Layered chiral metallic meta-materials

Nikolay I. Zheludev[*a], Aris Papakostas[ab], Adrian Potts[ab], Harry J. Coles[a] and Darren M. Bagnall[ab]

[a]Department of Physics and Astronomy and [b]Department of Electronics and Computer Science.
University of Southampton, SO17 1BJ, UK.

ABSTRACT

Using electron beam lithographic techniques we have manufactured left and right-handed forms of an artificial medium consisting of high densities of microscopic planar chiral metallic objects distributed regularly in a plane. In this artificial medium we have for the first time observed optical manifestations of planar chirality in the form of handedness-sensitive rotation of the polarization state and elliptization of visible light diffracted from the structure. Applications of such media in functional materials are discussed.

Keywords: planar chirality, optical activity, metallic nanostructures.

1. INTRODUCTION

Optical structures artificially engineered on a mezoscopic level such as photonic bandgap crystals[1], periodically altered dielectric materials[2, 3], holey fibers[4], microsculptured films[5] and composite media are attracting tremendous attention because of their potential importance in optoelectronic technologies. They are collectively known as "metamaterials". Recently a new group of layered planar and quasi-planar metamaterials has emerged which promise unique electromagnetic properties. Metallic bilayered chiral structures with inductive coupling are predicted to show huge optical polarization rotatory power, and possibly a photonic bandgap effect in a single layer of "fully metallic molecules"[6]. In addition, planar chiral structures, such as metallic figures of certain symmetries (for example gammadions), are predicted to show distinct polarization properties and photonic band-gap anomalies when placed randomly on a plane[7]. Layered metallic microstructures could play a special role in future technology, as they can be manufactured on a sub-optical wavelength scale using, well-established microelectronics technologies, and other non-traditional techniques. To date, however, research on layered chiral metallic microstructures has been confined to theoretical analysis with only a few experiments having been performed in the microwave range of frequencies. In particular, designs based on metallic non-chiral planar split ring resonators have been shown to exhibit frequency regions with simultaneously negative magnetic and electric permittivities as well as other intriguing and useful electromagnetic characteristics[8, 9].

In this paper we draw attention to planar chiral structures. The ability of left-right asymmetrical (chiral) three-dimensional helical molecules to rotate the polarization state of light is a cornerstone optical effect known as optical activity[10, 11]. Formally, the concept of chirality also exists in two dimensions[7, 12]. A planar object is said to be chiral if it cannot be brought into congruence with its mirror image unless it is lifted from the plane. However, the questions remains as to whether such a chiral medium consisting of flat chiral "elements" distributed in a plane could affect the polarization state of light in a manner similar to three-dimensional chiral media and what other optical characteristics could one expect for two-dimensional (planar) chiral structures? There have been a few theoretical publications on the optical manifestations of planar chirality. Hecht and Barron[13], predicted incoherent circular differential Rayleigh and Raman light scattering from an ensemble of planar chiral molecules and showed that genuine strong chiral scattering phenomena could be generated through pure electric dipole interactions (in comparison with the much weaker processes involving magneto-dipole interaction in three-dimensional chirality) and Arnaut *et al*, have calculated the scattered

*n.i.zheludev@soton.ac.uk; phone +44 (0) 2380 593566; fax +44 (0) 2380593910; http://www.nonlinear-optics.phys.soton.ac.uk; Department of Physics and Astronomy, University of Southampton. Southampton. SO17 1BJ, UK.

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fields from a planar metallic wire gammadion and found rotation of the polarization azimuth of the scattered field[7]. There have as yet been no reports of experimental observations of any optical manifestation of planar chirality and its application in functional materials has not yet been analyzed.

2. EXPERIMENTAL OBSERVATIONS AND DISCUSSION

In this paper we report the first experimental observations of optical manifestations of planar chirality in artificial media consisting of microscopic chiral metallic objects, namely gammadions. The gammadions we re arranged in regular two-dimensional square gratings to produce a structure of 442 wallpaper group symmetry (Fig.1).

