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Circular dichroism of planar chiral magnetic metamaterials

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We propose, fabricate, and study a double-layer chiral planar metamaterial that exhibits pronounced circular dichroism at near-infrared wavelengths. The antisymmetric oscillation modes of the two coupled layers allow local magnetic-dipole moments and enhanced polarization effects compared with similar single-layer systems where only electric-dipole moments occur. Experiment and rigorous theoretical calculations are in good agreement. © 2007 Optical Society of America

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Planar chiral metallic structures have recently been introduced.[1–4] These structures are composed of periodically arranged metallic building blocks that are chiral, i.e., that cannot be brought into congruence with their mirror image (the so-called enantiomer) unless lifted off the substrate. Thus, the optical response of these structures is different for right-handed circularly polarized (rcp) and left-handed circularly polarized (lcp) incident light, respectively. Recent work[3,4] has shown that the polarization behavior neither violates reciprocity nor time-reversal symmetry, in contrast to earlier claims.[2] This means that the polarization behavior is roughly similar to optical activity (though clearly not isotropic) but is distinct from the Faraday effect. Intuitively, this behavior is directly connected to the planar nature of these structures: while a three-dimensional spiral or helix keeps its handedness when looked at from the other side, a planar chiral structure obviously changes its handedness.

The optics of these planar chiral structures could be understood in terms of plasmon modes leading to local electric-dipole moments oscillating in a particular manner. Here, we discuss a novel structure where two metal layers are separated by a dielectric spacer [Fig. 1(a)]. As a result, strong magnetic-dipole moments (or, equivalently, electric quadrupoles) can occur as well. We show that the resulting polarization effects are yet stronger than for single-layer structures. Our spectroscopic experiments are in good agreement with rigorous theoretical calculations.

In our samples, the chiral building blocks or "photonic atoms" are arranged in a square lattice with lattice constant $a=340$ nm. We emphasize that the operation wavelength λ is large with respect to the lattice constant such that our structures can be viewed as effective materials in the same spirit as, e.g., magnetic[5,6] or negative-index photonic metamaterials.[7–9] Precisely, we have $\lambda/a>2$ ($\lambda/a=2$ is the fundamental Bragg condition), whereas previous work had $\lambda/a\approx 0.16$ (Refs. 1 and 2) and $\lambda/a\approx 1$ (Ref. 3), respectively. The photonic atoms [see Fig. 1(a)] consist of a sandwich of 25 nm gold, 25 nm magnesium fluoride (MgF_2), and 25 nm gold on a glass substrate, coated with a 5 nm thin film of indium tin oxide to prevent charging effects during electron-beam lithography. All layers are produced via electron-beam evaporation. The lateral patterning uses standard electron-beam lithography and a lift-off procedure. The footprint of all samples is $100\ \mu\text{m}\times 100\ \mu\text{m}$. Typical oblique-view electron micrographs of the samples fabricated along these lines are shown as insets in Figs. 2 and 3. The minimum feature sizes are of the order of 50 nm. We have also fabricated gammadion structures with angles other than 90° between the long and the short arms (e.g., 45°). These structures show smaller polarization effects and are not discussed here. Indeed, from symmetry it is obvious that structures with 0° and 180° angles would not exhibit any circular dichroism (CD) at all.

For the optical characterization we perform broadband linear-optical transmittance spectroscopy with circularly polarized incident light, enabled by a 100 W halogen lamp and a Glan–Thomson polarizer followed by a superachromatic quarter-wave plate (Bernhard Halle RSU 2.4.15, 600–2700 nm wavelength). In this fashion, we essentially measure effects due to the imaginary part of the refractive index, while previous work has focused on differences in the real part of the refractive index by measuring rotation angles or polarization states.[1–3] Our home-built setup allows for spectroscopy on small samples at a 5° half-opening angle of the incident light.

Spectroscopic results of the double-layer structures are shown in the left-hand side of Fig. 2. The spectra reveal four different resonances, the physics of which will be discussed below. The transmittances T_{rcp} and

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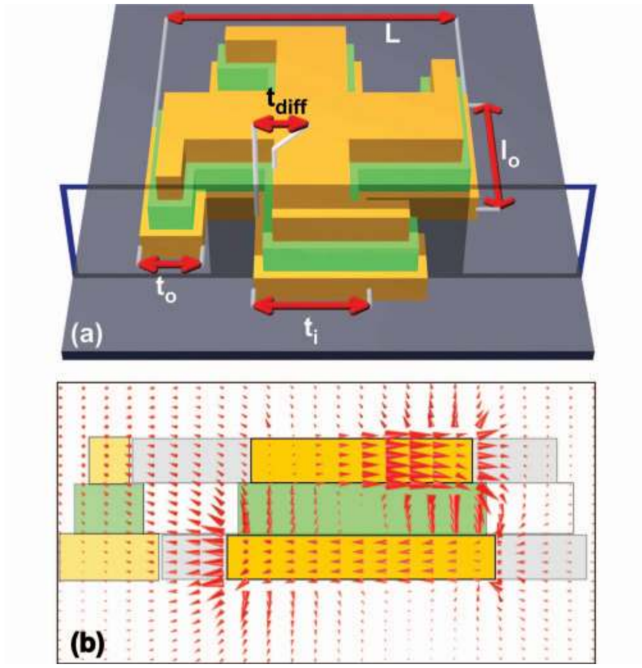

