Accuracy of local field enhancement models: toward predictive models?

Dominique Barchiesi, B. Guizal, Thomas Grosges

Accuracy of local field enhancement models: toward predictive models?

D. BARCHIESI[1,✉]
B. GUIZAL[2]
T. GROSGES[1]

[1] University of Technology of Troyes, Laboratory of Nanotechnology and Optical Instrumentation – Charles Delaunay's Institute FRE CNRS 2848, 12 rue Marie Curie – BP 2060, 10010 Troyes cedex France
[2] University of Franche-Comté, Optical Department of FEMTO UMR CNRS 6174, 13 route de Gray, 25030 Besançon cedex France

ABSTRACT An accurate computation of field enhancement in the vicinity of metallic nanostructures is fundamental for the prediction of different physical phenomena such as SERS or fluorescence, and also for the design of nanostructures for specific applications. Several numerical models have been developed and are used to compute the field enhancement. Nevertheless, its evaluation can be very tedious and boring due to the plasmon resonance increasing the intensity level, and to the discontinuity of the field near the material edges. The behavior of commonly used computational codes is investigated in order to identify the convergence problems, and to propose some solutions to control the accuracy in the computation of the field enhancement.

1 Introduction

Since the early developments of near-field optical microscopies, the variety of their applications has not ceased to widen. Recently, a new area of the nanotechnology appeared: the plasmonic. The applications concerned with this field go from telecommunications to biology, passing by the physics of the mesoscopic domain. In plasmonics, the use of electromagnetic field enhancement obtained either by the antenna effect, dependent on the electromagnetic singularities around material tips or by plasmon resonance, opens the way to local measurements of interaction between radiation and matter [1].

However, a direct measurement of this enhancement is not yet possible and the lithography techniques used to build the nanostructure are rather expensive. Therefore, it is necessary to use theoretical forecasts to optimize and functionalize the produced nanostructures. The field enhancement is strongly dependent on physical parameters like wavelength, shape, size, polarization, angle of incidence, and material properties. In the particular case of SNOM (scanning near-field optical microscopy) many papers are dedicated to the computation of the near-field diffracted by a nanometric probe. The computed intensity enhancement varies from 3000 [2] to 10 000 [1], between 10 and 160 000 [3] and it has been shown to be ten orders of magnitude in amplifying medium than what would be expected for plasmon resonance in a passive medium [4]. The accurate computation of the field enhancement is necessary to carry out predictions and to design the resonant structures for SERS, and TERS.

In the near-field optical community, several numerical methods have been used for a long time. In principle, all the methods which make possible the computation of the optical near-field around metallic nanostructures, illuminated by an external source, could be used. Nevertheless, only a few of them are able to deal with high local variations of the electric field. This requirement is directly connected with the numerical convergence and the discretization of the problem. Moreover, the discretization required to compute the solution is in the direct space for methods like finite difference time domain (FDTD) or the finite element method (FEM), or in the Fourier space for the spectral methods (like the differential, coupled wave (CWM), volume or surface integral (Green) methods). Let us note that the difference between the methods is sometimes subtle due to the fact that they can be close to each other or even mixed. As an example, the CWM could be considered as a modification of the classical differential methods, or the finite element method can be applied, with some refinements, to any set of differential equations [5].

Various methods (commercial or free of use codes) must be carefully handled especially in the case of high field enhancement computation. To illustrate this important fact, we will present an example of an application in Sect. 2. Section 3 will be devoted to investigating the convergence of three numerical methods, before concluding and proposing possible improvements of the methods.

2 The plasmonic design: the resolution of the inverse problem

An initial problem that has to be solved before designing nanostructures is the measurement of their true permittivity and shape. At present, the bulk permittivities are commonly used in simulations to compute the field enhancement. Moreover, the measurement of the topography of metallic nanostructures with atomic force microscopy (AFM) suffers from the convolution of the AFM probe shape by that of the nanostructure. Therefore, the metrology applications must be performed through an inversion procedure to deduce

✉ Fax: +33-(0)3-25-71-84-56, E-mail: dominique.barchiesi@utt.fr

values from the data recorded experimentally, as it is done in the medical domain. To take into account the important processes of the image formation, an accurate model has to be developed. The principle of an inversion procedure is based on the use of an accurate model to deduce the unknown parameters from experimental data (Fig. 1).

