

High contrast switching of distributed-feedback lasing in dye-doped H-PDLC transmission grating structures

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Abstract: Electrically switched distributed-feedback (DFB) lasing action is presented in a Pyrromethene 580 lasing dye-doped holographic polymer dispersed liquid crystal (H-PDLC) transmission grating structure. This design, when compared with the previously utilized H-PDLC reflection grating structure, has the advantage of a greatly enlarged gain length (10 mm) and a low concentration of liquid crystal (20%) while maintaining sufficient refractive index modulation. The experimental results demonstrate that the emitted laser bandwidth (~5 nm) can be obtained with a pump energy threshold of ~0.3 mJ at three different wavelengths, 561 nm, 569 nm and 592 nm, corresponding to three different grating spacings. The near- and far-field measurements have shown a high directionality of the lasing output. The lasing can be electrically switched off by an applied field of 30V/ μm . The temporal, spectral, and output/input properties of the laser output are also presented.

OCIS codes: (090.2890) Holographic optical element; (140.2050) Dye Lasers

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1. Introduction

Photofabrication of periodic structures using holographic interferometry has been widely studied in photopolymer/liquid crystal (LC) mixtures. The structure made by holographic polymer dispersed liquid crystal (H-PDLC) materials has already demonstrated applications in photonic areas [1, 2]. Moreover, one-, two-, and three-dimensional periodic structures (such as face centered cubic (FCC) or diamond-like structures) are readily fabricated using the interference of two or more laser beams [3]. The most attractive feature of the H-PDLC system is that the LC provides a mechanism to switch the optical state of the structure from "on" to "off" by applying a suitable electric field [4]. Recently stimulated emission from a dye-infiltrated (dye-doped) H-PDLC reflection grating has been reported [5, 6]. The emitted lasing wavelength coincides with the edge of the bandgap structure due to the fact that the photon group velocity approaches zero and through this effect the gain (G) of the reflection grating is enhanced [7, 8]. These reports provide results similar to the lasing action occurring within a periodic structure or distributed feedback (DFB) cavity in a dye-doped polymer film [9, 10]. In addition, an H-PDLC reflection grating film can also be employed as a reflective cavity element that provides narrow spectral-band feedback [11, 12].

In the above mentioned cases of using a dye-doped H-PDLC reflection grating film to generate lasing with a vertical microcavity [9, 10], the grating planes are parallel to the film surface and the lasing is emitted normal to the substrate. In such a case, the overall gain length is limited to the thickness of the film. In order to increase the lasing efficiency and reduce the lasing spectral linewidth, it is desirable to increase the film thickness and therefore increase the total number of feedback layers. However, in the reflection geometry, an increase of film thickness results in an increase of the required switching electric field. Thus, a tradeoff between cavity length and switching field is necessary. Stimulated emission from a DFB structure is enhanced by Bragg scattering due to the spatial refractive index modulation of the gain medium. For the condition where the gain G exceeds the threshold value at the center wavelength by the factor of 2, the lasing threshold is exceeded over a bandwidth ($\Delta\lambda$) given by:

$$\frac{\Delta\lambda}{\lambda} \cong \left(\frac{\lambda}{4\pi\Delta nL} \right) \times \ln(G), \quad (1)$$

where λ is the pumping wavelength, L is the length of gain medium, n is the refractive index, and Δn is refractive index modulation [13].

