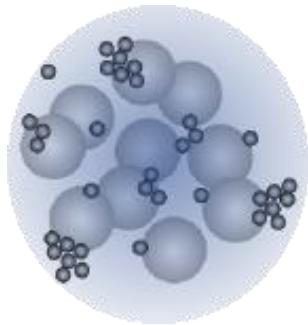


# Sticking between Fine Particles by Capillary Bridges

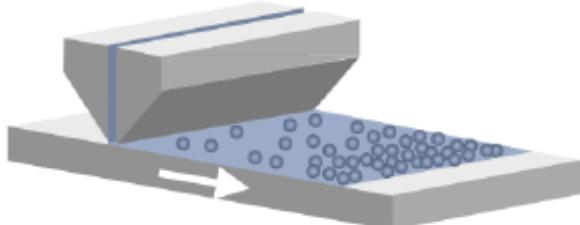
液架橋による粒子間固着作用

- 辰巳 恵 (東大環安セ)  
小池 修 (PIA)
- 山口由岐夫 (PIA)
- 辻 佳子 (東大環安セ/東大院工)

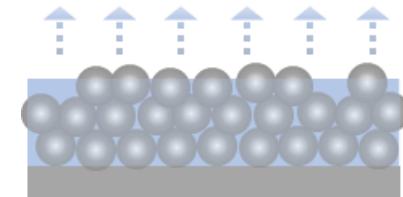
# Material Fabrication from Colloidal Suspensions<sup>2</sup>



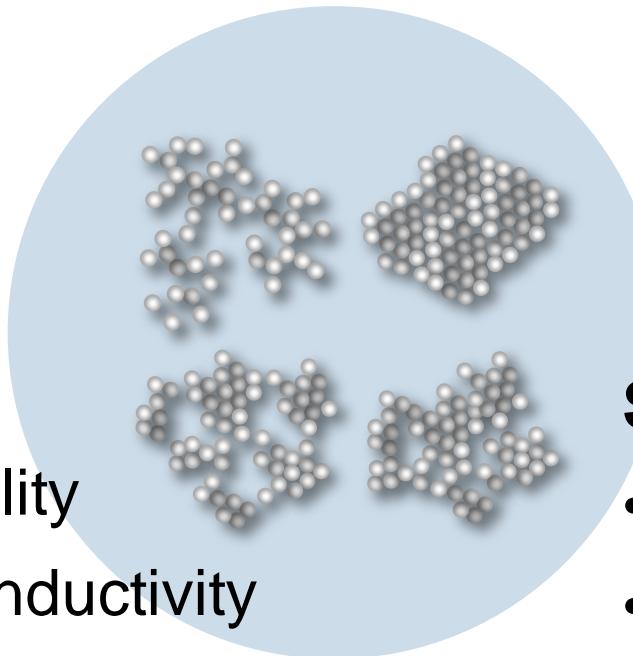
Dispersing



Coating



Drying



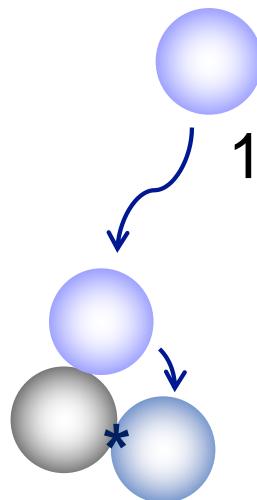
## Functions

- Strength
- Permeability
- Electrical/Thermal conductivity
- Optical property

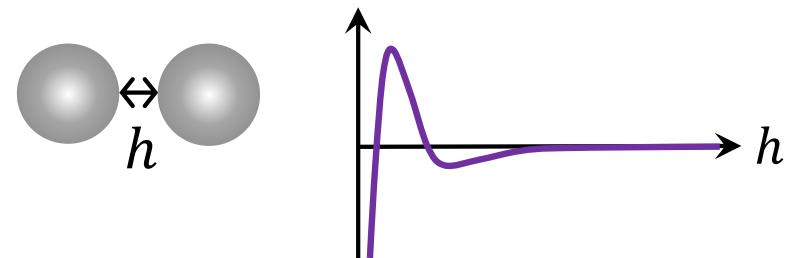
## Structures

- Porosity
- Contact network

# Aggregate Formation

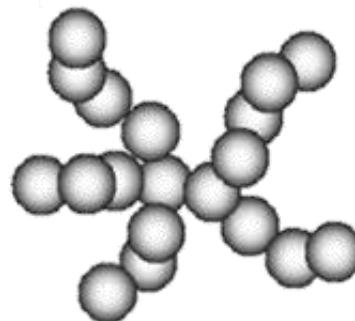


1. Collision (Diffusion) ← Interparticle potential

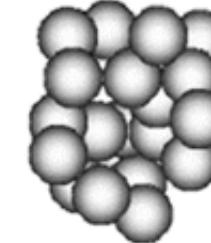


2. Sticking (Reaction): Fixation of contact points

← Interparticle slip/rolling friction

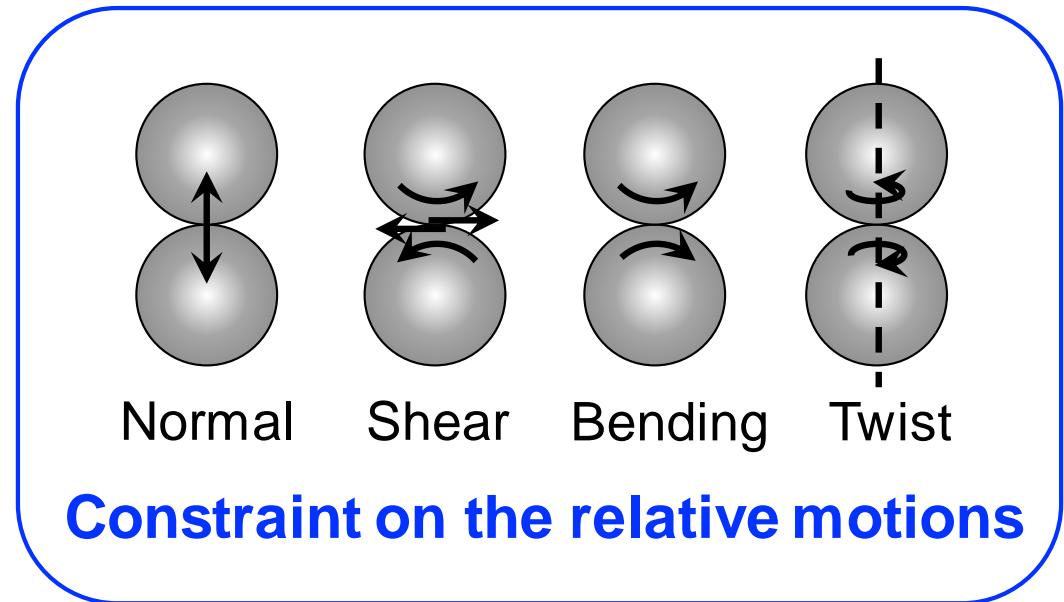
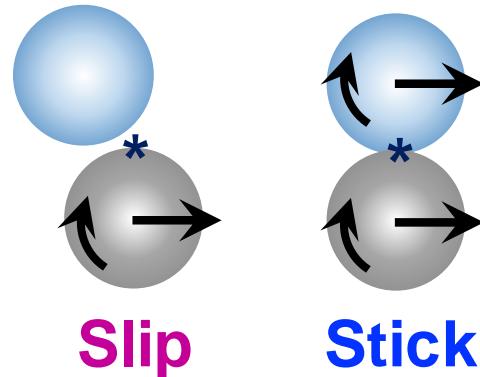


