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Electromagnetic Characterization Of NAnostructured Materials

PUBLIC CONSOLIDATED OVERVIEW OF THE STATE-OF-THE-ART AND MOST PROMISING ANALYTICAL AND NUMERICAL CHARACTERIZATION TECHNIQUES FOR NANOSTRUCTURED MATERIALS

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Abstract

This public overview summarizes the analysis of electromagnetic characterization techniques for nanostructured materials done by the consortium during the whole project duration. The text is written primarily **for a wide audience of non-specialists** and includes an introduction which explains the main concepts of complex electromagnetic materials. Experts in electromagnetic characterization of nanostructures can find more details in Deliverables D1.1.1-1.1.5 or use this document as a guide to original literature listed in the reference list at the end of this document.

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1 Introduction

1.1 Nanostructured electromagnetic materials and metamaterials

The design and manufacturing of nanostructured materials has been made possible thanks to progress made in nanotechnologies, material science and electrical engineering. With the help of advanced bottom-up manufacturing techniques, a large number of micrometer scale designs can now being scaled down to nanometer scale and therefore completely new optical materials can be created and investigated. Majority of new nanostructured materials for electromagnetic applications refers to so-called *metamaterials* which exhibit exceptional properties, defined by their geometrical nanostructures. 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 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 so-called photonic crystals which are discussed below and from so-called frequency-selective surfaces which are surface analogues of photonic crystals. These are periodic structures whose useful and unusual electromagnetic properties originate mainly from their internal periodicity. In contrast to photonic crystals and frequency selective surfaces, metamaterials possess their properties due to specific electromagnetic response of their "artificial molecules" and not due to specific distances between them (the importance has only the smallness of these distances). Furthermore, the electromagnetic properties of "molecules" are determined not only by their chemical composition, but their geometrical shape plays an important and in most cases the decisive role. The chemical composition is usually chosen so that the response of the inclusion to electromagnetic waves is high (conductive materials, high permittivity materials, ferromagnetic materials) and losses are reasonably small. Specific engineered properties are designed primarily by choosing the inclusions shape and their mutual arrangement.

The research and engineering tools needed to create metamaterials include electromagnetic modelling, geometry and property design, manufacturing, and structural and electron microscopy characterization at the nanoscale. The final and critical stage of design of new materials for radio, microwave, and optical applications is *electromagnetic characterization*, that is, measurement of electromagnetic properties of material samples.

This report is aimed to help the reader to learn more about the place of metamaterials among other nanostructures and especially about their electromagnetic characterization, which represents a fundamental problem of the modern electromagnetic science. Electromagnetic properties of nanostructured metamaterials are determined by the shape of the constituent nanoinclusions, by their concentration, by their geometric arrangement and by the material parameters of their constituents. Due to this highly increased complexity the electromagnetic characterization of metamaterials has become a branch of science in itself. Effective material parameters are often extracted from numerically simulated or experimentally measured reflection and transmission coefficients. Traditionally, these effective parameters are the basic phenomenological material parameters: permittivity and permeability. In the case of natural materials (and also for composite bulk media which cannot be referred to metamaterials but behave as effectively continuous structures) these parameters usually give a good description of the electromagnetic behavior. But metamaterials cannot always be characterized in this simple

and traditional way. Many metamaterials need more parameters for their characterization and, in general, the physical meaning and practical applicability of material parameters is to be carefully examined before these parameters can be used in design of various applications.

1.2 Electromagnetic waves in materials

From Maxwell's equations which are governing electromagnetic fields it follows that a plane electromagnetic wave incident on a half-space filled with a continuous material partially refracts into it and partially reflects. The refracted and reflected waves are also plane waves. The incidence of a plane wave on a half-space is one of the simplest boundary problems. The next grade of complexity for the boundary problem is the case of a plane wave incident on a planar layer. In this case the refracted wave experiences multiple internal reflections in the layer and partially penetrates behind the layer. The wave behind the layer is called *transmitted wave*, and it is also a plane wave as well as all partial waves corresponding to the inner reflection events.

A time-harmonic plane wave propagating along the z-axis with the wave number q is described as $A\exp(j\omega t - jqz)$, where A is the wave phasor (complex amplitude) which can refer to any component of the field vectors $\mathbf{E}, \mathbf{H}, \mathbf{D}$ or $\mathbf{B}$, ω is the frequency, and $j = \sqrt{-1}$. The real time-dependent field which is physically observable is calculated as

$$E(t,z) = \mathrm{Re}[A\exp(j\omega t - jqz)] = |A|\cos[\omega t - qz + \arg(A)]. \tag{1}$$

Since the factor $\exp(j\omega t)$ is common for all field vectors it is usually omitted. The time dependence $\exp(j\omega t)$ is usually adopted by electrical engineers, while physicists usually prefer the choice of $\exp(-i\omega t)$ (where the imaginary unit is denoted as i). Then the physically observable field of the plane wave is calculated as $\mathrm{Re}[A\exp(-i\omega t + iqz)]$. If a wave propagates obliquely with respect to the coordinate axis, one presents the plane wave through the wave vector $\mathbf{q}$. Then for a point with the radius-vector $\mathbf{r}$ one has $A\exp(-j\mathbf{q}\cdot\mathbf{r})$ in the "engineering notation" and $A\exp(i\mathbf{q}\cdot\mathbf{r})$ in the "physical notation".

Plane waves which can exist in a given medium in the absence of sources are called the medium eigenmodes. The concept of eigenmodes is also valid for periodical structures (lattices) as it is explained below. In the general case the time-harmonic wave field created by a given set of sources can be presented as a combination of plane waves (eigenmodes). This combination (which is rarely discrete, more often it is continuous) is called the spatial spectrum. An electromagnetic pulse can be presented through time harmonics as its frequency spectrum. So, in the general case the time-varying electromagnetic field is the double spectrum comprising both frequency spectrum and the spatial spectrum – spectrum of eigenmodes.

1.3 Why do we need effective material parameters?

Introduction of effective electromagnetic parameters of a material is called *homogenization*. In this approach a heterogeneous structure is replaced by an effectively homogeneous one. Any homogenization theory is an approximation, because it employs a finite number of parameters to describe samples with huge numbers of molecules (for artificial materials – inclusions). In the case of metamaterials the response of these inclusions is dispersive, i.e., the inclusions respond to a pulse excitation with some time delay. In terms of effective parameters for time-harmonic fields, this means that the parameters (permittivity, permeability,...) significantly depend on

the frequency of excitation. To solve exactly the electrodynamic problem for an array comprising a huge number of inclusions would require huge computational and time resources even with the use of supercomputers, still not giving any hints on what will happen if we change the sample shape, for instance. Long computation time strongly limits the opportunity to analyze the electromagnetic phenomena that occur in the material and moreover to understand its electromagnetic properties. To design a new electromagnetic material, the researcher should find the optimal shape and chemical content of inclusions, their optimal size and mutual arrangement, the optimal distance between them, etc. This means that many realizations of the material should be considered before the optimal design is obtained. If this process is based on the exact numerical simulations the optimal design becomes a hopeless task. The homogenization allows one to avoid the exact simulation of each realization, each size and each shape of material samples.

Homogenization means that material parameters of an artificial material are obtained and constitutive relations in which these parameters enter are known. Then one can analytically solve macroscopic Maxwell's equations complemented these constitutive relations. This solution for bulk materials is usually done assuming the infinite space filled with the material in the absence of sources. This way one determines all the eigenwaves that can exist in the infinite artificial medium. By enforcing the appropriate boundary conditions, the amplitudes of these eigenmodes are found for a finite-size sample of the medium. For nanostructured materials in many practically relevant situations samples are parallel-plate layers. If a nanostructured layer can be considered as a layer of an effectively bulk medium, the field inside it can be expressed as a superposition of the medium eigenmodes. The amplitudes of the eigenmodes used to decompose the scattered fields (usually reflected and transmitted plane waves) can be obtained, and a solution to the entire electromagnetic problem can be found. However, the approach based on the bulk material eigenmodes cannot be applied to artificial "surface materials" (or *sheets*) where the number of inclusions across the layer is small, which is especially true for a monolayer (a single grid of nanoinclusions on a dielectric substrate). For such structures one needs to introduce special material parameters relevant for an "artificial surface" (which is called metasurface if the constitutive elements possess resonant response). However, in any case the knowledge of material parameters is an obvious pre-requisite for solving the electrodynamic boundary problem as well as for understanding how the new materials will perform in various device applications.

2 Some important concepts of the electromagnetics of materials

In order to understand the modern approaches to electromagnetic characterization of nanostructured materials, we need to know some basic concepts of electromagnetics of materials, discussed next.

2.1 Continuous and effectively continuous magneto-dielectric materials

The simplest type of isotropic homogeneous media is the vacuum. The constitutive relations in the vacuum are expressed by the basic linear relations

$$\mathbf{D}(r,t) = \varepsilon_0 \mathbf{E}(r,t), \quad \mathbf{B}(r,t) = \mu_0 \mathbf{H}(r,t), \tag{2}$$

where the universal physical constants $\varepsilon_0 = 8.85418782 \cdot 10^{-12}$ F/m and $\mu_0 = 4 \cdot 10^{-7}$ H/m are called the free space permittivity and permeability, respectively.

Vast majority of natural substances are electrically neutral at the macroscopic level. Even at the scale of one nanometer the electric charges in them are not separated. Only permanent magnets and electrets are known as exceptions. Macroscopic electrical neutrality of materials is the consequence of an internal equilibrium in collective interactions of microscopic charges. In good insulators, positive and negative charges in atoms are tightly tied together and immense external energy is necessary to break the bonds. However, when the electromagnetic wave impinges on the sample, external electric and magnetic fields cause oscillatory displacements of charges from their original positions. Effects of local separation of positive and negative charges in the external electric field manifests itself in form of induced electric *dipoles*, and it is called *polarization*.

Several different mechanisms are responsible for polarization phenomena, e.g. electron displacement (dominant in non-polar solid media), polar molecule reorientation, ionization (dominant in plasma), etc. Similar effects caused by magnetic field are called magnetization (magnetic polarization). Presence of dielectric or magnetic materials alters the flux densities $\mathbf{D}$ and $\mathbf{B}$ due to electric $\mathbf{P}$ and magnetic $\mathbf{M}$ polarizations of the medium unit volume. The constitutive relations of a continuous medium with electric and magnetic properties result from the definitions

$$\mathbf{D}(r,t) = \varepsilon_0 \mathbf{E}(r,t) + \mathbf{P}(r,t), \quad \mathbf{B}(r,t) = \mu_0 \mathbf{H}(r,t) + \mathbf{M}(r,t), \tag{3}$$

where $\mathbf{P}(r,t)$ and $\mathbf{M}(r,t)$ usually linearly depend on the fields $\mathbf{E}(r,t)$ and $\mathbf{H}(r,t)$. In majority of electromagnetically isotropic materials, the electric and magnetic responses of the medium can be separated. Then the susceptibilities of the isotropic material exposed to the time-dependent fields can be represented by the following linear relations

$$\mathbf{P}(r,t) = \varepsilon_0 \int_{-\infty}^{t} \kappa_e(r, t-t') \mathbf{E}(r,t') dt', \quad \mathbf{M}(r,t) = \mu_0 \int_{-\infty}^{t} \kappa_m(r, t-t') \mathbf{H}(r,t') dt', \tag{4}$$

where the scalar functions $\kappa_{e,m}$ are the electric and magnetic susceptibilities.

The time required to develop material reaction to the electromagnetic field exposure varies, and any response from matter may strongly depend on the frequency. For example, electronic polarization is more sensitive to higher frequencies than ionic polarization, and in artificial materials that are made of macroscopic artificial molecules the response may be very strong at some frequencies in the radio (especially, microwave) range or in the optical range, strongly dependent on the size and shape of the inclusions. The instantaneous polarization at time t is expressed as a convolution over the past history and it has rather complicated form in time domain. On the other hand, since the induced polarization is represented by a convolution in the time domain (4), its expression in the frequency domain is reduced to a simple multiplication operation:

$$\mathbf{P}(r,\omega) = \epsilon_0 \tilde{\kappa}_e(r,\omega) \mathbf{E}(r,\omega), \qquad \mathbf{M}(r,\omega) = \mu_0 \tilde{\kappa}_m(r,\omega) \mathbf{H}(r,\omega), \tag{5}$$

where $\tilde{\kappa}_{e,m}(\omega)$ are the Fourier transformed kernels $\kappa_{e,m}(t)$. This leads to the material (constitutive) relation between the electric flux density $\mathbf{D}$ and the electric field strength $\mathbf{E}$ and that the magnetic flux density $\mathbf{B}$ and the magnetic field strength $\mathbf{H}$ at frequency ω in the form:

$$\mathbf{D} = \varepsilon_0 \varepsilon(r, \omega)\mathbf{E}, \quad \mathbf{B} = \mu_0 \mu(r, \omega)\mathbf{H} \tag{6}$$

with $\varepsilon = 1 + \tilde{\kappa}_e$ and $\mu = 1 + \tilde{\kappa}_m$.

