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

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1 Introduction: Characteristic material parameters versus effective material parameters

Characterization in material science refers to the use of external techniques to probe into the internal material structure (geometric characterization) and material properties of a material such as elemental content, chemical properties, electric conductivity, static permeability and hysteresis, mechanical, thermal properties, etc. Results of characterization of a material should not depend on the sample shape and measuring setup.

Now let us introduce the concept of characteristic material parameters (CMP) which follows from the definition of characterization. CMP are not parameters specific for a given wave process in a given sample excited by a given source. Electromagnetic CMP should be applicable to different linear wave processes and different samples of the same material. In a maximalist approach, the term CMP can be granted only to those characteristic parameters which do not depend on the sample shape and size and on the electromagnetic field distribution in it, i.e. give the full condensed description of linear electromagnetic properties of the material from which the sample is prepared.

Retrieval of electromagnetic effective parameters (EMP) from measured data (also called post-processing) is necessary for experimental electromagnetic characterization since no any electromagnetic characteristic parameters of a material can be measured directly. For example, in the classical scheme of linear electromagnetic characterization of a bulk magneto-dielectric material it is assumed that the material may be characterized by effective material parameters ε and μ and usual Maxwell boundary conditions are satisfied at interfaces.

In practice if the same set of effective EMP is applicable to describe the interaction of the sample with at least a number of plane waves propagating in different directions, these EMP deserve to be referred to characteristic parameters even if they are not applicable for evanescent waves. To find complete CMP which would give a fully condensed description of MTM suitable for the whole infinite spatial spectrum of electromagnetic field is not a realistic task, especially for nanostructured MTM. We think that one can use one set of CMP applicable for propagating waves and another set of CMP for a most important spatial spectrum of evanescent waves. What is important: the definition of the electromagnetic characterization clearly forbids us to refer effective material parameters that are applicable for an only case of the wave propagation to characteristic parameters of the material.

2 Classification of nanostructured materials

In the literature and on the Internet there are many definitions of metamaterials, which usually stress their unusual electromagnetic properties. Perhaps in the most generic way, metamaterial (MTM) can be defined as an arrangement of artificial structural elements, designed to achieve advantageous and unusual electromagnetic properties. The concept of material implies homogeneity, i.e. the distance between elements should be small enough. If a metamaterial is a periodical structure, the lattice constants should be considerably smaller than the wavelength in the medium. This distinguishes metamaterials from photonic (electromagnetic) crystals and usual frequency-selective surfaces, whose useful and unusual electromagnetic properties originate mainly from the periodicity of their structure. In contrast to photonic crystals, metamaterials possess such properties due to specific electromagnetic response of their "artificial molecules" and not due to specific distances between them. Furthermore, the electromagnetic properties of "molecules" are determined not only by their chemical composition, but mostly by their geometrical shape. The chemical composition is usually chosen so that the response is high (conductive material, high permittivity material, ferromagnetic material) and losses are minimized. Specific engineered properties are designed primarily by choosing the inclusion shape.

In this definition, metamaterials are characterized by effective materials parameters, like permittivity, permeability, chirality parameter. Sometimes also such structures as photonic crystals or frequency selective surfaces are also called metamaterials. In these cases, the description of electromagnetic response is based on other concepts, such as isofrequency plots or grid impedance. Furthermore, the term "metamaterial" is used in the literature also in connection with artificial materials designed to exhibit advantageous and unusual properties for waves of other nature, for example there are works on phononic metamaterials designed to control sound waves. Sometimes waves of other nature have electromagnetic components (sound waves in piezoelectrics or spin waves in nanostructured magnetic materials), so there is no solid boundary between electromagnetic and "non-electromagnetic" materials. Analogous classification can be possibly suggested for non-electromagnetic metamaterials. Classification of spin-wave metamaterials is given below as a separate chapter.

Table 1. Nanostructures classified by their linear electromagnetic properties (q is the wave number in the structure and a is the size of the lattice unit cell.

Nanostructures (NS)	Optically dense $(qa) < 1$	Optically sparse $(qa) > 1$	Dense in one direction, in other direction(s) either sparse or with extended inclusions
3D bulk materials	Bulk MTM with small inclusions, bulk NSM without MTM properties	Photonic crystals, quasi-crystals, sparse random composites	Wire media, multilayer plasmonic structures (fishnets, solid metal-dielectric nanolayers)
2D, sheet materials	Metasurfaces (metafilms), nanostructured sheets without MTM properties	Plasmonic diffraction grids, optical bandgap and frequency selective surfaces	Artificial nanostructured surfaces with long inclusions or slots
1D, lines	Metawaveguides	Not yet known but possible	Impossible