Diffusion-limited



Reaction-limited

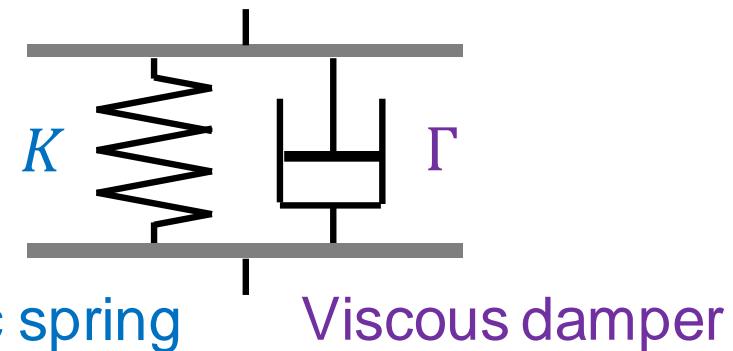
# Modeling of Sticking



Contact force/torque

$$F = K\delta + \Gamma\dot{\delta}$$

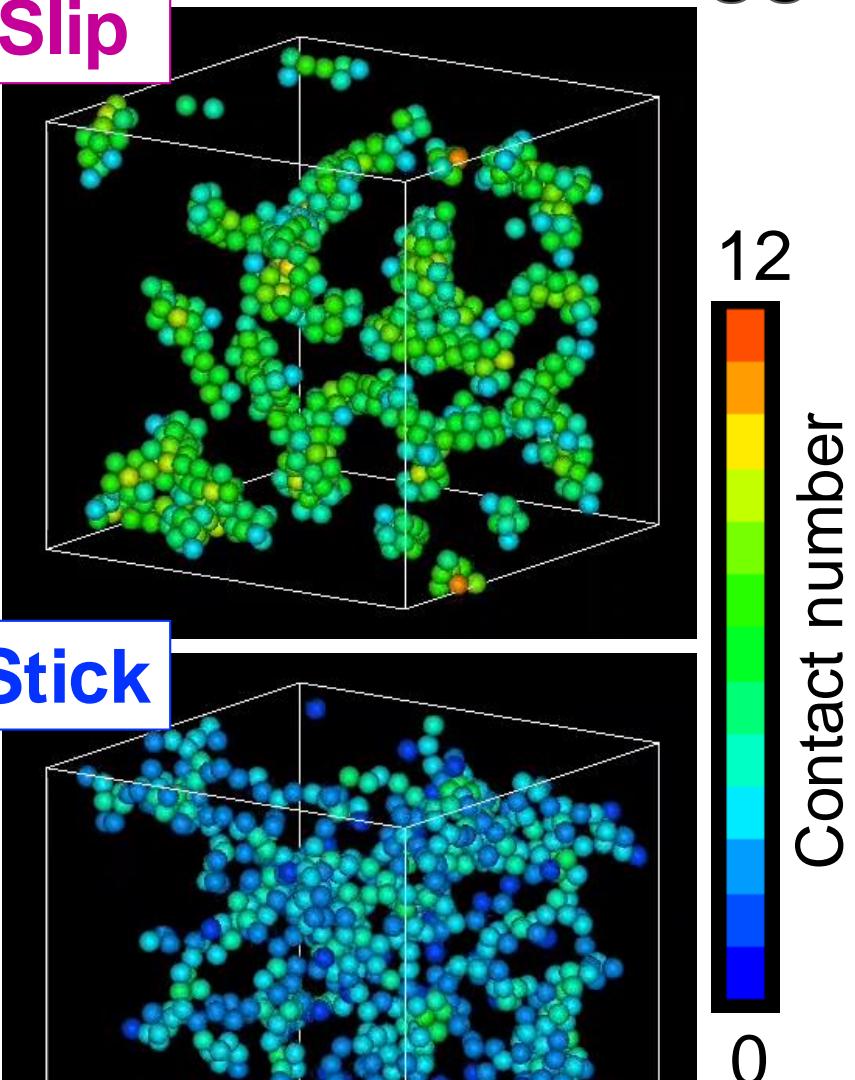
$\delta$ : Displacement



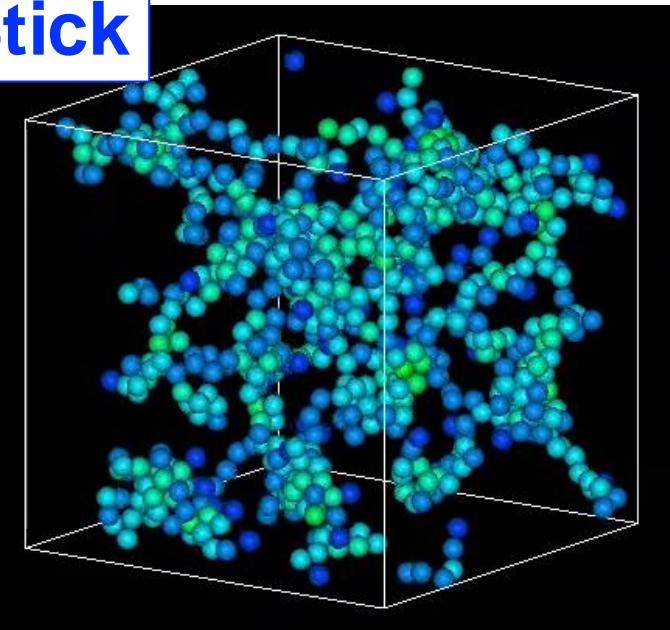
Langevin eq.

