Japanese Journal of Applied Physics
Vol. 45, No. 22, 2006, pp. L559–L561

Lasing from a One-Dimensional Photonic Crystal Made of Dye-Doped Holographic Polymer-Dispersed Liquid Crystal Gratings

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(Received January 2, 2006; accepted April 23, 2006; published online June 2, 2006)

Lasing action was observed in a one-dimensional photonic crystal made of rhodamine B dye-doped holographic polymer-dispersed liquid crystal gratings. The results showed that the lasing peak centered at about 604 nm, with a full width at half maximum of about 6.0 nm. The threshold pumping energy was about 80 μJ at the pumping wavelength of 532 nm. Theoretical simulation showed that the lasing from this structure happened at the photonic bandedge of the one-dimensional photonic crystal. The lasing modes were also investigated. [DOI: 10.1143/JJAP.45.L559]

KEYWORDS: polymer-dispersed liquid crystal, holographic grating, photonic crystal, lasing

Photonic bandgap materials have attracted great attention with new fundamental physics and practical applications.[1)] Dye-doped cholesteric liquid crystal (CLC) or ferroelectric liquid crystal (FLC) with helical structure, which can be viewed as one-dimensional photonic crystals (PCs), have been extensively investigated.[2–5)] PCs have photonic bandgap that is similar to the electronic bandgap in semiconductors. At the edges of the bandgaps, the group velocity approaches zero, so the gain is greatly enhanced.[6)] Holographic polymer-dispersed liquid crystal (H-PDLC)[7,8)] as a new photonic bangap material has attracted great attention recently. Using holography technique, one-, two-, and three-dimensional (1-, 2-, and 3-D) PCs with periodic structures have also been fabricated.[9)] An H-PDLC Bragg grating, fabricated by two interfering laser beams, is composed of parallel, periodic polymer-rich and LC-rich lamellae.[10)] It is actually a 1-D PC. Recently, light amplification and lasing emission in dye-doped H-PDLC reflection gratings were investigated.[11–13)] The lasing emission from dye-doped H-PDLC transmission grating was also observed.[14,15)] Compared to the reflection grating, the transmission grating offers a longer gain length and stronger distributed feedback. In this paper, we shall report laser emission from a 1-D PC based on dye-doped H-PDLC transmission gratings, with a low threshold pumping energy of about 80 μJ.

In our experiments, the recipe of materials to fabricate the 1-D PC made of dye-doped H-PDLC transmission grating consisted of 47.0 wt % monomer, trimethylolpropane triacrylate (TMPTA), 6.0 wt % cross-linking monomer, N-vinylpyrrollidone (NVP), 0.8 wt % photoinitiator, rose bengal (RB), 1.2 wt % coinitiator, N-phenylglycine (NPG), 10.0 wt % surfactant, octanoic acid (OC), and 1.0 wt % lasing dye, rhodamine B (RhB) (all from Sigma-Aldrich), and 34.0 wt % LC, E7 (from Merck). The LC used has an ordinary refractive index of $n_o = 1.521$, and birefringence of $\Delta n = 0.225$. The lasing dye was dissolved in prepolymer 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 LC E7) to form a homogeneous mixture in dark.

The mixture was sandwiched in a cell which was formed by two pieces of indium–tin-oxide (ITO) coated glass, and then the cell was placed behind the base of a right angle prism to record the hologram pattern, which was obtained by interference of two beams 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 ensure complete consumption of monomers. The thickness of the samples was 30 μm and the recorded grating area was about 6×10 mm^2. For scanning electron microscopy (SEM) analysis, the sample was broken with the ITO glass on one side removed, soaked in ethanol for more than 12 h to remove LC, and finally dried. The pumping source for lasing action is a linearly polarized, Q-switched, and frequency doubled 532 nm Nd:YAG pulsed laser (Spectra Physics DCR3) with a pulse duration of 7 ns and repetition rate of 10 Hz.

The schematic setup of the lasing measurement is shown in Fig. 1. The 532 nm 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 region along the grating vector with an area of about 10×0.5 mm^2. This focused line can cover thousands of periods of the grating, and thus ensure a sufficient feedback even though the thickness of the grating film is as thin as that of the reflection grating film. The detector was placed in the direction, where the laser outputs, to collect the lasing signal, which was fed to a spectrometer and monitored real-time by a computer.

