Transition from thin-film to bulk properties of metamaterials

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Effective material parameters for metamaterials are usually extracted for a single thin layer only. We show that, in general, these parameters cannot directly be assigned to a multilayered (bulk) metamaterial. The principal issue is the presence of higher-order Bloch modes. These modes are usually negligible for a single layer but can have considerable influence on the results for a periodically stacked structure. In particular, we show examples where a stack of single-layer negative refractive index metamaterials exhibits a positive index. Furthermore, on the basis of the dispersion relation of the respective Bloch modes, we derive criteria for the validity of the effective parameter approach.

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I. INTRODUCTION

Optical metamaterials (MMs) acquire their principal properties through their structure rather than by the intrinsic properties of the constituents. If the MM consists of unit cells much smaller than the wavelength of light, effective material parameters permittivity/permeability or refractive index/impedance can be derived because the light does not resolve the details of the structure. The geometry as well as the material of the unit cell can be varied to control the dispersion of these effective parameters to a certain extent. A variety of unit cell geometries have been suggested and fabricated. These include, e.g., the split-ring resonator, the single wire, the cut-wire pair, or the fish-net structure.[1–7] A frequently used approach for deriving the effective parameters is to invert the analytical expressions for the complex transmission and reflection coefficients of a respective homogeneous film.[8] These coefficients are usually calculated or measured from a single MM layer that consists of an identical unit cell periodically arranged in two dimensions. Nonetheless, this periodic arrangement is not a strict requirement, but merely made for practical purpose, in order to simplify the numerical characterization as well as the fabrication of such materials. For a true MM, the effective material parameters have to be independent of the period but should depend only on the filling fraction. A genuine bulk MM, which is the ultimate goal, can be constructed from a stack of such layers. Effective medium arguments would then imply that the same effective parameters can be assigned to this bulk MM. The realization of such a bulk MM at optical frequencies is one of the fundamental challenges in current research.[9] Recently, it was theoretically suggested to fabricate a bulk MM by stacking perforated metallic films which are mutually separated by dielectric spacers.[10] It was found that the structure supports at least one higher-order Bloch mode, which prevents an unambiguous definition of an effective refractive index. The structure was subsequently investigated experimentally.[11] There, the spectral domain, where a negative refractive index was observed, depended sensitively on the number of layers. This was explained by the conical shape of the functional layers. Nevertheless, the stacking approach seems presently to be the most promising approach to obtain bulk MMs, as recent experiments in the terahertz domain also suggested.[12]

In this paper, we analyze the properties of MMs at the transition from a thin-film MM, made of a single layer, to a bulklike structure that consists of multiple stacks of such MM layers. In particular, we systematically examine the effect of higher-order Bloch modes on the optical response of bulk MMs and the consequences for retrieving effective material parameters. To fully understand the response of the structure, the dispersion relation for the zeroth- and the first-order Bloch modes in the bulk MM is calculated. When deriving the effective material parameters for a single MM layer, the contribution of higher-order Bloch modes is frequently negligible. However, an external light field can couple to these Bloch modes. If these higher-order Bloch modes possess a lower imaginary part of their propagation constant, they are less attenuated upon propagation through the bulk medium and gain importance relative to the zeroth-order Bloch mode. Therefore, these modes may have a dominant effect on the optical reflection (transmission) behavior for the bulk MM. Because an appreciable influence of higher-order modes violates the assumptions of the retrieval algorithm, it is then inappropriate to derive, e.g., an effective refractive index.[13] In particular, this modification of the response can be so substantial that the bulk material may virtually exhibit a refractive index with an opposite sign when compared with the single layer.

A short remark has to be made concerning the use of the expression Bloch modes. In a periodic media with the period $\mathbf{R}$, the solutions of the wave equation obey the Floquet-Bloch theorem. Therefore, the eigenmodes (Bloch modes), which are connected to a wave vector $\mathbf{k}$ of the first Brillouin zone of the reciprocal lattice, can be written as modulated plane waves $\mathbf{E}_{\mathbf{k}}(\mathbf{r}) = e^{i\mathbf{kr}}\mathbf{u}_{\mathbf{k}}(\mathbf{r})$, e.g., for the electric field. Due

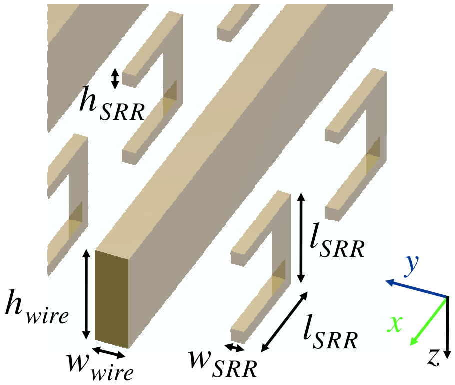

FIG. 1. (Color online) Outline of the geometry under consideration.

to the discrete translational symmetry, the amplitudes $\mathbf{u_k}(\mathbf{r}) = \mathbf{u_k}(\mathbf{r}+\mathbf{R})$ of the Bloch modes are periodic with the lattice.[14] Arbitrary wave fields that are propagating in such media can be written as a superposition of Bloch periodic waves.

