Electrohydrodynamics-driven droplet dynamics in an oil-in-oil emulsion

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NSERC CRSNG

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Background

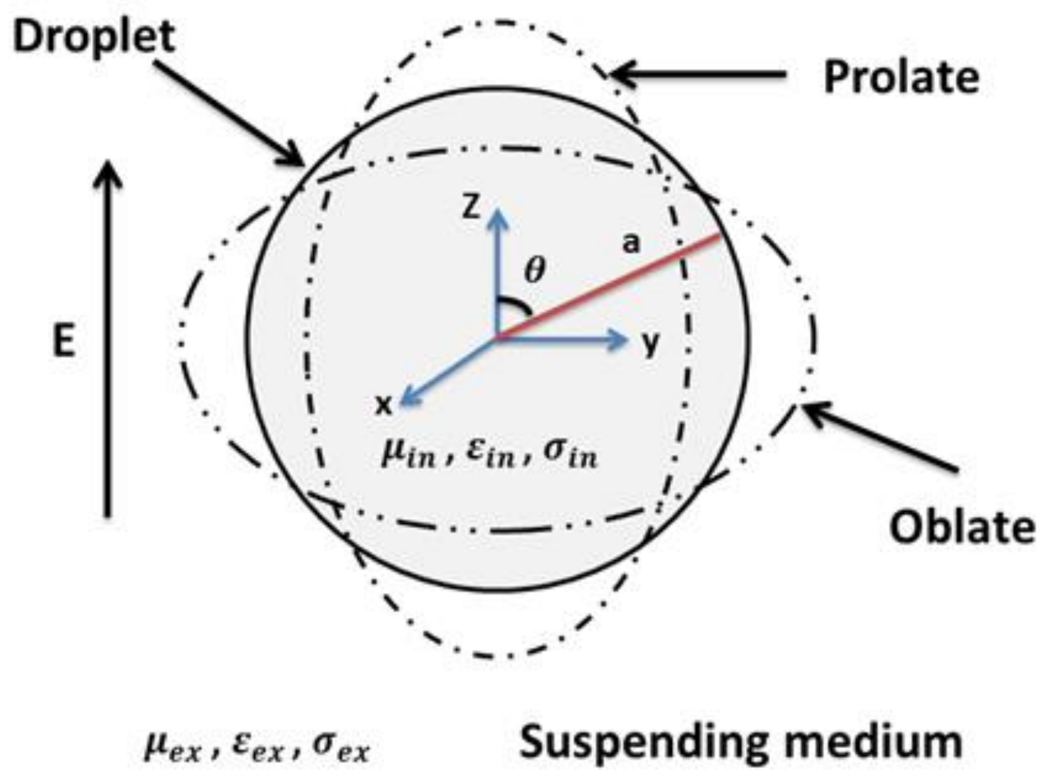
Leaky dielectric liquid:

A dielectric liquid with some free charge carriers, ions.

Leaky dielectric model (LDM):

Leaky dielectric liquid imports **normal** and **tangential** stresses which in turn creates **prolate** or **oblate** deformations.

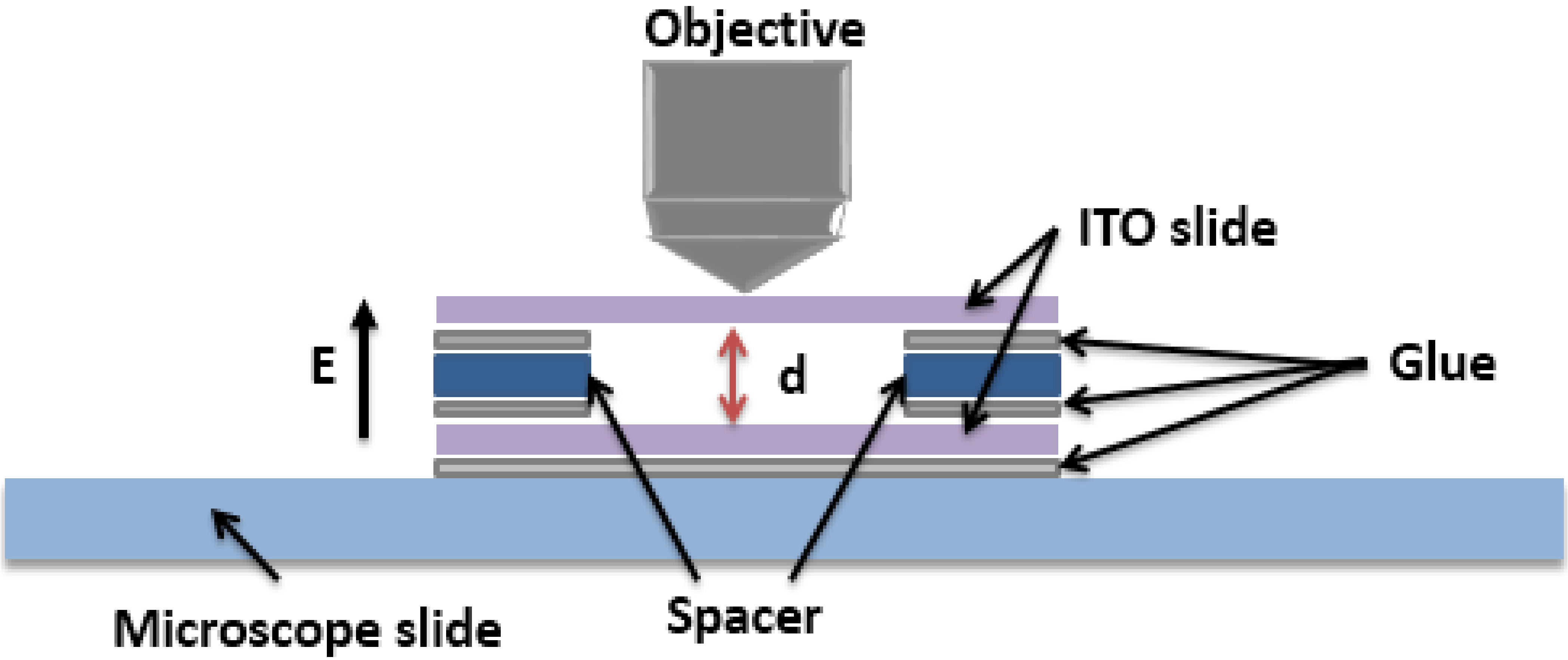
- ✓ Melcher & Taylor, Ann. Rev. Fluid Mech. 1969.
- ✓ Saville. Ann. Rev. Fluid Mech. 29:27-64, 1997.
- ✓ E. K. Zholkovskij, J. H. Masliyah, and J. Czar- necki. J. Fluid Mech., 472:1–27, 2002.
- ✓ P. F. Salipante & P. M. Vlahovska. Phys. Fluids, 22:112110, 2010.



- ❖ Angular variation of stresses at the interface
  ----------> shape deformation

- ❖ Strength of the applied electric field
  --------> breakup and instability

Experimental Setup



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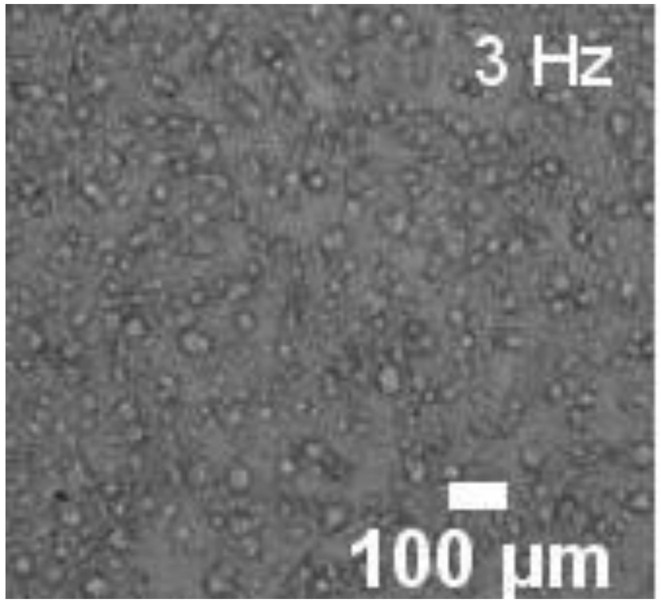
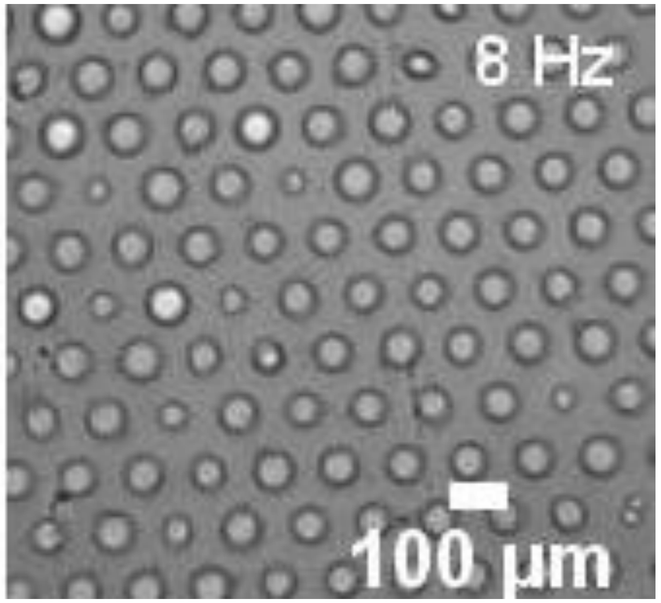
Electro-crystallization and electro-melting





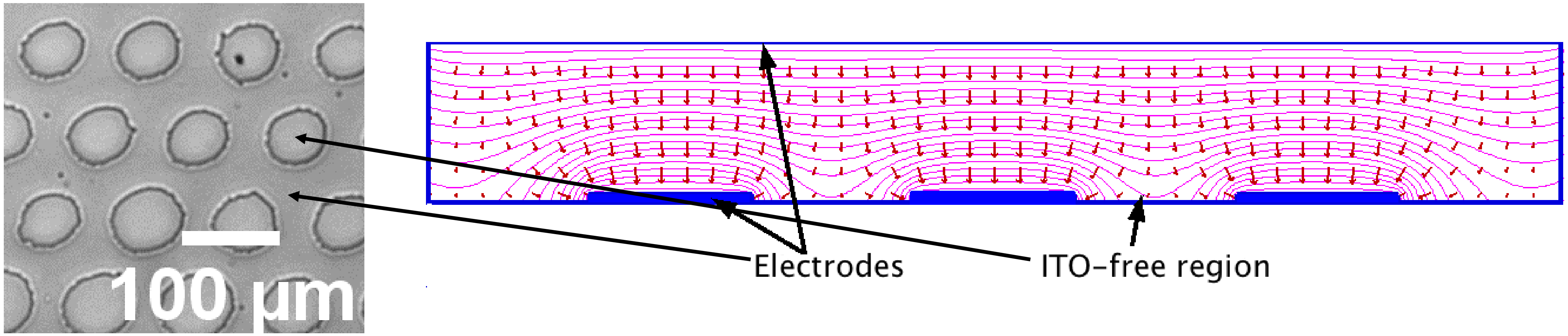
Electro-crystallization and electro-melting
An emulsion of silicone oil drops in leaky dielectric castor oil
AC electric field
d = 140 μm
Stroboscopic imaging White light microscopy Fluorescence microscopy

Goal

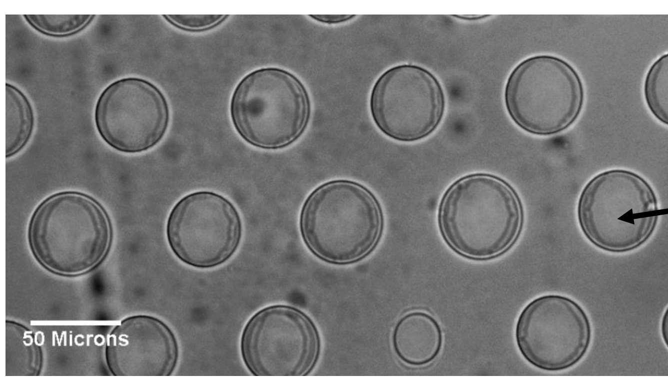
Construction of a phase diagram of an order to disorder non-equilibrium phase transition by two control parameters, f and E.

