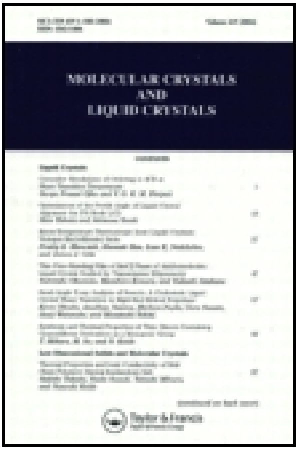

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Thermal Indexing in Cholesteric Liquid Crystals

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Thermal Indexing in Cholesteric Liquid Crystals

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We have theoretically worked out the effects of thermal indexing in cholesteric liquid crystals. We find that for light propagation parallel to the twist axis in a right (left) handed cholesteric, the right (left) circularly polarized state exhibits a large nonlinear optical (NLO) coefficient of the order of 10^{-4} cm^2/W. On the other hand the NLO coefficient for the left (right) circularly polarized state is of the order of 10^{-6} cm^2/W. We find that in both the cases the NLO coefficient is positive or negative depending upon whether the pitch of cholesteric increases or decreases with laser intensity. Further, due to variation in the average refractive index, the NLO coefficient for the right (left) circularly polarized state changes sign as we approach the Bragg band. In the case of laser beams with a Gaussian intensity profile we get self-focusing, self-divergence and self phase modulation. In the Mauguin limit we find a defect structure with a periodic array of disclination loops within the Gaussian beam.

Keywords: cholesterics; circular dichroism; laser absorption; nonlinear optics

1. INTRODUCTION

Nonlinear optics of liquid crystals has attracted a great deal of attention in recent times. The process of director reorientation in liquid crystals due to optical torque in a laser beam results in very high optical nonlinearities and has been studied in detail [1,2]. In one case [3] the nonlinear optical (NLO) coefficient found for a dye doped nematic liquid crystal was as high as 6 cm^2/W, which is really enormous. In these materials the dye molecules result in a space charge, which in turn leads to an enormous torque on the director. Very recently a colossal NLO coefficient of the order of 10^3 cm^2/W

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has been found in dye doped nematic liquid crystals due to surface induced nonlinear effect [4]. Such high optical nonlinearities in nematic liquid crystals find applications in optical image processing [5]. Cholesteric liquid crystals have many interesting optical properties like selective reflection of circularly polarized light, high optical rotation and circular dichroism. Applications based on these properties includes polarizers, apodizers, band pass and band rejection optical filters [6]. Nonlinear optics of such media is also of great technological importance. Winful [7] has predicted bistability in the reflection band of a cholesteric liquid crystal due to the torque of electric field of the laser beam on the local director. Lee *et al*. [8] have seen self-focusing in the Bragg reflection mode in a cholesteric liquid crystal. This self-focusing arises from the distortion of the cholesteric structure induced by the laser beam through the process of director reorientation. Optical nonlinearities can also arise from the heating of the material due to laser absorption. Here the nonlinearity is due to change in intrinsic refractive index due to laser heating, which is often referred to as thermal, indexing. Thermal indexing in nematics have been studied [9,10,11] widely. The nonlinear optical coefficient due to thermal indexing is very large near nematic to isotropic transition point. However, thermal indexing in cholesteric liquid crystals does not appear to have been studied in great detail.

We address ourselves to this process in this paper. We point out here that the absorption of a laser beam can lead to large optical nonlinearities in cholesteric liquid crystals. The mechanism involved here is entirely different from the director reorientation process found in other liquid crystals [1,2,7,8]. This thermal indexing effect is neither due to laser induced director reorientation nor due to changes in principal refractive indices of the material. It is entirely due to the change in pitch of the structure resulting from laser heating. We find that in a right (left) handed cholesteric, for a right (left) circularly polarized light wave the NLO coefficient is more than a billion times that found in usual nonlinear media like CS_2. On the other hand, for a left (right) circularly polarized light wave NLO coefficient is more than a million times than that found in usual nonlinear media. The sign of NLO coefficient is positive or negative depending on whether the pitch increases or decreases with laser intensity. We have also worked out the consequences of variation in principal refractive indices with intensity due to laser absorption. We find that under such circumstances the NLO coefficient of the right (left) circularly polarized wave changes sign near the Bragg reflection band.

