Validity of effective material parameters for optical fishnet metamaterials

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Although optical metamaterials that show artificial magnetism are mesoscopic systems, they are frequently described in terms of effective material parameters. But due to intrinsic nonlocal (or spatially dispersive) effects it may be anticipated that this approach is usually only a crude approximation and is physically meaningless. In order to study the limitations regarding the assignment of effective material parameters, we present a technique to retrieve the frequency-dependent elements of the effective permittivity and permeability tensors for arbitrary angles of incidence and apply the method exemplarily to the fishnet metamaterial. It turns out that for the fishnet metamaterial, genuine effective material parameters can only be introduced if quite stringent constraints are imposed on the wavelength/unit cell size ratio. Unfortunately they are only met far away from the resonances that induce a magnetic response required for many envisioned applications of such a fishnet metamaterial. Our work clearly indicates that the mesoscopic nature and the related spatial dispersion of contemporary optical metamaterials that show artificial magnetism prohibits the meaningful introduction of conventional effective material parameters.

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The introduction of nanostructured metamaterials (MMs) into optics potentially opens the door to a fairly comprehensive control of light propagation. During the past several years much effort has been devoted to achieve this goal and two major research fields may be distinguished. On the one hand, advances in nanotechnology provide ever smaller and more complex structures which constitute quite involved nanostructured media. On the other hand, optics in media with unprecedented effective material equations has been investigated purely theoretically and surprising effects have been revealed.[1] The desirable assignment of effective material parameters to a specific MM would bridge the gap between both approaches and allow to link a fabricated structure to a particular effective constitutive relation.[2–4] Furthermore it would appreciably facilitate the description of light propagation in optical MMs and their combination with other optical materials because canonical Maxwell boundary conditions could be applied. If we wish to understand under a MM an artificial medium made of periodically or nonperiodically arranged meta atoms which allows to control the properties of light propagation predominantly by the chosen geometry of the meta atoms, many different MMs can be envisioned which all have peculiar aspects if effective properties shall be designed.[5–7] To avoid any misunderstanding, we wish to restrict our considerations in the following on metamaterials that show the effect of an artificial magnetism and which shall operate at optical frequencies. Such property is often at the focus of interest since the media would enable optical phenomena that contradict our common perception of how light propagates.

For such special type of MM, there is, however, a serious issue which might prevent this simplified description in terms of effective material parameters. In general, typical optical MMs are mesoscopic where the vacuum wavelength is only a few times larger than the unit cell. In such systems the optical response may be reasonably described by induced currents, which nonlocally depend on the electric field. In Fourier space this leads to a spatially dispersive conductivity, as discussed in the work of Serdyukov *et al.*[8] At this stage it is not required to distinguish between polarization ($\sim\frac{\partial}{\partial t}\mathbf{P}$) and magnetization currents ($\sim\nabla\times\mathbf{M}$), however, this becomes important if either of them becomes resonant in the nanostructure. Provided that this spatial dispersion is weak, the constitutive relation between $\mathbf{j}$ and $\mathbf{E}$ can be expanded up to the second order. Since the fields $\mathbf{D}$ and $\mathbf{H}$ cannot be defined uniquely, spatial derivatives of $\mathbf{E}$ may be replaced in favor of $\mathbf{B}$. As a result, two constitutive relations $\mathbf{D}(\mathbf{E},\mathbf{B})$ and $\mathbf{H}(\mathbf{E},\mathbf{B})$ emerge with tensorial, but only frequency-dependent coefficients.[8] First-order terms lead to magnetoelectric ($\mathbf{E}\rightleftarrows\mathbf{H}$) coupling (bianisotropy and chirality). Second-order terms to anisotropic, but spatially nondispersive relations between both $\mathbf{D}$ and $\mathbf{E}$ and $\mathbf{H}$ and $\mathbf{B}$. To sum up, these so-called bianisotropic constitutive relations are the most general ones for a weak spatially dispersive conductivity in a nanostructured material. From a physical point of view it is appealing that the nonlocal relation between the electric field and the induced currents is the very source for the effective chiral and magnetic $[\hat{\mu}(\omega)]$ properties of MMs. From a technical point of view, with these spatially nondispersive constitutive relations at hand, standard boundary conditions[9] can be used to solve macroscopic Maxwell's equations in layered media. This has a big advantage compared to the rather involved procedure for spatially dispersive constitutive relations which require the use of so-called additionally boundary conditions.[10] In mirror-symmetric (nonchiral) media first order terms in the expansion vanish and the magnetoelectric coupling disappears. The MM may then be described by two material tensors $\hat{\varepsilon}(\omega)$ and $\hat{\mu}(\omega)$.

