15. M. Tanyeri, R. Perron, and I. M. Kennedy, "Lasing droplets in a microfabricated channel," Opt. Lett. **32**(17), 2529–2531 (2007).
16. S. K. Y. Tang, Z. Li, A. R. Abate, J. J. Agresti, D. A. Weitz, D. Psaltis, and G. M. Whitesides, "A multi-color fast-switching microfluidic droplet dye laser," Lab Chip **9**(19), 2767–2771 (2009).
17. G. Aubry, Q. Kou, J. Soto-Velasco, C. Wang, S. Meance, J. J. He, and A. M. Haghiri-Gosnet, "A multicolor microfluidic droplet dye laser with single mode emission," Appl. Phys. Lett. **98**(11), 111111 (2011).
18. I. M. White, J. Gohring, Y. Sun, G. Yang, S. Lacey, and X. Fan, "Versatile waveguide-coupled opto-fluidic devices based on liquid core optical ring resonators," Appl. Phys. Lett. **91**(24), 241104 (2007).
19. J. D. Suter, Y. Sun, D. J. Howard, J. A. Viator, and X. Fan, "PDMS embedded opto-fluidic microring resonator lasers," Opt. Express **16**(14), 10248–10253 (2008).
20. Y. Sun, S. I. Shopova, C.-S. Wu, S. Arnold, and X. Fan, "Bioinspired optofluidic FRET lasers via DNA scaffolds," Proc. Natl. Acad. Sci. U.S.A. **107**(37), 16039–16042 (2010).
21. I. M. White, H. Oveys, and X. Fan, "Liquid-core optical ring-resonator sensors," Opt. Lett. **31**(9), 1319–1321 (2006).
22. S. I. Shopova, J. M. Cupps, P. Zhang, E. P. Henderson, S. Lacey, and X. Fan, "Opto-fluidic ring resonator lasers based on highly efficient resonant energy transfer," Opt. Express **15**(20), 12735–12742 (2007).
23. H. Song, M. R. Bringer, J. D. Tice, C. J. Gerdts, and R. F. Ismagilov, "Experimental test of scaling of mixing by chaotic advection in droplets moving through microfluidic channels," Appl. Phys. Lett. **83**(22), 4664–4666 (2003).
24. Y. Sun and X. Fan, "Highly Selective Single-Nucleotide Polymorphism Detection with Optofluidic Ring Resonator Lasers," in *CLEO/QELS*(Optical Society of America, Baltimore, MD, 2011), p. CWL6.

1. Introduction

Optofluidic lasers have emerged in the past few years as a promising technology for the development of micrototal analysis systems (μTAS), due to their compact size, compatibility with microfluidic components, and capability of providing integrated light sources [1–3]. To date, various optofluidic lasers with different cavities have been demonstrated, including Fabry-Pérot (FP) resonators [4,5], distributed feedback gratings [6–8], and optofluidic ring resonators (OFRRs) [9,10]. However, despite the nL-sized cavity volumes of those devices, the actual consumption of samples (such as gain media or biological/chemical analytes) is quite large, since a continuous flow of liquid is used. As a result, a large fraction of samples is wasted and the full potential of the optofluidic laser is not realized. Particularly, in the case where samples are very expensive or extremely low in abundance (such as in biological applications), utility of optofluidic lasers may be significantly hindered by the high cost associated with the sample consumption.

Microdroplets have been investigated for years for biological/chemical sample delivery, analysis, and manipulation [11–13]. They naturally form liquid reaction chambers of extremely low volumes (pL-nL). Moreover, using mature microfluidic technologies, microdroplets can be generated and stored in a large quantity, and can be manipulated (merged and split) individually on-demand. Introduction of microdroplets will render the optofluidic laser the highly desirable capabilities in sample delivery, manipulation, and controlled biological/chemical reaction.

