Retrieving effective material parameters of metamaterials characterized by nonlocal constitutive relations

Karim Mnasri,1 Andrii Khrabustovskyi,2 Michael Plum,3 and Carsten Rockstuhl1,4

1Institute of Theoretical Solid State Physics, Karlsruhe Institute of Technology, Wolfgang-Gaede-Str., 1 76131 Karlsruhe Germany
2Institute of Applied Mathematics, Graz University of Technology, Steyrergasse 30 8010 Graz Austria
3Institute for Analysis, Karlsruhe Institute of Technology, Englerstr. 2 76131 Karlsruhe Germany
4Institute of Nanotechnology, Karlsruhe Institute of Technology, P.O. Box 3640 76021 Karlsruhe Germany

(Dated: August 3, 2018)

The parameter retrieval is a procedure in which effective material properties are assigned to a given metamaterial. A widely used technique bases on the inversion of reflection and transmission from a metamaterial slab. Thus far, local constitutive relations have been frequently considered in this retrieval procedure to describe the metamaterial at the effective level. This, however, is insufficient. The retrieved local material properties frequently fail to predict reliably the optical response from the slab in situations that deviate from those that have been considered in the retrieval, e.g. when illuminating the slab at a different incidence angle. To significantly improve the situation, we describe here a parameter retrieval, also based on the inversion of reflection and transmission from a slab, that describes the metamaterial at the effective level with nonlocal constitutive relations. We retrieve the effective material parameters at the example of a fishnet metamaterial. We demonstrate that the nonlocal constitutive relation can describe the optical response much better than local constitutive relation would do. Our approach is widely applicable to a large class of metamaterials.

PACS numbers:

PACS numbers: 41.20.Jb,42.70.-a,78.20.Bh,78.20.Ci,78.67.Pt

I. INTRODUCTION

Optical metamaterials constitute a novel class of materials that can control the propagation of light in a way inaccessible with natural materials. Optical metamaterials are mostly complicated structures with a spatially distributed permittivity \( \epsilon(x, y, z, k_0) \) that is, very often, periodic in space. To describe the propagation of light through such materials, full-wave numerical solvers of Maxwell’s equations that take into account all the fine details of the spatially dependent permittivity are always an option. Examples for such numerical solvers are the Fourier Modal Method (FMM) or the finite element method (FEM) or the finite-difference time-domain (FDTD) method. These approaches share the heavy request on computational resources. Therefore, to effectively consider optical metamaterials in the design of functional devices, we shall not describe them at the mesoscopic level, i.e. while considering the fine details of the unit cell, but rather at an effective level. By treating them as effectively homogeneous, we put metamaterials on an equal footing to ordinary materials. To make this homogenization, the assignment of effective material parameters is of paramount importance. This is done in a process called the parameter retrieval.

The starting step in the parameter retrieval is the agreement on a particular constitutive relation that shall describe the metamaterial at the effective level. Frequently, for centro-symmetric material with no magneto-electric coupling on which we will concentrate here and inspired by the way we treat natural materials, local constitutive relations are assumed, i.e., \( D(r, k_0) = \epsilon(k_0)E(r, k_0) \) and \( B(r, k_0) = \mu(k_0)H(r, k_0) \). The effective material parameters are the electric permittivity \( \epsilon(k_0) \) and the magnetic permeability \( \mu(k_0) \). \( k_0 = \frac{\omega}{c} \) is the free space wavenumber and \( \omega \) and \( c \) are the frequency of the considered time-harmonic field and the speed of light in vacuum, respectively. We refer to the description of a metamaterial with these two parameters only, as the Weak Spatial Dispersion (WSD) or local approximation. It is a local constitutive relation since the electric displacement \( D(r, k_0) \) and the magnetic induction \( B(r, k_0) \) depend only locally on the electric field \( E(r, k_0) \) and the magnetic field \( H(r, k_0) \), respectively.

However, metamaterials are usually made from building blocks, also called meta-atoms, that have a size in the order of several tens or even hundreds of nanometers, while being designed to operate at optical or near-infrared wavelengths. This is in stark contrast to natural materials that have critical dimensions of merely a fraction of one nanometer. The disparate length scales between critical feature size and operational wavelength for natural materials justifies their treatment with local constitutive relations. Indeed, it is quite a challenge to trace signatures of a nonlocal character with natural materials. The assumption of a local medium, however, ceases to be applicable for optical metamaterials when their critical length scale is no longer much smaller than the wavelength but only smaller. Then, nonlocal effects can no longer be neglected.

