Strong optical activity from twisted-cross photonic metamaterials

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Following a recent theoretical suggestion and microwave experiments, we fabricate photonic metamaterials composed of pairs of twisted gold crosses using two successive electron-beam-lithography steps and intermediate planarization via a spin-on dielectric. The resulting two effective resonances of the coupled system lie in the 1–2 μm wavelength regime and exhibit pronounced circular dichroism, while the circular polarization conversion is very small. In between the two resonances, we find a fairly broad spectral regime with strong optical activity, i.e., with a pure rotation of incident linear polarization. The measured optical transmittance spectra agree well with theory. © 2009 Optical Society of America

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Throughout the past five years, metamaterials have brought magnetism to the field of optics and photonics [1,2]. For example, this allows for obtaining very large chiral optical effects [3], which result specifically from the interplay of electric/magnetic dipoles and the magnetic/electric component of the incident light field [4]. As a result, a negative phase velocity for circular polarization of light may result even if electric permittivity and magnetic permeability are positive at the same time [5,6]. In addition, an incident linear polarization can experience a rotation per unit propagation length that is orders of magnitude larger than in, e.g., a solution of chiral sugar molecules.

At optical frequencies, several structures exhibiting strong optical activity and/or circular dichroism have been realized [7–9]. Importantly, chirality is a three-dimensional phenomenon, and, hence, the chiral effects from planar single-layer structures excited under normal incidence of light are extremely weak [9] (compare Figs. 2 and 3 therein). In this Letter, for the first time, to our knowledge, we investigate a chiral double-layer design [10] at optical frequencies, the individual layers of which are simple metal crosses. Unlike previous structures [11,12], ours do not reveal planar chirality [7]. In fact, our structures can be viewed as the chiral version of the well-known double-cut-wire magnetic metamaterial approach [13,14]. We choose crosses rather than cut wires in order to avoid undesired linear birefringence that otherwise occurs for few-layer structures. We note in passing that related nonchiral complementary double-cross structures have recently been discussed [15].

The twist angle between the crosses shown in Fig. 1 is 22.5°, as neither 0° [9] nor 45° lead to a chiral unit cell. Hence, we have chosen an intermediate value. Fabrication of this overall nonplanar structure (see Fig. 1) obviously requires advanced nanofabrication tools. Here, we follow the approach of [16]. We use electron-beam lithography for the first layer, subsequent planarization via a 500-nm-thick commercially available spin-on dielectric (IC1-200, Futurrex, Inc.) and thinning via reactive-ion etching (SF_6, Plasmalab80Plus, Oxford Instruments Plasma Technology), followed by a second step of electron-beam lithography, aligned relative to the first layer. Variations of this approach have recently been published by another group [17]. Electron micrographs of structures that we have fabricated are also shown in Fig. 1. The arms of the gold double crosses have a thick-

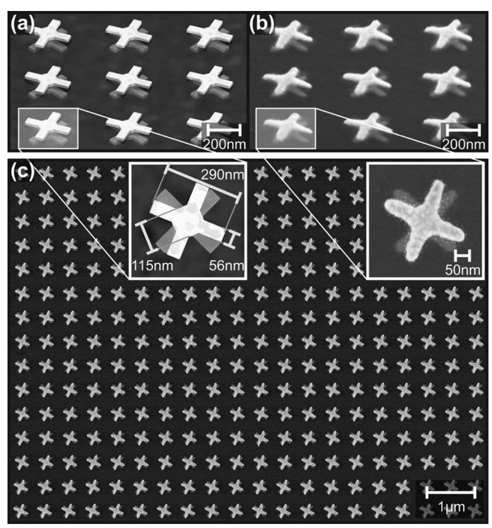

Fig. 1. (a) Scheme of the chiral metamaterial structure composed of right-handed twisted gold crosses. (b) Oblique-view electron micrograph of a fabricated sample and (c) normal-view large-area electron micrograph of a right-handed structure. The insets show close-ups.

ness of 25 nm, a full width of 56 nm, and a length of 315 nm. The center of the crosses is somewhat overexposed owing to the proximity effect. The double crosses are arranged on a simple square lattice with lattice constant $a=500$ nm. It becomes obvious from Fig. 1 that the sample quality is very high. In particular, the in-plane misalignment between the two crosses in each pair is smaller than 10 nm over the entire sample footprint of 100 μm $\times$ 100 μm. All samples are fabricated on a glass substrate covered with a 5 nm thin film of indium-tin-oxide (ITO). Both, left- and right-handed twisted-cross structures have been fabricated and characterized optically.

