On effective electromagnetic parameters of artificial nanostructured magnetic materials

C.R. Simovski and S.A. Tretyakov
Department of Radio Science and Engineering / SMARAD Center of Excellence, Aalto University, School of Science and Technology, 00076, Aalto, Finland

Abstract
In this paper we discuss effective material parameter description of new nanostructures designed to perform as artificial magnetic materials for visible light. Among these structures there are various split-ring resonators, dual-bar structures, fishnet layers and other geometries. Artificial magnetic response in these structures appears due to weak spatial dispersion effects, and it is important to study the conditions under which the magnetic response can be adequately measured with effective permeability tensors. On the examples of dual bars and split rings we show that this is possible only under some quite restrictive conditions. In the general case, more complicated constitutive relations with more effective material parameters need to be developed.

Keywords: metamaterial, electromagnetic characterization, effective material parameters, permeability, spatial dispersion, multipole media, bianisotropic media

1. Introduction

Conventionally, the electromagnetic properties of bulk materials are described by their permittivity ($\varepsilon$) and permeability ($\mu$) tensors. There are no means for the direct measurement of these electromagnetic parameters, which are for this reason dependent on the model used for their extraction from the measurement data. Approaches to measurements of even these fundamental electromagnetic characteristics of materials vary dramatically for electromagnetic waves of different frequencies, e.g. radio waves and light, and require
specialized measurement techniques. These difficulties are dramatically escalated in electromagnetic characterization and metrology of nanostructured materials with complex geometries of unit cells and exotic electromagnetic response. For example, let us consider a device that operates at the wavelength of 500-600 nm, with the structural periodicity of artificial material in the range of 50-100 nm, while the sizes of resonant inclusions are about 30-70 nm. Such structures, if they are bulk (3D) lattices, can still be described in terms of effective parameters (permittivity and permeability) of an equivalent homogeneous medium. However, the conventional models based on quasi-static homogenization procedures normally applied at the atomic level are not applicable here, and the resulting effective phenomenological parameters (like permittivity and permeability) often have quite different physical meaning as compared with conventional materials – natural ones and composites of non-resonant inclusions.

The bulk material parameters $\varepsilon$ and $\mu$ of an effective medium (isotropic or anisotropic) are classically defined so that they relate vectors $\mathbf{D}$ and $\mathbf{B}$ of the electromagnetic field in a medium with macroscopic (averaged) vectors $\langle \mathbf{E} \rangle$ and $\langle \mathbf{H} \rangle$, respectively:

$$
\mathbf{D} = \varepsilon_0 \langle \mathbf{E} \rangle + \mathbf{P} \equiv \varepsilon_0 \varepsilon \cdot \langle \mathbf{E} \rangle, \quad \mathbf{B} = \mu_0 \langle \mathbf{H} \rangle + \mathbf{M} \equiv \mu_0 \mu \cdot \langle \mathbf{H} \rangle
$$

Here $\mathbf{P}$ and $\mathbf{M}$ are the bulk electric and magnetic polarizations resulting from microscopic electric and magnetic dipole moments of particles averaged over the same volume as the electric and magnetic fields. For regular arrays of atoms, molecules or inclusions this is usually the volume $V$ of the lattice unit cell. Parameters defined by formulas (1) are applicable for solving boundary problems with the Maxwell boundary conditions [1, 2, 3].

Physically sound local material parameters should satisfy the following basic requirements:

- **Passivity** (for the temporal dependence $e^{-i\omega t}$ it implies $\text{Im}(\varepsilon) > 0$ and $\text{Im}(\mu) > 0$ simultaneously at all frequencies; for the $e^{i\omega t}$ time dependence convention the sign of both $\text{Im}(\varepsilon)$ and $\text{Im}(\mu)$ should be negative). Violation of this condition in passive media (no sources of electromagnetic energy at frequency $\omega$) means the violation of the second law of thermodynamics;

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

- Absence of radiation losses in electrically dense arrays with uniform distribution of particles. This means that in lossless arrays the electromagnetic parameters should take real values.

