Nanoimprinted polymer lasers with threshold below 100 W/cm^2 using mixed-order distributed feedback resonators

Yue Wang,[1] Georgios Tsiminis,[1] Alexander L. Kanibolotsky,[2] Peter J. Skabara,[2] Ifor D.W. Samuel[1,3] and Graham A. Turnbull[1,*]

[1] *Organic Semiconductor Centre, SUPA, School of Physics and Astronomy, University of St Andrews, North Haugh, St Andrews, KY16 9SS, UK*
[2] *WestCHEM, Department of Pure and Applied Chemistry, University of Strathclyde, Thomas Graham Building, Glasgow, G1 1XL, UK*
[3]*idws@st-andrews.ac.uk,*
[*] *gat@st-andrews.ac.uk*

Abstract: Organic semiconductor lasers were fabricated by UV-nanoimprint lithography with thresholds as low as 57 W/cm^2 under 4 ns pulsed operation. The nanoimprinted lasers employed mixed-order distributed feedback resonators, with second-order gratings surrounded by first-order gratings, combined with a light-emitting conjugated polymer. They were pumped by InGaN LEDs to produce green-emitting lasers, with thresholds of 208 W/cm^2 (102 nJ/pulse). These hybrid lasers incorporate a scalable UV-nanoimprint lithography process, compatible with high-performance LEDs, therefore we have demonstrated a coherent, compact, low-cost light source.

OCIS codes: (310.6628) Subwavelength structures, nanostructures; (140.3480) Lasers, diode-pumped; (140.3490) Lasers, distributed-feedback; (110.4235) Nanolithography; (160.4890) Organic materials.

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#187978 - $15.00 USD Received 29 Mar 2013; revised 30 May 2013; accepted 30 May 2013; published 10 Jun 2013

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1. Introduction

Organic semiconductor lasers are compact visible sources which combine solution processability and wavelength tunability [1]. Over the last decade, much of the work in this field has focused on improving gain media [2–4] and laser resonators [5–8] with the aim of reducing the laser thresholds to give an inexpensive and practical laser source. Low threshold optically-pumped organic lasers have been demonstrated with cheap inorganic diode lasers [9–12] and even with light emitting diodes (LEDs) [13, 14] as the pump source. Such indirect electrically-pumped organic lasers provide a convenient way to substantially reduce the complexity, size and expense of the laser system, and are therefore encouraging for the prospects of manufacturing compact and low-cost colour-tunable lasers.

Distributed feedback (DFB) periodic structures play a very important role in these high performance organic semiconductor lasers [1]. Simple techniques such as solvent-assisted micro-molding [15], two-beam holography [16] and nanoimprinted lithography (NIL) [17–20] are used to introduce periodic corrugations, on either the organic gain medium or substrate. Among these patterning methods, NIL provides parallel processing with a high throughput, leading to low cost fabrication of large-area nano-structures (with resolution down to a few tens of nanometres) on various types of substrates [21, 22].

Recently, we demonstrated that UV-NIL can be used to make polymer lasers with second-order DFB resonators and a threshold of 770 W/cm^2 [14]. In this paper, we use mixed-order DFB gratings [7], a composite structure with mixed orders allowing

#187978 - $15.00 USD Received 29 Mar 2013; revised 30 May 2013; accepted 30 May 2013; published 10 Jun 2013

extremely low laser threshold with surface emission, which are capable of being pumped by inorganic LEDs reliably and routinely.

2. DFB lasers on silica grating

In our work, we used the polymer poly[2,5-bis(2′,5′-bis(2″-ethylhexyloxy)phenyl)-*p*-phenylene vinylene] (BBEHP-PPV) to make DFB lasers. This polymer has been reported as a good laser gain material with a very low optical waveguide loss [23, 24], and was synthesised according to the method reported in reference [14]. The polymer has an absorption peak at 430 nm and emission maxima at 495 nm and 528 nm. We initially studied DFB laser operation on a fused silica substrate to characterise the tunability of the laser emission and wavelength for lowest threshold.

