Francois Frenkiel Award Lecture

Thermocapillary migration of interfacial droplets

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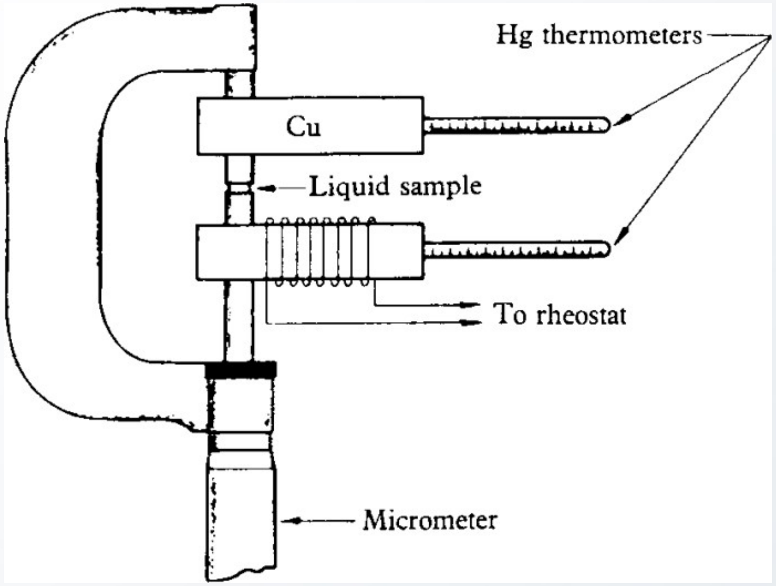
Historical Background

- Why does the drop move?
  - *The motion of bubbles in a vertical temperature gradient*
    Young, Goldstein and Block
    Journal of Fluid Mechanics 6 (3): 350-356 **1959**

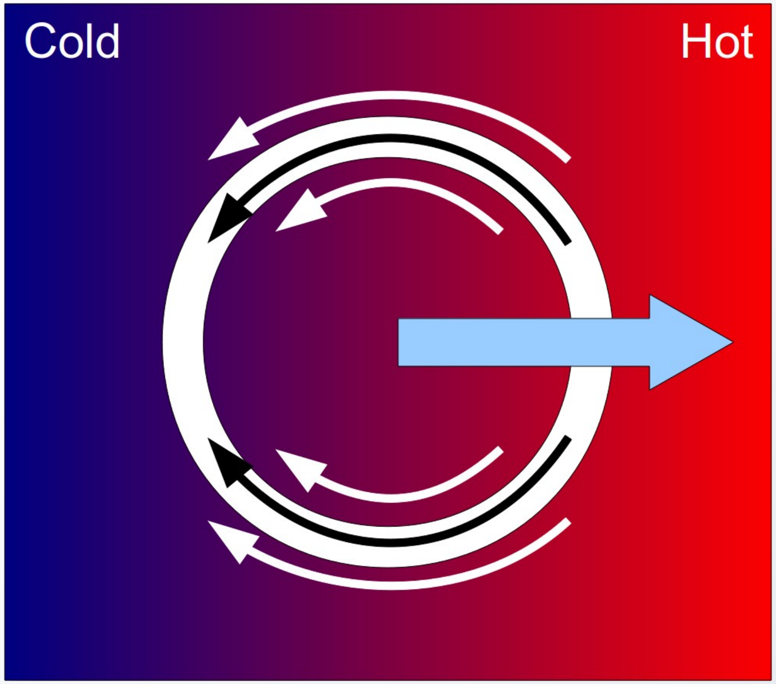
1. **Small spherical bubbles were observed to collect at the warmer anvil**
2. **By adjusting the temperature gradient bubbles could be moved up or down in the liquid sample**
3. **For a specific temperature gradient, bubbles could be suspended motionless in the sample**



The thermocapillary effect

- How was this observation explained?
  - **Surface tension is temperature dependent**

$$\sigma(T)=\sigma_0+\sigma'(T-T_0)$$



  - For most fluids, surface tension decreases with temperature
  - Variations in surface tension due to temperature gradients drive flow near the interface

$$\hat{n}\times(\Sigma_{\text{in}}-\Sigma_{\text{out}})\cdot\hat{n}=-\sigma'(\hat{n}\times\nabla T)$$

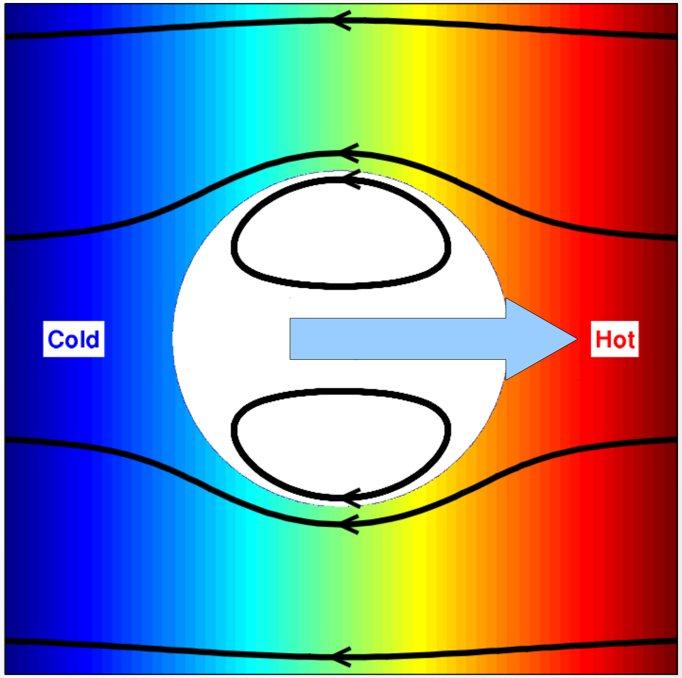
  - **This has the effect of migrating the droplet in the direction of increasing temperature**

Thermocapillary flow

- Young, Goldstein and Block examined the case of small bubbles and small temperature gradients
  - Stokes flow in both fluids, ignored heat advection

$$\nabla \cdot \vec{V} = 0 \quad \& \quad \mu \nabla^2 \vec{V} = \nabla p$$

$$\nabla^2 T = 0$$



  - **Inside the bubble the flow is dipole like (*Hill's Spherical Vortex*)**
  - **The migration velocity (in the absence of gravity)**

$$U_{\mathrm{YGB}} = \frac{2 r_0 k_{\mathrm{out}} (\sigma') |\nabla T_\infty|}{(2\mu_{\mathrm{out}} + 3\mu_{\mathrm{in}})(2k_{\mathrm{out}} + k_{\mathrm{in}})}$$

Bubbles in microgravity

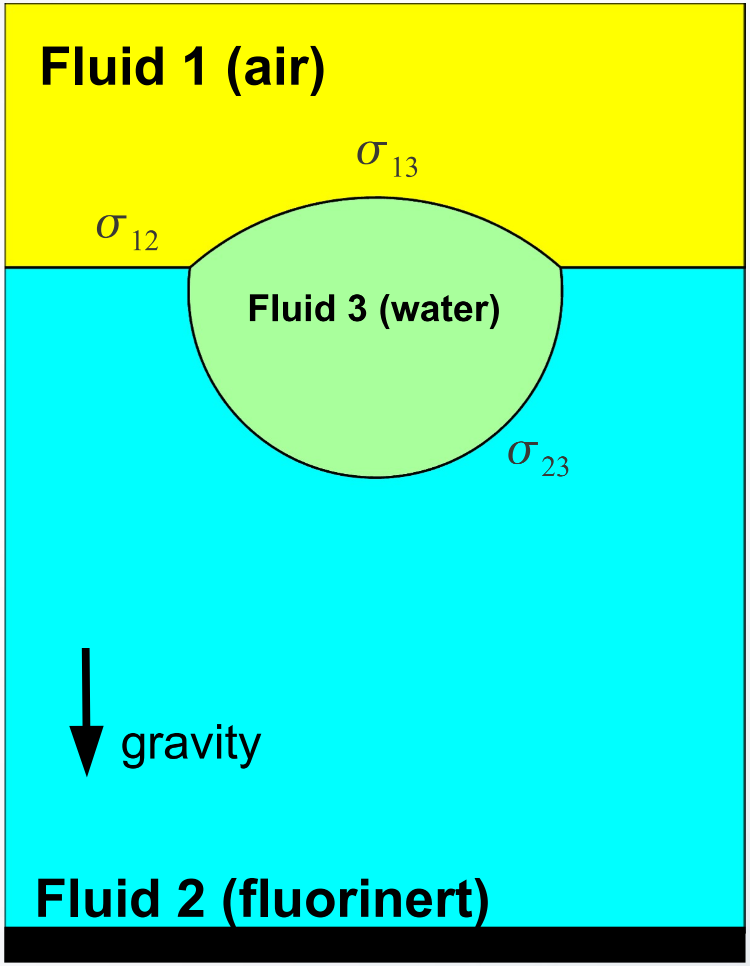
- In the early 1970s gas bubbles were found incorporated in materials solidified aboard the US Skylab

  - **This ignited a renewed interest in the migration of small drops due to the thermocapillary effect**

Space Shuttle Columbia
The second International Microgravity Laboratory (IML-2)

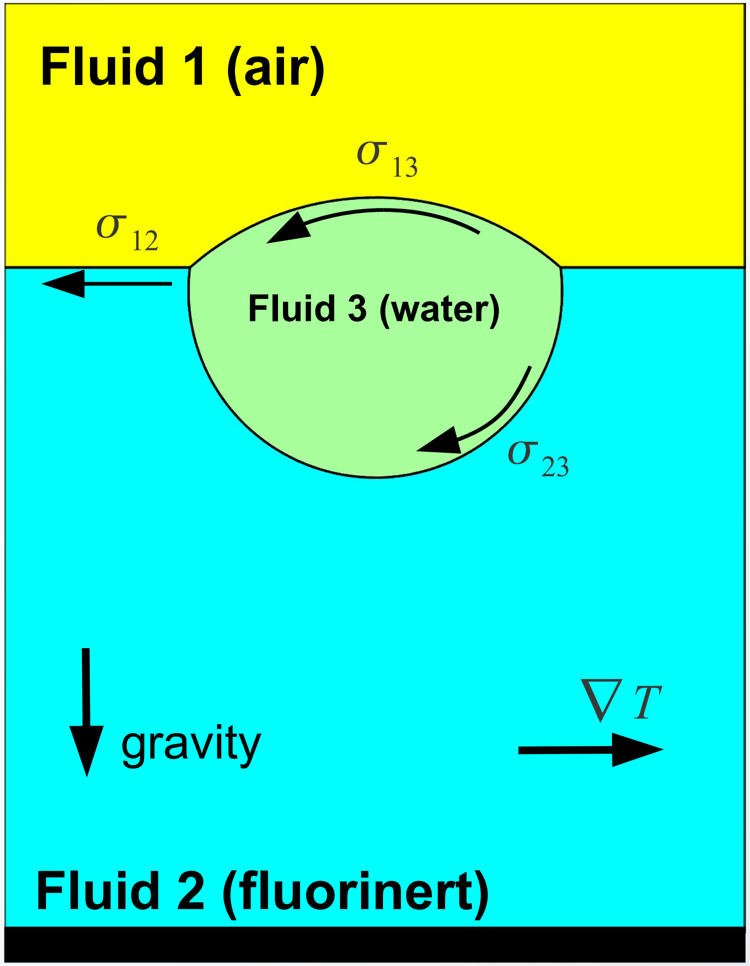
Interfacial Droplets

- **What's different?**
  1. Droplet is confined to an interface
  2. Droplet has a non-spherical shape