The first two requirements (passivity and causality) are most known and can be mathematically expressed through the Kramers-Kronig relations (see e.g. in [1, 2, 4]). These basic physical requirements must be satisfied to ensure that the use of the effective medium description does not lead to nonphysical results. Furthermore, locality of the model implies that the parameters

- are independent of the spatial distribution of fields excited in the material sample,
- are independent of the geometrical size and shape of the sample.

This ensures independence of material parameters on the wave vector \( \mathbf{q} \), if one uses a plane wave as a probe field to determine the material parameters. For a given frequency this means independence of effective parameters on the wave propagation direction [4]. These two requirements are difficult to satisfy for many nanostructures, in part because the samples usually contain only a few (or even one) layers of inclusions or patterned surfaces across the sample thickness. This introduces limitations on the applicability area of the effective parameters and demands the use of alternative descriptions in terms of surface impedance or grid impedance. If the last two conditions are not satisfied, the material parameters can be used basically only for the same excitation environment as in the characterization experiment.

Recently, considerable efforts have been devoted to the design and realization of nanostructures behaving as artificial magnetic materials at terahertz and optical frequencies, with the main motivation to create double-negative materials (e.g., [5, 6, 7]). These structures are usually characterized using a probe plane electromagnetic wave normally incident at planar samples. Then, the classical Nicolson-Ross-Weir method [8, 9] is used to extract the effective permittivity and permeability of the sample. However, in many situations one can observe that the resulting parameters do not satisfy the basic physical requirements of local material parameters (parameters have nonphysical signs of the imaginary parts in some frequency regions, do not
satisfy the Foster theorem (e.g., [10]) in low-loss regions, depend on the incidence angle of the probing wave, etc.) In a recent paper [11] it was shown that for typical optical fishnet structures the effective medium description in terms of permeability is not valid in the whole range where the backward-wave regime is observed. While these parameters still correctly restore the reflection and transmission coefficients for plane waves which were used in the characterization, they fail to describe the material for other excitations and sample shapes. This dramatically reduces their value in the design of applications which would utilize the effective magnetic properties of the new materials. Thus, it is of scientific and practical importance to understand the limitations of the effective parameter models and study the reasons for nonphysical behavior of extracted parameters. In this paper we make a step in this direction analyzing magnetic response of some simple inclusion geometries, conventionally used in the design of artificial magnetics.

2. Effective medium description of bulk optically dense metamaterials

In this paper we concentrate on effective-parameter characterization of optically dense bulk nanocomposites formed by electrically small scatterers. Even in this case the homogenization is a difficult task that obviously implies answering to the following questions:

- How to introduce (define) material parameters of composite media, what is the physical meaning of them and in which electrodynamic problems such material parameters are applicable?

- What are frequency bounds in which these material parameters keep their physical meaning and applicability?

- What are the physical limitations that should be imposed on these material parameters and can be used as a check of calculations, measurements, and, finally, in practical applications?

Frequency dispersion in metamaterials formed by electrically (optically) small inclusions can be strong when particles are excited in a vicinity of their resonant frequencies. This implies dramatic shortening of the wavelength inside the composite, which means that spatial dispersion effects can appear even in optically dense metamaterials. If the effective wavelength $\lambda/n_{\text{eff}}$, 


where \( n_{\text{eff}} \) is the effective refraction index, is close to or smaller than the lattice period, the local material parameters cannot be introduced. This situation can be easily detected experimentally or numerically, because in this case the nonlocality in response is visible also in the properties of the effective refraction index \( n_{\text{eff}} \) and wave impedance \( Z_{\text{eff}} \). In the following, we assume that the lattice period is electrically small, and concentrate on the analysis of the influence of the inclusion shape on the effective properties.

2.1. Artificial magnetism in composites of nanodimers

It has been known since early fifties \([12]\) that composites containing electrically small conductive inclusions of complex shapes can exhibit properties of artificial magnetics, usually described by magnetic polarizability of inclusions \( \alpha_{\text{mm}} \). This parameter is defined as

\[
m = \alpha_{\text{mm}} \cdot H
\]

where \( m \) is the induced magnetic moment and \( H \) is the local magnetic field at the center of the particle. After appropriate averaging, magnetic response of individual inclusions defines the effective permeability of the composite. This magnetic response is a manifestation of spatial dispersion effects, because the response to magnetic field basically means the response to the nonuniform component of the electric field (in form of \( \nabla \times E \)) \([13, 14]\). The key pre-requisite for the validity of the effective permeability model is that the response to other combinations of spatial derivatives of \( E \) can be neglected \([13, 16]\) (see also Chapter 2-1 of \([17]\)). Also, electric quadrupole as well as other higher-order polarization moments should be negligibly small as compared with the magnetic dipole \([16]\). This very much depends on the inclusion shape. In fact, only for some specific shapes these conditions can be satisfied.

