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# Monolithic optofluidic ring resonator lasers created by femtosecond laser nanofabrication

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### Lab on a Chip

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#### <sup>25</sup> Introduction

The fusion of optics and fluidics has given birth to the flourishing field of optofluidics, in which optical elements and fluidic frameworks are amalgamated on a common substrate to synergistically deliver a broad range of functionalities and high performances.<sup>1–3</sup> Optofluidic lasers are among the branches of optofluidics that promise solutions to the presently insurmountable problems.<sup>1,4,5</sup> They have justified themselves to be outstanding nominees for on-chip tunable coherent light sources,<sup>6–8</sup> bio-controlled lasers,<sup>9</sup> and sensitively analyzing biomolecules.<sup>5,10–14</sup> The earlier debuts of optofluidic lasers<sup>15–18</sup> with notable features have sparked a resurgence of research interest in exploring various optical cavity architectures, device materials, fluidic gain media, and fabrication technologies to promote broader applications.

From a manufacturing point of view, a successful deployment of optofluidic lasers as practical sub-modules in lab-ona-chip (LOC) design requires a simple process that can reliably produce the desirable apparatuses while providing a great degree of design freedom. The ideal fabrication process should be compatible with rapid prototyping and relatively low cost. From a device application standpoint, a platform that has high mechanical strength, broad chemical compatibility, and good thermal stability is highly favorable.

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## Monolithic optofluidic ring resonator lasers created by femtosecond laser nanofabrication<sup>†</sup>

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We designed, fabricated, and characterized a monolithically integrated optofluidic ring resonator laser that is mechanically, thermally, and chemically robust. The entire device, including the ring resonator channel and sample delivery microfluidics, was created in a block of fused-silica glass using a 3-dimensional femtosecond laser writing process. The gain medium, composed of Rhodamine 6G (R6G) dissolved in quinoline, was flowed through the ring resonator. Lasing was achieved at a pump threshold of approximately 15  $\mu$ J mm<sup>-2</sup>. Detailed analysis shows that the *Q*-factor of the optofluidic ring resonator is  $3.3 \times 10^4$ , which is limited by both solvent absorption and scattering loss. In particular, a *Q*-factor resulting from the scattering loss can be as high as  $4.2 \times 10^4$ , suggesting the feasibility of using a femtosecond laser to create high quality optical cavities.

> In the last decade, several research groups have demon-25 strated Bragg gratings based optofluidic lasers, such as distributed feedback (DFB) laser on a chip.<sup>8,19-22</sup> These devices exhibit certain compelling characteristics, such as low pump thresholds, tunable wavelengths, and single mode of operations. However, the manufacturing of these devices is not 30 straightforward and rather expensive as electron-beam (ebeam) lithography is used for patterning the gratings in conjunction with additional multilayer processing steps. Microfluidic dye lasers that integrate a Fabry-Pérot cavity with two fiber-based mirrors have shown single mode emissions and 35 digital programmability.<sup>6,17,23,24</sup> Nevertheless, the coating process of the Fabry-Pérot cavity is an arduous process, which is difficult to implement in a monolithically integrated system. Furthermore, this type of optofluidic laser entails a multipart assembly process, which degrades the compactness 40 and robustness of the device. Fabry-Pérot microcavity fluidic dye lasers with metallic mirrors have also been reported previously.16,25 While these devices can be expediently integrated with other microfluidic systems without adding further processing steps, the overall fabrication and assembly processes 45 either are relatively long and complicated,<sup>16</sup> or have to be accomplished manually by adding spacer beads.<sup>25</sup> Furthermore, the polymer bonding process may raise issues associated with imprecisions and device sturdiness. More recently, an optofluidic laser based on a two-dimensional photonic 50 crystal<sup>26</sup> was demonstrated. This device was patterned by a laser interference lithography<sup>27</sup> technique. It reveals enhanced spontaneous emissions and a low laser threshold of  $9 \times 10^3$  nJ cm<sup>-2</sup>. Although the laser interference lithography seems to have evaded the necessity of employing expensive and tedious 55 e-beam lithography process, it demands very customized

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and laborious optical alignments to pattern each device with particular dimensions. This patterning method may be suitable to accurately realize a large array of devices with high resolutions and optical sub-wavelength dimensions for various laboratory experiments, but it is very challenging to adopt this process to manufacture and integrate practical devices as this type of lithography technique is incapable of patterning arbitrary shapes.

The ring-shaped optical cavities that operate based on the 10 whispering gallery modes (WGM) are known to have relatively high quality factors (O-factors) even when they are fabricated on a chip.<sup>28-30</sup> They have a simple form factor, miniscule size, ability to support a broad range of lasing emission wavelengths, and do not demand space consuming arrays of identical nanoscale structures which require e-beam patterning 15 such as Bragg grating based devices. Optofluidic ring resonator dye lasers that have compact sizes and single mode of operations have been previously presented.31-33 However, they were built on materials (such as polydimethylsiloxane) with very modest flexural rigidity and limited chemical com-20 patibility, making it challenging to operate these devices under practical conditions. More importantly, the planar lithography method that is used to pattern those devices makes it extremely difficult to deliver liquid gain medium into the ring resonator without degrading its optical perfor-25mance, including its Q-factor.

