Giant Gyrotropy due to Electromagnetic-Field Coupling in a Bilayered Chiral Structure

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We report experimental evidence that electromagnetic coupling between physically separated planar metal patterns located in parallel planes provides for extremely strong polarization rotatory power if one pattern is twisted with respect to the other, creating a chiral object. In terms of a rotary power per sample thickness equal to one wavelength, the bilayered structure rotates 5 orders of magnitude stronger than a gyrotropic crystal of quartz in the visible spectrum.

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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. Materials with strong rotatory power or circular dichroism are always in demand as polarization controllers and circular polarizers for optoelectronic, life science microscopy, and display applications and are widely used in photography. The recent explosive increase in interest in gyrotropic media is driven by an opportunity for the development of negative index metamaterials, where simultaneous electric and magnetic responses of gyrotropic structures are required to achieve negative refraction [1–7]. However, naturally available gyrotropic materials showing negative refraction are not yet identified. Sculptured helical pillars for the optical part of the spectrum [8], helical wire springs [9,10], and twisted Swiss-role metal structures [2] for microwave applications have been discussed as possible candidates for achieving strong artificial gyrotropy. However, from the metamaterial perspective it would be very desirable if chirality could be achieved by surface patterning using well-established planar technologies, thus making nanofabrication of such structures for the optical part of the spectrum a practical proposition. The opportunity of creating true 3D chirality in noncontacting 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 a chiral object and thus provide for gyrotropy.

In this Letter we present an experiential demonstration that unexpectedly strong, giant gyrotropy can be achieved in a bilayered chiral structure through electromagnetic coupling between the layers and that there is no need to sculpture continuous helixlike volume three-dimensional chiral objects to achieve strong polarization rotatory power. 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 fourfold rotational symmetry located in parallel planes, as presented on Fig. 1. The fourfold symmetry of the rosette ensures that the structure is isotropic for observations at normal incidence and therefore shows no birefringence. Because of curved lines, the rosettelike structure can exhibit resonant properties at wavelengths larger than the overall size of the design. The latter would be important for achieving a nondiffracting 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 ($\varepsilon = 3.77 + i0.03$).

We studied circular birefringence and dichroism of the structure in the 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

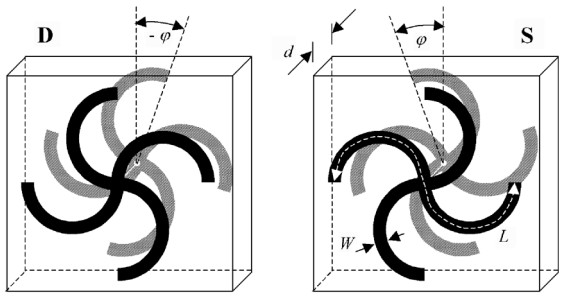

FIG. 1. Dextral (**D**, right-handed) and sinistral (**S**, left-handed) enantiomeric helicoidal bilayered structures constructed from planar metal rosettes separated by a dielectric slab of thickness d. L is the end-to-end length of the rosette's strip and W is its width. The rosettes forming the enantiomeric structures are mutually twisted around the axis of fourfold symmetry on angles $-\varphi$ and φ, respectively.

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 delay of the wave transmitted through the polarimeter.

In our experiments we studied enantiomeric forms of the structure (designated as type **D** and **S** in Fig. 1) with various angles of mutual twist, φ, in the range from $\varphi = 0°$ to $\varphi = 45°$. 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 of, for example, sinistral structure 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 on one side of the dielectric substrate from the same metal film ($d = 0$). We also manufactured a bilayered 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 the gyrotropic characteristics of the structure on the value of φ. The complete picture of the dependence of gyrotropy on φ may be obtained by measuring it in the interval of φ between 0° and 45°. Indeed, due to the fourfold symmetry of the rosettes, the structure with φ between 0° and 45° is equivalent to the structures twisted on $\varphi \pm n \times 90°$, where n is an integer number. While the structure with φ in between 45° and 90° is equivalent to its enantiomeric form with mutual twist of $\varphi \pm n \times 90°$ and therefore has an opposite sign of gyrotropy.

Characteristic spectral dependencies of Δ and δ exhibited by the sinistral (**S**) structure are presented in Fig. 2(a) for $\varphi = 15°$. They show two resonances located below and above 6 GHz. They will be called resonance A and B correspondingly. From Fig. 2(b) it follows that in case of the sinistral 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(c). 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 λ_g is the guided wavelength in the waveguide and electromagnetic wave is presented in the form $e^{i(\omega t+kx)}$). In accordance with Ref. [2] this is a signature, or the necessary condition of negative refraction in chiral media, indicating that a low-loss metamaterial consisting of regular arrays of bilayered chiral elements with electromagnetic coupling shall be closely investigated for the effect. In the proximity of the resonance B, there are possible signs of negative refraction of nonchiral nature. We believe that absorption associated with losses in the dielectric can be further reduced, at least in the microwave part of the spectrum, by using freestanding metal structures.