Historically, the investigation of microdroplet lasers can be dated back in 1980's, when the free-falling or levitated droplets are used [14]. Recently, a few studies have been carried out on the microdroplet lasers inside a microfluidic channel using two immiscible liquids. For example, Tanyeri *et al.* obtained laser emission from a microdroplet of liquid dye surrounded by a low refractive index (RI) carrier fluid [15]. In this case, the droplet forms a ring resonator and supports the whispering gallery mode (WGM) that circulates along the droplet surface to provide the optical feedback. Similarly, through the T-junction microfluidics, rapid generation of the microdroplets and subsequent demonstration of laser emission are achieved, allowing for fast switching of the laser wavelength up to kilo-Hz [16]. However, in the above designs, a few serious drawbacks still exist. First, the droplet is required to have a higher RI than the carrier fluid and no laser emission can be achieved for the droplet of lower RI [15,16]. Such requirement significantly limits the selection of the droplet liquid and the carrier fluid. In particular, for most biological applications, samples need to be the aqueous phase (RI = 1.33). Finding an immiscible carrier fluid with an RI sufficiently lower than that of water would be

#153247 - $15.00 USD Received 22 Aug 2011; revised 11 Sep 2011; accepted 11 Sep 2011; published 22 Sep 2011

very challenging. Second, the laser emission from those droplets can be collected only through scattering, thus greatly impairing the light detection efficiency. Third, the lasing spectrum may fluctuate due to the droplet size variations. Very recently, laser emission from microdroplets in combination with an FP cavity is realized [17]. This design employs the metal-coated facets of two optical fibers placed inside the microfluidic channel to form an FP cavity. It is capable of handling droplets of any RI and of coupling the droplet laser emission into the optical fiber with high efficiency. Consistent lasing spectrum can be obtained, as the cavity does not depend on the droplet size. However, due to the low Q-factor of the FP cavity, such a device usually has a high lasing threshold.

Here, we investigate a novel nL-sized optofluidic laser scheme utilizing the capillary based OFRR that is connected to the upstream microdroplet generator (see Fig. 1). The OFRR consists of a thin-walled glass capillary. When the capillary is filled with solution, the circular cross section of the OFRR forms the ring resonator. The WGM circulates along the OFRR circumference and has the electric field present in both the liquid solution and the solid wall [18], thus providing the optical feedback for the gain medium flowing inside the capillary. The capillary based OFRR has a number of highly desirable features that overcome the aforementioned issues associated with other droplet lasers. First, it retains the advantage of the small sample volumes (~nL) of the droplet. The OFRR cavity volume is on the order of only 1-10 nL. A single droplet can fill the entire cavity easily. Second, the lasing of the dye solution can be achieved regardless of the solution RI, even in the presence of the high RI carrier fluid [10,18]. Third, the lasing emission can be out-coupled efficiently into a waveguiding component (such as a waveguide or tapered optical fiber) in contact with the OFRR [10,18,19]. Fourth, due to the high Q-factor of the OFRR [9,20], a much lower lasing threshold can be obtained. Fifth, since the microdroplets utilize a solid-state OFRR cavity, it is free from the size and shape variation, resulting in consistent lasing characteristics. Last, thanks to the nature of the capillary, the OFRR is highly compatible with the upstream and downstream microfluidics, thus providing convenient in-line sample analysis capability without interruption of the droplet flow. In this paper, we first fabricate and characterize the droplet OFRR laser, and then show the lasing emission can be achieved with the low RI solution and with a lasing threshold >6 times lower than the state-of-the-art. Finally, we demonstrate the capability of the OFRR droplet laser in sample manipulation/analysis and in lasing wavelength tuning by merging two droplets containing respectively the donor and acceptor dyes.

