

Fluidic fibre dye lasers

A. E. Vasdekis, G. E. Town,¹ G. A. Turnbull, I. D. W. Samuel

*Organic Semiconductor Centre & Ultrafast Photonics Collaboration, SUPA, School of Physics and Astronomy,
University of St Andrews, St Andrews, Fife, KY16 9SS, UK*

ids@st-andrews.ac.uk
<http://www.st-andrews.ac.uk/~osc>

¹*Department of Electronics and Centre for Lasers & Applications,
Macquarie University, NSW 2109, Australia.*

<http://www.elec.mq.edu.au/gwopr>

Abstract: We report the demonstration of compact fluidic fibre lasers based on capillary tubes and photonic crystal fibres, featuring single channel and multiple laterally integrated fluidic lasers respectively. Their preparation was based on capillary action and lasing occurred without the need for external mirrors or lithographically defined microstructures. The fibre lasers were found to be tunable by varying the chromophore density in the liquid core and a functional wavelength selectivity mechanism inherent in both types of lasers provided a long free spectral range that does not correspond to the length of the fibres. The enhanced mode spacing is attributed to a Vernier resonant effect.

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References and links

1. D. Psaltis, S.R. Quake, C. Yang, "Developing optofluidic technology through the fusion of microfluidics and optics," *Nature* **442**, 381-386 (2006).
2. D.-Y. Zhang, N. Justis, Y.-H. Lo, "Fluidic adaptive lens of transformable lens type," *Appl. Phys. Lett.* **84**, 4194-4196 (2004).
3. S.C.-McGreehin, T.F. Krauss, K. Dholakia, "Integrated monolithic optical manipulation," *Lab on a Chip* **6**, 1122-1124 (2006).
4. P. Mach, M. Dolinski, K.W. Baldwin, J.A. Rogers, C. Kerbage, R.S. Windeler, B.J. Eggleton, "Tunable microfluidic optical fiber," *Appl. Phys. Lett.* **80**, 4294-4296 (2002).
5. D. Erickson, T. Rockwood, T. Emery, A. Scherer, D. Psaltis, "Nanofluidic tuning of photonic crystal circuits," *Opt. Lett.* **31**, 59-61 (2006).
6. Y. Xia, G.M. Whitesides, "Soft lithography," *Angew. Chem. Int. Ed.* **37**, 550-575 (1998).
7. S.R. Quake, A. Scherer, "From micro- to nanofabrication with soft materials," *Science* **290**, 1536-1540 (2000).
8. Z. Li, Z. Zhang, A. Scherer, D. Psaltis, "Mechanically tunable optofluidic distributed feedback dye laser," *Opt. Express* **14**, 10494-10499 (2006).
9. M. Gersborg-Hansen, A. Kristensen, "Tunability of optofluidic distributed feedback dye lasers," *Opt. Express* **15**, 137-142 (2007).
10. T. Kobayashi, W.J. Blau, "Laser emission from conjugated polymer in fibre waveguide structure," *Electron. Lett.* **38**, 67-68 (2002).
11. D.V. Vezenov, B.T. Mayers, R.S. Conroy, G.M. Whitesides, P.T. Snee, Y. Chan, D.G. Nocera, M.G. Bawendi, "A low-threshold, high-efficiency microfluidic waveguide laser," *J. Am. Chem. Soc.* **127**, 8952-8953 (2005).
12. P. Russell, "Photonic crystal fibers," *Science* **299**, 358-362 (2003).
13. P. Steinvurzel, B.T. Kuhlmeier, T.P. White, M.J. Steel, C. Martijn de Sterke, B.J. Eggleton, "Long wavelength anti-resonant guidance in high index inclusion microstructured fibers," *Opt. Express* **12**, 5424-5433 (2004).
14. B. Helbo, S. Kragh, B.G. Kjeldsen, J.L. Reimers, A. Kristensen, "Investigation of the dye concentration influence on the lasing wavelength and threshold for a micro-fluidic dye laser," *Sens. Actuators A* **111**, 21-25 (2004).

