Chapter 7

NANOIMPRINTED CIRCULAR GRATING DISTRIBUTED FEEDBACK DYE LASER

7.1 Introduction

In recent years, polymer dye lasers have attracted much attention due to their low-cost processing, wide choice of emission wavelengths, and easy fabrication on flexible substrates. Several waveguide dye lasers have been studied with emission wavelengths ranging from the ultraviolet to the near-infrared [1, 2]. By simply changing the fluorophore doped in the polymer these lasers can be used as the tunable sources for various applications, including spectroscopy [3]. The one dimentional (1-D) distributed feedback (DFB) resonator structure is widely employed and has been previously demonstrated for polymer lasers [4]. Operating characteristics can be significantly improved within two-dimensional structures. Here, a circular grating distributed feedback structure was used to obtain low threshold operation, a well-defined output beam, and vertical emission perpendicular to the device plane. Although surface emitting circular grating lasers using semiconducting polymers have been previously demonstrated by Bauer et al. [5] and Turnbull et al. [6, 7], their lasers were fabricated by depositing the organic gain material onto pre-patterned dielectric substrates, limiting the depth and the accuracy of the shape of the grating. To enable the mass production, the nanoimprint lithography [8] was chosen as a direct patterning method. A hard mold is used to transfer patterns with high fidelity into target polymers, and this technique has become an attractive approach to define nanofabricated optical resonator structures. Conjugated polymer lasers fabricated by hot embossing have been studied by Lawrence et al. [9], and 1-D DFB lasers based on organic oligomers using a room-temperature nanoimprint method were reported by Pisignano et al. [10].

In this chapter, we report a circular grating distributed feedback laser fabricated within a dye-doped poly(methylmethacrylate) (PMMA) film on a glass substrate. The laser circular grating structure was fabricated using a low-cost and manufacturable nanoimprint method. Lasing with single mode surface emission at 618.52 nm and a linewidth of 0.18 nm was measured from the polymer dye laser exhibiting a threshold value of 1.31 nJ/mm^2. The laser operation characteristics of the circular grating resonator are improved through the high accuracy and aspect ratio nanoimprint pattern transfer. Moreover, the mold can be re-used repeatedly, providing a convenient mean of mass production and large scale fabrication of low cost polymer dye laser arrays.

7.2 Chip design and fabrication

The circular grating structure proposed by Erdogan and Hall [11] provides a natural two-dimensional extension of the basic DFB structure. It allows feedback to be applied in all in-plane directions, and the second-order grating couples the emitted radiation perpendicularly out of the surface of the sample. Figure 7.1 shows a general design of a circular grating distributed feedback structure. A theoretical analysis of circular grating is described in detail elsewhere [5, 12-14] predicting that only the radial propagating components define the modes in the circularly symmetric grating. The design parameters of the circular gratings fabricated for this chapter are selected based on electromagnetic mode calculations and experimental results. A grating period of 440 nm is chosen to match the second order Bragg condition. The center defect is a 440 nm diameter gain region. The 400 nm groove

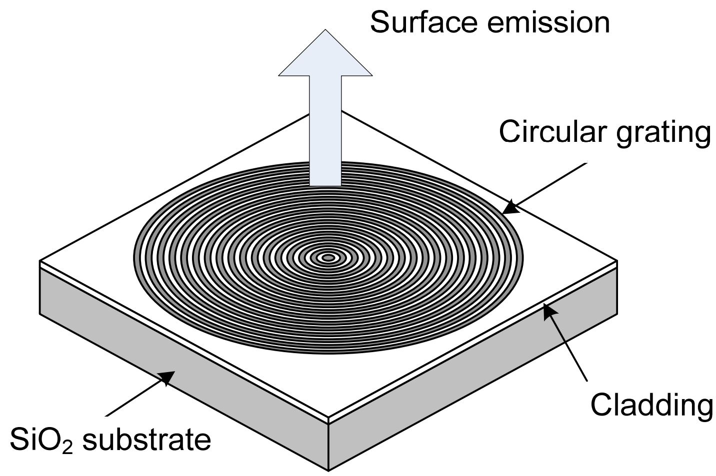

Figure 7.1. General design of a circular grating distributed feedback structure.

