Elliptical dichroism: operating principle of planar chiral metamaterials

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We employ a homogenization technique based on the Lorentz electronic theory to show that planar chiral structures (PCSs) can be described by an effective dielectric tensor similar to that of biaxial elliptically dichroic crystals. Such a crystal is shown to behave like a PCS insofar as it exhibits its characteristic optical properties, namely, co-rotating elliptical polarization eigenstates and asymmetric, direction-dependent transmission for left/right-handed incident wave polarization.

Metamaterials show promise for a wide range of unusual physical phenomena rare or absent in nature.1,2 Following a pioneering work by the group of N. Zheludev in 2006,3 planar chiral structures (PCSs) were recently introduced as a distinct class of metamaterials. They consist of planar elements4–6 that have a sense of “twist” and cannot be superimposed with their in-plane mirror image. PCSs are asymmetric in electromagnetic wave propagation for right-handed (RH) vs. left-handed (LH) circularly polarized incident wave. Unlike 3D or bilayer chiral metamaterials4–6,10, which resemble biaxial or gyrotropic media, PCSs change their properties if wave propagation direction is reversed.11 They also differ from Faraday media (also known to have related asymmetry) because PCSs have co-rotating elliptical polarization eigenstates7,12 while in bi-isotropic or Faraday media the eigenstates always come in pairs of RH and LH polarization.

In this Letter, we interpret optical properties of PCSs in terms of elliptical dichroism. The properties of chiral metamaterials are usually investigated on the level of resonant electromagnetic response in an individual element.8 9,10,12,13 This heavily depends on the element shape, a wide variety of which was reported lately.7,8,9,12,13 Although a recent work13 offers a generalized explanation based on polarization-sensitive excitation of electric and magnetic dipoles, such a treatment should still be performed separately for different PCS designs. Thus, there is a need for a “macroscopic” description that would make planar chirality available for studies on an abstract crystallographic level, similar to other optical phenomena like birefringence or optical activity.

The idea of regarding elliptical dichroism as a mechanism behind the optical manifestation of planar chirality is suggested by earlier experimental accounts of circular dichroism in PCSs.2,13 We confirm its existence using the Lorentz-theory homogenization scheme in chiral split-ring (CSR) structures.13,14,15 Moreover, we show that a bulk elliptically dichroic medium exhibits all the characteristic properties of PCS, namely (i) co-rotating elliptical polarization eigenstates, (ii) asymmetric transmission for RH vs. LH circular polarization, and (iii) enantiomeric asymmetry and change of properties for different wave propagation direction.

We begin by considering a CSR unit cell (Fig. 1b). For planar (as opposed to 3D) elements, the electric response is dominant because the magnetic moment is parallel to the wave propagation direction.7,11 Hence, one can arrive at homogenized material properties by considering the response within the framework of the electronic Lorentz theory by using \( \hat{\varepsilon}_{\text{eff}} \cdot \mathbf{E} = \mathbf{E} - (4\pi/\omega) \cdot \mathbf{E} \cdot \langle N \mathbf{v} \rangle \). The electrons in the dielectric substrate are bound and their concentration and velocity \( \langle N \mathbf{v} \rangle \) are position-dependent in a structured material. To derive the effective dielectric tensor \( \hat{\varepsilon}_{\text{eff}} \), one needs to average over the unit cell. If \( d \ll R \), the electrons in the ring can only be mobile in the azimuthal direction so that \( \mathbf{v}_m = R \hat{\phi} \mathbf{e}_\phi \) and \( \hat{\phi} + \gamma \hat{\phi} + \omega_m^2 \hat{\phi} = (e/Rm)_\phi \mathbf{e}_\phi \cdot \mathbf{E}_m \). The electrons in the dielectric substrate are bound and their velocity \( \mathbf{v}_d \) in response to the electric field is position independent. The averaging is then performed as

\[
\langle N \mathbf{v} \rangle = L^{-2} \left( \int_{\text{metal}} N_m \mathbf{v}_m(\phi) \, d^2 \mathbf{r} + \int_{\text{dielectric}} N_d \mathbf{v}_d \, d^2 \mathbf{r} \right).
\]

