STATE-OF-THE-ART AND MOST PROMISING ANALYTICAL AND NUMERICAL CHARACTERIZATION TECHNIQUES FOR NANOSTRUCTURED METAMATERIALS

by Prof. Constantin Simovski

(EU FP7 Contract No. 218696-ECONAM)

2010

1 A breakthrough in the electromagnetic characterization of nanostructured materials is coming

1.1 Bulk nanostructured metamaterials

1.1.1 State-of-the-art from our previous report

In [1] we introduced the concept of characteristic material parameters (CMP) which follows from the definition of characterization. We indicated the link between CMP and electromagnetic effective parameters (EMP) retrieved from measured data or data of exact numerical simulations (this retrieval is also called post-processing). It was stated that CMP are always EMP but the opposite assertion is not always correct. From our previous reports [1, 2, 3, 4], it followed that for nanostructured metamaterials EMP retrieved in many known works cannot be referred to as CMP. Meanwhile, it follows from the classification presented in [1] that metamaterials form broad and promising class of nanostructured materials.

Usually, for evaluating the EMP of bulk material layers one applies the exact numerical simulations of the wave reflection and transmission by the layer. Then one theoretically retrieves EMP from these data. The algorithm of this retrieval is called Nicholson-Ross-Weir (NRW) method. Recently, it was shown that the NRW method is suitable for theoretical or experimental characterization of material layers only in the case when the particles forming the material are not resonant [5, 6, 7, 8, 9, 10]. This is not the case of metamaterials, where the constitutive elements are resonant, and this method fails. More exactly, for metamaterials the NRW method leads to specific EMP which are obviously inconsistent with the definition of characterization.

Alternative method for obtaining EMP of bulk metamaterials which apparently leads to EMP compatible with the concept of CMP was suggested in 2007-2009 by C. Simovski [5, 6, 7]. It was also an indirect retrieval procedure, however it implied more involved calculations and even comprised the NRW retrieval algorithm as the first step. The method by Simovski is based on the representation of a composite slab as a 3-layer structure. The central layer is characterized by bulk material parameters ε and μ (which can be tensors). Two interface layers are dynamic analogues of Drude transition layers. They are characterized by their specific refraction index and characteristic impedance (neither ε nor μ). The method is in principle equally suitable as for theoretical characterization of bulk (thick enough) metamaterials as for their experimental characterization. However, it is applicable only to a special class of bulk regular metamaterials. These particular metamaterials were called Bloch lattices. Moreover, even for this class of metamaterials this method has been insufficiently developed. Namely, its application to obliquely incident waves has not yet been considered. Therefore, at present one has not proved that the components of ε and μ tensors tangential to the interface which were retrieved for the normal incidence of a plane wave are really applicable to other boundary problems, e.g. for the arbitrary plane-wave incidence. Also, it was not explained how to retrieve the normal (to the interface) components of these tensors if these components are also resonant. Finally, this method was never applied for the experimental characterization of metamaterials.

A different method was suggested by A. Scher and E. Kuester in 2009 [16, 21]. This method introduces EMP of a metamaterial using the quasi-static model, i.e. the Maxwell Garnett or Bruggeman formulas expressing the permittivity and permeability through individual electric and magnetic polarizabilities of constituent particles. However, individual polarizabilities of particles are calculated or retrieved beyond the quasi-static approximations through sheet electric and magnetic susceptibilities of a unit layer (monolayer) from the same particles with same interparticle distances. For this monolayer (metasurface) the reflection R and transmission T coefficients should be simulated exactly. Then the surface susceptibilities are retrieved through these R and T and the individual particle polarizabilities are found from these susceptibilities. The method leads to local material parameters of MTM, however its accuracy for practical metamaterials is doubtful. The static model of the bulk MTM is not consistent with their expectedly dynamic behavior. The results of this approach for arrays of Mie-resonant spheres strongly contradict to those of the dynamic approach suggested by Simovski and performed for the same array.

