Computation of the Field Enhancement by Small Facet Angles of Metallic Nanoparticles: Adaptive Remeshing for Finite Element Method

F. Mezghani, D. Barchiesi, A. Cherouat, T. Grosges, and H. Borouchaki
Project Group for Automatic Mesh Generation and Advanced Methods
Gamma3 Project (UTT-INRIA), University of Technology of Troyes, France

Abstract— The accurate calculation of the electromagnetic field enhancement around nanoparticles that exhibit facets with small angles is of interest to characterize their efficiency, given the experimental reproducibility of such structures. The finite element method is efficient to compute the enhancement of the field intensity at the surface of nanoparticles although the numerical results strongly depends on the mesh of the domain. An adaptive remeshing method is shown to be robust where the classical refinement method fails. The adaptive refinement method uses *a posteriori* error estimator with interpolation method of the physical field of interest, based on the residual method. The strategy of this method is to remesh the domain of calculation on the basis of the map of the physical and geometrical size that will ensure compliance with the initial geometry and the accuracy of the physical solution. The numerical application shows that the intensity enhancement computed near the vertex between facets with small angles (5°) can reach 23%.

1. INTRODUCTION

The outstanding optical properties of metal nanoparticles are used extensively in nanoscience [1]. As a matter of fact a huge local electric field is generated under excitation by an electromagnetic field (laser source). This field enhancement due to plasmon resonance in metallic nanoparticles produces a light source of nanometric size that is of interest among all in biology and nanotechnology applications, such as cancer therapy, drug delivery, fluorescence or spectroscopy enhancement. The optical response depends strongly on the size and on the shape of the nanoparticle [1, 2]. Therefore intense research activities occur on the synthesis of nanoparticles with control of their size and shape, and on related simulations of the field enhancement for their optimization. In some experimental cases, the shape of gold nanoparticles exhibits facets that are related to their mode of elaboration [3, 4]. Consequently an accurate method of calculation of the field around such nanostructures could help to their optimization.

The finite element method is suitable for the calculation of the electromagnetic field around complex geometries. It allows the control of the accuracy while ensuring the convergence of calculation through the adaptation of mesh. The adaptation of mesh can use various types of methods. These different techniques of adaptation lead to the refinement (or coarsening) of the mesh to meet the accuracy target deduced from a physical or geometrical estimator of error [5–7]. However the efficiency of the remeshing loop for the FEM depends on the error estimator and the adaptation strategy. In this study the remeshing process uses the interpolation of the physical solution of the problem obtained from the Finite Element Method at step t to calculate an approximation of the solution on the new mesh (step $t + 1$). An *a posteriori* error estimate enables the generation of a size map associated to the mesh elements. Then the construction of an adapted mesh conforming to this size map and the interpolation of the solution between the new and old meshes is repeated until the desired accuracy is obtained [8–11]. The application of this technique for electromagnetic modeling was shown to be efficient [8–11] but the calculation of the field enhancement near facets with small angles has never been investigated despite the fact that experimental shapes of cristalline structures exhibit such pattern. The purpose of this study is to evaluate accurately the field enhancement produced by the apex of the triangle constituted by two adjacent facets. The problem is hard, the angle between the considered facets being 5°. Indeed this problem cannot be solved with Finite Difference Time Domain (FDTD), Discrete Dipole Approximation (DDA) nor Green's tensor methods that are not able to give the field enhancement exactly on the surface of nanoparticles with accuracy and therefore to describe such small changes of slope in the shape of the nanoparticle.

The Section 2 is devoted to the description of the electromagnetic model used and its formulation for Finite Element Method. In Section 3 the refinement and the remeshing adaptive methods are compared. The numerical simulation of the field intensity enhancement is proposed before concluding.