Clockwise and anti-clockwise gammadions provide two enantiomeric forms of the structure. The 442 symmetry group was chosen because of the absence of any optical birefringence in the structure at normal incidence. In all of the arrays we studied, the gammadions had sides of length ξ =1.4μm, but different internal angles α (see Fig. 2). Here the positive values of α correspond to clockwise gammadions, while the negative values correspond to anticlockwise. In all cases the pitch of the gratings was 4.0μm in both directions so that the density of gammadions was 6.2×10^{6} cm^{-2}. Each grating provided an area of approximately 1.0×1.0 mm^2 available for optical measurements.

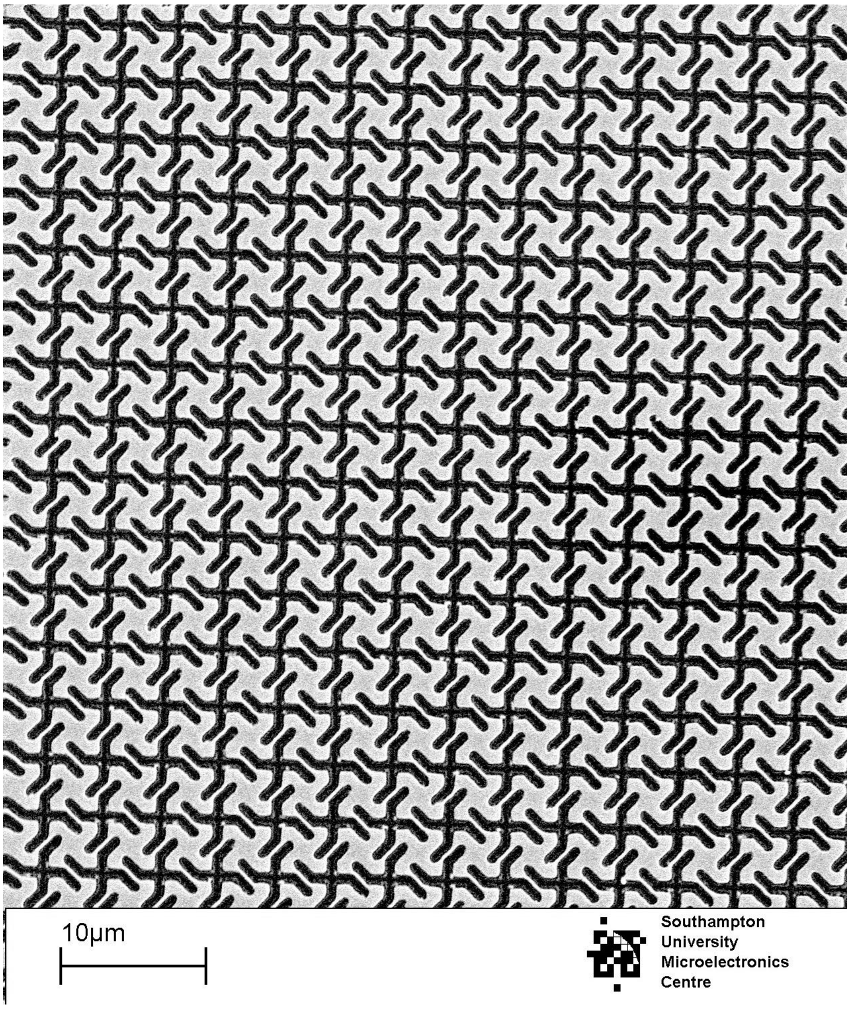

Fig. 1: An SEM image of a planar chiral structure of the 442 symmetry group manufactured using a combination of direct-write electron beam lithography and ion-beam milling to etch into a titanium/gold/titanium layer (20nm/100nm/20nm thick) deposited on a silicon.

Light scattered from the chiral structures shows well-defined rectangular diffraction patterns. The polarization states of diffracted waves are noticeably different from those that would be expected for diffraction from a non-chiral grating. Experiments have been performed at a wavelength of 632nm with polarization parameters of the diffracted wave measured using the "rotating wave plate" polarimetric technique. The structures were oriented to the incident beam in such a way that the "plane of incidence" (plane defined by the incident wave and the normal to the sample) was perpendicular to the gammadion array (see Fig. 2). In this installation diffracted waves propagate in both the plane of incidence, and at various angles to it. In this paper we discuss the polarization states of waves diffracted in the plane of incidence. If the diffracted wave remains in the plane of incidence and the incident wave is polarized either in the plane of incidence or perpendicular to the plane, no polarization change would be expected for a non-chiral grating. However, we have found that if the incident wave is linearly polarized, say, in the plane of incidence, the polarization states of the diffracted waves are, in general, elliptical, with the polarization azimuth rotated by some angle to the initial polarization state. In addition, for a given angle of incidence β, the polarization states of various diffraction orders are found to be different. We concentrated on characterizing the polarization state of the first-order diffracted wave and measured the difference $\Delta\Theta$ between the polarization azimuth of the incident wave φ and that of the diffracted wave. We also measured the ellipticity angle η of the diffracted wave (where η =0 corresponds to a linearly polarized wave, and $\eta = \pm 45^O$ correspond to right and left circular polarizations, respectively).

