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Laser Emission from Dye-Doped Liquid Crystal Gratings Formed by Polarization Holography

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Laser emission from a nematic liquid crystal system whose grating structure is induced by a linear photopolymerizable alignment layer is reported. The grating structure is written on the alignment layer through polarization holography, inducing periodic boundary conditions on the laser dye-doped nematic liquid crystal. We report on the presence of laser emission at wavelengths predicted by coupled wave theory and the switchability of emission in these systems through the application of an electric field. Sensitivities to incident polarizations are also discussed.

Keywords: dye; holography; H-PDLC; laser; liquid crystal; photonic crystal

Active research into dye-doped liquid crystal (LC) structures as lasing sources has increased in recent years due to the attractive ability to tailor and control the morphology of these materials. The structure of a liquid crystal material is easily manipulated through surface treatment techniques [1], holographic methods [2] and confining geometries; many of these techniques result in one-, two- or three-dimensional photonic crystals [3]. Lasing from a dye-doped liquid crystal has been achieved in cholesteric liquid crystal mixtures [4–6]. Low-threshold cholesteric liquid crystal lasers have been demonstrated in the visible [7] and ultraviolet regimes [8]; additionally, these

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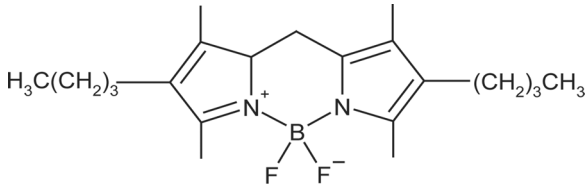
laser sources have been shown to be tunable over a range of wavelengths through electrical [9], thermal [10], and phototunable [11] methods. Lasing has also been shown in cholesteric elastomers with electromechanical tunability [12]. Liquid crystal and polymer composite structures have been demonstrated as distributed feedback cavities for lasing in the form of polymer dispersed [13,14] and holographic-polymer dispersed liquid crystals (H-PDLCs) [15–18]. Liquid crystal systems provide advantages over other photonic crystals in that they employ simple, rapid fabrication processes using well-established surface alignment and holographic techniques.

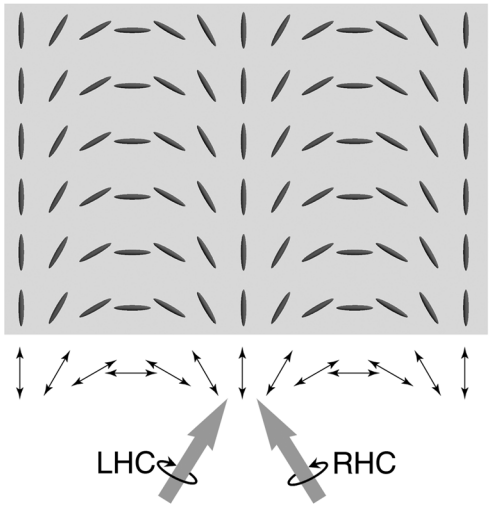
Lasing in all of these structures results from the presence of a photonic band gap (PBG) within the material. In the case of cholesteric or ferroelectric liquid crystals, the intrinsic band gap, caused by the helical pitch of the material, creates a stop band for circularly polarized light with identical handedness to the helical pitch of the liquid crystal molecules. The PBG in an H-PDLC arises from the index mismatch between the Bragg layers. This mismatch creates an effective potential for incident photons, akin to the periodic potential an electron experiences in a crystalline structure. Plane wave approximations, Bloch formalism and coupled wave theory have all been applied to photonic crystals in order to understand their band structures and other properties. Kogelnick and Shank [19,20] theoretically described lasing in one-dimensional periodic structures using a coupled wave theory. Kopp and colleagues present a sound explanation of lasing in chiral media [21] based on the works of Dowling and Bowden [22,23]. In all of the above-mentioned structures, the laser action occurs at the edge of the PBG due to the long interaction times between the lasing medium and pump source in this regime.