2. FORMULATION OF THE ELECTROMAGNETISM PROBLEM FOR FINITE ELEMENT METHOD

We consider a nanoparticle with an overall spherical but polygonal shape and center O. The metallic gold nanoparticle has complex relative permittivity ϵ_1 and is immersed in a dielectric medium with a relative permittivity ϵ_2. Both media are linear, homogeneous and isotropic. This nanoparticle is illuminated by a monochromatic polarized plane wave $\mathbf{E}_i = \mathbf{E}_0 \exp(\imath k_0 ct)$ with $\mathbf{E}_0$ the amplitude vector of the incident electric field, $k_0 = \frac{2\pi}{\lambda_0}$ the modulus of the wave vector of the illumination (λ_0 being the wavelength in vacuum), and c the speed of light in vacuum. The problem consists in solving the 3D vectorial Helmholtz equation (Partial Differential Equation of elliptic type, corresponding to time harmonic wave equation) in each non magnetic material, using the curl operator $\nabla\times$ [12]:

$$\nabla \times (\nabla \times \mathbf{E}) - k_0^2 \epsilon_i \mathbf{E} = \mathbf{0} \tag{1}$$

At the interface between two materials (n_{12} being the outgoing normal vector), the boundary conditions corresponds to the continuity of the tangential component of the electric field and at the external border of the domain of calculation Ω, the radiation condition corresponds to the free propagation of the scattered field and plane wave illumination [12]:

$$n_{12} \times (\mathbf{E}_2 - \mathbf{E}_1) = 0 \quad \text{and} \quad \nabla \times \mathbf{E}_2 = \imath[k_0 n_{12} \times (\mathbf{E}_2 - \mathbf{E}_i) + \mathbf{k}_0 \times \mathbf{E}_i] \tag{2}$$

The field intensity enhancement is the maximum of the square of the intensity ($I_{\mathbf{r}} = \|E\|^2$) over the domain Ω.

Numerical data: we consider a metallic gold nanoparticle with complex relative permittivity $\epsilon_1 = -11 + 1.33\imath$ ($\lambda_0 = 632.8$) as in a previously published 2D study [14]. The amplitude of the illuminating field is $\mathbf{E}_0 = 8.10^6(1, 0, 0)\,\mathrm{V/m}$ along the x axis. The mean radius of the spherical polygon shape particle is $r = 20\,\mathrm{nm}$. The radius of the domain of calculation is $R_\Omega = 1400\,\mathrm{nm}$.

3. ADAPTATION METHODOLOGIES AND NUMERICAL RESULTS

The accuracy of the FEM solutions depends on the quality of the mesh of domain [5–11]. The first step (#0) of any resmeshing method is the meshing of the geometry of the domain. Nodes are put on surfaces between materials and in the volume Ω with respect to a geometrical map using quality of elements. The result of computation on this mesh is shown in Fig. 1. The small angle between facets is not "seen" by the numerical simulation: no field enhancement is observed near the vertex between two adjacent facets.

Therefore mesh adaption is necessary. Amongst the various methods, the r-method moves the nodes from the initial mesh. The hp-method is a combination of h- and p-methods that can change both the size of elements on the mesh (h-method) and the dimension of space of functions describing their shape (p-method) [5–7]. The refinement of mesh consists in adding nodes to the initial one that is based on geometry properties of objects. Therefore the adaption leads to an increase of the number of elements and then requires more memory and computational time to obtain the solution

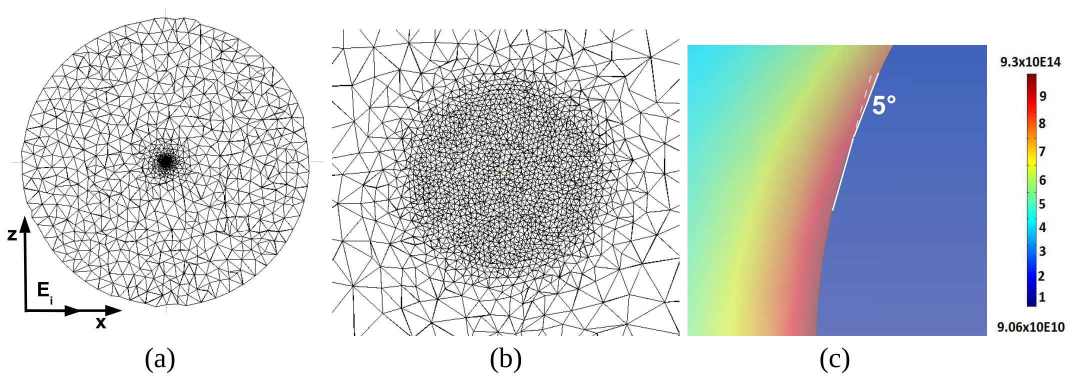

Figure 1: (a) Cut in the median plane $y = 0$ of the mesh at step #0 without remeshing, (b) zoom on the mesh around the nanoparticle) and (c) the distribution of the intensity field.