In this equation $\mathbf{E}$ and $\mathbf{H}$ are the so-called complex amplitudes of the *macroscopic* electric and magnetic fields which result from the averaging of the actual (microscopic) electric and magnetic field vectors over the unit cell of the material. This unit cell for simple arrays, such as a simple cubic lattice, includes only one particle forming the material, e.g. one atom or molecule (or an "artificial atom or molecule" of a metamaterial). For complex arrays the unit cell can contain many particles.

Material parameters ε and μ in (6) are called relative permittivity and relative permeability, respectively. They can be functions of not only frequency ω, but also of spatial coordinates. However, the latter dependence exists only in inhomogeneous materials with spatially variable parameters. In homogeneous media the permittivity and permeability are spatially invariant and, therefore, their coordinate dependencies can be dropped.

Now let us consider artificial materials. They operate in the radio, microwave, or optical frequency ranges, where the host materials (as a rule, some natural dielectrics) can be with the highest achievable accuracy considered as electromagnetically continuous media. Incorporating many small inhomogeneities called "inclusions" or particles in such a host material one can obtain artificial dielectrics whose permittivity can be very different from that of the host medium. This is so if the dielectric properties (permittivity) of inclusions are different enough from that of the background medium. Particles immersed in a homogeneous host material may be arranged in periodic or quasi-periodic lattices, or randomly dispersed. The response of such *composite medium* to electromagnetic exposure is predominantly determined by the particle polarization which in its turn together with the array geometry determines the possibility to introduce macroscopic description of the whose array in terms of its effective material parameters. The basic properties of artificial media fundamentally depend on the electrical dimensions of inclusions and the lattice periods as compared with the wavelength of the applied field. When the particle sizes and inter-particle distances are small enough compared to the wavelength, this is the key prerequisite for the possibility of homogenization, i.e. of the representation of the artificial medium as an effectively continuous one (this is the case of "electromagnetic materials").

Most often optically small inclusions behave in the applied electromagnetic field as electric dipoles whose amplitude is proportional to the applied field. If the dipole moment of inclusions in the frequency range of our interest does not experience a resonance, the permittivity of the whole composite can be calculated using static *mixing rules*. In the simplest case when the concentration of inclusions is small or the contrast is not high, this effective permittivity is just the averaged value, in which the permittivities of the matrix (host medium) and of particles enter with appropriate weight coefficients. The weight coefficient here is the volume fraction ratio of the medium in the composite. Dispersive dielectric properties of nanostructured materials appear mainly due to non-trivial (e.g., negative) permittivity of the material from which the inclusions are made. The shape and arrangement of inclusions can strongly influence to the resulting ε.

Inclusions can be also made of a natural magnetic, then the composite can possess magnetic properties. Magnetic properties of materials are described usually by permeability μ. Most of

the current research in metamaterials mainly concerns with the design of magnetic properties using non-magnetic constitutive materials. This is so because nanostructured materials exhibit their useful electromagnetic properties mainly in the optical range where the natural magnetism vanishes. Therefore, magnetic properties of nanostructured materials refer to the so-called "artificial magnetism". This phenomenon is a special case of the so-called *weak spatial dispersion*. In this case one can think about magnetic materials consisting of artificial molecules rather than of artificial atoms. Basically, these can be arrays of specially arranged particles with more than one particle in the unit cell (whereas particles within each unit cell interact mainly among themselves). Alternatively, these can be arrays of complex-shaped inclusions (such as split metal rings) where circulating conduction or polarization currents are induced in every inclusion by external electromagnetic field. In the literature, one can also find a number of attempts to exploit uniform magnetic resonances in nanoinclusions of natural magnetic materials, e.g. ferromagnets and antiferromagnets. These nanostructures operate in the radio frequency range, since in the optical range natural magnetics lose their magnetic properties. In all these cases magnetic materials have a non-trivial permeability, which is dispersive, i.e., strongly depends on the frequency.

Metamaterials, being effectively homogeneous media, can acquire unusual properties, e.g. artificial magnetism. For example, high permittivity small dielectric spheres embedded in a homogeneous dielectric matrix form an artificial medium in which the magnetic permeability is substantially different from unity, whereas the permeability of both constituent materials is unity. This is so because a small dielectric sphere under the condition of a high optical contrast can scatter the incident wave as if it were a magnetic dipole scatterer. This phenomenon is called the magnetic Mie resonance.

2.2 Conducting, diamagnetic and paramagnetic materials

In contrast to dielectrics where tightly bound charges experience only displacement in the applied field, in good conductors such as copper or silver, there are conduction electrons that are free to drift in the material. Therefore, external electric field causes directed motion of free electrons along the field, i.e., electric current. In most of good conductors, the current density $\mathbf{J}$ and electric field $\mathbf{E}$ are related by the Ohm law which in its general form can be written as

$$\mathbf{J}(\omega, \mathbf{r}) = \sigma(\omega, \mathbf{r})\mathbf{E}(\omega, \mathbf{r}), \tag{7}$$

where σ is the electric conductivity of the material expressed in Siemens per meter. Conductivity of a material is an intrinsic property of the material determined by mobility of free electrons. The motion of individual conduction electrons is exceedingly complex and is defined by the material microstructure. Conductivity may be anisotropic, i.e., dependent on the current direction. In semiconductors, it is usually a nonlinear function of $\mathbf{E}$. The conduction electron mobility is responsible for conductivity of conductors at frequencies below 10 GHz. At higher frequencies, the polarization effects should be taken into account when one calculates σ. Real materials are neither perfect conductors nor insulators, i.e., actual dielectrics have some conductivity whilst conductors demonstrate (at high frequencies) relative permittivity which is different from unity. Both these characteristics can be combined by introducing *complex permittivity* of the material for time harmonic electromagnetic field into a complex permittivity $\varepsilon = \varepsilon' - j\varepsilon''$ for the time dependence $\exp(j\omega t)$ or $\varepsilon = \varepsilon' + i\varepsilon''$ for the time dependence $\exp(-i\omega t)$. In good conductors $\varepsilon'' = \sigma/(\omega\varepsilon_0)$. In many practical situations, it is convenient to use the idealizations of perfect conductor ($\sigma \to \infty$) and perfect (lossless) dielectric ($\sigma = 0$).

The criteria of treating the medium as a perfect conductor or a lossless dielectric are inferred from comparison of conduction and displacement current magnitudes.

Permeability of most dielectrics and conductors is very close to unity. However, there are two types of weakly magnetic materials – diamagnetic with $\mu < 1$ and paramagnetic with $\mu > 1$. For example, copper is diamagnetic with $\mu = 0.999991$, whilst aluminium is paramagnetic with $\mu = 1.00002$. Natural magnetics such as ferromagnetic materials have very high permeability at dc ("direct current" at zero frequency), e.g. iron has static permeability $\mu(0) = 5,000$. However, already at 1 GHz, permeability of iron falls practically to unity. Permittivity, permeability and conductivity of materials vary significantly with the frequency. However, in many practical applications they may be considered frequency independent until the operating wavelengths remains substantially greater than the characteristic scale of the material microstructure or nanostructure. If the frequencies of electromagnetic fields approach the intrinsic resonances of matter, the characterization of the medium by its static parameters becomes inadequate even for natural materials.

2.3 Anisotropic and bianisotropic media

The main distinctive feature of isotropic media is that the electromagnetic response is invariant of the propagation direction and polarization of the electromagnetic field. Therefore the material parameters for isotropic media can be reduced to scalar coefficients (in simple media, permittivity and permeability). Models of isotropic homogeneous media provide valuable insight to the main properties of artificial electromagnetic materials. However, many practical materials are not isotropic while remaining reasonably homogeneous. They are called anisotropic materials. For example, most of known metamaterials are anisotropic as their internal microstructure or nanostructure has certain lattice-like arrangements with distinctive axes directions. This implies that the field orientation with respect to the internal structure affects the medium response. Often the linear relations exist between the four vectors of the electromagnetic fields, and the constitutive relations, introduced in the previous subsection, should be generalized by introducing the medium parameters in the form of the permittivity and permeability tensors.

Another complication, also related to complex geometry of nanostructures is electromagnetic coupling, when electric field applied to a (natural or artificial) molecule excites not only electric dipole moment, but also magnetic dipole moment. Likewise, magnetic field can in the general case excite both magnetic and electric polarizations. Materials with magneto-electric coupling are called *bi-isotropic materials* if their response is isotropic or *bianisotropic materials*, if they are anisotropic. In bianisotropic materials the electric polarization and magnetization can be expressed in the following form:

$$\mathbf{P} = \mathbf{P}_0 + \varepsilon_0 \overline{\overline{\kappa}}_e(\omega) \cdot \mathbf{E} + \sqrt{\varepsilon_0 \mu_0}\, \overline{\overline{\kappa}}_{em}(\omega) \cdot \mathbf{H}, \tag{8}$$

$$\mathbf{M} = \mathbf{M}_0 + \mu_0 \overline{\overline{\kappa}}_m(\omega) \cdot \mathbf{H} + \sqrt{\varepsilon_0 \mu_0}\, \overline{\overline{\kappa}}_{me}(\omega) \cdot \mathbf{E}, \tag{9}$$

where $\mathbf{P}_0, \mathbf{M}_0$ are the static electric polarization and magnetization which exist in the absence of external alternating fields, e.g., in ferroic and multi-ferroic materials, the dimensionless tensors $\overline{\overline{\kappa}}_{e,m}(\omega)$ are the electric and magnetic susceptibilities of an anisotropic medium, and the dimensionless tensors $\overline{\overline{\kappa}}_{em}$ and $\overline{\overline{\kappa}}_{me}$ describe the electric polarization of a bianisotropic medium induced by a magnetic field or magnetization caused by an electric field. The latter terms describe the linear magneto-electric effect which is observed in several oxide materials as well as in artificial bianisotropic materials which all refer to the class of metamaterials.

Although the polarization and magnetization expansions above can be further extended with higher-order (multipole) terms, the presented form proved to be sufficient for the description of the major phenomena encountered in the vast majority of electromagnetic materials, both natural and artificial. The general constitutive relations are represented as the linear interrelations between the vector electric and magnetic flux densities and vector electric and magnetic field strengths. The respective proportionality coefficients expressed in the tensor form are usually taken as the extrinsic (phenomenological) material parameters. From relations (8), (9) and (6) we arrive to the constitutive relations of bianisotropic media:

$$\begin{aligned} \mathbf{D} &= \varepsilon_0 \overline{\overline{\varepsilon}} \cdot \mathbf{E} + \sqrt{\varepsilon_0 \mu_0}\, \overline{\overline{\kappa}}_{em}(\omega) \cdot \mathbf{H}, \\ \mathbf{B} &= \mu_0 \overline{\overline{\mu}} \cdot \mathbf{H} + \sqrt{\varepsilon_0 \mu_0}\, \overline{\overline{\kappa}}_{me}(\omega) \cdot \mathbf{E}, \end{aligned} \tag{10}$$

where $\overline{\overline{\varepsilon}} = \overline{\overline{1}} + \overline{\overline{\kappa}}_e$, $\overline{\overline{\mu}} = \overline{\overline{1}} + \overline{\overline{\kappa}}_m$ and it is assumed that $\mathbf{P}_0 = \mathbf{M}_0 = 0$. In the case of a simply anisotropic medium $\overline{\overline{\kappa}}_{em} = \overline{\overline{\kappa}}_{me} = 0$. It can be shown that in the lossless medium all the components of tensors $\overline{\overline{\varepsilon}}$ and $\overline{\overline{\mu}}$ are real, whereas $\overline{\overline{\kappa}}_{em}$ and $\overline{\overline{\kappa}}_{em}$ are purely imaginary.

