Giant optical gyrotropy due to electromagnetic coupling

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The authors demonstrate a chiral photonic metamaterial with chirality provided by electromagnetic coupling between mutually twisted unconnected layers. In the visible and near-IR spectral ranges, the material exhibits polarization rotatory power of up to $2500°/\mathrm{mm}$ and shows relatively low losses and negligible circular dichroism, making it a promising candidate for the development of chiral negative index media. © *2007 American Institute of Physics*. [DOI: 10.1063/1.2745203]

Artificial gyrotropic chiral media that rotate the polarization state of electromagnetic radiation are known from the pioneering work of Bose, who in 1898, using bundles of twisted fiber jute, a spark millimeter wave source, and a wire grid polarimeter, observed that "the twisted structure (of jute) produces an optical twist of the plane of polarization."[1] Artificial materials exhibiting strong gyrotropy recently have started to attract a lot of attention as potential candidates for achieving negative refraction and for applications in microwave and optoelectronic devices.[2–10] In particular, films of mesoscopic sculptured helical pillars manufactured by self-assembly have been investigated as a promising photonic chiral material.[11] Recently microwave gyrotropy of a layered structure with electromagnetic coupling between unconnected and mutually twisted planar elements was demonstrated.[12] This design has the important advantage of allowing its fabrication using planar technologies. Moreover, in the microwave part of the spectrum, the structure has shown not only exceptionally strong gyrotropy but also a signature of negative refraction for circularly polarized waves. In fact, this structure is a chiral version of a metamaterial consisting of pairs of parallel rods in which optical negative refraction has been observed.[13]

In this letter, we demonstrate a chiral *photonic* metamaterial based on the principle of chiral electromagnetic coupling between unconnected layers showing strong rotatory power in the optical and near-IR parts of the spectrum. The metamaterial is formed by a double-periodic planar array of bilayered nanoscale three-dimensional chiral metamolecules [Fig. 1(a)]. It was manufactured by electron-beam lithography. After the first layer of rosettes was formed on a 500 μm silica substrate, a 50 nm spacer layer of silicon nitride was deposited by plasma enhanced chemical vapor deposition to cover it. Aluminum strips in individual rosettes had width and thickness of 50 nm, while the overall size of the template structure was $700\times 700\ \mathrm{nm}^2$. The sample area of $250\times 250\ \mu\mathrm{m}^2$ was covered with 10^5 rosettes arranged in a regular square grid. The process was then repeated to manufacture the second layer of rosettes on the silicon nitride spacer. The rosettes in two layers were coaxial but mutually twisted by 15°. The main achievement in manufacturing the bilayered rosette structure was in accurate aligning of the layers, which across the whole sample should have been better than a small fraction of the individual rosette size. Otherwise the chiral electromagnetic coupling between layers would be distorted and strong birefringence would appear. The alignment procedure included die-by-die registration using special gold alignment dies highly visible in electron scattering and placed on the sample with high density. Great care was also taken to avoid misalignment due to shadow evaporation. An alignment accuracy better than 10 nm was achieved across the whole sample. We have also manufactured a reference sample with only a single layer of rosettes [Fig. 1(c)].

The polarization and transmission properties of the metamaterial structures were studied for normally incident linearly polarized light using a supercontinuum laser source [Fig. 2(a)]. The bilayered metal structure shows somewhat stronger absorption than the single-layered structure. Both are transparent in the visible with absorption increasing at longer wavelengths. The bilayered chiral structure shows a broad absorption resonance at 1.55 μm, while the strongest absorption for the reference single-layered structure is seen at 1.45 μm.

Although the design of both metamaterial structures features fourfold rotational symmetry, which prohibits anisotropy at normal incidence, in reality small manufacturing asymmetries lead to residual anisotropy of the structure that manifests itself as a small birefringence for the normally incident beam: accurate measurements of the sample gyrotropy shall involve separating the polarization effect due to the residual anisotropy from the effect of chirality. For that purpose all polarization measurements were performed for different orientations of the incident polarization Φ_0. Single transverse mode semiconductor laser sources at 660, 980, and 1310 nm and an automated full Stokes parameter rotating wave-plate polarimeter were used. To describe the results of polarimetric measurements, we used the degree of ellipticity η and polarization azimuth Φ, calculated from the Stokes parameter data: $\eta=\frac{1}{2}\arcsin(S_3/S_0)$ and $\Phi=\frac{1}{2}\arctan(S_2/S_1)$. Here S_i are Stokes parameters of the wave transmitted through the structure.