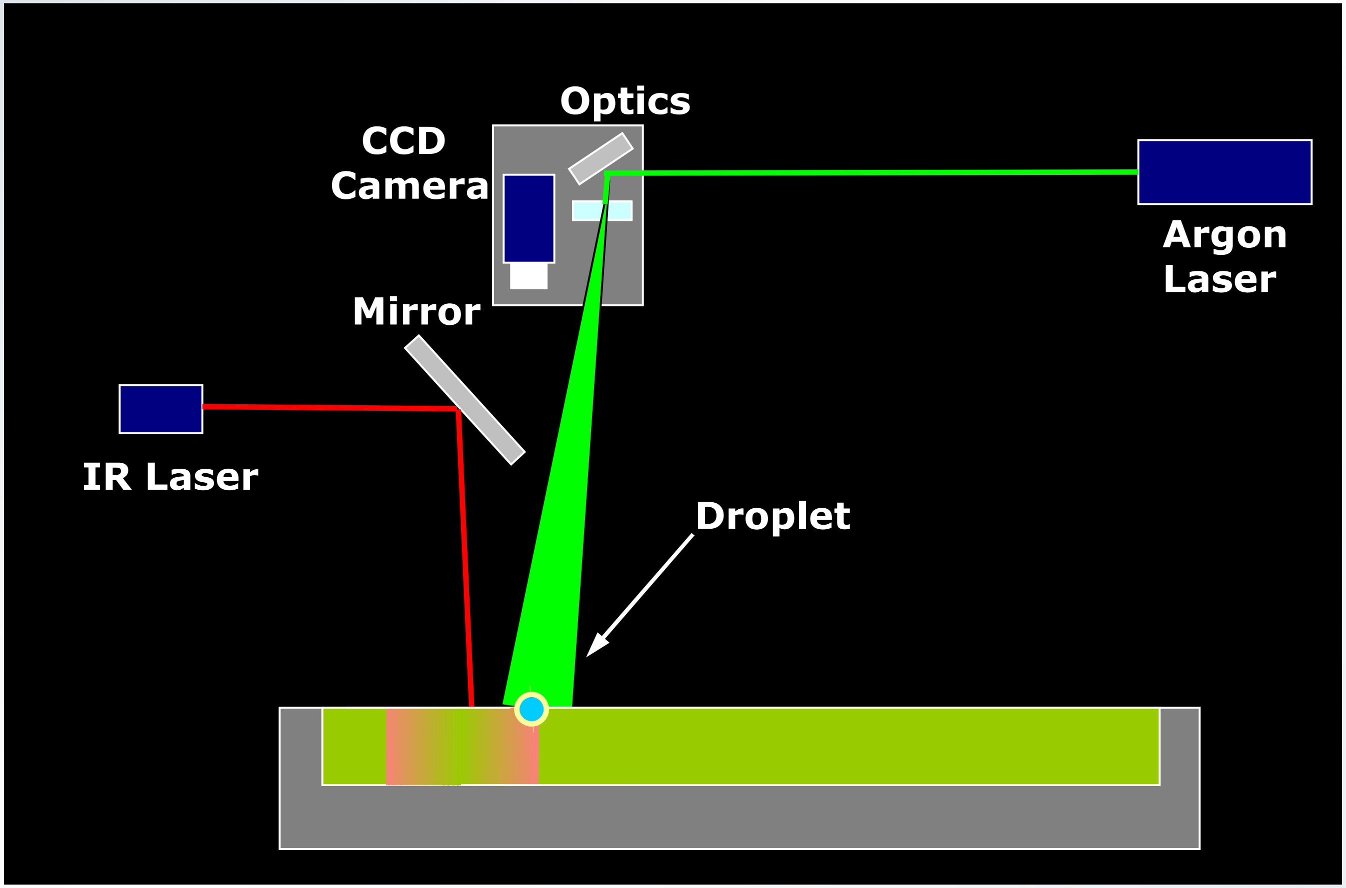


Interfacial Droplets

- **What's different?**
  1. Droplet is confined to an interface
  2. Droplet has a non-spherical shape
  3. Migration velocity is dominated by the substrate flow



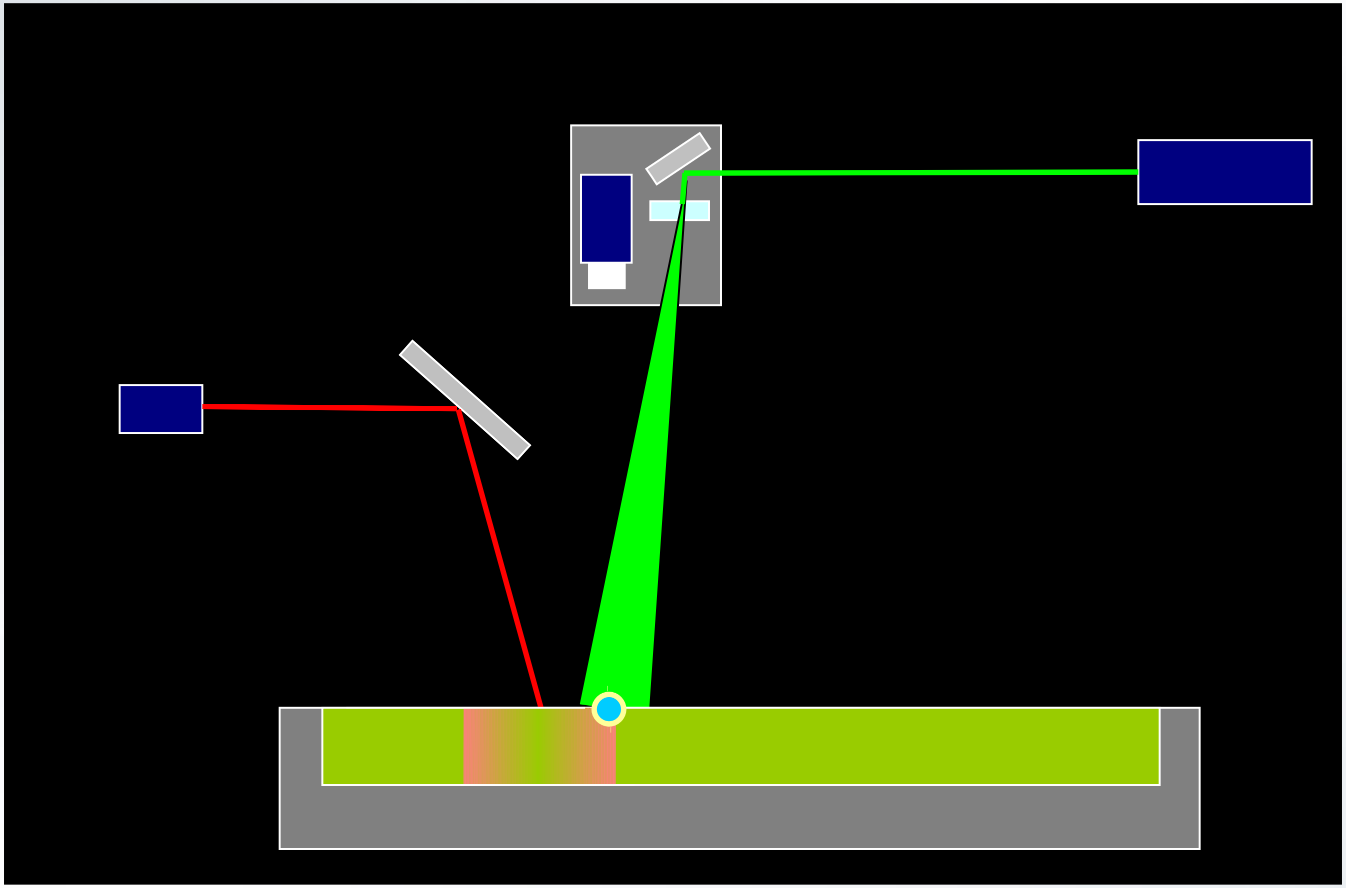
Experimental Setup



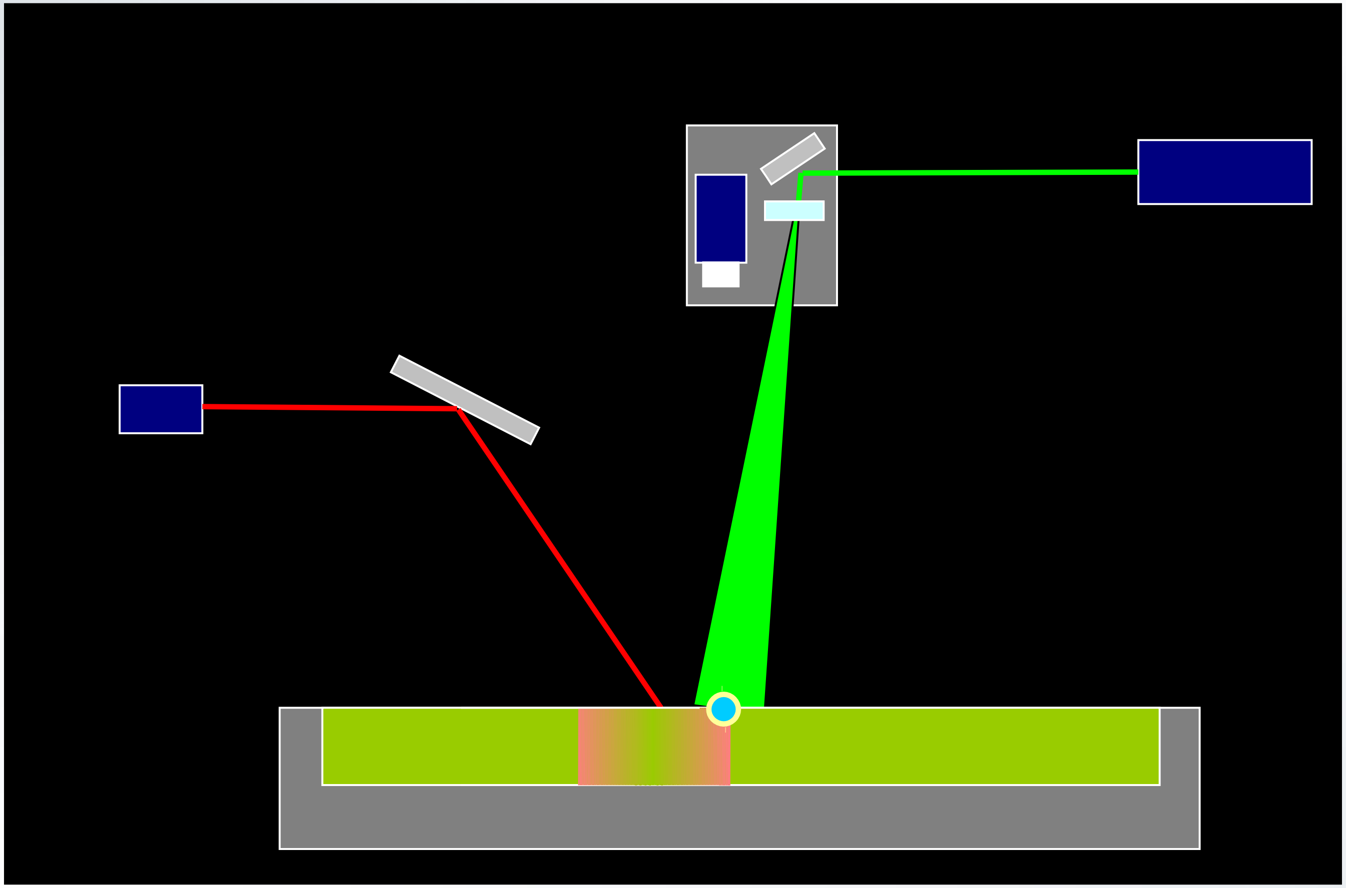
R.O. Grigoriev, V. Sharma, and M. Schatz

