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High symmetry versus optical isotropy of a negative-index metamaterial

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Optically isotropic metamaterials (MMs) are required for the implementation of subwavelength imaging systems. At first glance one would expect that their design should be based on unit cells exhibiting a cubic symmetry being the highest crystal symmetry. It is anticipated that this is a sufficient condition since it is usually assumed that light does not resolve the spatial details of MM but experiences the properties of an effective medium, which is then optically isotropic. In this work we challenge this assumption by analyzing the isofrequency surfaces of the dispersion relation of the split cube in carcass negative index MM. We show that this MM is basically optically isotropic but not in the spectral domain where it exhibits negative refraction. The primary goal of this contribution is to introduce a tool that allows to probe a MM against optical isotropy.

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I. INTRODUCTION

Driven by the desire and the opportunity to have optical materials with tailored properties at hand, the field of metamaterials (MMs) attracts a steady increasing share of research interest. To loosely define the field, one may understand MMs as artificial structures made of subwavelength unit cells, called meta-atoms. They predominantly affect the light propagation by a careful choice of their geometry and their arrangement; and not by the intrinsic properties of the materials they are made of. MMs allow to tailor the flow of light well beyond what would be possible with naturally occurring materials by mimicking unprecedented optical properties. A large variety of new optical phenomena were predicted and experimentally proven on the base of MMs, where negative refraction attracted potentially the largest amount of interest.1–6 Metamaterials providing a negative effective refractive index are the essential ingredient to fabricate a perfect lens with an optical resolution limit well below Abbe’s prediction.7 However, and this is the major requirement currently not met by most of present MMs, this effective refractive index can only be meaningfully introduced if the dispersion relation is isotropic, i.e., for lossless media the isofrequency surface of the dispersion relation in three-dimensional (3D) \( k \) space must be spherical. Moreover, this isotropy has to hold also for evanescent waves since they carry the subwavelength information for the desired super-resolution. To overcome this obstacle various approaches for obtaining isotropic MMs have been put forward.8–14

A systematic approach to design an isotropic magnetically active MM was proposed by Baena et al.15 The first step is to choose a highly symmetric MM. Such MM is constructed based on a cubic unit cell arranged at a cubic lattice. It is evident that MMs with this high-symmetry behave isotropically in the quasistatic limit. Frequently the optical properties of such MMs have only been investigated for normal incidence (zero-transverse wave vector) and then extrapolated toward finite transverse wave vectors, assuming isotropic medium response due to the symmetry of the structure.

However, and this is crucial, even if the structure is operated in the subwavelength domain, the required optical isotropy is not straightforward, since the typical resonance wavelengths, evoking magnetic effects, are comparable to the structure size. Here a structure is meant to be subwavelength if the unit cell is smaller than \( \lambda/2 \) with \( \lambda \) being the wavelength in the outer domains. If this condition is met, only the zeroth diffraction order is propagating in transmission as well as in reflection for real-valued angles of incidence. There are only a few attempts where the MMs are probed by obliquely incident fields and for different polarizations, mostly restricted to transmission and reflection measurements.16–20 Since even for an isotropic medium the reflection and transmission coefficients evidently depend on the angle of incidence, it is hardly possible to draw conclusion with respect to the isotropic behavior of the respective MM. To ultimately verify whether a highly symmetric metamaterial behaves optically isotropic, it is necessary to calculate the dispersion relation \( \omega = \omega(k_x, k_y, k_z) \) where its isofrequency surface \( k_z = k_z(k_x, k_y, \omega = \text{const.}) \) governs the diffraction and refraction properties of the MM.51 Here, \( \omega, k_x, k_y, \) and \( k_z \) are the frequency, the transverse and the longitudinal wave-vector components, respectively, where the latter is frequently termed propagation constant. The aim of this work consists just in evaluating this isofrequency surface.

