Nonlinear Reflection in Cholesteric Liquid Crystals: Mirrorless Optical Bistability

Herbert G. Winful
Advance Technology Laboratory, GTE Laboratories, Inc., Waltham, Massachusetts 02254
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Exact elliptic-function solutions are presented for intense light waves in cholesteric liquid crystals. Light-induced changes in the pitch of the cholesteric helix lead to a bistable reflection characteristic even in the absence of external reflectors.

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The linear propagation of light in cholesteric liquid crystals has been studied extensively by several authors.[1] The remarkable rotatory power, circular dichroism, and iridescence displayed by these liquid crystals have been explained on the basis of a spiral arrangement of anisotropic molecular layers which produces selective Bragg scattering of circularly polarized light of the appropriate helicity and wavelength. In all these theories it has been tacitly assumed that the properties of the cholesteric helix do not change under the action of light waves. Recently, however, an extremely large orientational optical nonlinearity has been predicted in the mesophase of nematic liquid crystals,[2] and effects such as self-focusing, degenerate four-wave mixing, optical field-induced birefringence, and the Fréedericksz transition[3] have been observed in nematics with cw laser intensities of less than 200 W/cm^2. Since cholesterics are locally indistinguishable from nematics, one would expect a similarly large orientational nonlinearity in the former. Furthermore, the large-scale helical ordering of cholesterics should give rise to nonlinear optical phenomena unique to that mesophase.

In this Letter I consider light-induced distortions of the cholesteric helix for waves in the Bragg regime. One novel result is that the reflection coefficient of a cholesteric film is a multivalued function of the input intensity. This leads to an intrinsic, mirrorless optical bistability in contrast to the classic bistability of nonlinear Fabry-Perot interferometers.[4] The physical principle is similar to the bistability predicted for distributed-feedback structures[5] and in the degenerate four-wave mixing process.[6]

DeGennes[7] and Meyer[8] have shown that static electric and magnetic fields can distort the cholesteric helix and increase its pitch. An extension to high-frequency traveling waves has been suggested by Dmitriev.[9] However, he assumed a rather special form for the pitch dilation, and his linearized theory (linear in intensity) applies only to infinite cholesteric media and for waves far from the Bragg condition. In the present work no assumption is made concerning the form of the static distortion. Instead we solve the coupled nonlinear Euler-Lagrange and Maxwell equations in a self-consistent manner which yields the steady-state configuration of the cholesteric helix. The reaction of the distorted helix on the light field emerges in a natural way from this calculation which is exact within the slowly-varying-envelope approximation.

We consider a cholesteric slab of length L whose helix axis is oriented along $\hat{z}$. The average orientation of the elongated liquid-crystal molecules is described by the director $\hat{n}(z)$ whose components are

$$n_x = \cos\theta(z), \quad n_y = \sin\theta(z), \quad n_z = 0. \tag{1}$$

The director rotates about the z axis, and in the absence of external perturbations the angle θ is given by $\theta = q_0 z$, where q_0 is the unperturbed wave number of the helix whose pitch is $p = 2\pi/q_0$. Under the action of intense light waves the cholesteric director assumes a new configuration which may be found by minimizing the total free energy[10]

$$F = \tfrac{1}{2}\int d^3r\{K_{11}(\nabla\cdot\hat{n})^2 + K_{22}(\hat{n}\cdot\nabla\times\hat{n}+q_0)^2 + K_{33}(\hat{n}\times\nabla\times\hat{n})^2 - \vec{E}\cdot\vec{D}/4\pi\}. \tag{2}$$

Here K_{11}, K_{22}, and K_{33} are the Frank elastic constants that describe the basic distortions of splay, twist, and bend, respectively. The electric displacement is related to the electric field E through

$$\vec{D} = \epsilon_\perp \vec{E} + \epsilon_a \hat{n}(\hat{n}\cdot\vec{E}), \tag{3}$$

where $\epsilon_a = \epsilon_\parallel - \epsilon_\perp$, $\epsilon_\parallel$ and $\epsilon_\perp$ being the dielectric constants parallel and perpendicular to the local director.

