Low-threshold and narrow-linewidth lasing from dye-doped holographic polymer-dispersed liquid crystal transmission gratings

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

H. P. Li, J. Mi, and W. Ji
Department of Physics, National University of Singapore, 2 Science Drive 3, Singapore 117542

X. H. Zhang
Institute of Materials Research and Engineering, 3 Research Link, Singapore 117602

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Optically pumped lasing with low threshold and narrow linewidth was observed in a 4-dicyanomethylene-2-methyl-6-p-dimethylaminostyryl-4H-pyran dye-doped holographic polymer-dispersed liquid crystal transmission grating structure. The results showed that the lasing peak centered at about 609 nm, with a full width at half maximum of about only 1.8 nm. The threshold pumping intensity was about 120 μJ under the excitation of a frequency-doubled Nd:yttrium–aluminum–garnet laser operating at a wavelength of 532 nm. Theoretical calculation showed that the lasing from this structure happened at the band edge of the photonic band gap. The lasing modes were also investigated. The transmission grating investigated enjoys a much larger gain length compared to the reflection one. © *2006 American Institute of Physics*. [DOI: 10.1063/1.2172161]

In recent years, holographic polymer-dispersed liquid crystal (H-PDLC) materials [1] have attracted much attention, not only for their physical properties, but also for a wide range of potential applications, such as reflective flat-panel displays,[2–4] switchable lenses,[5] optical switches for communications,[6,7] reflective strain gauges,[8] application-specific lenses,[9] spatially patterned devices,[10] image capture systems,[11] and remote sensing.[12] A H-PDLC Bragg grating, fabricated by two interfering laser beams, is composed of parallel, periodic polymer-rich and liquid crystal-rich lamellae.[13] It is actually a one-dimensional (1D) photonic crystal. Using holography technique, two- and three-dimensional periodic structures have also been fabricated.[14] Recently, light amplification in dye-doped H-PDLC was investigated by Lucchetta *et al.*[15] Lasing from a dye-doped H-PDLC reflection grating has also been reported.[16,17] Hsiao *et al.* reported the lasing emission from dye-doped H-PDLC transmission grating.[18] Compared with the reflection grating, the transmission grating enjoys a longer gain length, which facilitates low threshold lasing.

In this letter, we shall report lasing emission from dye-doped H-PDLC transmission grating. The lasing peak centered at about 609 nm, and its full width at half maximum (FWHM) is about 1.8 nm and the threshold pumping energy is about 120 μJ. The experimental results showed that the emitted laser linewidth was much narrower than that reported in Ref. 18.

As previously reported,[19] our H-PDLC transmission gratings were fabricated from a formulation consisted of 41 wt% monomer, trimethylolpropane triacrylate, 16 wt% cross-linking monomer, N-vinylpyrrollidone, 0.7 wt% photoinitiator, rose bengal (RB), 1.3 wt% coinitiator, N-phenylglycine, 7 wt% surfactant, octanoic acid, all from Sigma-Aldrich, and 34 wt% 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-p-dimethylaminostyryl-4H-pyran (DCM) (from Sigma-Aldrich), was dissolved in pre-polymer mixture as an active material at a concentration of 1 wt%. All the materials were mechanically blended according to the appropriate weight ratio at 65 °C (higher than the clearing point of the liquid crystal E7) to form a homogeneous mixture in dark condition.

The mixture was sandwiched in a cell that was formed by two pieces of indium tin oxide (ITO) coated glass. The cell was then placed behind the base of a right angle prism to record the hologram pattern, which was obtained by interference of two beams split from an Ar^+ laser operating at 514.5 nm. The exposure intensity on the sample was about 20 mW/cm^2 of each beam and the exposure time was 2 min. After exposure, the samples were further cured for 5 min by a mercury lamp to stabilize the polymerization. The thickness of the samples was $\sim$30 μm and the recorded grating area was about 6×10 mm^2. A Q-switched frequency-doubled Nd:yttrium–aluminum–garnet pulsed laser (Spectra Physics DCR3) operating at 532 nm with a pulse duration of 7 ns and repetition rate of 10 Hz was used as the pumping light source for the lasing experiment. For scanning electron microscopy (SEM) analysis, the test samples were broken with the ITO glass on one side removed, soaked in ethanol for more than 12 h to remove liquid crystal, and finally dried.