Lab on a Chip 6 (10): 1369-1372, 2006

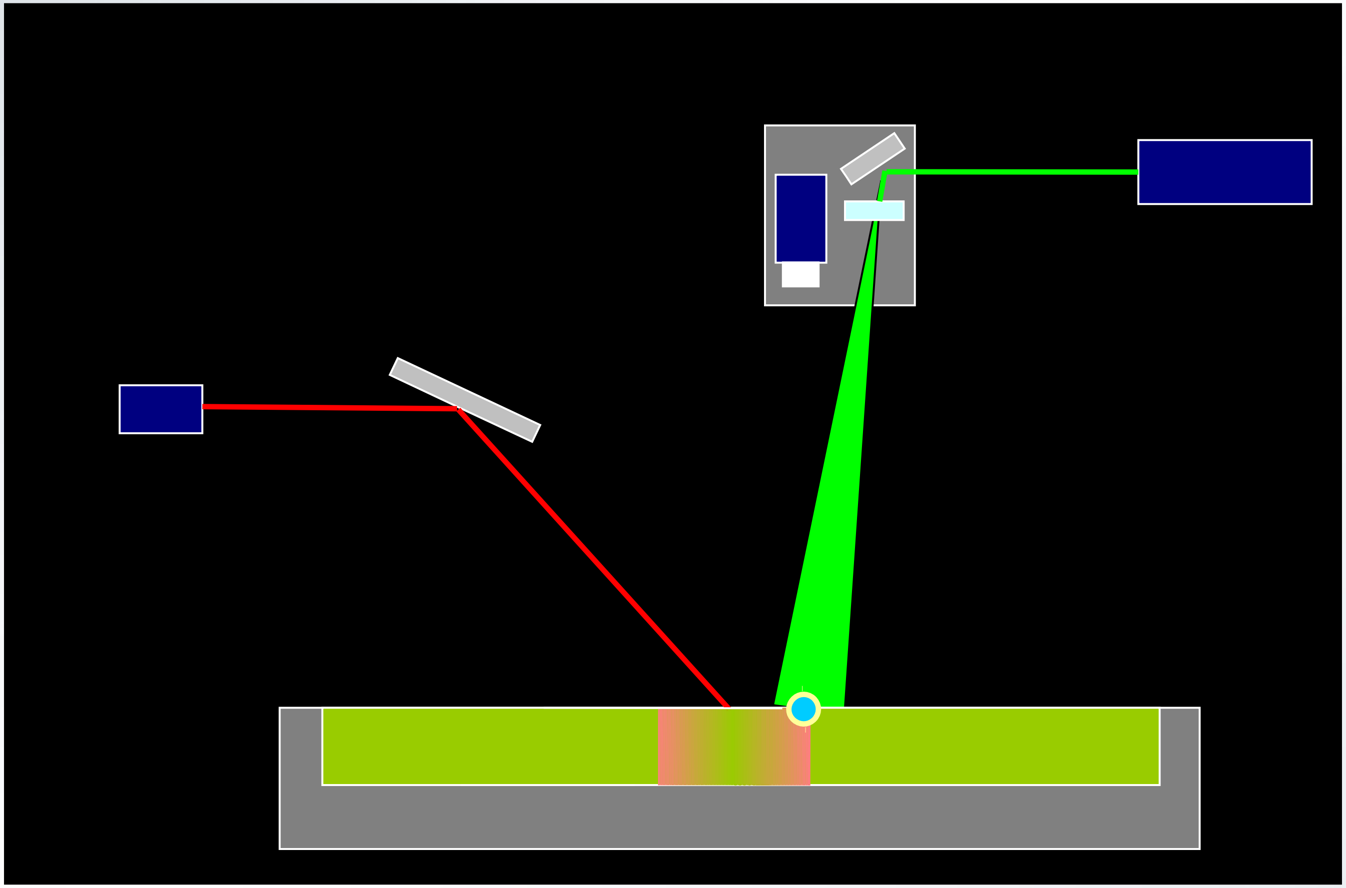
Experimental Setup



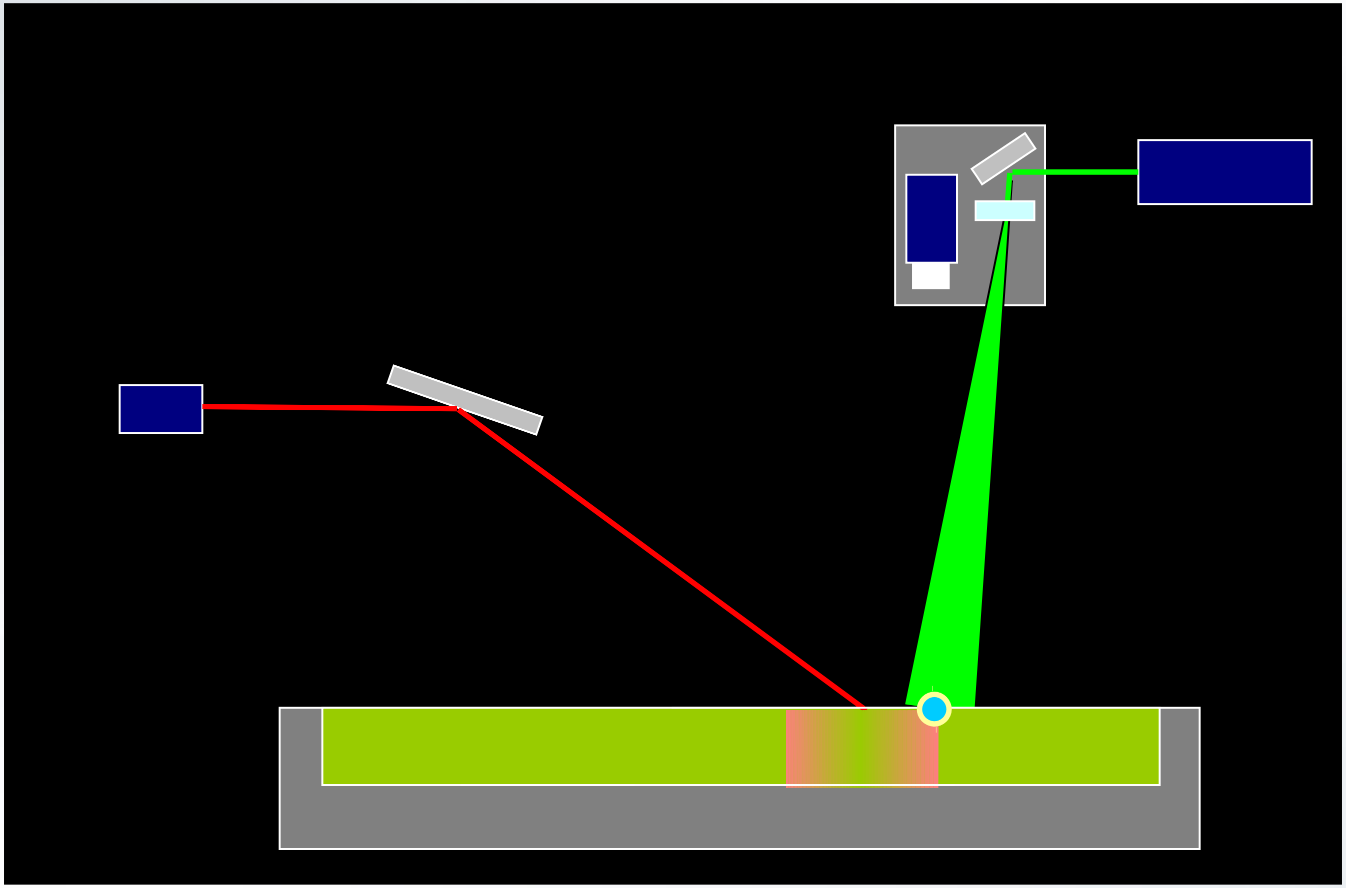
Experimental Setup



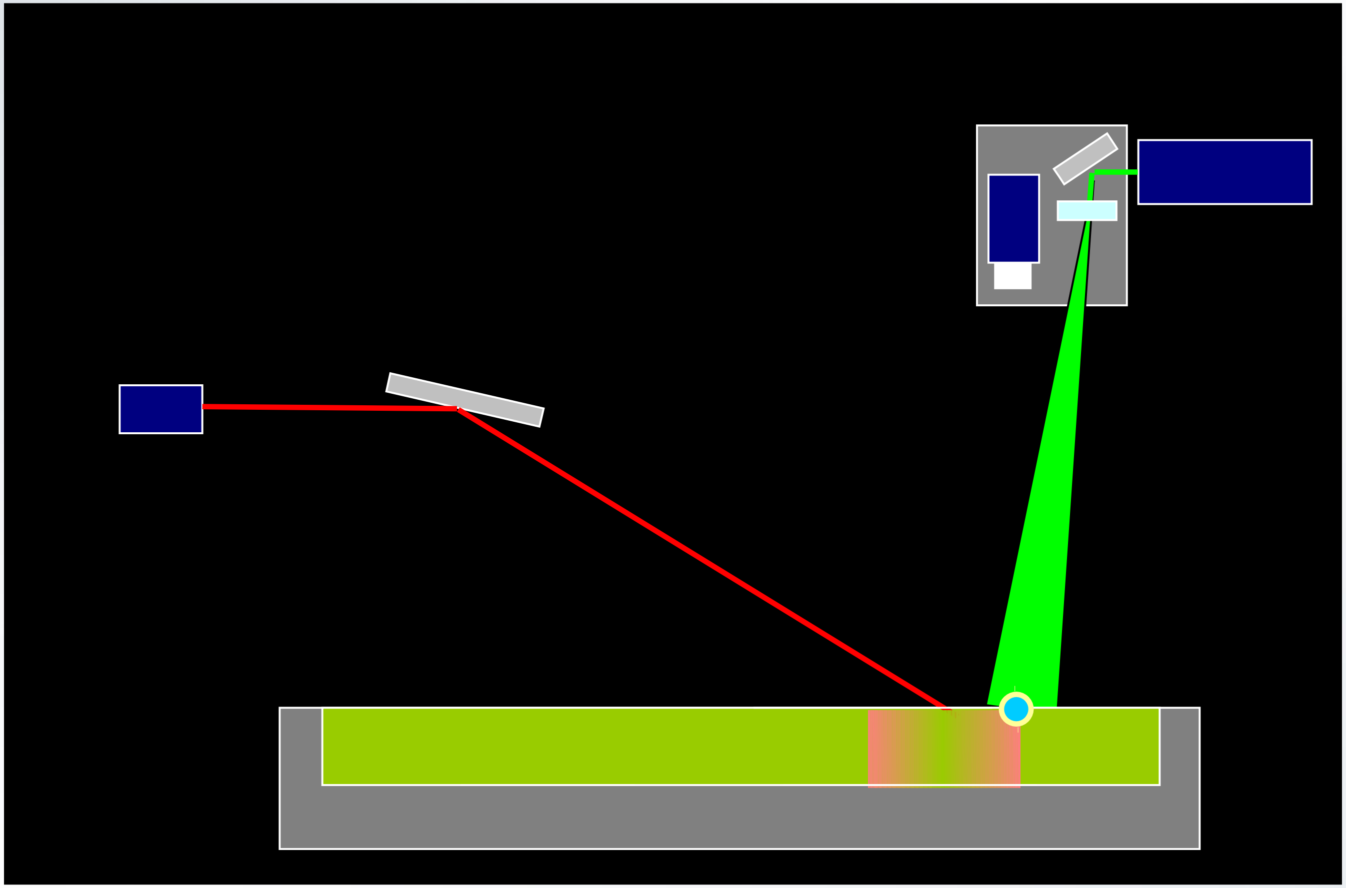
Experimental Setup



Experimental Setup



Experimental Setup



Asymptotic fields

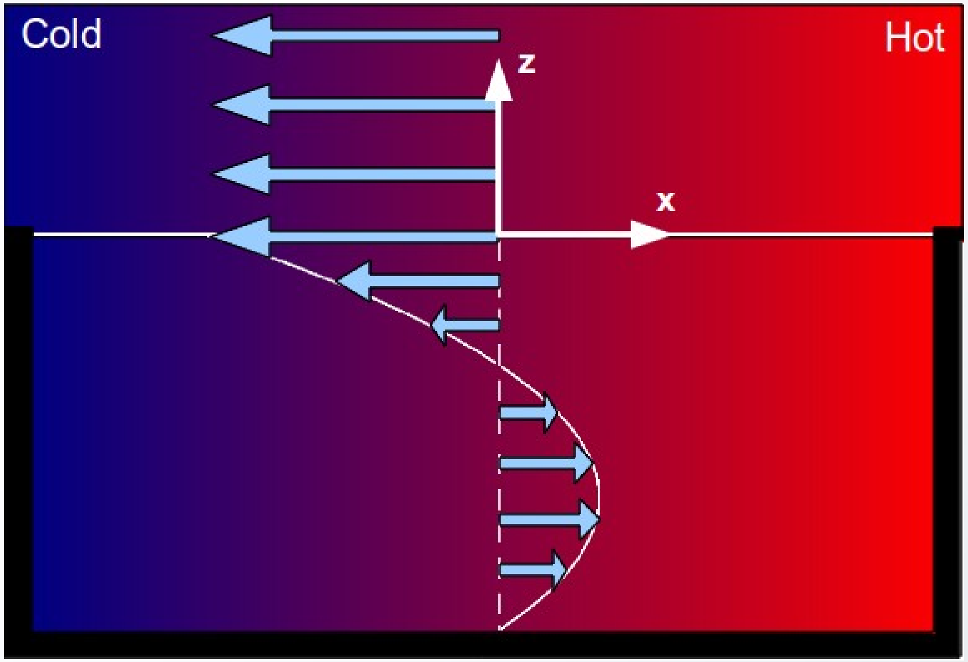
- A uniform temperature gradient

$$T_1 = T_2 = T_0 + |\nabla T_\infty| x$$

- The velocity profile is constant above and quadratic below:

$$V_1 = \frac{\sigma_{12}' H |\nabla T_\infty|}{4\mu_2} \hat{x}$$

$$V_2 = \frac{\sigma_{12}' |\nabla T_\infty|}{\mu_2} \left(\frac{3}{4\mathrm{H}} z^2 + z + \frac{H}{4} \right) \hat{x}$$



Geometry

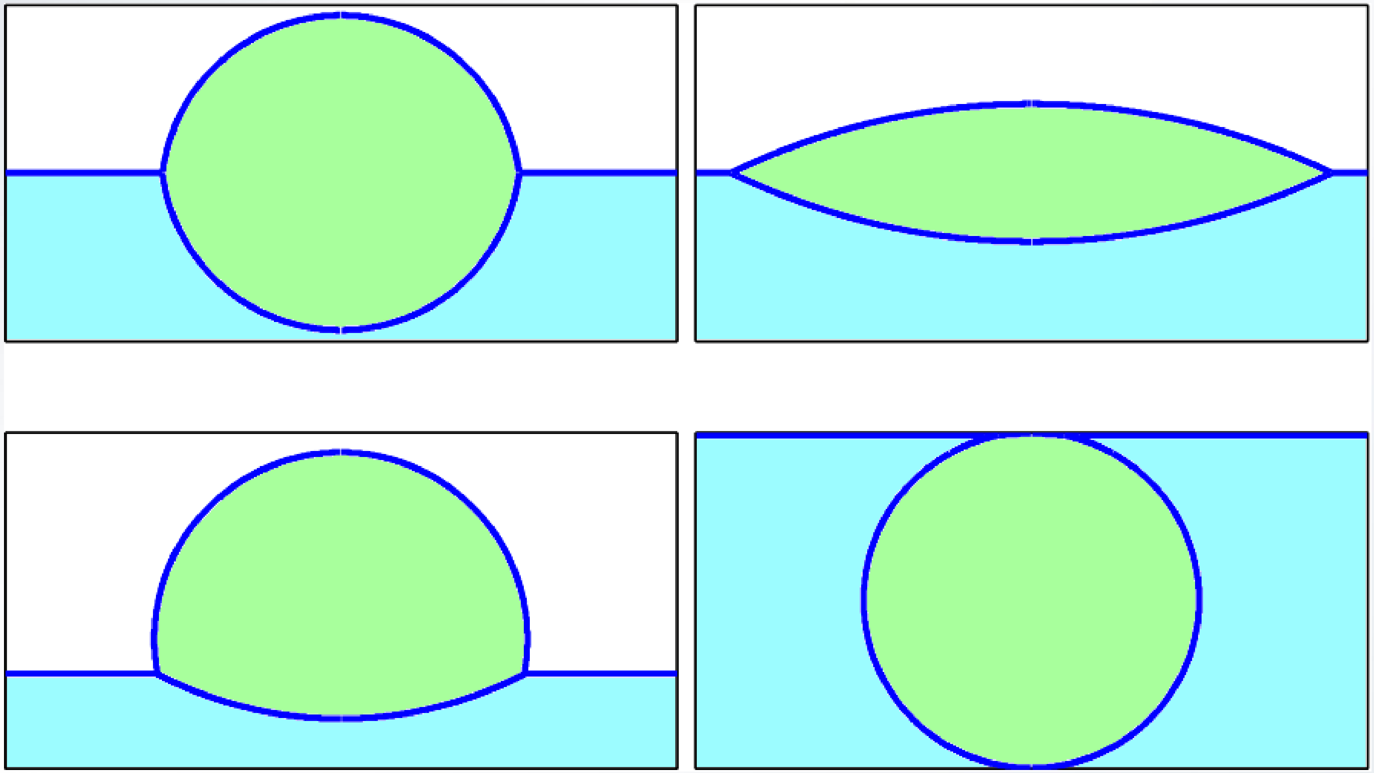
- We are only interested in very small droplets
  - **The limit where the Capillary and Bond number are small**

$$\mathrm{Ca} \equiv \frac{\mu_0 v_0}{\sigma_0} \qquad \mathrm{Bo} \equiv \frac{\rho_0 g r_0^2}{\sigma_0}$$

  - Deformations to the substrate and droplet interfaces are small and can be ignored
    - **In this limit each interface has constant curvature**
      - Two spherical caps for the droplet
      - The substrate interface is flat

The shape of the droplet

- The shape and position of the droplet is determined by a force balance at the contact line for a prescribed droplet volume