For optical characterization, we employ normal-incidence transmittance spectroscopy with either circular or linear polarization of the incident light. Our dedicated setup [9] based on super-achromatic quarter-wave plates (Bernhard Halle RSU 2.4.15, 600–2700 nm wavelength) further allows for analyzing the emerging polarization of light, i.e., the conversion of circular as well as of linear incident polarization into the corresponding orthogonal polarization state. In each case, we present results for left-handed (lh) and for right-handed (rh) twisted-cross structures. The spectra shown in Fig. 2 reveal two distinct resonances. In close analogy to our previous discussion [9,13], these two resonances correspond to the two effective modes of the otherwise degenerate fundamental electric-dipole Mie resonances of the two coupled crosses. Within the effective-medium limit, it is clear that a single cross has a polarization-independent optical response.

First, by comparing the results for incident left-handed circular polarization (LCP) and right-handed circular polarization (RCP) on left- and right-handed

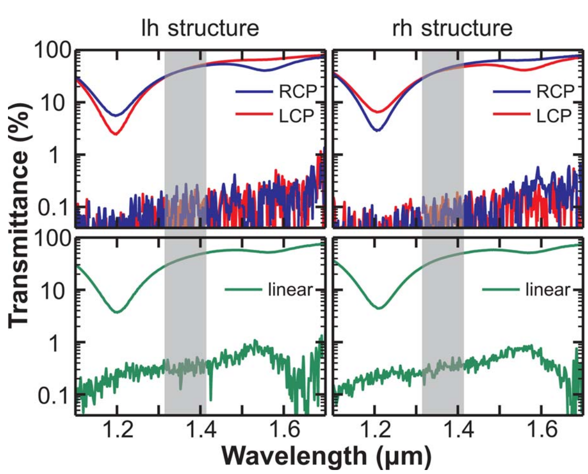

Fig. 2. (Color online) Measured normal-incidence transmittance and conversion spectra (logarithmic scale) of the twisted-cross structure shown in Fig. 1. The left column is for an lh structure, the right column for an rh structure. Transmittance for LCP and RCP is plotted in gray (red online) and black (blue online), respectively. The conversion into the other circular polarization is extremely small and, hence, shown by the same color. In contrast, we do find significant conversion of linear polarization (second row) in between the two resonances, where the intensity transmittances for the two incident circular polarizations are closely similar. This regime highlighted in gray delivers pure and large optical activity.

metamaterial structures in Fig. 2, it becomes obvious that reproducibility is excellent and that fundamental symmetries are obeyed. Second, intensity conversion of incident LCP and RCP is smaller than 10^{-3}, a value that corresponds to our experimental measurement limit. Hence, LCP and RCP are the eigenpolarizations of our structures throughout the entire spectral range shown in contrast to other recently presented chiral metamaterials [17] that have exhibited wavelength-dependent elliptical eigenpolarizations. Third, as desired for a chiral material, coupling to the two effective resonances is strongly dependent on the handedness of the incident light; i.e., we find huge circular dichroism. Note that, in between the two resonances (see gray areas), the amplitudes of the two circular transmittances are very nearly identical. Thus incident linearly polarized light is expected to remain linearly polarized, but it is rotated because of the different refractive indices n_{LCP} and n_{RCP} for circular polarization. Indeed, the spectra for linear polarization of the incident light in Fig. 2 show significant conversion in this regime. The measured conversion of 2×10^{-3} at 1.36 μm wavelength is compatible with a rotation of the linear polarization axis of 4.0° for just $l=87.5$ nm total thickness of the metamaterial. As usual, the polarization rotation angle φ due to propagation over length l at free-space wavelength λ is given by

$$\varphi=(n_{\mathrm{LCP}}-n_{\mathrm{RCP}})\frac{\pi}{\lambda}l, \tag{1}$$

which leads us to estimate $|n_{\mathrm{LCP}}-n_{\mathrm{RCP}}|\approx 0.35$.