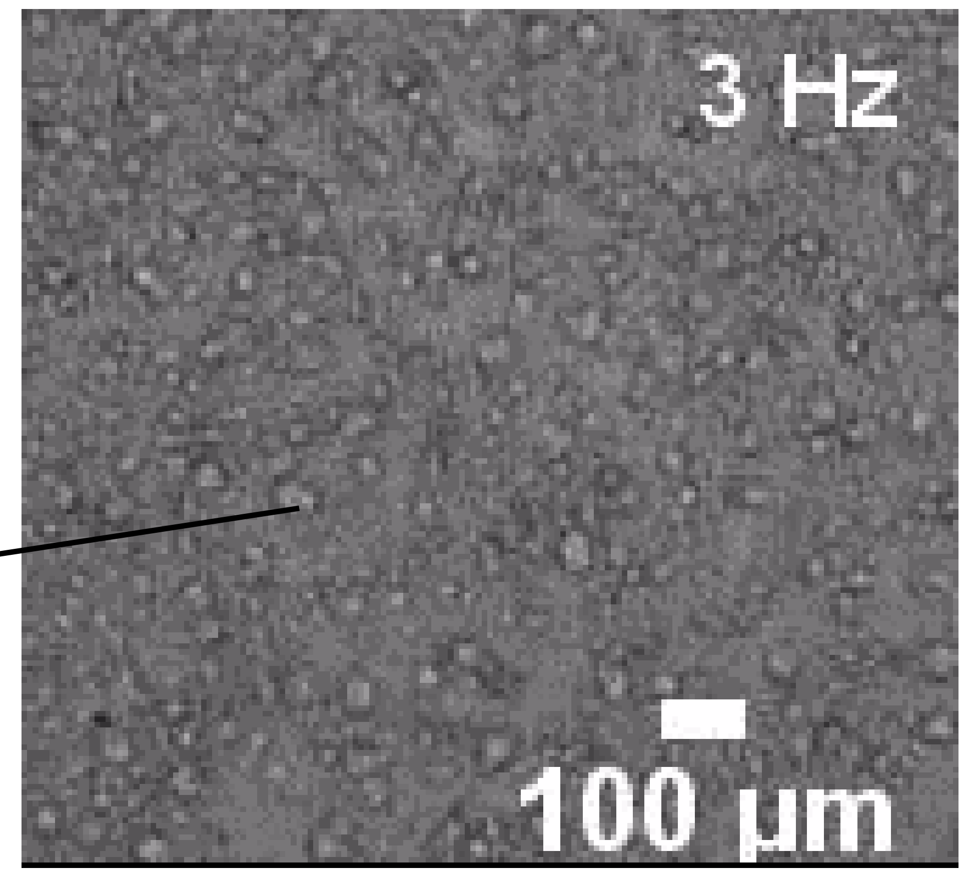
S. Khajehpour Tadavani, A. Yethiraj, Tunable hydrodynamics: A field-frequency phase diagram of a non-equilibrium order-to-disorder transition, Submitted, 2017

Pattern Formation by Negative DEP Force



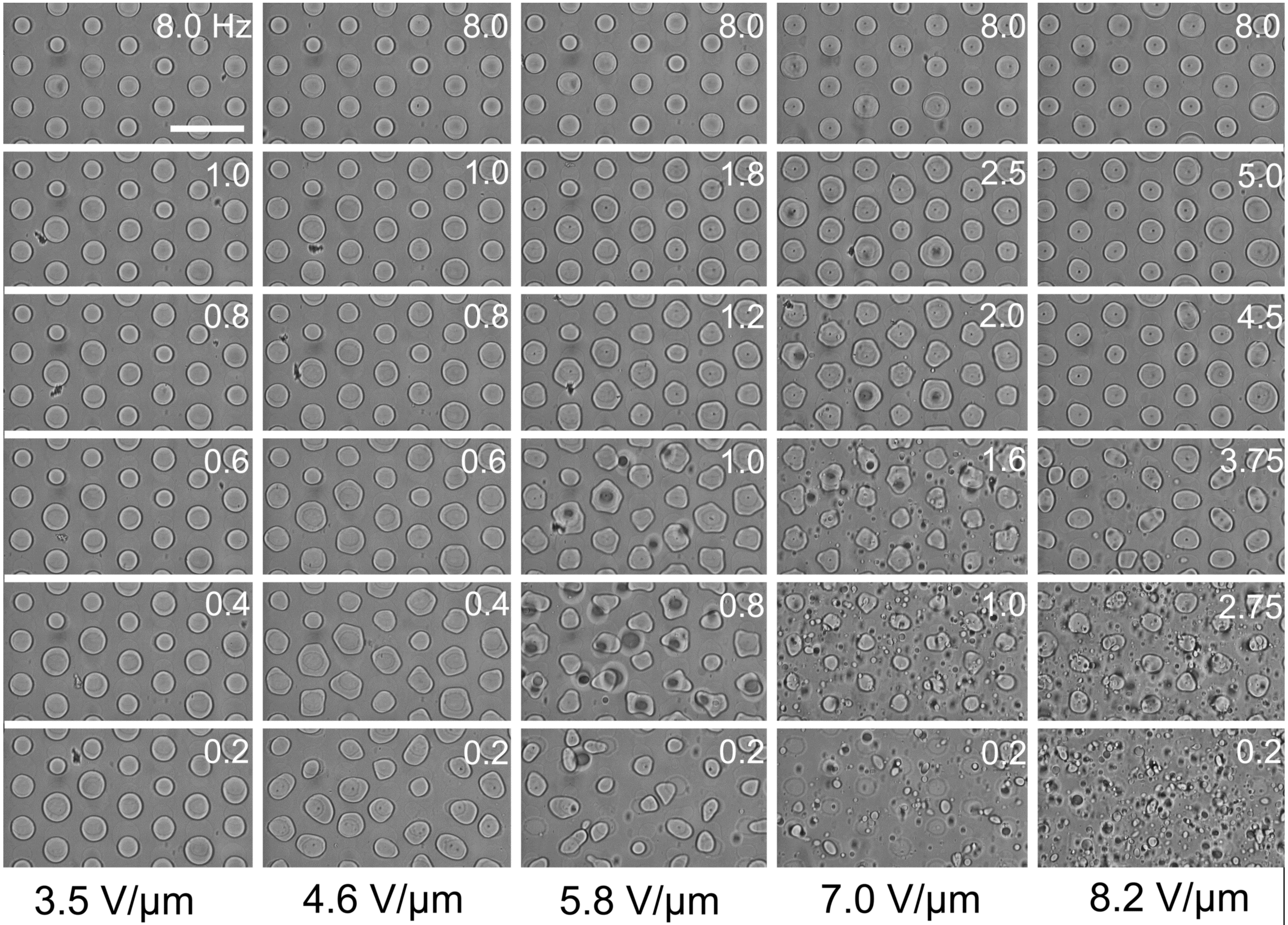
$$F_{DEP} = \frac{1}{2}\left(\varepsilon_{in} - \varepsilon_{ex}\right)\nabla\left(\mathbf{E}\cdot\mathbf{E}\right)$$



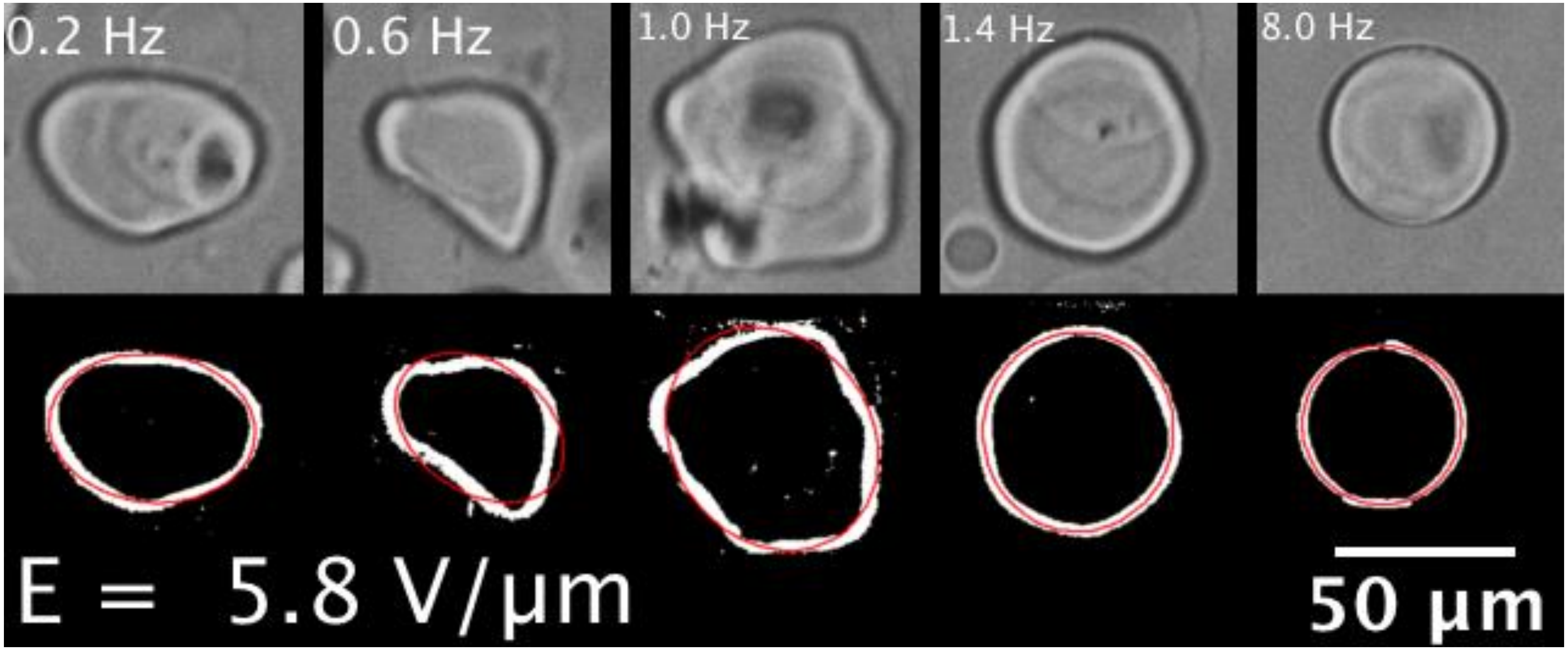


A. Varshney, S. Gohil, S. Khajehpour Tadavani, A. Yethiraj, S. Bhattacharya, S. Ghosh, Lab on a Chip, 2014, 14, 1330

Shape Deformation (Optical Micrograph)