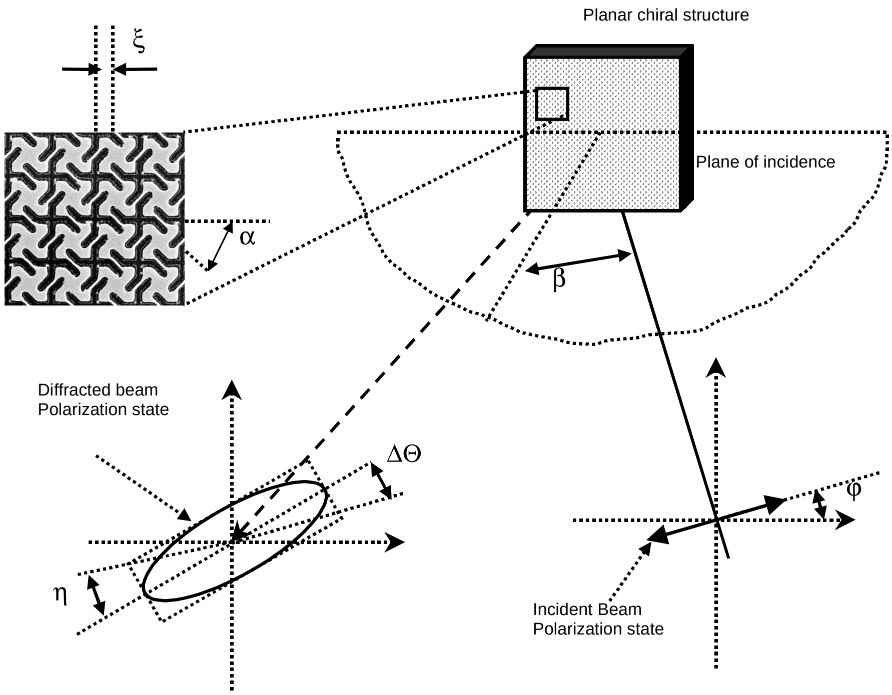

Fig. 2: A schematic illustration of the experiment to study optical manifestations of two-dimensional chirality in artificial planar structures. The inset shows a scanning electron micrograph of a typical chiral structure (TiAu/Ti@Si).

We first measured the polarization change of the diffracted beam for three pairs of enantiomeric chiral structures with different internal angle α, for an incident wave that was linearly polarized in the plane of incidence. These measurements were performed for different incident angles, β. Fig. 3, shows the polarization azimuth rotation and the ellipticity angles for two enantiomeric structures corresponding to α = +45^0 and α = -45^0. The polarization azimuth rotation $\Delta\Theta$ and the degree of ellipticity η, strongly depend on the angle of incidence, β. However, whatever the value of β, $\Delta\Theta$ and η, the waves diffracted from enantiomeric planar chiral structures were found to be of the same amplitude, but of strictly opposite signs. A comparison of results for the polarization azimuth rotation (Fig.3) for enantiomeric pairs with different angle α illustrates that the strongest polarization effect is seen in the "open" gammadion structures with $\alpha = \pm 45^O$ and steadily decreases with increasing α. In all cases an opposite sign for rotation and ellipticity is observed for enantioneric pairs. We also compared the polarization azimuth rotation for positive and negative incident angles, $\pm\beta$. Although some minor differences in the polarization response for incident angles of opposite signs were observed, the polarization rotation and the ellipticity are found to be largely insensitive to the sign of the incident angle. This is not an obvious result as planar chiral structures installed with positive and negative angles of incidence are not equivalent.