To support our experimental findings, we compare with theoretical modeling using a finite-element frequency-domain approach provided by the commercially available computer program Comsol MultiPhysics. We have found consistent results (not shown) using the finite-integral time-domain approach provided by CST MicroWave Studio. As usual, the gold is modeled by the free-electron Drude model with plasma frequency $\omega_{\mathrm{pl}}=2\pi\times2159$ THz and collision frequency $\omega_{\mathrm{coll}}=2\pi\times25$ THz plus a background dielectric constant of $\epsilon_b=9.07$. The refractive indices of the glass substrate and the spin-on dielectric are taken as 1.45 and 1.41, respectively, and the thin ITO film is neglected. The lateral geometrical parameters are shown in Fig. 1(a) and the insets in Fig. 1(c); the gold thickness is 25 nm, and that of the spacer layer is 37.5 nm. The calculated results in Fig. 3 are presented just like the experiment shown in Fig. 2. Obviously, the overall agreement is very good in all aspects discussed above. In particular, we find very little conversion of circular polarization and significant conversion of linear polarization corresponding to optical activity. From the calculations (see Fig. 4), we deduce a maximum rotation angle $\varphi=4.0°$ at $\lambda=1.36$ μm wavelength with a tangent of the ellipticity angle [18], e, smaller than 1% (e is the ratio between the semiminor and the semimajor axis of the polarization ellipse, and hence a number between −1 and +1, $e=0$ corresponds to linear polarization).

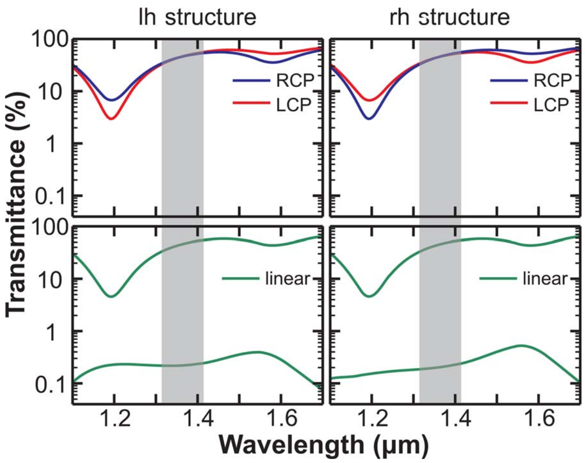

Fig. 3. (Color online) Calculated normal-incidence transmittance and conversion spectra of the twisted-cross structure shown in Fig. 1(a). The conversions for LCP and RCP are below 5×10^{-5}.

Finally, we have also applied the chiral effective-parameter retrieval discussed in [19] to our structures (not depicted). This leads to refractive indices for, e.g., the right-handed structure of $n_{RCP}=2.33$ and $n_{LCP}=1.99$ at $\lambda=1.36$ μm, hence leading to $n_{LCP}-n_{RCP}=-0.34$, which is consistent with our above estimates. The (real parts of the) refractive indices stay positive throughout the entire spectral range, unlike their microwave counterparts [10]. Yet, as usual, effective-parameter retrieval for metamaterial structures even thinner than one lattice constant should be taken with a grain of salt.

In conclusion, we have fabricated and characterized three-dimensional photonic metamaterials composed of pairs of twisted gold crosses. In a spectral region around 1.36 μm wavelength, we find a rotation of an incident linear polarization of up to 4° with little polarization conversion. This rotation for a total metamaterial thickness of just 87.5 nm (45,714°/mm) can be translated into a difference of the refractive indices for the two circular polarizations as large as $|n_{RCP}-n_{LCP}|\approx0.35$.

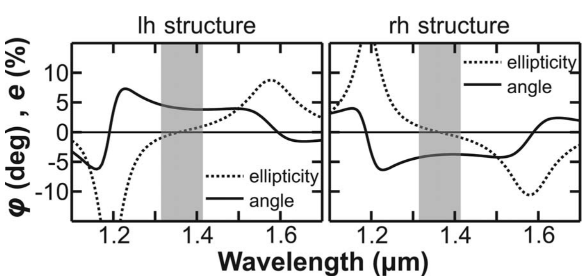

Fig. 4. Calculated rotation angle φ (solid) and tangent of the ellipticity angle e (dashed) of lh and rh structures for linearly polarized incident light.

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