2. Experimental

The droplet generation system, schematically shown in Fig. 1(a), is adapted from the work by Trivedi *et al.* [13]. It consists of plastic tubings (Upchurch, P1476, inner diameter (ID) = 150 μm) and a T-junction (Upchurch, P-890). The hydrophilic phase consists of Rhodamine 6G (R6G) (Exciton Inc.) dissolved in methanol (RI = 1.33). The hydrophobic carrier fluid is silicone oil (Dow Corning 200 Fluid, RI = 1.53) with viscosity of 10 centistokes. The methanol phase and the carrier fluid are injected by the syringe pumps (KD Scientific) at a respective flow rate of Q_d = 3 μL/min, and Q_o = 10 μL/min. When continuous liquid flows meet at the T-junction, a series of methanol droplets separated by immiscible silicone oil form at a rate of 0.4 Hz. Each droplet is approximately 125 nL in volume and fills the entire cross section of the tubing. Figure 1(b) illustrates the droplet generation and mixing system, which consists of a second T-junction downstream. R6G and LDS 722 (Exciton Inc.), both dissolved in methanol, are used as Dye 1 and Dye 2, respectively, with a flow rate of 3 μL/min for each. As both R6G and LDS 722 have the same solvent (methanol), they can easily mix together and form a larger droplet. Figure 1(c) shows, as an example, the picture of a series of R6G droplets stored inside a plastic tubing.

#153247 - $15.00 USD Received 22 Aug 2011; revised 11 Sep 2011; accepted 11 Sep 2011; published 22 Sep 2011

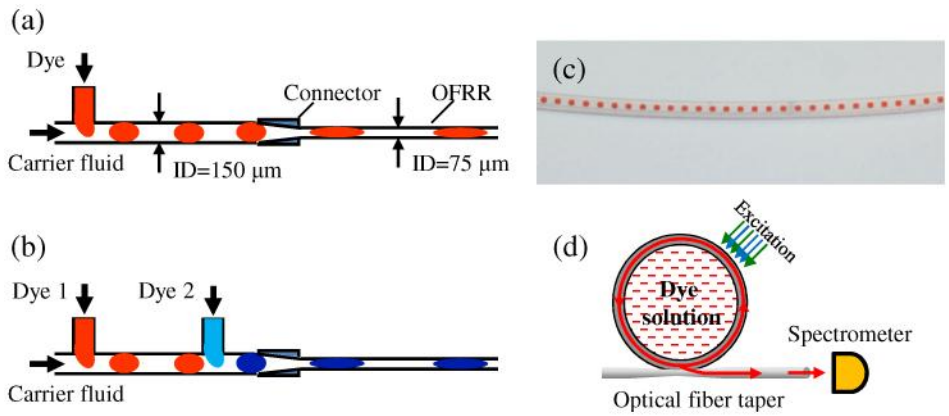

Fig. 1. (a) Schematic of the droplet generating system consisting of a T-junction. The carrier fluid is silicone oil. The dye is dissolved in methanol. The plastic tubing and the OFRR have an ID of 150 μm and 75 μm, respectively. (b) Schematic of the droplet mixing system consisting of a second T-junction. (c) Picture of R6G droplet series stored inside a plastic tubing. (d) Cross-sectional view of the OFRR, the laser excitation, and out-coupling. A tapered optical fiber is placed in contact with the OFRR to couple the lasing emission into a spectrometer. Note that in the illustration, the dye solution fills the entire cross section of the OFRR. The oil that may be present between the dye solution and the wall is not shown.

We fabricate the capillary OFRR by the conventional drawing method [9,21]. A fused silica capillary with the original ID of 536 μm and the wall thickness of 40 μm is mechanically pulled under CO_2 laser irradiation. The resultant OFRR is approximately 3 cm long, 75 μm in ID, and about 7 μm in wall thickness. It can be connected easily to the outlet of the plastic tubing, as illustrated in Figs. 1(a) and (b). Figure 1(d) depicts the cross-sectional view of the OFRR. Inside the OFRR, the droplet becomes elongated (due to the small inner diameter in comparison with the plastic tubing) and fills the entire cross section of the OFRR. An optical parametric oscillator (Continuum, 532 nm, 5 ns pulse width, 20 Hz repetition rate) is focused on the side of the OFRR. An optical fiber taper is in contact with the OFRR and couple the laser emission efficiently into a spectrometer (Horiba iHR550, resolution of 0.06 nm) at the distal end.

