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Aggregation of Molecules is Controlled in Microdroplets

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Table of Contents

1 Experimental section:

1.1 Microdroplet imprinting experiment. The droplet generation and deposition setup are already discussed in our recent work.[1] Briefly, a custom-built setup was used for droplet generation and imprinting. The electrospray sonic ionization (ESSI) set up with a transfer tube, as seen in the schematic in Fig. S1, was clamped together and mounted on an XYZ translational stage so that the emitter tube of the ESSI source could go inside the transfer tube. The dye-containing solution was kept in a syringe and purged through a fused silica capillary with a 150 µm internal diameter using a syringe pump. We used N_2 for nebulization with a gas pressure of 20 psi for most of the experiments otherwise mentioned. The droplets were imprinted on clean glass slides (SCHOTT nexterion®). All the droplet imprinting and fluorescence imaging experiments were conducted in a dark environment to prevent the photobleaching of the dye.

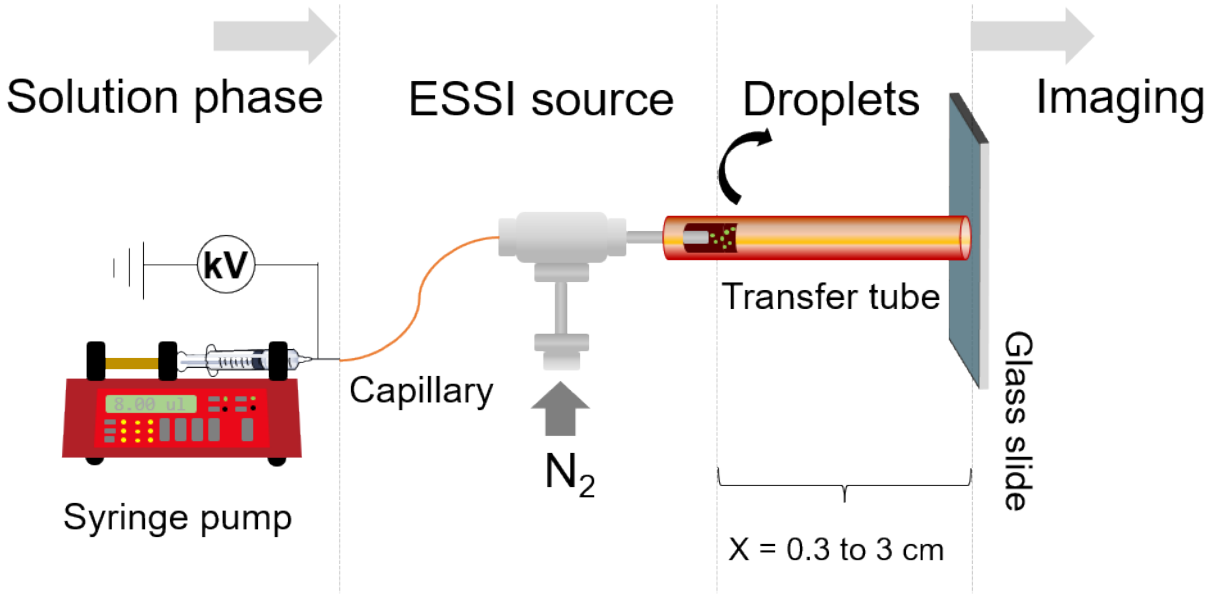

Fig. S1 Schematic of microdroplet generation and deposition experiment. It has been segmented into four parts; Solution phase, ESSI source, droplet flight path, and deposition for imaging. A high voltage DC power supply was connected to the syringe for the potential to apply optionally.

1.2 Microdroplet imaging experiment. The imprinted droplets were immediately transferred to the micromechanical stage of the dark field microscope (CytoViva™) for fluorescence imaging. The microscope has a high-resolution dark-field oil immersion condenser lens and a 100x oil immersion objective (UPLFLN, Olympus). We used a white light source for illuminating the dye with an L1090-halogen lamp from International Light Technologies Inc. Fluorescence imaging of microdroplets was also done on the CytoViva™ microscope with a triple bandpass filter for the green emission. A Dage Excel M cooled CCD (Charge coupled Device) camera was used for capturing the images. Clean glass slides (SCHOTT nexterion®) were used for all the imaging experiments.