# Aggregation

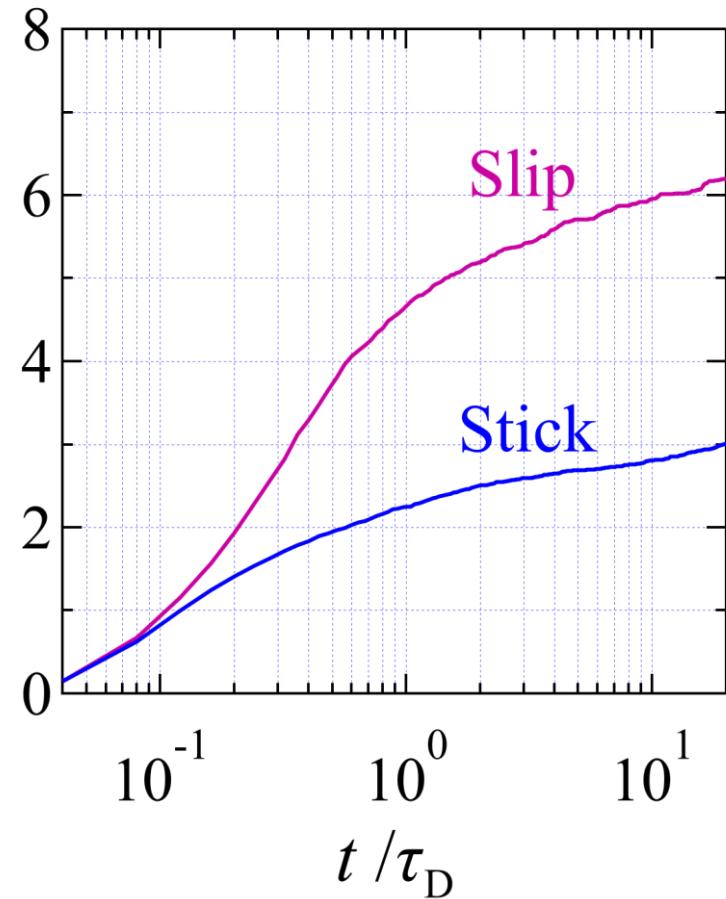
Slip



Stick



Average contact number

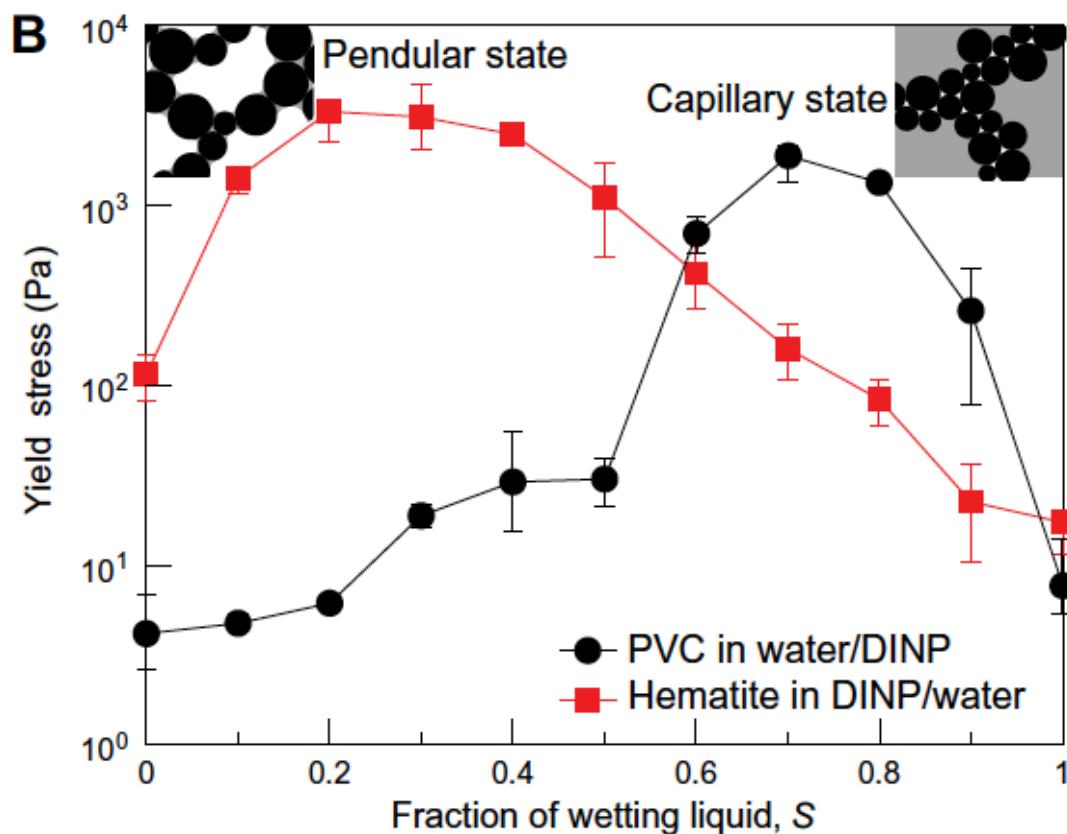
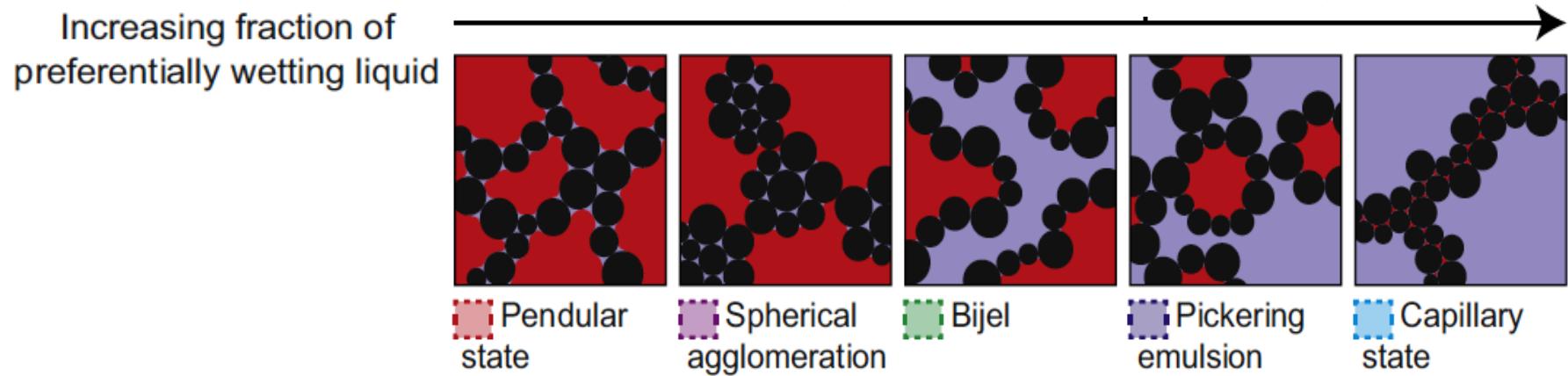