Fig. 1. (a) Scheme of the double-layer magnetic metamaterial. The geometrical parameters are indicated and given by $L=274$ nm, $t_i=90$ nm, $l_o=135$ nm, $t_o=50$ nm, and $t_{\text{diff}}=15$ nm (b) Snapshot of the $\vec{E}$-field at 0.86 μm wavelength for lcp incident light. The cutting plane is indicated in (a).

T_{lcp} are different for rcp and lcp incident light. The differences in transmittances $\Delta T_{\text{rcp,lcp}}$, multiplied by a factor of ten, are shown as green curves in Fig. 2. Obviously, the effects are on the level of about ±6%. The spectra for the enantiomeric structures (second row) simply reveal a reversed sign of the transmittance difference—as expected from symmetry. This observation rules out a major influence of unintentional linear birefringence. To allow a direct comparison with corresponding single-layer structures, we have also fabricated identical lateral structures, however, with just a single 50 nm thin gold layer. Obviously, the CD spectra in Fig. 3, which are arranged just as in Fig. 2, show strongly reduced effects, if any. Also, only two resonances occur. Spectra for a single 25 nm thin gold layer (not shown) reveal no significant circular dichroism. This comparison between double-layer (Fig. 2) and single-layer (Fig. 3) samples shows that the differences are due to design rather than due to the amount of metal.

The physical origin of this difference lies in the fact that the coupling in the double-layer structure allows for symmetric and antisymmetric charge-oscillation eigenmodes. The latter can be interpreted as part of a ring current [see Fig. 1(b)], leading to a local magnetic-dipole moment. This aspect has recently also been the key for realizing magnetic[5] and negative-index metamaterials.[7–9] The antisymmetric oscillation mode gives the combined plasmon mode a certain twist into the propagation direction of light, hence increasing the circular dichroism. Clearly, this twist oscillates with the frequency of light and occurs in linear optics already. The long and short arms of the gammadion give rise to two pairs of resonances. These four resonances are visible as transmittance minima in Fig. 2, whereas only two resonances appear in Fig. 3.

To support this interpretation, we have obtained rigorous numerical solutions of the three-dimensional vector Maxwell equations based on a finite-element frequency-domain approach implemented in a commercial software package (Comsol Multiphysics). The geometrical parameters are taken from the experiment (see above), the refractive index of the glass substrate is taken as 1.5, that of magnesium fluoride as 1.38, and gold is described as a Drude metal with plasma angular frequency $2\pi \times 2081$ THz and collision frequency (damping) $2\pi \times 35$ THz. The calculated spectra are shown in the right-hand columns of Figs. 2 and 3 for the double- and the single-layer samples, respectively. The overall qualitative agreement is very good. In particular, four resonances occur for the double-layer structure, whereas only two resonances occur for the single-layer structure. Also, the magnitude of the circular dichroism is nicely reproduced. Furthermore, the spectral positions of all resonances agree with experiment. A snapshot of the electric field distribution of the antisymmetric mode of the short arms of the double-layer structures is shown in Fig. 1(b).

