Grahn, Patrick; Shevchenko, A.; Kaivola, M.

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Internally twisted non-centrosymmetric optical metamaterials

P. Grahn, Andriy Shevchenko, Matti Kaivola


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Internally twisted non-centrosymmetric optical metamaterials

P. Grahn, A. Shevchenko, M. Kaivola
Department of Applied Physics, Aalto University, P.O. Box 13500, FI-00076 Aalto, Finland

ABSTRACT

In optical metamaterials, the scatterers are usually aligned symmetrically with respect to the unit cells of the material. In this work, we consider metamaterials in which the “metamolecules” can be non-centrosymmetric and have an arbitrary, but common, orientation in the unit cells. Such internally twisted crystalline structures are difficult to find in natural materials, but metamaterials of this type can be designed and fabricated at will. Here we present a theoretical method that enables a detailed analysis of internally twisted non-centrosymmetric metamaterials. The method establishes a connection between the optical properties of a metamaterial and the plane-wave optical response of a single two-dimensional array of metamolecules. In this theory, the effective wave parameters, such as the refractive index and wave impedance, are retrieved. Using the model, we show that these parameters can dramatically depend on the wave propagation direction and metamolecular orientation in a metamaterial. This dependence provides a possibility to adjust and control the plane-wave content of optical beams propagating in the material.

Keywords: metamolecular orientation, propagationally anisotropic metamaterials, non-centrosymmetric scatterers, spatial dispersion, wave parameters

1. INTRODUCTION

In recent years, a wide range of artificial optical materials have been designed with the aim of manipulating light in desired ways. Based on the size of the unit cells, these materials can be conditionally divided into three categories. Photonic crystals compose one of them. They are periodic structures characterized by highly symmetric constituents, such as spheres and holes, with the unit-cell size being on the order of \( \lambda/2 \) (\( \lambda \) is the wavelength of light). These materials exhibit photonic band gaps used to confine and guide optical waves. Photonic crystals are often characterized by Bloch modes and the corresponding dispersion diagrams, such as isofrequency surfaces and contours. Light propagation in a photonic crystal is governed by diffraction and depends on the propagation direction. The second category includes materials with unit cells that are several orders of magnitude smaller than \( \lambda \). The optical properties of such a material depend only on the local strength and orientation of the electric field component of light. These materials can be described in terms of an electric permittivity tensor or refractive index ellipsoid using effective medium theories.\(^1\)\(^,\)\(^2\) They are not optically magnetic and their properties are independent of the light propagation direction. We thereby call these materials propagationally isotropic metamaterials. The third category of artificial materials, which is the type considered in this work, consists of those which fit in the range between the mentioned photonic crystals and the propagationally isotropic metamaterials. In these materials, the metamolecules have designed nontrivial shapes and the unit cell size is shorter than \( \lambda/2 \), but not much. As a result, while no diffraction orders can appear in any direction, the material’s optical response is influenced by the direction of the wave propagation. These metamaterials are spatially dispersive and we therefore call them propagationally anisotropic metamaterials. Their effective refractive index and wave impedance are well-defined. Furthermore, in such metamaterials, the moments of higher-order multipoles, such as electric quadrupoles or magnetic dipoles, are not negligible. Consequently, the effective wave impedance is no longer determined by the refractive index. Therefore, the characterization of these metamaterials requires both the effective refractive index and wave impedance along with their dependence on the propagation direction.

By using propagationally anisotropic metamaterials one can achieve many extraordinary optical phenomena similar to those observed in photonic crystals, but without diffraction effects. These phenomena include the superprism phenomenon,\(^3\)\(^,\)\(^4\) self-collimation\(^5\)\(^,\)\(^6\) and negative refraction with positive refractive index.\(^7\) They can appear due to non-circular isofrequency contours caused by spatial dispersion. The spatial dispersion, in turn, can be enhanced by breaking the symmetry of the material. Previously, we have designed a material in which the
metamolecules are not centrosymmetric, which makes the material bifacial.\textsuperscript{9,10} In this work, we consider non-centrosymmetric metamolecules, which are tilted with respect to the crystal lattice axes. This further enhances the propagational anisotropy of the metamaterial. Since the metamolecules no longer share their symmetry axis with the lattice, we coin the material as internally twisted.