Analysis: A Fit-ellipse Method



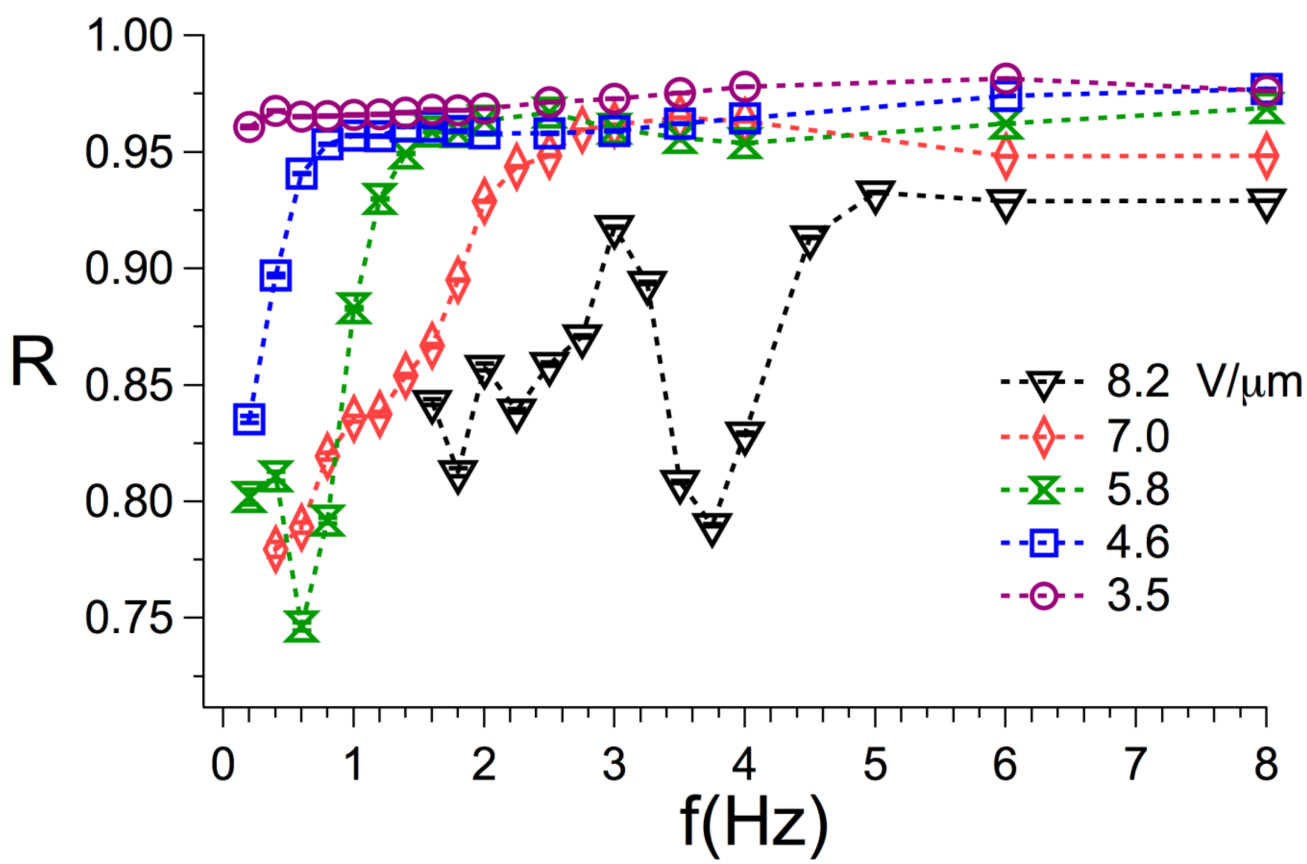
The output consists of

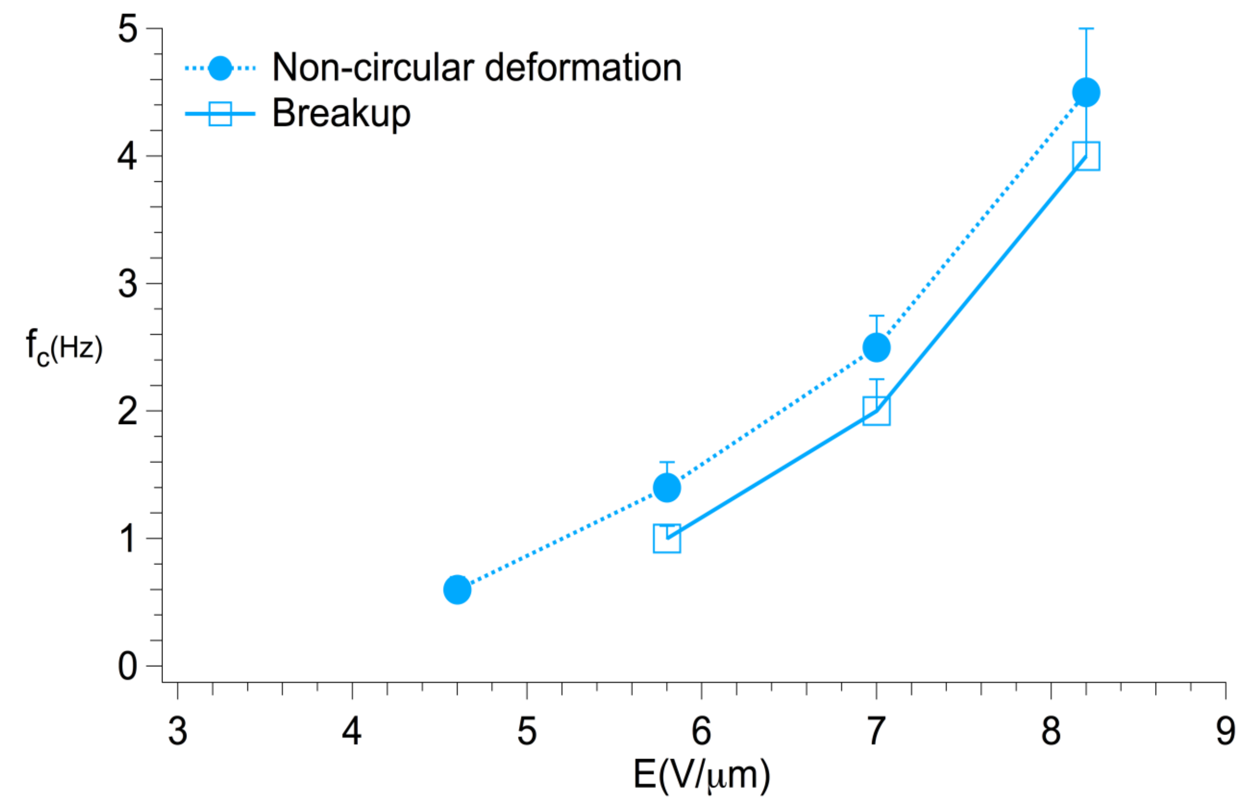
1. Major axis (M)
2. Minor axis (m)
3. Centroid (x,y)
4. Angle of the major axis with respect to the x-axis (θ_x).

A Quantitative Study of Shape Deformation

Fit with ellipse ⟶ $$R = \frac{4 \times Area}{\pi \times (Major\ Axis)^2}$$

For a perfect circle drop R = 1





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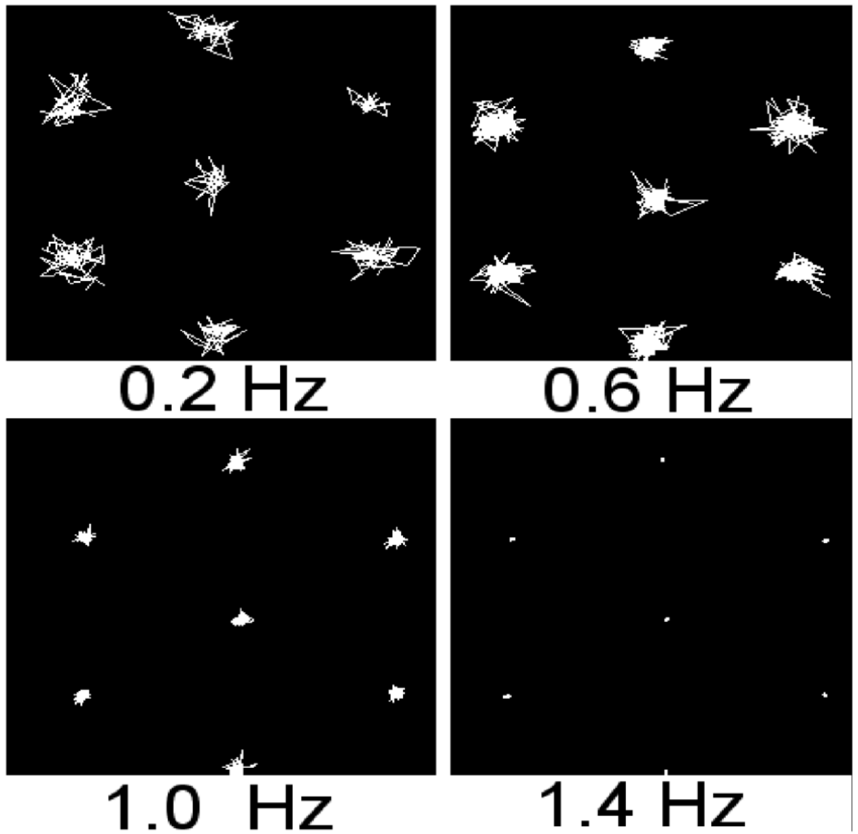
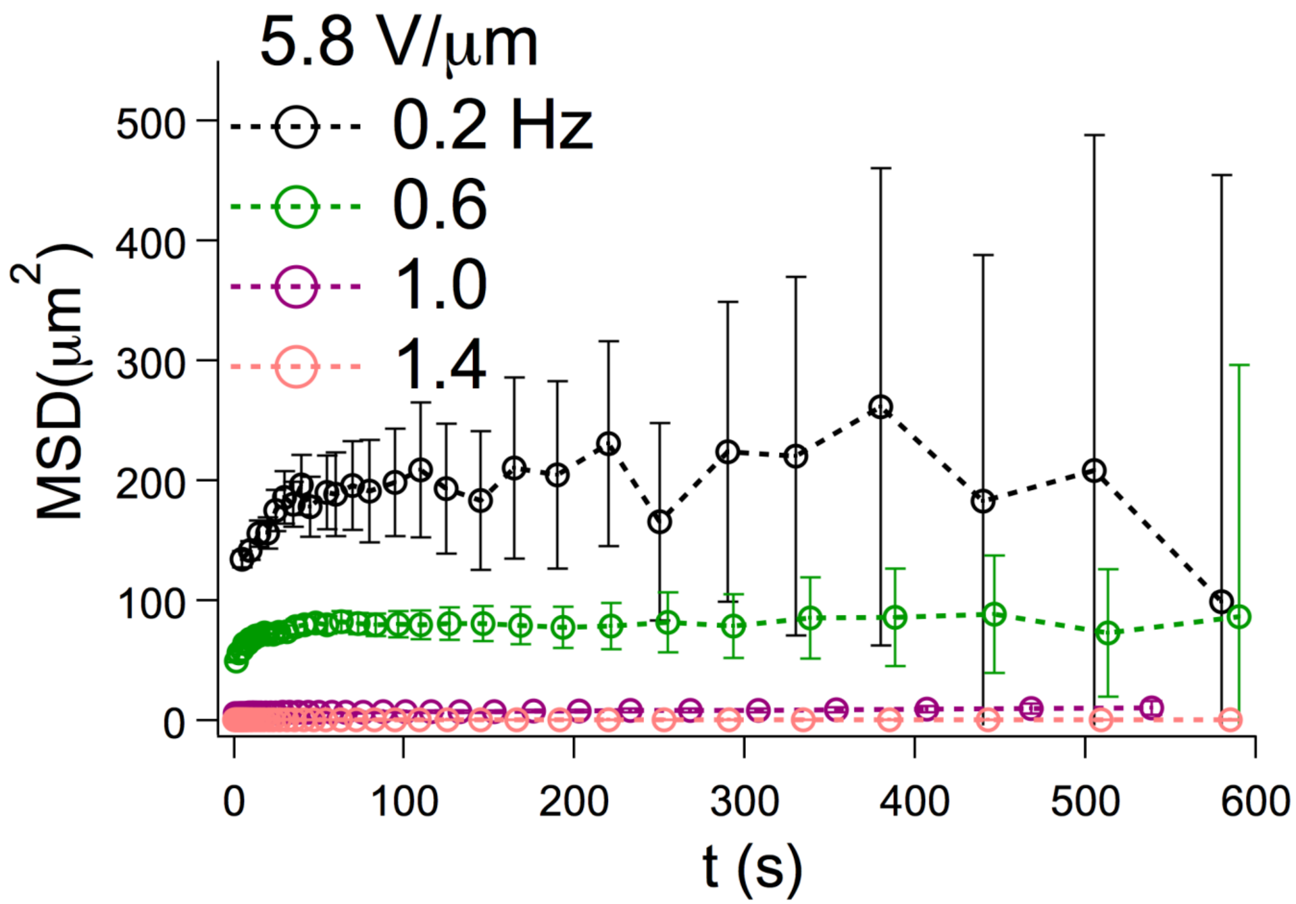
Dynamics of the Array





In 2D $$MSD = <\Delta r^2> = \frac{\beta k + \frac{1}{4Dt}}{\left(\frac{1}{2}\beta k + \frac{1}{4Dt}\right)^2}$$