Let us, for example, consider arrays of dual plasmonic nanoparticles (dimers): dual spheres, dual bars or other nanostructures in which the resonant magnetic response of a unit cell is related with the phase shift of the exciting wave over the distance \( a \) between two plasmonic elements. In one of the resonant modes of such pairs the electric polarizations induced in two particles are out of phase, which corresponds to high magnetic moment of the pair. Usually, composites formed by many such inclusions are modelled by effective permeability and permittivity, as in \([18–25]\) and a number of other papers. However, we will see from the following that in this case the
Figure 1: (Color online) A unit cell of a lattice of pairs of plasmonic spheres. If the wave propagates along $x$, the magnetic field induces a resonant magnetic moment (and an electric quadrupole moment). If the wave propagates along $y$, the same magnetic field at the same frequency induces only an octupole moment, which is negligible for optically dense lattices. Such a lattice cannot be described only in terms of $\varepsilon$ and $\mu$.

physical meaning of the permeability introduced via averaging of the induced magnetic moments is different from the conventional definition, which limits the possible use of this effective parameter.

Fig. 1 represents a plasmonic nanodimer. Let this nanopair be a unit cell of a nanostructured metamaterial. The magnetic moment of the medium unit cell of volume $V$ is defined as [2, 4]

$$m = \frac{1}{2} \int_V \mathbf{J} \times \mathbf{r} \, dV$$  \hspace{1cm} (3)

Here $\mathbf{r}$ is the radius-vector referred to the particle center, and $\mathbf{J} = -i\omega\varepsilon_0(\varepsilon - \varepsilon_h)\mathbf{E}$ is the polarization current density (in our case that inside the plasmonic spheres). Metal nanospheres are non-magnetic, and their permittivity is denoted as $\varepsilon$. $\varepsilon_h$ is the permittivity of the host medium. The response of the pair of nanospheres to the time-varying magnetic field is in fact the response of the nanopair to spatially varying electric field [13, 14]. When the local electric field $\mathbf{E}$ acting on different nanospheres is different, the dimer can acquire magnetic moment. Since for the centers of two nanospheres the radius vector is equal to $\mathbf{r} = \pm(a/2)\mathbf{x}_0$, magnetic moment $m$ of the nanopair given by (3) is always $z$-directed and for any direction of the wave propagation equals to $m = m_z = -(i\omega a/2)[p_y(B) - p_y(A)]$. The action of the local magnetic field $\mathbf{H}$ to the nanopair is in this way represented as action of electric fields $\mathbf{E}(A)$ and $\mathbf{E}(B)$ to nanospheres A and B.
To quantify the magnetic effect, we assume that the dimer is probed by a pair of plane waves that form a standing wave. This allows us to position the dimer center at the point where the incident electric field is zero and this way find the response to quasi-uniform magnetic field [13, 14]. If the exciting waves propagate along $x$ ($q = q_x$ in Fig. 1), the local electric field is $y$-polarized, $E(B) = -E(A)$, and $p(B) = -p(A)$. Respectively, $m = -i\omega p(B)/2$. The local magnetic field $H = H_z$ at the nanopair center is related to the electric fields $E(A)$ and $E(B)$ through Maxwell’s equations:

$$i\omega \mu_0 H_z = \frac{\partial E_y}{\partial x} - \frac{\partial E_x}{\partial y} = \frac{\partial E_y}{\partial x} \approx \frac{E(B) - E(A)}{a} = \frac{2E(B)}{a}$$  \hspace{1cm} (4)

