Lasing Thresholds of Cholesteric Liquid Crystals Lasers

W. Cao[1], A, Marino[2], G. Abbate[2], P. Palffy-Muhoray[1], B. Taheri[1]

1) Liquid Crystal Institute, Kent State University, Kent, OH 44242, USA
2) Dipartimento di Fisica, Università di Napoli "Federico II", 80125 Napoli Italy

Abstract

Cholesteric liquid crystals (CLCs) are one dimensional photonic band-gap materials. Due to distributed feedback, low threshold mirrorless lasing can occur in dye-doped and pure CLCs. In order to optimize the lasing conditions, we have studied the dependence of the lasing threshold on dye concentration and sample thickness. In particular, we have studied dye concentrations in the range of 0.1-3.0*wt%*, and cell thicknesses are in the range of 5-50μm. We have found that the system has a shallow lasing threshold minimum and can operate efficiently in the range of dye concentrations of 0.3-2.4*wt%* and sample thicknesses of 10-50μm. We discuss the physical processes responsible for the observed behaviour.

Keywords: mirrorless lasing, lasing threshold, cholesteric liquid crystal, photonic band-gap, density of states

Introduction

Due to their birefringence and periodic structure, cholesteric liquid crystals (CLCs) in the helical cholesteric phase are one-dimensional photonic band gap materials [1]. As a consequence, the fluorescent emission in these materials is suppressed in the reflection band and is enhanced at the band edges. This gain enhancement and distributed feedback effect can give rise to low threshold mirrorless lasing at the band edge in a variety of liquid crystal materials [2-15].

Until recently, research on lasing in CLCs has focused primarily on feasibility and fundamental aspects in different materials. With the first demonstration of a CLC fiber laser for sensor application [16], research is needed at the technological front, to enable the design and optimization of CLC lasers. In this paper, we report the results of our study of the dependence of the lasing threshold on dye concentration and liquid crystal sample thickness in dye-doped CLC lasers.

Materials and Setup

The liquid crystal used in this study was a mixture consisting of 78*wt%* cholesteric BLO61 and 22*wt%* nematic E7 (EM Industries). The mixture was doped with the laser dye DCM (4-(dicyanomethylene)-2-methyl-6-(4-dimethyl-amino-styryl)-4-H-pyran) with

concentration in the range of 0.1*wt%* to 3.0*wt%*. A glass cell, with windows treated to produce homogeneous alignment, was filled with this mixture. On cooling slowly from the isotropic phase, the sample showed a defect-free planar texture. The material has a right-handed helical structure, with a reflection band between 545*nm* to 608n*m* that overlaps the maximum of the fluorescence spectrum of dye. For the study of the dependence of lasing threshold on dye concentration, the thickness of the CLC sample in the cell was 23μm. For the study of the dependence of lasing threshold on sample thickness, a wedge-shaped cell, with thickness ranging from 5μm to 50μm, was used. The dye concentration for the wedge cell was 0.5*wt%*. The cells were pumped either by a Q-switched Nd-YAG laser delivering 7.5*ns* pulses, or a mode-locked Nd-YAG laser delivering 40*ps* pulses, at 532*nm*. The pump beam was focused by a lens to a beam waist of 25μm at the sample. Details of the experimental setup and instrumentation have been described elsewhere [7].

Results and Discussions

Threshold Measurements

In our well-aligned dye-doped CLC samples, the lasing peak with the lowest lasing threshold is at the low energy edge of the reflection band. We measured the intensity of this lasing peak when it was stable; measurements were averaged over 10 pulses. Fig. 1 shows typical behaviour of laser intensity as function of pump energy. The intensity was measured at the laser peak at 608.6*nm*. The lasing threshold, determined from the intersection of the two straight line fits to the intensity below and above threshold, is 64.0*nJ* for this sample, as shown.