βk : Potential well's curvature , D: Effective diffusion coefficient

β: $(k_B T)^{-1}$, k: Spring constant

J. A. Weiss, A. E. Larsen, and D. G. Grier. J. Chem. Phys. 109(19):8659–8666, 1998.

Dynamics of the Array

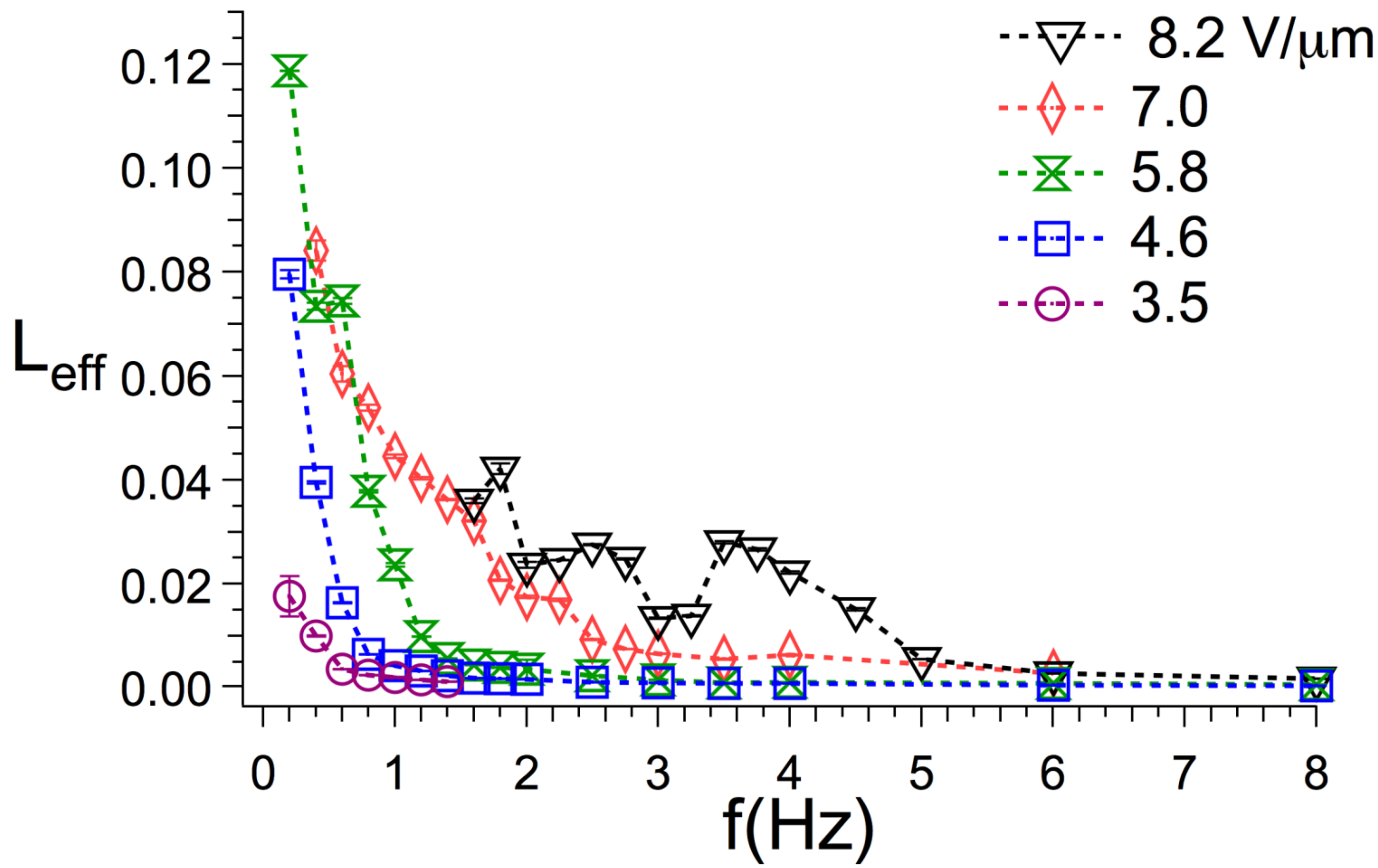
$$t \to \infty \leftrightarrow \ < \Delta r^{\,2} > \ = \frac{4}{\beta k}$$

Effective Lindemann parameter $\boldsymbol{L_{eff}}$: A Dimensionless measure of drop fluctuations

$$\boldsymbol{L_{eff}} = \frac{\mathbf{1}}{\boldsymbol{d_{nn}}}\sqrt{\frac{\mathbf{3}}{\mathbf{4}} < \Delta r^{\,2} >_{t\to\infty}}$$

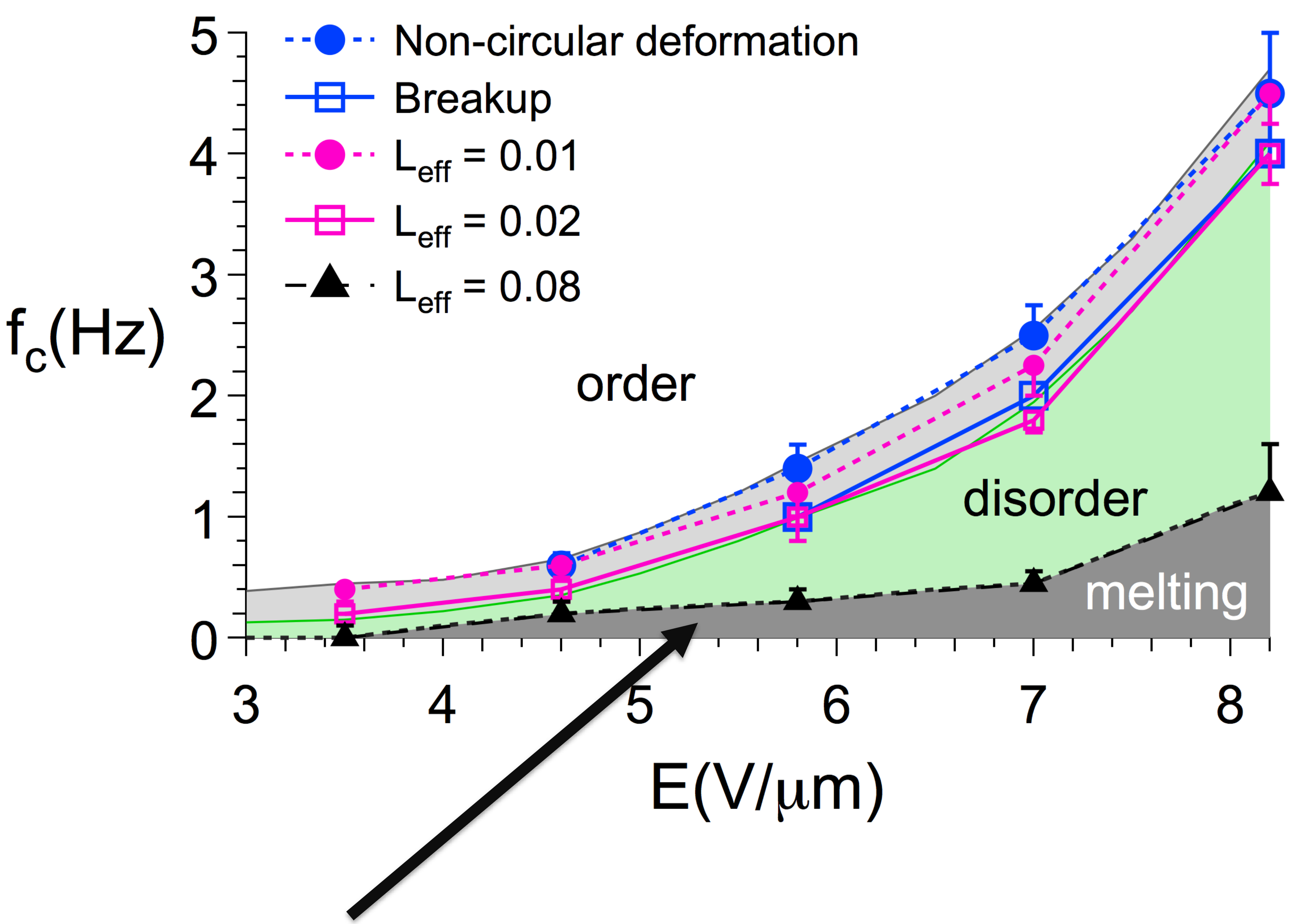
$d_{nn} = 100\ \mu m$, is the crystal nearest-neighbor distance

A. M. Alsayed, M. F. Islam, J. Zhang, P. J. Collings, and A. G. Yodh. Science, 309(5738):1207–1210, August 2005.



Non-equilibrium Phase Transition: A Phase Diagram

- Onset of melting in 2D lattice ------> $L_{eff} \geq 0.1$ (in unit of $\boldsymbol{d_{nn}}$)

