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Twist defect in an imprinted cholesteric elastomer

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We have found that a chiral twist defect inserted in a cholesteric elastomer gives rise to circularly polarized localized modes of both handedness. This defect enhances the resonant mode amplitude whose handedness is opposite to that of the cholesteric helix for high cross-linked density, whereas for low cross-linked density, the same mode is decoupled with the defect and thus the resonant mode disappears. Finally, the resonant mode of the same handedness as the elastomer helix is maintained for both high and low cross-linked density. © *2009 American Institute of Physics*.
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Cholesteric elastomers (CEs) are formed by monomers of liquid crystals cross-linked to polymeric chains. This union produces a flexible material whose molecular order is similar to cholesteric liquid crystals (CLCs) with the advantage that in this new material, it is possible to change the optical properties by means of macroscopic deformations.[1] The study of the optical properties of these materials have recently grown due to the achievement managed by Kim and Finkelmann[2] who found a procedure to obtain monodomain nematic and CEs.

In the past decade, it has been shown that dye-doped CEs can show laser emission at the band edge. To this aim, they have been synthesized by anisotropic deswelling method[3] and UV cross linking of a cholesteric side-chain polymer.[4] In a former work,[5] it was shown that on application of a biaxial stress, the selective reflection band of the same handedness as the helix shifts to shorter wavelengths and gives rise to laser emission when the reflection band overlaps the dye's emission maximum.

A different mechanism to achieve lasing is introducing defects in these materials in order to break the anisotropic periodicity of the elastomers. For instance, in Ref. 6 it has been reported the construction of a device in which an isotropic film doped with Rhodamine dye was inserted within an elastomer slab. There, laser emission was observed when the Rhodamine fluorescent emission and the selective reflection gap of the cholesteric are overlapped. A simpler type of defect has been introduced in conventional CLC by inserting a discontinuity in the helix. This insertion produces quasilocalized resonant modes,[7] which can be utilized to build low threshold lasers, low loss waveguides, and narrow band filters.[8]

The distorted CEs are chiral materials, which have a reflection band for each circularly polarized component as it has been theoretically shown in previous works.[5,9,10] This has been also recently confirmed experimentally[6] where a reflection band of polarization opposite to helix of elastomer for high chemical cross-linked density was found. The local anchoring effect of cross link in CEs is essential for the director response to deformation, as the chemical cross-linked density is related with the chiral parameter order.

The purpose of this work is to analyze the influence of a twist defect in the band structure of CE and find the resulting resonant modes as a function of the chemical cross-linking density.

A twist defect can be created by rotating around the helix axis, one part of the elastomer with respect the other in such way that the helix presents a discontinuity on its director's angle, as is illustrated in Fig. 1. A chiral-imprinted elastomer can be attained experimentally by reacting a mesogenic nematic group and a cross-linker doped with a fixed concentration of total weight of a chiral compound diluted in a solvent. After evaporation of the solvent and completion of cross linking, CEs can be obtained with the same helical pitch irrespective of the cross-linking density. Finally, the chiral dopant is removed by placing the material in a large volume of nonchiral solvent leading to a diffusion of the chiral dopant.[1]

There are two competing mechanisms in the elastomer: (i) the twist of the gel which has its minimal energy at cross linking, whose energy density is given by $K_2(d\phi/dx)^2/2$ and (ii) the anchoring of the director to the configuration at the moment of cross linking, whose change in energy density due to rotation is $D_1\sin^2(\phi-q_0x)/2$, where $D_1=\mu(r-1)^2/r$. Here, K_2 is the twist elastic constant, D_1 is the local anchoring of the director to the rubbery network, r measures the anisotropy of the average chain shape spheroid, and μ is the linear shear modulus of the rubber.[1]

The configuration of this imprinted CE is found by minimizing the sum of both free energy contributions, which yields

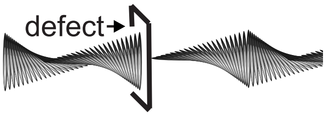

FIG. 1. Schematic plot of the twist defect introduced in the CE helix.

