Large-scale electromagnetic modelings based on high-order methods: nanoscience applications

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Abstract. This paper presents large-scale computations and theoretical or computational aspects of the spectral element methods for solving Maxwell’s equations that have potential applications in nanoscience for surface-enhanced Raman scattering (SERS) and solar cell devices. We study the surface-enhanced electromagnetic fields near the surface of metallic nanoparticles using spectral element discontinuous Galerkin method. We solve Maxwell’s equations in time-domain and provide accuracy and efficiency of our method compared to the conventional finite difference method. We demonstrate light transmission properties for nanoslab and nanoslits, and time-averaged electric fields over the cross sections of nanoholes in a hexagonal array.

1. Introduction
This paper presents numerical algorithms for solving strongly enhanced scattering fields refracted from metallic device at nanoscale. An important contribution to the enhanced fields is due to surface plasmon (SP) excitations near the metal surface. SP excitations change light transmission or reflection properties when the metallic nanostructures such as nanohole or nanowell arrays exposed to different chemicals. The surface sensitive technology will be able to detect unique signatures of chemical and biological structures at molecular level.

The numerical challenge is to efficiently resolve the enhancement interactions with as few as grid points possible and to accurately represent the sharp jumps in material properties that arise at metal interfaces. Lower-order methods such as first- or second-order approximation, including the popular finite-difference time-domain (FDTD) method, have limitations in representing the materials accurately for complex geometries because of staircase discretization errors. Optimizing response of such nanomaterial devices involves many variables in the structure with size Very small grid spacing and very large computational resource are required to obtain accurate fields. Our studies shows that FDTD method faild to provide reasonable profiles near the metal surface even with very fine resolution.

Here we use the spectral element discontinuous Galerkin (SEDG) method with meshes conforming the complex geometry of nanostructured materials with sharp edges and nanosholes with multilayer composite of metals and dielectrics. The enhanced local field are dependent on the polarization of the incident light, nanoparticle size, the thickness of layers, etc. We will demonstrate numerical studies of the different parameters and compared with FDTD results.
2. Formulation

We consider the Maxwell’s equations with Drude model in SI unit. With an auxiliary differential equation (ADE) for the Drude model defined as in [3], [4], our governing equations are written as follows:

\[
\frac{\varepsilon}{\partial H}{\partial t} = -\nabla \times E, \tag{1}
\]

\[
\frac{\mu}{\partial E}{\partial t} = \nabla \times H - J, \tag{2}
\]

\[
\frac{\partial J}{\partial t} = \alpha J + \beta E, \tag{3}
\]

where the field vectors are defined as \( E = (E_x, E_y, E_z) \), \( H = (H_x, H_y, H_z) \), and \( J = (J_x, J_y, J_z) \) for electric density, magnetic density, and current density, respectively. The phenomenological parameters in dielectric media are

\[
\epsilon = \epsilon_0 \epsilon_r, \quad \mu = \mu_0 \mu_r, \quad \alpha = 0, \quad \text{and} \quad \beta = 0 \tag{4}
\]

and in the metallic region are

\[
\epsilon = \epsilon_0 \epsilon_r, \quad \mu = \mu_0 \mu_r, \quad \alpha = -\Gamma_p, \quad \text{and} \quad \beta = \epsilon_0 \omega_p^2. \tag{5}
\]

The parameters \( \epsilon_0, \mu_0, \epsilon_r, \mu_r, \Gamma_p, \) and \( \omega_p \) indicate permittivity of free space, permeability of free space, relative permittivity, relative permeability, Drude damping coefficient, and plasmon frequency. The relative permeability is \( \mu_r = 1 \) for most of the cases and the parameters \( \epsilon_r, \mu_r, \Gamma_p, \) and \( \omega_p \) are defined as follows:

\[
\epsilon_r = \begin{cases} 
1.0 & \text{(air)}, \\
1.33^2 & \text{(water)}, \\
1.5^2 & \text{(glass)},
\end{cases} \quad \text{and} \quad \epsilon_r = \begin{cases} 
1.0, & \Gamma_p = 0.2367 \text{ eV}, \quad \text{and} \quad \omega_p = 7.32318 \text{ eV (gold)}. \tag{6}
\end{cases}
\]

Our simulations are based on the spectral element discontinuous Galerkin (SEDG) code, NekCEM, which has been designed for computational electromagnetics on large-scale parallel platforms. Our SEDG method uses tensor-product of \( N \)-th order Legendre-Lagrange interpolation polynomials based on the Gauss-Legendre-Lobatto grids, resulting in \( n \approx EN^3 \) degrees of freedom for each field component [1].

3. Numerical convergence

We present validation of NekCEM with light transmission calculations for 2D nanoslab and 2D nanoslits provided with comparison to the FDTD results. Figures 1 shows nanoslab with 24 nm thickness. Figure 2 shows nanoslit with 24 nm thickness and 100 nm distance between slits. The nanoslab and nanoslits are placed between air and glass. For the configuration of Figure 1, we can obtain analytic solutions for the transmission coefficient for various range of wavelength. Figures 3 and 4 show the transmission coefficient curves for the wavelengths between 300 and 1000 on different mesh sizes with NekCEM and FDTD. We observe that NekCEM results of \( N = 5 \) are equivalent to the results of FDTD with 0.5 nm case. From the Figures 1-2, one can see the spectral element mesh. For \( N=5 \), it is approximately equal to 4 or 5 nm mesh size in FDTD. But the level of accuracy is close to the case of 0.5 nm of FDTD. Tables 1 and 2 show the convergence of NekCEM and FDTD for the wavelength \( \omega=501.11017 \text{ nm} \). The coarse grid with \( N = 3 \) or \( N = 5 \) already reached to 6 digit accuracy for NekCEM, whereas FDTD results are only 3 digit accuracy even with 5 times finer grids with 0.5 nm grid spacings.
Figure 1. 2D nanoslab with size of 24 nm.

