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Ultraviolet lasing in cholesteric liquid crystals

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We report the observation of stimulated emission and mirrorless lasing in pure cholesteric liquid crystals. The lasing action is attributed to the combination of the fluorescence and the distributed feedback that are due to the inherent periodic structure of the liquid crystal. If the reflection band matches the intrinsic emission of the cholesteric liquid crystal, the crystal becomes a natural laser material, which will self-lase, without any optical elements or the addition of dyes, under picosecond excitation at 355 nm. Samples have been made to lase at different wavelengths in the near UV by shifting of the edge of the reflection band in the range of 385–405 nm. Typical linewidths observed are of the order of 0.5 nm. © 2001 Optical Society of America

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Cholesteric liquid crystals (CLC's), with their natural periodicity and large birefringence, can be viewed as one-dimensional photonic bandgap materials. Mirrorless lasing in dye-doped CLCs was proposed^{1,2} and, recently, unambiguously demonstrated on nanosecond³ and picosecond^{3,4} time scales. The primary role of the CLC in these systems is as a distributed cavity host for active materials such as DCM laser dyes. Lasing in dye-doped CLC systems occurs at the band edges^{3,4}; this is consistent with distributed-feedback laser systems.⁵ However, fluorescent emission in the near-UV region can be observed from certain liquid crystals. This suggests the possibility that the CLC may act both as the active material and as a distributed cavity host. When it is excited, such a system can lase without mirrors or any other optical elements and without the use of dyes. We term such behavior self-lasing. Preliminary evidence of self-lasing at the edge of the reflection band in an undoped CLC was reported recently.^{6,7}

In this Letter we report observations of stimulated emission and self-lasing in a pure homogeneous CLC under picosecond excitation at 355 nm. In these samples, the reflection band was matched to the emission and excitation spectra of the CLC hosts. We observed self-lasing in the region 385–405 nm.

Mixtures of the commercial CLC mixture BLO61 (EM Industries) with right-handed helicity and the right-handed chiral dopant 4-(2-methylbutyl)-4-cyanobiphenyl (CB15) were studied. These constituents were selected because their optical properties, including absorption, reflection, emission, and excitation spectra, were characterized previously.^{7,8} Samples were prepared by addition of <10-wt. % CB15 to the BLO61 mixture. By variation of the CB15 concentration, the reflection band of the samples could be tuned from the visible to the near UV. Cells consisting of parallel glass plates coated with buffed ~50-nm-thick polyimide alignment layers and separated by 25- μ m spacers were filled with the CLC mixture.

Continuous fluorescence and excitation spectra were obtained with a Fluorolog 3 (Jovin Yvon–Spex) spectrofluorometer equipped with a 400-W xenon lamp and fitted with a Hamamatsu R-928 photomultiplier.

This spectrofluorometer provides corrected emission and excitation spectra in the range 200–800 nm. For our experiments, excitation consisted of ~35-ps pulses at 355 nm from a tripled Q-switched and mode-locked Nd:YAG laser (Continuum PY61C). The beam intensity was controlled by use of a polarizing prism and a $\lambda/2$ plate. The polarization state of the pump beam was controlled by a $\lambda/4$ plate. By use of a focusing lens, the pump-beam diameter at the sample could be varied from ~300 μ m ($f = 20$ cm) to 5 mm ($f = \infty$). Typically, the pump beam was incident at ~2° from normal, to prevent the transmitted pump beam from entering the collection optics. The light emitted from the exit side of the sample (where the pump beam exits) was collected and focused by a lens ($f = 5.5$ cm) onto the entrance slit of a TRIAX 550 (Jovin Yvon–Spex) spectrometer. The emission was recorded with an i-Spectrum ONE intensified CCD (Jovin Yvon–Spex) detector operated in the continuous mode, which captures a 72-nm band. With an integration time of 0.01 s, the defined spectral region was scanned five times during a single experiment, and the signal was subsequently averaged. The spectral resolution of the system was 0.03 nm.

Figure 1 shows the unpolarized and left circularly polarized emission from a CLC sample whose periodicity is such that the low-energy edge of the reflection band is near the peak of the emission spectrum. The emission spectra were normalized to demonstrate alteration in the line shape. The effect of the reflection band is to suppress the emission of the nonpropagating right circularly polarized mode in the bandgap and to enhance right circularly polarized fluorescent emission at the band edges.^{3–7} The fluorescence spectrum of the BLO61 mixture without CB15 is also shown for comparison. Since the host liquid crystal BLO61 is a commercial mixture, the detailed origin of its characteristic UV fluorescent emission is not well understood. Mechanisms of UV emissions observed in similar nematic liquid crystals are discussed in the literature.⁹

Figure 2 shows the reflection band for normally incident unpolarized light, the fluorescent emission from excitation at 355 nm, and the laser line when the pump intensity is just above threshold at 692 nJ. The

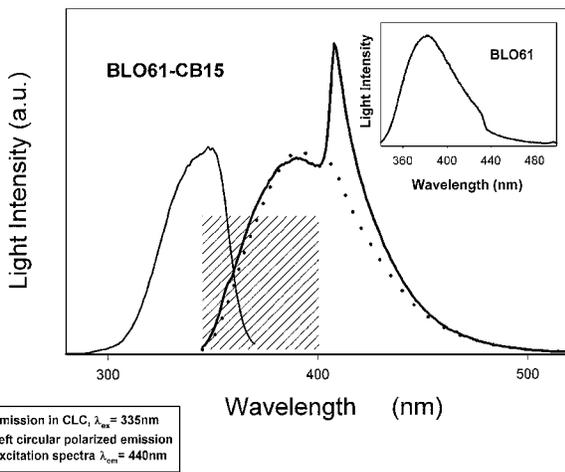


Fig. 1. Excitation spectrum and emission spectra for unpolarized and left circularly polarized modes. The hatched region indicates the reflection band. The inset shows the fluorescence spectrum of the BLO61 mixture.