For computing isofrequency surfaces of a three-dimensional MM we take the complex permittivity of the material into account. This is crucial because most the bandstructure solvers assume lossless media which is certainly incorrect for metamaterials. We use instead a plane-wave-expansion technique that solves Maxwell’s equations in the frequency domain for the periodic structure. The technique solves the respective eigenvalue problem for the generally complex propagation constant of the Bloch modes.25 Computing the propagation constant as a function of the transverse wave vector for a fixed frequency provides the isofrequency surfaces. By using this method we specifically show
for the split cube in carcass (SCiC)-MM with cubic symmetry that its optical response is isotropic only at low frequencies. By contrast this isotropy disappears in the spectral domain where the propagation constant and hence the effective refractive index is negative.

After having introduced the system under consideration in Sec. II, we show in Sec. III that the proposed MM fulfills all requirements usually imposed for its homogenization. In Sec. IV we discuss the isofrequency surface for the negative refraction regime and introduce a simple measure to quantify the optical isotropy across the entire subwavelength frequency domain.

II. SPLIT CUBE IN CARCASS DESIGN

For our investigations we use the split cube in carcass structure, which is a simplified version of the split cube in cage structure containing less number of fine details. Using the nested structures approach the SCiC is designed such that one element of the unit cell that exhibits magnetic properties is inserted into another element which shows specific dielectric response. The SCiC unit cell consists of two silver parts embedded in silica ($\varepsilon = 2.25$). Silver is regarded as Drude metal with a plasma frequency $\omega_p = 1.37 \times 10^{16}$ rad/s and collision frequency $\omega_c = 8.5 \times 10^{13}$1/s. These parameters were adjusted to emulate the real experimental situation in simulations.

The outer part, the Carcass, is a three-dimensional wire medium and provides a negative permittivity. The inner part, the split cube, is a hollow cube with slits in the middle of the facets. It is the logical 3D extension of the symmetric split ring resonator concept and provides a magnetic resonance (“negative permeability” $\mu$). The details regarding the structure’s sizes are indicated in Fig. 1.

The SCiC was chosen because it has the highest possible symmetry for a periodically arranged MM. In particular, it is mirror symmetric with respect to three orthogonal axes. This excludes any effects resulting from first-order spatial dispersion-like chirality. Due to the cubic symmetry the three main propagation directions are equivalent. Hence, the optical response of the SCiC is supposed to be described by scalar effective material parameters in the quasistatic limit, i.e., the optical response should be isotropic. From these symmetry considerations one usually draws the conclusion that the SCiC might be an ideal candidate for an isotropic negative index metamaterial.

III. HOMOGENEITY

To avoid any misinterpretations we will argue at first what we will understand as homogeneity. A periodic medium is said to be homogenizable if, at first, it consists of subwavelength unit cells. This first condition simply requires the period to be smaller than half the wavelength to suppress propagation of higher diffraction orders. At second, light propagation inside the structure is governed by the properties of a single Bloch mode only. And at third, light impinging onto the structure predominantly couples to this particular mode.

Under these assumptions the considered medium is indistinguishable to a homogeneous medium with an arbitrary complex dispersion relation when its properties are probed from an outer medium. Its optical response may be described by nonlocal material parameters which are strongly spatially dispersive in general. The isofrequency surfaces of the dispersion relation, i.e., the normal-mode surfaces can have an almost arbitrary complex shape. Only if the spatial dispersion is weak the material parameters can become local and the isofrequency surfaces are simply elliptical. Then the so-called quasistatic limit is reached for small ratios of period over wavelength, i.e., $a/\lambda$. The latter assumption is obviously more demanding than homogeneity in general. Hence optical isotropy, i.e., spherical isofrequency surface of the dispersion relation, is attainable only for electrically small objects with cubic symmetry. Therefore a homogeneous medium is not necessarily describable by local material parameters but vice versa.

So to describe a MM as an effectively homogeneous medium, it is necessary to assure at first that light propagation within the MM is exclusively governed by a single Bloch mode. To be sure that this requirement is met, we also have to prove that the incident light field couples only to this particular mode. This is done by comparing the effective plane-wave-propagation constant, as retrieved from reflection and transmission data of a finite structure, with that obtained from the dispersion relation of the lowest-order Bloch mode.