An inversion procedure based on an evolutionary scheme was recently used to perform the recovery of unknown experimental parameters [6]. Also, it was shown theoretically that the measurement of the permittivity of nanostructure is possible [7]. The direct method used in this inversion procedure was FDTD including, an improvement of the model of dispersion [8]. The use of classical FDTD (with Cartesian mesh, commonly used in commercial codes with regular or non-regular Yee's cells) to compute the field enhancement around a metallic nano-wire has been investigated in [9]. It has been shown that to achieve realistic spectroscopic studies, the models must take into account both the geometric and dispersive characteristics of the complex materials. In particular, an appropriate refinement of the grid must be used in order to assure the stabilization and the convergence to the physical solution, especially in the regions where a strong confinement of the light around the nanostructure occurs. In FDTD, artificial plasmons are generated at the vertices of metallic cells, which may prevent computation of the reliable field enhancement [9–11]. In plasmonics, the problem of accuracy and the difficulty of obtaining convergence of the models is illustrated in Fig. 2 of [12], where the computed transmission of a square coaxial aperture can vary by more than 10% between FDTD and the Fourier modal method.

In the following section, three popular methods for field computation will be investigated in the case of metallic nanostructures.

3 Convergence of CWM, FEM and Green methods

To simplify the discussion, the field enhancement is computed in the vicinity of a nano-wire with rectangular shape ($200 \times 20\,\mathrm{nm}^2$) with relative permittivity $\varepsilon = -10$. This permittivity corresponds to gold material at a wavelength $\lambda = 632.8\,\mathrm{nm}$. Due to the negative value of the relative permittivity, the excitation of plasmon resonance (which depends on the geometry of the object) can occur. The imaginary part of the permittivity is neglected in order to bench the numerical models with high requirements. Actually, the imaginary part would contribute in widening the resonance peaks and therefore would facilitate the convergence of the methods. Nevertheless, this hypothesis is not restrictive, and all the numerical models used here can deal with complex permittivities.

The nanostructure is illuminated at normal incidence, with a *p*-polarized plane wave. The computation of the electric field is achieved in reflection, 2 nm above the nanostructure. The intensity enhancement is the square modulus of the electric field and strongly varies at the edges of the nanostructure. Therefore, high order evanescent diffracted waves are required to converge to the solution. Moreover, in order to compare the efficiency of the three investigated methods, the Matlab codes run on the same personal computer.

In the following subsections, the convergence of the Coupled Wave Method, the Volume Integral Method, and the Finite Element Method will be investigated.

3.1 *The coupled wave method (CWM)*

The coupled wave method (CWM) is widely used due to its implementation simplicity and effectiveness, especially in the case of lamellar structures. This method is a modification of the differential method [13]. In [14] the method was fully described and was applied to the case of a glass aperiodic structure. The code used for the present CWM simulations is the same as in [14], but in this work, the sample is illuminated with a plane wave rather than a gaussian beam. Therefore, the nanostructure is a grating made of gold wires with rectangular shape with a 400 nm period (Fig. 2). The convergence of this spectral method depends only on the number of diffracted waves involved in the computation.