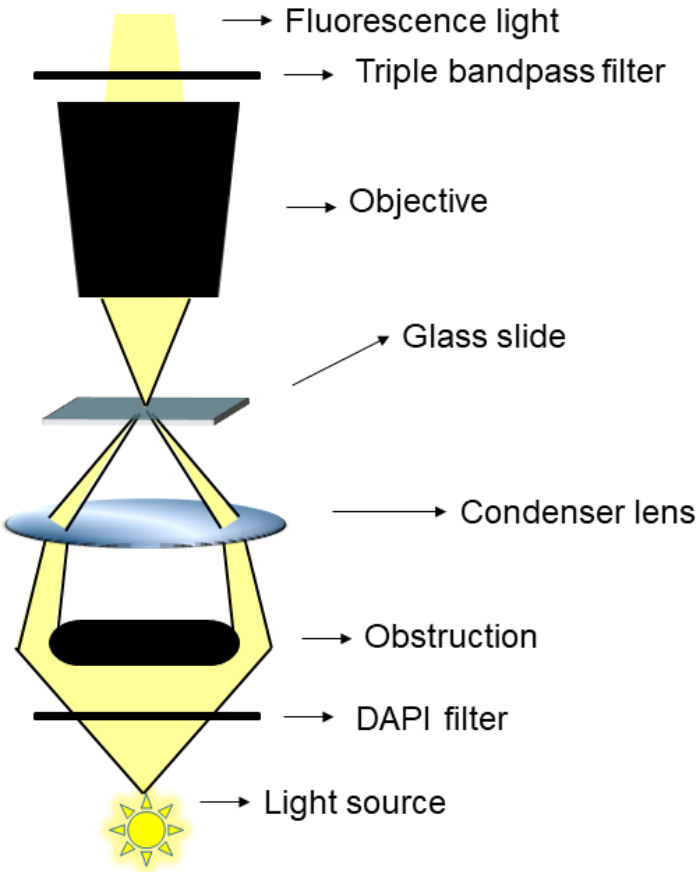

Fig. S2 Schematic view of Fluorescence microscopy. (DAPI - 4',6-diamidino-2-phenylindole).