In our previous reports another theoretical retrieval method suggested independently in 2007-2008 by C. Tserkezis and by C. Smigaj and B. Gralyak was reviewed. This method led to EMP whose physical meaning was not clear. One introduced EMP of arbitrary lattices defined through the refraction index and the wave impedance of a given eigenwave (no matter if strong spatial dispersion exists in the lattice or not). The refraction index and the wave impedance are deduced from the theoretical study of the dispersion of an infinite lattice which is complemented by the simulations or measurements of S-parameters of the finite-thickness lattice (slab). This approach links the dispersion of the infinite lattice to R and T of the slab in a rather heuristic manner. However the retrieved EMP turn out to be violating the locality limitations and, therefore, are not consistent with the definition of CMP.

Other known methods for theoretical calculation of CMP of metamaterials which were reviewed in our previous reports do not refer to the retrieval of EMP from R and T coefficients of a metamaterial slab. In the best case they refer to the theoretical calculations of CMP i.e. to the homogenization of bulk MTM layers and contain the link to the boundary problem. This is, for example, the method by M. Silveirinha. Theoretically obtained EMP fit our concept of CMP since the information on obliquely propagating waves is included and they can be used for solving boundary problems. The role of the experiment is, however, not an indirect measurement of ε and μ but the validation of the theory which predicts such ε and μ together with other crucial characteristics (namely additional boundary conditions). The use of additional boundary conditions in this method makes the link between the measurements of R and T coefficients and calculation of ε and μ parameters very difficult. Briefly, this theoretical method is hardly suitable for the experimental characterization. No experimental results which would confirm this method for nanostructured materials are available. Other methods (by D. Felbacq et al., by G. Shvets and A. Urzhumov, by J.B. Pendry et al. [11, 12, 13, 14, 15, 25, 18, 19, 20]) refer only to eigenwaves in the infinite lattice and contain no link to the boundary problem. Therefore the EMP calculated using these methods cannot be considered as CMP.

To sum up, the conclusion was done in our previous report as well as in our booklet [22] that the theory of bulk metamaterials is developed not enough to recommend any procedure of theoretical (analytical or numerical) calculation of EMP for theoretical and practical electromagnetic characterization of nanostructured metamaterials. This conclusion was rather pessimistic because it stated the very weak development of the microscopic theory of bulk nanostructured metamaterials. Therefore even prerequisites for a correct and reliable procedure of electromagnetic characterization of any nanostructured metamaterial were absent.

1.1.2 How the situation has changed during last year

During the last period the situation has being drastically changing. A new trend in the literature devoted to the bulk material parameters of metamaterials can be easily noticed if we inspect papers published in 2010. This trend has following features:

- Scientific groups which have gained extremely high reputation over the world by their theoretical and experimental works (Profs. G. Shvets, Yu. Kivshar, F. Lederer, C. Holloway, E. Kuester and some others) have suggested new homogenization models especially addressed to nanostructured metamaterials [25, 26, 10, 27, 28, 29, 30, 31];

- In these papers the theoretical models have been linked to possible experiments, in other words, algorithms of possible experimental retrieval of EMP have been suggested.

- Special attention has been paid to the applicability of the theoretically retrieved material parameters to other cases of wave propagation. In other words, authors concentrate on those EMP which can be called characteristic parameters, their goal is now the electromagnetic characterization. This is probably the most important feature of the aforementioned papers.

In some recent papers (e.g. [26]) results which had been previously obtained in the field of electromagnetic characterization of nanostructured metamaterials have been criticized independently on our criticism from our previous reports. It has been shown that the previously retrieved material parameters of nanostructured metamaterials with claimed magnetic properties are not applicable for other cases of the wave propagation but only for the case for which they were retrieved and their physical meaning is not clear. Further, in works by other independent researchers [10, 27, 21, 28] the physical effect which makes the NRW method not applicable to metamaterials (which had been earlier pointed only in works by Simovski and Tretyakov) has been also noticed and widely discussed. This effect is the jump of tangential components of bulk macroscopic fields at the interface of the resonant material. This jump leads to the significant difference between the surface impedance of the material and its wave impedance[1].