The field enhancement at the edges of the nanostructure is clearly related to the singularity of the electric field in the region where the permittivity abruptly changes. In CWM, the convergence parameter is the truncation order M, which limits the number of diffracted plane waves involved in the computation [14]. Figure 3 shows the convergence of the method with M, by focussing on the maximum of the intensity in the computational window. The regular discretization step in Fourier space is essential to describe resonance in metals and to assure the convergence of the method (which is slower than in the dielectric case [14]). Figure 3a–c show the intensity computed with $M = 80$, $M = 360$ and the convergence as a func-

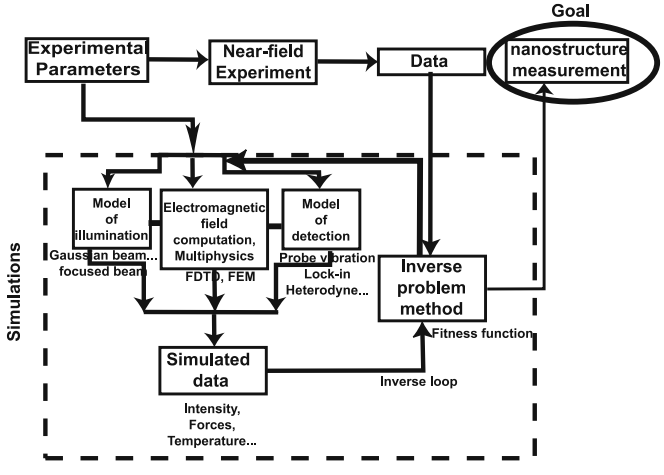


FIGURE 1 Principle of the inverse scheme to measure unknown parameters from experimental data. An accurate model of the imaging process and an efficient inversion procedure are needed

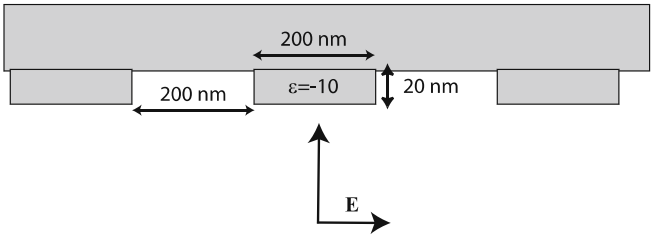
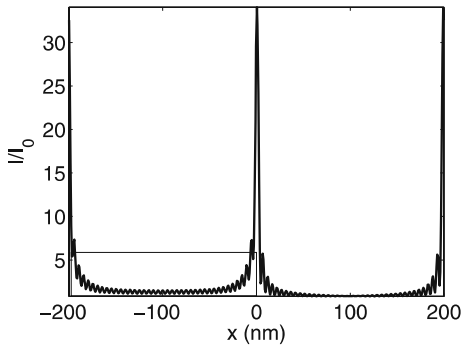
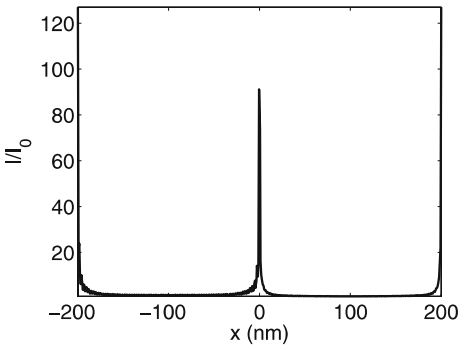


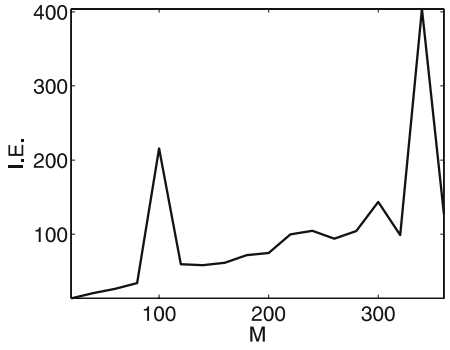
FIGURE 2 Schematic of the gold grating with aspect ratio 0.5, height 20 nm, and period 400 nm. The incoming light is a plane wave



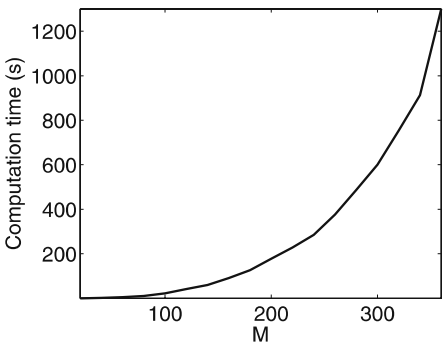
a Intensity profile ($M = 80$).



b Intensity profile ($M = 360$).



c Intensity enhancement as a function of M.



d Computation time as a function of M.

