Speed of flow of non-wetting droplets in capillaries of circular cross-section - theory

Karol Makuch, Jean-Baptiste Gorce, Piotr Garstecki

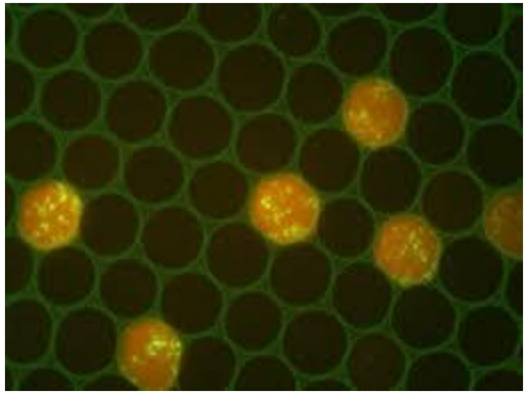
Microfluidics and Complex Fluids Group

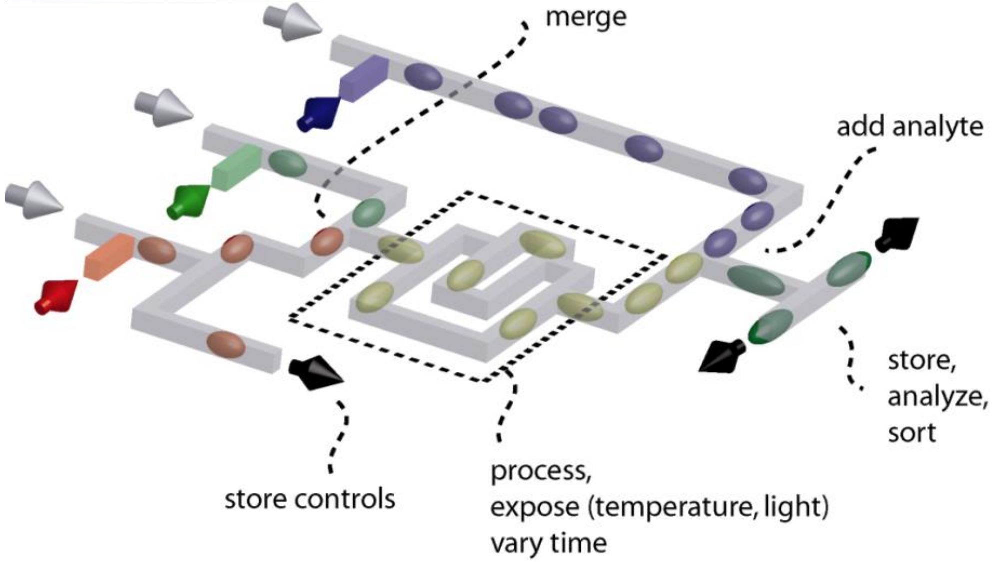
Institute of Physical Chemistry
Polish Academy of Sciences

IChF group meeting 15.03.2018

Motivation

Compartmentalization:
-chemical reactions
-encapsulate and cultivate microorganisms



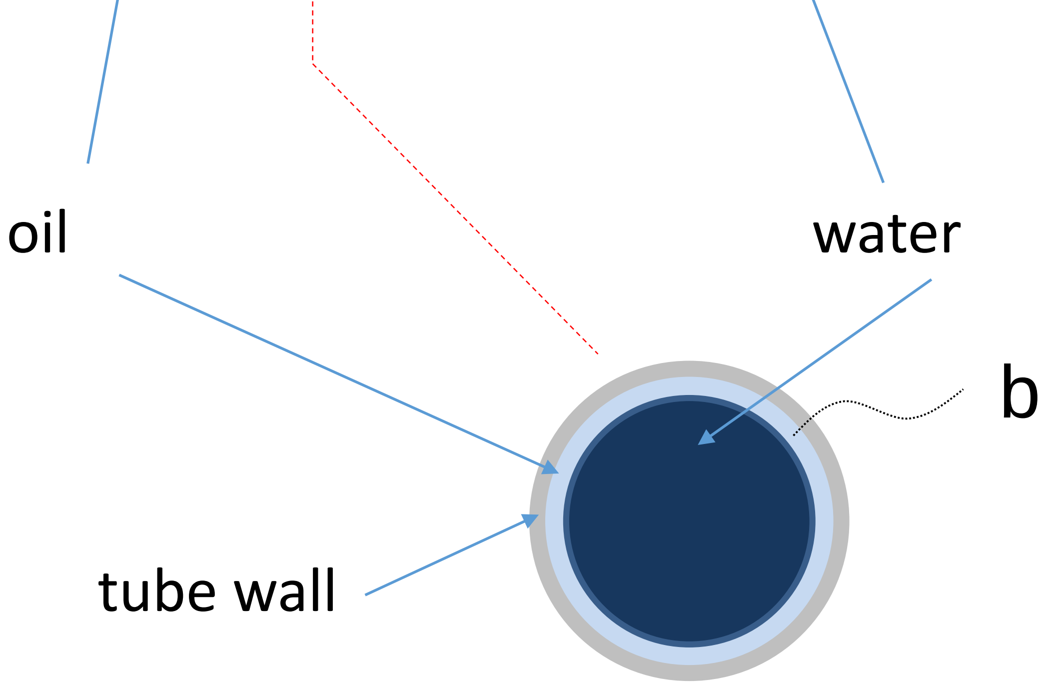


What from measurements of velocity of moving droplets?

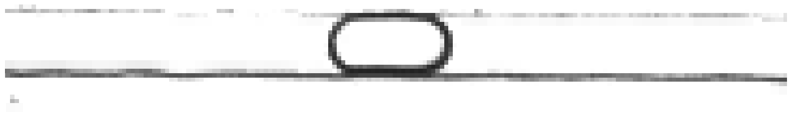
Goal

to understand speed of flow of droplets in microfluidic channel

- non-wetting (thin layer)
- Long (Taylor)



Goal: theoretical explanation of speed of a moving droplet



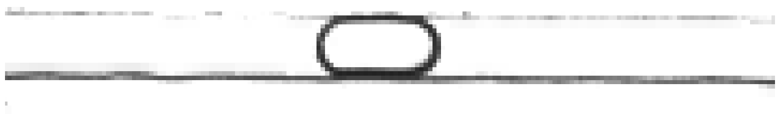
Mobility of droplet:

$$\beta = \frac{U}{V}$$

velocity of droplet

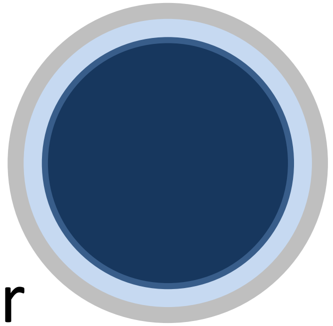
average velocity of continuous phase

Factors which influence speed of droplet



- average speed of flow of oil
- length of droplet
- viscosity of the droplet
- viscosity of the continuous phase
- interfacial tension
- presence/absence of surfactant (oil, droplet)
- gravity field
- …

Fairbrother and Stubbs (1935)



