

Chiral photonic film and flake

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Abstract: We show that chiral photonic flake has potential as a physical effect colorant that contributes both bright interference colors and a characteristic polarization spectrum. An analysis of the polarizing properties of chiral film and flake indicates that the Stokes spectrum s_3 v. λ is suitable for characterization. s_3 is shown to be invariant both to azimuthal rotation of a flake and to incoherent summation of the light from an array of flakes with random orientations. We form chiral photonic flake experimentally by scraping film material from nanoengineered chiral films on glass. Three basic architectures are used, a standard chiral medium that supports a single Bragg resonance, a threaded chiral medium that supports right-handed and left-handed resonances at different wavelengths and a threaded chiral medium that supports two right-handed resonances at different wavelengths. In a separate set of experiments a twist defect is added to each basic structure. Experimental measurements of s_3 spectra from film and flake show the expected signatures of the circular Bragg resonances and of the spectral holes caused by the defects.

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1. Introduction

Physical effect colorants [1, 2], such as optical interference coatings on foils and flake derived from coatings, are finding diverse applications from packaging to security of documents and

bank notes to color-shift paint for automobiles. A basic premise for inhibition of counterfeiting is that substantial technical resources are needed to produce the reflected color and variation with tilt that we see when the coating is illuminated with white light. In this brief article we show that flake can be made from inorganic chiral films that offer similar angle-dependent bright colors and provide characteristic polarization spectra for an additional level of security.

In a parallel study of natural and nanoengineered [3] structurally chiral reflectors we have been inspired by the architectures of scarab beetles. Both structures can be described as a periodic birefringent medium with in-plane principal refractive indices n_2 and n_3 that twist steadily with increasing thickness. In the engineering process, serial bideposition of titanium oxide from an e-beam source on to a computer-controlled tilted glass substrate generates a normal columnar biaxial film [3]. Azimuthal rotation of the substrate at a suitable rate adds corkscrew twisting to the columns [4] and when combined with serial bideposition yields columns with the form of a double-start screw [3]. Left-handed structural chirality contributes a left circularly polarized (LCP) Bragg resonance at $\lambda_{Br} = 2n_{av}P$, where $n_{av} = (n_2 + n_3)/2$ and the dielectric pitch P is the distance over which the fast axis of the birefringent medium twists through an angle of 180° . There is a narrow stopband for LCP light in the transmittance spectrum, and a corresponding bright LCP peak in the reflectance spectrum. Apart from the polarizing properties, and an absence of periodicity with wavenumber, the reflectance spectrum is similar to that from an isotropic narrowband interference filter [5]. In addition, angular analogues of structural perturbations such as chirping of layer thickness (pitch) to increase bandwidth and the introduction of a spacer layer (twist defect) to cause a spectral hole to appear at the center of the stopband have been found in beetles.

The surface of a beetle is usually textured, in some cases with an hexagonal lattice of epithelial cells [3]. At the cell boundaries the fibrous structures that produce the chirality appear to thread through each other, giving rise to the concept of threaded chiral media. Here we progress the idea to the field of nanoengineered chiral structures, in order to introduce additional spectral signatures to the polarized reflected light. In particular we introduce a threaded chiral medium in which a structure A with pitch $P_A = \lambda_{BrA}/2n_{av}$ weaves through a second structure B of the same material but with different pitch $P_B = \lambda_{BrB}/2n_{av}$ and the same or opposite handedness. Our aim is to make chiral photonic flake from such films with or without a twist defect and show that we can detect the characteristic polarization spectra.

2. Polarizing properties of chiral films

In this section we consider characteristic spectral reflection from chiral films illuminated by polarized light and by unpolarized light. The properties of a chiral film illuminated by polarized light can be expressed in terms of a set of amplitude reflection coefficients, r_{RR} , $r_{RL} = r_{LR}$ and r_{LL} . Here we are using RCP and LCP basis vectors, and a subscript pair such as LR indicates LCP out from RCP in. Given knowledge of the film architecture and materials, the amplitude reflection coefficients can be computed using Berreman 4×4 matrix theory [6].