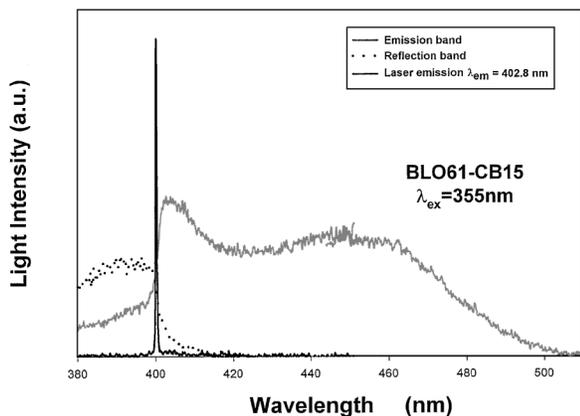


Fig. 2. Reflection band (unpolarized), fluorescent emission spectrum (5-mm pump-beam diameter), and laser line (300- μ m pump-beam diameter) for the BLO61-CB15 mixture. A different intensity scale was used for each of the three curves; they are shown in the same graph for the purpose of comparison.

laser linewidth is ~ 0.3 nm. The shoulder in the fluorescence at 460 nm is believed to be associated with formation of excimers (exciplex), which have been observed in liquid crystals with a similar structure.¹⁰⁻¹² Experiments are currently being conducted to determine the origin and role of excimers in the lasing process.

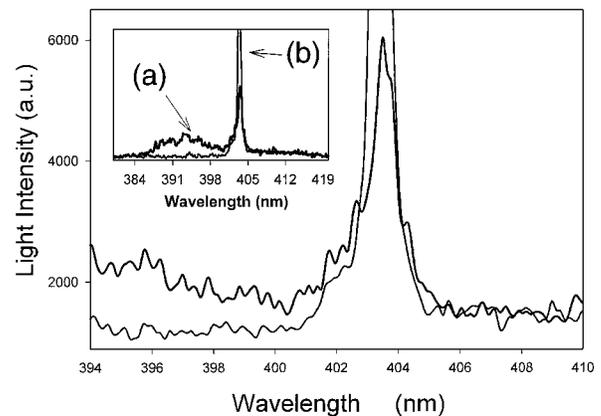
Focusing the pump beam with an $f = 20$ cm lens resulted in a spot diameter of ~ 300 μ m on the sample. Above the threshold, a slight change in the input power resulted in a nonlinear increase in the laser output power. Figure 3 shows an example of the fluorescent emission below threshold (input energy $E_a = 325$ nJ) and the laser emission above the laser threshold (input energy $E_b = 1.262$ μ J). The sudden spectral narrowing in the emission indicates the onset of stimulated emission.

Above the laser threshold, strong directional emission was observed at the edge of the reflection band,

as shown in Fig. 2, similar to that reported earlier.³⁻⁷ The narrowing of the linewidth, nonlinear gain in fluorescence, and directional emission at the band edge indicate coherent emission and lasing.

Since the lasing occurs at the edge of the reflection band, shifting the edge of the reflection band changes the wavelength of laser emission. We observed lasing at a number of wavelengths in the range ~ 385 – ~ 410 nm in CLC mixtures with various CB15 concentrations. The FWHM of the laser lines range from ~ 1.1 nm at 387 nm to ~ 0.3 nm at 402 nm. Examples of laser emission observed in our pure CLC samples are shown in Fig. 4.

The experimental results demonstrate lasing in pure (dye-free) CLC thin films under picosecond pulse excitation. The possibility of UV-blue laser emission in thin CLC films is significant for both scientific and technological reasons and may be useful in realizing compact laser light sources at short wavelengths. Experiments to gain better understanding of light amplification in CLC and related systems are under way.



$\lambda_{\text{pump}} = 355\text{ nm}, 35\text{ ps}$
input power ($E_b > E_a$)

Fig. 3. Line narrowing and increase in output power with increasing pump intensity. The curves correspond to input energy $E_a = 325$ nJ and $E_b = 1.262$ μ J.

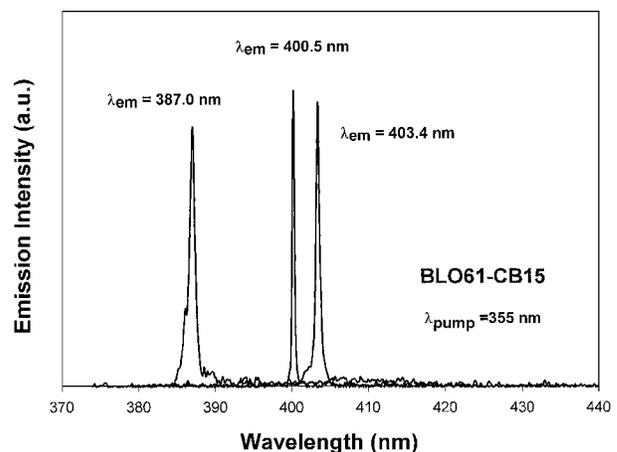


Fig. 4. Laser lines observed in CLC mixtures with various CB15 concentrations.

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