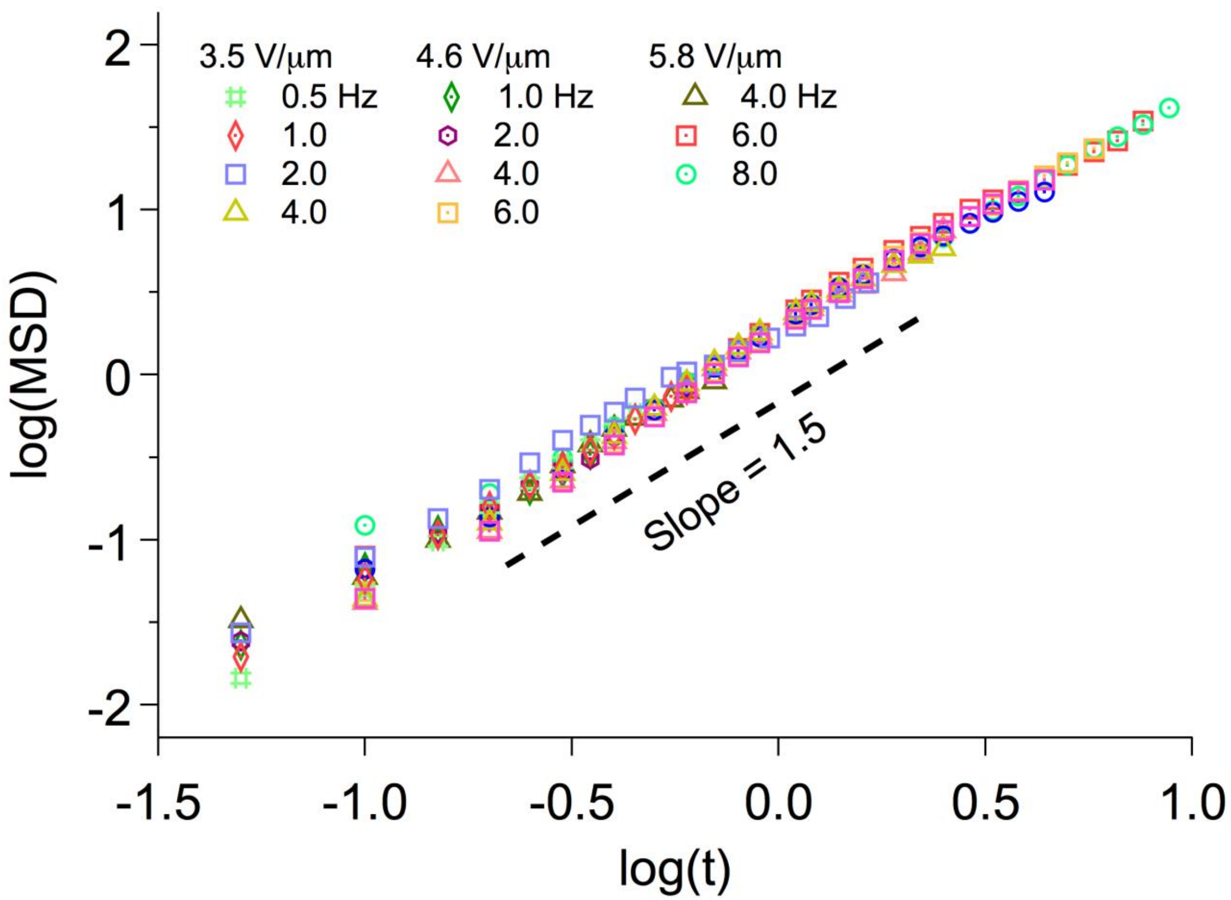


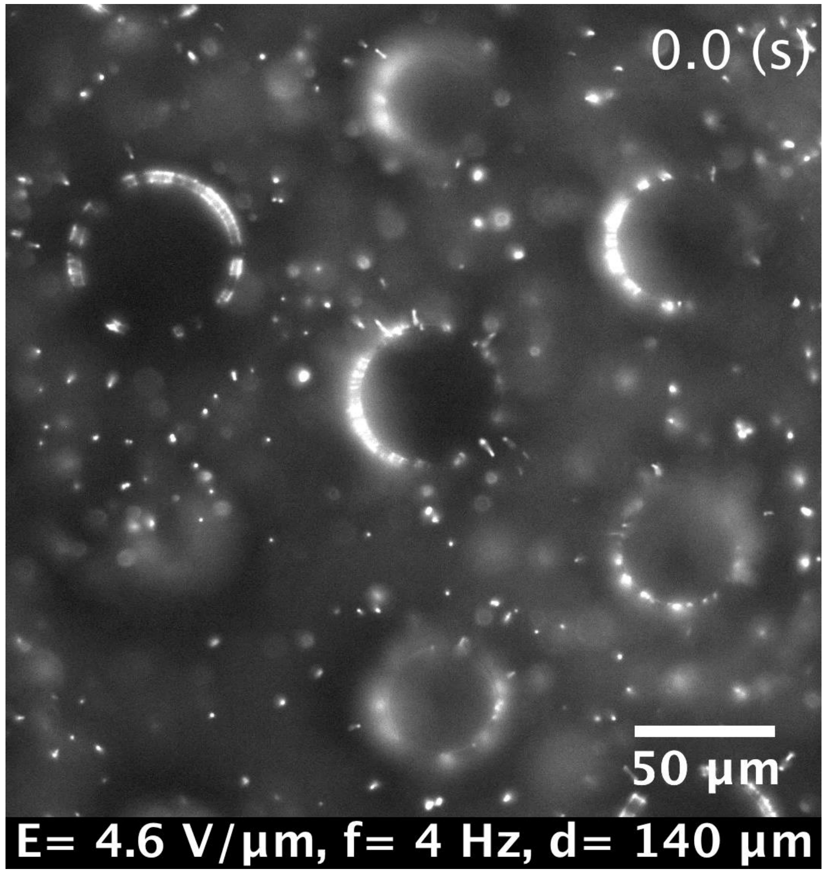
$L_{eff} \approx$ 0.08 – 0.12 (dark grey region) corresponds to a classic criterion for melting.

Mechanism of the Non-equilibrium Thermodynamics: Pseudo- Random Fluid Velocity Fields

$$MSD \propto t^{3/2}$$

$$\text{Log(MSD)} \propto 1.5 \log(t)$$





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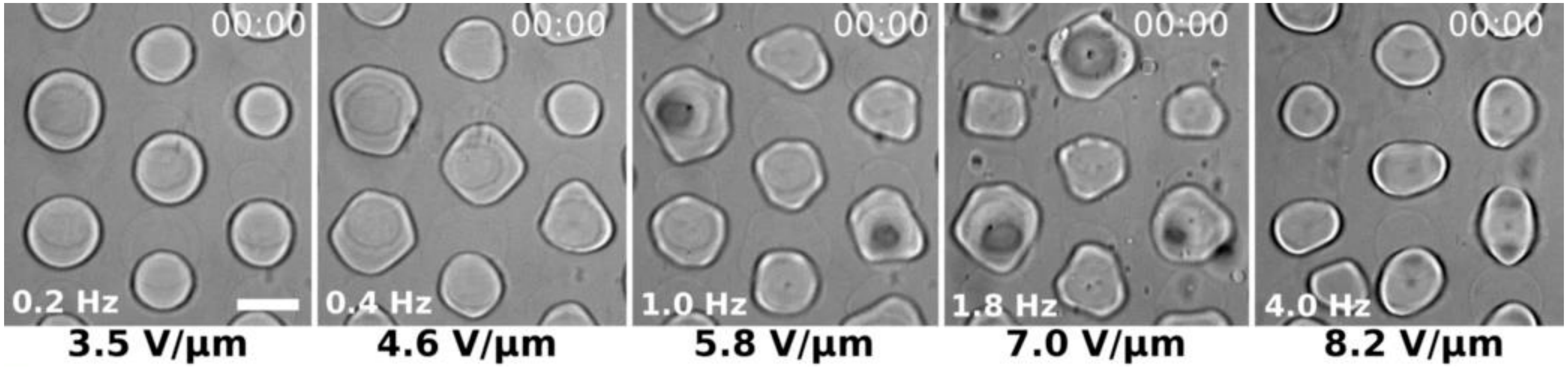
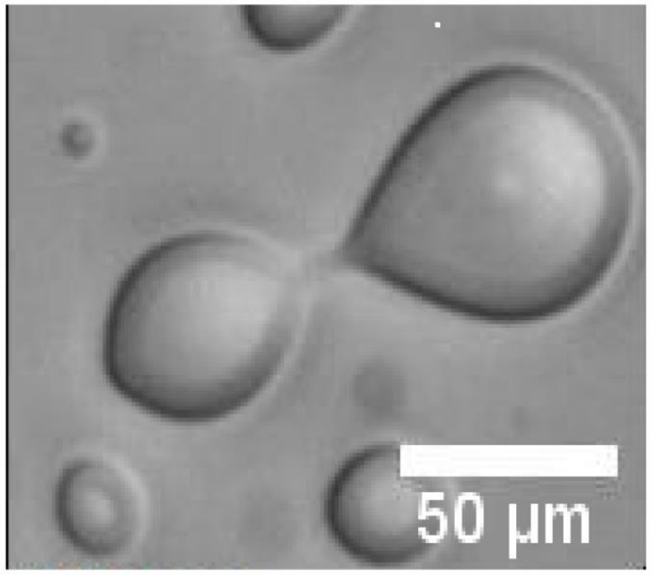
Summary

1. Dynamics and shape deformation of a roughly mono-disperse array of droplets are control with f and E.
2. A phase diagram of an order to disorder non-equilibrium phase transition is constructed.
3. The mechanism of the non-equilibrium thermodynamics is related to fractional, super-diffusive dynamics in the outer fluid of the oil-in-oil emulsion.

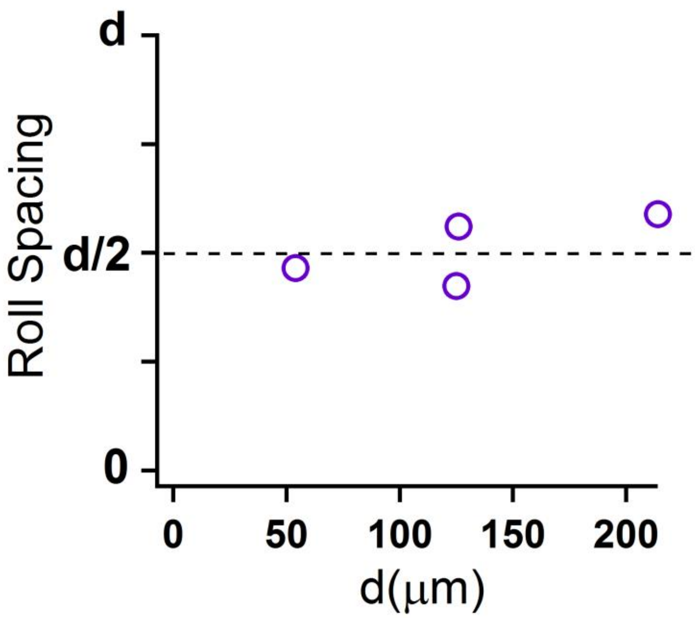


Part 2: Dimensionality and Convective Instability

Breakup



Instability



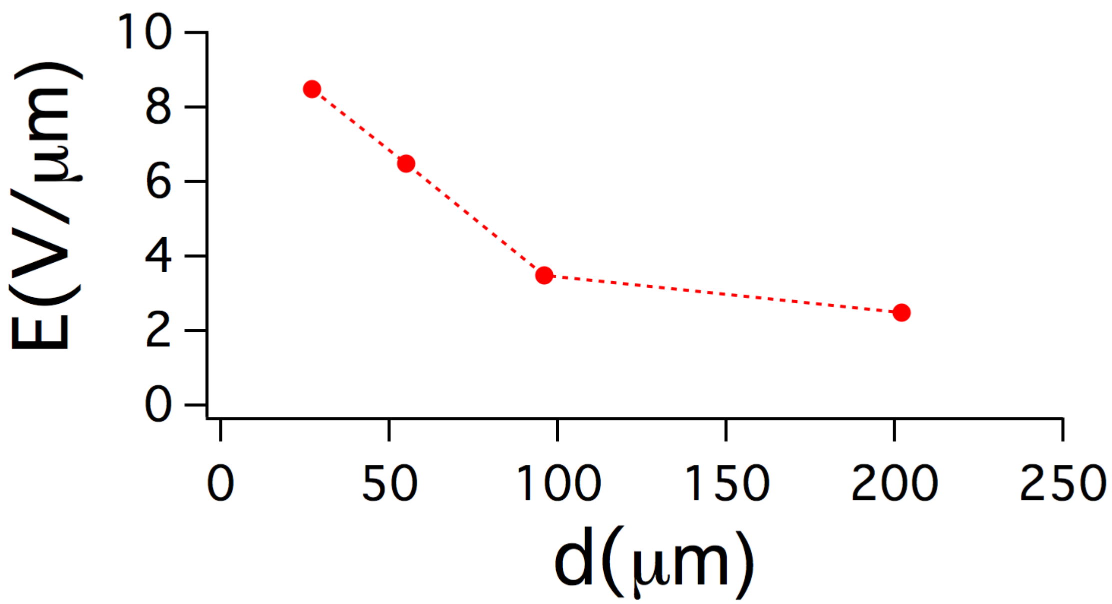
Dimensionality and multi-scale flow
An emulsion of silicone oil drops in castor oil
DC electric field
d = 27, 55, 96, 202 μm
White light microscopy Fluorescence microscopy

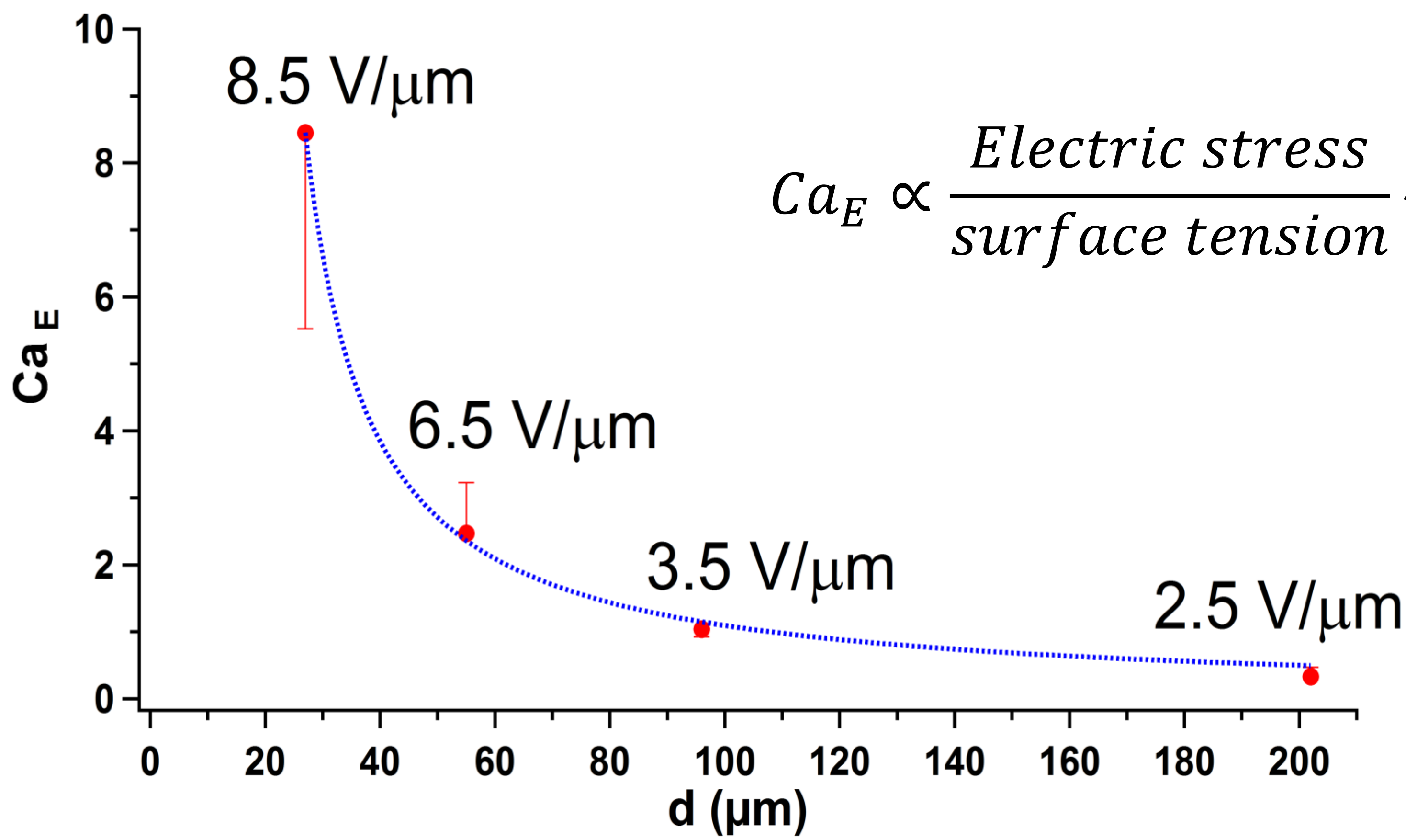
Goal

Effect of cell thickness on the breakup of drops and convective instability.

