Nanoimprinted circular grating distributed feedback dye laser

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(Received 23 April 2007; accepted 21 June 2007; published online 31 July 2007)

The authors demonstrate an optically pumped surface emitting polymer dye laser fabricated by nanoimprint lithography. The laser is based on an organic dye hosted within a poly(methylmethacrylate) matrix coated on a transparent substrate, and the laser cavity consists of a second order circular grating distributed feedback structure. The authors achieved lasing at 618 nm with 0.18 nm linewidth and 1.31 $\mu J/mm^2$ pump threshold. The nanoimprinted solid-state dye laser offers a low-cost coherent light source for laboratory-on-chip spectroscopy systems. The laser also has a low pump threshold and a geometry well matched to light-emitting diode pump sources, which provide an interesting alternative for constructing portable polymer laser devices. © *2007 American Institute of Physics*. [DOI: 10.1063/1.2757600]

Within recent years, the development of polymer dye lasers has progressed to higher levels of performance and functionality. The most attractive advantages of polymer dye lasers include 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 ultraviolet to near infrared.[1] By simply changing the fluorophore doped in the polymer, these lasers can be used as the tunable sources for various applications including spectroscopy.[2] The one-dimensional (1D) distributed feedback (DFB) structure is a widely employed resonator geometry, and has been previously demonstrated for polymer lasers.[3] Operating characteristics can be significantly improved within two-dimensional structures. Here, we choose a circular grating distributed feedback structure 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.*[4] and Turnbull *et al.*,[5] their lasers were fabricated by depositing the organic gain material onto a prepatterned dielectric substrates, limiting the depth and the accuracy of the shape of the grating. For better geometric control, we choose nanoimprint lithography[6] 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.*,[7] and 1D DFB lasers based on organic oligomers using a room temperature nanoimprint method were reported by Pisignano *et al.*[8,9]

In this letter, we report a circular grating distributed feedback laser fabricated on dye-doped poly(methylmethacrylate) (PMMA) films. The laser was fabricated on a glass substrate using a low-cost and manufacturable nanoimprint method. Surface emission lasing with single frequency at 618 nm and a linewidth of 0.18 nm was measured from the polymer dye laser exhibiting a threshold value of 1.31 $\mu J/mm^2$. The laser operation characteristics of circular grating resonator are improved through the high accuracy and aspect ratio nanoimprint pattern transfer. Moreover, the mold can be reused repeatedly, providing a convenient way of mass production and large scale fabrication of low-cost polymer dye laser arrays.

The circular grating structure proposed[10] and demonstrated[11] by Erdogan and Hall 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 1 shows a general design of a circular grating distributed feedback structure. A theoretical analysis of circular grating lasers is described in detail elsewhere[12–16] 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 letter are selected based on the 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 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.[17] In our experiments, silicon dioxide (SiO_2) was used as the mold ma-

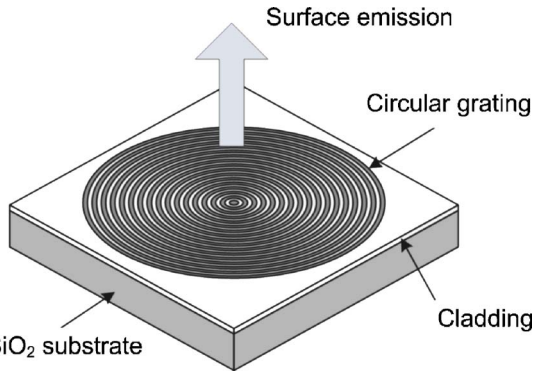

FIG. 1. General design of a circular grating distributed feedback structure.

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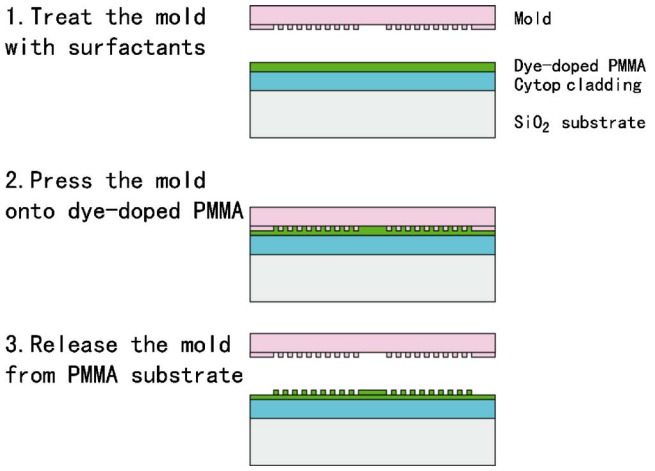

FIG. 2. (Color online) Schematic fabrication process of circular grating polymer dye laser.

terial 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.[18]

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, $n=1.49$) 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. However, degradation of light emission efficiency of the organic materials during air exposure at high temperatures presents a challenge in nanoimprint lithography.[19] To solve this problem, a modified nanoimprint method is used to prevent this degradation of the dye-doped PMMA film by sealing the mold and the PMMA substrate into a curable polymer during the imprinting process. A mold release reagent such as $1H,1H,2H,2H$-perfluorodecyl-trichlorosilane (Alfa Aesar) was also deposited on the dye from the vapor phase 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 Fig. 2.

Figure 3 shows scanning electron microscopy (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. Photoluminescence spectra confirm that there is no degradation of the luminescence performance of the polymer.