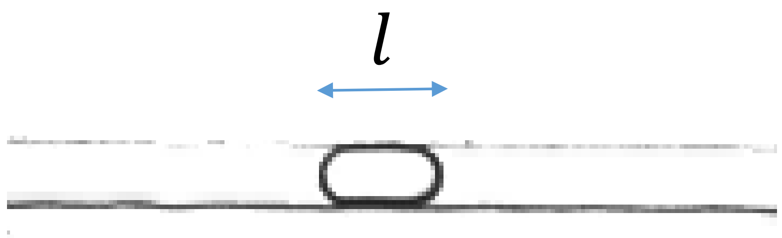
Inviscid droplet (bubble):

-velocity of bubble different than the velocity of continuous phase

$$U > V$$

-length of bubble insignificant when larger than 3/2 of tube diameter (the effect of length readily observed for $l \approx 2R$)

$$l \geq 3R$$



Empirical law:

$$\beta = 1 + Ca^{\frac{1}{2}}$$

$$Ca = \frac{\mu_c V}{\sigma}$$

Taylor (1961)

Inviscid droplet (bubble):
Validation of Fairbrother and Stubbs formula $\beta = 1 + 1.0 * Ca^{\frac{1}{2}}$
for

$$Ca < 0.09$$

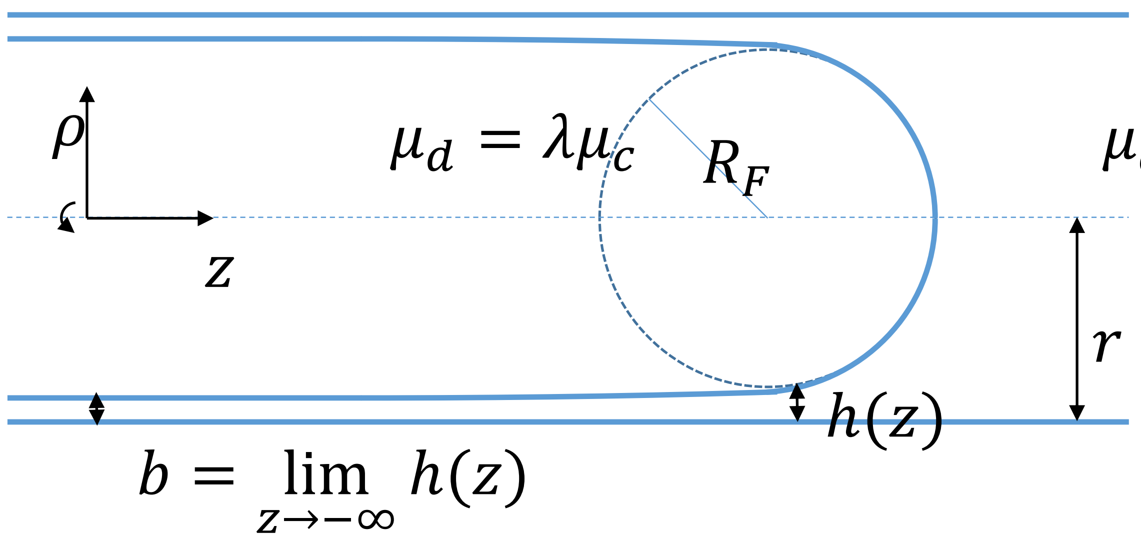
$$Ca = \frac{\mu_c V}{\sigma}$$

e.g. water droplet in FC-40 oil:
≈ 0.001 for $V = 1\,\frac{\text{cm}}{s}$

Criticism of empirical ‘1.0’ factor and ‘1/2’ exponent in the above law.

Bretherton (1961) – assumptions

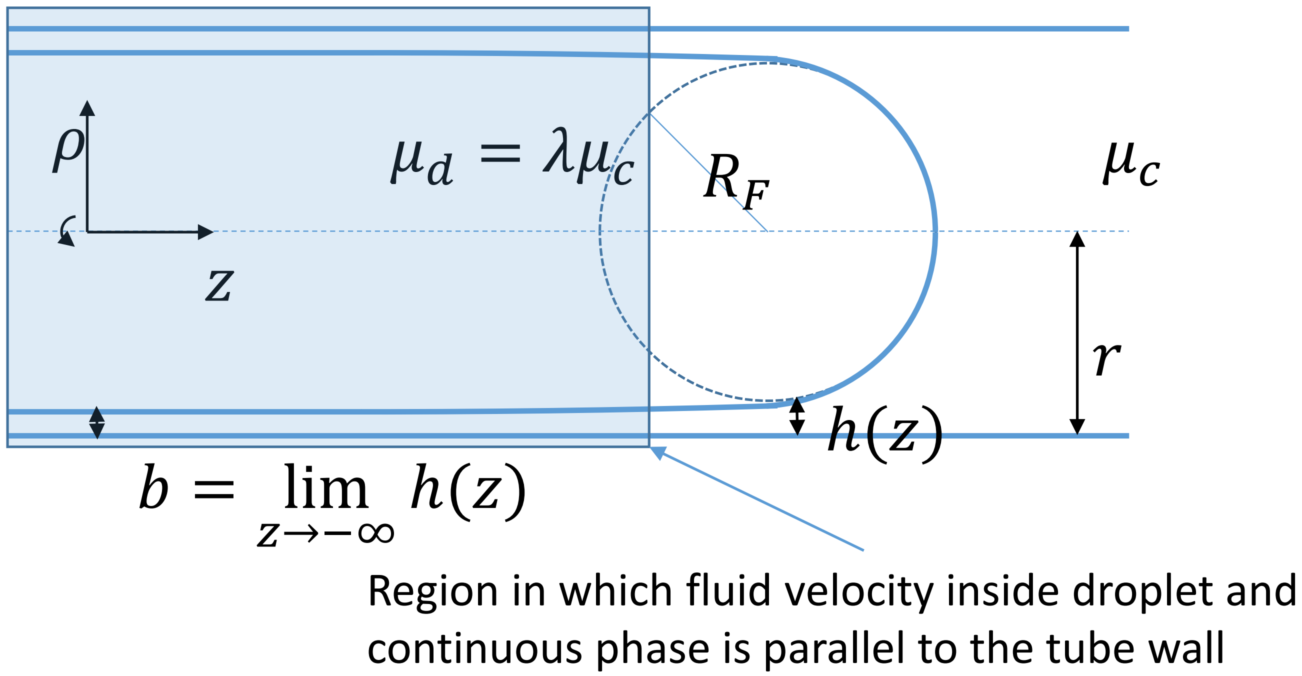
- Second Newton's law for fluid for small velocities (Stokes equations) + b.c.
- Incompressible fluids
- Jump of the pressure at the interface: $\sigma\kappa(z)$