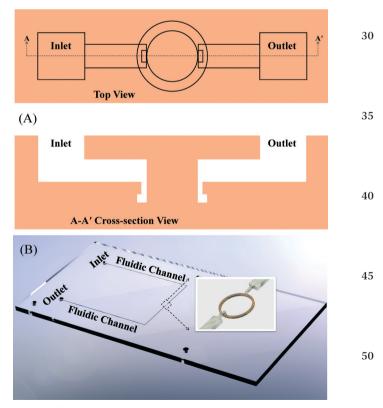
The present work focuses on the design, fabrication, and characterization of a monolithically integrated optofluidic ring resonator laser that meets the requirements put forth earlier. We fabricated the entire device, including the ring resonator and the microfluidics, with a relatively simple femtosecond laser direct-writing, followed by a wet chemical etching, and a final thermal bonding. This integration concept is appealing for many lab-on-a-chip applications: it has manifold advantages, such as reproducible fabrication, straightforward assembly, and does not require postfabrication processes such as dicing and packaging. These are all originated from the fact that we are engaged to a single piece of material.34 In addition, the 3-D fluidic design enabled by the femtosecond laser fabrication allows to deliver the liquid gain medium to the ring resonator without affecting significantly its optical performance or degrading the cavity Q-factor as discussed later. Note that previously Cheng et al. demonstrated dye lasers with four micro-mirrors machined using a femtosecond laser.<sup>15</sup> Nonetheless, the cavity occupied a relatively large area and has a low Q-factor due to the stringent requirements for the micro-mirror's surfaces imposed by the device configuration.

Our optofluidic ring resonator laser was characterized using Rhodamine 6G (R6G) dye dissolved in a quinoline solution (n = 1.62) as the gain medium and optically pumped by nanosecond pulses generated by a 532 nm optical parametric oscillator (OPO). The laser emission emerged at 575 nm wavelength with a relatively low pump threshold fluence of ~15 µJ mm<sup>-2</sup> per pulse. We observed stable laser emissions for different dye concentrations. Furthermore, by analyzing the lasing threshold, a bare cavity Q-factor of  $3.3 \times 10^4$  is deduced. Lasing with R6G in chlorobenzene (n = 1.524) was also demonstrated.

#### Design and fabrication

The optofluidic ring resonator laser in this work predominantly comprised of a ring-shaped channel to define the liquid cavity and connecting microfluidic channels used to flow the gain medium solution. All components that build the laser were monolithically incorporated in a piece of fusedsilica glass with 3-D freedom. The top and cross-sectional views of the device are illustrated in Fig. 1. The ring cavity was designed to have inner and outer radii and depth of 150  $\mu$ m, 170  $\mu$ m, and 40  $\mu$ m, respectively.

Owing to the capability of the femtosecond laser to tailor the microfluidic channels in three dimensions, we are able to introduce fluidic channels to transport the gain medium from the *inner* circumference of the ring where virtually no WGM is present (Fig. 1(A)). This design concept, in contrast to the ring cavity in ref. 32 where the liquid waveguide channel has to merge with the liquid ring resonator channel for sample delivery, eliminates the possible source of *Q*-factor degradation.<sup>35</sup> In addition, the vertical fluidic channels with direct contact to the ring are built to have smaller crosssectional dimensions (10  $\mu$ m × 20  $\mu$ m) than the ring to serve



**Fig. 1** (A) Top and cross-sectional view of the monolithic optofluidic ring resonator. (B) CAD design of the optofluidic system. Inset shows the details of the ring resonator and the fluidics that delivers the gain solution into the ring resonator.

The machining process was started with two fused silica 5 substrates with identical dimensions of 1 inch  $\times$  2 inch. The optofluidic components were initially machined separately in these two substrates, with one piece containing the microring and the other encompassing the remaining microfluidic ele-10 ments, including the input and output fluidic ports. The fabrication process is comprised of three main steps: in the first step, the external boundaries of the various microfluidic elements are written in their respective substrates. This writing step consists of exposing the substrates to femtosecond laser pulses from a Ti:sapphire laser operating at 800 nm. The 15 pulse width is approximately 100 fs and the repetition rate is set at 250 kHz. The beam is focused into the sample with a long-working distance microscope objective. The laser writing speed ranges from 100  $\mu$ m s<sup>-1</sup> to 5 mm s<sup>-1</sup> depending on the local geometry of the optofluidic element. The laser pulse 20 energy is adjusted so that there is no material ablation associated with this first step. Both transverse and longitudinal polarizations of the laser beam are used for the exposure. After the laser exposure is completed, the fused-silica substrates are immersed in a low-concentration aqueous solution 25of hydrofluoric acid. The specimens are polished and rinsed multiple times. Finally, the two substrates are aligned in a clean room and fused together in a high-temperature furnace to form a monolithic optofluidic system. The photograph of the laser-machined device and the microphotograph of the 30 microring cavity are shown in Fig. 2. Fig. 2(B) and (C) present the images of the microring and adjoining fluidic channels taken at different focal lengths. The optical measurements suggest that the separation distance between the ring and

