Highly Reproducible, Isotropic Optofluidic Laser Based on Hollow Optical Fiber

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(Invited Paper)

***Abstract*—We report a reproducible optofluidic laser (OFL) for multichannel biochemical sensing. A hollow optical fiber (HOF) serves as both microring resonator for lasing and microfluidic channel. The lasing mechanism is analyzed in spectral domain. Thanks to the precise control of the fiber geometry and the rotational symmetry, the HOF-OFL emission is uniformly distributed in the angular direction ($\sigma = 0.6\%$) and can be employed conveniently for disposable or arrayed use. An array of ten OFLs is demonstrated with good reproducibility ($\sigma = 3.9\%$) in laser threshold. The arrayed sensing capability of the OFL is also investigated. The HOF-OFLs are highly reproducible, easy to integrate in chip-scale, and have great potential for biochemical detection.**

***Index Terms*—Optofluidic laser, hollow optical fiber, whispering gallery mode, biochemical sensor.**

I. Introduction

OPTOFLUIDIC laser (OFL) integrates optofluidics [1]–[6], microcavity [7]–[9] and the gain medium to generate laser emission and has been developed as a powerful tool in bioanalysis [10]–[14]. Ultrahigh sensitivity for bioassay [15]–[17] has been achieved due to the enhanced light-matter interaction in optical microcavities of OFLs. However, achieving high reproducibility and high throughput arrays of OFLs remain challenging.

Initial strategy for optical feedback in OFLs is based on microstructured resonators embedded in microfluidic chips, including on-chip Fabry-Perot (FP) cavities [18], [19], distributed feedback microstructures [20]–[22], microrings [23], microgoblets [24], [25] or droplets [26]–[28]. An external FP cavity, using two reflective mirrors sandwiching the microfluidic channel, is an excellent candidate to enhance the sensitivity of OFL sensors. Recently Wang *et al.* [29] designed an optofluidic laser array based on micro wells on FP cavity. However, the microwells on the mirror cannot be employed as isolated sampling channels. FP cavity is also bulky, which makes it hard to integrate with multi-channels. Sandwiching an array of capillaries between two mirrors may introduce misalignment of the cavity [15]. Thin-walled glass capillaries are also outstanding candidates for providing both a high Q factor microring resonator and an integrated microfluidic channel [30], [31]. However, the fabrication protocol results in high performance but stand-alone capillary device and is not suitable for mass production of highly reproducible micro resonators.

Fiber optofluidic lasers that utilize optical fibers as optical micro resonators were recently developed [32], [33]. The outer boundary of any round optical fibers can be employed as the microring for optical resonance [33]. Photonic bandgap in optical fiber [36] and microring in microstructured optical fiber [34], [35] can play the same role. Optical fibers were designed for optical fiber communications at the beginning. The fabrication process has been commercially optimized for mass production with low cost as well as very high precision of the fiber geometry in order to control their transmission properties. Gong *et al*. developed a reproducible fiber optofluidic laser based on the low cost, mass-produced twin-hole microstructured fiber (TMF). The TMF was axisymmetric so that the laser emission was angular-dependent, which made it difficult to achieve a high reproducibility.

In this paper, we report a reproducible fiber OFL array using hollow optical fibers (HOFs). The HOF is fabricated by a commercial fiber draw tower that guarantees the highly precise control of its diameter and thickness. The rotational symmetry of HOF enables an angular independent laser emission with high repeatability. We use the reproducible fiber optofluidic lasers to construct an array of ten channels with very good consistency.

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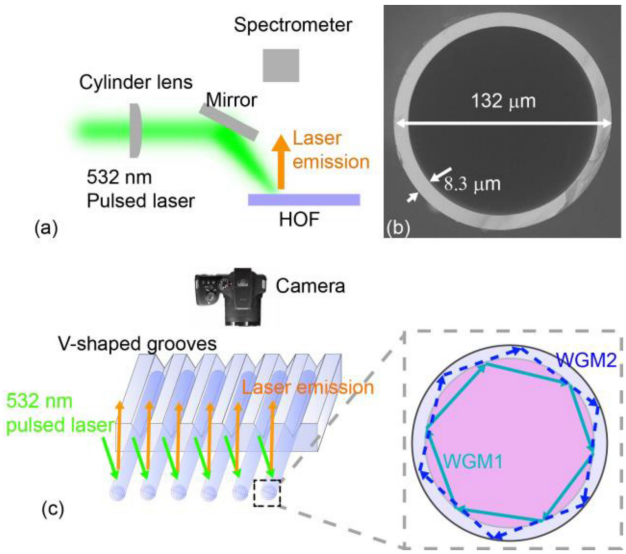

