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Nanostructured Metal Film with Asymmetric Optical Transmission

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ABSTRACT

We demonstrate for the first time a nanostructured planar photonic metamaterial transmitting light differently in forward and backward directions.

In optics it is impossible to distinguish between experiments in which light passes through a medium in forward and backward directions. Isotropic, birefringent, and chiral media interact with light in a way that does not depend on the direction of propagation. This fundamental symmetry is only violated in magnetized media, where it matters whether light propagates parallel or antiparallel to the direction of magnetization. In this Communication, we demonstrate experimentally for the first time an artificial photonic planar metamaterial (a thin metal film regularly patterned on the nanoscale) for which propagation experiments in opposite directions yield different results in the absence of a magnetic field: the total intensity of initially circularly polarized light transmitted through the metamaterial film depends on the direction of propagation. To enable the asymmetric transmission effect reported here, the pattern of the film must possess a sense of twist (planar chirality), which is reversed when it is observed from the opposite side. The pattern must also be anisotropic and the metamaterial itself must be lossy.

The growing interest in artificial optical chiral media is currently driven by the desire to create metamaterials with strong optical activity (gyrotropy),[1−3] since they also show "left-handed" behavior and negative refraction of light,[4] as predicted by Pendry and others.[5,6] This paper deals with a different type of chirality belonging to essentially planar objects which are said to be planar chiral (or 2D-chiral) if they cannot be superimposed with their mirror images (enantiomers) obtained by reflection against a line in the plane of the structure. Such structures show enantiomerically sensitive polarization effects for light diffracted on the structure,[7,8] but optical enantiomerically sensitive and asymmetric transmission phenomena for light propagating through the medium without diffraction have not been reported for planar chiral media before.

The most general and universal tool for assessing the symmetry of propagation phenomena in optics is the Lorentz Reciprocity Lemma. In simple terms, it states that, if a signal polarized at P_1 and passing through a medium is received through analyzer P_2, the efficiency of transmission will be exactly the same as if the signal were polarized by P_2, sent in the opposite direction through the medium, and received through analyzer P_1, or in terms of the intensity transmission matrix $\overrightarrow{T}_{ij} = \overleftarrow{T}_{ji}$, where the arrow indicates the light wave direction and i and j denote polarization states. The Lorentz Reciprocity Lemma is not applicable to a magnetized medium placed in between polarizers, thus allowing for optical Faraday isolators, but in all other cases, it works perfectly well. For instance, in compliance with the lemma, the attenuation and retardation of a right-handed circularly polarized light wave (RCP, +) sent through a three-dimensional (3D) optically active medium such as sugar solution are identical in opposite directions, i.e. $\overrightarrow{T}_{++} = \overleftarrow{T}_{++}$. The same applies also for the case of left-handed circularly polarized waves (LCP, −).

The fundamental importance of the Lemma led optical scientists to believe that asymmetric propagation is not possible in optics unless magnetization is involved. Indeed, no materials were known that could challenge this assessment. However, the Lorentz Lemma does provide leeway for asymmetric transmission. The total transmission in opposite directions may be different if the medium between the polarizers partially converts the polarization state of the wave: the transmission intensity, for example, of a RCP wave in the forward direction, will then be determined by $\overrightarrow{T}_{++}$

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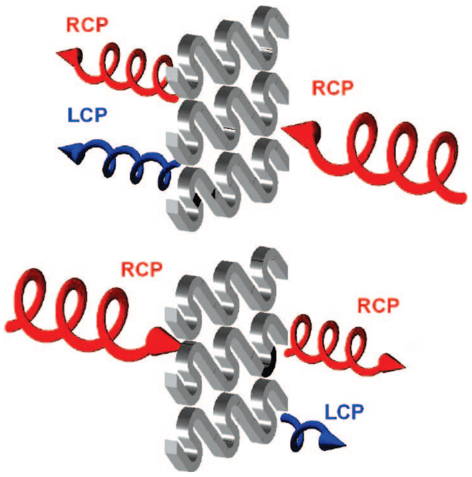

Figure 1. Illustration of transmission and polarization conversion of circularly polarized light by a metallic planar chiral metamaterial. Incident right-handed circularly polarized light (red spiral) is partially converted to the left-handed (blue spiral) polarization, as it propagates through the chiral grid. The ratio of conversion is different for opposing directions of propagation.

