Electromagnetic characterization of planar and bulk metamaterials: A theoretical study

Dmitry Morits and Constantin Simovski
Department of Radio Science and Engineering, School of Science and Technology, Aalto University, P.O. Box 13000, FI-00076 AALTO, Finland

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Using an example of a bilayer of plasmonic nanospheres, we generalize a method of electromagnetic characterization of metasurfaces (metafilms) recently suggested in the literature. We theoretically demonstrate that the results of this characterization method are suitable to predict scattering parameters of bilayer metasurfaces. Further, we develop an original electrodynamic approach which allows one to extract the effective material parameters of bulk lattices from reflection and transmission coefficients of a single generic metasurface. The results of this retrieval are compared with the results of an alternative method based on the quasistatic homogenization model. The difference between these results is discussed from the point of view of the electromagnetic interaction of scatterers forming the lattice.

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I. INTRODUCTION

In recent years, a great interest of researchers has been dedicated to composite electromagnetic structures called metamaterials (MTMs). Metamaterials are artificial materials engineered to achieve advantageous and unusual electromagnetic properties not normally met in ordinary structures.[1] The most convenient way to describe the electromagnetic behavior of MTMs is the utilization of so-called effective material parameters, obtained by a homogenization procedure. However, in the modern scientific literature there is no well-established method of homogenization and existing solutions often give controversial and ambiguous results. In this situation, we consider the development of the clear and handy homogenization technique as one of the most important goals of the modern metamaterial science.

Let us assume that an MTM under study can be adequately homogenized in terms of two bulk material parameters ε and μ, perhaps tensors that we somehow retrieved from scattering parameters of the MTM sample. Since most part of practical MTM are performed as parallel-plate layers, we restrict our consideration by the case when the MTM sample is a layer. We assume that the external sources exciting the layer are located far enough and their field can be presented as a set of plane waves. Then the scattering parameters of the layer are plane-wave reflection R and transmission T coefficients, which must be obviously related to parameters ε and μ. In order to give a condensed description of bulk electromagnetic properties of the layer, these two parameters must be independent of the distribution of the incident electromagnetic field. Otherwise these parameters are applicable only to same special case of the wave incidence from which they were retrieved, in other words are redundant. Since the field in the layer is the superposition of plane waves the concept of useful (nonredundant) material parameters implies their independence on the wave vector $\mathbf{q}$ for fixed frequency ω. This property in the general theory is called locality.[2]

It is well known that the locality of the medium of separate particles corresponds to the frequency region in which the medium unit cell is rather optically small. Let us accept for simplicity that we deal with lattices whose unit cell possess both electric and magnetic (p-m) dipole moments and operate in the range $(qa) \lesssim 1$. In this range, the value of q is uniquely related with ω for given propagation direction and wave polarization. Therefore, locality in this range implies the independence of material parameters ε and μ on the angle of the wave incidence to the layer.

Two following consequences of the locality of ε and μ were derived in the classical electrodynamics of media:[2–4] (1) *Passivity*. For the temporal dependence $e^{-i\omega t}$, it implies $\mathrm{Im}(\varepsilon)>0$ and $\mathrm{Im}(\mu)>0$ 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 (no generators of the electromagnetic oscillations at frequency ω) means the violation of the second law of thermodynamics.[2]

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

Both passivity and causality can be mathematically unified in the form of the well-known Kramers-Kronig relations (see, e.g., in Refs. 2–4). If these mandatory locality requirements are not respected for material parameters retrieved from R and T coefficients over the essential frequency range this obviously indicates the dependence of these parameters on the spatial field distribution, and these extracted material parameters are redundant.

For media formed by inclusions or their groups located in a homogeneous nondispersive host medium experiencing an electric resonance and a magnetic resonance, these two requirements are consistent with Lorentz-type formulas for bulk material parameters,

$$\varepsilon=\varepsilon_0\left(\varepsilon_{\mathrm{h}}+\frac{A_{\mathrm{e}}\omega_{\mathrm{e}}^2}{\omega_{\mathrm{e}}^2-\omega^2+j\omega\gamma_{\mathrm{e}}}\right), \tag{1}$$

$$\mu=\mu_0\left(1+\frac{A_{\mathrm{m}}\omega^2}{\omega_{\mathrm{m}}^2-\omega^2+j\omega\gamma_{\mathrm{m}}}\right). \tag{2}$$

Here ε_h is the matrix relative permittivity, A_e, A_m are dimensionless positive values independent of the frequency, ω_e, ω_m are frequencies of electric and magnetic resonances, respectively, γ_e, γ_m are also positive values with dimension of frequency (in the initial Lorentz's model they are constant and called damping frequencies, in more advanced models γ_e, γ_m are dependent on ω). The time dependence in Eqs. (1) and (2) is assumed to be $\exp(j\omega t)$. Formula (1) follows from the classical microscopic theory of molecular media. Formula (2) describing the artificial magnetism was initially introduced in 1950s by Schelkunoff for bulk arrays of parallel split-ring resonators (see, e.g., in Ref. 5). Later it was generalized to other kinds of artificial magnetic media in both radio and optical frequency ranges. Formulas (1) and (2) are applicable also to anisotropic magnetodielectric composites where they describe resonant components of tensors ε and μ. A more detailed discussion of these formulas can be found in Ref. 1.