The optical properties of an arbitrary metamaterial can be fully characterized in terms of the metamaterial's response to optical plane waves of different frequencies, polarizations and propagation directions. For crystalline metamaterials, the plane-wave response can be efficiently evaluated using a recently proposed interferometric approach.\textsuperscript{10} Within this approach, the metamaterial crystal is divided into planar arrays of metamolecules, as shown in Fig. 1. The overall interaction of light with each array is determined by plane-wave transmission and reflection coefficients that in general depend on the light frequency, polarization and propagation direction. The interaction picture is obtained by considering the multiple reflections and transmissions of light by the consecutive metamolecular layers, as shown in Fig. 1.

To explain the approach in more detail, we consider a plane wave propagating at an angle $\theta$ with respect to the $z$-axis (see Fig. 1). The crystal planes are periodically separated by the lattice constant $\Lambda_z$. The finite reflection coefficient of the sheets, with which we replace the horizontal arrays of molecules, produces a second wave that propagates at an angle $180^\circ - \theta$ with respect to the $z$-axis. In general, the transmission and reflection coefficients $\tau_2$ and $\rho_2$ experienced by this reflected wave differ from $\tau_1$ and $\rho_1$ of the original wave. Only for propagation along the $z$-axis does the equality $\tau_1 = \tau_2$ necessarily hold. Here, we consider arbitrary propagation directions and polarizations, for which $\tau_1 \neq \tau_2$ and $\rho_1 \neq \rho_2$. Our consideration also includes metamaterials that are not symmetric with respect to the $z$-axis. The metamolecules are twisted (their symmetry axis is tilted by an angle $\alpha$ with respect to $z$), resulting in the inequality $\tau_1 \neq \tau_2$. A theory suitable for such metamaterials has not been introduced previously. Hence, we do this in the following section.
2. THEORETICAL DESCRIPTION

For a metamaterial, a recursive relation that connects the fields between adjacent crystal planes can be written as (see Eq. (3) in Ref. 10)

$$\beta U_{j+1} + U_{j-1} - \alpha U_j = 0, \quad (1)$$

where $U_j$ denotes the amplitude of the forward-propagating plane wave after the $j$'th sheet of the particles. The parameters $\alpha$ and $\beta$ are related to the phase-shifted transmission and reflection coefficients $f_1 = \tau_1 \exp(ik_z\Lambda_z)$, $f_2 = \tau_2 \exp(ik_z\Lambda_z)$, $g_1 = \rho_1 \exp(ik_z\Lambda_z)$ and $g_2 = \rho_2 \exp(ik_z\Lambda_z)$ by

$$\alpha = f_2 + f_1^{-1}(1 - g_1g_2), \quad (2)$$

$$\beta = f_2/f_1. \quad (3)$$

Here $k_z$ denotes the $z$ component of the wave vector in the host medium that surrounds the scatterers. The wave number in the effective medium is denoted by $\gamma$ and the $z$ component of the corresponding wave vector by $\gamma_z$. We require that translation by one unit cell in the effective medium satisfies the Bloch periodicity condition

$$U_j = U_{j-1} \exp(i\gamma_z\Lambda_z). \quad (4)$$

Inserting Eq. (4) into Eq. (1) allows us to solve for

$$\gamma_z\Lambda_z = -i\ln \left[ \frac{\alpha}{2\beta} \pm \left( \frac{\alpha^2}{4\beta^2} - \frac{1}{\beta} \right)^{1/2} \right] + 2\pi m, \quad (5)$$

where $m$ is an integer. At an interface between the metamaterial and the host medium, the wave vector components that are tangential to the interface are preserved, i.e., $\gamma_x = k_x$ and $\gamma_y = k_y$. Thus, the effective refractive index $n_{\text{eff}}$ can straightforwardly be calculated from

$$\gamma^2 = n_{\text{eff}}^2 k_0^2 = k_x^2 + k_y^2 + \gamma_z^2, \quad (6)$$

where $k_0$ denotes the wave number in vacuum. Together, Eqs. (5) and (6) enable the determination of the effective refractive index from the transmission and reflection coefficients of a single planar array of nanoscaters. This approach considers the evanescent-wave coupling between the metamolecular layers as negligible.

