Effect of liquid crystal concentration on the lasing properties of dye-doped holographic polymer-dispersed liquid crystal transmission gratings

Y. J. Liu and X. W. Sun[a)]
School of Electrical and Electronic Engineering, Nanyang Technological University, Nanyang Avenue, Singapore 639798, Singapore

H. I. Elim and W. Ji
Department of Physics, National University of Singapore, 2 Science Drive 3, Singapore 117542, Singapore

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Optically pumped single-mode lasing was achieved from a 4-(dicyanomethylene)-2-methyl-6-(4-dimethylaminostyryl)-4*H*-pyran dye-doped holographic polymer-dispersed liquid crystal transmission grating with various liquid crystal concentrations, which played an important role in the lasing generation and wavelength selection. With the decrease of the liquid crystal concentration, under the excitation of a frequency-doubled Nd:yttrium aluminum garnet laser operating at 532 nm, the lasing wavelength was blueshifted, and the full width at half maximum of the lasing peak became narrower. The lowest threshold pumping energy was found to be about 5 μJ/pulse at a liquid crystal concentration of 19.7 wt %. The lasing emission was thermally switchable due to the change of the refractive index modulation. © *2007 American Institute of Physics*. [DOI: 10.1063/1.2426885]

In recent years, liquid crystal (LC)-based photonic band gap materials have received considerable interest for optoelectronic applications. With the existence of a band gap, it is possible to achieve lasing with the use of proper dyes and pumping sources.[1] The dye-doped LC-based materials exhibit large optical gain over broad spectra in the visible range. Moreover, LC can be electrically or thermally tuned. These features are well suitable for use as tunable lasers. The most familiar LC-based lasing medium is the cholesteric LC with helical structure, a one-dimensional (1D) photonic crystal (PhC), which has been extensively investigated.[2–6] Yablonovitch theoretically described the lasing action from 1D structure, and other PhCs, through the use of distributed feedback theory.[7]

More recently, attention has been given to lasing in holographic polymer-dispersed liquid crystals (H-PDLCs). Bunning and co-workers observed laser emission from dye-doped H-PDLC reflection gratings,[8,9] which was subsequently followed by Luchetta *et al.*[10,11] and others.[12] The lasing action from dye-doped H-PDLC transmission gratings was also investigated recently.[13–15] Compared to the reflection grating, the transmission grating has a longer gain length, which facilitates low threshold lasing with narrow linewidth. Generally, two-dimensional (2D) and three-dimensional (3D) PhCs can create stronger coupled electromagnetic bands that lead to low group velocity dispersion and consequent local field enhancement, which favors low threshold lasing. A much better lasing performance has been reported in 2D square-lattice H-PDLC PhCs compared to the same materials in 1D PhCs.[16] Although the lasing properties can be improved much in 2D or 3D H-PDLCs, there are still rooms to improve the lasing performance in 1D H-PDLC structure, especially in H-PDLC transmission gratings.

In this letter, we report the effect of LC concentration on the lasing properties based on H-PDLC transmission gratings. We found that 35 wt % LC concentration was an approximately critical value to get a single-mode lasing from dye-doped H-PDLC gratings. With the decrease of the LC concentration, the lasing wavelength was blueshifted. At a LC concentration of 19.7 wt %, the lowest threshold of 5 μJ/pulse was obtained. In addition, the lasing can be thermally switched.

The prepolymer syrup used consisted of monomer, trimethylolpropane triacrylate (TMPTA), cross-linking monomer, *N*-vinylpyrrolidinone (NVP), photoinitiator, rose bengal (RB), coinitiator, *N*-phenylglycine (NPG), surfactant, octanoic acid (OC), all from Sigma-Aldrich, and liquid crystal, E7, from Merck. The E7 liquid crystal used has an ordinary refractive index of $n_o=1.521$ and a birefringence of $\Delta n=0.225$. The lasing dye, 4-(dicyanomethylene)-2-methyl-6-(4-dimethylaminostyryl)-4*H*-pyran (DCM) (from Sigma-Aldrich), was dissolved in prepolymer mixture as an active material. The wt % of every component in the syrup was tabulated in Table I. The fabrication process and setup for lasing measurement can be found elsewhere.[15] In addition, in the lasing measurement, a heater was used to control the temperature of the samples. The controllable temperature ranges from 20 to 80 °C, with an accuracy of about 3 °C.

Figure 1 shows the measured lasing threshold and lasing wavelength for the dye-doped H-PDLC as a function of the LC concentration. It can be seen from Fig. 1 that the LC concentration plays an important role in the lasing threshold and the lowest threshold of pumping energy is about 5 μJ/pulse at a LC concentration of 19.7 wt %. From Fig. 1, we can also see that with the decrease of the LC concentration from 34.2 to 14.6 wt %, the lasing wavelength is blueshifted from 627 to 584 nm. It is worth mentioning that, different from our previous report,[15] at almost the same LC concentration of 34 wt %, the lasing peak wavelength was 627 nm (compared to 609 nm in Ref. 15). The difference in lasing wavelength is due to the concentration of cross-linking monomer, NVP, which plays a crucial role in controlling the LC droplet size.[17] The relatively larger amount of

[a)]Author to whom correspondence should be addressed; electronic mail: exwsun@ntu.edu.sg

TABLE I. wt % of each component used in the prepolymer syrup for samples 1–6.