Bretherton (1961) – approximations

- Lubrication approximation: $\vec{v}(\vec{r}) \approx \hat{e}_z v(\rho, z)$
- Jump of the pressure at the interface: $\sigma\kappa(z)$

$$\kappa(z) \approx \frac{1}{r - h(z)} + \frac{d^2h(z)}{dz^2}$$

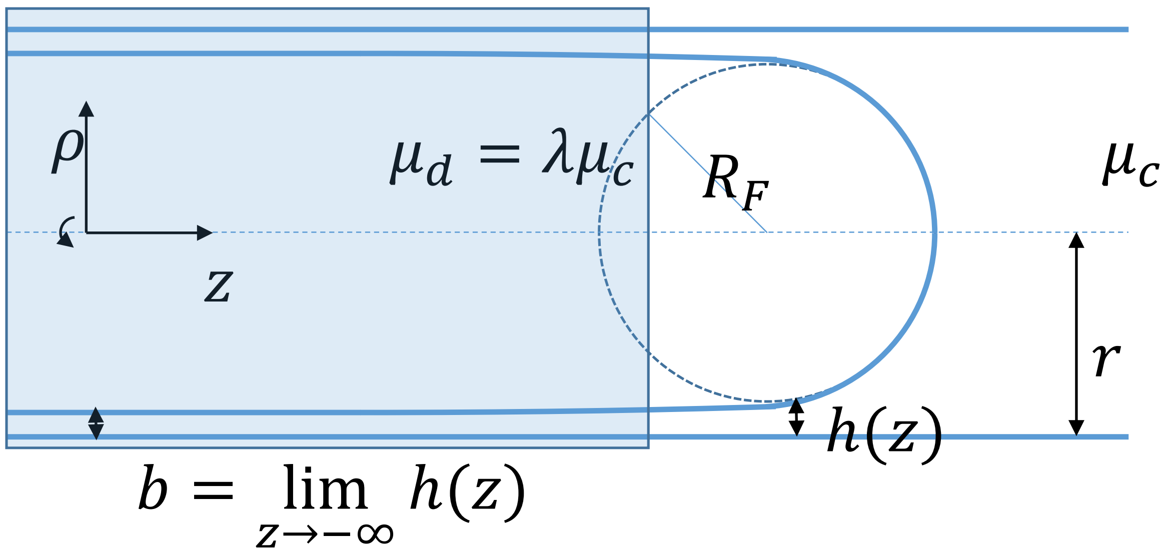


Bretherton (1961) – results and approximations

- Equation for shape of the interface

$$\eta(Z) \equiv \frac{h(z)}{b}$$

$$Z \equiv \frac{z}{b}(3Ca)^{\frac{1}{3}},$$



Landau-Levitch equation

$$\frac{d^3\eta(Z)}{dZ^3} = \frac{\eta - 1}{\eta^3}$$

Ondulations of the profile at the rear of the droplet

Parabola at the front: $\eta(Z) \approx A + BZ + PZ^2/2$

Bretherton matching:

$$\frac{1}{R_F(\approx r)} = \frac{d^2h(z)}{dz^2}$$

P from numerical integration of

Bretherton (1961) – result for mobility

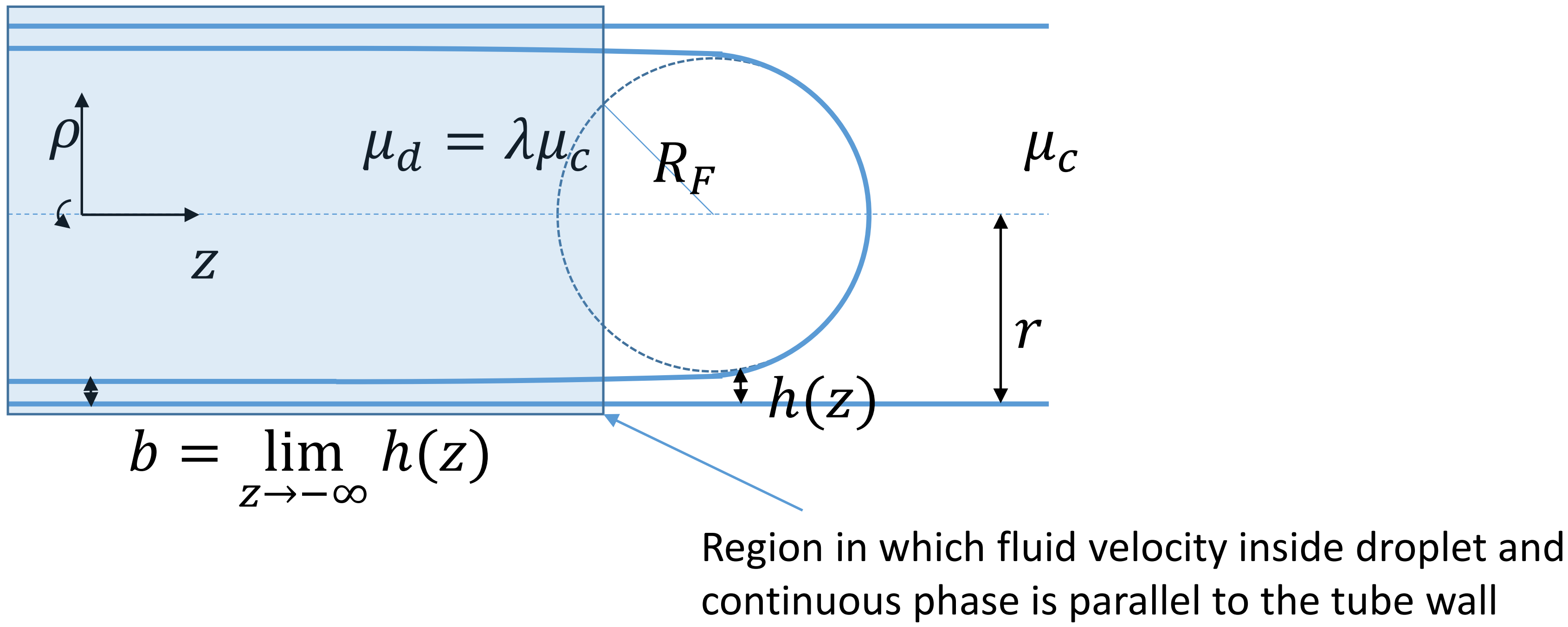
Inviscid droplet ($\lambda = 0$), low capillary number

(Bretherton, F., JFM, 1961, 10, 166-188)

$$\beta = 1 + 1.29(3Ca)^{\frac{2}{3}}$$

$$Ca = \frac{\mu_c V}{\sigma}$$

Goldsmith and Mason (1963)



By solving hydrodynamic (Stokes) equations in 'parallel region' they found relation between mobility and film thickness for viscous droplet:

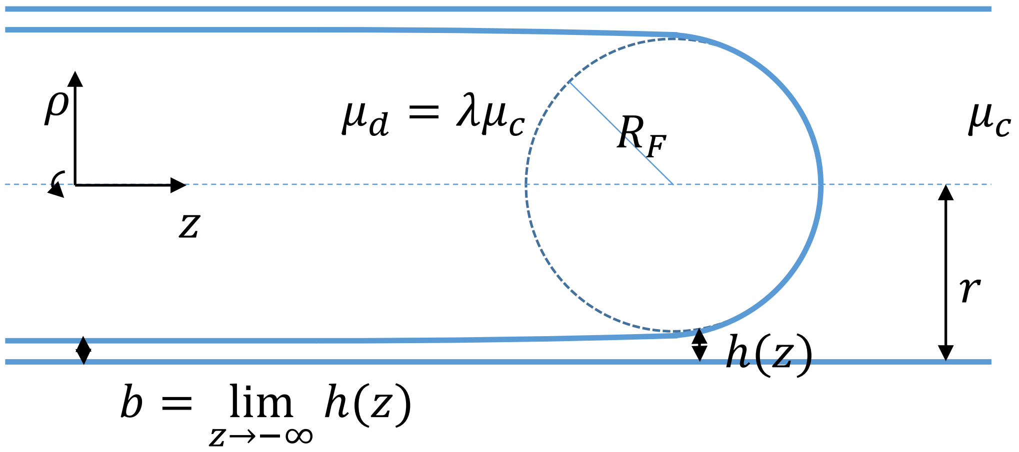
$$\frac{V}{U} = \frac{1 + \left(4\frac{b}{r} - 6\left(\frac{b}{r}\right)^2 + 4\left(\frac{b}{r}\right)^3 - \left(\frac{b}{r}\right)^4\right)(-1+\lambda)}{1 + \left(2\left(\frac{b}{r}\right) - \left(\frac{b}{r}\right)^2\right)(-1+2\lambda)}$$

