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## TUNABLE MICROFLUIDIC DYE LASER

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### ABSTRACT

We present a tunable microfluidic dye laser fabricated in SU-8. The tunability is enabled by integrating a microfluidic diffusion mixer with an existing microfluidic dye laser design by Helbo et al. By controlling the relative flows in the mixer between a dye solution and a solvent, the concentration of dye in the laser cavity can be adjusted, allowing the wavelength to be tuned. Wavelength tuning controlled by the dye concentration was demonstrated with macroscopic dye lasers already in 1971, but this principle only becomes practically applicable by the use of microfluidic mixing. With presently available dyes, the lasing wavelength can be tuned in an interval between 400 nm and 900 nm, depending on the specific dye. In this first demonstration, the lasing wavelength was tuned between 568 nm and 574 nm, using a solution of  $10^{-2}$  mol/L Rhodamine 6G in ethanol.

### INTRODUCTION

Since the first realizations in the 1960's, dye lasers have been used in many different applications in the visible region [1]. The organic dye molecules have a broad gain profile, which make dye lasers easily tunable over a wide range of wavelengths. Today, commercially available laser dyes cover the wavelength range from app. 400-900 nm. In liquid dye lasers, where the dye is dissolved in a suitable solvent, cooling and renewal of the active medium is achieved by a regenerating flow of dye through the laser cavity. Single mode operation and wavelength tunability is commonly achieved by actuated bulk optical components inside the laser cavity. The lasing wavelength can also be tuned by changing the concentration of dye molecules in the solvent [4].

A miniaturized liquid dye laser – a micro-cavity fluidic dye laser – that can readily be integrated with polymer based “lab-on-a-chip”-microsystems, was recently demonstrated [2,3]. In the present paper we present a tunable microfluidic dye laser. Integrating the micro-cavity fluidic dye laser with a microfluidic diffusion mixer enables the tunability. By controlling the relative flows in the mixer between a dye solution of high concentration and a solvent, the concentration of dye in the laser cavity can be adjusted, allowing the wavelength to be tuned. Although the wavelength tunability controlled by the dye concentration was discovered with macroscopic dye lasers already in 1971 [4], this principle only becomes practically applicable by the use of a microfluidic mixer.

### DESIGN

**Micro-cavity fluidic dye laser.** The micro-cavity fluidic dye laser is based on the design of Helbo et al. [2,3], see Fig. 1. The laser consists of a micro-fluidic channel fitted with metallic mirrors, which form the laser cavity. The laser dye flows through the device, which is pumped optically by an external laser. Dye laser output is emitted vertically through the semi-transparent top mirror. The micro-fluidic channel structure is formed in 10  $\mu\text{m}$  thick SU-8 photo-resist, and sandwiched between two Pyrex glass substrates. In the design of Helbo et al [2], the wafers were bonded together using a bonding layer of 10  $\mu\text{m}$  thick SU-8. The present design relies on a new adhesive bonding technique, using an intermediate bonding layer of 5  $\mu\text{m}$  thick poly-methylmethacrylate (PMMA). This is described further in the section on processing.

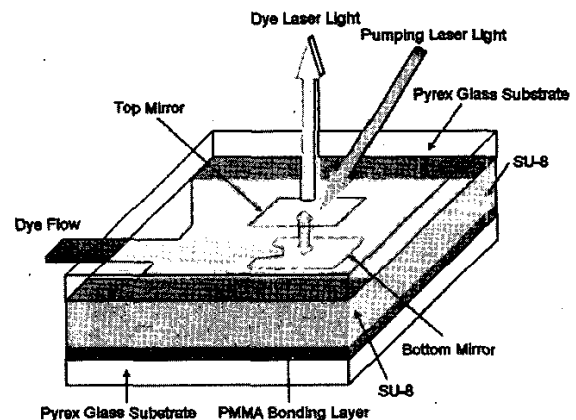


Fig. 1. Schematic drawing of the vertically emitting micro-cavity fluidic dye laser.