The morphology of liquid crystal systems, as mentioned earlier, is easily controlled using surface and holographic techniques. We report on gain-narrowed emission from a nematic liquid crystal with a periodic structure induced solely by an alignment layer with periodic boundary conditions; a technique making use of both surface alignment methods and holography. In previously reported results, a linear photopolymerizable polymer (LPP) was used to create a polarization grating on a surface alignment layer for a nematic liquid crystal, which propagated into the bulk nematic [24,25]. These reports present grating structures with pitch lengths on the micrometer scale; the emissive systems presented here make use of a much smaller pitch, comparable to the wavelength of visible light.

The nematic liquid crystal BL038 ($n_e = 1.7999$, $n_o = 1.527$, $\Delta\varepsilon = +16.4$, *EM Industries*) is used as the host solvent for an organic laser dye molecule Pyrromethene 580 (PM580, *Exciton, Inc.*), whose



(a)



(b)

FIGURE 1 The molecular structure of the laser dye Pyrromethene 580 (a); illustration (top view) of the alignment induced on the liquid crystal molecules by the constant intensity, polarization grating created by the interference of two circularly polarized beams with opposite handedness (b).

molecular configuration is shown in Figure 1(a). The liquid crystal was chosen for its large birefringence and dielectric anisotropy. This laser dye is characterized by broadband absorption around 520 nm and broadband fluorescence between 550 and 600 nm [15]. A low concentration, <0.5 wt.%, of the laser dye was added to the liquid crystal material and mixed for approximately one hour using a stirbar to ensure a homogeneous mixture.

The LPP material (*ROLIC*) was spun coat onto two glass substrates coated with an anti-reflection coating on one side and a thin layer of indium tin oxide (ITO) followed by an index matching layer on the other. Fiber spacers of 5 μm thickness were placed on one substrate

and the substrates were fixed together with epoxy. When exposed to ultraviolet radiation, the LPP molecules polymerize with their molecular axes parallel to the direction of incident polarization. By using a two-beam holographic exposure to create a constant-intensity, rotating polarization grating, a periodically rotating alignment layer is locked into the surface. The two-incident beams are circularly polarized with opposite handedness and result in the periodic structure depicted in Figure 1(b). The dye-doped liquid crystal mixture, when capillary filled into the exposed cell, will align parallel to the local molecular direction of the polymerized LPP material and result in a grating structure. The resulting grating possesses a sinusoidal modulation of both its index of refraction and dielectric anisotropy along one axis of the sample, which gives rise to the photonic band gap and enables enhanced emission in the material.

The periodicity, or pitch, of the polarization grating was selected such that an edge of the band gap would exist within the fluorescence band of the laser dye, resulting in ideal conditions for distributed feedback emission. According to Kogelnick's coupled wave theory [15,20], the enhanced emission from this Bragg grating structure follows the relationship $\lambda_{\text{laser}} = 2n_{\text{eff}}\,\Lambda/m$ where n_{eff} is an effective index of refraction of the gain material, Λ is the pitch of the periodic medium and m is a diffraction order of the grating. Hsiao and coworkers have reported on lasing H-PDLC gratings with experimental emission wavelengths within 2% of those predicted by this model [15].

Targeting a wavelength for emission of 570 nm, and using $n_{\text{eff}} = 1.623$, an average of n_e and n_o for the liquid crystal ($n_{\text{eff}} = \sqrt{(n_e^2 + 2n_o^2)/3}$), and $m = 2$, second order diffraction, the above-mentioned coupled wave theory suggests an optimal pitch of $\sim$351 nm. The angle of incidence within the material for the two writing beams can be calculated from the theoretical pitch by $\theta_i = \sin^{-1}(\lambda_w/2\Lambda)$ where λ_w is the wavelength of the laser used to fabricate the grating, in our case an Ar^+ laser (*Coherent* Innova 70) operating at $\lambda_w = 351$ nm with average beam intensities of 50 mW/cm^2 was used for sample fabrication; the resulting necessary angle of incidence for these samples was 30°.

