Asymmetric Transmission of Linearly Polarized Light at Optical Metamaterials

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We experimentally demonstrate a three-dimensional chiral optical metamaterial that exhibits an asymmetric transmission for forwardly and backwardly propagating linearly polarized light. The observation of this novel effect requires a metamaterial composed of three-dimensional chiral meta-atoms without any rotational symmetry. Our analysis is supported by a systematic investigation of the transmission matrices for arbitrarily complex, generally lossy media that allows deriving a simple criterion for asymmetric transmission in an arbitrary polarization base. Contrary to physical intuition, in general the polarization eigenstates in such three-dimensional and low-symmetry metamaterials do not obey fixed relations and the associated transmission matrices cannot be symmetrized.

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During the past several years optical metamaterials (MMs) have attracted an enormous amount of interest since they promise to allow for a manipulation of light propagation to a seemingly arbitrary extent. MMs are usually obtained by assembling subwavelength unit cell structures called meta-atoms. Initial studies on MMs were based on rather simple and highly symmetric meta-atoms [1–3]. Recently, more and more sophisticated structures were explored in order to achieve customized functionalities such as, e.g., a negative refractive index due to chirality [4–6]. Also, a large variety of plasmonic meta-atoms were investigated that evoke a huge polarization rotation like gammadions, omega-shaped particles, or helices [7–9]. Studying the characteristics of light propagation in such low-symmetry MMs also revealed unexpected phenomena like asymmetric transmission for circularly polarized light [10–14]. Although at first sight this effect of nonreciprocal transmission, to date not observed for linearly polarized light, is counterintuitive, it does not violate Lorentz's reciprocity theorem. This asymmetric transmission of circularly polarized light was demonstrated at so-called planar chiral MMs. Such MMs are composed of meta-atoms without structural variation in the principal propagation direction. They preserve symmetry in this direction and are only chiral in the two-dimensional space [15]; thus, strictly speaking, they are intrinsically achiral in three dimensions since the mirror image of a structure is congruent with the structure itself if operated from the back. The remaining mirroring plane is perpendicular to the propagation direction.

In this Letter we theoretically and experimentally demonstrate a novel MM design which breaks the latter symmetry. For the first time our approach reveals that the very structures exhibit asymmetric transmission for linearly polarized light. We emphasize that also in this case the reciprocity theorem is not violated since only reciprocal materials are involved. Since our approach is solely based on symmetry considerations it is, in particular, valid for lossless systems, too.

Prior to any further considerations we concisely discuss the effect of a potential MM substrate that is, after all, in most cases required for fabricating planar MMs. Generally speaking, just this supporting substrate breaks the mirror symmetry for any planar structure perpendicular to the propagation direction [16]. However, the effect of this symmetry breaking is almost negligible when compared to the impact of a strong structural anisotropy that leads, e.g., to asymmetric transmission of circularly polarized light [10,11]. On the other hand, if no structural anisotropy is present, such as, e.g., for gammadions, the chirality of the planar metatom-substrate system yields a measurable polarization rotation [17]. So, although asymmetric transmission for linearly polarized light may have been observed already, its magnitude was too weak and it was merely attributed to an insufficient measurement accuracy rather than to a true physical mechanism. To unambiguously enhance the effect towards a measurable extent, it is necessary to deliberately break the symmetry in the third dimension of the meta-atom itself and not just rely on the rather perturbative effect of a supporting substrate. Exactly this goal is pursued in this work.