It is often thought that Eqs. (1) and (2) are only possible dependencies for local material parameters ε and μ. However, it is not so. Formulas (1) and (2) follow from the quasistatic theory which is hardly adequate for MTMs because most part of bulk MTMs operate in the range $(qa)>0.1$. Meanwhile, the locality limitations can meet dynamic homogenization models. In works,[6,7] examples were presented when the quasistatic model gives different frequency dependencies for ε and μ than the dynamic model. The last one, however, respects the locality of material parameters. In the present paper the concept of dynamic ε and μ is developed. Below we consider a nanostructured MTM operating in the visible for which homogenization model combining a retrieval procedure and a dynamic theory of dipole lattices gives a specific resonant dispersion which is local but qualitatively different from Eqs. (1) and (2). In precedent works,[8–11] the Maxwell Garnett approximation leading to Eqs. (1) and (2) has been used for such MTM. In the present paper, we will see how strongly the dynamic interaction between clusters of nanoparticles changes the material parameters. Namely, we will show that the permittivity of MTM can resonate at the frequency where the magnetic dipole of the lattice unit cell resonates and the permeability of MTM can resonate at the frequency where the electric dipole of the lattice unit cell resonates. This effect has nothing to do with the so-called "antiresonance of metamaterials" which is, definitely, an artifact related to an inadequate retrieval procedure (see, e.g., in Ref. 12). The "antiresonance" refers to the most widely applied method, namely, to the method suggested in Refs. 13 and 14 as an evolution of the classical method by Nicholson, Ross, and Weir of characterization of continuous media (see, e.g., in Refs. 15–17). In this method, the effective material parameters $\varepsilon_{\rm eff}$ and $\mu_{\rm eff}$ are extracted from the R and T coefficients of a composite slab which is replaced by a uniformly homogeneous layer of the same thickness. The application of this simplistic procedure to MTMs obviously leads to the violation of locality in the retrieved material parameters which was noticed in Refs. 18 and 19 and later criticized in works.[6,7,12,20]

The second goal of this paper is to develop the method of the electromagnetic characterization of metasurfaces (metafilms), i.e., resonant composite layers with 1–2 unit cells across them. In the dominant literature metasurfaces are treated as if they were bulk media, however as it was properly noticed in Refs. 21 and 22 this is a special and very important class of MTMs which requires the special characterization approach.

Effective material parameters for grids of small resonant inclusions were introduced in works.[21,22] Metasurfaces considered in these works contain one resonant inclusion across the layer which possess simultaneously electric and magnetic dipole moments whose resonances can overlap over the frequency axis. Material parameters of metasurfaces correspond to their replacement by effective current sheets and describe the interaction of metasurfaces with tangentially averaged electric and magnetic fields. In general, these material parameters are called electric and magnetic tangential and normal surface susceptibilities.[21] All these susceptibilities were defined through jumps of tangential and normal components of averaged electric and magnetic fields across the physical metasurface thickness (which was assumed to be very optically small). The averaged fields were obtained by averaging of microscopic fields over periods of the grid at two sides of the metasurface. The characterization approach suggested in Ref. 21 was developed in Ref. 22 for monolayers of solid magnetoelectric particles isotropic in the tangential plane. Then four scalar parameters give the condensed description of the metasurface. For two specific angles of incidence (say 0° and 45°), one simulates R and T coefficients and from these four complex parameters one extracts four material parameters.

In this paper, we show that this approach is applicable to bilayers of resonant electric dipoles, in our example, silver nanospheres. We consider a bilayer formed by two doubly periodic planar grids of nanospheres as a metasurface whose unit cells possess both electric and magnetic moment. The whole bilayer is then replaced by a sheet of effective electric and magnetic currents and the method from Ref. 22 is applied. We show that material parameters of the metasurface extracted from R and T coefficients simulated for two angles of incidence are applicable to predict R and T for other incidence angles with suitable accuracy.

This part of our paper is tightly related with the problem formulated in the first part of this section, i.e., with the dynamic characterization of a bulk material. This is so because we consider a bulk lattice as that obtained by repetition of such generic bilayers. In work,[23] it was suggested to use retrieved susceptibilities of the generic grid (monolayer) metasurface to predict bulk material parameters of the three-dimensional lattice. Namely, it was recommended to use specially derived relations between these susceptibilities and individual polarizabilities (electric and magnetic) of particles forming the grid. After that bulk material parameters of the lattice can be found with the use of the Maxwell Garnett algorithm.[23] Below we show that the first part of this task is justified also for bilayers. Namely, the extraction of electric and magnetic polarizabilities of the bilayer unit cell from the previously retrieved susceptibilities of this metasurface gives fully adequate results. However, as it has been already mentioned above, the second stage of the characterization procedure recommended in Ref. 23, i.e., application of the Maxwell Garnett formulas is not an adequate task for MTMs if

$(ka)\sim 1$, especially for those formed by clusters of resonant electric dipoles.

So, in the present work we expand the characterization procedure suggested in Ref. 22 suggested for a grid of solid magnetodielectric dipole scatterers to the case when the metasurface is a grid of regular clusters of electric dipoles and apply the results to the dynamic retrieval of ε and μ of a bulk lattice formed by such clusters.

II. CHARACTERIZATION OF A BILAYER GRID OF PLASMONIC NANOSPHERES

A. General approach

In works,[21,22] authors considered metasurfaces performed as a planar grid of solid resonant particles. It was shown that the electromagnetic behavior of these metasurfaces is described by two planar tensors called tangential surface susceptibilities $\bar{\bar{\chi}}_{\rm ES}$ and $\bar{\bar{\chi}}_{\rm MS}$ and two scalar values called normal surface susceptibilities $\chi_{\rm ES}^{zz}$ and $\chi_{\rm MS}^{zz}$. These values can be called characteristic material parameters of these metasurfaces because they do not depend on the incidence angle. They are defined through the generalized sheet transition conditions (GSTCs),

$$\mathbf{z}_0\times\mathbf{H}\big|_{z=0^-}^{0^+}=j\omega\varepsilon\bar{\bar{\chi}}_{\rm ES}\cdot\mathbf{E}_{t,\rm av}\big|_{z=0}+\mathbf{z}_0\times\nabla_t[\chi_{\rm MS}^{zz}H_{z,\rm av}]_{z=0}, \tag{3}$$

$$\mathbf{z}_0\times\mathbf{E}\big|_{z=0^-}^{0^+}=j\omega\mu\bar{\bar{\chi}}_{\rm MS}\cdot\mathbf{H}_{t,\rm av}\big|_{z=0}-\mathbf{z}_0\times\nabla_t[\chi_{\rm ES}^{zz}E_{z,\rm av}]_{z=0}, \tag{4}$$

where the subscript "av" represents the average of the field quantity on either side of the metafilm, ε and μ are the material parameters of the host medium.