3. Results and discussion

Since the OFRR wall (fused silica) has a higher RI than the dye solution (methanol), the majority of the WGM resides inside the wall and only the evanescent field of the WGM interacts with the liquid gain medium. Thus, it is imperative that the high RI layer of the carrier fluid (silicon oil) sandwiched between the dye solution and the OFRR wall be sufficiently thin (less than several micrometers) so that the strong WGM-dye interaction can be sustained. No lasing emission is expected to occur when the droplet is surrounded by a thick layer of higher RI oil (the reverse RI contrast case) or even slightly lower RI oil (the low RI contrast case) [15,16]. Since the OFRR inner surface is hydrophilic, we expect that, in the presence of methanol, hydrophobic oil should be expelled away from the OFRR inner surface. As a result, the dye solution should fill nearly the entire cross section of the OFRR (see Fig. 1(d)). To test this, we carry out two experiments. The first experiment is shown in Fig. 2(a). When the 1 mM R6G/methanol droplet in the carrier fluid flows through the OFRR, under the external excitation the typical multi-mode laser emission ranging from 550 nm to 580 nm emerges, which can be out-coupled by a tapered optical fiber in contact with the OFRR (see the inset of Fig. 2(a)). The lasing threshold, based on the maximum intensity of the lasing signal as a function of the pump energy, is measured to be approximately 1.54 μJ/mm^2. The results presented above show that using our OFRR system the droplet lasing can be achieved

#153247 - $15.00 USD Received 22 Aug 2011; revised 11 Sep 2011; accepted 11 Sep 2011; published 22 Sep 2011

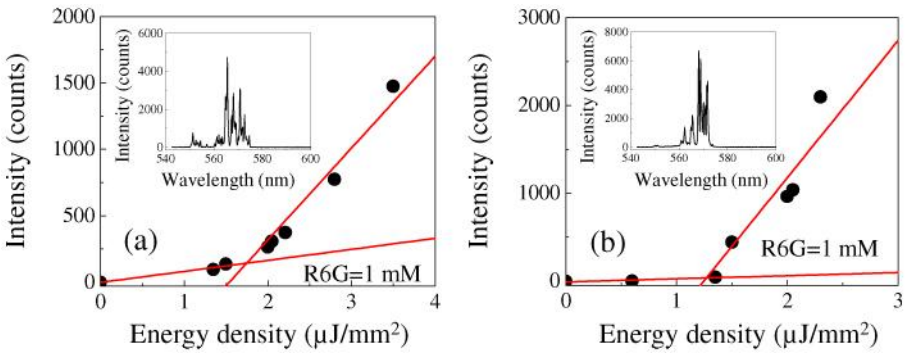

Fig. 2. (a) Lasing intensity from a microdroplet flowing through the OFRR as a function of pump energy density. The droplet is formed by 1 mM R6G dye dissolved in methanol immersed in silicone oil. The lasing threshold is approximately 1.54 μJ/mm^2. Inset shows the lasing spectrum at the pump energy density of 6.5 μJ/mm^2. (b) Lasing intensity from a conventional continuous flow OFRR laser in the absence of carrier fluid. The dye solution is the same as in (a). The threshold is approximately 1.25 μJ/mm^2. Inset shows the lasing spectrum at the pump energy density of 6.2 μJ/mm^2.

even in the presence of high RI oil and with a much lower lasing threshold (>6 times), which represent a significant advance over the droplet laser systems previously reported [15–17].

