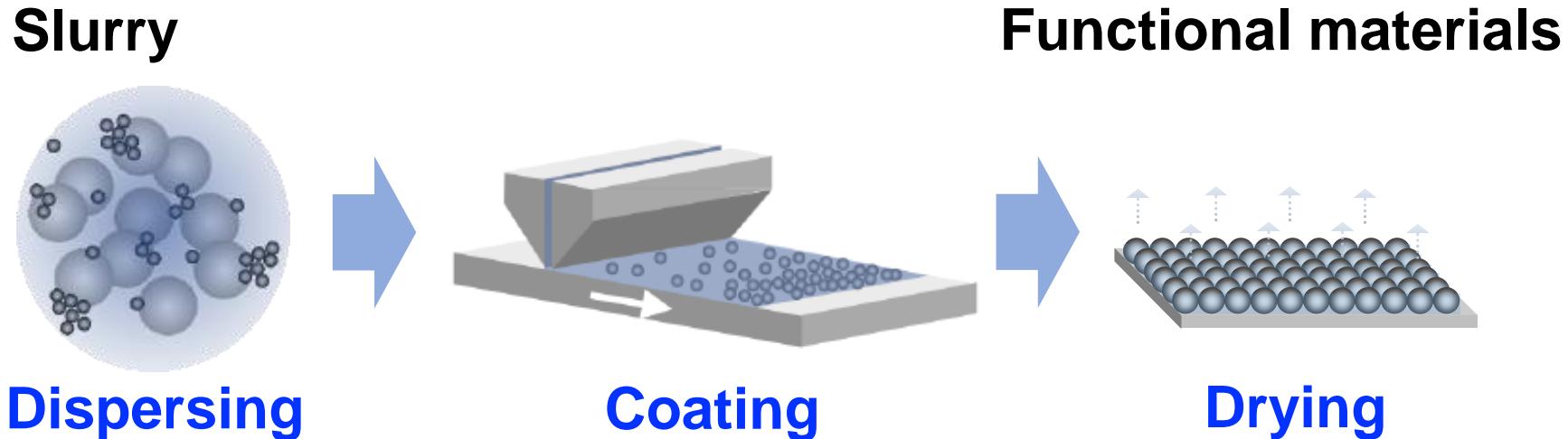


# Mechanism of the viscoelastic behavior of slurry

## スラリーの粘弹性挙動のメカニズム

- 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)

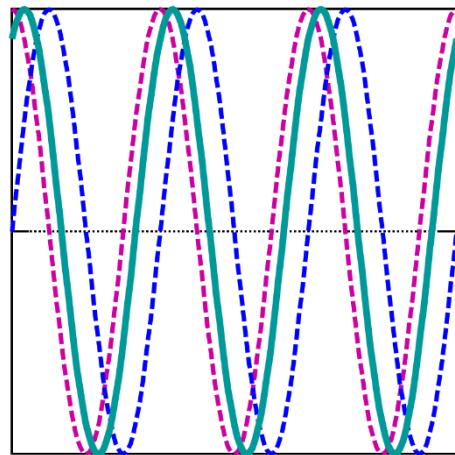
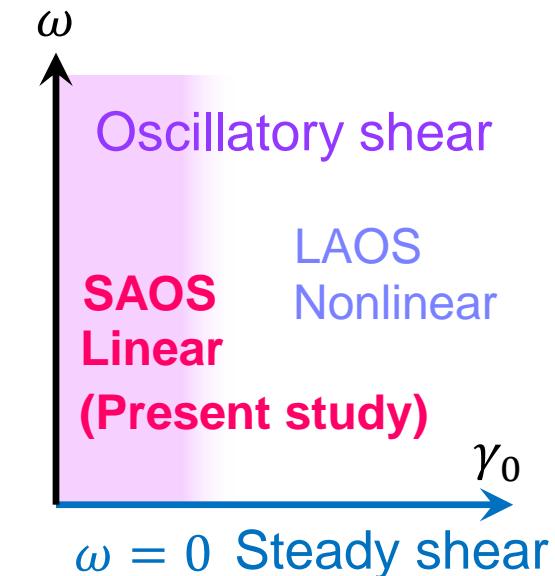
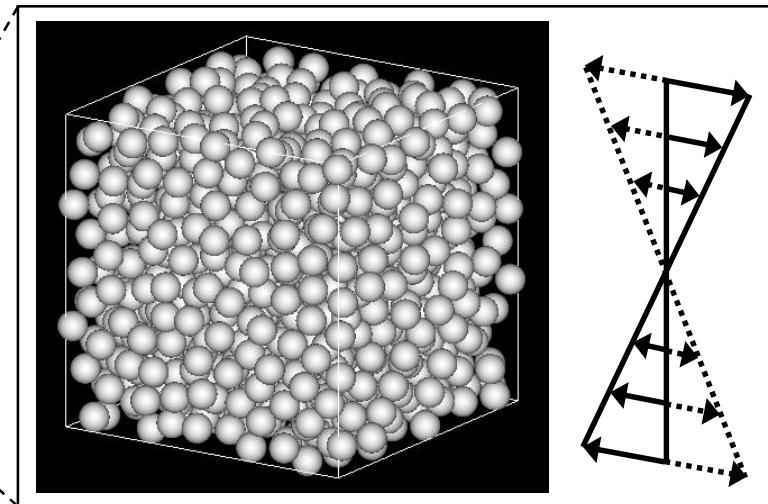
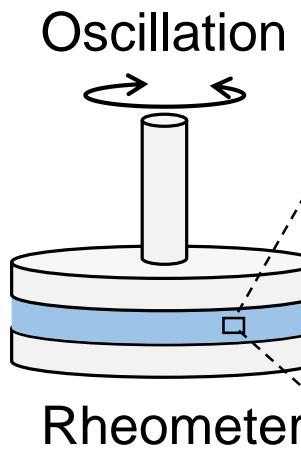
# Material Fabrication from Slurry



**Viscoelasticity can be an index of process**

- Structure of particles (aggregation/dispersion)
- Coating properties
- Influence of dispersing/coating on post-process

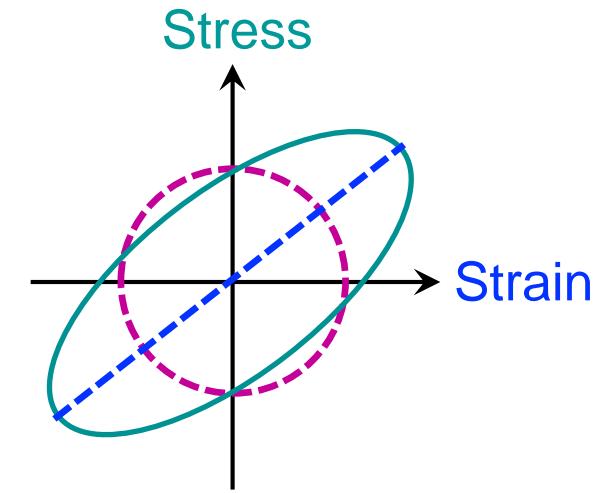
# Viscoelasticity



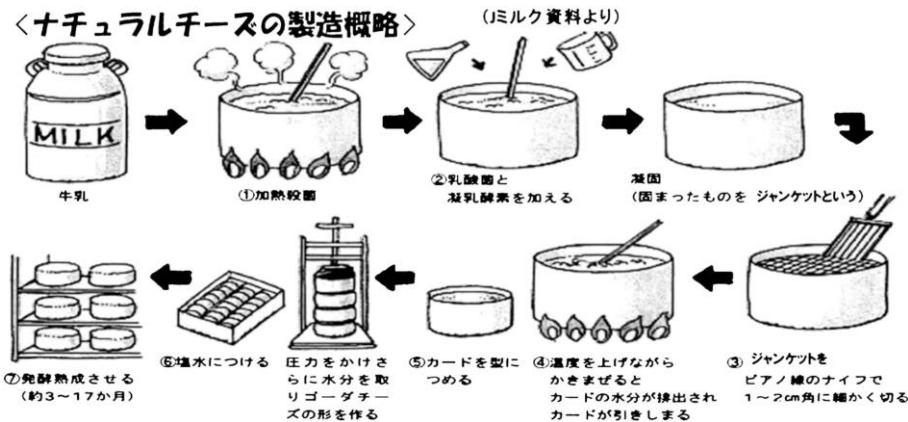
Strain:  $\gamma_0 \sin \omega t$   
Shear rate:  $\gamma_0 \omega \cos \omega t$