FIGURE 3 The intensity enhancement computed from CWM for a grating with aspect ratio 0.5, height 20 nm, and period 400 nm. The angle of incidence of the incoming light is 0. The intensity enhancement is computed 2 nm above the grating tracks

tion of M, respectively. With the computer used (512 Mo RAM), a memory overflow occurs if M is greater than 360. As shown in Fig. 3c, the convergence is still not reached, even if the energy conservation is satisfied with an accuracy better than 10^{-6}. The computation time increases drastically with M (Fig. 3d). This result confirms the study of convergence in [14], where $M = 600$ is needed to reach convergence, in the case of a dielectric sample. Let us note that the method converges more rapidly and much less harmonics are necessary to reach convergence if either the near-field, a few nanometers away from the sample, or the far-field is required to be diffracted in a given direction (or a scattering section).

In this section, it has been shown that controlling the convergence is essential in the case of computation of the intensity enhancement in the vicinity of the edges of a periodic nanostructure. The investigated case corresponds to a pure reciprocal space method, where the number of plane waves involved in the model of diffraction is the key parameter of the convergence.

Another class of methods uses the Green tensors. In the following section, the classical volume integral method is used to investigate the influence of both the mesh of the nanostructure geometry and the truncation order for the computation of the intensity enhancement.

3.2 *The volume integral or green volumic method (VIM)*

The volume integral method (VIM) is based on the computation of the Green tensors associated with the discretization of a defect above an interface. The convergence of the method depends on the number of cells or dipoles used to mesh the defect and on the discretization in Fourier space used to compute the Green tensor. In this paper, the same numerical method as described in [15], is used to model the diffraction by the nanostructure depicted in Fig. 4.

Like in the CWM case, the goal is to compute the field enhancement 2 nm above the edges of the nanostructures, where a high gradient of electric field is expected. In the following, 2 nm square cells or dipoles are used to mesh the nanostructure. It must be noticed that the size of the dipoles is much smaller than the $\lambda/10$ discretization step commonly used in far-field simulations. This small size is necessary to reach convergence. Figure 5c shows the convergence as a function of the discretization used to compute the Green tensor previously defined as the truncation order M. The value of the intensity enhancement cannot be computed with $1 \times 1\ \text{nm}^2$ square dipoles, due to the insufficient available memory. The intensity enhancement is equal to 24.6082 with $M = 180$

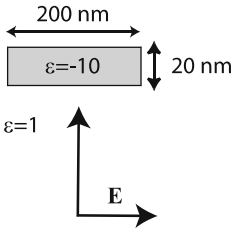
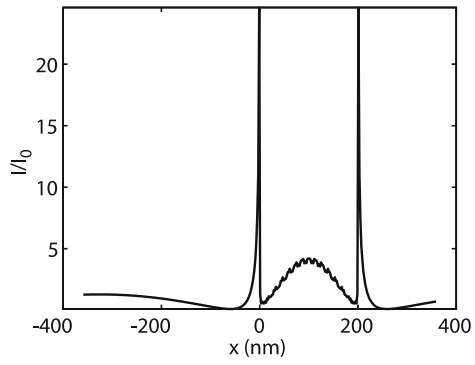
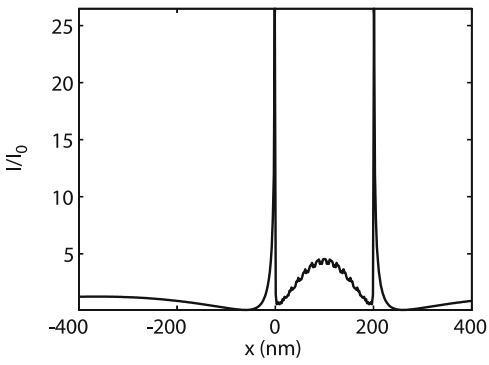