Inspecting different types of existing nanostructured materials [1]– [7] and prospective nanostructured materials (NSM) which are under discussion in the current literature we can find many types of NSM which satisfy the above definition of MTM. To show the place of nanostructured MTM among all NSM it is instructive to suggest a classification of NSM. Possible classification is presented in a form of a table and is related to the characterization of linear electromagnetic properties of NSM. Different types of NSM should be characterized by different sets of electromagnetic parameters. This classification takes into account the internal geometry of NSM and most important linear electromagnetic properties of constituents and effective material formed by them. The left upper cell has rich content which is detailed as a chart in Fig. 1. The most important criterion of the classification is the dimension of the array of constituents which form the nanostructured material. 3D or bulk materials correspond to structures with a large number of constitutive elements in the array along any direction. 2D or surface materials

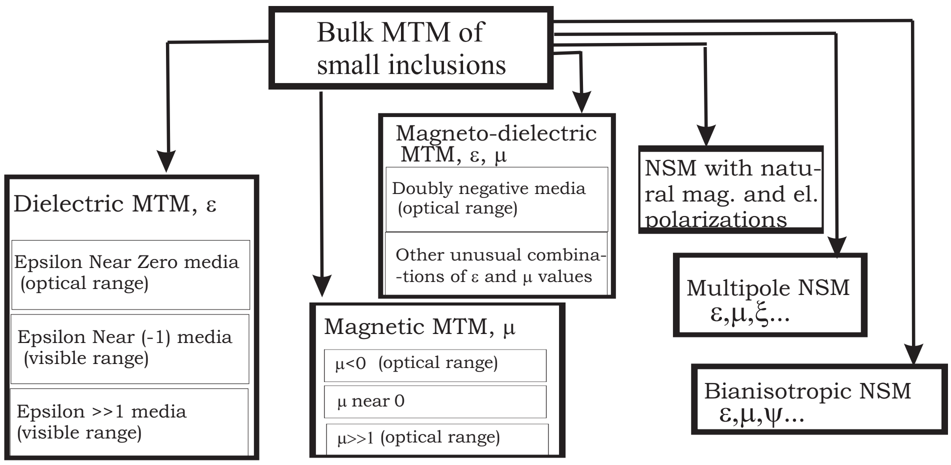

Figure 1: Classification of bulk nanostructured MTM of small inclusions. Optical range in this scheme by definition covers IR, visible and near UV ranges.

correspond to the case when the artificial material is a layer including only 1-3 constitutive elements across it.

MTM of the surface type were called metasurfaces in [8, 9] and metafilms in papers [10, 11]. Notice that in these papers the consistent method of the electromagnetic characterization of metasurfaces (metafilms) comprising obe inclusion across the sheet was suggested.

Linear (directional) structures with nanosized inclusions are optical waveguides with nanoinclusions, plasmonic and polaritonic nanochains. They are called in this table metawaveguides, but are not referred to MTM since the concept of material is usually not applied to 1D structures.

The second criterion of the classification is the optical size of the unit cell in the visible and near-IR frequency range where known NSM operate (possesses useful and unusual electromagnetic properties). Large optical size practically implies $(qa) > 1$. In this range the structure cannot be characterized by local material parameters (neither bulk nor surface ones). In the opposite case $(qa) < 1$ the structure in principle can be characterized by local parameters and can be referred to MTM. The same structures at different frequencies can be referred either to optically dense or to optically sparse types of the structure.

The third criterion is the presence or absence of properties which allow us to refer or not to refer an optically dense material to MTM. Usually though not always these useful unusual properties are granted by the resonance of inclusions.

Let us give some comments on Table 1. Nanostructured composites can be designed for whatever other purpose and not possess any interesting or unusual electromagnetic properties. Instead they can possess useful thermal and mechanical properties. They can have specially engineered electric conductivity. The combinations of these properties are used for example in thermoelectric elements. Such materials are not referred to MTM in this table. Optically sparse nanostructures are not referred to MTM since they do not fit the concept of material.