In order to obtain the effective wave impedance of a metamaterial, one must average the electric and magnetic fields inside the material. According to Ref. 10, the effective wave impedance can be expressed as

$$Z_{\text{eff}} = Z \left( \frac{k\gamma_z}{k_z\gamma} \right)^p \frac{\langle U_j \exp(ik_zz) + U'_j \exp(-ik_zz) \rangle}{\langle U_j \exp(ik_zz) - U'_j \exp(-ik_zz) \rangle}, \quad (7)$$

where $Z$ is the impedance of the host medium and $p$ is equal to +1 and −1 for TE- and TM-polarized light, respectively. The factor $(k\gamma_z/k_z\gamma)^p$ accounts for the fact that the propagation direction of the effective wave differs from that in the host medium. The variable $U'_j$ denotes the amplitude of the backward propagating plane wave after the $j$'th sheet and is related to $U_j$ through

$$U_j = f_1U_{j-1} + g_2U'_j. \quad (8)$$

Performing the spatial averaging over $z$ in Eq. (7) and using Eq. (8) we obtain the following expression for the effective wave impedance,

$$Z_{\text{eff}} = Z \left( \frac{k\gamma_z}{k_z\gamma} \right)^p \frac{g_2 + [1 - f_1 \exp(-i\gamma_z\Lambda_z)]}{g_2 - [1 - f_1 \exp(-i\gamma_z\Lambda_z)]}. \quad (9)$$

The effective wave parameters $n_{\text{eff}}$ and $Z_{\text{eff}}$, along with their dependence on the wave frequency, propagation direction and polarization, form a complete macroscopic description of a twisted crystalline metamaterial. In order to calculate the reflection, transmission and refraction at a metamaterial boundary, one needs to know the Fresnel coefficients. In Ref. 9, these coefficients are generalized for uniaxial bifacial metamaterials. In order to allow for the wave internally reflected from the material boundary to have a refractive index that differs from that...
of the incident wave, we further generalize these coefficients to cover also such twisted metamaterials. Consider a boundary at \( z = 0 \) between two materials of which either one can be a twisted metamaterial. Suppose that a plane wave is incident from the first material with a wave vector \( (\gamma_x, \gamma_y, \gamma_z) \), wave number \( \gamma \) and wave impedance \( Z_i \). In general, we can express the characteristics of the wave reflected by the interface as \( (\gamma_x, \gamma_y, -\gamma_z) \) and \( Z_r \). Whereas for conventional materials, one would obtain \( \gamma_{z,r} = \gamma_{z,i} \), \( \gamma_t = \gamma_i \) and \( Z_r = Z_i \), these equalities will not necessarily hold for metamaterials. For example, for uniaxial bifacial metamaterials the impedances can be different. For the internally twisted metamaterials, on the other hand, also the refractive indices and, therefore, the wave vectors can have different values. For the wave transmitted by the interface we define the characteristics \( (\gamma_x, \gamma_y, \gamma_z) \), \( \gamma_t \) and \( Z_i \). Applying the electromagnetic boundary conditions, we obtain the Fresnel transmission and reflection coefficients, \( \tau_F \) and \( \rho_F \), for internally twisted metamaterials to be

\[
\tau_F = \frac{\gamma_{z,i}/(\gamma_i Z_i^r) + \gamma_{z,t}/(\gamma_t Z_t^r)}{\gamma_{z,i}/(\gamma_i Z_i^r) + \gamma_{z,t}/(\gamma_t Z_t^r)} \left( \frac{Z_i}{Z_t} \right)^{(\sigma-1)/2},
\]