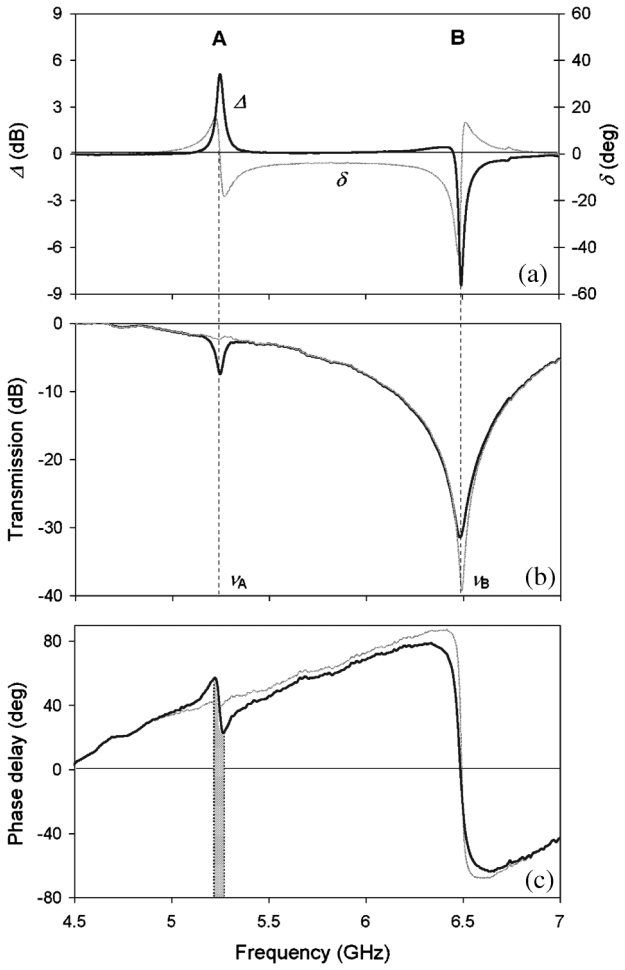

FIG. 2. Electromagnetic properties of the bilayered sinistral chiral structure with mutual twist $\varphi = 15°$. Frequency dependencies of: (a) circular dichroism, Δ (black line) and circular differential phase delay, δ (gray line); (b) transmission losses for left circularly polarized wave (black line) and right circularly polarized wave (gray line); (c) the phase delay for left circularly polarized wave (black line) and right circularly polarized wave (gray line). The shaded area in (c) represents a frequency range where the phase velocity v_p and group velocity v_g have opposite signs for right circular polarization, which could be a signature of negative refraction.

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 η and polarization azimuth rotation θ of elliptically polarized light [12], we calculated polarization changes of the linearly polarized wave incident on the sinistral structure as follows: $\eta = \frac{1}{2} \times \arcsin(\frac{\Delta}{|t^S_{++}|^2+|t^S_{-}|^2})$, $\theta = -\frac{1}{2}\delta$. This is presented in Fig. 3 for different values of φ. The peak values of both rotation and ellipticity initially increase with φ. They reach their absolute maxima at about $\varphi = 15°$ with $\eta = -30°$ at frequency ν_B ($\eta = -45°$ 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 ν_B rotation reaches $\theta_{B^+} = 28°$. Between the peaks, in the spectral range of low losses and virtually zero dichroism, we observe a pure rotation of polarization azimuth of about $\theta_0 = 3°$. With further increase of φ, resonances A and B move closer to one another and the dispersions of θ and η change. Peak value of rotation and ellipticity decreases as well as rotatory power at frequencies between the peaks. Gyrotropy completely collapses at $\varphi = 45°$ as should be expected.

It shall be noted that the observed rotation induced by the artificial bilayered 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 rotary power per sample thickness equal to one wavelength, bilayered structure rotates *five orders* of magnitude stronger than a gyrotropic crystal of quartz, *four orders* of magnitude stronger than cinnibar at the absorbtion edge, and correspondingly *three* and *two orders* of magnitude stronger than cholesteric liquid crystals and sculpture thin films in the visible spectrum (specific rotatory power of quartz, cinnibar, liquid crystals, and sculptured films is about $20°/\text{mm}$, $600°/\text{mm}$, $10^3°/\text{mm}$ [13], and $6 \times 10^3°/\text{mm}$ [14], respectively). Rotatory power of the bilayered system is also 2 orders of magnitude stronger that in the recently introduced metal-on-dielectric chiral system, where resonant rotation of about 1° was seen in a sample $1/6$ of the wavelength thick [15].