Figure 1 shows the schematic setup of the lasing measurement. The laser beam was focused onto the surface of the grating film by a cylindrical lens, which has a focal length of $f=40$ cm, to form a narrow strip gain area along the grating vector 0.5 mm wide and 10 mm long. This focused line can cover thousands of periods of the grating, and thus ensure sufficient distributed feedback. A fiber pigtail

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

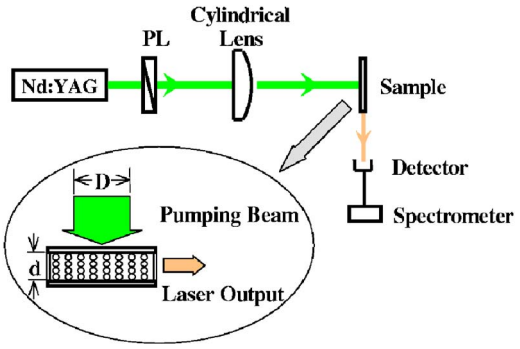

FIG. 1. (Color online) Schematic setup of the lasing measurement.

collector was placed facing the laser outputs to collect the lasing signal, which was fed into to a spectrometer that was monitored real time by a computer.

The absorption and photoluminescence (PL) spectra of the DCM-doped H-PDLC films are shown in Fig. 2. It can be seen from Fig. 2 that, the DCM-doped H-PDLC film exhibits a broad fluorescence emission band (from 520 to 750 nm) and the absorption band is in the UV range (high absorption, not shown here) and visible range with wavelength less than 600 nm. In addition, in the spectral range of 500–590 nm there is obvious overlap between the absorption band and the PL band. Therefore, the lasing action will not occur in this spectral range due to the reabsorption of the emitted fluorescence signals. In contrast, lasing above 600 nm wavelength can be expected, where the absorption is negligible and the gain is relatively high. In our experiments, the writing wavelength was 514.5 nm, as indicated in Fig. 2. At this specific wavelength, the photoinitiator, RB, will absorb more light than the DCM dye, which ensures not only the complete polymerization, but also an as low as possible photodegradation of the dye. Moreover, a low intensity UV light was used to avoid the photodegradation of the dye during the postcure of the gratings.

In our experiments, the wavelength used for writing the grating was 514.5 nm. According to the geometrical structure of exposure, the grating pitch can be calculated using the following formula:

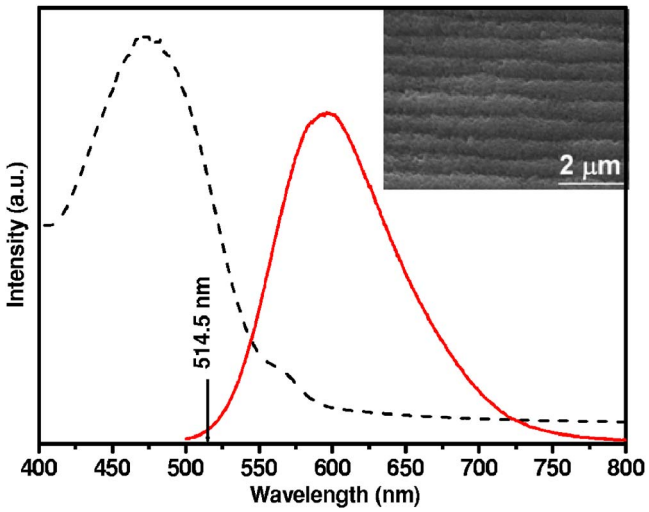

FIG. 2. (Color online) Absorption (dashed) and PL (solid) spectra of DCM-doped H-PDLC grating films. The wavelength used to write the gratings, 514.5 nm, is indicated in the figure. The inset is a SEM image showing the surface morphology of the grating.