The four tensors involved in the constitutive relations in a general case of bianisotropic medium contain in general 36 complex (nine in each tensor and four tensors) numerical parameters. However, not all these parameters are independent due to the reciprocity conditions that impose certain conditions on the tensors. Also the lattice symmetry of the material microstructure may restrict certain interactions and thus significantly reduce the number of nonzero and distinctive tensor components. It is noteworthy that magneto-electric coupling may exist in isotropic materials where electric field causes the magnetic response and magnetic field excites the electric polarization, where both these effects are direction independent. Such bi-isotropic materials are characterized by four scalar parameters instead of four tensors. Finally, it is necessary to remark that most metamaterials are described by the general bianisotropy theory, but even the use of so large number of parameters might be insufficient to adequately characterize the properties of many effectively continuous artificial materials.

2.4 Reciprocity and non-reciprocity

The principle of reciprocity is fundamental for the electromagnetic media, and all electromagnetic materials can be cast in the two categories: reciprocal and nonreciprocal media. The notion of reciprocity is directly related to the symmetry with respect of the time inversion operation. Most natural materials are reciprocal, except for naturally biased magnetic materials, such as hard ferrites and antiferromagnetics, and externally biased media, e.g. magnetized plasma, voltage biased semiconductor junctions, etc. In practical terms, reciprocity means an invariance of the media response when the positions of the field source and probe are interchanged. For bianisotropic medium to be reciprocal, its constitutive parameters must obey the following relations:

$$\overline{\overline{\varepsilon}} = \overline{\overline{\varepsilon}}^T, \qquad \overline{\overline{\mu}} = \overline{\overline{\mu}}^T, \qquad \overline{\overline{\kappa}}_{me} = -\overline{\overline{\kappa}}_{em}^T \equiv j\,\overline{\overline{\kappa}}, \tag{11}$$

where superscript T denotes the transpose operation. The tensor $\overline{\overline{\kappa}}$ (real for lossless media) is usually called the *chirality parameter*. From these relations we see that the permittivity and permeability of reciprocal media are symmetric tensors, whereas the tensors of magneto-electric cross-coupling are negatively transposed. It is noteworthy that the reciprocity conditions above reduce the number of the parameters describing bianisotropic medium from 36 to 21 complex valued quantities.

In the general case of non-reciprocal bianisotropic medium the constitutive relations take the form

$$\mathbf{D} = \varepsilon_0 \overline{\overline{\varepsilon}} \cdot \mathbf{E} + \sqrt{\varepsilon_0 \mu_0}\,(\overline{\overline{\chi}} - j\overline{\overline{\kappa}}) \cdot \mathbf{H}, \tag{12}$$

$$\mathbf{B} = \mu_0 \overline{\overline{\mu}} \cdot \mathbf{H} + \sqrt{\varepsilon_0 \mu_0}\,(\overline{\overline{\chi}} + j\overline{\overline{\kappa}})^T \cdot \mathbf{E}. \tag{13}$$

Permittivity and permeability of general non-reciprocal media contain antisymmetric parts: they are Hermitian tensors with complex conjugated non-diagonal elements. The additional magnetoelectric tensor $\overline{\overline{\chi}}$ describes the non-reciprocal cross-polarization effect and is called *Tellegen's tensor*. The anti-symmetric parts of permittivity and permeability as well as the Tellegen tensor are nonzero only if there is an external field which changes sign (breaks the symmetry) under the time-reversal operation. This stimulus can be an external magnetic bias field or internal magnetization of the medium.

All bianisotropic materials can be categorized into 14 classes based upon 7 reciprocal and 7 nonreciprocal components of their material parameter tensors which are related to the reciprocal or non-reciprocal nature of their constituents [5].

2.5 Piezoelectric and magnonic materials

Piezoelectricity is an electro-mechanical effect in solids when the electric field is generated in response to the applied mechanical stress. This phenomenon is associated with displacement of positive and negative charges caused by the lattice strain in certain crystals (e.g., quartz, tourmaline) and ceramic materials (e.g. $BaTiO_3$, PZT) with the special crystalline symmetry. The piezoelectric effect is reversible, i.e., voltage applied to the material inflicts the lattice strain and linear deformations of the crystals. Polarization of piezoelectric materials is obtained from the coupled electro-mechanical equations relating stress-strain and stress-charge (direct piezoelectric effect). Since the mechanical stresses and strains themselves are represented by the tensors of the 2nd rank, their relations with electric flux density are generally expressed as the tensors of rank 4. Depending on the crystal class and symmetry of piezoelectric material, the stress-strain and strain-charge tensors contain only a few nonzero components. Nevertheless the constitutive relations for piezoelectrics are much more complicated than for ordinary anisotropic materials.

Nanostructured magnetic materials can be characterized as effectively continuous media in the long-wavelength limit. Their effective parameters are represented by the Hermitian permeability tensor and the diagonal tensor of permittivity. Owing to the crystalline microstructure of the magnetically active materials, boundaries of the crystal domains and grains create internal magnetic bias which causes resonant precession of the magnetic dipoles in applied high-frequency magnetic fields. The precessing magnetic dipoles in the known ferrimagnetic structures exhibit resonances at GHz frequencies and support spin waves which manifest themselves in very short waves of nonuniform precession travelling through the ensemble of magnetic dipoles. The theoretical analysis of the periodic structures containing ferrimagnetic inclusions in dielectric matrix has shown that the response of such artificial media is determined by the uniform ferrimagnetic resonances of the constituent particles and limited to the GHz frequency range. All the resonant phenomena in such composite materials are described by the Hermitian tensor $\overline{\overline{\mu}}$. In contrast to anisotropic materials with the symmetric tensor $\overline{\overline{\mu}}$, the Hermitian tensor represents a unique notion of gyrotropic medium. The latter feature of magnetically active composite materials is of particular importance for enabling nonreciprocal propagation

of both spin and bulk electromagnetic waves. Historically, magnetic nanostructures (called magnonic media) have been associated with excitation of slow spin waves (called magnons) and attempts to increase the frequencies where the spin-wave resonances could be exploited. However, to bring the spin wave phenomena in nanoscaled artificial materials to the THz and optical ranges, it is necessary to resolve the two principal problems of (i) the efficient coupling of electromagnetic fields to non-uniform spin waves and (ii) high losses of spin waves at these frequencies.

2.6 Effectively continuous surface materials

If we deal with very optically thin nanostructures impressed into uniform material or located at its surface, the boundary problem can be formulated in terms of the so-called transition boundary conditions. Consider first an optically thin layer consisting of a highly conducting non-magnetic material. The conducting layer of optically small thickness d illuminated by an electromagnetic wave can be presented as an infinitesimally thin induced current sheet. The surface density of the electric current in this sheet is $\mathbf{J} = \tau\mathbf{E}_t$, where $\mathbf{E}_t$ is the tangential component of the total electric field in the layer (the total field is the sum of the incident and reflected ones) and τ is the sheet material parameter called *surface susceptibility*, which is related to the bulk conductivity σ as $\tau = d\sigma$. The electric field $\mathbf{E}$ in the layer can be calculated as the average of electric fields at the layer surfaces $\mathbf{E}(d)$ and $\mathbf{E}(0)$: $\mathbf{E} = [\mathbf{E}(d) + \mathbf{E}(0)]/2$. From the Maxwell equations it follows that the surface current is equal to the jump of the tangential magnetic field across it. Therefore, the equation for the surface current induced in the layer (sheet) can be described through parameter τ in the form of a transition boundary condition:

$$H_t(d) - H_t(0) = \tau[E_t(d) + E_t(0)]/2. \tag{14}$$

This condition relates the electric and magnetic fields taken at the two sides of the surface "material". It is invariant with respect to the incidence angle. Together with the known permittivity of the substrate (or that of the host dielectric matrix) (14) allows one to find the reflection and transmission coefficients for incident plane electromagnetic waves.

The case of a lossy dielectric layer with complex relative permittivity ε is more complicated in what concerns the relation between τ and ε, namely $\tau = \omega\varepsilon_0(\varepsilon - 1)d$. However, the relation (14) holds. Similar situation holds in the case when the layer has also magnetic properties. However, in this case the electric field is also discontinuous across the layer and (14) should be complemented by relation

$$E_t(d) - E_t(0) = \chi[H_t(d) + H_t(0)]/2. \tag{15}$$

For a non-magnetic layer $\chi = 0$. For a magnetic one it is related to the bulk permeability as $\chi = \omega\mu_0(\mu - 1)d$. For anisotropic magneto-dielectric media parameters ε and μ are tensors, and therefore in the general case τ and χ can be tensors, too.

The description of a natural surface material in terms of surface material parameters τ and χ is equivalent to its description in terms of bulk material parameters ε and μ. The last ones result in the same solution of any boundary electromagnetic problem if the layer finite thickness d is properly taken into account. However, this equivalence holds only because for a natural magneto-dielectric layer there are very many atoms across the layer and the material is anyway inherently bulk. Its representation as an effective surface is an additional approximation which simplifies the solution of the boundary problems.

For nanostructured surface materials, say, for a grid of nanoparticles located on a substrate or embedded into a dielectric matrix the presentation in terms of the continuous sheet can be called "surface homogenization". Such structures were called metasurfaces or metafilms in [28, 29]. Bulk material parameters ε and μ cannot be introduced for a metasurface by a number of reasons. Among them the following reason appears to be the main one. Bulk material parameters imply the refraction coefficient, i.e., the phenomenon of the wave refraction should be observed. However, a monolayer of small scatterers does not refract the incident wave: Behind the layer the wave propagates in the same direction as if the monolayer were absent. However, induced electric dipoles of the metasurface cause a jump of the magnetic field given by equation (14), and magnetic dipoles results in a jump of the electric field given by equation (15). Thus, nanostructured surface materials can be electromagnetically characterized by surface material parameters τ and χ (tensorial in the general case) but cannot be described by bulk material parameters.

2.7 Band-gap structures (photonic crystals)

During the last few decades much attention has been attracted to electrodynamic properties of periodical structures where permittivity and/or permeability are periodical functions of the coordinates. Following quantum mechanics such structure are referred to as "photonic crystals" (PC) or "band-gap structures". There is an analogy between Maxwells equations for electromagnetic wave propagation and Scrödingers equation for electrons propagation in a periodic potential. Consider a monochromatic electromagnetic wave of frequency ω propagating in a medium whose permittivity varies from point to point in space as $\varepsilon(\mathbf{r}) = \varepsilon(\mathbf{r}+\mathbf{a})$. Then $\varepsilon(\mathbf{r}) = \varepsilon_{\text{av.}} + \Delta\varepsilon(\mathbf{r})$, where the average value of $\Delta\varepsilon(\mathbf{r})$ over the lattice period is zero. The last term (more exactly $k^2\Delta\varepsilon$, where k is the wave number in the medium of permittivity $\varepsilon_{\text{av.}}$) plays the role of the scattering potential in Schrödinger's equation.

An electron in a semiconductor crystal (periodic arrangement of atomic potentials) and a photon in a PC (periodically modulated dielectric or magneto-dielectric medium) possess a variety of band gaps, where the propagation is forbidden for certain ranges of wavelengths. Using different materials (different dielectric constants) and by adjusting geometrical parameters, the propagation of light can be modified in a controllable manner.

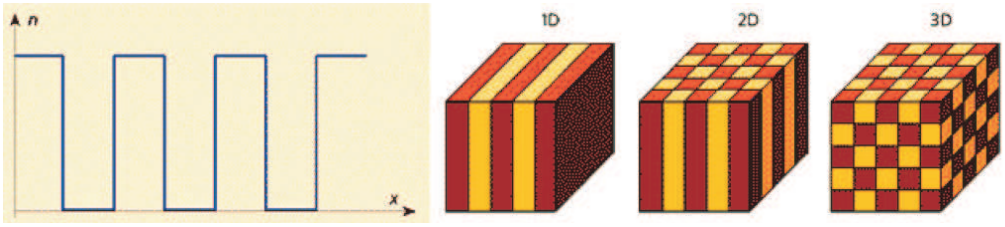

Figure 1: Left – The coordinate dependence of the local refraction index n (n is the square root of permittivity $\varepsilon(\mathbf{r})$) in photonic crystals shown in the right panel. Right panel – the schemes of typical 1D, 2D and 3D photonic crystals.

Band gaps (BG) appear due to the so-called Bragg reflection. The condition of this reflection is an integer number of half wavelengthes equal to the the lattice period along the propagation direction. The light is reflected from the crystal planes in phase and as a result totally reflects from the surface of a PC. In an infinite crystal such a wave cannot propagate, in other words, its wave number is imaginary and it should exponentially decay if it is excited by any source.