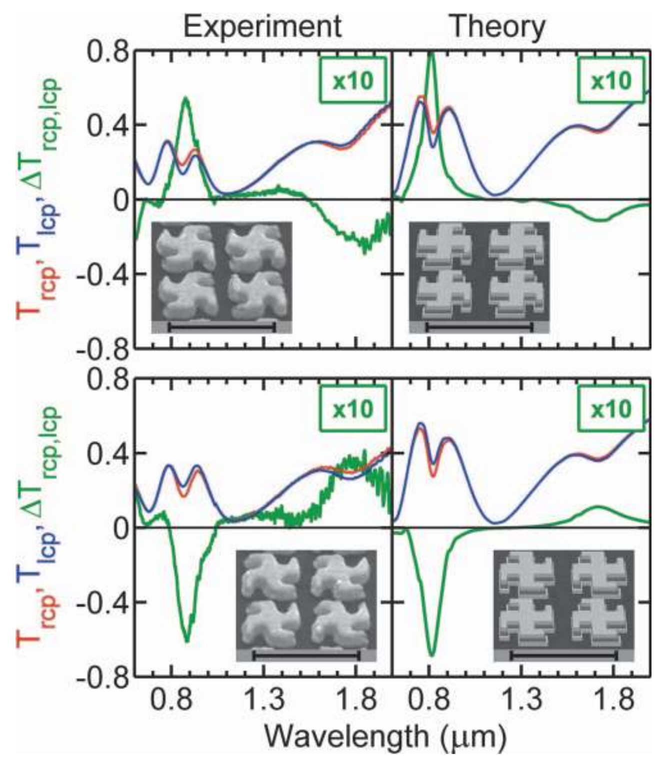

Fig. 2. Normal-incidence linear-optical transmittance spectra of the double-layer chiral metamaterial. The first (second) row corresponds to right-handed (left-handed) gammadions. The left column is experimental, and the right column is calculated. Transmittance spectra are shown for left-circular incident polarization of light (blue) as well as for right-circular incident polarization (red). The difference between the two is multiplied by a factor of ten and depicted as the green curves. The corresponding oblique-view electron micrographs and the geometry used in our calculations, respectively, are shown as insets. The scale bar is 500 nm.

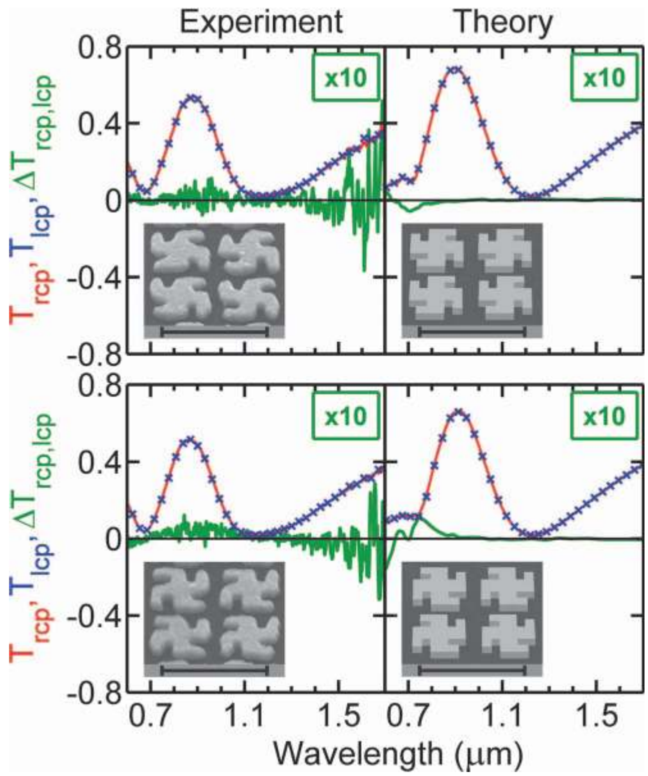

Fig. 3. Normal-incidence linear-optical transmittance spectra of the single-layer chiral metamaterial. The representation is identical to that of Fig. 2. Note that the thickness of the single gold layer (50 nm) is identical to the sum of the thicknesses of the two gold layers in Fig. 2.

Finally, we relate our optical results to recent microwave results on structures with two layers of gammadions,[10] however, with a larger spacing between the two layers compared with the wavelength of electromagnetic radiation. The authors of Ref. 10 find no circular dichroism for identical orientation of the two gammadions (but strong effects if the gammadions include an angle). This result does not contradict ours. As discussed previously,[5] the choice of the spacer thickness is a compromise: for zero spacer thickness (Fig. 3), the two layers merge into one and weak CD results. For too thick spacers (which is the case for Ref. 10), the coupling between the two layers via their evanescent fields becomes too weak. Again, weak CD results. For intermediate spacer thicknesses (Fig. 2), strong coupling results and the antisymmetric mode of the coupled system acquires appreciable oscillator strength in the optical spectrum, leading to pronounced CD. This compromise is closely similar to that for obtaining a pronounced magnetic response in magnetic metamaterials, for which a systematic study on the influence of the relative spacer thickness has been published by our group.[5]

In conclusion, we have realized a novel double-layer chiral planar metamaterial that exhibits larger circular dichroism than similar single-layer systems. This effect is due to the antisymmetric eigenmodes of the coupled system that lead to a certain twist into the propagation direction—to some extent mimicking three-dimensional chirality. Antisymmetric eigenmodes are directly connected to local magnetic-dipole moments. Indeed, we find that the (antisymmetric) magnetic-dipole resonances of our metamaterial structure lead to much larger circular dichroism than the (symmetric) electric-dipole resonances. Our experiments at near-infrared frequencies are in good agreement with corresponding numerical solutions of the three-dimensional vector Maxwell equations.

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