In order to maintain a narrow bandwidth of $\Delta\lambda$ (~ 1 nm) one requires either a large refractive index modulation or a large gain medium length. However, large modulations in refractive index are difficult to achieve. As an example, a DFB with a typical $\Delta n \sim 10^{-5}$ and a gain $G=100$ requires a gain length of 10 mm when operating at $\lambda=0.63$ μm to produce sufficiently narrow spectral bandwidths [13]. In this paper a dye-doped H-PDLC transmission grating film that is transversely pumped is used as the DFB lasing device structure. This

transversely pumped geometry allows one to (a) achieve a long gain medium length, (b) obtain sufficient feedback by using a large number of grating periods with a small modulation in refractive index, (c) reduce the electric field necessary to electrically switch the lasing, and (d) reduce the optical scattering due to the LC since the small index modulation requires a low concentration of liquid crystal. In this case, the grating layers are perpendicular to the surfaces of the film and the substrate, and the laser emission is from the edge of the film (perpendicular to the grating layers). To accomplish this, a thin stripe pump laser beam (10 mm long) is created by focusing via a cylindrical lens onto the film surface along the direction perpendicular to the grating layers (inset of Fig. 1). Obviously, in this design, the effective optical cavity with around thousands of periods is chosen by the transverse pump length, and the film thickness can be kept as thin as that for reflection grating films. In this letter the preliminary results of this transverse optically pumped and electrically switchable lasing device, fabricated using a dye-doped H-PDLC transmission grating film, are presented.

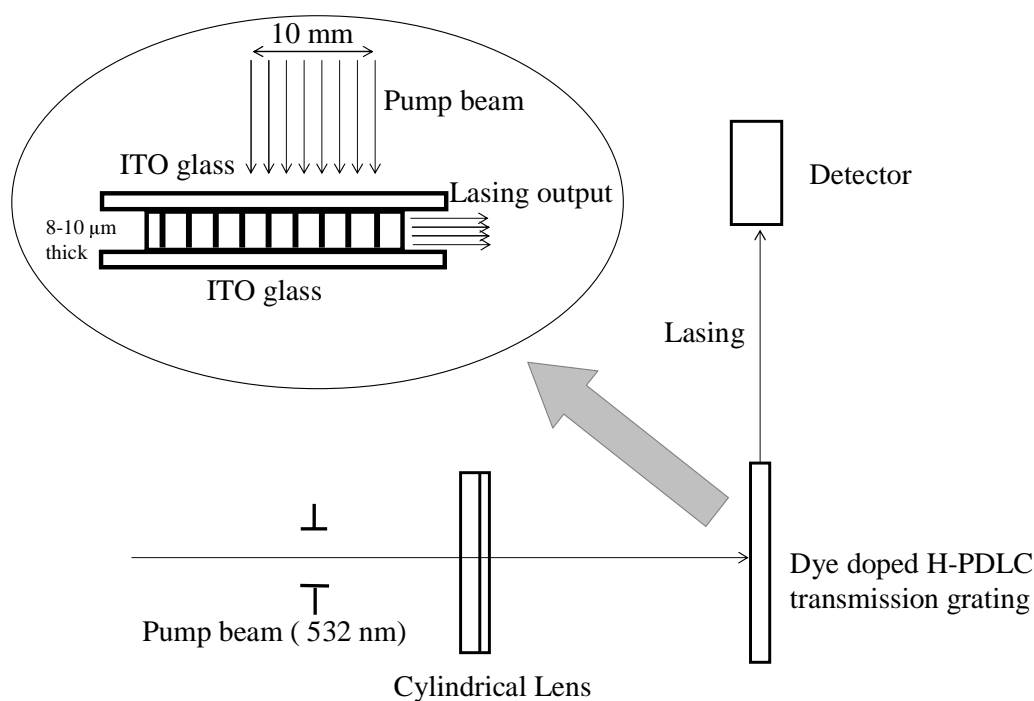


Fig. 1. Schematic diagram of the lasing experiment.