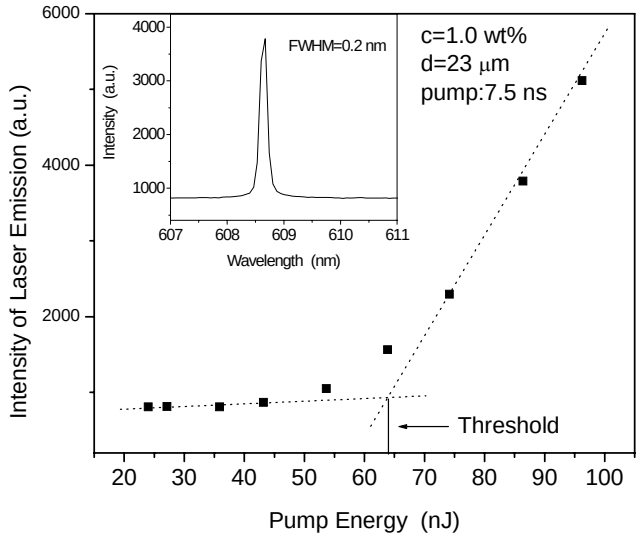

Fig. 1 Intensity of CLC laser emission as function of pump energy.

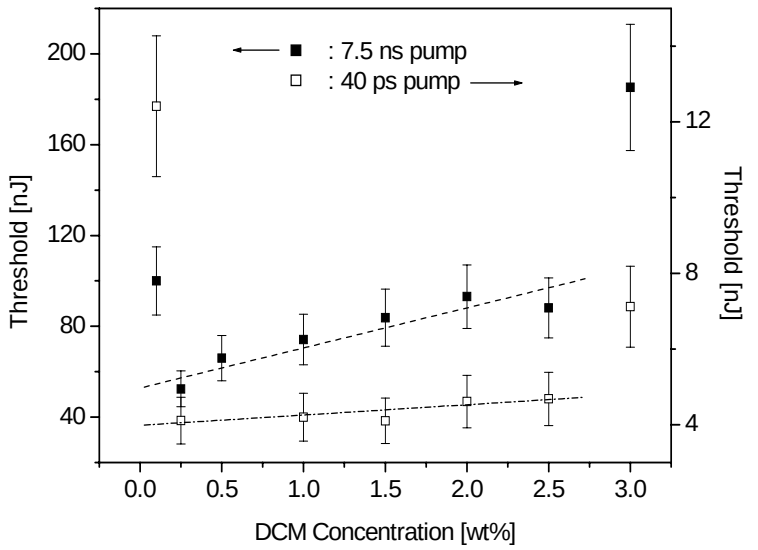

Fig. 2 Dependence of lasing threshold on dye concentration with nanosecond and picosecond pump pulses. The dotted lines are linear fits in the concentration region of 0.25 *wt%* to 2.5 *wt%*.

Threshold dependence on dye concentration

The threshold dependence on dye concentration is shown in Fig. 2. The threshold exhibits similar behaviour with both nanosecond and picosecond pump. At both the low (0.1*wt%*) and high (3.0*wt%*) ends of the concentration range, thresholds are significantly higher. We propose the following explanation. In order to lase, the medium has to provide gain greater than the gain threshold to overcome loss. Since nominally the gain is proportional to dye concentration, there can be no lasing if the dye concentration is below the concentration corresponding to the gain threshold. The lasing threshold therefore diverges as this critical dye concentration is approached from above.

At high dye concentration, it appears that the liquid crystal mixture becomes saturated with the dissolved dye. Above a certain concentration, dye crystals, coexisting with the liquid, can exist in the medium. At 3.2*wt%* concentration, dye crystals can be clearly seen in the mixture under a microscope. The scattering and absorption of both pump beam and lasing beam by these dye crystals are likely the cause of the observed strong increase of the lasing threshold at high dye concentrations.

In the intermediate dye concentration region, between 0.25 *wt*% and 2.5 *wt*%, the lasing threshold increases linearly with dye concentration. This increase is likely due to quenching of the fluorescence by the dye. The dye concentration in conventional dye lasers using DCM as active medium is typically of the order of 10^{-4} *[M]* while in our samples, the DCM concentration is of the order of 10^{-2} *[M]*. At these relatively high concentrations, bimolecular process involving dye molecules, such as excimer formation, are likely important, resulting in quenched fluorescence as shown in Fig. 3. The fluorescence spectra were measured for samples with different dye concentrations. With the same experimental conditions (pump energy, pump position, spectrometer

parameters), the fluorescence intensity at 608.5*nm* is quenched, by a factor 2.6, when the dye concentration is increased from 0.5*wt%* to 2.5*wt %*. Correspondingly, the lasing threshold is increased, by a factor of 1.6.