We start by deriving a criterion for the occurrence of asymmetric transmission in an arbitrary polarization base and particularly for linearly polarized light. The transmission of coherent light through any dispersive optical system can be described by means of complex Jones matrices T [10,12]. We consider an incoming plane wave that propagates in positive z direction

$$\mathbf{E}_i(\mathbf{r}, t) = \begin{pmatrix} I_x \\ I_y \end{pmatrix} e^{i(kz-\omega t)},$$

with frequency ω, wave vector k, and complex amplitudes

I_x and I_y. The transmitted field is then given by

$$\mathbf{E}_t(\mathbf{r}, t) = \begin{pmatrix} T_x \\ T_y \end{pmatrix} e^{i(kz-\omega t)},$$

where it is assumed that the medium is symmetrically embedded, e.g., in vacuum. The T matrix relates the generally complex amplitudes of the incident field to the complex amplitudes of the transmitted field:

$$\begin{pmatrix} T_x \\ T_y \end{pmatrix} = \begin{pmatrix} T_{xx} & T_{xy} \\ T_{yx} & T_{yy} \end{pmatrix}\begin{pmatrix} I_x \\ I_y \end{pmatrix} = \begin{pmatrix} A & B \\ C & D \end{pmatrix}\begin{pmatrix} I_x \\ I_y \end{pmatrix} = \hat{T}^f_{\text{lin}}\begin{pmatrix} I_x \\ I_y \end{pmatrix}, \tag{1}$$

where we have replaced T_{ij} by A, B, C, D for convenience. The indices f and lin indicate propagation in forward direction and a special linear base with base vectors parallel to the coordinate axes, i.e., decomposing the field into x- and y-polarized light. Applying the reciprocity theorem delivers the T matrix $\hat{T}^b$ for propagation in $-z$ or backward direction,

$$\hat{T}^b_{\text{lin}} = \begin{pmatrix} A & -C \\ -B & D \end{pmatrix}, \tag{2}$$

characterizing the transmission in a fixed coordinate system with the sample being rotated by 180° with respect to either the x or the y axis. The form of Eq. (2) results from the reciprocity of four-port systems and the definition of the coordinate system [18]. In general, all components A, B, C, and D are mutually different and strongly dispersive. They obey fixed relations only for certain symmetries. For example, for planar, anisotropic 2D chiral meta-atoms the mirror image coincides with that seen from the back side, i.e., $B = C$, and therefore

$$\hat{T}^f_{\text{lin}} = \begin{pmatrix} A & B \\ B & D \end{pmatrix} \quad \text{and} \quad \hat{T}^b_{\text{lin}} = \begin{pmatrix} A & -B \\ -B & D \end{pmatrix}. \tag{3}$$

On the other hand, for gammadions on a substrate (or trapezoidally shaped) with a C_4 symmetry with respect to the z axis, we have $D = A$ and $C = -B$, and therefore

$$\hat{T}^f_{\text{lin}} = \hat{T}^b_{\text{lin}} = \begin{pmatrix} A & B \\ -B & A \end{pmatrix}. \tag{4}$$

For analytical as well as for experimental purposes it is advantageous to also have at hand the T matrix for circularly polarized light. It can be obtained by a change of the base vectors from linear to circular states resulting in

$$\hat{T}^f_{\text{circ}} = \frac{1}{2}\begin{pmatrix} [A + D + i(B - C)] & [A - D - i(B + C)] \\ [A - D + i(B + C)] & [A + D - i(B - C)] \end{pmatrix}, \tag{5}$$

connecting the amplitudes of circularly polarized incident light with those of circularly polarized transmitted light:

$$\begin{pmatrix} T_+ \\ T_- \end{pmatrix} = T^f_{\text{circ}}\begin{pmatrix} I_+ \\ I_- \end{pmatrix} = \begin{pmatrix} T_{++} & T_{+-} \\ T_{-+} & T_{--} \end{pmatrix}\begin{pmatrix} I_+ \\ I_- \end{pmatrix}.$$

By using Eq. (2) we find for propagation in $-z$ direction

$$T^b_{\text{circ}} = \begin{pmatrix} T_{++} & T_{-+} \\ T_{+-} & T_{--} \end{pmatrix}.$$

Now we introduce the overall transmission in an arbitrary base. The base vectors denoted by $\mathbf{e}_1$ and $\mathbf{e}_2$ are assumed to be normalized and linearly independent. The overall transmission may then be written as

$$\mathbf{T}^{f,b} = T_1^{f,b}\mathbf{e}_1 + T_2^{f,b}\mathbf{e}_2 = \hat{T}^{f,b}\{I_1\mathbf{e}_1 + I_2\mathbf{e}_2\}.$$