2. Experimental

To fabricate the dye doped transmission grating in the H-PDLC structure, a pre-polymer syrup consisted of 20 wt% LC (TL038, Merck), 2 wt% photo-initiator (Rose Bengal, Spectral Group), 3 wt% co-initiator (*N*-phenyl glycine, Aldrich), 15 wt% chain extender (*N*-vinyl pyrrolidinone, Aldrich), 58 wt% monomer (dipentaerythritol penta/hexa acrylate, monomer, Aldrich), and 2 wt% Pyrromethene 580 lasing dye (PM 580, Exciton). The writing geometry is accomplished by interference of two coherent s-polarized laser beams from a 514 nm Ar ion laser source and the grating spacing can be changed by adjusting the angle between the two interference beams as dictated by the Bragg condition. The thickness of the film is controlled

by 8 μm spacers and the grating area is maintained at the area of 10 x 10 mm^2 . The technical details of the fabrication of the grating samples with different grating periods were previously described [4]. The pump laser source for lasing experiment is a frequency doubled 532-nm Nd:YAG pulsed laser with a pulse duration of ~ 10 ns, beam size of ~ 10 mm, divergence angle of ~ 1 mrad, repetition rate of 1-10 Hz, and a pulse energy of 0.3-2 mJ. The schematic setup of the lasing experiment is shown in Fig. 1. The 532-nm laser beam is focused through a cylindrical lens of $f=10$ cm onto the surface of the grating film to form a narrow (~ 0.1 mm wide) strip gain area of ~ 10 mm long along the direction perpendicular to the grating layers.

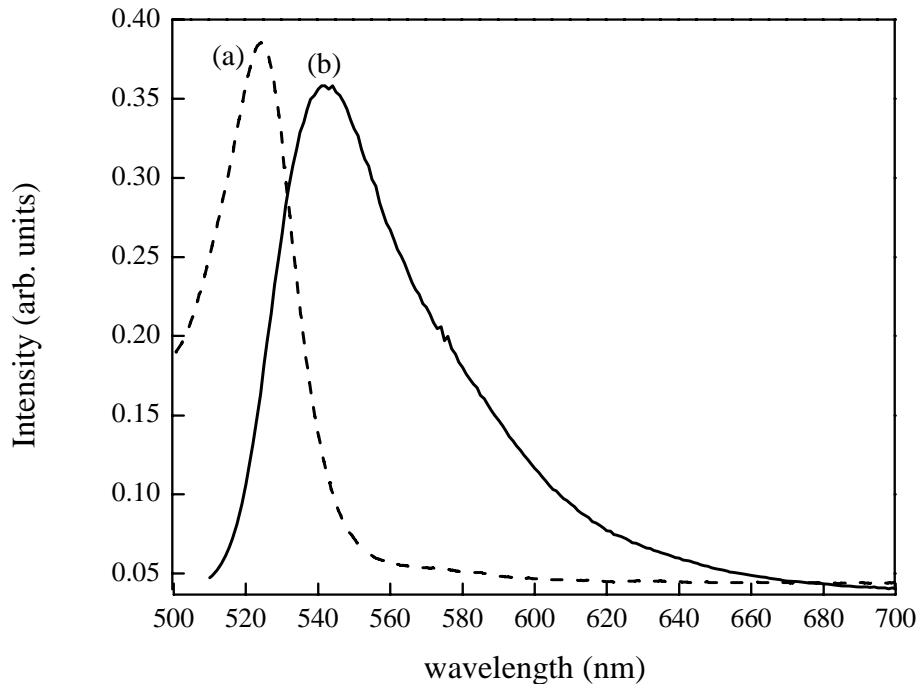


Fig. 2. Linear absorption spectrum (a) and fluorescence emission spectrum (b) of dye doped polymer H-PDLC film. The emitted lasing wavelength is expected to be between 540 nm to 600 nm.