15. H.G. Danielmeyer, "Effects of drift and diffusion of excited states on spatial hole burning and laser oscillation," *J. Appl. Phys.* **42**, 3125-3132 (1971).
16. S. Yokoyama, T. Nakahama, S. Mashiko, "Amplified spontaneous emission and laser emission from a high optical-gain medium of dye-doped dendrimer," *J. Luminescence* **111**, 285-290 (2005).
17. D. Ouyang, R. Heitz, N.N. Ledentsov, S. Bognar, R.L. Sellin, Ch. Ribbat, D. Bimberg, "Lateral-cavity spectral hole burning in quantum dot lasers," *App. Phys. Lett.* **81**, 1546-1548 (2002).
18. E.P. O'Reilly, A.I. Onischenko, E.A. Avrutin, D. Bhattacharyya, J.H. Marsh, "Longitudinal mode grouping in InGaAs/GaAs/AlGaAs quantum dot lasers: origin and means of control," *Electron. Lett.* **34**, 2035-2037 (1998).
19. M. Sugawara, K. Mukai, Y. Nakata, H. Ishikawa, "Effect of homogeneous broadening of optical gain on lasing spectra in self-assembled In_xGa_{1-x}As/GaAs quantum dot lasers," *Phys. Rev. B* **61**, 7595-7603 (2000).
20. M. Horowitz, R. Daisy, B. Fischer, J.L. Zyskind, "Linewidth-narrowing mechanism in lasers by nonlinear wave mixing," *Opt. Lett.* **19**, 1406-1408 (1994).
21. R.C. Polson, G. Levina, Z.V. Vardeny, "Spectral analysis of polymer microring lasers," *Appl. Phys. Lett.* **76**, 3858-3860 (2000).
22. Y.H. Ja, "Optical Vernier filter with fibre grating Fabry-Perot resonators," *Appl. Opt.* **34** 6164-6167 (1995).
23. A.J. Poustie, N. Finlayson, P. Harper, "Multiwavelength fiber laser using spatial mode beating filter," *Opt. Lett.* **19**, 716-718 (1994).
24. L.M. Blinov, G. Cipparrone, P. Pagliusi, V.V. Lazarev, S.P. Palto, "Mirrorless lasing from nematic liquid crystals in the plane waveguide geometry without refractive index or gain modulation," *Appl. Phys. Lett.* **89**, 031114 1-3 (2006).

1. Introduction

In recent years, opto-fluidic devices have emerged as a tool for sensing, imaging and spectroscopy application as they uniquely combine the non-intrusive interaction capabilities of photons and the fluidic delivery of biological samples [1]. Several types of devices have been demonstrated including adaptive lenses, optical manipulators of particles and sensors based on photonic crystal fibres and waveguides [2-5]. The practicality of most systems is significantly enhanced by cost-effective fabrication techniques, such as soft lithography [6, 7]. Increasing interest has been focused additionally on fluidic organic lasers that can act as efficient sources of narrow linewidth emission in the visible spectral range [8-11]. These sources can be tuned over 45 nm for a single chromophore and their integration within fluidic networks can be easily implemented achieving significant progress towards lab-on-a-chip applications.

In this paper, we demonstrate fluidic lasers based on optical fibres. We explore two geometries realised in single and multiple cores fibres based on low cost and commercially available capillaries and photonic crystal fibres respectively [12]. The latter demonstrates the potential of multiple laterally integrated fluidic lasers, approximately 50 in our experiments. In both structures, lasing is achieved without the need of external mirrors or lithographically defined microstructures, featuring thus a rapid fabrication technique of compact laser sources. Long interaction lengths are achieved in both geometries, even for the small volumes of the gain medium (~ nL), where in addition an advantageous spectral selectivity is observed despite the long cavity lengths.

2. Methodology

The single core capillaries had a core and fused silica cladding diameters of 2 μm and 128 μm respectively (Polymicro Technologies). The photonic crystal fibre was the ESM-12-01 (Crystal Fibre), which consists of a hexagonal array of capillaries with diameter of 3.68 μm at a period of 8 μm, a defect silica core and a silica cladding of 12 μm and 125 μm in diameter respectively. The inner side of the capillary walls was not chemically treated and the fibres' coating was stripped both in the single and multiple cores (polyimide and acrylate coating respectively). Both types of fibres were examined using an SEM and shown in Fig. 1.