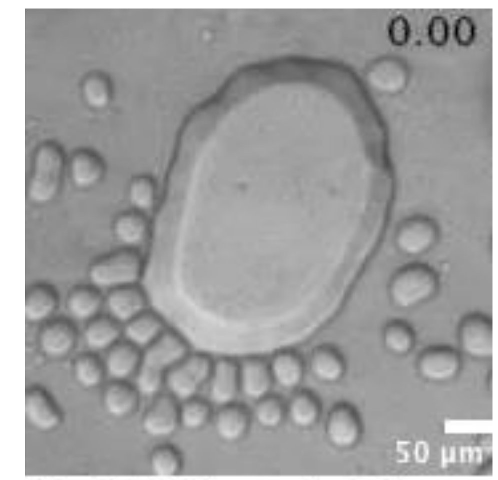
S. Khajehpour Tadavani, J. R. Munroe, and A. Yethiraj. The effect of confinement on the electrohydrodynamic behavior of droplets in a microfluidic oil-in-oil emulsion. Soft matter, 12(45):9246–9255, 2016.

Role of Substrate in Breakup

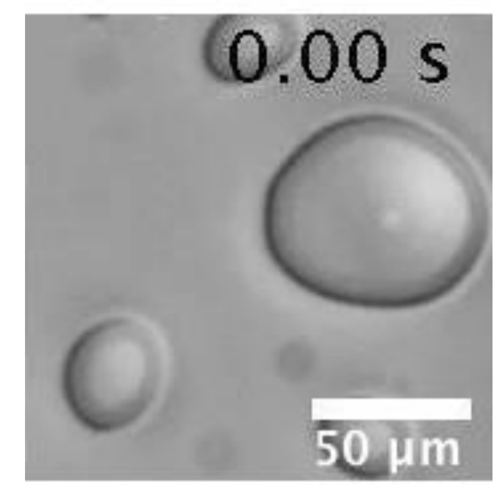




$$Ca_E \propto \frac{Electric\ stress}{surface\ tension} \sim \frac{aE^2}{\gamma}$$



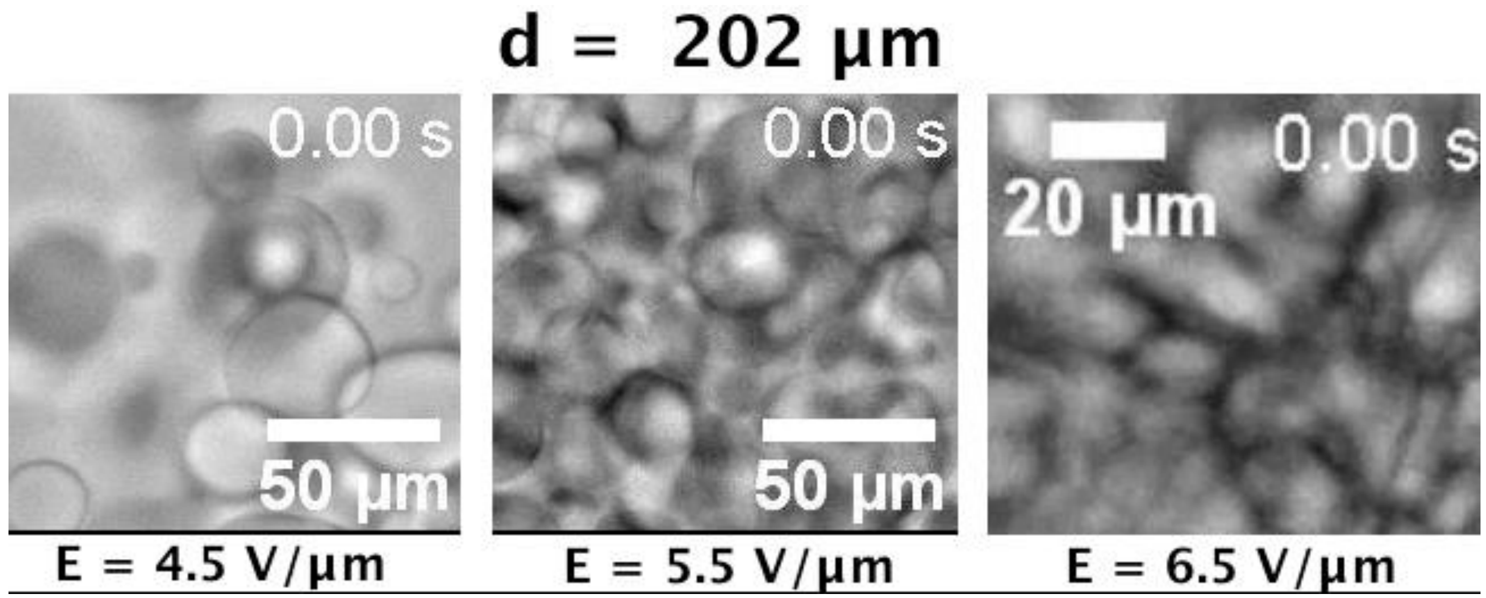
E = 9.5 V/μm, d= 27 μm

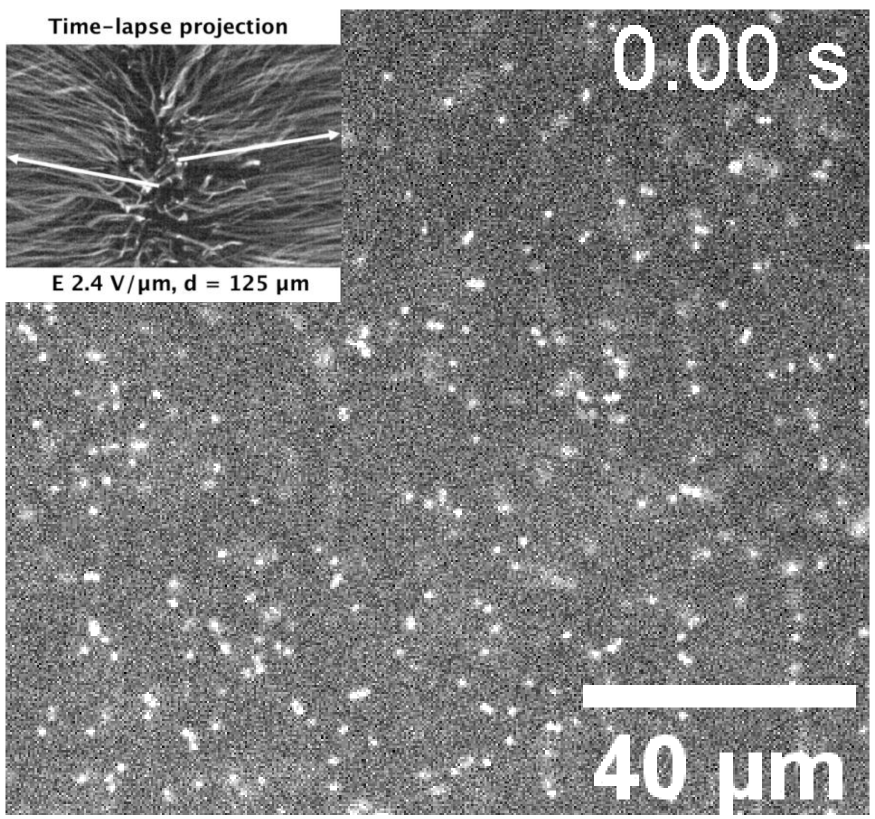


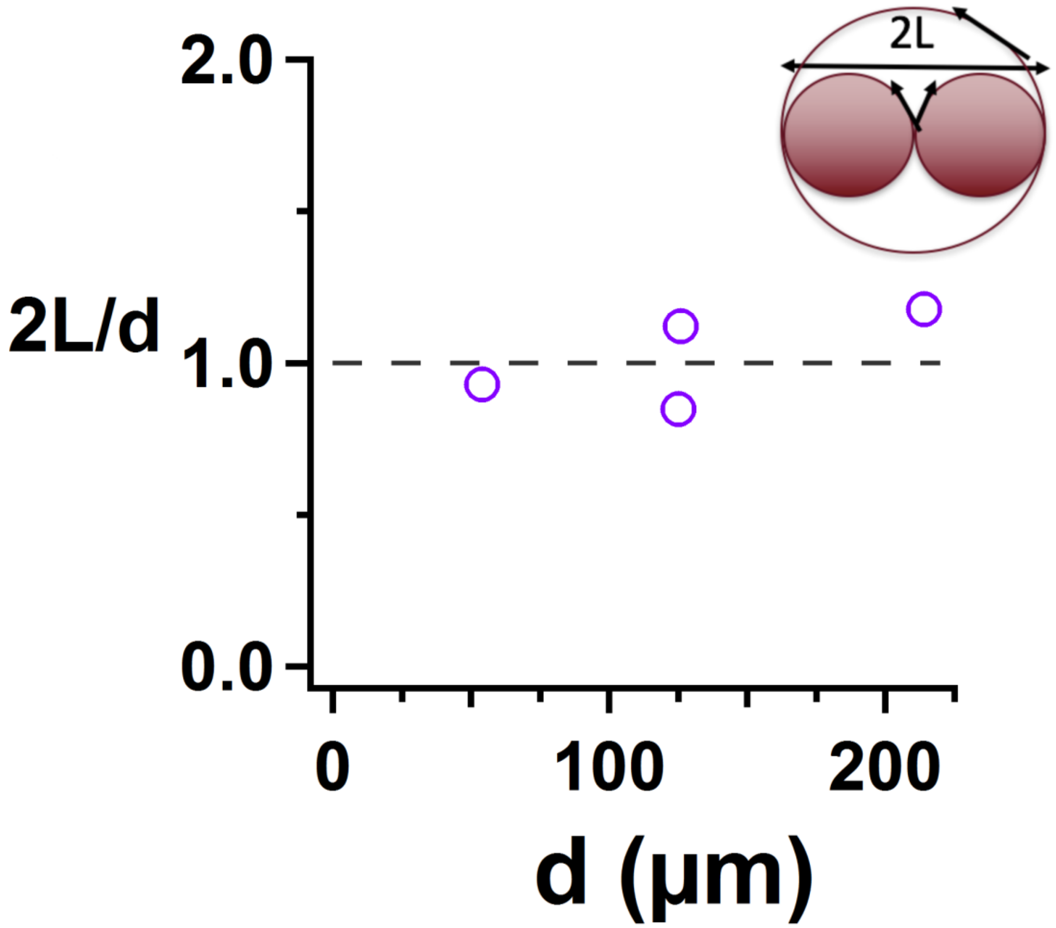
E = 8.5 V/μm, d= 55 μm

Substrate interactions are less important above 100 μm.