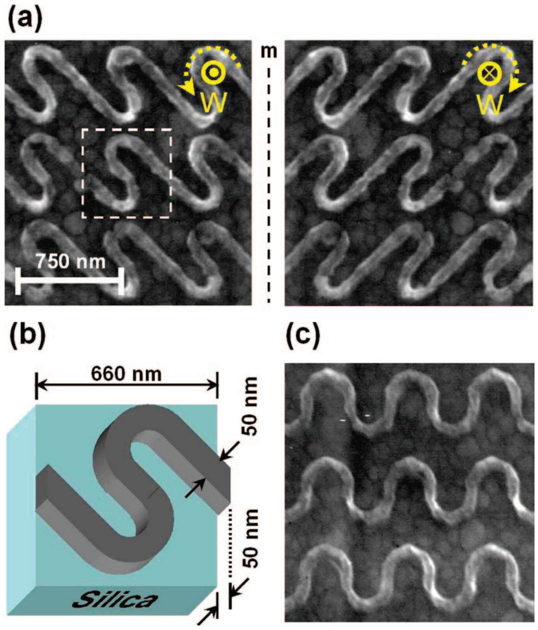

Figure 2. Photonic planar metamaterials. (a) SEM images show the two enantiomeric forms of the investigated planar metamaterial with asymmetric transmission, which are aluminum nanowire structures supported by a glass substrate. The sense of twist and direction of the twist vector **W** are shown in the upper-right corner of each image. The patterns are interconnected by reflection against mirror line “m”. (b) Schematic of the chiral fish-scale metamaterial’s unit cell indicated by the dashed box in panel a. (c) SEM image of a reference achiral metamaterial.

and $\overrightarrow{T}_{-+}$, while in the opposite direction it will depend on the values of $\overleftarrow{T}_{++}$ and $\overleftarrow{T}_{-+}$. While the lemma requires that $\overrightarrow{T}_{++} = \overleftarrow{T}_{++}$, it imposes no restriction on the relative values of $\overrightarrow{T}_{-+}$ and $\overleftarrow{T}_{-+}$. These in fact can be different, making total transmission asymmetry possible for opposite directions (see Figure 1).

Recently, asymmetric transmission through a planar chiral structure was demonstrated for circularly polarized microwave radiation.[9] In this type of metamaterial, the transmission field matrix for circular polarizations χ can be presented as a sum $\chi = \chi_0 + i \cdot \xi \text{ sign}(k \cdot W)g$. Here, χ_0 is a symmetric matrix that describes the anisotropy of the structure; W is its twist vector (see Figure 2); k is the wave vector of the incident wave; g is a unitary antisymmetric matrix, and ξ is the polarization conversion term. The antisymmetric part is proportional to the pseudoscalar combination $(k \cdot W)$ and changes sign upon reversal of the propagation direction. Consequently, it gives rise to the difference between $\overrightarrow{\chi}$ and $\overleftarrow{\chi}$ and is therefore responsible for total direction-dependent transmission $\overrightarrow{T}$ and $\overleftarrow{T}$.[10] Importantly the antisymmetric part is also proportional to the polarization conversion term ξ, which can only exist for anisotropic patterns of low symmetry and which vanishes for structures possessing a 4-fold symmetry axis. Moreover, only in dissipative systems can ξ not be eliminated by the choice of an appropriate coordinate system. In other words, transmission asymmetry is only possible in anisotropic, dissipative, planar chiral structures, and therefore it is fundamentally different from the truly symmetric effects of conventional optical activity observed recently in isotropic pseudoplanar nanostructures with three-dimensional chiral symmetry.[1−3] It should also be noted that the new effect presents a clear violation of the time reversal symmetry. Indeed, since time reversal sends light waves in the opposite direction without affecting their handedness, any optical phenomenon that is different for contra-propagating waves of the same handedness must violate the time reversal symmetry, which in the case of asymmetric transmission observed here is related to dissipation.

In this Communication, we report on the first experimental observation of the asymmetric transmission of light through a photonic planar metamaterial with a two-dimensional (2D) chiral pattern. The metamaterial’s pattern is a continuous anisotropic meander, which exists in two enantiomeric forms (with correspondingly left and right twist) related by reflection across a mirror line in the plane of the patterns (see Figure 2a). A “twist” vector W governed by the “corkscrew law” may be associated with each of the forms according to the handedness of their twist: as illustrated, for the left-twisted pattern, the vector points toward the reader, while for the right-twisted pattern W reverses its direction. Importantly, the direction of W is independent of the side of the pattern from which the definition is applied.