Fig. 1. (a) Schematic diagram of the experimental setup for HOF based fiber optofluidic laser. (b) SEM image of the cross section of HOF. Dimensions are also given. (c) The schematic diagram of the HOF-OFL array. Inset, illustration of the lasing mechanism.

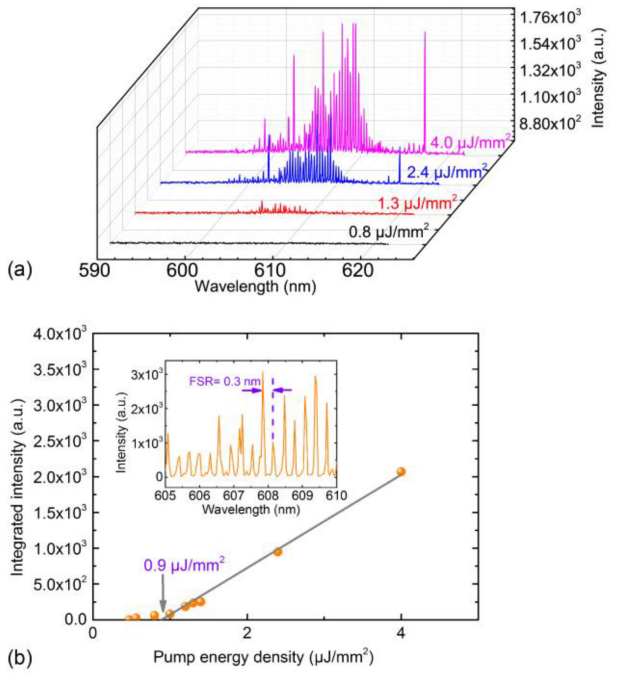

Fig. 2. (a) Laser spectra of fiber OFL with different pump energy density. (b) The spectrally integrated intensity as a function of pump energy density. The intensity is integrated between 591 nm and 619 nm to include all the laser peaks. Inset, the enlarged spectrum with a pump density of 6.0 μJ/mm^2.

The performance of fiber OFL is characterized by both spectroscopic and imaging techniques. The arrayed sensing capability of the fiber OFL is also investigated by concentration detection of dye in different channels with a good linearity. The proposed OFL array offers a new way to conduct high throughput bioassay.

II. Experimental Setup

Fig. 1(a) shows the experimental setup for HOF based fiber OFL. The 532 nm pulsed laser (Continuum, 532 nm, 5 ns pulse width, 20 Hz repetition rate) was expanded and focused by a cylindrical lens, forming a strip of 9.0 mm $\times$ 0.1 mm. The emission from fiber OFL was detected by a commercial camera (Nikon, D3400, 24 million pixels). The laser spectra were measured using a spectrometer (Andor, SR500IA). A long pass filter (cutting wavelength, 550 nm) and a neutral density filter (optical density, O.D. $= 2$) were used to filter out the pump and to attenuate the fluorescence.

The HOF was fabricated on the fiber drawing tower in UNSW. It was drawn from a quartz tube (Heraeus, F300, 25 $\times$ 22 mm) mounted on the feeder of fiber draw tower. The tube was slowly fed into the graphite furnace heated up to $\geq$1880 °C and melted into fiber with capstan drawing. The dimension of the HOF is controlled by the drawing temperature T, tube feeding rate V_f, fiber drawing rate V_d as well as the internal pressure P applied by the flow rate of purged high purity nitrogen. In the fabrication, $T = 1885$ °C, $V_{\rm f} = 0.5$ mm/min, $V_{\rm d} = 20.9$ m/min and $P = 2.7$ mbar. HOF with an outer diameter of 132 μm and a thickness of 8.3 μm (Fig. 1(b)) was used in this experiment.