Prior Instability: Evidence of a Convective (Rayleigh-Benard like) Instability







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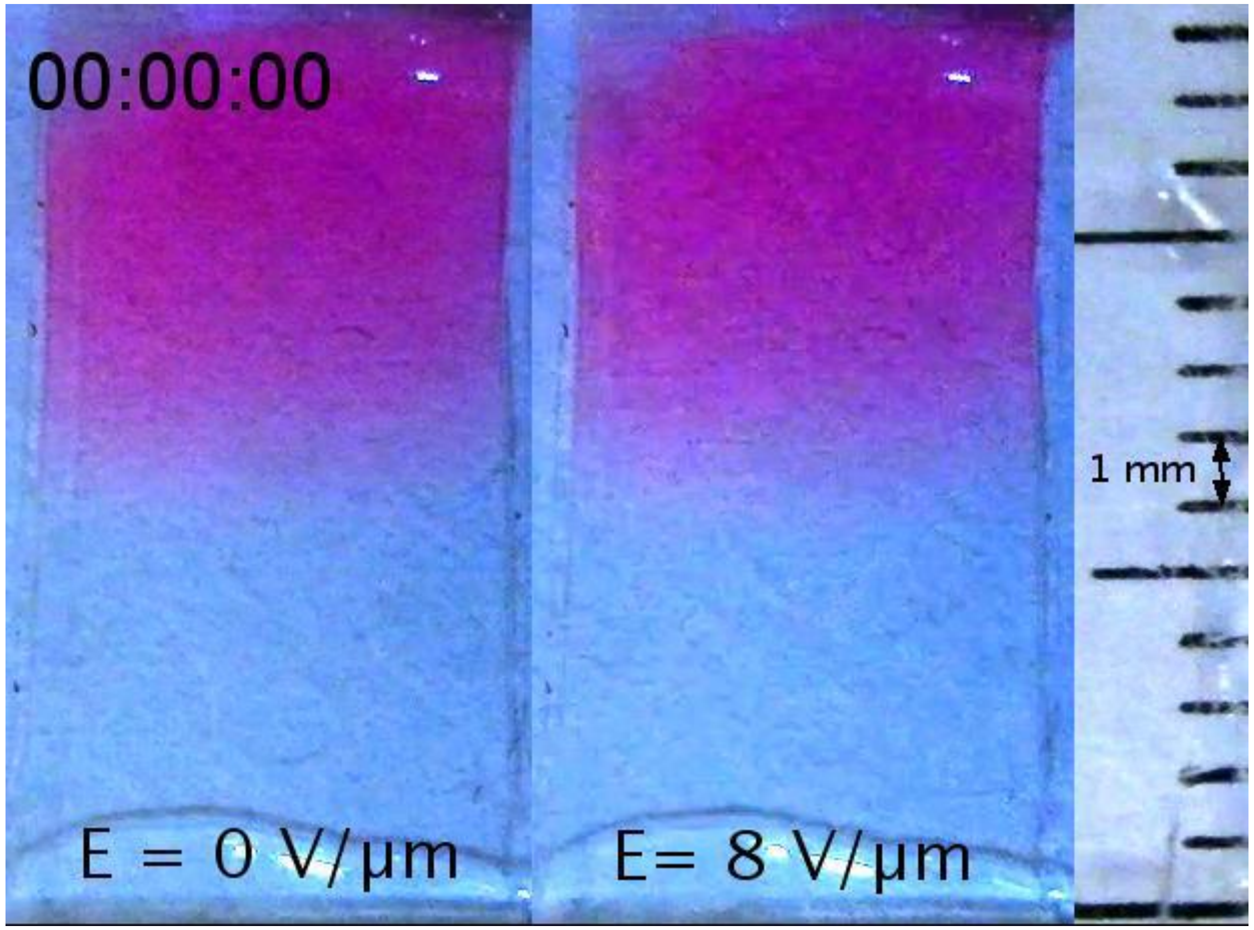
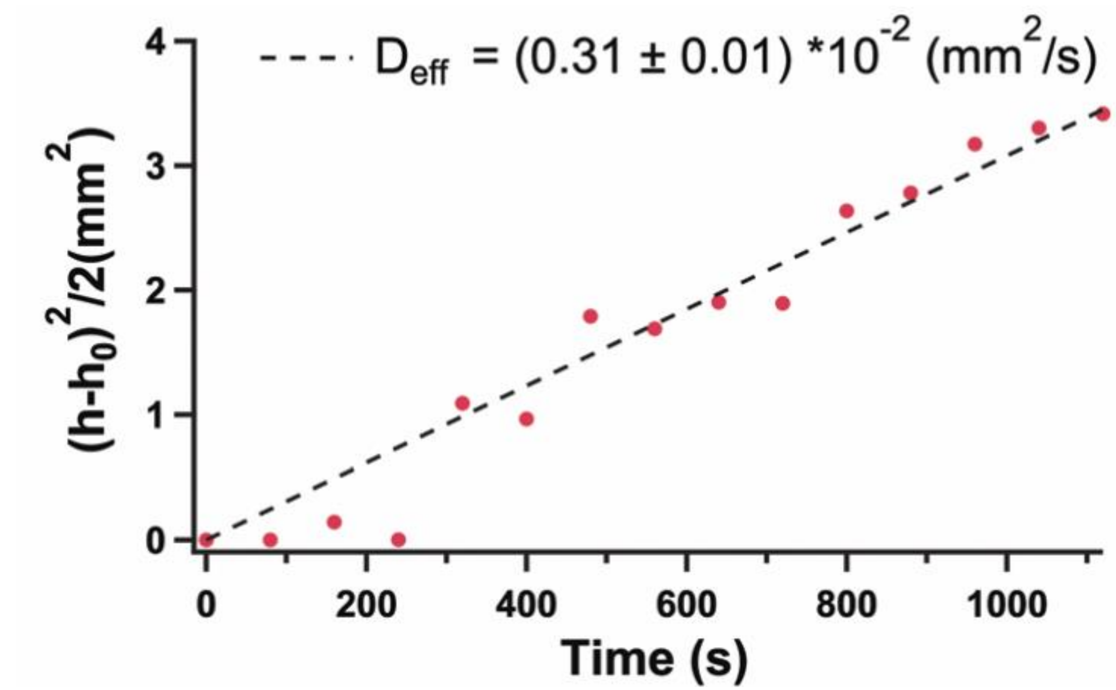
Vertical Overturning Flow: Validity of the Leaky Dielectric Picture?!



$$2D_{\text{eff}}^{E\neq 0}t = (h - h_0)^2$$

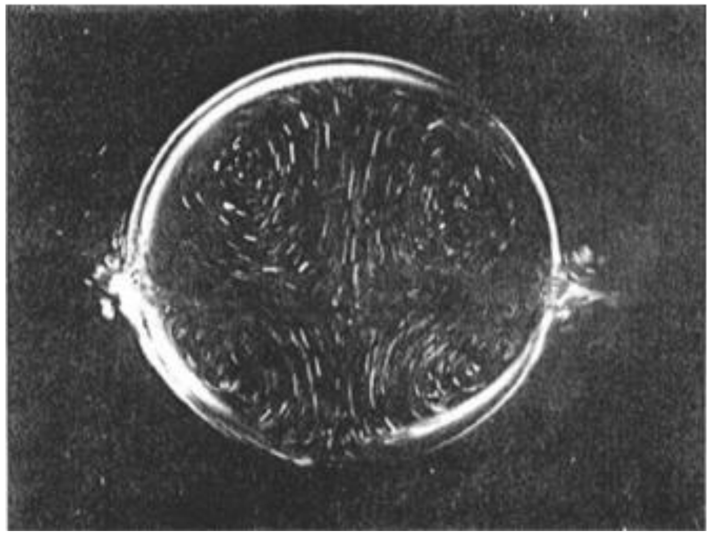


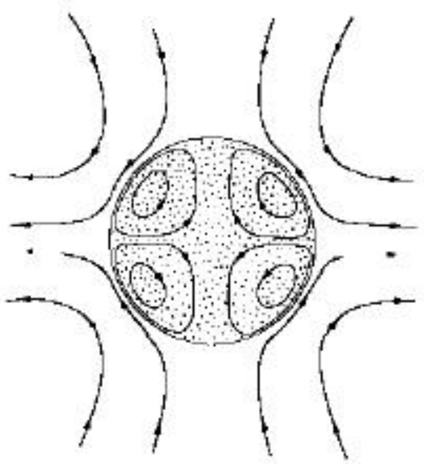
Enhanced dynamics, 5500 times faster, in the presence of the field.

$$D_{E=0} \cong 0.56 \times 10^{-6}\,\frac{mm^2}{s}, \quad D_{E\neq 0} \cong 0.31 \times 10^{-2}\,\frac{mm^2}{s} \;\rightarrow$$

$$\frac{D_{E\neq 0}}{D_{E=0}} \sim 5500$$

Part 3: Underlying Flow Mechanism





J. R. Melcher and G. I. Taylor. Annu. Rev. Fluid Mech., 1(1):111–146, 1969.

Underlying Flow Mechanism
Emulsion seeded with PMMA particles
DC and AC electric field
Differnt thicknesses
White light microscopy Fluorescence microscopy

Upshot:

A study of the effective thermodynamics.

Summary

Part 1: Electro-crystallization and electro-melting

1. Dynamics and shape deformation of a roughly mono-disperse array of droplets are control with f and E.
2. A phase diagram of an order to disorder non-equilibrium phase transition is constructed.

Part 2: Dimensionality and convective instability

1. Ca_E at the threshold of drop breakup is of order unity for cell thicknesses of 100 μm and thicker, but much larger for thinner cells.
2. As field is increased, convective instability precedes onset of strong hydrodynamics regime
3. Enhanced dynamics even in the absence of drops or particles: a mechanism beyond the leaky dielectric model.

Part 3:Underlying Flow Mechanism

1. The mechanism of the non-equilibrium thermodynamics is related to fractional, super-diffusive dynamics in the outer fluid of the oil-in-oil emulsion.

Focuses of Interest

Part 1: Electro-crystallization and electro-melting.

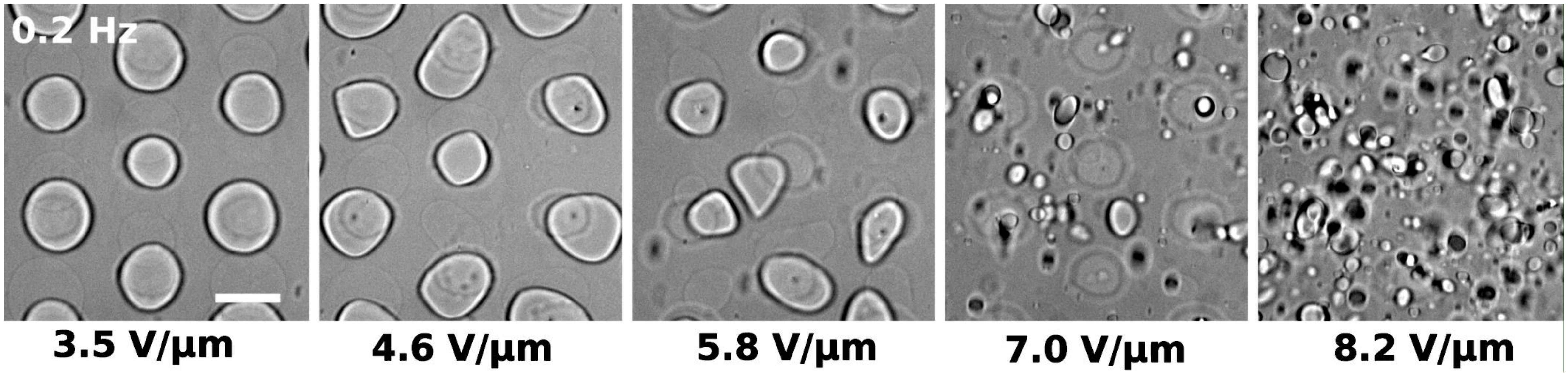
- ❖ Studying shape deformation and dynamics of an array of roughly monodisperse drops by tuning two control parameters, amplitude and frequency.
- ❖ Construct a phase diagram of an order to disorder non-equilibrium phase transition.

Part 2: Dimensionality and convective instability.

- ❖ Effect of cell thickness on the breakup of drops and convective instability.

