Metamaterials: Optical Activity without Chirality

E. Plum,[1] X.-X. Liu,[1,2] V. A. Fedotov,[1] Y. Chen,[3] D. P. Tsai,[2] and N. I. Zheludev[1,*]

[1]*Optoelectronics Research Centre, University of Southampton, SO17 1BJ, United Kingdom*
[2]*Department of Physics, National Taiwan University, Taipei, 10617, Taiwan, Republic of China*
[3]*Rutherford Appleton Laboratory, Chilton, Didcot, Oxon, OX11 0QX, United Kingdom*

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We report that the classical phenomenon of optical activity, which is traditionally associated with chirality (helicity) of organic molecules, proteins, and inorganic structures, can be observed in artificial planar media which exhibit neither 3D nor 2D chirality. We observe the effect in the microwave and optical parts of the spectrum at oblique incidence to regular arrays of nonchiral subwavelength metamolecules in the form of strong circular dichroism and birefringence indistinguishable from those of chiral three-dimensional media.

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The phenomenon of optical activity, that is the ability to rotate the polarization state of light, is a fundamental effect of electrodynamics which is traditionally associated with mirror asymmetry (chirality) of organic molecules. The effect has enormous importance for analytical chemistry, crystallography, molecular biology, and is also a signature effect used to detect life forms in space missions. The recognition of chirality as a source negative refraction of light [1–9] needed for the creation of a perfect lens [10] inspired intense work in developing microwave and optical artificial chiral metamaterials [11–15] and yielded a demonstration of negative index due to chirality [11,16,17]. In this Letter, we present a somewhat surprising result that very strong optical activity may be seen in a metamaterial system consisting of metamolecules that itself are not chiral. Here, chirality is drawn extrinsically from the mutual orientation of the wave propagation direction and the two-dimensional metamaterial. We demonstrate the effect in both the microwave and optical parts of the spectrum using artificially created nonchiral planar metamaterial structures and show that they behave indistinguishably from 3D-chiral molecular systems manifesting resonant circular birefringence and dichroism. Our experiments also indicate that in such metamaterials, extrinsic chirality may also lead to negative refraction of circularly polarized electromagnetic waves.

The recent effort in creating artificial metamaterials with strong optical activity was focused on different types of arrays of 3D-chiral metamolecules [11–21]. It is significantly less acknowledged that the effect can also be seen when oriented nonchiral molecules make a chiral triad with the wave vector of light (extrinsic chirality). This mechanism was first described by Bunn [22] and detected in liquid crystals [23]. Here, we show that extrinsic chirality can lead to exceptionally large optical activity and circular dichroism in microwave and photonic *planar* metamaterials that possess neither 2D chirality [24] nor 3D chirality, and which are much simpler to fabricate than metamaterials based on arrays of 3D-chiral metamolecules.

We argue that to manifest optical activity, metamolecules of a planar metamaterial may have a line of mirror symmetry, but shall lack an inversion center, i.e., they will posses a polar direction s (see Fig. 1). A regular oriented array of such metamolecules will not show optical activity at normal incidence. However, the metamaterial will become optically active at oblique incidence provided that the plane of incidence does not contain the polar direction. Indeed, in this case, the wave vector k, normal to the metamolecule plane n and polar vector s, constitute a