But how can we judge, which effective description is appropriate? Well, first of all and with the purpose to treat the material as effectively homogeneous, we require it to be sub-wavelength under all circumstances. A fre-
quent situation, adapted in its geometry also to experimental constraints, is the availability of the metamaterial as a slab with a finite thickness. To perceive such metamaterial from the outer world as homogeneous, we require the absence of a first diffraction order at oblique incidences. This requires the period $a$ of the metamaterial to be smaller than half the operational wavelength $\lambda$, i.e. $\lambda > 2a$ (cf. Ref. [14]).

Next, in the retrieval procedure the reflection and transmission from the metamaterial illuminated with a linearly polarized plane wave are calculated with a full wave solver. By inverting the expressions for reflection and transmission from a homogeneous slab characterized by a specific constitutive relation, the effective material parameters can be retrieved. However, these retrieved properties shall predict the optical response from the same metamaterial also in situations that have not been considered in the retrieval. If they fail, the effective material properties would be useful to reproduce reflection and transmission for the same situation in which they have been retrieved but they could not be used for anything else. This would be strongly against the idea of a material parameter.

When considering local constitutive relation, it has been shown in the past, and for comparative purpose we also show below at a specific example for a metamaterial, that the retrieved local material parameters are insufficient to cope with this requirement [18,19]. Local constitutive relations can reasonably explain the optical response for waves close to near-normal incidence but they fail to describe the optical response at angles beyond the paraxial regime [20]. The situation is of course more severe at wavelengths close to the resonance. These are clear indications that local constitutive relations are insufficient to capture the optical response from metamaterials. Instead, nonlocal constitutive relations have to be considered. The general importance of considering nonlocality for a reasonable parameter retrieval has been also pointed out by other authors and can be considered to be accepted by now in the literature [21,22].

In the past we introduced a specific form of nonlocal constitutive relations and showed that it can capture very well the bulk properties of a given fishnet metamaterial [23]. In particular, the dispersion relation of the eigenmodes of the fishnet metamaterial could be correctly reproduced with the nonlocal constitutive relation. However, the actual material parameters could not be retrieved as they appear in the dispersion relation only as products or ratios (see coefficients of Eqs. (3) and (4)).

Here, we significantly advance this approach and outline a procedure to retrieve the actual nonlocal material parameters. We rely for this purpose on reproducing the reflection and transmission coefficients from a slab of a given metamaterial with those calculated under the assumption of a homogeneous but nonlocal metamaterial. We show that the retrieved nonlocal material parameter can correctly describe the metamaterial at the effective level beyond the paraxial regime. We also show that artefacts that have been controversially discussed in the initial research period on metamaterials, e.g. anti-Lorentz resonances and a negative imaginary part in the effective permittivity [24,25] at the wavelength of the magnetic resonance, vanish when nonlocal constitutive relations are considered. Therefore, as often speculated but now demonstrated, we deem these artefacts to be associated to the non-adapted description of the metamaterial at the effective level with local constitutive relations. We stress upfront that while we can mitigate these features from the local material parameters, we continue to encounter them in the nonlocal material parameters. At the moment, we can only speculate that when considering higher order terms in the nonlocal terms, these problems in lower order terms will vanish as well.

In the following Sec. II we discuss the basic system considered and describe all the theoretical background to predict the optical response from a slab of a homogeneous metamaterial characterized by non-local constitutive relations. The technical details of the actual retrieval procedure will be outlined in Sec. III. We retrieve angle-independent material parameters for a fishnet metamaterial also in Sec. III and discuss them in depth. We compare our results to those obtained with a local model and quantify the improvement. Finally, we conclude and summarize our work in Sec. IV.
been lately introduced in Ref. [24] in the frequency domain that reads

$$D[E] = \begin{cases} E & \text{for } r \in \Omega_- \cup \Omega_+, \\ \epsilon E + \nabla \times \alpha (\nabla \times E) + \nabla \times \nabla \times \gamma (\nabla \times \nabla \times E) & \text{for } r \in \Omega, \end{cases}$$

(1)

with the tensorial and frequency-dependent effective material parameters $\epsilon$, $\alpha$, and $\gamma$. We skip here and in the following the frequency and space dependency in the arguments to simplify the notation. However, it is implicitly considered. We assume that the material is intrinsically nonmagnetic, such that $B(r, k_0) = H(r, k_0)$. However, as a consequence of the finite size and the sophisticated geometry of metamaterial’s building-blocks, currents in a closed loop can be induced. They lead to an artificial effective magnetic response that is linked to the second term in the constitutive relation of the electric field above via $\alpha_i = \frac{\mu_i^{-1}}{k_0^2 \mu_i}$, where $i$ refers to a spatial coordinate. Hence, the parameter $\alpha$ can be re-interpreted as a local, effective magnetic permeability $\mu$ and potentially leads to a negative index behaviour. As this second term can be recast to appear as a local magnetic response, the model with $\epsilon$ and $\alpha$ only, or alternatively $\epsilon$ and $\mu$ only, will be denoted as the Weak Spatial Dispersion (WSD) or the local model. In contrast, the model that also includes the nonlocal material parameter $\gamma$ will be denoted as the Strong Spatial Dispersion (SSD) or the nonlocal model. Throughout this manuscript, we will treat both WSD and SSD models simultaneously, with careful consideration of the limit $\gamma \to 0$.