Second-order DFB lasers were made by spin-coating the BBEHP-PPV (from 15 mg/ml solution in chlorobenzene) onto a 1D holographic silica grating with a period of 355 ± 5 nm. The architecture of the laser is presented in Fig. 1. In a second-order DFB laser, the optical feedback wavelength is determined by the Bragg equation $\lambda_{Bragg} = 2n_{eff}\Lambda/m$, where m = 2, and second order diffraction provides in-plane feedback while first order diffraction gives rise to a surface-emitted output coupling. By changing the thickness of the gain material, the effective refractive index varies; therefore, the laser wavelength can be tuned. Lasers with light emitting layers of thickness from 145 nm to 500 nm were made. The BBEHP-PPV lasers were encapsulated by spin-coating a thin layer of CYTOP (Asahi Glass, Japan), a chemically robust and optically transparent amorphous fluoropolymer, directly onto the polymer waveguides. The polymer laser was excited by 450 nm pulses (FWHM 4 ns) at 20 Hz repetition rate from a Nd:YAG laser pumped optical parametric oscillator (OPO). Figure 1 shows the variation in laser threshold density with operation wavelength. The lowest laser threshold of 120 W/cm^2 (0.48 μJ/cm^2) was obtained with a 170 ± 10 nm thick BBEHP-PPV film (which supports TE$_0$ and TM$_0$ transverse modes), with the DFB resonator providing laser feedback for the TE$_0$ mode at the peak wavelength of the gain, i.e. 533 nm.

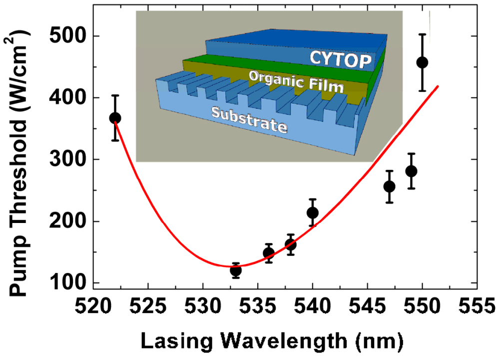

Fig. 1. Threshold of second order DFB lasers made with a holographic grating (period of 355 nm), as a function of output wavelength, the solid line is a guide for the eye. Insert: the CYTOP-encapsulated laser structure.

3. Mixed-order UV-NIL polymer lasers

The second-order holographic grating has a sinusoidal profile naturally resulting from a two-beam interference exposure of the photoresist layer followed by a chemical etching process used to transfer the pattern into the substrate. The efficient surface emission represents a loss mechanism in the second-order DFB lasers. To reduce the laser threshold, one standard route is to eliminate the surface output loss by using a first-order DFB resonator. However, the edge-emitting first-order resonator has its own limits, including highly divergent output beam and requiring high quality facets [23, 25, 26]. Instead, a mixed-order resonator, consisting of a short second-order region for surface output coupling between two first-order grating regions to provide in-plane feedback, takes advantage of both first and second order gratings [7, 8]. In such resonators, it is

#187978 - $15.00 USD Received 29 Mar 2013; revised 30 May 2013; accepted 30 May 2013; published 10 Jun 2013

possible to control vertical out-coupling of the laser and hence adjust the balance of feedback and output coupling.

Figure 2(a) shows a schematic drawing of a BBEHP-PPV laser, with four periods in the second-order region and multiple first-order gratings on each side. The gratings were initially fabricated in silicon by electron beam lithography followed by a reactive ion etch using fluorine based chemistry (Kelvin Nanotechnology Ltd). The gratings were 1 mm by 1 mm in dimension with most of this area filled with first order gratings and 4, 8, 15 and 30 periods of second-order gratings respectively in the central region. A conventional second-order DFB grating of the same area with a period of 350 nm was also fabricated on the same wafer as a reference grating.

UV-NIL was then used to replicate the master structures, as schematically shown in Fig. 2(b). This consists of three stages – first, an anti-sticking monolayer, perfluoro-octyl-trichlorosilane (Sigma Aldrich), was coated onto the master wafer to make it highly hydrophobic; second, a soft stamp was made on a glass substrate as a replica of the silicon master, and finally this was used to transfer the nanostructures into a UV-NIL resist mr-UVCur06 (Microresist Technologies). The soft stamp could be used for multiple imprinting. Prior to completing the laser fabrication by spin-coating the polymer gain medium, the final imprinted structures in mr-UVCur06 were characterized using scanning electron microscopy (SEM), see Fig. 2(c)-(e).