Part 3: Underlying Flow Mechanism.

- ❖ A study of the effective thermodynamics.


0.2 Hz
3.5 V/μm
4.6 V/μm
5.8 V/μm
7.0 V/μm
8.2 V/μm

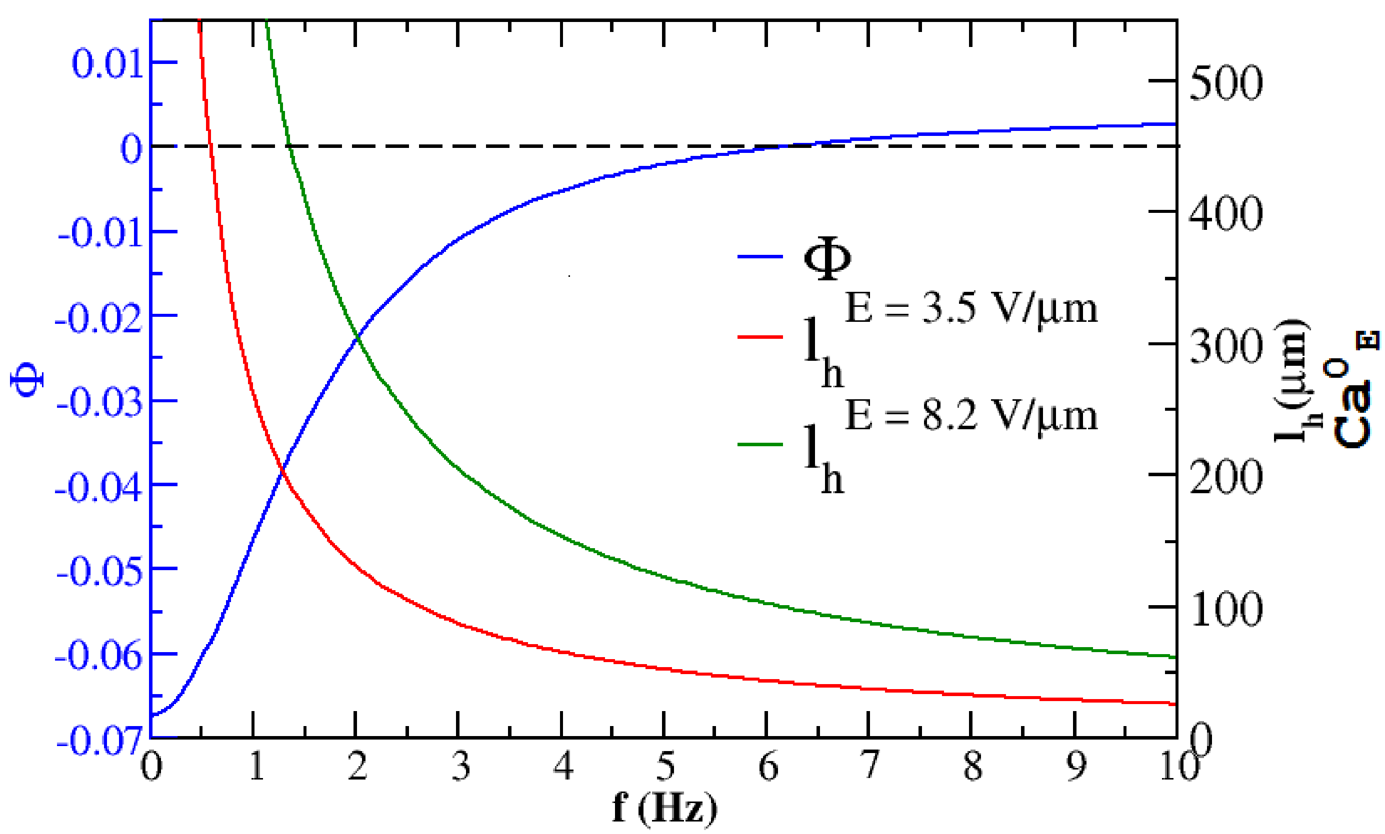
Background

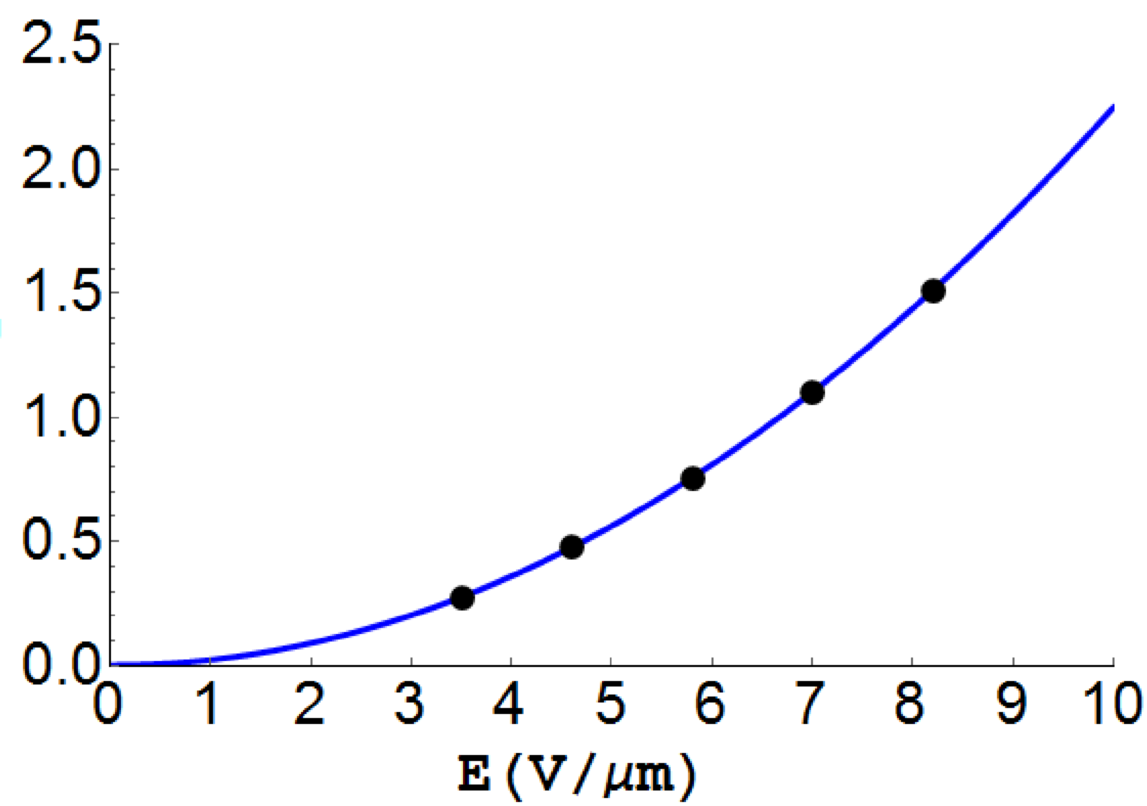
$$\Phi = \frac{9}{16}\left(1 - \frac{H^{-1}(11N+14) + H^{-2}\Big(15(N+1) + S(19N+16)\Big) + 15\tau^2\omega^2(1+N)(1+2S)}{10(1+N)\Big((2H^{-1}+1)^2 + \tau^2\omega^2(S+2)^2\Big)}\right)$$

$$S = \frac{\varepsilon_{in}}{\varepsilon_{ex}} = 0.67, H = \frac{\sigma_{in}}{\sigma_{ex}} = 0.10, N = \frac{\eta_{in}}{\eta_{ex}} = 0.17, \tau = \frac{\varepsilon_{ex}}{\sigma_{in}} = \frac{\varepsilon_0 k_{ex}}{\sigma_{in}} \simeq 0.81, \omega = 2\pi f$$

$$l_h = v_d/f, v_d = \mu_E E$$
$$\mu_E \simeq 75\ \mu\mathrm{m}^2\mathrm{V}^{-1}\mathrm{s}^{-1}$$

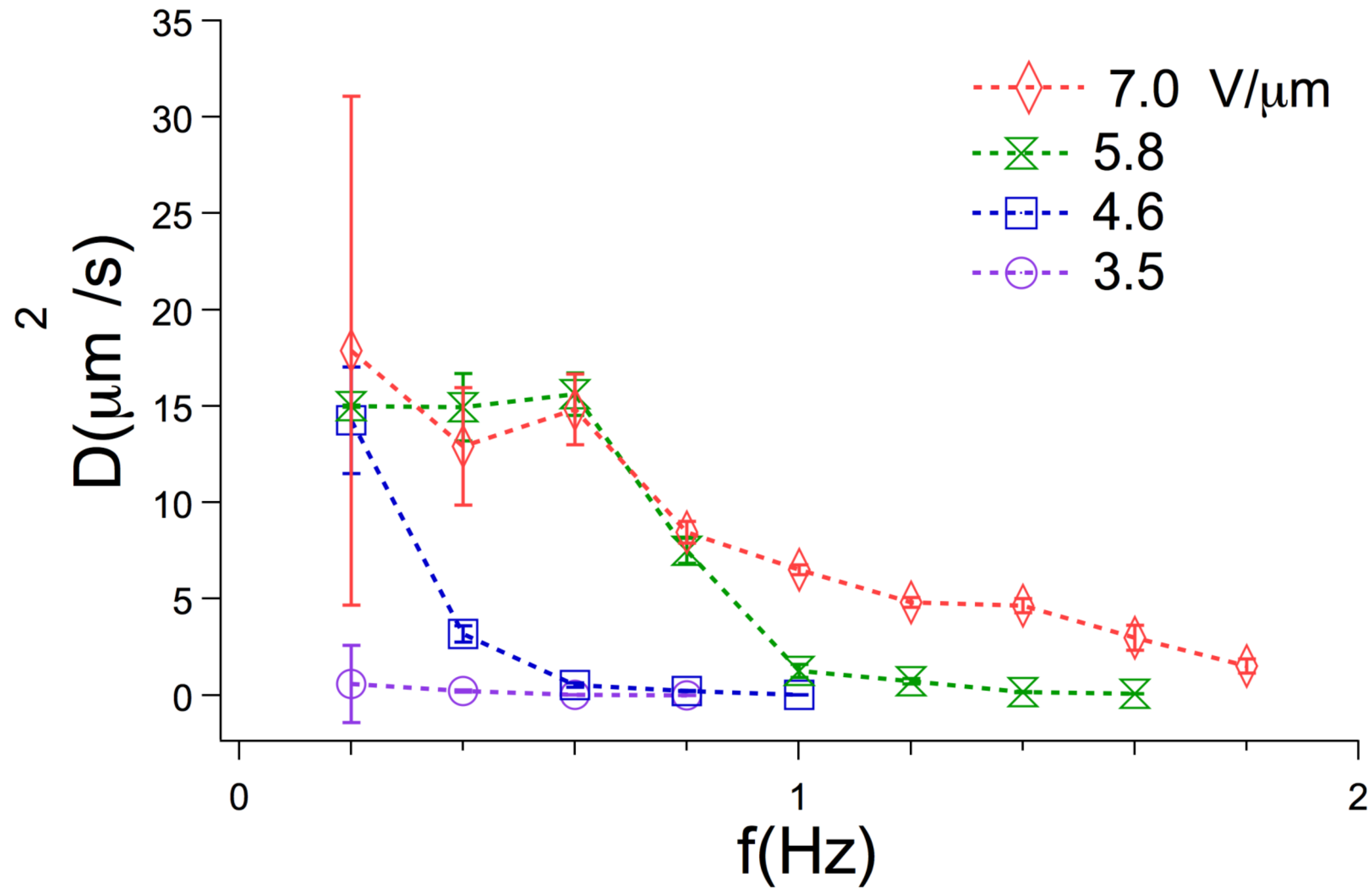
$$Ca_E^0 \equiv \frac{9\mid S^{-1}H - 1\mid N^{-1}}{10\,(H+2)^2\,(N^{-1}+1)}\,\frac{\varepsilon_{in} a E^2}{\gamma}$$





Dynamics of the Array

$t \to 0 \leftrightarrow \; < \Delta r^{2} > = 4D_{eff}t \leftrightarrow$ effective diffusivity



EHD forces are proportional to diffusive forces.

Strong Hydrodynamic regime ---------> low frequencies $(l_h = \frac{V}{f})$