In (4) we have taken into account the small optical size of the nanopair ($|k|a < 1$). The nanopair magnetic polarizability $\alpha_{mm} = \alpha_{zz}^{zz}$ is then equal to

$$\alpha_{mm} \equiv \frac{m}{H} = Z_0(ka)^2 \frac{p_B}{E(B)} = Z_0(ka)^2 \alpha$$ \hspace{1cm} (5)

where $Z_0$ is the free-space wave impedance and $\alpha = E(B)/p(B) = E(A)/p(A)$ is the electric dipole polarizability of the individual plasmonic particle. The magnetic polarizability in this special case of propagation turns out to be resonant at the same frequency as the plasmon resonance of an individual particle. Actually, the magnetic resonance is slightly red-shifted with respect to that of the single sphere (our simplistic model does not take into account the mutual coupling of spheres), but it is not important for our purposes.

On the other hand, if the probing waves propagate along $y$ ($q = q_y$ in Fig. 1), the response of the nanopair to the same magnetic field $H$ at the dimer center corresponds to zero induced magnetic moment. In this case we have

$$i\omega \mu_0 H_z = \frac{\partial E_y}{\partial x} - \frac{\partial E_x}{\partial y} = -\frac{\partial E_y}{\partial y}$$ \hspace{1cm} (6)

The magnetic moment of the unit cell induced by the magnetic field is zero because $E_x$ is zero at the centers of nanoparticles and their electric dipole moments vanish. Magnetic field $H$ induces opposite electric polarizations of the upper and lower halves of both spheres as it is shown in Fig. 1. This effect is practically negligible and has nothing to do with magnetic polarization, i.e., in this case $\alpha_{mm} = 0$.

We observe a dramatic dependence of the magnetic polarizability $\alpha_{mm}$ on the direction along which the incident field is changing. For example, in paper [25] this effect is treated as that of strong spatial dispersion, resulting
in both permittivity and permeability strongly dependent on the propagation direction. However, the optical size of the dimers and the lattice period are assumed to be optically very small, and there are no physical reasons for strong spatial dispersion in the effective medium response. The absence of strong spatial dispersion means that the permeability tensor \( \mu \) (as well as \( \varepsilon \)) cannot depend strongly on the direction of the vector \( \mathbf{q} \).

The answer to this paradox is given in the theory of multipole media, also called media with weak spatial dispersion [15, 16, 17, 26]. Multipole media depend on higher-order multipole response accompanying the magnetic dipole response of complex particles. In the present case this higher-order multipole response is the electric quadrupole susceptibility. In [25] it was correctly noticed that the quadrupole polarization of plasmonic dimers is as essential as their electric and magnetic dipole polarizations. In general, media with resonant quadrupole moments as any other multipole media cannot be described only in terms of \( \varepsilon \) and \( \mu \). Physically sound material equations for multipole media contain besides macroscopic fields \( \mathbf{E} \) and \( \mathbf{H} \) also spatial derivatives of \( \mathbf{E} \) [15, 16, 26]. Respectively, more material parameters are required. In this theory the unit cell magnetization can be properly described without involving spatially dispersive permittivity \( \varepsilon(\mathbf{q}) \) and permeability \( \mu(\mathbf{q}) \), which describe only excitation by plane waves traveling along one particular direction.

For a medium formed by dimers we can write for any direction of the wave propagation

\[
m = m_z = -i\omega a \frac{p_y(B) - p_y(A)}{2} = -i\omega a^2 \alpha \frac{E_y(B) - E_y(A)}{2} \approx 2\nu \nabla_x E_y (7)
\]

where \( \nu = i\omega a^2 \alpha/4 \). This relation is more adequate than the proportionality between \( m \) and \( H \), which is only a special case of (7). We can present (7) as the sum of the symmetric and antisymmetric derivative forms:

\[
m_z = \nu(\nabla_x E_y - \nabla_y E_x) + \nu(\nabla_x E_y + \nabla_y E_x) \quad (8)
\]

or, in the index form,

\[
m_\alpha = \Gamma_{\alpha\beta} H_\beta + \kappa_{\alpha\beta\gamma} \nabla_\beta E_\gamma \quad (9)
\]