Timescale of particle diffusion:

$$\tau_D = d^2 / D$$

Particle diameter:  $d$  Diffusion coefficient:  $D$

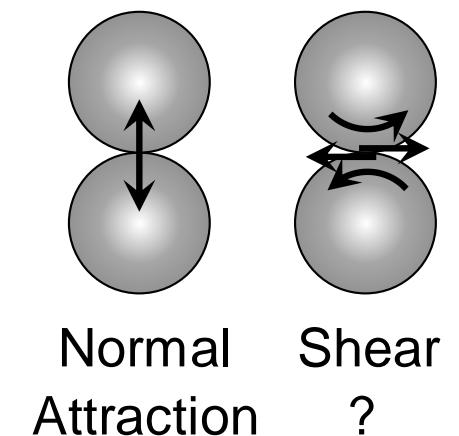
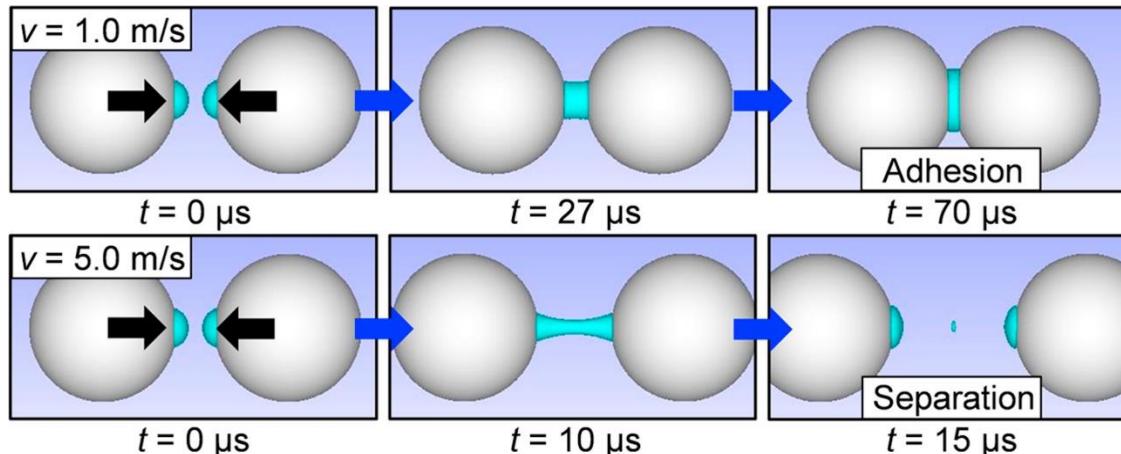
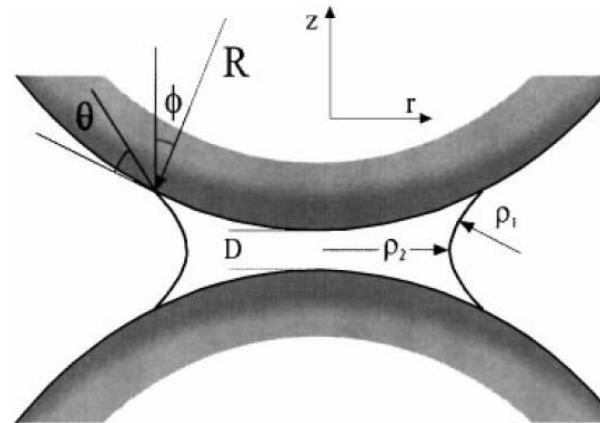
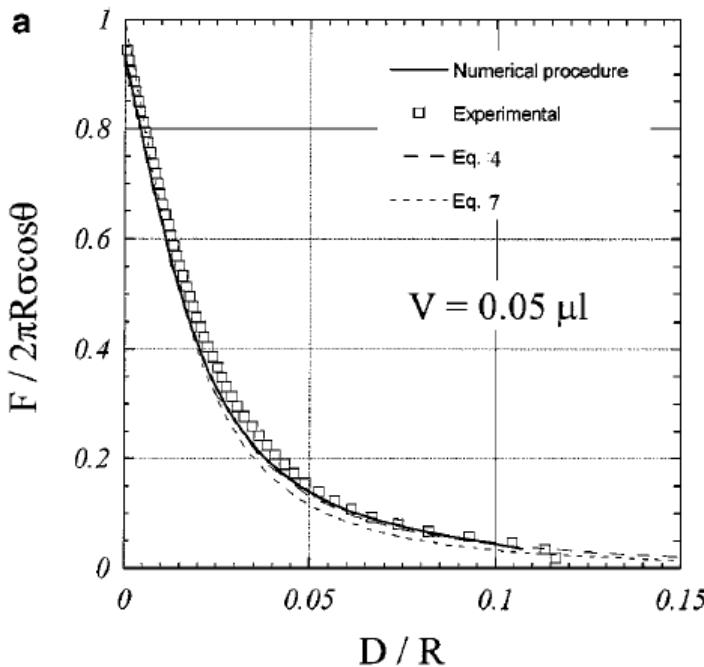
# A Possible Origin of Sticking



Network formation by capillary bridges

E. Koos,  
Curr. Opin. Colloid Interface Sci.  
**19** (2014).

# Capillary Bridge Force

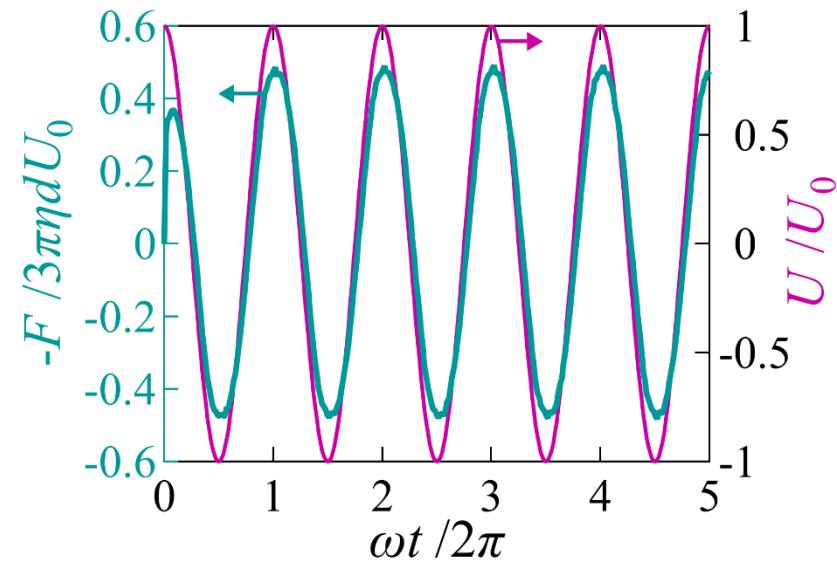
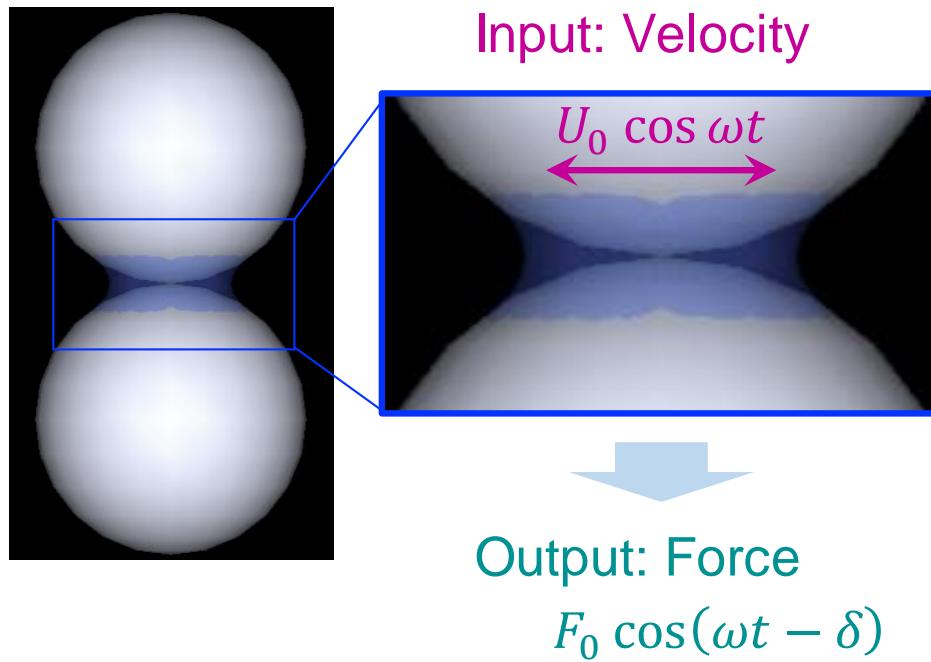