Optically dense surface nanostructures are, for example, plasmonic island nanofilms and chemically roughened metal surfaces. The constituents are metal islands or random nanocorrugations. They are used mainly in sensing applications: molecular clusters and even separate molecules utilizing the so-called surface-enhanced Raman scattering (SERS) scheme. This scheme is based on the effect of the local field enhancement in the vicinity of such a surface. The property of plasmon resonance which is responsible for this effect is definitely useful, however, it is rather usual from the electromagnetics point of view. Huge literature has been devoted to such structures since the discovery of SERS in 1970s. On random plasmonic surfaces the local field enhancement is the same as for a single plasmonic nanoparticle and is observed only at points on top of the particle (or at the edges of a nanoisland). Plasmon resonance in metal particles has been known for a long time. Therefore (this our point fits the content of books [1]–[9]) we do not refer these structures to MTM. However, if the nanostructured plasmonic surface is regular, it possesses an unusual property: the strong enhancement of the averaged field in such structures. Such artificial surfaces probably refer to metasurfaces. Nanostructured surfaces of vertical (aligned) metal nanorods grown on the metal substrate enhance at the plasmon resonance the field at the plane corresponding to the upper edges of the rods [12]. Such structures should be also referred to metasurfaces. In general, nanostructured metasurfaces are self-resonant grids possessing certain regularity either in the arrangement of resonant elements or in their orientation. Such grids can comprise separate plasmonic scatterers on a dielectric substrate or plasmonic corrugations. They also can be performed as solid metal screens of nanometer thickness with slots or holes. For example, metal nanolayers with subwavelength slots support surface plasmon polariton waves whose dispersion is determined by these slots (see e.g. in Chapters 25 and 26 of [9]). Another important example of a metasurface is a monolayer optical fishnet (see e.g. in Chapter 29 of [9]). This monolayer is formed by a pair of parallel silver or gold periodically slotted nanolayers with nanogap between them. The slots in fishnet structures are non-circular and are rather optically large.

Abundant literature is devoted to nanostructured waveguides (metawaveguides in our classification). Their useful and unusual property is the subwavelength channel for the guided wave observed in plasmonic nanochains (see e.g. in [13]) and nanostructured fibers with plasmonic insertions (see e.g. in [14]). They also can be used for frequency filtering of optical signals on the nanoscale level (e.g. in [15]).

Multilayer optical fishnets and multilayer metal-dielectric nanostructures (see e.g. [16]–[21]) are important types of MTM. Multilayer optical fishnets demonstrate not only the negative phase shift of the wave across the structure (i.e. backward wave propagation), in them one obtained negative group velocity (i.e. inverse direction of the pulse peak velocity with respect to the energy transport direction). In multilayer nanostructures of alternating continuous metal and dielectric nanolayers one experimentally demonstrated the subwavelength optical imaging in the far zone of the optical object (e.g. [22]). Note that known fishnets do not offer subwavelength imaging in the backward-wave frequency range, while stacks of continuous layers do not support backward waves, so none of them can make a Pendry perfect lens. These structures are referred to MTM in our table since the thickness of their layers is much smaller than the wavelength. However, they possess strong spatial dispersion for waves propagating obliquely to the structure or along it because the constituents are extended and the tangential periods (in fishnet structures) are large.

For another type of such MTM - wire media - the effect of spatial dispersion is crucial for their unusual properties (see e.g. in Chapter 15 of [8]). Wire media in the optical range are optically dense arrays of optically long metal nanowires or carbon nanotubes. Wire media can be grown on the substrate or prepared by embedding nanowires into porous glass or porous plastic fiber matrix. If nanowires are grown on a substrate, they can be optically short and then refer to metasurfaces (see above). Arrays of aligned nanorods and nanotubes should be referred to nanowire media if the optical length of cylinders is enough. Nanostructured wire media are spatially dispersive in the infrared range [24]. In the visible range optically dense lattices of parallel metal nanowires become a uniaxial dielectric medium without strong spatial dispersion [23].

Below we give a brief comment on Fig. 1. Bulk MTM are classified judging upon their material parameters. Optically dense nanocomposites can be artificial dielectric media with unusual permittivity, i.e. close to 0 or to (-1) or very high in the visible range where usual materials (except some liquid crystals existing only in the form of thin films) have rather low permittivity. Artificial magnetic media, possessing negative permeability in the optical range are also MTM. Not only media with negative deserve to be referred to MTM, media with =0 or =2 in the optical range are also MTM. Media with both negative permittivity and permeability represent the most known type of MTM, however if e=0 and m=2 in the visible range such NSM should be also referred to magneto-dielectric MTM. Optically dense nanocomposites with plasmonic inclusions refer to MTM for both cases of regular and random arrangements. Random nanocomposites possess scattering losses whereas regular plasmonic composites do not. Therefore regular plasmonic composites are more promising. They can be also resonant multipole media and resonant bianisotriopic media formed by nanoparticles. In both these cases the number of material parameters describing such media is larger than two. The difference between magneto-dielectric nanostructured media, multipole and bianisotropic media is discussed in Chaper 2 of [8].

The correct classification of nanostructured MTM is helpful to avoid the most dangerous pitfall in their electromagnetic characterization: the inconsistent classification of the material under study. In the most part of known works describing metasurfaces, i.e. MTM layers with only 1-2 resonant inclusions over the layer thickness, authors of [16]–[18], [24]–[32] and other similar works treat these metasurfaces as bulk media. Multipole and bianisotropic bulk media were described as simple magneto-dielectrics (ε and μ), for example in works [**?**]–[45].