For convenience, we align the laboratory frame with the principal axes of the metamaterial. The material parameters are, therefore, diagonal and read

$$\epsilon = \begin{pmatrix} \epsilon_x & 0 & 0 \\ 0 & \epsilon_y & 0 \\ 0 & 0 & \epsilon_z \end{pmatrix}, \quad \alpha = \begin{pmatrix} \alpha_x & 0 & 0 \\ 0 & \alpha_y & 0 \\ 0 & 0 & \alpha_z \end{pmatrix}, \quad \gamma = \begin{pmatrix} \gamma_x & 0 & 0 \\ 0 & \gamma_y & 0 \\ 0 & 0 & \gamma_z \end{pmatrix}.$$  (2)

For technical convenience and without loss of generality, we shall assume that the incident plane is either in the $xz$-plane or in the $yz$-plane and the propagation direction is normal to the slab, i.e., in the positive $z$-direction. The incident wave-vector is, therefore, either $k^i = (k_x, 0, k_z)$ or $k^i = (0, k_y, k_z)$. In addition, since the material is centro-symmetric, no optical activity takes place and the polarization of the fields is preserved. It is thus sufficient to consider linearly polarized waves only and to decompose the eigenmodes into decoupled TE (transverse electric) and TM (transverse magnetic) modes. For the sake of notation, we shall denote the transverse component of $k$ by $k_t$, where $t$ is either $x$ or $y$ and $t^* = x$ for $t = y$ and $t^* = y$ for $t = x$.

In the regions outside the slab, i.e., in $\Omega_- \cup \Omega_+$ the dispersion relation is $(k_{z,\sigma}^{(i)}(k_0, k_t))^2 = k_0^2 - k_t^2$, where the superscript $i \in \{I, R, T\}$ represents the incident, reflected, or transmitted fields, respectively. In region $\Omega$, the solutions to the wave equation, i.e., the dispersion relations $(k_{z,\sigma}(k_0, k_t))^2$ are multiple, with $\sigma = \pm 1$, are derived and discussed in depth in Ref. [23]. The polarizations in this case differ remarkably such that the TE and TM polarizations show different functional dependency of $k_{z,\sigma}(k_0, k_t)$. This also severely affects the Fresnel equations and finally the reflection and transmission coefficients. In the case of the TE polarization the dispersion relation reads

$$(k_{z,\sigma}^{TE}(k_0, k_t))^2 = -k_t^2 + p_0^{TE} + \sigma \sqrt{(p_0^{TE})^2 - q_1^{TE} + 2(p_1^{TE} - p_0^{TE}) k_t^2}$$

(3)

with $\sigma = \pm 1$ and the coefficients $p_0^{TE} = [2k_0^2 \gamma_t \mu_t]^{-1}$, $p_1^{TE} = [2k_0^2 \gamma_t \mu_t]^{-1}$ and $q_1^{TE} = \frac{\alpha_t}{\gamma_t}$. Whereas for the TM polarized field we obtain

$$(k_{z,\sigma}^{TM}(k_0, k_t))^2 = - \frac{1}{2} (q_0^{TM} + q_1^{TM}) k_t^2 + p_0^{TM} + \sigma \sqrt{(p_0^{TM} + q_0^{TM} - q_1^{TM}) k_t^2} - p_1^{TM}$$

(4)

with $\sigma = \pm 1$ and the coefficients $p_0^{TM} = [2k_0^2 \gamma_t \mu_t]^{-1}$, $p_1^{TM} = \frac{\alpha_t}{\gamma_t}$, $q_0^{TM} = \frac{\gamma_t}{\alpha_t}$ and $q_1^{TM} = \frac{\alpha_t}{\gamma_t}$. We shortly want to recall that the dispersion relation for a local medium can be reproduced by considering the limit $\gamma \to 0$. Here, one has to be just careful since one of the $k_{z,\sigma}$ asymptotically behaves like $\frac{1}{\sqrt{\gamma}}$ as $\gamma \to 0$. Without loss of generality let $k_{z,\sigma}$ be the divergent solution.