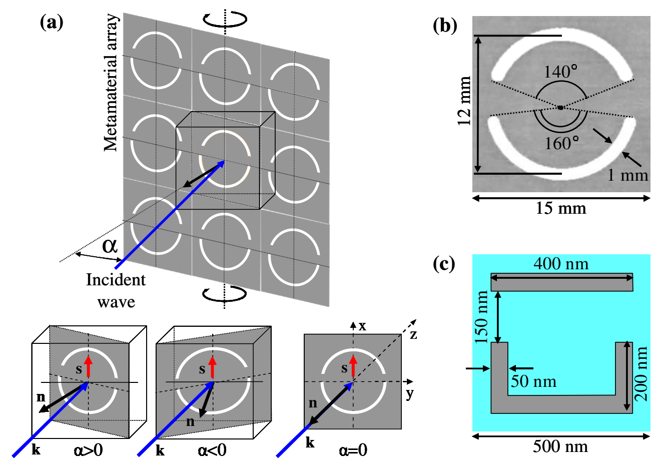

FIG. 1 (color online). (a) Planar metamaterials based on an array of asymmetrically split rings manifest optical activity and circular dichroism at oblique incidence of light. The direction of asymmetry is represented by a polar vector s (long to short arc). Optical activity is seen when the metamaterial plane is tilted around the x-axis so that the sample normal n and the incident wave vector k form an angle $\alpha \neq 0$. Configurations $\pm\alpha$ are enantiomeric arrangements showing optical activity of opposite signs. Configuration $\alpha = 0$, i.e., normal incidence, shows no optical activity. (b) Unit cell of the microwave metamaterial: an asymmetrically split ring aperture in a 1 mm thick aluminum sheet. (c) Unit cell of the photonic metamaterial containing 50 nm thick aluminum wires placed on a 500 μm-thick glass substrate.

3D-chiral triad. The enantiomeric configurations of these vectors corresponding to optical activity of opposite signs are created by tilting the plane of the structure in opposite directions with respect to the incident wave vector (compare $\alpha > 0$ and $\alpha < 0$ in Fig. 1).

The origin of the effect in such a planar nonchiral structure may be readily seen by considering a "unit cell," which contains a single tilted split ring (Fig. 1). Using the terminology of crystallography, the direction of light propagation will be a "screw direction" of the unit cell (that is to say, it will have a sense of twist), if several conditions are met [25]. First, the unit cell itself shall not have an inversion center. This is assured by an asymmetry of the split ring. Second, there should be no reflection symmetry in the plane perpendicular to the propagation direction, which is provided by oblique incidence. Third, there should be no inversion or mirror rotation axis along the propagation direction. This is provided by oblique incidence and the asymmetric split. And finally, there should be no reflection symmetry for any plane containing the propagation direction. This requirement is only fulfilled if the split is not perpendicular, and therefore vector s is not parallel, to the incidence plane yz. Therefore, with reference to Fig. 1(a), in cases $\alpha > 0$ and $\alpha < 0$, the direction of light propagation is a screw direction and supports optical activity. On the contrary, case $\alpha = 0$, i.e., normal incidence, fails the second, third, and forth conditions of the "screw direction" test. For instance, at normal incidence, there is a plane of reflection symmetry containing the propagation direction.

We observed optical activity in microwave and photonic metamaterials based on regular arrays of asymmetrically split rings. Each split ring has a line of mirror symmetry along the x-axis but has no axis of twofold rotation, which enables the introduction of a polar vector s that points towards the short arc [see Fig. 1(a)]. The microwave metamaterial is a self-standing aluminum plate with a size of $\approx 220 \times 220$ mm^2, which is perforated with split ring slits [see Fig. 1(b)]. The period of perforation is 15 mm rendering the structure nondiffracting at normal incidence for frequencies below 20 GHz. The photonic metamaterial consists of aluminum split nanorings manufactured by e-beam lithography on a glass substrate and has size of 500×500 μm^2 [see Fig. 1(c)]. The period of the nanostructure is 500 nm, which ensures no diffraction in the near IR.

The microwave metamaterial has a number of intriguing and useful properties. Being essentially a perforated sheet of metal, it is not transparent apart from a narrow spectral range around the resonant frequency, at which the wavelength is approximately twice the slit length. Transmission at the resonance is "extraordinarily" high and substantially exceeds the fraction of the area taken by the slits. As Joule losses in metals at microwave frequencies are negligible, the incident energy is split between reflected and transmitted radiation, and at the resonance reflection is low [26]. As illustrated below, the structure shows a strong bell-shaped resonance of circular birefringence leading to strong polarization rotation, while circular dichroism is zero at the resonance. This very useful feature is in striking contrast with optical activity in most molecular systems, where characteristically strong resonant polarization rotation is accompanied by substantial circular dichroism resulting in elliptical polarization. Moreover, at the optical activity resonance, the system shows *no linear birefringence* (anisotropy), and eigenstates are therefore two circular polarizations with equally moderate losses.

