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Second-order distributed feedback polymer laser based on holographic polymer dispersed liquid crystal grating

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ABSTRACT

We report on the fabrication and characterization of a second-order distributed feedback (DFB) polymer laser based on a holographic polymer dispersed liquid crystal (HPDLC) transmission grating. The fine organic grating is fabricated on top of the homogeneous conjugated polymer layer in a one-step process. The device shows surface-emitting, single mode laser emission with a threshold of 13.3 μJ/cm^2, and the working characteristics merely degrade after 10 months of storage in ambient atmosphere. We further explain the dependence of threshold on pumping length, and demonstrate the small refractive index modulation of this all-organic grating is sufficient to maintain efficient DFB laser action. This simple working structure, combined with large processing area provided by the holographic polymerization technique, is extremely promising in realizing ultra-low cost plastic lasers.

1. Introduction

Organic semiconductor lasers (OSLs) have attracted a broad scientific interest since the first demonstration of stimulated emission in semiconducting polymer in 1992 [1]. Thanks to the salient features of organic materials, OSLs are cost effective, tunable across the whole visible spectrum and mechanically flexible [2,3]. Due to some additional loss mechanisms under electrical injection [4,5], there has been no demonstration of organic laser diodes. Fortunately, indirect electrical pumping with cheap inorganic diode lasers [6] or light emitting diodes [7] is possible as OSLs exhibit very low operational thresholds. Distributed feedback (DFB) resonator configuration has attracted the most attention in OSLs [3], because it exhibits several advantages, such as compatibility with processing technique of organic thin film, no need of cleaved edge for output, excellent mode selection and low working threshold. In publications about organic DFB lasers, two kinds of configurations are commonly used: the active medium is over-coated onto the corrugated substrate [8] or the grating structure is directly written into the active layer [9]. In either case, the thickness of active layer is strongly modulated, and the grating depth in the active layer has been demonstrated as a complex parameter to be optimized [10]: (1) The DFB coupling mechanism is complex and uncertain, as the corrugations in the active layer provide both gain and index modulation. In some cases, the feedback is attributed to gain coupling because the lasing peak locates at the dip of band gap [8]. While in some cases, the feedback is attributed to index coupling as the lasing peak locates at the long wavelength edge of the band gap [11]. This uncertainty brought by grating depth in the active layer rends laser threshold reduction through optimizing coupling coefficient difficult. (2) The effective thickness and refractive index of the waveguide core layer which are essential to estimate the location of lasing wavelength and number of lasing modes are uncertain. They are inevitably modulated by the amount of other

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materials filling in the corrugations, which in turn are affected by the grating depth in the active medium. The significance of this effect varies from device to device, and is extremely pronounced when the grating depth is comparable to the active medium thickness.

In addition to the working structure, there is still potential in developing new techniques for fabricating the periodic microstructures used in organic lasers. Electron beam lithography in combination with dry etching techniques [12,13] give very sharp edged gratings with narrow periods, but it requires sophisticated experimental equipment and a sequence of fabricating steps. These techniques are not compatible with the initial motivation of easy processing of organic materials. As for the various methods in nanoimprint lithography, hot embossing will bring deleterious effect to fluorescence property of the gain medium [14,15], and liquid imprinting [9] or solvent-assisted [16] techniques cannot give high quality gratings. The UV nanoimprint lithography [17] may be the most promising, but it does not provide an easy way to alter the grating period. Additionally, the laser ablation technique [18,19] requires a careful selection of the gain medium.