We will determine the effective propagation constant in the SCiC by using the $S$-parameter retrieval method. The complex reflection and transmission coefficients are numerically calculated by the Fourier modal method where $31 \times 31$ Fourier orders were retained to achieve convergent results. By inversion of these scattering data the effective propagation constant $k = k_z$ and the effective impedance $Z$ can be determined. To certify that such an assignment of effective parameters to structures composed of only a small number of functional layers is meaningful and that the homogenization is valid, it is necessary to investigate the convergence of the parameters toward their bulk values therefore we determine the propagation constant of the Bloch eigenmodes of the infinite structure as well.
This is done by calculating the $T$ matrix for a single period and solving the eigenvalue problem

$$
\hat{T} \begin{pmatrix} E \\ H \end{pmatrix} = \exp(ik\Lambda) \begin{pmatrix} E \\ H \end{pmatrix}
$$

to obtain the propagation constant $k = k_{\text{Bloch}}$ of the Bloch modes, where $\Lambda$ is the period in the main propagation direction.

The results for the real and imaginary part of both propagation constants (finite homogeneous and infinite periodically structured media) and the formally introduced effective refractive indices $n = ck/\omega$ are shown in Fig. 2. For the periodic bulk material we get in general the propagation constants for an infinite number of Bloch modes (which remains nevertheless finite because of the numerical truncation of the number of plane waves retained in the plane-wave expansion) but only the zeroth-order mode, i.e., the one with the smallest imaginary part is shown. Clearly the values obtained from the finite effectively homogeneous structure are converging astonishingly fast toward the values for the periodic bulk medium. Thus, two important conclusions can be drawn, first the homogenization of the periodic MM is feasible and second the optical response of a SCiC-MM consisting of only a few layers equals that of a bulk medium.

The design purpose of the SCiC is a negative effective propagation constant, and thus an negative effective index of refraction which is clearly achieved for frequencies around 170 THz [figure of merit $-\text{Re}(n)/\text{Im}(n) = 0.35$, ratio of period to vacuum wavelength $a/\lambda = 1/7$]. Note that the effective permeability is dispersive but positive in the investigated frequency range. Hence, the SCiC is a single negative MM resulting in the fairly small figure of merit. For frequencies less than 150 THz the SCiC is rather a strong absorber due to the large metal fraction.

Since for the scattering problem at the finite system the coupling to different Bloch modes was rigorously considered, we can conclude that the propagation of light through the structure as well as the coupling process is almost entirely dictated by the fundamental Bloch mode only, again underlining the validity of the homogenization procedure. This is clear as the propagation constant values for the finite and the infinite structures coincide. Although not shown here for the sake of brevity the propagation constants for oblique incidence are also rapidly converging toward the bulk values. Also the effective impedance is converging for normal incidence as well as for oblique incidence.

Hence, we can fully rely on describing light propagation in terms of the dispersion relation assuming that light will couple to this fundamental mode only. Note that from the results above we can also conclude that the SCiC fulfills all requirements for the homogenization of the MM, namely, the structure is subwavelength (compared to the wavelength of the environment) and light propagation inside the structure is determined by a single Bloch mode to which an external light field predominantly couples.

**IV. DISPERSION RELATION AND OPTICAL ISOTROPY**

The main property we are interested in is the optical isotropy of the SCiC, in particular, in the spectral region around 170 THz where the propagation constant is negative. To judge this we will proceed in calculating the isofrequency surface.

Due to the symmetry of the structure the complete isofrequency surface can be constructed by only calculating the dispersion relation in the irreducible Brillouin zone. The real and imaginary parts of the propagation constant for a fixed frequency of 170 THz are shown in Figs. 3(a) and 3(b). For comparison we calculated Fig. 3(c) and 3(d) the analytically calculated isofrequency surface of an isotropic medium, which has the same refractive index as SCiC at normal incidence.