No.	TMPTA	NVP	NPG	RB	OC	DCM	E7
1	67.1	7.8	0.9	0.6	8.0	1.0	14.6
2	63.1	5.6	0.7	0.5	9.2	1.2	19.7
3	57.8	6.0	0.7	0.5	9.5	1.2	24.3
4	53.6	6.5	0.8	0.5	7.4	1.4	29.8
5	47.6	7.4	0.9	0.6	8.1	1.2	34.2
6	34.1	6.3	0.7	0.6	8.4	1.1	48.8

NVP in Ref. 15 leads to smaller droplets and thus smaller anisotropy of the LC droplets. Therefore, the index modulation (the refractive index difference between LC-rich lamellae to polymer-rich lamellae) in H-PDLC in Ref. 15 was smaller than that reported here, resulting in a blueshifted lasing wavelength.[18] It is worth mentioning that for the dye-doped H-PDLC transmission grating using material syrup No. 6 with 48.8 wt % LC, only amplified spontaneous emission (ASE) was obtained regardless of how high the pumping energy was.

Figure 2 shows the normalized lasing spectra for H-PDLC samples with various LC concentrations excited by about two times of the corresponding threshold. It can be seen from Fig. 2 that the linewidth of the lasing peak becomes narrower with the decrease of the LC concentration. The full width at half maximum (FWHM) of the lasing peak centering at 584 nm was less than 0.8 nm. The transmission curves along the grating vector direction are also theoretically calculated, as shown in Fig. 2. The refractive index of polymer matrices was assumed to be 1.54. The other data used in our calculations were tabulated in Table II. It is worth mentioning that in our calculations, the wavelength dispersion of the refractive indices of LC and polymer used is not considered because the refractive index changes of LC (Ref. 19) and polymer[20] are small enough to be ignored. It is evident that the lasing action happens at one edge of the reflection band. The inset of Fig. 2 shows a typical emission photograph of the dye-doped H-PDLC, where a sharp lasing point can be clearly observed. The estimated divergence angles from the far-field pattern were ~15 and ~65 mrad in the directions perpendicular and parallel to the substrates, respectively.

As reported previously,[16] lasing properties of H-PDLC transmission gratings are strongly dependent on the morphology of the grating. In our previous report,[21] the morphology significantly changed in H-PDLC transmission gratings as the LC concentration changed. At a higher concentration, the volume fraction of LC-rich lamella increases and the LC droplets become larger. As a result, a larger scattering loss will be induced and the lasing has a larger threshold and wider FWHM. At a lower concentration, the volume fraction of LC-rich lamellae decreases and the LC droplets become smaller. Within the LC-rich lamellae, a large amount of polymer exists and the index modulation is lowered accordingly. As a result, only a higher threshold can sustain an enough distributed feedback, and a narrower FWHM is obtained due to the smaller scattering loss. Moreover, lower LC concentration reduces the index modulation, resulting in a blueshifted band gap. Therefore, the lasing wavelength is blueshifted accordingly. In our experiments, we found that the

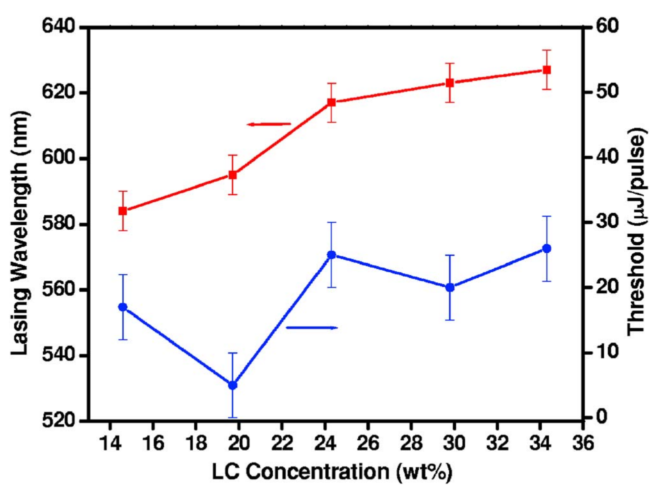

FIG. 1. (Color online) Measured lasing threshold and lasing wavelength from the dye-doped H-PDLC as a function of the LC concentration. The accuracies for the threshold and lasing wavelength are around ±5 μJ/pulse and ±6 nm, respectively.