O. Pitois, P. Moucheront, X. Chateau, J. Colloid Interface Sci. **231** (2000).

H. Kan, H. Nakamura, S. Watano, Chem. Eng. Sci. **138** (2015).

# Objective

- ◆ Examination of sticking by capillary bridges
- ◆ Calculation of the viscoelasticity of shear force induced by capillary bridges



# Equations

**Fluid flow**     $\nabla \cdot \boldsymbol{v} = 0$

$$\rho \left( \frac{\partial \boldsymbol{v}}{\partial t} + \boldsymbol{v} \cdot \nabla \boldsymbol{v} \right) = \nabla \cdot \boldsymbol{\sigma} + \Phi \boldsymbol{K}$$

$$\boldsymbol{\sigma} = -p \boldsymbol{I} + [\nabla \boldsymbol{v} + (\nabla \boldsymbol{v})^T] + \gamma (\boldsymbol{I} - \hat{\boldsymbol{n}} \hat{\boldsymbol{n}}) |\nabla f|$$

Surface tension

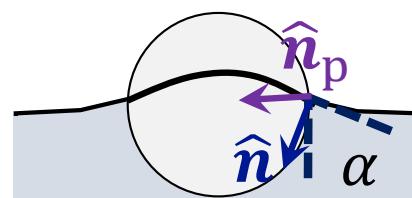
Imposition of velocity  
in particle domains:  $\Phi \boldsymbol{v} = \Phi \boldsymbol{V}$

**Free surface**

$$\frac{\partial f}{\partial t} + \boldsymbol{v} \cdot \nabla f = 0 \quad \text{Contact angle condition}$$

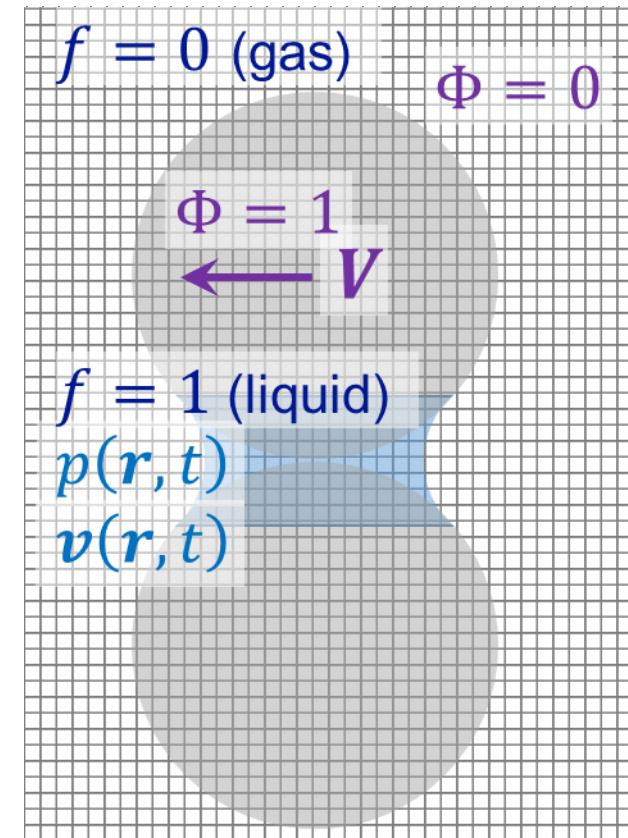
$$\hat{\boldsymbol{n}} \cdot \hat{\boldsymbol{n}}_p = \cos \alpha$$

$$\hat{\boldsymbol{n}} = \nabla f / |\nabla f|$$



**Drag force**

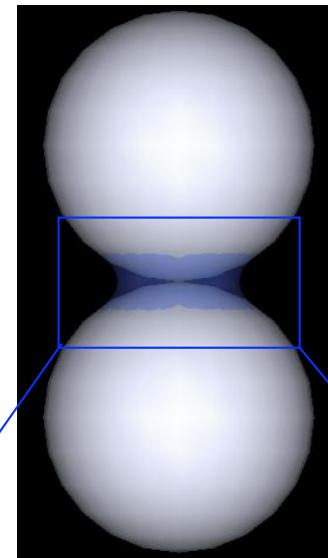
$$\boldsymbol{F} = \int \Phi \nabla \cdot \boldsymbol{\sigma} d\boldsymbol{r}$$



# Simulation Conditions

## Particles

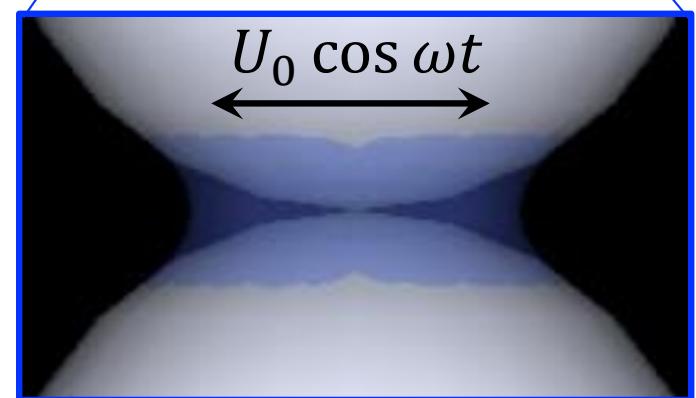
- Diameter:  $d = 100 \text{ nm}$
- Contact angle:  $0^\circ$



## Fluid: water

- Volume:  $0.012d^3$
- Surface tension:  $\gamma^* = 0.18 - 7.2$

$$\gamma^* = \frac{\rho d}{\eta^2} \gamma \quad \begin{matrix} \text{Fluid density: } \rho \\ \text{Viscosity: } \eta \end{matrix}$$



## Motion

- Displacement:  $\delta_0 = 0.01d$
- Velocity amplitude:  $U_0 = \delta_0 \omega$
- Frequency:  $\omega \tau_{\text{vis}} = 3 - 300$

$$\tau_{\text{vis}} = \frac{\rho d^2}{\eta} \quad \begin{matrix} \text{Timescale of momentum diffusion by viscosity} \end{matrix}$$