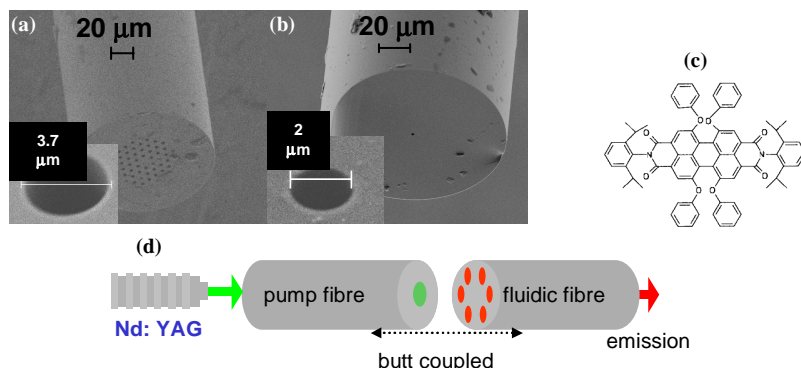


Fig. 1. (a), (b). SEM images of the fibres; inset shows the lateral dimensions of the cores. (c). the molecular structure of the organic dye Perylene Red. (d). A schematic of the set-up for the longitudinal excitation of the fluidic fibres.

The gain medium was a toluene solution of the ionized organic dye Perylene Red (BASF) at a concentration of 0.5 mg/ml (Fig. 1(c)). The solvent was chosen based on its higher refractive index than fused silica and its appropriate wetting properties with the capillary walls. The preparation of the fibre lasers involved the cleavage of the fibres at appropriate lengths and their subsequent infilling by capillary action from dipping them in the dye solution. The infilling process was relatively rapid and was measured to be 1 $\mu\text{m}/\text{sec}$ on average for a 25 mm long test capillary tube with a 5 μm core diameter. This technique is promising for high throughput preparation; however it is characterised by the presence of on average 0.5 mm long air-plugs on either side of the fibres, resulting in a mismatch of termination of the fluidic regions with respect to the fibre facets [13]. To delay the solvent evaporation and the photo-oxidation of the organic chromophore, the fibre ends were end-capped by dipping the fibres in a viscous solution of the transparent polymer CYTOP®.

The optical pump source was a frequency-doubled diode pumped Nd: YVO₄ laser with repetition rate and pulse duration of 200 Hz and 7.5 nsec respectively. The laser beam was coupled to an 8 μm core diameter silica fibre and butt-coupled to the fluidic lasers to longitudinally excite them (Fig. 1(d)). It was found that the diameter of the pump fibre was critical both in terms of threshold and quantum efficiency of the fluidic lasers. The 8 μm core diameter pump fibre provided the optimal combination for both the single core capillary and the holey fibre. For the aforementioned concentration, the excitation is absorbed in the first 800 μm of propagation.

3. Characterisation

The axial emission from the fibres was focused using a 25 \times microscope objective on to a fibre coupled CCD spectrometer with a 40 μm resolution. Typical emission spectra above and below threshold are depicted in Fig. 2 for a 41 mm long single core capillary (a) and a 39 mm long photonic crystal fibre (b). As both types of lasers exceed threshold, the broadband emission becomes characterised by a series of narrow linewidth peaks. The near-field images above and below threshold for the multi-channel fluidic laser are shown in (c). The transverse intensity variation above threshold designates the localization of the light within the cores, indicative of stimulated emission.

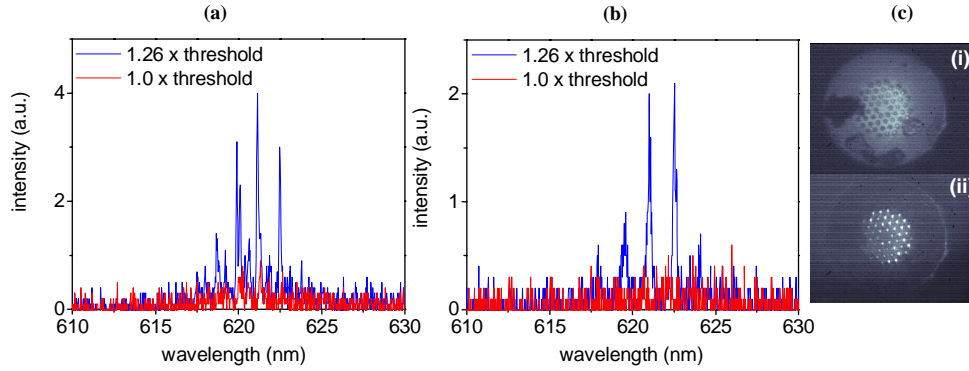


Fig. 2. The emission spectra at threshold and 1.26 times above threshold for the 2 μm diameter capillary (a) and for the photonic crystal fibre (b). In (c), the near field images are depicted below (i) and above (ii) threshold.