Schwartz, Princen and Kiss (1986)

Extension of Bretherton's approach for viscous droplets:

$$\eta(Z) \equiv \frac{h(z)}{b} \qquad Z \equiv \frac{z}{b}\left(3Ca\, s(\delta)\right)^{\frac{1}{3}},$$

$$\delta \equiv \frac{\lambda b}{r} \qquad s(\delta) = \frac{1 + 2\delta - \frac{2\delta}{1+4\delta}}{1+\delta},$$



Equation for shape (lets call it viscous Landau-Levitch equation):

$$\frac{d^3\eta(Z)}{dZ^3} = \frac{\eta - 1}{\eta^3}\,\frac{1 + 2\delta\eta - \frac{2\delta}{1+4\delta}}{1 + \delta\eta}\,\frac{1+\delta}{1 + 2\delta - \frac{2\delta}{1+4\delta}}$$

Schwartz, Princen and Kiss (1986)

Ondulations at the rear (rescaled by viscosity od droplet)
Parabolic behavior at the front
Bretherton matching

$$\frac{b}{r} = (3Ca)^{\frac{2}{3}} P\left(\lambda \frac{b}{r}\right),$$

$P(x)$ simple fit given

The above formula allows to numerically find the film thickness (and therefore the droplet mobility)
For every viscosity ratio and capillary number the numerical solution is needed.
Is there a simple way to represent $\frac{b}{r} = \frac{b}{r}(Ca\,,\lambda)$?

Ballestra, Zhu, Gallaire (2017)

For theoretical part they start where Schwarz, Princen and Kiss finished:

$$\frac{b}{r} = (3Ca)^{\frac{2}{3}}\, P\left(\lambda \frac{b}{r}\right)$$

and introduce phenomenological approximation:

$$\frac{b}{r} = (3Ca)^{\frac{2}{3}}\, \bar{P}(\lambda)$$

$$\bar{P}(\lambda) = \frac{0.643}{2}\left\{1 + 2^{\frac{2}{3}} + \left(2^{\frac{2}{3}} - 1\right) tanh[1.28 \log_{10} \lambda - 2.36]\right\}$$

Therefore they have simple formula for the film thickness and thus mobility
-with another approximation that goes beyond Bretherton approximation

Our results

We start where Schwarz, Princen and Kiss finished:

$$\frac{b}{r} = (3Ca)^{\frac{2}{3}} P\left(\lambda \frac{b}{r}\right)$$

But we rewrite it: $\lambda \frac{b}{r} = \lambda (3Ca)^{\frac{2}{3}} P\left(\lambda \frac{b}{r}\right)$ $\quad \delta = \lambda (3Ca)^{\frac{2}{3}} P(\delta) \quad \delta \equiv \lambda \frac{b}{r}$

Value of δ depends solely on $\lambda (3Ca)^{\frac{2}{3}}$ parameter!!!
Numerical solution gives:

$$\delta \equiv \lambda \frac{b}{r} = c\left(\lambda (3Ca)^{\frac{2}{3}}\right)$$

$$c_{fit}(g) = t(0) + \frac{g + b_4 g^2 + \left(t(0) 2^{2/3} - t(0)\right) g^3}{b_1 + b_2 g + b_3 g^2 + g^3}$$

$$b_1 = 4.109, b_2 = 8.906, b_3 = 10.144, b_4 = 3.575.$$

Easy to use expression for mobility
(lubrication approximation + Bretherton matching)

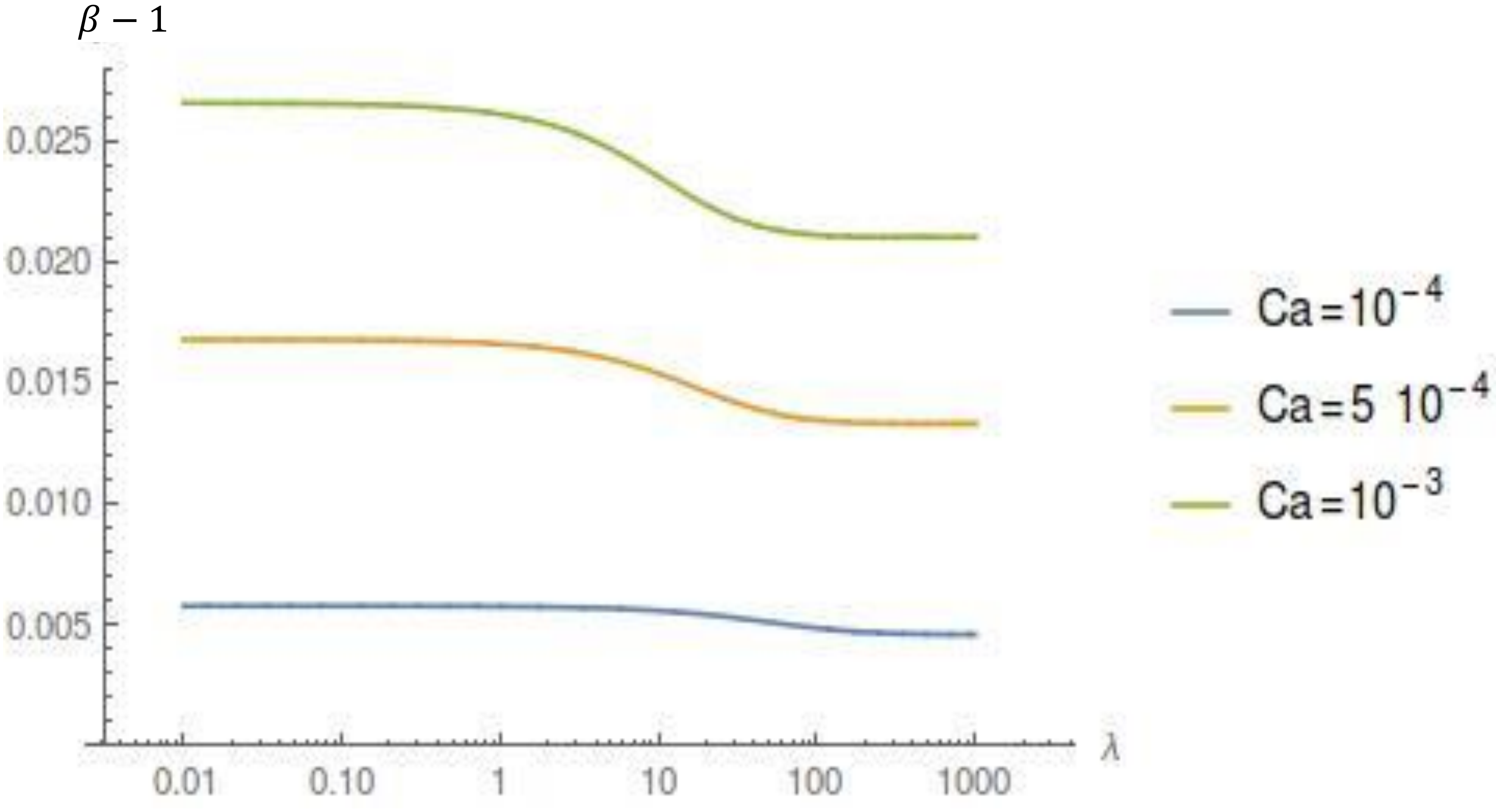
$$Ca = \frac{\mu_c V}{\sigma} \qquad \lambda = \frac{\mu_d}{\mu_c} \qquad g \equiv \lambda(3Ca)^{\frac{2}{3}}$$