For microwaves, we measured the complex transmission matrix t relating the incident E^{in} and transmitted E^{out} circularly polarized electric fields and defined as $E_i^{\text{out}} = t_{ij}E_j^{\text{in}}$, where subscripts $+$ and $-$ denote right and left circularly polarized waves correspondingly. Our measure-

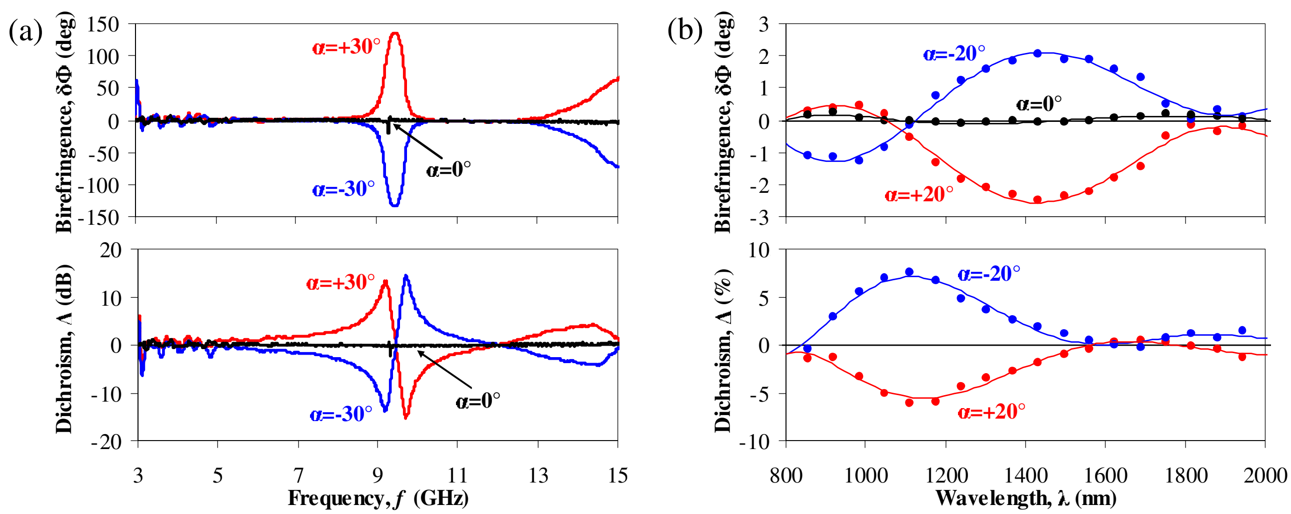

FIG. 2 (color online). Circular birefringence $\delta\phi$ and circular dichroism Δ observed in transmission for different tilt angles α: (a) microwave metamaterial (measured in an anechoic chamber using broadband horn antennas and a vector network analyzer) and (b) photonic structure (measured in a microspectrophotometer using linear polarizers and a superachromatic wave plate).

ments show that t_{++} and t_{--} are generally not equal, indicating true optical activity. The difference between their magnitudes $\Delta = |t_{++}|^2 - |t_{--}|^2$ is a measure of circular dichroism, while the corresponding phase difference $\delta\phi = \arg(t_{++}) - \arg(t_{--})$ is a measure of circular birefringence (see Fig. 2). t_{-+} and t_{+-} are equal within experimental accuracy, which indicates the expected presence of some linear anisotropy but also shows a complete absence of the asymmetric transmission effect recently discovered in planar chiral structures [24]. Importantly, the metamaterials' gyrotropic properties cannot be explained by linear anisotropy, which does not contribute to Δ and $\delta\phi$. Particularly, while linear anisotropy causes a polarization state dependent modulation of azimuth rotation, it has no effect on the material's average polarization rotary power, which is only determined by $\delta\phi$. For the photonic metamaterial, we measured the transmission difference Δ for right and left circular polarizations and the average polarization azimuth rotation $\delta\phi/2$ directly. In all cases, experiments performed in opposite directions of wave propagation show identical results.