Asymmetric transmission for a given base vector $i \in \{1, 2\}$ can then be defined as the difference between the transmitted intensities for different propagation directions as

$$\Delta(I_i = 1; I_j = 0) \doteq \Delta^{(i)} = |\mathbf{T}^f|^2 - |\mathbf{T}^b|^2. \tag{6}$$

For the linear and the circular base we obtain

$$\Delta^{(x)}_{\text{lin}} = |C|^2 - |B|^2 = -\Delta^{(y)}_{\text{lin}} \tag{7}$$

and

$$\Delta^{(+)}_{\text{circ}} = |T_{-+}|^2 - |T_{+-}|^2 = -\Delta^{(-)}_{\text{circ}}, \tag{8}$$

respectively.

Obviously, the effect of asymmetric transmission depends on the specific base. For both bases given here it is exclusively determined by the difference between the off-diagonal elements of the respective T matrices. For structures obeying, e.g., Eq. (3), we have $\Delta_{\text{lin}} = 0$, whereas $\Delta_{\text{circ}} \neq 0$. Note that for an arbitrary base the diagonal elements of the T matrix for different propagation directions are not identical and will also contribute to asymmetric transmission.

From this analysis we can conclude that for MMs featuring $A \neq B \neq C \neq D$ asymmetric transmission will be observable in any base and, in particular, for linearly polarized light, too. To prove this statement we designed a simple meta-atom that is essentially three dimensional, nonsymmetric, and consists of strongly coupled plasmonic elements. To rule out asymmetric effects due to the presence of a substrate, the meta-atoms are completely embedded in an index-matched dielectric host. The geometry of the structure is shown in Fig. 1(a). It consists of two closely spaced layers. The first layer comprises an L-shaped metallic particle and the second one a single nanowire. The L-shaped structure on its own causes an anisotropic response if both arms are dissimilar, giving rise to asymmetric transmission for circularly polarized light. Adding the nanowire breaks the remaining symmetry in the principal propagation direction and leads to a genuine three-dimensional chiral meta-atom. Note that the particles themselves, in general, sustain three first-order plasmonic resonances; one may be associated with the nanowire and the two others with the L-shaped particle. The coupling of these particles results in a complex response featuring various resonances. The specific hybrid plasmonic eigen-