As a control experiment, R6G in methanol alone is flowed through the same OFRR continuously. The lasing characteristics of the conventional OFRR dye laser are presented in Fig. 2(b). It has a similar lasing threshold of 1.25 μJ/mm^2, and shows nearly the same lasing spectrum as in our OFRR droplet dye laser. Comparison between the results in Figs. 2(a) and (b) suggests that, while the exact thickness of the oil layer between the dye solution and the OFRR wall is unknown, it must be optically thin (less than one wavelength) and the existence of the oil layer (if any) does not alter noticeably the interaction between the WGM and the dye. Consequently, the OFRR droplet laser should be similar to the continuous flow OFRR laser and a number of advantageous characteristics of the OFRR, such as lasing emission with low RI solution, the high Q-factor (>10^7 [9,20]), and the high out-coupling efficiency (>50% [19]), can be kept.

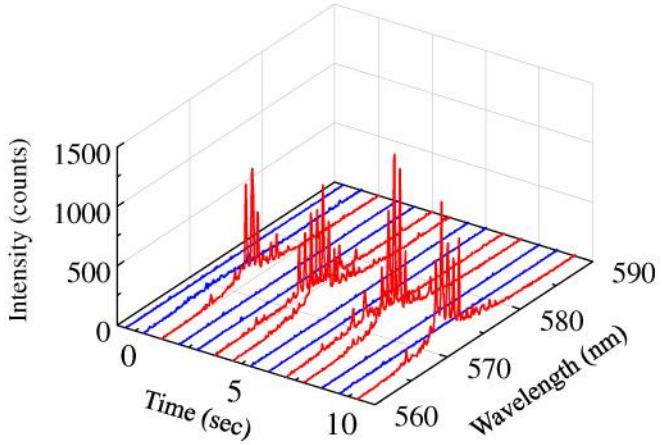

Fig. 3. Lasing spectra from the microdroplet laser as a function of time. As the R6G/methanol droplet flows through the capillary OFRR, the laser shows pulsed lasing signal at a frequency of approximately 0.4 Hz (red curves). Note that the signals from the carrier fluid gap between different droplets are zero (blue curves) and that the variations in the laser emission are due mainly to the power variations of the pump laser and the imperfect synchronization between the pump laser and the droplet generation.

#153247 - $15.00 USD Received 22 Aug 2011; revised 11 Sep 2011; accepted 11 Sep 2011; published 22 Sep 2011

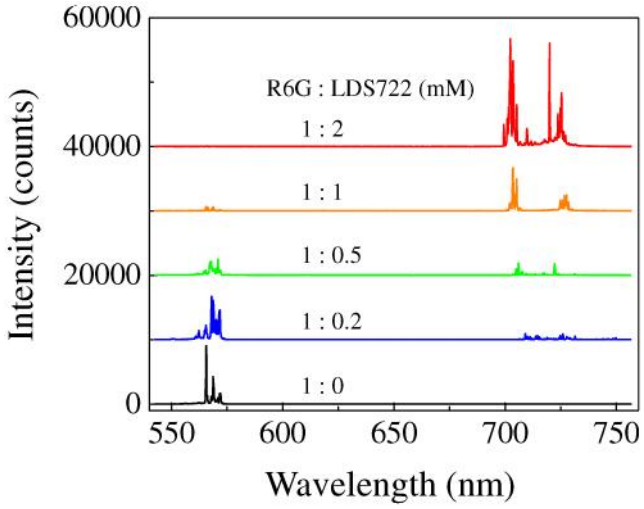

Fig. 4. FRET lasing spectra from merged droplets with various donor/acceptor concentrations. R6G and LDS722 are used as the donor and the acceptor, respectively. The concentration of the donor is fixed at 1 mM and the concentration of the acceptor varies from 0 to 2 mM as indicated. Curves are vertically shifted for clarity. The pump energy density is 14.6 μJ/mm^2 for all curves. As the concentration of the acceptor increases, the donor lasing signal around 570 nm diminishes. Meanwhile, the acceptor lasing signal ranging from 700 nm to 740 nm arises.