For the structure shown in Fig. 1 integration in Eq. (1) results in:

\[
\langle N \mathbf{v} \rangle = L^{-2} \left( \int_{\text{metal}} N_m \mathbf{v}_m(\phi) \, d^2 \mathbf{r} + \int_{\text{dielectric}} N_d \mathbf{v}_d \, d^2 \mathbf{r} \right).
\]

Fig. 1. (a) Bilayer (3D) vs. monolayer (planar) chiral metamaterial. (b) Unit cell of a CSR structure.13
For non-magnetic media and normal incidence states can be recovered from its normal refraction tensor rotation of polarization eigenstates then becomes present), planar chirality \[14, 15\]. Otherwise (when planar chirality is

in a complex symmetric tensor \(\hat{\varepsilon}_{\text{eff}}\)

\[
\hat{\varepsilon}_{\text{eff}}(\omega) = \begin{bmatrix}
\varepsilon_x & \varepsilon_{xy} & 0 \\
\varepsilon_{xy} & \varepsilon_y & 0 \\
0 & 0 & \varepsilon_z
\end{bmatrix}.
\]

(2)

Its components depend on dielectric constants of metal \(\varepsilon_m(\omega) = 1 + \omega_0^2/\left(\alpha_0^2 - \omega^2 - i\omega\gamma\right)\) and dielectric \(\varepsilon_d(\omega) = 1 + \Omega_p^2/\left(\Omega_0^2 - \omega^2 - i\omega\Gamma\right)\), as well as on the geometrical parameters, e.g., the angles \(\alpha_{1,2}\) and \(\beta_{1,2}\) (Fig. 1). Fig. 2 shows the dependencies \(\varepsilon_{x,y}(\omega)\). If the split ring is symmetric \((\beta_2 = 0, \alpha_1 = \alpha_2, \text{ or } \beta_1 = \beta_2)\), then \(\varepsilon_y = 0\) and the tensor \(\hat{\varepsilon}\) supports two linearly polarized eigenwaves different in their attenuation (linear dichroism). Such CSRs display no planar chirality \([14, 15]\). Otherwise (when planar chirality is present), \(\varepsilon_y \neq 0\) and the eigenpolarizations are elliptical, (the effective medium is elliptically dichroic). We further rewrite the tensor \(\hat{\varepsilon}_{\text{eff}}\) in axial representation (assuming a monoclinic crystal as a particular case of biaxial media \([17]\)) as

\[
\hat{\varepsilon} = \varepsilon_o + (\varepsilon_e - \varepsilon_o) c \otimes c, \quad c = c' + ic''
\]

where the vectors \(c'\) and \(c''\) determine the optical axes, and \(c \otimes c\) denotes the outer dyadic product \((c \otimes c)_ij = c_ic_j\).

At any fixed frequency, the medium’s polarization eigenstates can be recovered from its normal refraction tensor \(\varepsilon_l\) \([18]\). For non-magnetic media and normal incidence

\[
\varepsilon_l^2 = q^2 \hat{\varepsilon} q = (\hat{\varepsilon} \hat{\varepsilon})^{-1} q^2 \hat{\varepsilon} q = \hat{\varepsilon} q q^\ast,
\]

(4)

where \(q\) is a unit vector normal to the plane and \(q^\ast\) is defined as \((q^\ast)u = q \times u\). The eigenvectors \(H_{1,2}\) of \(\varepsilon_l^2\) represent the polarization states of the field that are preserved as the wave propagates along \(q\). If the polarization is circular or elliptical, the handedness of \(H_j\) is determined by the sign of the product \(q \cdot [\text{Re} H_j \times \text{Im} H_{j'}]\) \([17]\). The condition for co-rotation of polarization eigenstates then becomes

\[
\eta = (q \cdot [\text{Re} H_1 \times \text{Im} H_1]) (q \cdot [\text{Re} H_2 \times \text{Im} H_2]) > 0.
\]