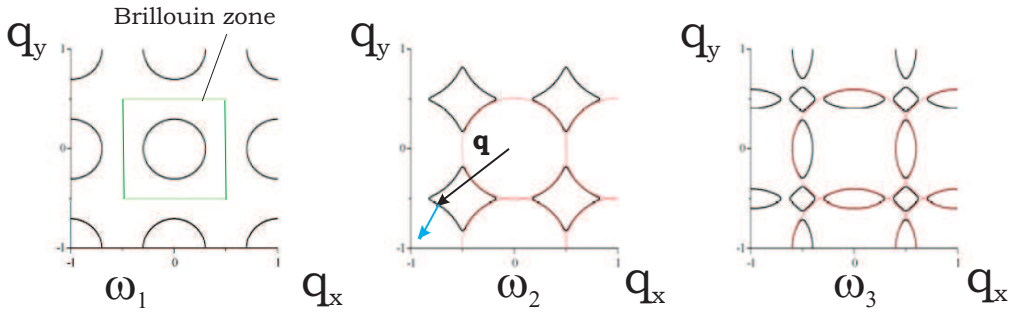

Figure 2: Typical isofrequency contours for a PC of at three different frequencies above the first BG.

For finite-thickness PC the BG are important if the thickness is larger than the decay distance D. The last one is related to the BG width $\Delta\omega$, lattice period a and operating frequency ω as [90]:

$$D = \frac{a\omega}{\pi\Delta\omega}. \tag{16}$$

For a harmonically varying permittivity $\Delta\varepsilon(z) = A\cos\Omega z = A\cos(2\pi z/a)$ one has [90]:

$$\Delta\omega = \omega\frac{A}{\varepsilon_{\text{av.}}}. \tag{17}$$

An important feature of PC is that the propagating eigenwaves in them are so-called Bloch waves. A Bloch wave is a product of the factor describing a propagating plane wave with wave number q, i.e. $\exp(-jqz)$ and a certain function $u(\mathbf{r})$ which is periodic with the lattice period a. In the case when the wave propagates not along the lattice coordinate axis, the plane wave is described as $\exp(-j\mathbf{q}\cdot\mathbf{r})$ and $u(\mathbf{r})$ is periodic with a vector period $\mathbf{a}(\mathbf{a_x}, \mathbf{a_y}, \mathbf{a_z})$. The wave vector $\mathbf{q}$ in periodical media is called the Bloch wave vector.

To understand the properties of PC and their difference from natural materials it is convenient to employ the so-called isofrequency surfaces (or their sections by a coordinate plane called isofrequency contours). Isofrequency surfaces were introduced in the beginning of XIX century by Fresnel and have been since that time widely used in optics of anisotropic materials. An isofrequency surface is a locus of the ends of wave vectors (the Bloch wave vectors in the case of PC) at a fixed frequency. The isofrequency surface is a periodic pattern. In Fig. 1 besides a 1D PC (periodical stack) the so-called chess-board PC are shown as examples of 2D and 3D PC. PC are not obviously chess-board structures, all artificial periodic arrays can be referred to PC. However, the chess-board PC are typical. Isofrequency contours of a 3D chess-board PC depicted in Fig. 1, right panel, for three different frequencies are shown in Fig. 2. Two frequencies refer to the 2d pass-band (laying above the lowest BG), and the third one refers to the 3d pass-band (above the 2d BG). The group velocity of the eigenwave is directed along the normal to the isofrequency surface and points to the direction of the isofrequency shift with frequency increase. It is shown as a blue arrow in Fig. 2.

PC for which these isofrequencies are calculated is geometrically isotropic. For continuous isotropic media all isofrequency contours are obviously circular [119], but the PC is not a continuous medium. We can see that isofrequency contours of PC deviate from circular ones very strongly. This difference is the manifestation of the general phenomenon called strong spatial dispersion. In this situation the dispersion of signals in PC is very different from that in continuous media. Other manifestations of strong spatial dispersion are band gaps

(Bragg reflection of waves whose frequencies belong to band gaps) and the existence of multiple scattered plane waves.

In PC one observes many other interesting and useful phenomena such as all-angle negative refraction, which leads to the parallel-plate focusing almost without aberrations, extraordinary dispersion (so-called photonic super-prism is a class of optical components based on this effect), modes concentration in cavities, defect waveguiding, surface states, non-linear effects such as modification of radiation spectra of quantum emitters and many others. Most of these effects are tightly related to the strong spatial dispersion in these structures. Strong spatial dispersion in PC occurs at frequencies above the lower edge of the first low-frequency band gap. This frequency is called the lowest Bragg frequency or frequency of the first spatial resonance of the lattice. At lower frequencies band-gap structures behave nearly as continuous media with uniform permittivity $\varepsilon(\mathbf{r}) = \varepsilon_{\text{av}}$. Therefore at so low frequencies artificial lattices are not called PC and are referred to as artificial dielectrics. If their constitutive elements are magnetic they are referred as composite magnetics. An exception is the class of metamaterials which are as a rule also lattices operating at frequencies below the first spatial resonance. Due to the resonance response of their inclusions metamaterials cannot be considered as media whose permittivity is averaged over the total volume of the unit cell. This makes their electromagnetic characterization so problematic.

Effective material parameters ε and μ can be formally introduced for PC also at high frequencies above the first band gap using definitions (3) and (6). However, as it is known for media with strong spatial dispersion [119], these material parameters (though they can be used for studying the eigenwaves in the unbounded lattices) become useless for solving boundary problems. Therefore they cannot be used as characteristic material parameters. There is little physical meaning in these parameters since in the photonic crystal regime the electric response of the lattice unit cell to the propagating electromagnetic wave cannot be separated from the magnetic response. The wave interaction with any unit cell is fully dynamic. Therefore permittivity and permeability of the discrete medium in formulas (6) above the first band gap of a PC are functions of the wave vector $\mathbf{q}$.

2.8 Resonant and non-resonant materials

In some frequency regions the permittivity and permeability of materials can exhibit resonances, where these parameters abruptly vary with the frequency of applied fields. For resonant structures their permittivity and permeability (if the last one is also resonant) suffer large losses within the resonant band. For non-resonant structures permittivity and permeability change slowly when the frequency is varied, and usually have much smaller losses. Whilst the name might suggest otherwise, non-resonant composite materials do have frequencies at which their particles (for composite media – inclusions) are resonant, but these resonances occur at higher frequencies. Conversely to resonant nanostructures, non-resonant ones have a broad bandwidth at which their relative permittivity can make a strong contrast to the free space (significantly different from unity). In the optical range these materials are metals and semiconductors. This broadband contrast is their primary advantage in design of various devices. Their main disadvantage is that they have small dynamic range of material parameters. Provided they do not have negative permittivity or very large positive one, simple-shape particles such as spheres or ellipsoids are non-resonant. In the case of metamaterials, the resonant frequencies strongly depend on the shape and the size of inclusions, in addition to their chemical composition.

2.9 Quasi-static limit

In the quasi-static limit the optical size of the medium unit cell is by definition at least 100 times smaller than the wavelength in the medium. In this case the spatial variation of the local exciting field over the unit cell is negligible even for complex-shaped particles, and the phenomena of artificial magnetism and bianisotropy in reciprocal media disappear. This situation corresponds to most natural media in the optical frequency range (moreover, even in the radio frequency range). Artificial magnetism is never observed in natural media, only bianisotropy is not negligible for some natural materials in the visible frequency range (chiral materials, like sugar solution, for example).

For composite materials of non-magnetic inclusions in the quasi-static limit only the permittivity remains different from that of the host medium. If inclusions are magnetic (for nanostructured materials this corresponds to arrays of nanomagnets), the permeability in the quasi-static limit can be different from unity. In the literature one can meet opinions that the correct electromagnetic characterization of materials in the condensed form of a few material parameters is possible only in the quasi-static limit when the frequency dispersion of the medium response is absent (see e.g. [131, 132, 133, 134, 135]. Following to these authors we should have recognized the concept of artificial magnetism and bianisotropy as incorrect ones. However, if the medium satisfies the locality limitations, the basic theory does not inhibit the electromagnetic characterization of the medium in terms of permittivity, permeability and magnetoelectric coupling tensors. The only valid limitation to the electromagnetic characterization of composite media in a condensed form is, in our opinion, the locality limitation (see more detailed discussion below) .

3 Classification of nanostructured materials

Inspecting different types of existing nanostructured materials (NSM) (and prospective nanostructured materials which are under discussion in the current literature) we can find many types of them which satisfy the above definition of metamaterial (MTM). To show the place of nanostructured MTM among all nanostructured materials it is instructive to introduce 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 described 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 medium formed by them. The left upper cell has rich content which is detailed in Table 1. The most important criterion of the classification is the dimension of the array of constituents which forms 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 correspond to the case when the artificial material is a layer including only 1-3 constitutive elements across it. Analogous classification can be possibly suggested for non-electromagnetic MTM.

MTM of the surface type were called metasurfaces in [40, 41] and metafilms in papers [42, 43]. Notice that in these papers a consistent method of electromagnetic characterization of metasurfaces (metafilms) comprising one inclusion across the layer thickness 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 as “materials” since the concept of material is usually not applied to

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

Nano-structures (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 (meta-films), nanostructured sheets without MTM properties	Plasmonic diffraction grids, optical bandgap and frequency selective surfaces	Artificial nano-structured surfaces with long inclusions or slots
1D, lines	Metawaveguides	Not yet known but possible	Impossible

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 nanostructures usually operate (i.e. possess useful and unusual electromagnetic properties). Large optical size practically implies $|q|a > 1$. In this range the structure cannot be characterized by local material parameters (neither bulk nor surface ones). In the opposite case $|q|a \ll 1$ the structure in principle can be characterized by local parameters and can be referred to as MTM. The same structure at different frequencies can be referred either to optically dense or to optically sparse types.

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 enabled by the resonance of inclusions.

Let us give some comments on Table 1. Nanostructured composites can be designed for whatever purpose and not always possess any interesting or unusual electromagnetic properties. Instead they can possess useful thermal and mechanical properties, for example, or 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 (photonic crystals) 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. Extensive 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 we do not refer these structures

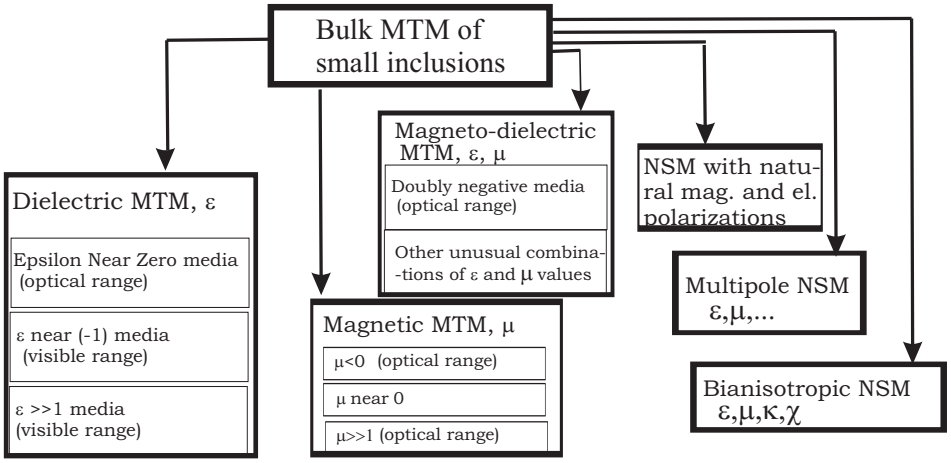

Figure 3: Classification of bulk nanostructured MTM of small inclusions. Optical range in this diagram by definition covers infrared, visible and near ultraviolet ranges.

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 at the plasmon resonance of the rods enhance the field at the plane corresponding to the upper edges of the rods [44]. 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 [41]). Another important example of a metasurface is a monolayer optical fishnet (see e.g. in Chapter 29 of [41]). 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 [45]) and nanostructured fibers with plasmonic insertions (see e.g. in [46]). They also can be used for frequency filtering of optical signals on the nanoscale level (e.g. in [47]).

Multilayer optical fishnets and multilayer metal-dielectric nanostructures (see e.g. [48]–[53]) 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), the negative group velocity has been observed as well (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. [54]). Note that known fishnets do not offer subwavelength imaging in the backward-wave frequency range, 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 [40]). 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 [55].