at each step of the refinement loop. On the contrary, advanced methods of remeshing allow to improve the quality of the solutions by minimizing the overall error in the solutions obtained on the new mesh, with the possibility to increase or decrease locally the density of nodes. To reach this goal the error estimator indicates if the mesh density is too fine or too coarse. An efficient estimator of error must not be expensive in memory and CPU time. In this study we compare the refinement S_1 and the remeshing S_2 whose adaptation strategies are following. The first geometrical meshing (#0) is about the same for both methods. Then two methods S_1 (refinement) and S_2 (remeshing) are applied. The same solver for the finite element calculation of the electric field is used in both case.

- Refinement S_1 uses adaptation on the residual of the physical solution with h-adaptive mesh refinement (part of the software Comsol Multiphysics). The basic steps are:
  - Solve the problem on the mesh;
  - Evaluate the residual of the PDE on all mesh elements;
  - Refine a subset of the elements based on their size or of the local error indicators (the minimum and maximum element size are set automatically);
  - Repeat these steps until the requested number of refinements or the maximum number of elements are reached.
- Remeshing S_2 uses adaptation on the residual of the geometry and of the interpolation of the physical solution with h-adaptive mesh refinement and coarsening (OPTIFORM software). The basic steps are:
  - Solve the problem on the mesh;
  - Calculation of the interpolation error estimator to generate a unit metric on the physical map. It is based on the interpolation error depending on the Hessian of the finite element solution that is approximated by a local calculation of the solution.
  - Generation of an adapted meshing that meets the requirements of the metric physical card and the metric geometry for respecting the initial geometry ($h_{\min}$ and $h_{\max}$ are the minimum and maximum sizes of the elements respectively, ε is the maximum interpolation error that is tolerated in the solution);
  - Repeat these steps until the interpolation error is greater than ϵ.

  The generation of an adapted meshing can be considered as an optimization problem which consists in minimizing a norm function (L2-norm or H1-norm) related to the distance between the new and the previous solutions.

The criteria of comparison between S_1 and S_2 are the accuracy of the solution, the memory (RAM) and the CPU time. Tables 1 and 2 gives the results of both methods S_1 and S_2: for each refinement or remeshing steps: the number of nodes and elements, the parameters of the method and the maximum of the computed intensity ($\max\|E\|^2$) that corresponds to the field intensity enhancement.

Table 1: Number of nodes, tetrahedra, $\max\|E\|^2$, RAM, and computation time for the refinement strategy S_1.

Refinement step	0	1
Nodes	49 285	208 934
Tetrahedra	289 609	1 148 123
$\max\|E\|^2$ [V/m]^2	$9.3.10^{14}$	$9.28.10^{14}$
RAM [GB]	10.48	71
Time [s]	213	7 381

S_1 is efficient in the $2D$ case but we met difficulties when applied to $3D$ problem because of the calculation cost. In the investigated case of wide domain with small object (the ratio of the mean radius of particle to that of the whole domain of calculation is $1,4\%$) only one refinement is

Table 2: Number of nodes, elements (tetrahedra), h_{min}, ε, max $\|E\|^2$, RAM, and computation time for the remeshing strategy S_2.

Remeshing step	0	1	2	3	4	5
Nodes	48 781	51 851	76 796	317 642	377 681	404 107
Tetrahedra	289 605	308 333	459 179	1 922 512	2 277 531	2 432 568
h_{min}		0.35	0.35	0.35	0.35	0.35
ϵ		0.1	0.05	0.01	0.01	0.01
max $\|E\|^2$ [V/m]^2	$9.88.10^{14}$	$1.06.10^{15}$	$1.14.10^{15}$	$1.22.10^{15}$	$1.22.10^{15}$	$1.22.10^{15}$
RAM [GB]	8.72	9.57	12.89	54.53	65.95	66.14
Time [s]	214	208	292	1 568	1 942	1 932