\[
\rho_F = \frac{\gamma_{z,i}/(\gamma_i Z_i^r) - \gamma_{z,t}/(\gamma_t Z_t^r)}{\gamma_{z,i}/(\gamma_i Z_i^r) + \gamma_{z,t}/(\gamma_t Z_t^r)} \left( -\frac{Z_i}{Z_t} \right)^{(\sigma-1)/2},
\]

where \( \sigma = \pm 1 \) for the TE and TM polarization, respectively. Here a positive real-valued \( \rho^{TM}_F \) indicates that the incident and reflected waves have their tangential components of the electric field in phase. The derived Fresnel coefficients can be used to calculate, e.g., the transmission and reflection coefficients of slabs of arbitrary crystalline metamaterials.

### 3. A DISC DIMER METAMATERIAL

As an example of an internally twisted metamaterial, we consider a cubic lattice of asymmetric paired silver discs with a 120 nm lattice constant [see Fig. 2(a)]. These metamolecules are found to exhibit significant magnetic dipole and electric quadrupole polarizabilities\(^{11}\) and spatial dispersion\(^{12}\) in the visible spectral range. Using electromagnetic multipole theory,\(^{13}\) it can be verified that the dominant higher-order multipole excitation in the particles is composed of linear currents in the two discs oscillating out-of-phase with respect to each other. We choose the larger disc to have a radius of 40 nm and the smaller one 25 nm. Both discs have a thickness of 10 nm and they are separated by a surface-to-surface distance of 20 nm. The dimer axis is allowed to be tilted by an angle \( \alpha \) in the \( yz\)-plane. The angle of incidence of a plane wave is denoted by \( \theta \), such that the plane of incidence coincides with the \( yz\)-plane. Also, when \( \theta \) is equal to \( \alpha \), the wave is incident from the smaller-disc side. A dielectric host medium of refractive index 1.5 is assumed throughout this work.

In order to calculate the effective wave parameters of the metamaterial, we first numerically calculate the transmission and reflection coefficients of a single planar array of the dimers. The numerical calculations are performed using the computer software COMSOL Multiphysics and the values for the refractive index of silver are taken from Ref. 14. For each incidence angle \( \theta \), two calculations are required, one for a wave incident at \( \theta \) (giving \( \tau_1 \) and \( \rho_1 \)) and one for a wave incident at an angle of \( 180^\circ - \theta \) (giving \( \tau_2 \) and \( \rho_2 \)). Then, using Eqs. (5) and (9), we calculate the effective wave parameters for the metamaterial crystal. Since the material is both spatially dispersive and anisotropic, the retrieved parameters depend on the polarization and propagation direction of light. In Figs. 2(b) and (c) we show the spectra of the wave parameters retrieved for the simplest case, where \( \theta = 0 \) and \( \alpha = 0 \). In the refractive index spectrum, one can distinguish the dipole resonances of the two discs composing the dimer. For \( \alpha = 0 \) we can have \( \rho_1 \neq \rho_2 \), but the equality \( \tau_1 = \tau_2 \) must be the case for all choices of \( \theta \). Indeed, for a counter-propagating wave (\( \theta = 180^\circ \)), the refractive index is the same as for the original wave, but the impedance shown by the red lines in Fig. 2(c) is different. Thus, this metamaterial is optically bifacial.