It is important for the present study that the light propagation in the third direction can only be described in terms of Bloch modes fulfilling the full three-dimensional periodicity if a sufficiently large number of layers are used to make the considered MM a true bulk medium, and the problem of how an external light field can couple to these modes is of no importance. Only these fully evolved three-dimensional Bloch modes allow for the retrieval of meaningful effective material parameters for the bulk MM.

II. PROPERTIES OF THE SINGLE CONSTITUENTS

As an example, we study a MM consisting of split-ring resonators (SRRs) and metallic wires. The structure was chosen because it is assumed to be well understood. It allows for an easy arrangement in a three-dimensional unit cell of sufficient small spatial extension in all directions and the action of this unit cell is easily decomposed into two separate elements whose properties can be studied independently.[19] The metallic wire exclusively affects the effective permittivity of the MM, leaving the permeability unaltered. The SRR affects in the relevant spectral domain dominantly the effective permeability, leaving the permittivity unaltered. The effective refractive index of these MMs can then be derived from this single material parameter.

The basic geometry defining all relevant parameters is shown in Fig. 1. The wires have a width of $w_{\text{wire}}=100$ nm and a height of $h_{\text{wire}}=300$ nm. The square U-shaped SRRs have a size of $l_{\text{SRR}}=300$ nm, and both their width w_{SRR} and height h_{SRR} are 40 nm. The SRRs are oriented such that the electric field component is in the x direction and the wave vector in the z direction. Both are therefore in the SRR plane (see top of Fig. 2). The wave illuminates the structure at normal incidence. The unit cells are periodically arranged with a period of 400 nm in both lateral directions. The spatial extension of the layer in the z direction is assumed to be likewise 400 nm. This spatial extension was also taken into account for retrieving the effective material parameters of a hypothetically homogeneous slab. Substrate or dielectric host media were omitted because they will not qualitatively alter the optical properties analyzed in this work. For a dielectric host which does not show a strong dispersion in the spectral range of interest, the only impact is a shift of the resonance frequencies and a slight modification of the resonance strength. All simulations were performed by using the Fourier modal method.[15] A sufficiently large number of orders were retained in the simulation in order to ensure convergence of all quantities derived from the calculation. The material properties of gold were taken from Ref. 16.

The amplitude transmissions as functions of the number of layers for the two basic elements (metallic wire and SRR) are shown in Figs. 2(a) and 2(d). For both individual structures, the retrieved relevant effective material parameters are shown in Fig. 2 as a function of the number of layers.

For the wire medium, an exponential decrease of the transmission with increasing number of layers is observed. The effective permittivity is independent of the medium's thickness. The structure acts like a diluted metal and, in accordance with Drude's theory, the effective plasma frequency of this artificial metal is redshifted when compared to the bulk Au metal.[2] This holds for both the real and the imaginary parts, which are shown separately in Figs. 2(b) and 2(c).

For the SRRs, essentially three dips are observed in the analyzed spectral domain. The transmission drops exponentially with a linearly increasing MM thickness. These dips correspond to the first three plasmonic eigenmodes excited in the SRR.[17] The resonance at $\bar{\nu}_1=5000\ \text{cm}^{-1}$ is associated with a magnetic dipole response.[1] Thus, the effective permeability exhibits a Lorentz-type resonance. The second plasmonic eigenmode at $\bar{\nu}_2=8000\ \text{cm}^{-1}$ alters the effective permittivity due to an induced electric dipole. In the case of circular SRRs, the occurrence of these and further higher-order resonances was shown rigorously and quasianalytically in Ref. 18. However, these higher-order resonances are of no importance for further considerations.

III. PROPERTIES OF THE COMBINED CONSTITUENTS

A. Effective refractive index retrieved from reflection and transmission for a structure with a thick wire

The combination of an SRR and a metallic wire in a unit cell may result in a negative effective refractive index.[19] Figure 3 shows the transmitted amplitude and Fig. 4 demonstrates the real and the imaginary parts of the retrieved effective refractive index depending on the number of layers. This is in contrast to examples shown in Fig. 2. For the unit cell that combined the SRR and the thick wires, the transmission shows an exponential attenuation with an increasing MM thickness. Depending on the spectral position, the transmission changes as a function of the MM thickness, but the resonance positions, appearing now as transmission peaks, remain constant. For a single MM layer, a negative index of $n_{\text{eff}}=-2.2$ is obtained. However, with an increasing number of layers, the dispersion of the retrieved n_{eff} changes strongly and does not converge. This is in contrast to the examples before where the relevant effective material parameter was nearly independent of the number of layers. The spectral