With the information above, one can solve the bulk problem and retrieve the wave parameters, i.e., dispersion relation and the effective refractive index. Despite that, the real utility of the effective medium description of a metamaterial is not to reproduce the wave parameters only, but rather, the real utility of effective medium description is also in relating the structure of a metamaterial to its transmission and reflection and to retrieve the effective material parameters. It is therefore important to understand reflection and transmission of light through a slab of a
metamaterial surrounded by air. To this end, and after we have set up the pieces above together, we can write down the Fresnel matrix that has been rigorously derived for the model in Ref. [24]

\[
\mathbf{F}^{(P)} = \begin{pmatrix}
    r_1^{(P)} & a_x^{(P)} & a_y^{(P)} & d_x^{(P)} & d_y^{(P)} & a_z^{(P)} & a_{z,\perp}^{(P)} & 0 \\
    r_2^{(P)} & b_x^{(P)} & b_y^{(P)} & d_x^{(P)} & d_y^{(P)} & b_z^{(P)} & b_{z,\perp}^{(P)} & 0 \\
    0 & c_x^{(P)} & c_y^{(P)} & e_x^{(P)} & e_y^{(P)} & c_z^{(P)} & c_{z,\perp}^{(P)} & 0 \\
    0 & a_x^{(P)} e^{i\psi_0^{(P)}} & a_y^{(P)} e^{i\psi_0^{(P)}} & a_z^{(P)} e^{i\psi_0^{(P)}} & a_{z,\perp}^{(P)} e^{i\psi_0^{(P)}} & b_z^{(P)} e^{i\psi_0^{(P)}} & b_{z,\perp}^{(P)} e^{i\psi_0^{(P)}} & 0 \\
    0 & b_x^{(P)} e^{i\psi_0^{(P)}} & b_y^{(P)} e^{i\psi_0^{(P)}} & b_z^{(P)} e^{i\psi_0^{(P)}} & b_{z,\perp}^{(P)} e^{i\psi_0^{(P)}} & c_z^{(P)} e^{i\psi_0^{(P)}} & c_{z,\perp}^{(P)} e^{i\psi_0^{(P)}} & 0 \\
    \end{pmatrix}
\]

and \( \mathbf{I}^{(P)} = (i_1^{(P)}, i_2^{(P)}, 0, 0, 0, 0)^T \). The matrix elements are very long, and therefore, summarized in Tab. I. We note that P indicates the polarization.

The Fresnel matrix above has to be understood as follows. The first three rows correspond to the interface conditions at the first interface at \( z = 0 \) and the last three ones to the second interface of the slab, where \( z = d_{\text{slab}} \) and the fields accumulate a phase \( e^{i\psi_0^{(P)}} \), where \( \psi_0^{(P)} = k_{z,\perp}^{(P)} d_{\text{slab}} \). The first and sixth columns contain information regarding the reflected and transmitted fields, respectively. The second and fourth columns pertain the terms for the forward propagating modes inside a slab with \( \Im(k_{z,\perp}^{(P)}) > 0 \) while the third and fifth columns are associated to the modes that propagate backwards inside the slab with \( \Im(k_{z,\perp}^{(P)}) < 0 \). To reconstruct the Fresnel matrix for the WSD, one has to consider the limit \( \gamma \to 0 \). Two rows and columns will contain the divergent \( k_{z,\perp}^{(P)} \) which exponentially damps the field amplitudes and, therefore, do not contribute neither to reflection nor to transmission. The effective dimension of the Fresnel matrix in the case of WSD reduces to a \( 4 \times 4 \)-matrix, as expected.

Using the Fresnel matrix above, we obtain the complex-valued reflection and transmission coefficients by taking the first and the last components, respectively.

The formulas for both \( \rho^{(P)} \) and \( \tau^{(P)} \) are very long and will, for the sake of readability not written explicitly. Yet, they will be used in the next section for the parameter retrieval and evaluated for comparison to the reflection and transmission coefficient of the actual material to be homogenized.

### III. RETRIEVAL PROCEDURE

In general, light-matter interaction depends on the polarization and the incident plane of the light. Therefore, the electromagnetic fields couple to different material parameters in each scenario. Due to the anisotropy of the structure, one can only retrieve all effective material pa-
FIG. 1. Fishnet metamaterial. We consider a biperiodic structure with periods \( \Lambda_x = \Lambda_y = 600 \text{ nm} \) and consider an extension of the thin film in \( z \)-direction of 200 nm. The fishnet consists of rectangular holes with the width \( w_x = 100 \text{ nm} \) and \( w_y = 316 \text{ nm} \). They consist in a stack of layers made of two 45 nm Ag layers separated by a thin dielectric spacer, 30 nm of MgF\(_2\), with \( n_{\text{MgF}_2} = 1.38 \). The rest is filled with air.