Here we aim at introducing a simple criterion that tells us if this condition is fulfilled. In general, one has to develop an approach to retrieve these tensor elements from reflection/

transmission data. However, it will turn out that the very calculation of these parameters is not required. Only if the criterion is fulfilled one has to proceed with the retrieval algorithm to calculate effective parameters which are then independent of the incidence angle and may be termed effective *material parameters*. On the other hand, if the criterion is not fulfilled the assignment of an effective permittivity $\hat{\varepsilon}(\omega)$ and permeability $\hat{\mu}(\omega)$ is pointless. This means physically that the assumption of weak spatial dispersion is violated and the effective material parameters would become spatially dispersive making the approach used inconsistent. Thus the aim of this work is not the retrieval of parameters but to evaluate if a certain MM may be described by effective material parameters.

But in any case the recently introduced retrieval approach for isotropic materials at arbitrary incidence[11] has to be generalized toward anisotropic media. The main advantage of the present approach is that all tensor elements can be determined without requiring explicitly that the propagation direction coincides with a crystallographic axis. Hence, the approach may be applied to all currently fabricated MMs.

To start with, we assume that the metallic structures (split ring, fishnet, etc.) involved are reciprocal and not intrinsically magnetic ($\mu_{\text{metal}}=1$). Their response to the electromagnetic field can be completely described by a current nonlocally induced by the electric field as[8]

$$\mathbf{j}(\mathbf{r},\omega) = \int_V \hat{R}(\mathbf{r},\mathbf{r}',\omega)\mathbf{E}(\mathbf{r}',\omega)d\mathbf{r}', \tag{1}$$

where the dyadic $\hat{R}(\mathbf{r},\mathbf{r}',\omega)$ describes the nonlocal response of the medium. However this approach is not practical. For weak nonlocality (or spatial dispersion) one may rather expand Eq. (1) up to the second order. For improving the readability of the paper we provide our derivation in following the lines in Ref. 8. We obtain

$$j_k(\mathbf{r},\omega) \approx i\omega\left[a_{kl}E_l + b_{klm}\frac{\partial E_l}{\partial x_m} + c_{klmn}\frac{\partial E_l}{\partial x_m\,\partial x_n} + \cdots\right], \tag{2}$$

where Einstein notation is applied and $b_{klm}=-b_{lkm}$. The factor $i\omega$ has been introduced for convenience, so that the terms in the brackets represent the induced polarization. Now the constitutive relations read as

$$D_k(\mathbf{r},\omega) = (\varepsilon_0\delta_{kl} + a_{kl})E_l + b_{klm}\frac{\partial E_l}{\partial x_m} + c_{klmn}\frac{\partial^2 E_l}{\partial x_m\,\partial x_n}, \tag{3}$$

$$\mathbf{H} = \mathbf{B}/\mu_0. \tag{4}$$

The first term leads to the anisotropic permittivity $\varepsilon_{kl}=\delta_{kl}+a_{kl}/\varepsilon_0$. The second term accounts for magnetoelectric coupling and vanishes for media with three orthogonal planes of mirror symmetry, i.e., media that are purely anisotropic in the quasistatic limit. To proceed we require the coefficients c_{klmn} to obey the following relation:

$$c_{klmn}\frac{\partial^2 E_l}{\partial x_m\,\partial x_n} \stackrel{!}{=} [\nabla \times (\hat{\gamma}\nabla \times \mathbf{E})]_k \tag{5}$$

where the components γ_{ij} are related to the coefficients c_{klmn}. Since $c_{klmn}=c_{lkmn}$ holds, the tensor $\hat{\gamma}$ is symmetric ($\gamma_{ij}=\gamma_{ji}$).