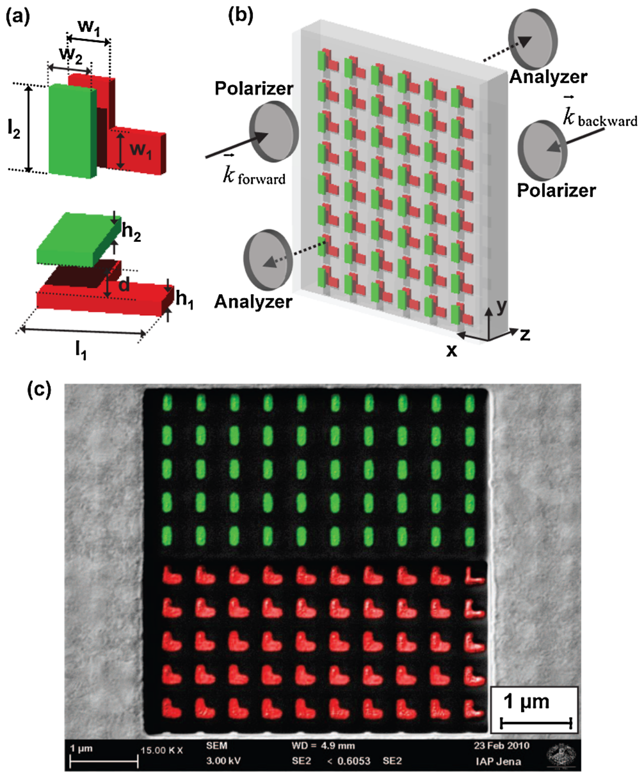

FIG. 1 (color online). (a) Two sketches of the MM unit cell from different perspectives with the definition of the geometrical parameters: $l_1 = l_2 = 290$ nm, $w_1 = w_2 = 130$ nm, $h_1 = h_2 = 40$ nm, $d = 80$ nm. (b) Schematic of the experimental setup. Normally incident light was linearly polarized before propagating through the MM and subsequently analyzed by means of a second linear polarizer. (c) Normal view electron micrograph with false colors of the fabricated MM. The meta-atoms were completely embedded in an index-matched dielectric. Focused ion beam slicing reveals the two layers composing the MM. Green and red colors represent the nanowires and the L structures in the top and bottom layer, respectively. The periods in both the x and y directions are 500 nm.

modes are of minor importance for our purpose and are beyond the scope of this work, though they can be easily accessed within the framework of an oscillator model [19]. The meta-atoms are periodically arranged on a square lattice in the x-y plane with periods smaller than the wavelength so that only the zeroth diffraction order propagates. A schematic of the experiment is shown in Fig. 1(b), where we measured the transmitted intensities through a single MM layer at normal incidence for both forward and backward propagation direction, corresponding to positive and negative z direction. Note that resonant, lossy meta-atoms enhance considerably the effect of asymmetric transmission but are not required to observe the very effect. However, the effect can be hardly measured in lossless media.

The MM was fabricated as follows. Both layers were defined in gold using standard electron-beam lithography (Vistec SB350OS) and a liftoff technique. Each single layer was planarized with spin-on glass resist (Dow Corning XR-1541) matching the refractive index of the fused silica substrate. Atomic force microscopy confirmed that the resulting surfaces were ideally planar within a measurement accuracy of 1 nm. For the writing of the top layer, multiple alignment marks were used to ensure an alignment accuracy of better than 20 nm to the bottom layer [Fig. 1(c)].

The squared moduli $t_{ij} = |T_{ij}|^2$ of the T matrix of the MM were determined experimentally [Figs. 2(a) and 2(b)]. Clearly t_{xy} and t_{yx} interchange for opposite propagation directions proving that all elements in the linear base are different in general. Additional deviations between the forward and backward propagation are negligible and can be attributed to a nonideal alignment of the polarizers. For comparison, the complex transmission coefficients T_{ij} are computed by using the Fourier modal method [20] and shown in Figs. 2(c) and 2(d). The spectral dispersion of gold was properly accounted for [21] using the bulk permittivity. Hence, possible quantum effects resulting in a size dependent permittivity of the metal are neglected because of the relatively large dimensions of the gold particles. The experimental and numerically computed intensities are in almost perfect agreement. Minor deviations are attributed to an approximative modeling of the structure with geometrical parameters as deduced from electron microscopy images. As expected from the design of the meta-atoms, the spectra reveal a complex multiresonant behavior, which will be elucidated in another contribution.

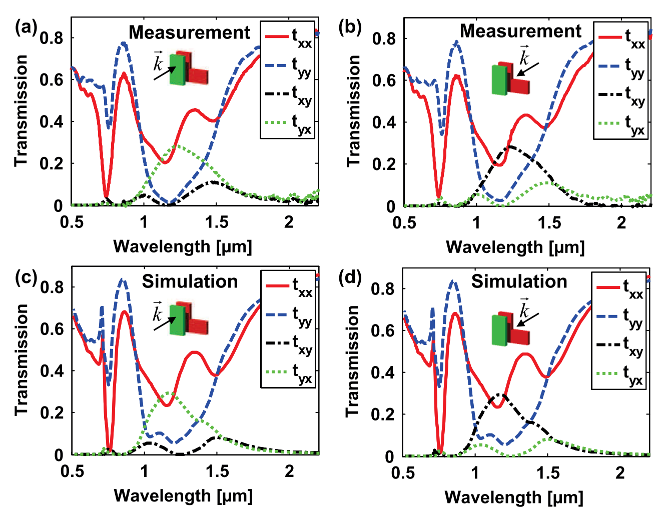

FIG. 2 (color online). Squared moduli of the four T-matrix components of the MM. (a),(b) Measurements and (c), (d) simulations. (a),(c) The k vector of the plane waves illuminating the MM was directed in (a), (c) positive and (b), (d) negative z direction. Note that t_{xy} and t_{yx} interchange when k is reversed.