parameters by considering all four possible illuminations (TE,TM) \( \times (k_x, k_y) \). The relevant material parameters for a fixed illumination condition are summarized in Table II. The nonlocal parameter \( \gamma \) behaves like the permittivity \( \epsilon \), i.e., it couples only to the electric field components that appear in the considered polarization. For instance, in the TM-\( k_z \) polarization, the electric field is \( \mathbf{E} = (E_x, 0, E_z) \) and couples to the double \( (\epsilon_x, \epsilon_z) \) and to \( (\gamma_x, \gamma_z) \), while the magnetic field \( \mathbf{H} = H_y \hat{\mathbf{e}}_y \) couples to \( \mu_y \).

TABLE II. Relevant material parameters that couple to light depending on the polarization and the incidence plane.

| \( P = 0 \) (TE) | \( P = 1 \) (TM) |
|----------------|----------------|
| \( (k_x, 0, k_z) \) | \( (0, k_y, k_z) \) |
| \( (k_x, 0, k_z) \) | \( (0, k_y, k_z) \) |
| \( \epsilon_y \) | \( \epsilon_x \) |
| \( \mu_x \) | \( \epsilon_z \) |
| \( \mu_z \) | \( \mu_x \) |
| \( \gamma_y \) | \( \gamma_x \) |
| \( \gamma_z \) | \( \gamma_y \) |

As an exemplary metamaterial of current interest, which we consider in the following retrieval, we choose the fishnet metamaterial. It exhibits a negative refraction in a specific frequency range with both a dispersive permittivity and a dispersive permeability. The basic geometry is shown in Fig. 1. The geometrical parameters are taken from literature. It consists of a centro-symmetric unit cell with side lengths of \( \Lambda_x = \Lambda_y = 600 \text{ nm} \) being replicated in the \( xy \)-plane and a stacking of air-metal-dielectric-metal-air nanowires in \( z \)-direction with a total thickness of 200 nm. The nanowires are perpendicularly aligned and form rectangular holes with widths of \( w_y = 100 \text{ nm} \) and \( w_x = 316 \text{ nm} \). The metal (silver) layers have a thickness of 45 nm. Silver is described by a Drude model for the permittivity that reads

\[
\epsilon_{\text{Ag}} = 1 - \frac{\omega_p^2}{\omega^2 + \Gamma \omega},
\]

with the plasma frequency \( \omega_p = 13700 \text{ THz} \) and the relaxation rate \( \Gamma = 85 \text{ THz} \). The silver layers are separated by a nondispersive magnesium fluoride spacer with \( \epsilon_{\text{MgF}_2} = 1.9044 \) and a thickness of 30 nm. This metamaterial admits a negative index in the TM-\( k_z \) polarization for frequencies around \( k_0 = 4.3 \text{ \mu m}^{-1} \). Since we are interested in this negative index property, we perform the retrieval for this polarization. The retrieval procedure for the other three illumination directions advances similarly and will not be shown here, as we are mainly interested in the material parameters that are linked to the negative index behaviour.

To obtain the numerical data for reflection and transmission from a slab of the fishnet, we perform a full-wave simulation by using a Fourier Modal Method (FMM). In this method, we expand the eigenmodes of the \( xy \)-periodic structure into Bloch modes and in \( z \)-direction into plane waves for wavelengths \( \lambda \) in the near IR-range with \( \lambda \in (\frac{2\pi}{4 \text{ nm}}, \frac{2\pi}{2 \text{ nm}}) \text{ \mu m} \). Outside the metamaterial the fields are expanded into plane waves. By matching the interface conditions between substrate and cladding and the individual layers that form the fishnet metamaterial, respectively, all amplitudes of all modes are found. In the numerics, a sufficient large number of modes has been taken into account to achieve convergent results for reflection and transmission.

Since the structure is sub-wavelength, only the 0th-diffraction order in transmission and reflection is propagating and will carry energy into the cladding (\( z > d_{\text{slab}} \)) or the substrate (\( z < 0 \)), respectively. Higher-order diffraction contributions are, therefore, suppressed and homogenization is feasible. The 0th-order reflection and transmission coefficients obtained from a fishnet slab with \( d_{\text{slab}} = 200 \text{ nm} \) will be represented by \( \rho^\text{FMM} \) and \( \tau^\text{FMM} \), respectively and are shown in Fig. 2(a) and (d), for \( k_0 \in (3.8, 4.768) \text{ \mu m}^{-1} \) and the angle of incidence from \( 0^\circ \) to \( 90^\circ \), meaning \( \forall k_0 \in (3.8, 4.768) \text{ \mu m}^{-1} \). The amplitudes are shown but of course the values are complex. These are the amplitudes that have to be reproduced in the homogeneous description.