Some researchers in this situation suggest novel algorithms of the retrieval of CMP which are not related to the simulation or measurement of R and T coefficients. For example, methods suggested in works [25, 28, 29] refer to the measurements or simulations of the fields inside the metamaterial sample. For example, the wave propagation retrieval method [28, 29] seems to be perfect for the theoretical calculation of CMP of metamaterials, even chiral ones [29], which require an additional tensor material parameter for the description of their bulk electromagnetic properties. However, the problem appears with the applicability of these CMP to practical boundary problems since the properties of the surface are not taken into account by this method. In fact, the method delivers only CMP of an infinite lattice, i.e. what was done in first works by Simovski and Tretyakov [5, 6]. The set of characteristic parameters retrieved in this way is therefore not complete. The method in its present form is hardly appropriate for the experimental characterization of nanostructured metamaterials. It implies the measurements of the amplitudes of Cartesian components of the electric field provided the metamaterial is a very thick layered structure with gaps between layers into which the probe can be introduced. So thick nanostructured metamaterials are not yet known, and such gaps would be challenging for fabrication. A similar drawback can be noticed if we consider the method suggested in [25]. This method also introduces a quite unusual description of any metamaterial, even a resonant artificial dielectric through additional tensor material parameters, whose physical meaning is

[1] The surface impedance and refraction index are retrieved by the NRW method and used for finding bulk material parameters.

not fully clear. However, these works are important as the illustration of the constructive trend in the literature and hopefully these new methods will be further developed by their authors.

We have to notice that in recent works [21, 30] the model of the dynamic homogenization of the infinite lattice of electric and magnetic dipoles suggested (to our knowledge) in work by Simovski [32] and applied for the theoretical electromagnetic characterization of metamaterials in his further works [5, 6, 7, 8] has been strongly developed. Namely, the model was expanded from lattices in which the near-field interaction between adjacent crystal planes is negligible (so-called Bloch lattices) to lattices in which this interaction is significant. In the report [31] A. Alu suggested a modification of the model [32] which allows one to fully remove the small deviation of the lattice material parameters from locality.This deviation occurs in the model [32] in a very narrow frequency range in lossless arrays near the lower edge of the resonance band. In works [5, 6, 7] this effect was practically eliminated by introduction of small losses. Though recent works [**?**, 21, 30, 31] do not consider surface effects and the retrieved EMP are not enough to solve boundary probelms, they evidence that the theorists have started to understand the importance of the dynamic interparticle interaction for the proper characterization of metamaterials.

Finally, in work [10] the algorithm previously suggested by Simovski which takes into account both dynamic interaction effects and surface effects has been improved. Instead of transition layers (suggested 100 years ago by Drude and revisited by Simovski) the authors suggested their compressed version – sheets of effective electric and magnetic currents which should be introduced at the interfaces of the metamaterial layer. This approach which seems (for us) to be the most promising one needs further development compared to [10]. In this paper authors could not satisfy the locality limitations in their retrieval procedure. The possible reason of this result is probably the small thickness of the sample for which the surface current sheets interact by near fields and their susceptibilities become mesoscopic. Also, in [10] there is no link to the problem of the oblique incidence which should show that the retrieved EMP are really CMP of this metamaterial.

However, we consider the apparition of all these cited works within the period under reporting as a key prerequisite of the real breakthrough in the electromagnetic characterization of bulk metamaterials we expect in next future. We hope that the ECONAM activity has played some role in this positive trend.

