

## **Electronic Supplementary Information:**

### **A fast and efficient microfluidic system for highly selective one-to-one droplet fusion**

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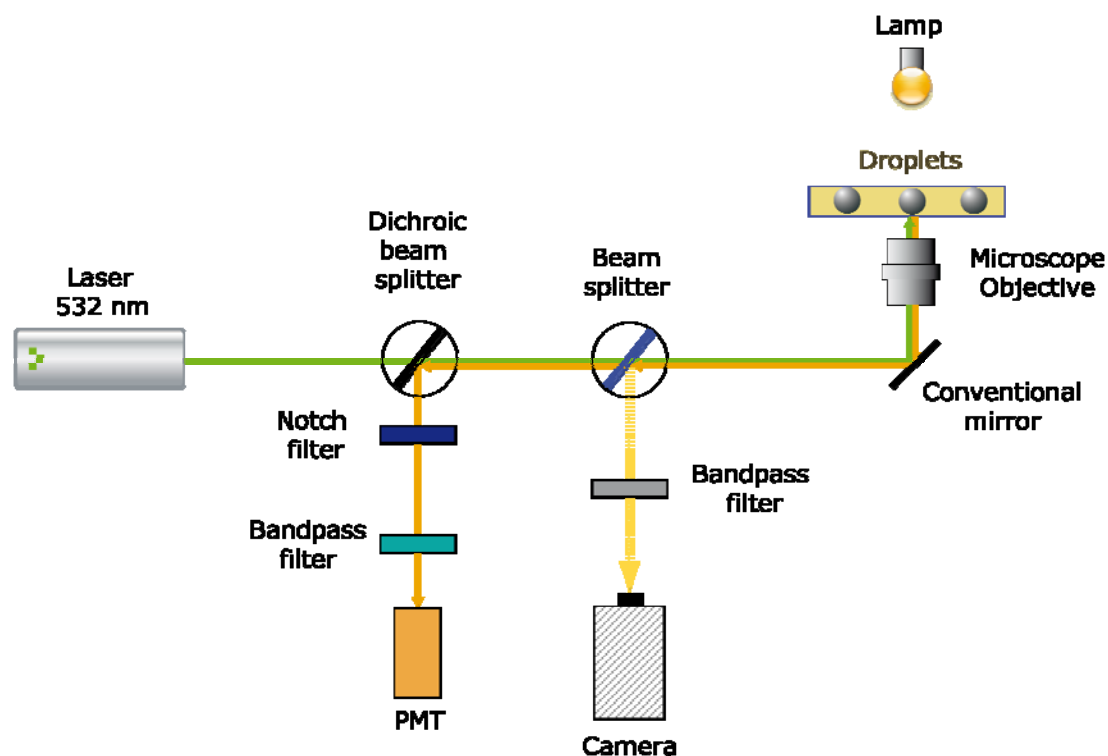
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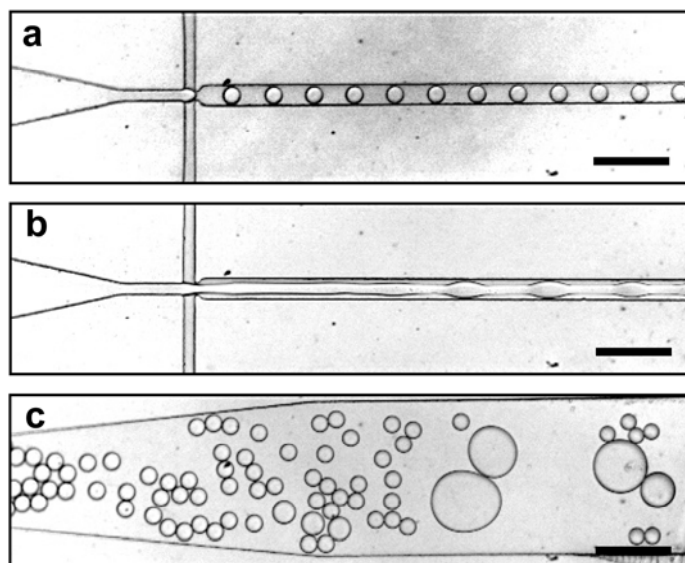
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**Figure S1. Schematic representation of the optical set-up.** Droplet fluorescence was induced using a 532 nm (25mW) solid-state laser (Newport-Spectraphysics) guided to the side camera port of the Axiovert 200 inverted microscope (Carl Zeiss SAS) by a series of periscope assemblies (Thorlabs GmbH). Inside the microscope, the laser light was reflected up into a LD Plan Neofluar 40×/0.6 microscope objective (Carl Zeiss SAS) and focused in a channel within the microfluidic device. A Phantom v4.2 high-speed digital camera (Vision Research) was mounted on the top camera port of the microscope to capture digital images during droplet production, fusion and re-injection. A 562/40 BrightLine® bandpass filter (Semrock Inc.) positioned in front of the camera protected the camera's sensor from reflected laser light. Light emitted from fluorescing droplets was captured by the objective and channeled back along the path of the laser into the system of periscope assemblies. The emitted light was separated from the laser beam by a 488/532/638 nm-wavelength transmitting dichroic beam splitter (Semrock Inc.), and filtered through a notch (NF01-532U-25) and bandpass (FF01-617/73-25; Semrock Inc.) filter for detection on an H5784-20 photomultiplier tube (PMT, Hamamatsu Photonics KK). The signal output from the PMT was analysed using a PCI-7831R Multifunction Intelligent DAQ card (National Instruments Corporation) executing a program written in LabView 8.2 (FPGA module, National Instruments Corporation) which allowed the identification of droplets by peaks in fluorescence, as well as the width of each droplet. The data acquisition rate for the system was 100 kHz.



**Figure S2. Droplet production in the presence and absence of surfactant.** (a) Light micrograph of droplet production when the carrier oil contained 0.05% EA-surfactant. (b) Light micrograph of droplet production when the carrier oil was without surfactant. (c) Light micrograph showing droplet coalescence in the stability measurement chamber when the carrier oil contained 0.55% EA-surfactant. Scale bars 100  $\mu\text{m}$ .

**Supplementary movies:**

**Movie M1** shows selective droplet fusion between 3 pL re-injected droplets (black) and 9 pL droplets produced on-chip (clear). Carrier oil (FC40) used to create droplets on-chip and to space re-injected emulsion contained 0.55% EA-surfactant. Stabilization oil contained 2.8% EA-surfactant.

**Movie M2** shows droplet coalescence within a channel lacking zig-zag structures. Only ~70% of droplet pairs coalesce. Carrier oil used to create droplets on-chip and to space re-injected emulsion contained 0.55% EA-surfactant. Stabilization oil contained 2.8% EA-surfactant.

**Movie M3** shows fused droplets in the stability measurement chamber. Fused droplets (12 pL) are grey due to methylene blue from the 3 pL re-injected droplets (black). Carrier oil used to create droplets on-chip and to space re-injected emulsion contained 0.55% EA-surfactant. Stabilization oil contained 2.8% EA-surfactant.

**Movie M4** shows selective droplet fusion in multiple droplet clusters. 3 pL re-injected droplets are black and 9 pL droplets produced on-chip are clear. Droplets fuse selectively one-to-one (one re-injected droplet with one droplet generated on-chip). Carrier oil used to create droplets on-chip and to space re-injected emulsion contained 0.55% EA-surfactant. Stabilization oil contained 2.8% EA-surfactant.