Giant gyrotropy due to electromagnetic coupling

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We report the first experimental evidence that electromagnetic coupling between physically separated planar metal patterns located in parallel planes provides for exceptionally strong polarization rotatory power if one pattern is twisted in respect to the other, creating a 3D chiral object. In terms of optical rotary power per sample of thickness equal to one wavelength, the bi-layered structure rotates five orders of magnitude stronger than a gyrotropic crystal of quartz in the visible spectrum. We also saw a signature of negative refraction for circularly polarized waves propagating through the chiral slab.

The ability to rotate the polarization state of light (gyrotropy) by chiral molecules is one of the most fundamental phenomena of electrodynamics. It was discovered by F. Aragot in 1811 and is now widely used in analytical chemistry, biology and crystallography for identifying the spatial structure of molecules. Recent explosive increase in the interest in gyrotropic media is driven by an opportunity for the development of negative index metamaterials, where simultaneous electric and magnetic response of gyrotropic structures are required to achieve negative refraction. Sculptured helical pillars for the optical part of the spectrum, helical wire springs and twisted Swiss-role metal structures for microwave applications have been discussed as possible candidates for achieving strong artificial gyrotropy that can be used for implementing negative refraction. However, from meta-material prospective it would be very desirable if chirality could be achieved by planar patterning using well-established planar technologies, thus making nano-fabrication of such structures for the optical part of the spectrum a practical proposition. The opportunity of creating true 3D chirality in non-contacting layers of planar metal structures was first identified in Ref. 11. It was suggested that inductive coupling between two identical mutually twisted metal patterns can create an optically active chiral object and thus provide for gyrotropy.

In this letter we show the first experimental demonstration that giant optical gyrotropy can be achieved in a bi-layered chiral structure through electromagnetic coupling between the layers and that there is no need to sculpture continuous helix-like volume three-dimensional chiral objects to achieve strong polarization rotatory power. We also saw clear evidence of negative refraction in the structure. The experiments were performed in the microwave part of the spectrum. Although we expect the effect to be seen with a large variety of patterns, we investigated a structure consisting of two identical metal rosettes of 4-fold rotational symmetry located in parallel planes, as presented on Fig. 1. The 4-fold symmetry of the rosette ensures that the structure is isotropic for observations at normal incidence and therefore shows no birefringence. Due to curved lines rosette-like structure exhibit resonant properties at wavelengths larger than the overall size of the design. The latter would be important for achieving a non-diffracting regime if two-dimensional arrays of such structures were used to form planar metamaterial sheets and volume structures. The rosettes were etched from 35 μm flat copper film on both sides of a dielectric substrate. The rosettes had the length \( L = 53 \) mm and strip width \( W = 0.4 \) mm and were spaced by a homogeneous dielectric layer of thickness \( d = 1.5 \) mm (\( \varepsilon = 3.77 + i 0.03 \)).

We studied circular birefringence and dichroism of the structure in 4.5 – 7.0 GHz frequency range (wavelength range 4.3 – 6.7 cm) using a microwave waveguide polarimeter. The polarimeter included a 480 mm long circular waveguide with a diameter of 41.5 mm and two high quality circular polarizers of either same or opposite handedness (series 64 by Flann Microwave) attached on both sides of the waveguide. Each sample was placed in the middle of the waveguide, perpendicular to its axis. A full S-parameter vector network analyzer (model E8364B by Agilent) was used to measure both magnitude and phase of the wave transmitted through the polarimeter.

In our experiments we compared pairs of enantiomeric forms of the structure (designated as type D and S in
the mutual twist $\phi$. Frequency dependencies of: (i) circular dichroism, $\Delta$, (black line) and circular differential phase delay, $\delta$, (gray line); (ii) transmission losses for right circularly polarized wave (black line) and left circularly polarized wave (gray line); (iii) phase delay for right circularly polarized wave (black line) and left circularly polarized wave (gray line). The shaded area in section (iii) represents a frequency range where the phase velocity $v_p$ and group velocity $v_g$ have opposite signs for right circular polarization, which is a signature of negative refraction.

In Fig. 1) with various angles of mutual twist, $\varphi$, in the range from $\varphi = 0^\circ$ to $\varphi = 45^\circ$. To describe the results of polarimetric measurements we will define transmission of a sample, $t$, measured by the polarimeter as follows: the superscript index refers to the type of the sample, $D$ or $S$, while subscript indices refer to the state of polarizer and analyzer. In these terms circular dichroism is defined as $\Delta = |t_{++}^S|^2 - |t_{--}^S|^2$ while circular differential phase delay (responsible for circular birefringence) is defined as $\delta = \arg(t_{++}^S) - \arg(t_{--}^S)$. To eliminate any possible polarization effects, which could have resulted not from three-dimensional chirality, but from anisotropic imperfections of polarimeter and/or sample, we performed experiments at different mutual orientations rotating the sample around the axis of the cylindrical waveguide, and found virtually no dependence of the observed effects on the orientation.