In (9) indices \( \alpha, \beta, \gamma \) correspond to the Cartesian coordinates and we use the notations

\[
\Gamma_{zz} = -i\nu \omega \mu_0, \quad \kappa_{zxy} = \kappa_{zyx} = \nu \quad (10)
\]
Components with all the other combinations of indices $\alpha, \beta, \gamma$ equal zero. From (9) it is clear that the magnetic polarization contains two parts. The first part is the magnetic moment of the dimer induced by the magnetic field which results in the resonant effective permeability of the composite medium. The second part describes the magnetic moment of the dimer which is proportional to all the other combinations of spatial derivatives of the electric field. When the wave propagates along $y$, the second term cancels out with the first one and the induced magnetic moment vanishes. The dependence of the magnetic moment on the propagation direction is not a feature of strong spatial dispersion because the polarizability tensors $\Gamma_{\alpha\beta}$ and $\kappa_{\alpha\beta\gamma}$ do not depend on the propagation direction. This feature of weak spatial dispersion was discussed in books [15, 16]. The material equation for the magnetic field resulting from (9) and the quasi-static averaging procedure can be written in the index form as

$$B_{\alpha} \equiv \mu_0 H_{\alpha} + M_{\alpha} = \mu_0 \mu_{\alpha\beta} H_{\beta} + \chi_{\alpha\beta\gamma} \nabla_{\beta} E_{\gamma}$$

(11)

Here tensor $\mu_{\alpha\beta}$ results from $\Gamma_{\alpha\beta}$ and tensor $\chi_{\alpha\beta\gamma}$ results from $\kappa_{\alpha\beta\gamma}$.

A similar consideration can be done for the quadrupole moment of the dimer. The vectors of the electric dipole and magnetic dipole moments of the dimer and the tensor of its quadrupole moment are resonant. This means that the electric displacement vector $D$ essentially includes the contribution of the electric quadrupole polarization $Q_{\alpha\beta}$. This evolves one more material parameter in the material equation which takes the form [15, 16, 17, 26]:

$$D_{\alpha} \equiv \varepsilon_0 E_{\alpha} + P_{\alpha} + \frac{1}{2} \nabla_{\beta} Q_{\alpha\beta} = \varepsilon_0 \varepsilon_{\alpha\beta} E_{\beta} + \xi_{\alpha\beta\gamma\delta} \nabla_{\beta} \nabla_{\gamma} E_{\delta}$$

(12)

Material equations (11) and (12) corresponds not only to dimers. They refer to all composites whose inclusions possess resonant quadrupole polarization in the absence of bianisotropy, see e.g. [30, 31, 32].

In this formalism the boundary conditions should be revised as compared to the formalism of initial equations (11) and (12), because the set of Maxwell’s boundary conditions is not enough to solve boundary problems for such media. Additional boundary conditions should be derived as it was explained in books [15, 16]. However, to our knowledge, for media described by equations (11) and (12) boundary conditions have not been derived. In the known papers composites of dimers and other multipole media are described by only $\varepsilon$ and $\mu$. That description implies the use of Maxwell’s boundary
conditions, but can be applied only to the plane-wave incidence at the angle used in the extraction of the effective parameters.

### 2.2. Artificial magnetism in bianisotropic composites

Next we will discuss the artificial magnetism in composites based on split rings (SRR), with the emphasis on the effect of bianisotropy. In particular, we consider an array of U-shaped metal SRRs experiencing plasmonic resonance in the optical range. Similarly to the previous section, we use a pair of plane waves to probe the particle response to external magnetic fields. First, let us show that magnetic polarizabilities are different for the two cases: when the waves propagate with vector $\mathbf{q} = q\mathbf{x}_0$ along $x$ and the electric field associated to the local magnetic field $\mathbf{H}$ is $y$-polarized, and when $\mathbf{q} = q\mathbf{y}_0$ and the same magnetic field is related to $x$-polarized electric fields.

The following consideration is based on the method of induced electromotive force (IEF) as it was formulated in [27] for conducting wire scatterers. A formula for the IEF more appropriate to the case of a plasmonic SRR is more involved and corresponding derivations are cumbersome, whereas the result will be definitely similar to that obtained below.