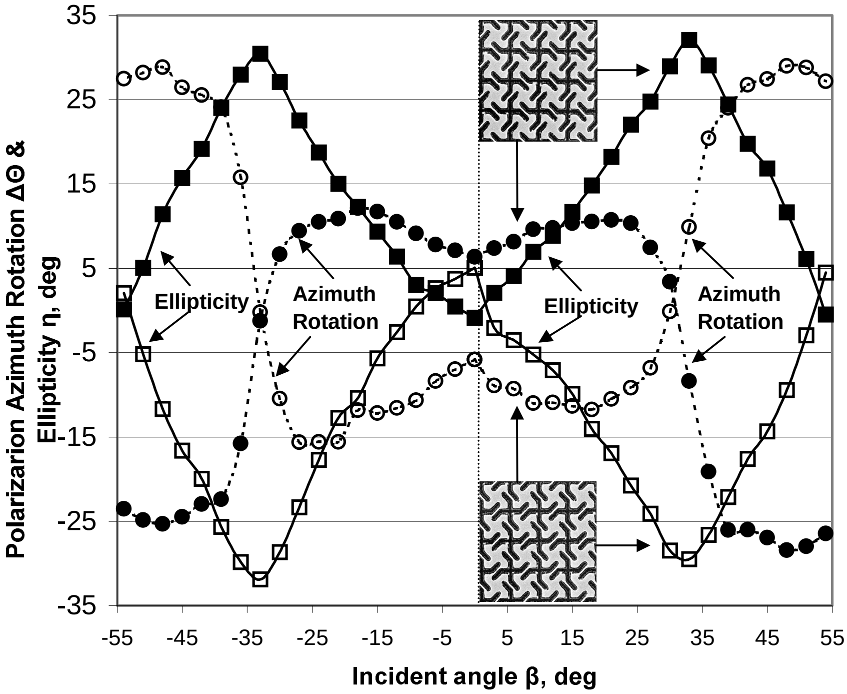

Fig. 3: Optical manifestations of planar chirality in the diffraction regime. Polarization azimuth rotation (●, ○) and change in ellipticity (■, □) of light wave diffracted from left (○, □) and right (●, ■) handed gammadion arrays plotted as functions of the incident angle. The incident angle β is measured between the direction of the incident beam and normal to the sample, as presented on fig.2. The two enantiomeric forms correspond to $\alpha = -45^0$ (left-handed) and $\alpha = +45^0$ (right-handed). Note the change of sign for the polarization effects due to left- and right-handed structures.

Our results unambiguously show that polarization changes in light diffracted from a chiral planar structure are a true effect of planar chirality. Indeed, to be a true optical manifestation of two-dimension chirality, the following test must be satisfied: the interaction of light with the two enantiomeric forms of the structure must yield different polarization changes, and these changes should not be observed if the structure lacks chirality. More generally, the magnitude of the polarization change and its sign must have a correlation with the geometrical chirality of the structure, in particular with its handedness. The results presented in Fig. 3 and 4 prove that enantiomeric structures can, in fact, give opposite polarization changes where non-chiral structures would show no polarization change at all. Indeed, the wave polarized in the plane of incidence represents the polarization eigenstate of the non-chiral structure and will be unaffected by diffraction in the plane of incidence. Additionally, Fig. 4 illustrates that the polarization effect is also sensitive to the geometrical chirality of the structure and its enantiomeric form, as predicted theoretically for fields scattered by a single conductive gammadion[14]. Independence of the polarization effect on the sign of the incident angle would appear to indicate that decreasing the grating pitch until diffraction disappears (i.e. all the diffracted beams merge into the zero order direction) would yield a true chiral net polarization effect for light propagating through or reflected from the structure.