# Dynamic Drag Coefficients

Input: Velocity

$$U(t) = U_0 \cos \omega t$$

Output: Force

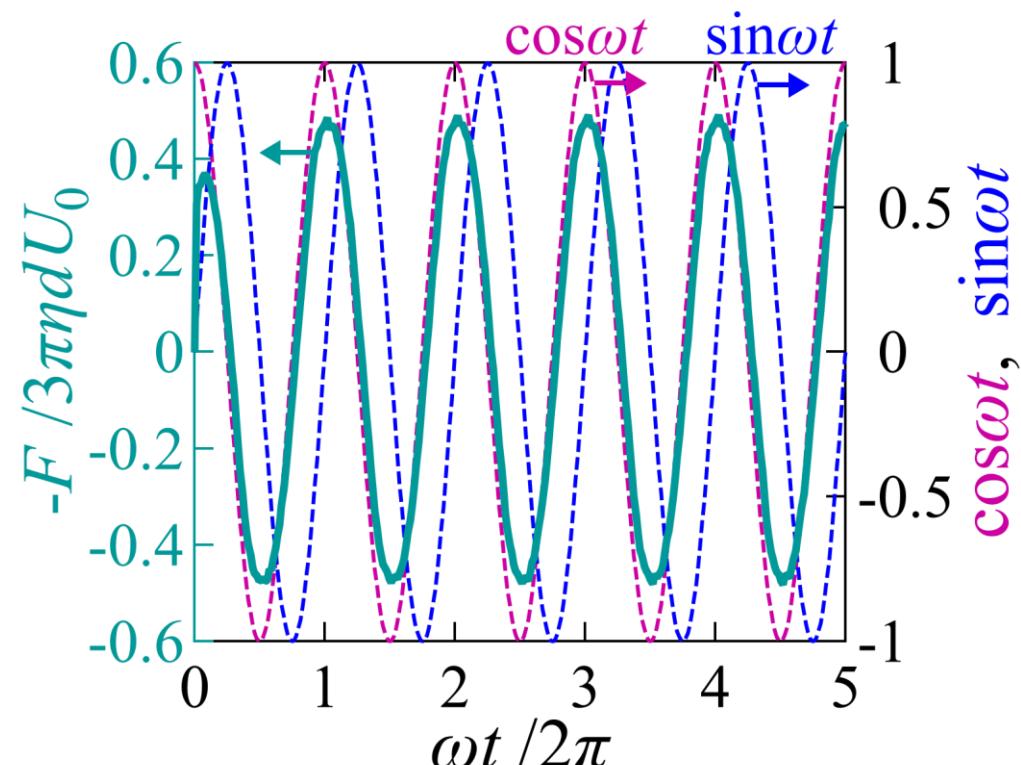
$$F(t) = F_0 \cos(\omega t - \delta)$$

$$= -U_0 (\zeta' \cos \omega t + \zeta'' \sin \omega t)$$

$$\zeta' = \frac{F_0}{U_0} \sin \delta \quad \zeta'' = \frac{F_0}{U_0} \cos \delta$$

Viscous part

Elastic part

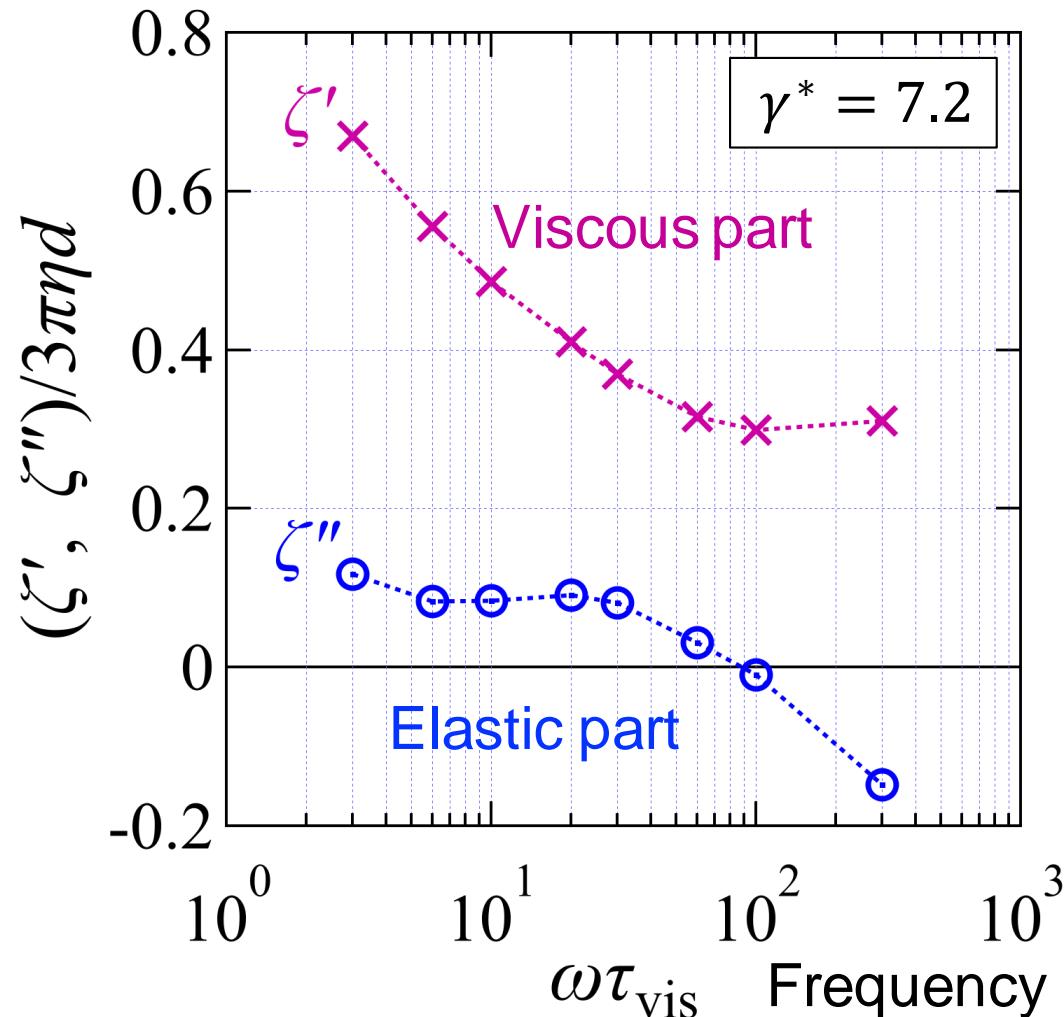


Calculation of dynamic drag coefficients

$$\zeta'(\omega) = -\frac{\omega}{\pi U_0} \int_0^{2\pi/\omega} F(t) \cos \omega t \, dt$$

$$\zeta''(\omega) = -\frac{\omega}{\pi U_0} \int_0^{2\pi/\omega} F(t) \sin \omega t \, dt$$