To identify the underlying nature of the observed polarization effect we recall the classical model of gyrotropy developed by Born and Kuhn [16]. 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 reemit 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

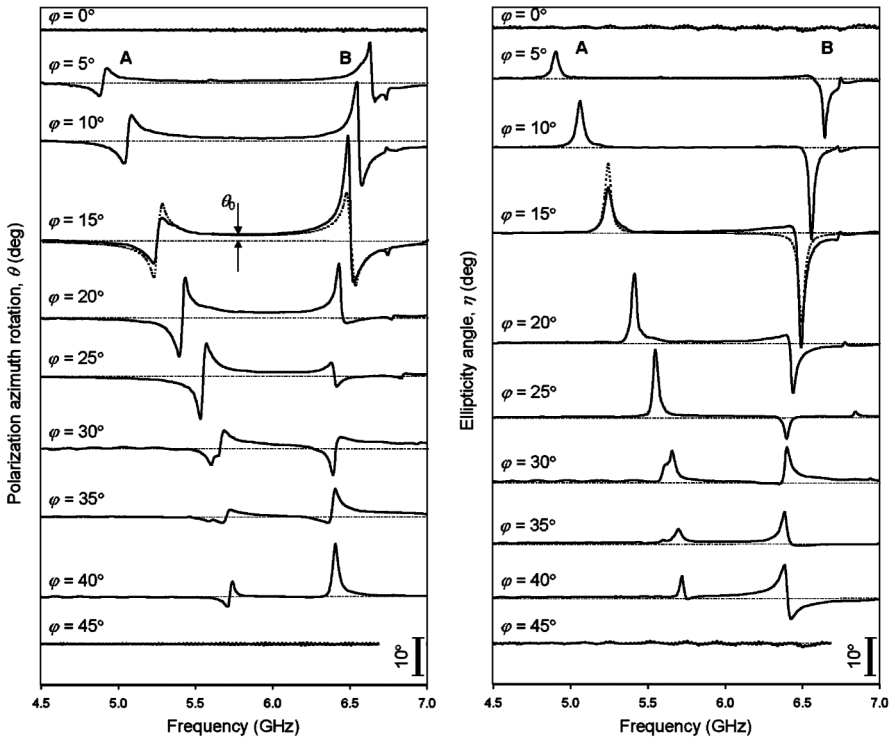

FIG. 3. Frequency dependencies of polarization azimuth rotation θ (left column) and ellipticity, in terms of the ellipticity angle η (right column) that a linearly polarized wave would acquire upon transmission through a sinistral bilayered chiral structure for various twist angles φ. Vertical scale is shown in the right bottom corners. Dotted line at $\varphi = 15°$ shows the frequency dispersion of the effect as predicted by the Born-Kuhn model.

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 reemitted 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 $\binom{\theta}{\eta} \propto \binom{\mathrm{Re}}{\mathrm{Im}} \times \frac{\xi\nu^2}{(\nu_0^2+\xi-i\gamma\nu-\nu^2)^2-\xi^2}$, where ν_0 and γ are the resonant frequency and damping parameter of individual oscillator and ξ is the Hooke coefficient of elastic force between the oscillators [16]. Here, ν_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 Borh-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 ξ. Indeed, in our experiments the peak separation is at maximum for small φ when the electromagnetic interaction between rosettes is strong. It decreases with increasing φ 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 bilayered structure as may be see in Fig. 3, where theoretical dispersion curves are plotted as a dotted line for $\varphi = 15°$ ($\nu_0 = 5.247$ GHz, $\gamma = 4.4 \times 10^7$ s^{-1}, $\xi = 4.6 \times 10^{19}$ s^{-2}). Importantly, the model indicates strong coupling between rosettes in the bilayered system, where the 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 the 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 mechanisms of enantiomericly sensitive interactions with the bilayered 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 directions, creating enantiomericly 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 enantiomericly sensitive scattering that creates a complex frequency dispersion of the effect at various φ.

In summary, we have observed exceptionally strong gyrotropy provided by electromagnetic coupling in a bilayered chiral structure made of physically separated metallic patterns. We expect that optical activity of this nature will also be displayed by appropriately scaled subwavelength nanostructures and suggest that bilayered chiral structures are promising bulk media for the search of negative refraction due to chirality, although losses can be a problem in the optical part of the spectrum. We also expect the structure to show resonant polarization plane rotation in reflection, similar to sculptured thin films [8].

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