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This letter describes the design and operation of a polymer-based third order distributed feedback (DFB) microfluidic dye laser. The device relies on light confinement in a nanostructured polymer film where an array of nanofluidic channels is filled by capillary action with a liquid dye solution which has a refractive index lower than that of the polymer. In combination with a third order DFB grating, formed by the array of nanofluidic channels, this yields a low threshold for lasing. The laser is straightforward to integrate on lab-on-a-chip microsystems where coherent, tunable light in the visible range is desired. © *2006 American Institute of Physics.* [DOI: 10.1063/1.2345602]

Integration of optical and fluidic functionalities on a chip has recently been investigated by several research groups to realize novel optofluidic laser devices, microfluidic dye lasers, suitable for lab-on-a-chip microsystems.[1–4] These optically pumped devices consist of microfluidic channels with an embedded optical resonator and a liquid laser dye is used as active gain medium. In general, these devices can be added to a microfluidic chip without adding additional fabrication steps,[5] thus offering a simple way of integrating optical transducers to lab-on-a-chip microsystems,[6] and pave the way for advanced integrated sensor concepts.[7]

The threshold for lasing is a key parameter for the feasibility of these devices in a future technology. Distributed feedback (DFB) laser resonators have proven particularly suited in this respect. A DFB microfluidic dye laser was first demonstrated by Balslev and Kristensen,[2] who used a high order Bragg grating in an 8 $\mu$m thick polymer film to obtain feedback. Single mode lasing with a threshold fluence of approximately 20 $\mu$J/mm$^2$ was obtained due to mode selective losses in the multimode structure where light was not guided in the fluidic segments. Vezenov *et al.*[3] obtained liquid-core waveguiding using a low refractive index polymer and a high refractive index liquid. Li *et al.*[4] exploited this concept to realize a 15th order DFB laser with a record-low threshold fluence for lasing of approximately 8 $\mu$J/mm$^2$.

However, the 15th order Bragg grating still gives rise to scattering of light out of the chip plane.[8] These losses can be reduced by employing a lower order Bragg grating. Ideally, a first order Bragg grating combined with planar waveguiding in the polymer film could be used. The requirement of a high refractive index liquid and low refractive index polymer[3,4] may be relaxed by reducing the dimensions of the resonator segments as the subwavelength regime is entered.[9]

In this letter, we present the design and operation of a polymer-based third order DFB microfluidic dye laser. The device is fabricated by a flexible lithography technique, combining electron beam lithography (EBL) and UV lithography (UVL) in a single polymer film, enabling fast prototyping of device designs. In the device, see Fig. 1, a third order DFB resonator is formed by infiltrating a periodic array of nanofluidic channels with a liquid laser dye. The filling is mediated by capillary action, thus simplifying the operation of the

device as opposed to conventional setups with external syringe pumps. Replenishing of the dye in the cavity region is achieved for hours through capillary filling of a 16 cm long, 100 $\mu$m wide meandering channel following the cavity region. The speed of the advancing fluid front in the meandering channel decreases with time.[10] The dye solution in the cavity region is replaced in 30 s–5 min during the first 30 min of experiments (ethylene glycol solution). The flow rate in the $500 \times 500$ $\mu$m$^2$ cavity region can be further increased by increasing the width of the meandering channel. By employing a third order Bragg grating, we demonstrate lasing with a threshold fluence comparable to the results of Ref. 4. The subwavelength dimensions of the third order DFB grating yield a low coupling loss for the light when traversing the dye-filled nanofluidic channels, and combined
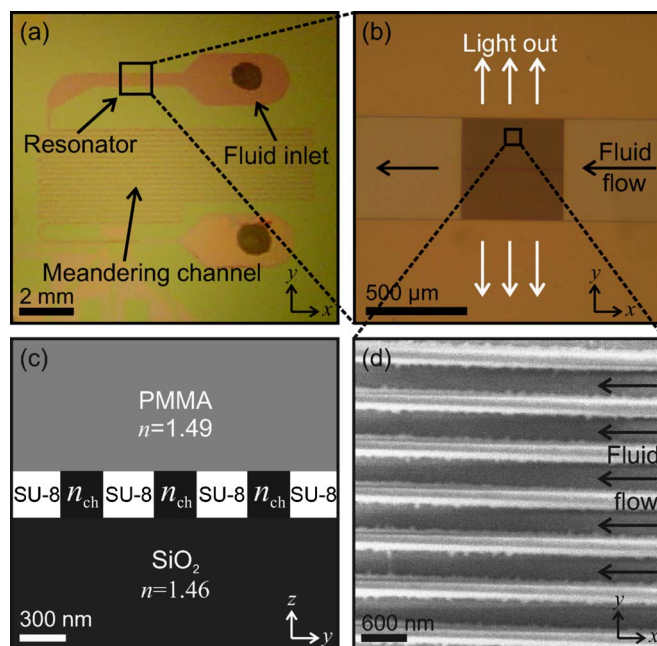


FIG. 1. (Color online) Overview of the fabricated microfluidic dye laser. (a) Top-view picture of the overall layout of the fabricated chip. The shallow 300 nm high meandering channel facilitates capillary filling of the embedded laser resonator. (b) Optical micrograph of the DFB laser resonator embedded in the shallow meandering channel. (c) Side-view sketch showing the layers of the DFB laser resonator. (d) Scanning electron micrograph showing the third order Bragg grating of nanofluidic channels which constitutes the DFB laser resonator (prior to bonding).