The polymer laser chip was optically pumped with 6 ns Q-switched Nd:YAG (yttrium aluminum garnet) laser pulses

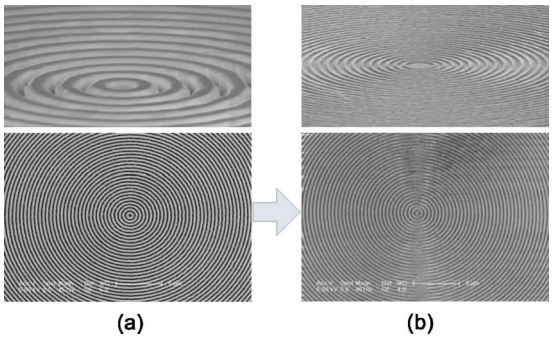

FIG. 3. SEM images of (a) the SiO_2 mold and (b) the imprinted PMMA film.

at 532 nm wavelength, focused through a 20× objective to the back side of the chip. A 10× microscope objective was used to collect the emission from the top surface of the chip and deliver it to a fiber coupled charge coupled device (CCD)-array based spectrometer with 0.1 nm resolution (Ocean Optics HR4000). A typical single frequency lasing spectrum is shown in Fig. 4. The lasing wavelength is 618.52 nm, and the measured linewidth is 0.18 nm. Lasing occurs near the Bragg resonance, determined by the equation $m\lambda_{\mathrm{Bragg}}=2n_{\mathrm{eff}}\Lambda$, 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 linewidth near threshold is measured as 0.20 nm, which results in a cavity quality factor (Q) of over 3000.

Figure 5(a) represents the far-field image of the emission pattern recorded by a CCD camera, and Fig. 5(b) shows the far-field radiation patterns of the laser passing through a linear polarizer with different orientation angles. The laser is expected to be azimuthally polarized,[20] as illustrated in the polarization patterns. The azimuthal polarization also results in a zero electrical field (a dark spot) at the center of the laser.[11] In the lasing process, many spatial modes can be excited with their mode thresholds very close to each other.[12] The fundamental mode is normally the favored one, because higher order modes do not overlap well with the gain region.

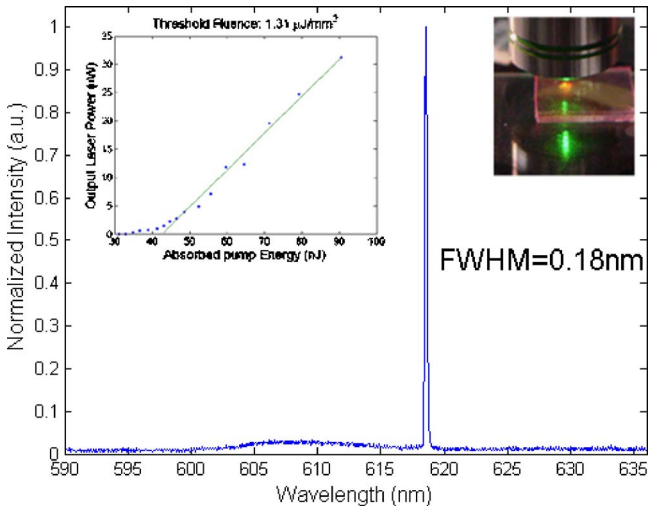

FIG. 4. (Color online) Nanoimprinted circular grating DFB dye laser spectrum. The measured linewidth is 0.18 nm. Left inset: the output laser power vs the absorbed pump energy curve. The threshold pump fluence is 1.31 μJ/mm^2. Right inset: polymer laser chip excited by Nd:YAG 532 nm laser pulse.

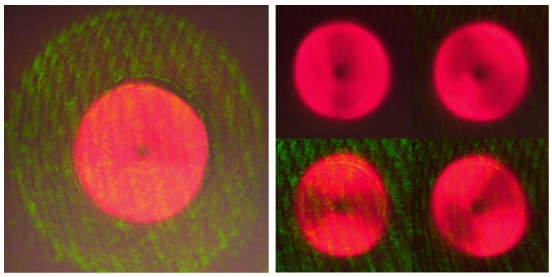

FIG. 5. (Color online) (a) Far-field image of the emission pattern recorded by a CCD camera. (b) Circular grating DFB laser far-field radiation patterns through a linear polarizer with different orientation angles. The laser emits an azimuthally polarized, well-confined circular beam.

The left inset of Fig. 4 shows the variation of the output laser power as a function of absorbed pump energy. With the absorbed threshold energy of 41.3 nJ, the threshold pump fluence is estimated to be 1.31 $\mu J/mm^2$. 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 Fig. 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 match well with the output beams of high power light-emitting diode (LED) and LD. 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.[21] The lifetime of polymer dye laser can last over 10^6 shots of pump laser pulse, and if the characterization of the device is carried out under vacuum to inhibit photooxidation, the lifetime can be further extended.[22] 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[23,24] of the circular grating dye laser which allow us to constantly change the dye to increase the device lifetime and to tune the wavelength.[25]

In summary, we demonstrate 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 $\mu J/mm^2$ and full width at half maximum (FWHM) 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 laboratory-on-a-chip applications such as sensing and spectroscopy. Future work will be focused on improving the laser cavity Q values with better electromagnetic design, optimizing the dye concentration, and fabricating smoother surfaces. The ultimate goal is to reduce the lasing threshold to enable the use of LEDs as integrated and inexpensive pump sources for on-chip polymer lasers.

This research was supported by the Caltech DARPA Center for Optofluidic Integration under Contract No. HR0011-04-1-0032 and by the Boeing Corporation.

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