Optical manifestation of planar chirality may be understood if one considers the polarizability of a gammadion. If a conductive gammadion interacts with the electric field of the incident wave, polarized for instance, along one of the gammadion legs, the positive and negative carriers are shifted into opposite legs. As the conductive strip is bent in a chiral fashion, charges will be shifted not only along the direction of the field, but also into the bent ends of the legs, thereby inducing a polarization perpendicular to the driving electric field with sign that depends on the direction of the bend, and therefore on the chirality of the gammadion. When fields are re-emitted by the chiral currents, a rotation of the polarization state of the scattered field is observed. We suggest that two mechanisms may be relevant to the dependence of the polarization effects on β. The first mechanism is due to the delay with which the incident wave reaches the opposing legs of a gammadion, thus creating a phase lag in the re-emission of secondary waves from opposing legs. The second mechanism of the dependence of the polarization effect on the angle of incidence may be relevant to the excitation of guided waves in the periodic structure of gammadions, which is known to yield sharp dependencies in the efficiency of the diffraction process on the incident angle[15]. Finally, the difference between the dielectric properties of media that surround the chiral structure (silicon and air) should also be very important in forming the optical response, in particular because the presence of the interface breaks the inversion symmetry of the structure as a whole[16].

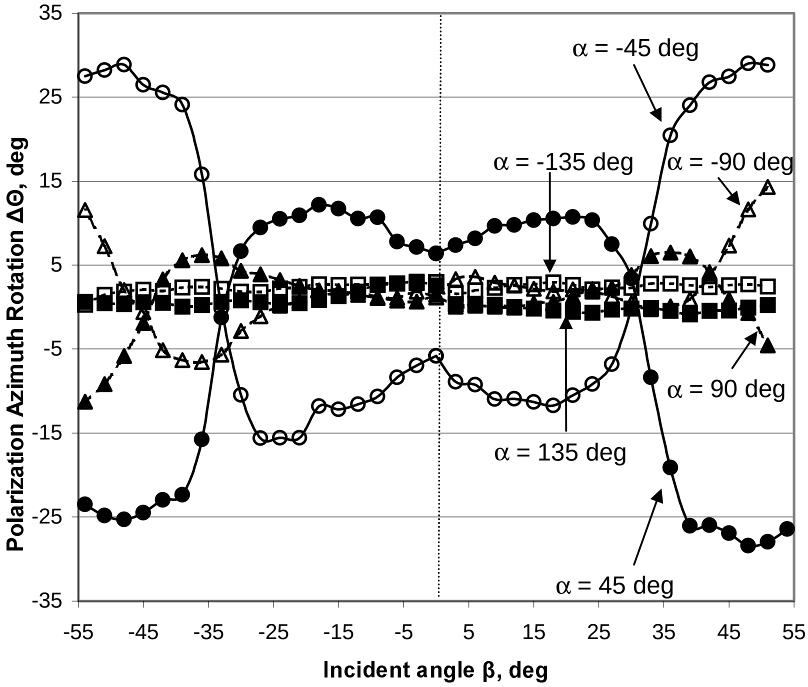

Fig. 4: Polarization azimuth rotation of light wave diffracted from left-handed (○,△,□) and right-handed (●,▲,■) gammadion arrays of different chirality plotted as functions of the incidence angle. The different plots correspond to different internal angle α = -45^O (○), α = -90^O (△), α = -135^O (□), α = +45^O (●), α = +90^O (▲), α = +135^O (■). Note that the polarization effects increase with decreasing internal angle.

3. CONCLUSION

To summarize we have observed for the first time unambiguous evidence of optical chirality in a two-dimensional planar system. So far our observations have been confined to the far-field diffraction regime. However, our results suggest that a significant polarization effect will be seen at wavelengths above the diffraction limit, confirming predictions coming from direct calculations of scattered fields[14]. The observed polarization changes are in the scale of several tenths of degrees and therefore promise potential applications. We believe that planar chiral meta-materials could find applications in polarization beam splitters, optical polarizes, spectral filters; polarization and wavelength sensitive apertures; diffraction/polarization routers and WDMs. Possibly the most intriguing feature of planar chirality in non-diffracting systems is that the sign of chirality reverses if the structure is observed from different sides of the plane. This nonreciprocal feature resembles the optical Faraday effect of polarization plane rotation in magnetized media, and contrasts with conventional optical activity where polarization azimuth rotation caused by three-dimensional chirality does not depend on the direction of propagation. As reversing of the propagation direction is equivalent to the time reversal T operation, polarization azimuth rotation on transmission through a planar chiral structure would be a relatively rare manifestation of a T-odd effect in optics. The polarization effects of planar chirality, however, fully obey simultaneous application of time and space inversion, also referred to as enantiomeric reversality. If strong nonreciprocal effect is found in artificially created planar structures, this property could find applications in unidirectional optical valves, similar to that based on the Faraday effect.

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