3 Electromagnetic characterization of bulk metamaterials

The procedures of the theoretical and experimental electromagnetic characterization of bulk layers of metamaterials with a regular inner structure were analyzed in our previous surveys. The list of these procedures is presented below. We do not give here bibliographic references since in our previous reports the detailed review of these methods was presented.

- Methods used in the literature for the characterization of MTM layers:
  1. The Nicolson-Ross-Weir (NRW) method is suitable for both theoretical (exact numerical simulations of S-parameters) and experimental characterization (measured complex S-parameters) of material layers. For MTM the NRW method leads to specific EMP which are inconsistent with the definition of characterization.
  2. The method suggested by C. Simovski is the indirect retrieval procedure including the NRW method as the first step. The method includes the representation of a composite slab as a 3-layer structure. The central layer is characterized by bulk material parameters ε and μ (perhaps, tensors). Two interface layers (dynamic analogues of Drude transition layers) are characterized by transmission line characteristic parameters (refraction index and characteristic impedance). The method is

in principle suitable for both theoretical and experimental characterization of a special class of bulk MTM which were called Bloch lattices. However this method has been insufficiently developed and never applied for experimental characterization.

3. The method of M. Silveirinha is based on the special averaging procedure for fields and polarizations and specially derived additional boundary conditions. This method is suitable only for theoretical characterization (i.e. homogenization) of bulk MTM layers.
4. The method of C. Tserkezis, C. Smigaj and B. Gralyak introduces EMP through refraction index and the wave impedance of a given eigenwave (no matter does the strong spatial dispersion exist in the material or not). The refraction index and the wave impedance are retrieved from theoretical study of the infinite lattice dispersion complemented by the simulations or measurements of S-parameters of the layer. This approach for MTM layers leads to non-local retrieved material parameters which are not consistent with the definition of characteristic parameters.
5. The method by T. Driscoll et al. retrieves EMP as the result of the approximate fitting of unknown parameters in Lorentz's dispersion laws for permittivity and permeability to the results of measurements or simulations of S-parameters of a layer. The method was applied to both surface and bulk metamaterials. In the first case it fails due to the inconsistent classification of the material, in the second case it fails because the problem of boundary conditions beyond the quasi-static limit is ignored.
6. The method of A. Scher and E. Kuester introduces EMP of a metamaterial using the quasi-static model, i.e. 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). These susceptibilities are retrieved through S-parameters of the monolayer. The method leads to local material parameters of MTM, however its accuracy and practical applicability are not yet clear. The static theoretical characterization of the bulk MTM is not fully consistent with the dynamic retrieval procedure of a specially prepared monolayer of MTM.

- Methods suitable only for theoretical characterization (homogenization) of unbounded regular MTM:

1. The method of D. Felbacq et al. is based on unusual operator definitions of the effective permittivity and permeability and on the exact solution of the cell problem for an infinite lattice.
2. The method of G. Shvets and A. Urzhumov is suitable for unbounded metamaterial lattices with densely packed plasmonic constituents. The bulk EMP in this method are calculated semi-analytically taking into account the typical features of the polarization current distribution in such structures. The method also ignores the problem of boundary conditions and is suitable only for homogenization of unbounded regular MTM.
3. The method of J. B. Pendry defines EMP very unusually through different averaging procedures for different microscopic field vectors $\mathbf{E}$, $\mathbf{H}$ and $\mathbf{B}$, $\mathbf{D}$.

From this list it is clear that known methods of the electromagnetic characterization of finite-thickness metamaterials either have been developed not sufficiently or are not consistent with the definition of the characterization i.e. retrieved EMP are not CMP.

4 Conclusions

The retrieval of effective material parameters with the purpose of material characterization implies, obviously, that the these parameters

- have clear physical meaning which is adequate to the structure they should describe;
- give a condensed description of the structure properties (for the electromagnetic characterization are applicable to a number of wave processes);
- are really material, not sample parameters (for bulk layers this means the independence on the layer thickness).
- demonstrate the agreement between the theory and experiment.

In the literature devoted to the electromagnetic characterization of nanostructured metamaterials all these expectations have not been yet satisfied simultaneously (to our knowledge). The reason of this unsatisfactory situation is the weak development of existing microscopic theories of metamaterials. Promising approaches which could be potentially recommended progress slowly since the efforts of their authors have not been supported. Instead, the classical approach to the material electromagnetic characterization (inapplicable for nanostructured metamaterials) has been used extensively for both bulk and sheet materials multiplying the mess in the literature and leading the metamaterial science to the impasse. The activity of our expert group tried to resist to this tendency and the present report is our modest contribution into this struggle.

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