Stress:  $\sigma_0 \sin(\omega t + \delta)$   
 $= \gamma_0(G' \sin \omega t + G'' \cos \omega t)$

Storage modulus:  $G'$   
Loss modulus:  $G''$



# Example 1: foods (cheese)



(a) LT-milk with *H. erinaceum*

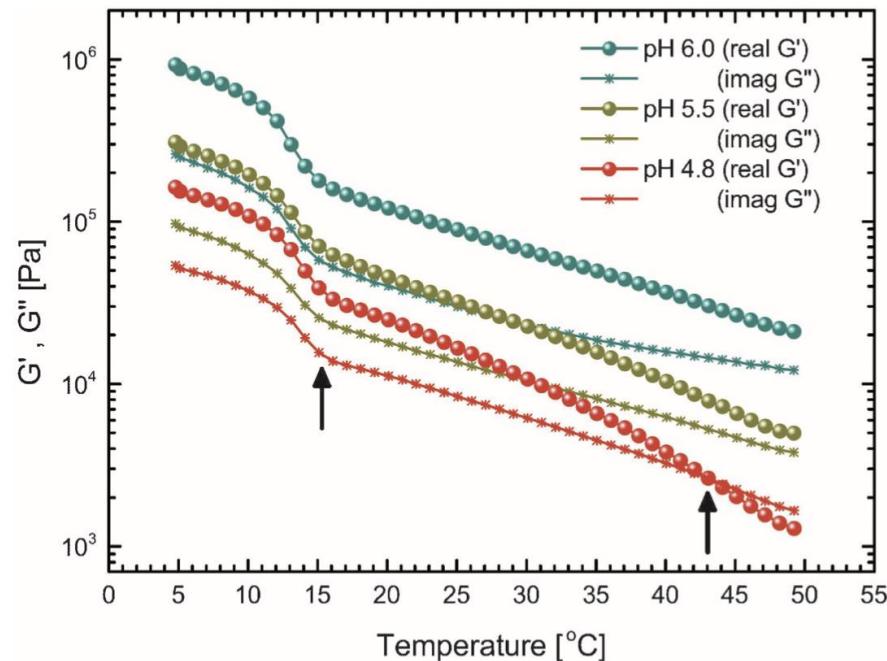
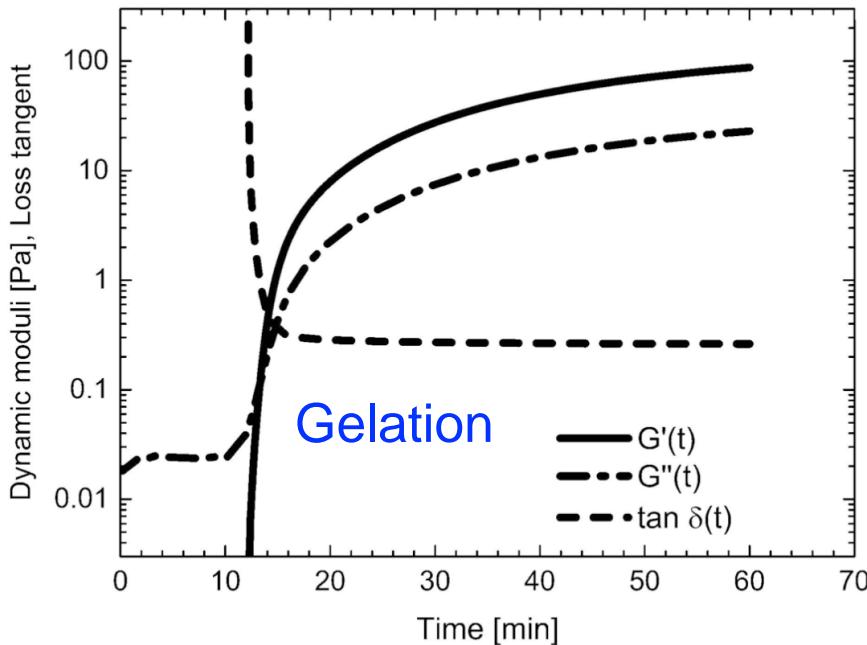
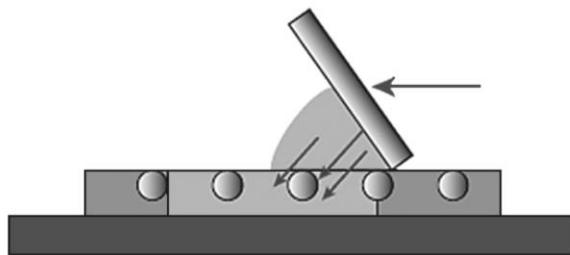


Fig. 8 Temperature dependences of the visco-elasticities of curds under different pH conditions. Each upward arrow indicates the sol-gel transition point (right) and the shoulder caused by lipid-globule melting (left).

# Example 2: screen printing



B スキージによる  
新しいインクの  
供給

Fig. 2 スクリーン印刷でのインク供給のメカニズム

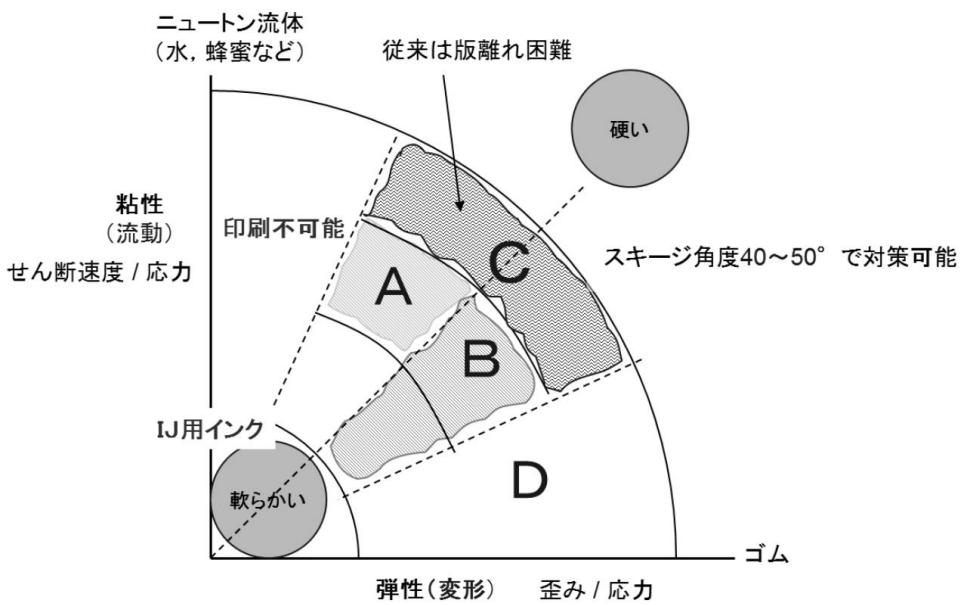


Fig. 6 「粘弾性マップ」でのスクリーン印刷用ペーストの印刷性

佐野康, 日本写真学会誌 77, 65 (2014).