Figure 2. 2D nanoslits with size of 24 nm.

Figure 3. 2D nanoslab: Transmission $T$ for wavelengths $\omega=300-1000$.

Figure 4. 2D nanoslits: Transmission $T$ for wavelengths $\omega=300-1000$.

Table 1. Convergence comparison for 2D nanoslab

| NekCEM | FDTD  |
|--------|-------|
| N=3    | T=0.33743298 err=8.45e-05 |
| N=5    | T=0.33751212 err=5.43e-06 |
| N=9    | T=0.33751590 err=1.66e-06 |
| N=13   | T=0.33751589 err=1.67e-06 |
| N=15   | T=0.33751589 err=1.67e-06 |

4. Nanohole applications and parallel performance
We use meshes that conform to the material with high-order polynomial approximations [1] which enable us to handle complex geometry of nanostructured materials with sharp edges with
Table 2. Comparison for 2D nanoslits

| NekCEM          | FDTD                                      |
|-----------------|-------------------------------------------|
| $T = N/A$ (analytic solution) for $\omega = 501.11017$ nm | N= 3 $T = 0.1090288060495094$             |
|                 | N= 5 $T = 0.1081468232317703$             |
|                 | N= 9 $T = 9.9669399705373168E-002$       |
|                 | N=11 $T = 9.8431225244869594E-002$       |
|                 | N=13 $T = 9.7112908185410532E-002$       |
|                 | $T = 0.1090288060495094$ (dx=4.0 nm)      |
|                 | $T = 9.9669399705373168E-002$ (dx=4.0 nm) |
|                 | $T = 9.8431225244869594E-002$ (dx=2.0 nm) |
|                 | $T = 9.7112908185410532E-002$ (dx=1.0 nm) |
|                 | $T = 9.6578478E-02$ (dx=0.5 nm)           |

multilayer composite of metals. The enhancement factor and local field distribution are studied depending on the polarization of the incident excitation light, nanoparticle size, the thickness of layers, etc. We have simulated a unit-cell of 3D nanohole system on hex-lattice. The radii of nanoholes are 50 nm and their center-to-center distances are 200 nm. The thickness of nanogold is 25 nm with the radii of nanoholes of 50 nm and their center-to-center distances of 200 nm. The thin nanosilver with holes are placed on the glass substrate and filled with water on the top. Figure 5 shows time-averaged $E^2$ profile at the bottom of silver with y-polarized incident field. Our SEDG simulations show that the SP excitations around the metallic nanohole surface are well-captured with physically reasonable profiles as shown in the Figures 5-6 and have good agreement with the results from FDTD method on finer grids.

![Figure 5. 3D nanohole with radius of 50 nm.](image1)

![Figure 6. 3D nanohole with radius of 50 nm.](image2)

Table 3 shows its parallel performance and scalability for NekCEM. NekCEM achieves 73 percent efficiency at $P = 4096$ for $n/P = 2734$, which is a relatively small number of points per processor. Scaling studies of the latest version of Nek5000 reported in the 2008 SciDAC article showing > 50 percent efficiency for Navier-Stokes computations on $P = 65,536$ for
\(n/P = 7,300\) \cite{2}. For similar \(n\) and \(P\), we expect NekCEM to achieve > 75 percent efficiency given the improved communication characteristics of the DG formulations which has no edge or corner-exchanges) and absence of elliptic solves in NekCEM.

Table 3. Parallel Performance of NekCEM for a Strong-Scaling Test

| nProc | Time (sec) | Ideal | Speedup (Ideal) | Efficiency |
|-------|------------|-------|-----------------|------------|
| 64    | 3.305E+3   | 3.305E+3 | 64.0 (64)      | 1.000      |
| 128   | 1.702E+3   | 1.652E+3 | 124.2 (128)    | 0.970      |
| 256   | 9.520E+2   | 8.262E+2 | 222.1 (256)    | 0.867      |
| 512   | 4.814E+2   | 4.132E+2 | 439.5 (512)    | 0.858      |
| 1024  | 2.831E+2   | 2.065E+2 | 747.1 (1024)   | 0.729      |
| 2048  | 1.400E+2   | 1.032E+2 | 1510.9 (2048)  | 0.737      |
| 4096  | 6.870E+1   | 5.164E+1 | 3078.9 (4096)  | 0.751      |

5. Conclusion
We studied the surface-enhanced electromagnetic fields near the surface of metallic nanoparticles using spectral element discontinuous Galerkin method. We solve Maxwell’s equations in time-domain and provide accuracy and efficiency of our method compared to the conventional finite difference method. We demonstrate time-averaged electric fields over the cross sections of nanoholes in a hexagonal array. We demonstrate parallel performance on the ALCF BG/P showing 75 percent efficiency.

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