Asymmetric transmission of electromagnetic radiation where you do not expect it: when passing through a two-dimensional array of highly symmetric particles

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We report that directional asymmetry of transmission of circularly polarized waves, which previously had only been observed in two-dimensional arrays of planar chiral particles, may occur in a much larger class of planar periodic structures. If the particles constituting such periodic arrays are not chiral, asymmetric transmission can still be observed if planar enantiomorphism is imposed on the structural level, by a particular arrangement of particles in the array. Moreover, in order to see asymmetric transmission at oblique incidence planar enantiomorphism is required not for the structure itself, but only for the projection of the structure onto the plane normal to the incident beam. Our findings confirmed by a series of model microwave experiments show that asymmetric transmission may occur in highly symmetric structures such as arrays of spherical semiconductor quantum dots at oblique incidence or in two-dimensional arrays of oriented elliptical plasmonic metal particles at normal incidence.

"I call any geometrical figure, or group of points, chiral, and say it has chirality, if its image in a plane mirror, ideally realized, cannot be brought to coincide with itself." Lord Kelvin (1904) [1]. This famous definition of enantiomorphism by Lord Kelvin is usually applied in three dimensions, where it defines objects like helices, DNA, sugar molecules and the crystal lattice of quartz as 3D-chiral. 3D chirality, which leads to optical activity with its numerous technological applications, is well-studied. It is much less acknowledged that enantiomorphism can also be defined in two dimensions, where it describes the twisted nature of planar patterns like spirals or S-elements. In contrast to 3D-chiral objects, 2D-chiral (planar chiral) patterns have the peculiar property that their sense of twist is reversed for observation from opposite directions. Thus waves incident on the front and back of a planar chiral interface see "different" structures, which could have different optical properties. As twisted planar patterns are rare in nature, it was discovered only a few years ago in metamaterials that 2D chirality indeed gives rise to a new fundamental effect of electromagnetism termed asymmetric transmission [2]. The phenomenon manifests itself as a difference in normal incidence transmission and retardation of a circularly polarized wave for opposite directions of propagation. It was observed in regular sub-wavelength arrays of anisotropic intrinsically 2D-chiral meta-molecules [2–5] as well as isolated plasmonic nano-structures [6] and has been linked to directionally asymmetric absorption losses [5].

Here we demonstrate for the first time that asymmetric transmission can be exhibited by regular arrays consisting of meta-molecules that are not 2D-chiral. The effect is attributed to 2D chirality of the entire array, so-called structural chirality [7, 8]. While structural chirality as a source of asymmetric transmission was not known, its intrinsic form had previously attracted attention as a source of chiral effects in *diffraction*, where it was shown to lead to polarization azimuth rotation in diffracted beams [9, 10].

Like molecular chirality [11], structural chirality can arise in two ways: intrinsically or extrinsically. As illustrated in Figs. 1(a) and (b), the intrinsic form of structural 2D chirality results from the orientation of achiral meta-molecules placed in a planar regular array, when the lines of mirror symmetry of the molecules and the mirror lines associated with the array's lattice do not coincide. Note that in this case mirror-forms of the array cannot be superimposed by translations and rotations in the plane, which makes the entire structure 2D-chiral.

As shown by Figs. 1(c) and (d), structural 2D chirality can also be imposed extrinsically even in a regular array containing meta-molecules of the highest symmetry. This is achieved by tilting the array around any in-plane axis that does not coincide with one of the array's lines of mirror symmetry. It is easy to see that in this case the metamaterial's projection onto the plane normal to the incidence direction becomes structurally 2D-chiral and anisotropic. Note that chirality here arises from the arrangement of the meta-molecules, rather than their internal structure. This implies that asymmetric transmission being essentially a 2D-chiral effect may be observed in any planar regular array containing identical particles of any symmetry. Thus asymmetric transmission, which has been previously perceived as an exotic effect specific to metamaterials, may in fact be a common phenomenon.