If the incident laser beam has a radial intensity distribution then, we get a pitch profile within the beam width. When the pitch is not very large compared to the wavelength of light, the base states are right and left circular waves [12] whose refractive indices depend on the pitch. Therefore, inside the medium we get a refractive index profile for left and right circularly polarized waves. Thus, for an incident right or left circularly polarized plane wavefront the output is not a plane wavefront. This results in a self phase modulation leading to self-focusing or self-divergence of the circularly polarized beam. Lee *et al*. [8] have experimentally found self-focusing in the retro reflection mode of a cholesteric but the effect is due to a torque on the director by the optical field and not due to pitch change resulting from the absorption of the laser beam.

When the pitch is very large compared to the wavelength of light, the base states are linear vibrations polarized along and perpendicular to the local director. For the linear vibration along the local director the radial intensity distribution of laser beam results in a radial variation in refractive index again leading to self phase modulation. Here, again we do not have any torque on director. This effect is an analogue in cholesterics of an effect detected in nematic by Ono [13]. Finally, in the same geometry in compensated cholesterics pitch varies rapidly near the nematic point with intensity and can lead to a new defect structure.

2. THEORY

A cholesteric liquid crystal can be looked upon as a helically twisted birefringent medium. It has a periodically varying dielectric constant along its helical axis (z axis). If the light is propagating along z axis then, the Maxwell's equations for such a system becomes [12]

$$\frac{d^2}{dz^2}\begin{pmatrix} E_x \\ E_y \end{pmatrix} = -\left(\frac{\omega}{c}\right)^2 \hat{\varepsilon}(z)\begin{pmatrix} E_x \\ E_y \end{pmatrix}$$

The dielectric tensor $\hat{\varepsilon}(z)$ is given by,

$$\hat{\varepsilon}(z) = \frac{\varepsilon_\parallel + \varepsilon_\perp}{2}\begin{pmatrix} 1 & 0 \\ 0 & 1 \end{pmatrix} + \frac{\varepsilon_\parallel - \varepsilon_\perp}{2}\begin{pmatrix} \cos(2q_0 z) & \sin(2q_0 z) \\ \sin(2q_0 z) & -\cos(2q_0 z) \end{pmatrix}$$

where, ω is the frequency of light, $\varepsilon_\parallel$ and $\varepsilon_\perp$ are the principal components of the dielectric tensor whose principal axes are along and perpendicular to the local director which is taken to be the major axis of the dielectric tensor and q_0 is the wave vector corresponding to the pitch. The dispersion relation is given by:

$$\left(-k_0^2 + l^2 + q_0^2\right)^2 - 4q_0^2 l^2 - k_1^4 = 0$$

where $q_0 = 2\pi/P$, $k_1 = \frac{2\pi}{\lambda}(\bar{n}\delta\bar{n})^{1/2}$, $k_0 = \frac{2\pi}{\lambda}\bar{n}$, $\bar{n} = (n_e + n_o)/2$, $\delta\bar{n} = (n_e - n_o)$. Here P is the pitch of the cholesteric, n_e and n_o are the refractive indices parallel and perpendicular to the local director, λ is the wavelength of light and $(l + q_0)$ and $(l - q_0)$ are the wave vectors of right i.e., $(E_x + iE_y)$ and left i.e. $(E_x - iE_y)$ circular waves respectively.

In the two regimes $P < \left(\frac{\lambda}{\bar{n}}\right)$ and $\left(\frac{\lambda}{\bar{n}}\right) < P < \left(\frac{\lambda}{\delta\bar{n}}\right)$, the base states are right and left circularly polarized waves. The refractive indices n_R and n_L for these two base states are given by:

$$n_R = \mp\frac{\lambda}{2\pi}\sqrt{k_0^2 + q_0^2 - 2k_0q_0\left(1 + \frac{k_1^4}{4k_0^2q_0^2}\right)^{1/2}} + \frac{\lambda}{2\pi}q_0 \tag{1}$$

$$n_L = \frac{\lambda}{2\pi}\sqrt{k_0^2 + q_0^2 + 2k_0q_0\left(1 + \frac{k_1^4}{4k_0^2q_0^2}\right)^{1/2}} - \frac{\lambda}{2\pi}q_0 \tag{2}$$