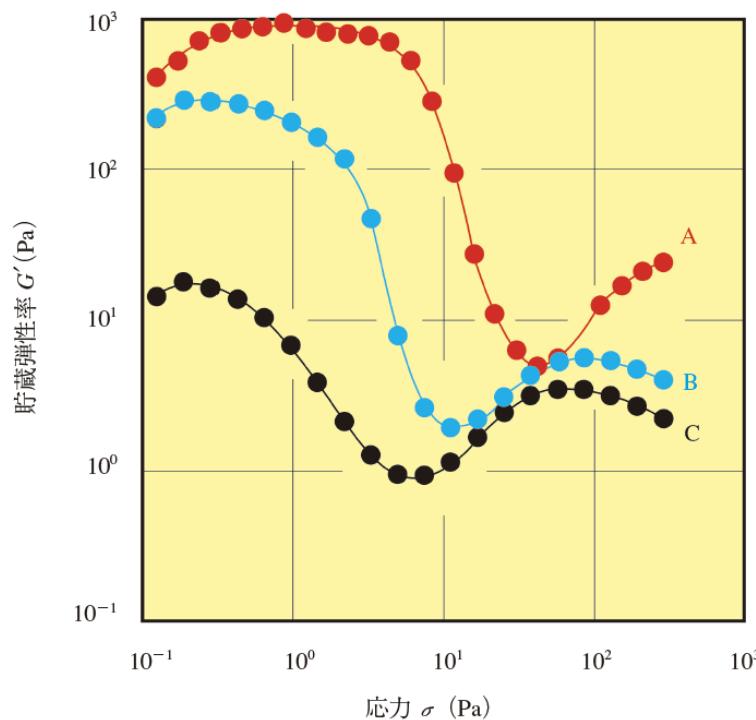


図 5 異なるバインダー樹脂で調製した蛍光体インキにおける貯蔵弾性率の応力依存性

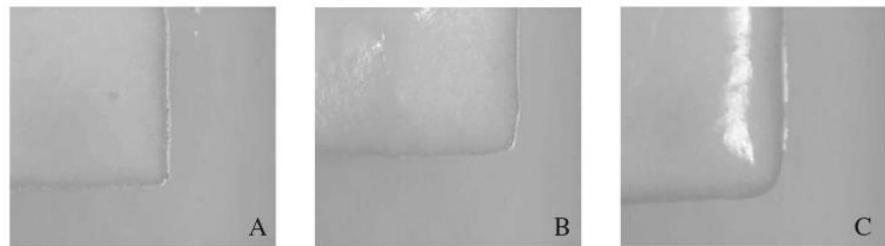
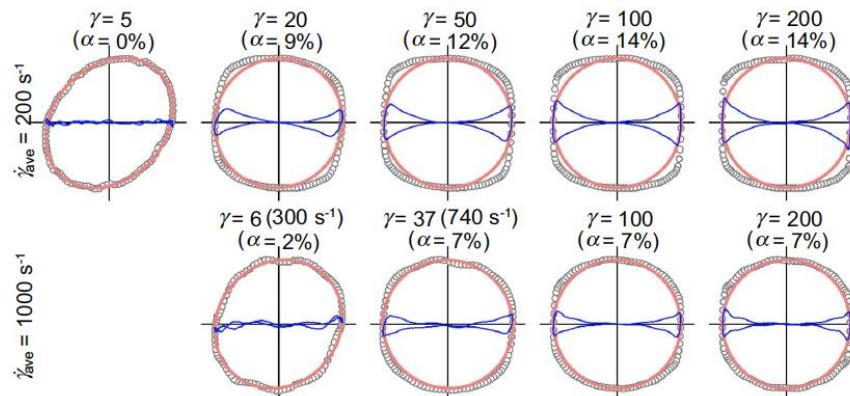
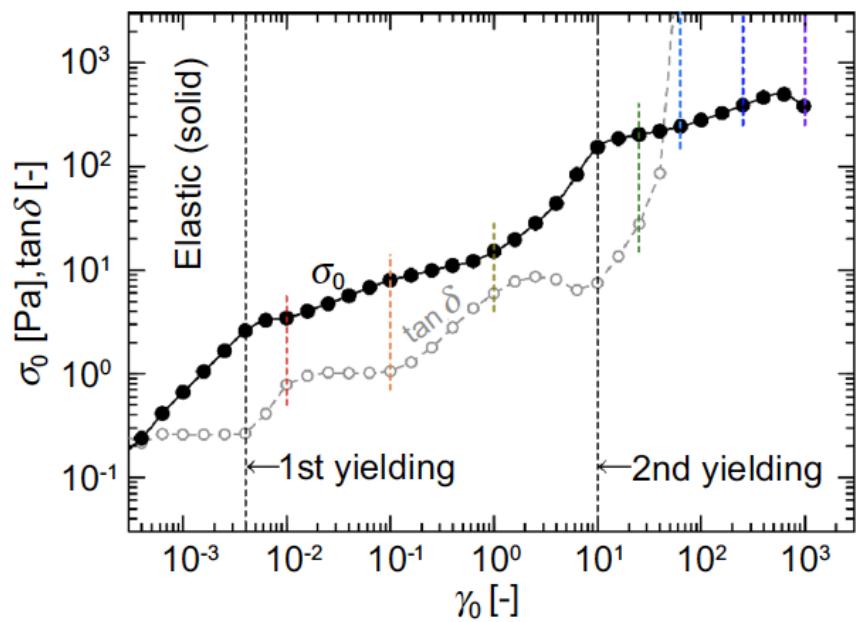
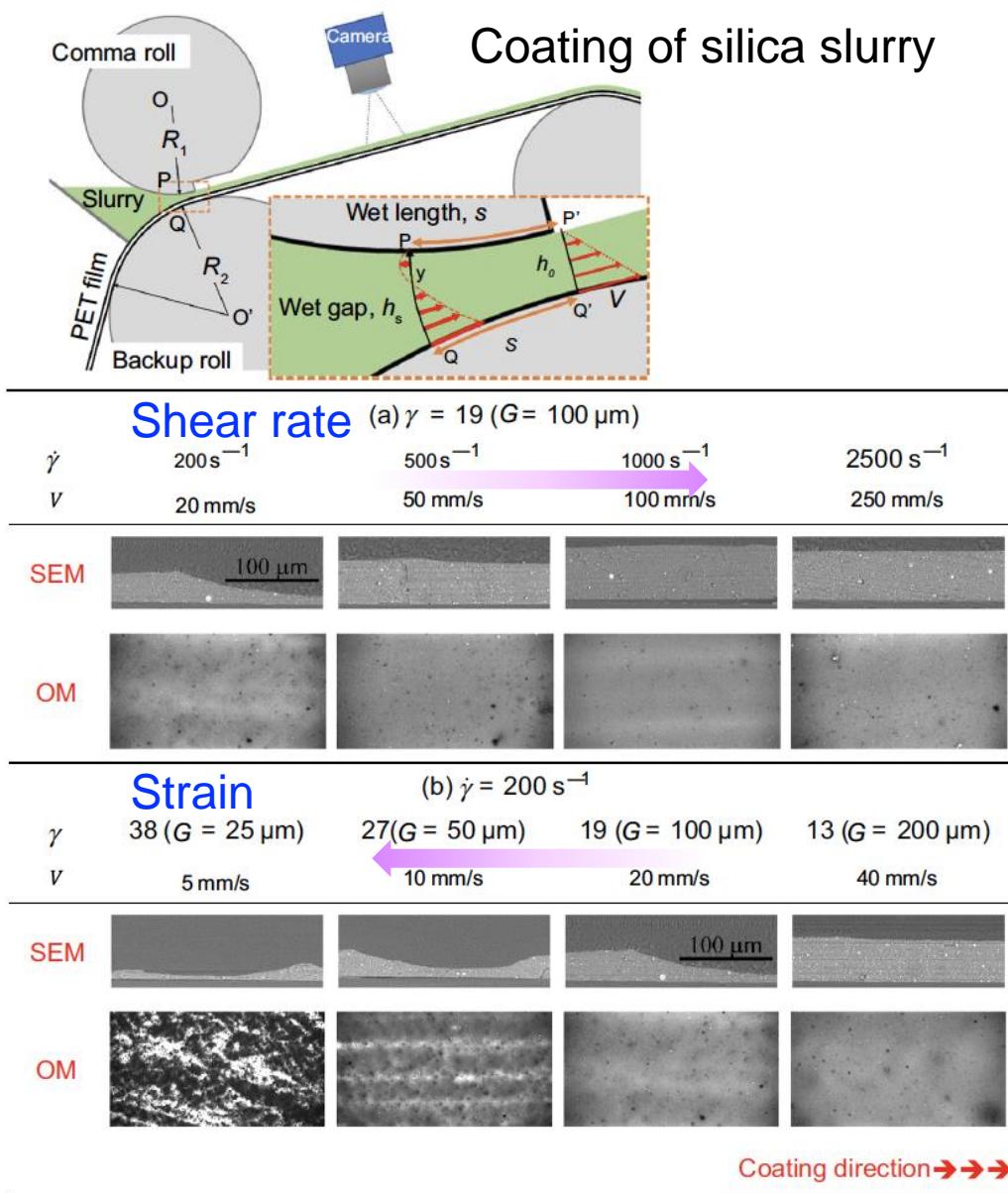


図 6 印刷パターンの顕微鏡写真

大坪泰文, 成形加工 22, 392 (2010).