At first we notice that both the real and the imaginary parts of the propagation constant of the MM are not rotation-
ally symmetric and monotonically changing with increasing absolute value of the tangential component $k_t$ as for the isotropic medium. In particular, the dependency on the transverse wave vector is tremendously different when compared to the isotropic medium. While the real part is monotonically increasing with $k_t$ in the $\Gamma X$ direction it is nonmonotonously decreasing in the $\Gamma M$ direction. Also the imaginary part is strongly increasing in the $\Gamma M$ direction and only slowly varying in the $\Gamma X$ direction. Hence, any formally introduced effective refractive index would explicitly depend on the transverse wave vector $k\parallel=(k_x,k_y)$. Therefore, the introduction of a global effective refractive index is pointless since no additional information is obtained. Nevertheless for paraxial wave propagation the introduction of a local effective refractive index is feasible. Near the $\Gamma$ point the isofrequency surface is approximately spherical where the validity of this approximation strongly depends on the frequency and the wavelength to cell-size ratio as discussed later in detail.

It should be mentioned that the choice of real valued $k_x$ and $k_y$ is arbitrary to a certain extent. In lossy medallike metamaterials, in general, the complex nature of the dispersion relation cannot be neglected. Hence the dispersion relation could also be calculated for complex valued $k_t$. On the other hand it is pointless to provide these values as they are not accessible in any experiment. The tangential wave-vector components are continuous at boundaries and only real-valued plane-wave solutions can exist in free space, therefore this choice reflects experimental constraints.

To more quantitatively evaluate the optical isotropy of the MM, we monitor in the following the relative deviation between the numerically obtained propagation constant and the ideal spherical isofrequency surface. To quantify this deviation we assume that the effective refractive index at normal incidence is valid also for any other propagation direction. From symmetry considerations and the exemplary results in Fig. 3 we conclude that it is sufficient to investigate the dependency in the high-symmetry $\Gamma X$ and $\Gamma M$ directions assuming that for a fixed value of $|k_t|$ these points are extremes of the isofrequency surface. In Fig. 4 the real part of the propagation constant is shown as a function of the frequency and the angle of incidence where the plane of incidence is parallel to the coordinate axes. (b) The same as (a) with plane of incidence rotated by $\pi/4$, i.e., the $\Gamma M$ direction. The black isoerror lines to the ideal ones is slightly smaller in the $X$ direction but not significantly.

Obviously in the resonant regime the MM can be only considered isotropic if the ratio $a/\lambda$ is much less than in the nonresonant regime. In general the situation is identical for both planes of incidence ($\Gamma X$ and $\Gamma M$ directions). However, for larger frequencies the deviation of the calculated isofrequency lines to the ideal ones is slightly smaller in $\Gamma M$ than in $\Gamma X$ direction but not significantly.

Note that the introduction of the quantity $\Delta$ is only one option to quantify the deviation of the pertinent isofrequency surface from the ideal one. Other measures are also possible, where, for example, the deviation of the length of the actual wave vector compared to the length of the wave vector in an isotropic medium yields qualitatively the same results.
Before we proceed to discuss the results, some remarks concerning the investigated parameter space are in order. Here, the dispersion relation is not calculated up to the edge of the Brillouin zone but for fixed tangential wave vectors which translate for propagating waves into certain angles of incidence. This is more useful since for very low frequencies it is not possible to provide incident plane waves with tangential wave-vector components $k_t$ that are in the order of $\pi/a$ where $a$ is the lattice constant. Furthermore, plane waves with wave vectors in the dimension of the lattice vector will always sense the details of the periodicity. In this case the issue of an optically isotropic structure is pointless. Of course, also for small frequencies the isosurface are not rotationally invariant but these domains are, in general, not accessible to the experiment and are negligible. Assuming the outer medium to have a permittivity $\varepsilon$ then the line given by $k_t/k_0=\sqrt{\varepsilon}$ corresponds to grazing incidence and to the angular domain accessible from free space.

