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Band structure controlled by chiral imprinting

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Using the configuration of an imprinted cholesteric elastomer immersed in a racemic solvent, the authors find the solution of the boundary-value problem for the reflection and transmission of incident optical waves due to the elastomer. They show a significant width reduction of the reflection band for certain values of nematic penetration depth, which depends on the volume fraction of molecules from the solvent, whose handedness is preferably absorbed. The appearance of nested band gaps of both handednesses during the sorting mixed chiral process is also obtained. This suggests the design of chemically controlled optical filters and optically monitored chiral pumps. © 2007 American Institute of Physics. [DOI: 10.1063/1.2784174]

Chiral materials of the periodically nonhomogeneous kind are usually found as chiral nematic, as chiral smectic liquid crystals,¹ as chiral sculptured thin films,² or as chiral elastomers.³ The keynote of these materials is the display of the circular Bragg phenomenon, whereby a normally incident, circularly polarized plane wave of a specific handedness is highly reflected in a certain wavelength regime, whereas a similar plane wave but of the reverse handedness is not. This polarization-discriminatory filtering characteristic of structurally chiral materials is very attractive in optical technology.⁴ In a recent paper⁵ and references within, it has been shown that the band gap, transmittance, and reflectance of a chiral sculptured thin film can be controlled by applying a dc electric field parallel to the helix axis of these materials. Furthermore, the dc electric field was found to optically switch on the band gap, since it creates a Bragg regime even if the sculptured chiral material lacks band gap in its absence.

A polarization-universal filter based on periodically twisted nematics has been proposed,⁶ in which the nematic's director does not rotate uniformly around the helix axis, but oscillates with respect to a certain direction following a predetermined modulation. Under specific conditions, the same structure is obtained from chiral elastomers subjected to transverse stress.⁷ For both systems, the polarizationdiscriminatory feature of a standard chiral material is almost lost and both handednesses of circular polarization exhibit a very similar band gap. The fact that imprinted cholestericelastomer's configurations are also susceptible to the presence of racemic and chiral solvents⁸ opens up the door not only to a mechanism for chemically controlling these optical devices but also to an optical procedure for determining quantitatively the degree of chiral segregation performed by an imprinted elastomer. The purpose of this work is to study the transmission and reflection of normally incident light on a cholesteric elastomer slab immersed in a solvent and surrounded by vacuum, as a function of the concentration of the absorbed molecules with preferred chirality.⁹

A cholesteric liquid crystal without distortion possesses a locally uniaxial structure given by the director vector $\hat{\mathbf{n}} = [0, \cos \phi_0(x), \sin \phi_0(x)]$, where $\phi_0(x) = q_0 x$ is the angle between director vector and the y axis in the x-y plane, describing a periodic helical structure characterized by a pitch $p = \pi/q_0$. The energy for an elastomer formed under the presence of a cholesteric solvent, which is subsequently replaced by an achiral one, can be expressed as,¹⁰

$$F = \frac{1}{2}D_1 \int \left[\xi^2 \left(\frac{d\phi}{dx}\right)^2 + \sin^2(\phi - q_0 x)\right] dx,\tag{1}$$

where the nematic penetration depth in the rubbery network is $\xi = \sqrt{K_2/D_1}^3 K_2$ is the twist elastic constant and D_1 is the local anchoring of the director to the rubbery network which depends on the cross-linked density.¹¹ The first term in Eq. (1) which comes from the Frank-free energy contributes to unwind the cholesteric helix whereas, the second term tends to twist the helix. A spectacular property of imprinted networks is their capacity to preferentially absorb and retain right or left molecules from a racemic solvent which renders them chiral pumps.