The absorption and photoluminescence spectra of the dye-doped grating film are shown in Fig. 2. It can be seem from Fig. 2 that, the RhB-doped H-PDLC film exhibits a broad fluorescence emission band (from 550 to 700 nm) and the absorption band is in the range of UV (high absorption, not shown here) and visible range (less than 600 nm). In addition, in the spectral range of 500–600 nm, there is obvious overlap between the absorption band and the photoluminescence band. Therefore, the lasing wavelength should be expected above 600 nm, 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 RhB, which ensures not only a complete polymerization, but also a low photodegradation of

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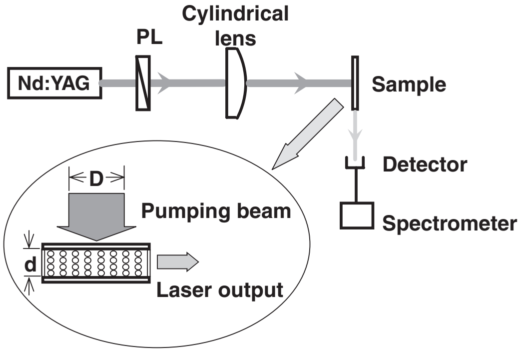

Fig. 1. The schematic setup of lasing measurement.

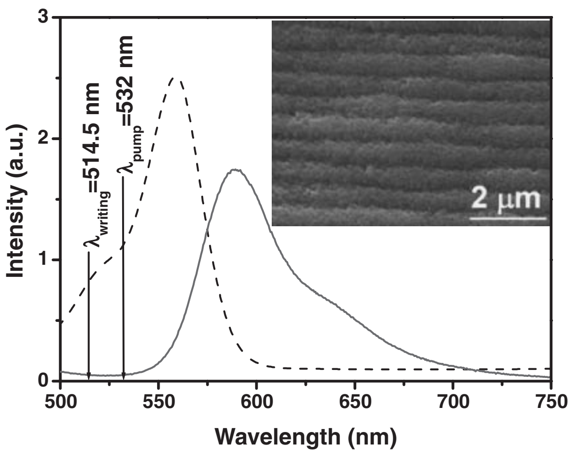

Fig. 2. The absorption and PL spectra of RhB. The dash line and the solid line denote the absorption curve and the PL curve, respectively. The writing wavelength, 514.5 nm, and the pumping wavelength, 532 nm, are shown in the figure. The inset shows the surface morphology of the 1-D PC made of RhB-doped H-PDLC.

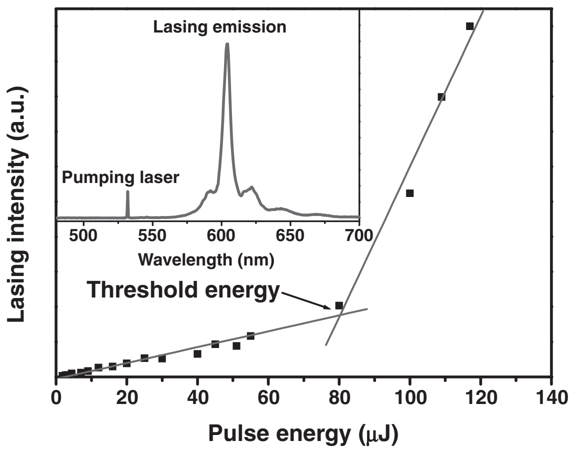

Fig. 3. The output lasing intensity from RhB-doped H-PDLC 1-D PC as a function of the pumping energy. The inset shows the lasing spectrum captured together with the pumping laser at the pumping energy of 109 μJ.

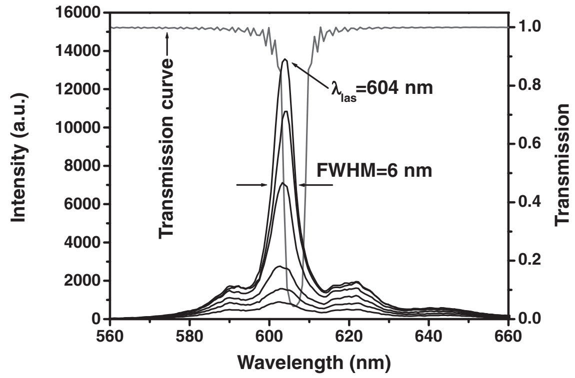

Fig. 4. The lasing emission spectra with various pumping energies for RhB-doped H-PDLC 1-D PC. The pumping energies are 40, 55, 80, 100, 109, and 117 μJ from lower to higher emission, respectively. The theoretically calculated transmission curve along the grating vector direction is also shown in the figure.

the dye. The inset shows the surface morphology of the RhB-doped H-PDLC 1-D PC. It can be seen that, a good periodic structure is formed and the period is about 0.58 μm.