As shown in Fig. 1(c), the fiber OFL array was constructed by arranging HOFs in parallel in arrayed V-shaped grooves. Right before use, the polymer coating of the HOF was removed by sonicating in acetone for 5 min. The fiber was further cleaned in plasma (140 W) for 10 min to make its surface hydrophilic. For practical application, the plasma treated fiber can be stored by immersing it in acetone so that the fiber can be automatically dried in air due to acetone evaporation and then ready for test. Then, the liquid gain material, i.e., 2 mM of R6G in quinoline ($n = 1.63$) was filled into the HOF by the capillary effect. The HOF supports whispering gallery modes (WGMs) that interacts with the liquid gain medium. The lasing mechanism is schematically illustrated in the inset of Fig. 1(c) and will be discussed in detail together with the spectral characteristics of fiber OFL.

III. Results and Discussion

The laser threshold of the fiber OFL was characterized at first. The emission spectra were recorded as the pump energy density increases. As shown in Fig. 2(a), laser peaks appear around the threshold and then the intensity increases sharply above the threshold, indicating lasing. The integrated intensity as a function of pump energy density, i.e., the threshold curve, is given in Fig. 2(b), showing a threshold of 0.9 μJ/mm^2. The low threshold is due to the high Q-factor of optical resonator in the HOF and the good performance of the laser dye. The inset of Fig. 2(b) shows an enlargement of laser spectrum with a pump density of 6.0 μJ/mm^2. The linewidth of the HOF-OFL was around 0.06 nm. The free spectral range (FSR) was experimentally measured to be 0.3 nm by the spectra.

Due to the higher refractive index of quinoline than silica, lasing mode was excited in the inner surface (WGM1, noted in orange in the inset of Fig. 1(c)). However, to our surprise, the theoretical FSR of WGM1, calculated to be 0.6 nm using

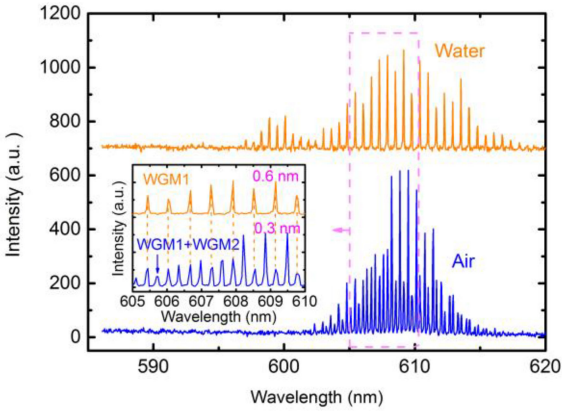

Fig. 3. Comparison of the laser spectra of fiber OFL immersed in water and air. Inset, the enlargement of spectra.

$\mathrm{FSR} = \lambda^2/(n\pi d)$, is two times larger than the measured FSR, 0.3 nm. In the calculation, $\lambda = 608$ nm is the lasing wavelength. $n_1 = 1.63$ is the refractive index of liquid gain material in the core. $d_1 = 115.4$ μm is the inner diameter of HOF. Then we presume that there was another lasing mode, WGM2, which is the higher order WGM supported by the outer surface of HOF. The FSR of WGM2 (blue in the inset of Fig. 1(c)) is very close to that of WGM1, 0.6 nm, with $n_2 = 1.45$ for the refractive index of silica and $d_2 = 132$ μm for the outer diameter of HOF. Because of the thin wall (~ 8.3 μm), we inferred that the WGM2 can also interact with the liquid gain in the core. These sets of WGMs, both having FSR about 0.6 nm, co-exist in the cross section of HOF and provide optical feedback for lasing, resulting in a final FSR of 0.3 nm.

In order to confirm this hypothesis, we designed an experiment by immersing the same fiber OFL in water and in air to change the surrounding index. The relative index difference between silica and water is small so that WGM2 vanished (Fig. 3). In this case, a FSR of 0.6 nm was observed, in good agreement with calculation. When the fiber OFL was put in air, two sets of lasing WGMs co-existed and a FSR of 0.3 nm was observed. The peak positions of WGM1 in both cases was highly consistent with each other. These observations not only verify the lasing principle of HOF-OFL, but provide a straightforward sensing mechanism based on the extinction of resonant peaks.