The absorption and fluorescence spectra of the dye doped grating film are shown in Fig. 2. The linear absorption spectrum (curve a) is measured by using a scanning spectrophotometer (UV-3101PC from Shimadzu) with a 3-nm spectral resolution. The fluorescence spectrum (curve b) is measured by a CCD-camera connected to a grating spectrometer (HoloSpec from Kaiser Optical System) with a spectral resolution ~ 1 nm and with the sample excited by an unfocused 532-nm laser beam. The PM 580 doped polymer film exhibits a broad fluorescence emission band (from 520 to 610 nm). Moreover, in the spectral range of 520-550 nm there is obvious overlap between the long wavelength tail of the absorption band and the short wavelength tail of the emission band. Therefore, one should expect no lasing to occur in this spectral range due to the re-absorption of the emitted fluorescence signals. In contrast, a lasing wavelength should be expected above 560 nm, where the linear absorption is negligible and gain is reasonably high. Moreover, if the DFB (grating) structure selectively provides optical feedback (reflectivity) for a specific wavelength, a narrow band DFB lasing output should be expected even in the longer wavelength range (> 560 nm), where there is little linear absorption but still sufficient gain.

3. Results and discussion

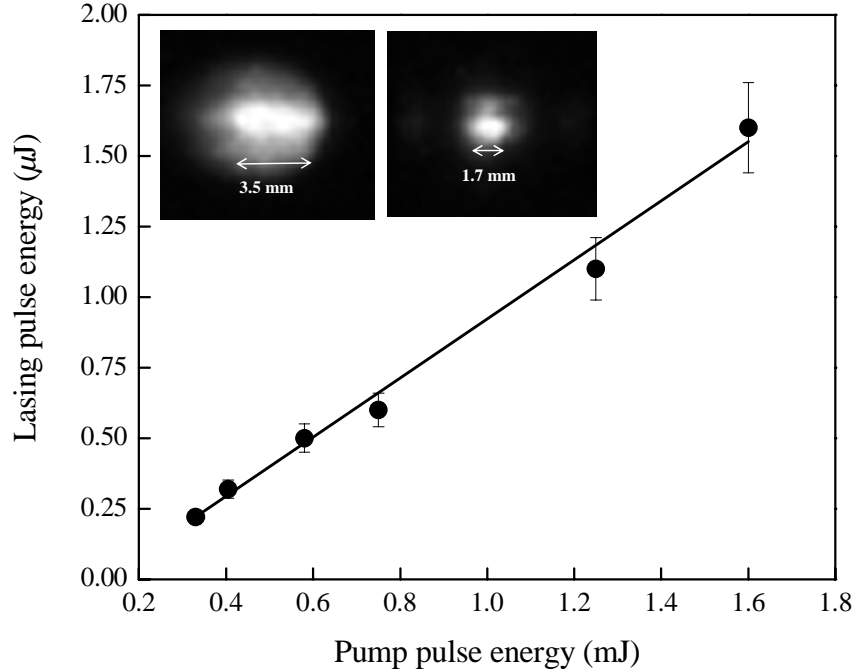


Fig. 3. Emitted 569 nm lasing energy as a function of the 532 nm pump energy, and images (inset) of near-field (left) and far-field (right) of emitted lasing

The measured output lasing pulse energy from the dye-doped H-PDLC cavity as a function of the pump pulse energy for energies above the laser threshold are shown in Fig. 3. It was observed that the pump energy had to be increased above the pump threshold value (≥ 0.3 mJ) for lasing to be observed, and the emitted laser power increased with increasing pump energy. At a pump energy of 1.6 mJ, the efficiency of conversion of pump input to the lasing output is $\sim 0.10\%$. The inset of Fig. 3 presents photographs of the near-field and far-field patterns for the output laser beam at 569 nm. To measure the near-field pattern, a ground glass screen was placed 3 cm from the sample edge. To measure the far-field pattern, the output lasing beam was re-collimated by an $f=7$ cm lens and then focused via an $f=30$ cm lens onto the same glass screen. In both cases, the beam image on the screen was recorded by a CCD camera. The spot size of the output lasing beam was measured to be ~ 3.5 mm and 1.7 mm in near- and far-field, respectively. The estimated divergence angle from the far-field pattern was ~ 5.6 mrad, consistent with unidirectional lasing from the DFB geometry of the dye-doped H-DPLC transmission grating.