Laser emission in the dye-doped LC samples was generated with the use of a Brilliant model frequency doubled Nd:YAG pulsed laser (*Quantel*) operating at a wavelength of $\lambda = 532$ nm, a repetition rate of 10 Hz and a maximum pulse energy of 200 mJ. The beam passed through an attenuator, which reduced the average pulse energy to between 5 μJ and 5 mJ. The sample was oriented with its surface normal at 45° to the pump beam. A long pass laser line filter with 100% transmission above 540 nm was placed between the emissive cell and the spectrometer to completely block the pump beam from the

recorded spectra. When strong emission was measured, a gradient neutral density filter was used to diminish the intensity of the incident signal. The resulting spectra were measured with a fiber spectrometer (*Ocean Optics*) and recorded to a computer. A schematic of the optical setup for measuring emission from the edge of the sample is shown in Figure 2.

The fluorescence spectra, obtained by pumping with a low power constant wave Verdi Nd:YAG laser (*Coherent*) operating at $\lambda = 532\,\text{nm}$ and an energy of 25 mW, underwent significant line narrowing with the application of the pulsed pump beam, as seen in Figure 3. The fluorescence spectra possessed a full width at half maximum (FWHM) of approximately 50 nm, while emission generated by the pulsed pump beam possessed a FWHM of $\sim 5\,\text{nm}$. This line narrowing is indicative of laser emission in a gain environment. As the pump energy was increased from 50 μJ to 225 μJ, the intensity of emission rose slowly; above a threshold of approximately 225 μJ, the rate of change in the intensity of emission as a function of pump beam energy rose much more sharply. Additionally, the FWHM decreased significantly as the pump energy increased from 35 μJ and approached the threshold point near 225 μJ. This threshold behavior and line narrowing in a gain environment is indicative of lasing and is shown in Figure 4.

The emission was also measured as a function of incident linear polarization relative to the grating vector of the periodic structure. S-polarization is defined as a linear polarization parallel to the grating vector, Figure 5(a); p-polarization is defined as a linear polarization perpendicular to the grating vector, Figure 5(b). As the polarization

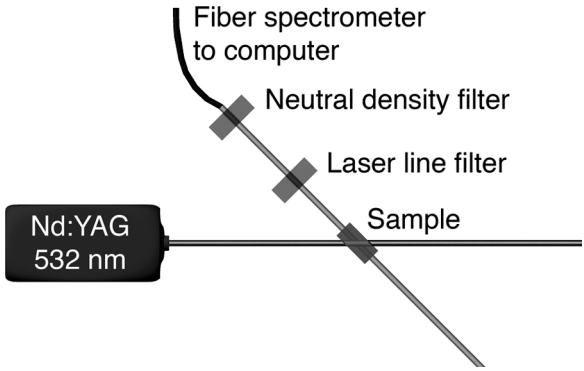


FIGURE 2 Schematic of the liquid crystal laser pumping setup for emission from the edge of the sample.

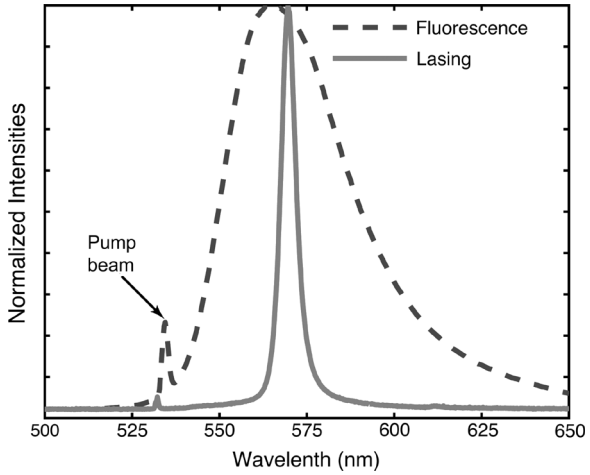


FIGURE 3 Enhanced emission from a dye-doped liquid crystal polarization grating (solid), compared to the fluorescence emission (dashed) of the laser dye in the liquid crystal material.

is rotated from linear s- to linear p-, the intensity of emission for a fixed pump energy increases by $\sim$50%, as shown in Figure 5(c). This dramatic increase suggests a high degree of sensitivity for the