Because Maxwell's equations are invariant with respect to the transformations[8] $\mathbf{D}'=\mathbf{D}+\nabla\times\mathbf{Q}$, $H'=\mathbf{H}-i\omega\mathbf{Q}$ we can rewrite Eqs. (3) and (4) in using $\mathbf{Q}(\mathbf{r},\omega)=-\hat{\gamma}\nabla\times\mathbf{E}=i\omega\hat{\gamma}\mathbf{B}$ to obtain the ultimate constitutive relations

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

These equations represent our point of departure. They reflect that a mesoscopic metallic structure with a weak nonlocal response (weak spatial dispersion) can be likewise treated as an effective homogeneous, anisotropic but magnetic medium where the magnetic properties merely originate from the nonlocal response $\hat{R}(\mathbf{r},\mathbf{r}',\omega)$,

$$\mu_{kl} = (\delta_{kl} - \mu_0\omega^2\gamma_{kl})^{-1}. \tag{7}$$

Only if the constitutive relations of a medium without bianisotropy can be cast in the form of Eqs. (6), the usual boundary conditions are applicable where the tangential components E_t and H_t as well as the normal components D_n and B_n are continuous.

The strategy of our work is as follows: we develop a retrieval algorithm for anisotropic, homogeneous, and local media which relies on the rigorously calculated reflection/transmission data from a MM slab for transverse wave vectors that extend even into the evanescent domain. Within this algorithm we identify a quantity $\alpha(\omega)$ which contains the essential information. If this quantity depends only on frequency and not on the wave vector, the effective permittivity and permeability tensor elements, not calculated at this step, will exhibit the same feature and can be calculated in a further step and assigned to the metamaterial. If this is not the case the ratio unit cell size/wavelength has to be decreased until this criterion is met. Here we restrict ourselves to media where both material tensors can be simultaneously diagonalized as

$$\hat{\varepsilon} = \text{diag}\{\varepsilon_x,\varepsilon_y,\varepsilon_z\}, \quad \hat{\mu} = \text{diag}\{\mu_x,\mu_y,\mu_z\}. \tag{8}$$

We align the coordinate system and therefore all interfaces to the crystallographic axis of the effective anisotropic medium. The incident light shall consist of monochromatic plane waves whose wave vector is perpendicular to at least one coordinate axis. Then the eigenmodes of the medium can be decomposed into decoupled TE and TM modes and the reflection/transmission problem is equivalent to that of an isotropic medium. The only difference is the different propagation constant for each eigenmode. For a particular eigenpolarization the reflection and transmission coefficients in terms of the normal wave-vector component have the same form as in the isotropic case. For varying the incidence plane and the polarization only certain quantities have to be exchanged as indicated in Table I. The transmission and reflection coefficients for the electric field read as

TABLE I. Substitution table for the relevant coefficients depending on the polarization and the incidence plane.

	TE		TM	
	$k_x=0$	$k_y=0$	$k_x=0$	$k_y=0$
α	$1/\mu_y$	$1/\mu_x$	$1/\varepsilon_y$	$1/\varepsilon_x$
β	$\varepsilon_x\mu_y$	$\varepsilon_y\mu_x$	$\varepsilon_y\mu_x$	$\varepsilon_x\mu_y$
γ	μ_y/μ_z	μ_x/μ_z	$\varepsilon_y/\varepsilon_z$	$\varepsilon_x/\varepsilon_z$

$$T(k,\xi) = \frac{2k_s\xi A}{\xi(k_s+k_c)\cos(kd) - i(\xi^2+k_sk_c)\sin(kd)}, \quad (9)$$

$$R(k,\xi) = \frac{\xi(k_s-k_c)\cos(kd) + i(\xi^2-k_sk_c)\sin(kd)}{\xi(k_s+k_c)\cos(kd) - i(\xi^2+k_sk_c)\sin(kd)}, \quad (10)$$

where the following abbreviations have been used,

$$k_{s,c} = \alpha^{s,c}k_z^{s,c}, \quad k = k_z^f, \quad A_{\mathrm{TE}} = 1, \quad A_{\mathrm{TM}} = \sqrt{\frac{\varepsilon^s\mu^c}{\varepsilon^c\mu^s}}, \quad (11)$$

$$\text{with } \xi = \alpha^f k_z^f, \quad \text{where } k_z^i = \sqrt{\frac{\omega^2}{c^2}\beta^i - k_t^2\cdot\gamma^i}, \quad (12)$$

is the normal component of the wave vector in medium "i" and $k_t=(k_x,k_y)$ is its conserved tangential component. The superscripts $i\in\{s,f,c\}$ denote substrate, film and cladding. In what follows we assume that substrate and cladding are isotropic and nonmagnetic where $\alpha_{\mathrm{TE}}^{s,c}=1$, $\alpha_{\mathrm{TM}}^{s,c}=1/\varepsilon^{s,c}$, $\beta^{s,c}\equiv\varepsilon^{s,c}$, and $\gamma^{s,c}\equiv 1$. For the sake of clarity we drop the superscript 'f' and write $\alpha^f(\omega)=\alpha(\omega)$, $\beta^f(\omega)=\beta(\omega)$, and $\gamma^f(\omega)=\gamma(\omega)$. These coefficients are related to different combinations of tensor components of the permittivity and permeability depending on the polarization and the incidence plane, see Table I.