Let us choose the coordinate system as shown in Fig. 2 and locate an imaginary port in the origin. The electromotive force induced by the local electromagnetic field at this imaginary port is equal to [12, 27]

$$\mathcal{E} = \frac{1}{I_0} \int_l E_l I_{\text{rad}}(l) \, dl$$

(13)

Here $E_l$ is the component of the local electric field tangential to the scatterer’s contour, $I_{\text{rad}}$ is the complex amplitude of the linear current distributed over this contour in the radiation regime, when an external voltage source is connected to the port, and $I_0 = I_{\text{rad}}(0)$. In the case $q = q_x$ from Maxwell’s equation (4) we obtain

$$E_l = \pm E_y|_{x=\pm \frac{d}{2}} = \pm i \frac{\omega \mu_0 H d}{2}$$

(14)

Since the U-shaped scatterer is optically small and the currents at the ends of the stems vanish, the distribution of the induced current over the scatterer’s contour $I_{\text{rad}}(l)$ can be approximated as a linear function of the contour coordinate $l$:

$$I_{\text{rad}}(l) = I_0 \frac{b + \frac{d}{2} - l}{b + \frac{d}{2}}$$

(15)
Figure 2: (Color online) A sketch of U-shaped SRRs for two cases of the wave propagation. The incident magnetic field at the particle center is the same in both cases. Introducing an imaginary port at the center of the horizontal bar and applying the method of induced electromotive force we prove the strong difference of the particle magnetic polarizabilities on the propagation direction for these two cases.
where \( l = 0 \) at the port, \( l = |x| \) for \(-d/2 < x < d/2\), \( y = 0 \) and \( l = y + d/2 \) for \( x = \pm d/2, \ y < b \). Upon substitution of (14) and (15) formula (13) can be rewritten in the case \( q = q_x \) as

\[
E = i \omega \mu_0 H d \int_{0}^{b} (b - y) \ dy = i \omega \mu_0 H b^2 d \quad \text{(16)}
\]

The same electromotive force \( E \) can be expressed through the input impedance \( Z_{\text{in}} \) of the scatterer referred to the imaginary port: \( E = I_0 Z_{\text{in}} \). Equating these two representations one can express the ratio \( (I_0/H) \) through \( Z_{\text{in}} \):

\[
\frac{I_0}{H} = i \omega \mu_0 H b^2 d \quad \text{at } Z_{\text{in}}(2b + d) \quad \text{(17)}
\]

When we calculate \( \alpha_{mm} \), we use the definition (3), and take into account that the current distribution induced by the local field is different from the distribution of the current excited by an external voltage in the port. Therefore, instead of (15) we use a more adequate approximation which is smooth at the coordinate origin:

\[
I(l) \equiv I_0 F(l) = I_0 \left( \frac{b + d}{2} \right)^2 - l^2 \quad \text{at } (b + d/2)^2 \quad \text{(18)}
\]

Using the definition of the magnetic moment (3) and the distribution function \( F(l) \) we can write the magnetic polarizability of the SRR in the form:

\[
\alpha_{mm} = \frac{m}{H} = \frac{\mu_0 I_0}{2H} \left[ b \int_{-d/2}^{d/2} F(x, y = 0) \ dx + 2 \int_{0}^{b} F \left( x = \frac{d}{2}, y \right) \ dy \right] \quad \text{(19)}
\]

Substituting relation (18) for function \( F(l) \) into (19) we finally obtain

\[
\alpha_{mm} = i \omega \mu_0^2 b^3 d^2 (6b + d) \quad \text{at } 2Z_{\text{in}}(2b + d)^2 \quad \text{(20)}
\]

In the case \( q = q_y \) from Maxwell’s equation (6) we find

\[
E_l = E_x(y = 0) = \pm i \frac{\omega \mu_0 H b}{2} \quad \text{(21)}
\]
Substituting (21) and (13) into the main formula (13) we obtain

\[ \mathcal{E} = i \frac{\omega \mu_0 H b}{b + \frac{d}{2}} \int_0^{d/2} \left( \frac{d}{2} + b - x \right) dx = i \frac{\omega \mu_0 H bd(4b + d)}{8(2b + d)} \] (22)

Only the horizontal bar gives contribution into the induced electromotive force, while the integrals over the stems vanish. For the ratio \( I_0/H \) we have in this case

\[ \frac{I_0}{H} = i \frac{\omega \mu_0 bd(4b + d)}{8Z_{in}(2b + d)} \] (23)

