Supplementary Information
Electrohydrodynamic behavior of droplets in an microfluidic oil-in-oil emulsion

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I. DROP BREAKUP AND COALESCENCE

Breakup: Movie 1 (left panel, $E = 9.5$ V/μm) shows a time series for the breakup of a large pancake-shaped drop. Breakup events can be spotted at 7.4 s, 14.6 s, and 16.9 s Cell thickness $d = 27$ μm. The middle panel ($E = 8.5$ V/μm) shows the breakup a drop in a thicker ($d = 55$ μm) cell. Here the drop is close to spherical, and breakup occurs by deformations in the plane perpendicular to the field. Breakup occurs at 3.60 s.

Coalescence: Movie 1 (right panel, cell thickness $d = 96$ μm, $E = 3.5$ V/μm) shows a drop coalescence event. The third dimension is crucial to the coalescence event, with a drop detaching from a bounding substrate at around 0.7 s, moving above a larger drop. Both drops are oblate, so this configuration allows their flat faces to touch. Coalescence occurs at ≈ 2.5 s.

II. DROP DYNAMICS

Movie 2 (left panel) shows a time series for the short-time dynamics of drops for $t < 0.9$ s. In this regime, the mean-square displacements are linearly dependent on time. $d = 55$ μm. $E = 12.5$ V/μm

Movie 2 (right panel) shows the long-time dynamics ($t < 9$ s) for the same sample conditions. Some directed motion is observable.

Supplementary Figure 1 shows the mean square displacements as a function of time for the thinnest cell ($d = 27$ μm). This dependence can be fitted to a linear form for intermediate electric fields, and to MSD $= K_1 t + K_2 t^2$ for larger electric fields. There is no motion at all for low fields. The prefactor to the quadratic term yields the advection: $v = \sqrt{K2}$. Since the square of this speed is proportional to the kinetic energy, and E^2 is proportional to the injected electric field energy, one would expect that $v \propto E$.

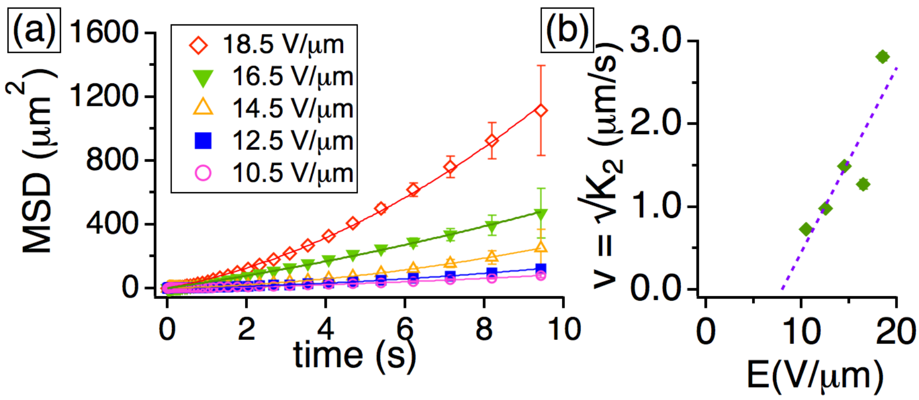

FIG. 1: **Drop Dynamics.** (a) Mean-squared displacement as a function of time shows a roughly linear dependence at intermediate fields and quadratic dependence at higher fields. (b) The advective term can be obtained from the fits in (a). A linear fit is shown.

III. FIELD THRESHOLDS

Two field thresholds are identified from the thickness dependence study and shown in FIG. 2. The first is the field threshold, as a function of cell thickness, where drop breakup occurs. The second is the field threshold, as a function of cell thickness, when drops are displacing and changing shape rapidly enough that particle tracking is not feasible.

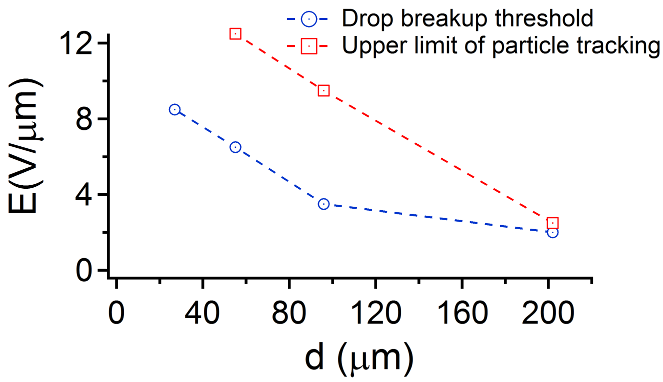

FIG. 2: **Field Thresholds.** Field thresholds for drop breakup and for vigorous drop motion (evaluated by inability to track drops) as a function of cell thickness. For the $d = 27$ μm cell, particle tracking is always possible.

IV. CONVECTION WITH DROPLETS AND PMMA COLLOIDS

Movie 3 shows convection of droplets (left panel, $E = 4.5$ V/μm) and particles (PMMA colloids, right panel, $E = 2.4$ V/μm) close to the instability threshold.

V. MIXING OF DYED AND NON-DYED CASTOR OIL IN THE PRESENCE OF THE ELECTRIC FIELD

Movie 4 shows the dynamics of the interface, obtained with time-lapse macro scale photography and white light illumination, between dyed castor oil (top) and non-dyed castor oil (bottom), without and with an applied electric field. Left panel: With $E = 0$ V/μm, there is no distinguishable motion of the interface over a period of 3 hours. Right panel: with the field on, $E = 8$ V/μm, rapid blurring of the interface between the dyed and non-dyed regions is observed over a time scale of ≈ 15 minutes.