The planar chiral metamaterial samples were fabricated using e-beam lithography and comprised a double-periodic grid of aluminum nanowires with a cross section of 50×50 nm^2 supported by a 500 μm thick silica substrate (see Figures 2a and 2b). The unit cell of each grid was a square with the size of 660×660 nm^2, ensuring that in air the nanostructures did not diffract light of wavelength longer that 660 nm, that is, in the red and infrared part of the spectrum. The metamaterial structures had lateral dimension of $\sim 500 \times 500$ μm^2 and consisted of about 6×10^5 fish-scale unit cells. We also manufactured a reference, nonchiral planar metamaterial with similar specifications based on a “straight” fish-scale pattern (see Figure 2c).

The optical transmission properties of the fish-scale metamaterials were studied at normal incidence for both right and left circularly polarized light with the wavelengths in the 700−1700 nm range using a supercontinuum laser source and an optical spectral analyzer. The incident light was circularly polarized by a superachromatic quarter-wave plate, while detection of the transmitted light was polarization insensitive.

Figure 3 presents the relative difference in the total transmission of left and right circularly polarized light measured with respect to the average level of the total transmission through the planar structures, which reads for forward direction:

$$\overrightarrow{\Delta} = 2\frac{\overrightarrow{T_+} - \overrightarrow{T_-}}{\overrightarrow{T_+} + \overrightarrow{T_-}} = 2\frac{(T_{++} - T_{--}) - (\overrightarrow{T_{+-}} - \overrightarrow{T_{-+}})}{(T_{++} + T_{--}) + (\overrightarrow{T_{+-}} + \overrightarrow{T_{-+}})} \tag{1}$$

while in the opposite direction

$$\overleftarrow{\Delta} = 2\frac{\overleftarrow{T_+} - \overleftarrow{T_-}}{\overleftarrow{T_+} + \overleftarrow{T_-}} = 2\frac{(T_{++} - T_{--}) - (\overleftarrow{T_{+-}} - \overleftarrow{T_{-+}})}{(T_{++} + T_{--}) + (\overleftarrow{T_{+-}} + \overleftarrow{T_{-+}})} \tag{2}$$

Here the difference $(T_{++} - T_{--})$ is related to the conventional effect of 3D chirality, which does not depend on the direction of propagation (hence no arrows are introduced in the formulas), while the difference $(T_{+-} - T_{-+})$ represents the circular conversion dichroism associated with 2D chirality of the patterns, which is expected to change sign on reversal of the propagation direction.

Indeed, the experimental data clearly demonstrate the existence of the asymmetric transmission effect for the planar chiral nanostructures, which peaks at a wavelength of around 1.4 μm and shows the following three main characteristics (see Figures 3). First, reversing the direction of propagation reverses the asymmetry of total circular transmission thus confirming the presence of circular conversion dichroism, $(T_{+-} - T_{-+}) \neq 0$ and the absence of conventional optical activity, $(T_{++} - T_{--}) = 0$ (Figure 3a). Second, replacing the planar chiral metamaterial with its enantiomeric form also reverses the asymmetry of total circular transmission (Figure 3b). Although the magnitudes of conversion in both cases, as well as spectral location of the maxima, are slightly different, this shall be attributed to manufacturing difficulties in obtaining true mirror-like left- and right-tilted fish-scale patterns with the nanowires having exactly the same thickness and width on different parts of the wafer. Thus, whenever the handedness of the polarization state or the pattern's twist is changed, the sign of the transmission asymmetry is reversed. Third, the nonchiral structure shows no transmission asymmetry (Figure 3c) and no such effect would be seen for linear polarizations.