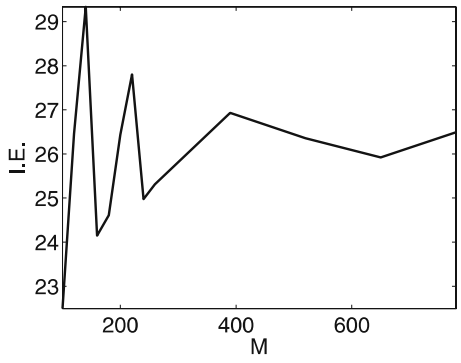
FIGURE 4 Schematic of the gold nanostructure with height 20 nm, and period 400 nm. The incoming light is a plane wave



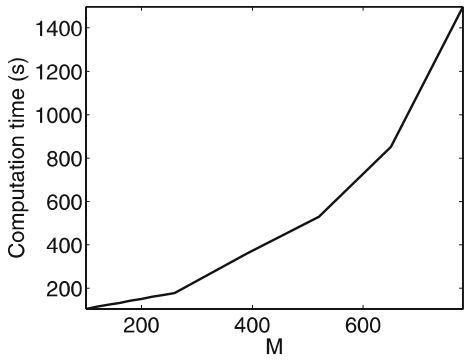
a Intensity profile ($M = 180$).



b Intensity profile ($M = 360$).



c Intensity enhancement as a function of M.



d Computation time as a function of M.

FIGURE 5 The intensity enhancement computed with VIM for a nanostructure with height 20 nm, and lateral size 200 nm. The angle of incidence of the incoming light is 0 degrees. The intensity enhancement is computed 2 nm above the nanostructure. The size of the mesh cells is $2 \times 2\,\text{nm}^2$

and 26.4890 with $M = 360$, respectively. These results were obtained close to the maximum of the available memory in the PC.

Figure 5d can be compared to Fig. 3d: the computation time increases rapidly with the order M of truncation. This parameter has to be included in the choice of the models. The performance of the processor influences the computation time. Let us notice that the considered nanostructure is described in a 2 dimensional geometry. Of course, modeling 3D nanostructures would require more computing time and memory resources.

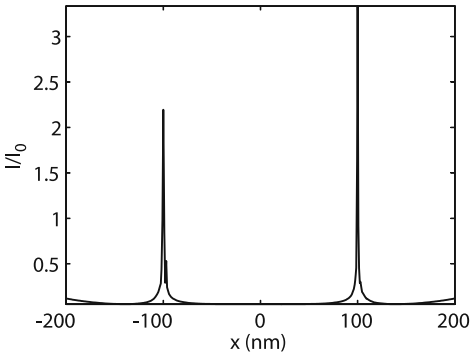
After the study of periodic structure with the CWM, where only the order of truncation was involved in convergence, and those of isolated nanostructures were investigated with the VIM, and where both spatial and truncation influenced the computation of the intensity enhancement, the convergence problem will be illustrated using a pure direct space method: the finite element method applied to the propagation equation.