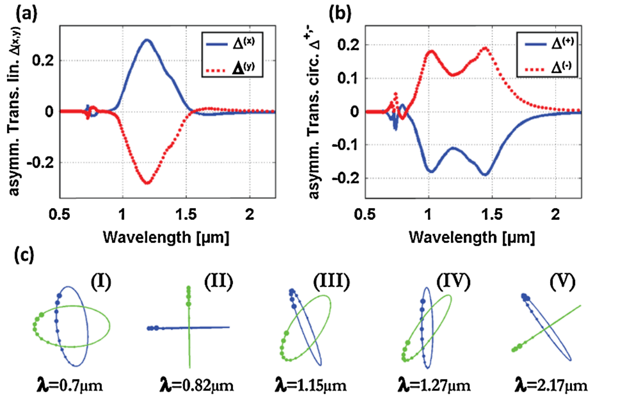

FIG. 3 (color online). Values for the asymmetric transmission Δ for linear (a) and circular (b) states as determined by the numerical data. The color of the lines indicates the particular input state. (c) Eigenstates of the polarization at different wavelengths. The rotation direction is indicated by increasing spot sizes.

The values for the asymmetric transmission Δ as determined from numerical data are calculated according to Eqs. (7) and (8) in the linear as well as in the circular base [Figs. 3(a) and 3(b)]. Clearly, they are different for different bases and achieve values up to 25% in the linear base. The asymmetry for a certain state is of course identical to the asymmetry for the complementary state with negative sign since the total transmission for unpolarized light from both sides has to be the same in a reciprocal medium. Figure 3 represents the main result of our work, which is an experimental proof of asymmetric transmission for linearly polarized light in a truly three-dimensional chiral meta-atom.

Any system made of complex meta-atoms is characterized by its eigenstates, i.e., those polarization states that do not change upon transmission. For these eigenstates, being in general not orthogonal, the T matrix is diagonal when transformed into that eigenbase. For planar quasichiral structures with additional rotational symmetries like gammadions, the eigenstates are circular counterrotating, whereas for anisotropic, 2D chiral structures the eigenstates are elliptical corotating with fixed phase relations [22]. For three-dimensional chiral systems without any additional symmetry like those considered here, the eigenstates can be arbitrarily complex. Both elliptical corotating and counterrotating as well as linear and circular states and combinations of them are expected. In Fig. 3 we exemplarily show the eigenstates of the present MM for different wavelengths. Most notably, elliptical corotating states (I and IV) as well as elliptical counterrotating states (III), almost linear states (II), and combined ones (V) may be observed. Even the angle between the polarization ellipses obeys no fixed relation. For structures lacking any symmetry, the eigenstates of the polarization have obviously no preferred orientation and rotation direction. Note that the principle form of eigenstates of the T matrix is invariant to any proper rotation with respect to the z axis. Hence, their complex behavior is a clear indication that it is not possible to symmetrize the T matrix by a proper rotation since there is no preferable orientation of the meta-atoms. We suggest that our findings can be exploited for the design of a purely MM-based optical isolator basing on the rich variety of possible combinations of complex T matrices in low-symmetry and three-dimensional chiral meta-atoms.

In conclusion, we reported the first experimental observation and theoretical analysis of asymmetric transmission for linearly polarized light in a 3D low-symmetry MM. Contrary to previous works focusing exclusively on circularly polarized light, the key for asymmetric transmission in any polarization base is the complete symmetry breaking of the meta-atom. The composition of three-dimensional chiral meta-atoms considerably enriches the variety of transmission functionalities and offers yet a new freedom of design for photonic MMs. In particular, optical isolation based on MMs seems to be in reach.

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