In equation (1) the negative sign is taken for $P < \left(\frac{\lambda}{\bar{n}}\right)$ and the positive sign is taken for $\left(\frac{\lambda}{\bar{n}}\right) < P < \left(\frac{\lambda}{\delta\bar{n}}\right)$. It can be clearly seen that for $q_0 > 0$ i.e., for a right handed cholesteric structure n_R becomes complex within a range of pitches $\left(\frac{\lambda}{n_o}\right) > P > \left(\frac{\lambda}{n_e}\right)$. In this region the right circular wave does not propagate through the medium but gets reflected back. However, outside this range of pitches it propagates through the medium. On the other hand, left circular wave propagates unaltered through the medium in the two pitch regimes. We emphasize here that when a right or left circularly polarized laser light is going through the medium, there is no optical torque acting on the director. When, $P \gg \left(\frac{\lambda}{\bar{n}}\right)$ the base states are found to be linear vibrations parallel and perpendicular to the local director [12]. For the linear vibration parallel to the local director not only the absorption and hence thermal effects will be a maximum but also there will not be any optical torque on the director. Thus, in all the cases considered here we would get only the pure thermal indexing effects. Due to laser absorption the temperature of the medium increases. We can show (see Appendix) that this increase in temperature in a 100 μm thick sample is of the order of:

$$\delta T = I/A \tag{3}$$

Here, $A \approx 100\, W/\mathrm{cm}^{2\circ}\mathrm{C}$ and I is the laser intensity. We know that the wave vector q_0 of cholesteric is usually a linear function of temperature [18] and hence laser intensity. Therefore,

$$\frac{\partial q_0}{\partial I} = \beta \text{ (a constant)} \tag{4}$$

β can be a positive or negative number. From equations (1) to (4) we can calculate the NLO coefficient $\partial n/\partial I$ as a function of laser intensity.

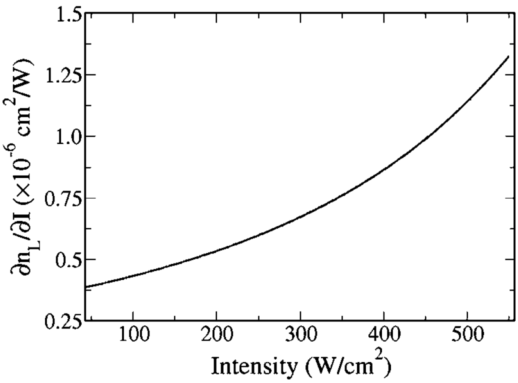
3. RESULTS AND DISCUSSION

The thermal indexing in a cholesteric depends on the way the pitch and other parameters vary due to laser absorption with concomitant raise of temperature. It is important to emphasize here that the optical absorption also affects the optics of cholesteric medium. Our calculations indicate in view of the smallness of absorption coefficient ($\alpha \sim 0.1\,\mathrm{cm}^{-1}$) for all practical purposes the dispersion curves for right and left circular waves remain unaltered. Therefore, the absorption coefficient (α) results only in a change in the pitch of the structure and does not enter into the optics of cholesteric. Further, in principle, right and left circular waves are absorbed to slightly different extents. Again, our calculations imply that the heating effect is nearly the same for both and is largely due to the average absorption. Throughout our discussion we consider only a right handed cholesteric and very similar results are valid *mutatis mutandis* for a left handed cholesteric. We consider here some special cases of thermal indexing in cholesterics.

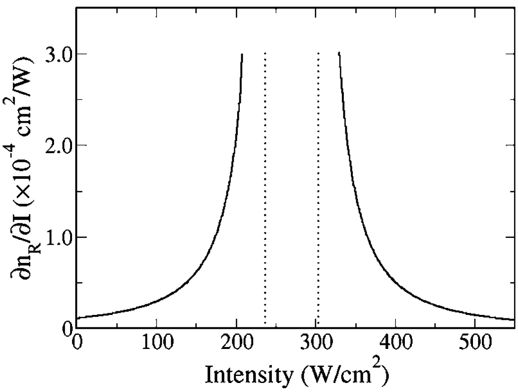
3.1. Large Optical Nonlinearity in Cholesteric