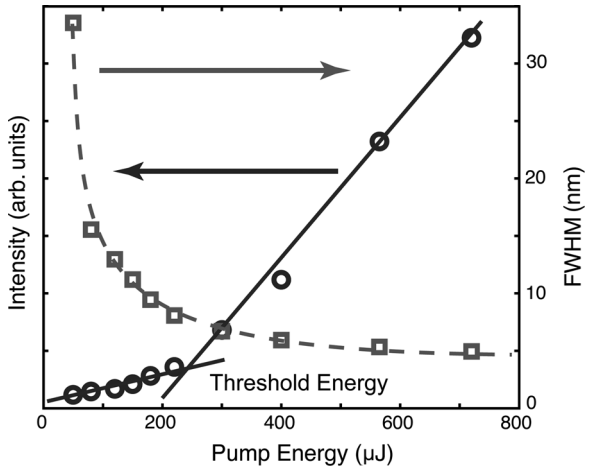


FIGURE 4 The threshold for laser emission occurs at a pump energy of approximately 225 μJ; as the pump energy is increased to the threshold point, the FWHM of emission sharply decreases to under 5 nm.

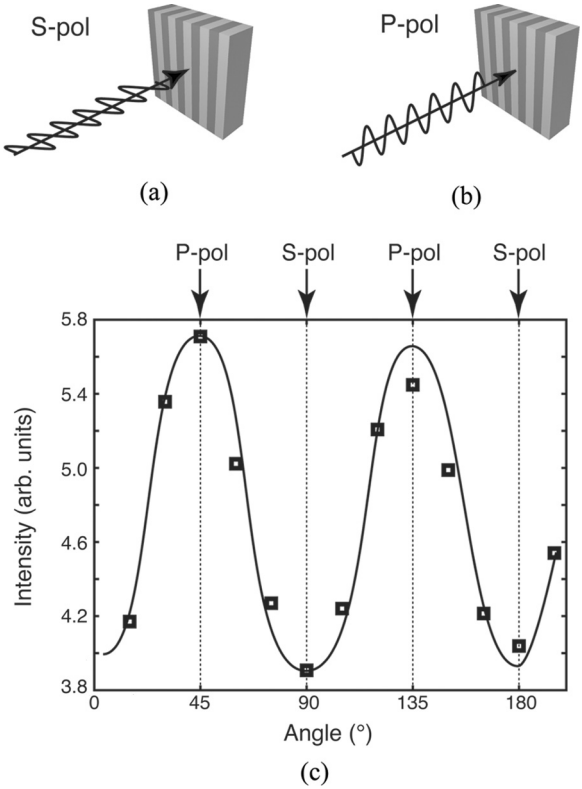


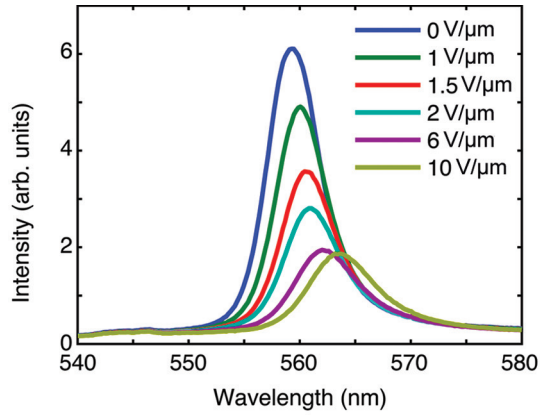
FIGURE 5 Depictions of s- (a) and p-polarizations (b) relative to the grating structure. The emission increases by ~50% when the incident polarization is rotated from incident s- to incident p- (c).

emission relative to the incident polarization. This result, along with the threshold behavior mentioned earlier, gives strong credence to the presence of laser emission.