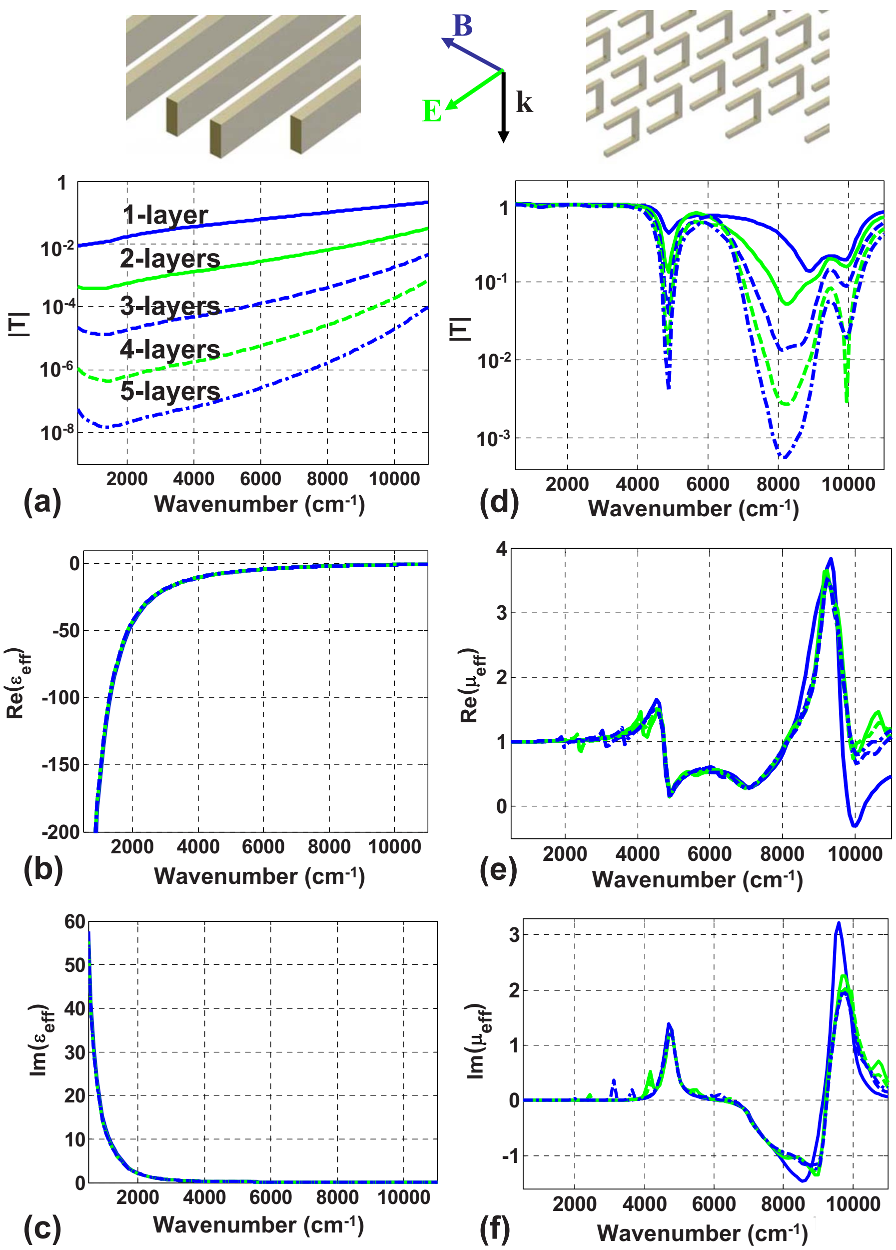

FIG. 2. (Color online) Transmitted amplitudes for a MM consisting of (a) metallic wires and (d) SRRs as a function of the number of layers. The real parts of the effective material parameter (permittivity or permeability) altered by each unit cell are shown in (b) and (e). Their respective imaginary parts are shown in (c) and (f). The same colors and types of lines as used in (a) appear in all subsequent figures to denote the number of layers (one layer = solid blue line; two layers = solid green line; three layers = dashed blue line; four layers = dashed green line; five layers = dashed-dotted blue line).

domain with $n_{\mathrm{eff}} < 0$ shifts toward lower frequencies. The dip in n_{eff} where the refractive index takes negative values becomes distorted and ultimately changes sign. For a MM made of four or five layers, n_{eff} is always positive. Although n_{eff} seems to converge toward a dispersion curve for a larger number of layers, these bulk properties cannot be derived unambiguously.

B. Ratio of the strength of Bloch modes

The origin of this behavior is the essential contribution of higher-order Bloch modes. The coupling of light to these higher-order modes is assessed by analyzing the amplitudes of higher diffraction orders, which are the Bloch modes of the subsequent free space. Although evanescent, their amplitudes may exceed those of the fundamental mode which corresponds to the zeroth diffraction order. The conclusions to be drawn are twofold. In general, it is not possible to deduce the effective refractive index of a bulk MM consisting of a stack of MM layers from the effective index of the single layer. Furthermore, it might be meaningless to assign an effective index to a bulk MM. This is not related to the particular retrieval algorithm but entirely caused by an intrinsic MM property, namely, the excitation of higher-order Bloch modes. Figure 5(a) shows the ratio of the amplitude of the first- and the zeroth-order transmitted Bloch modes as a function of the number of layers. For a single layer, the ratio reaches a maximum of 1.5. However, for an increasing number of layers, this ratio becomes as large as 13.