After minimizing Eq. (1), we get $\xi^2(d^2\phi/dx^2)$ +sin $2(q_0x-\phi)=0$, whose solution is given by $\phi(x)=q_0x$ $-Am(cx/\xi,1/c^2)+\pi/2$, where Am(x,m) is the Jacobian amplitude, *c* is a constant related to the reduced elastic energy,¹⁰ $g=2\xi F/TD_1$, as is shown in Fig. 1, with *T* the period of the Am(x,m) function, and $\alpha=\xi q_0$ is the imprinting efficiency. Notice how $g(\alpha,c)$ increases when either values of α or *c* get large and exhibit a critical value at $c_c=1$, where for $\alpha \leq 2/\pi$ all the curves have a sharp peak. In contrast, for $\alpha \geq 2/\pi$, the peak is smoothed and a minimum appears in the region $c_c \geq 1$ whose position moves to the right as α gets bigger. By increasing ξ and *c* from zero, $\phi(x)$ presents oscillations around $\phi_0(x)$ whose spatial period and amplitude in-

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FIG. 1. Reduced energy density as function of c for $\alpha = 0.2$, $2/\pi$, and 1.

crease, respectively, until each of these parameters reach the critical values: $\xi_c=3$ and $c_c=1$. After these values, $\phi(x)$ deviate from ϕ_0 and enlarge its effective q value for $\xi \ge \xi_c$ or diminishes for $c \ge c_c$ which is equivalent to having an untwisted or a more twisted helix than that of ϕ_0 .

To solve the electromagnetic problem, we chose the x axis as the axis of chiral nonhomogeneity. Then, the optical relative permittivity matrix of this material may be stated as

$$\overline{\epsilon}(x) = \begin{pmatrix} \epsilon_{\perp} & 0 & 0\\ 0 & \epsilon_{\perp} + \epsilon_a \cos^2 \phi(x) & \epsilon_a \sin \phi(x) \cos \phi(x)\\ 0 & \epsilon_a \sin \phi(x) \cos \phi(x) & \epsilon_{\perp} + \epsilon_a \sin^2 \phi(x) \end{pmatrix},$$
(2)

where $\epsilon_a = \epsilon_{\parallel} - \epsilon_{\perp}$ is the dielectric anisotropy and ϵ_{\parallel} and ϵ_{\perp} are the dielectric constants parallel and perpendicular to the director $\hat{\mathbf{n}}$, respectively.

Maxwell's equations (in MKS units) for a medium whose dielectric tensor is only x dependent are given by



FIG. 2. Circularly polarized reflectance R_{RR} and R_{LL} against λ and c for normal incidence and $\alpha=1$. Other parameters are $\epsilon_{\perp}=1.91$, $\epsilon_{\parallel}=2.22$, h=1, p=218 nm, and $L/\Omega=60$.

$$\nabla \times \mathbf{E}(x, y, z) = i\omega\mu_0 \mathbf{H}(x, y, z)$$

$$\nabla \times \mathbf{H}(x, y, z) = -i\omega\epsilon_0 \overline{\epsilon}(x) \cdot \mathbf{E}(x, y, z)$$

$$0 < x < L, \quad (3)$$

where ϵ_o and μ_o are the permittivity and the permeability of free space. If a plane wave impinges normally on the cholesteric elastomer, we can describe the fields⁵ in terms of the column vector $\overline{\psi}(x) = [e_y(x), e_z(x), h_y(x), h_z(x)]^T$. Thus, the essential part of the Maxwell curl equations can be stated as

$$d\bar{\psi}(x)/dx = i\bar{A}(x) \cdot \bar{\psi}(x), \quad 0 < x < L, \tag{4}$$

where $\overline{A}(x)$ is given by

$$\bar{A}(x) = \omega \begin{pmatrix} 0 & 0 & \mu_0 \\ 0 & 0 & -\mu_0 & 0 \\ -(\epsilon_0/2)\epsilon_a \sin 2\phi(x) & -\epsilon_0(\epsilon_\perp + \epsilon_a \cos^2 \phi(x)) & 0 & 0 \\ \epsilon_0(\epsilon_\perp + \epsilon_a \sin^2 \phi(x)) & \epsilon_0/2\epsilon_a \sin 2\phi(x) & 0 & 0 \end{pmatrix}$$

$$\begin{pmatrix} 0 & \mu_0 \\ \mu_0 & 0 \\ 0 & 0 \\ 0 & 0 \end{pmatrix} .$$
 (5)