depth is defined to ensure maximum confinement, whereas the 200 μm overall diameter of the circular grating and the 50% duty cycle are used to reach the maximum coupling strength [6]. In our experiments, silicon dioxide (SiO_2) was used as the mold material and the grating pattern was defined by electron beam lithography on a LEICA EBPG 5000+ electron beam writer with proximity correction. The pattern was subsequently transferred into a SiO_2 substrate via reactive ion etching using CHF_3 plasma. PMMA was selected as the polymer matrix because of the solubility of the dye in PMMA, as well as its low optical absorption within the wavelength range for activating the dye molecules, and its excellent properties for nanoimprint lithography [15].

To construct the dye laser, a glass substrate was spin-coated with 5 μm of Cytop, a low refractive index material (n=1.34, Asahi Glass) as the lower cladding to ensure the vertical optical confinement. Then, 500 nm Dye(Rhodamine 640, Exciton)-doped PMMA (30 mM) with the refractive index about 1.48 at emission wavelength was spin-coated on top of the Cytop layer as the gain medium. An oxygen plasma treatment (Anatech SP100) of the Cytop was necessary for good adhesion of Cytop to the PMMA.

Nanoimprint lithography exploits the glass transition of polymers to achieve high-fidelity pattern transfer. In order to reduce the degradation of photoluminesence efficiency of the organic materials at high temperatures during the nanoimprint lithography, a vacuum environment nanoimprint was demonstrated to have less than 5% reduction of degradation [16]. In our experiment, a simple alternative was used to reduce the degradation by sealing the imprinting mold and the PMMA substrate into an elastic PDMS mold during the imprinting process. This elastic mold serves as pressure balancing to reduce the inhomogeneous pressure due to the non-parallel presser heads. A mold release agent (surfactant) such as 1H,1H,2H,2H-perfluorodecyl-trichlorosilane (Alfa Aesar) was also deposited on the PMMA to reduce the resist adhesion to the mold. Then, the mold was pressed into the PMMA film by using an automatic mounting press machine (Buehler SimpliMet 1000) at a temperature of 150°C (above PMMA's glass transition temperature) and a pressure of 1200 psi. After sample cooling, the mold could be easily separated from the patterned polymer laser chip. The fabrication process is schematized in Figure 7.2.

Figure 7.3 shows SEM images of the mold and the imprinted PMMA. From these pictures, we can observe that the structure on the SiO_2 mold is faithfully replicated on the PMMA substrate surface with high resolution. The resolution of the nanoimprint could be as high as sub 10 nm. Photoluminescence spectra confirm that there is no degradation of the luminescence performance of the polymer.

7.3 Measurement and laser characteristics

The polymer laser cavity was optically pumped with 6 nanosecond Q-switched Nd:YAG