In the experiments reported here we studied two different types of planar metamaterials based on asymmetrically split rings and pairs of concentric rings respectively, with the dimensions that are specified in Fig. 2. Both metamaterial structures were formed by a square array of about 200 meta-molecules separated by 15 mm, which rendered our structures non-diffracting below 13 GHz for angles of incidence of up to 30°. The patterns were etched on 1.6 mm thick lossy FR4 printed circuit boards (Im $\varepsilon \sim 0.1$) covered with a 35 μm copper layer using

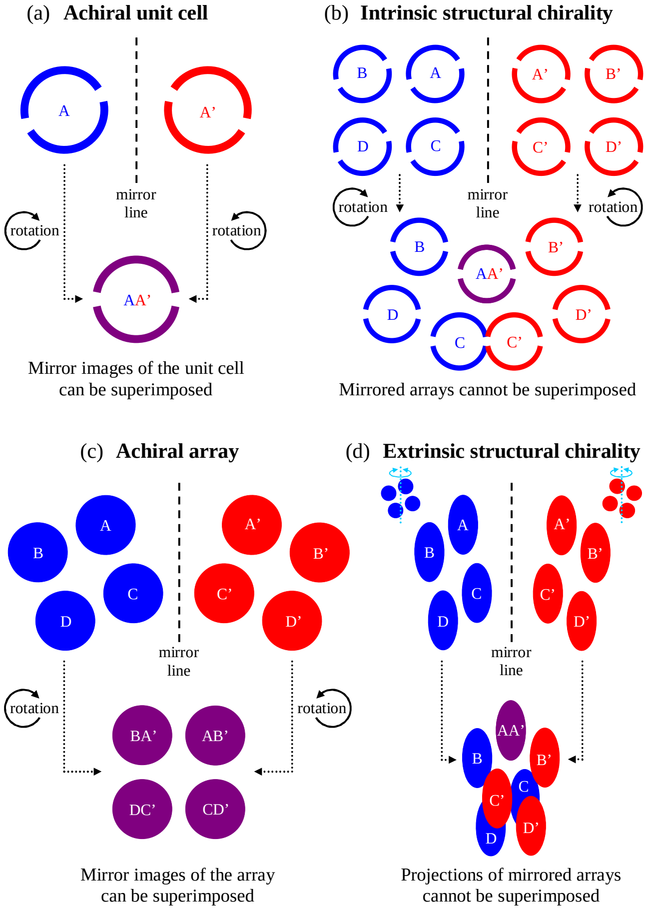

FIG. 1: (Color online) **Intrinsic structural chirality:** Anisotropic achiral meta-molecules (a) can form a structurally planar chiral array (b). While a single meta-molecule (blue) and its mirror image (red) can be superimposed by translation and rotation (purple), certain arrays of such meta-molecules show planar enantiomorphism: congruency can only be achieved for one meta-molecule in the array (purpule) while the rest of the mirrored arrays does not coincide. **Extrinsic structural chirality:** An array (c) of highly symmetric meta-molecules (blue) is congruent with its mirror image (red) and therefore does not have intrinsic chirality. However (d), when it is tilted with respect of the observation direction its projection onto the plane normal to this direction becomes planar chiral.

standard photolithography. The transmission properties of the metamaterials were measured between 5 and 12 GHz in a microwave anechoic chamber using a vector network analyzer (Agilent E8364B) and linearly polarized broadband horn antennas (Schwarzbeck BBHA 9120D) equipped with lens concentrators. In particular, we measured the structures' transmission matrix $E_i^0 = t_{ij}E_j^t$. To study chirality-related effects the matrix was transformed to the circular polarization basis, where indices i and j denote the handedness of the circularly polarized components: “+” for right-handed (RCP) and “-” for left-handed (LCP). In terms of power the transmission and polarization conversion levels are given by $T_{ij} = |t_{ij}|^2$.

