Low-threshold lasing at the edge of a photonic stop band in cholesteric liquid crystals

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Low-threshold lasing is observed at the edge of the stop band of a one-dimensional structure—a dye-doped cholesteric liquid-crystal film. The mode closest to the edge has the lowest lasing threshold. The rates of spontaneous and stimulated emission are suppressed within the stop band and enhanced at the band edge. The ratio of right to left circularly polarized spontaneous emission is in good agreement with calculated density of photon states. © 1998 Optical Society of America

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In analogy with electronic bandgaps in semiconductors, three-dimensional (3-D) photonic bandgaps have been observed in dielectric materials with sufficiently high modulation of the refractive index.[1–6] Yablonovitch[1] predicted that spontaneous emission would be inhibited within such a photonic bandgap. Since the depletion of the excited state by spontaneous emission is minimized, low-threshold lasing would occur at defect modes introduced into the gap. An enhancement of nonlinear phenomena including superradiant emission was also predicted at the edge of a 3-D photonic bandgap.[7] There has therefore been a considerable effort to produce structures possessing a 3-D bandgap.[3,4] In contrast with the difficulty of producing samples with a 3-D bandgap, one-dimensional (1-D) and two-dimensional (2-D) bandgap materials are readily fabricated and have also been studied extensively. For 1-D periodic structures with sufficiently large refractive-index modulation, Dowling *et al.* predicted that lasing should occur at the edge of a photonic stop band.[8] Tocci *et al.* found that the presence of a stop gap in a 1-D structure results in an alteration of the spontaneous-emission spectrum of a GaAs light-emitting diode sandwiched between stacks of distributed Bragg reflectors.[9] Although a wide variety of periodic structures possessing photonic bandgaps has been produced in one, two, and three dimensions at frequencies ranging from the microwave to the optical, lasing in photonic bandgaps has not been demonstrated to our knowledge.

In this Letter we report lasing at the edge of a stop band in a 1-D periodic structure. In measurements of dye-doped cholesteric liquid-crystal (CLC) films we find that the rate of spontaneous emission is inhibited within the band and enhanced at its edge. Because light of opposite chirality is unaffected by the periodic structure in this system, we are able to make a direct measurement of the density of photon states by comparing the emission spectra of oppositely polarized radiation. The observed suppression of the density of states within the band and the sharp rise at the band edge are in good agreement with the calculated density of states in a 1-D structure.

These results demonstrate that lasing in dye-doped CLC's is a consequence of the photonic band structure. Lasing was observed in these structures more than 20 years ago[10] and was explained by use of distributed feedback theory.[11] Based on the assumption that the dielectric function is weakly modulated in this structure, it was presumed that lasing occurs at the center of the reflection band near the Bragg frequency. Subsequent observations of lasing in CLC's far from the center of the reflection band were ascribed to distortions of the helical structure.[12] However, the amplitude of the index modulation in CLC's is large enough to produce a reflection band for radiation with the same chirality as the structure,[13] and the optical properties of this system should therefore be described in terms of the photonic bandgap.

The rod-shaped molecules in the 1-D host used in the present study form a periodic helical structure with pitch P that can be either right or left handed. For sufficiently thick films the reflectance of normally incident, circularly polarized light, with the same sign of rotation as the CLC structure, is nearly 100% within a band centered at $\lambda_c = nP$. The reflected light has the same sign of rotation as the incident beam. The bandwidth is $\Delta\lambda \approx \lambda_c \Delta n/n$, where $n = (n_o + n_e)/2$ is the average of the ordinary and extraordinary refractive indices of the medium and $\Delta n = n_e - n_o$.[13,14]

Measurements were carried out on two CLC samples with different host compositions doped with laser dye PM-597 (1,3,5,7,8-pentamethyl-2, 6, -di-*t*-butylpyrromethene-difluoreborate complex), which gave rise to an absorption peak at 530 nm and an emission peak near 570 nm. Samples 1 and 2 had right- and left-handed helical structures, respectively. The transmittance and reflectance spectra for normally incident unpolarized light for sample 1 are shown in Fig. 1. In sample 2 the band center is shifted to

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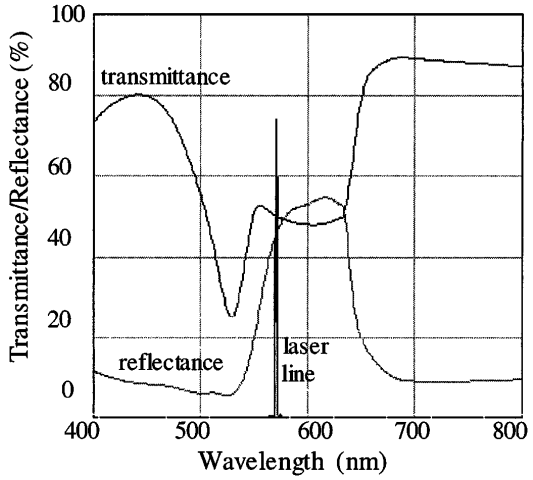

Fig. 1. Transmittance and reflectance spectra for unpolarized light of dye-doped CLC for sample 1 with superimposed laser emission.

the red relative to sample 1 by approximately 40 nm (Fig. 3, below). CLC films 1 and 2 have thickness L of approximately 20 and 30 μm, respectively. In these samples $n \sim 1.6$ and $\Delta n \sim 0.1$.