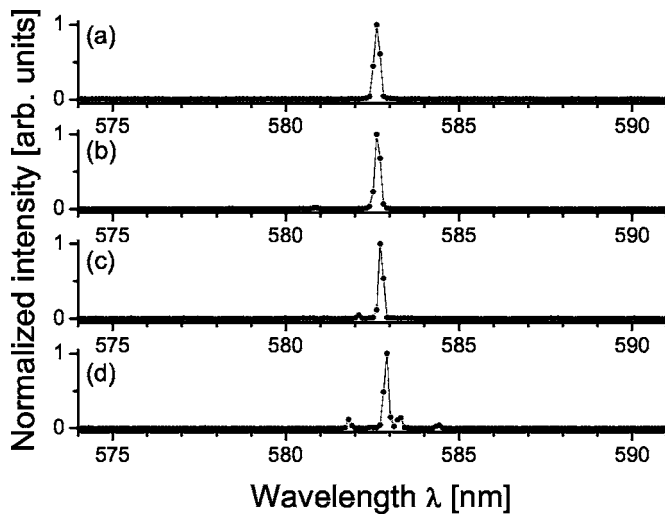[a)]Electronic mail: ak@mic.dtu.dk; URL: www.mic.dtu.dk/nil

FIG. 2. Normalized laser spectra from four nominally identical chips demonstrating wafer-scale spectral reproducibility of the laser. On each chip, the DFB laser resonator is filled by capillary action with rhodamine 6G (R6G) dissolved in ethylene glycol. The average laser wavelength of the chips is 582.72 nm with a standard deviation of 0.14 nm.
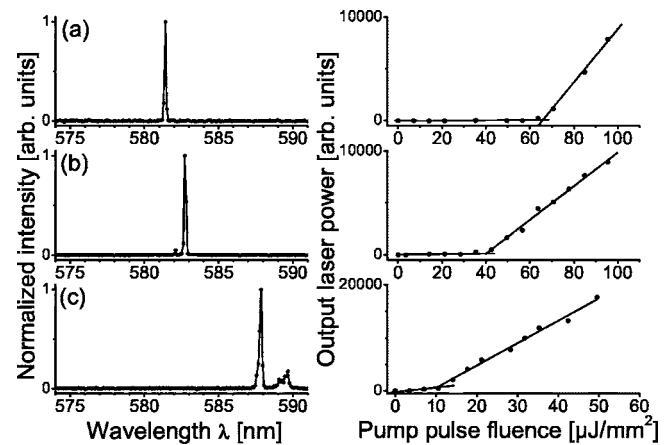


FIG. 3. Normalized laser spectra and corresponding laser output power curves as a function of pump pulse fluence for R6G in three different solutions: (a) R6G in ethanol, (b) R6G in ethylene glycol, and (c) R6G in a 1:1 mixture of ethylene glycol and benzyl alcohol. The results are summarized in Table I.

with the feedback of the DFB grating, this yields an efficient laser device.

The laser is based on a planar waveguide structure supporting a single propagating TE-TM mode. The basic waveguide structure consists of a $SiO_2$ (refractive index $n=1.46$) buffer substrate, a 300 nm thick core layer of the negative-tone resist SU-8 ($n=1.59$), and a top cladding of polymethyl-methacrylate (PMMA) ($n=1.49$). The device structure is formed by nanofluidic channels defined lithographically in the SU-8 film. The laser resonator consists of an array of 300 nm high nanochannels of period $\Lambda=601$ nm which comprises a third order Bragg grating with a central $\pi/2$ phase shift, embedded in a 500 $\mu$m wide shallow nanochannel.

To fabricate the devices, a 430 nm thick film of SU-8 2000 resist[11] is spin coated onto a Si substrate with a 2.5 $\mu$m thick thermally grown oxide layer. The wafer is baked at 90 °C for 1 min. The device structure is defined by combined EBL and UVL in the SU-8 film. The nanostructures of the third order DFB grating are defined by 100 kV electron beam exposure (JEOL-JBX9300FS, dose of 3 $\mu$C/cm$^2$). The total writing time for all 32 devices on a 4 in. wafer is 15 min. After electron beam exposure, the micron-sized structures (meandering channel and reservoirs) are defined in the same polymer film by UV exposure (20 s at 8.9 mW/cm$^2$), the wafer is postexposure baked at 90 °C for 1 min, and the micro and nanostructures are developed simultaneously in propylene glycol monomethyl ether acetate for 30 s, followed by an isopropyl alcohol rinse. The wafer is subsequently subjected to a 20 s soft oxygen plasma[12] to remove any residues of SU-8 at the $SiO_2$ surface in the nanochannels of the DFB resonator. After the plasma treatment, the structures are 300 nm high, and the heights of the EBL and UVL defined structures are matched within 30 nm. Fluidic access holes are formed by micropowder blasting[13] and the channels are sealed by a glass lid, using adhesive bonding by means of a 5 $\mu$m thick PMMA film (2200 N, 140 °C, 10 min).[14]