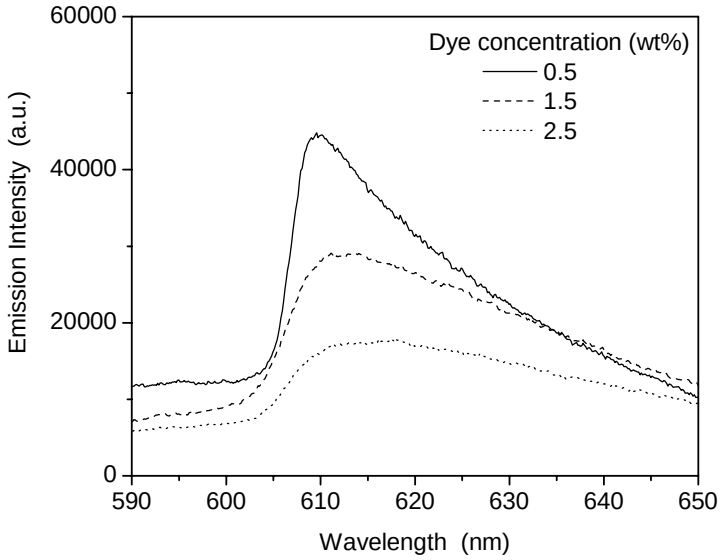

Fig. 3 Fluorescence quenching: fluorescent emission for different dye concentrations. The pump pulsewidth is 7.5*ns*.

This quenching of the fluorescence, together with the shifting of the emission peak to longer wavelengths (Fig. 3) is characteristic of excimer formation. DCM molecules are polar, due to the electron-acceptor (-CN) and the electron-donor (-N) groups attached to the conjugated molecular skeleton. The molecular dipole moment is 6.1*D*. The excited state of DCM is even more polar, with a dipole moment of 26.3*D* [17]. An excited DCM molecule and a ground state DCM molecule can form an excimer due to charge transfer interaction (Fig. 4) [18]. The excimer can decay by returning to an excited DCM and a ground state DCM, or by emission of a photon or by dissociation. In the latter two cases, the excited population of DCM molecules is quenched. Using a picosecond pump and a streak camera, we measured the fluorescence decay at 608*nm* as shown in Fig. 5. For dye concentration of 0.5*wt%*, the decay curve was fit well by a single exponential, with a lifetime of 1.25*ns*. This is the typical lifetime of an excited DCM molecule. For dye concentration of 2.5*wt%*, the decay is double exponential, with lifetimes of 1.20*ns* and 0.32*ns*. The much shorter-lived excited state species, with a lifetime 0.32*ns*, is the DCM excimer.

In the intermediate dye concentration region from 0.25 *wt*% to 2.5 *wt*%, the threshold energy increases from 4.13*nJ* to 4.72*nJ* for picosecond pump, while it increases from 53*nJ* to 95*nJ* for the nanosecond pump. The less pronounced dependence of lasing threshold on dye concentration for the picosecond pump is consistent with the picture of fluorescence quenching by excimer formation. Since the lifetime of excimer is 320*ps*, the excimer can only form once after 40*ps* pump, while the quenching process can likely repeat a number of times during the 7.5*ns* pump pulse.

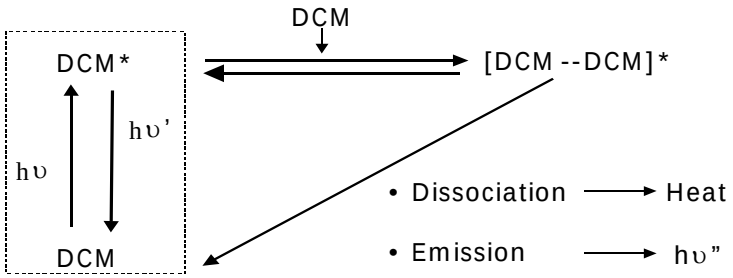

Fig. 4 Excimer formation and decay. DCM*: excited DCM molecule, [DCM--DCM]*: DCM excimer. The dotted frame shows the process when dye concentration is low.

