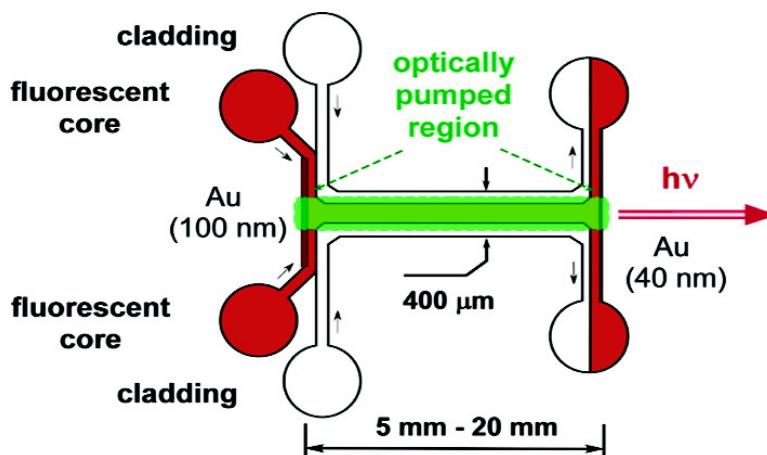Communication

A Low-Threshold, High-Efficiency Microfluidic Waveguide Laser

Dmitri V. Vezenov, Brian T. Mayers, Richard S. Conroy, George M. Whitesides,
Preston T. Snee, Yinthai Chan, Daniel G. Nocera, and Moungi G. Bawendi

More About This Article

Additional resources and features associated with this article are available within the HTML version:

- Supporting Information
- Links to the 9 articles that cite this article, as of the time of this article download
- Access to high resolution figures
- Links to articles and content related to this article
- Copyright permission to reproduce figures and/or text from this article

View the Full Text HTML

A Low-Threshold, High-Efficiency Microfluidic Waveguide Laser

Dmitri V. Vezenov,[†] Brian T. Mayers,[†] Richard S. Conroy,[†] George M. Whitesides,*,[†]
Preston T. Snee,[‡] Yinthai Chan,[‡] Daniel G. Nocera,[‡] and Moungi G. Bawendi[‡]

*Department of Chemistry and Chemical Biology, Harvard University, 12 Oxford Street,
Cambridge, Massachusetts 02138, and Department of Chemistry, Massachusetts Institute of Technology,
77 Massachusetts Avenue, Cambridge, Massachusetts 02139*

Received March 18, 2005; E-mail: gwhitesides@gmwgroup.harvard.edu

We describe the use of a liquid−liquid (L^2) waveguide as the basis of a dye laser suitable for integration with microfluidic systems. This system (i) provides low threshold using a long optical cavity, (ii) simplifies the liquid handling and optical setup compared to conventional dye lasers (by omitting movable mirrors, prisms, and gratings), and (iii) integrates the laser into microfabricated fluidic networks. Vertical-cavity surface-emitting lasers can be monolithically integrated into microfluidic systems; however, they lack the tunability of the emission wavelength.[1] Conventional dye lasers provide sharp, bright spectral lines that are tunable over a range of wavelengths (>30 nm for a single dye).[2] They have two disadvantages: (i) they need high-power optical pump sources that are bulky and expensive and, thus, not readily amenable to miniaturization and (ii) they require rapid replenishment of the dye to avoid bleaching, and thus they consume large volumes of dye-containing solutions. Using microfluidics and L^2 waveguides, we can address these disadvantages.