This increase is evoked at first by a coupling of the illuminating field to the zeroth-order and the higher-order Bloch modes. Although light couples dominantly to the zeroth-order Bloch mode, the losses of the higher-order Bloch modes are weaker than those of the zeroth-order Bloch mode. This will be shown subsequently by calculating the

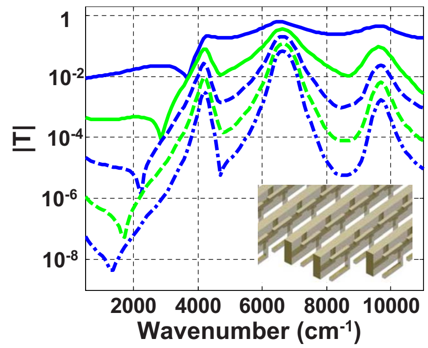

FIG. 3. (Color online) Transmission for a medium whose unit cell comprises an SRR and a thick metallic wire (see inset).

dispersion relation of the two lowest-order Bloch modes for the bulk MM. Therefore, although both modes (zeroth and first orders) show a decrease in the transmitted amplitude, the zeroth-order mode decreases quicker with increasing number of layers. Because the assumption of a single forward and backward propagating plane wave in any parameter retrieval algorithm is obviously violated, it is not meaningful to extract effective material parameters in this operational regime.

There are two solutions to this issue. Either one has to prevent the excitation of higher-order modes by an appropriate MM design, if one aims to retain the concept of effective material parameters, or one has to describe the optical properties of the bulk MM entirely by the dispersion relation of normal modes in this medium, similarly as for photonic crystals.[14]

Here, we demonstrate two examples for the first possible solution, namely, the increase of the losses associated with higher-order Bloch modes to retain the concept of effective material parameters. As the qualitative change of the effective material parameters was not observed by analyzing the two unit cells' constituents alone, we may attribute the low attenuation of higher-order Bloch modes to the electromagnetic coupling of the wire and the SRR in their combined unit cell. This coupling is well pronounced in the present geometry, as the plasmonic eigenmode of the SRR shows a strong field confinement in the SRR side arms.[20] Because the size of the wires is rather large, they are in close proximity to these side arms. The perturbation of the SRR eigenmode is therefore strong and coupling results. The distance between the SRR arms and the wire medium can be identified as the critical parameter. For a reduced wire thickness or a larger period, the coupling becomes weaker.

C. Effective refractive index retrieved from reflection and transmission for a structure with a thin wire

We show first that the losses of the higher-order modes can be increased if the height of the wires is reduced. Figure 5(b) shows the amplitude ratio of the first- to the zeroth-order Bloch mode for a reduced height of the wires of 100 nm [instead of 300 nm as in Fig. 5(a)]. Although for a larger number of layers the amplitude ratio grows slightly, it never exceeds 0.5. The effective refractive index of the medium n_{eff} and its impedance are depicted in Fig. 6. Although slight modifications are observable for n_{eff}, the real part of the refractive index converges for an increasing number of layers toward −0.8. Likewise, the impedance shows a quick convergence toward a bulk value. As required for a passive medium, the imaginary part of the effective refractive index and the real part of the effective impedance are positive in the entire spectral domain. The effective material parameters have converged if at least three layers are present. Thus, the conclusion to be drawn is that light propagation in this bulk MM is essentially governed by the zeroth-order Bloch mode.

D. Effective refractive index retrieved from the dispersion relation for a structure with a thick wire

To verify the assumption that the presence of higher-order Bloch modes with a low imaginary part in the propagation constants prohibits the assignment of effective material parameters from a single-layer MM, we have calculated the dispersion relation of these Bloch modes of the bulk MM. The analysis starts with the MM where the unit cell is comprised of the SRR and the thick wire. Their dispersion relation will be compared with that of a MM where thick wires were replaced by thin ones. This corresponds exactly to the previously discussed cases.

In this simulation, the unit cell is periodically arranged in all directions (period of 400 nm); hence, the true bulk MM properties can then be derived. In order to calculate the dispersion relation, we have extended the Fourier modal method to calculate first the transfer matrix of the unit cell in Fourier space and second the eigenvalues and eigenvectors of this transfer matrix. For a frequency ω and a wave vector with given in-plane components (k_x and k_y), the algorithm calculates the corresponding longitudinal wave vector component k_z of the eigenmodes. The advantage of the method as compared to classical plane wave expansion techniques is the formulation of the problem in frequency space. This permits the retention of the dispersion of an experimental dielectric function of gold.