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$$\frac{\alpha^2}{q_0^2}\frac{d^2\phi}{dx^2}+\sin 2(q_0x-\phi)=0, \qquad (1)$$

where $\alpha=\sqrt{K_2/D_1}q_0$ is the chiral order parameter, $q_0=\pi/p$ is the wave number, and p is the pitch. Variations in the chiral order parameter produce variations in the density of cross links in the network (affecting in turn D_1) and changing the robustness of chiral imprinting.[11]

The solution of Eq. (1) for $\alpha=0$ is $\phi_0=q_0x$ corresponding to an ideal cholesteric helix. The general solution without defect is given by $\phi(x)=q_0x-Am(cxq_0/\alpha,1/c^2)+\pi/2$, where $Am(x,m)$ is the Jacobian amplitude and c is a constant related with the reduced elastic energy.[12] When we introduce a chiral twist defect of 90° in the middle of the sample, the configuration is described by $\phi(x)=q_0x-Am(cxq_0/\alpha,1/c^2)+3\pi/4+(\pi/4)\text{sign}(x)$, where sign (x) is the sign function.

We have expressed the electromagnetic problem in terms of a 4×4 matrix first order differential equation, which we have solved numerically by using the piecewise constant approximation method, whose formulation is available elsewhere.[5] We have chosen a right-handed real CE (Ref. 13) including a 90° twist defect of the same handedness.

In Ref. 5, it has been shown that a distorted CE has reflection bands for both polarizations and values of $\alpha<\alpha_c=2/\pi$, i.e., those values for which the chemical cross-linked density and the efficiency imprinting of helix are high. While a conventional CLC in absence of electric field does not have a reflection band for the polarization of handedness opposite to its helix, the distorted CE exhibits a reflection band for that polarization, which has a bandwidth larger for oblique incidence.[9] These two bands have been observed experimentally in the CE submitted to a strain in presence of cross-linking for oblique incidence.[6] When a chiral twist defect of 90° is introduced in a CE distorted appears a resonant mode for both polarizations. In Fig. 2(a), it is shown how the reflection band is influenced by the presence of a chiral twist defect of 90° for $\alpha=0.3$; consequently, we have found a resonant mode at the center of reflection band of both polarizations. Additionally, the defect enhances the resonant mode amplitude for the left circularly polarization light. This behavior has been observed in conventional CLC with a chiral twist defect,[8] where the resonant mode amplitude for circularly polarized light of handedness opposite to its helix augments for thickness larger than the crossover length L_{co}, despite that a conventional CLC with a twist defect[8] does not have reflection band for this polarization.

For α near to the critic value α_c, the chemically cross-linked density diminishes and the resonant mode of the polarization opposite to cholesteric helix disappears, that is the incident left circularly polarization light and the defect mode are decoupled in the resonant frequency, as is shown in Fig. 2(b). While the resonant mode for right circularly polarization is kept at the band center, two reflection bands appear for the left circularly polarized light at the edges of the right reflection band.

This behavior is qualitatively maintained for values of α far from α_c with the difference that the bands amplitudes diminish until they finally disappear. We have found a similar behavior to that of Fig. 2(a) for $c=0.5$ and $c=0.6$. Additionally, the chiral twist defect enhances the resonant mode amplitude for all the values of c until $c=1.6$, where finally the resonant modes disappear for both polarizations.

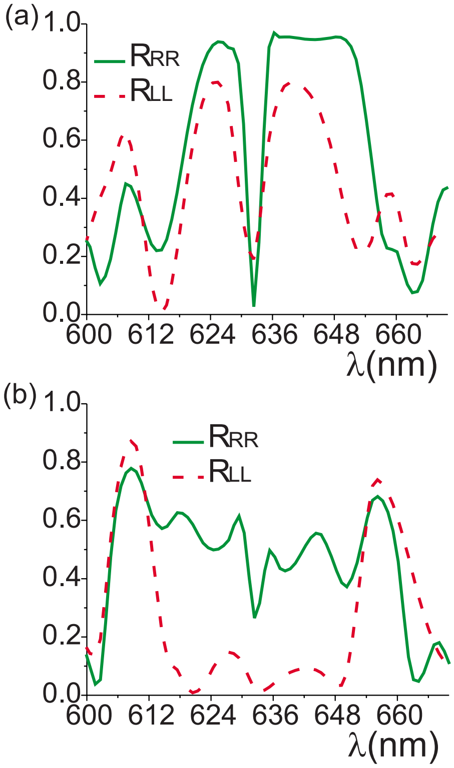

FIG. 2. (Color online) Circularly copolarized reflectances right-R_{RR} (solid line) and left-handed R_{LL} (dashed line) vs wavelength λ for a CE distorted with a chiral twist defect of 90° for $c=0.5$ and (a) $\alpha=0.3$ and (b) $\alpha_c=2/\pi$ at resonant wavelength of 632 nm. The dielectric ordinary and extraordinary constants are $\epsilon_0=1.91$ and $\epsilon_e=2.22$. Other parameters are $p=218$ nm and sample thickness $l=60p$.