3.3 *The finite element approach to solve the Helmholtz' equation*

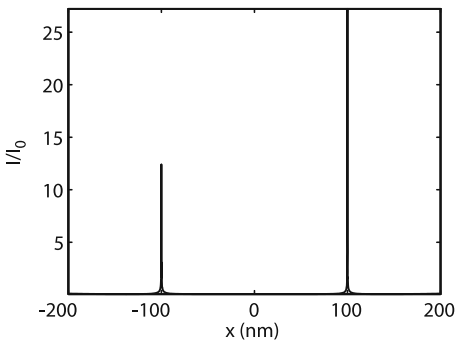
The last method considered in this study is widely used to solve different kinds of differential equations (such as in mechanical systems, thermic, fluid mechanics etc.) and has been used to compute the near-field around nanostructures in the Green formalism [16]. It is based on the variational formulation of the differential equation and the resolution is achieved by its minimization on an adaptive non-regular non-Cartesian mesh of the computation window, taking into account the boundary conditions on the edges of this window (external boundary conditions). In this paper, the FEM is used to solve the Helmholtz' equation [17], the boundary conditions are imposed on the incoming plane wave field (Dirichlet's conditions) on the edges of the computational domain [18]. The drawback of the finite volume element method is that the whole domain of computation must be meshed, therefore a huge number of cells is needed. Due to the use of an adaptive non-regular non-Cartesian mesh, the refinement of the grid is effective only where high gradients of the solution are located. This may help to spare computer memory. This point has been addressed in [9] by comparison of the FEM with the classical FDTD with regular/non-regular Cartesian Yee's cells.

In this study, the incoming plane wave amplitude is set on the edges of the external boundaries. Consequently, the diffraction of the nanostructure is neglected in this region which assures the unicity of the solution. Nevertheless, to be valid, a sufficiently wide domain of computation must be used (relatively to the scattering cross-section of the nanostructure). In this study, the domain of computation is a 3λ square domain and also the studied nanostructure, situated at the center of the domain is a rectangle of width 200 nm and heigth 20 nm. The intensity in Fig. 6 is computed 2 nm above the nanostructure.

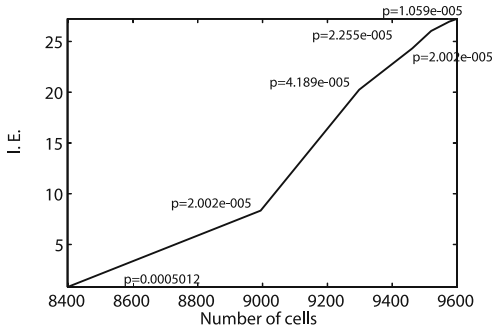
Figure 6 shows the influence of the gridding in the adaptive mesh process. Figure 6a–b shows the confinement of the computed intensity with the initial mesh and after seven re-



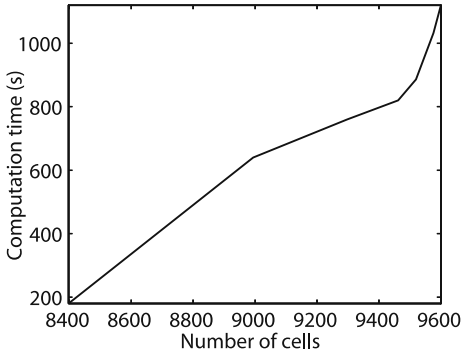
a Intensity profile (8398 cells).



b Intensity profile (9600 cells).



c Intensity enhancement as a function of the number of cells to reach a given accuracy p.



d Computation time as a function of the number of cells.

FIGURE 6 The intensity enhancement computed from FEM for a nanostructure with height 20 nm, and period 400 nm. The angle of incidence of the incoming light is 0 degrees. The intensity enhancement is computed 2 nm above the nanostructure

gridings. Even if the problem is symmetric, the height of the peaks is not the same in the plot due to a small decay between the left cell and the right one above the nanostructure edges. As expected, the model is very sensitive to the mesh refinement. The accuracy, obtained for the computation of the field is indicated in Figure 6c and is the maximum of the variation of the field between two adjacent cells. This accuracy is automatically computed in the region of the intensity enhancement, where the gradient of the field is maximum. The advantage of the adaptive non-Cartesian mesh is shown in Figs. 6c–d: the computation time and the number of cells varies slowly with the accuracy p (or the intensity enhancement) and is mainly related to the iterative inversion algorithm process. The smallest cell size reaches 0.1 nm to obtain stabilization of the solution. In this case as for the previous ones, the convergence is probably not completely reached before the memory overflow, but the lateral size of the peaks is smaller, due to the non-regular and non-Cartesian mesh. Let us note that the intensity enhancement is close to those given through the VIM.