$$b_1 = 4.109, b_2 = 8.906, b_3 = 10.144, b_4 = 3.575, \mathrm{t}(0) = 0.643$$

$$c_{fit}(g) = t(0) + \frac{g + b_4 g^2 + \left(t(0)2^{2/3} - t(0)\right) g^3}{b_1 + b_2 g + b_3 g^2 + g^3}$$

$$\epsilon \equiv \frac{b}{r} = (3Ca)^{\frac{2}{3}}\, c_{fit}\left(\lambda(3Ca)^{\frac{2}{3}}\right),$$

$$\beta = \frac{U}{V} = \frac{1 + (-2\epsilon + \epsilon^2)(1 - 2\lambda)}{1 + (4\epsilon - 6\epsilon^2 + 4\epsilon^3 - \epsilon^4)(-1 + \lambda)}$$



$\beta - 1$
0.025
0.020
0.015
0.010
0.005
0.01
0.10
1
10
100
1000
λ
Ca=10^-4
Ca=5 10^-4
Ca=10^-3

Schwartz, Princen and Kiss (1986)

Extension of Bretherton's approach for viscous droplets:

Between parallel plates:

Teletzke, G. F.; Davis, H. T. & Scriven, L. Revue de Physique Appliquee, 1988, 23, 989-1007

In channels of circular cross-sections:

Hodges, S.; Jensen, O. & Rallison, J., JFM, 2004, 501, 279-301

$$\beta = 1 + 1.29(3Ca)^{\frac{2}{3}}$$

Dominant effects

water droplets in FC-40 oil
tube diameter: $d = 0.8\,mm$

Viscous and interfacial forces: $Ca = \frac{\mu_c V}{\sigma}$ ← ≈ 0.001 for $V = 1\,\frac{\text{cm}}{s}$

Inertial and viscous effects: $Re = \frac{\rho_c d V}{\mu_c}$ ← ≈ 3.6 for $V = 1\,\frac{\text{cm}}{s}$

Gravitational and interfacial effects: $Bo = \frac{\Delta\rho d^2}{\sigma} \approx 0.01$

Theoretical model: stationary Stokes equations

Governing parameters:

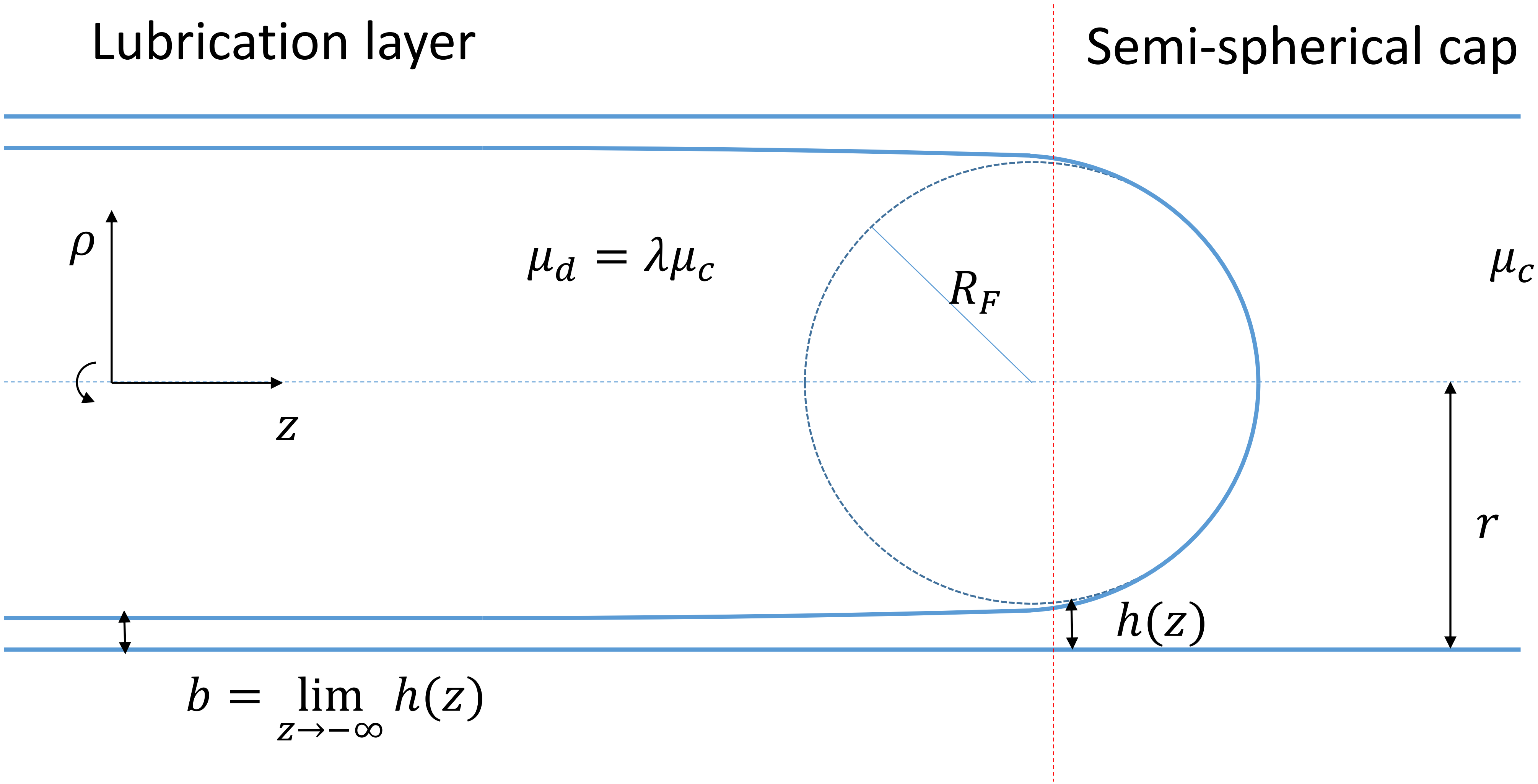
- Ratio of viscosities $\lambda = \frac{\mu_d}{\mu_c}$
- Capillary number $Ca = \frac{\mu_c V}{\sigma}$
- Length of droplet l