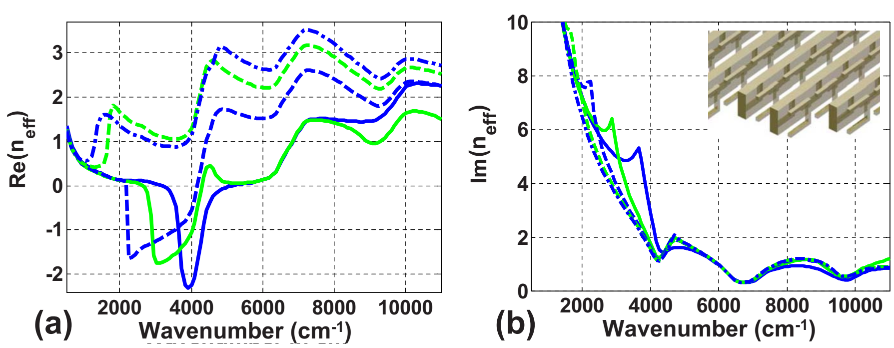

FIG. 4. (Color online) (a) Real and (b) imaginary parts of the effective refractive index retrieved from a medium whose unit cell comprises an SRR and a thick metallic wire (see inset).

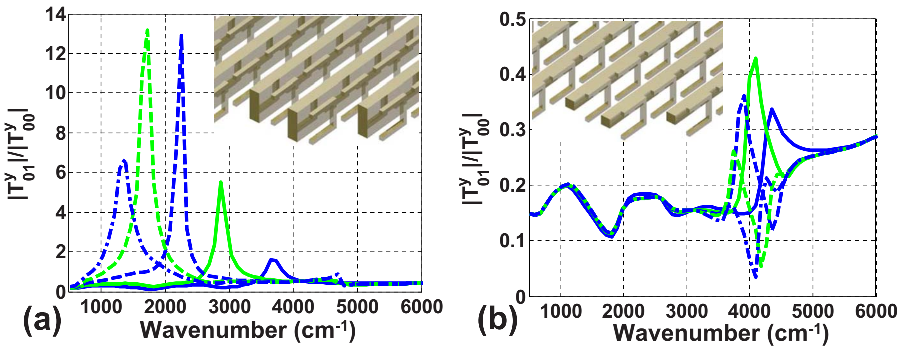

FIG. 5. (Color online) Ratio of the amplitudes of the first-order Bloch mode, which is evanescent, and the zeroth-order Bloch mode for a medium made of SRRs and thick wires in (a) and for a medium made of the same SRRs but with thinner wires in (b).

Figures 7(a) and 7(b) show the real and imaginary parts of the wave vector for the two lowest-order eigenmodes of the unit cell with a thick wire. We restrict the propagation constant shown in the figure to those having a positive sign in the imaginary part. Wave vectors having the opposite sign appear as well but are omitted. The eigenmodes were labeled in accordance with their increasing imaginary part of the wave vector at small frequencies. The sketch on top of the figure indicates the geometry. We have chosen a point in k space with $k_x=0$ and $k_y=0$, being identical to the illumination direction of the aforementioned configuration. The electric field polarization of the eigenmodes is dominantly parallel to the SRR. The eigenmodes for the orthogonal polarization are only weakly influenced by the structure in the relevant spectral domain and follow closely a linear relation. This corresponds to the light line in a medium with a homogeneous refractive index without resonances in the dispersion relation.

For the structure comprising the SRR and the thick wires, the dispersion relation indicates that the first-order eigenmode (green stars) with wave numbers slightly above $1000\ \mathrm{cm}^{-1}$ up to $5000\ \mathrm{cm}^{-1}$ has indeed a smaller imaginary part in the propagation constant compared to the zeroth-order eigenmode (blue circles). Therefore, this higher-order mode dictates the properties of light propagation in the structure. It is therefore not justified to assign effective material parameters to the structure in this spectral domain. This is the same conclusion which we have drawn from the analysis of the amplitude of the transmitted higher-order Bloch mode, as shown in Fig. 5. The lowest-order mode dominates only for frequencies smaller than approximately $1200\ \mathrm{cm}^{-1}$. Therefore, this is the only spectral domain where effective material