To solve Eq. (4) we chose a piecewise homogeneity approximation method.² The final result is the computation of a transfer matrix \overline{U}' such that $\overline{\psi}(L) = \overline{M}^T(L) \cdot \overline{U}' \cdot \overline{M}(0) \cdot \overline{\psi}(0) = \overline{U} \cdot \overline{\psi}(0)$. The solution of the boundary value problem for the reflection and transmission of an obliquely incident plane wave in cholesterics is a well known problem.² Here, we state that if a_L and a_R are the amplitudes of the left- and right-circularly polarized components of the incident plane wave's components, and t_L and t_R are those of the transmitted plane wave's components, then they are related by

$$(t_R \ t_L \ r_R \ r_L) = (a_R \ a_L) \begin{pmatrix} t_{RR} \ t_{LR} \ r_{RR} \ r_{LR} \\ t_{RL} \ t_{LL} \ r_{RL} \ r_{LL} \end{pmatrix},$$
(6)

where t_{ij} and r_{ij} are the transmission and reflection coefficients. The copolarized transmittances are denoted by $T_{LL} = |t_{LL}|^2$ and $T_{RR} = |t_{RR}|^2$, and the cross-polarized ones by $T_{LR} = |t_{LR}|^2$ and $T_{RL} = |t_{RL}|^2$, and similarly for the reflectances $R_{RR} = |r_{RR}|^2$, etc. These quantities depend on the wavenumber k_0 , in addition to the constitutive parameters of the elastomer cholesteric and its thickness. The calculation parameters used in this work were obtained from a real cholesteric elastomer material.¹²

plane wave's components, then they are related by In Fig. 2 we show circularly polarized reflectances R_{RR} Downloaded 10 Dec 2008 to 169.234.241.179. Redistribution subject to AIP license or copyright; see http://apl.aip.org/apl/copyright.jsp



FIG. 3. Circularly polarized reflectance R_{RR} and R_{LL} vs λ and α for normal incidence and c=0.5. We use the same parameter as that of Fig. 2.

and R_{LL} against the light wavelength λ and c for normal incidence. For c=0 we have an ideal helix so there is only a band reflection for R_{RR} . The bandwidth and amplitude of R_{RR} remain almost constant until c is around 0.4, where these quantities diminish considerably to about 70% for $c \simeq 0.75$. For this value, the reflection band loses its well defined robust band showing a minimum at its center. For even larger values of c, the band gets wider and larger and attains values of their width and amplitude for c=1, very similar to those shown by an undistorted helix (c=0). For c>1 the bandwidth reduces rapidly but the amplitude decreases slowly. Complementary, R_{LL} is practically null for c < 0.4, but outside this interval its amplitude grows as much as 0.2 for c>0.7. A very similar behavior is displayed by R_{LR} . The transmittances show consistently the complementary part of the band gap, even though there exist reflection bands for both handednesses for a complete interval of c, just at c=0.75 both bands have the same amplitude but different widths. As a consequence, the elastomer slab exhibit nested band gaps similar to those found for an elastomer subjected to a mechanical stress applied perpendicularly to the elastomer-helix axis.' That is, a wide partial reflection band for left-circularly polarized waves contains a thinner total reflection band where the system behaves actually as a polarization-universal filter, as the one proposed by Sarkissian *et al.*⁶

In Fig. 3 we plot R_{RR} and R_{LL} versus λ and α also for normal incidence. At $\alpha=0$, R_{LL} recovers again the undistorted helix case but for larger values increases and widens abruptly to reveal a wide and short band reflection at α =0.25 whose amplitude is roughly 0.2, but its bandwidth first increases and then diminishes. On the other hand, R_{RR} has roughly the same width for all α values while its amplitude decreases until 0.25 for $\alpha = 0.6$. Mao and Warner⁸ performed calculations of volume fraction of right-handedsolvent molecules in a gel as a function of the right-handed species in the solvent outside the gel. They showed that during an inverse imprinting process, the elastomer can lead to chiral separation whose maximum resolving power is obtained when $\alpha = 2/\pi$. Thus, by measuring these optical spectra we can determine either the value of c or α . Conversely, this system may function as a chemically handled filter whose bandwidths and reflectance amplitudes are controlled by the concentration of molecules with preferably absorbed chirality.

In summary, we have obtained the circularly polarized reflectances and transmittances of a cholesteric elastomer immersed in a racemic solvent. The bandwidths of the reflectances for both handednesses under the presence of the solvent, change considerably in such way that their variations may be experimentally detected. Thus, the resolving power of the chiral segregation performed by the elastomer may be also determined.

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