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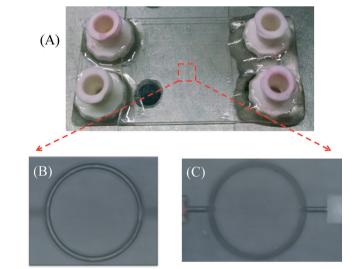


Fig. 2 (A) Picture of the optofluidic ring resonator laser system. (B) and (C) Microscopic pictures of the optofluidic ring resonator focused on the ring and the fluid delivery channel, respectively.

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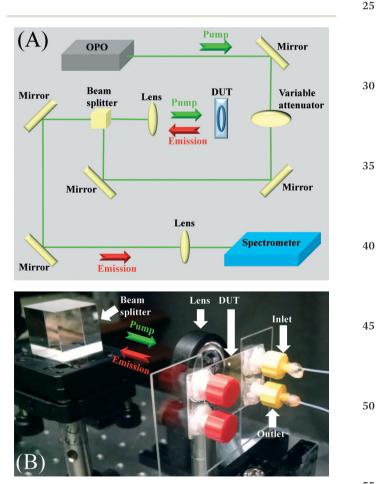
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parallel fluidic channels is approximately 100 µm, which is in agreement with the design specification. Fig. 2(C) also evidently points out that the orthogonal fluidic channels which directly contact the ring are routed from the inner circumference of the ring.

#### **Experimental**

The optofluidic laser was tested according to the measurement setup outlined in Fig. 3. Chemically inert polytetrafluoroethylene (PTFE) tubes with an inner diameter of 300 µm were attached to the input and output fluidic ports as shown in Fig. 3(B). The RG6 dye was dissolved in quinoline: the concentration was 1 mM. The gain solution was then injected into the fluidic channel at a constant flow rate of 3  $\mu$ L min<sup>-1</sup> using a syringe pump. The aqueous cavity was formed as the active solution with a higher refractive index (1.62) than that of the circumambient fused-silica (1.46) inhabits the ring channel. Next, nanosecond pulses at 532 nm wavelength with 20 Hz repetition rate were launched from an optical parametric oscillator to excite the cavity. The pump beam was focused to a spot area of 0.33 mm<sup>2</sup> on the sample by using a cylindrical lens. The coherent light output was emitted from the optofluidic cavity and recorded by a



55 Fig. 3 (A) Schematic of the measurement setup. (B) A close-up view of the device under test during measurement.

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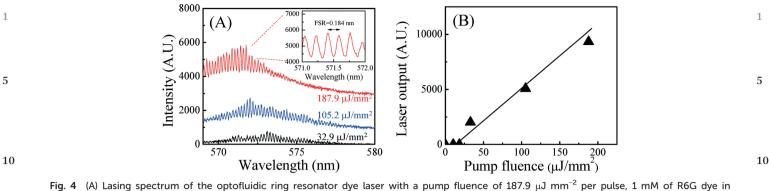


Fig. 4 (A) Lasing spectrum of the optofuluic ring resonator dye laser with a pump fluence of 187.9  $\mu$ J mm<sup>-</sup> per pulse, 1 mM of R6G dye in quinoline, and a flow rate of 3  $\mu$ L min<sup>-1</sup>. The spot size of the OPO pumping laser beam was approximately 0.33 mm<sup>2</sup>. A 2400 g mm<sup>-1</sup> grating was used to achieve high spectral resolution (~0.05 nm). Broad fluorescence background is removed. Spectra are vertically shifted for clarity. Inset: a magnified portion around 572 nm, showing a free spectral range of 0.184 nm. Wider spectral coverage lasing spectra below and just above the threshold are given in Fig. S1.† (B) Spectrally integrated (569–576 nm) laser output as a function of pump fluence extracted from (A). The lasing threshold is approximately 15  $\mu$ J mm<sup>-2</sup>. Solid line is the linear fit above the threshold.

spectrometer (Horiba iHR550, 2400 g mm<sup>-1</sup> grating, spectral resolution: ~0.05 nm, unless otherwise noted) (Fig. 3(A)).