Another hypothesis in this work is that the HOF based OFL can achieve isotropic laser emission, in order to avoid the angular adjustment problem suffered from the twin-hole microstructured fiber based OFL [30]. Thus, we experimentally demonstrated the merit of angular independent HOF laser using the same setup as in our previous work. The laser spectra were recorded by rotating the HOF with a step of 30° (Fig. 4(a)). The emission spectra at various orientations are almost the same. The spectrally integrated intensity is plotted in polar coordinates (Fig. 4(b)). As expected, the fiber OFL shows a very uniform intensity distribution in the angular domain, with a standard deviation of 0.6%. The angular independence of laser emission is dominated by the rotational symmetry of the HOF geometry.

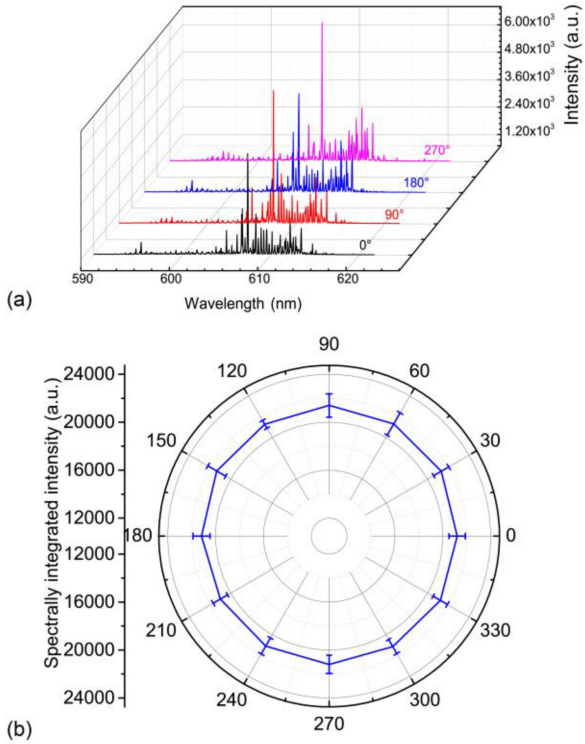

Fig. 4. (a) The laser spectra at various angles. (b) The integrated intensity as a function of orientation angle in polar coordinate system. The pump density was 10 μJ/mm^2. Error bars are obtained based on triplicate measurements.

An array of ten fiber OFLs was constructed by integrating the fiber OFLs on the V-type grooves. The good consistency of the laser emission from ten channels was characterized and confirmed by imaging (inset of Fig. 5(a)). 2 mM of R6G in quinoline was used. At each pump density, the laser intensity was uniform among different channels. With an increase of pump density, the brightness of the fiber OFLs increased synchronously. The threshold of ten fiber OFLs were measured at the same time by imaging. The laser emission of each channel was normalized by its pump intensity in order to eliminate the spatial fluctuations of pump. The normalized intensity as a function of pump density is shown in Fig. 5(a). An inflection was observed to indicate an average threshold of about 0.8 μJ/mm^2 that was close to the spectral observations. The slight difference in this experiment with Fig. 2 is induced by the deviation in solution preparing in two measurements.

The threshold of ten channels are given separately in Fig. 5(b), showing a good consistency with a statistical deviation of $\sigma = 3.9\%$. The repeatability was improved compared with the fiber optofluidic laser based on the twin-hole microstructured fiber [30]. This residual deviation was mainly derived from the inhomogeneity in the liquid gain material, and the position of HOFs, which can be further optimized.

The parallel sensing capability of the fiber OFL array was demonstrated by detecting the concentration of R6G through six channels. The calibration curve was measured by withdrawing R6G in quinolone with 2-fold serially diluted concentrations (0.125 mM, 0.25 mM, 0.5 mM, 1 mM, 2 mM and 4 mM) into