The metamaterial becomes internally twisted if the dimers are tilted, e.g., by an angle \( \alpha = 45^\circ \). By calculating the transmission and reflection coefficients for \( \theta = 45^\circ \) and \( \theta = 135^\circ \) – such that the wave still propagates along the symmetry axis of the dimers – we obtain the effective parameters for these waves. These parameters are shown in Fig. 3 for TE-polarized plane waves propagating in the directions of \( \theta = 45^\circ \) and \( \theta = 135^\circ \). We notice that in this case the two waves experience different refractive indices and different wave impedances. It can be seen that at certain wavelengths, blue detuned from the dipole resonances, the real part of the refractive index takes values close to zero and even becomes negative. This feature shows that the effect of a twist can be remarkable.
Figure 2. (a) Transmission and reflection of a plane wave by a single plane of metal nanodimers composing the metama-terial crystal. In the spectra, the black lines show the calculated effective (b) refractive index and (c) wave impedance (normalized to that of vacuum) of the metamaterial for waves propagating at $\theta = 0$, when $\alpha = 0$. The wave impedance for a counter-propagating wave ($\theta = 180^\circ$) is shown with red lines. The real and imaginary parts are shown by solid and dashed lines, respectively.

Figure 3. Effective wave parameters for a metamaterial with the dimers tilted by $\alpha = 45^\circ$. The effective refractive index and wave impedance are shown for waves propagating at (a,c) $\theta = 45^\circ$ and (b,d) $\theta = 135^\circ$. The real and imaginary parts are shown by solid and dashed lines, respectively.
3.1 Verification of the theory

In order for a metamaterial to be treatable as a homogeneous material, it is necessary that the calculated wave parameters do not depend on the size of the crystal. This is equivalent to the requirement that the evanescent-wave coupling between adjacent crystal planes is weak.\textsuperscript{10} According to the practical criterion proposed in Ref. 10 for noble-metal metamolecules, the gap $d$ between the particles in the $z$-direction must be larger than $\Lambda_{\text{max}}/2$, where $\Lambda_{\text{max}}$ is the largest lattice constant of the crystal plane. For the considered dimer metamaterial, we have $d \approx 80$ nm and $\Lambda_{\text{max}}/2 = 60$ nm, so that the metamaterial is expected to be homogenizable.

In order to verify the correctness of the approach, we compute directly the transmission and reflection through five molecular layers of a metamaterial with $\alpha = 45^\circ$ at $\theta = 45^\circ$. The interaction geometry is depicted in Fig. 4(a). The obtained intensity transmission and reflection spectra are shown in Fig. 4(b) by solid lines. Then, inserting the previously calculated effective wave parameters (see Fig. 3) into Eqs. (10) and (11), we obtain the Fresnel coefficients for an interface between the metamaterial and the host dielectric for waves propagating at $\theta = 45^\circ$ and $\theta = 135^\circ$. The transmission and reflection by a homogeneous slab characterized by these parameters and having the same five-layer thickness is then obtained by using these Fresnel coefficients in the standard equations for a Fabry-Perot etalon (see, e.g., equations (25) and (26) in Ref. 9). The intensity transmittance and reflectance calculated in this way are shown by stars in Fig. 4(b). The obtained agreement between the analytical results and the direct numerical calculations confirms the homogenizability of the metamaterial and the correctness of our calculation approach.
Figure 5. The angle $\alpha$ between the orientation of the dimers and the $z$-axis is increased from $0^\circ$ to $360^\circ$. The wave is TE-polarized and it propagates in the positive $z$-direction ($\theta = 0^\circ$). The calculated wave parameters, $n_{\text{eff}}$ and $Z_{\text{eff}}/Z_0$, are shown as functions of $\alpha$ for vacuum wavelengths of (b) $\lambda_0 = 525$ nm, (c) $\lambda_0 = 580$ nm, (d) $\lambda_0 = 730$ nm, (e) $\lambda_0 = 790$ nm and (f) $\lambda_0 = 1000$ nm. The real and imaginary parts are shown by solid and dashed lines, respectively. The radius in the polar plots indicates the magnitude, whereas a blue (red) color indicates whether the value is positive (negative).