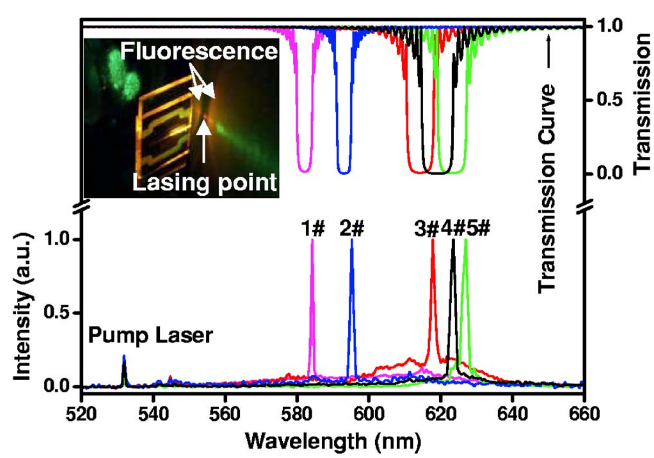

FIG. 2. (Color online) Normalized lasing spectra for H-PDLC samples with various LC concentrations excited by about two times of the corresponding threshold. The inset shows a typical emission photograph of the dye-doped H-PDLC.

TABLE II. Data used for the transmission curve calculation. Λ_G is the grating pitch, $V_{LC}:V_p$ is the volume ratio of LC-rich lamellae to polymer-rich lamellae, and Δn_1 is the index modulation.

No.	Λ_G (μm)	$V_{LC}:V_p$	Δn_1
1	0.564	16:84	0.05
2	0.572	25:75	0.07
3	0.583	45:55	0.09
4	0.584	50:50	0.10
5	0.584	55:45	0.11

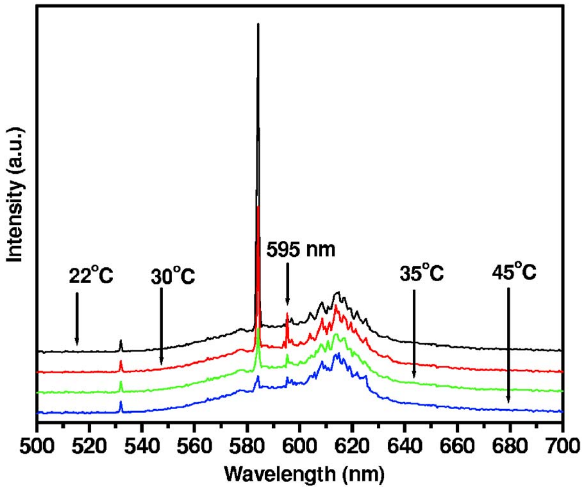

FIG. 3. (Color online) Emission spectra from dye-doped H-PDLC gratings at different temperatures.

LC concentration of around 35 wt % was close to the critical value to obtain a single-mode lasing from dye-doped H-PDLC transmission grating. At a higher concentration of 48.8 wt %, only ASE was observed. Furthermore, to achieve a lasing emission with better properties from H-PDLC transmission gratings, the LC concentration of around 20 wt % was found to be the best trade-off between sufficient refractive index modulation and optical scattering in the samples.

For the devices based on H-PDLC, a distinct advantage is that they can be tuned or switched by an external stimulus. Hsiao *et al.* reported the electrically switchable lasing emission from the dye-doped H-PDLC transmission gratings.[13] Besides electric tuning, the lasing can be also thermally switched. Figure 3 shows the emission spectra of dye-doped H-PDLC gratings using No. 1 material syrup at various temperatures with a fixed pumping energy of 40 μJ/pulse. From Fig. 3, we can see that the lasing can be thermally switched. It is interesting to note that at a temperature of 30 °C, another lasing peak centered at 595 nm appears in the emission envelope (Fig. 3), which indicates that at this temperature, a sufficient gain can also be obtained for 595 nm. The switching mechanism of the H-PDLC grating is due to the index modulation of the phase separated LC droplets. Generally, H-PDLC gratings are slightly anisotropic because of the volume shrinkage during the grating formation.[22,23] With the increase of the temperature, the optical anisotropy reduces, and accordingly the effective refractive index of the LC changes. At a certain elevated temperature, the effective refractive index of the LC may match closely with the refractive index of the polymer matrix. As a result, the lasing peak disappears. Unlike Ref. 16, where the lasing wavelength was shifted by about 5 nm, in our experiments, there is no shift in lasing wavelength. From Fig. 3, we can see that at the temperature of 45 °C, both lasing peaks nearly disappear. After cooling down, the lasing emission appeared again.

In conclusion, the effect of the LC concentration on the lasing properties based on dye-doped H-PDLC transmission gratings was investigated. With the decrease of LC concentration, the lasing wavelength showed a blueshift, and the FWHM of the lasing peak became narrower. The lowest threshold pumping energy was found to be about 5 μJ/pulse at a LC concentration of 19.7 wt % among five different LC concentrations studied in our experiment. A LC concentration of around 20 wt % was found to be the best trade-off between sufficient refractive index modulation and optical scattering in our samples. In addition, the lasing emission was thermally switchable due to the change of the refractive index modulation.

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