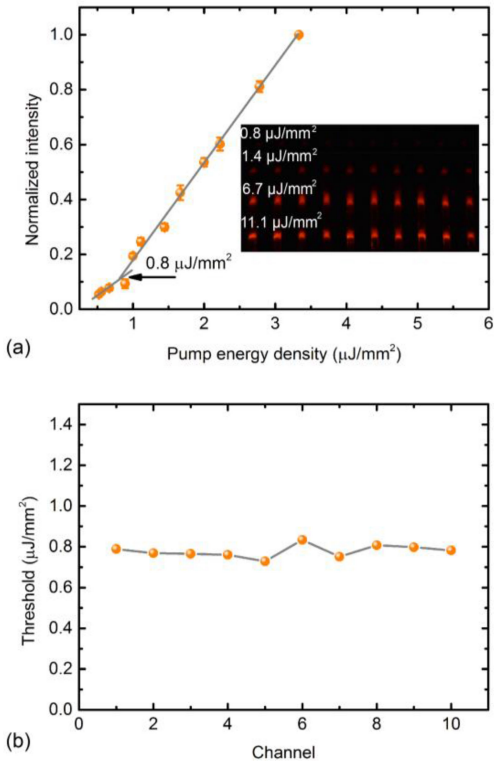

Fig. 5. (a) Normalized intensity as a function of pump density. The data are obtained by averaging the normalized intensity of ten channels, with error bars corresponds to the intensity deviation of each channel. Inset, The image of the fiber OFL array at various pump energy density. (b) The laser threshold of each channel. The deviation is statistically determined to be 3.9%.

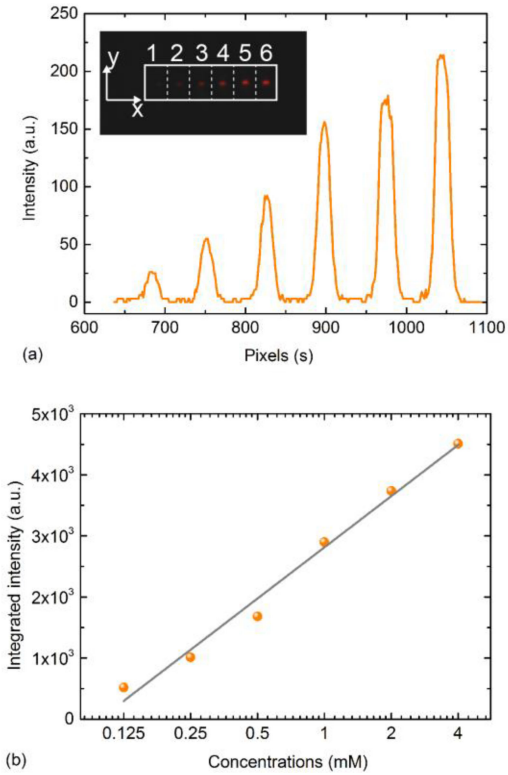

Fig. 6. (a) Intensity distribution of six parallel optofluidic lasers along the x-axis. The pumping density was 10 μJ/mm^2. Inset, the corresponding photo that the data in (a) was obtained. (b) Integrated intensity versus concentration.

each channel. The image of the lasing fiber OFL array is given in the inset of Fig. 6(a). In each case, the spectra were measured in order to confirm the intensity from camera was from laser emission. Obviously, the brightness of the lasing channels varies with the concentration of liquid material in the HOF. Fig. 6(a) shows the intensity distribution along x-axis obtained from the image. Six intensity peaks were observed, corresponding to six lasing channels. The laser intensity, calculated by the sum of pixels with each channel, increases with a good linearity of 98% as a function of the R6G concentration (Fig. 6(b)).

For biochemical sensing, further improvements could be made in three aspects. First, over 100 channels can be integrated on a chip with a size of 3 cm $\times$ 3 cm (with a separation of 100 μm between each channel). High-throughput sensing can be achieved by imaging the laser array simultaneously. The high-throughput sensing chip can be utilized for biological diagnostics [37]–[39], DNA sequencing [40], [41] and drug screening [42], [43]. Second, a disposable biochemical sensor can be realized by using the HOF-OFL as a low cost, single-use element. The disposable sensors [33], [44] are of great importance in biochemical detection for its intrinsic safety, ease of use and mass production with low cast. Due to the precise geometry control by the fiber draw tower, the properties of the HOF and thus the HOF-OFLs are highly reproducible. Third, reducing the thickness of the HOF may further enhance the laser-matter interaction and improve the sensing performance.

IV. Conclusion

We have developed a fiber optofluidic laser with isotropic and reproducible emission. The lasing mechanism was revealed by analyzing the spectral characteristics. An array of fiber OFLs was constructed and arrayed sensing of R6G concentration was demonstrated by imaging. The proposed fiber optofluidic laser is promising for wide use of sensitive biochemical detection.

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