The structure of L^2 waveguides is defined by the laminar flow of multiple liquid streams in a single microfluidic channel in an arrangement where two streams of a low refractive index fluid, the liquid cladding, envelop a third stream of high refractive index, the liquid core.[3,4] L^2 waveguides are dynamic systems and can be reconfigured by changing the dye, the solvent composition, or the flow rates of the liquids. Using L^2 waveguides, one achieves, in real time, direct control of the emission wavelength, numerical aperture, absorbance, size, geometry, and modal content of the fluorescent waveguide. In addition, the optical interfaces forming the waveguide are the smooth boundaries defined by liquids under laminar flow. Accordingly, the requirements for the roughness of the channels are substantially lower than those for conventional waveguides.[3,5,6]

We fabricated the microfluidic laser (Figure 1) using soft lithography.[7] The 5−20-mm-long active region is terminated at both ends with T-junctions, which were coated with thin layers of gold to act as mirrors for the optical cavity. Pressure-driven core streams containing fluorescent dye and cladding streams flowed in from both sides of a single T-junction and down the length of the active region, forming the L^2 waveguide. Mixing of the core and cladding streams occurs by diffusion only (Reynolds number, Re, ∼0.001−0.02, see the Supporting Information, SI).

For initial characterization, we used 2 mM rhodamine 640 perchlorate in methanol as the core stream and pure methanol as the cladding stream. At the rate of flow chosen in our experiments (32−4 mL/h), the incoming pump pulse (50 Hz repetition rate) excited a given volume of the dye solution within the fluorescent core between 2 and 20 times (channel traversal times of 45−360 ms). The dependence of the power of the output light on the flow
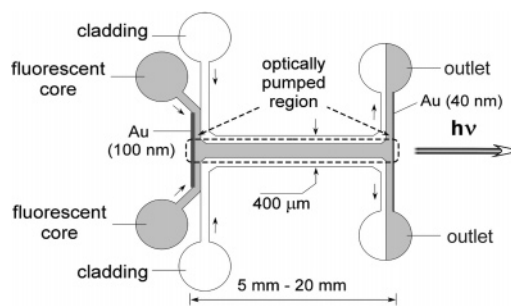

Figure 1. Top-view scheme for an L^2 waveguide laser consisting of a liquid cladding and fluorescent liquid core flowing laminarly in a microfluidic channel in PDMS (n_D = 1.41). The dimensions of the central channel were 100 μm × 400 μm × 10 mm (height × width × length). The 532-nm laser beam (frequency doubled Nd:YAG, 50 Hz repetition rate, 16-ns pulse) was elongated with a cylindrical lens, and the optical pumping region covered the full length of the waveguiding region for a 10-mm-long channel.