Substituting (23) into formula (19) we obtain

\[ \alpha_{mm} = i \frac{\omega \mu_0^2 b^2 d^2 (6b + d)(4b + d)}{16Z_{in}(2b + d)^2} \] (24)

This result clearly differs from (20). The ratio of magnetic polarizabilities corresponding to the two cases of propagation is as follows:

\[ \frac{\alpha_{mm}^{(q=x)}}{\alpha_{mm}^{(q=y)}} = \frac{8b}{4b + d} \] (25)

For \( b = d \) these magnetic polarizabilities differ by the factor of 1.6. Though in both cases of the wave propagation direction the magnetic polarizability is nonzero, this example clearly shows that the magnetic polarizability \( \alpha_{mm} \) of a U-shaped SRR is an ambiguous value, similarly to the case of a dimer. Respectively, one should take into account the electric quadrupole polarization.

2.3. **Bianisotropic material relations for nanostructured metamaterials**

When some local magnetic field acts on a U-shaped SRR, the same polarization current which forms dipole moments of the stems (whose vector sum vanishes) also creates some noncompensated resonant dipole moment of the horizontal bar. Thus, local magnetic field in this case creates resonant magnetic dipole moment, resonant electric quadrupole moment, and resonant electric dipole moment. The effect of electric dipole polarization of the medium unit volume by magnetic field can be expressed as the second term in the right-hand side of the equation

\[ P_\alpha = \Lambda_{\alpha \beta} E_\beta + \Psi_{\alpha \beta} H_\beta \] (26)
This effect in reciprocal media is complemented by the effect of the magnetic polarization of the medium by electric fields:

\[ M_\alpha = \Theta_{\alpha\beta} H_\beta + \Psi'_{\alpha\beta} E_\beta + \kappa_{\alpha\beta\gamma} \nabla_\beta E_\gamma \] (27)

This couple of effects (tensors \( \Psi \) and \( \Psi' \) are uniquely related: \( \Psi'_{\alpha\beta} = -\Psi_{\beta\alpha} \)) is well known in the electromagnetic theory and engineering (e.g., [15]) and is called bianisotropy. Media formed by parallel U-SRR particles possess resonant bianisotropy. Even dual SRRs introduced in [28] are bianisotropic, and their bianisotropy was widely discussed in the literature after the publication of paper [29]. The bianisotropy of dual SRRs is an important factor that can suppress the backward-wave regime in metamaterials [29]. Moreover, U-shaped particles show stronger bianisotropic effects than dual SRRs. Since bianisotropy is a stronger effect than artificial magnetism (first- and second order effects in terms of \( ka \)), characterization of such metamaterials only by two parameters \( \varepsilon \) and \( \mu \) is clearly not adequate.

From (26) and (27) it is clear that two additional material parameters should be introduced compared to multipole media without bianisotropy. Material equations (11), (12) for bianisotropic multipole media generalize to the form:

\[ B_\alpha = \mu_0 \mu_{\alpha\beta} H_\beta + \psi_{\alpha\beta} E_\beta + \chi_{\alpha\beta\gamma} \nabla_\beta E_\gamma \] (28)

\[ D_\alpha = \varepsilon_0 \varepsilon_{\alpha\beta} E_\beta - \psi_{\beta\alpha} E_\beta + \xi_{\alpha\beta\gamma\delta} \nabla_\beta \nabla_\gamma E_\delta \] (29)

The material parameter describing the bianisotropic effect in equations (28) and (29) is tensor \( \psi \), called \textit{magnetoelectric coupling} parameter in the theory of reciprocal bianisotropic media (e.g. in [15, 16, 33]).

Two special cases of bianisotropic media are well known: chiral media when tensor \( \psi \) is symmetric, and omega media when tensor \( \psi \) is antisymmetric. According to the classification [34, 15] arrays of parallel U-SRRs belong to the class of omega media. The known theory of omega media (see e.g. in [15, 35, 36, 37]) was developed for the special case when tensors \( \chi \) and \( \xi \) in (28) and (29) are negligible. Fabrication of nanostructured arrays of U-SRRs [38] will hopefully motivate researchers to develop the theory of multipole bianisotropic media, and the first task should be the derivation of boundary conditions for media described by equations (11) and (12) and for media obeying equations (28) and (29).