Considering the complexity and the nonlinearity of Eq. 4 and of Eqs. 6 and 7, the retrieval cannot be performed by inverting and solving

\[
\rho^\text{TM}(k_0, k_x, \epsilon, \mu, \gamma) \doteq \rho^\text{FMM}(k_0, k_x) \quad \text{and} \quad \tau^\text{TM}(k_0, k_x, \epsilon, \mu, \gamma) \doteq \tau^\text{FMM}(k_0, k_x).
\]

For this reason, the retrieval is based on fitting the analytically derived reflection and transmission formulas, i.e., Eqs. 6 and 7 to the numerically calculated reflection and transmission coefficients of the reference material. In this procedure we consider each frequency individually, as the material parameters are functions of the frequency. Given reflection and transmission as a function of the angle of incidence, one can minimize a merit function \( \delta \) w.r.t. to the effective material parameters to capture the reflection and transmission coefficients of the structure. The merit-function \( \delta \) is a measure of how well a model applies to homogenize a structure. It rewards the ability
FIG. 2. (a)-(c) amplitude of the reflected light $|\rho|$ and (d)-(f) of the transmitted light $|\tau|$ from one fishnet layer with thickness $d_{\text{slab}} = 200$ nm using different approaches. The left figures ((a) and (d)) correspond to the full-wave simulation of the actual fishnet slab as done with the FMM. This can be considered as the reference data. The centered figures ((b) and (e)) are the fitted reflection and transmission amplitudes from a homogeneous slab with the same thickness as the fishnet using the WSD, i.e., the local approach. The figures on the right ((c) and (f)) are obtained from considering a homogeneous slab with SSD, i.e., retaining nonlocal effects in the effective description. The figure indicates the improvement in capturing the reflection and transmission of the reference material ((a) and (d)) using SSD (nonlocal) compared to WSD (local).

of the constitutive relation to recover the electromagnetic response of the slab and is explicitly defined as follows:

$$
\delta(k_0) = \min_{\epsilon, \mu, \gamma} \sum_{k_x = 0}^{k_0} \frac{w(k_x)}{2} \left( \left| 1 - \frac{\rho^{TM}(k_0, k_x, \epsilon, \mu, \gamma)}{\rho^{FMM}(k_0, k_x)} \right| \right) + \left| 1 - \frac{\tau^{TM}(k_0, k_x, \epsilon, \mu, \gamma)}{\tau^{FMM}(k_0, k_x)} \right|, \quad (8)
$$

where $w(k_x)$ is a weight function that is centered in the paraxial regime, i.e., $k_x \approx 0$. We chose here an exponentially decaying dependency, such that

$$
w(k_x) = e^{-\alpha k_x}, \quad (9)
$$

with $\alpha = 2.5 \Lambda_x$, with the lateral period of the fishnet $\Lambda_x = 0.6$ nm. The purpose of giving more importance to the weight function in the paraxial regime is our requirement to be able to reproduce at least reflection and transmission at normal incidence and look afterwards how far we can stretch this regime of applicability also to oblique incidence angles.

Using this approach, we retrieve by minimizing the merit function at each frequency individually the material parameters that can describe best the optical response from the fishnet metamaterial as a function of the angle of incidence. The retrieval has been done individually for both the WSD and the SSD. The reflection and transmission coefficients as calculated with the retrieved effective properties are shown in Fig. 2. Evidently, it reveals the improvement in capturing the reflection and transmission of the reference material ((a) and (d)) using SSD ((c) and (f)) compared to WSD ((b) and (e)).

From these results (Figs. 2 and particularly Fig. 3) we mainly observe two outcomes. With retaining nonlocality we can not only increase the agreement with the reference curves, but also the functional behaviour, i.e., the curvature of reflection and transmission w.r.t. the angle of incidence, seems to be more realistic than in the case when WSD only is considered. This is a solid confirmation in favor for the relevance of including nonlocal material parameters into the effective description of metamaterials.

To quantify the findings and to allow for a better discussion, we show in Fig. 4 in % the absolute deviation between the amplitude of reflection and transmission as calculated with the FMM to those fitted using either the WSD (Fig. 4 (left)) or the SSD model (Fig. 4 (right)), respectively. For a better discussion of the deviations, the color axis has been truncated to 10%. The figures show two regimes of interest. The blue regime is the regime where reflection and transmission can be captured quite well with the retrieved material parameters and, hence, the homogenization is meaningful. In contrast, the red regions refer to deviations above 10% from the reference, i.e., the region where the model fails to capture the electromagnetic response adequately which leads to the failure of the homogenization approach. We
FIG. 3. Reflection (top) and transmission (bottom) coefficients at a selected frequency of $k_0 = 4.2414 \mu m^{-1}$. The solid (dashed) curves represent the real (imaginary) part. The reference curves obtained from the FMM are in black while the blue curves are obtained from considering WSD and the red ones for the case of SSD. The red curves are obtained from fitting Eqs. (6) and (7) to the reference curve and show a good agreement up to $50^\circ$. Meanwhile, the blue curves, which are obtained from WSD, are showing only an agreement within the paraxial regime.