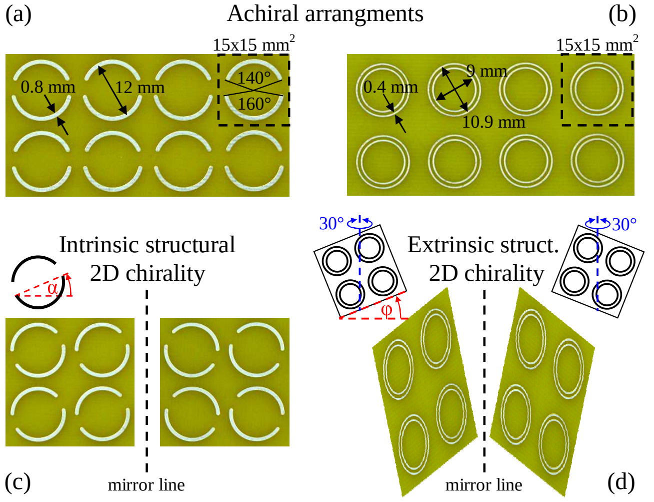

FIG. 2: (Color online) **Metamaterial samples.** Panels (a) and (b) show achiral arrangements of asymmetrically split rings and double rings respectively. (c) Rotation of the split rings by an angle $\alpha \neq n \cdot 45°$, $n \in \mathbb{Z}$ leads to intrinsic chirality of the array, which becomes different from its mirror image. (d) At oblique incidence, even a regular array of rings can become extrinsically 2D-chiral: For orientations $\varphi \neq n \cdot 45°$, $n \in \mathbb{Z}$ the projection of the double ring array onto the plane normal to the direction of incidence is 2D-chiral.

Eight different versions of the asymmetric split ring array were studied at normal incidence, where the orientation of the split α was varied in steps of $11.25° = \pi/16$ relative to the achiral arrangement shown in Fig. 2(a). As illustrated in Fig. 2(c), $\pm\alpha$ correspond to structural planar chirality of opposite handedness, while rotations by α and $\alpha + 90°$ yield identical metamaterial arrays.

The double ring array was characterized at 30° oblique incidence for different orientations φ of the array relative to the plane of incidence. For $\varphi \neq n \cdot 45°$ ($n \in \mathbb{Z}$) the projection of the entire pattern onto the plane normal to the propagation direction becomes 2D-chiral. Similarly to the case of asymmetrically split rings, orientations $\pm\varphi$ correspond to extrinsically 2D-chiral arrangements of opposite handedness, while an in-plane rotation of the metamaterial by 90° results in an identical experimental configuration.

Fig. 3 presents typical spectra of direct transmission intensities T_{++}, T_{--} and circular polarization conversion T_{-+}, T_{+-} for achiral and chiral arrangements of both types of planar metamaterial. In all studied cases, the direct transmission intensities (as well as field transmission coefficients t_{++} and t_{--}) did not depend on the handedness or propagation direction of incident circularly polarized waves. In particular this confirms the expected absence of the 3D-chiral phenomena of optical activity $\arg(t_{++}) - \arg(t_{--})$ and circular dichroism $T_{++} - T_{--}$ (after all neither the metamaterial patterns

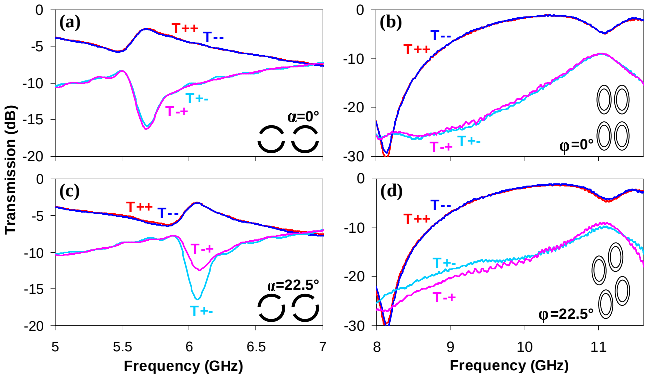

FIG. 3: (Color online) Direct transmission T_{++}, T_{--} and circular polarization conversion T_{-+}, T_{+-} spectra for: (a) Normal incidence onto an achiral array of asymmetrically split rings ($\alpha = 0°$). (b) 30° oblique incidence onto the double ring array oriented in a way which does not lead to extrinsic chirality ($\varphi = 0°$). (c) Normal incidence onto an array of asymmetrically split rings which has intrinsic structural planar chirality ($\alpha = 22.5°$). (d) 30° oblique incidence onto the double ring array rotated to become extrinsically structurally 2D-chiral ($\varphi = 22.5°$). Insets show the metamaterial patterns projected onto the plane normal to the incident beam.

