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Broadband Luminescent Dye-Doped Temperature-Tunable Cholesteric Liquid Crystal Laser

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Abstract—We have synthesized a luminescent dye that exhibits good solubility in cholesteric liquid crystals and broadband spectral emission. By incorporating this dye into cholesteric liquid crystal and using a nitrogen laser beam as the pumping source, a stable laser emission from the dye-doped cholesteric liquid crystal has been generated. We conducted a temperature-controlled tuning of the laser lines, achieving an extra-wide spectral range of over 172nm, which is the broadest spectral interval so far, using a single luminescent dye.

Fluorescent and luminescent materials are revolutionizing applications in a range of fields, including chemical sensing, biological imaging, lighting, displays, photodynamic therapy, and information storage. Today's technology and photonics are based on newer organic dyes with high luminescent properties [1−2]. One of the most popular and promising applications of luminescent dyes (LDs) is the cholesteric liquid crystals (CLCs), formed by chiral molecules, which represent a self-assembled, one-dimensionally periodic helical structure [3]. The selective reflection band (SRB) of a CLC is observed in the spectral range bandwidth $\Delta\lambda=P_0\Delta n$, where $\Delta n=n_e-n_o$ determined by the pitch and the ordinary n_o and extraordinary n_e refractive indices. The SRBs are centered at $\lambda_p=\check{n}P_0$, where $\check{n}=(\check{n}_e+\check{n}_o)/2$ is the average refractive index, which in CLC can be altered by numerous external and internal stimuli, including light, temperature, mechanical deformation, electric field, chemical substances, pH changes, humidity, and biomolecules [4−5]. LD-doped CLCs (DDCLCs) are self-assembled, mirrorless structures that can function as distributed-feedback lasers with low threshold requirements [6−7]. In DDCLC lasers, it is essential to tune the laser lines across a broad spectral range, which, along with other factors, crucially depends on the width of the LD's emission spectrum [8]. Several approaches were used to broaden the emission spectrum of LDs. For example, in [9], a CLC mixture comprising two highly efficient LDs was investigated as an active medium to broaden the tunability of laser emission. However, in DDCLC lasers, it is desirable to use a single LD with broad spectral emission, enabling uniform and continuous tuning of the laser lines across a wide spectral range. Therefore, to achieve this goal, synthesizing new broadband LDs is one of the most desired tasks for DDCLC lasers. In this study, we synthesized an LD and dissolved it in a CLCs mixture. Upon excitation with a nitrogen laser, we achieved stable, reversible, and extremely broadband $\Delta\lambda$=172nm tuning of the laser emission lines, controlled by temperature. The synthesized LD is chemically stable and does not degrade upon photo- and thermo- exposure.

To prepare DDCLC, two initial materials were used: the nematic matrix (ZLI-1184 + ZLI-1185) and the optically active dopant ZLI-811. Polyvinyl alcohol (PVA) was used to achieve a uniform distribution of CLC mixtures over the glass plates. All materials were purchased from Merck. The 2D structure of LD synthesized at Georgian Technical University is shown in Fig. 1, labeled as ELD.

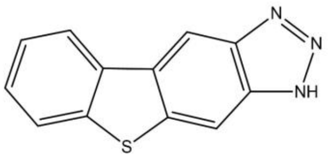

Fig. 1. 3H-benzothieno[2,3-f] benzotriazole (ELD).

The final DDCLC composite contains the following compounds by weight percentages: {99.8 wt% [68 wt% [50 wt% (ZLI-1184 + ZLI-1185)] + 32 wt% ZLI-811]+0.2 wt% ELD}. The prepared DDCLC mixture was stirred evenly for 15 minutes in the isotropic phase at 95°C to ensure thorough blending. To fabricate a DDCLC optical cell, two 2cm × 2cm glass plates were used. The plates were first double-cleaned with deionized water. The PVA solution was applied by dropping it onto the glass substrates, then dried in an oven for 60 minutes at 80°C, and subsequently rubbed in opposite directions. After that, an optical cell was assembled with a 30μm gap using the Teflon films. A DDCLC mixture was capillary-

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infiltrated into the optical cell in the isotropic phase. The imaging technique involved was a fiber-optic spectrometer (Avaspec-2048, Avantes) with 1nm resolution. A hot stage with 0.1°C accuracy was used to control the temperature of the CLC and DDCLC mixtures. For optical excitation of the samples, a Nitrogen laser (MNL-100) with a pulse duration of 3ns, a pulse energy of 170μJ, a repetition rate of 5Hz, and a wavelength of 337.1nm was used. To measure the output laser power, a laser power meter (TS5+TP) was used.

First, we recorded the temperature-dependent spectral tuning of the SRBs of the CLCs. The optical cell was placed on a hot stage to smoothly and continuously control the temperature. A spectrometer was used to record the SRBs of the CLC. In Fig. 2, the spectral tuning of the SRBs exhibits consistent behavior during both heating and cooling, with no changes in thermo-optical parameters after multiple cycles.

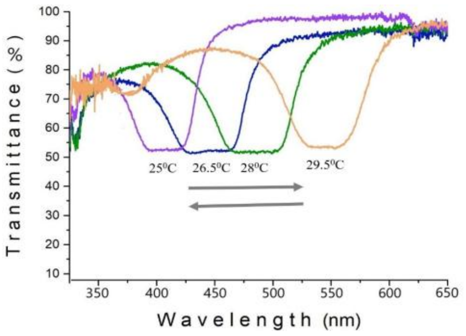

Fig. 2. Temperature-dependent reversible tuning of CLC SRBs.