In our pervious publication [20], we report on the first fabrication of third-order DFB polymer lasers based on holographic polymer dispersed liquid crystal gratings. The organic grating is fabricated separately on top of the homogeneous conjugated polymer layer. This technique is a variation of the holographic polymerization, and has the advantages of large processing area, low cost, easiness to vary the grating period. It also does not bring any deleterious effect to the gain medium and can be applied to any kind of active medium, as the fabrication requires no harsh conditions. Moreover, the working characteristics are good, with a threshold of 21 μJ/cm^2 and a bandwidth of 0.7 nm. In this paper, we report on the fabrication and characterization of a second-order DFB polymer laser using a small period HPDLC grating. In addition to all the advantages of the third-order one, this polymer laser shows surface-emitting laser emission with a threshold of 13.3 μJ/cm^2. We further give explanations on the threshold dependence on pumping length, and find the comparatively low refractive index modulation afforded by this organic grating is sufficient to maintain efficient optical feedback. Besides, the laser performances of these devices merely degrade after 10 months of storage in ambient atmosphere, indicating the stability of the organic grating in providing feedback. Thus, we think this "organic" approach is extremely compatible with organic semiconductors, and can provide ultra-low cost plastic lasers.

2. Device fabrication

Conjugated polymer Poly(2-methoxy-5-(2′-ethyl-hexyloxy)–p-phenyl-enevinylene) (MEH–PPV) (Average molecular weight ~120,000; photoluminescence quantum yield ~10%) was purchased from Jilin OLED Material Corporation and used as received. The solution of MEH-PPV in xylene (6 mg/ml) was deposited onto a glass substrate by spin-coating (1700 rpm), resulting in a homogeneous thin film of MEH–PPV with a thickness of ~80 nm measured by a Dektak Profilometer. The thickness here is chosen as the optimized value, which enables sufficient evanescent wave spread into the grating layer for feedback [20]. Then an empty cell was made with another uncoated glass substrate, where the cell gap was controlled as ~6 μm by Mylar spacers. After that, the prepolymer syrup containing monomers, liquid crystal (LC) and photo-initiator was injected into the cell via capillary effect in the dark. For a direct comprehension of above processes or detailed chemical composition of the mixture, readers may refer to [20].

The writing setup for HPDLC grating is shown schematically in Fig. 1. A frequency-doubled, continuous wave Nd-YAG laser at λ_w of 532 nm is used as the writing light source. After filtered and collimated by the beam expander, the laser beam is divided into two beams with equal intensity. The two beams are then directed by two mirrors and recombine at the cell surface with an intersecting angle of θ. The grating period Λ can be calculated and precisely controlled according to $\Lambda = \frac{\lambda_w}{2\sin\theta/2}$. The DFB lasing wavelength λ_{las} should obey the Bragg equation $\lambda_{las} = 2n_{eff}\Lambda/m$, where n_{eff} is the effective refractive index of the laser mode and m is the Bragg order. n_{eff} mainly depends on the thickness of MEH–PPV layer and refractive indices of three different layers, which is calculated as ~1.60 previously by us [20]. Considering the gain spectrum of MEH–PPV is around 630 nm [11], the grating periodicity for the third-order laser and second-order one should be selected as 596 nm (corresponding to an intersection angle of 53°) and 394 nm (corresponding to an intersecting angle of 85°), respectively. During the holographic exposure process, monomers would polymerize preferentially in bright fringes, causing a chemical potential gradient both in monomer and LC across the fringe patterns. Then monomers would diffuse to bright regions to continue photo-polymerization, and the unreacted LC would diffuse to dark regions. At some point, the chemical miscibility between monomer and LC breaks, leading to the phase separation of LC. For efficient LC phase separation, the choice of curing intensity is very important. The optimal curing condition for the narrower grating is different due to two reasons: first, an increase in the intersecting angle would bring an increase in the effective exposure area in the sample, resulting in decreased exposure

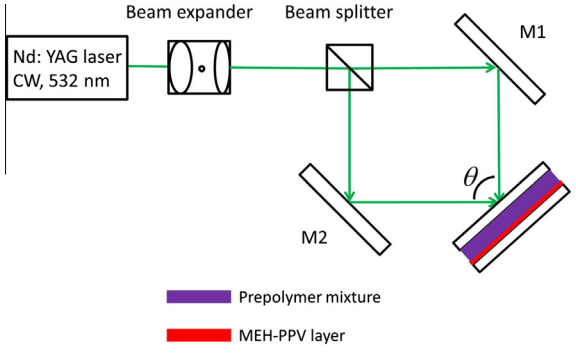

Fig. 1. Schematic experimental setup for the fabrication of HPDLC gratings.