Reduced-Threshold Emission of Capillary Filled by Doped-Dye Cholesteric Liquid Crystals With Photo-Alignment Polyimide Films

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Abstract: A microcavity laser is reported using dye-doped cholesteric liquid crystals (CLCs) filled in a glass capillary with photo-alignment polyimide films. This laser runs based on the properties that a CLC has a refractive index much higher than that of glass, so whispering-gallery-mode (WGM) laser and distributed-feedback-mode lasers are included in the emission spectrum. We obtain a low threshold energy of 4.5 μJ · pulse⁻¹ · mm⁻², establish the shift of 3.04 nm of the WGMs in the temperature range 43–48 °C, and investigate electric field tunability.

Index Terms: Liquid crystal, whispering gallery mode, dye laser, tunable laser.

1. Introduction

Liquid crystals (LCs) have been widely used as tunable dye-doped lasers. One of the most attractive aspects of an LC laser is its tunability by various external conditions, such as temperature [1], light [2], electric field [3], [4], and mechanical strain [5]. There are many categories of reported LC dye lasers, the most important of which is based on the self-organized helical structure of cholesteric LCs (CLCs). Some of these, based on LC cells, include generated-distributed-feedback (DFB) lasers [6]–[8], while others, based on LC droplet generation, include whispering-gallery-mode (WGM) lasers [9]–[12]. Another category includes random lasers based on nematic LCs (NLCs) [13]–[15]. DFB lasing using dye-doped CLCs (DDCLCs) have constituted a main direction of previous research, with most studies based on cells of a parallel glass substrate. Ferjani et al. [16] investigated a core-resonance WGM laser based on dye-doped NLCs filled in a glass capillary. Nagai et al. [17] injected NLCs into a silica capillary and investigated core-resonance cylindrical WGM lasers. Lin et al. [18] investigated manipulation of the random lasing characteristics of dye-doped polymer-dispersed liquid crystals in capillary tubes. Barna et al. [19] observed the lasing
effect in axial and radial direction of cylindrical microcavity which is filled with dye-doped helixed liquid crystals and the lasing emission wavelength can also be tuned by temperature. Microcavity lasers with capillaries filled with DDCLCs can confine the light into a small volume by either the helical periodic structure of the CLCs or by total internal reflection at the interfaces of the CLCs and capillary wall [20]. The lasers based on capillaries filled with DDCLCs have several advantages, including omnidirectional emission, low threshold, and tunability, as well as the ability of WGM and DFB-mode lasers to coexist. To our knowledge, however, no research exists on a capillary emitting laser with a polyimide (PI) film for alignment of LC molecules.

In this paper, we present an experimental investigation of the emission of a capillary filled by DDCLCs with photo-alignment PI films. We propose that optically controlled PI films can effectively control the order and uniform alignment of LC molecules and make CLC molecules more evenly arranged, reduce lasing threshold energy, and optimize sample performance. In addition, we also study temperature and electric field tuning.

2. Sample Preparation and Experiment

We prepared tunable capillary laser samples filled with DDCLCs of different inner diameters (100, 200, and 300 μm). During preparation, the DDCLCs were prepared by mixing a nematic liquid crystal (NLC), a chiral additive (S811), and DCM laser dye [4-dicyanomethylene-2-methyl-6-(4-dimethylstyryl)-4H-pyran] at a weight ratio of 79.2:19.8:1. All experimental materials were supplied by Beijing Bayi Space Liquid Crystal Science and Technology Co. (China). The concentration of chiral additive depended on the position of the dye's fluorescence spectra, so that the fluorescence spectra covered the CLC’s reflection forbidden band in its entirety. For the NLC (BHR33200), the ordinary refractive index and extraordinary refractive index under normal temperature (20 °C) were 1.522 and 1.692, respectively, and the clearing-point temperature was 61.2 °C.

To reduce the threshold voltage and enhance slope efficiency, the orientation of LC molecules should be well controlled. However, aligned films on the capillary inner wall are hardly controlled using the rubbing method. Therefore, in this study, we used photo-alignment PI films, which were diluted by N-methyl-2-pyrrolidinone at a weight ratio of 50%, and then heated to 120 °C for drying. We evacuated and coated the inner capillary by photo-alignment PI films with a thickness of 100–200 nm. The capillary is immersed in the PI solvent for 10 minutes, then a vacuum pump is used to suck out the PI solvent in the capillary, the PI on the inner surface of the capillary will be remained, the residual volume of the PI depends on the air pressure of the inner capillary and on the polyimide concentration. And then vertically exposed the capillary to polarized ultraviolet (UV) light, during which time we used a LUYOR-3120 UV flashlight (Manufactured by AmericaLuyor company) and an exposure intensity of 20 mW/cm². The polarization direction was along the capillary tube, and the orientation was initialized after UV irradiation for 10 min. Alignment direction of LC molecular on inner surface of the capillary is perpendicular to axis of the capillary. A double-frequency laser pump (Dawa-100 Nd:YAG, manufactured by Beamtech Optronics Co., Ltd., China) was used for all experiments. Specifically, the laser was operated at a wavelength of 532 nm with a maximum laser power of 50 mJ, a repetition rate of 10 Hz, and a pulse width of 8 ns. The emission spectra were tested using a fiber optic spectrometer with a resolution of 0.09 nm (PG2000, Idea Optics Co., Ltd., Shanghai, China). Energy meter (FieldMaxII-TOP, Coherent, INC, America) and temperature probe (50-II, fluke CO., LTD, America) were used in the experimental.