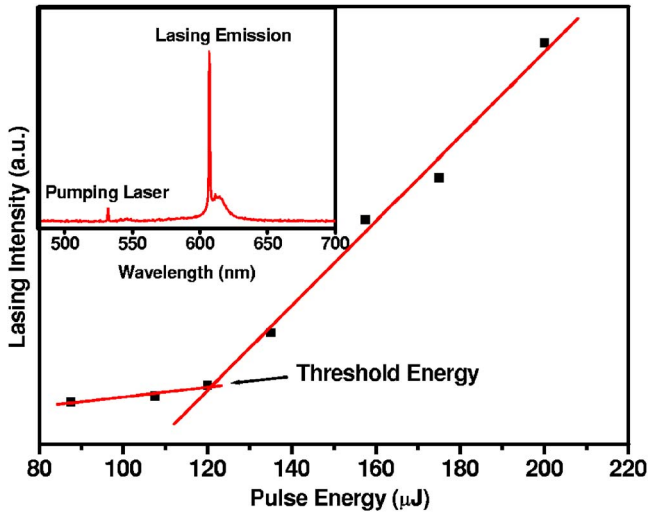

FIG. 3. (Color online) Output lasing intensity from DCM-doped H-PDLC transmission grating as a function of the laser pumping energy. The inset shows the lasing and the pump laser spectra.

$$\Lambda = \frac{\lambda}{2n\cos[\alpha + \sin^{-1}(n^{-1}\cos\alpha)]}, \tag{1}$$

where λ is the recording wavelength, n is the refractive index of the prism, and α is the incident angle. In our experiments, $\lambda = 514.5$ nm, $n = 1.52$, and $\alpha = 45°$, therefore, the calculated grating pitch is about 0.57 μm. The inset in Fig. 2 shows the surface morphologies of the DCM-doped H-PDLC grating. It can be seen that a clear and relatively uniform grating structure is formed and the grating period is about 0.58 μm, which is in good agreement with the theoretical calculation.

The measured output lasing intensity from the dye-doped H-PDLC as a function of the pumping laser energy is shown in Fig. 3. It can be seen from Fig. 3 that, the threshold pumping energy is about 120 μJ, which is considerably lower than previously reported.[18] A possible reason for the low threshold excitation intensity may be due to the uniform grating, which provides uniform coherent distributed feedback. We found that the uniformity strongly affected the performance of the lasing emission. The higher uniformity of the grating decreases the scattering, increases the coupling of

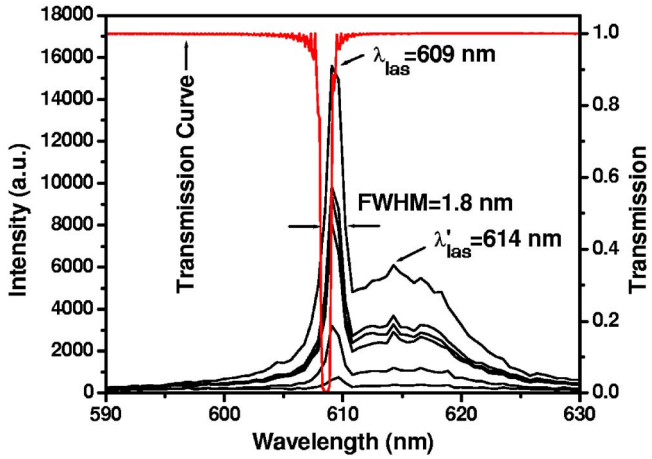

FIG. 4. (Color online) The lasing spectra obtained at various pumping intensities for DCM-doped H-PDLC transmission gratings. The simulated transmission spectrum of the grating along the grating vector direction is also shown in the figure.