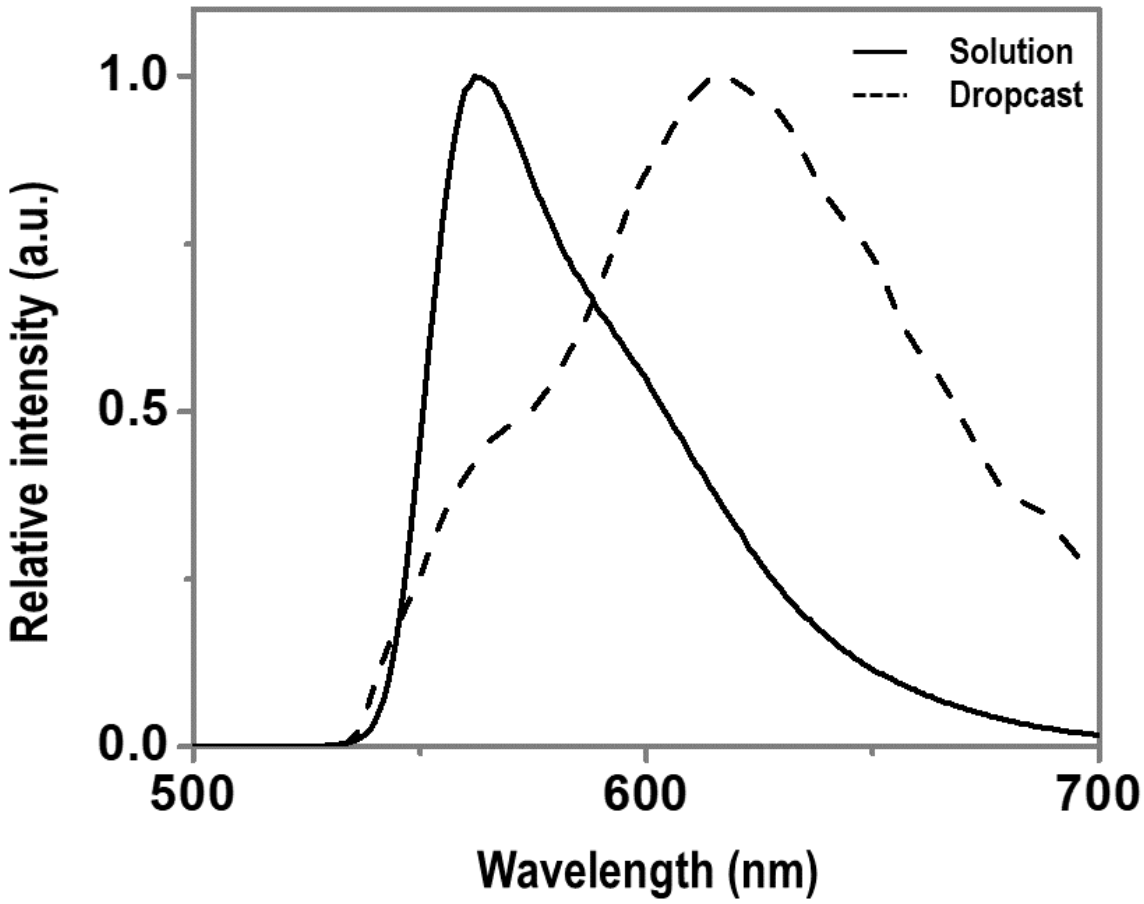

Fig. S3 Fluorescence spectra collected from the dropcast film and bulk solution of R6G.

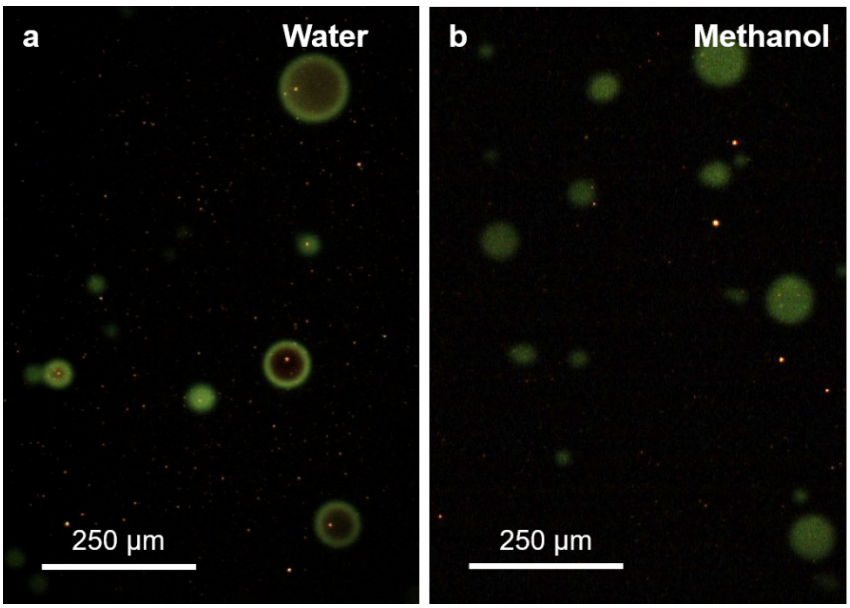

Fig. S4 Fluorescence microscopy images of microdroplets containing 100 µM R6G in a) water and b) methanol.