The helical structure of the cholesteric liquid crystal suggests the use of circularly polarized basis fields; hence we take the electric vector as

$$\vec{E} = \mathrm{Re}\{[E_-(z)(\hat{x}+i\hat{y})/\sqrt{2} + E_+(z)(\hat{x}-i\hat{y})/\sqrt{2}]e^{-i\omega t}\}, \tag{4}$$

where $E_\pm = (E_x \pm iE_y)/\sqrt{2}$. For transverse electromagnetic waves propagating along the z axis, only the twist term (proportional to K_{22}) contributes to the distortion free energy and thus minimization of the free energy leads to the Euler-Lagrange equation

$$\frac{d^2\theta}{dz^2} = \frac{\epsilon_a}{8\pi K_{22}}[\mathrm{Re}(E_+E_-^*)\sin 2\theta - \mathrm{Im}(E_+E_-^*)\cos 2\theta]. \tag{5}$$

The field amplitudes are functions of z and must be found from the Maxwell equations which, by use of (3) and (4), can be written[10]

$$-d^2E_\pm/dz^2 = k_0^2E_\pm + k_1^2E_\mp e^{\pm i2\theta}, \tag{6}$$

where $k_0^2 = (\omega/c)^2(\epsilon_\parallel + \epsilon_\perp)/2$ and $k_1^2 = (\omega/c)^2\epsilon_a/2$.

In the linear theory, circularly polarized light whose wavelength lies in the Bragg regime of the cholesteric structure is almost totally reflected if its helicity matches that of the liquid crystal.[1] The reflected light is also circularly polarized with the same sense as the incident light, and with a wave vector close to that of the cholesteric structure. (In contrast, ordinary mirrors reverse the helicity of an incident wave.) Thus we may take the field within the medium as a sum of counterpropagating right circularly polarized waves of the form $E_\pm = |\mathcal{E}_\pm(z)|\exp[i\varphi_\pm(z) \pm iq_0z]$. Then in the slowly-varying-envelope approximation Eqs. (6) become

$$d|\mathcal{E}_+|/dz = \kappa|\mathcal{E}_-|\sin\Psi, \tag{7a}$$

$$d|\mathcal{E}_-|/dz = \kappa|\mathcal{E}_+|\sin\Psi, \tag{7b}$$

$$d\Psi/dz = 2(q_0 + \Delta k) + \kappa(|\mathcal{E}_-|/|\mathcal{E}_+| + |\mathcal{E}_+|/|\mathcal{E}_-|)\cos\Psi - 2d\theta/dz. \tag{7c}$$

Here $\Psi = \varphi_+ - \varphi_- + 2q_0z - 2\theta$, $\kappa = k_1^2/2q_0$, and $\Delta k = (k_0^2 - q_0^2)/2q_0$. Without loss of generality I choose boundary conditions such that the director is constrained at the input $[\theta(0) = 0]$ and free at the exit $(d\theta/dz|_L = q_0)$. For the field I assume zero-reflection boundary conditions so that $|\mathcal{E}_-(L)| = 0$. (Discrete dielectric reflections may be included trivially but these do not introduce any new physics.) The simultaneous solution of Eqs. (5) and (7) yields the self-consistent electric field and director distributions.

Three integrations of the set are used to obtain a single equation for the forward flux in the cholesteric:

$$du/dz = 2\kappa[Q(u)]^{1/2}, \tag{8}$$

where

$$Q(u) = (u-J)[u - (u-J)(\Delta k/\kappa - J + u)^2], \tag{9}$$

with $u = \gamma|\mathcal{E}_+|^2$, $J = \gamma|\mathcal{E}_T|^2$, the normalized transmitted intensity, and $\gamma = \epsilon_a/32\pi K_{22}\kappa^2$. The solution of Eq. (8) is the distribution of the forward flux,

$$u(z) = u_3 + \frac{u_2 - u_3}{1 - (u_1-u_2)(u_1-u_3)^{-1}\mathrm{sn}^2[2\kappa(z-L)/g, k]}, \tag{10}$$