Clearly see that for all frequencies $k_0$, the nonlocal approach pushes the agreement to higher incidence angles. Of course, as we also expected, around the resonance frequency $k_0 = 4.3 \mu m^{-1}$ the homogenization becomes less reasonable. Accordingly, in the blue regions of Fig. 4 the metamaterial can be effectively characterized.

The effective material parameters that were obtained in the retrieval are shown in Fig. 5. The parameters that appear in both the local and the nonlocal constitutive relations that result from the retrieval with the WSD and the SSD are shown within one figure. The parameters that appear only in the nonlocal constitutive relation are shown alone. There are a few things worth to discuss.

First of all, using the local approach, we note that the permittivity $\epsilon_x$ has an anti-Lorenzian shape around the resonance frequency $k_0 = 4.3 \mu m^{-1}$, leading to a negative imaginary part. The permittivity $\epsilon_x$ therefore has a complex pole in the upper complex $k_0$ half-plane, and hence, violates causality. This unphysical anti-resonance is no longer existing when the nonlocal constitutive approach is considered. Moreover, as physically expected, the permittivity $\epsilon_x$ shows a Drude type behaviour. This is the usual response expected for a diluted metal; which the fishnet metamaterial is actually. Second, the permeability $\mu_y$ is in both models nearly identical and shows a Lorentzian functional dependency. This behaviour is expected due to the structure of the fishnet. When light in TM-$k_x$ polarization couples to the metallic nanowires, circular currents are induced and a magnetization occurs. The magnetization is driven into resonance at $k_0 = 4.3 \mu m^{-1}$ and the Lorentzian dispersion is centered around that frequency. Third, the parameter that is hard to interpret is the $z$-component of the permittivity, $\epsilon_z$. It exhibits a quite strange behavior at some frequencies where $\Im(\epsilon_z)$ is negative in both the WSD and the SSD. However, we stress that this parameter is hard to capture in the present geometry. At normal incidence, for example, the electromagnetic field does not couple at all to this $z$-component. If only the response at normal incidence is considered, this component cannot be retrieved. At oblique incidence the situation improves somewhat, but after all it turns out to be rather insensitive. This can be explained by the fact that the wavenumber in the metamaterial is quite larger. Hence, even though excited at oblique incidence from the surrounding, the plane waves that are the eigenmodes in the metamaterial propagate inside in a paraxial direction. Therefore, they do not fully probe the $z$-component of the permittivity tensor.

Fourth, considering the nonlocal parameters $\gamma_x$ and $\gamma_z$, they show a resonance at a frequency around $k_0 = 4.3 \mu m^{-1}$, where the negative index has its min-

FIG. 4. Deviations from the reflection and transmission coefficients in % for (left) the local and (right) the nonlocal approaches for frequencies $k_0 \in [3.8, 4.768] \mu m^{-1}$ and the angle of incidence from $0^\circ$ to $90^\circ$. In comparison to the local approach, the nonlocal one covers a larger parameter space (blue region) where the homogenization is meaningful and effective material parameters can be retrieved. The colorbar is truncated to 10% to indicate a threshold of applicability.
FIG. 5. Real and imaginary parts of the effective permittivities $\epsilon_x$ and $\epsilon_z$, effective permeability $\mu_y$, and the effective nonlocal parameters $\gamma_x$ and $\gamma_z$ as a function of the frequency $k_0$ using both local (dashed blue) and nonlocal (solid red) approaches. The results are obtained from fitting reflection and transmission coefficients (6) and (7) by means of absolute deviations from the exact data as defined in Eq. (8). Note that the nonlocal parameters are always at least one order of magnitude smaller than the local parameters. Ways how to design metamaterials such that $\gamma$ is maximized is still an open question and subject to study. Nevertheless, we can summarize that with those nonlocal material parameters we can significantly improve our ability to describe the optical response from the considered fishnet metamaterial in a slab geometry, as mainly evidenced in Fig. 4.

