A multi-color fast-switching microfluidic droplet dye laser

Supplementary Information

Sindy K.Y. Tang,[a] Zhenyu Li,[b] Adam R. Abate,[c] Jeremy J. Agresti,[c] David A. Weitz,[c] Demetri Psaltis,[d] and George M. Whitesides[a*]

[a] *Department of Chemistry and Chemical Biology, Harvard University, Cambridge, MA 02138. E-mail: gwhitesides@gmwgroup.harvard.edu; Fax: +617-495-9857; Tel: +617-495-9430*

[b] *Department of Electrical Engineering, California Institute of Technology, Pasadena, CA 91125*

[c] *School of Engineering and Applied Sciences/Department of Physics, Harvard University, Cambridge, MA 02138.*

[d] *Optics Laboratory, School of Engineering, Swiss Federal Institute of Technology Lausanne (EPFL), CH-1015 Lausanne, Switzerland*

**Figure S1.** Setup for optical excitation and characterization
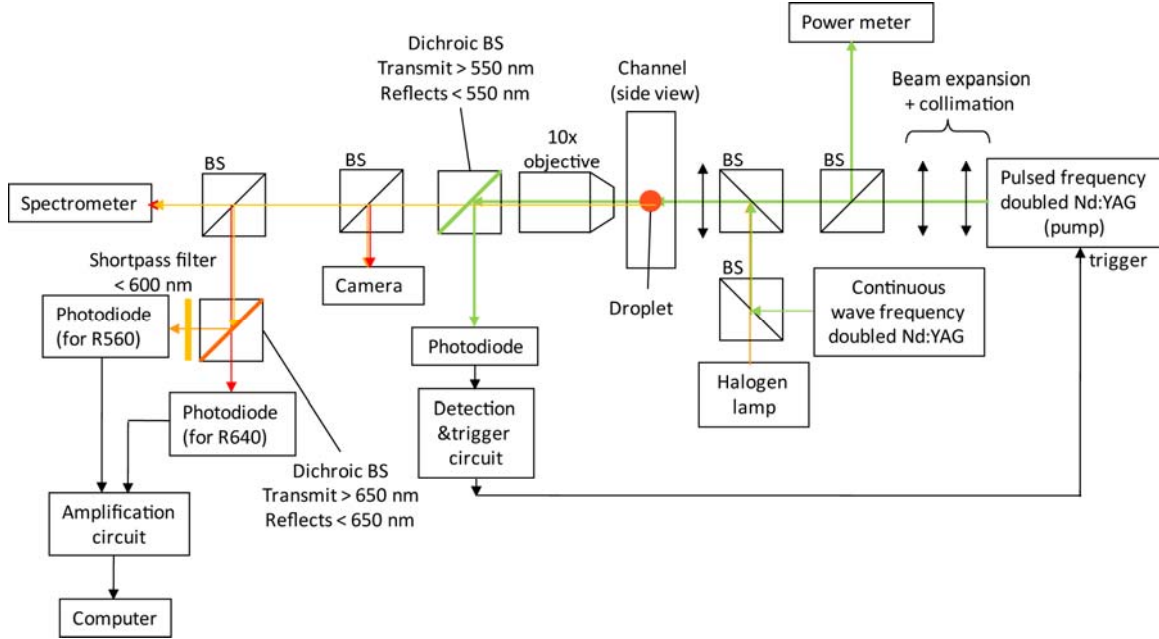
Optical excitation. We used a pulsed frequency-doubled Nd:YAG laser at 532 nm as the source of excitation (Photonix DC150-532). The pump beam was expanded, and refocused onto the microfluidic channel to cause lasing of the droplets in whispering gallery modes. We split the beam in half with a 50/50 beam splitter, and measured the power of the excitation laser using a power meter. We also introduced broadband light from a halogen lamp onto the channel for imaging the drops. This light was blocked during the acquisition of emitted light from the drops.

Optical characterization. We collected the optical output from the droplets via a 10x objective (NA 0.25). This output was split into two via a 50/50 beam splitter. One beam was coupled to a fast camera (Vision Research Phantom v7.3) for imaging the droplets, the other was coupled to another beam splitter for characterization of lasing: half of this beam was coupled to a spectrometer (Ocean Optics HR4000, 0.1 nm or 0.3 nm resolution) for analysis of intensity as a function of wavelength at low switching speeds

of drops (< 130 Hz). The other half passed through a dichroic beam splitter (transmits > 650 nm, reflects < 650 nm) and were coupled to two photodiodes (Thorlabs DET36a) for characterization of the rate of switching between the lasing wavelengths at higher speeds (> 130 Hz). The photodiode for characterization of R560 drops was also mounted with a short pass filter (< 600 nm). The voltage output from the photodiode was amplified, and then acquired and recorded using National Instruments Labview FPGA module at 200k samples per second.