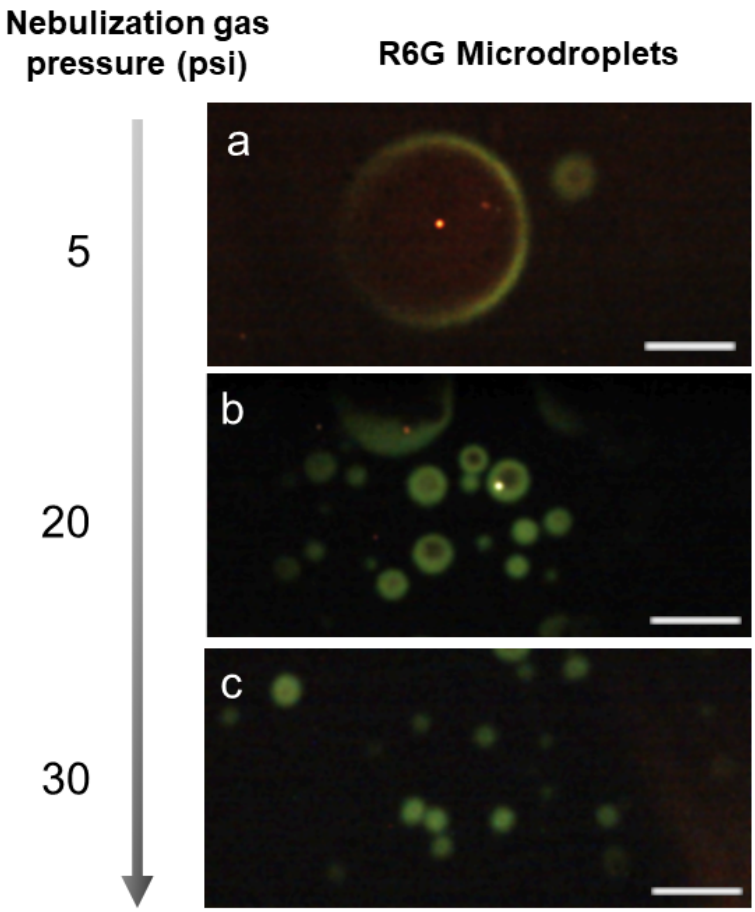

Fig. S5 Pressure dependency of the fluorescence emission of the microdroplets. The scale bar is 250 μm.