The threshold energy and the slope efficiency were found to be different for the single and multiple core fluidic lasers of similar lengths (Fig. 3(left)). A number of factors may contribute to such an effect, including the efficiency of the coupling of the pump light into the liquid core and also the fact that in the photonic crystal fibre the active area is larger and multiple cores are excited simultaneously. In addition, the laser threshold and slope efficiency were found to be dependent on the fibre length. This effect is shown in Fig. 3(right), where the input-output curves are plotted for the photonic crystal fibre lasers of different length. Such behaviour is indicative of the length mismatch between the amplification section and the total propagation length. The emitted photons experience gain only in the initial section of the fibre where the excitation is absorbed, while the rest of the liquid waveguide acts as a source of loss due to re-absorption, reducing thus the output intensity.

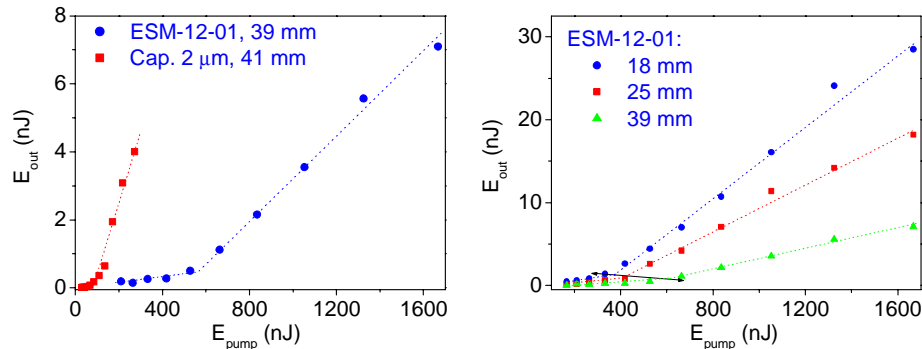


Fig. 3. Comparative study of the input-output relationship for a single and multi-channel fibre laser (left) and for a multi-channel laser of different lengths (right). In the right figure the black arrow designates the threshold energies for the three lasers.

The laser emission was also found to be tunable by varying the concentration of the dye in the liquid gain medium. An increase in the chromophore density resulted in increased absorption losses at the shorter wavelengths of the fluorescence spectrum due to the finite Stokes' shift in the electronic structure of the dye molecules. The narrowing of the fluorescence spectra for increasing dye concentration is illustrated in Fig. 4(a). Such red-shift of the blue-end of the edge-emitted fluorescence results in a convenient gain-tuning mechanism since the gain peak red-shifts accordingly for increasing chromophore densities [14]. The tuning of the centre wavelength of the laser emission spectrum is illustrated in Fig. 4(b). At different chromophore densities, lasing occurs at the vibronic of the highest net-gain,

while the laser performance does not degrade significantly as indicated by the slope efficiency measurements for the same concentrations (Fig. 4(c)).

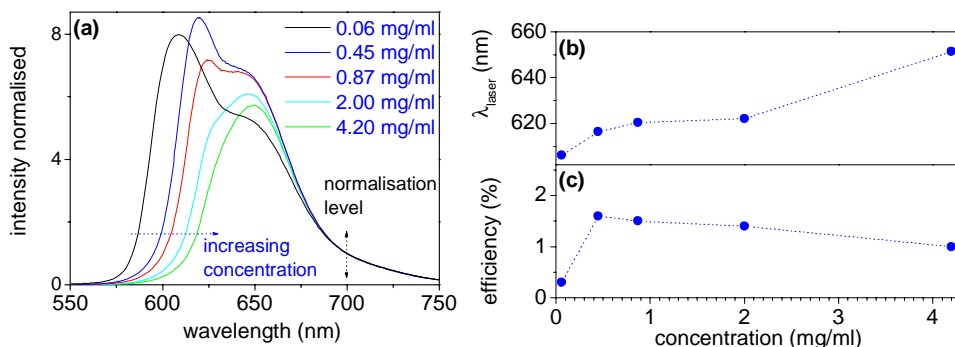


Fig. 4. For increasing chromophore densities, the blue-end of the fluorescence spectra red-shifts (a) and the centre wavelength of the laser emission spectrum is accordingly tuned (b). Under the same conditions, the laser efficiency does not vary significantly (c).