4 Conclusion

In this study, it has been shown that modeling the intensity enhancement near nanostructures edges with classical codes can induce slow convergence (i.e. a great amount of memory, and a large computation time is needed in both the periodic or non periodic cases). The three methods used in this paper use the experimental values of permittivities in contrast to FDTD for which an analytical model of dispersion is required. Therefore, situations where gain or non linearities exist may be described is spite of a probably larger computational time.

Some requirements to accurately compute the intensity enhancement near nanostructures are:

- the use of adaptive non-regular and non-Cartesian mesh. Indeed, the use of Cartesian grid in FDTD or VIM will result in failure to reach convergence.
- the use of a remeshing process and control of the accuracy of the solution. High field gradients are expected near the nanostructures edges and therefore high spatial frequencies or high order diffracted modes are needed to achieve convergence. Moreover, cell sizes much smaller than $\lambda/10$ are required (in contrast to those commonly used) where the intensity enhancement is located near the nanostructure. In Fourier space, the contribution of the diffracted plane waves with high spatial frequency cannot be neglected.
- the use of the surface methods, for which only the surface of the nanostructure must be discretized. This way may be a memory sparing solution and could be more efficient if a non regular discretization of the surface can be considered.
- to smooth the nanostructure shape models in order to avoid the mathematical singularity of the electric field.

Let us note that the CWM, VIM and the FEM have been compared in the same conditions. The language of computation, and the computer are the same, and a similar nanostructure is considered. In this context, the codes have not reached the convergence before a memory overflow. The computation of intensity enhancement requires high performance computers, and a careful control of the convergence. These considerations are critical to tend to accurate computations of the intensity enhancement and predictive plasmonics.

REFERENCES

1 N. Calander, M. Willander, J. Appl. Phys. **92**, 4878 (2003)
2 L. Novotny, R.X. Bian, X. Sunney Xie, Phys. Rev. Lett. **79**, 645 (1997)
3 J.P. Kottmann, O.J.F. Martin, D.R. Smith, S. Schultz, Opt. Express **6**, 213 (2000)
4 N.M. Lawandy, Appl. Phys. Lett. **85**, 5040 (2004)
5 P. Harscher, S. Amariand, R. Vahldieck, IEEE Trans. Microw. Theory Technol. **50**, 433 (2002)
6 D. Macías, D. Barchiesi, Opt. Lett. **30**, 2557 (2005)
7 D. Macías, A. Vial, D. Barchiesi, J. Opt. Soc. Am. A **21**, 1465 (2004)
8 A. Vial, A.-S. Grimault, D. Macías, D. Barchiesi, M.L. de la Chapelle, Phys. Rev. B **71**, 85 416 (2005)
9 T. Grosges, A. Vial, D. Barchiesi, Opt. Express **13**, 8483 (2005)
10 R.G. Milner, D. Richards, J. Microsc. **202**, 66 (2001)
11 P. Leuchtmann, C. Fumeaux, D. Baumann, Adv. Radio Sci. **1**, 87 (2003)
12 A. Moreau, G. Granet, F. Baida, D. Van Labeke, Opt. Express **11**, 1131 (2003)
13 G. Granet, B. Guizal, J. Opt. Soc. Am. A **13**, 1019 (1996)
14 B. Guizal, D. Barchiesi, D. Felbacq, J. Opt. Soc. Am. A **12**, 2274 (2003)
15 A. Sentenac, J. Greffet, J. Opt. Soc. Am. A **9**, 996 (1992)
16 J.K. Kottmann, O.J.F. Martin, D.R. Smith, S. Schultz, J. Microsc. **202**, 60 (2001)
17 M. Born, E. Wolf, *Principles of Optics* (Pergamon, Oxford, 1992)
18 J. Jin, *The Finite Element Method in Electromagnetics* (Wiley, New York, 1993)