

Asymmetric Transmission of Light and Enantiomerically Sensitive Plasmon Resonance in Planar Chiral Nanostructures

V. A. Fedotov,* A. S. Schwanecke, and N. I. Zheludev†

Optoelectronics Research Centre, University of Southampton, Highfield, Southampton SO17 1BJ, United Kingdom

V. V. Khardikov and S. L. Prosvirnin

Institute of Radio Astronomy, National Academy of Sciences of Ukraine, Kharkov 61002, Ukraine

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ABSTRACT

We show that in the visible to near-IR part of the spectrum, normal incidence transmission of circularly polarized light through a nanostructured anisotropic planar chiral metamaterial is asymmetric in the opposite directions. The new effect is fundamentally different from the conventional gyrotropy of bulk chiral media and the Faraday effect. It has a resonance nature associated with a new type of excitation in the metal nanostructure: the enantiomerically sensitive plasmon.

The sense of twist of a planar structure such as the Archimedes spiral drawn on a sheet of transparent substrate is reversed when the sheet is observed from the opposite side. This brings up an intriguing question on whether optical transmission and scattering effects depend on the direction of propagation through planar structures containing twisted patterns. Indeed, it has recently been shown theoretically that the propagation of circularly polarized light through a hole of twisted shape with 4-fold rotational symmetry is sensitive to the handedness of the incident wave: when the hole size is close to the wavelength of light, the intensity and polarization state distribution at the hole and beyond, in the diffraction zone, are dramatically different depending on the mutual handedness of the hole's twist and the incident light's polarization state. These results show broken enantiomeric symmetry for excitation with circularly polarized light and imply that light scattering on a small twisted hole is different in opposite directions. However, if the incident wave falls on the twisted, 4-fold symmetric hole at normal incidence, the total intensity of light leaking through the hole does not appear to depend on the direction of propagation.^{1–3} Nevertheless, when electromagnetic radiation *diffracts* on a regular array of chiral elements into a *nonzeroth* order, i.e., when losses associated with diffraction into the other multiple

beams are present and the high rotational symmetry of the entire process is broken (because of the change in the direction of the diffracted beams), the polarization eigenstates of the diffraction process *do* depend on the direction of propagation.⁴ Moreover, it has recently been demonstrated experimentally in the microwave part of the spectrum that, even *in the absence of diffraction*, the intensity of electromagnetic waves normally transmitted through a regular array of subwavelength planar chiral elements depends on the direction of wave propagation, providing the structure is simultaneously *anisotropic* and *lossy*.⁵

In this Letter, we show that asymmetric transmission through a planar metal nanostructure consisting of twisted elements can be observed in the optical part of the spectrum. The asymmetry has a resonant nature linked to the enantiomerically sensitive excitation of plasmons in the metal nanostructure. A planar structure is said to be chiral (twisted) if it cannot be brought into congruence with its mirror image (reflected across a line in the plane of the structure) unless it is lifted off the plane. The asymmetric effect in some ways resembles the famous nonreciprocity of the Faraday effect in magnetized media but requires no magnetic field for its observation and is fully compliant with the Lorentz reciprocity principle. Both in the Faraday effect and in that produced by planar chirality, the transmission and retardation of a circularly polarized wave is different in opposite directions.

* Corresponding author. E-mail: vaf@orc.soton.ac.uk.

† URL: www.nanophotonics.org.uk/niz.

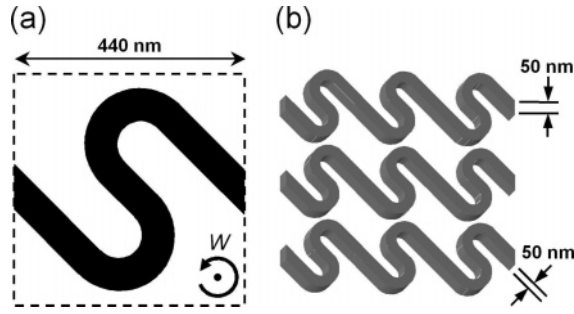


Figure 1. Schematics of the modeled nanostructured chiral fish-scale metamaterial. (a) Unit cell of the chiral fish-scale pattern. The pattern has an overall anticlockwise twist (left tilt), which corresponds to the twist vector \mathbf{W} pointing toward the reader. (b) Free-standing fish-scale aluminum grid.