### 3.2 Influence of metamolecular orientation

The influence of the dimer orientation can be analyzed in more detail by calculating the wave parameters as a function of the angle $\alpha$ at a fixed propagation direction, e.g., for $\theta = 0$ [see Fig. 5(a)]. The incident light is assumed to be polarized along the $x$-axis, which ensures that for all choices of $\alpha$ the incident field points in the same direction relative to the discs, keeping the effect of optical anisotropy fixed. For the calculations, we select five particular wavelengths:
λ₀ = 525 nm (blue-detuned from the resonances),
λ₀ = 580 nm (nearly on-resonance with the smaller disc),
λ₀ = 730 nm (between the two resonances),
λ₀ = 790 nm (nearly on-resonance with the larger disc),
λ₀ = 1000 nm (considerably red-detuned from the resonances).

For each of these wavelengths, we evaluate and present nₑffective and Zₑffective as functions of α in the polar plots (b)-(f) of Fig. 5. Considering first the case of λ₀ = 1000 nm, shown in Fig. 5(f), we see that the wave parameters are independent of α. At this long a wavelength, the material responds like a spatially nondispersive dielectric with a refractive index of 2. Similarly, the response at λ₀ = 525 nm, shown in Fig. 5(b), is nearly independent of α. However, at this wavelength, the excitation in the dimers is essentially out-of-phase with the incident field, leading to a refractive index of nₑffective = 1. This is the value of the refractive index of vacuum. However, the material is not quite impedance matched to vacuum. This means that, while optical refraction is absent, some reflection from the material will still take place. The situation changes dramatically when the wavelengths approaching the dipole resonances of the discs are considered. For λ₀ = 580 nm, shown in Fig. 5(c), both wave parameters significantly depend on α and have large imaginary parts. The real part of the effective refractive index is observed to be negative when the dimer axis is about perpendicular (90° ± 50°) to the propagation direction of light (see the red line). In contrast, at the resonance wavelength of the larger disc, λ₀ = 790 nm [see Fig. 5(d)], the real part of the refractive index is positive and quite large (nₑffective ≈ 3). At this wavelength, however, the wave impedance is essentially imaginary, leading to a high reflectivity. Finally, we consider the excitation at λ₀ = 730 nm [see Fig. 5(e)]. At this wavelength, the imaginary parts of the wave parameters are smaller, and at certain values of α, light can penetrate relatively deeply into the material. The refractive index and especially the impedance are found to depend strongly on α. At α = 75°, the imaginary part of Zₑffective crosses zero and becomes negative (see the red dotted line). Note that for all five wavelengths, the conditions Im{nₑffective} > 0 and Re{Zₑffective} > 0 hold for all values of α, as they should. From the obtained results, it is clear that the orientation of the dimers significantly affects the wave parameters when light is close to one of the resonance frequencies of the discs. We emphasize that this phenomenon has nothing to do with optical anisotropy and, therefore, can be considered as an additional tool for adjusting the properties of artificial optical materials.