#### Results and discussion

Fig. S1<sup>†</sup> plots the evolution of the emission from the ring resonator when the pump intensity increases. At low pump 25intensity, only broad fluorescence centered around 567 nm appears. With further increase in pump, multiple laser emission peaks emerge at the red-side of the fluorescence spectrum. Fig. 4(A) shows the high resolution lasing spectra, which allows us to distinguish each lasing peak. The free-30 spectral-range (FSR) of approximately 0.184 nm is clearly seen near the wavelength of 572 nm. This FSR is very close to the theoretical prediction of 0.189 nm using FSR =  $\lambda_m^2/(2\pi r n_{\rm Eff})$ , where  $\lambda_{\rm m} = 572$  nm, r = 170  $\mu$ m,  $n_{\rm Eff} = 1.62$ , which suggests 35 that the laser mode travels along the outer rim of the ring resonator, as expected. At pump intensity well above the threshold, a small blue shift in the lasing peak is observed, consistent with what was previously reported.<sup>36</sup> Fig. 4(B) displays the spectrally integrated laser emission as a function 40 of the pump intensity. The lasing threshold is approximated 15 µJ mm<sup>-2</sup>. To further confirm the laser emission,

additional experiments were performed with lower concentrations of RG6 dye in quinoline. Blue shifts in the laser emis-20 sions accompanied by attenuations in output peak intensity were recorded as the concentration of RG6 was lowered from 1 to 0.5 and 0.38 mM while preserving the same pump intensity as shown in Fig. 5(A)-(C). The dependence of emission Q3 wavelengths on the dye concentrations has been previously 25 studied<sup>36,37</sup> and can be accounted for by the dye laser theory presented in the ESI.<sup>†</sup> The versatility of the optofluidic ring resonator laser lies in its capability to conveniently change the solvent (and gain medium as well). Fig. S8<sup>†</sup> shows that the lasing emission can easily be achieved with R6G 30 dissolved in chlorobenzene (n = 1.524).

Success of ring resonator fabrication by femtosecond laser writing technology is reflected in the resonator's bare cavity Q-factor,  $Q_0$  (*i.e.*, the Q-factor in the absence of the gain medium). The Q-factor in the previous work, although not reported, is expected to be rather low due to the low reflectivity of the mirrors.<sup>15</sup> In the current work, it is difficult to directly measure  $Q_0$ . However, as detailed in the ESI,† the lasing threshold analysis is able to provide quite accurate  $Q_0$  estimation. Based on the lasing fluence threshold of 15 µJ mm<sup>-2</sup>, we estimate  $Q_0 = 3.3 \times 10^4$ .

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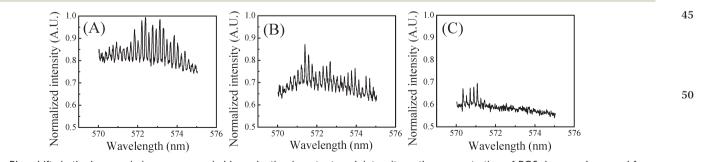


Fig. 5 Blue shifts in the laser emissions accompanied by reduction in output peak intensity as the concentration of RG6 dye was decreased from 1 mM (A) to 0.5 mM (B) and finally to 0.38 mM (C) while keeping the same pump fluence (105.2  $\mu$ J mm<sup>-2</sup>). All spectra are normalized with respect to the highest intensity peak recorded in Fig. 5(A).

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It is well known that  $Q_0$  is primarily determined by:

 $\frac{1}{O_0} = \frac{1}{O_{\text{red}}} + \frac{1}{O_{\text{red}}} + \frac{1}{O_{\text{red}}},$ 

where  $Q_{\text{Rad}}$ ,  $Q_{\text{Sol}}$ , and  $Q_{\text{Sc}}$  are the Q-factors due to the radia-

tive loss, solvent absorption, and loss resulting from wall

scattering. Among the three contributing Q-factors, the loss

due to radiation is known to be very small  $(Q_{\text{Rad}} > 10^{11})^{38}$ due to the relatively large ring radius (170 µm) and high

refractive index contrast ( $\Delta n \sim 0.17$ ).  $Q_{\rm Sol} = 2\pi n_{\rm Sol}/\lambda_{\rm L}\alpha_{\rm Sol}$ 

where  $\lambda_{\rm L}$  and  $\alpha_{\rm Sol}$  are respectively the lasing wavelength and

the absorption coefficient of the solvent. For quinoline,  $\alpha_{sol}$  =

1.1 cm<sup>-1</sup> (see Fig. S6<sup>†</sup>), which results in  $Q_{Sol} = 1.6 \times 10^5$ . Thus,

we can conclude that  $Q_{\rm Sc} = 4.2 \times 10^4$  and  $Q_0$  is mainly deter-

mined by the losses due to light scattering from the fused-

silica walls and absorption in quinoline. Note that although

 $Q_0$  (or  $Q_{\rm Sc}$ ) is 20–30 times lower than the best previously

reported femtosecond laser fabricated ring resonator (Q =

 $10^6$ ),<sup>39</sup> which underwent CO<sub>2</sub> laser annealing, it still repre-