In case of the lossless microwave structure, the observed effect has a resonant nature and is strongest around the resonance between 9 and 10 GHz, where the average arc length corresponds to approximately half of the free-space wavelength. For the photonic metamaterial, the effect is weaker and the resonances are broader due to the increase of losses in the metal wires. The following characteristic features of the effect have been observed in the experiments: i) no circular birefringence or dichroism is seen at incidence normal to the metamaterial array ($\alpha = 0$); ii) equal tilt in opposite directions yields circular dichroism and circular birefringence of opposite sign. On the absolute scale, the effect due to extrinsic chirality (polarization azimuth rotation, $|\delta\Phi/2|$, exceeds 60° for microwaves and 1° in optics) appears to be even stronger than that exhibited by microwave and photonic artificial 3D-chiral rosette structures [11,14,16]. Metamaterials of the proposed type, being essentially planar structures, are generally much easier to fabricate than the existing artificial chiral media, especially in the visible, and therefore are capable of superseding the latter as powerful ultrathin circular polarizers and polarization rotators.

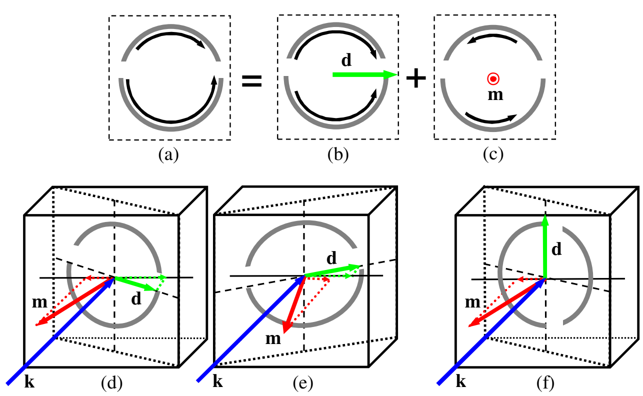

FIG. 3 (color online). Electric and magnetic responses in an asymmetrically split wire ring. Oscillating currents in the split ring (a) can be represented as a sum of symmetric (b) and antisymmetric (c) currents that correspond to the induced electric dipole in the plane of the ring *d* (green arrow) and magnetic dipole perpendicular to the plane *m* (red arrow). For tilted asymmetrically split rings, polarization rotation is strongest and of opposite sign if the projections of *d* and *m* onto the plane perpendicular to the *k*-vector (correspondingly green and red dashed arrows) are either antiparallel (d) or parallel (e). Optical activity is only absent if these projections are orthogonal (f).

The microscopic origin of optical activity in extrinsically chiral split wire rings can be easily understood (see Fig. 3, also see [27]). As with conventional optical activity exhibited by chiral molecules, the effect must result from the presence of both electric and magnetic responses. As illustrated in Fig. 3(a), a wave polarized along the split induces unequal oscillating currents in the upper and lower arches of the ring. This may be represented as a sum of symmetric and antisymmetric currents corresponding to the induced electric dipole in the plane of the ring and magnetic dipole perpendicular to the ring [see Figs. 3(b) and 3(c)]. Now we shall consider non-normal incidence of the wave onto the structure [see Figs. 3(d)–3(f)]. Here blue, red, and green solid arrows represent the wave vector *k* and induced magnetic *m* and electric *d* dipoles of the metamaterial's unit cell, while dashed arrows show projections of the corresponding dipole moments onto the plane perpendicular to the wave vector. The structure shows optical activity if the split is not perpendicular to the plane of incidence. Maximum optical activity is observed when the split is parallel to the plane of incidence; in this case, the wave vector and induced magnetic and electric dipoles are coplanar. The mutual phase difference between the electric and magnetic responses and thus the sign of optical activity depends on the sign of the tilt [compare projections of electric and magnetic dipoles in Figs. 3(d) and 3(e)]. Similarly to how it happens in conventional chiral media, when the wave vector and induced magnetic and electric dipoles of the "metamolecule" are coplanar, the oscillating dipole components perpendicular to the *k*-vector create scattered electromagnetic waves with orthogonal polarizations in the direction of wave propagation, and therefore the polarization of the transmitted wave rotates. On the contrary, if the split is perpendicular to the plane of incidence, the induced magnetic and electric dipoles as well as their projections are orthogonal and the structure does not show any optical activity [see Fig. 3(f)]: the oscillating magnetic and electric dipoles emit electromagnetic waves of the same polarization that propagate along the direction of the incident wave. According to Babinet's principle, the slit metamaterial, which is the Babinet complementary structure to the wire pattern discussed above, will exhibit similar polarization resonances in the same frequency band.