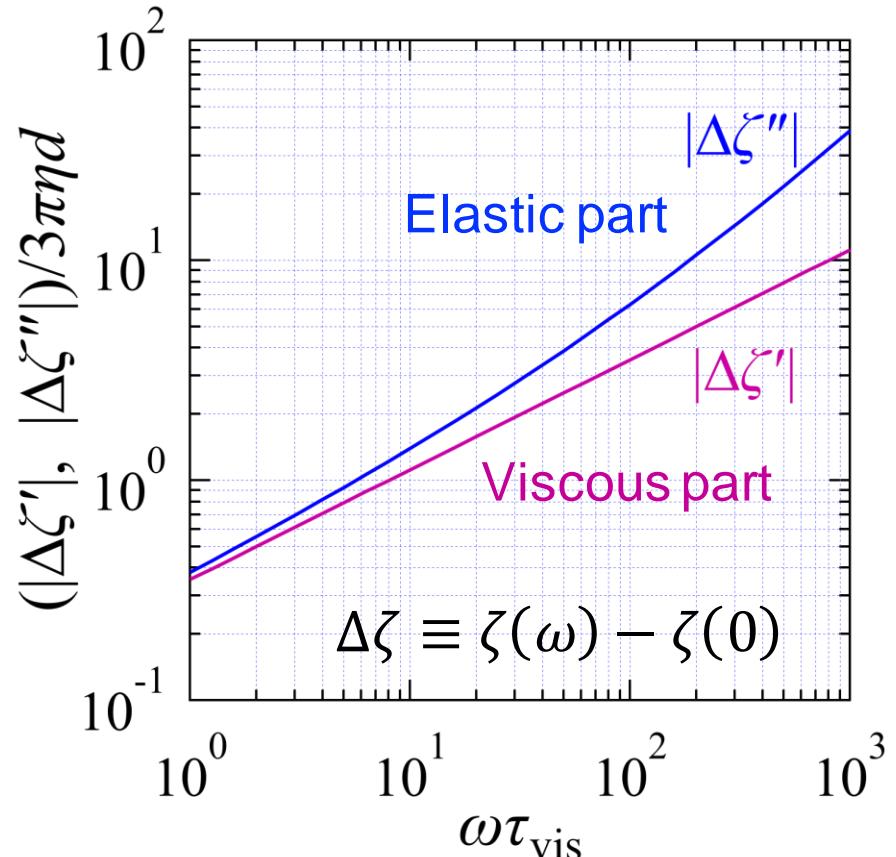
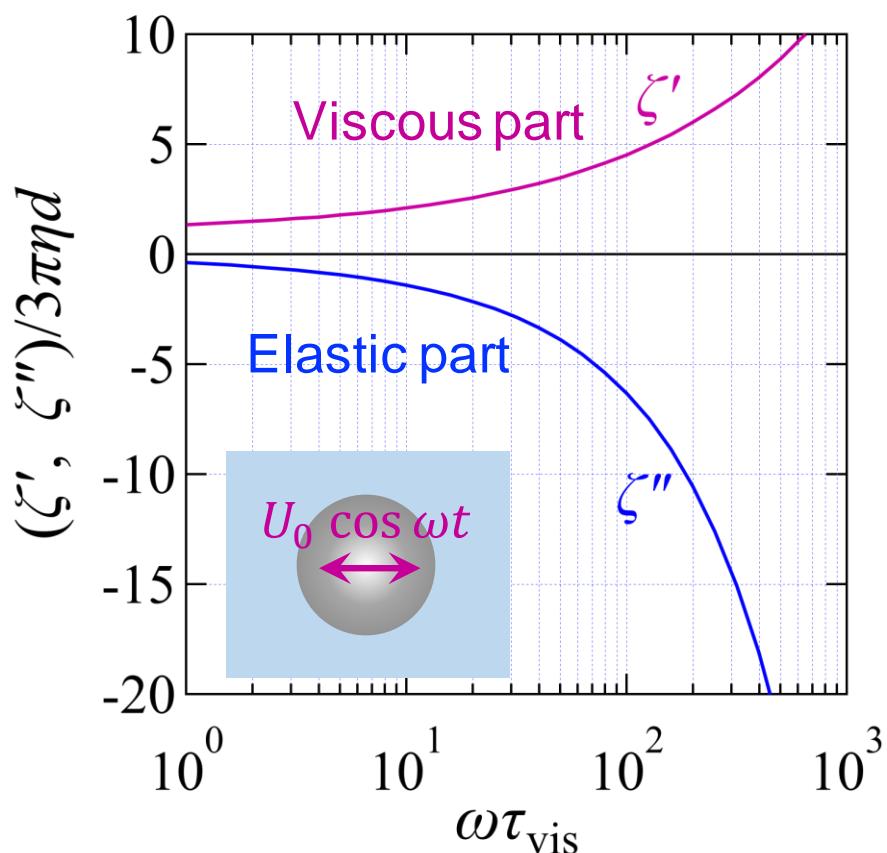
# Dynamic Drag Coefficients



- Viscoelasticity of shear force by capillary bridges
- Negative elastic part at high frequency