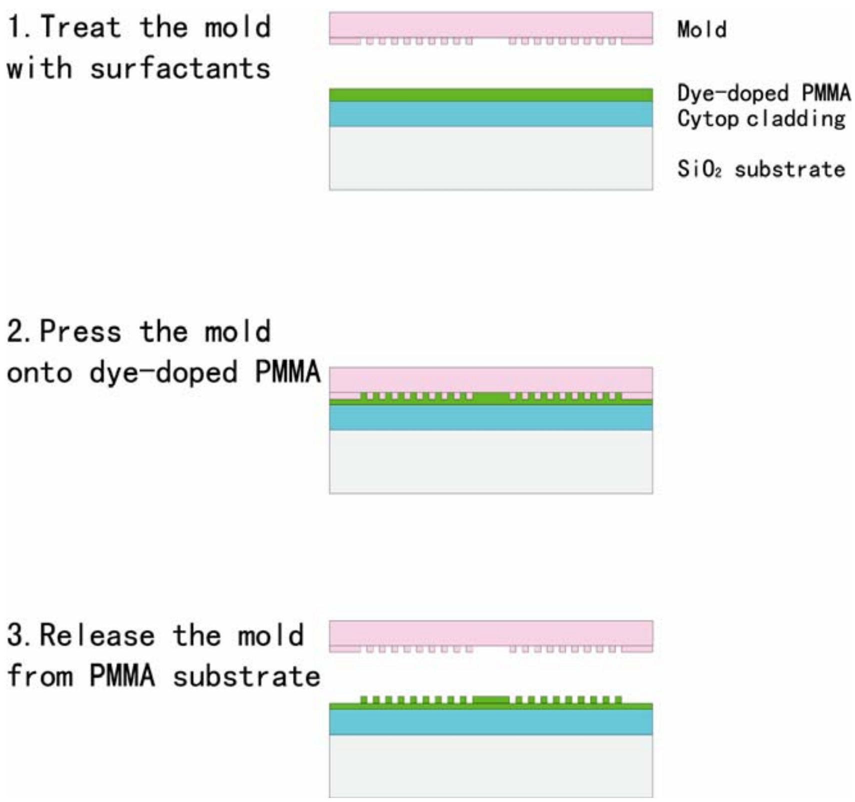

Figure 7.2. Schematic fabrication process of circular grating polymer dye laser

laser pulses at 532 nm wavelength, focused through a 20X objective to the back side of the chip. A 10X microscope objective was used to collect the emission from the top surface of the chip and deliver it to a fiber coupled CCD-array based spectrometer with 0.1 nm resolution (Ocean Optics HR4000). A typical single mode lasing spectrum is shown in

Figure 7.4. The lasing wavelength is 618.52 nm, and the measured linewidth is 0.18 nm.