The structure’s three-dimensional chirality and thus its gyrotropic characteristics should depend strongly on the mutual orientation of the rosettes and distance $d$ between them. To verify this we manufactured a truly planar version of the structures by etching both rosettes from the same metal film ($d = 0$) and sandwiching them between two layers of dielectric for symmetry. We also manufactured a bi-layered structure with no twist between rosettes ($\varphi = 0$, $d \neq 0$). No circular dichroism or differential phase delay was observed in both cases. We also studied the dependence of gyrotropic characteristics of the structure on the value of $\varphi$. The complete picture of the dependence of gyrotropy on $\varphi$ may be obtained by measuring it in the interval of $\varphi$ between 0 and 45°. Indeed, due to the four-fold symmetry of the rosettes, the structure with $\varphi$ between 0 and 45° is equivalent to the structures twisted on $\varphi \pm n \cdot 90^\circ$, where $n$ is an integer number. The sinistral structure with $\varphi$ in between 45° and 90° is equivalent to the dextral structure with mutual twist of $\varphi - 90^\circ$ and therefore has an opposite sign of gyrotropy.

Characteristic spectral dependencies of $\Delta$ and $\delta$ exhibited by the sinistral structure are presented in Fig. 2(i) for $\varphi = 15^\circ$. They show two resonances located below and above 6 GHz. They will be called resonance $A$ and $B$ correspondingly. From Fig. 2(ii) it follows that on a sinistral (left) structure, losses at resonance $A$ for right circular polarization are smaller than for left circular polarization. However, losses at resonance $B$ for right circular polarization are stronger than for left circular polarization. Phase delay $\psi = \arg(t)$ is presented on Fig. 2(iii). Provided that an electromagnetic wave is presented in the form $e^{i(\omega t+kx)}$, in the proximity of resonance $A$ the group velocity $v_g = d/(\partial \psi/\partial \omega)$ for right circular polarization in the sinistral structure has opposite sign to the phase velocity $v_p = c\psi\lambda_g/2\pi d$ (here $\lambda_g$ is guided the wavelength in the circular waveguide). In accordance with Ref. 2, this is a signature of negative refraction in chiral media. In proximity of resonance $B$, negative refraction is observed for both circular polarizations on the background of high losses. Availability of negative refraction is especially important in the proximity of resonance $A$, where overall losses are small.

In general, a linearly polarized wave transmitted through the structure will become elliptical on transmission and its polarization azimuth will rotate. Using standard definitions of the degree for ellipticity $\eta$ and polarization azimuth rotation $\theta$ of elliptically polarized light, we calculated polarization changes of the linearly polarized incident wave as follows: $\eta = \ldots$
FIG. 3: Frequency dependencies of polarization azimuth rotation $\theta$ (left column) and ellipticity, in terms of the ellipticity angle $\eta$ (right column) that a linearly polarized wave would acquire upon transmission through a sinistral bi-layered chiral structure for various twist angles $\varphi$. Vertical scale is shown in the right bottom corners. Dotted line at $\varphi = 15^\circ$ shows the frequency dispersion of the effect as predicted by the Born-Kuhn model.

$$\frac{1}{2} \arcsin \left( \frac{\Delta}{|t_{S}^{+} + t_{D}^{+}|^2} \right), \quad \theta = -\frac{1}{2} \delta.$$ This is presented in Fig. 3 for different values of $\varphi$. The peak values of both rotation and ellipticity initially increase with $\varphi$. They reach their absolute maxima at about $\varphi = 15^\circ$ with $\eta = -30^\circ$ at frequency $\nu_B$ ($\eta = -45^\circ$ corresponds to perfectly left-handed circularly polarized light). At the exact resonance no polarization rotation is seen as its dispersion passes zero, but in proximity of resonance $\nu_B$ rotation reaches $\theta_B = 28^\circ$. Between the peaks, in the spectral range of low losses and virtually zero dichroism, we observe a pure rotation of polarization azimuth of about $\theta_0 = 3^\circ$. With further increase of $\varphi$, resonances A and B move closer to one another and the dispersions of $\theta$ and $\eta$ change. Peak value of rotation and ellipticity decreases as well as rotatory power at frequencies between the peaks. Gyrotropy completely collapses at $\varphi = 45^\circ$ as should be expected, see Ref. [11].