Emission in these samples was studied by use of the second harmonic of a Q-switched Nd:YAG laser with and without mode locking. Individual mode-locked pulses were approximately 70 ps long. Single Q-switched pulses were 150 ns long with maximum pulse energy of ~1 mJ. The energy of the pump laser pulse was controlled by use of an electro-optic attenuator. The pump beam was approximately 5 mm in diameter at the focusing lens, which resulted in spot diameters of approximately 40 and 20 μm for the 30- and the 14-cm focal-length lenses, respectively. A lens with a focal length of 5.5 cm was used to collect the emitted light and to focus it onto the entrance slit of the spectrometer; this corresponds to a collection angle of ~30° in air and ~18° within the CLC film. The emission was dispersed in a spectrometer and recorded with a CCD detector that captured a 74-nm band with a resolution of 0.075 nm.

A comparison of lasing, reflectance, and transmittance spectra in Fig. 1 shows that lasing occurs at the edge of the photonic stop band. We find that shifting the edge of the stop band shifts the lasing frequency from approximately 560 to 610 nm in different hosts doped with the same dye. The divergence of the laser beam emitted from sample 1 is found to be ~0.06 and ~0.11 rad for pumped spot diameters of 40 and 20 μm, respectively; this corresponds to a diffraction-limited divergence for a coherent beam generated by spots with diameters of ~12 and ~7 μm. This result is probably consistent with a single coherent laser source, which is smaller than the pump diameter as a result of the nonlinear nature of lasing action. The laser radiation is right circularly polarized (RCP) from sample 1 and left circularly polarized (LCP) from sample 2, in conformity with the chirality of the CLC structure. Approximately 4% of the laser intensity is of opposite sign polarization, as expected as a result of the change in chirality of light on Fresnel reflection from the rear glass plate.

RCP laser emission spectra from sample 1 at different pump powers are shown in Fig. 2(a) for Q-switched pump pulses. At low pump power a single laser line with a width of approximately 0.2 nm is observed at the stop band edge at 571.5 nm. Even at high pump powers only a small number of closely spaced modes within a total width of ~1 nm are involved in lasing. The center of the laser emission at higher powers shifts from the band edge to wavelengths at which the output-coupling coefficient is improved. The energy-conversion efficiency from the pump to the laser beam is as high as 25% at a pump pulse energy of ~0.1 mJ. The spacing between modes shown in Fig. 2(a) is considerably less than the mode spacing of $\delta\lambda \approx \lambda_c^2/2Ln = 5$ nm for a 20-μm-thick film, which is consistent with the increased density of states expected at the edge of the stop band.[7,13]

The dependences of the output energy on pump power for modes near the band edge for sample 1 are shown in Fig. 2(b). For comparison, the linear dependence of the spontaneous emission integrated over the spectrum from 547 to 622 nm is also shown. Mode 1 at 571.5 nm, which is closest to the band edge, has the lowest lasing threshold. Lasing is observed at the lowest pump energies at which reliable spectral measurements are possible, 0.3 μJ. The thresholds for modes 2, 3, and 4, which peaked near 571.1, 570.5,

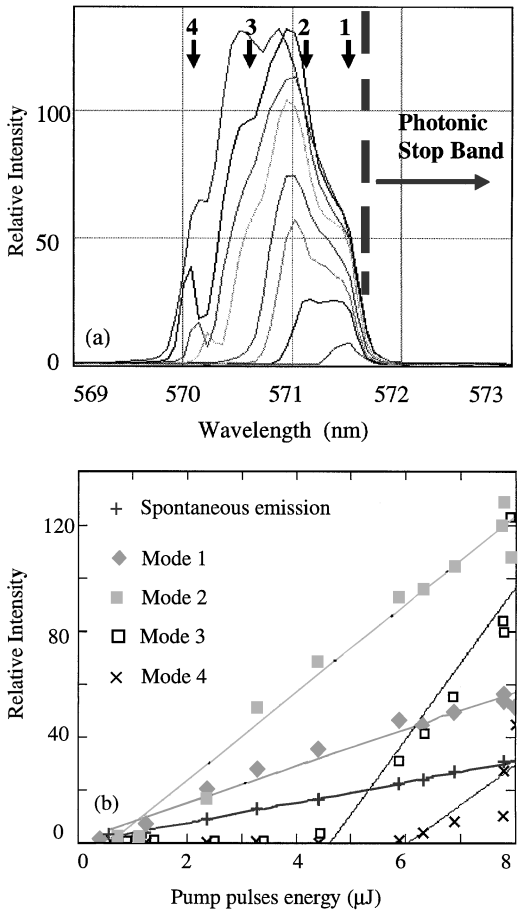

Fig. 2. (a) Emission spectrum with peaks at wavelengths 1–4 from sample 1 at different pump powers. (b) Relative intensities of emission at the wavelength corresponding to each mode.

