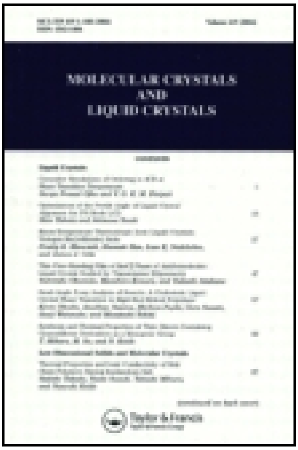

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Low Threshold Lasing in Cholesteric Liquid Crystals

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Cholesteric liquid crystals, due to their periodic structure and large birefringence, are 1-d photonic band-gap materials. Circularly polarized light of the same handedness as the cholesteric structure cannot propagate in the reflection band. We have studied the effects of this band-gap structure on the fluorescence spectrum of dyes dissolved in the cholesteric liquid crystal, as well as on the fluorescence spectrum of the liquid crystal itself. We have found that emission is enhanced at the band edges, and, above a certain pump threshold, lasing occurs both in dye-doped and in pure liquid crystals. Extremely low lasing thresholds and high efficiencies were observed in dye-doped systems. The results suggest an active energy transfer mechanism between the LC host and the fluorescent dye molecules.

INTRODUCTION

Microcavity effects in light emitting materials have received considerable attention in the recent years. It has been shown that the presence of a cavity can enhance fluorescent emission at the resonant wavelengths of the cavity[1]. Studies have been performed on a variety of organic materials, including liquid crystals, using external resonators. Lasing in these systems is of great interest, since they present great potential for achieving low threshold and high efficiency lasing.

In materials whose dielectric constant varies periodically in space, a range of frequencies exists for which classical electromagnetic wave propagation is forbidden. In this band-gap, the emission of photons is suppressed; this can lead to photon localization[2]. Conversely, at the band edge, where the propagation length can become infinite, emission is enhanced. Materials with periodic structure are therefore expected to give rise to these distributed feedback and cavity effects, where the fluorescent emission is suppressed in the band gap and enhanced at the band edges. These materials can therefore be used as efficient laser materials. The use of periodic dielectric structures to exploit these mechanisms has been proposed and investigated in the field of semiconductor lasers[3].

Cholesteric liquid crystals (CLC), with their periodic helical structure, exhibit a selective reflection band. Their large birefringence results in strong Bragg reflection of light with the same helicity as the cholesteric. This intrinsic characteristic has led to their use in display applications[4]; they have also been used as end mirrors in laser cavities[5]. The optics of cholesterics has been the subject of a considerable investigation. It is straightforward to show that, for propagation along the helix axis, eigenmodes of the optical field at the band edges are two counterpropagating circularly polarized waves with the same helicity as the cholesteric and with infinite propagation length. These form a circularly polarized standing wave. The presence of standing waves suggests that these materials may be well suited for use as hosts in distributed feed back lasers. In the reflection band, emission from a fluorescent center is suppressed due to band gap structure of the host, resulting in photon trapping. In the appropriate configuration and with suitable pumping, stimulated emission can occur, and the system can be made to lase. The possibility of lasing in CLC was first proposed by Goldberg and Schnur[6] in 1973; experimental investigations have been carried out subsequently[7]. Lasing in dye-doped CLC has been unambiguously demonstrated for the first time during the past year by V. Kopp et al. at Queen's College at CUNY [8], and by us at the Liquid Crystal Institute[9]. The enhancement of fluorescent emission at the band edges in these systems can be understood on the basis of the two equivalent descriptions: photonic band gap[8] and distributed feed back systems[10]. We have recently observed, for the first time, mirrorless lasing in a pure liquid crystal system[11].

We have carried out a systematic study of the response of our systems on dye concentration, configuration and pumping conditions. This enabled optimization of system parameters resulting in an increase in efficiency and record low lasing thresholds, more than an order of magnitude lower than reported earlier[8,9]. Our observations indicate energy transfer between the dye and liquid crystal host. Based on these results, we were able to achieve mirrorless lasing in a pure liquid crystal system. Here we present the results of our experiments demonstrating the alteration of the fluorescence spectrum of dyes by cholesterics, lasing in dye-doped cholesterics, and finally, lasing in pure liquid crystals.

EXPERIMENTAL SETUP

Several 25μm thick cells were fabricated using glass coated with ITO and with a Nissan 2555 alignment layer. The cells were filled with a cholesteric liquid crystal and DCM laser dye mixture. In addition to having a high quantum yield, DCM showed good solubility in our liquid crystals. Several mixtures were prepared, with DCM dye concentrations ranging from 0.5% to 3% in mixtures of BLO61 and E7. The location of the reflection band with respect to the emission band of the dye was adjusted by varying the concentrations of the liquid crystal constituents.