3.1.1. Rapid Variation of Pitch with Intensity

If β is a large number then the cholesteric pitch varies rapidly with the laser intensity or equivalently temperature. Then to a good approximation the accompanying temperature change is not much. We can then assume that the average refractive index ($\bar{n}$) and linear birefringence ($\delta\bar{n}$) do not change in the small range of intensities considered. To a first approximation we can take them to be constants and only the pitch of the structure varies with intensity. It is possible to have a cholesteric with the pitch either increasing or decreasing with an increase of temperature or equivalently laser intensity. In Figures (1) and (2) we show the theoretical dependence on laser intensity of NLO coefficient $\partial n_L/\partial I$ and $\partial n_R/\partial I$ for the left and right circular waves, respectively. We find that, even away from the reflection band, the NLO coefficient can be as high as $10^{-4}\,\mathrm{cm}^2/\mathrm{W}$ for the right circular wave. This is truly a large nonlinear optical effect being a billion times more than what we get in a usual nonlinear medium. On the other hand, for the left circular wave it is about $10^{-6}\,\mathrm{cm}^2/\mathrm{W}$ which, though much smaller than that for the right circular wave, is still very high i.e., a million times than that of a normal nonlinear material. Both the NLO



(a)



(b)

FIGURE 1 The variation of the NLO coefficient with intensity for left circularly polarized wave in 1(a) and for right circularly polarized wave in 1(b), when pitch increases rapidly with intensity i.e., β is a large negative number. The value of β is -157.1 cm/W. In the intensity range considered here the pitch varies from 0.3 μm to 0.54 μm. Here, $\lambda = 0.6$ μm, $\bar{n} = 1.5$ and $\delta\bar{n} = 0.1$. The region between dotted lines represents the reflection band.

coefficients are positive if pitch increases with intensity (Fig. (1)) and negative if pitch decreases with intensity (Fig. (2)). The NLO coefficient of the left circular wave monotonically increases with intensity when the pitch increases with intensity (see Fig. 1(a)); it exhibits a monotonic approach to saturation when the pitch decreases with intensity (see Fig. 2(a)). It may be remarked here that for both the circular waves with a radial intensity profile, we get self-divergence when NLO coefficient is negative and we get self-convergence, when NLO coefficient is positive. We again stress here that these NLO effects are consequences of heating

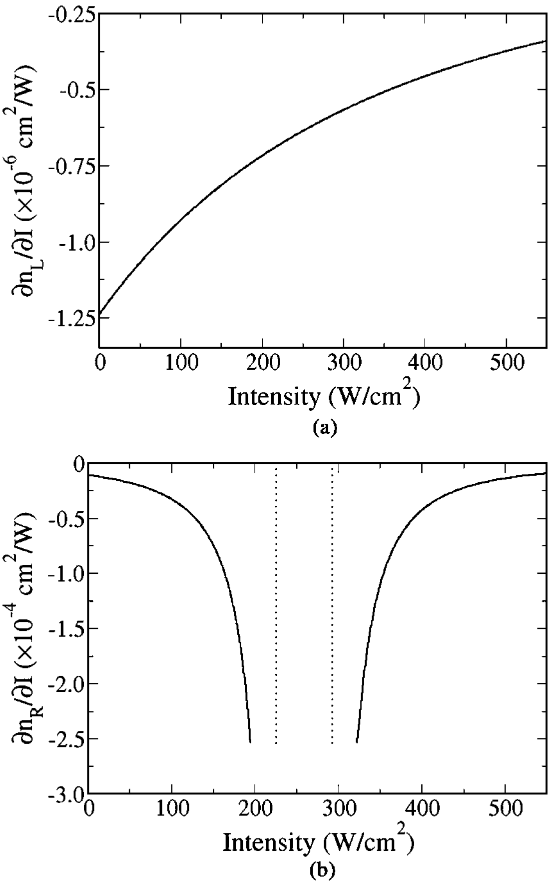


FIGURE 2 The variation of the NLO coefficient with intensity for left circularly polarized waves in 2(a) and right circularly polarized wave in 2(b), when pitch decreases rapidly with intensity i.e., β is a large positive number. The value of β is $+157.1\,\mathrm{cm/W}$. Other parameters are same as Figure (1).

due to laser absorption and not due to any torque exerted by electric field of laser beam on the local director.