The procedure of extracting these material parameters from R and T coefficients of the metasurface is as follows. Assume that we have obtained amplitudes and phases of R and T for the normal incidence ($\theta=0$) from broadband measurements or numerical simulations. Then we obtain the surface susceptibilities using the expressions derived in Ref. 22 (for the TM polarized wave),

$$\chi_{\rm ES}^{xx}=\frac{2j}{k}\frac{R(0)-T(0)+1}{R(0)-T(0)-1}, \tag{5}$$

$$\chi_{\rm MS}^{yy}=-\frac{2j}{k}\frac{R(0)+T(0)-1}{R(0)+T(0)+1}, \tag{6}$$

where $k=k_0\sqrt{\varepsilon_{\rm h}}$ is the wave number in the host medium which is assumed to be uniform dielectric space of permittivity $\varepsilon_{\rm h}$. Notice that in work,[22] signs in front of the expressions in the right-hand side are opposite.

On the base of the boundary conditions (3) and (4), we rederived the equations connecting the reflection and transmission coefficients of the grid and surface susceptibilities, already obtained in Ref. 22. This way we found several misprints in expressions (7)–(10) of paper.[22] For the TM polarization the reflection and transmission coefficients for the incidence angle θ have been deduced as follows:

$$R(\theta)=\frac{(jk/2\cos\theta)(\chi_{\rm MS}^{yy}+\chi_{\rm ES}^{xx}\cos^2\theta-2\chi_{\rm ES}^{zz}\sin^2\theta)}{1-(jk/2\cos\theta)(\chi_{\rm MS}^{yy}-\chi_{\rm ES}^{xx}\cos^2\theta-2\chi_{\rm ES}^{zz}\sin^2\theta)+(k/2)^2\chi_{\rm ES}^{xx}(\chi_{\rm MS}^{yy}-2\chi_{\rm ES}^{zz}\sin^2\theta)}, \tag{7}$$

$$T(\theta)=\frac{1-(k/2)^2\chi_{\rm ES}^{xx}(\chi_{\rm MS}^{yy}-2\chi_{\rm ES}^{zz}\sin^2\theta)}{1-(jk/2\cos\theta)(\chi_{\rm MS}^{yy}-\chi_{\rm ES}^{xx}\cos^2\theta-2\chi_{\rm ES}^{zz}\sin^2\theta)+(k/2)^2\chi_{\rm ES}^{xx}(\chi_{\rm MS}^{yy}-2\chi_{\rm ES}^{zz}\sin^2\theta)}. \tag{8}$$

In Eqs. (7) and (8), there are factors 2 in front of the term $\chi_{\rm ES}^{zz}$ which were missed everywhere in formulas (9) and (10) of Ref. 22. Also the sign in front of the expression in the right-hand side of formula (10) from Ref. 22 is inverse.

Characteristic parameters $\chi_{\rm MS}^{yy},\chi_{\rm ES}^{xx}$ can be extracted from $R(0)$ and $T(0)$ using formulas (5) and (6) whereas $\chi_{\rm ES}^{zz}$ can be found from one of formulas (7) and (8) for any specific angle θ, say $\theta=45°$. However, for better accuracy it is reasonable to use for this extraction both $R(\theta)$ and $T(\theta)$ and apply formula (16) of Ref. 22 which takes form

$$\chi_{\rm ES}^{zz}=-\frac{\chi_{\rm MS}^{yy}}{\sin^2\theta}-\frac{2j\cos\theta}{k\sin^2\theta}\frac{R(\theta)+T(\theta)-1}{R(\theta)+T(\theta)+1}. \tag{9}$$

After obtaining all three scalar surface susceptibilities corresponding to the incidence of a TM polarized wave, we can calculate the reflection and transmission coefficients for any angle of incidence of the TM wave. Similarly, one can retrieve and utilize three remaining scalar surface susceptibilities describing the TE polarized waves.

In the present paper, we generalize this approach to a bilayer of silver nanospheres depicted in Fig. 1(a). Two identical planar grids of nanospheres forming the metasurface are double periodic in the case $a\neq 2b$ (distances a and b are shown in this figure). Then the unit cell of the metasurface of area a^2 in the tangential (x-y) plane includes four nanospheres. It is clear that in this bilayer structure the incident wave will induce both resonant electric and magnetic polarization. Therefore the unit cell should possess both electric and magnetic resonant dipole polarizabilities. To explain this fact in more details, we have to refer to seminal paper[8] (ideas of this paper have been developed in many works, among

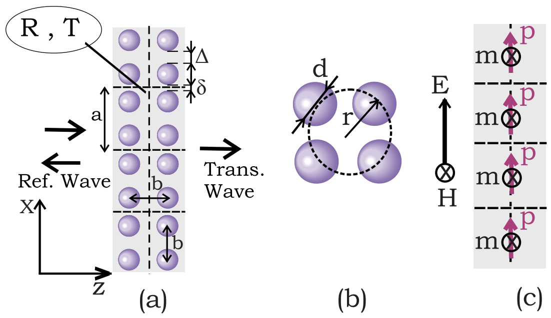

FIG. 1. (Color online) (a) An original structure performed as a bilayer of nanospheres. (b) An effective nanoring of four plasmonic spheres. (c) An effective metasurface formed by electric (p) and magnetic (m) dipoles induced in every unit cell.

them we have to mention the very recent work[24]). Consider a cluster of four nanospheres lying in a cross section of the unit cell as shown in Fig. 1(b). When four plasmonic spheres arranged in a such way are excited by a time-varying magnetic field the electric dipoles induced in them are oriented in the azimuthal direction and form a discrete analog of a circular polarization current[8] which is shown in Fig. 1(b) as a circle of radius r. In work,[8] clusters of nanospheres located at the corners of a regular polygone were called effective nanorings. Effective nanorings as a design solution of the artificial magnetism and their variants developed in works[9–11] are in our opinion very promising for obtaining isotropic negative permeability in the optical range.

An incident plane wave excites both electric and magnetic modes in the unit cell of the original structure. Every unit cell can be therefore replaced by a pair of electric and magnetic dipoles **p** and **m** as shown in Fig. 1(c). These dipoles are oriented tangentially for a normally incident wave. Such a p-m grid can be replaced by an effective sheet with electric and magnetic currents if $a \ll \lambda$, where λ is the operation wavelength.[22] So, the structure shown in Fig. 1(a) seems to be a variant of a magnetodielectric metasurface studied in Refs. 21 and 22, where solid p-m particles are replaced by p-m clusters. From R and T coefficients referred to the central plane of the structure [shown in Fig. 1(a) by a straight dashed line], we apparently can retrieve its characteristic parameters.