rate within the above range of low rates was minimal (variation < 20% for averaged power output). The diffusional mixing between core and cladding was also minimal (see SI). The rapid replacement of the dye, coupled with the short (16 ns) pulse length, eliminated potential problems associated with the absorption of the laser radiation by the long-lived triplet state and bleaching of the dye at power levels used (up to 0.15 mJ/pulse). While microfluidic lasers have been reported,[8−10] this communication describes the first microfluidic waveguide laser with long path length (in a single pass) through the gain medium; a 10-mm path is equivalent to 500 round trips in a 10-μm fluidic microcavity. Indeed, we observed considerable line width-narrowing and amplified spontaneous emission (ASE) even in devices without gold mirrors at the front and back wall of the channel (see SI). Figure 2A displays the transition from spontaneous emission to lasing in the spectral output of a 10-mm-long device with increasing optical pump intensity. Figure 2B is a plot of output power and line width as a function of pump power for the device shown in Figure 1. At low pump power (<5 μJ/pulse), the line width (full width at half-maximum, fwhm) of the emission is ∼45 nm, with λ_{max} centered at 625 nm; these values are similar to those obtained for a cuvette of rhodamine 640 perchlorate in methanol using a fluorimeter. The fwhm drops by an order of magnitude to ∼4 nm between pump powers of 7 and 16 μJ. The rapid decrease in fwhm and the drastic change in the slope in the plot of output power versus pump power indicate the onset of ASE at pulse energies of ∼10 μJ (see SI). The line width of the output above the threshold is comparable to that of other microfluidic dye lasers.[8−10] The plot of output light intensity versus pump power for L^2 waveguide without mirrors is well-described by the asymptotic formulas for ASE at low and high pump intensities.[11] The divergence angle obtained from such fits is 80 mrad and is in reasonable agreement with direct experimental
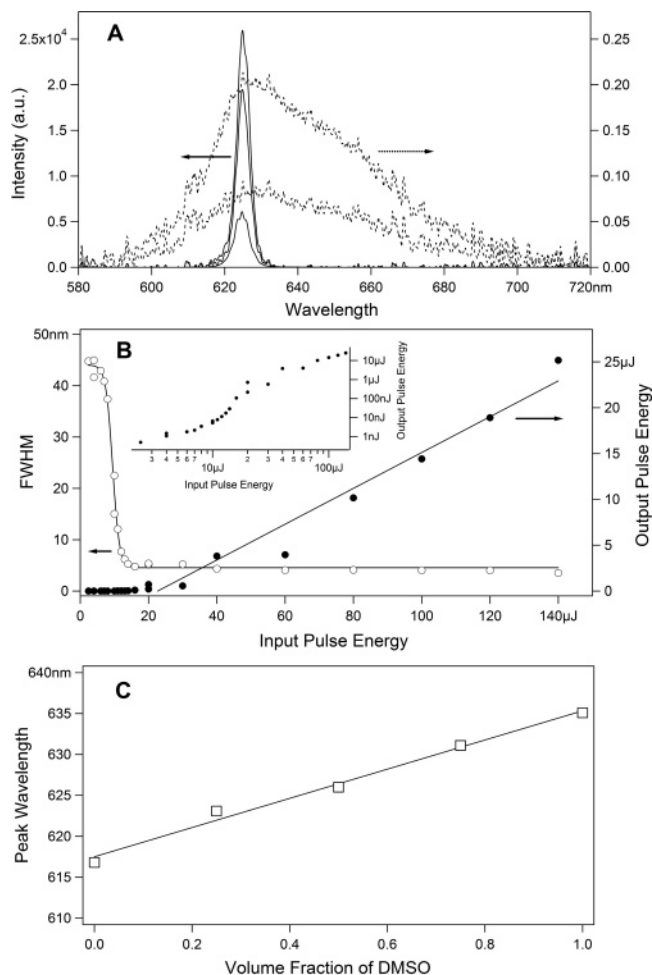
Figure 2. Output of the L^2 waveguide of Figure 1 containing 2 mM rhodamine 640 perchlorate in methanol—the core—and pure methanol—the cladding. (A) Emission spectra from L^2 fluorescent light source at different pump powers measured on-axis (note the 10^5 difference in scale for spectra at input powers below (dashed) and above (solid) threshold. (B) Dependence of the intensity of the optical output and line width (fwhm) on the energy of the pump pulse. The curves are drawn to guide the eye. Inset: output vs input power on a log—log scale from data on integrated peak area. (C) Adjustment of the peak wavelength of the light output by changing the composition of the mixed methanol—DMSO liquid core with constant concentration of the dye (2 mM).

measurements of beam divergence (25—45 mrad). The threshold for lasing occurs at 22 μJ pulse energy (corresponding to 70 kW/cm^2 instantaneous or 55 mW/cm^2 average power density). This L^2 waveguide system displays a slope efficiency of ~20%, calculated as the ratio of measured power emitted by the dye along the waveguide axis to the power of the pump laser absorbed by the device (e.g., incident power less ~3% reflected by the poly-(dimethylsiloxane) (PDMS)—air interface and ~7% transmitted through the device).

We could modify the properties of the waveguide by adjusting the refractive index contrast ($\Delta n = n_{core} - n_{cladding}$) between the core and the cladding liquids. We could also assess the contribution of residual reflectivity at the interface between the liquid core and the wall of the PDMS to transition to ASE in microchannels without mirrors. Interestingly, above and below threshold, the output characteristics—intensity and line width—were very similar for methanol-core/methanol-cladding ($\Delta n = 0.0001$) and for ethylene glycol (EG)-core/methanol-cladding ($\Delta n = 0.1$), as well as for a series of DMSO—methanol mixtures with methanol cladding ($\Delta n = 0.0001-0.15$), including the case where the liquid core was