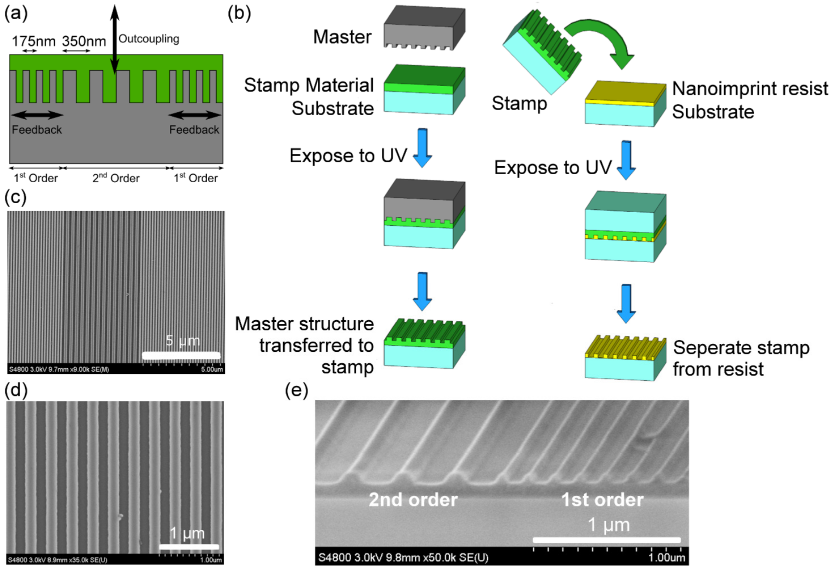

Fig. 2. (a) Schematic drawing of a mixed-order grating with four periods of second-order gratings in the outcoupling region; (b) Schematic fabrication steps for the DFB structure. A rigid nanostructure template in silicon, a 'master,' is pressed onto a perfluoro polyether polymer film to form a negative of the master, a 'stamp'. Finally, the stamp is used to print the nanostructures into a UV-NIL resist; (c) SEM image of the master structure with fifteen periods of second order gratings in the middle; (d) SEM image of the reference master grating; (e) A cleaved edge SEM image with 75° viewing angle of a UV-NIL mixed-order grating imprinted from the master in (c). The groove depths of the imprinted first and second-order gratings are 65 ± 5 nm and 85 ± 5 nm respectively.

The conventional second-order BBEHP-PPV laser made by UV-NIL had a threshold of 126 W/cm^2 (0.50 µJ/cm^2, 5 nJ/pulse), see Fig. 3. This is comparable to the optimized threshold on silica gratings and is a factor of 4 lower than the lowest reported threshold densities for nanoimprinted organic semiconductor lasers [14] [27]. An even greater reduction in laser threshold was achieved with the nanoimprinted mixed-order gratings. A UV-NIL laser with 30 periods of second-order grating had a threshold of 57 W/cm^2 (230 nJ/cm^2), see Fig. 3, which is to the best of our knowledge the first demonstration of a threshold under 100 W/cm^2 in an organic NIL laser. We observed similar thresholds for the mixed-order lasers with 4, 8 and 15 periods of second-order gratings in the range of

#187978 - $15.00 USD Received 29 Mar 2013; revised 30 May 2013; accepted 30 May 2013; published 10 Jun 2013

52 W/cm^2 to 65 W/cm^2. In principle, the fewer periods of second-order grating, the lower the lasing threshold due to the smaller out-coupling loss. The similar thresholds for these different laser cavities suggest that the surface output loss is small compared with other losses in the resonators.

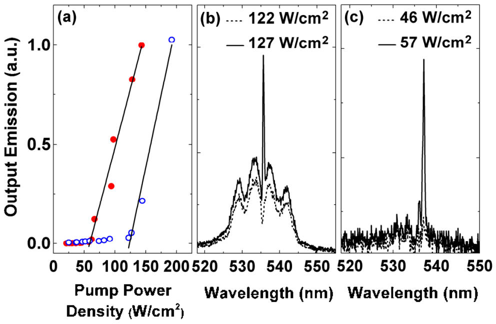

Fig. 3. (a) Output intensity as a function of pump power for NIL second-order DFB (open circles) and mixed-order DFB (closed circles); surface emission spectra below (dashed line) and above (solid line) the lasing threshold for (b) second-order and (c) mixed-order DFB. The lasing wavelengths are 536 and 537 nm respectively.

4. LED-pumped polymer lasers

UV-NIL lasers with such low thresholds have the potential to be optically pumped by commercial inorganic LEDs. Figure 4 presents a schematic drawing of an LED-pumped organic NIL laser geometry. To match the BBEHP-PPV thin-film absorption (peaked at 430 nm), we used an Indium Gallium Nitride (InGaN) LED (Philips LUMILED) with maximal emission at 448 nm. The light-emitting surface of the LED (1.3 mm by 1.3 mm) was butt-coupled to the CYTOP encapsulation layer to ensure the best possible power density at the polymer gain medium.