1.2 Surface nanostructured metamaterials

1.2.1 State-of-the-art from our previous report

Surface nanostructured metamaterials (metasurfaces) following to our previous reports is an important class of surface nanostructured materials. The most part of known nanostructured materials are artificial surfaces. Among them metasurfaces are most promising for applications. However, their electromagnetic characterization was almost ignored in the previous literature. We stated in the precedent report that many researchers characterized metasurfaces as if they where bulk materials. Some researchers simply treated metasurfaces as layers of bulk materials. The others retrieved their EMP as the result of the approximate fitting to the results of measurements or simulations of S-parameters of a layer. For example, one wrote Lorentz's dispersion laws for permittivity and permeability through unknown dimensionless parameters and tried to find them through such fitting. Only in works [23, 24] it had been properly noticed that metasurfaces or metafilms form a special and very important class of metamaterials which requires the special characterization approach. Effective material parameters for grids of small resonant inclusions were introduced in these works. Metasurfaces considered in these works contain one resonant inclusion across the layer which possess simultaneously electric and magnetic dipole moments whose resonances can overlap over the frequency axis. Material parameters of metasurfaces correspond to their replacement by effective current sheets and describe the interaction of metasurfaces with tangentially averaged electric and magnetic fields. In general, these material parameters are called electric and magnetic tangential and normal surface susceptibilities. All these susceptibilities were defined through steps of tangential and normal components of so-called transversally averaged electric and magnetic fields across the physical metasurface thickness (which was assumed to be very optically small). Averaged fields were obtained by surface averaging of microscopic fields – integration over periods of the grid. This integration is performed at two sides of the metasurface and the averaged value of these two integrals is the transversally averaged field. This characterization approach was developed in these works only for monolayers of solid magneto-electric particles isotropic in the tangential plane. Then the condensed description of the metasurface is given by four scalar parameters. For two specific angles of incidence (e.g., 0 and 45°) one simulates R and T coefficients and from these four complex parameters one extracts four complex material parameters. These works presented the only (to our knowledge) fully physical and constructive approach to the characterization of some metasurfaces available from the literature.

In spite of works [23, 24], our conclusion on the state-of-the-art in this field from the precedent report was still quite pessimistic. Really, the theory was presented in the form suitable only for a monolayer of solid particles. Metasurfaces like optical fishnets and metasurfaces whose constitutive elements are not dipole scatterers cannot be described by this model. Physically sound results were obtained only for a non-realistic case of very optically small spheres with very large internal permittivity and permeability. And, what is most important, the model implies the absence of any substrate. Particles forming the grid should be located in a homogeneous matrix whose interfaces are well distanced from the grid. Such metasurfaces are difficult to implement.

1.2.2 What has changed since that time

A progress in this direction is modest, and, to our knowledge, it is related only to the study by Simovski and Morits performed on summer 2010 [33]. Using an example of a bilayer of plasmonic nanospheres we generalized the method of electromagnetic characterization of monolayer magneto-dielectric metasurfaces (metafilms) suggested in [23, 24]. We theoretically demonstrated that the results of this characterization method are suitable to predict scattering parameters of bilayer metasurfaces. We expanded the model [23, 24] to realistic nanostructured metasurfaces. Since we considered different angles of incidence and theoretically demonstrated that the retrieved material parameters are applicable for all of them, we have done a longer leap in the characterization of metasurfaces than we have done since 2007 in the characterization of bulk nanostructured metamaterials.

However, an important theoretical problem is not yet resolved. The approach [23, 24] does not allow one to take into account the dielectric substrate. The theory is applicable only to grids of particles well distanced from all interfaces. Namely, the minimal distance should be larger than the grid period. To generalize the approach to the realistic case when the particles are located at the dielectric interface is not an easy task. However, there is a strong need in such a theory and we expect in the next future a breakthrough also in this question.

2 Conclusions

In this report we have presented an update to our overview of recent constructive works devoted to the theoretical electromagnetic characterization of nanostructured metamaterials. These works have appeared in the corresponding literature, hopefully, not without the influence of the efforts of ECONAM partners (and the project officer [22]) in the dissemination of the scientific knowledge of our experts. The analysis of this literature allows us to conclude that the vector which shows the accumulated knowledge changed the direction. From the depression, i.e. accumulation of waste data obtained with the use of an obsolete and inadequate homogenization model it turned to the development, i.e to the search of adequate, really working theoretical models. This new trend allows us to expect the approaching breakthrough in the electromagnetic characterization of nanostructured metamaterials.