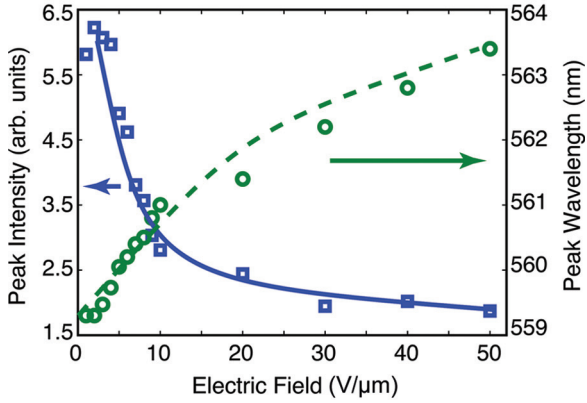
The emission from the edge of the cell was focused to a single spot with the use of a convex lens – with a focal length of 100 mm. The focused emission was measured with both the spectrometer and a power meter. The spectrometer recorded a spectrum comparable to that shown in Figure 2, with only slightly decreased emission. The power meter measured an output of approximately 1 μJ with a pump intensity of 500 μJ – yielding an efficiency for emission of ~0.2%.

The response of the laser emission was also measured in the presence of an electric field. Electrodes were attached to the ITO layers of the glass substrates and a function generator (*HP* 33120A) with a

high voltage power supply (*Trek* Model 50/750) to apply a 1.0 kHz square wave across the sample at field strengths between 0 V/μm and 10 V/μm. The resulting emission spectra are shown in Figure 6 (incident p-polarization) and Figure 7 (incident s-polarization). In the zero field case, the peak emission intensity for an s-polarized pump beam is half that of the peak emission intensity for a p-polarized pump beam. For both polarizations, the emission red shifts as the applied field strength increases. The total shift in emission for a p-polarized pump beam is 4 nm; whereas the shift for s-polarized light is 7 nm.



(a)



(b)

FIGURE 6 Emission as a function of applied electric field for p-polarization (a); the emission drops by ~60% and slightly red shifts (b).

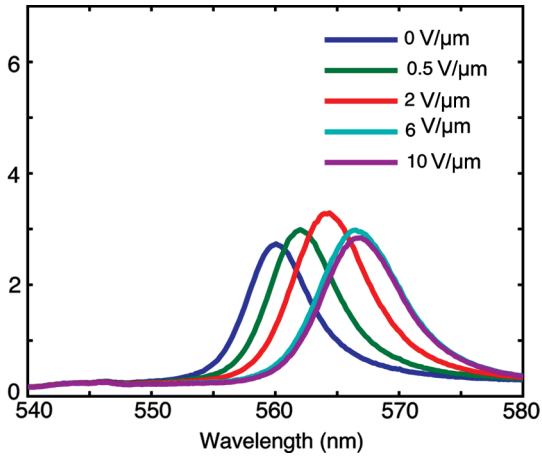


FIGURE 7 Emission as a function of applied electric field for s-polarization; the emission intensity remains constant while the peak intensity red shifts.

For both pump beam polarizations the linewidth of emission increases by $\sim$2 nm. The most noticeable difference between s- and p-polarized pump beams is the "switching off" nature of the emission pumped by a p-polarized beam; a feature not present for an s-polarized pump beam. The peak emission intensity falls off by approximately 60% with an application of a strong electric field. The emission intensity of the p-polarized beam in the presence of an electric field is comparable to that of the s-polarized beam, suggesting the possibility of amplified spontaneous emission being the dominant process, over laser emission for the s-polarization pumped cell and the p-polarization in the presence of an applied field. Further contributions will develop a phenomenological model of the laser emission in these grating structures in the presence of an applied electric field.

In conclusion, we have presented a report of laser emission from a dye doped-nematic liquid crystal whose grating structure is induced solely by an alignment layer. The periodic grating structure on the alignment layer was fabricated through polarization holography and a linear photopolymerizable polymer. The emission exhibited a narrow width ($\sim$5 nm), a threshold pump energy of approximately $\sim$225 μJ and was switchable and tunable through the application of an electric field across the sample. The efficiency of the laser system was measured to be approximately 0.2%. Further studies will investigate the effect of the birefringence, Δn, of the liquid crystal, as well as methods

to further reduce the linewidth, and, in turn, increase the quality of the emission.

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