In Fig. 3 bulk MTM are classified based on their material parameters. Optically dense nanocomposites can be artificial dielectric media with unusual permittivity, i.e. ε close to 0 or to (-1) or very high ($\varepsilon > 10$ in the visible range where usual materials (except some very lossy semiconductors and liquid crystals) have rather low permittivity ($\varepsilon < 6-7$). Artificial magnetic media, possessing negative permeability in the optical range are also MTM. Not only media with negative μ deserve to be referred to MTM, media with $\mu \approx 0$ or even $\mu = 2$ in the optical range are also MTM. Media with both negative permittivity and permeability represent the most known type of MTM, however if $\varepsilon = 0$ and $\mu = 2$ in the visible range such material 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 additional 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 Chapter 2 of [40].

The consistent classification of nanostructured MTM is helpful to avoid the most dangerous pitfalls in their electromagnetic characterization: the inconsistent classification of the material under study. In many publications describing metasurfaces, i.e. MTM layers with only 1-2 resonant inclusions over the layer thickness, authors of [48]–[50], [56]–[64] 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 [65]–[77].

4 Introduction to electromagnetic characterization of complex nanostructured materials

4.1 Preliminary notes

In spite of the fast development of nanoscience and nanotechnologies the problem of the electromagnetic characterization of nanostructured materials has been scantly studied, and methods and techniques of this characterization are barely developed. This is so due to several reasons. First, a significant part of known nanostructures are not materials in the common meaning of the term. Nanowaveguides, nanoantennas, nanocircuits [1, 2] and other nanodevices (logical nanoelements and optical memory nanocells, surface-plasmon lasers called SPASERs, etc. [3]) are

devices, not materials. The theoretical electromagnetic characterization of these devices (e.g. calculations of the input impedance and pattern of a nanoantenna) does not face the basic theoretical problems. Their characteristic parameters are unambiguous, and only difficulty there is their accurate computation. The attention of specialists in the electromagnetic characterization is strongly attracted to nanodevices and not enough attention is paid to nanomaterials.

As to nanostructured *materials*, the electromagnetic methods of their chemical and structural characterization are well developed. Chemical characterization of constituents can be done using methods of optical spectral analysis, which are well known: Surface-Enhanced Raman Scattering (SERS) and fluorescence and luminescence spectral measurements. Electrostatic and magneto-static properties of some nanostructured materials (e.g. the conductivity of arrays of carbon nanotubes [4]) are well studied and there are no theoretical problems with similar characterization of other nanomaterials. However, when the goal is to characterize the *electromagnetic* properties of nanostructured materials, researchers encounter a very basic difficulty. It is, in principle, more difficult to characterize an artificial material than a device because artificial materials are extended heterogeneous structures comprising a huge number of interacting constitutive elements. For a significant part of nanomaterials it is not only unclear how to calculate (and moreover, how to measure) their electromagnetic characteristic parameters. Even a list of these characteristic parameters has not been conclusively established at present time. One researcher characterizes some nanostructured materials by only dielectric permittivity. Another one claims for the same material some non-trivial permeability. The third proves that both of them are wrong since these parameters applied to those materials have no physical meaning. The fourth researcher asserts that within certain bounds they perhaps can characterize some electromagnetic properties but it is so only if permittivity and permeability are complemented by some additional material parameters.

Even for a specialist in the electromagnetic theory it is sometimes very difficult to judge who of them is wrong and who is right. For example, in many works simple cubic lattices of plasmonic nanospheres are electromagnetically characterized by an only scalar complex parameter – isotropic complex permittivity (e.g. [6, 7]). Meanwhile, in some other papers (e.g. [8]) it is noticed that these lattices are photonic crystals, their permittivity is spatially dispersive and therefore cannot be isotropic. Moreover, in [9] the same lattices are characterizes by four tensor material parameters which are all nonzero and non-trivial (i.e., totally 12 scalar parameters).

This situation in the electromagnetic characterization of nanostrucrtured metamaterials complicates the writing of the state-of-the-art overviews and makes the choice of the best technique very problematic because these problems are not fully understood at this time.

4.2 Locality requirement for effectively continuous media

If all the material parameters depend only on the frequency and possibly on coordinates (assuming slow variations without abrupt changes) but are invariant to the direction into which the applied field is changing in space (direction of the wave propagation, in the case of plane-wave excitation), one tells that the medium is *local*. In other words, for local media the susceptibilities entering (8) and (9) depend only on the material properties (including the material frequency dispersion) and do not depend on the position of external sources, provided they create the same applied field at the observation point.

For effectively continuous media the locality requirements are fulfilled [119], they can be not fulfilled only for discrete media, e.g. for band-gap structures. For isotropic magneto-dielectric media the locality implies (see e.g. in [119]) the system of following conditions:

- Passivity (for the temporal dependence $e^{-i\omega t}$ it implies $\mathrm{Im}(\varepsilon) > 0$ and $\mathrm{Im}(\mu) > 0$ simultaneously at all frequencies, for $e^{j\omega t}$ the sign of both $\mathrm{Im}(\varepsilon)$ and $\mathrm{Im}(\mu)$ should be negative). The violation of passivity in the energetically inactive media (without any electromagnetic field sources at frequency ω) means the violation of the 2nd law of thermodynamics;

- Causality (for media with negligible losses it corresponds to the conditions $\partial(\omega\varepsilon)/\partial\omega > 1$ and $\partial(\omega\mu)/\partial\omega > 1$. This also means that in the frequency regions where losses are negligibly small, the material parameters obviously grow versus frequency: $\partial(\mathrm{Re}(\varepsilon))/\partial\omega > 0$ and $\partial(\mathrm{Re}(\mu))/\partial\omega > 0$);

These mandatory properties of effectively continuous media follow from the independence of the material parameters from the wave vector $\mathbf{q}$ (for a given frequency this means the independence of electromagnetic parameters (EMP) on the propagation direction) [119]. For anisotropic reciprocal media similar requirements of locality can be formulated for all components of tensor material parameters if they are written in a suitable system of coordinates where these tensors are diagonal. It is more difficult but also possible to formulate the locality limitations for bianisotropic and non-reciprocal media.

It is worth noticing that the local bianisotropic constitutive relations model not only local dielectric response, but also effects of chirality and artificial magnetism which are physically slightly non-local effects [119]. In fact, chirality and artificial magnetism are manifestations of the so-called weak spatial dispersion which allows the condensed description of some slightly non-local media in terms of local material parameters by the cost of introducing two additional material parameters (permeability and chirality tensors) [5]. In the framework of the weak spatial dispersion models, the locality requirement assumes that the spatial dispersion is so weak that the constituent particles of the composite medium interact only through their near fields. The wave interaction between particles is not important. Only particles inside a rather optically small volume around the reference particle effectively influence to the local field acting to the reference particle. This volume is often called the Lorentz sphere, though for many possible array geometries it is not a spherical region. The contribution of particles beyond the Lorentz sphere (i.e. located practically in the far zone of the reference particle) is not so important for the response of the reference particle. In other words, the polarization response of the medium calculated at a reference point is local because it is determined by the polarization within the Lorentz sphere. This sphere is an optically small volume around the reference point and the phase of the wave over this volume is effectively uniform. This is the meaning of the term locality. It is clear why for effectively discrete media the locality does not hold. Particles adjacent to the reference one in the effectively discrete media are located in its far zone, and the Lorentz sphere cannot be introduced.

It is interesting to note that in the case of the bianisotropy and artificial magnetism we cannot neglect the spatial variations of the field acting to the reference particle. These effects appear namely due to the small spatial variation of the local field. However, the concept of the Lorentz sphere (over which the phase shift of the wave is negligible) is qualitatively applicable for bianisotropic media and for artificial magnetics. There is no contradiction in the model because the bianisotropy and artificial magnetism appear only when optically small particles have a complex internal geometry, and therefore they feel even a very small spatial variation of the local field.

The two locality requirements are complemented by the requirement on absence of radiation losses in optically dense arrays with uniform concentration of particles [5]. Such arrays do not scatter the incident wave, they only refract it. If particles of the arrays are lossless, i.e. their

material parameters are real, there is no dissipation in particles and no radiation losses in the array. Then the refracted wave must propagate without attenuation. This means that the effective material parameters of lossless materials with regular internal structure should take real values.

4.3 Direct and inverse (retrieval) homogenization approaches

As already indicated in the preceding section, it is of prime importance to define and specify the applicability domain and the physical meaning of every material parameter that is used to characterize a structured material. We recall that only the parameters that describe the material properties in a condensed way consistently and unambiguously can be called characteristic material parameters. Measured quantities are given the name effective parameters, however, in many literature examples as it was shown in [15] these parameters are not uniquely defined and their meaning and applicability limits are not clear. The effective material parameters result from a homogenization procedure, however, only adequate homogenization models guarantee that the effective material parameters fit the concept of the electromagnetic characterization, i.e., give a practically accurate condensed description of the material response to electromagnetic waves.

There are two main approaches to the homogenization of bulk lattice structures. One approach can be called direct homogenization. It is a theoretical approach. It starts from the known polarizabilities of individual particles and goes through averaging of microscopic fields and microscopic (local) polarization and magnetization. The result of the averaging in the sense of the averaging of Maxwell equations is a set of material equations (6) (or in the general case formulas (12) and (13)) where the EMP are expressed through individual polarizabilities of particles. The relations expressing EMP through polarizabilities of individual particles and other parameters of the original heterogeneous structure are the main results of this direct approach. However, to assess whether these EMP are representative of the characteristic parameters, it is necessary to analyze the properties of waves with different polarizations of the electric field vector, propagating at different directions in the lattice.

At first, the same set of EMP should describe eigenwaves propagating in different directions with respect to the lattice axes. However, this is still not enough – applicability of these EMP to the boundary problems (at least the interface problem for layers) should also be checked. If the same set of EMP is also suitable for describing the wave reflection and transmission at the interface at different angles of incidence, then we can finally conclude that this set of EMP is really adequate and these EMP can be called electromagnetic characteristic parameters of the medium.

All these issues have been addressed in the classical homogenization model of natural crystals developed in 1910s-1950s by P. Ewald, S. Oseen, M. Born, D. Sivukhin and others. This model refers to the case when the optical size of the unit cell is very small $|q|a < 0.01$ where q is wave number and a is the lattice constant. For composite media, especially for MTM which normally operate in the range $0.01 < qa < 1$ a similar thorough theory is not developed yet. Moreover, many researchers working with MTM (especially, with nanostructured MTM) prefer the inverse approach suggested in 2002 in works [109, 110]. This approach can be called heuristic homogenization. In this approach the sample formed by an array of artificial particles in the dielectric matrix is heuristically replaced by a body of the same shape filled with an isotropic uniform continuous magneto-dielectric medium with unknown ε and μ to be determined. The latter two complex quantities are retrieved from measurement or from

accurate numerical simulation of two scattering parameters of the body at a specific angle of the wave incidence and wave polarization type (reflection and transmission coefficients). Then it is assumed that the sample composed of the actual MTM will have all scattering characteristics identical to those of the effective continuous magneto-dielectric medium with such ε and μ as were found for the specific case of a particular plane-wave incidence.

Is this approach justified? Any model, even crude, is justified if it is predictive and works in the design of devices. However, the authors of the present report do not know any work, which would prove that ε and μ retrieved in this way for any nanostructured MTM are applicable to all angles of the wave incidence and for both polarizations of exciting waves. On the contrary, from the available literature (see the review in [15]) one can infer that these quantities are in general apparently applicable only to the same case of the wave incidence in which they were retrieved. Therefore they do not fit the concept of the electromagnetic characterization.

This heuristic homogenization was however often claimed successful [15], and this claim has some reasonable grounds. Indeed, works based upon the aforementioned parameter retrieval often refer to a special class of MTM samples, composed of finite-thickness orthorhombic lattices of small resonant scatterers. These scatterers are not bianisotropic and at most frequencies (beyond the ranges of quadrupole and other multipole resonances) they can be adequately modelled by electric and magnetic point dipoles. The finite thickness lattice forms a layer (infinite in a plane and finite in the normal direction) with an integer number N of unit cells across it. Its scattering parameters are complex valued reflection R and transmission T coefficients. The retrieval of EMP from R and T coefficients of a layer of a continuous magneto-dielectric medium is the standard method (called NRW method), which is discussed below in a separate section of this review. In many papers it was found that EMP retrieved for this class of MTM by the NRW method do not depend on N, i.e. they remain unchanged for any layer thickness if divisible by the lattice period $d = Na$. One may conclude that these EMP are representative of the internal properties of the layer because they are independent of the layer thickness, and they characterize the material inside the layer. Formally, this is the case. For a special class of MTM the parameters ε and μ retrieved by the NRW method do indeed comply with the definition of EMP. This class of lattices was called in [11] the Bloch lattices. This name is related to the possibility to easily introduce the so-called Bloch impedance for these lattices. However, even for Bloch lattices these retrieved material parameters unfortunately do not fit the concept of the characteristic material parameters, because the locality limitations are violated for them. This is a clear indication, that the retrieved ε and μ do not fully fit the characterization concept. The non-local parameters obtained for the case of the normal wave incidence cannot be applied to other excitation cases. Even in the case of normal incidence they become inapplicable, if the same MTM layer is mounted on different substrates.