Fig. 1(a) shows the experimental platform. Fig. 1(b) shows the results of observation of a capillary filled with CLCs with and without PI film obtained with a polarizing optical microscope (POM). It is evident that the capillary with PI films shows obvious uniformity in brightness and the capillary without PI films exhibits no uniformity in brightness. These results indicate that photo-alignment PI films can effectively control the order and uniform orientation of LC molecules in the capillary and make the arrangement of CLC molecules more uniform. The cross-section of a capillary filled with CLCs with PI films is shown in Fig. 1(c), from which it can be observed that the direction of pitch is perpendicular to the capillary, indicating that the emission direction of the DFB-mode laser was also perpendicular to the capillary. Meanwhile, the LC molecules have an average refractive index
3. Experimental Results and Discussions

Fig. 2(a) shows the threshold for lasing of the capillary filled by DDCLCs with a 100-μm inner diameter without and with photo-alignment PI films. The green trigonal dots in Fig. 2(a) are the full width at half maximum (FWHM) of the lasing emission with the pumped energy. The threshold energy per pulse of the emitted laser without PI was 26.2 μJ · mm⁻². In contrast, after irradiation-based orientation of the photo-alignment PI films filling the capillary, the threshold energy per pulse of the emitted laser was 4.5 μJ · mm⁻², which is 21.7 μJ · mm⁻² lower than the value without photo-alignment PI films. Meanwhile, radius of pumping light which is focused by lens is about 300 μm, the spot area is bigger than the real excitation areas on the capillaries, so real lasing threshold is lower than 4.5 μJ · mm⁻². The threshold is much lower than that reported for NLC [16], CLC...
Fig. 3. Emission threshold as a function of input intensity with inner diameters of 100, 200, and 300 μm.

DFB-mode [17], and PDLC random lasers [18]. Light loss of the laser with PI was smaller than that without PI, because light scattering was decreased when LC molecules have a better alignment in the capillary with PI. Therefore, threshold energy of the laser with PI is lower than that without PI. Thus, we can set up stable laser emission at a lower pump-pulse energy. Fig. 2(b) shows the emission spectra without and with PI. The results show that the emission spectrum with PI has stronger emission intensity than that without PI under pumping light of the capillary with PI smaller than that without PI. The latter experimental samples were oriented using photo-alignment PI films. The circular capillary microcavity was optically pumped laterally or axially. A WGM resonant cavity was then formed between the LCs and capillary wall. Meanwhile, because of the photo-alignment on the capillary inner wall, helical structures were formed in the CLC, and thereby, cylindrical symmetry DFB-mode laser emission was produced. By operating in these two emission modes, the lasers were perpendicular to the capillary wall outward, which resulted in the enhancement of laser emission intensity with a range of 0–360° space angle covered.

To research the impact of the inner diameter of the capillary on emission threshold, we selected different inner diameters of the capillary. Fig. 3 shows the emission thresholds of the capillary laser, in which the inner diameter of the capillary was set as 100, 200, and 300 μm, respectively. It can be observed that the emission threshold for the capillary with an inner diameter of 100 μm was the lowest, when the pump energy exceeded the threshold energy, the laser output pulse energy increased rapidly with pump energy, and the laser's slope efficiency was great. In the thinner-inner-diameter capillary, LC molecules would have a stronger anchoring force because of the effect of the PI film, so the LC molecules would have better alignment than that in the thick-inner-diameter capillary. The thinner-inner-diameter capillary means the lower light loss and the lower emission threshold. Therefore, we selected a capillary with an inner diameter of 100 μm as the microcavity for the latter lasing experiment.