Governing Equations (Temp)

- Similar to Young *et al.* we restrict our attention to the slow migration of small droplets
- **The thermal Peclet (Marangoni) number characterizing the flow around the droplet is small**

$$\mathrm{Pe} \equiv \frac{r_0 v_0 \rho_0 C_p}{k_0}, \qquad v_0 \equiv \frac{\sigma_0' r_0}{\mu_0} \left| \nabla T_\infty \right|$$

  - **Velocity and Temperature are decoupled and in Steady State**
  - Near the droplet the temperature field is determined by solving Laplace's equation subject to asymptotic boundary conditions
    - Far from the drop the temperature gradient is horizontal and uniform

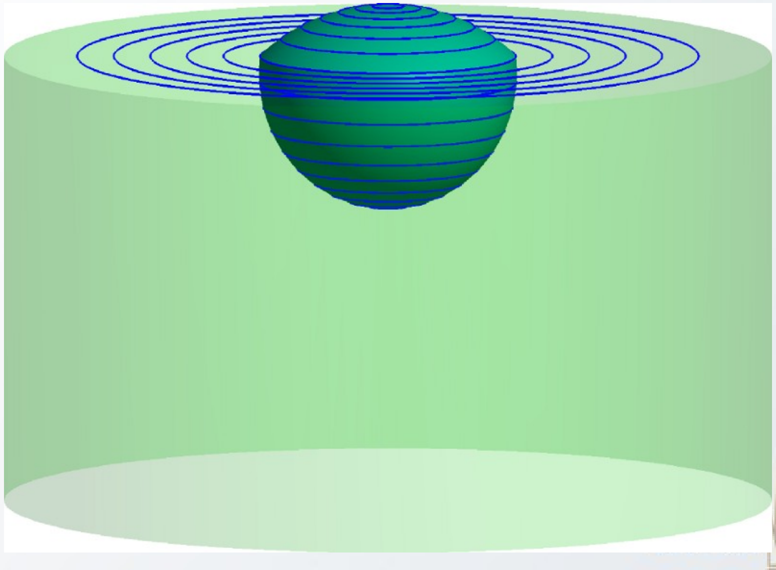
Governing Equations (Velocity)

- The Reynolds number is small
  - Steady state; the droplet migrates with constant speed
  - Near the droplet the velocity field is determined by solving Stokes equation subject to asymptotic boundary conditions
    - Far from the droplet the velocity field is the undisturbed flow of the substrate
    - At each interface normal velocities vanish and tangential velocities are continuous (*in the frame of the droplet*)
- **We satisfy the tangential component of the stress boundary conditions but not the normal component**
  - We have prescribed the shape and must instead specify that the net force on the droplet is zero