(5)

For anisotropic media without absorption, \(\eta = 0\) (the eigenstates have linear polarization). For bi-isotropic chiral or Faraday media, it can be shown that \(\eta < 0\), so circular or elliptical polarization eigenstates are counter-rotating. However, for \(\hat{\varepsilon}\) of Eq. (3) where \(c' = e_c \cos \phi + e_e \sin \phi\) and \(c'' = e_c \cos \psi + e_e \sin \psi\), Eqs. (4) and (5) result in

\[
\eta = \frac{4k^2 \sin^2(\phi - \psi)}{(1 + k^2)^2 - (\cos 2\phi + k^2 \cos 2\psi)^2} \geq 0.
\]

(6)

Next, we consider the transmission spectra of an elliptically dichroic layer, which can be obtained by constructing the wave evolution operator in a generalized transfer matrix formalism \([18]\). The Lorentzian resonance is assumed to be broad enough so that material dispersion can be neglected. Fig. 3 shows the spectra for the LH/RH circularly polarized incident wave. The transmission of LH vs. RH polarization is clearly asymmetric. For higher frequencies, the spectra become flat. This is caused by the absence of material dispersion, so that spectrally uniform elliptical dichroism results in spectrally uniform planar chiral behavior. Real metamaterials are dispersive and thus elliptically dichroic only in a narrow spectral range close to resonance (see Fig. 3). Hence real PCSs display planar chirality in a resonant manner, in accordance with earlier findings \([4, 13]\).

If the material is replaced with its enantiomeric counterpart \((\phi \rightarrow -\phi, \psi \rightarrow -\psi)\), or if the wave propagates in the opposite direction \((q \rightarrow -q)\), the LH/RH plots are exchanged (Fig. 4). This agrees with experimental results for PCSs \([13]\) and with the Lorentz homogenization scheme.

When an initially circularly polarized wave travels through the layer, the wave is first seen to experience polarization mixing similar to what happens in a birefringent crystal (LH \(\rightarrow\) RH). This is associated with the oscillatory portion in the spectra (Fig. 4). As the wave propagates further, it assumes the polarization matching one of the eigenstates in orientation and both eigenstates in handedness. So, a wave
whose handedness does not match that of the eigenstates undergoes polarization conversion, as reported earlier [7]. This can be understood from the combined action of anisotropy (which creates polarization mixing) and dichroism (which diminishes the circular polarization component not matching the eigenstates in handedness).

The results obtained are complementary to a recent account [13] linking planar chirality with polarization-sensitive excitation of resonant modes in CSRs. There, e.g., LH and RH waves were shown to excite an electric dipole (well coupled to the field) and a magnetic dipole (poorly coupled), respectively. For the latter, the energy is “trapped” in the resonant mode and dissipates, resulting in circular dichroism. The range of PCS designs where dichroism is brought about by this mechanism remains to be determined. It is also interesting to note that in high-$T_c$ "anyon superconductors" circular dichroism was reported [19] yet no sign of Faraday-like non-reciprocity was found [20], similar to PCSs. This striking similarity may have fundamental physical reasons given that such superconductors are known to possess layered geometrical structure.

To summarize, we have shown that bulk media with elliptical dichroism exhibit optical properties characteristic for PCSs. First, the elliptical polarization eigenstates are co-rotating (Fig. 3). Second, transmission for LH/RH-polarized incident wave is asymmetric and is exchanged if the material is replaced with its enantiomeric counterpart (Fig. 3). Finally, an elliptically polarized wave propagating in such a material undergoes circular polarization conversion so as to match the polarization eigenstates. The medium is described solely by a complex symmetric dielectric tensor whose structure is given by homogenization of a split-ring PCS. This way, it is shown that a crystallographic approach is possible in studying PCSs, offering a theoretical starting point for analyzing planar chirality as an optical phenomenon. One can use existing bianisotropic multilayer solvers [21] to efficiently model PCS-based optical devices, with possible applications in polarization-sensitive integrated optics.

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