The dependence on the dimensionless thickness $L=l/p$ of reflectance (solid line) and transmittance (dashed line) at the resonant peak for left and right circular polarizations is displayed in Fig. 3(a). We have depicted the crossover length for high chemical cross-linked density corresponding to $\alpha=0.3$ in both polarizations. The crossover length is obtained in numerical simulations by finding the sample thickness at which reflectance and transmittance of the same handedness coincide.

Finally, in Fig. 3(b) we have presented the inverse bandwidth at resonant wavelength versus the sample thickness for different values of the chiral order parameter of right polarization. Figure 3(c) shows the same as that of Fig. 3(b) but for left circularly polarized light. The curves grow and exhibit saturation values like typical curves for laser devices. For values around the critic value α_c, the resonant mode for left circularly polarized light wave vanishes so the localized mode become decoupled for a low chemical cross-linked density as is shown in Fig. 3(c) for $\alpha=0.8$.

We have found a resonant mode in the center of reflection band for both polarizations in an imprinted distorted CE including a 90° chiral twist defect, for high chemical cross-linked density. Also, the defect enlarges the resonant mode amplitude for left circularly polarization, and for all c-values and some α-values.

In contrast to CLC with a chiral twist defect for which the right circularly mode is decoupled from the defect mode after of crossover length, this system has two resonant modes of different circular polarizations, which are kept coupled

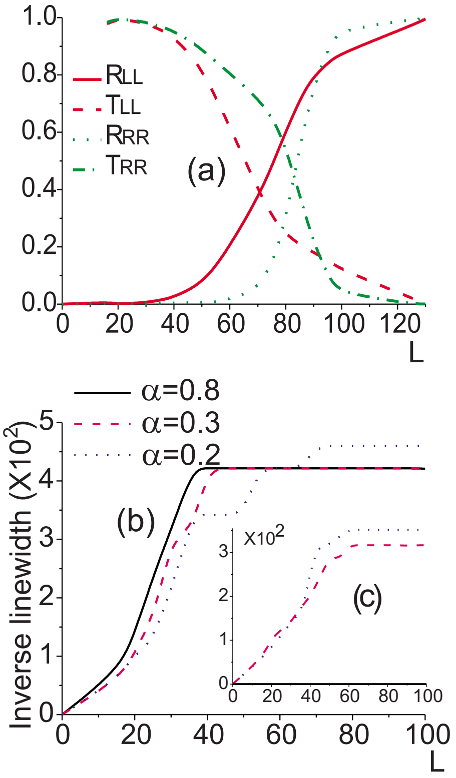

FIG. 3. (Color online) (a) Circularly copolarized reflectances R_{RR}, R_{LL} (solid line), transmittances T_{RR}, and T_{LL} (dashed line) vs L at the resonant wavelength for high chemical cross-linked density where $\alpha=0.3$ and $c=0.5$. (b) Inverse linewidth at resonant wavelength for right and (c) left circularly polarized light as function of L for $c=0.5$ and $\alpha=0.2$ (dotted line), 0.3 (dashed line), and 0.8 (continuous line) corresponding to various values of the chemical cross-linked density.

with the defect mode for high chemical cross-linked density beyond the crossover length.

When the chemical cross-linked density is lower in distorted CE around the critic value α_c, the resonant mode for left circularly polarized light quits to be coupled and disappears because then, the dynamic constraints force the CE helix to accommodate the shape change by varying the helix pitch so that it will not return to its original pitch value without distortion.

In conclusion, once having a lasing system for both mentioned modes, CEs can be used to improve the laser feedback by making it more efficient. They also have the advantage of being micrometer films systems that can be inserted in smaller spaces. Moreover, having two resonant modes inside of one system at resonant frequency provides a higher brightness as it occurs in the broad band of devices with reflection for both handedness of polarized light.[14]

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