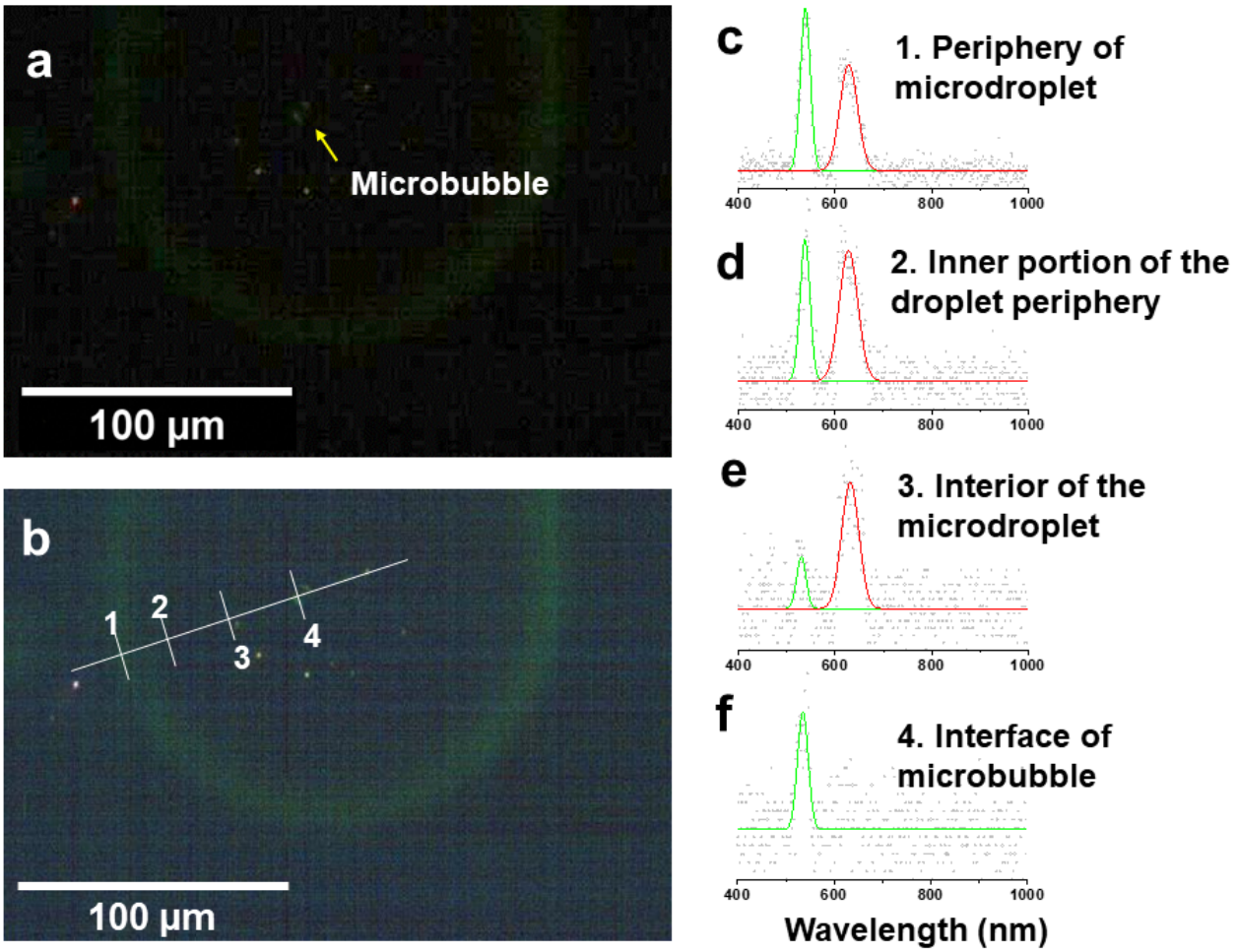

Fig. S6 Scattering spectrum of the microdroplet at the different regions from the droplet. a) and b) Fluorescence microscopy image of a droplet using two different filters. c-f) Fluorescence spectra of R6G at different locations of the droplet as indicated in b.

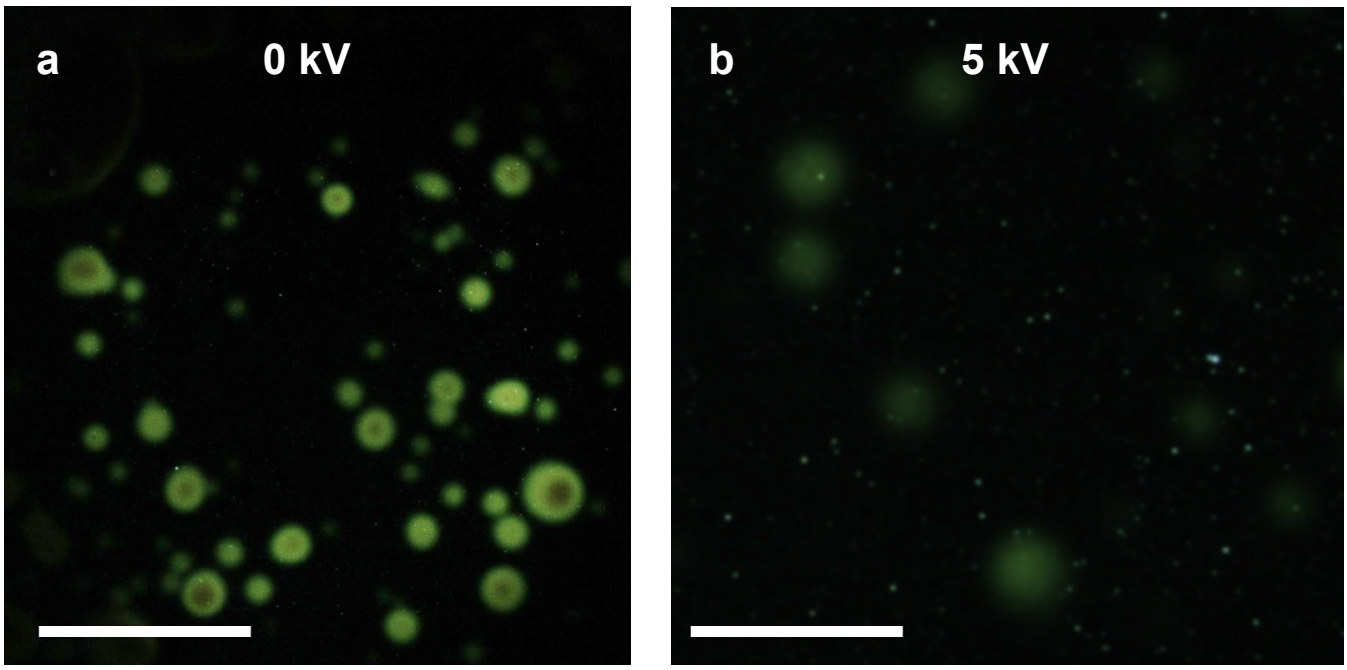

Fig. S7 The potential effect on microdroplets sprayed with R6G at 100 μM (at a spray distance of 1.5 cm). Scale bar is 250 μm.

References:

1. P. Basuri, A. Chakraborty, T. Ahuja, B. Mondal, J. Shantha Kumar and T. Pradeep, *(Submitted)*, 2022.