nor the experimental arrangements had 3D chirality). The presence of circular polarization conversion indicates an anisotropic metamaterial response in all cases. Figs. 3(a) and 3 (b) show that for both arrays of split rings and double rings in the absence of structural 2D chirality the intensities of circular polarization conversion are identical and independent of the propagation direction, indicating the complete absence of asymmetric transmission. When, however, the split rings were rotated by an angle α to form a structurally 2D-chiral array, normal incidence transmission through the metamaterial showed a resonant region around 6 GHz where the right-to-left and left-to-right conversion efficiencies were different from each other, $T_{-+} \neq T_{+-}$, as illustrated in Fig. 3(c) for $\alpha = 22.5°$. Similarly, when the double ring array was rotated in its plane by an angle φ, so that for oblique incidence its projection onto the plane normal to the wave propagation direction became 2D-chiral, a broad band of asymmetric circular polarization conversion appeared (as shown in Fig. 3(d) for 30° incidence and $\varphi = 22.5°$). Furthermore, our data indicate that in both cases the conversion efficiencies are simply interchanged for opposite directions of propagation, $\overrightarrow{T}_{ij} = \overleftarrow{T}_{ji}$ and thus, for example, RCP waves incident on front and back of the metamaterials will experience different levels of circular polarization conversion, $\overrightarrow{T}_{-+} \neq \overleftarrow{T}_{-+}$. Given that the direct transmission terms are independent of the propagation direction, the total transmission (i.e. transmission that would be measured with polarization insensitive detector) $T_{+} = T_{++} + T_{-+}$ is different for the circularly polarized waves incident on front and back of the metamaterial arrays.

Fig. 4 illustrates the dependance of the asymmetric effect on the arrangement of the meta-molecules in both types of planar metamaterial array. When the split rings were rotated by a multiple of 45°, structural 2D chirality of the array was absent and asymmetric transmission could not be detected. Other orientations of the split rings used in our experiments led to a 0.2 GHz wide band of asymmetric transmission observed between 5.5 and 6.5 GHz [see Fig. 4(a)], whose exact spectral position and magnitude were controlled by the slit's orientation α. The largest asymmetry was observed for $\alpha = \pm 22.5° = \pm\pi/8$, where the difference in circular polarization conversion $T_{-+} - T_{+-}$ was about 4 dB. Importantly, as we mentioned earlier orientations $\pm\alpha$ correspond to mirror forms of the split-ring array and should lead to asymmetric transmission of opposite sign, which was exactly what we observed in our experiments [see Fig. 4(a)]. The double ring array was extrinsically 2D-chiral at oblique incidence for in-plane orientations φ excluding multiples of 45° and, as Fig. 4(b) illustrates, exhibited relatively wide bands of asymmetric transmission with the sign of the effect being reversed for the enantiomeric forms of the array's projection.

Thus, our data confirm the existence of asymmetric transmission due to both intrinsic and extrinsic structural chirality in arrays of achiral meta-molecules. While asymmetric transmission was initially only known for normal incidence onto lossy arrays of anisotropic and intrinsically 2D-chiral meta-molecules [2], we are now able to identify a much larger class of structures exhibiting the effect: *asymmetric transmission can occur for any lossy array of particles, when its projection onto the plane normal to the direction of incidence is 2D-chiral and anisotropic.* At oblique incidence any regular array of even perfectly symmetric particles can become planar chiral and anisotropic in projection, while at normal incidence - when the structure and its projection coincide - planar chirality and anisotropy must be properties of the array itself.

Importantly, asymmetric transmission due to structural planar chirality is a direct consequence of electromagnetic interactions between meta-molecules, as there can be no first-order contribution from individual achiral meta-molecules. The structures studied here present the two limiting cases of strong and weak inter-molecular coupling.