3.3 Influence of light propagation direction

The relative orientation between the light propagation direction and the direction of the metamolecular alignment does not uniquely determine the effective wave parameters, because also the orientation of the lattice is important. Consequently, the wave parameters depend on the propagation direction θ in a different way than on the metamolecular orientation α considered in the previous subsection. Let us twist the metamolecules in the crystal cells to an angle α = 45° and calculate nₑffective and Zₑffective as functions of θ at the five spectral locations selected previously. We still consider the TE-polarized light (polarized along the x-axis) to keep constant the influence of optical anisotropy. The obtained wave parameters are shown in the polar plots of Fig. 6. Far to the red from the resonances, at λ₀ = 1000 nm [see Fig. 6(f)], the wave parameters are, as expected, independent of θ. Also at λ₀ = 525 nm [Fig. 6(b)], the refractive index is independent of θ. However, at this wavelength we have nₑffective ≈ 1 and, therefore, any beam of light will propagate inside the material approximately as in vacuum [note the similarity with Fig. 5(b)]. The wave impedance is also seen to depend on θ, but quite weakly, referring to an insignificant spatial dispersion in the metamaterial. The situation changes near the resonance wavelengths of the individual discs. At λ₀ = 580 nm [Fig. 6(c)], the wave parameters are extremely sensitive to θ. Here the real part of the refractive index is close to zero when light propagates along the dimer axis (θ = 45° or θ = 225°), and then it changes abruptly to the value of 2 and stays constant within a range spanning approximately 60°. Also the wave impedance is extremely sensitive to θ at this wavelength. However, the large imaginary part of Zₑffective makes the metamaterial highly reflective when the material is surrounded by an ordinary dielectric medium. The same holds also around the second resonance, at λ₀ = 790 nm, where Im{Zₑffective} ≫ Re{Zₑffective} [see Fig. 6(e)]. The large refractive index at λ₀ = 790 nm varies only slightly with θ, which could be anticipated from the locally flat spectra shown earlier in Figs. 3(a) and (b). Between the resonances, at the wavelength of λ₀ = 730 nm [Fig. 6(d)], the wave impedance is large, almost real-valued, and it dramatically depends on θ.
Figure 6. The wave propagation direction in the host medium $\theta$ is changed from $0^\circ$ to $360^\circ$. The orientation of the dimers is fixed to $\alpha = 45^\circ$ and the wave is TE-polarized. The calculated wave parameters, $n_{\text{eff}}$ and $Z_{\text{eff}}/Z_0$, are shown as functions of $\theta$ for vacuum wavelengths of (b) $\lambda_0 = 525$ nm, (c) $\lambda_0 = 580$ nm, (d) $\lambda_0 = 730$ nm, (e) $\lambda_0 = 790$ nm and (f) $\lambda_0 = 1000$ nm. The real and imaginary parts are shown by solid and dashed lines, respectively. The radius in the polar plots indicates the magnitude, whereas a blue (red) color indicates whether the value is positive (negative).

It is obvious that the dependence of both the real and imaginary parts of $n_{\text{eff}}$ and $Z_{\text{eff}}$ on $\theta$ considerably differs from the dependence of these quantities on $\alpha$ obtained in the previous subsection. This means that the optical response of the metamaterial at a particular incidence angle $\theta$ can be efficiently tuned by the metamolecular alignment. The “twist” indeed makes the response strongly depend on the propagation direction via the phenomenon of spatial dispersion.

4. CONCLUSIONS

In this work, we have introduced a general technique for calculating the effective wave parameters of internally twisted crystalline metamaterials composed of non-centrosymmetric metamolecules that are tilted with respect to the lattice. In these propagationally anisotropic metamaterials, both the effective refractive index $n_{\text{eff}}$ and
wave impedance $Z_{\text{eff}}$ significantly depend on the propagation direction of light, even if the light polarization is such that the optical anisotropy does not contribute to this dependence. Thus, to characterize these materials it is necessary to consider the wave parameters as functions of not only the frequency and polarization, but also of the propagation direction of light. Previously used Fresnel transmission and reflection coefficients are not valid for such materials. We have therefore generalized these coefficients to allow for the incident and reflected waves to “see” different optical media.

The applicability of the technique was demonstrated by investigating an optical metamaterial composed of paired silver discs embedded in glass. In the metamaterial, the common orientation of the dimers was considered to be independent of the orientation of the crystal lattice, which provided a control over the spatial dispersion of the material. For this metamaterial, we have verified that the introduced wave parameters do not depend on the number of molecular layers in the material and are therefore defined unambiguously. We have studied in detail the dependence of $n_{\text{eff}}$ and $Z_{\text{eff}}$ on the orientation of the metamolecules and found that, close to the resonance wavelengths of the discs, both the refractive index and the wave impedance are significantly influenced by this orientation. We have also analyzed the dependence of $n_{\text{eff}}$ and $Z_{\text{eff}}$ on the propagation direction of light. This dependence was shown to give a powerful tool for adjusting the metamaterial properties in addition to classical optical anisotropy. This fact opens up new opportunities for designing functional metamaterials, which are purposefully tailored to reflect, refract or guide light beams by harnessing the dependence of their plane-wave components on the propagation direction.

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