However, these expectations become not so evident if we take into account two important factors. The first factor is the rather large optical size of the cluster. In the design of Ref. 8, the size of the effective nanoring ($2r+d$ in our notations) is equal to $2r+d \approx \lambda_{res}/3$ in the best possible geometry, where λ_{res} is the wavelength in the host medium at the central frequency of the magnetic resonance band. In this frequency range arguments in favor of the p-m model of the effective nanoring (the model replacing an individual cluster by an electric dipole and a magnetic dipole) become crucial for the whole concept. The analogy of our structure to a very optically dense grid of very small solid scatterers[22] does not seem evident anymore.

The second factor concerns the interaction of adjacent unit cells which is even more important factor for the model replacing the original structure by an effective planar p-m grid. Solid spheres of magnetodielectric material operating in the microwave range which were considered in work[22] are definitely p-m pairs (pairs of electric and magnetic dipoles referred to the centers of spheres). First, they are optically very small (their resonances are due to high permittivity and permeability of the material of spheres achievable in the range of radio frequencies). Second, they are well distanced from one another and clearly interact as p-m pairs. In the present structure, the situation is very different. The distance between the edges of two adjacent effective nanorings δ which is equal to $a-b-d$ is several times smaller than the distance $\Delta = b-d$ between spheres forming the effective nanoring. In other words, the distance between the edges of two adjacent clusters is smaller than the cluster size. This dense package of effective nanorings is necessary to obtain a considerable magnetic resonance of the whole structure.

It is not evident that splitting the whole structure to effective nanorings so that the distance between them is smaller than their size, we can describe the interaction between unit cells in terms of only p-m dipoles avoiding high-order multipoles. Notice, that taking into account higher multipoles would modify the model[22] so strongly that the characterization procedure will become hardly feasible. To sum up, the applicability of the simple characterization approach suggested in Ref. 22 to double-periodic bilayers of electric dipoles is very nontrivial and needs to be checked. It is especially important to check it in the present case when these dipoles are plasmonic nanospheres and the expected effect is the resonant optical magnetism.

B. Polarizabilities of individual nanorings

Following to Refs. 21 and 22, we can retrieve electric and magnetic polarizabilities of the metasurface (from solid p-m particles arranged in a square grid with unit-cell area a^2) through the surface susceptibilities. Formulas expressing these electric and magnetic polarizabilities through surface susceptibilities are given in Ref. 21,

$$\alpha_{ee}^{xx} = \frac{1}{a^2 \chi_{ES}^{xx} + \frac{1}{4sa^3} + \frac{jk^3}{6\pi}}, \tag{10}$$

$$\alpha_{mm}^{yy} = \frac{1}{-a^2 \chi_{MS}^{yy} + \frac{1}{4sa^3} + \frac{jk^3}{6\pi}}. \tag{11}$$

Here $s=0.6956$ is a quasistatic interaction constant of planar dipole grid. The term $jk^3/6\pi$ corresponds to the compensation of radiation losses which are forbidden in regular arrays (see, e.g., in Ref. 3) including two-dimensional arrays.[25] It is clear from relations (10) and (11) that the retrieval of individual polarizabilities suggested in Ref. 21 is a quasistatic procedure. At the first glance, it is difficult to expect its applicability for our structure whose unit cells are formed by clusters of electric dipoles and the size of these clusters is as large as $\lambda/3$. However, let us apply formulas (10) and (11) to our bilayer plasmonic structure and see the result.

To obtain the reflection and transmission coefficients of a bilayer of silver nanospheres, we performed a full-wave nu-

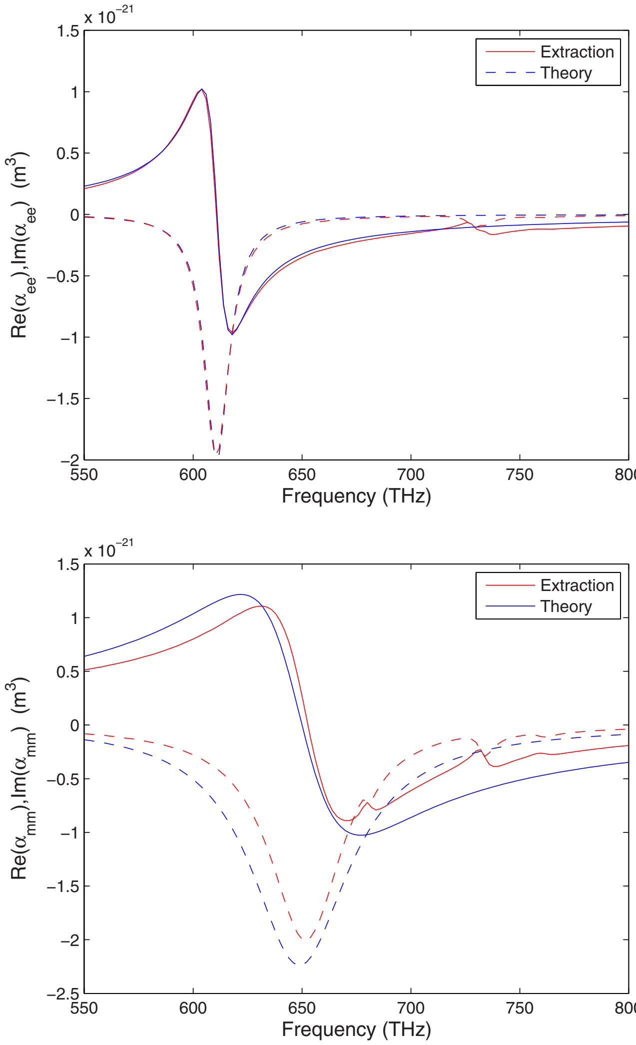

FIG. 2. (Color online) (a) Electric and (b) magnetic polarizability of an effective ring of four plasmonic spheres with $d=32$ nm and $r=38$ nm. Red curve—extracted from the reflection and transmission coefficients of an individual grid; blue curve—calculated using the theory from Ref. 8. Real and imaginary parts are shown by solid and dashed lines, respectively.

merical simulation in high-frequency structure simulator (HFSS) and then compared the results of formulas (10) and (11) to results predicted by the model of an individual effective nanoring presented in Ref. 8.