(numerical simulations: mobility does not change for $l > 2d$)

tube diameter

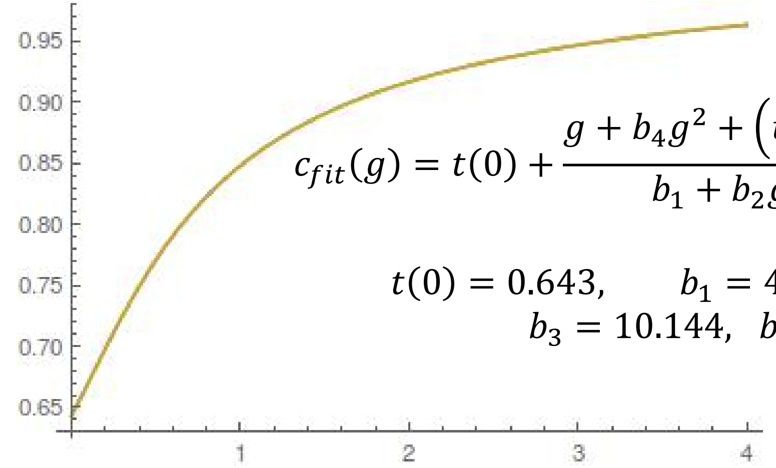
Lac, E. & Sherwood, J., JFM, 2009, 640, 27-54

Theoretical model: stationary Stokes equations



Our theoretical results

Consequent application of low Ca condition leads to scaling, and mobility of droplet ($\lambda = \frac{\mu_d}{\mu_c}$, $Ca = \frac{\mu_c V}{\sigma}$,) can be calculated as follows:



$$c_{fit}(g) = t(0) + \frac{g + b_4 g^2 + \left(t(0)2^{2/3} - t(0)\right) g^3}{b_1 + b_2 g + b_3 g^2 + g^3}$$

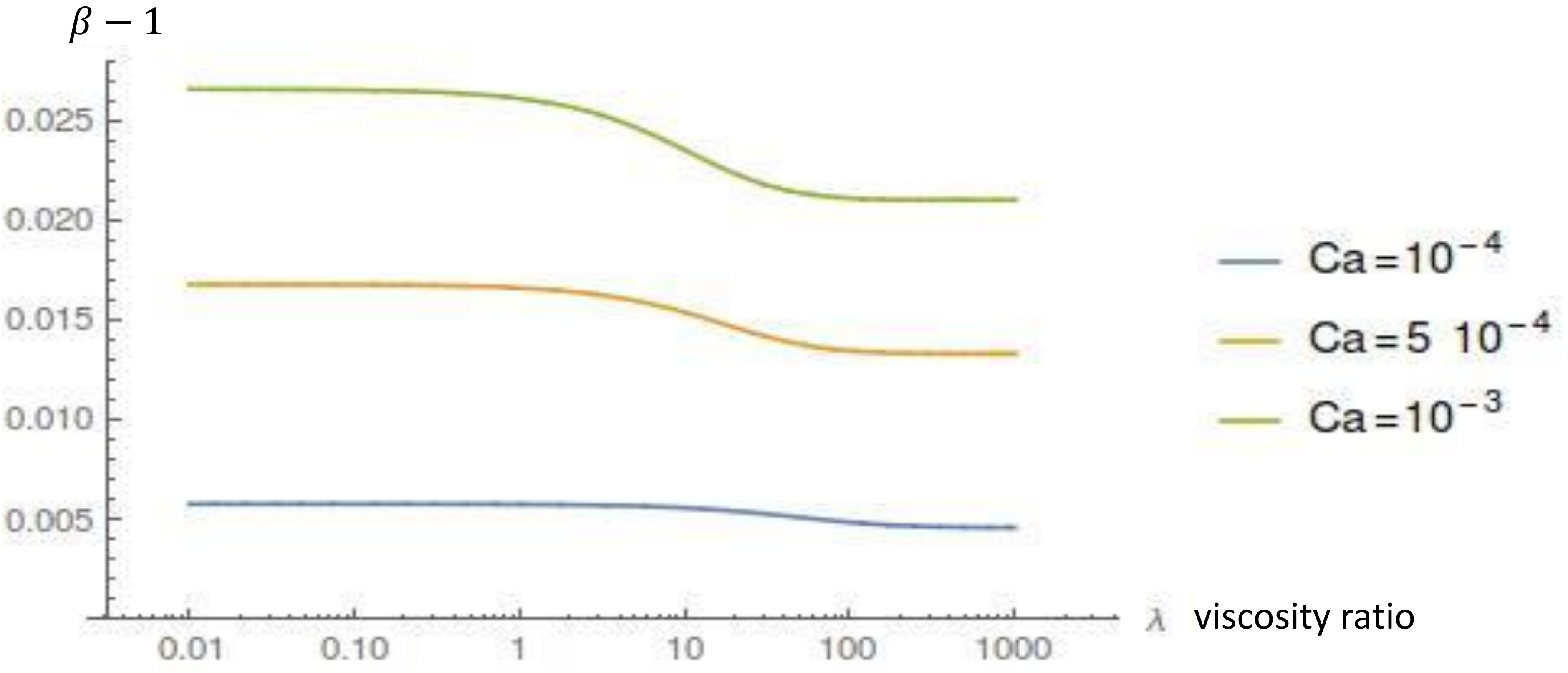
$t(0) = 0.643,\quad b_1 = 4.109,\quad b_2 = 8.906,$
$b_3 = 10.144,\quad b_4 = 3.575.$

Film thickness:

$$\frac{b}{r} = (3Ca)^{\frac{2}{3}} c\left(\lambda (3Ca)^{\frac{2}{3}}\right),$$

$$\beta \equiv \frac{U}{V} = 1 + 2\frac{b}{r}\frac{1 + 2\frac{b}{r}\lambda}{1 + 4\frac{b}{r}\lambda}.$$

Our theoretical results



→ To observe the above changes with viscosity, relative error of measurement of mobility should be less than about 1/1000

Summary

- Theory: practical formula for mobility of long, non-wetting droplets
- Preliminary experimental verification (error of measurements < 1/1000)

Further goals:

- broader range of capillary numbers and viscosities
- can that be used to measure viscosity, surface tension? what accuracy?

In collaboration with Michał Horka, Jean Baptiste-Gorce and Piotr Garstecki

Stokes equations in cylindrical system

$$\partial_z p_d(\rho, z) = \mu_d \rho^{-1} \partial_\rho \rho \partial_\rho v_d(\rho, z),$$