# Example 3: uniformity of coated layer



# Objective

- ◆ Constructing a simple particle-scale model that describes the viscoelastic behavior of slurry
- ◆ Visualizing the motion of particles that is related to the viscoelastic behavior
- ◆ Investigating the effects of interparticle interactions on the viscoelastic behavior

# Equation of motion of particles

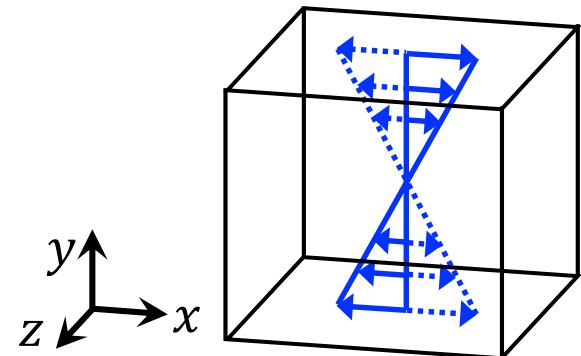
$$M\dot{V} = -\zeta(V - V_{\text{ex}}) + F^{\text{cnt}} + F^{\text{DLVO}}$$

Fluid                          Interparticle

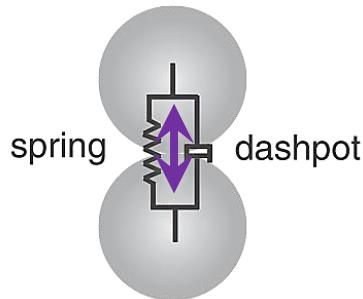
- **Hydrodynamic drag:**  $-\zeta(V - V_{\text{ex}})$

Oscillatory shear flow

$$V_{\text{ex}} = \dot{\gamma}(t)y \mathbf{e}_x \quad \dot{\gamma}(t) = \gamma_0 \omega \cos \omega t$$



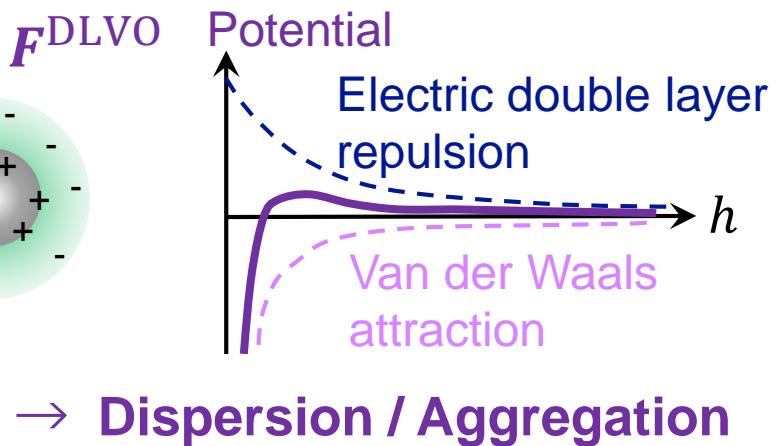
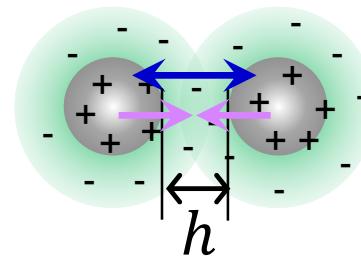
- **Contact force:**  $F^{\text{cnt}}$



- **Boundary conditions**

$x, z$ : Periodic,  $y$ : Lees-Edwards

- **DLVO force:**  $F^{\text{DLVO}}$



# Dynamic modulus

Input

$$\text{Shear strain } \gamma(t) = \gamma_0 \sin \omega t$$

$$\text{Shear rate } \dot{\gamma}(t) = \gamma_0 \omega \cos \omega t$$

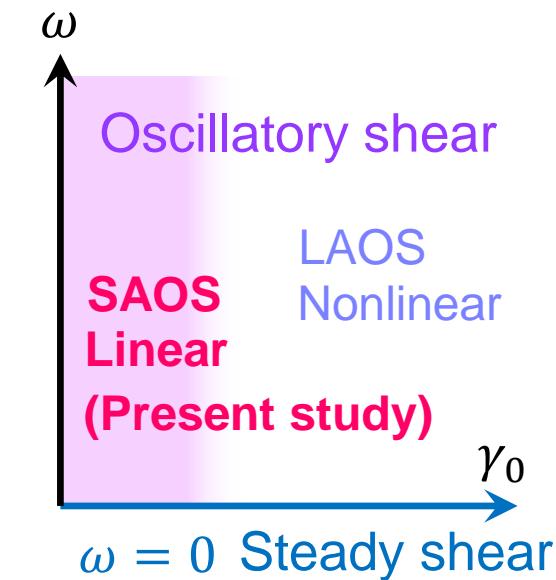
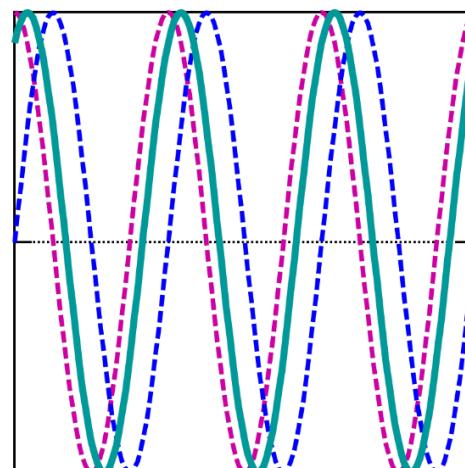
Output

$$\text{Stress } \sigma(t) = \sigma_0 \sin(\omega t + \delta)$$

$$= \gamma_0 (G' \sin \omega t + G'' \cos \omega t)$$

$$G' = \frac{\sigma_0}{\gamma_0} \cos \delta \quad G'' = \frac{\sigma_0}{\gamma_0} \sin \delta$$

----- Shear strain  
 ----- Shear rate  
 — Stress



Time

## Dynamic modulus

- Storage modulus (Elastic)

$$G'(\omega) = \frac{\omega}{\pi \gamma_0} \int_0^{2\pi/\omega} \sigma(t) \sin \omega t \, dt$$

- Loss modulus (Viscous)

$$G''(\omega) = \frac{\omega}{\pi \gamma_0} \int_0^{2\pi/\omega} \sigma(t) \cos \omega t \, dt$$

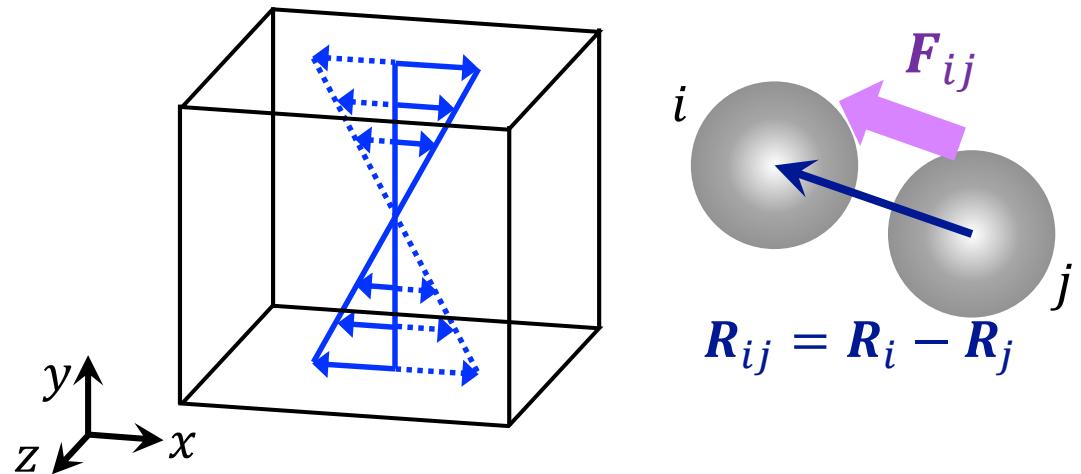
# Stress

$$\sigma = \sigma_f + \sigma_p = \eta \left( 1 + \frac{5}{2} \phi \right) \dot{\gamma} - \frac{1}{V} \sum_{i < j} F_{ij}^x R_{ij}^y$$

Fluid                          Interparticle  
(Einstein's viscosity equation)