4. Spectral selectivity

Both in the single and in the multi-core fluidic lasers, the measured free spectral range is approximately 300 times longer than expected for a Fabry-Perot resonator formed by the liquid waveguide and the solvent-air interfaces. The laser emission appears to be strongly modulated by an envelope function, allowing the appearance of only certain longitudinal modes. This intracavity wavelength selectivity mechanism is unanticipated though conveniently provides a functional mode spacing that does not correspond to the fibre length.

A wide range of processes have been reported to affect the mode spacing in waveguide lasers. One is the excitation diffusion along the cavity axis, which in our experiments is very limited and thus ineligible as an origin of this effect [15]. The presence of an additional resonance constraint that imposes this broader spectral selectivity is more likely. Similar spectral observations have been reported for lasers based on a dye solution in a cuvette and a quantum-dot waveguide, and in both cases the lateral dimensions of the waveguide were considered responsible for this wavelength selectivity [16,17]. In other work, longitudinal mode grouping has been attributed to interference effects with substrate modes leading to a wavelength dependent depletion of the net-gain, but also linked to the linewidth of the homogeneously broadened laser transition [18,19]. Saturable absorber based resonances have been also considered as a linewidth-narrowing mechanism in fibre lasers [20].

In order to further investigate the effect in our system, we measured the dependence of the laser mode spacing with the length of the liquid waveguide. The effective optical cavity lengths were determined for both the single and multiple core lasers by Fourier transforming the emission spectra above threshold and are plotted in Fig. 5 as a function of the physical length of the fibres [21]. In contrast to the aforementioned reports, we found a linear dependence between the mode separation and the fibre length, indicating a primarily longitudinal origin of this spectral selectivity. The linearity is present in both the single capillary and photonic crystal fibre lasers at approximately the same rate. The observed linear dependence indicates that the spectral selectivity is due to a Vernier type of resonant effect [22]. Such a resonant selection mechanism requires the simultaneous presence of two (or more) cavities of different free spectral ranges. In such a 'superstructure', the spectral overlap of the resonances for each cavity can lead to a combined response of longer modal spacing.

In our experiment, the resonances are attributed to different order transverse modes. The transverse modes each have a distinctive group index and hence different effective cavity lengths; differences between these scale linearly with the fibre length, as observed [23]. Indicatively, for the single capillary, there are two bound core modes and we calculate their group indices to be 1.498 ± 0.002 and 1.506 ± 0.001 , giving $\Delta n_g = 0.008 \pm 0.002$. Considering only the core modes, the Vernier effect would increase the mode spacing [22] by a factor $n_g/\Delta n_g = 188 \pm 50$. This factor is slightly lower than the measured value of 309 ± 8 . We believe that this discrepancy would be further reduced by additionally considering the contribution of cladding modes, as recently proposed for planar structures [24].

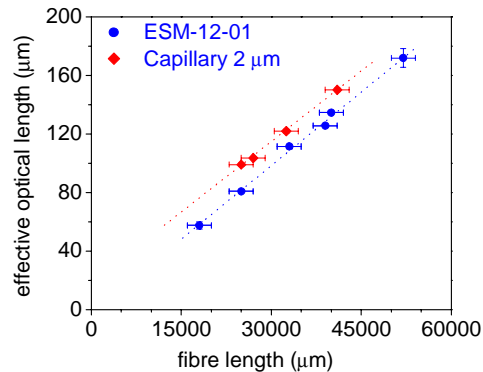


Fig. 5. The linear dependence of the cavity mode spacing with the fibre length for the capillary (\blacklozenge) and the photonic crystal fibre (\bullet).

5. Conclusions

In conclusion, we have demonstrated fluidic lasers based on optical fibres. Single and multi-channel fluidic lasers were realized in a single capillary and a photonic crystal fibre respectively. The lasers were simply and rapidly prepared by capillary action. Laser emission is observed when the fluidic fibres are longitudinally pumped above threshold and manifests itself as an axially emitted beam with a spectrum of a series of narrow peaks. The threshold and quantum efficiency were found to be dependent on the coupling efficiency of the pump light and the fibre length. The practicality of these optical sources is greatly enhanced by the possibility of tuning their emission by varying the chromophore density within the liquid core and an advantageous spectral selectivity was observed and attributed to a Vernier resonant mechanism between transverse electromagnetic modes of the fluidic laser. In addition, the photonic crystal fibre features a hexagonal array of approximately 50 fluidic lasers and could be used for the formation of an optical lattice. This type of laser is promising for opto-fluidic sensing and spectroscopy applications, and provides a convenient platform for rapid investigation of new types of liquids and organic dye molecules.

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