As the pump laser is focused only on a small portion of the OFRR, the device should exhibit the lasing emission only when the droplet passes through the focusing point. Figure 3 plots the lasing signal from the OFRR droplet laser as a function of time, when 1 mM R6G droplet and the carrier fluid are flowed through the OFRR. With the flow rate of 13 μL/min (Q_d = 3 μL/min, Q_o = 10 μL/min), the lasing signal at the frequency of 0.4 Hz is obtained. It is clearly seen that the lasing signal is observed only from the dye droplet and the signals from the carrier fluid gap between two adjacent droplets are virtually zero. Therefore, it is possible to distinguish two different droplets by the carrier fluid gap in our system, which is essential for individually analyzing and manipulating the droplets. Note that although the tuning frequency reported here is much lower than previous studies [16, 17], when needed, it can be increased drastically by using different microfluidic channel geometries, flow rates, and pump light sources.

As discussed earlier, microdroplets serve as an excellent chemical and biological reaction chamber and can be manipulated conveniently. The droplet and the OFRR in combination will provide powerful capability in sample generation, delivery, processing, and in-line analysis. Here, we demonstrate the rapid sample mixing of donor dye (R6G) and acceptor dye (LDS722) in two different droplets and subsequently achieve the tuning of the lasing wavelength through the fluorescence resonance energy transfer (FRET) between the donor and the acceptor. The experimental setup is described in Fig. 1(b) where two different dyes are merged into a single microdroplet by two T-junctions. During the experiment, the LDS722 concentration is varied from 0 mM to 2 mM, whereas the R6G concentration remains at 1 mM. Since the Förster

distance between R6G and LDS722 is about 6.2 nm [22], significant energy transfer is expected to occur when LDS722 concentration is above 1 mM. Figure 4 shows the lasing spectra from merged droplets with various R6G/LDS722 ratios. In the absence of LDS722, the droplet OFRR laser shows a typical R6G lasing signal in the 550 to 580 nm range. With the increased LDS722 concentration, due to the FRET process, the lasing emission in the LDS722 range (710 – 740 nm) starts to emerge whereas the R6G lasing emission decreases. With 2 mM LDS722 concentration, the donor emission is completely suppressed and strong

#153247 - $15.00 USD Received 22 Aug 2011; revised 11 Sep 2011; accepted 11 Sep 2011; published 22 Sep 2011

lasing emission from LDS722 is observed. While in the above experiment, only 150 nm wavelength tuning is demonstrated, larger wavelength tuning is certainly possible by implementing cascade energy transfer processes where three or more dyes can be mixed. Note that, due to the nature of the droplet, sample mixing is rapid [23], as evidenced by the decrease (or disappearance) of the donor lasing emission and the concomitant emergence of the acceptor lasing emission within a few seconds after mixing.

4. Conclusion

In summary, we demonstrate a novel microfluidic laser that combines microdroplets and the capillary based optofluidic ring resonator with a high Q-factor. This device has nL-sized sample and cavity volume and is versatile in handling solutions of various RIs. In particular, the lasing emission can be obtained even in the presence of high RI carrier fluid. A lasing threshold of 1.54 $\mu J/mm^2$ is achieved, much lower than the state-of-the-art. By mixing two droplets containing respectively the donor dye and acceptor dye, we not only demonstrate the OFRR droplet laser's capability in tuning or switching of the lasing wavelength, but also open the door to the potential biosensing applications of the OFRR droplet laser [20, 24].

Acknowledgments

W. Lee and Y. Luo contribute equally to this paper. The work is supported by the NSF under ECCS 1045621 and CBET 1037097. Y. Luo is supported by NSFC under grant 61008057 and 60877044, by Fundamental Research Funds for Central Universities under grant 21609507, and by Shenzhen Key Laboratory of Sensor Technology Open Project under grant SST201012.

#153247 - $15.00 USD Received 22 Aug 2011; revised 11 Sep 2011; accepted 11 Sep 2011; published 22 Sep 2011