- Storage modulus

$$G' = G'_p$$



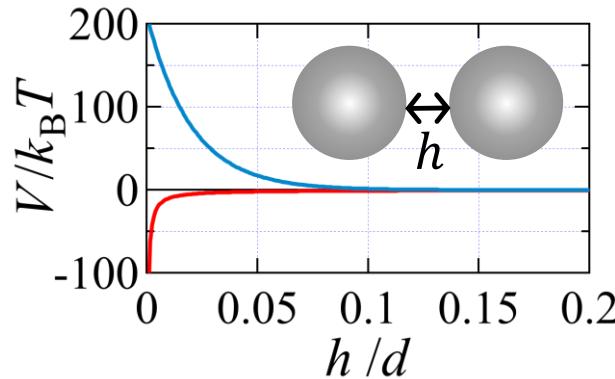
- Loss modulus

$$G'' = G''_f + G''_p = \eta \omega \left( 1 + \frac{5}{2} \phi \right) + G''_p$$

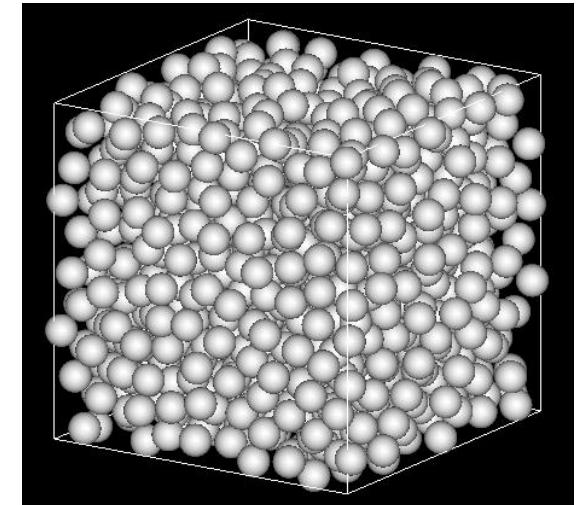
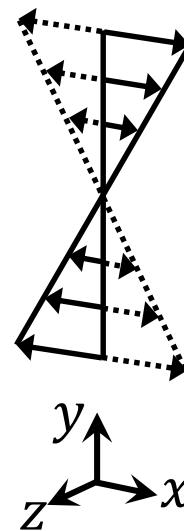
# Simulation Conditions

## Particles

- Diameter:  $d = 1 \mu\text{m}$
- Concentration: 45 vol%
- Zeta potential: 0 mV, 20 mV



$$U_x(t) = \dot{\gamma}(t)y$$



Side length:  $22.7d$

## Fluid: Water

- Ionic strength: 0.23 mM

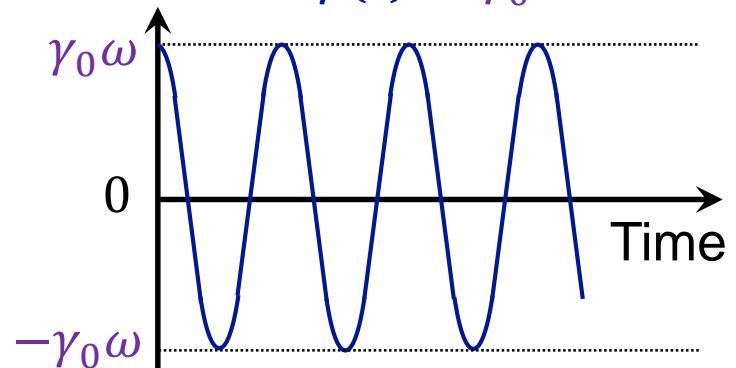
## Shear flow

- Strain:  $\gamma_0 = 1 \times 10^{-3} - 3$
- Frequency:  $\omega\tau = 3.6 \times (10^{-3} - 10^2)$

$$\tau = 3\pi\eta d^2/F$$

$F$  : Force between contacting particles

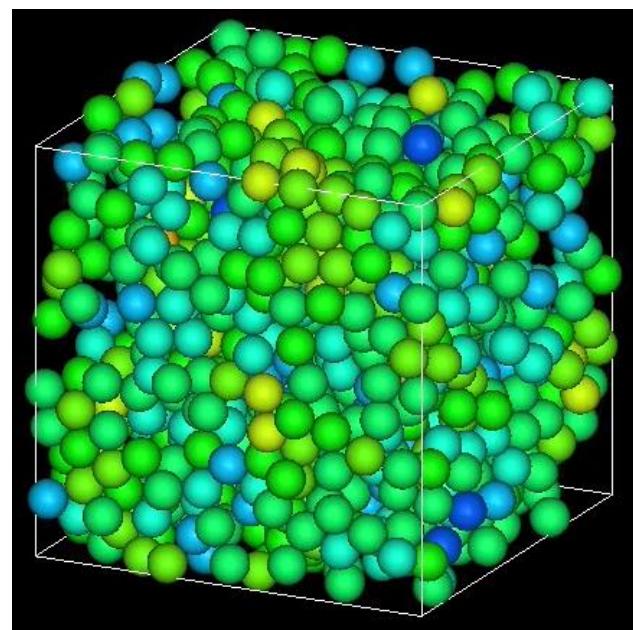
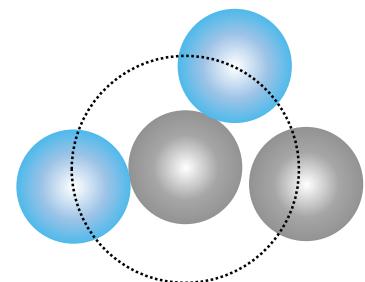
Shear rate  $\dot{\gamma}(t) = \gamma_0\omega \cos \omega t$