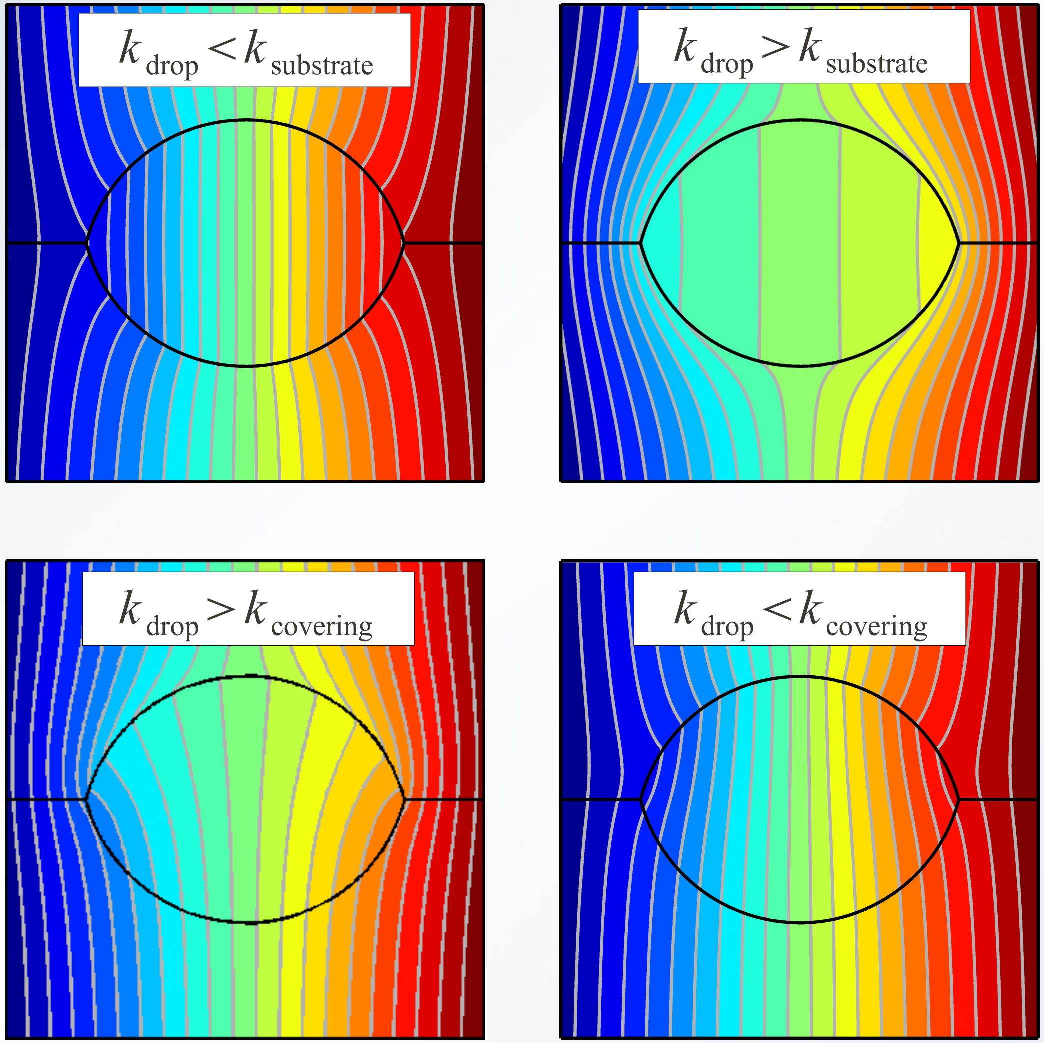
$$\vec{f}_{\text{body}} + \vec{f}_{\text{surface}} + \vec{f}_{\text{line}} = 0$$

Numerical solution procedure

- **We use Lamb's general solution for Stokes flow and spherical harmonics for the temperature field**
  - The solution in terms of Lamb's expansion has superior properties for computing interior streamlines
- **Satisfy boundary conditions only at rings on the surface of the droplet**
  - This reduces the boundary conditions to a linear system of equations with constant coefficients
  - We use an overdetermined system in order to improve numerical stability



Sample temperature fields



The Migration Velocity

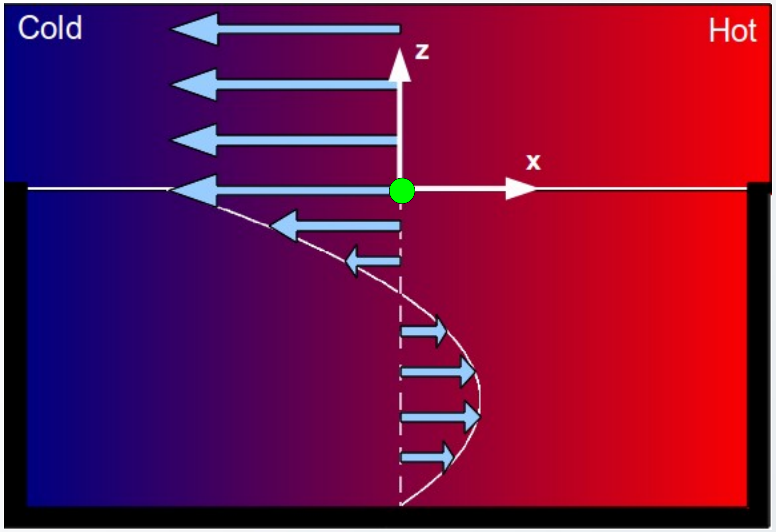
1. Thermocapillary stress at the surface of the droplet will push it in the direction of **warmer fluid**
2. Thermocapillary stress at the substrate interface will advect the droplet towards **cooler fluid**
   - **These speeds usually differ by orders of magnitude**

$$\frac{V_{\text{interface}}}{U_{\text{YGB}}} \sim \frac{H}{r_0}$$

   - **Mobility function**

$$M = \frac{U_{\text{migration}} - V_{\text{interface}}}{U_{\text{YGB}}}$$

   - Variation in the mobility illustrate the effect of confinement at the interface, droplet shape, etc.



Modeling Mobility

- We can derive an estimate for the mobility function by considering two corrections to the advection velocity at the substrate interface
  1. **Thermocapillary migration relative to substrate**
  2. **Drag due to the shear flow in the substrate**

$$U_{\text{migration}} \approx V_{\text{interface}} + U_{\text{thermo}} + U_{\text{shear}}$$

- These terms are approximated by substitution of averaged external fluid properties into analytical results for spherical unbounded droplets

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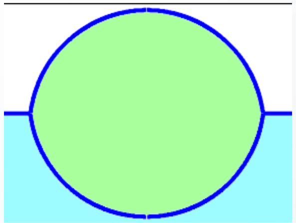
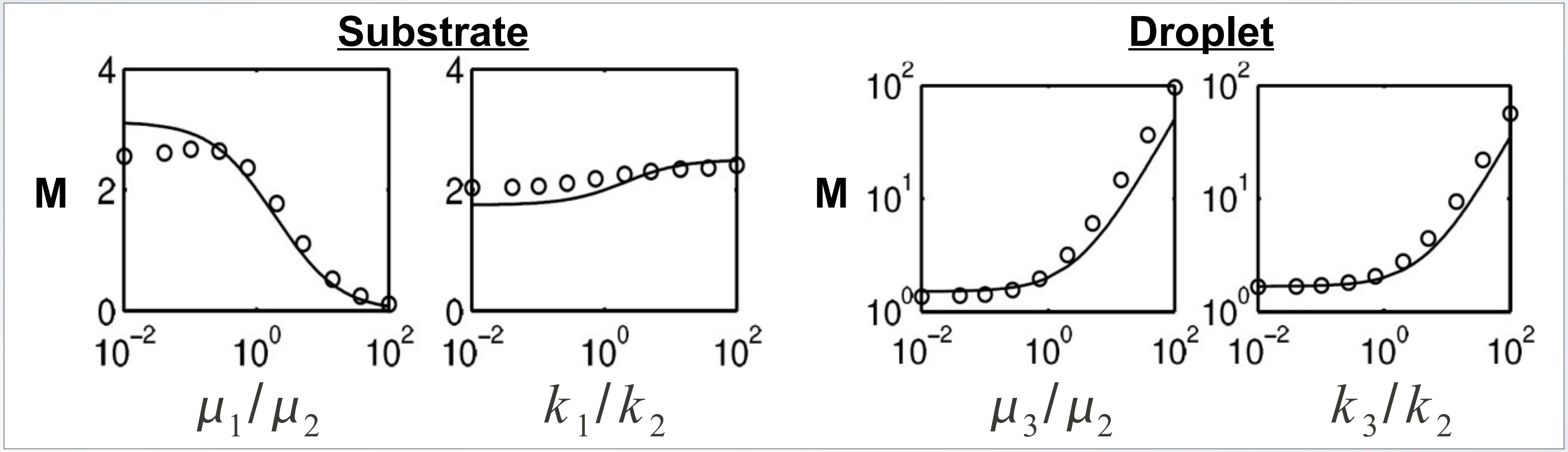
Mobility (fluid properties)

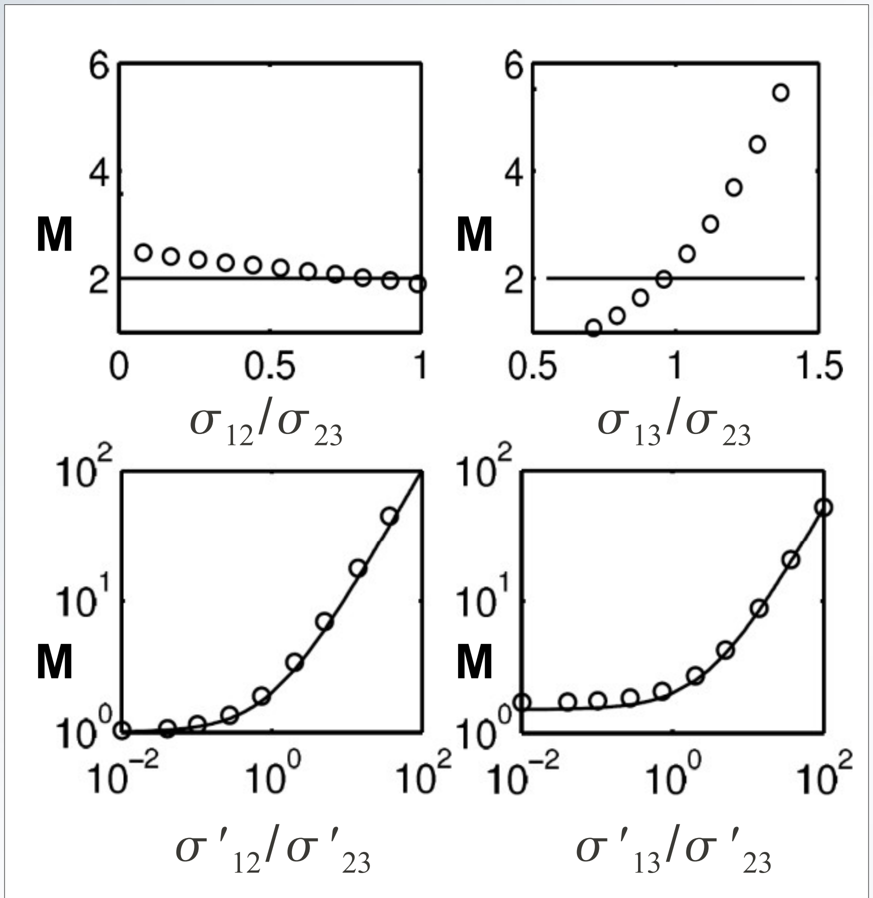
- We hold the droplet shape fixed and vary the ratio of fluid properties
  - **We find reasonable agreement between the numerical solution (**circles**) and our model of the mobility function (**line**)**

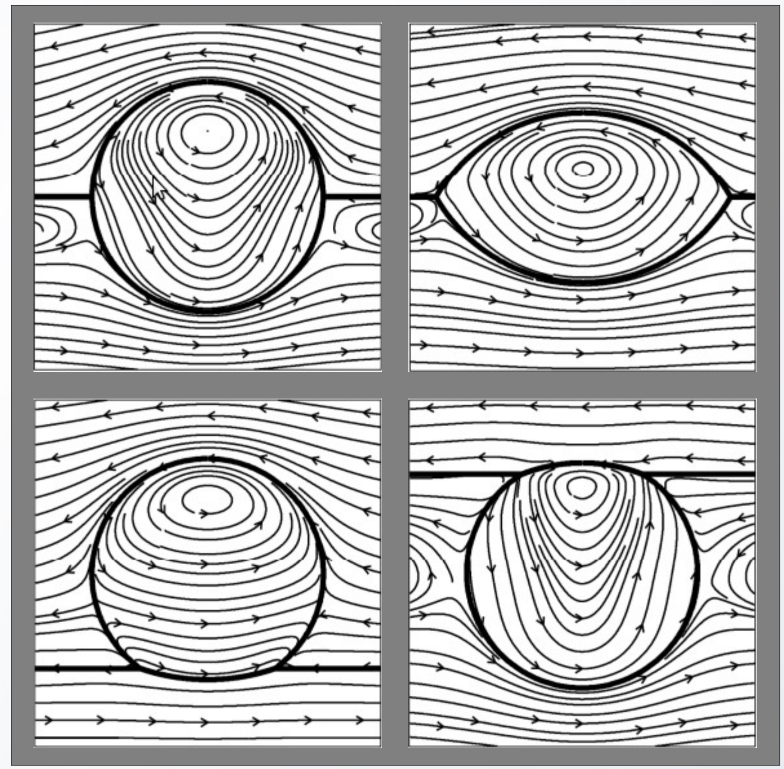




Mobility (surface tension)