IV. CONCLUSIONS

In this contribution, we investigated the response of a fishnet metamaterial in the frequency range where it undergoes a resonant coupling with light yielding a negative index behaviour in the TM-$k_x$ polarization. We retrieved both local and nonlocal material parameters emerging in the TM-$k_x$ polarization and sketched the procedure for the other three cases as well in Table II. The retrieval was performed by fitting the analytically derived reflection and transmission coefficients for both weak and strong spatial dispersion and compare these to the complex-valued reflection and transmission coefficients of the heterogeneous slab. We clearly see that for all simulated frequencies, the nonlocal approach pushes the agreement to higher angles of incidence than the local approximation and that the reflection and transmission from a slab can be captured more efficiently using the nonlocal approach. In addition, the permittivity $\epsilon_x$ shows unphysical behaviour around the resonance frequencies in the retrieval using WSD model. This has been lifted by introducing the nonlocal material parameters. This is another indication that it is important to retain nonlocality for a more realistic homogenization of optical metamaterials. These findings have been obtained using the fishnet structure as a test subject, which sustains a negative index and therefore of utmost importance in applications. However, the procedure can be readily applied to other metamaterials.

ACKNOWLEDGMENTS

We gratefully acknowledge financial support by the Deutsche Forschungsgemeinschaft (DFG) through CRC 1173. K.M. also acknowledges support from the Karlsruhe School of Optics and Photonics (KSOP).

1 N. Engheta, in Advances in Electromagnetics of Complex Media and Metamaterials (Springer, Dordrecht, 2002) pp. 19–37.
2 M. G. Silveirinha, A. Alù, and N. Engheta, Phys. Rev. B 78, 075107 (2008).
3 A. Monti, A. Alù, A. Toscano, and F. Bilotti, in Photonics, Vol. 2 (Multidisciplinary Digital Publishing Institute, 2015) pp. 540–552.
4 S. Guenneau and S. A. Ramakrishna, C. R. Phys. 10, 352 (2009).
5 E. E. Narimanov and A. V. Kildishev, Appl. Phys. Lett. 95, 041106 (2009).
6 L. Li, J. Opt. Soc. Am. A 14, 2758 (1997).
7 L. Li, J. of Opt. A: Pure and Appl. Opt. 5, 345 (2003).
8 J.-M. Jin, The finite element method in electromagnetics (John Wiley & Sons, 2015).
9 A. Taflove and S. C. Hagness, Computational electrodynamics: the finite-difference time-domain method (Artech...
10 C. R. Simovski, J. of Opt. 13, 013001 (2010).
11 A. I. Căbuz, D. Felbacq, and D. Cassagne, Phys. Rev. Lett. 98, 037403 (2007).
12 A. Ciattoni and C. Rizza, Phys. Rev. B 91, 184207 (2015).
13 V. Agranovich and Y. Gartstein, Metamaterials 3, 1 (2009).
14 P. Lalanne and D. Lemercier-Lalanne, J. of Mod. Opt. 43, 2063 (1996).
15 S. V. Zhukovsky, A. Andryieuski, O. Takayama, E. Shkondin, R. Malureanu, F. Jensen, and A. V. Lavrinenko, Phys. Rev. Lett. 115, 177402 (2015).
16 A. Andryieuski, A. V. Lavrinenko, and S. V. Zhukovsky, Nanotechnology 26, 184001 (2015).
17 C. Menzel, T. Paul, C. Rockstuhl, T. Pertsch, S. Tretyakov, and F. Lederer, Phys. Rev. B 81, 035320 (2010).
18 M. G. Silveirinha, Phys. Rev. E 73, 046612 (2006).
19 I. Tsukerman, J. Opt. Soc. Am. B 28, 2056 (2011).
20 A. Alù, Phys. Rev. B 84, 075153 (2011).
21 C. Rizza and A. Ciattoni, Photonics 2, 365 (2015).
22 C. Fietz and C. M. Soukoulis, Phys. Rev. B 86, 085146 (2012).
23 M. A. Gorlach and P. A. Belov, Phys. Rev. B 92, 085107 (2015).
24 K. Mnasri, A. Khrabustovskyi, C. Stohrer, M. Plum, and C. Rockstuhl, Phys. Rev. B 97, 075439 (2018).
25 T. Koschny, P. Markoš, D. R. Smith, and C. M. Soukoulis, Phys. Rev. E 68, 065602 (2003).
26 D. R. Smith, D. C. Vier, T. Koschny, and C. M. Soukoulis, Phys. Rev. E 71, 036617 (2005).
27 G. Dolling, C. Enkrich, M. Wegener, C. M. Soukoulis, and S. Linden, Opt. Lett. 31, 1800 (2006).
28 A. Alù, A. D. Yaghjian, R. A. Shore, and M. G. Silveirinha, Phys. Rev. B 84, 054305 (2011).