**Microfluidic mixer.** A diffusive microfluidic mixer is placed between the fluidic inlet and the laser cavity, see Fig. 2. We consider the laser dye Rhodamine 6G dissolved in ethanol. The small cross sectional area of the microfluidic mixer ( $300 \times 10 \mu\text{m}^2$ ), the small flow rate,  $Q = 10 \mu\text{L}/\text{hour}$ , and the viscosity of ethanol,  $\eta_{\text{ethanol}} = 1.197 \text{ mPas}$  ( $20^\circ\text{C}$ ) [5], gives a Reynolds number of 0.231, hence the flow in the mixer is laminar. Mixing of the ethanolic Rhodamine 6G solution and the pure ethanol thus relies on diffusion. The bulk diffusion constant,  $D$ , of Rhodamine 6G in water has earlier been investigated by photon burst analysis by Schuster et al. [6], and is found to be  $2.5 (\pm 1.7)$

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$\times 10^{-6} \text{ cm}^2/\text{s}$  at room temperature. According to the Stokes-Einstein relation,

$$D = \frac{k_B T}{6\pi\eta a} \quad (1)$$

the diffusion constant,  $D$ , of a molecule in a liquid is inversely proportional to the viscosity,  $\eta$ , of the liquid. Here  $k_B$  is Boltzmann's constant,  $T$  is the temperature, and  $a$  is the spherical diameter of the molecule. The diffusion constant of Rhodamine 6G in ethanol can therefore be found as the diffusion constant in water times the ratio between the viscosities of water and ethanol. The viscosity of water at 20°C is 1.0019 mPas [5]. This gives a diffusion constant of Rhodamine 6G in ethanol of  $2.1 (\pm 1.4) \times 10^{-6} \text{ cm}^2/\text{s}$  at room temperature. A value of  $1 \times 10^{-6} \text{ cm}^2/\text{s}$  is used to calculate the required length of the microfluidic mixer channel.

The necessary diffusion time,  $\tau$ , to ensure mixing, is calculated from the Einstein-Smoluchowski relation

$$\tau = \frac{d^2}{2D} \quad (2)$$

Here  $d$  is the diffusion length. The dye molecules are homogeneously distributed in one half of the channel width at the beginning of the mixer channel. At the end of the mixer channel, the dye should be uniformly distributed along the full channel width. In order to achieve this, Rhodamine 6G dye molecules must diffuse a distance of up to half the channel width, 150  $\mu\text{m}$ . This gives a diffusion time of 112.5 s, corresponding to a minimum channel length of 104 mm, for a flow rate,  $Q = 10 \mu\text{L}/\text{hour}$ . The final layout of the microfluidic channel structure with integrated diffusion mixer and fluidic micro-cavity dye laser is shown in Fig. 2. The length of the diffusion mixer channel is app. 122 mm.

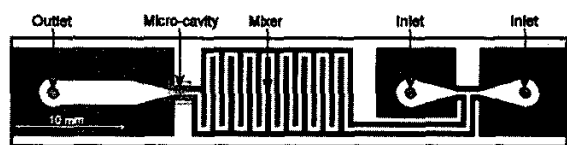


Fig. 2. Final layout of tuneable microfluidic dye laser chip. The dark area is the 10  $\mu\text{m}$  high SU-8 structures, and the white area is the channel structures. The length of the mixer channel is app. 122 mm, and the width is 300  $\mu\text{m}$ .

The flow resistance,  $R$ , of the microfluidic mixer can be estimated by using the following expression for a straight channel with a length,  $L$ , and with a rectangular cross section where the height,  $H$ , is smaller than the width,  $W$  [7]

$$R = \frac{96\eta L}{HW^3} \left[ 1 - \frac{192W}{\pi^2 H} \sum_{m=1,3,5,\dots}^{\infty} \frac{1}{m^5} \tanh\left(\frac{\pi m H}{2W}\right) \right]^{-1} \quad (3)$$

Expanding to lowest order in  $(H/W)$  gives

$$R \approx \frac{96\eta}{H^3} \left( \frac{L}{W} \right) \quad (4)$$

This expression, which is accurate within 1% when  $W/H > 100$ , gives  $\Delta P = R \times Q = 1.3 \text{ atm}$ , for the relevant parameters:  $H = 10 \mu\text{m}$ ,  $W = 300 \mu\text{m}$ ,  $L = 122 \text{ mm}$ ,  $\eta = 1.197 \text{ mPas}$ , and  $Q = 10 \mu\text{L}/\text{hour}$ . In order to be able to control the relative flow in the two inlets to the mixer with an acceptable relative pressure, the flow resistance of each inlet branch should be larger than the flow resistance of the mixer.

## PROCESSING

The tunable microfluidic laser devices are fabricated by standard micro fabrication techniques, as illustrated in Fig. 3. Before processing, the Pyrex wafers are cleaned with Triton X100 soap and 7-up etch (sulphuric acid and ammoniumperoxodisulphate at 80°C), and dehydrated at 220°C for 12 hours.