It shall be noted that the observed rotation induced by the artificial bi-layered structure (which has a thickness $d$ of only about 1/30 of the wavelength) is huge. To appreciate its magnitude it shall be compared with the gyrotropy of natural optical active materials. Indeed, in terms of optical rotatory power per sample thickness equal to one wavelength, bi-layered structure rotates five orders of magnitude stronger than a gyrotrropic crystal of quartz and three orders of magnitude stronger than cholesteric liquid crystals in the visible spectrum (specific rotatory power of quartz and cholesteric liquid crystals is about $20^\circ$/mm and $10^3^\circ$/mm respectively). Rotatory power of the bi-layered system is also two orders of magnitude stronger that in the recently intro-
duced metal-on-dielectric chiral system, where resonant rotation of about 1° was seen in a sample 1/6 of the wavelength thick \[14\].

To identify the underlying nature of the observed polarization effect we recall the classical model of gyrotropy developed by Born and Kuhn \[15\]. In this model two spatially separated charged oscillators moving along orthogonal directions have an elastic binding between them. Excitation of one of them by the incident electromagnetic wave is then transferred by the elastic coupling to the other. Induced oscillations of its charge then re-emit a wave at a different polarization and with some delay, thus ensuring both polarization azimuth rotation and dichroism. Analogously, current driven in the rosette arm by the incident wave is inductively (or capacitively) coupled to the current in the rosette arm of the second layer. The induced current in the second layer is then re-emitted into the transmitted wave with a different polarization state providing for gyrotropy. It is remarkable how well the Born-Kuhn model is suitable for describing this process. It gives the following dispersion of the effect \(\phi_0 \propto \left( \frac{\text{Re}}{\text{Im}} \right) \frac{\xi^2}{\nu^2 + \xi^2} \tau_{01}^2 \), where \(\nu_0\) and \(\gamma\) are the resonant frequency and damping parameter of individual oscillator and \(\xi\) is the Hooke coefficient of elastic force between the oscillators \[15\]. Here, \(\nu_0\) is analogous to the resonant frequency of the dipole interaction of an electromagnetic wave with the arm of an individual rosette, which shall be about \(c/L = 5.7\) GHz. The Hooke elastic interaction between the oscillators in the Born-Kuhn model is analogous to the electromagnetic coupling between rosettes. According to above formula, the Born-Kuhn model predicts a two-peak dispersion of rotatory power and circular dichroism with the peak spectral separation increasing with coupling \(\xi\). Indeed, in our experiments the peak separation is at maximum for small \(\phi\) when the electromagnetic interaction between rosettes is strong. It decreases with increasing \(\phi\) when rosette overlapping diminishes and separation of the peaks reduces accordingly. The Born-Kuhn dispersion accurately describes the main features of the rotatory power and circular dichroism in the bi-layered structure as may be seen in Fig.3, where theoretical dispersion curves are plotted as a dotted line for \(\phi = 15^\circ\). \(\nu_0 = 5.247\) GHz, \(\gamma = 4.4 \cdot 10^7 s^{-1}\), \(\xi = 4.6 \cdot 10^{19} s^{-2}\). Importantly, the model indicates strong coupling between rosettes in the bi-layered system, where figure of merit is \(\xi/\nu_0^2 = 0.26\), i.e. the energy of the interaction between rosettes amounts to a quarter of the energy of interaction between individual rosette and field, which explains incredibly strong gyrotropy of the system in chiral configurations. The Born-Kuhn model is less accurate in giving a correct ratio of the effect magnitude in the peaks. This small discrepancy is not surprising as elastic coupling is not really equivalent to the electromagnetic one, and there are other mechanism of enantiomerically sensitive interactions with the bi-layered structure, which are not covered by the Born-Kuhn model. For instance, one can see the twisted rosette pair as an enantiomeric sensitive scattering object. Scattering, could happen in all direction creating enantiomerically sensitive losses for the wave propagating in the forward direction. In the reality of the confined environment of the waveguide we perhaps see a strong interplay between the Born-Kuhn like gyrotropy and enantiomerically sensitive scattering that creates a complex frequency dispersion of the effect at various \(\phi\).

In summary, we provided the first experimental evidence that strong gyrotropy can be achieved by electromagnetic coupling in chiral bi-layered disconnected metal structures and saw a signature of negative refraction for circularly polarized waves on the chiral bi-layered structure. We expect that optical activity of this nature will also be displayed by appropriately scaled sub-wavelength nanostructures in the optical part of the spectrum.

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