As it is evident now, the locality violation in the retrieved parameters was an indication, that the heuristic homogenization was flawed, and the prime attention should be paid to alternative (direct and inverse) homogenization models. However, this problem was not duly understood before 2006. The absence of locality in the retrieved parameters of Bloch lattices was interpreted in the numerous papers as a sign of strong spatial dispersion. Meanwhile these media as a rule behave as effectively continuous ones. Locality of the medium response can be easily checked through the frequency dependence of the retrieved refractive index n and wave impedance Z. The two locality limitations formulated above for permittivity and permeability can be also formulated for n and Z. Causality requires growth of the refractive index n with the frequency in the low-loss frequency regions (where the imaginary part of n is negligible). This is called the Foster theorem. Additionally, passivity requires the sign of the imaginary part of n and of

the real part of Z to be consistent with lossy medium, i.e. positive for the optical selection for the time dependence $\exp(-i\omega t)$ and negative for the engineer selection $\exp(j\omega t)$.

Inspecting many papers devoted to Bloch lattices of small separate (non-bianisotropic) scatterers, it was observed that within the frequency region $|q|a < 1$ locality is satisfied for n and Z and therefore strong spatial dispersion does not occur [10, 11, 12, 15]. Therefore, the non-local material parameters are retrieved because the simplistic inverse homogenization model [109, 110] fails. It was shown in [10, 11, 12, 15] that the failure happens because Maxwell's boundary conditions which are implied by this model can be insufficient for macroscopic fields. Maxwell's boundary conditions are equations of continuity for tangential components of fields $\mathbf{E}$ and $\mathbf{H}$ at interfaces of two different materials. For truly continuous media there is no difference between microscopic and macroscopic fields and these conditions are obviously satisfied. If the medium is formed by strongly dispersive (resonant) particles, these conditions are satisfied only for microscopic fields. For macroscopic ones it is not necessarily the case.

It is therefore clear that new, more advanced, inverse homogenization models for MTM are needed. Unfortunately, progress in the theoretical homogenization of MTM up to 2009-2010 has been very modest. Only a few works are known, which link microscopic theoretical models of nanostructured MTM with a possibility of experimental electromagnetic characterization. As already noticed, in order to find characteristic parameters of an artificial material, its theoretical model has to explicitly include boundary conditions. As a result, it should establish the relationships between material parameters and scattering parameters, which can be experimentally measured, e.g. R and T coefficients of a layer. When measured in the far-field zone, these two coefficients contain only macroscopic (averaged) information about the structure properties. The contribution of evanescent waves which contains microscopic information cannot be uniquely extracted from the far field, i.e., from the reflected and transmitted waves. Only for a special class of MTM – for the Bloch lattices – such incomplete information can be sufficient for their characterization [13]. However, it is not yet fully clear if it is really so even for these lattices, since the study of their electromagnetic properties for oblique incidence of waves has not yet been done.

4.4 Retrieval of material parameters for bulk nanostructures with non-resonant constitutive elements

As was mentioned above, the response of most isotropic bulk media to an incident plane wave can be described, in the long-wavelength limit, by a frequency dependent electrical permittivity, ε, and magnetic permeability, μ. These can be obtained using the scattering data from a finite slab of such material, considering the material as a homogeneous medium. This approach can be applied for natural and composite media, but fails in two cases. First, it fails when the wavelength becomes comparable to the material unit cell size (practically when the unit cell maximal size a becomes not optically small, for example when it exceeds $\lambda/4$. This situation corresponds to the frequencies close to the first band gap of the lattice, which in this case should be considered as a photonic crystal rather than as a composite medium. In our definition, photonic crystals do not refer to materials and consequently are not metamaterials. Second, the standard retrieval procedure fails for MTM at frequencies where the material is resonant.

Consider now the frequency ranges in which the composite material is not resonant. For non-resonant and optically dense ($a < \lambda/4$) composite media the standard approach to electromagnetic characterization can be used. This is so because a layer of such a medium in spite of its heterogeneous internal structure can be considered as being effectively continuous

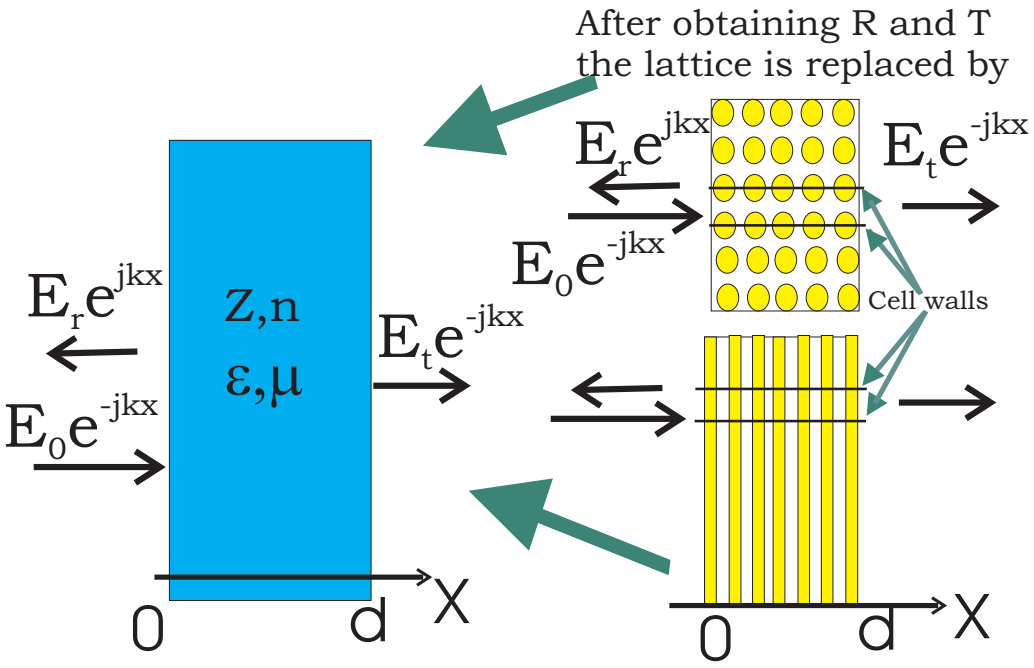

Figure 4: The configuration used in the NRW method for the determination of effective material parameters from reflection and transmission data for a non-resonant composite slab. The structure period across the slab is small. The structure can comprise inclusions which are small in all three dimensions (the upper one). Alternatively, inclusions can be extended in the plane parallel to the slab boundaries (the lower one). In the last case one can refer to a partial (conditional) electromagnetic characterization.

between its physical boundaries. There are two main methods of the experimental electromagnetic characterization of non-resonant composite media. One is the RT-retrieval method, also called the Nicholson-Ross-Weir (NRW) method. It is also often used for the theoretical electromagnetic characterization of nanostructured materials in numerous works (see discussion below). Another is the modern ellipsometric method called VASE which is probably not applicable to theoretical characterization of composite media, however, it is widely used as a powerful experimental method to characterize nanostructured materials (see e.g. in [56]–[64]). More information on this can be seen in the comprehensive review [15].

4.5 RT-retrieval method (Nicholson-Ross-Weir method)

The RT-retrieval refers to the normal incidence of the probing plane wave and is a quite simple procedure, since the transmission and reflection of a plane wave by a homogeneous medium are relatively simple functions of the refractive index, n, and the wave impedance, Z, of the effective medium, whose bulk material parameters can be in their turn found as $\varepsilon = n/Z$ and $\mu = nZ$. The NRW method principally operates by analyzing the transmission and reflection signals for the calculation of the dielectric and also magnetic properties of bulk layers. Its main advantage is that it can be applied in a wide frequency range providing a broadband description of the medium dispersive properties.

Considering the scattering configuration shown in Fig. 4, where an incident plane wave of the form $\mathbf{E}_{\text{inc}} = \mathbf{y}_0 E_{\text{inc}} \exp[j(\omega t - kx)]$ impinges normally on a slab of a homogeneous material in vacuum, one can find the refractive index n and the wave impedance Z through measured (or

numerically simulated) reflection R and transmission T coefficients of the slab using inverted Fresnel-Airy formulas:

$$\cos(\omega\sqrt{\varepsilon_0\mu_0}nd) = \frac{1-R^2+T^2}{2T}, \quad z = \pm\sqrt{\frac{(1+R)^2-T^2}{(1-R)^2-T^2}}. \tag{18}$$

Here the transmission coefficient refers to the back surface of the slab. There are many works on the proper choice of the inverse cosine branch for n and of the sign of the real part of Z. We do not discuss these details here.

The most important advantage of the NRW method for the theoretical electromagnetic characterization of nanostructured layers is related to the fact that these layers are periodic. This allows one to perform exact numerical simulations of R and T coefficients using commercially available software. The periodicity in the plane parallel to the layer interfaces yields the boundary problem with the plane wave incidence to the so-called cell problem. Then one can select a unit cell of the structure and ascribe to the cell walls so-called periodic boundary conditions. For a lattice of inclusions which is infinite along one direction, (as it is in the case of the wire medium, for example) the unit cell size can be chosen arbitrary. The periodical conditions at the walls of a unit cell allow one to consider only a single unit cell and to calculate the fields only inside it. In the infinite structure around the selected cell the same fields are periodically replicated. Even if the unit cell contains 10-20 particles, such a problem is not challenging for modern electromagnetic software. One calculates R and T, retrieves from them n and Z using (18) and finally finds ε and μ.

Let us conclude this subsection by two important comments. The NRW method implies the normal propagation of the wave in the material slab. Therefore, the obvious condition of optical smallness of the structure period a refers only to the period across the slab. To consider the medium as effectively continuous is possible not only if the slab is formed by optically small inclusions. It can be an array of long inclusions (e.g. wires), if they are parallel to the slab boundary. This possibility seems to broaden the scope of applicability for this method. However, it is not so simple. In fact, the retrieved effective material parameters can be treated as characteristic parameters only in the case when the particles are optically small and isotropic. If the inclusions are not small in the transverse direction, material parameters, retrieved from the measurements of R and T coefficients for the normal incidence, can be applied for condensed description of the materials only for this particular excitation. For a slab of a wire medium or for a structure of alternating metal and dielectric layers this simple approach is not applicable. Here the oblique propagation of waves obeys different laws than the normal propagation. The interaction of the obliquely propagating wave with such media cannot be considered in terms of the permittivity which was retrieved for the normal incidence. Moreover, in the case of the oblique propagation of the wave such media are spatially dispersive and to relate their effective material parameters to R and T coefficients so-called additional boundary conditions are needed (see e.g. in [82]). However, if there is a reliable theoretical model of the structure and a minimal knowledge on its geometrical parameters (e.g. the period a across the slab), the NRW retrieval will be not useless. For example, for a wire medium from the retrieved permittivity one can find the so-called plasma frequency which can be further used for calculating the spatially dispersive material parameters. This case can be referred as partial or conditional electromagnetic characterization.

The second comment refers to the obvious requirement of the absence of resonances. In fact, the NRW method is inapplicable in the frequency ranges of the Fabry-Pérot resonances

(thickness resonances of the slab) [14]. At these resonances the method, if applied, leads to the violation of physical laws in the retrieved material parameters.

4.6 Are the retrieved effective material parameters of nanostructured materials always representative of their characteristic parameters?

The retrieval of material parameters of an effective homogeneous medium evidently implies that we replace the original heterogeneous structure by an equivalent homogeneous medium. In what sense this equivalence can be understood? It is clear that the homogeneous model should imitate the electromagnetic properties of the original inhomogeneous material. But it is also clear that all electromagnetic properties cannot be imitated since the homogenization is always an approximation. So, which properties are important to imitate?