3.1 Temperature Tuning

To investigate the tunability of the capillary laser emission filled with CLCs with photo-alignment PI films, we measured the emission spectra of the capillary under different temperatures and voltages. We detected the output laser spectrum at different temperatures. As shown in Fig. 4(a), with a temperature increase from 25 to 30 °C, the emission spectra do not show wavelength shift obviously, but the lasing intensity changes, which is caused by different pumping power. And multi-mode DFB lasing in the emission spectra was dominant. This is consistent with Yoshiaki Uchida’s report [21], and the detailed mechanism responsible for the multi-mode lasing is not clear up to now. Multi-mode DFB lasing disappeared when the temperature increased to 35 °C as a result of the destruction of the helical structure of the CLCs and WGM lasing in the emission spectra appeared. The entire
heating process becomes increasingly better. This is mainly due to decreased light scattering when the temperature increased and the resonant power was enhanced. This indicated that threshold of the WGM lasing was lower than that DFB lasing. As the temperature was increased to 43 and 48 °C, WGM lasing became dominant. As shown in Fig. 4(b) and (c), which shows excellent WGM lasing, the free spectral range (FSR) is 1.05 nm and the corresponding Q factor is \(4 \times 10^3\), which is 1 order of magnitude better than reported [17].

The mode number \(l\) of the lasing wavelength can be simply expressed as \(l = \frac{2\pi n_{\text{eff}}}{\lambda}\), where \(n_{\text{eff}}\) is the effective refractive index of LCs, which decreased when the temperature was increased; \(r\) is the inner radius of the capillary; and \(\lambda\) is the resonant wavelength. Here, it can be assumed that \(n_{\text{eff}}\) and \(r\) is constant for different values of \(\lambda\) at the same temperature, so the mode number can be calculated as shown in Fig. 4(b) and (c). The shift of the lasing wavelength of WGMs with increasing temperature from 43 to 48 °C for mode number 563 is 3.04 nm.

### 3.2 Electric Field Tuning

We measured the emission spectra of the capillary filled with CLCs under different applied voltages, and the results and the electric field were applied as shown in Figs. 5(a) and 1(c), respectively. It can be observed that the single-mode DFB lasing in Fig. 5(a) [denoted by symbols] coexists with the WGM laser in the emission spectrum [21]. The wavelength of the single-mode DFB lasing showed a redshift from 608.56 to 610.80 nm, with a tuning range of 2.24 nm. The tuning range is smaller than that reported previously for a CLC DFB-mode laser [22]. The lasing wavelength of a single-mode DFB laser can be calculated by \(\lambda = n_e p\), where \(n_e\) and \(p\) are the extraordinary refractive index and the helix pitch, respectively, and the both were relative with orientation and magnitude of the electric field.

Additionally, the FSR of the WGM laser shifted from 1.3 to 2.1 nm, as shown in Fig. 5(b). The lasing wavelength and FSR of WGM can be calculated by \(\lambda = \frac{2\pi n_{\text{eff}}}{l}\) and \(\Delta \lambda = \frac{\lambda^2}{2\pi n_{\text{eff}}},\) respectively, and here \(n_{\text{eff}} = n_o n_e/\sqrt{n_r^2 \cos^2 \theta + n_r^2 \sin^2 \theta}\). At the boundary, the thickness of the anchoring layer that exists in the capillary because of the anchoring force of the photo-alignment PI film is nearly 10 nm [23], as shown by “h” in Fig. 6(a). The alignments of the LC molecules of the anchoring layer were barely changed under the electric field, and, if they were changed, the electric field tunability of the device would be increased. The LC molecules in the bulk of the capillary would be rotated under
the electric field, as shown in Fig. 6(b). There are two possible WGM polarizations in the capillary [24]: transverse-electric (TE) polarization, in which the electric field oscillates parallel to the surface of the sphere (Fig. 6(a), symbol), and transverse-magnetic (TM) polarization, in which the electric field is perpendicular to the surface [see Fig. 6(a)]. Before applying voltage, the TM modes sense the lower index $n_o$ and the TE modes sense the effective index $n_{eff}$. When voltage was applied, the TM modes sense the indexes $n_e$ and $n_o$ at position A, C and B, D, respectively, and the TM modes sense the index $n_{eff}$ at other positions. The TE modes sense the index $n_o$ at all positions. In short, for the TM modes, the index becomes slightly larger; for the TE modes, the index becomes smaller. For this reason, the electric field tunability of the device was not good. Therefore, if good tunable results are desired, the electric field structure needs to be improved. There is an improved electric field design that radial electric field matches radial helix alignment of LC molecular in this manuscript as shown as Fig. 6(c), so this design could be effective use of LC’s birefringence.

4. Conclusion
This paper, for the first time to our knowledge, presents the results of an investigation of a capillary microcavity laser filled with CLCs with photo-alignment PI films through theoretical analysis and
experiments. Experimental results show that capillary lasers with PI films have lower threshold energies than those without PI films, with the threshold energy per pulse obtained being 4.5 μJ · mm⁻². Moreover, capillaries with thinner-inner-diameters have a lower emission threshold, and a capillary laser shows excellent WGM under changing temperature. The paper also discussed electric field tunable results. These results collectively indicate that capillary microcavity lasers have potential application in temperature sensing device or analytical microdevices.

References