We used the following geometrical parameters of our structure (see Fig. 1): $d=32$ nm, $r=b/\sqrt{2}=38$ nm, and $a=96$ nm. Permittivity of silver was taken from the Drude model as in works[8,9] (though the Drude model does not meet known experimental data for silver in the visible) in order to better compare our results with those obtained in these works. The plots presented in Fig. 2 reveal the striking agreement between the results of two independent models. This is a surprising result of this study. The distances b between spheres in an individual nanorings is comparable with their diameter d. The distances between the edges of two adjacent nanorings is significantly smaller than the nanoring radius r. However, the dipole model keeps adequate, i.e., the p-m dipolar interaction is dominant in the structure. In other words, the quasistatic p-m dipole model of extraction[21] of polarizabilities keeps valid for grids of plasmonic nanoclusters like shown in Fig. 1(a) and the quasistatic p-m dipole model of an individual effective nanoring[8] keeps adequate as well.

It should be noticed that the description of the bilayer structure in terms of effective rings is not the only possible approach. The alternative way is a description in terms of a superlattice of individual spheres. One can consider such structure as two double-periodic planar arrays of spheres. Response of each planar array can be obtained using the well-known expression for the polarizability of an individual sphere. The interaction between arrays is then described taking into account high-order Floquet harmonics because $b \ll \lambda$. We have not verified it but we believe that scattering matrix calculated with this approach will be the same as we obtained considering the structure as a grid of effective nanorings and will also fit numerical simulations. However, in our opinion, the effective-ring model is more physical in the sense that the description in terms of electric and magnetic dipoles is rather clear and understandable. Moreover, this approach is much more simple and appropriate for practical use, what is extremely important for description of metamaterials in general.

C. Applicability of extracted susceptibilities for arbitrary incidence angles

Figure 3 allows one to compare theoretical and simulated R and T of our bilayer metasurface for $\theta=30°$. Theoretical curves were calculated with the use of formulas (7) and (8) and susceptibilities extracted from $R(0)$, $T(0)$ and $R(45°)$, $T(45°)$ which were obtained in their turn by full-wave simulations in HFSS. In the region below 700 THz, where the electric and magnetic resonances hold, the compliance between the theory and simulation is surprisingly good. Notice that the size of the unit cell a approaches at 700 THz to $\lambda/3$ and approximations implied by the homogenization model are quite rough. On the higher frequencies (not shown on the plot) the results are worse, which is clearly can be explained by overcoming the limit of applicability of the theory.

We may conclude that utilization of surface susceptibilities[21,22] is a very efficient tool to characterize resonant grids far beyond the quasistatic limit which is applicable also for grids formed by clusters of resonant dipoles.

III. BULK LATTICE AND ITS CHARACTERIZATION

Now let us consider an infinite three-dimensional lattice obtained by repetition of the same bilayer with the step a. To obtain the effective material parameters of such a composite authors of Ref. 8 suggested to use a quasistatic homogenization procedure, i.e., well-known Maxwell Garnett formulas for ε and μ. It was shown in Refs. 8 and 9 that with this approach (and with the use of the Drude model of the permittivity of silver) it is possible to achieve negative permeability. Moreover, even both effective permittivity and permeability can be negative in the visible range.

In the work,[23] one presented a characterization procedure of the bulk orthorhombic lattice of solid p-m pairs based on