In both cases, the polarization eigenstates, i.e., polarization states conserved on propagation, are elliptical (circular). There are also essential differences between the two phenomena. The asymmetry of the Faraday effect applies to the transmission and retardation of the *incident* circularly polarized wave itself, whereas the planar chirality effect leads to partial conversion of the incident wave into one of opposite handedness, and it is the efficiency of this *conversion* that is asymmetric for the opposite directions of propagation. The effect that we describe here is also radically different from conventional gyrotropy in 3D-chiral media (such as sugar solution or quartz), which is completely symmetric for waves propagating in opposite directions. It is linked to the excitation of enantiomerically sensitive plasmons in the metal nanostructure.

We investigated the new propagation effect in nanostructured planar chiral metamaterial in the visible to near-IR part of the spectrum. The metamaterial is a continuous chiral “fish-scale” pattern, periodic in both directions (see Figure 1a). Such a pattern can also exist in an enantiomeric form obtained by reflection across a mirror line in the plane of the pattern. A “twist” vector \mathbf{W} governed by the corkscrew law may be associated with each of the forms according to the handedness of their twist (tilt) and is independent of the side of the pattern to which the definition is applied (see Figure 1a). The structure of the considered metamaterial comprises a free-standing grid of aluminum wires with a square cross-section of 50 nm \times 50 nm (see Figure 1b). The grid has a square unit cell of 440 nm \times 440 nm, which ensures that the metamaterial does not diffract at wavelengths longer than 440 nm.

Using the pseudospectral time domain (PSTD) numerical simulation method,⁶ with periodic boundary conditions applied to the unit cell, we studied the transmission properties of the planar chiral metamaterial in the 500–1700 nm wavelength range for the case of normal incidence of circularly polarized light. For the purpose of our calculations, we used widely accepted values of the dielectric constant of aluminum.⁷

The results of our calculations can be presented in terms of a circular transmission 2×2 matrix χ , whose indexes “+” and “−” denote right (RCP) and left (LCP) circular polarizations, respectively. For a wave incident on the

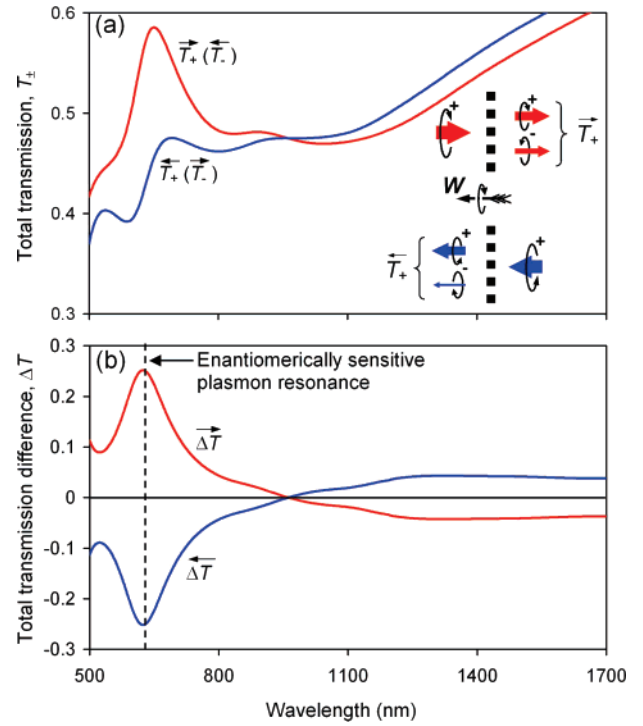


Figure 2. Asymmetric transmission effect in the aluminum chiral fish-scale grid. Red and blue curves correspond respectively to forward and backward directions of propagation. Part (a) shows total transmission of circularly polarized light. The insets illustrate polarization conversion in the metamaterial for RCP light propagating in opposite directions. Part (b) shows the relative enantiomeric difference in the total transmission.

structure’s front side, against its twist vector \mathbf{W} (as defined by Figure 1a), matrix χ will be denoted as $\bar{\chi}$, while $\bar{\chi}$ corresponds to the wave entering the structure from the opposite side. Our calculations showed that, within the computational accuracy, all the diagonal elements of both matrices are the same, i.e., $\bar{\chi}_{\pm\pm} = \bar{\chi}_{\mp\mp}$. This means that the structure does not manifest polarization effects of the same symmetry as conventional optical activity or the optical Faraday effect of bulk media. However, it displays the intriguing effect of *asymmetric transmission*.