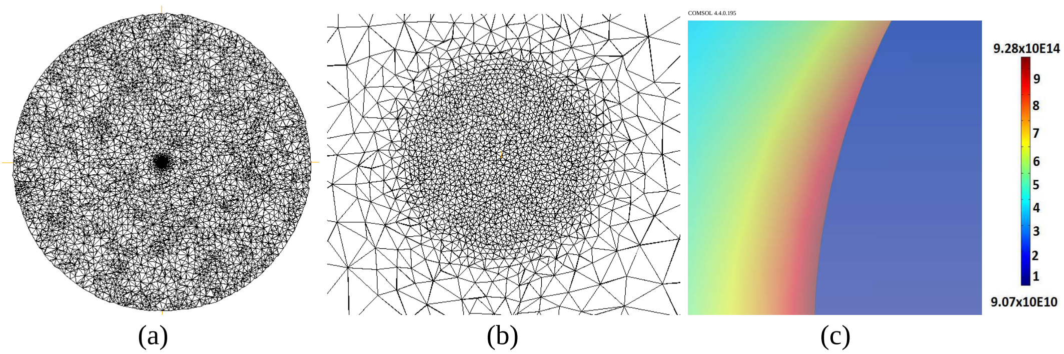

Figure 2: (a) Cut in the median plane $y = 0$ of the mesh at step 1 for the strategy S_1, (b) zoom on the mesh around the nanoparticle, and (c) the distribution of the intensity field.

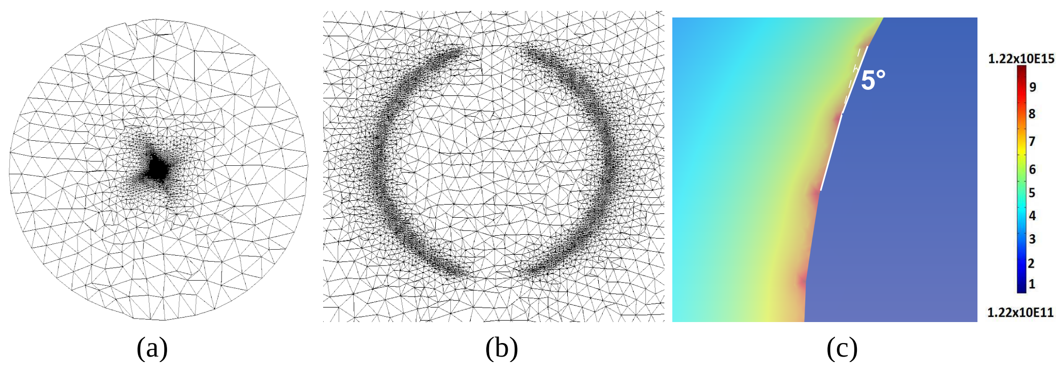

Figure 3: (a) Cut in median the plane $y = 0$ of the mesh at step 5 before remeshing for the strategy S_2, (b) zoom on the mesh around the nanoparticle and (c) the distribution of the intensity field.

possible before divergence of calculation due to the huge increase of elements. The computational time at step #1 of S_1 is about four times that of each step of S_2.

S_2 produces five new meshes to reach convergence toward the maximal error ε to 1%. The first iteration was performed to reach $h_{\min}$. The computational time remains around half an hour and the number of nodes obtained from refinement (step #1 by S_1) is reached after two remeshing steps with S_2. The value of the field intensity enhancement is stable from mesh #3 to #5. The corresponding increase of the number of elements is about 10%. The refinement S_1 guided by the physical error estimator of residual type increases the number of elements in the regions far from the particle where the field variation is smoother than in the vicinity of the particle (Fig. 2(a) compared to Fig. 1(a)). This explains the huge increase of nodes in the refinement method that prevent to reveal the field enhancement around the vertex. The performance of S_2 is illustrated in Fig. 3. The field enhancement appears in Fig. 3(c) using S_2. Near vertex the size corresponding to the minimum element size $h_{\min}$, it is about 0.35 nm and the error of the solution interpolated in each element is below the threshold of 0.01. The mesh is coarser far from the particle (Fig. 3(a)) and therefore the calculation requires less computational time and memory, the method giving a new mesh at each step, with optimized positions of nodes. Consequently, S_2 reveals that the small angle at the junctions between two facets produces an increase in the value of the intensity enhancement around 23% for an angle of tilt between the two facets of 5° (Fig. 3).