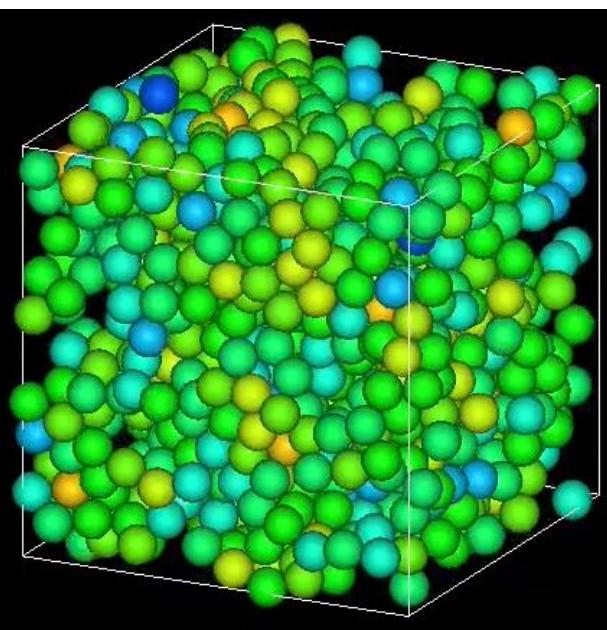
# Structure of particles

Aggregation

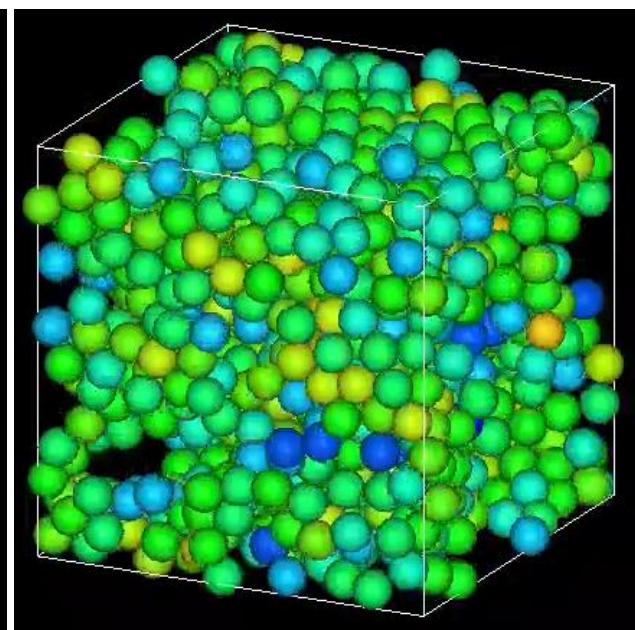
Contact number



$$\gamma_0 = 0.01$$



$$\gamma_0 = 0.1$$



$$\gamma_0 = 1$$

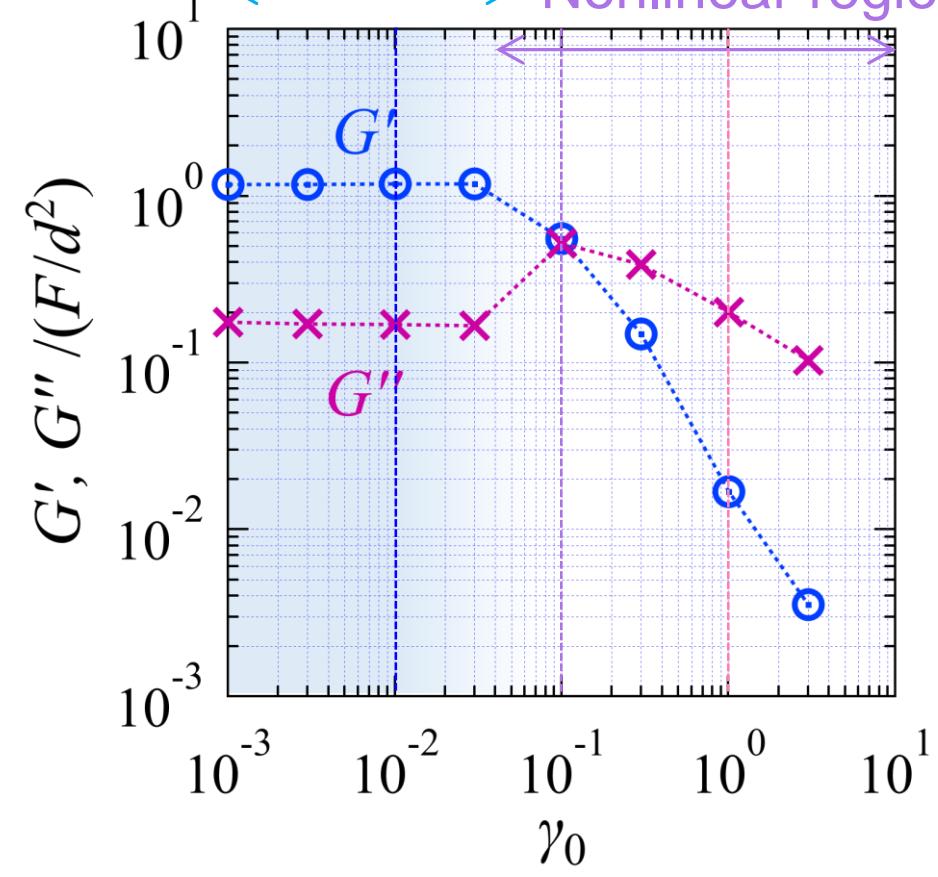
# Strain dependence

Aggregation

Structure change

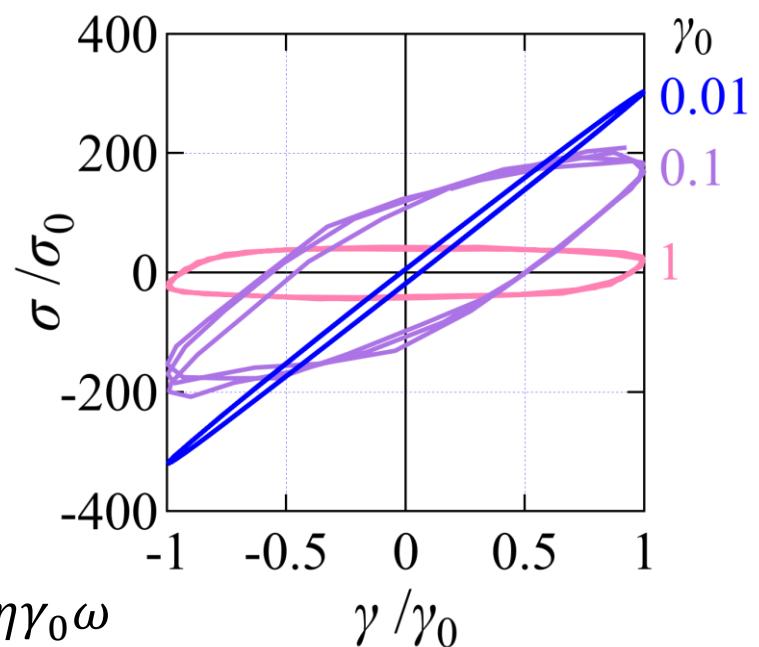
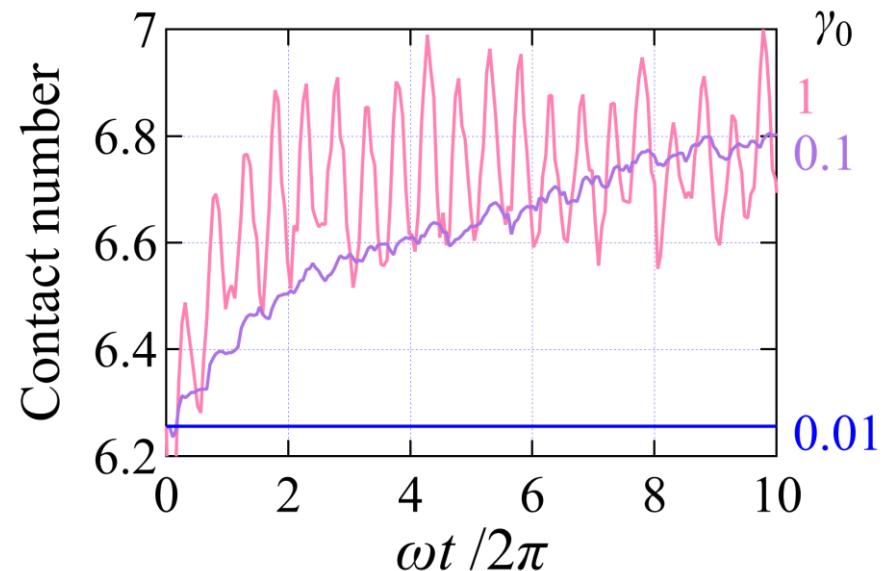
Linear region

Nonlinear region



$$\omega\tau = 3.6 \times 10^{-2}$$

$$\sigma_0 = \eta\gamma_0\omega$$



# Structure of particles

Dispersion

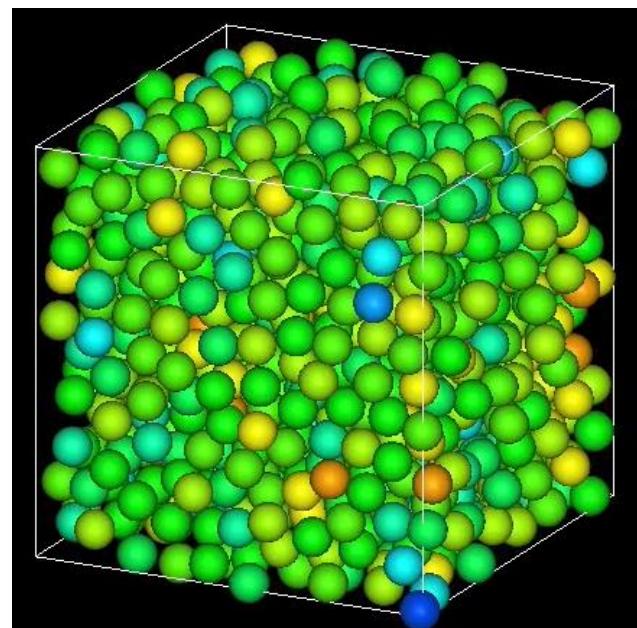
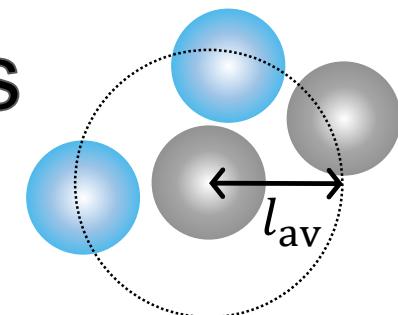
Coordination number