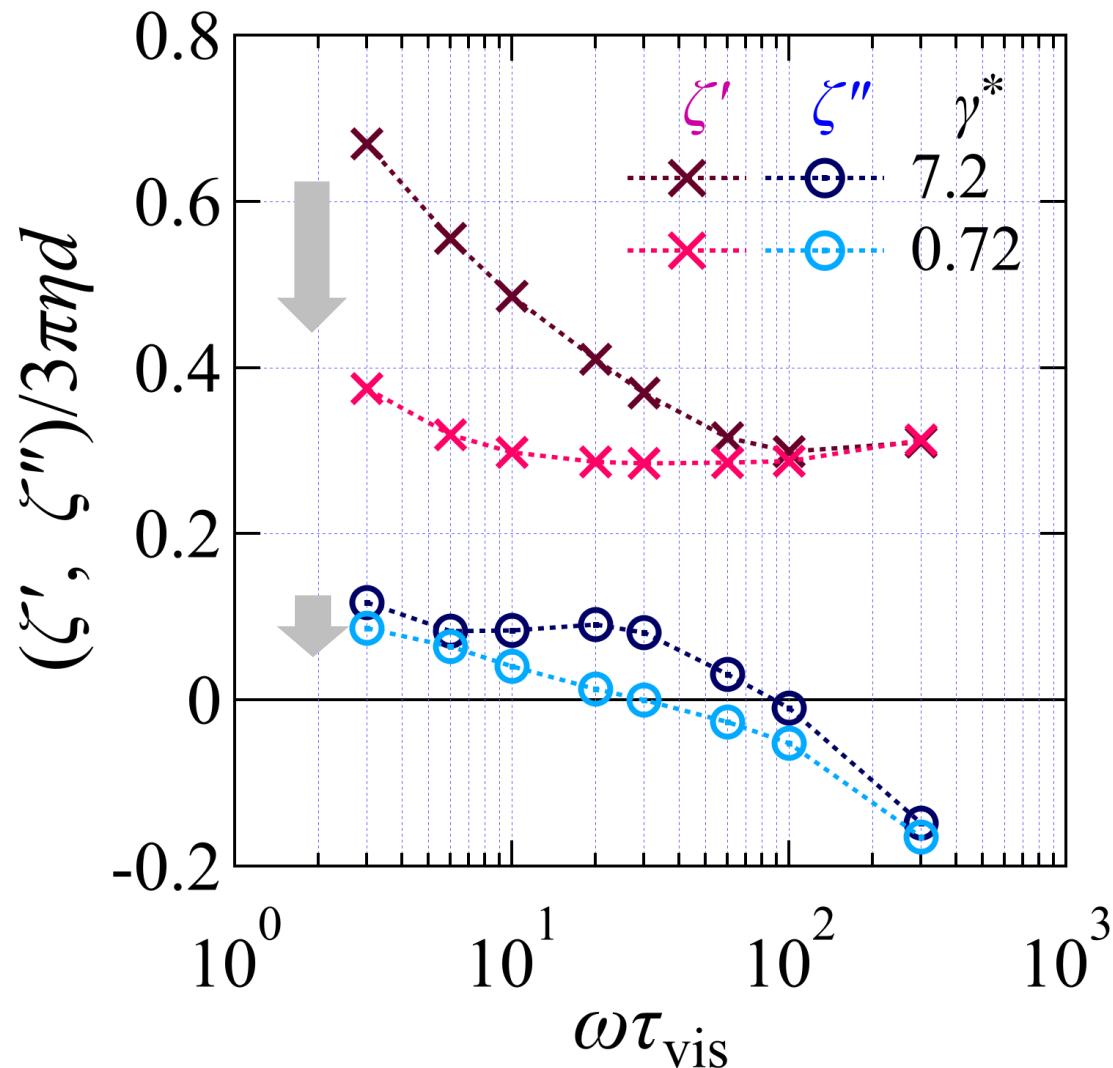
# Drag Coefficients in Bulk Fluid

Oscillatory motion of a sphere    G. G. Stokes (1851).

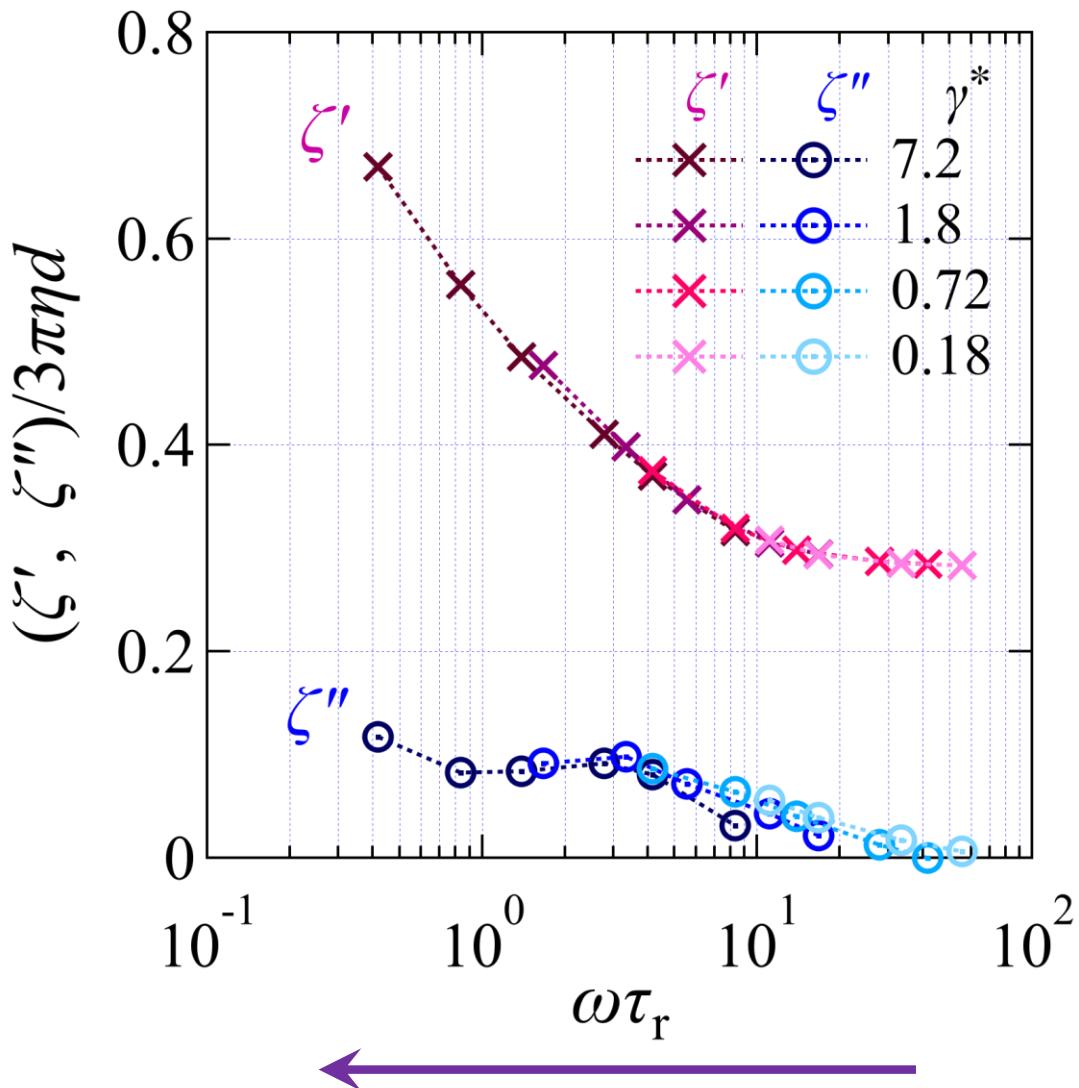


Negative elastic part  $\leftarrow$  Unsteady flow effects

# Effects of Surface Tension



# Scaling by Relaxation Time



$$\tau_r = \frac{\eta d}{\gamma} \text{ Relaxation time}$$

$$\frac{\eta d \delta_0}{\tau_r} \sim \gamma \delta_0$$

Viscous force  $\sim$  Elastic force

$$\omega\tau_r = \frac{\eta d \omega}{\gamma} \propto \frac{\eta U_0}{\gamma}$$

Capillary number

Larger drag coefficients  $\leftarrow$  Larger surface tension, Lower frequency

# Summary

- ◆ Modeling of sticking in mechanics
  - Constraint on relative motions between contacting particles
    - Fixation of contact points
  - Constraint imposed by viscoelastic force
- ◆ Examination of sticking by capillary bridges
  - Calculation of dynamic drag coefficients in shear motion
    - Viscoelasticity of capillary bridges
  - Larger elastic/viscous drag coefficients
    - ← Larger surface tension, Lower frequency