The planar chiral metamaterial is more transparent to a circularly polarized light from one side than from the other. The data presented in Figure 2a illustrates such directional asymmetry in terms of total transmission. For instance, for incident RCP light, the total transmitted intensity in the forward direction is given by $\bar{T}_+ = |\bar{\chi}_{++}|^2 + |\bar{\chi}_{-+}|^2$, while in the opposite direction $\bar{T}_+ = |\bar{\chi}_{++}|^2 + |\bar{\chi}_{-+}|^2$ and $\bar{T}_+ \neq \bar{T}_+$. The difference in the total transmission comes from the fact that the conversion of a circular polarized wave into one of opposite handedness is asymmetric for opposite directions of propagations, i.e., $|\bar{\chi}_{-+}|^2 \neq |\bar{\chi}_{-+}|^2$, while $|\bar{\chi}_{++}|^2 = |\bar{\chi}_{++}|^2$. The relative enantiomeric difference in the total transmission $\bar{\Delta T} = (\bar{T}_+ - \bar{T}_+)/\bar{T}_+$ appears to be resonant, reaching maximum value of 25% at about 630 nm and changes sign upon reversal of the propagation direction (see Figure 2b). The asymmetry of total transmission is

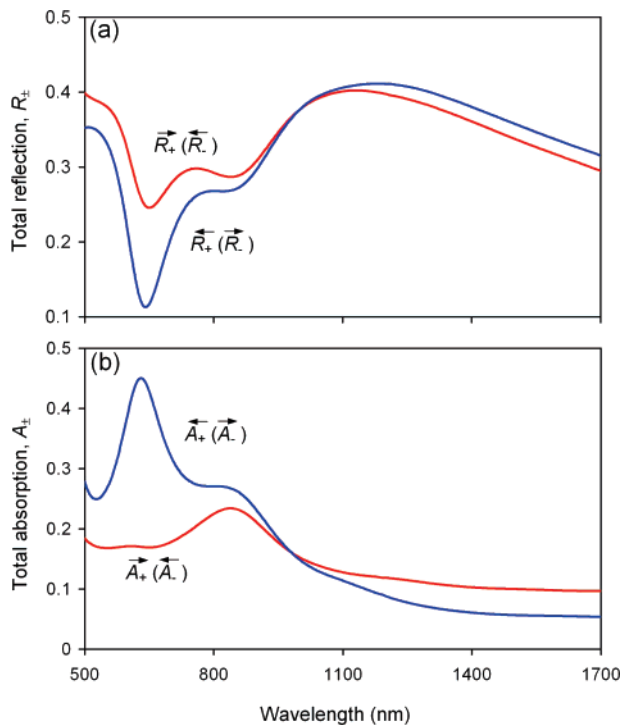


Figure 3. Asymmetry of total reflection (a) and total absorption (b) in the aluminum chiral fish-scale grid. Red and blue curves correspond respectively to forward and backward directions of incidence/propagation.

accompanied by similar asymmetries in total reflection and absorption, as shown in Figure 3.

The asymmetric transmission phenomenon does not mount any challenge to the validity of the reciprocity lemma, which requires $\bar{\chi}_{ij} = \bar{\chi}_{ji}$, but does not require the equality of $\bar{\chi}_{ij}$ and $\bar{\chi}_{ji}$, the difference between which underpins the asymmetric transmission. In comparison, for the Faraday magneto-optical effect $\bar{\chi}_{++} \neq \bar{\chi}_{++}$ and reciprocity does not hold, nor indeed is it supposed to in the presence of magnetic field.

The origin of the asymmetric interaction may be traced to the structure of the transmission matrix χ , which can be presented as a sum $\chi = \chi_0 + i \xi \text{sign}(\mathbf{k} \cdot \mathbf{W}) \mathbf{g}$.⁵ Here χ_0 is a symmetric matrix that describes the anisotropy of the structure, \mathbf{k} is the wave vector of the incident wave, and \mathbf{g} is a unitary antisymmetric matrix. The antisymmetric part is proportional to the pseudoscalar combination $(\mathbf{k} \cdot \mathbf{W})$ and changes sign upon reversal of the propagation direction. Consequently, it gives rise to the difference between $\bar{\chi}$ and $\bar{\chi}$ and is therefore responsible for the direction-dependent transmission. The antisymmetric part is also proportional to the conversion term ξ , which can only exist for anisotropic patterns of low symmetry and 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. In the present case, planar chirality and anisotropy are provided by the topography of the structure, while losses are associated with plasmonic dissipation.