Regular arrays of asymmetrically split rings are known to exhibit a strong resonant response of collective nature established through strong interactions between split rings [12]. When excited by a circularly polarized wave this interaction appears to be *resonantly* perturbed by the 2D-chiral arrangement of the array in a way that depends on the incident wave's handedness and propagation direction. Such polarization sensitive excitation previously observed in planar chiral meta-molecules has

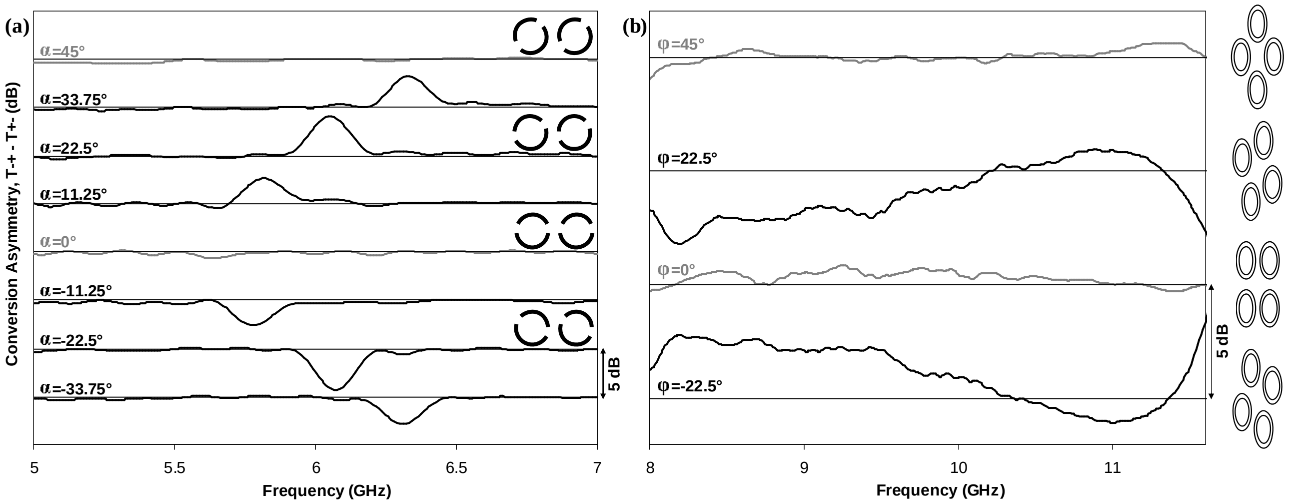

FIG. 4: (a) Conversion asymmetry, $T_{-+} - T_{+-}$, for normal incidence onto arrays of asymmetrically split rings as a function of the slit's orientation α. (b) Conversion asymmetry for 30° oblique incidence onto the double ring metamaterial as a function of its in-plane orientation φ. Insets show the metamaterial patterns as they are seen by an observer looking along the incident beam.

been called enantiomerically sensitive plasmon [3]. The sensitivity of this enantiomerically sensitive resonance to changes in the inter-molecular coupling conditions is particularly evident in the substantial frequency-shift of the resonance with changing orientation α of the split, as presented in Fig. 4(a).

The array of double rings presents an example of an incoherent metamaterial system with weak inter-molecular coupling [12]. The source of structural structural 2D chirality in such case has a non-resonant, broadband nature leading to a relatively weak asymmetry in transmission [see Fig. 3(d)].

In summary, we have experimentally demonstrated asymmetric transmission due to intrinsic and extrinsic structural chirality in arrays of achiral meta-molecules. Our results greatly expand the range of natural and artificial materials in which the phenomenon may be expected, making asymmetric transmission a mainstream electromagnetic effect rather than a curiosity of planar chiral metamaterials. Indeed, while only few natural examples of intrinsically 2D-chiral interfaces are known, regular arrays of simple particles are much more common and much easier to manufacture. This indicates that asymmetric transmission should be observable in natural or self-assembled structures. Prime candidates for asymmetric transmission at oblique incidence would be square arrays of plasmonic spheres or semiconductor quantum dots and lossy double-periodic gratings, which have the same symmetry, as the double ring array studied here.

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