3.1.2. Slow Variation of Pitch with Intensity

If β is rather small then the variation of pitch with laser intensity is slow. Then to get a considerable change in pitch we have to have a large intensity change (i.e., temperature variation). Then, over this intensity range the average refractive index ($\bar{n}$) of the medium cannot be taken to be a constant. An exact calculation of the variation of the average refractive index with the temperature is difficult for cholesterics [14]. All that one can say is that the average refractive index varies almost linearly with temperature. We have estimated the temperature variation of $\bar{n}$ from the experimental data of Demus and Wartenberg [15]. In a similar fashion absorption coefficient (α) is also temperature dependent. It can be assumed to vary exactly like average refractive index i.e., linear decrease with an increase of temperature. However, over the intensity (i.e., temperature) range of our interest we find the correction to absorption to be negligibly small and thus the temperature and pitch of the structure are to a good approximation determined by a constant absorption coefficient. Our results for intensity dependence of NLO coefficients are shown in Figures (3) and (4). For the left circularly polarized wave it is of the order of $10^{-5}\,\text{cm}^2/\text{W}$ and for right circularly polarized wave it is of the order of $10^{-4}\,\text{cm}^2/\text{W}$. It is clear from Figure 3(b) that when pitch increases with intensity, near the reflection band the NLO coefficient not only increases considerably for the right circular wave but also changes sign. It may be remarked here that we have not taken the variation of birefringence with intensity. This is reasonably a good approximation since we have considered the system to be far from cholesteric to isotropic transition.

It should be remarked here that we have excluded completely the region of the reflection band in our calculations. Also, on both sides of reflection band we have stopped our calculations at a point at which the polarization is nearly circular and the transmission coefficient is around 95% In the regimes considered here, the base states are right and left circular to a very good approximation (ellipticity is greater than 0.9).

3.2. Finite Beams

The theory and discussions so far pertained to the wide beam limit. We will now briefly discuss the effects in the presence of finite beams.

3.2.1. Spatial Solitons

It is well known that a finite beam undergoes self diffraction. As a result of which the beam spreads out as it propagates through the medium. However, in the presence of optical nonlinearity there will be self phase modulation. Thus, the nonlinear effect can compensate for diffraction process, resulting in a beam, which travels inside the medium without any change in its spatial intensity profile. Such beams are referred to as spatial solitons. Spatial solitons due to various NLO processes in different liquid crystals have been discussed in literature [16]. The exact compensation between the two processes takes place at

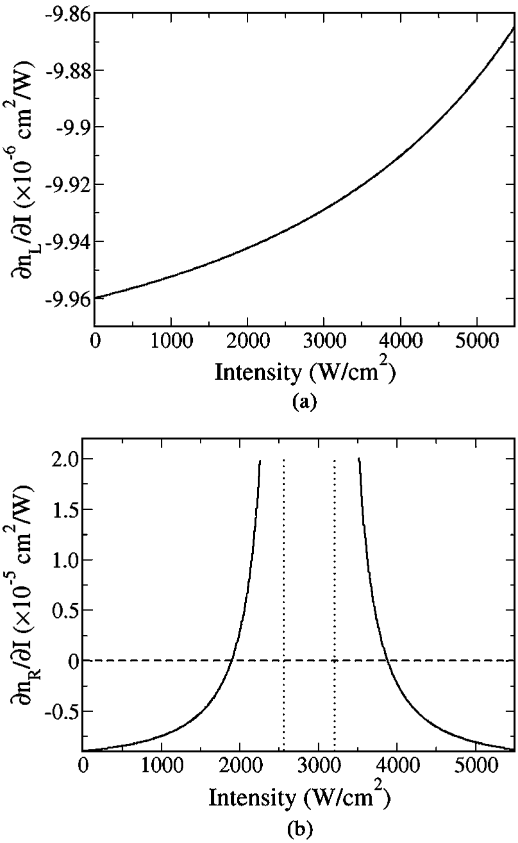
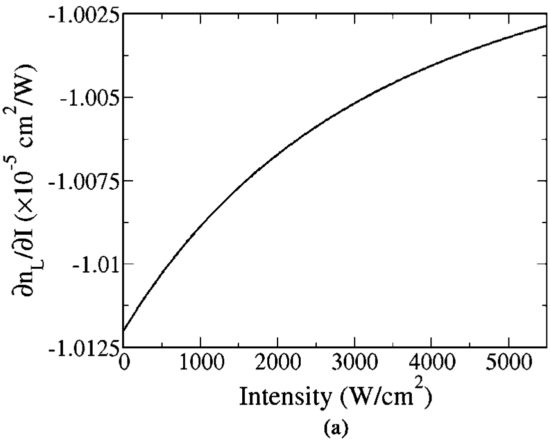