Employing the pseudospectral time-domain (PSTD)[11] method, we numerically modeled the effect. Using the complex dielectric constants of bulk aluminum, we found good qualitative agreement with our experiments (see Figure 3d). The modeled dispersions of the relative asymmetry of the total circular transmission Δ show slightly narrower and blue-shifted peaks, which is a typical discrepancy often seen in the modeling of metallic metamaterials. It is normally attributed to a departure of the dielectric properties of nanogranulated metals from those of bulk materials. The high-frequency oscillation on the dispersion curves are due to interference effects in the substrate. Numerical modeling also revealed that the phase dispersion corresponding to the calculated spectra is characteristically resonant and results in the excitation of localized plasmons, that is, collective oscillations of free electrons and optical fields coupled to the metallic nanostructure. Furthermore, because of 2D-chiral asymmetry of the metallic grid, such plasmonic excitation strongly depends on the handedness of the polarization state of light incident on the nanostructures, as well as on the direction of light propagation. We argue that the resonance feature observed in the spectral dependence of the effect corresponds to the excitation of enantiomerically sensitive plasmons, a new class of plasmonic excitations recently described in ref 10. Importantly, since asymmetric transmission is a dissipative effect, the lossy nature of the plasmonic interactions is absolutely essential for the observation of the asymmetry.

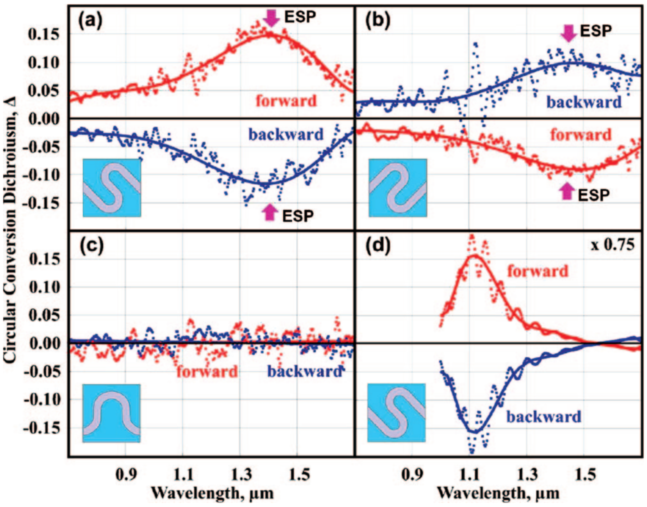

Figure 3. Asymmetric optical transmission through planar chiral metamaterials. The graphs show the relative asymmetry of total circular transmission Δ for forward and backward propagation through left-tilted (a) and right-tilted (b) forms of the metamaterial. Arrows indicate the enantiomerically sensitive plasmon resonances (ESP). For a test (nonchiral) structure transmission asymmetry disappears (c). Panel (d) shows the theoretically calculated dispersion of Δ for propagation through a left-tilted metamaterial (the dispersion for its right-tilted form is identical, while the sign of the effect is reversed).

A comparison of Figures 3a and 3b allows us to conclude that the circular transmission asymmetry is controlled only by the perceived sense of rotation (i.e., 2D chirality) associated with the metallic pattern alone and not the substrate. Furthermore, the asymmetry of the effect does not depend on whether the pattern is placed in front of or behind the silica substrate, or whether the substrate is present at all, which is in complete agreement with the prediction of ref 10. In other words, it only matters if the wave vector of the incident light wave is parallel or antiparallel to the vector of the pattern's twist *W* (see Figure 2a). This is in sharp contrast to conventional optical activity in 3D-chiral media, where the observable effect does not depend on the direction of propagation.

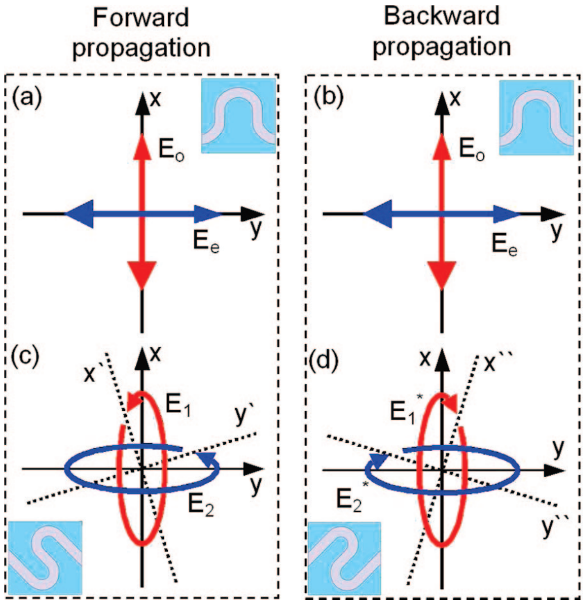

Figure 4. Polarization eigenstates in planar anisotropic lossy metamaterials. Eigenstates E_o and E_e are linear for a nonchiral metamaterial pattern and are identical in the forward (a) and backward (b) directions of propagation. In the case of chiral metamaterial patterns, eigenstates E_1 and E_2 are a pair of co-rotating ellipses with a handedness that is reversed for forward (c) and backward (d) propagation.