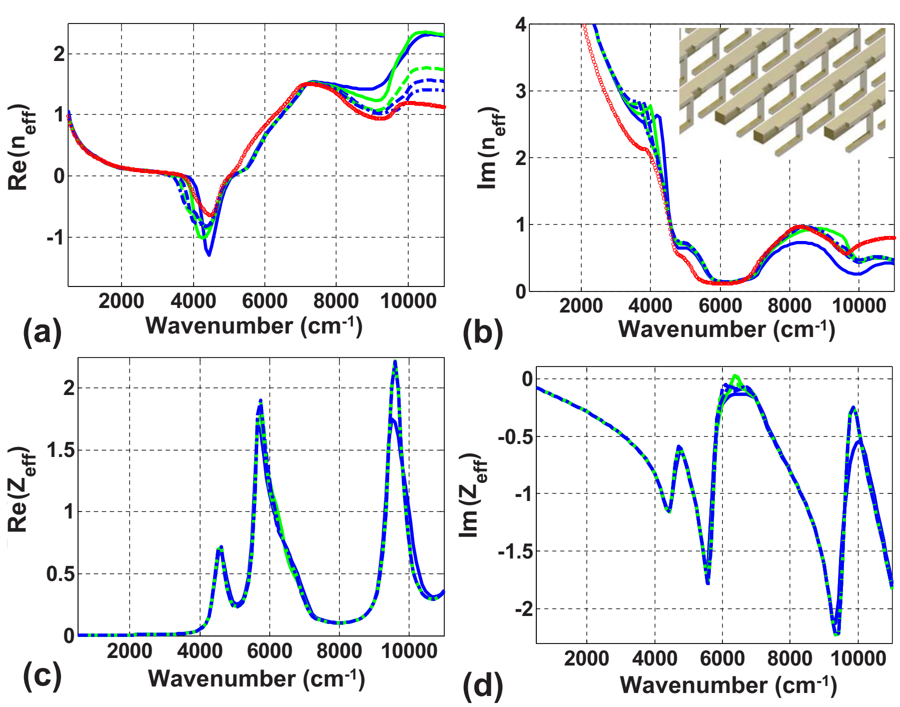

FIG. 6. (Color online) (a) Real and (b) imaginary parts of the effective refractive index and (c) real and (d) imaginary parts of the effective impedance retrieved for a medium whose unit cell comprises SRRs and metallic wires, as shown in the inset. The parameter retrieval was performed for several numbers of layers. The red dots in (a) and (b) denote the refractive index as deduced from the dispersion relation of the zeroth-order Bloch mode.

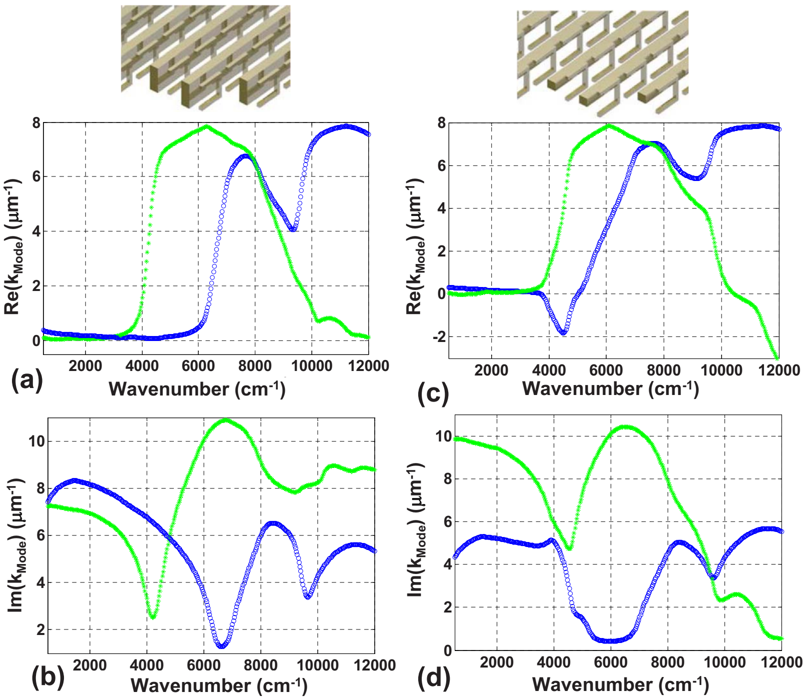

FIG. 7. (Color online) Effective propagation constant of the two lowest-order Bloch modes propagating in the unit cell comprising [(a) and (b)] the SRR and the thick wire and [(c) and (d)] the SRR and the thin wire. Real parts of the propagation constants are shown in (a) and (c) and imaginary parts are shown in (b) and (d). Blue circles correspond to the zeroth-order Bloch mode and green stars to the first-order Bloch mode.

parameters can be assigned to the structure. This can also be deduced from Fig. 4, where the refractive index shows no variation as a function of the number of layers below 1200 cm^{-1}. For the sake of simplicity, we have shown in Fig. 7 only the two lowest-order eigenmodes. Nevertheless, at frequencies larger than 5000 cm^{-1}, further modes exist. Their imaginary part is comparable to the lowest-order Bloch mode. This prohibits the assignment of effective material parameters in this spectral domain. A further result of the dispersion relation is the real part of the propagation constant, as shown in Fig. 7(a). Although it cannot be interpreted as an effective refractive index multiplied by the free space wave number, the real part for this propagation constant remains entirely positive.

E. Effective refractive index retrieved from the dispersion relation for a structure with a thin wire

We have found that the assignment of an effective refractive index is not possible for the structure that is composed of the SRR and the thick wire in the unit cell. We have shown that the effective refractive index as deduced from reflection and transmission does not converge with an increasing layer number. This behavior was attributed to the presence of higher-order Bloch modes with smaller imaginary parts in the propagation constant when compared to the lowest-order Bloch mode.