$$\partial_\rho p_d(\rho, z) = 0$$

$$\frac{1}{\rho} \partial_\phi p_d(\rho, z) = 0,$$

$$\nabla \cdot \vec{v}_d = 0$$

Boundary conditions

$$v_c(\rho, z)\Big|_{\rho=r-h(z)} = v_d(\rho, z)\Big|_{\rho=r-h(z)},$$

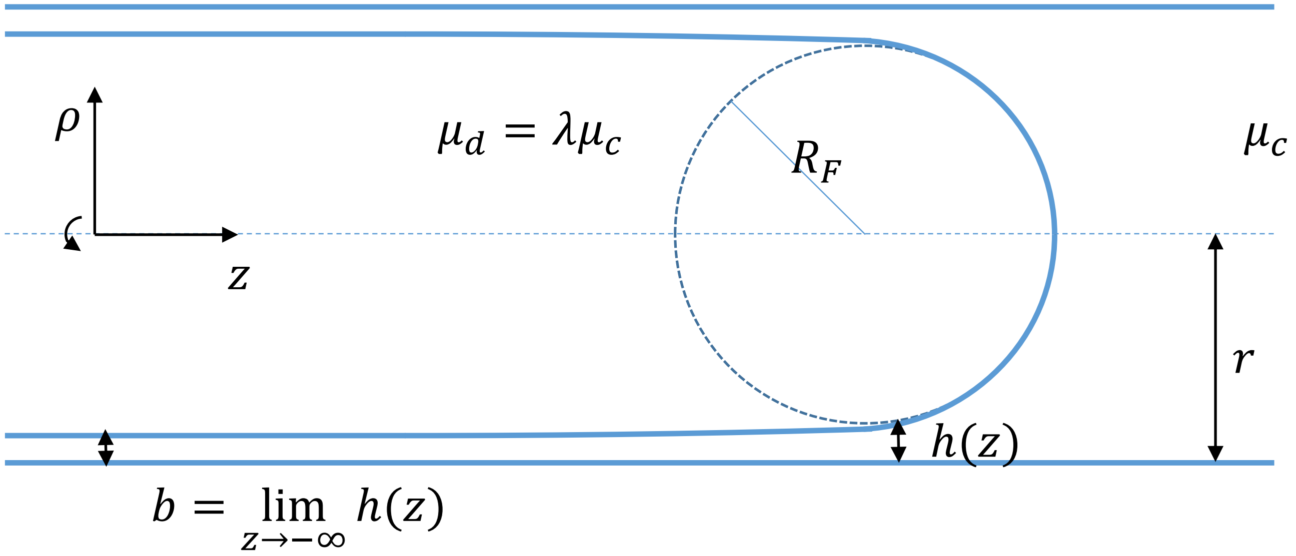
$$\mu_c \partial_\rho v_c(\rho, z)\Big|_{\rho=r-h(z)} = \mu_d \partial_\rho v_d(\rho, z)\Big|_{\rho=r-h(z)},$$

$$p_c(\rho, z)\Big|_{\rho=r-h(z)} + \sigma\kappa(z) = p_d(\rho, z)\Big|_{\rho=r-h(z)},$$

$$v_c(\rho, z)|_{\rho=r} = 0.$$

In lubrication region pressure does not depend on ρ.
It simplifies boundary condition for pressure jump.

$$\kappa(z) \approx \frac{1}{r - h(z)} + \frac{d^2 h(z)}{dz^2}.$$



$$\frac{dp_c(z)}{dz} + \sigma \frac{d^3 h(z)}{dz^3} = \frac{dp_d(z)}{dz}$$

..which we use in Stokes equation for z direction (for droplet and continuous phase):

$$\frac{\mathrm{d}p_d(z)}{\mathrm{dz}} = \mu_d \rho^{-1} \partial_\rho \rho \partial_\rho v_d(\rho, z),$$

..which with boundary condition gives $v_d(\rho, z)$ and $v_c(\rho, z)$ for given z:

Velocity field in lubrication regime

$$v_d\,(\rho,z) = \frac{1}{4\mu_d}\frac{dp_d(z)}{dz}\left(\rho^2 - (r-h)^2\right) + \frac{1}{4\mu_c}\left(\frac{dp_d(z)}{dz} - \sigma\frac{d^3h(z)}{dz^3}\right)\left((r-h)^2 - r^2\right) + \frac{1}{2\mu_c}\sigma\frac{d^3h(z)}{dz^3}(r-h)^2\ln\left(\frac{r-h}{r}\right)$$

$$v_c(\rho,z) = \frac{1}{4\mu_c}\left(\frac{dp_d(z)}{dz} - \sigma\frac{d^3h(z)}{dz^3}\right)(\rho^2 - r^2) + \frac{1}{2\mu_c}\sigma\frac{d^3h(z)}{dz^3}\ln\left(\frac{\rho}{r}\right)$$

…but $\frac{dp_d(z)}{dz}$ and $\frac{d^3h(z)}{dz^3}$ are unknown (we may treat z as parameter)

Two unknows $\frac{dp_d(z)}{dz}$ and $\frac{d^3h(z)}{dz^3}$ are determine from two conditions for flux:

Total flux through the cross section is the same for all z.

$$\pi r^2 V = 2\pi \int_0^{r-h(z)} d\rho\, \rho\, v_d(\rho, z) + 2\pi \int_{r-h(z)}^{r} d\rho\, \rho\, v_c(\rho, z)$$

In the coordinate system in which droplet is at rest,
the flux of the droplet phase vanishes.

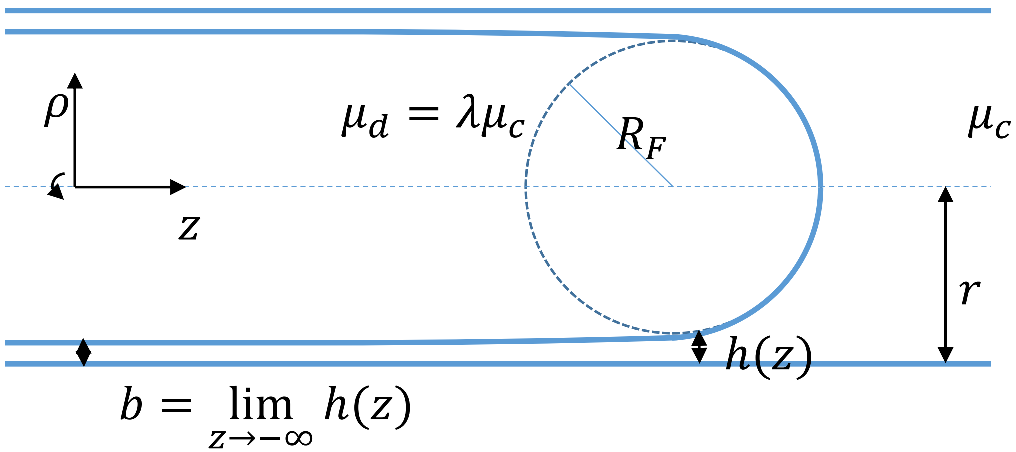
$$2\pi \int_0^{r-h(z)} d\rho\, \rho\, (\, v_d(\rho, z) - U\,) = 0$$

