Numerical Investigation of Light Scattering by Coupled Plasmonic Nanospheres Using a High-Accuracy Multidomain Legendre Pseudospectral Time-Domain Method

Shih-Yung Chung¹, Chih-Yu Wang², Chun-Hao Teng³, Chung-Ping Chen⁴, and Hung-chun Chang⁵

¹Graduate Institute of Electronics Engineering, National Taiwan University, Taipei, Taiwan 10617, R.O.C., E-mail: d95943006@ntu.edu.tw
²Graduate Institute of Electronics Engineering, National Taiwan University, Taipei, Taiwan 10617, R.O.C., E-mail: d95943034@ntu.edu.tw
³Department of Applied Mathematics and Center of Mathematical Modeling and Scientific Computing, National Chiao Tung University, Hsinchu, Taiwan 30010, R.O.C., E-mail: chunhao.teng@gmail.com
⁴Graduate Institute of Electronics Engineering and Department of Electrical Engineering, National Taiwan University, Taipei, Taiwan 10617, R.O.C., E-mail: cchen@cc.ee.ntu.edu.tw
⁵Department of Electrical Engineering, Graduate Institute of Photonics and Optoelectronics, and Graduate Institute of Communication Engineering, National Taiwan University, Taipei, Taiwan 10617, R.O.C., E-mail: hcchang@cc.ee.ntu.edu.tw

Abstract

A high-numerical-accuracy multidomain Legendre pseudospectral time-domain (PSTD) method is utilized to study the three-dimensional problem of light scattering by coupled metallic nanospheres and associated plasmonic resonances. With the multidomain technique, the simulation environment is partitioned into curvilinear hexahedral subdomains that well match the geometrical profile and material interfaces, which is an essential treatment to assure numerical accuracy, in particular, in near-field calculations. Cross sections are accurately calculated for studying the effects of different sphere radii and spacings. Interactions between two silver spheres are investigated for different incident wave propagation directions and polarizations. Electric near fields at plasmon resonances are examined.

1. Introduction

The interaction between closely spaced metallic nanoparticles, such as silver or gold ones, under optical wave incidence has been a fundamental problem in plasmonics [1]. It is important particularly due to that plasmon resonance induced strong field enhancement in the gap between the particles may occur, which offers useful applications such as the surface enhanced Raman scattering (SERS) technique for biomedical detection [2]. Therefore, accurate near-field calculation is essential for understanding the optical properties of such coupled structures. The Mie theory [1] and the multiple scattering methods [3] have been used for analytically or semi-analytically calculating lightwave scattering by spheres. But for plasmonic objects having more general geometries, analysis with suitable numerical methods can provide more flexibilities. In this paper, we employ the multidomain Legendre pseudospectral time-domain method (PSTD) method [4] to calculate and study the three-dimensional (3D) problem of scattering and near-field properties of coupled plasmonic nano-spheres, with emphasis on two closely placed spheres interacting with incident optical waves of different directions and polarizations. In particular, we demonstrate the excellent agreement of our numerical results with those using analytical approaches given in [1] and [3], and thus the PSTD method as a useful tool for simulating plasmonic problems with high-accuracy.

2. The PSTD method

The recently developed 3D multidomain Legendre PSTD method [4] is employed here as the simulation tool for studying the plasmonic behaviors of metallic nanospheres. With the multidomain technique, the simulation environment and structure are partitioned into curvilinear hexahedral subdomains that well match the outline of the geometry and material interfaces. The mesh division, subdomains, and coordinates for one-sphere problem used in this study are depicted in Fig. 1(a), where one eighth of the sphere is removed for illustrating the meshing inside, and those for the two-sphere structure are shown in Fig. 1(b). Legendre polynomials are used as interpolation bases, and the penalty
scheme is utilized to handle boundary conditions between adjacent subdomains. Also, each subdomain has its own collocation grid points which are defined by the Legendre-Gauss-Lobatto quadrature (LGL) grid points. Based on these distinct collocation points, a set of global degree-$N$ Lagrange interpolation polynomials can be further established to approximate a function and its spatial derivatives. And the temporal derivatives (time-marching) are managed by the fourth-order Runge-Kutta scheme [4].

Fig. 1. The mesh division, subdomains, and coordinates for (a) the one-sphere problem and (b) the two-sphere structure.

Dispersive characteristic of the metal at the visible light frequency range is approximated in this paper by the Drude-Lorentz model, with its relative permittivity as the function of the angular frequency formulated as

$$\varepsilon(\omega) = \varepsilon_\infty + \sum_{n=1}^\infty \varepsilon_n = \varepsilon_\infty - \frac{\omega_p^2}{\omega^2 + i \omega \gamma} - \sum_{n=1}^\infty \frac{\Delta\varepsilon_n \omega_n^2}{\omega_n^2 - \omega^2 + i \omega \gamma_n},$$

(1)

Curve-fitting is generally used to find out suitable values for the parameters in Eq. (1) that could properly match the experimentally measured dielectric constant of the real metal [5]. We exploit the genetic algorithm (GA) and choose a proper curve-fit of these parameters for silver, obtaining $\varepsilon_\infty = 1.0$, $\omega_p = 1.3877 \times 10^{16}$ rad/s, $\gamma = 1.87 \times 10^{15}$ rad/s, $\Delta\varepsilon_1 = 0.089$, $\omega_1 = 3.254 \times 10^{15}$ rad/s, $\gamma_1 = 1.165 \times 10^{15}$ rad/s, $\Delta\varepsilon_2 = 2.066$, $\omega_2 = 7.758 \times 10^{15}$ rad/s, and $\gamma_2 = 3.46 \times 10^{14}$ rad/s, which provide well fit in the optical wavelength range from 350 nm to 1100 nm.