This discussion changes for the structure whose unit cell is made of the SRR and a thin wire. The dispersion relations of the real and imaginary parts of the wave vectors for two lowest-order eigenmodes are shown in Figs. 7(c) and 7(d), respectively. Most importantly, the imaginary part of the wave vector is smallest for the zeroth-order eigenmode in the relevant spectral domain, although at the resonance the imaginary parts have nearly the same strength for both modes. Furthermore, higher-order modes have an even larger imaginary part in the entire spectral domain. This dominance of the zeroth-order Bloch eigenmode allows for the assignment of effective material parameters. Figure 7(c) shows that the wave vector's real part of the zeroth-order Bloch mode changes its sign in the frequency domain between 4000 and 5000 cm^{-1}. This is exactly the spectral domain where the parameter retrieval based on reflection and transmission predicted a negative refractive index. Indeed, extracting the refractive index from the effective propagation constant of the lowest-order Bloch mode, as shown in Figs. 6(a) and 6(b), reveals that the effective refractive index of the bulk medium is in excellent agreement with the refractive index deduced from the structure with an increasing number of layers. The agreement is better for the structures with an increasing layer number, as naturally expected.

We conclude, from the considerations of these two examples, that only for those MMs made of SRR and thin wires, effective material parameters can be meaningfully deduced. For the structure with thick wires, the coupling between the unit cell constituents is too strong. This coupling mediates the excitation of higher-order Bloch modes with a smaller imaginary part of their wave vectors. As they tend to dominate the spectral properties of this MM, effective material parameters cannot be deduced.

F. Influence of the coupling among the unit cell constituents

An alternative approach to reduce the coupling between the single elements is to increase the distance between the two constituents of the unit cell. This can be achieved by increasing the period (note that the wires are always placed in the center between two SRRs) but retaining otherwise all geometrical parameters. Therefore, we have chosen a period of 600 nm instead of 400 nm as before. Results of the pa-

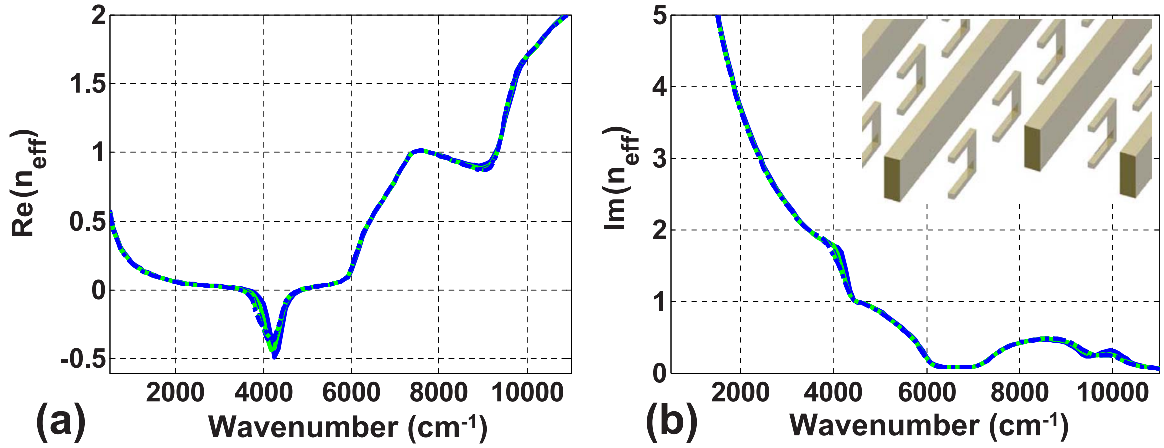

FIG. 8. (Color online) (a) Real and (b) imaginary parts of the effective refractive index retrieved for a medium whose unit cell comprises SRRs and thick metallic wires, as shown in the inset. Geometrical details of the elements in the unit cell were not altered but only the period was increased by 200 nm as compared to the results shown in Fig. 4; hence, the period was 600 nm. The parameter retrieval was performed for several numbers of layers the MM consists of.

rameter retrieval as a function of the layer number are shown in Fig. 8.

For such a structure, where the coupling among the constituents is reduced, the effective material parameters as derived from a single thin-film layer are identical to those derived from a multilayer stack. The strength of the negative real part of the refractive index is lower because the metal content in the structure was further reduced by increasing the period. This lowers the resonance strength in the dispersion. An additional band structure analysis revealed that for this structure, the fundamental mode has the lowest imaginary part of the propagation constant. Hence, the assignment of effective material parameters is possible. For this structure, the parameters as deduced from a single layer are in good approximation equal to those derived for the bulklike multilayer structure.

IV. REMARKS ON THE GENERALITY OF THE ANALYSIS

Although the analysis was done with the example of a negative index material whose unit cell comprises an SRR and a metallic wire medium, the conclusions drawn apply to every type of MM. Namely, effective material parameters of a bulk MM cannot necessarily be deduced from the properties of a single MM layer. The parameters agree better if the propagation losses of higher-order Bloch modes are larger.