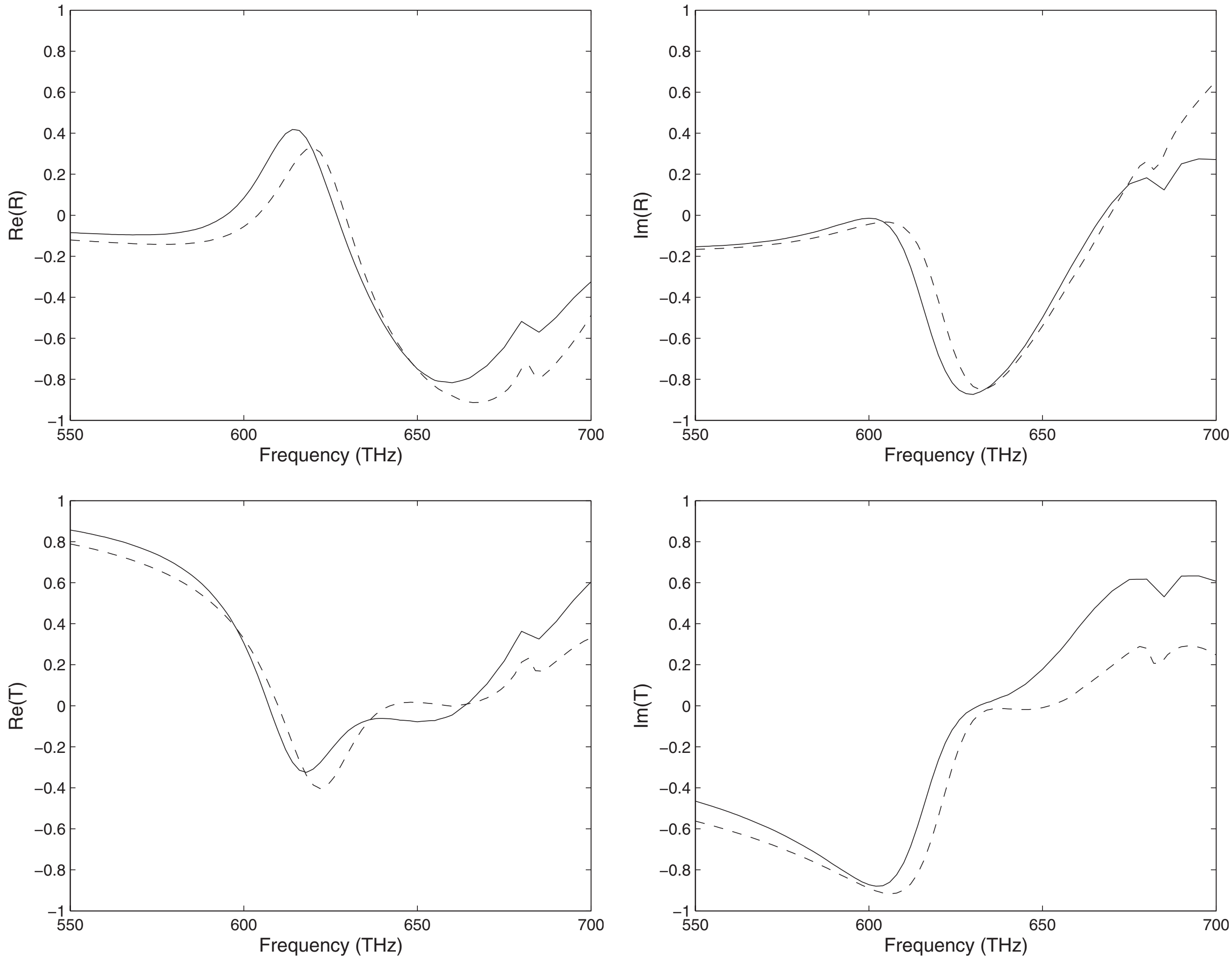

FIG. 3. Reflection and transmission coefficients of the grid of effective nanorings for the TM polarization, angle of incidence is 30°. Solid line—theoretical results calculated by the use of Eqs. (7) and (8); dashed line—results of full-wave simulation in HFSS.

the retrieval of their individual polarizabilities for a single grid which is generic for the bulk lattice. It was suggested to put the retrieved α_{ee} and α_{mm} into Maxwell Garnett equations to find the material parameters of the bulk lattice. This approach gave good results for the structure composed of a cubic array of very small magnetodielectric spheres with very large permittivity and permeability. In the example from Ref. 23, the lattice constant a was about 30 times smaller than the wavelength at the resonance of spheres. Is Maxwell Garnett model adequate also for a lattice of effective nanorings with optical sizes $\lambda/3$? To check it we use a dynamic procedure of the material parameters retrieval.

A. Dynamic model of an orthorhombic p-m lattice

Our method of extraction of bulk material parameters is based on a model (suggested in Ref. 6) of an infinite orthorhombic lattice of p-m dipoles [the response of each inclusion is modeled by electric (**p**) and magnetic (**m**) dipole moments]. Let the wave propagate along the crystal axis which corresponds to the lattice period a and let p-m inclusions be embedded into a dielectric medium with relative permittivity ε_h. Neglecting the interaction of adjacent crystal planes through higher-order Floquet harmonics one can derive the dispersion equation for the wave number q,[12]

$$\cos qa = \cos ka - (1 + \sqrt{1-\Phi})\left(\frac{G+X}{4}\right)\sin ka, \quad (12)$$

where $k = k_0\sqrt{\varepsilon_h}$ and it is denoted as

$$\Phi = \frac{GX \cos ka}{2 \ \sin^2 ka}\left(\frac{4 + GX/2}{G+X}\cos ka - \sin ka\right). \quad (13)$$

Parameters jG and jX are called, respectively, shunt sheet admittance and series sheet impedance of an individual grid formed by p-m pairs, where p-dipoles are responsible for nonzero G and m-dipoles are responsible for nonzero X. In Ref. 20, it was proved that for an individual p-grid parameter G is connected to the jump of the tangential magnetic field across the grid (located, e.g., at the plane $z=0$),

$$H^{TA}(-0) - H^{TA}(+0) = jG\frac{E^{TA}(0)}{\eta}. \quad (14)$$

Here the index TA (transverse averaging) means the microscopic field averaging over the unit-cell surface S and η is the wave impedance of the host medium. Similarly, for the m-grid parameter X defines the jump of the tangential electric field across the grid,

$$E^{\mathrm{TA}}(-0) - E^{\mathrm{TA}}(+0) = jX\eta H^{\mathrm{TA}}(0). \quad (15)$$

The relations (14) and (15) refer to individual grids. It can be easily shown that in the case of normal incidence GSTCs, Eqs. (3) and (4), obtained in Ref. 21 fully coincide with Eqs. (14) and (15), there parameters χ_{ES} and χ_{MS} are connected with G and X as follows:

$$G = k\chi_{\mathrm{ES}}, \quad X = -k\chi_{\mathrm{MS}}. \quad (16)$$

Notice that G and X were derived in the electrodynamic model of p-m grids developed for the case when these grids (crystal planes) have the period d satisfying to the restriction $kd<1$ and the distance between these grids a satisfies to the restriction $qa<1$. Much more restrictive limitation $kd\ll 1$ is implied by the quasistatic model.[22] The equivalence of sheet admittance jG and sheet impedance jX to, respectively, χ_{ES} and χ_{MS} shows that the quasistatic model keeps valid well beyond its initial limitations.

The reason of this result is as follows. We consider the case when the wave propagates normally to crystal planes. A planar grid of in-phase p-dipoles does not produce magnetic field in its own plane. A planar grid of in-phase m-dipoles does not produce electric field in its own plane. Therefore, there is no p-m interaction (electromagnetic interaction between p-dipoles and m-dipoles) on the level of an individual crystal plane. Since the grid is optically rather dense ($kd<1$), the p-p interaction and the m-m interaction are mainly quasistatic,[25] i.e., the same as in the case $kd\ll 1$. Therefore the quasistatic model[22] turns out to be consistent with the model of the bulk lattice to describe the response of every crystal plane.

Now the characterization procedure becomes clear. As in the previous case, we start from R and T coefficients of an individual grid for the normal incidence. Formulas (5) and (6) together with Eq. (16) allow us to extract parameters G and X. In the next step we find the wave number q solving the Eq. (12) and find the normalized wave impedance Z_w of the lattice through the auxiliary parameter γ, obtained in Refs. 6 and 7,

$$\gamma = \sqrt{\frac{G}{X}}\sqrt{\frac{\cos qa - \cos ka - 0.5X\sin ka}{\cos qa - \cos ka - 0.5G\sin ka}}. \quad (17)$$

$$Z_w = \eta\frac{\gamma k_0 + q}{\gamma q + k_0\varepsilon_{\mathrm{h}}}. \quad (18)$$

Bulk material parameters of the composite are expressed through the wave number and wave impedance as follows:

$$\varepsilon_l = \frac{q}{\omega Z_w}, \quad \mu_l = \frac{qZ_w}{\omega}. \quad (19)$$

Index l in these formulas means that parameters found in this way have to satisfy the locality limitations in all frequency regions where the strong spatial dispersion is absent.[12] This distinguish bulk material parameters retrieved through the *wave* impedance of the homogenized lattice [Eq. (18)] from nonlocal material parameters resulting from the same wave number q but the *surface* impedance as in the standard Nicholson, Ross, and Weir method of retrieval (see more details in Ref. 12).