2.4. Artificial magnetism without resonant multipole polarization

Finally, let us discuss which inclusion shapes can potentially offer artificial magnetism property without strong excitation of higher multipoles. If the
ring shown in Fig. 2 were closed, the electric dipole moments in each side would be in all cases of the wave propagation cancelled by that in the opposite arm. In an electrically small closed loop with uniform current distribution, the induced electromotive force is the same for any direction of propagation of incident waves. This induced electromotive force is equal to the negative time derivative of the applied magnetic flux. No bianisotropy and no quadrupole moment arise, and the array of closed rings (at enough low frequencies, of course) behaves an effective diamagnetic medium.

If the open part of the ring is very small compared to its perimeter, the dependence of the induced magnetic moment on the propagation direction is also small and, respectively, quadrupole moments are negligible. However, the bianisotropy remains strong and resonant, since the induced current distribution is not uniform, which results in both electric and magnetic dipole moments of the loop. The bianisotropy of the array related to the open part of the loop can be suppressed in two ways. The first way is to prepare the unit cell from at least two SRRs so that the bianisotropy of the unit cell is compensated. That way was suggested in [29]. A similar approach was developed in [39, 40] where a practically isotropic doubly-negative composite was designed. The unit cell in these works was formed by six properly arranged $\Omega$-shaped metal particles. A dense stack of split rings with interrupting positions of splits was introduced in [41]. In all these design the governing idea is to realize uniform distribution of induced current along the inclusion perimeter.

However, the above designs refer to the microwave range. Realistic approaches to the artificial magnetism without strong spatial dispersion, high-order multipoles and bianisotropy in the optical range were suggested more recently. Suitable design solutions are, for example, as follows: four-split optical SRRs suggested in [42] and effective nanorings of plasmonic spheres [43]. In [43] an effective ring is formed by plasmonic nanospheres located at the corners of a regular polygon. The minimal number of nanospheres when the higher multipoles can be (with a qualitative accuracy) neglected is four. In [42] each of four plasmonic scatterers forming the effective ring is a planar L-shaped nanoparticle. In [43] these four plasmonic scatterers are spheres. It is clear that in both these design solutions the physical mechanism of the artificial magnetism is the same.

Notice, that the concentration of these effective rings should be very high in order to obtain a strong resonance of permeability. Results presented in [43] correspond to almost touching nanorings. Therefore the magnetism
of such nanorings can be realized only in the anisotropic variant. The re-
requirement of the high concentration makes a composite of randomly oriented
effective rings of nanospheres hardly feasible (the same concerns three mutually orthogonal arrays of effective rings). The fully isotropic solution based on core-shell magnetic nanoclusters \cite{44} is probably more practical.

3. Conclusion

In this paper we have considered the problem of effective medium description of nanostructures designed to emulate properties of magnetic materials in the visible range. The artificial magnetic effect can be achieved due to specific shapes of nanoinclusions and results from weak spatial dispersion in the composite. In general, spatial dispersion produces a variety of effects, including resonant bianisotropy, and the artificial magnetism is only one of them. Care should be taken in introducing effective permeability in such a way that it indeed measures the averaged magnetic polarization and can be used in solving boundary problems for composite samples. If the inclusion geometry is such that bianisotropic effects are allowed, appropriate terms should be added to the material relations: bianisotropic effects are usually stronger than the artificial magnetism. Except some very specific geometries, induced electric quadrupoles are of the same order as magnetic moments and they must be included in the analysis. Furthermore, not only the response to the curl of electric field (that is, to magnetic field), but also to all other combinations of the first-order spatial derivatives of electric field should be included for adequate description of the effective material properties. In other words, characterizing bulk optically dense nanostructured metamaterials starts from the analysis of multipoles and bianisotropy. The insight on adequate material equations for particular nanocomposite is mandatory for physically sound characterization. Unfortunately, the effective medium theory of weakly dispersive media is not complete at this stage, and further investigations are necessary (especially in what concerns boundary conditions at interfaces), before we will be able to use the effective parameters in the design of optical devices.

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