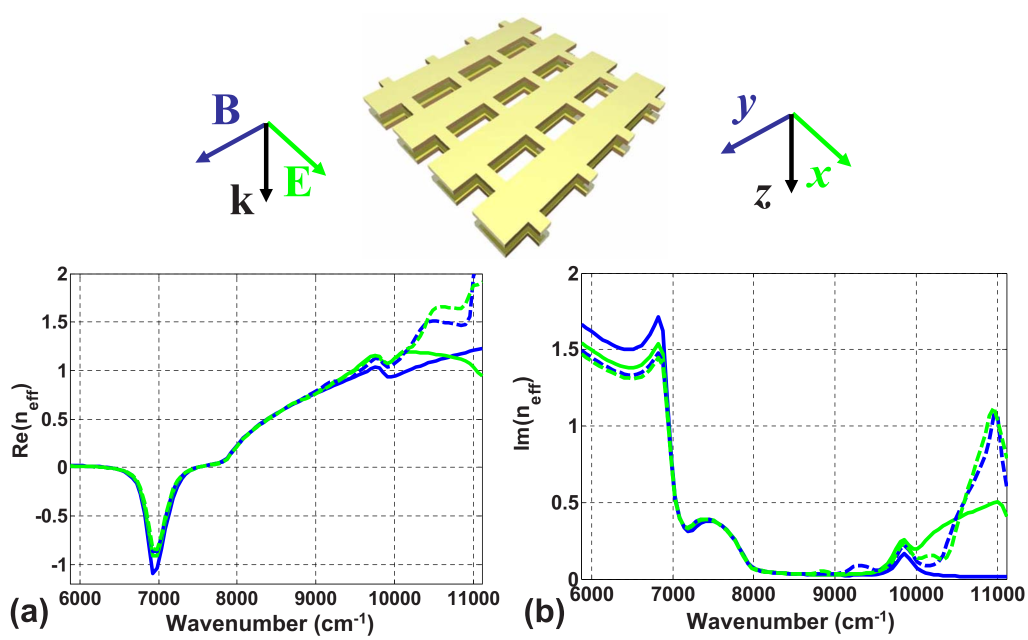

FIG. 9. (Color online) (a) Real and (b) imaginary parts of the effective refractive index retrieved for a fish-net structure as a function of the number of the layers (one layer=solid blue line; two layers=solid green line; three layers=dashed blue line; four layers=dashed green line). The coordinate system, the chosen polarization, and the geometry are shown on top of the figure. Geometrical details of the structure were adopted from values published in literature (Ref. 23) and are detailed in the text.

Only in this case can the effective material parameters of the MM be deduced from the dispersion relation of the Bloch mode with the lowest imaginary part of the propagation constant.

The dominance of a single Bloch mode was found to be a property of the fish-net structure. This will be shown briefly below. This structure is of particular relevance as it is easy to fabricate, and design parameters were found to cause a negative refractive index at truly optical frequencies.[21,22] This structure seems, in addition, to be a promising candidate to be realized as a three-dimensional bulk MM.[11]

Figure 9 shows the retrieved effective material parameters as a function of the number of layers for a fish-net structure. Geometrical parameters were taken from literature.[23] The structure is a three layer system made of Ag (h_{Ag}=45 nm), MgF_2 (h_{MgF_2}=30 nm, n_{MgF_2}=1.38), and Ag (h_{Ag}=45 nm). The fish net is made of two perpendicular wires having widths of w_x=316 nm and w_y=100 nm. The period of both lateral directions is $\Lambda_x=\Lambda_y$=600 nm. This three layer structure was stacked in the third dimension with a period of Λ_z =200 nm. A substrate was omitted in the present consideration.

Although slight modifications occur in the spectrum, the retrieved effective refractive index converges in both real and imaginary parts if three MM layers are present. Although not shown, this refractive index is in excellent agreement with the refractive index as deduced from the dispersion relation of the lowest-order Bloch mode. Only for frequencies larger than 9000 cm^{-1} a convergence is not observed. This is the spectral domain where higher-order Bloch modes contribute equally to the light transport in the bulk MM. Then, effective material parameters cannot be meaningfully retrieved.

Nevertheless, problems in the assignment of bulk material parameters can always occur if they are deduced from those of a single film. Therefore, one must be careful in such assignments, independent of the particular implementation of the unit cell and the spectral range. Clear discrepancies have been encountered in the literature, e.g., in the gigahertz domain, as reported in Ref. 24.

V. CONCLUSIONS

In conclusion, we have addressed the issue of deducing bulk MM effective parameters from their single-layer counterparts. It was shown that the concept of effective material parameters can only be retained if the amplitudes of higher-order Bloch modes do not exceed that of the fundamental mode. It is important to underline that this behavior might not be observable in the transmission (reflection) response of a single MM layer. This is particularly true if the fundamental mode is more strongly attenuated than the higher-order Bloch modes. This can be deduced from the calculation of the Bloch mode dispersion relation. These results will be useful for the design of bulk MMs, which is a mandatory step toward the implementation of real-world MM applications, e.g., for the design of cloaking devices or perfect lenses at optical frequencies.

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