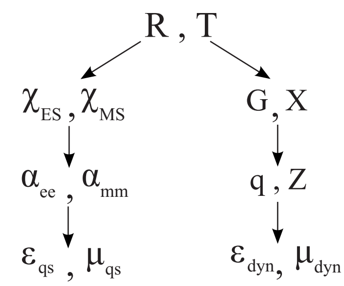

FIG. 4. Schematic algorithms of extraction of quasistatic (left) and dynamic (right) material parameters.

B. Retrieval of bulk material parameters

Thus, we have two algorithms of extraction of material parameters of volumetric metamaterial from the reflection and transmission coefficients of a single grid. The first one is the quasistatic retrieval suggested in Ref. 23. The second one suggested in the present paper is dynamic. Both are schematically shown in Fig. 4. In this paper, we will apply these algorithms to the lattice of effective nanorings introduced in Ref. 8.

To extract the material parameter, we got the reflection and transmission coefficients of a single layer of nanorings using full-wave numerical simulation with HFSS. Then the effective material parameters of both types of composites were obtained in compliance to the quasistatic and dynamic approaches schematically shown in Fig. 4. Their results are compared in Fig. 5. The dispersion plot presented in Fig. 5(b) helps to outline two resonant stop bands. The first one is related to the resonance of p-dipoles (nearly 580–625 THz). The second one is related to the resonance of m-dipoles (nearly 635–680 THz).

First, very important notice we can make from the plots is that retrieved quasistatic and dynamic material parameters are causal and passive for the both structures. As we already said this is a major reference point which tells us that both procedures give physically adequate results. Meanwhile, for the both structures the difference between quasistatic and dynamic effective material parameters is dramatic. First difference concerns the magnitudes of the electric and magnetic resonances, which are overestimated in the quasistatic model. This is very important particularly for the composites designed to achieve negative effective material parameters.

The next important difference is that the resonances of electric permittivity and magnetic permeability are not independent in the dynamic model. In the dynamic theory, we take into account the p-m interaction. It is a wave interaction between p-dipoles and m-dipoles, belonging to different crystal planes. For the permittivity, the main resonance (580–625 THz) which is nearly Lorentzian is supplemented with a