At all wavelengths upon transmitting through the metamaterial sample, the polarization azimuth Φ_0 of the initially linearly polarized light rotates, as illustrated in Fig. 2(b). These data allow unambiguous separation of the effects of anisotropy and gyrotropy. Indeed, in terms of effective medium parameters, propagation of light through a lossy me-

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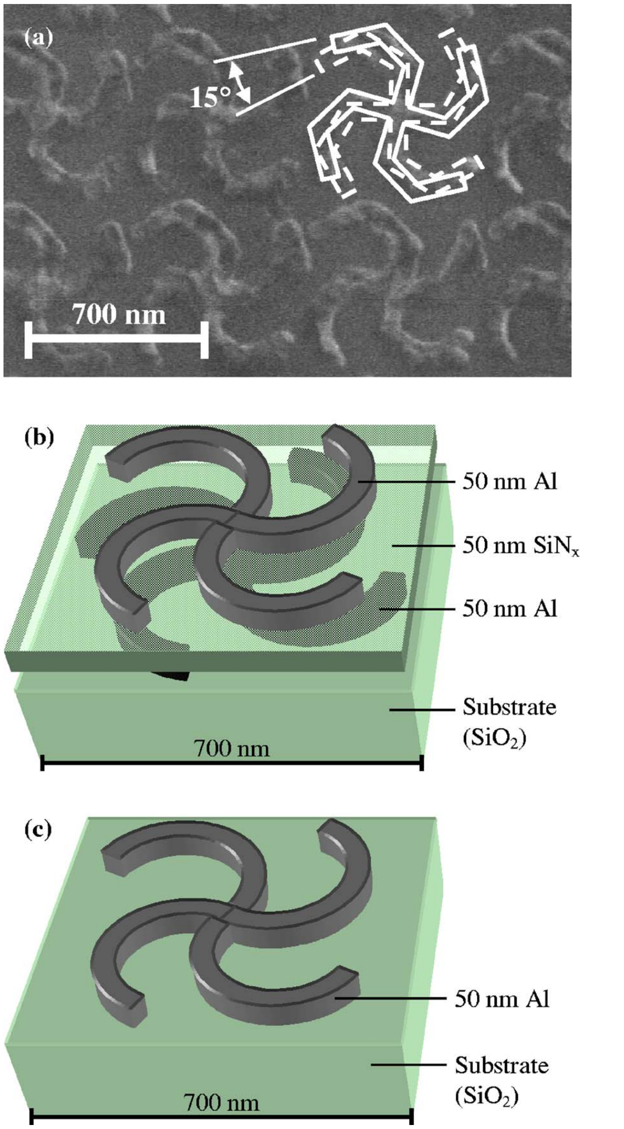

FIG. 1. (Color online) (a) SEM micrograph of the bilayered metamaterial. Note that the rosettes in two layers are mutually twisted and that their centers are aligned with high precision. (b) Schematics of the bilayered metamaterial's square unit cell. (c) Schematics of the monolayered reference metamaterial's square unit cell.

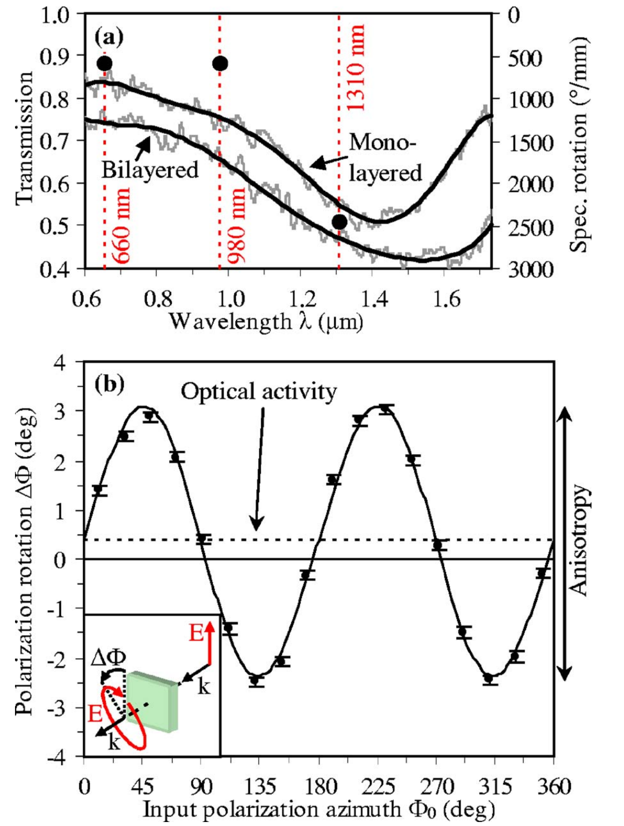

FIG. 2. (Color online) (a) Transmission spectra of the bilayered and reference monolayered metamaterial structures. Red dashed lines indicate wavelengths for which the optical activity has been measured and black dots mark the specific rotation of the bilayered metamaterial at these wavelengths. (b) Polarization rotation induced by the bilayered metamaterial at 1310 nm as a function of azimuth of the incident linearly polarized light. The solid line is a numerical fit. The dashed line indicates the true level of the optical activity component in the rotation.