The measured output lasing intensity from the dye-doped H-PDLC cavity as a function of the pumping energy is shown in Fig. 3. It can be seen from Fig. 3 that, the threshold pumping energy is about 80 μJ per pulse, which is much lower than that reported in ref. 14 with the same structure. A possible reason may induce this low threshold, the uniformity of grating. We found that the uniformity strongly affected the performance of the lasing emission. The higher uniformity of the grating decreases the scattering and increases the coupling of the pumping light, thus, resulting in lower threshold. Above the threshold, the emitted laser intensity linearly increases with the increase of the pumping energy.

Figure 4 shows the lasing emission with different pumping energy. It can be seen from Fig. 4 that, the lasing peak centers at about 604 nm, with a FWHM of about 6.0 nm. It is worth mentioning that, the lasing peak was not absolutely accurate because of the resolution (only about 0.6 nm) of the spectrometer used. The laser linewidth is a bit broader than some reported previously.[13–15] However, from Fig. 4, we can clearly see optical confinement with two small shoulders due to the distributed feedback of the grating structure, which is different from the gain narrowing obtained in dye-doped PDLC with randomly distributed LC droplets.[16] So lasing is used in our discussion. As previously reported,[3,5,13] lasing is generally generated at the edge of the bandgap, because the photon group velocity approaches zero and the gain of the grating is enhanced greatly at the edge of bandgap.[6] The transmission grating can also be considered as a 1-D PC along the grating vector direction. Although difficult to measure experimentally, the transmission spectra can be theoretically calculated,[13] as shown in Fig. 4. The data used in our calculation are that, the grating pitch, Λ, is 0.58 μm, the effective refractive indices of LC-rich and polymer-rich channels are, $n_{\mathrm{LC}} = (2n_{\mathrm{o}} + n_{\mathrm{e}})/3 = 1.60$ and $n_{\mathrm{p}} = 1.53$,[17] respectively. The volume ratios of LC and polymer, V_{LC} and V_{p}, are 0.4 and 0.6, respectively. It is clearly seen that the lasing emission located at one edge of the stop band, which is in good agreement with the theoretical calculation. In our experiment, the lasing preferred to happen at the higher energy edge of the bandgap because of the higher gain at this edge, which can be judged from the PL spectra in Fig. 2.

The lasing modes from the dye-doped H-PDLC 1-D PC

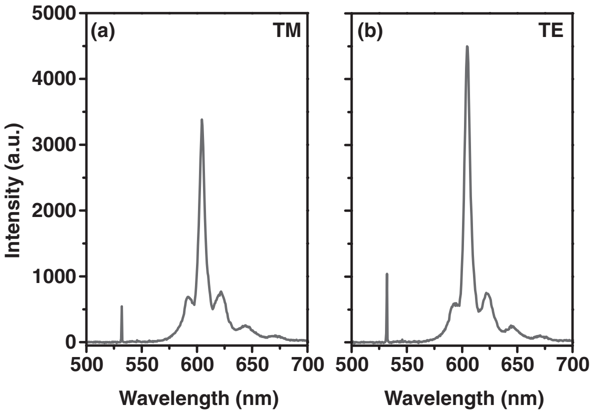

Fig. 5. (a) TM and (b) TE polarized emission spectra observed for RhB-doped H-PDLC 1-D PC.

were also investigated (Fig. 5). From Fig. 5, we can see that, both transverse magnetic (TM) and transverse electric (TE) modes exist in the H-PDLC structure and the intensity of TE mode is slightly stronger than that of the TM mode. The film thickness of H-PDLC plays an important role in selecting the lasing modes. It has been reported that, for a very thin film (less than 0.5 μm), only TE mode was observed while TM mode was suppressed.[18)] For our case, although the H-PDLC film is much thicker (about 30 μm), slight difference might still be induced between the coexisted TE and TM modes. On the other hand, for H-PDLC, there is a known shrinkage existed during the photo-induced polymerization,[19)] which makes the droplets anisotropic because the LC director in the droplet has a preferred direction. As a result, our H-PDLC 1-D PC should exhibit an anisotropic distributed feedback. Therefore, the TE and TM modes should have different cutoff thickness, resulting in small intensity difference.

In conclusion, lasing action was observed in 1-D PC made of RhB-doped H-PDLC. One-step holographic exposure simplified the fabrication. Compared with the refection mode gratings, the transmission mode structure offers a much larger gain length, more sufficient distributed feedback and low threshold. The emitted laser centered at about 604 nm with a linewidth of about 6.0 nm.

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