c Fig. 2c, respectively

on side surface than in the gap due to the incident electric field is along y-axis. Similar results are also shown in Fig. 3c for the transverse polarization (E along y axis, k along x axis), but the corresponding SCS peak of single particle is a little higher than those obtained from other Ag nanosphere pair. The SCS is almost the same as the single particle case. The x-z sectional plane of scattered field patterns at light wavelengths $\lambda=359$ nm, which corresponds to the SCS peaks shown in the Fig. 2c is shown in Fig. 3c, revealing weaker field intensity in second Ag nanosphere due to the wave propagation direction is along x-axis.

The field distribution at the vicinity of the Ag nanosphere pair is also very important. In Fig. 4, we show the near-field scattering field pattern between the particles along the dashed line indicated in Fig. 4a. The same geometry, as in the inset of Fig. 1a, is considered. The Ag nanosphere pair is illuminated with a unit-amplitude plane wave ($E=1$ V/m). The amplitudes shown in Fig. 4b,c give the enhancement caused by the Ag nanosphere pair. The scattering field distributions are computed for the wavelength corresponding to the maximum SCS, so a different wavelength is used for each Ag nanosphere pair's dimensions. We observe very different behavior depending on the illumination direction. For the x-polarization (the incident field E along the x-axis) as shown in Fig. 4b, the field in the gap is strongly enhanced for very small particles (for example, $d=20$ and 35 nm). For $d=20$ nm, the electric field amplitude reaches 40 times that of the incident plan wave. For larger particles (for example, $d=75$ nm and 100 nm), the field in the gap between Ag nanosphere pair decreases. This phenomenon can be explained from the SCS shown in Fig. 1a that the maximum SCS increases for larger particles, the corresponding resonance broadens immensely, indicative of a weaker near field [32].

On the other hand, for the y-polarization, a symmetrical field distribution is observed from Fig. 4c. It can be seen in Fig. 4c that the amplitude enhancement displayed such a noisy function is due to the shift in the resonance frequency. The amplitude distribution is approximately symmetrical, with a lower value of amplitude distribution in the gap and two maxima on each side of x-axis, which can be related to the polarization charges depicted in the inset of Fig. 1c. Note that the field amplitude distribution in the gap is different from those obtained from [11], which has a minimum field amplitude distribution (approximately zero) in the gap.

The discrepancy of field amplitude distribution in the Ag nanosphere pair gap between our 3D simulation model and [11] by 2D model can be attributed to the length of y-axis in a Cartesian coordinate system. For our 3D nanosphere pair, the finite length of nanosphere diameter d in the y-direction, which can be relative to the polarization charges

Fig. 4 The near-field scattering field pattern between the particles along the dashed line indicated in (a) same geometry as in Fig. 1. Two incident polarizations are considered: **b** x-polarization and **c** y-polarization

depicted in the inset of Fig. 1c. In contrast to the 2D model depicted in [11], the 2D Ag cylinder model with no polarization charges distribution in the third direction, which causes approximately zero value of amplitude distribution in the gap.

The field distributions and properties discussed above can help us to understand how the light passes through the Ag nanosphere pair. The strong fields in the gap and side

Fig. 5 **a** SCS vs. incident wave at different gap distance for the nine Ag nanosphere chains aligned parallel to the incident electric field (x-polarization). **b** Field pattern of nine Ag nanospheres aligned parallel to the incident electric field (x-polarization)

edge surface of the entrance and the exit are due to the SPR on both surfaces of the Ag nanosphere pair. The SPRs enhance the field at two ends and gap of Ag nanospheres to couple the energy into and out of the nanospheres. Therefore, the SPR play an important role on transmission enhancement through the subwavelength nanospherical array. The concentration and enhancement of electric fields in nanometer scale on the gaps and sides of the nanosphere pair may be useful in the application to optical nanodevices.

For the complex case, we present nine Ag nanosphere chains of diameter $d=50$ nm as an example. Figure 5a shows the SCS vs. incident wave at different gap spacing for the nanoparticle chains aligned parallel to the incident electric field (x-polarization). It is shown that as the gap of particles decreases, the intensity of SCS increase, and the resonant peaks are red-shifted and become broadened. In the range of shorter wavelengths, the additional higher-

order modes are excited as g decreases. These features are similar to those for particle pairs with decreasing the interparticle spacing [18, 33]. Note that the red shift becomes significant as the spacing g decreases further, and the SCS patterns are roughly alike for different gap widths. It can be also observed from Fig. 5a that the resonant peak wavelengths are $\lambda=354,496$ nm for $g=2$ nm, $\lambda=351,405,441$ nm for $g=5$ nm, and $\lambda=408$ nm for $g=10$ nm. The x-y sectional plane of scattered field patterns at resonant peak wavelengths $\lambda=351,405,441$ nm, which corresponds to the SCS peaks for $g=5$ nm is shown in Fig. 5b, revealing strong field intensity between the gaps due to the strong localized charges can occur near the gap and distorted intra particle charge distribution. Notices that the strong attractive interaction between these charges across the gap reduces the intra particle restoring forces and lowers the resonant energies.