References

[1] FP7 contract No. 218696-ECONAM, Deliverable 1.1.4, State-of-the-art and most promising analytical and numerical characterization techniques for nanostrcutured metamaterials, Due date $T0 + 24$, March 2010

[2] FP7 contract No. 218696-ECONAM, Deliverable 1.1.3, State-of-the-art and most promising analytical and numerical characterization techniques for nanostrcutured metamaterials, Due date $T0 + 18$, October 2009

[3] FP7 contract No. 218696-ECONAM, Deliverable 1.1.3, State-of-the-art and most promising analytical and numerical characterization techniques for nanostrcutured metamaterials, Due date $T0 + 15$, June 2009

[4] FP7 contract No. 218696-ECONAM, Deliverable 1.1.3, State-of-the-art and most promising analytical and numerical characterization techniques for nanostrcutured metamaterials, Due date $T0 + 9$, February 2009

[5] C.R. Simovski, S.A. Tretyakov, *Phys. Rev. B* **75** (2007) 195111

[6] C.R. Simovski, *Metamaterials* **1** (2007) 62

[7] C.R. Simovski, *Metamaterials* **2** (2008) 169

[8] C.R. Simovski, *Optics and Spectroscopy* **107** (2008) 726

[9] Kh. Chalapat, K. Sarvala, J. Li, G.S. Paraoanu, *IEEE Transactions on Microwave Theory and Techniques* **57** (2009) 2257

[10] S. Kim, E. F. Kuester, C. L. Holloway, A.D. Scher, J. Baker-Jarvis, Boundary effects on the determination of metamaterial parameters from normal incidence reflection and transmission measurements, submitted to *IEEE Trans. Antennas Propag.*, available at http://arxiv.org/abs/1009.5927v2

[11] G. Bouchitte, C. Bourel and D. Felbacq, *Comptes Rendus, Acad. Sci. Paris*, Ser. I 347 (2009) 571.

[12] A. I. Cabuz, D. Felbacq, and D. Cassagne, *Phys. Rev. Lett.* 98 (2007) 037403.

[13] D. Felbacq, G. Bouchitte, *Opt. Lett.* 30 (2005) 10.

[14] D. Felbacq, G. Bouchitte, *Phys. Rev. Lett.* 94 (2005) 183902.

[15] G. Bouchitte, D. Felbacq, *Comptes Rendus, Acad. Sci. Paris*, Ser. I 339 (2004) 337.

[16] A.D. Scher, E.F. Kuester, *Metamaterials* **3** (2009) 44

[17] Ya. A. Urzhumov and G. Shvets, *Solid State Communications* 146 (2008) 208

[18] J.B. Pendry, A.J. Holden, D.J. Robins, and W.J. Stewart, *J. Phys. Condens. Matter*, **10** (1998) 4785

[19] J.B. Pendry, A.J. Holden, D.J. Robins, and W.J. Stewart, *IEEE Trans. MTT*, **47** (1999) 207

[20] D. Smith and J. B. Pendry, *JOSA B*, **23** (2006) 391

[21] A.D. Scher, E.F. Kuester, *Progress In Electromagnetics Research B* **14** (2009) 341

[22] *Nanostructured Metamaterials: Exchange between experts in electromagnetics and material science*, A. de Baas, Editor, European Commission, Brussels, 2010

[23] E.F. Kuester, M.A. Mohamed, M. Piket-May, C.L. Holloway, *IEEE Trans. Antennas Propag.* **51** (2003) 2641

[24] C.L. Holloway, A. Dienstfrey, E.F. Kuester, J.F. O'Hara, A.A. Azad, A.J. Taylor, *Metamaterials* **3** (2009) 100

[25] C. Fietz, G. Shvets, Physica B **405**(2010) 2930

[26] C. Menzel, T. Paul, C. Rockstuhl, T. Pertsch, S. Tretyakov, F. Lederer, Physical Review B **81** (2010), 035320

[27] D.A. Powell, Yu.S. Kivshar, *Applied Physics Letters* **97** (2010) 091106

[28] A. Andryieuski, R. Malureanu, A.V. Lavrinenko, *Phys. Rev. B* **80** (2009) 193101

[29] A. Andryieuski, R. Malureanu, A.V. Lavrinenko, *Optics Express* **18** (2010) 15498

[30] R.A. Shore, R.D. Yaghjian, *IEICE Transactions on Communications* **E91.B** (2010) 1819

[31] A. Al, Abstracts of Metamaterials'2010, Karlsruhe, Germany, September 16-19, 2010, (invited talk)

[32] C.R. Simovski, S. He, *Physics Letters A* **311** (2003) 254

[33] D. Morits, C. Simovski, *Physical Review B* **82** (2010) 165114