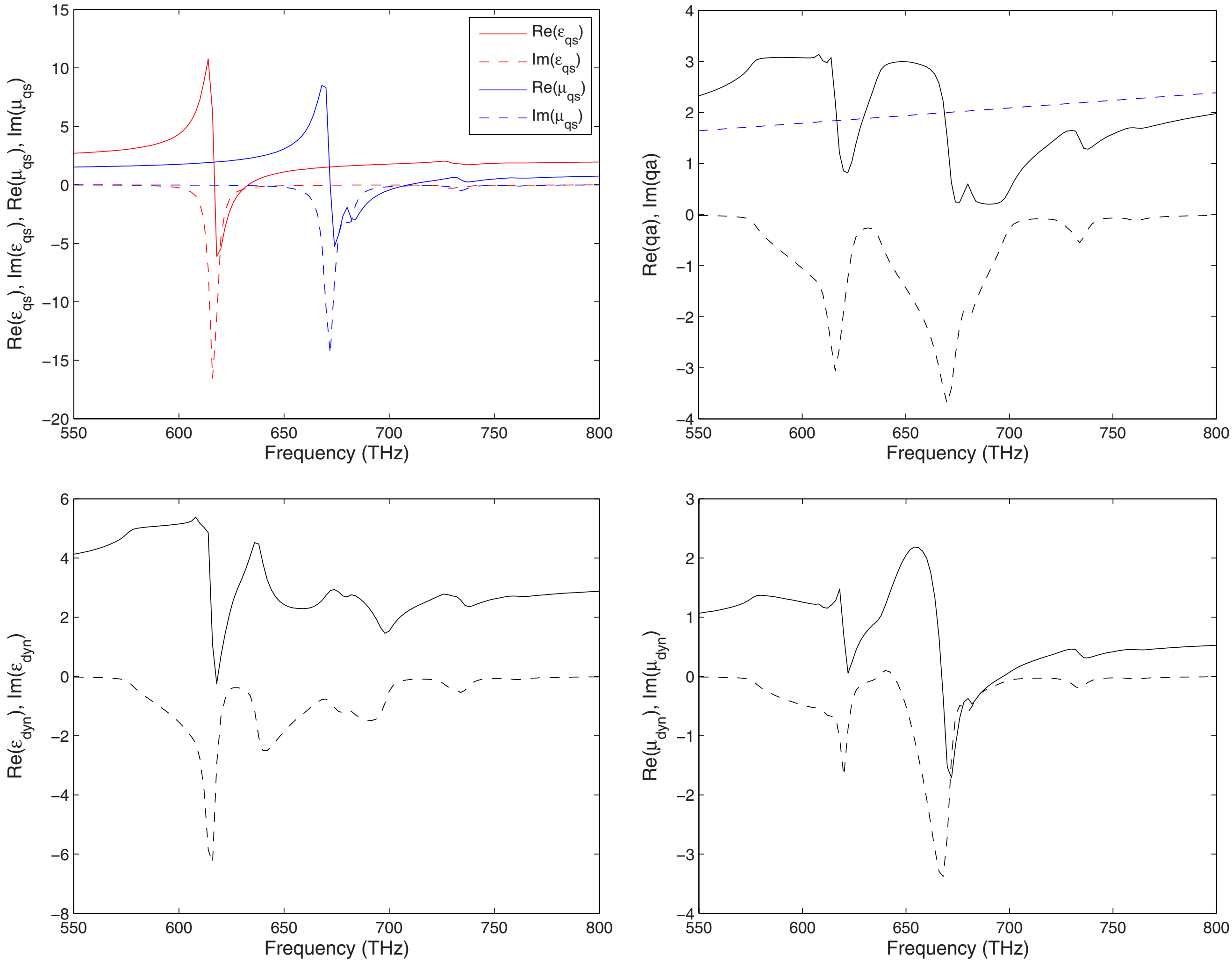

FIG. 5. (Color online) Effective material parameters of the composite media of effective nanorings of silver spheres with parameters $d=32$ nm, $r=38$ nm, and $a=96$ nm: (a) effective permittivity and permeability obtained in the quasistatic retrieval procedure (Ref. 23). (b) Normalized wave number versus frequency obtained in the dynamic retrieval model. The light line is shown by blue dashed color. (c) Effective permittivity obtained using the dynamic model. (d) Effective permeability obtained using the dynamic model. Real and imaginary parts of all parameters are depicted by solid and dashed lines, respectively.

weaker but rather pronounced resonance which corresponds to magnetic dipoles resonating at 650 THz. The third resonance of extracted permittivity which occurs above 700 THz is probably a spacial resonance of the lattice (where particles are not resonant). In other words, at frequencies higher than 670–680 THz the strong spatial dispersion appears and the extracted material parameters have no physical meaning.

The main resonance of permeability is supplemented with an additional broadband resonance produced by electric dipoles. Since the resonance bands of electric and magnetic dipoles of effective nanorings are close to one another, the overall shape of the frequency dispersion for both material parameters looks non-Lorentzian. We are not familiar with similar results in the literature, when both permittivity and permeability of any natural or artificial material are causal and passive but non-Lorentzian. The possibility for local material parameters to be non-Lorentzian gives us a distinct insight and seems to be an important result of the study.

Notice that the strong influence of the p-m interaction in the lattice of effective nanorings is not surprising when we recall that at 650 THz the lattice period a exceeds one third of the wavelength in the host medium. Comparing the period with effective wavelength $\lambda_{\mathrm{eff}}=2\pi/q$ (where q is taken beyond the resonance stop band) we can see in Fig. 5(b) that at 630 THz $qa=2.5$, i.e., $a=0.39\lambda_{\mathrm{eff}}$. However, even for so large (qa) in the passband 625–635 THz extracted material parameters are causal and passive.

Two important conclusions can be done. First, the quasistatic approximation which keeps adequate over the whole resonance band for a single metasurface of effective nanorings fails for the three-dimensional lattice. Second, the resonances of permittivity and permeability of the lattice of effective nanorings are related with one another due to the p-m interaction.

IV. CONCLUSIONS

This paper concerns the homogenization of metasurfaces related to the surface averaging approach and the homogenization of bulk lattices obtained by repetition of these metasurfaces, which is related to the bulk averaging approach. Using an example of a plasmonc bilayer, we have generalized a recently suggested procedure of the electromagnetic characterization of metasurfaces[22] to the case when the unit cell if formed by a group of resonant dipoles. We have shown that it is possible to consider such a bilayer as an array of effective nanorings of plasmonic nanospheres. It was checked that an individual effective nanoring can be considered as a group of p-dipoles forming a p-m pair, i.e., the pair of resulting electric and magnetic dipoles. High-order multipoles can be neglected in spite of the small distances (on the order of the sphere radius) between adjacent nanospheres in one nanoring and in spite of the small distances between adjacent effective nanorings (several times smaller than the size of cluster itself). The quasistatic approximation keeps valid with an acceptable accuracy well beyond the assumed initial quasistatic limit, practically up to the frequency at which the unit-cell size approaches to $0.34\lambda_{\mathrm{eff}}$ in the host medium.

The important result is that the close periodicity does not disqualify the effective description of metasurfaces in terms of the surface susceptibilities. However, the difference between metasurface and bulk medium layer is dramatic. One clearly fails to present a bilayer as an effective layer of a continuous bulk material. For the case of normal incidence, one can always introduce effective ε and μ which will be suitable only for this special case. However, they will not correctly describe the angular dependence of reflection and transmission coefficients. We would like to stress that in our opinion the description of metasurfaces in terms of ε and μ is physically incorrect and practically pointless.

For bulk lattices, we have suggested to extract the effective material parameters from reflection and transmission coefficient of a single grid representing a crystal plane of the lattice. This procedure is an expansion of the recently suggested method[23] to the dynamic case when the quasistatic approximation becomes inapplicable. The reason why the quasistatic model is not adequate for bulk lattices is the p-m interaction (that between electric and magnetic dipoles) which is carried by partial plane waves produced by crystal planes. It was shown that the suggested method gives effective material parameters which satisfy the locality conditions. However, the frequency behavior of the dynamic material parameters is more complex than simple Lorentzian dispersion predicted by the quasistatic approximation. Particularly, the resonances of electric permittivity and magnetic permeability become coupled with one another.

Now let us better clarify the ultimate aim of our retrieval process. As we already said, we want to obtain for the p-m lattice physically adequate material parameters satisfying the locality principle applicable beyond the quasistatic limitation. The main reason for utilization of these parameters is that we should be able to describe the electromagnetic properties of the composite for any angle of incidence. Otherwise our material parameters are redundant. The limitations of causality and passivity are necessary conditions of the locality of material parameters, i.e., if they are respected it does not guarantee that the same parameters keep for the oblique incidence. However, we hope to prove this in our next study in the same way as we did for a single grid of effective nanorings. Our expectation are related not only with locality limitations satisfied for retrieved material parameters. In this paper, we have shown that the same surface susceptibilities describe with acceptable accuracy the electromagnetic behavior of the metasurface of effective rings also for the oblique incidence.

The main problem with the application of bulk material parameters to a boundary problem is related to the necessity of transition layers. Beyond the quasistatic limit case $qa \ll 1$ bulk material parameters of a homogenized lattice are not consistent with Maxwell boundary conditions.[12] This inconsistency can be overcome using transition layers (or transition sheets) as it was done in Ref. 12 for a lattice of ceramic cylinders and for a lattice of split-ring resonators. For a lattice of effective nanorings transition layers require a serious separate study.

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