On the other hand, Fig. 6a shows very different behaviors for the same Ag nanosphere chains aligned normal to the incident electric field (y-polarization). It can be clearly seen from Fig. 6a that the interparticle spacing

Fig. 6 **a** SCS vs. incident wave at different gap distance for the nine Ag nanosphere chains aligned normal to the incident electric field (y-polarization). **b** Field pattern of nine Ag nanospheres aligned normal to the incident electric field (y-polarization)

Fig. 7 The Scattering near-field patterns of **a** 3, **b** 9, and **c** 15 identical Ag nanospheres (with the polarization of incident plane wave along x-polarization and excitation wavelength $\lambda=381$ nm)

has relatively little effect on the resonant peaks. For a larger g , i.e., $g=5$ and 10 nm, there is only one and the same resonant peak $\lambda=365$ nm, but as g decreases to 2 nm, two resonant peaks appear. The average wavelength of the two resonant peaks ($\lambda=356$ and 373 nm) for $g=2$ nm is very close to that of the one resonance peak 365 nm for $g=5$, 10 nm. Figure 6b shows the x–y sectional plane of scattered field patterns at resonant peak wavelengths $\lambda=356, 373$ nm for y-polarization, which corresponds to the SCS peaks for $g=2$ nm, revealing a marked enhancement occurs at the edge of Ag nanosphere chains. The higher field intensity near the rims of spherical chains (edge enhancement) can excite a small intensity of evanescent wave contribution. Note the localized electric field enhancement at the edge along the silver nanosphere chains extending in several ten nanometer range from the Ag nanosphere chains. These near-field optical properties of Ag nanosphere have nonlinear near field optical properties, which were previously found in our experiments [5, 27, 28].

The main different between the two principle polarizations are, hence, the degree of confinement which is favorable for x-polarization (for the case of Fig. 5) and the available electric field distribution which is favorable for y-polarization (for the case of Fig. 6). It is important to note here that the intensity of y-polarized field in this case plays a leading role in edge enhancement. If we observe again the field maps in Fig. 6b, we can see that the light is generated mostly by the Ag nanosphere chains rim due to the excitation of the metal surface and the presence of a local dipole along the Ag nanosphere rim. In contrast, for x-polarization as shown in Fig. 5b, large amount of light

between the Ag nanospheres is responsible for the nano-particle chains aligned parallel to the incident electric field, and the free electrons of an Ag nanosphere chains oscillate collectively in resonance with the incident light. The interaction between the incident light and Ag nanospheres may constitute a surface plasmon polariton.

On the basis of our simulations, the evolution of SPR processing or nano-photonic device design of metal nanosphere chains with the structural variations can be designed in a controlled manner. Figure 7 shows the scattering near-field patterns of (a) 3, (b) 9, and (c) 15 identical Ag nanospheres, respectively, with the polarization of incident plane wave along x-polarization (k along negative z axis), $d=50$ nm, $g=10$ nm and resonant peak wavelength $\lambda=381$ nm. An interesting phenomenon is observed when the particle number of Ag nanospheres is three times of N , i.e., $3N$, where $N=1, 3, 5, \dots$. As can be seen in Fig. 7c, the Ag nanospheres of first to third, seventh to ninth, and 13th to 15th exist large amount of light in the gap than those of fourth to sixth and tenth to 12th. It is worthy to note again that the field intensity of local field in the gaps between the Ag nanospheres is quite sensitive to the gap spacing. The simulation results of Fig. 7 may be suitable for designing of SPR processing or nano-photonic device which can be regard as a neon light in this case. Figure 7 (for $g=10$ nm) which, compared to Fig. 5 (for $g=5$ nm), clearly show that the size variations of gap spacing may yield different field intensities and indicate that the intensities of local fields exhibit a nonlinear optical response with increasing nano-particle gap spacing. One of the reasons of the discontinuous intensities in the gaps for $g=10$ nm case may be attributed to the increasing gap spacing between Ag nanospheres, which are less interaction between the incident light and the Ag nanospheres and form the resonant cavity in Ag nanosphere chains with three Ag nanospheres as a set. Turning to the y-polarization case as shown in Fig. 8, where (a) $\lambda=359$ nm, k along negative z -axis, $d=50$ nm, $g=10$ nm and (b) $\lambda=361$ nm, k along x -axis (long particle pair axis), $d=50$ nm, $g=10$ nm, different concentration and enhancement of electric field patterns are shown with different k directions, and the light is generated mostly by the Ag nanosphere chains rim.

Fig. 8 The Scattering near-field patterns of 15 identical Ag nanospheres with different wave-vector k of incident plane wave: **a** $\lambda=359$ nm, k along negative z -axis and **b** $\lambda=359$ nm, k along x -axis

Conclusion

In conclusion, scattering field interactions and SPR in coupled silver nanospheres are simulated by FEM on the basis of 3D calculation models and their features are discussed, which includes the influences of near field enhancements of electric field by the particle sizes, separation distances, propagation directions, as well as the polarizations of the incident wave. Simulation results show some optical properties in our 3D calculation model are different from those of 2D models. For the case of two identical Ag nanospheres, the proposed structures exhibit a red- and blue-shifted that can be tuned by varying the particle sizes and the separation distances, respectively. We found that a distinct red shift of the surface plasmon peak with increasing particle size for the x-polarized light, but the smaller red shift for the y-polarized light. Also the propagation direction (wave vector k) can influence the SCS and ACS and found a second peak in the spectrum when the particle sizes larger than $d=75$ nm. For the case of identical Ag nanosphere chains, the main difference between the two principle polarizations are the degree of confinement of field distribution which distributes in the gap between nanoparticles for x-polarization and occurs at the edge of Ag nanosphere chains for y-polarization. The size variations of gap spacing may yield different field intensities and indicate that the intensities of local fields exhibit a nonlinear optical response with increasing nanoparticle gap spacing. The concentration and enhancement of electric fields in nanometer scale on the gaps and edge surface of the Ag nanospheres may be useful in the application to optical nanodevices, such as near-field optical imaging [34, 35], chemical and biological sensors [15], and nonradiative optical transport [36], and the combination of the near-field and dipole coupling as a possible way to optimize the sensitivity of metal particle-based sensors [37, 38]. Further, the strong frequency sensitivity of these coupled resonances could be used to produce frequency-selective devices [11].

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