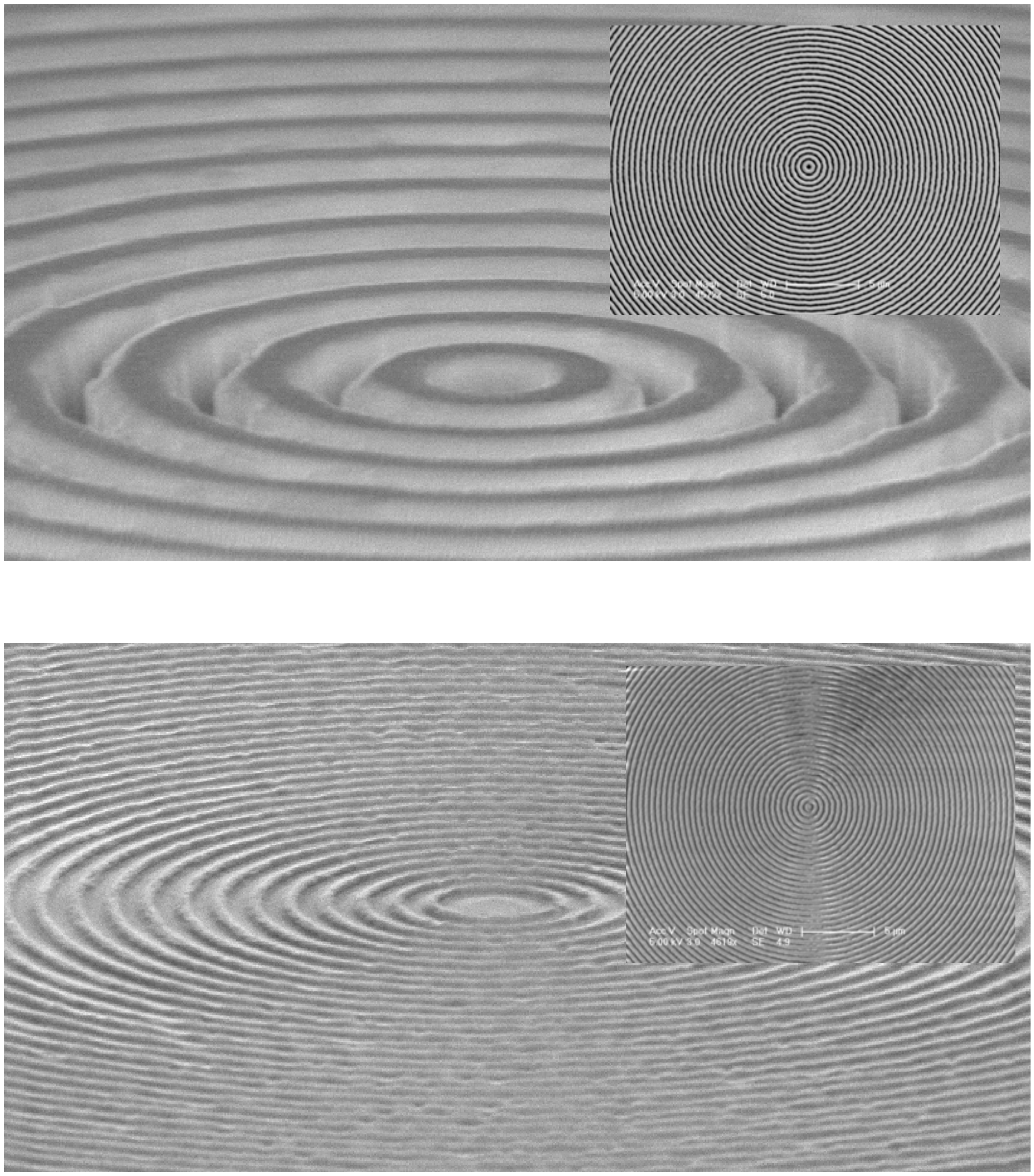

Figure 7.3 SEM images of angled view of (a) the SiO_2 mold and (b) the imprinted PMMA film with circular grating structure with top view of insets.

Lasing occurs near the Bragg resonance, determined by the equation $m\lambda_{Bragg} = 2n_{eff}\Lambda$, where m=2 is the order of diffraction, n_{eff} is the effective refractive index of the propagation mode, and Λ is the grating period. The lasers exhibit single mode emission, as higher order modes do not appear, due to the smaller overlap between the mode profile and the gain region. The linewidth near threshold is measured as 0.20 nm, which results in a cavity quality factor (Q) of over 3000. The left inset of Figure 7.4 shows the variation of the output laser power as a function of the absorbed pump pulse fluence, and a laser threshold pump fluence of 1.31 nJ/mm^2 is observed. The absorbed pump pulse fluence was calculated