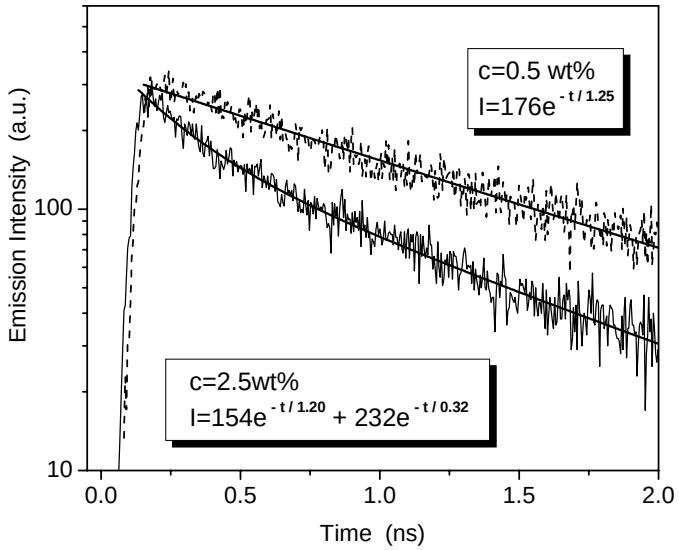

Fig. 5 Fluorescence decay at 608*nm* with DCM concentrations of 0.5*wt%* and 2.5*wt%*. The pump pulsewidth is 40*ps*.

Threshold dependence on cell thickness

We have measured the lasing threshold as function of sample thickness at the fixed dye concentration of 0.5*wt%*. The results are shown in Fig. 6. The threshold behaviours are the same for both nano- and picosecond pump pulses: with increasing cell thickness, the threshold decreases to reach a minimum, and then gradually increases. In addition, with increasing cell thickness, the lasing wavelength blue-shifts several nanometers, towards the expected band edge for a semi-infinite sample.

The observed behaviour suggests competition between processes with opposite thickness dependence. In order to understand this, we make comparison with the threshold of a typical Fabry-Perot (F-P) cavity laser. The threshold gain constant of a F-P cavity laser is [19]:

$$\gamma_{th}^{F-P} = \alpha - \frac{\ln(r^2)}{d} \tag{1}$$

where α is the absorption coefficient, r is the reflection coefficient of the mirrors and d is the cavity length. The gain must therefore compensate for both absorptive and cavity losses to achieve lasing. The threshold pump energy is $E_{th}^{F-P} = A\gamma_{th}^{F-P} d$ [19], where A is a constant related to the pump properties. It is straightforward to show that in a F-P cavity, the maximum value of the density of states (DOS) is related to the reflection coefficient by

$$DOS_M = \frac{1+r^2}{1-r^2} \tag{2}$$

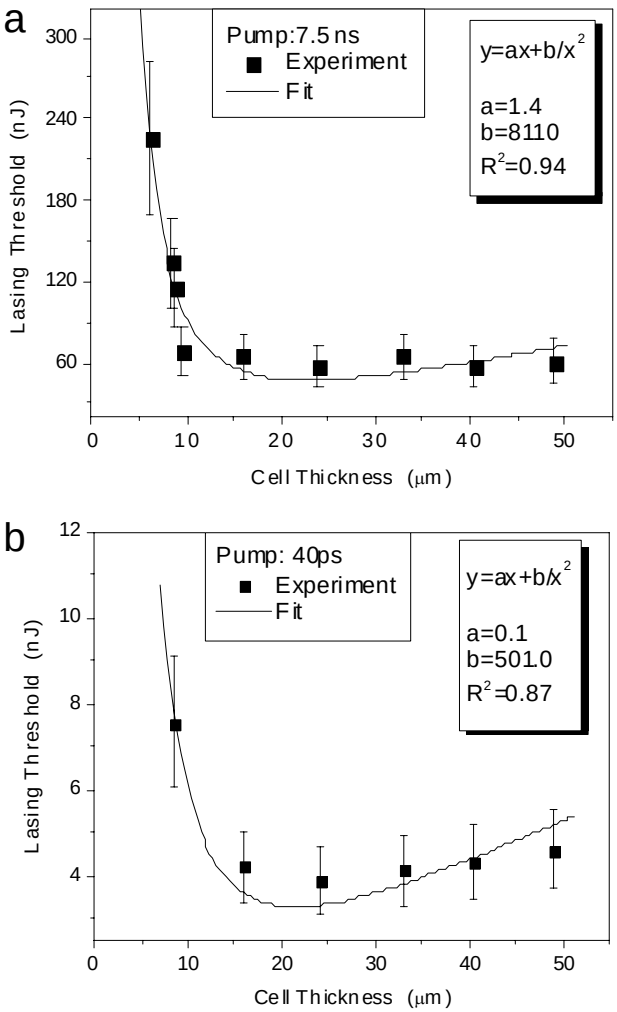

Fig. 6 Lasing threshold as function of sample thickness. (a) nanosecond pump (b) picosecond pump