Another important property of the asymmetric transmission, which makes it fundamentally different from the Faraday effect and optical activity is that it has *co-rotating* elliptical polarization eigenstates, while both 3D-chiral and magnetized media support *contra-rotating* circular polarization eigenstates. This, and the origin of the asymmetric propagation, can be understood by comparing the transmission matrices for lossy anisotropic chiral and nonchiral metamaterial structures (see Figure 4). In a nonchiral anisotropic structure, the transmission matrix can be diagonalized by the choice of coordinate system XY, as illustrated in Figure 4a. Here, the eigenstates are two orthogonal linear polarizations E_o and E_e which remain the same for the opposite propagation direction (Figure 4b). For the planar chiral lossy anisotropic structure, the real and imaginary parts of the transmission matrix can only be diagonalized in different coordinate systems XY and $X'Y'$ (see Figure 4c). Here, the polarization eigenstates E_1 and E_2 are elliptical with handedness determined by the sign of rotation from XY to $X'Y'$ which is linked to the perceived sense of twist in the pattern (compare the unit cells of the pattern as perceived by an observer looking along the direction of light propagation). For the opposing propagation direction, the diagonalizing coordinate systems XY and $X''Y''$, are also linked to the perceived sense of twist in the pattern, and connected by a rotation of the opposite sign: the polarization eigenstates E_1^* and E_2^* are again elliptical but of the opposite handedness (see Figure 4d).

In conclusion, we provide the first experimental observation of asymmetric transmission of circularly polarized light through metallic nanostructured film, and link the effect to the excitation of enantiomerically sensitive plasmon.

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References

(1) Deckerand, M.; Klein, M. W.; Wegener, M.; Linden, S. *Opt. Lett.* **2007**, *32*, 856–858.
(2) Plum, E.; Fedotov, V. A.; Schwanecke, A. S.; Zheludev, N. I.; Chen, Y. *Appl. Phys. Lett.* **2007**, *90*, 223113.
(3) Konishi, K.; Sugimoto, T.; Bai, B.; Svirko, Y.; Kuwata-Gonokami, M. *Opt. Express* **2007**, *15*, 9575–9583.
(4) Rogacheva, A. V.; Fedotov, V. A.; Schwanecke, A. S.; Zheludev, N. I. *Phys. Rev. Lett.* **2006**, *97*, 177401. Plum, E.; Dong, J.; Zhou, J.; Fedotov, V. A.; Koschny, T.; Soukoulis, C. M.; Zheludev, N. I. *arXiv.org, 0806.0823v1* (2008).
(5) Pendry, J. B. *Science* **2004**, *306*, 1353–1355.
(6) Tretyakov, S.; Sihvola, A.; Jylha, L. *Photonics Nanostruct. Fundam. Appl.* **2005**, *3*, 107.
(7) Papakostas, A.; Potts, A.; Bagnall, D. M.; Prosvirnin, S. I.; Coles, H. J.; Zheludev, N. I. *Phys. Rev. Lett.* **2003**, *90*, 107404.
(8) Prosvirnin, S. L.; Zheludev, N. I. *Phys. Rev.* **2005**, *E71*, 037603.
(9) Fedotov, V. A.; Mladyonov, P. L.; Prosvirnin, S. L.; Rogacheva, A. V.; Chen, Y.; Zheludev, N. I. *Phys. Rev. Lett.* **2006**, *97*, 167401.
(10) Fedotov, V. A.; Khardikov, P. L.; Prosvirnin, S. L.; Schwanecke, A. S.; Zheludev, N. I. *Nano Lett.* **2007**, *7*, 1996–1999.
(11) Gao, X; Mirotznik, M. S.; Shi, S.; Prather, D. W. *J. Opt. Soc. Am.* **2004**, *A 21*, 777.

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