FIGURE 3 The variation of the NLO coefficient with intensity for left circularly polarized wave in 3(a) and right circularly polarized wave in 3(b), when pitch increases slowly with intensity i.e. β is a small negative number. The value of β is $-15.71\,\mathrm{cm/W}$. Here, to bring out the same change in pitch one needs 10 times more intensities, so average refractive index ($\bar{n}$) cannot be taken to be a constant with intensity. Other parameters are same as Figure (1). The value of $\partial\bar{n}/\partial I = 10^{-5}\,\mathrm{cm^2/W}$.

a critical laser power. If the NLO coefficient is positive we get a bright soliton with central peak intensity and if the NLO coefficient is negative we get a dark soliton with a central dip in intensity.

In our case in the low pitch cholesterics the base states are right and left circularly polarized waves with accompanying positive or negative NLO coefficients. Hence, we can expect the said soliton structure in cholesterics as well. Of course, they will exist only at a certain critical laser power.



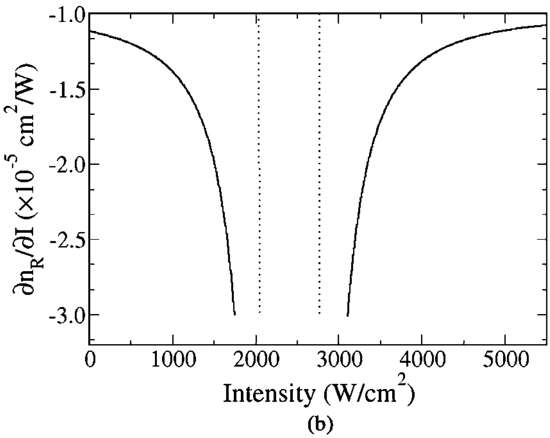


FIGURE 4 The variation of the NLO coefficient with intensity for left circularly polarized wave in 4(a) and right circularly polarized wave in 4(b), when pitch decreases slowly with intensity i.e. β is a small positive number. The value of β is $+15.71\,\mathrm{cm/W}$. Other parameters are same as Figure (1). The value of $\partial\bar{n}/\partial I = 10^{-5}\,\mathrm{cm^2/W}$.

3.2.2. New Beam Structures

In section 1 we restricted ourselves to the cases where $\left(\frac{\lambda}{\bar{n}}\right) < P < \left(\frac{\lambda}{\delta\bar{n}}\right)$ or $P < \left(\frac{\lambda}{\bar{n}}\right)$. In both the cases the base states namely right and left circular waves propagates through the structure. However, inside the reflection band centered at $P = \left(\frac{\lambda}{\bar{n}}\right)$ the right (left) circularly polarized light gets reflected back for a right (left) handed cholesteric. It is easy to see that if we have a right (left) circularly polarized beam with a peak intensity profile whose peak is inside the reflection band