and it follows that

$$\ln r^2 = \ln \frac{1 - DOS_M}{1 + DOS_M} \simeq -\frac{2}{DOS_M} \tag{3}$$

for large DOS_M. In CLC, the salient optical properties of the system are contained in the DOS. In analogy with the F-P laser, we write the threshold gain constant of the CLC laser as

$$\gamma_{th}^{CLC} = \alpha + \frac{\beta}{DOS_M \bullet d} \tag{4}$$

where α is the absorption coefficient. We have calculated the DOS for cells with different thicknesses shown in Fig. 7. The details of the calculation will be published elsewhere. The highest DOS peak is close to the reflection band edge and lasing occurs at this wavelength. We have found that here $DOS_M = cd^2$, where c is a fitting constant. This result is similar to that of Dowling [20], who analytically calculated the DOS as function of sample thickness in a dielectric medium with alternating refractive indices. We then write for the pump energy at lasing threshold:

$$\begin{aligned} E_{th}^{CLC} &= B\gamma_{th}^{CLC} d \\ &= B(\alpha + \frac{\beta}{DOS_M \bullet d})d = B(\alpha + \frac{\beta'}{d^3})d \\ &= (B\alpha)d + \frac{(B\beta')}{d^2} \end{aligned} \tag{5}$$

This describes the dependence of lasing threshold on sample thickness d. We have fit experimental data to this expression, and found good agreement, as shown in Fig. 6. The first term accounts for absorption loss, while the second term accounts for the cavity loss. For a distributed feedback laser without absorption losses, Kogelnik and Shank found that the threshold gain constant $\gamma \approx c'/d^3$, c' is a constant related to material and lasing wavelength, and d is the cavity length [21]. Thus the dependence of threshold on cavity length agrees with our results.

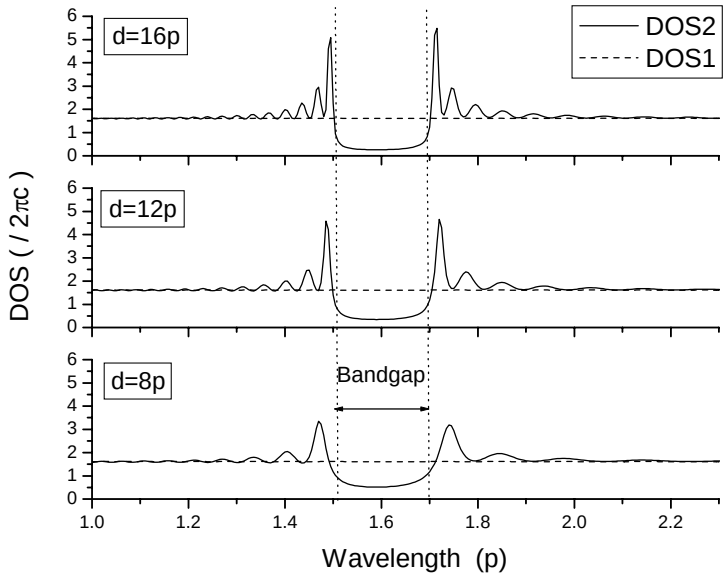

Fig. 7 Calculated DOS for the two normal modes as a function of cell thickness. The dotted lines are the reflection band edges of a semi-infinite sample, $n_o = 1.51$, $n_e = 1.70$.

Conclusions

We have studied the lasing thresholds of CLC lasers as a function of dye concentration and of sample thickness. Our results show that the lasing threshold is a minimum when the dye concentration is in the range of 0.1-3.0w*t%*, and the sample thickness is in the range of 5-50μm. We have proposed mechanisms to explain theobserved behaviour. We have shown that the DOS can be used to quantify the lasing behaviour, and expect that this approach can be useful in the design of CLC lasers for device applications.

Acknowledgement: This work was supported in part by the NSF under DMR-0132611.

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