Figure 4 shows the lasing spectra of three samples with different grating spacings measured by the spectrometer. The emitted lasing wavelengths are 561 nm, 569 nm and 592 nm, respectively, corresponding to three different grating spacings of the films. The full width at half maximum (FWHM) of the emitted lasing spectra is ~ 5 nm and that is ~ 10 times narrower than the spontaneous fluorescence. Furthermore, in Fig. 4 there is a relatively lower residual emission peak around 565 nm for the two structures with grating spacings of 368 nm and 382 nm. This secondary, weak, spectral peak is due to residual stimulated emission (amplified spontaneous emission, ASE) that does not experience sufficient gain suppression within the grating structure. This reveals that there is either (i) imperfections within the grating samples or (ii) non uniform (spatial) pumping. In fact, if we moved the sample position and let the focused pump illuminate the sample area where there is no grating

structure, directional ASE output along the pump line direction could be observed. The spectral measurement shows that the peak ASE position is located at 565 nm with a slightly broader (~10 nm) linewidth.

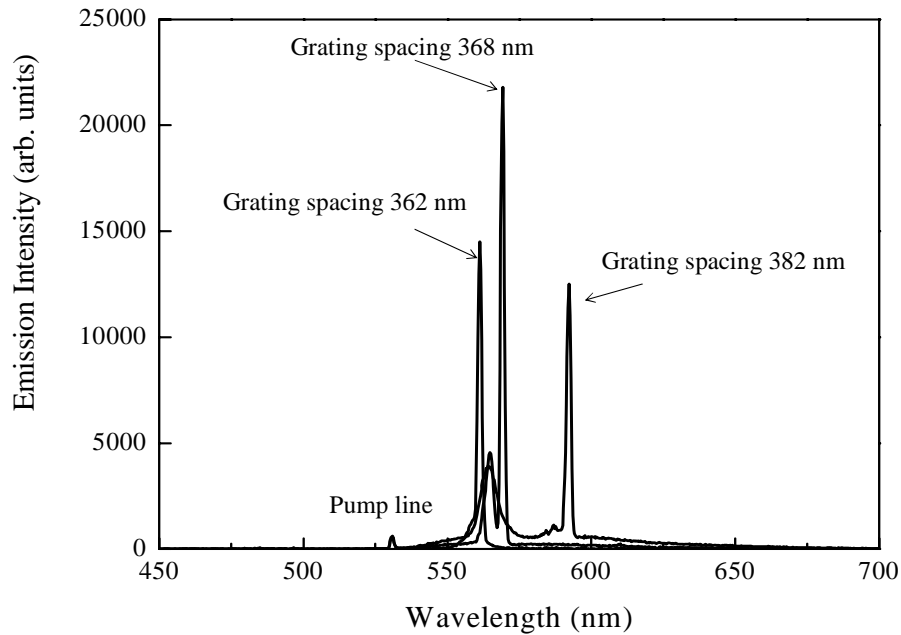


Fig. 4. The lasing spectra showing the narrowing effect of lasing behavior in the dye doped H-PDLC grating with three different grating spacings. The pump threshold energy is ~2.0 mJ.

As indicated by Kogelnik's coupled-wave theory of DFB lasers [14], the lasing wavelength should satisfy the equation

$$\lambda_{las} = \frac{2n_{eff}\Lambda}{m}, \quad (2)$$

where λ_{las} is the laser wavelength emitted from the grating, n_{eff} is the effective index of the grating at that wavelength, Λ is the grating spacing, and m is the diffraction order related to the grating. The three samples reported here have three different grating spacings that were calculated from the equation given by Ref.15 as 362 nm, 368 nm and 382 nm. Based on these grating spacing values and using Kogelnik's coupled-wave theory (m value = 2, second order diffraction, and $n_{eff}=1.52$ [16]), the calculated lasing wavelengths should be 550 nm, 559 nm and 580 nm. These calculated wavelengths are only 2% shorter than the experimental values of emitted lasing wavelengths of 561 nm, 569 nm and 592 nm, shown in Fig. 4. The shrinkage of the film during the polymerization process could be the explanation for this small error.