sents one of the best cavities in optofluidic lasers (for exam-

bility of using a femtosecond laser to create high quality opti-

An integrated optofluidic ring resonator laser in a fused-silica

glass substrate has been successfully designed, fabricated,

and characterized. The manufacturing procedure of this

optofluidic laser is unique and compelling for various appli-

cations as it offers maskless and flexible fabrication process,

easy assembly, excellent device robustness, no post-

fabrication processes, quick prototyping, and relatively low

cost. The bare cavity Q-factor of  $3.3 \times 10^4$  has been achieved,

suggesting that the femtosecond writing technology can be

entitled for further investigations. First, we will incorporate

another liquid ring resonator nearby to achieve single mode

of laser operation via the Vernier effect.<sup>31,32</sup> In addition, since

each ring resonator can be independently controlled, a

wavelength-tunable laser will be built by changing the solu-

tion in one of the rings. A waveguide can also be fabricated

in the vicinity of the ring so that the laser emission can be

directionally coupled out. Furthermore, we will explore other

high index and low absorption solvents<sup>40</sup> as alternatives to

quinoline (see example in Fig. S8<sup>†</sup>). With those solvents, we

expect  $Q_0$  to approach  $Q_{Sc}$ , which is caused during femtosec-

ond laser fabrication. Post-fabrication annealing will also be

employed to further reduce scattering and increase  $Q_{Sc}$ .

Finally, biolasers will be explored where gain media such as

nanoparticles, biological molecules, and cells will be flowed

Several characteristics of this proof-of-concept device are

Conclusions and future work

used to create high quality cavities.

Acknowledgements

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#### ple, the Q-factor for DFB and Fabry-Pérot are usually on the order of 10<sup>3</sup> or lower). Such achievement suggests the feasi-

cal cavities.

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through the ring resonator.<sup>5</sup>

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#### **Supplementary Information**

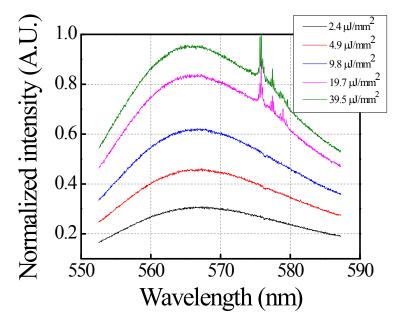
#### Monolithic Optofluidic Ring Resonator Lasers Created by Femtosecond Laser Nanofabrication

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**Supplementary Figure 1:** Emission spectra as a function of pump fluence below and just above the lasing threshold. A 1200 g/mm grating was used to achieve wider spectral coverage. For comparison purposes, all emission spectra are normalized to the highest peak value. High resolution (0.05 nm) spectra using a 2400 g/mm grating are plotted in Fig. 4(A).

Parameters	Definitions	Units
$\lambda_{ m P}$	Pump wavelength	nm
$\lambda_{ m L}$	Lasing wavelength	nm
$\lambda_{\mathrm{E}}$	Fluorescence peak wavelength	nm
n <sub>Sol</sub>	Refractive index of Solvent	-
n <sub>T</sub>	Rhodamine 6G (R6G) dye concentration	cm <sup>-3</sup>
n <sub>X</sub>	Dye concentration at excited state	cm <sup>-3</sup>
$\sigma_a(\lambda_P)$	Absorption cross-section at $\lambda_P$	cm <sup>2</sup>
$\sigma_a(\lambda_L)$	Absorption cross-section at $\lambda_L$	cm <sup>2</sup>
$\sigma_{e}(\lambda_{L})$	Emission cross-section at $\lambda_L$	cm <sup>2</sup>
Ε(λ)	Fluorescence quantum distribution	nm <sup>-1</sup>
q	Quantum yield of RG6 in quinoline	-
$q_{\text{Ref}}$	Quantum yield of RHB in ethanol	-
FL-Slope <sub>Sol</sub>	Fluorescence efficiency of RG6 in quinoline	nM <sup>-1</sup>
FL-Slope <sub>Ref</sub>	Fluorescence efficiency of RHB in ethanol	nM <sup>-1</sup>
σ	Standard deviation of RG6 fluorescence distribution	ns
$\epsilon_{Sol}$	Extinction coefficient of RG6	$M^{-1}cm^{-1}$
$\epsilon_{Ref}$	Extinction coefficient of RHB	$M^{-1}cm^{-1}$
$ au_{ m F}$	Fluorescence lifetime	ns
А	$\sigma_{\rm A}(\lambda_{\rm L})/\sigma_{\rm A}(\lambda_{\rm P})$	-
В	$\sigma_{\rm A}(\lambda_{\rm L})/\sigma_{\rm E}(\lambda_{\rm L})$	-
С	$Q_{Dye}/\eta Q_0$	-
h	Planck constant	Js
c	Speed of light in vacuum	m/s
$\Delta t$	Pulse width of the pump laser	ns
η	Light coupling efficiency	-
$\mathbf{P}_{\text{th}}$	Lasing photon density threshold	#photons/m <sup>2</sup>
$\Phi_{ m th}$	Lasing fluence threshold	$\mu J/mm^2$
$Q_{\text{Dye}}$	Q-factor due to dye absorption	-
$Q_{Sol}$	Q-factor due to solvent absorption	-
$Q_{\text{Rad}}$	Q-factor due to radiation loss	-
$Q_{Sc}$	Q-factor due to scattering loss	-
Q <sub>0</sub>	Q-factor of the laser cavity in the absence of gain medium	-