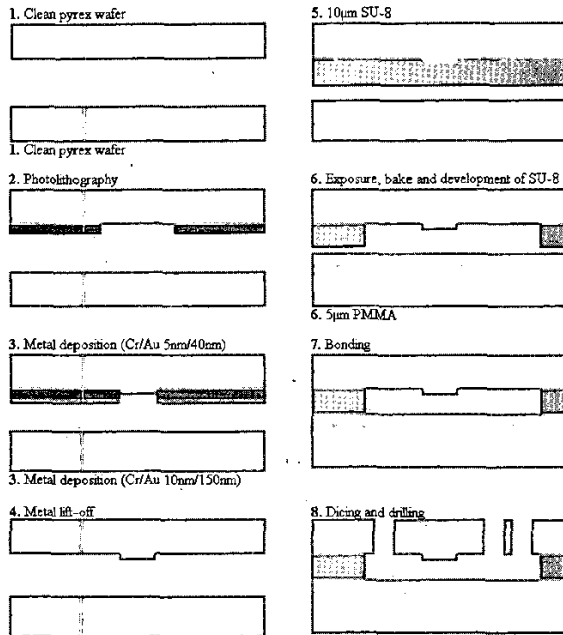
The Cr/Au metal mirrors for the laser cavity, are formed on the top and bottom Pyrex glass wafers by UV lithography, e-beam deposition and lift-off. The bottom mirror, 10 nm Cr and 150 nm Au, is designed to have maximum reflectance while the top mirror, 5 nm Cr and 40 nm Au, is designed to be semi-transparent with a transmittance of approximately 5% in the yellow part of the optical spectrum. The thickness of the metal layers was found by calculating the reflectance and transmittance from the refractive indices of the metals [8] and dielectric materials using an iterative procedure [9].

The microfluidic channel structure is defined in SU-8 photoresist [10] on the top wafer. The wafer is dehydrated at 120°C for 12 hours. A 10  $\mu\text{m}$  thick SU-8 layer is spun onto the wafer and pre-exposure baked on a hotplate (65°C for 2 min, 90°C for 2 min). The SU-8 photoresist is patterned by UV-lithography, post-exposure baked (65°C for 2 min, 90°C for 15 min), and finally developed in propylene glycol monomethyl ether acetate (PGMEA) for 3 min.

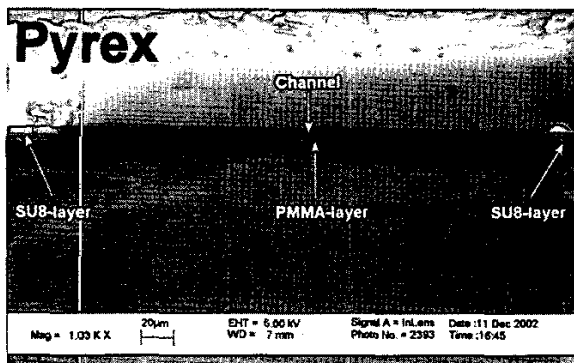
The microfluidic channels are sealed by a new adhesive bonding technique using PMMA [11] as an intermediate material. After dehydration at 120°C for 12 hours a 5  $\mu\text{m}$  thick PMMA layer is spun onto the bottom wafer and baked at 170°C for 30 min in order to remove the solvent. The two wafers are placed on top of each other with the SU-8 and PMMA face to face and with a bonding force of 5 N distributed over the full 4-inch sandwiched structure. The sandwiched structure is baked at 170°C for 12 hours with the bonding force applied. Finally the chips are diced and inlet/outlet holes to the microfluidic channels are drilled.

The scanning electron micrograph in Fig.4, shows the cross section of the 300  $\mu\text{m}$  wide, 10  $\mu\text{m}$  high microfluidic

channel on a cleaved, finished device. There are no signs of PMMA blocking the microfluidic channel.



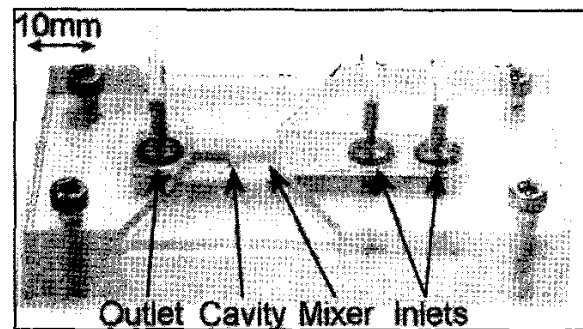
**Fig. 3.** Process overview. 500 µm thick, 4-inch diameter Pyrex wafers are used as top and bottom substrates. 1-4: Metal mirrors are deposited on the top and bottom wafers by standard UV-photolithography, electron-beam metal vapour deposition and lift-off. 5-6: The microfluidic channels are formed in a 10 µm thick SU-8 photoresist layer by spin-casting, pre-exposure baking, UV-photolithography, post-exposure baking and development. A 5 µm thick PMMA layer is spin-casted onto the bottom wafer and baked to evaporate the solvent. 7: The two wafers are bonded together using PMMA as an intermediate material. 8: The chips are diced and inlet/outlet holes are drilled through the top substrate.