index-matched to the PDMS. We do, however, see a large difference in the slope of the transition to lasing for waveguides with different Δn (Figure S4 in SI). The transition is much sharper for L^2 waveguides having a higher numerical aperture (NA $\propto \Delta n$), whereas systems with small values of Δn show spectral line narrowing at values of input power much lower than threshold. In addition to having a sharper transition to lasing, the systems with high Δn, where the light is strongly confined to the waveguide, have higher efficiencies of conversion and wider divergence angles (e.g., 24% and 100 mrad, respectively, for EG-core and methanol-cladding) than the systems with low Δn (e.g., 10% and 40 mrad, respectively, for methanol-core and methanol-cladding), where light is weakly confined to the waveguide.

Changing the solvent in the fluorescent core provides a simple means of adjusting the wavelength of emission for a given dye without incorporating dispersive elements (prisms or gratings) into the optical cavity. For example, the wavelength of the light output for rhodamine 640 can be shifted by more than 20 nm by changing the composition of the core liquid; $\lambda_{max} = 617$ nm for methanol, 631 nm for EG, and 634 nm for DMSO. Further, the λ_{max} could be tuned continuously by adjusting the composition of a mixture of DMSO and MeOH in the core. Figure 2C displays the linear relationship between the DMSO/methanol ratio and λ_{max} at fixed dye concentration (2 mM) and path length (10 mm).

Before lasers are widely used in fluidic microsystems, they must be (i) simple and inexpensive to fabricate and operate, (ii) easy to integrate into microfluidic systems, (iii) amenable to miniaturization, and (iv) easily tunable in wavelength and intensity. The long-path microfluidic laser described here meets these conditions. It allows for narrow line width, low-threshold emission that is highly directional (even without the incorporation of mirrors). While in-line mixing of solvents provides a viable route to tuning the output wavelength in systems where dispersive elements (e.g., diffraction gratings or prisms) are difficult to implement, the relatively large spectral bandwidth and inability to achieve cw operation are limitations of the present setup.

Supporting Information Available: Experimental methods, diffusion modeling, and ASE modeling. This material is available free of charge via the Internet at http://pubs.acs.org.

References

(1) Thrush, E.; Levi, O.; Cook, L. J.; Deich, J.; Kurtz, A.; Smith, S. J.; Moerner, W. E.; Harris, J. S. *Sens. Actuators, B* **2005**, *105*, 393.
(2) Duarte, F. J. *Opt. Photonics News* **2003**, *14*, 20.
(3) Wolfe, D. B.; Conroy, R. S.; Garstecki, P.; Mayers, B. T.; Fischbach, M. A.; Paul, K. E.; Prentiss, M.; Whitesides, G. M. *Proc. Natl. Acad. Sci. U.S.A.* **2004**, *101*, 12434.
(4) Vezenov, D. V.; Mayers, B. T.; Wolfe, D. B.; Whitesides, G. M. *Appl. Phys. Lett.* **2005**, *86*, 041104.
(5) Stroock, A. D.; Dertinger, S. K. W.; Ajdari, A.; Mezic, I.; Stone, H. A.; Whitesides, G. M. *Science* **2002**, *295*, 647.
(6) Ma, H.; Jen, A. K. Y.; Dalton, L. R. *Adv. Mater.* **2002**, *14*, 1339.
(7) A Special Issue on Soft Lithography. *Adv. Mater.* **2004**, 16.
(8) Balslev, S.; Kristensen, A. *Opt. Express* **2005**, *13*, 344.
(9) Cheng, Y.; Sugioka, K.; Midorikawa, K. *Opt. Lett.* **2004**, *29*, 2007.
(10) Helbo, B.; Kristensen, A.; Menon, A. *J. Micromech. Microeng.* **2003**, *13*, 307.
(11) Svelto, O.; Taccheo, S.; Svelto, C. *Opt. Commun.* **1998**, *149*, 277.

JA0517421