 Table 1: Parameters used in the present work

#### **Q**-factor analysis

Based on the lasing theory and the experimentally measured lasing threshold, the cavity Q-factor in the absence of the gain medium (*i.e.*,  $Q_0$ ) can be calculated. The lasing threshold is given by:

$$n_X \sigma_E(\lambda_L) = (n_T - n_X) \sigma_a(\lambda_L) + \frac{2\pi n_{Sol}}{\lambda_L \eta Q_0}$$
(S1)

Equation (S1) can be written as:

$$\gamma \equiv \frac{n_X}{n_T} = \frac{\sigma_a(\lambda_L)}{\sigma_e(\lambda_L) + \sigma_a(\lambda_L)} \cdot (1 + \frac{Q_{Dye}}{\eta Q_0})$$
(S2)

The stimulated emission cross section is obtained from the following expression:

$$\sigma_e(\lambda_L) = \frac{\lambda_L^4 E(\lambda_L)}{8\pi c n_{Sol}^2 \tau_F}$$
(S3)

According to the four energy level model, at steady state,  $\gamma$  relates to the dimensionless pump intensity by:

$$\gamma = \frac{I}{I+1},\tag{S4}$$

and 
$$I = \frac{\gamma}{1 - \gamma}$$
, (S5)

where  $I = P \cdot \sigma_a(\lambda_p) \cdot \tau_F$ . P is the power density in units of (# of photons per m<sup>2</sup> per second).

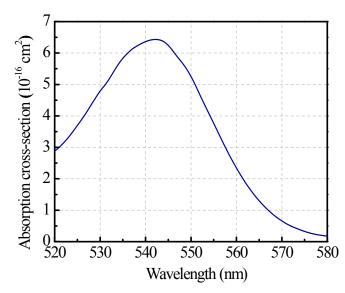
Equations (S2) and (S5) can be combined:

$$\frac{P_{th}}{\Delta t} \cdot \sigma_a(\lambda_P) \cdot \tau_F = \frac{\frac{\sigma_a(\lambda_L)}{\sigma_e(\lambda_L) + \sigma_a(\lambda_L)} \cdot [1+C]}{1 - \frac{\sigma_a(\lambda_L)}{\sigma_e(\lambda_L) + \sigma_a(\lambda_L)} \cdot [1+C]} = \frac{1}{\sigma_e(\lambda_L)} \frac{\sigma_a(\lambda_L) \cdot [1+C]}{1 - \frac{\sigma_a(\lambda_L)}{\sigma_e(\lambda_L)} \cdot C}$$
(S6)

P<sub>th</sub> can be expressed in a simpler form as:

$$P_{th} = \frac{\Delta t}{\sigma_e(\lambda_L) \cdot \tau_F} A \frac{1+C}{1-B \cdot C}$$
(S7)  
where  $A = \frac{\sigma_a(\lambda_L)}{\sigma_a(\lambda_P)}, B = \frac{\sigma_a(\lambda_L)}{\sigma_e(\lambda_L)}, \text{ and } C = \frac{Q_{Dye}}{\eta Q_0}.$   
 $Q_{Dye}$  can be calculated by:  
 $Q_{Dye} = \frac{2\pi n_{Sol}}{\lambda_L n_T \sigma_a(\lambda_L)}$   
P<sub>th</sub> is related to the excitation fluence at the pump wavelength by:  
(S8)

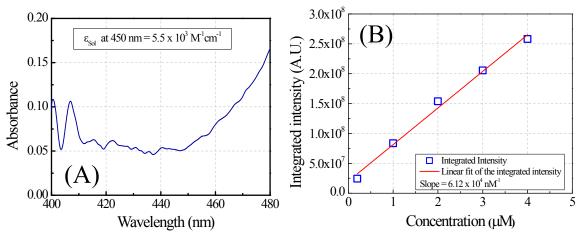
$$\Phi_{th} = P_{th} \cdot \frac{h \cdot c}{\lambda_P} = \frac{h \cdot c \cdot \Delta t}{\sigma_e(\lambda_L) \cdot \lambda_P \cdot \tau_F} A \frac{1 + C}{1 - B \cdot C}$$
(S9)



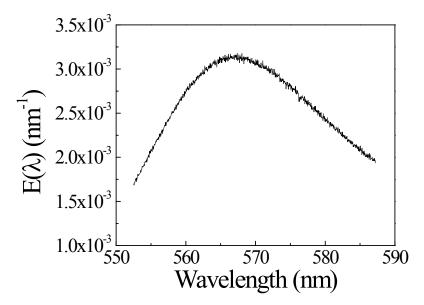
Supplementary Figure 2: Absorption cross-section of RG6 in quinoline based on absorbance measurement with  $10 \mu M$  solution.