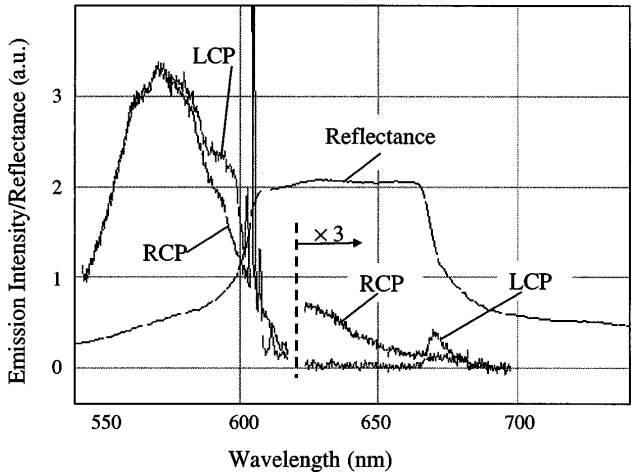

Fig. 3. Emission spectra from sample 2 for LCP and RCP emission with the reflectance spectrum superimposed.

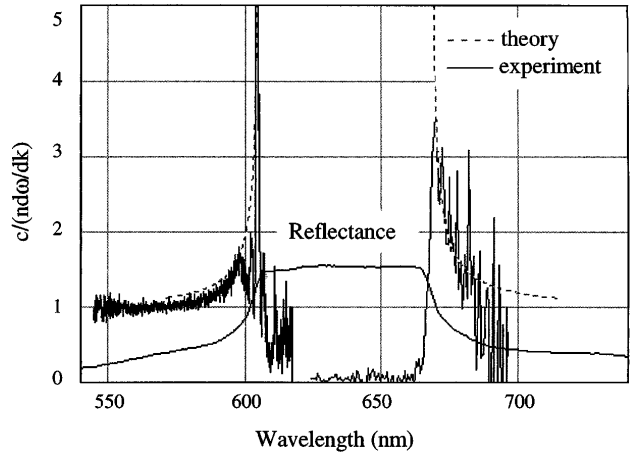

Fig. 4. Ratio of RCP to LCP emission spectra from sample 2 and theoretical curve for $(c/n)/(\mathrm{d}\omega/\mathrm{d}k)$ for the stop band.

and 570.2 nm, respectively, can be seen to increase with increasing frequency shift from the band edge. The rate of increase of output power can be seen in Fig. 2(b) to increase with mode number to mode 3. Presumably, the frequency-dependent output-coupling coefficient, which increases away from the band edge, is then optimal for the excitation energies used.

In sample 2, which has a stop band that is shifted away from the emission peak, lasing is observed only when the pump laser is both mode locked and Q switched. Polarized emission spectra from this sample are shown in Fig. 3. For reference purposes the unpolarized reflectance spectrum is also presented. LCP lasing again occurs at the blue edge of the reflection band, which is the closest edge to the emission peak. The peak intensity of the laser lines is 100 times greater than the maximum of the spontaneous emission. The RCP emission spectrum has a single peak and is similar to the emission spectrum that is expected for molecules within an unoriented host. However, LCP emission is suppressed in the stop band and enhanced above the level of RCP emission at both edges of the band.

Since both LCP and RCP spontaneous emission are emitted by the same dye in the same host and the periodic structure does not influence the dipole matrix element, their ratio gives the ratio of the density of photon states, which is shown in Fig. 4. In 1-D structures the density of states is proportional to $1/(\mathrm{d}\omega/\mathrm{d}k)$, where ω and k are the frequency and the wave vector of light, respectively. In contrast with that of 2-D and 3-D structures, the density of states diverges at the band edge of 1-D structures. Figure 4 shows good agreement between the ratio of LCP and RCP spontaneous emission and $(c/n)/(\mathrm{d}\omega/\mathrm{d}k)$, where c is the velocity of light in vacuum:

$$n\omega(k)/c = \mathrm{sign}(k - k_0)(k^2 - 2kk_0 + \omega_0{}^2)^{1/2} + \omega_0\,,$$

$\omega_0 = 2\pi nc/\lambda_c$, $\Delta\omega = \omega_0\Delta n/n$, and $k_0c/n = [\omega_0{}^2 - (\Delta\omega/2)^2]^{1/2}$.

We believe that low-threshold lasing at the band edge in a 1-D periodic structure is related to the singularity at the band edge in the density of photon modes that propagate perpendicular to the layers. The high density of states is associated with a small group velocity that tends to zero at the band edge. The lifetime of modes at the edge is thereby lengthened, and they experience high gain before leaving the sample. In conclusion, these results demonstrate that the bandgap properties of CLC's can be exploited to design effcient, low-threshold, mirrorless microlasers.

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