Equation for shape of the droplet in lubrication regime

Rescaling…

$$\eta(Z) \equiv \frac{h(z)}{b} \qquad \epsilon \equiv \frac{b}{r}$$

$$\delta \equiv \lambda\epsilon$$



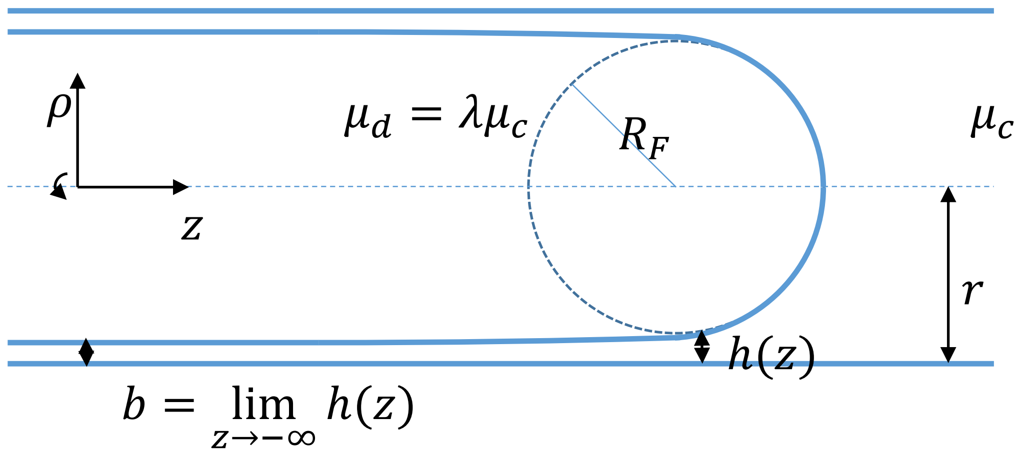
$$s(\delta) = \frac{1 + 2\delta - \dfrac{2\delta}{1+4\delta}}{1+\delta}, \qquad Z \equiv \frac{z}{b}\left(3Ca\, s(\delta)\right)^{\frac{1}{3}},$$

Equation for shape (lets call it viscous Landau-Levitch equation):

$$\frac{d^3\eta(Z)}{dZ^3} = \frac{\eta - 1}{\eta^3} \frac{1 + 2\delta\eta - \dfrac{2\delta}{1+4\delta}}{1+\delta\eta} \frac{1+\delta}{1 + 2\delta - \dfrac{2\delta}{1+4\delta}}$$

Linearized equation for shape of the droplet

$$\eta(Z) \equiv \frac{h(z)}{b}$$



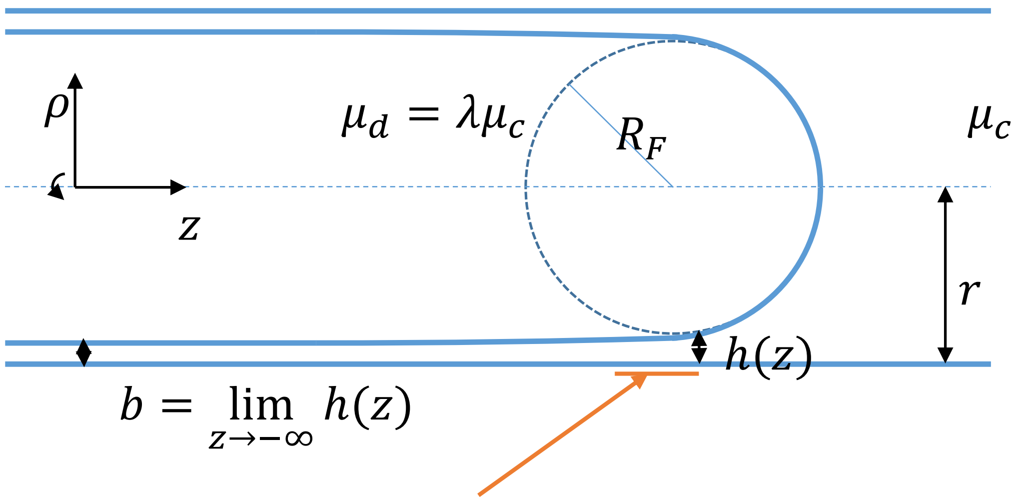
Linearized viscous Landau-Levitch equation (for $\eta(Z) - 1 \ll 1$)

$$\frac{d^3\eta(Z)}{dZ^3} = \eta - 1$$

$$\eta(Z) = 1 + \alpha_1 e^{Z} + \alpha_2 e^{-\frac{1}{2}Z} \cos\frac{\sqrt{3}}{2}Z + \alpha_3 e^{-\frac{1}{2}Z} \sin\frac{\sqrt{3}}{2}Z$$

Linearized equation for shape of the droplet

$$\eta(Z) \equiv \frac{h(z)}{b}$$



Linearized viscous Landau-Levitch equation (for $\eta(Z) \gg 1$, but $\eta(Z) \ll \frac{r}{b}$)

$$\frac{d^3\eta(Z)}{dZ^3} \approx 0$$

$$\eta(Z) = \frac{t(\delta)}{2}\left(Z - Z_0(\delta)\right)^2 + q(\delta)$$

Therefore viscous LL equation

$$\frac{d^3\eta(Z)}{dZ^3} = \frac{\eta - 1}{\eta^3} \frac{1 + 2\delta\eta - \frac{2\delta}{1+4\delta}}{1+\delta\eta} \frac{1+\delta}{1 + 2\delta - \frac{2\delta}{1+4\delta}}$$

may be solved numerically for given $\delta \equiv \frac{\lambda b}{r}$ starting from function

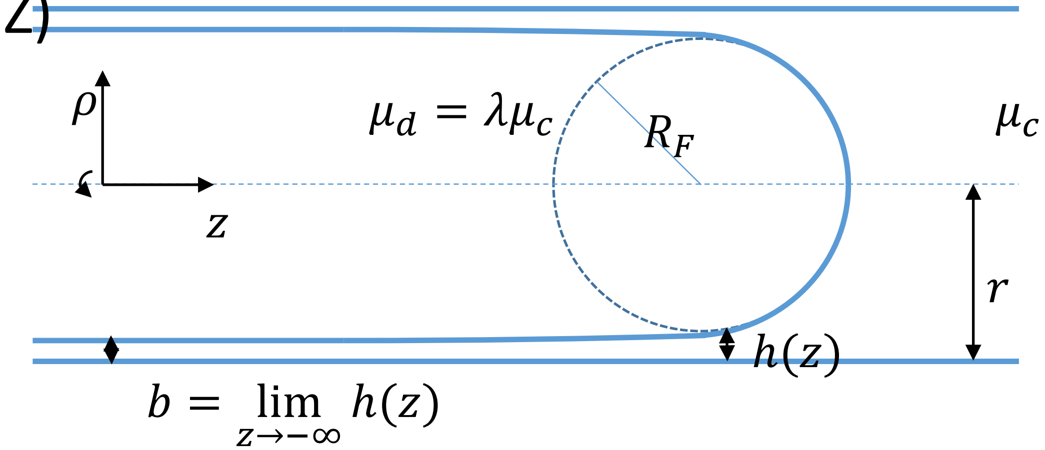
$$\eta(Z) = 1 + e^Z \quad \text{(small Z)}$$

for small Z. In this way we can determine $t(\delta)$ in large Z behavior:

$$\eta(Z) = \frac{t(\delta)}{2}\left(Z - Z_0(\delta)\right)^2 + q(\delta) \quad \text{(large Z)}$$

This determines curvature which is matched to semi-spherical cap

$$\frac{1}{R_F(\approx r)} = \frac{d^2h(z)}{dz^2}$$



Matching condition leads to the following equation for δ

$$\delta \equiv \frac{\lambda b}{r}$$

$$\delta = \underline{\lambda(3Ca)^{\frac{2}{3}}}\, t(\delta)\, s(\delta)^{\frac{2}{3}}$$

$$s(\delta) = \frac{1 + 2\delta - \frac{2\delta}{1+4\delta}}{1+\delta}$$

Lets denote it by $g \equiv \lambda(3Ca)^{\frac{2}{3}}$.

Given numerically from the solution of viscous LL equation.

Value of δ depends solely on g parameter!!!