In order acquire necessary parameters to analyze  $Q_0$  in equation (S9), we performed several measurements and data extractions. We measured the Rhodamine 6G (R6G) dye absorption cross-section as presented in Fig. S2 and obtained  $\sigma_a(\lambda_P)$  and  $\sigma_a(\lambda_L)$ .  $Q_{Dye}$  was calculated to be 9.07x10<sup>3</sup> according to equation (S8).

Next, we need to determine the fluorescence quantum distribution  $E(\lambda)$  and fluorescence lifetime ( $\tau_F$ ) of the RG6 in quinoline to calculate  $\sigma_e(\lambda_L)$  in equation (S3). In order to compute  $E(\lambda)$ , we need to first calculate the quantum yield (q) and measure the fluorescence spectrum of the dye solution. To determine the quantum yield, we performed absorption and fluorescence measurements of RG6 dye in quinoline and compared the results against a reference dye solution (Rhodamine B (RhB) in ethanol) which has a known  $q_{Ref}$  value<sup>1</sup>. Fluorescence test was done with FluoroMax-4



**Supplementary Figure 3:** (A) Absorbance of RG6 dye in quinoline as a function of wavelength. (B) Integrated fluorescence intensity as a function of RG6 dye concentration.



**Supplementary Figure 4:** Fluorescence quantum distribution,  $E(\lambda)$ .

spectrofluorometer (Horiba Scientific). Absorption test was performed with NanoDrop 2000c spectrophotometer (Thermal Scientific). The excitation wavelength for the absorption measurement was 450 nm. The measurement results shown in Fig. S3 indicate  $\epsilon_{Sol}$  and FL-Slope<sub>Sol</sub> of 5.5 x 10<sup>3</sup> M<sup>-1</sup>cm<sup>-1</sup> and 6.12 x 10<sup>4</sup> nM<sup>-1</sup>, respectively.

Using the equation:

$$q = q_{Ref} \times \frac{FL - Slope_{Sol}}{FL - Slope_{Ref}} \times \frac{n_{Sol}^2}{n_{Ref}^2} \times \frac{\varepsilon_{Ref}}{\varepsilon_{Sol}}$$
(S10)

the fluorescence quantum yield, q, of 0.12 was obtained.

RG6 fluorescence quantum distribution,  $E(\lambda)$ , can be obtained through the fluorescence spectrum in Fig. S1(B) by considering that

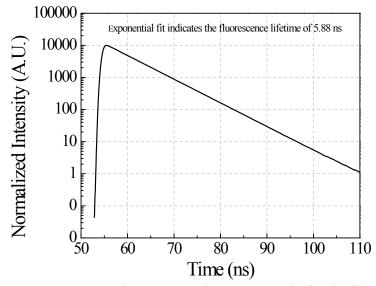
$$\int E(\lambda) \cdot d\lambda = q. \tag{S11}$$

(S12)

According to Fig. S4 E(575 nm) =  $0.0028 \text{ nm}^{-1}$ .

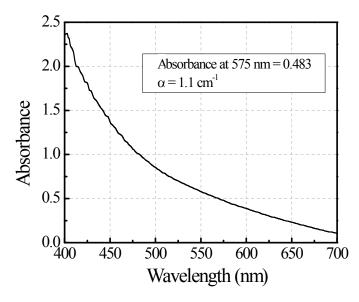
In the next step, we measured the fluorescence lifetime,  $\tau_F$  of the RG6 in quinoline. The measurement result of the time correlated photon counting experiment performed using a fluorescence lifetime spectrometer (TemPro, Horiba) is shown in Fig. S4. An exponential fit reveals 5.88 ns fluorescence lifetime of RG6 dye in quinoline. We substituted the measured parameters to equation (S3), and determined the stimulated emission cross-section,  $\sigma_e$ , of 2.61x10<sup>-17</sup> cm<sup>2</sup>.

We have now all the parameters needed to extract  $Q_0$  from equation (S9). Considering an input pump fluence threshold ( $\Phi_{TH}$ ) of 15  $\mu$ J/mm<sup>2</sup> during experiment, we calculate  $Q_0$ to be 3.3x10<sup>4</sup>.



**Supplementary Figure 5:** Fluorescence decay curve obtained with 500  $\mu$ M RG6 dye in quinoline. The exponential fit reveals 5.88 ns fluorescence lifetime.