For samples which can have different shapes this is the equivalence of the overall scattering properties which is described in the electromagnetic theory by the so-called scattering matrices. Two bodies of the same shape (one of which is made of the original inhomogeneous material and the other one of the corresponding homogenized material) should have nearly identical scattering matrices. Fortunately, most nanostructured materials are prepared as layered structures and we can consider them as layers of finite thickness, whereas in the interface plane they are practically infinite. Then the equivalence of scattering matrices reduces to the equivalence of reflection and transmission coefficients for plane waves. However, the crucial moment is that this equivalence should hold for any incidence angle, since homogenous material is characterized by material parameters which are invariant with respect to the incident wave propagation direction. Moreover, material parameters of continuous media such as ε and μ obey the basic physical limitations of locality as explained above. If the retrieved effective material parameters turn out to be different for different angles of the wave incidence or if they violate the physical limitations then this homogeneous material model cannot be equivalent to the material under study.

The retrieved effective material parameters can be obtained wrongly due to several reasons. First, the structure can be spatially dispersive in the frequency range where the retrieval of material parameters is done. Many metamaterials behave as spatially dispersive (effectively discrete) structures within some narrow frequency interval(s) laying inside the resonance band of inclusions but outside this (these) narrow sub-band(s) they behave as continuous media. Then outside these regions it is possible to properly retrieve the effective material parameters which characterize the medium, but within them it is impossible and the retrieved parameters have no physical meaning. Another reason can be a wrong retrieval model. First, the researcher can try to characterize an anisotropic medium with tensor ε and μ by scalar ε and μ. Since the scattering by anisotropic layers obeys different law than the scattering by isotropic layers this approach delivers wrong scalar material parameters. Third, if the retrieval procedure which is valid only for natural materials or for composites of non-resonant constitutive elements is applied to arrays of resonant elements, it also delivers wrong material parameters. This situation is most difficult and it is discussed below in more detail.

Usually one retrieves effective material parameters for one angle of incidence and hopes that they are applicable to other angles. However, these hopes are not always grounded for MTM. Below we present a typical list of mistakes in retrieved material parameters for nanostructured MTM which can happen at some frequencies within the resonance band (or even over the whole

resonance band):

- Retrieved parameters depend on the sample size and on the surrounding environment (e.g. [91])
- Retrieved parameters have nonzero imaginary parts in the absence of real dissipation (e.g. [92])
- The sign of the imaginary part of one of retrieved material parameters is opposite to the sign of another one (e.g. [93])
- The frequency dispersion of retrieved material parameters violates the causality limitations (e.g. [94])

In all these cases the retrieved material parameters are wrong and therefore should be different for different incidence angles (see e.g. in [95]). Therefore they cannot be considered as electromagnetic characteristic parameters.

5 State-of-the art of the retrieval of material parameters for metamaterials in 2008-2009 years

The modern history of MTM starts from paper [100] where the goal to create the so-called *perfect lens* was claimed by J.B. Pendry. The development of MTM showed that composite media with extraordinary material properties are suitable not only for subwavelength focusing and resolution and they found a lot of other applications. Simultaneously the concept of MTM was generalized. Now, transmission line networks with periodical loads and resonant artificial surfaces (metasurfaces) also refer to MTM [96, 97, 98]. MTM definitely deserves the special attention paid to them in the modern literature. Early surveys on the electromagnetic characterization of metamaterials (MTM) can be found in books [96, 97, 98] (also see review paper [99]). However, in many papers devoted to MTM the reader can find some misinterpretations, as often happens when a research field is developing very quickly. Especially this concerns electromagnetic characterization of MTM and interpretation of results obtained for effective material parameters.

In a focused literature review [81] it was shown that in more than 50% of papers on MTM published in leading physical journals effective material parameters experimentally retrieved for MTM violated basic physical laws in their frequency dependencies and in their values. These non-physical results became possible because the retrieval methods did not take into account the peculiarities of MTM.

Recall that MTM are usually composed by arrays of resonant scatterers whose characteristic size δ and period a at the resonance are though smaller than the wavelength in the host medium λ but comparable with it (practically $(a, \delta)/\lambda = 0.05...0.2$). The resonance of lattice particles happens at these comparatively low frequencies and this distinguishes MTM lattices from photonic crystals. However, this resonance also differentiates MTM from previously known artificial magneto-dielectrics and enables unusual properties. Moreover, MTM can be also a combination of two (or more) periodic building blocks. One of them can be formed by small magnetic scatterers often in form of split-ring resonators (see e.g. [101, 102]), another can be an array of long wires [103]. Some interesting phenomena in MTM arise due to spatial dispersion (e.g. in aforementioned wire media when the wave propagates obliquely to the wires or even

along them). These peculiarities of MTM should have been taken into account in the procedures of their electromagnetic characterization. Though this evident point discussed spontaneously in some conferences already in 2002-2003, in what concerns journal articles, this was noticed only in work [10] published in 2007. In 2000-2008 the most part of the literature devoted to the electromagnetic characterization of MTM practically ignored specific properties of these materials.

In this section we will discuss five methods of theoretical characterization of MTM (including nanostructured MTM) found in the literature and discuss the value of their outcomes. We will conclude this section with some recommendations for future research.

5.1 Five procedures for theoretical electromagnetic characterization of metamaterials

In the literature published in 2000-2009 one finds five main procedures of the theoretical electromagnetic characterization of MTM:

- Procedure 1. Effective material parameters are obtained by a direct RT-extraction of ε and μ from plane-wave reflection and transmission ($R-T$) coefficients of a composite slab, assuming the slab to be continuous and uniform medium. In other words, this is the theoretical NRW retrieval.

- Procedure 2. Effective material parameters are obtained by an extraction of ε and μ from plane-wave reflection and transmission ($R-T$) coefficients of a composite slab, assuming the slab to be a 3-layer structure. The central layer is characterized by ε_L and μ_L of the bulk medium which obey the Lorentz dispersion laws and therefore respect the aforementioned locality limitations. Two other layers (optically very thin) are located at the interfaces of the original slab and characterized by other effective material parameters (which correspond to surface materials).

- Procedure 3. Effective material parameters are introduced through sophisticated combination of line and surface averaging procedures for different vectors of the electromagnetic field. Line averaging is applied to define macroscopic vectors $\mathbf{E}$ and $\mathbf{H}$ through true (microscopic) fields. Surface averaging is applied to define macroscopic vectors $\mathbf{D}$ and $\mathbf{B}$. Effective material parameters can be obtained from an approximate model of the lattice unit cell or from exact simulations of the infinite lattice. These parameters are not retrieved from $R-T$ coefficients of the layer.

- Procedure 4. Effective material parameters are obtained from exact simulations of the electromagnetic wave propagation in the infinite lattice. They cannot be directly retrieved from $R-T$ coefficients of the layer, however can be obtained for solving boundary problems if complemented by additional boundary conditions.

- Procedure 5. Effective material parameters for thin MTM layers (1-3 scatterers across the layer) describe the electromagnetic response of the layer per unit area of the surface (i.e. the response over the whole layer thickness) correspond to the bulk response of the layer however the microscopic responses of elements as well as microscopic fields are averaged over the layer thickness. These material parameters combine the surface and bulk approaches and can be also retrieved from $R-T$ coefficients of a composite layer.

5.2 Discussion of Procedure 1: NRW retrieval for metamaterials

Procedure 1 was first applied for MTM layers in papers [109] and [110], and later in hundreds of papers (it is also described in all above cited books devoted to MTM). It is the most extensively used method which is considered as most successful. In fact, this is nothing but the standard procedure previously known as Nicolson-Ross-Weir (NWR) method of the extraction of material parameters of continuous magneto-dielectric media from the measured S-parameters. It was first suggested for retrieval of transmission-line characteristic impedance and refraction index in [125] and then developed for the characterization of layers of natural media in [126], [127] (dielectrics or magnetics) and in [128] (magneto-dielectrics). The NRW algorithm is based on the material Maxwell equations (implying the local permittivity and permeability) and Maxwell's boundary conditions. Applying to MTM we encounter the problem of non-locality of the slab electromagnetic response. This non-locality is not the same as the strong spatial dispersion in the infinite lattice. It results from the combination of the discreteness and finiteness of the structure and leads to the violation of Maxwell's boundary conditions for macroscopic fields. Mathematically, it is expressed in the difference between the so-called Bloch impedance that describes the reflection from the original lattice and the wave impedance of the homogenized medium that would describe this reflection in the absence of this boundary non-locality. These two impedances are equal one to another only in the static limit [11].

In [111]–[118] and many other works the authors claim that a composite slabs comprising a small number N of monolayers[1] and even a single monolayer $N = 1$ has the same effective material parameters as infinite or semi-infinite MTM lattices $N \to \infty$. It is considered in the literature (a detailed survey is given in the next section) as a proof that this indirect homogenization is correct. In other words, the indirect homogenization through extraction of material parameters through simulated or measured R and T is considered in the dominating literature on MTM as a correct procedure of homogenization **because** it gives the same result for $N = 1$ and for $N \to \infty$. However, a simple analysis shows that namely this fact means that it is an **incomplete** procedure.

It is clear that in a layer with $N > 1$ grids of point dipoles when the distance a between the grids is not optically negligible, the obliquely incident wave refracts. On the contrary, in the case when $N = 1$ the grid of electric and magnetic dipoles (optical thickness is negligible) does not refract the wave (it is commonly known that the interaction of the wave with any planar grid leads to the non-refractive reflection and transmission of waves). If the host medium in the monolayer does not differ from the media in front of and behind it (i.e. if the monolayer is the grid in free space layer of thickness a) the obliquely incident wave does not refract in it. But it definitely refracts inside the layer of $N \gg 1$ such grids separated by distance a. This is so, in spite of same material parameters attributed to these two different layers. Thus, the set of effective material parameters which are unique for layers with $N = 1, 2...\infty$ have little to do with the refraction of waves. It is clear from this discussion that the physical meaning of these effective material parameters is special and very different from that of Lorentzian effective material parameters[2]. Therefore these effective material parameters cannot be considered as physically fully sound.

Finally, let us list all the reasons which in our opinion restrict the NRW retrieval for MTM:

[1]Monolayer is a single grid of particles placed in the host medium slab of thickness a which is equal to the period of the MTM lattice obtained by the periodic repetition of the monolayer in the normal direction to its interface.

[2]Which evidently have no physical meaning for a single monolayer.

- An improper choice of the material equations. Not all media can be described by scalar ε and μ. The possible anisotropy, bianisotropy and non-reciprocal components of the tensorial material parameters (gyrotropy) should be taken into account.

- An improper choice of the material type. For example surface MTM should not be modelled as bulk ones.

- Strong spatial dispersion. It is observed when there are several reflected waves (more exactly several scattered waves together with the reflected one). It is also present when the dispersion of the wave propagating in the medium strongly differs from the dispersion in continuous media. Namely, it happens when the isofrequency contours strongly differ from elliptical or hyperbolic. No local material parameters can be introduced in this case. When one introduces effective material parameters for such media they may have only qualitative (illustrative) meaning.

- Ignoring the asymmetry of the internal sample geometry. This is a kind of weak spatial dispersion effect. For example, a slab of MTM may not have a plane of symmetry. As a consequence the transmission coefficients for the waves incident from the right to the left and from the left to the right may be different whereas for the homogeneous slab these coefficients are identical even for an anisotropic material of the slab. The problem can be fixed by describing a MTM slab as a layer of two or three media with different material parameters.

- Strong excitation of particles located near the interface. This results in the medium polarization which does not obey to the law of refraction. In other words the macroscopic field inside the material is not just one single spatial harmonic.

- The surface non-locality of resonant materials. It is discussed below in details. This is the most often effect for MTM of small scatterers. Below it is explained how this problem can be resolved.

5.3 Discussion of Procedure 2: Characterization of MTM by local material parameters

Procedure 2 was developed in 2007-2008 in works [10, 11, 12, 122]. It allows to find effective material parameters of MTM whose frequency dispersion obeys the Lorentz resonance law and therefore respects the locality limitations. In these works it was shown that the new retrieval procedure was a generalization of the well-known NRW retrieval to the resonant case. The procedure was developed only for lattices of reciprocal (without elements of natural magnetic media) not magnetoelectric inclusions. The restrictions of applicability (frequency bounds) are the same as for Bloch material parameters. Local effective material parameters keep the physical meaning of the medium unit volume response to the electric and magnetic fields even in the resonance band of inclusions [10, 11, 12], except special frequencies where the effects of strong spatial dispersion are essential.