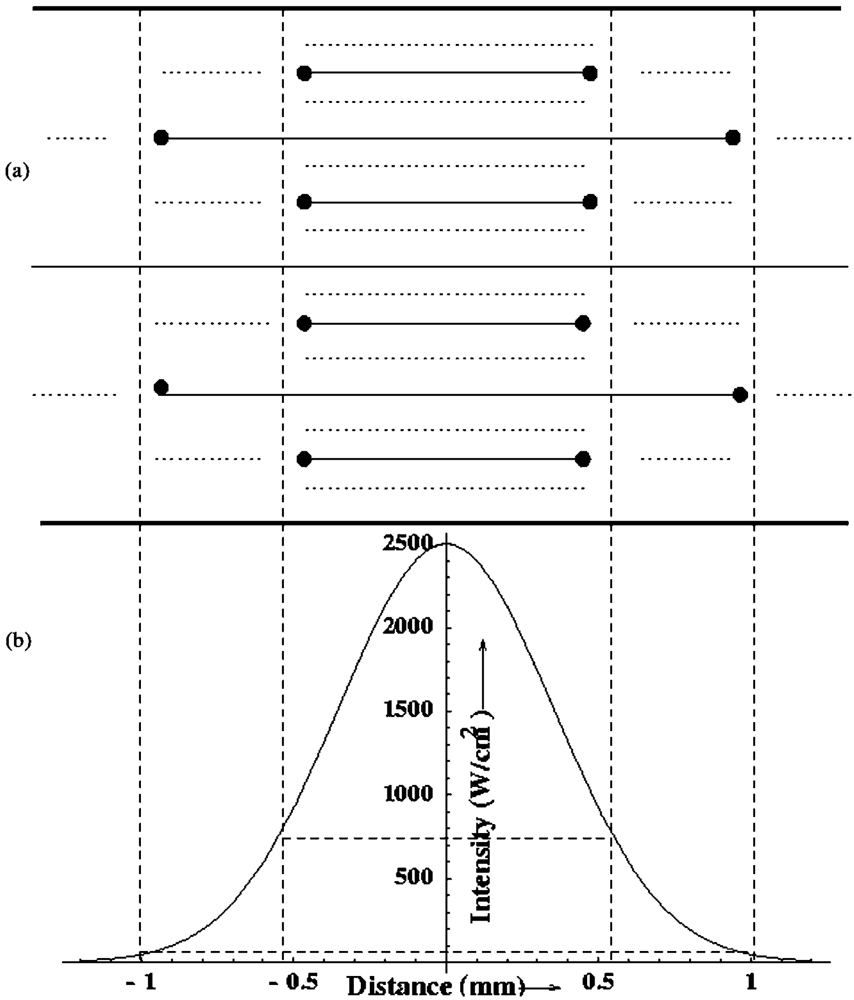


FIGURE 5 A typical calculation showing (in cross section) disclination loops in a cholesteric induced by a laser beam of Gaussian intensity profile. (a) cholesteric defect structure, (b) beam profile. Here, the pitch of cholesteric decreases with intensity. At zero intensity pitch is 4 μm and as the intensity reaches 2500 W/cm^2 at the peak of Gaussian beam, it becomes 1 μm. The cholesteric is assumed to be strongly anchored at the boundaries.

but with a tail outside the reflection band, then the central part of the intensity profile, will get completely reflected back and what comes out will be a ring like beam. Thus, new beam structures can be generated in such thermo-nonlinear cholesterics.

3.2.3. Defect Structure in Compensated Cholesterics

A compensated cholesteric liquid crystal [17] is a mixture of two or more cholesteric liquid crystals having different handedness. The peculiar property of such a cholesteric mixture is that it shows a structural transition with a change in handedness at a particular temperature at which it becomes a nematic. The pitch can also be changed by changing the concentration of individual components of the mixture. In these systems the pitch is extremely sensitive to the temperature and hence to the laser intensity if the system is also absorbing. The pitch variation is very large near the compensation or the nematic point. If the wavelength of light is very small compared to the pitch of structure near the nematic point then, as stated earlier the base states are linear vibrations parallel and perpendicular to the local director. If we consider a linear state parallel to the local director then there will not be any torque on the director but laser heating will be present. Hence, in a cholesteric strongly anchored at boundaries, in a finite beam with a peak central intensity we expect regions of large pitch differences. Such regions can be matched only by an array of twist disclination loops and not the usual χ or λ lines [12]. In the case of usual χ and λ lines the pitch of cholesteric does not alter. A typical representation of such a structure with disclination loops is shown in Figure (5). It may be pointed out that near a cholesteric to smectic transition also cholesteric pitch diverges. In such systems also we can find large variation in the pitch inside the beam and therefore, again we can expect a similar defect structure. However, in the wide beam limit where the intensity of light is almost constant over the whole wavefront, for a thick enough sample, the pitch may so vary that the handedness of the helix changes inside the medium. The intensity of light decreases progressively inside medium. Hence, the heating effect also progressively decreases. So, we can expect a right (left) handed cholesteric to go over to a left (right) handed cholesteric through a nematic state. For a cholesteric close to smectic transition in a similar geometry we expect a highly non uniformly twisted cholesteric. In both the cases, in low pitch limit we get the nonlinear optics of usual cholesterics as discussed already.

3.3. Effect of High Absorption

In our calculations we have considered the case of weak absorption. We find that even when the absorption coefficient (α) is 10 times larger there is no significant change in n_R, n_L and NLO coefficients. However, if α is very much more than this then the intensity and hence the pitch of the structure would not be constant through out the material and the simple theory that we have presented will not be applicable. Also, the heating effect may be so large as to melt the structure itself altogether at higher intensities. For these reasons we have not considered the case of strongly absorbing cholesterics in this investigation.