To study the effects of the band gap on the fluorescence characteristics of DCM, we used a Spex Fluorolog 5 spectrofluorometer. The fluorescence emission normal to the surface of the cell was studied as function of the excitation wavelength. The polarization of the emission was determined. The reflectance of the sample as function of wavelength was measured using an Ocean Optics spectrometer. Our initial experiments focused on the effects of the reflection band on the emission characteristics of the guest dye. Several cells were used to investigate the emission characteristics of the dye in the liquid crystal host. Fig.1 shows the emission characteristics of purified DCM in a cholesteric liquid crystal. The solid line shows the total emission, the dashed line shows left circular emission with handedness opposite to that of the cholesteric host. The salient feature is the suppression of emission in the band gap, and the enhancement of emission at the band edge.

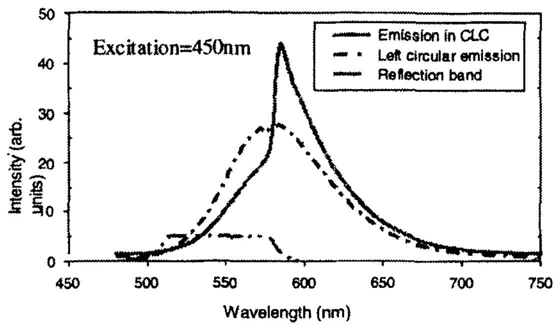

Fig 1. Effect of the reflection band on the emission characteristics of the dissolved dye. The dashed line shows the unaltered emission. The solid line is the emission in the CLC host.

For the lasing experiments, picosecond pulses from a Nd:YAG were frequency doubled and passed through a waveplate-polarizer attenuator, and focused on the cell with an $f = 10cm$ lens. Fig. 2 shows the experimental setup used in the lasing experiments. The laser emission was detected using a fiber connected to an Ocean Optics spectrometer. The efficiency and laser threshold measurements were performed with the same setup. Energies of the pump beam and laser emissions were measured using a Scientec energy meter. The energy of laser emission was measured as function of the pump intensity and dye concentration.

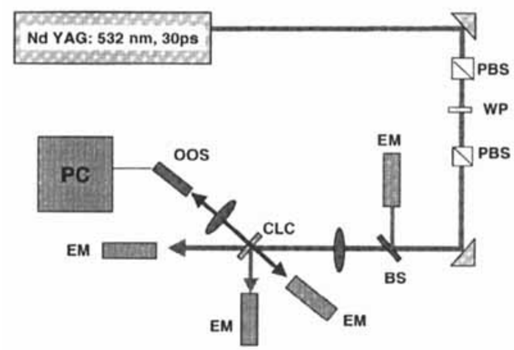

Fig.2. Experimental setup for generating lasing. PBS=Polarizing beam splitter, WP= $\lambda/4$ plate, BS= beam splitter, EM=energy meter, OOS=Ocean Optics spectrometer.

Samples were prepared so that the unperturbed peak emission coincided with either the high or the low energy band edge of the cholesteric liquid crystal. Fig. 3 shows the experimental results obtained with such a system. In the figure, the dashed line shows dye emission prior to lasing and the solid line is the laser emission. In addition to nonlinear gain and line narrowing, strong directional emission was observed.

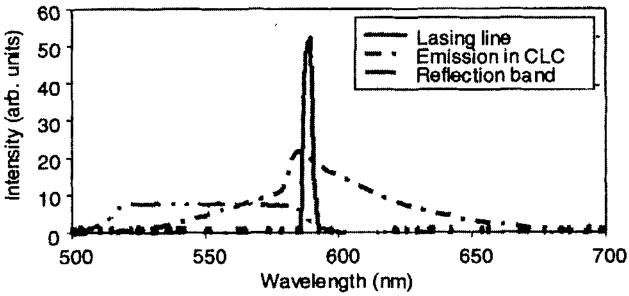

Fig. 3. Lasing of the dye-CLC system. The solid line shows the resolution limited emission from the dye-doped CLC. The long dash shows the emission characteristics at low pump energies below lasing threshold.

RESULTS

To maximize the efficiency of lasing in our system, we investigated the role of dye concentration. In our samples we found that lasing is most efficient at the low energy band edge. Fig. 4 shows measurements of emitted energy versus pump energy. In these measurements, the pump beam waist was $100\mu m$.