Up to here we have only discussed the properties of the fundamental Bloch mode with the smallest imaginary part since this mode essentially dictates light propagation. While considering the coupling of an external field to this Bloch mode we had to suppose that only this mode is excited thus restricting ourselves to a certain polarization state of the incident light. Nevertheless, in the general case of arbitrarily polarized incident light one has to discuss both eigenstates of the polarization separately, as both may exhibit different propagation constants. Assuming an isotropic homogeneous medium the eigensolutions are always degenerated, i.e., they have the same propagation constant and it is possible to define the eigenmodes as orthogonally and linearly polarized plane waves. This is also the case for normally incident light. For oblique incidence the situation changes dramatically because both linearly polarized eigenstates may encounter different structures. Both eigensolutions may have different eigenvalues for the propagation constant. One may compare the situation with a uniaxial crystal, where the optical axis is aligned with the $z$ axis. For normally incident light the structure can be considered as being isotropic whereas at oblique incidence the eigenvalues for the two eigenpolarizations are different. Note that the eigenpolarizations are always orthogonally, linearly polarized for planes of incidence being identical with mirror planes of the metamaterial.

Considering the SCiC as an effectively isotropic homogeneous medium, we expect that both the two fundamental Bloch modes are degenerated and the effective refractive index is isotropic. So as soon as the eigenvalues for both Bloch modes deviate from each other, the medium cannot be described as being effectively isotropic. This deviation can also serve as a measure for a meaningful MM homogenization.

In Fig. 5(a) we show the real and the imaginary parts of the propagation constant for the two Bloch modes with the smallest losses at a certain angle of incidence and in Fig. 5(b) their difference as function of the frequency. Clearly for low frequencies the propagation constants for both modes are identical. As soon as the frequency increases the propagation constants start to deviate from each other hence the SCiC cannot be considered as optically isotropic anymore. The deviation is of course the stronger the larger the angle of incidence is. At normal incidence they are identical as explained in detail before. The conclusions to be drawn from these figures are of course in line with those from the angular dependency of the first Bloch mode only. The structure can be considered as being isotropic only for small frequencies and a limited range of angles of incidence where this angular range is the larger the smaller the frequency is.

Finally, some further remarks are necessary in order to elucidate the significance of the obtained results for other spectral domains. Although we have investigated a specific high-symmetry metamaterial here designed to operate in the optical domain, we firmly believe that the conclusions to be drawn will not change significantly by modifying the operational wavelength. First of all, the structure sizes when compared to the vacuum wavelength are already quite small. At the frequency of interest the wavelength to cell-size ratio is approximately 7. Even in the gigahertz range this ratio does not exceed 10 considerably. From Fig. 4 it can be deduced that even in the nonresonant regime for $\lambda/a \approx 15$ the deviation between the expected response and the achieved one exceeds 2% already at $k_0/a=1$. The structure cannot be made significantly electrically smaller if resonant metallic particles are used to achieve the artificial magnetism. But since the unit-cell design itself is already of cubic symmetry the deviations of the expected isotropic optical response to the actually obtained one is hence only due to the insufficiently large wavelength to cell-size ratio. To overcome these obstacles it is maybe necessary to go completely different ways.

**V. CONCLUSION**

In this contribution we have investigated the optical response of a highly symmetric SCiC MM in the negative refraction regime. The SCiC exhibits cubic symmetry, i.e., the highest possible symmetry for periodic metamaterials and is therefore considered a very promising candidate for an isotropic negative refractive-index material. By investigating
at first a finite number of functional layers and the convergence of the corresponding effective parameters to the values obtained from the dispersion relation of the infinite structure we can conclude that the SCiC fulfills all requirements of a homogenizable metamaterial. Already a single functional layer can be described by its bulk properties as only a single Bloch mode determines light propagation inside the structure. Nevertheless our investigation of the isofrequency surface of the dispersion relation clearly shows that even this metamaterial with the highest symmetry is far away from having an optically isotropic response in the region of negative refraction. This nonisotropic response is rather due to the large electrical size of the resonant unit cells than due to an insufficient design and will unlikely to be solved by shifting the spectral domain of operation to the microwave regime. Hence, the key result of this contribution is that one must not conclude from high symmetry on an optically isotropic response. Hence, optical isotropy of a MM can be only deduced from a careful inspection of the dispersion relation by taking into account the material dispersion of metal too. This is an important message to the designers of isotropic optical metamaterials.

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