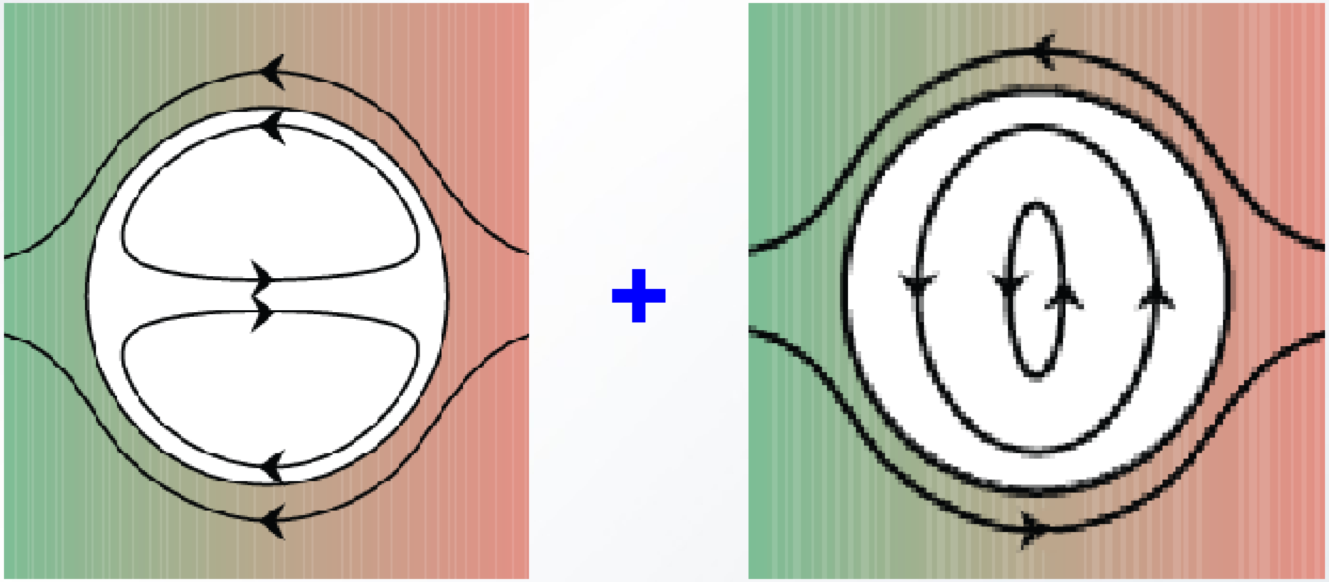
- We hold the fluid properties fixed and vary the surface tensions





A model flow

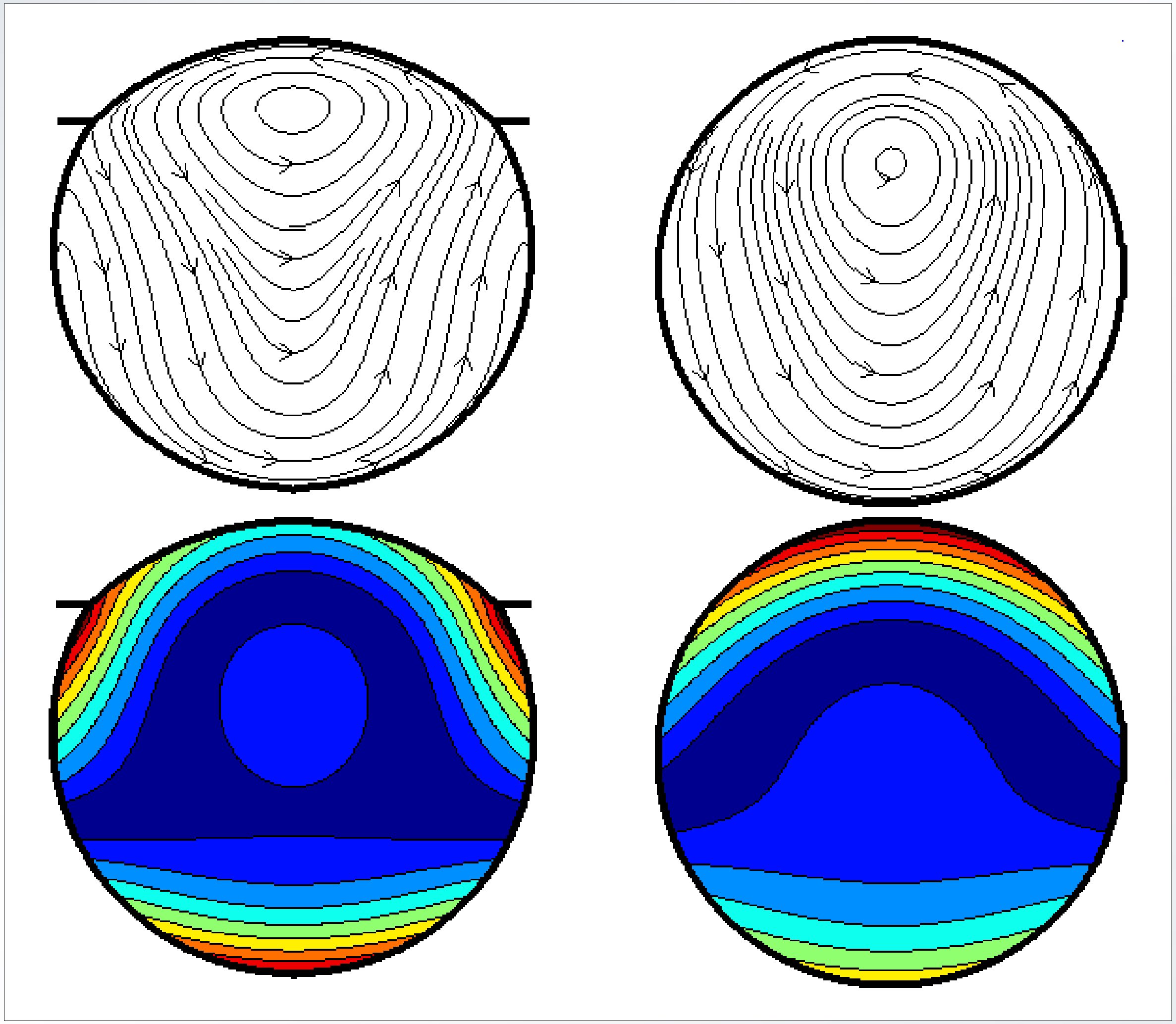
- **R.O. Grigoriev,**
  Phys. Fluids 17, 033601, 2005
  - By considering the combination of model flows, Grigoriev found that mixing was possible for a specific sets of parameters
  - Using parameters taken from the lab, the model is a combination of dipole and taylor flows; *no mixing expected*



Flow structures (predicted)

- The flow fields look qualitatively similar

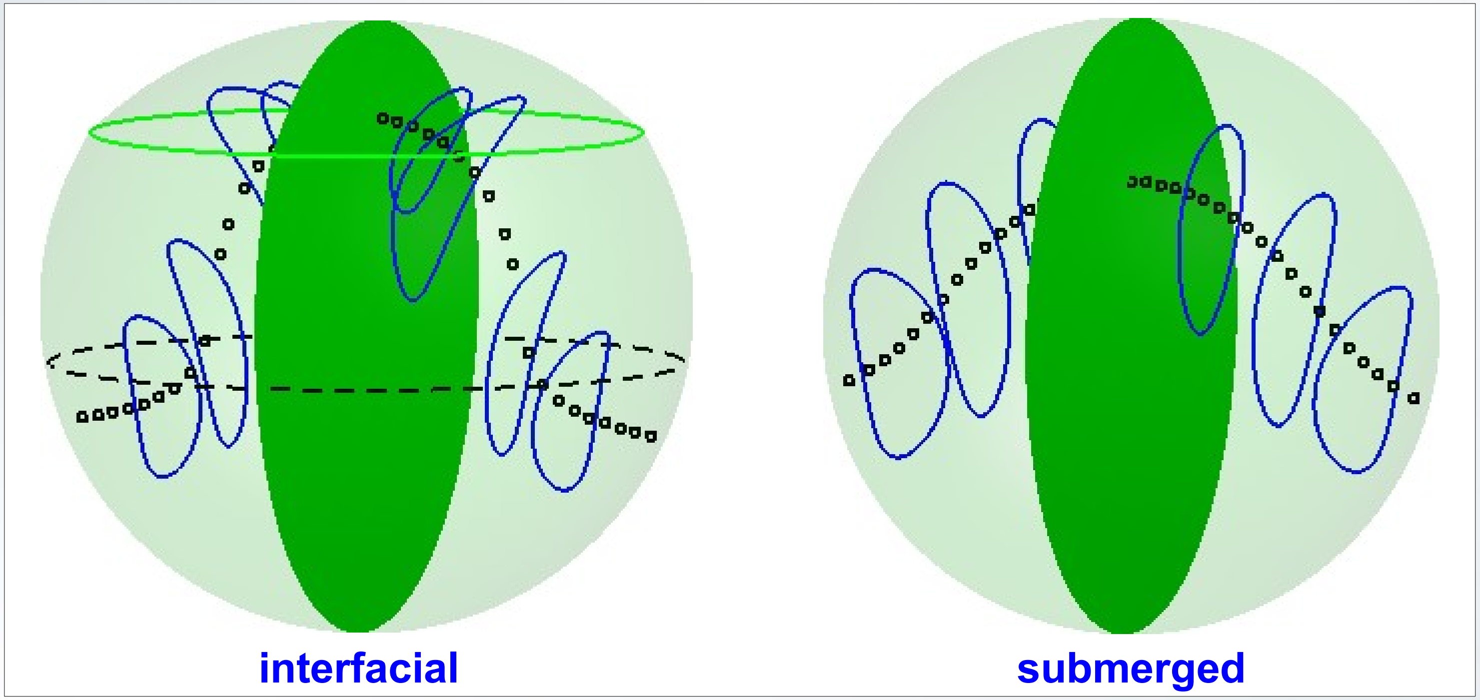
Streamlines longitudinal



Level sets of velocity transverse

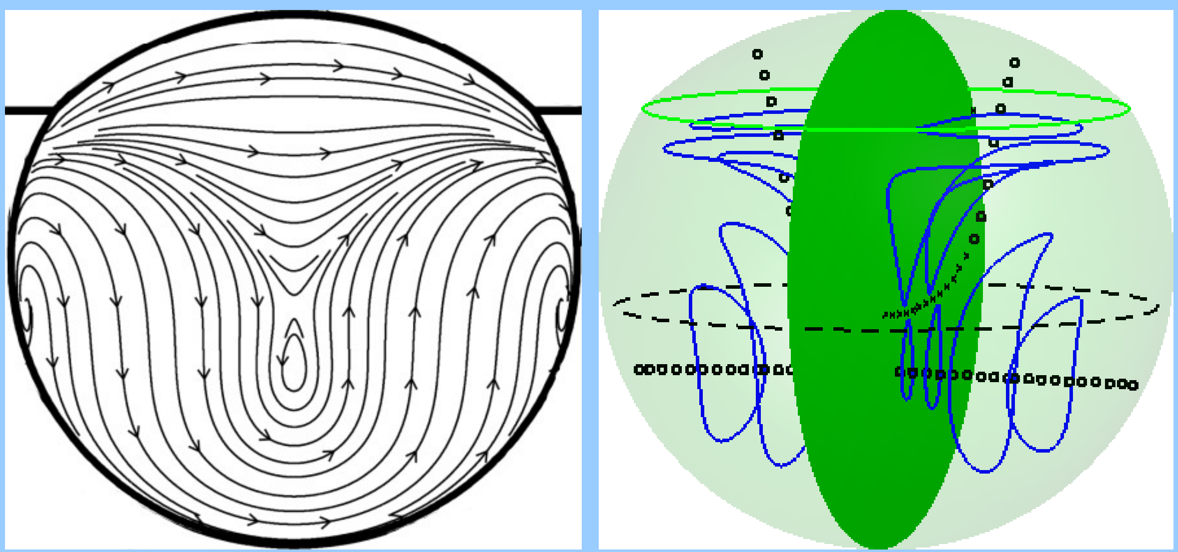
Fixed points

- We find that the fixed points of the flow characterize the topology of the flow structure
  - **Here we find that the interfacial droplet has a set of spiral fixed points connected by a heteroclinic orbit**