where $u_1 > u_2 > u_3 > u_4$ are the roots of $Q(u)$, and sn is a Jacobi elliptic function with $g = 2/[(u_1-u_3)(u_2-u_4)]^{1/2}$ and $k = [(u_1-u_2)(u_3-u_4)]^{1/2}g/2$. A final integration then yields the director distribution

$$\theta(z) = q_0z + 2\kappa(J-u_3)z - g(u_2-u_3)[\Pi(\varphi(z), \alpha^2, k) - \Pi(\varphi(0), \alpha^2, k)], \tag{11}$$

where Π is an elliptic integral of the third kind, with $\alpha^2 = (u_1-u_2)/(u_1-u_3)$, and $\varphi(z) = \sin^{-1}[\mathrm{sn}(\kappa(z-L)g)]$.

The main results of the theory are contained in Eqs. (10) and (11). For a given value of the transmitted intensity J, evaluation of u at $z = 0$ yields the incident intensity I. The relation between transmitted (or reflected) intensity and incident intensity is shown in Fig. 1. This relation is multivalued and thus the reflected (and transmitted) intensity will exhibit hysteresis and discontinuous jumps as the incident intensity is varied. As is the case with static electric and

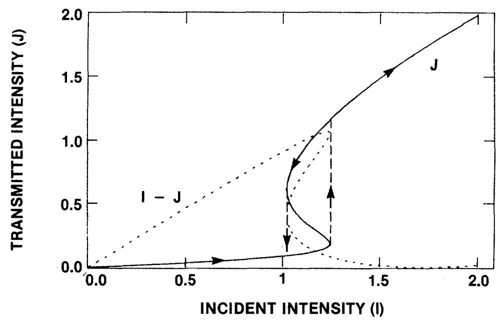

FIG. 1. Transmitted (solid line) and reflected (dashed line) intensities vs incident intensity for $\kappa L = 2$ and $\Delta k = 0$. For $\kappa L > 2$, a discontinuous jump from the lower to the upper branch occurs when $I \simeq 1$. Intensities are normalized by $32\pi K_{22}\kappa^2/\epsilon_a$.

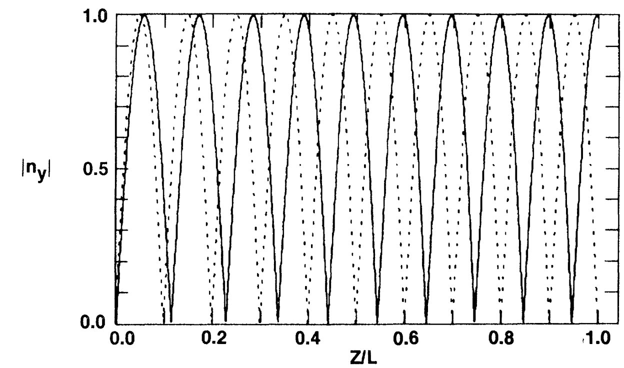

FIG. 2. The y component of the director for zero field (dashed line) and for an incident intensity of $I = 1.14$ (full line). The transmitted intensity is $J = 0.12$ which is on the lower branch of the bistability curve of Fig. 1. The normalized length of the medium is $q_0 L = 10\pi$.

magnetic fields, the time-averaged optical field couples to the local dielectric anisotropy to exert torques within the cholesteric which oppose the elastic torques responsible for its liquid-crystalline order. The resulting distortion of the cholesteric helix and the increase in its period means that a light wave which at low intensity satisfies the Bragg condition for the structure may not suffer Bragg reflections at higher intensities. Thus a transition from high reflection to high transmission occurs as the incident intensity is increased beyond a critical value. In the high-transmission state, the light field extends throughout the length of the medium and is able to maintain the director distribution in the perturbed condition until the intensity is reduced to a level considerably lower than the critical switch-on intensity. This explains the hysteresis observed in Fig. 1.