For the sake of brevity the frequency dependence will be kept in mind but not explicitly written in the following.

The effective material slab is then fully characterized by the parameters k and ξ. Note that throughout the paper k is the normal component of the wave vector in the slab. By inverting Eqs. (9) and (10) one obtains

$$kd = \pm\arccos\left(\frac{k_s(1-R^2)+k_c(T/A)^2}{(T/A)[k_s(1-R)+k_c(1+R)]}\right) + 2m\pi \quad (13)$$

with $m\in\mathbb{Z}$ and

$$\xi = \pm\sqrt{\frac{k_s^2(R-1)^2 - k_c^2(T/A)^2}{(R+1)^2-(T/A)^2}}. \quad (14)$$

The sign of k and ξ and the branch order m are determined by the usual physical constraints.[11] The quantities k and ξ can be uniquely determined and are the final *effective wave parameters*. They are independent of the thickness d of the slab, provided that they already converged toward the bulk data.[12] Then these wave parameters must coincide with those provided by the dispersion relation of the fundamental Bloch mode.[13] It is evident that these effective wave parameters describe properties of the fundamental Bloch mode in the infinite lattice formed by periodically arranging a single fishnet layer.[14,15] They still depend on the propagation direction, the angle of incidence and the polarization state as in any anisotropic medium.

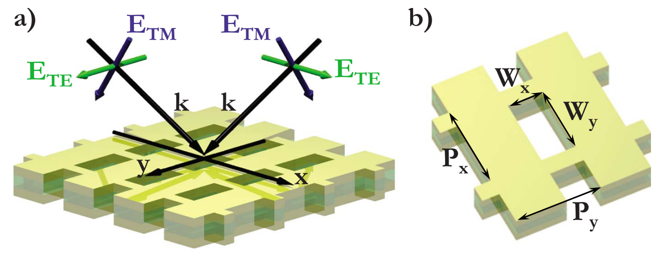

FIG. 1. (Color online) (a) Schematic view of the single fishnet layer together with the four principal directions for the retrieval. (b) Unit cell of the fishnet with $P_x=P_y=600$ nm, $W_x=284$ nm, and $W_y=500$ nm embedded in air. The thicknesses of the silver and the intermediate MgF_2 ($n=1.38$) layer are $d_{\mathrm{Ag}}=45$ nm and $d=30$ nm, respectively.

Now the criterion central to this work can be formulated. Since $\alpha(\omega)=\xi/k$ [see Eq. (12)] is related to the effective material parameter tensor components (see Table I) it has to be independent of the angle of incidence although ξ and k will strongly depend on it. Actually this is a very simple criterion that we can use to evaluate the validity of the effective material approach just by calculating ξ and k from the transmission/reflection data by using Eqs. (13) and (14).

Only in the case that the above criterion is fulfilled the very parameter retrieval can be performed, otherwise their assignment is pointless. This parameter retrieval may then be performed as follows. If the wave parameters k and ξ are determined for a certain polarization and incidence plane, the parameter α is given by $\alpha=\xi/k$. The parameter β follows then from Eq. (12) at normal incidence which reads as $k=\frac{\omega}{c}\sqrt{\beta}$. With β being constant, the remaining parameter γ can be retrieved as a function of k_t from Eq. (12) as $\gamma=\frac{k^2-\frac{\omega^2}{c^2}\beta}{k_t^2}$. Now having α, β, and γ, the classification in Table I can be used to retrieve the effective material parameters for the respective polarization and incidence plane. It can be recognized that each parameter can be retrieved twice; providing a possibility to double-check results. If both results agree the model of weak spatial dispersion holds and the effective material parameters are meaningful.