Intriguingly, in the slit metamaterial, in the resonance spectral band from about 9 to 10 GHz, phase velocity

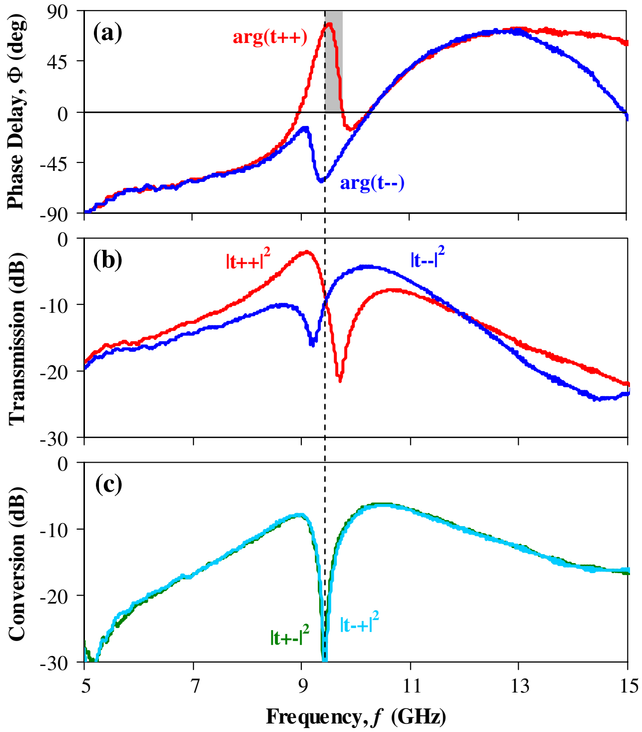

FIG. 4 (color online). (a) Dispersions of phase delay ϕ for transmitted left and right circularly polarized waves. The shaded area indicates the frequency range with almost circular eigenstates, where phase velocity v_p and group velocity v_g for right circular polarization have opposite signs. (b) Transmitted intensity of both left and right circularly polarized waves. (c) Efficiency of circular polarization conversion, which is a direct indication of anisotropy (linear birefringence) of the material response. All data correspond to incidence angle $\alpha = 30°$ onto the microwave metamaterial, see Fig. 1.

($v_p \sim \omega/\phi$, where $\omega = 2\pi f$) and group velocity ($v_g \sim d\omega/d\phi$) for right circular polarization have opposite signs indicating the appearance of a backward wave [see Fig. 4(a)]. In accordance with Pendry [1], this is a necessary condition or signature of negative refraction in bulk chiral media. Following Pendry, negative refraction should be seen at the resonance for one circular polarization only swapping to the other one in the medium's enantiomeric form. Indeed, our experiments show opposite signs of group and phase velocities for right circular polarization at $\alpha = 30°$ and for left circularly polarized waves for the enantiomeric arrangement, at $\alpha = -30°$. Importantly, linear anisotropy essentially disappears [negligible circular conversion $t_{+-} = t_{-+} = \frac{1}{2} \cdot (t_{xx} - t_{yy})$, see Fig. 4(c)]. Thus, the polarization eigenstates are very close to circular and in the k-vector direction, the material behaves as isotropic optically active medium. Moreover, in this spectral range, losses represented by $|t_{++}|^2$ and $|t_{--}|^2$ are relatively small [see Fig. 4(b)].

In conclusion, we have demonstrated strong optical activity and circular dichroism in nonchiral planar microwave and photonic metamaterials. The phenomena are due to extrinsic chirality, which arises from the mutual orientation of the metamaterial and the incident beam. The effect could be exploited for developing novel highly efficient polarization rotators and modulators, and vibration sensors, and may lead to the appearance of a new class of negative index metamaterials, in addition to the recently demonstrated conventional chiral negative index media [11,16,17].

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*niz@orc.soton.ac.uk
www.nanophotonics.org.uk/niz/

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