When a chiral film is illuminated by a beam of unpolarized white light at normal incidence the reflected beam is in general partially polarized and can be described by a set of Stokes parameters [7], s_0, s_1, s_2, s_3 . The Stokes parameters are defined to be amenable to experimental measurement using, as examples, a set of four polarizing filters or a spectral ellipsometer that records reflected intensity spectra after the beam passes through a rotating quarter-wave retarder plate and a fixed linear polarizer. The parameter s_0 represents the total intensity of the reflected beam. Linearly polarized light is indicated by s_1 and s_2 and circularly polarized light by s_3 . Unlike s_0 which is always positive, s_1, s_2 and s_3 may have positive or negative signs with the positives indicating linear along the y -axis rather than along z , linear at $+45^\circ$ rather than at -45° , and RCP rather than LCP.

Representing the unpolarized incident beam as the superposition of incoherent RCP and LCP beams of unit intensity allows the Stokes parameters to be expressed in terms of the reflection coefficients. Thus

$$s_0 = |r_{RR}|^2 + |r_{LR}|^2 + |r_{RL}|^2 + |r_{LL}|^2 \quad (1)$$

$$s_1 = 2\Im\{(r_{RR}r_{LR}^* + r_{RL}r_{LL}^*)e^{-i2\xi}\} \quad (2)$$

$$s_2 = 2\Re\{-(r_{RR}r_{LR}^* + r_{RL}r_{LL}^*)e^{-i2\xi}\} \quad (3)$$

$$s_3 = |r_{RR}|^2 - |r_{LR}|^2 + |r_{RL}|^2 - |r_{LL}|^2 \quad (4)$$

where ξ allows for azimuthal rotation of the sample relative to a reference orientation. Alternatively the partially-polarized reflected beam can be expressed as superposed polarized and unpolarized beams. In terms of the Stokes parameters, the total intensity of polarized light is $(s_1^2 + s_2^2 + s_3^2)^{1/2}$ and the intensity of unpolarized light is $s_0 - (s_1^2 + s_2^2 + s_3^2)^{1/2}$ [7].

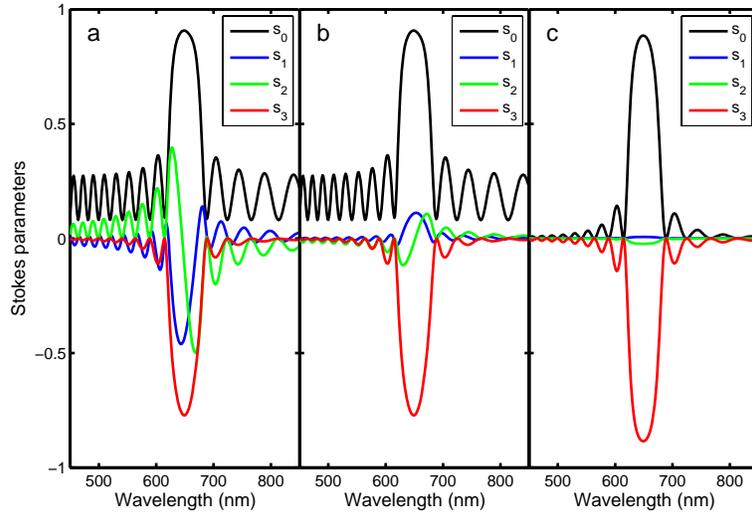


Fig. 1. Simulated Stokes parameters for (a) a chiral photonic film on glass, (b) 50 identical films with random azimuthal angles and (c) an index-matched film.

Now we consider an illustrative example, a left-handed chiral film with $n_2 = 1.75$, $n_3 = 1.85$ and $N = 20$. Figure 1(a) shows the Stokes parameters for a film bounded by air on one side and glass of refractive index 1.5 on the other side. Clearly all four Stokes parameters exhibit strong signatures of the circular Bragg resonance. However, to be useful for the characterization of randomly-oriented chiral photonic flakes, a reflected polarization spectrum should be invariant both to azimuthal rotation of a single flake and to random orientations of a set of flakes. Equations (1)-(4) imply that s_0 (which determines the color) and s_3 (the circular polarization) are invariant to azimuthal rotation, but s_1 and s_2 are not individually invariant. To investigate the second requirement we use Eqs. (1)-(4) to determine the effect of superposing incoherently the light from two or more flakes. For two identical flakes oriented at right angles the Stokes vectors

$$\begin{bmatrix} s_0 \\ s_1 \\ s_2 \\ s_3 \end{bmatrix} \text{ and } \begin{bmatrix} s_0 \\ -s_1 \\ -s_2 \\ s_3 \end{bmatrix} \text{ average to } \begin{bmatrix} s_0 \\ 0 \\ 0 \\ s_3 \end{bmatrix}. \text{ Overall the superposition maintains } s_0 \text{ (and hence}$$