The temporal behavior of the output lasing pulses from the transmission grating samples was measured using a high speed digital oscilloscope (Infinium 500 MHz from HP). The recorded waveforms for the input pump pulses, the fluorescence emission pulses at low pump level, and the DFB lasing pulses are shown in Fig. 5 respectively. Each curve is a result of averaging over 20 pulses. The temporal resolution of the system (photodiode detector + oscilloscope) is about 1 ns for a single pulse signal. From Fig. 4 the pulse shape of the DFB lasing signal is obviously different from that of the spontaneous fluorescence signal; there is a

much sharper rising edge and shorter decay tail for the lasing pulse due to the threshold requirement of the lasing process. In contrast, the spontaneous emission (fluorescence) signal exhibits a slower rise and much longer decay, consistent with a fluorescence decay time in the nanosecond range.

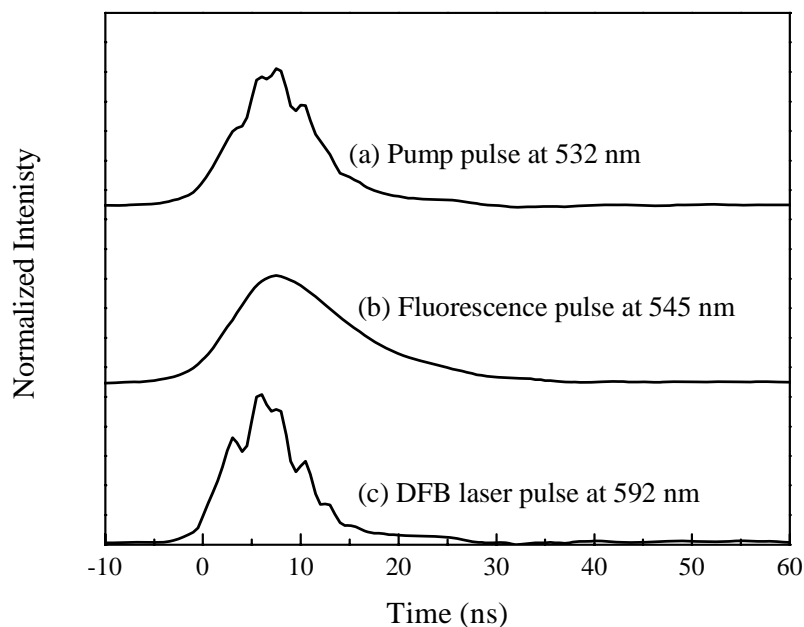


Fig. 5. (a) Temporal profile of the pump laser pulse, (b) temporal profile of fluorescence emitted from the grating sample, and (c) temporal profile of DFB cavity laser pulse from the same sample.

The most attractive reason to utilize the H-PDLC elements for photonics applications is their tunability and switchability. To demonstrate the switching behavior, the preliminary experiments by applying a dc electric field to an H-PDLC transmission sample sandwiched between two ITO-layers are presented. The switching ability of the PM580-doped H-PDLC film varies from 35% to 5% diffraction efficiency (DE) as shown in the Fig. 6 (inset). The switching mechanism of the H-PDLC grating is due to the index modulation of the phase-separated LC droplets. The axis of LC droplets with extraordinary refractive index ($n_e \sim 1.7$) follows the direction of applied field and the ordinary refractive index ($n_o \sim 1.51$) is matched closely with the polymer matrix. At this time the index modulation between LC lamella and polymer matrix is removed and light passing through the film is not affected [16]. The switchable behavior of lasing action is also shown in the same Fig. that is for the same grating sample of 382 nm spacing and 10 μm thickness. When the electric field is “off”, the grating sample produces a unidirectional laser beam from the edge of sample at a wavelength of 592 nm. After applying the electric field, the refractive index modulation of the sample is reduced and the cavity feedback is reduced. In this case, no lasing is observed at 592 nm when the electric field is increased to 30 $\text{V}/\mu\text{m}$. It should be noted that 30% LC concentration is typically used for the switchable H-PDLC transmission gratings to achieve a high DE¹⁶, however, 20% LC is used here. This concentration is found to be the best trade off between sufficient refractive index modulation and optical scattering due to the long period of the DFB geometry. High LC concentration in the H-PDLC structure results in high light scattering. The scattering increases the loss mechanism in the DFB cavity structure and the lasing threshold is substantially increased. Suitable index modulation is obtained in the grating film of 20% LC

