## **Supporting information**

## Sizing sub-diffraction limit electrosprayed droplets by structured illumination microscopy

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**Video 1**: Video of paper sprayed droplets depositing from a 3 mm tip-to-surface distance. The video was captured using a CCD camera attached to a confocal microscope. A piezoelectric discharge gun was used as the pulsed, high voltage power source. An electrospray was generated for approximately 500 ms whereby droplets were observed to significantly agglomerate on the microscope coverslip surface before evaporating. The spray solution was 1  $\mu$ M rhodamine B in 4:1 methanol:water. The video has been slowed down to better observe the evolution of droplets depositing.



**Figure S1**: Images of paper sprayed droplets containing 9:1 methanol:glycerol showing the effect of using (a) one, (b) two, (c) three, and (d) four Hough transformations for analysis. Droplet size distributions for (e) one, (f) two, (g) three, and (h) four Hough transformations.



**Figure S2**: Example super resolution images of fluorescent nanoparticles with nominal diameters of (a) 0.25, (b) 0.53, and (c) 0.84  $\mu$ m. Images of nanoparticles after detection using circular Hough transformations corresponding to (d) 0.25, (e) 0.53, and (f) 0.84  $\mu$ m nominal diameters. (Right) Overlaid histograms of the different sized nanoparticles. Each distribution was a collection of images of nanoparticles taken from different frames.



Figure S3: Manufacturer reported distribution for 0.25 µm nominal diameter fluorescent nanoparticles.



Figure S4: Manufacturer reported distribution for 0.53 µm nominal diameter fluorescent nanoparticles



**Figure S5**: Super resolution images of fluorescent nanoparticles (nominal dimeter =  $0.84 \mu$ m) showing the effect of image resizing on the accuracy of the droplet size measurement algorithm. Resized images allowed the algorithm to detect a greater number of droplets and provide more accurate size measurements compared to non-resized images.



**Figure S6**: Images of 0.53  $\mu$ m nominal diameter fluorescent nanoparticles shown at different normalization values (a-e). Images of detected particles (f-j) and the corresponding distributions of nanoparticles at each set of normalization values (k-o). The data show that no significant difference is obtained when using optimal Hough transformation conditions to analyze images with different normalization values.

## S-7



**Figure S7**: Evaporation study showing how the average diameter of droplets in a single frame changes with respect to time when (blue) continuously exposed to laser light and (red) exposed to laser light only during image acquisition.



**Figure S8**: Droplet size distributions acquired after the analysis of nanoelectrosprayed droplets generated from (a) 5, (b) 10, and (c) 20  $\mu$ m o.d. nanoelectrospray emitters. Each experiment was conducted using a 1 mM solution on 1:1 methanol:glycerol at a 3 mm distance using a 20 ms pulse duration.