In order to validate the method used here to extract the Q-factor (*i.e.*,  $Q_0$ ), we employ the same theoretical model to estimate the lasing threshold of an optofluidic ring resonator laser reported in Ref. 2., which Q-factor is known (*i.e.*,  $\eta Q_0 = 4x10^6$ ). Table 2 tabulates several key parameters used for this work and the work reported in reference<sup>2</sup>. Based on the parameters listed on the right column of Table 2, an estimated lasing fluence threshold of 22 nJ/mm<sup>2</sup> is obtained, very close to the experimentally measured result of 25 nJ/mm<sup>2</sup>. Therefore, our current approach to extract  $Q_0 = 3.3x10^4$  using the lasing fluence threshold is justified.



**Supplementary Figure 6:** Absorbance measurement of quinoline as a function of wavelength.

Parameters	Current work (1 mM R6G in quinoline)	Ref. 2 (2 mM R6G in ethanol)
$\lambda_{ m P}$	532 nm	532 nm
$\lambda_{ m L}$	575 nm	600 nm
n	1.62	1.40
n <sub>T</sub>	$6 \text{ x } 10^{17} \text{ cm}^{-3}$	$1.2 \text{ x } 10^{18} \text{ cm}^{-3}$
$\sigma_a(\lambda_P)$	5.18 x 10 <sup>-16</sup> cm <sup>2</sup>	$2 \text{ x } 10^{-16} \text{ cm}^2$
$\sigma_a(\lambda_L)$	$3.25 \text{ x } 10^{-17} \text{ cm}^2$	$1 \ge 10^{-19} \text{ cm}^2$
$\sigma_e(\lambda_L)$	$2.61 \text{ x } 10^{-17} \text{ cm}^2$	$1 \ge 10^{-16} \text{ cm}^2$
Ε(λ)	2.8 x 10 <sup>-3</sup> nm <sup>-1</sup>	5.7 x 10 <sup>-3</sup> nm <sup>-1</sup>
q	0.12	0.9
$ au_{ m F}$	5.88 ns	4.08 ns
А	6.27 x 10 <sup>-2</sup>	5 x 10 <sup>-2</sup>
В	1.25	1 x 10 <sup>-3</sup>
С	0.28	0.3
$\Delta t$	5 ns	5 ns
Q <sub>Dye</sub>	9.07 x 10 <sup>3</sup>	1.2 x 10 <sup>6</sup>
$\eta Q_0$	3.3 x 10 <sup>4</sup>	4 x 10 <sup>6</sup>
$\Phi_{ m th}$	15 μJ/mm <sup>2</sup>	22 nJ/mm <sup>2</sup>

 Table 2: Comparisons of the current work and the work in Ref. 2.

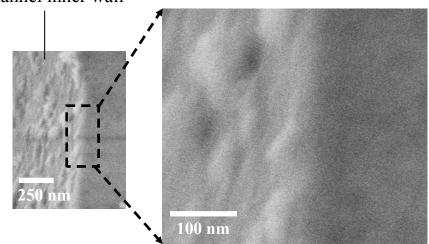
#### Estimation of Q<sub>Sc</sub>

The surface roughness of a pre-annealing device fabricated by the 3-D femtosecond laser writing process has previously been characterized to be 40-50 nm<sup>3</sup>. Fig. S6 shows the SEM image of the ring resonator inner wall taken after the ring resonator underwent thermal annealing. The roughness was reduced to approximately 30 nm.

The roughness is related the Q-factor by<sup>4</sup>:

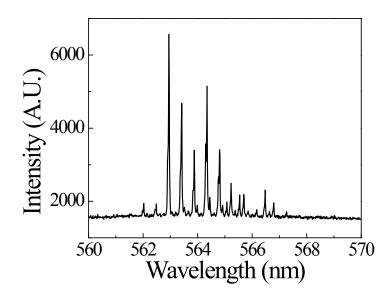
$$Q \propto \frac{1}{\sigma_{rough}^2 \cdot (n_{core}^2 - n_{cladding}^2)},$$
(S13)

where  $\sigma_{rough}$  is the surface roughness.  $n_{core}$  and  $n_{cladding}$  are the refractive index for the ring and the cladding, respectively. Previous work on polymer ring resonators, whose surface roughness and the Q-factor have been well characterized and directly measured, provides a means to estimate the scattering loss related Q-factor of our ring resonator. According to Ref. 4,5, a Q-factor of  $5x10^5$  was obtained at 780 nm for a polymer ring resonator with the surface roughness of 5-10 nm. Based on equation S13, the scattering loss related Qfactor of our ring resonator should range between  $3.2x10^4$  and  $1.3x10^5$ , in agreement with the scattering related Q-factor estimation ( $Q_{Sc} = 4.2x10^4$ ) that we extract based on the lasing threshold.



**Supplementary Figure 7:** SEM images of the channel inner wall. The surface roughness (rms) is about 30 nm.

Channel inner wall



**Supplementary Figure 8:** Lasing emission from 1 mM R6G in chlorobenzene (n=1.524) at a pump fluence of 90  $\mu$ J/mm<sup>2</sup>. Flow rate was 3  $\mu$ L/min.

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