The y component of the cholesteric director is shown in Fig. 2 for zero field (dotted) and for an input field of intensity somewhat less than the critical value. Because the intensity inside the helix is inhomogeneous, the light-induced pitch dilation is spatially nonuniform. In particular, near the exit where $|\mathcal{E}_-| = 0$, there is no change in pitch. The maximum pitch dilation occurs near the front of the cholesteric where the field is enhanced through Bragg reflections. An approximate expression for this maximum pitch dilation, to first order in intensity, can be shown from (11) to be

$$\frac{\delta p}{p} = -\frac{\delta q}{q_0} = \left(\frac{\omega}{c}\right)^2 \frac{\epsilon_a{}^2 |\mathcal{E}_{\text{in}}|^2 L^2}{64\pi K_{22} q_0{}^2}, \tag{12a}$$

where $\mathcal{E}_{\text{in}}$ is the amplitude of the incident right circularly polarized field. This result holds for thin cholesteric films such that $\kappa L \lesssim 1$. For thick samples ($\kappa L > 2$) the resulting pitch dilation is independent of the length of the medium and is given by

$$\delta q = -q_0 |\mathcal{E}_{\text{in}}|^2 / 4\pi K_{22}(\omega/c)^2. \tag{12b}$$

This lack of dependence on the dielectric anisotropy ϵ_a and sample length can be understood through the following reasoning: The orientational forces responsible for pitch dilation are proportional to $(\epsilon_a L)^2$ while the orienting field decays exponentially within a characteristic length L_c proportional to $1/\epsilon_a$. Thus, for $\kappa L > 2$, the field does not "see" the extra length of medium beyond L_c.

To estimate the critical intensity for bistability, recall that the jump in transmission occurs when the light wavelength no longer lies within the stop band of the distorted helix. The width of this reflection band is $\delta\lambda' = \epsilon_a/\epsilon$, where $\epsilon = (\epsilon_\parallel + \epsilon_\perp)/2$ and λ' is a reduced wavelength given by $\lambda/p\sqrt{\epsilon}$. Thus at the threshold for bistability, the relative light-induced change in pitch is of the order of the bandwidth, or $|\delta q/q_0| \simeq \epsilon_a/\epsilon$. By means of (12b) this leads to a critical intensity of

$$|\mathcal{E}_{\text{in}}|^2 = 4\pi(\omega/c)^2(\epsilon_a/\epsilon)K_{22}, \tag{13}$$

for circularly polarized waves which satisfy the Bragg condition for the cholesteric helix. For thick samples the intensity predicted by the expression (13) is indistinguishable from the exact value obtained from (10). Using typical values of

$\epsilon_a/\epsilon = 0.1$, $K_{22} = 10^{-6}$ dyn, and an incident wavelength of 1 μm, we find a critical intensity for bistable reflection on the order of 1 MW/cm^2.

The connection between bistable reflection in cholesteric liquid crystals and the problem of nonlinear distributed-feedback structures[5] alluded to in the introduction can now be formally established. In Ref. 5, intense light changes the refractive index of a grating whose period remains fixed. For the cholesteric, the effect of the strong field is to alter its period. Either effect may be described in terms of a variable phase function $\Phi(z)$ in a periodic perturbation of the form $\cos[Qz + \Phi(z)]$, or equivalently as a chirp in the spatial frequency given by $\delta Q = d\Phi/dz$. Under the assumption that the frequency chirp is proportional to the local intensity, $\delta Q = -\gamma|E|^2$, it is easy to show that the Maxwell equations for the cholesteric reduce to precisely the same equations that describe nonlinear propagation in distributed-feedback structures. Of course, the quantitative results of this heuristic argument differ from the exact treatment presented here since the response of the director to applied fields is not simply proportional to the local intensity. The correct form of the nonlinear interaction must be obtained by solving the coupled Euler-Lagrange and Maxwell equations as done here.

In conclusion, we have solved the problem of nonlinear propagation in cholesteric liquid crystals and discovered an intrinsic optical bistability in these remarkable materials. This bistability is a result of light-induced pitch dilation, and occurs even in the absence of external reflectors.

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