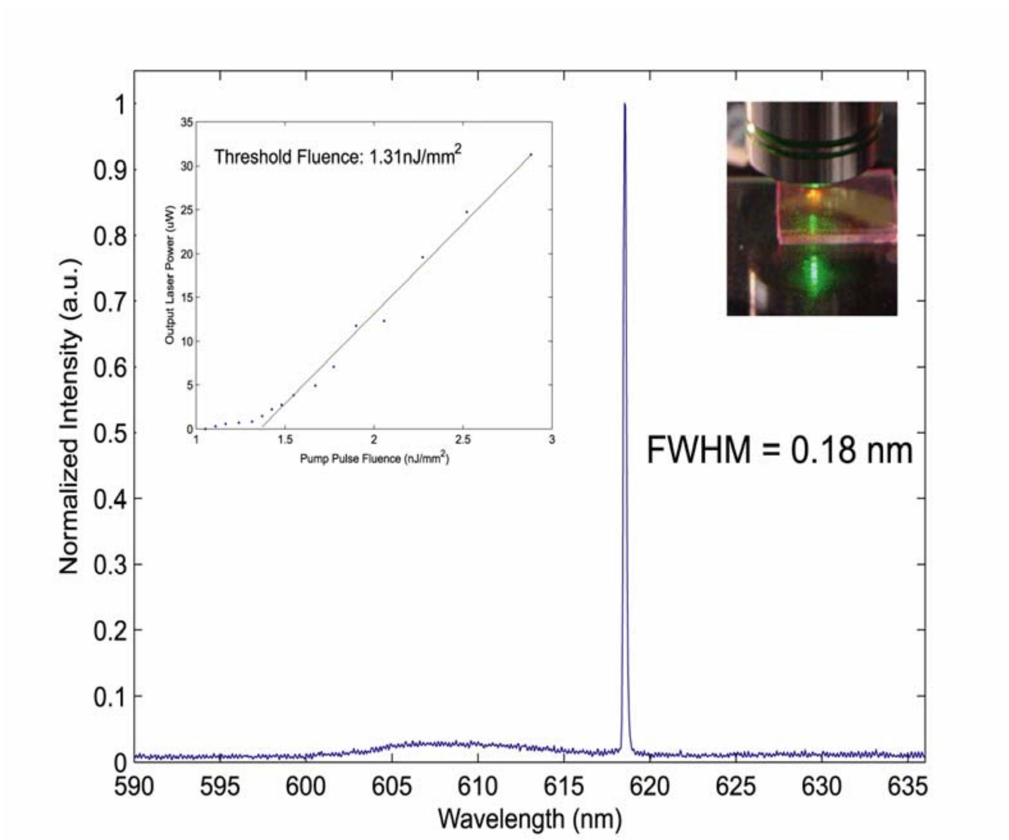

Figure 7.4. The typical nanoimprinted circular grating DFB dye laser spectrum. The measured linewidth is 0.18nm. Left inset: The pump laser pulse fluence vs. the absorbed pump pulse fluence. The threshold fluence is determined as 1.31 nJ/mm^2. Right inset: Polymer laser chip excited by Nd:YAG 532 nm laser pulses

with the pump pulse energy measured divided by the focal point which is about 600 μm in diameter and the output laser power was measured directly using the power meter. This pump intensity is well within the reach of commercial high power blue laser diodes (LDs), enabling a self-contained LD pumped device. The right inset of Figure 7.4 shows that the polymer laser is pumped from the back side and the lasing emission is collected from the surface of the chip. The transparency of the substrate, the size and geometry of the laser cavity and the low threshold also match well with the output beams of high power LEDs and LDs. Therefore the replication-molded ring geometry represents a very promising structure for the construction of compact LED or LD pumped portable dye lasers.

We observe decreases in the laser emission with increasing exposure time. This result is consistent with previous studies on polymer DFB structures [17]. The lifetime of polymer dye laser can last over 10^6 shots (of pump laser pulses), and if the characterization of the device is carried out under vacuum to inhibit photo-oxidation, the lifetime can be further extended [18]. Because of the low cost of materials and fabrication, replication molded devices are disposable and may not require a long lifetime. In the future, we plan to make an optofluidic version [4] of the circular grating dye laser which allows us to constantly change the dye to increase the device lifetime and to tune the wavelength.

7.4 Conclusion

In summary, we demonstrated a surface emitting polymer dye laser with a circular grating distributed feedback structure realized by nanoimprint lithography. We achieved excitation thresholds as low as 1.31 nJ/mm^2 and FWHM linewidths of 0.18 nm. The technique described here enables the fabrication of low-cost, high-quality and mass-producible laser arrays, which may be deployed as compact and inexpensive coherent light sources for lab-on-a-chip applications such as sensing and spectroscopy. The two weakness of this structure are: low vertical optical confinement (low refractive contrast) so the threshold would be reduced mainly through the increasing of the gain of the gain medium; degradation of the dye emission efficiency during the operation. The first weakness could be improved by using high index contrast design, and the second could be avoided by the dye circulation. The ultimate goal is to reduce the lasing threshold to enable the use of LEDs or LDs pumping as integrated and inexpensive pump sources for on-chip polymer lasers with wavelength tunability through different mechanisms which include mechanical tuning, temperature fine tune, and tuning by circulating different dyes.

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