4. SUGGESTED EXPERIMENTS

To observe the effect of thermal indexing cholesteric liquid crystals, we suggest an experiment. Compensated cholesterics [17] are the right systems to observe these effects since they exhibit tunable pitch variation with temperature. A Gaussian laser beam will undergo self-focusing when the pitch decreases with temperature. By changing the concentration of components in compensated cholesteric one can have a situation where pitch increases with temperature. In that case a Gaussian beam will undergo self-divergence. The NLO coefficient can be measured in principle by conventional methods. In both the cases it is possible to so select the system that the defect state does not appear in the medium. In order to see the defect structure induced by the laser beam one has to work near the nematic point where the pitch diverges with temperature.

5. CONCLUSION

We find that a simple model leads to large thermal nonlinearities in weakly absorbing cholesteric liquid crystals. We find that in a right (left) handed cholesteric the NLO coefficient $\partial n/\partial I$ for the right (left) circular wave is more than a billion times greater than that found in ordinary nonlinear materials. It is about $10^{-4}\,\mathrm{cm}^2/\mathrm{W}$ as compared to $10^{-14}\,\mathrm{cm}^2/\mathrm{W}$ in case of usual nonlinear materials. It increases in magnitude as we approach the reflection band. For the same structure the left (right) circular wave has a nonlinear optical coefficient of the order of $10^{-6}\,\mathrm{cm}^2/\mathrm{W}$, which again is million times greater than that found in the usual nonlinear materials. It monotonically increases when the pitch increases with laser intensity and shows a smooth saturation behavior when the pitch decreases with laser intensity. The NLO coefficient for both the waves is positive when the pitch increases with laser intensity and negative when pitch decreases with laser intensity. When the principle refractive indices also vary with laser intensity then we find that for both the circular waves the NLO coefficient is of the order of $10^{-5}\,\mathrm{cm}^2/\mathrm{W}$. Further, the NLO coefficient even changes sign for the right (left) circular wave as we approach the reflection band. In the case of laser beams with a Gaussian intensity we get within the beam a variation in the pitch of the structure. If the pitch variation is small compared to the intrinsic pitch then we find that in the de Vries limit both right and left circular states have a refractive index profile across the wavefront leading to self-focusing, self-divergence and self phase modulation. On the other hand, in the Mauguin limit the base states are linear vibrations along and perpendicular to the local director. For the vibration parallel to the local director the pitch variation can become enormous compared to the intrinsic pitch. Then we find a defect structure with a periodic array of disclination loops within the beam. New beam structures can be generated by using the selective reflection of cholesteric liquid crystals.

APPENDIX

Laser Heating

The pitch of a cholesteric is generally a sensitive function of temperature. Here, we establish a relation between temperature rise and the laser absorption. It depends on many experimental and material parameters like cell thickness, beam spot size, thermal conductivity etc. It is possible to obtain to a first approximation, the temperature increase from the well known heat conduction equation [19]

$$\kappa \nabla^2 T - \rho c_v \frac{\partial T}{\partial t} = -\alpha I \tag{5}$$

where α is the optical absorption coefficient, κ is the thermal conductivity, ρ is the mass density and c_v is the specific heat at constant volume. For a beam width larger than the sample thickness, under steady state conditions ($\frac{\partial T}{\partial t} = 0$), we can estimate the increase in temperature δT from Eq. (5). It is given by:

$$\delta T \approx \left(\frac{d^2 \alpha}{\kappa} \right) I \tag{6}$$

Here d is the sample thickness. Thus the temperature rise is a linear function of the laser intensity and has a square dependence on sample thickness. Typically, $\kappa \approx 10^{-3}\,\mathrm{W/^\circ C\,cm}$ for $\alpha \approx 0.1\,\mathrm{cm}^{-1}$ and in a $100\,\mu\mathrm{m}$ thick sample

$$\delta T \approx I/A \tag{7}$$

where, $A \approx 100\,\mathrm{W/^\circ C\,cm^2}$. Further, over a sample thickness of $100\,\mu\mathrm{m}$ the attenuation of the wave inside the medium is about 0.1%, which can be ignored. In principle we should also take into account the anisotropy in thermal conductivity and optical absorption in liquid crystals. Therefore, a proper analysis of heat conduction equation leads to involved calculations. As an example we may refer to the work of Janossy [20], who has studied the case of a homeotropic nematic.

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