concentration¹⁶ and according to equation 1, a narrow bandwidth of the emitting lasing is still achievable if the cavity length is sufficiently long. It should be pointed out that there is still an emission peak around 565 nm remaining in Fig. 6 even in the switched “on” (voltage applied) state. This peak is due to the residual ASE contribution as explained earlier.

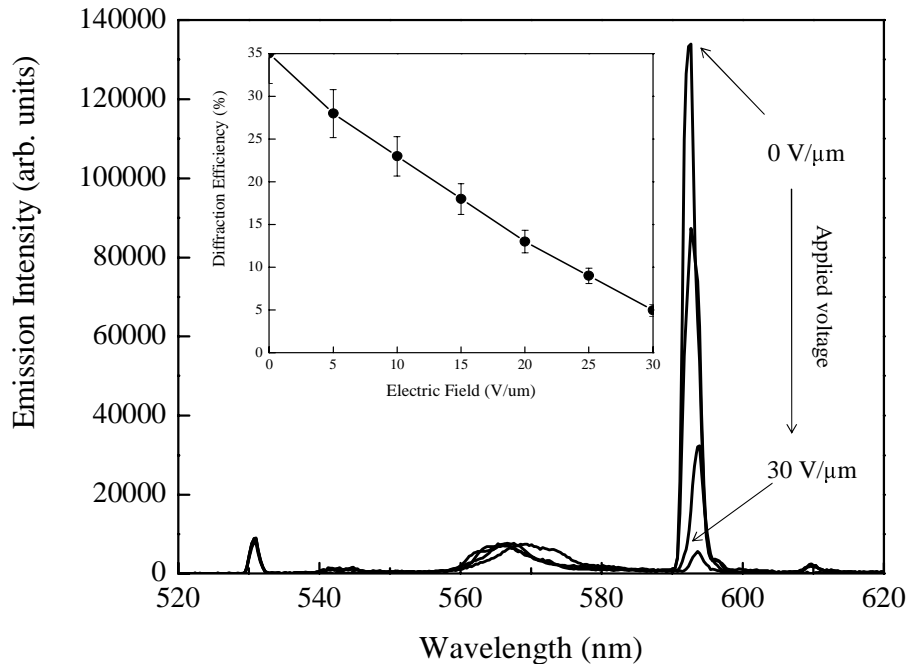


Fig. 6. The diffraction efficiency dependence of the PM580-doped H-PDLC grating with different applied electric field (inset), and the switching behavior of lasing in the same film.

4. Conclusions

Highly directional lasing behavior in a distributed feedback H-PDLC transmission grating structure has been studied in a transverse pump configuration. The advantage of this device geometry is that a long gain region and low switching voltage can be simultaneously achieved. In this specific case, the emitted laser spot size of 5 mm in diameter can have a narrowed spectral bandwidth of 5 nm and the emitted wavelength is selected by the angle of interference writing pattern. In addition, the emitted lasing can be switched on and off by applying a field of 30 V/μm sufficient to erase the index mismatch between the polymer and the LC regions. Thus, a robust switchable optically pumped polymer DFB cavity lasing structure can be easily achieved using the one-step fabrication process of a H-PDLC transmission grating.

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