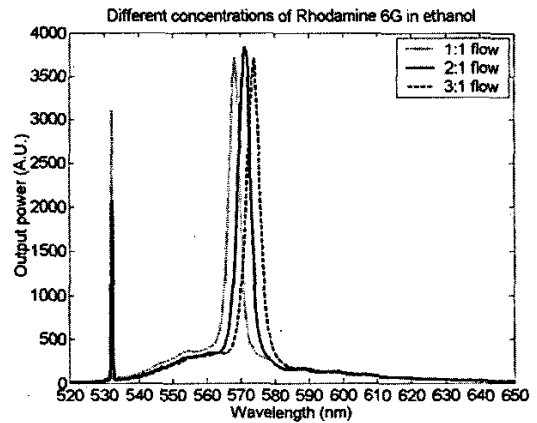
**Fig. 4.** Representative scanning electron micrograph of a cross section of the 300 µm wide and 10 µm high microfluidic channel after finished processing.

**OPTICAL MEASUREMENTS**

The tunable microfluidic dye laser chip is mounted in a polycarbonate sample holder, see Fig. 5. The sample holder is fitted with o-ring sealed fluidic connections, and a window giving direct access to the chip at the laser micro-cavity. The Rhodamine 6G solution and pure ethanol to be mixed is pumped through the two separate inlets by means of individual syringe pumps. The micro-cavity fluidic dye laser is pumped optically by a frequency doubled Nd:YAG laser (Continuum Surelite I-10), at a wavelength of 532 nm. The Nd:YAG laser is pulsed with a pulse width of 5 ns and a repetition frequency of 10 Hz. The pumping laser beam impinging on the laser device has a diameter around 1.5 mm. The emitted dye laser output is collected by an optical fiber, and analyzed in a spectrometer (Avantes, AVS-HR2000, 10 µm slit).



**Fig. 5.** Photo of the sample holder made from polycarbonate, and fitted with o-ring sealed fluidic connections, and a window giving direct access to the laser cavity region. The chip is fixed between a top and bottom plate.



**Fig. 6.** Graph showing how the peak lasing wavelength changes when the relative flow rate between a flow of  $10^{-2}$  mol/L ethanolic Rhodamine 6G solution and a flow of pure ethanol is varied. The first peak at 532 nm is the scattered pumping laser light and the three peaks at 568 nm, 571 nm and 574 nm are the dye lasing peaks for the corresponding relative flows (Rhodamine 6G:ethanol) 1:1, 2:1 and 3:1.

Spectra recorded with three different flow ratios are shown in Fig. 6. A peak from scattered pumping laser light is observed at 532 nm on all three spectra. The pumping intensity for the three spectra was  $\sim 300$  mW/cm<sup>2</sup>. This is above the lasing threshold for the device,  $\sim 34$  mW/cm<sup>2</sup>. A strong lasing peak emerges from the Rhodamine 6G dye fluorescence, around 570 nm. As seen, the lasing wavelength could be varied between 568 nm and 574 nm using a flow of an  $10^{-2}$  mol/L ethanolic rhodamine 6G solution diluted with a second flow of pure ethanol. The relative flows between rhodamine 6G solution and ethanol are 1:1, 2:1, 3:1 resulting in peak wavelengths of 568 nm, 571 nm and 574 nm, respectively.

### CONCLUSION

We have realized a micro-fabricated tuneable liquid dye laser, and demonstrated that wavelength tuning controlled by dye concentration is usable, when combined with microfluidic diffusive mixing. This was achieved by integrating an existing microfluidic dye laser design by Helbo et al. [2,3] with a microfluidic diffusion mixer on a SU-8 based microfluidic chip. In this first demonstration, the lasing wavelength was tuned between 568 nm and 574 nm, using a solution of  $10^{-2}$  mol/L Rhodamine 6G in ethanol, which was mixed with a pure ethanol in ratios from 1:1 to 3:1.

### Acknowledgments

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