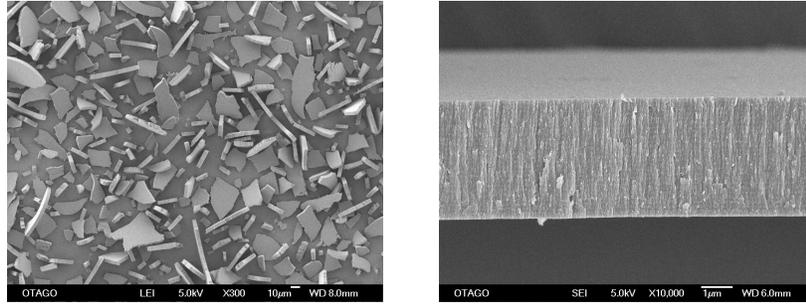


Fig. 2. Scanning electron micrographs of chiral photonic flake (left) and the edge of a single platelet (right).

the color), reduces the polarized light per flake from $(s_1^2 + s_2^2 + s_3^2)^{1/2}$ (elliptical) to s_3 (circular) and increases the unpolarized light per flake from $s_0 - (s_1^2 + s_2^2 + s_3^2)^{1/2}$ to $s_0 - |s_3| = R_{RL} + R_{LR}$. The latter result, that the unpolarized light is the sum of the cross-polarized reflectances, follows from Eqs. (1) and (4) with $R_{RR} \approx 0$ and $R_{LR} = R_{RL}$. Figure 1(b) shows that similar effects occur when the light from 50 randomly-oriented flakes is combined. Specifically, the s_0 and s_3 spectra are unchanged, s_1 and s_2 become less significant per flake and the polarization tends to circular. Hence of the various polarization parameters only s_3 has the required invariance to superposition. Finally we note that in principle the sidebands in the s_0 spectrum and the magnitudes of s_1 and s_2 can be reduced by index matching, as shown in Fig. 1(c), or with antireflection coatings [8]. Such a procedure reduces the polarized component of the reflected light, increases the strength of the s_3 resonance, and in turn leads to more saturated colors.

3. Results

We simulated and deposited six chiral photonic films on glass for use in the project using the deposition parameters:- substrate temperature 300°C , bidposition angle 65° , deposition rate 0.25 nm s^{-1} , oxygen backfill pressure $2 \times 10^{-4} \text{ mbar}$. Typically 25 dielectric periods were deposited. Anti-reflection coatings were not included in the designs. Following ellipsometric characterization of the films as they were illuminated by unpolarized white light at an angle of incidence of 20° we formed flake from each sample, by scraping with a firm scalpel blade and collecting the material on double-sided black adhesive carbon tape [9]. The scanning electron micrograph in the left side of Fig. 2 shows platelets with lateral size in the range $3 - 50 \mu\text{m}$, and the micrograph of the edge of a single flake in the right side of the same figure shows a normal columnar nanostructure, parallel sides and a thickness of about $3 \mu\text{m}$. Figure 3 shows left-handed flake that appears green when illuminated by unpolarized white light and observed directly (left side), and dull when observed through a filter that is opaque to LCP (right side).

Finally the light reflected from flake illuminated with unpolarized white light at 20° was characterized by ellipsometry. The collection area was a circle of nominal diameter $60 \mu\text{m}$. Figure 4 shows s_3 polarizing spectra recorded from chiral films and flake for three architectures:- (i) a standard medium that supports a single left-handed Bragg resonance, (ii) a threaded chiral medium that supports both a right-handed resonance and a left-handed resonance at a longer wavelength, and (iii) a threaded chiral medium that supports two right-handed resonances at different wavelengths.