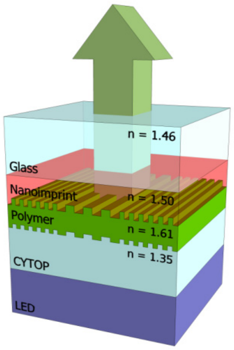

Fig. 4. Schematic drawing of a LED pumped polymer laser.

The InGaN LED was driven by a compact laser diode driver (module PCO-7110-120-15, Directed Energy Inc.), which is designed to provide high current (up to 120 A) pulses with FWHM of tens of nanoseconds. The pulse duration and peak current are dependent on the high voltage supplied to the LED driver. The trigger signal was supplied by a function generator, which also set the repetition frequency of the LED pulses. The optical pulse duration was measured using a fast photodiode (HSP-V2, Standa). The output pulse energy from the LED was measured with a calibrated energy meter for a range of driving currents. The LED power density is plotted against driving current in Fig. 5(a).

For an LED pumped UV-NIL second-order reference laser, the threshold was found to be approximately 500 W/cm^2 (260 nJ/pulse). Higher threshold for LED pumping than OPO pumping is believed to be associated with the longer pump pulse duration. A NIL mixed-order DFB laser with 30 periods of second-order gratings for out-coupling was pumped with the same LED. Laser action was observed at a wavelength of 537.1 nm at

#187978 - $15.00 USD Received 29 Mar 2013; revised 30 May 2013; accepted 30 May 2013; published 10 Jun 2013

pump power density of 210 W/cm^2 (100 nJ), as shown in Fig. 5(b)-(c). When the LED power was increased to about 260 W/cm^2 (130 nJ), a second band edge lasing mode at 534.1 nm came above threshold, see Fig. 5(d). The probable reason that the longer-wavelength mode had slightly lower threshold is that it had less surface output-coupling losses compared to the shorter-wavelength mode [5]. This is so far the lowest threshold reported for an LED pumped organic semiconductor laser. The result was found to be highly repeatable for multiple UV-nanoimprint lithography fabrication runs. The output pulse energy of an LED pumped second-order DFB laser was measured to be 3.15 nJ at an LED pulse energy of 385 nJ. The slope efficiency for the mixed-order laser was approximately half of the conventional second-order laser.

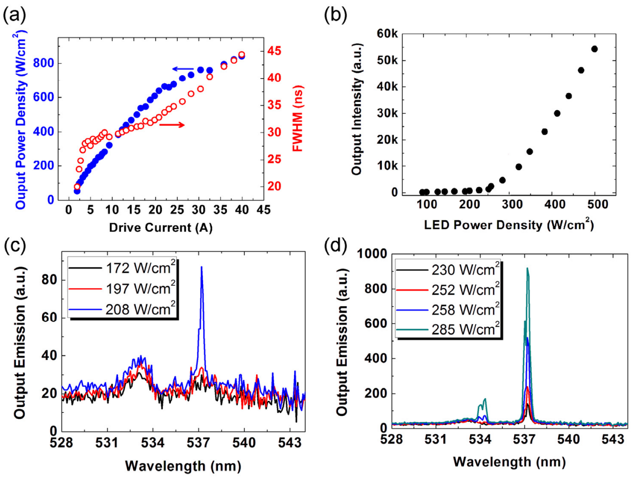

Fig. 5. (a) Output intensity (closed circles) from a pulsed LED as a function of driving current and the corresponding pulse duration (open circles); (b) surface emission of a mixed-order BBEHP-PPV laser at a wavelength of 537.1 nm as a function of LED intensity; (c) and (d) are the emission spectra at different LED pump intensities showing the onset of two band-edge lasing modes.

5. Conclusions

Distributed feedback lasers with resonators manufactured by UV nanoimprint lithography are presented. BBEHP-PPV was spun onto the resonator as the gain material and encapsulated by CYTOP. It was shown that a UV-NIL second-order DFB laser has a low threshold of 120 W/cm^2, while by employing a mixed-order resonator the threshold of the nanoimprinted lasers was reduced to below 60 W/cm^2. This allowed UV-NIL lasers to be pumped by a single LED with very low peak drive current, i.e. 5.6 A (approximately 330 A/cm^2). In conclusion, this work presents a simple route to indirect electrically pumped organic thin film laser devices. The very simple fabrication in combination with the low threshold and especially the possibility of LED pumping is very important as it paves the way to low-cost tunable visible lasers.

Acknowledgments

We would like to thank the EPSRC for financial support (grant number EP/F059922/1 and EP/F05999X/1). We also acknowledge EV Group for process support for the UV-nanoimprint lithography and Asahi Glass for the supply of CYTOP.