4. CONCLUSION

This paper presents a remeshing method allowing to detect the effects of the small angles in the junction between two facets in nanostructures. A comparison between refinement and remeshing methods shows that remeshing gives accurate solution unlike the refinement that fails: angles of 5° between two adjacent facets can increase the value of the intensity of the electric field of 23% near the vertex. The adaptive remeshing allows to increase the accuracy and the convergence toward a solution having physically meaning. This method can be applied to any other shapes of nanostructures, which present facets with any angle and also roughnesses.

REFERENCES

1. Hu, M., J. Chen, Z.-Y. Li, L. Au, G. V. Hartland, X. Li, M. Marquez, and Y. Xia, "Gold nanostructures: engineering their plasmonic properties for biomedical applications," *Chem. Soc. Rev.*, Vol. 35, 10841094, 2006.
2. Kessentini, S. and D. Barchiesi, "Quantitative comparison of optimized nanorods, nanoshells and hollow nanospheres for photothermal therapy," *Biomed. Opt. Express*, Vol. 3, No. 3, 590–604, 2012.
3. Liu, B., J. Xie, J. Y. Lee, Y. P. Ting, and J.-P. Chen, "Optimization of high-yield biological synthesis of single-crystalline gold nanoplates," *J. Phys. Chem. B*, Vol. 109, 15256–15263, 2005.
4. Daniel, M.-C. and D. Astruc, "Gold nanoparticles: Assembly, supramolecular chemistry, quantum-size-related properties, and applications toward biology, catalysis, and nanotechnology," *Chem. Rev.*, Vol. 104, 293–346, 2004.
5. Verfürth, R., *A Review of A Posteriori Error Estimation and Adaptive Mesh-Refinement Techniques*, Wiley and Teubner, 1996.
6. Ainsworth, M. and J. T. Oden, "A posteriori error estimation in finite element analysis," *Comput. Methods Appl. Mech. Engrg.* Vol. 142, 1–88, 1997.
7. Borouchaki, H., D. Chapelle, P.-L. George, P. Laug and P. Frey, "Estimateurs d'erreur géométriques et adaptation de maillages," *Maillage et Adaptation*, sous la direction de P.-L. George, Traité Mécanique et Ingiénerie des Matériaux, Hermès, France, 2001.
8. Grosges, T., H. Borouchaki, and D. Barchiesi, "Improved scheme for accurate computation of high electric near-field gradients," *Opt. Express* Vol. 15, No. 3, 1307–1321, 2007.
9. Borouchaki, H., T. Grosges, and D. Barchiesi, "Improved 3D adaptive remeshing scheme applied in high electromagnetic field gradient computation" *Finite Elements in Analysis and Design*, Vol. 44, Nos. 1–2, 84–95, 2010.
10. Grosges, T., H. Borouchaki, and D. Barchiesi, "New adaptive mesh development for accurate near field enhancement computation," *J. Microsc.* Vol. 229, No. 2, 293–301, 2008.
11. Borouchaki, H., T. Grosges, and D. Barchiesi, "Enhancement of the accuracy of numerical field computation using adaptive three dimensional remeshing scheme," *C. R. Meca.*, Vol. 338, No. 3, 127–131, 2010.
12. Jin, J., *The Finite Element Method in Electromagnetics.*, John Wiley and Sons, New York, 1993.
13. Barchiesi, D., E. Kremer, A. Cherouat, T. Grosges, and H. Borouchaki, "Dilation of nanonatennas induced by an electromagnetic source," *Adv. Electromagn.*, Vol. 1, No. 2, 48–57, 2012.
14. Barchiesi, D., T. Grosges, E. Kremer, and M. Lamy de la Chapelle, "Electromagnetic heat induced in meso-structures: Computation of temperature in metallic dimers," *PIERS Online*, Vol. 7, No. 5, 406–410, 2011.