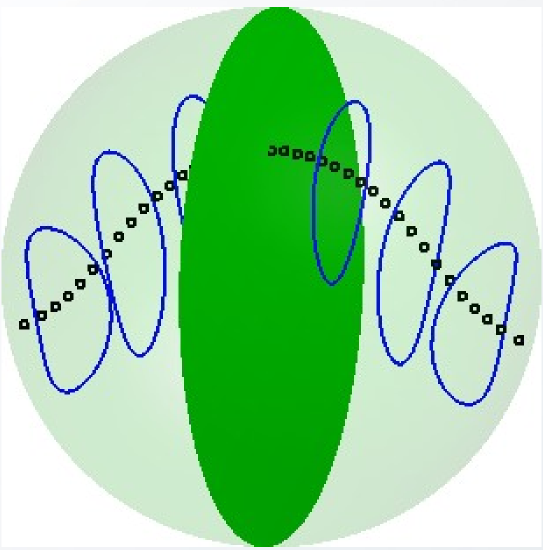


Another comparison

- There is disagreement when, for instance, the surface tension variation at the top cap is small

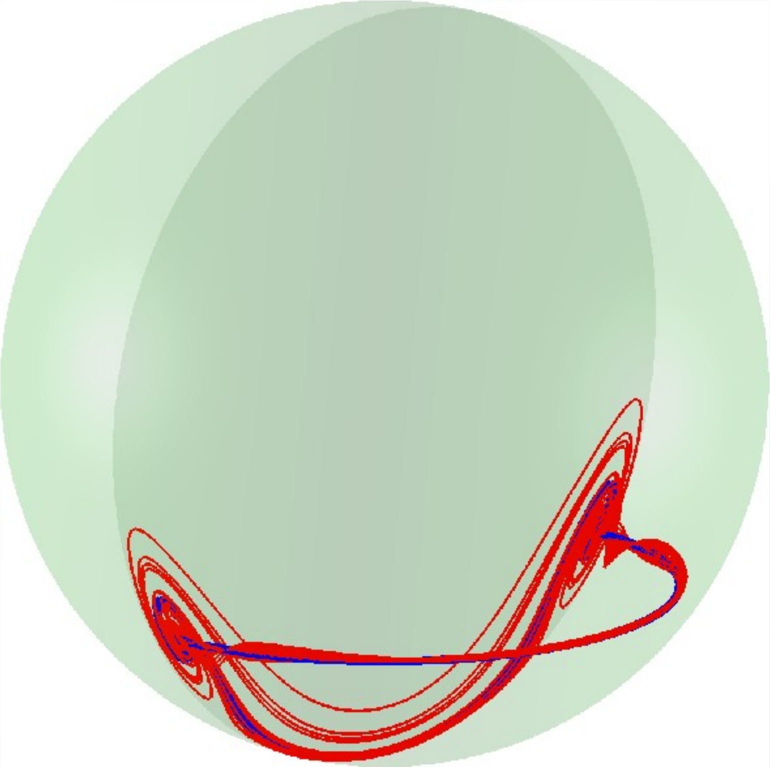


Interfacial droplet

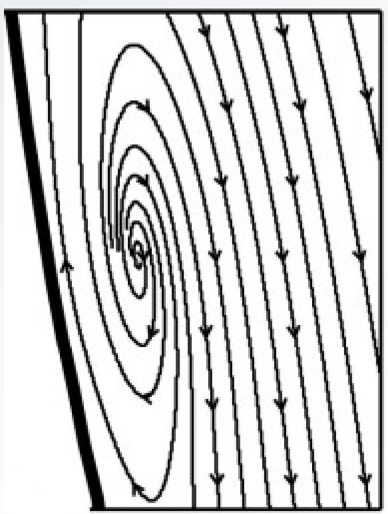


Spirals?

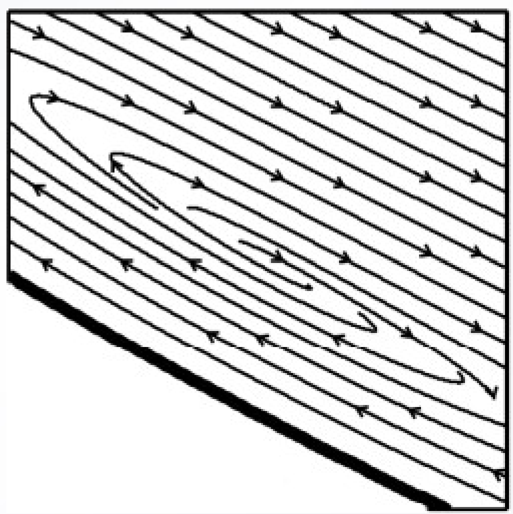
- Are the spiral fixed points unique to interfacial droplets?
  - **If you look for them in the submerged droplet you can find them**



Interfacial

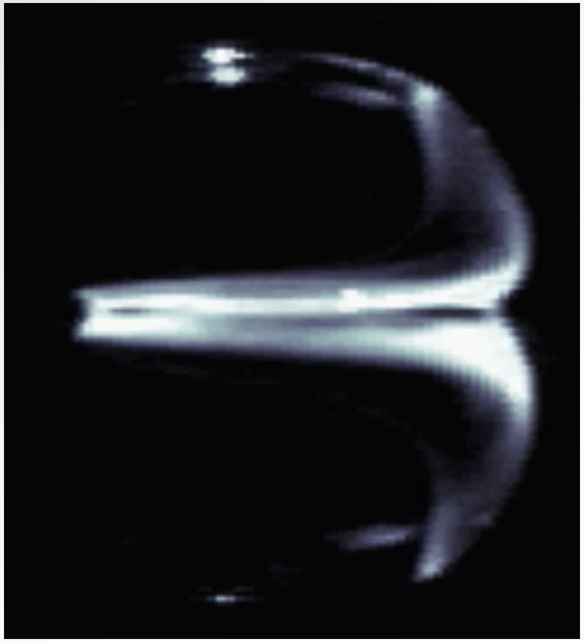


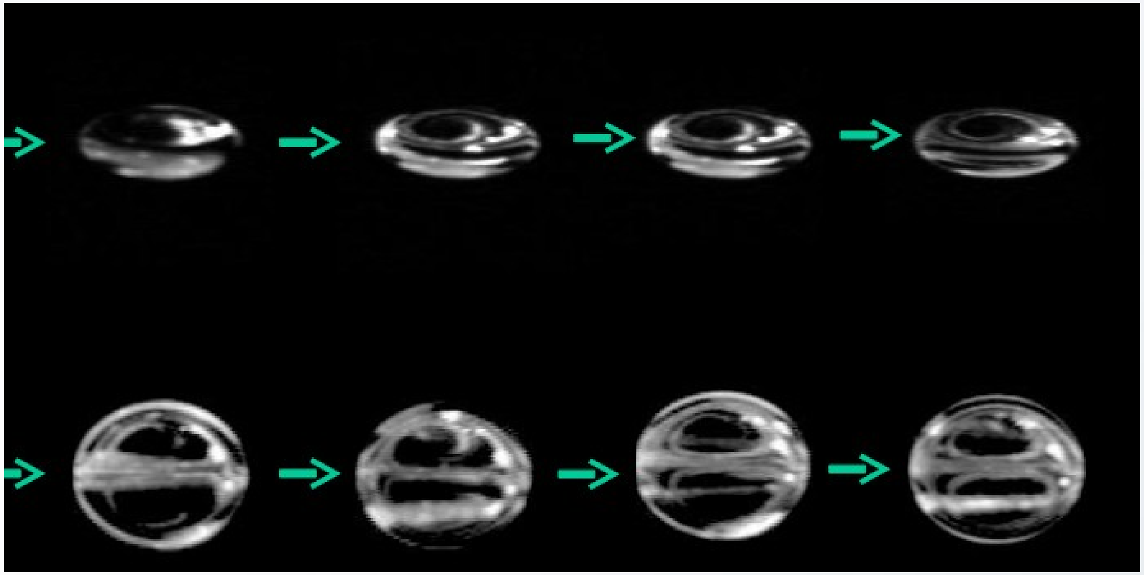
Submerged



From the lab

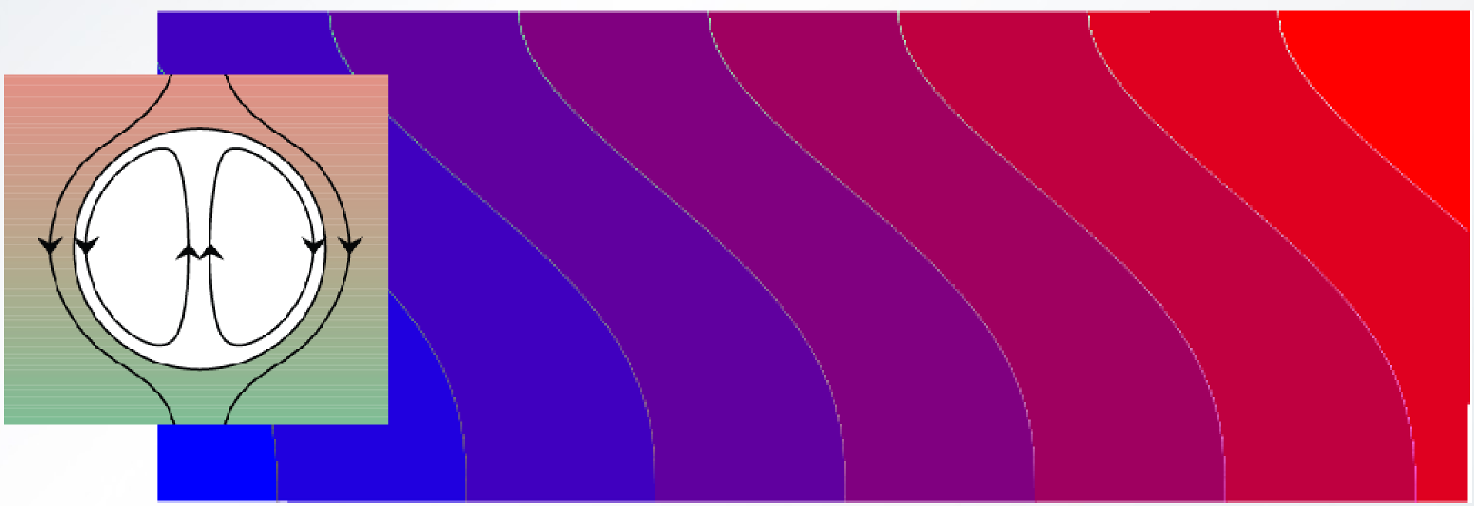
- Side and top view from a typical experiment
  - **Images and video provided by Daniel Borrero**
  - Water/Glycerin droplet (r ~ 100 μm) on FC-70 substrate exposed to air
  - Passive tracers: 0.5 micron fluorescent microspheres