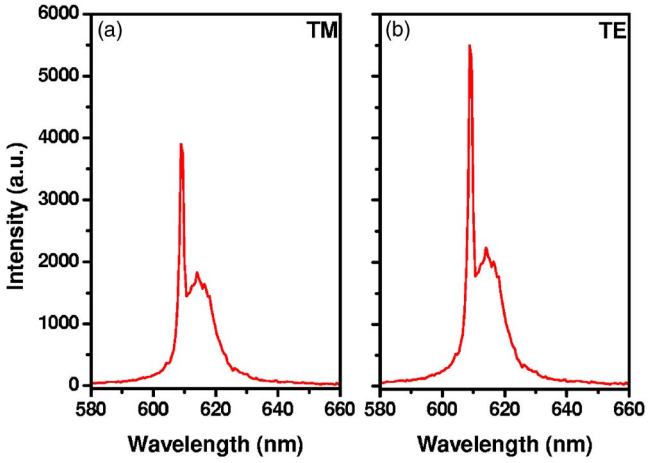

FIG. 5. (Color online) The TM (a) and TE (b) polarized emission spectra for the DCM-doped H-PDLC transmission grating.

the pumping light, and most importantly, provides uniform coherent distributed feedback, resulting in lower threshold. Above the threshold, we can see that, the emitted laser intensity linearly increases with increasing pumping laser energy. Far above the threshold energy, dye photodegradation was observed, which has been reported previously.[20,21]

Figure 4 shows the lasing spectrum excited with various pumping laser intensities. It can be seen from Fig. 4 that the lasing peak centers at about 609 nm with the FWHM of about 1.8 nm, which is the narrowest linewidth for the dye-doped H-PDLC gratings. It is worth mentioning that the lasing peak was not absolutely accurate because of the low resolution (only about 0.6 nm) of the spectrometer used. As previously reported for dye-doped reflection gratings, lasing is generally generated at the edge of the band gap of the 1D photonic crystal, because the photon group velocity approaches zero and the gain of the grating is enhanced greatly at the edge of band gap.[21,22] For our transmission H-PDLC grating, we have calculated the band gap of the grating. Shown in Fig. 4 is also the transmission spectrum of our transmission grating, a 1D photonic crystal. The data used in our calculation are as follows: the grating pitch, Λ, is 0.58 μm, the effective refractive indices, n_{LC} and n_p, of liquid crystal-rich and polymer-rich channels are 1.60 and 1.54, respectively, liquid crystal and polymer V_{LC} and V_p, are 0.3 and 0.7, respectively. It is clear from Fig. 4 that the lasing emission is located at one edge of the reflection band, which is in good agreement with the theoretical calculation. We can also see from Fig. 4 that, with the increase of pumping intensity, a new peak around 614 nm appears. This is because the grating structures cannot provide enough gain suppression to the residual stimulated emission, i.e., amplified spontaneous emission.

For lasers, an important parameter is the quality factor, Q, which can be described by

$$Q \approx \frac{\lambda_{\mathrm{las}}}{\Delta\lambda}, \tag{2}$$

where λ_{las} the laser wavelength emitted from the grating, and $\Delta\lambda$ is the linewidth. From Fig. 4, with $\lambda_{\mathrm{las}}=609$ nm and $\Delta\lambda=1.8$ nm, we can obtain the quality factor, Q, of about 338, which is comparable to the conventional lasers. However, the Q factor is lower compared with that of other microlasers,[23] due to the imperfection of the gratings.

The lasing modes from the dye-doped H-PDLC gratings were also investigated. Figure 5 shows the transverse magnetic (TM) and transverse electric (TE) lasing modes from DCM-doped H-PDLC structure. From Fig. 5, we can see that both TM and TE modes exist in the H-PDLC structure and the intensity of TE mode is a little larger than that of the TM mode.

In conclusion, lasing action was demonstrated in a DCM dye-doped H-PDLC transmission grating structure. The transmission mode structure offers a much larger gain length and more sufficient feedback than the reflection mode. The threshold pumping energy was about 120 μJ at the pumping wavelength of 532 nm. The lasing peak centered at about 609 nm, and its FWHM was about 1.8 nm. Theoretical calculation showed that the lasing from this structure happened at the band edge of the photonic band gap.

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