The introduction of Lorentz effective material parameters for lattices of resonant inclusions implies a more complicated three-layer representation of any finite-thickness MTM lattice: The inner layer with Lorentzian effective material parameters of the infinite lattice and two thin Drude layers at the two interfaces. This approach encounters a serious difficulty: In

general, the effective material parameters of Drude layers remained unknown. The transition layer properties were not retrieved in [10, 11, 12, 122]. This was done only in 2009 in [13]. The retrieval procedure has been developed in [10, 11, 12, 122, 13] quite weakly since only the normal incidence and only isotropic inclusions were considered. No proof that the same retrieved material parameters can be used to calculate the scattering matrix of the layer for other incidence angles was presented. Only in work [123] such a proof was first presented for surface MTM.

5.4 Discussion of Procedure 3: Flux-averaging homogenization

Here we briefly discuss Procedure 3 introduced and developed by J.B. Pendry and co-authors in [101, 104, 105]. Material parameters related to such a unusual averaging procedure were obtained as a mathematically intermediate result in the modelling of the transfer matrix of the unit cell of an arbitrary lattice of inclusions. These effective material parameters are helpful in semi-analytical calculations of eigenwaves in any infinite lattice, however we did not find any papers where these parameters would be used for anything else.

Though one can find many references to this method in the modern literature on MTM, this fact is sooner related to the name of J.B. Pendry than to the usefulness of his homogenization model. We know no one example of successful retrieval of these effective material parameters from measurements. One can find only direct theoretical calculations of these effective material parameters mainly with the purpose to demonstrate that they can be negative.

Physical meaning of these effective material parameters remains unclear even after a detailed discussion of their properties in [105]. Apparently, these material parameters violate the locality limitations even in the absence of spatial dispersion. At this stage it is not clear how to link these effective material parameters to $R-T$ coefficients of finite-thickness layers, beyond the static limit (where all five aforementioned procedures give the same result).

5.5 Discussion of Procedure 4: The non-local homogenization for MTM with weak spatial dispersion

Procedure 4 developed in works [106, 107, 108] is very accurate and allows one to take into account fine effects. The obtained material parameters are useful also for finite-thickness layers. These effective material parameters are extracted from simulations of an infinite lattice and are tensors describing the electric and magnetic response of the unit cell to the electric and magnetic fields averaged in a special way. One can show (though it is not explained in papers [106, 107, 108]) that this averaging is sharing out the fundamental Bloch mode of microscopic fields and polarizations. These effective material parameters depend obviously on the wave vector $\mathbf{q}$ at all nonzero frequencies, however this dependence is (in frequency regions where the structure behaves as an effectively continuous material) comparatively weak. It corresponds to the so-called weak spatial dispersion. In the static limit nonlocal effective material parameters [106, 107, 108] converge to static material parameters of the lattice.

At low frequencies where strong spatial dispersion in the infinite lattice is absent the $\mathbf{q}$-dependence of material parameters describes the presence of decaying (evanescent) eigenwaves of the lattice. They are lost in other known homogenization procedures. However, near the interface of the lattice these attenuating waves exist and can strongly influence the reflection and transmission phases. These waves are often called *polaritons*. It spite of their exponential decay

inside the lattice, polaritons can be not surface waves (the exception are so-called plasmon-polaritons). They can be excited by incident plane waves and their tangential wave number q_t is mainly determined by the lattice period and weakly influenced the projection of the incidence wave vector to the interface.

In the theory of diffraction gratings polaritons induced in the lattice crystal planes are called the *higher-order Floquet modes* of the grating.

To apply such non-local effective material parameters to the boundary problems with MTM layers (i.e. beyond the static limit) one has to deduce so-called additional boundary conditions (ABC) relating macroscopic fields at two sides of the interface. These ABC should refer to a properly chosen interface plane which is not obviously the same as the physical surface of the MTM slab. The successful choice of the interface is crucial for the whole method. We conclude that this method is fine for solving direct electrodynamic problem. However, it can be hardly applied to the experimental characterization of MTM lattices. Really, we do not know a priori which ABC we should apply and where the interface plane should be located. Moreover, these effective material parameters are assumed by definition to be non-local, i.e., they depend on the wave vector at all frequencies, which makes their experimental retrieval hardly possible. As to theoretical characterization of MTM lattices, it is more than justified for structures strongly influenced by polaritons, but is too complicated for many practical MTM.

5.6 Discussion of Procedure 5: The combination of bulk and surface homogenization models

Procedure 5 was introduced in [124] as an alternative to Procedure 2 for very thin layers comprising $N = 1 - 3$ unit cells across the layer. Here the bulk effective material parameters lose their physical meaning since the concept of the macroscopic field is not grounded. The effective material parameters defined by Procedure 4 were called *mesoscopic effective material parameters* since they evidently depend on the number of layers N. Since these effective material parameters really describe the layer unit cell electromagnetic response, they satisfy the locality requirements as well as Lorentzian bulk effective material parameters.

However, except the case of very simple inclusions (unloaded wires or patches), mesoscopic effective material parameters defined through the electric and magnetic response per unit area of the slab do not fit the $R - T$ coefficients and are of little practical use. Therefore, the definitions of mesoscopic effective material parameters from [124] have to be revised.

5.7 Conclusions from the discussions

In conclusion, there are five most known methods of the characterization of bulk MTM lattices of finite thickness through effective material parameters, from which three procedures are clearly related to interface phenomena. One of them, i.e. Procedure 1 is in fact the well-known NRW method applied to MTM. It is most popular, however, it is prone to improper applications and wrong interpretations. Procedure 3 is not applicable to boundary problems and therefore we cannot recommend it for the electromagnetic characterization of practical materials. Procedure 5 is recommended, however, only for specific MTM (strongly influenced by polaritons) and only for theoretical characterization. Two other procedures (numbered as 2nd and 4th ones) are not popular and therefore have been weakly developed. It is impossible up to now to judge whether it is related to their inherent shortcomings or simply concerned with their weak development. In this situation it is impossible to definitely make the choice of the best method for the

characterization of bulk MTM layers judging upon the publications up to 2009. However, it is obvious that it is necessary to promote the existing insights in the physical meaning of extracted material parameters, since misinterpreted results in the literature on MTM are, as a rule, related to incomplete theoretical knowledge. The key issue for future research is to find a simple and adequate description of interfaces and develop a model which would be free from non-physical artifacts in the retrieved bulk material parameters. Most likely, an appropriate model of interfaces will be a key for the adequate extraction of bulk parameters.

6 A breakthrough in the electromagnetic characterization of nanostructured materials is hopefully coming

6.1 Bulk nanostructured metamaterials. State-of-the-art in 2010

During the last year the situation has dramatically changed. A new trend in the literature devoted to the bulk material parameters of MTM can be easily noticed if we inspect papers published in 2010. This trend has the following features:

- Scientific groups which have gained extremely high reputation over the world by their theoretical and experimental works (Profs. A. Alu, C. Holloway, Yu. Kivshar, E. Kuester, F. Lederer, G. Shvets, and others) have suggested new homogenization models especially addressed to nanostructured MTM [23, 30, 16, 31, 32, 33, 34, 35, 36, 37];

- In these papers the theoretical models have been linked to possible experiments, in other words, algorithms of possible experimental retrieval of effective material parameters 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 effective material parameters 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. [30]) results which had been previously obtained in the field of electromagnetic characterization of nanostructured MTM have been criticized independently from the analytical critisism of the ECONAM overviews and conference presentations. It has been shown that in many cases previously retrieved material parameters of nanostructured MTM 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 [16, 31, 27, 32] the physical effect which makes the NRW method not applicable to MTM (which had been earlier pointed only in works by Simovski and Tretyakov, e.g. [10, 11, 12, 39]) has been also noticed and widely discussed. This effect is a jump of tangential components of bulk macroscopic fields at the interface of the resonant material. This jump leads to a significant difference between the surface impedance of the material and its wave impedance[3].

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

Some researchers in this situation suggested novel algorithms of the retrieval of characteristic material parameters which are not related to the simulation or measurement of R and T coefficients. Here the term characteristic material parameters is used instead of EMP in order to stress that these effective material parameters pretend to fit the concept of the electromagnetic characterization (see above). For example, methods suggested in works [23, 32, 33] refer to the measurements or simulations of the fields inside the metamaterial sample. For example, the wave propagation retrieval method [32, 33] seems to be perfect for the theoretical calculation of characteristic materila parameters of MTM, even chiral ones [33], which require an additional tensor material parameter for the description of their bulk electromagnetic properties. However, the problem appears with the applicability of these characteristic materila parameters to the practical boundary problems since the properties of the surface are not taken into account by this method. In fact, the method delivers only characteristic materila parameters of an infinite lattice, similarly to the earlier results in [10, 11]. The set of characteristic parameters retrieved in this way is therefore still not complete and the method in its present form is hardly appropriate for the experimental characterization of nanostructured MTM. A similar drawback can be noticed if we consider the method suggested in [23]. 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 not fully clear. Another important step forward was done by A. Alu in [36, 37], where an additional volumetric material parameter is used to remove the non-physical anti-resonances in retrieved permittivity and permeability. Unfortunately, at this stage this approach does not give a reasonably simple scheme for predicting properties of finite-size samples. Despite some still "missing links", all these recent works are very important as a clear indication of the constructive trend in the literature and hopefully these new methods will be further developed by their authors.

In recent works [27, 34] the model of the dynamic homogenization of the infinite lattice of electric and magnetic dipoles suggested (to our knowledge) in work by Simovski [38] and applied for the theoretical electromagnetic characterization of MTM in his further works [10, 11, 12, 13] 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 [35] A. Alu suggested a modification of the model [38] which allows one to fully remove the small deviation of the lattice material parameters from locality. This deviation occurs in the model [38] in a very narrow frequency range in lossless arrays near the lower edge of the resonance band. In works [10, 11, 12] this effect was practically eliminated by introduction of small losses. Though recent works [27, 34, 35] do not consider surface effects and the retrieved effective material parameters are not enough to solve boundary problems, they evidence that the theorists have started to understand the importance of the dynamic inter-particle interaction for the proper characterization of MTM.

In work [16] 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 some 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 needs further development compared to [16]. In this paper the authors could not satisfy the locality limitations in their retrieval procedure. A possible reason of this 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 [16] there is no link to the problem of the oblique incidence which should show that the retrieved effective material parameters are

really characteristic materila parameters of this metamaterial.

We consider the apparition of all these cited works within this recent period as a key prerequisite of a real breakthrough in the electromagnetic characterization of bulk MTM which we expect in the near future. We hope that the ECONAM activity has played some role in this positive trend.

6.2 Surface nanostructured metamaterials. State-of-the-art in 2010

A progress in this direction is still modest, and, to our knowledge, it is related only to the study by Simovski and Morits performed in summer of 2010 [39]. Using an example of a bilayer of plasmonic nanospheres this work generalized the method of electromagnetic characterization of monolayer magneto-dielectric metasurfaces (metafilms) suggested in [28, 29]. It has been theoretically demonstrated that the results of this characterization method are suitable for predicting scattering parameters of bilayer metasurfaces. Since the authors considered different angles of incidence and theoretically demonstrated that the retrieved material parameters are applicable for all of them, this is an important step in the characterization of metasurfaces.

However, an important theoretical problem is not yet resolved. The approach [28, 29] 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. Generalization of the approach to the realistic case when the particles are located at a dielectric interface is not an easy task. However, there is a strong need in such a theory and we expect in the near future a breakthrough also in this area.

7 Conclusions

In this overview we have summarized the available information on the electromagnetic characterization of nanostructured materials collected in our preceding reports [78, 79, 80, 81] and complemented it by explanations of he most important electromagnetic concepts similarly to that in [82]. In this report we have presented the information on the theoretical (analytical and numerical) characterization techniques for nanostructured materials in the form accessible for non-specialists in the electromagnetic characterization of materials. The concepts important for the electromagnetic characterization of nanostructures are not very well developed yet. Some of them are new and under development, which hopefully made our work useful also for specialists. This refers to the most important case when the nanostructured materials demonstrated useful and unusual (absent in natural media) properties, in other words, when they are metamaterials.

The analysis of very recent (2009-2010) literature on MTM allows us to conclude that the vector which shows the knowledge development has changed the direction. From the depression, i.e., accumulation of often waste data obtained with the use of obsolete and inadequate homogenization models (see the overview for 2008-2009) it turned to the development, i.e to the search of adequate, properly working theoretical models as enablers for proper interpretations of measured results. This new trend allows us to expect an approaching breakthroughs in the electromagnetic characterization of nanostructured MTM.

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