The outlined procedure is exemplarily applied to the fishnet MM. Geometrical parameters are given in Fig. 1. The metal layers are assumed to be made of silver.[18] To identify the limits of the effective parameter description, we varied the wavelength to unit cell size ratio by scaling the structure and calculated $\alpha=\xi/k$. Hence, all parameters defined in Fig. 1 are scaled by a factor f. Results of the numerical simulations are summarized in Fig. 2 where the real part of the propagation constant k is shown as a function of the wavelength and the scaling factor for normal incidence. The dark-colored area exhibits a negative propagation constant and can thus be considered magnetically active (antisymmetric

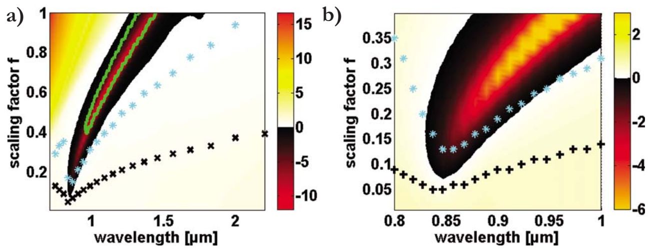

FIG. 2. (Color online) (a) Real part of the propagation constant k (in μm^{-1}) of the fishnet structure for normal incidence. Additionally the lines of constant relative deviation of α are shown (black cross −0.1%, cyan asterisk −5%). The relative deviation is calculated for varying $k_t = k_y$ in TE polarization ($\alpha = \mu_y^{-1}$) being the preferential operating polarization state for the fishnet. The green solid line indicates the region of $\Re(\mu) < 0$. (b) zoomed domain of interest from (a).

plasmon resonance). In the light-colored area the fishnet exhibits a plasmalike effective permittivity with negligible dispersion in the permeability. The green solid line indicates the region of $\Re(\mu) < 0$ at normal incidence, i.e., the region of double negativity.

It is evident that the smaller the structure the smaller the resonance wavelength and the weaker the resonance strength. For very small scaling factors ($f < 0.1$, i.e., unit cell size of less than 60 nm) where the quasistatic limit is reached, the resonance is almost wavelength independent but also tends to disappear; being in agreement with findings for split rings.[16,17] Having identified the area where resonances are occurring it is now interesting to disclose where the effective parameter description (α must be invariant) may be applied. To this end we calculated for discrete wavelengths the parameter α depending on the transverse wave vector ($k_t = 0..1.2k_0$) including at least partially the evanescent spectrum. The relative deviation $(\max|\alpha(k_t) - \alpha(0)|)/|\alpha(0)|)$ serves as a measure to characterize the variation in α and is displayed in Fig. 2. Two bounds for this deviation are considered, 0.1% (almost ideal assignment of effective material parameters possible) and 5% (assignment of effective parameters might be still feasible). Clearly, close to the resonance this deviation is strongest for a fixed scaling factor. It is evident that for a required deviation of 0.1% effective material parameters can be only introduced when there are no magnetic resonances (no left handedness). Since an effective description holds only for almost invariant α this deviation should be as small as possible but values of 5% might be tolerable at most. This condition requires a scaling factor of about 0.15 in the resonance region resulting in a wavelength to cell size ratio of $\lambda/P > 10$. Evidently, the resonance strength is very weak in this domain leading to a nonmagnetic response of the material (left handedness occurs only because of the large imaginary parts). This is consistent with the assumption that such small structures can be described in the quasistatic limit where no magnetic response is observed. Hence, the result of our studies is quite discouraging, namely: a sufficiently strong magnetic response ($\Re(\mu) < 0$) requires a certain minimum unit cell size/wavelength ratio (about 1:4 in case of the fishnet), but this mesoscopic structure must not be described by conventional frequency-dependent effective permittivity and permeability tensors. We have proven this for a fishnet structure, but since all present optical MMs rely on similar resonances we conclude that this tendency may hold in general.

To sum up, based on the assumption of a weakly spatially dispersive conductivity in MM unit cells we have developed a method to verify/falsify an effective anisotropic medium description of MMs. We have shown that a prototypical magnetically active, and thus potentially negative index, material, namely, the fishnet, cannot be described as a homogeneous anisotropic medium in the relevant resonance region. By varying the ratio of wavelength to cell size we have elaborated the limitations of the weak spatial dispersion assumption. There is a trade off: if the spatial dispersion is weak and the material parameters have the usual meaning, the antisymmetric plasmonic resonance, which is responsible for magnetic activity, is also weak or disappears. Our work clearly indicates that for optical MMs the commonly assigned effective parameters do not have the physical meaning of conventional material parameters.

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[18] $\varepsilon = 1 - \omega_p^2/\omega(i\gamma + \omega)$ with $\omega_p = 1.37 \cdot 10^{16}$ s^{-1} and $\gamma = 8.5 \cdot 10^{13}$ s^{-1}.