Synchronization of the drops with the excitation pump pulses. We focused a continuous-wave green laser (< 5 mM) at the same spot as the pump laser. We measured the transmitted intensity at 532 nm using a photodiode; the presence of a drop caused this transmitted intensity to decrease. Such dip in intensity (i.e. output voltage from the photodiode) triggered the pump laser to release one pulse to cause lasing from the droplet. The intensity of fluorescence from the drops caused by this CW laser was much lower than the WGM lasing intensity from the drops caused by the pump laser, and was below background noise of our spectrometer, and did not interfere with the lasing spectra.

Figure S2. Lasing spectra for conditions used in figure 4. The integration time was 3.8 ms (minimum of the spectrometer); multiple (about four) lasing spectra from R560 and R640 (switching at 1.18 kHz) were acquired and integrated during this integration period. Their summed intensities appeared on the same graph. The diameter of the drops was about 20 μm.
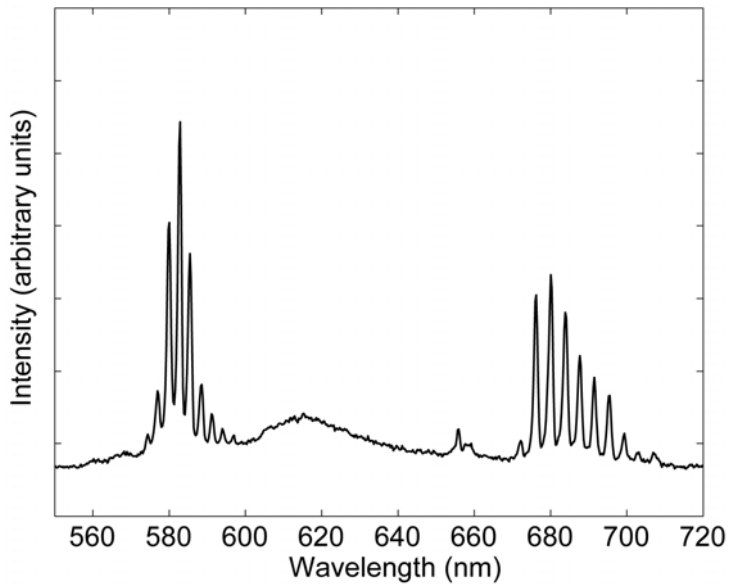
Discussion about the shift in the wavelength of the cluster of lasing modes shown in figure 3b.

The modes we measured here consisted of overlapping transverse electric (TE) and transverse magnetic (TM) modes, each with different mode numbers (n) and different mode orders (l). The positions of these modes overlap, and can usually be predicted by modeling only (reference 22).

A change in the size of the drop changes both the separation between the peaks and the position of the peaks. In general:

1) $FSR = \Delta\lambda = \lambda_{n,l} - \lambda_{n+1,l} \sim \dfrac{\lambda^2}{2\pi m r}$, where m = group index of the mode, and r = drop

radius. This $\Delta\lambda$ (or usually called free spectral range, FSR) is the separation between the peaks belonging to the same mode order (l) but different mode number (n).

2) $\dfrac{\Delta\lambda_{n,l}}{\lambda_{n,l}} = \dfrac{\Delta r}{r}$. This $\Delta\lambda_{n,l}$ is the shift in the position of a particular mode.

We believe the shift in the cluster of modes shown in figure 3b was due to small variations in the size of the drops generated. We cannot show at this point if the cluster of modes have shifted only ~ 1 nm or more (> 2 nm) and started overlapping with other modes (since the separation between different modes was only ~ 2 nm).

However, if we assume that the clusters of modes at different times in figure 3b shifted less than 2 nm in wavelength, using 2) above, the variation in drop size would be

$$\frac{\Delta r}{r} = \frac{\Delta\lambda_{n,l}}{\lambda_{n,l}} \sim \frac{1\,nm}{670\,nm} = 0.15\% \ .$$