Spectra recorded from similar architectures, but with a central 90° twist defect, are shown in Fig. 5. In these spectra the structural defect causes a spectral hole at the center of each Bragg

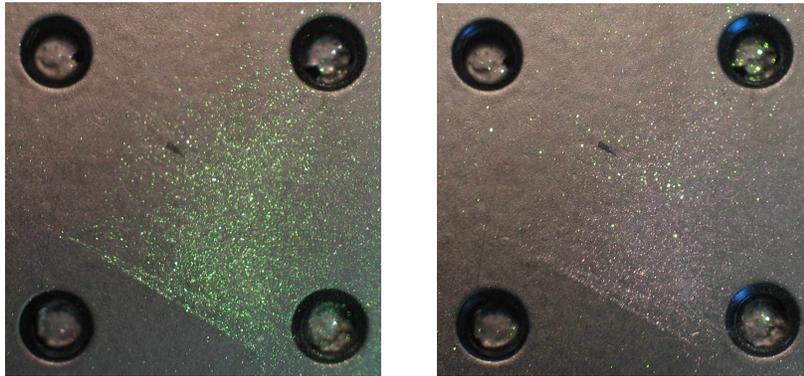


Fig. 3. Left-handed chiral photonic flake illuminated by unpolarized white light and photographed without a filter (left) and using a filter opaque to left circular light (right). The scale can be determined from the 25mm spacing of the screw holes.

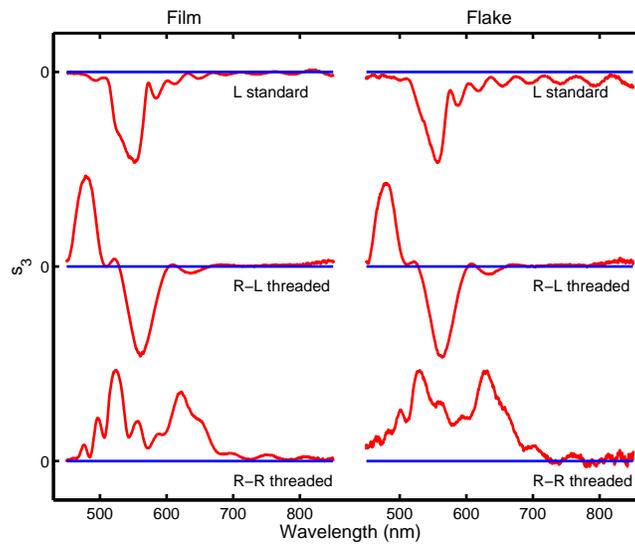


Fig. 4. Polarization spectra s_3 recorded for chiral photonic films on glass and from flake scraped from the films.

resonant peak. The spectral hole is somewhat less distinct in the flake from the standard chiral film, probably due to averaging over a range of angles of incidence. As well a tendency for more circular light from flake (relative to film) was noted in some samples. Average values of $n_3 - n_2 = 0.05$ and $n_{av} = 1.69$ were recorded for the six chiral films used in the study.

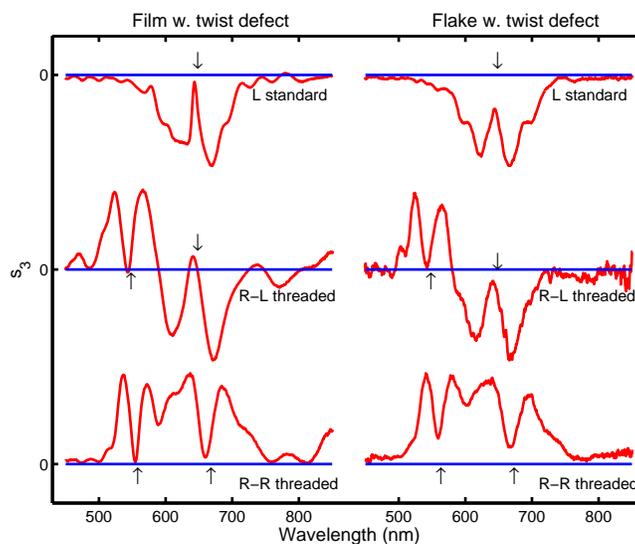


Fig. 5. Polarization spectra s_3 recorded for chiral photonic films deposited with a central twist defect of 90° on glass and from flake scraped from the films. In each case the arrows indicate the location of spectral holes caused by the defect.

4. Summary

We have discussed the design, fabrication and polarizing properties of inorganic chiral photonic films and flake derived from the films. The Stokes s_0 spectrum determines the color and the s_3 spectrum is shown to characterize the polarizing properties. Sample films were fabricated with threaded right-handed and left-handed architectures and with or without twist defects. Ellipsometric measurements of the films and flake showed s_3 spectral signatures of the circular Bragg resonances and the spectral holes. Overall our experiments show that chiral photonic flake has potential as a physical effect colorant with polarizing properties. Possible applications include document security and polarizing paint.

Acknowledgments

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