To record the absorption and emission spectra of the ELD, the DDCLC mixture was heated to the isotropic state. Figure 3 shows the absorption band of ELD in the ultraviolet/violet regions of the spectrum.

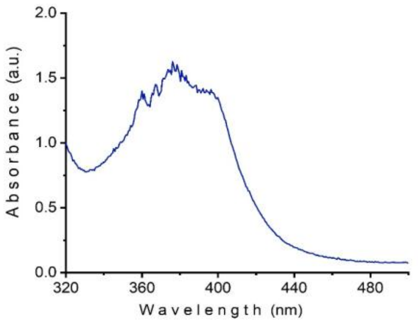

Fig. 3. Absorption spectrum of ELD.

Figure 4(a) shows the luminescent spectrum of the ELD, and Fig. 4(b) displays the DDCLC mixture inside a laboratory vial. Notably, the ELD's luminescence band spans a broad visible spectral range from 400 to 660nm.

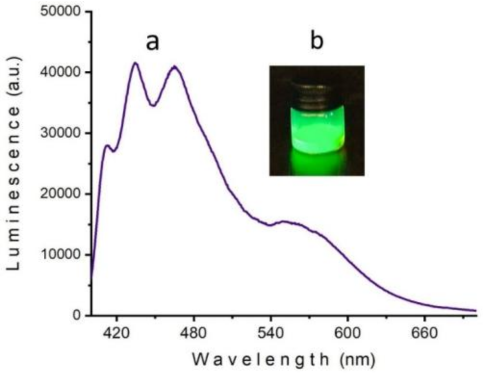

Fig. 4. Luminescence spectrum of ELD.

The next step of the experiments involved a temperature-controlled tuning of the laser lines. To achieve this, we constructed the setup shown in Fig. 5. The pumping beam from a nitrogen laser (1) is directed into an optical cell (3) embedded on a hot stage (2) for temperature control. The pumping beam stimulates laser emission from the DDCLC, which is then directed to a computer-connected spectrometer (4, 5)

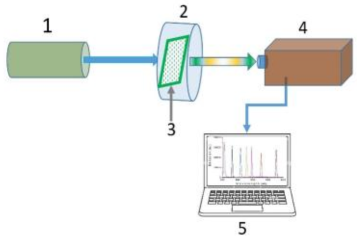

Fig. 5. Experimental set-up. Nitrogen laser (1), Hot stage (2), DDCLC optical cell (3), Spectrometer (4), Computer (5).

Figure 6 illustrates the tuning of laser lines emitted from a temperature-controlled DDCLC, spanning 172 nm. To the best of our knowledge, the DDCLC laser line tuning demonstrated in this work is the widest achieved using a single LD. On the upper part of Fig. 6, the DDCLC laser spots corresponding to the different wavelengths, projected on the screen, are shown.

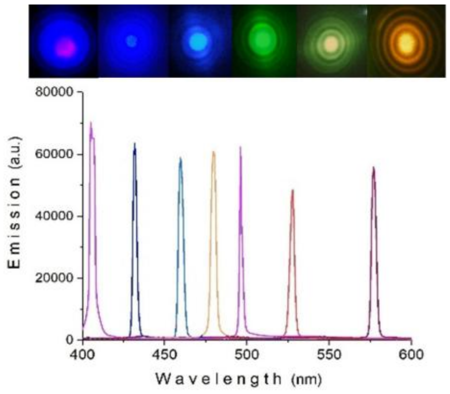

Fig. 6. Temperature-dependent tuning of laser lines of DDCLC.

A key parameter in DDCLC lasers is the relationship between pump energy density and output emission intensity. We conducted experiments on several laser lines across different spectral regions controlled by temperature. We found that the lasing threshold is lowest near the ELD luminescence peaks and increases toward the edges of the luminescence band. In Fig. 7, we demonstrate a laser line with a peak at 496nm. As shown in Fig. 7, the laser emission begins at a pulse energy of 30–40μJ.

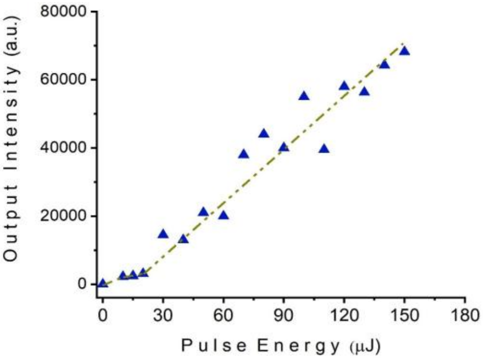

Fig. 7. Emission spectra as a function of pump energy for the laser line at λ=496nm.

It is noteworthy that the laser line tuning, besides the temperature control, is possible in DDCLC mixtures whose SRBs can be tuned by internal or external forces such as electric and magnetic fields, light, pressure, etc.

To conclude, we synthesized an LD that emits light across a broad spectrum. By incorporating this dye into a CLC mixture and exciting the DDCLC with a pumping laser beam, laser generation from DDCLC was observed. Besides, a reversible tuning of the laser emission lines over a wide range of the optical spectrum was obtained, controlled by temperature. The DDCLC-based lasers developed here hold promising applications in medicine, especially cancer detection and imaging, as well as in environmental pollution monitoring and communications.

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