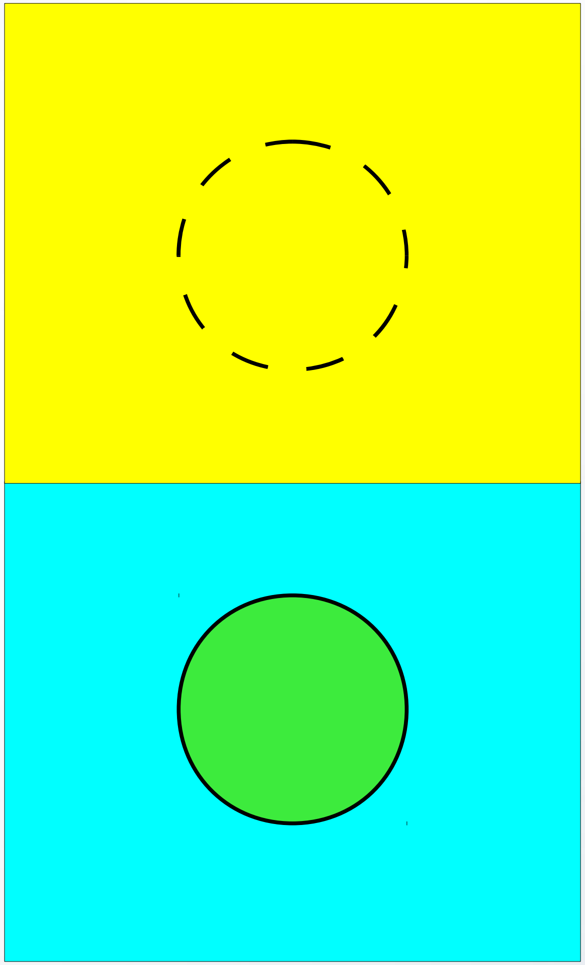
Future Work

- Improved substrate temperature model
- In lab the thermal Peclet number in the substrate was not negligible
  - **Pe ~ 35**
  - Advection introduces a different temperature profile
  - Near the droplet, thermal gradients introduce a vertical dipole flow



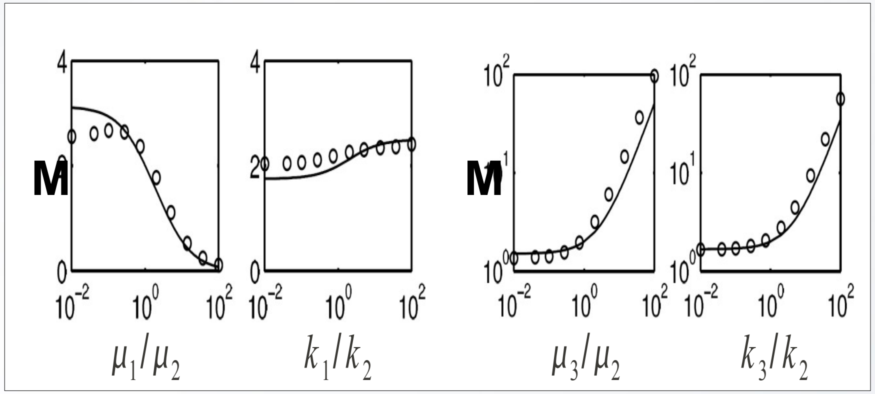
Future Work

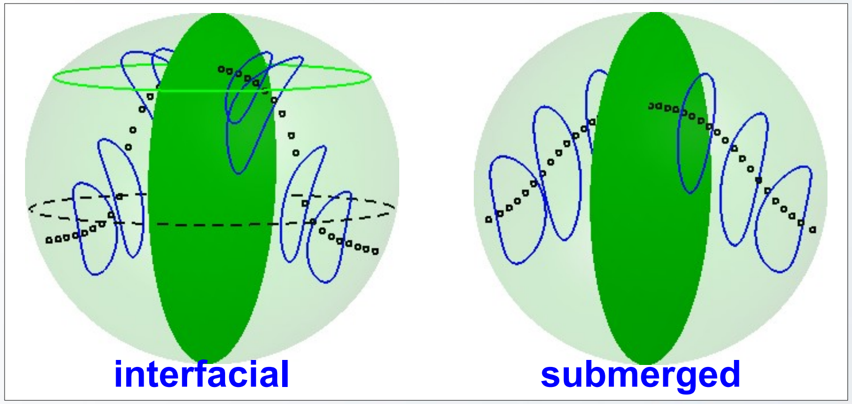
- A solution procedure by the method of reflections
  - Solve Stokes equation near the droplet
  - Reflect
  - Expand the reflection and truncate
  - Solve Stokes equation near the droplet for the truncated reflection
  - Repeat to desired order



In Summary

- A numerical method was developed to model the steady state migration of a small droplet floating at the surface of a fluid substrate
  1. **Migration velocities were modeled and compared to the classical result for an unbounded droplet**
  2. **Velocity fields inside the droplet were compared to those predicted by a submerged drop model**





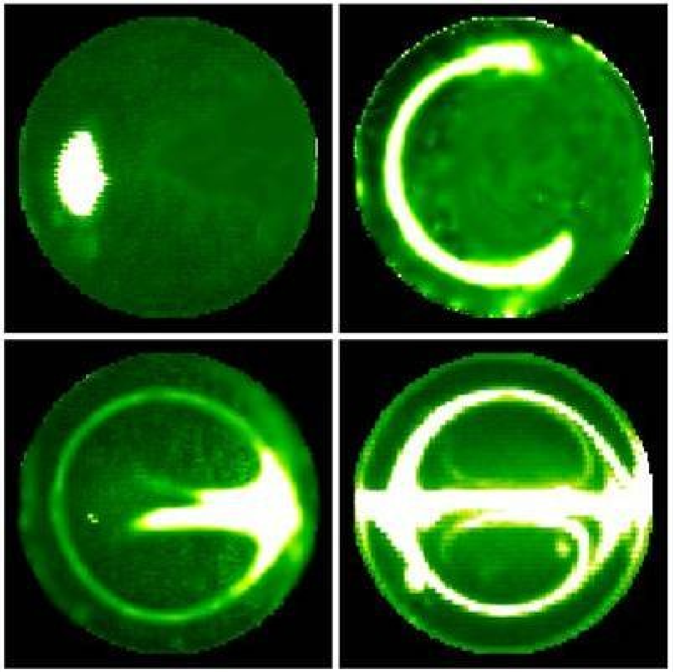
Thank You

http://en.wikipedia.org/wiki/Fran%C3%A7ois_N._Frenkiel

Optical Microfluidics

- **R.O. Grigoriev, V. Sharma, and M. Schatz**
  Lab on a Chip 6 (10): 1369-1372, 2006
  - **Mixing in microdroplets can be difficult**
  - Flows constrained to such small volumes typically have low Reynolds numbers
  - Their high degree of symmetry gives rise to invariant surfaces that trajectories cannot cross
  - They are too large for diffusion to be an effective mixing mechanism
  - **Mixing by chaotic advection**
    - Kneading Dough

Top-down view



Temperature in the substrate

No-flux

$$\nabla^2 T = 0$$

Continuous flux

$T = T_{\text{left}}$

$$\nabla^2 T = 0$$

$T = T_{\text{right}}$

No-flux

Velocity field in the substrate

Stress-free

$$\nabla \cdot \vec{V} = 0 \qquad \mu \nabla^2 \vec{V} = \nabla p$$

$$\hat{n} \times (\Sigma_{\text{in}} - \Sigma_{\text{out}}) \cdot \hat{n} = -\sigma' (\hat{n} \times \nabla T)$$

$$\nabla \cdot \vec{V} = 0 \qquad \mu \nabla^2 \vec{V} = \nabla p$$

No-slip

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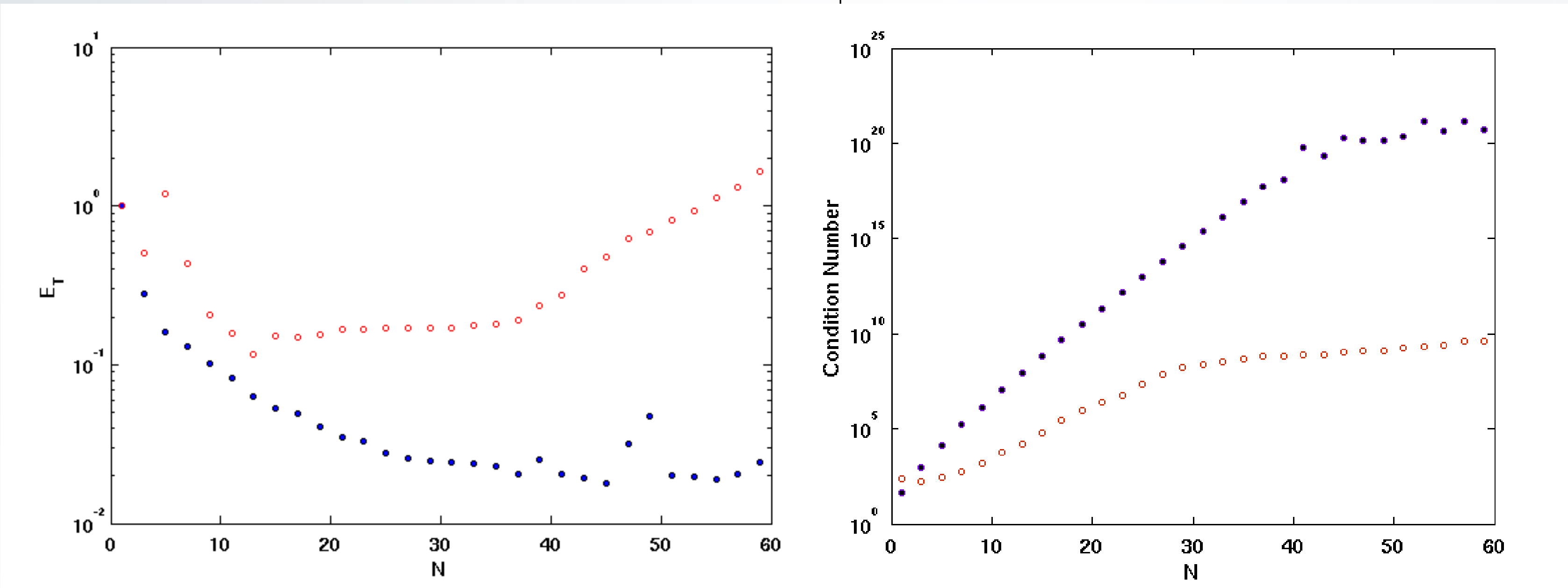
Optimization

- What is the total error in our solution?
  - **For a given choice of parameters integrate the error in each of the boundary conditions over the interface**
    - The net sum is the total error E_T



  - **Truncation order in solution is sensitive to drop shape**
  - Strongly Overdetermined and not very sensitive to location