0

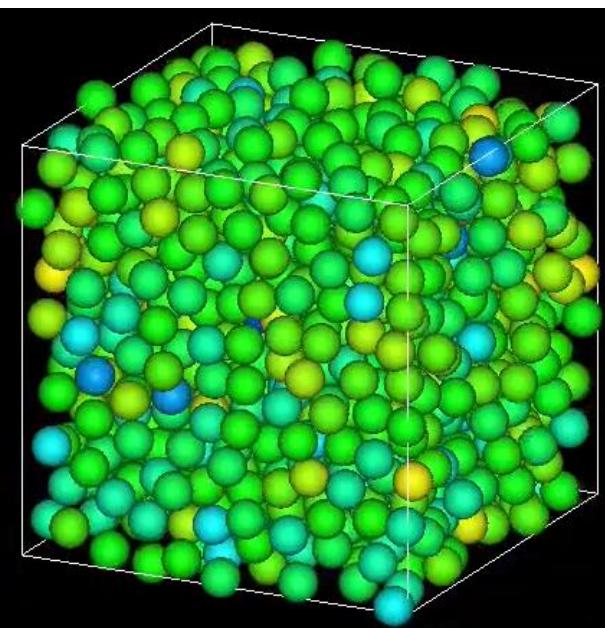


11

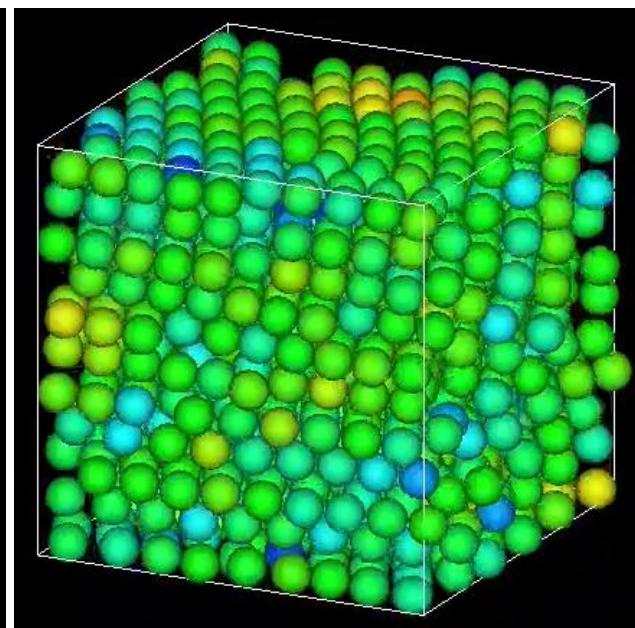
$$l_{av}/d = (\text{Volume fraction})^{-1/3}$$



$$\gamma_0 = 0.01$$



$$\gamma_0 = 0.1$$



$$\gamma_0 = 1$$

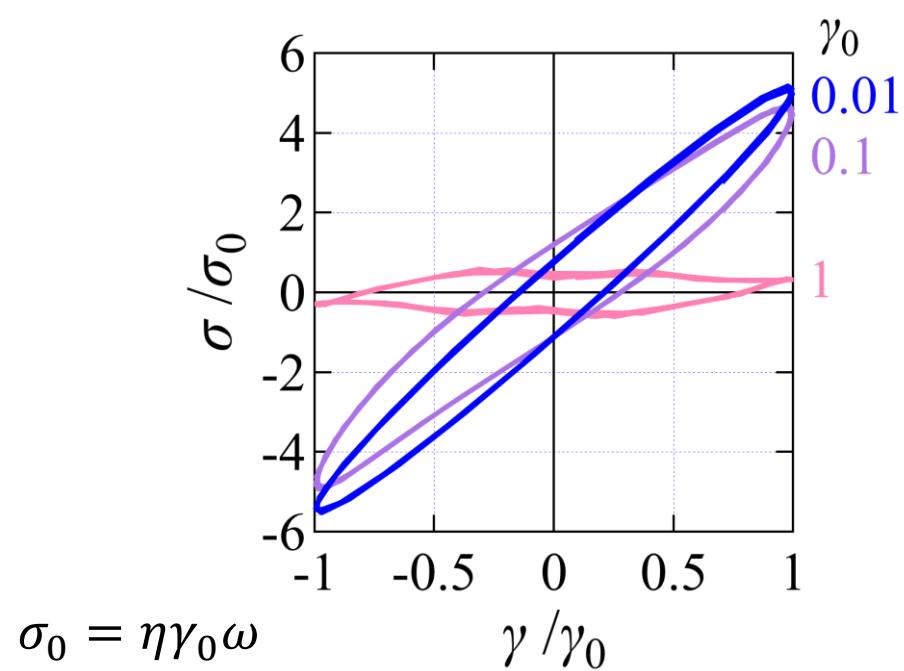
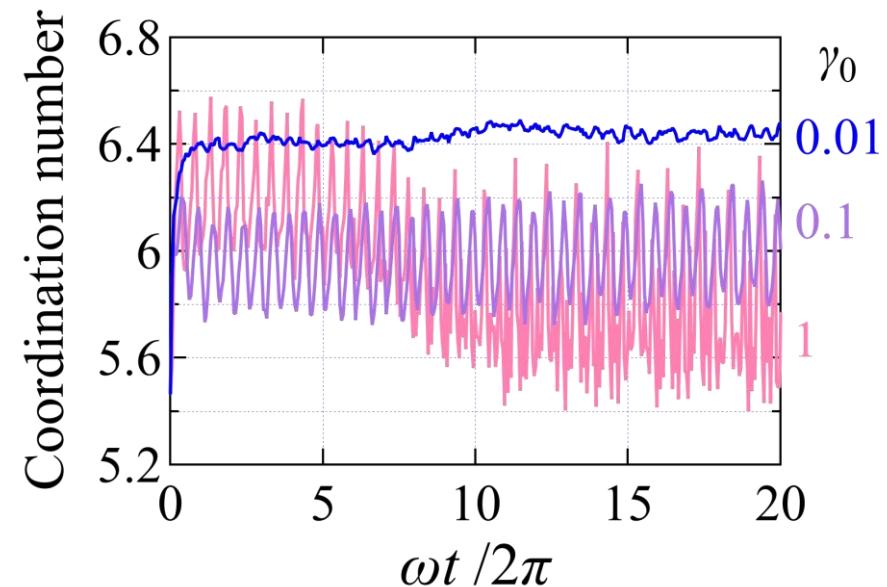
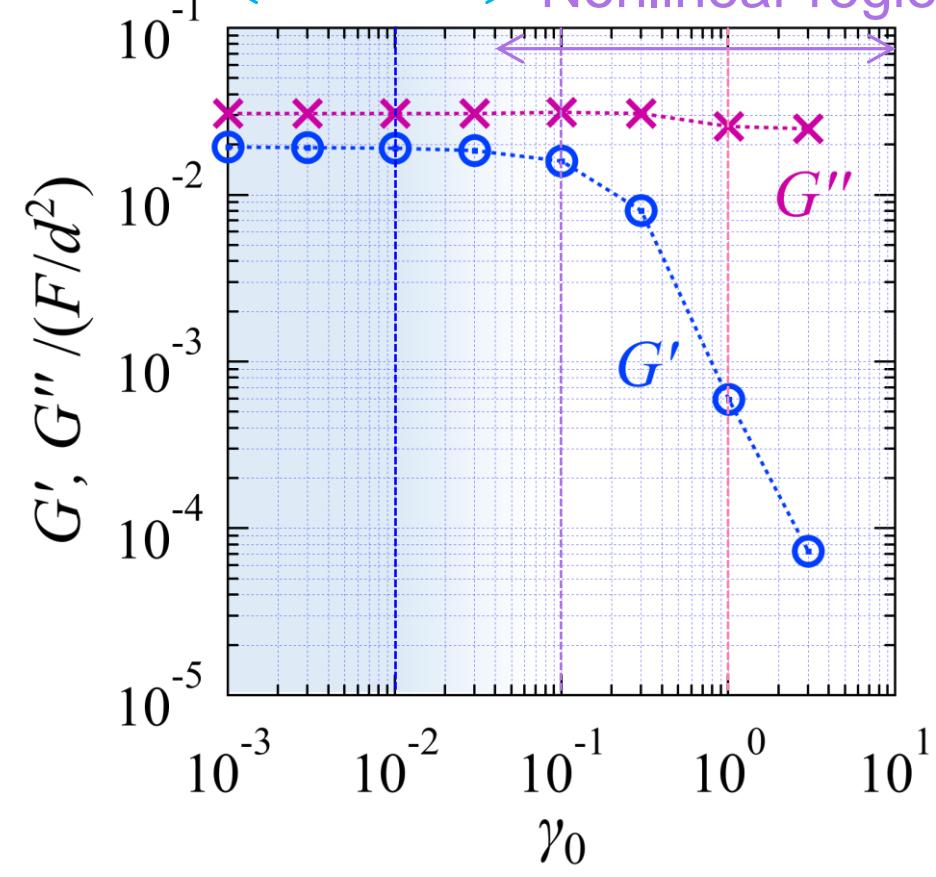
# Strain dependence