3. Numerical Results

First, the cross sections of a single silver sphere are computed by the PSTD method. The Drude-Lorentz model with parameters listed in the last paragraph for silver is used. The calculated extinction cross sections for three different radii, 25 nm, 50 nm, and 100 nm, are compared, respectively, with the analytic Mie theory results [1], as shown in Fig. 2(a). With the maximum values of the Mie theory normalized to 1.0, the maximum absolute errors corresponding to these three radii are found to be 0.012, 0.002, and 0.006, respectively, for grid resolution $N = 12$ (i.e., $N + 1$ points for each direction of the subdomain). These errors can be improved by increasing $N$. We also compute the corresponding extinction cross sections for the same silver spheres but each coated with a 10-nm thickness dielectric shell of relative

Fig. 2. (a) PSTD calculated and analytical (Mie theory) extinction cross sections for three silver spheres with different radii, 100 nm, 50 nm, and 25 nm, for pure silver spheres. (b) Same as (b) but for silver spheres coated with 10-nm thickness dielectric shell of $\varepsilon = 3$. 

permittivity $\varepsilon = 3$, and the results are shown in Fig. 2(b). It is known that the environmental material can cause the shift of the resonant wavelength, and it is found that red-shift occurs in Fig. 2(b) compared with Fig. 2(a). In these shell coated cases, the errors are about 0.005. The obtained cross sections in Fig. 2 indicate that larger sphere possesses more resonant peaks, i.e., more higher order modes.

Next, the field coupling between two closely placed silver spheres with radii 50 nm is studied. We place these two spheres along the $x$-axis with three different spacings, 10 nm, 25 nm, and 50 nm, and consider three situations with different incident wave directions (wave vectors) and polarizations, i.e., wave vector $k_x$ with $E_y$ polarization, $k_y$ with $E_x$ polarization, and $k_y$ with $E_z$ polarization. The extinction cross sections for these three polarizations are shown in Fig. 3(a), (b), and (c), respectively, for three different spacings. When the incident wave propagates toward the spheres, the incident electric field would lead the electrons in the metallic spheres to oscillate and then radiate energy outward. In Fig. 3(a), the sphere spacing has small effect on the interaction and the resonant cross section profile because the arrangement of spheres are perpendicular to the incident electric field. But, the induced $E_x$ field still causes strong interaction between the spheres, as can be seen in the $|E_x|$ and $|E_y|$ profiles in the $x$-$y$ plane in Fig. 4(a) for the 10-nm spacing and the main spectral peak at $\lambda = 414$ nm. On the contrary, the resonant spectral shift, as shown in Fig. 3(b), is related to the spacing between the two spheres when the incident wave propagates along the $y$-axis and is with $E_z$ polarization which is parallel to the sphere arrangement. There exists quite strong field enhancement in the gap as revealed in the $|E_x|$ and $|E_y|$ profiles in the $x$-$y$ plane in Fig. 4(b) for the 10-nm spacing and the main spectral peak at $\lambda = 481$ nm. The $|E_x|$ field is about 28 times larger than the incident field. But if the incident polarization is changed to $E_z$ (with $k_y$), the cross section again has no apparent relation to the sphere spacing, as shown in Fig. 3(c), and the electric field in the gap is weaker as seen in the $|E_x|$ and $|E_z|$ profiles in the $x$-$z$ plane in Fig. 4(c) for the 10-nm spacing and the

Fig. 3. Extinction cross sections of two silver spheres with 50-nm radius for different spacings, 10 nm, 25 nm, and 50 nm. Three kinds of wave propagation directions and polarizations are considered. (a) $k_x$ with $E_y$ polarization. (b) $k_y$ with $E_x$ polarization. (c) $k_y$ with $E_z$ polarization.

Fig. 4. Electric field component magnitude distributions at the main peak wavelength of the extinction cross section for 10-nm spacing in Fig. 3(a), (b), and (c), respectively. (a) $\lambda = 414$ nm (top view). (b) $\lambda = 481$ nm (top view). (c) $\lambda = 375$ nm (side view).
main spectral peak at $\lambda = 375$ nm. Note that although both the first and third cases are with perpendicular incident polarization against the sphere arrangement, much stronger coupling occurs in the first case, due to that the two spheres are placed symmetrically with respect to the direction of wave incidence in the third case so that $E_x = 0$ at the center of the gap.

### 4. Conclusion

Plasmon resonance and near field interaction between silver nanospheres and the incident light wave have been analyzed in this paper by the multidomain Legendre PSTD method. Cross-section computations of two coupled spheres with various spacings and radii are conducted for different normally incident wave propagation directions and polarizations. The dependences of plasmon resonance and field enhancement on the sphere radius, the spacing between the spheres, and the wave polarization with respect to the sphere alignment are examined and discussed. The excellent agreement of our numerical results with those using analytical approaches [1, 3] is presented, which demonstrates the PSTD method is a useful tool for simulating plasmonic problems with high-accuracy.

### Acknowledgments

This work was supported in part by the National Science Council of the Republic of China under grants NSC98-2628-M-002-009 and NSC98-2221-E-002-025-MY2. The authors would like to thank the National Center for High-Performance Computing in Hsinchu, Taiwan, and the Academia Sinica Computing Center in Taipei, Taiwan, for providing useful computing resources.

### References