It was found that the threshold decreased as the dye concentration was increased from 0.5 to 2.0%. The lowest threshold observed was at 10nJ pump energy. Beyond this, the threshold value increased with increasing dye concentration. The lasing efficiency increased with dye concentration and peaked at 20% for the 2% dye mixture. The lasing output had a saturated maximum of 30 nJ for all dye concentrations. The origin of this limit as well as the dependence of the lasing efficiency and threshold on dye concentration are currently under study.

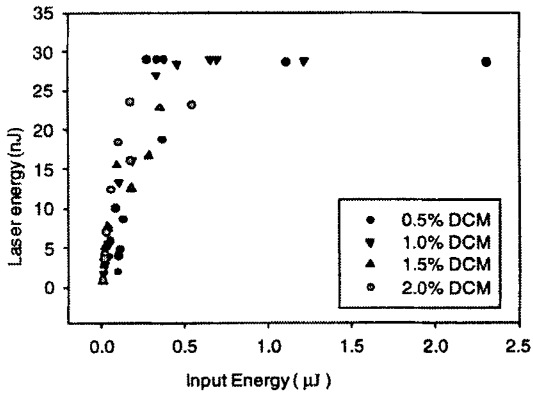

Fig. 4. Emission energy as function of input pump energy for different dye concentrations.

To investigate the effects of pump wavelength, we measured the excitation spectra of our systems. Fig. 5 shows the excitation spectra of one mixture for the 582nm emission line. The peak observed at 350nm corresponds to the peak absorption of the liquid crystal; it agrees with the peak in the excitation spectra of the 400nm emission line observed in our pure liquid crystal mixture without the dye. This suggests energy transfer between the liquid crystal host and the guest dye.

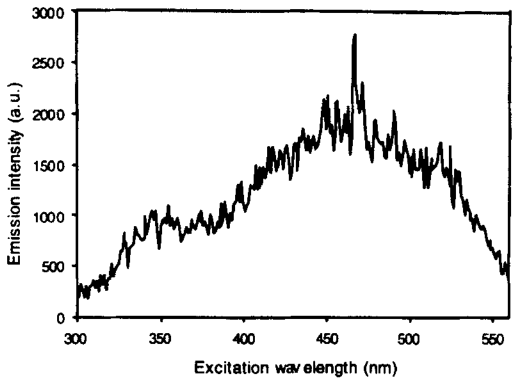

Fig. 5. Excitation spectra of the DCM dye dissolved in the CLC.

After measurements of the excitation spectra, the emission arising from pumping at 355nm was studied. Lasing by the dye could be observed under this excitation; however, the threshold was approximately one order of magnitude higher than that observed with 532nm. In addition, there was evidence of dye degradation at this wavelength.

Under 355nm excitation, strong blue/UV emission was observed from the pure liquid crystal. Several cells were prepared with liquid crystal mixtures without dye. These samples were pumped with third harmonic of our picosecond Nd:YAG laser in the configuration shown in Fig. 2. We observed mirrorless lasing in pure liquid crystals without any dye at 400nm when the low energy edge of the reflection band was tuned to this wavelength. Fig. 6 shows the laser line from the mixture as well as from the pump laser. The emission decreased with time, indicating degradation of the dye, and had a high threshold. Lasing in pure liquid crystals is under continued study.

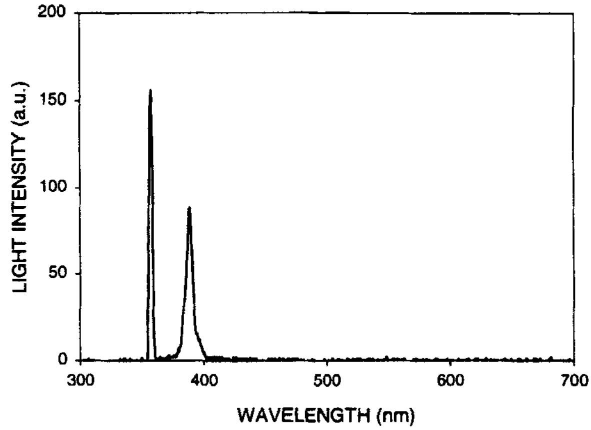

Fig. 6. Lasing emission in a pure liquid crystal material without dye. The left peak is the pump at 355nm.

SUMMARY

We have observed lasing with record low lasing thresholds in dye doped cholesteric liquid crystals. Mirrorless lasing occurred at several wavelengths, determined by the edges of the reflection band of the cholesteric liquid crystal mixtures. We have observed lasing in the range of wavelengths from 575nm – 614nm. Our experimental results indicate energy transfer between the liquid crystal host and the dissolved dye. We have observed, for the first time, mirrorless lasing in a pure liquid crystal material.

Acknowledgment

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