dium along an anisotropic and gyrotropic direction can be introduced by the following effective constitutive equation: $D_i=\epsilon_{ij}E_j+ik_n\Gamma_{ijn}E_j$. The response of the medium is fully determined by a set of two complex parameters and one real parameter largely responsible for gyrotropy, anisotropy, and losses:[14] $\Gamma^a e_{ijn}=\Gamma_{ijn}$, $\Omega_1^L=(\epsilon_{11}-\epsilon_{22})/n$, and $\mathrm{Im}\{4n+(\epsilon_{11}+\epsilon_{22}-2n^2)/n\}$. The magnitude of the oscillation is largely controlled by residual anisotropy, while the offset of the median of the oscillating curve from the zero level is largely due to the presence of chirality. At 1310 nm the best fit was obtained for $\Gamma^a=(3.7+0.3i)$ nm with an uncertainty of about 0.3 nm in both real and imaginary parts. The measurements of gyrotropy performed for light propagation through the sample in opposite directions gave results identical within the experimental tolerance. The imaginary part of Γ^a, which is responsible for circular dichroism and ellipticity of the transmitted light, is about 12 times smaller than its real part and is, in fact, below our reliable experimental resolution at all measured wavelengths, indicating that circular dichroism of the sample is small.

At 660 and 980 nm optical activity of the bilayered structure seems to depend weakly on the wavelength (Table I). However, close to the resonance wavelength, its value increases by four times reaching 0.37° at 1310 nm. This corresponds to a giant specific rotary power of 2500°/mm close to the absorption resonance and 600°/mm away from it. The level of optical activity exhibited by the single layer metamaterial at 660 and 980 nm falls below the resolution of our measuring technique. Nevertheless some small rotation of about −0.03° was seen at 1310 nm. Therefore the optical rotary power of the bilayered twisted structure is not only much larger than the rotation caused by the monolayered structure, but it is also of the opposite sign, thus indicating a different mechanism of rotation. It is interesting to examine the results within the framework of the Born-Kuhn model of optical activity that well describes chirality due to electromagnetic coupling.[12,14] In the Born-Kuhn model, $\mathrm{Re}(\Gamma^a)\approx\beta D(\varepsilon-1)$, where β is a figure of merit of coupling be-

TABLE I. Polarization rotation $\Delta\Phi$ exhibited by bilayered and monolayered metamaterial structures.

Structure	660 nm	980 nm	1310 nm
Bilayered	0.09° (600°/mm)	0.09° (600°/mm)	0.37° (2500°/mm)
Monolayered[a]	<0.01°	<0.01°	≃−0.03°

[a]<0.01° refers to the magnitude $|\Delta\Phi|<0.01°$.

tween the rosettes in the bilayered system and $D=150$ nm is the thickness of the "molecule." From here, assuming that the effective ϵ of the structure is close to that of silica ($\epsilon \sim 2.1$), the estimated value of coupling is $\beta=0.02$, i.e., the energy of the chiral interaction between rosettes amounts to about 2% of the energy of interaction between individual rosettes and the optical field.

The observed rotation of the bilayered structure (which has a thickness D of only about 1/9 of the wavelength) is huge in terms of specific rotatory power. Indeed, at 1310 nm, the specific rotary power of the bilayered structure is three orders of magnitude stronger than that of quartz. The polarization effect in our structure has a different origin to the polarization rotation in sculptured films. In sculptured films, as in cholesteric liquid crystals, strong specific rotation is observed at the Bragg diffraction regime when the pitch of the screw is equal to the wavelength of light and light scattering is strong. In the metamaterial reported here, the resonance seems to appear when the wavelength is about ten times larger than the thickness of the structure and light scattering is negligible.

Finally, our results may be compared with recent observations of chiral effects in bilayered metamaterial consisting of two layers of coaxial, mutually untwisted gammadions, where a chiral effect is archived by using gammadions of different sizes in two layers.[15] The metamaterials reported in Ref. 15 show much stronger absorption, and circular dichroism seems to be the main manifestation of chirality. This is in contrast with the metamaterial reported here where losses are relatively low, circular dichroism is undetectable, and optical polarization rotation is the main manifestation of chirality. Such a combination of low loss and high rotatory power is exactly what is needed for demonstrating negative refraction due to chirality.[4] The optical data presented here and in Ref. 15 as well as microwave results reported in Ref. 12 show a much smaller polarization effect in a single-layered rosette structure than in a double-layered structure. This is in partial contradiction with the interpretation of data reported in Ref. 16 that claims strong optical activity of a single array of gammadions at normal incidence. Such a discrepancy may have it roots in the presence of a ferroelectric chromium layer in the material reported in Ref. 16.

In conclusion, we have demonstrated a chiral photonic metamaterial exhibiting strong rotatory power and negligible circular dichroism in the visible to near-IR spectral range.

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