The laser is characterized by adding a droplet of dye solution to the fluid inlet, thus filling the DFB resonator structure by capillary action. In this letter, the laser dye

rhodamine 6G (R6G) is dissolved in ethylene glycol ($n=1.43$), ethanol ($n=1.33$), and in a 1:1 mixture of ethylene glycol and benzyl alcohol ($n=1.485$ by experimentally confirmed linear extrapolation of the refractive indices), all with a concentration of $2\times10^{-2}$ mol/L and a refractive index below that of the polymer surrounding the channel. The laser is optically pumped through the glass lid by a frequency doubled neodymium-doped yttrium aluminum garnet laser at 532 nm (5 ns pulse duration, 10 Hz repetition rate).

Figure 2 shows laser spectra from four different chips filled with R6G in ethylene glycol. The results demonstrate wafer-scale spectral reproducibility of the laser, exhibiting narrow-linewidth emission, polarized perpendicularly to the chip plane (TM). The average laser wavelength is 582.72 nm with a standard deviation of 0.14 nm, at the resolution limit of our spectrometer (0.15 nm). The deviation and the minor peaks in Figs. 2(c) and 2(d) may arise from grating imperfections due to fabrication defects. Due to limited spectrometer resolution, our equipment does not allow us to determine whether the laser is truly operating in a single mode, and the spectral shape and linewidth of the laser remain undetermined.

Figure 3 shows laser spectra from chips with R6G dissolved in different solvents, thus demonstrating tunability of the laser from 581.41 to 587.86 nm. A quantitative theoretical prediction of the shift in wavelength requires a full solution of Maxwell's equations in the fabricated structure, since the structures are of subwavelength dimensions.[9] In Fig. 3(c), the minor side mode shifted 1.8 nm from the main peak may be attributed to an additional phase shift introduced in the DFB grating by a stitching error during electron beam exposure. The dye laser output power is found by integrating the measured laser spectrum for each value of the average pump pulse fluence. The dye laser output power versus average pump pulse fluence follows the standard pump/output relation of two linear segments around a lasing threshold fluence $Q_{th}$.

The coupling loss $\gamma$ for a period of the DFB grating has been modeled by a finite-difference beam propagation method.[15,16] Light of the observed wavelength traveling in the TM mode of the single mode polymer waveguide is propagated through a 273 nm wide channel filled with a liq-

TABLE I. Summary of the results. For R6G solutions of larger refractive indices $n$, the laser wavelength $\lambda$ increases, and the threshold pump fluence $Q_{th}$ and coupling loss $\gamma$ decrease.

| Solvent | $n$ | $\lambda$ (nm) | $Q_{th}$ ($\mu$J/mm$^2$) | $\gamma$ |
|---|---|---|---|---|
| Ethanol | 1.33 | 581.41 | 65 | 0.191 |
| Ethylene glycol | 1.43 | 582.72 | 40 | 0.089 |
| Ethylene glycol/ benzyl alcohol | 1.485 | 587.86 | 10 | 0.043 |

uid of refractive index $n_{ch}$, corresponding to the value of the used solvent. The calculations yield the energy propagation loss corresponding to the coupling loss for a period of the DFB grating. The loss decreases for increasing $n_{ch}$, see Table I. To obtain lasing in the resonator, the amplification of the R6G must be equal to the loss. As expected, the experiments show that the threshold fluence for lasing decreases as the refractive index of the R6G solution increases, see Table I. Further, when considering the absorbed pump energy throughout the device, the effective threshold fluence may be considerably lower.

In summary, we have demonstrated a third order DFB microfluidic dye laser with a low threshold for lasing and operating with liquids of refractive index below the refractive indices of the surrounding polymer. Although experimental uncertainties make a direct comparison of lasing thresholds difficult, our results imply the importance of both liquid-core waveguiding[4] and low order Bragg reflection. The device relies on light confinement in a nanostructured polymer film where the individual resonator elements—nanofluidic channels and polymer walls—are of subwavelength dimensions. The resonator consists of an array of nanofluidic channels forming a third order DFB Bragg grating resonator. The laser is realized using a flexible fabrication technique, combining electron beam lithography and UV lithography in a single polymer film, followed by adhesive polymer wafer bonding. The laser is straightforward to integrate on lab-on-a-chip microsystems, e.g., for integrated sensors, where coherent, tunable light in the visible range is desired.

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