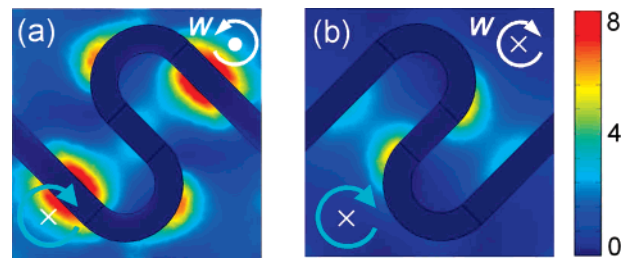


Figure 4. Enantiomerically sensitive plasmonic excitations in the aluminum chiral nanowire structure. The maps show the distribution of the intensity of the electric field (in arbitrary units) in the plane of the structure at the resonance ($\lambda = 630$ nm) calculated for left circularly polarized light incident on the wire grid from the opposite sides. Note that the field patterns are not mirror images.

Our calculations show that RCP light not only transmits but also converts through the planar chiral structure exactly the same way as LCP in the opposite direction, i.e., $\bar{\chi}_{ij} = \bar{\chi}_{ji}$. In fact, this symmetry is prescribed by fundamental space reversal symmetry, which demands a simultaneous reversal of the direction of light propagation and its handedness while leaving chirality of the planar sample unchanged. Indeed, in contrast with three-dimensional chiral objects switching enantiomeric forms on parity operation $x \rightarrow -x$, $y \rightarrow -y$, $z \rightarrow -z$, two-dimensional chiral objects are undisturbed by parity as $z \rightarrow -z$ does not affect planar object, while consecutive application of $x \rightarrow -x$ and $y \rightarrow -y$ switches enantiomeric form twice, restoring the original state. However, the fundamental symmetry of time reversal only allows the asymmetric propagation effect in dissipative media, as otherwise it prescribes the same transmission and conversion in opposite directions: reversing of time does not act on the sample, but reverses the direction of light propagation without altering handedness of the light wave.

In a continuous-wire structure at the resonance condition, electromagnetic radiation excites a standing wave of current oscillations.⁸ In the optical regime, this coupled oscillation of the electron density and electromagnetic field is a localized plasmon. It is similar in nature to the plasmonic excitation of a metal nanoparticle, with the only difference being that plasmonic excitation of an anisotropic chiral wire is enantiomerically sensitive: the excitation level and energy loss are higher when the handedness of the wire's twist and the polarization state of the incident light do not coincide (see Figure 2). Here, the lossy nature of the plasmonic interaction is essential for the asymmetric transmission effect. The striking difference between plasmonic excitations created by waves propagating in opposite directions may be seen in Figure 4, which shows electric field intensity maps in the plane of the structure. The maps reveal the strength and localization of the plasmonic excitations, which are more efficient, and therefore correspond to higher losses when the handedness of the wave is opposite to that of the structure (see Figure 4a).

Finally, we have also observed the asymmetric propagation phenomenon in the same planar chiral structure supported on a thick silica substrate. The results obtained closely resemble those presented in Figures 2 and 4. In the supported

structure, the transmission asymmetry is of the same order, while the resonance shifts to 746 nm. As in the free-standing sample, in the supported nanostructure we saw no evidence of polarization effects with the symmetries of conventional bulk gyrotropy of the Faraday rotation, i.e., even in the presence of a substrate $\overrightarrow{\chi_{\pm\pm}} = \overrightarrow{\chi_{\mp\mp}}$. Therefore, the asymmetric transmission effect is controlled *only* by the perceived sense of rotation associated with the chiral grid itself and does not depend on the presence of a substrate. 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** (as shown in Figure 1a). This is in sharp contrast with conventional gyrotropy in three-dimensional chiral media consisting of two layers of coaxial gammadions^{9–11} where in refs 9,10 the 3D chirality is introduced through mutual twist of gammadions, while in ref 11 it is achieved due to the different size of the gammadions in the top and bottom layers. As appropriate for volume chirality, the transmission phenomena observed in these 3D-chiral media are conventional optical activity and circular dichroism, which are identical for opposite directions of light propagation. They are not accompanied by any asymmetric intensity effect and therefore are completely different from the planar chirality phenomenon reported here.

In conclusion, we report the first analysis of asymmetric transmission of light through planar chiral nanostructured photonic metamaterials in the optical part of the spectrum and associate it with a new type of enantiomerically sensitive plasmon excitation. Numerous optoelectronic applications of

such metamaterials can be envisaged, in particular in polarization-sensitive devices. For instance, when placed into a ring resonator, they will enforce the generation of left and right elliptically polarized waves in opposite directions of circulation.

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