Dispersion

Structure change

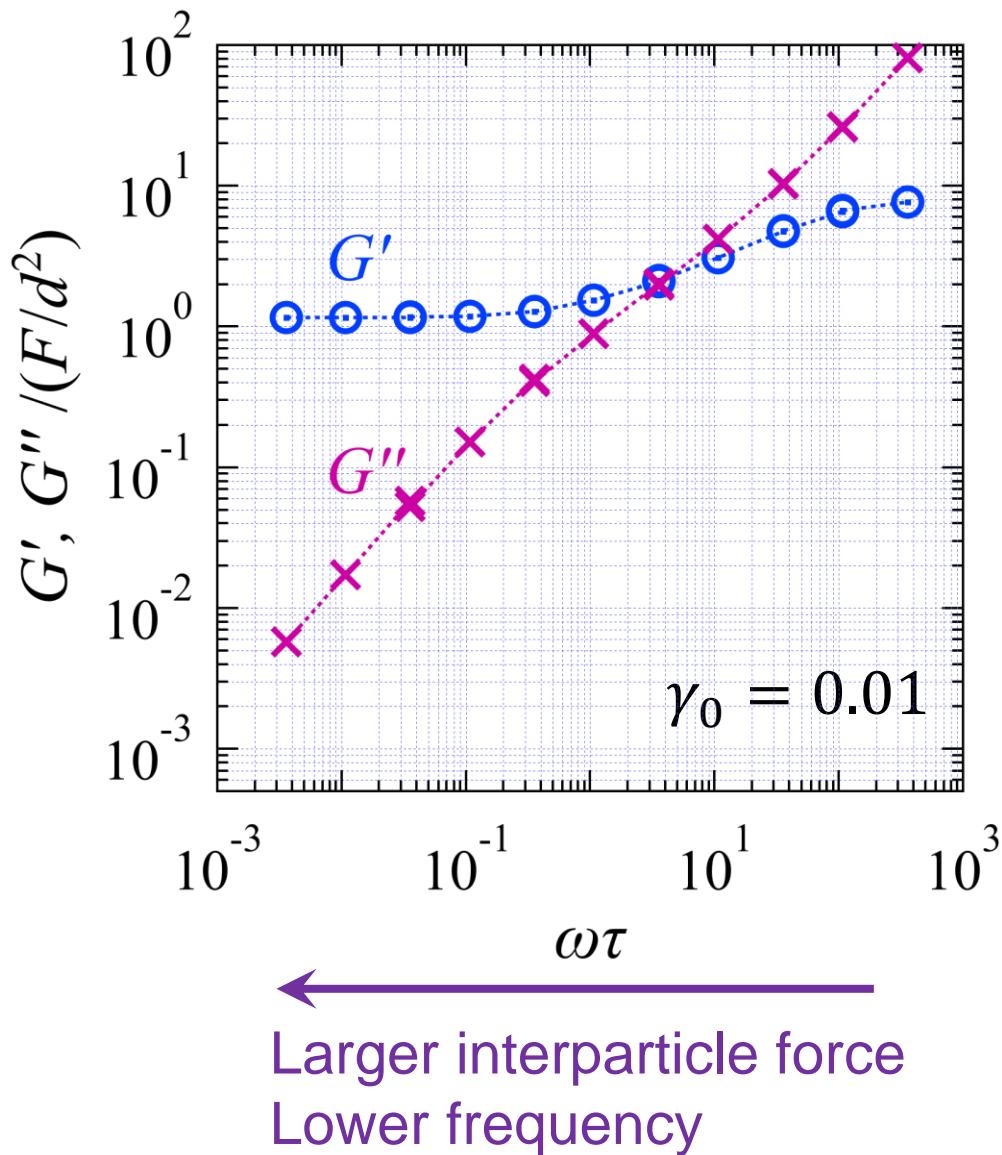
Linear region

Nonlinear region



# Frequency dependence

## Aggregation



Interparticle force = Drag force

$$F = 3\pi\eta dU$$

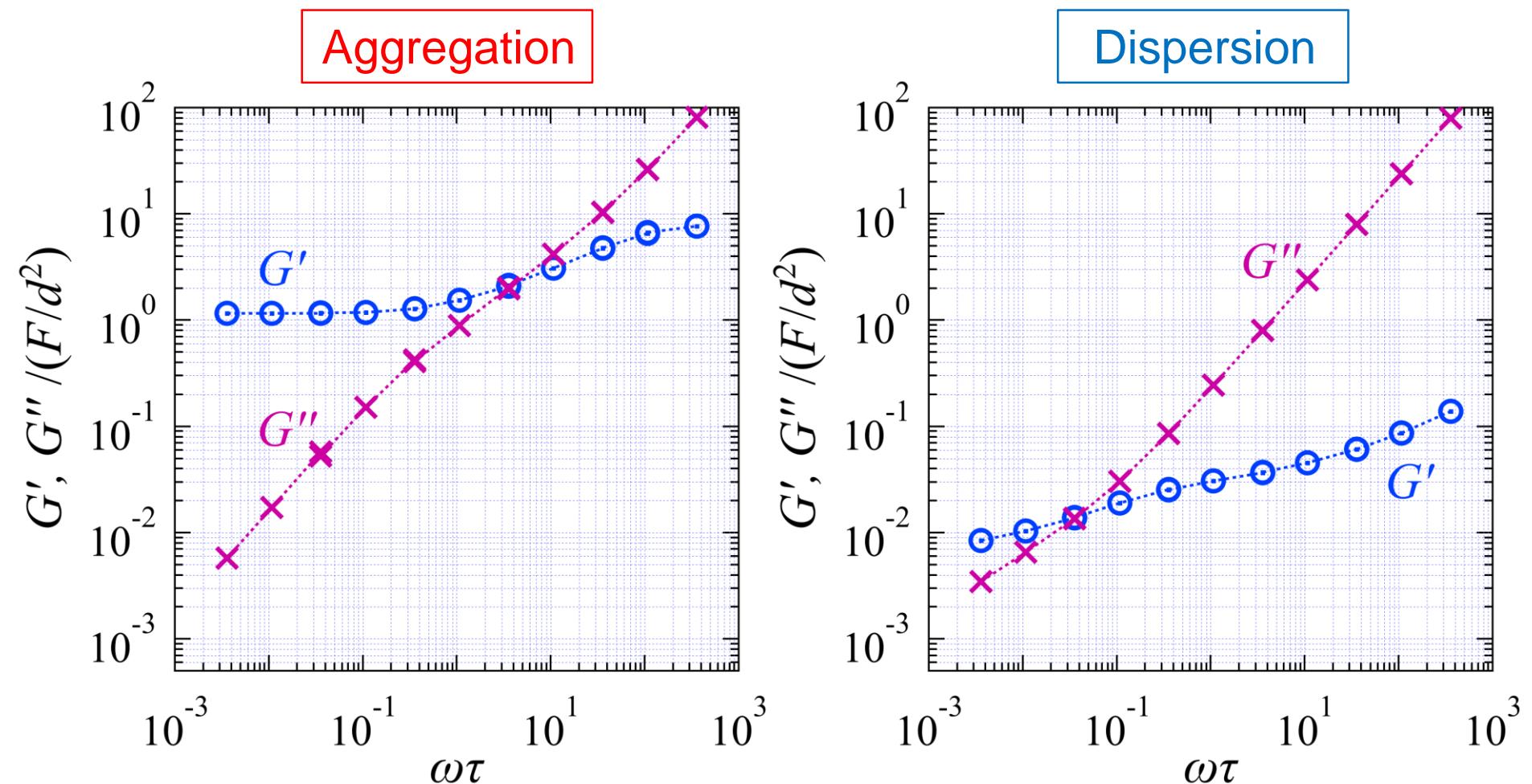
Velocity of adhesion

$$U = \frac{F}{3\pi\eta d}$$

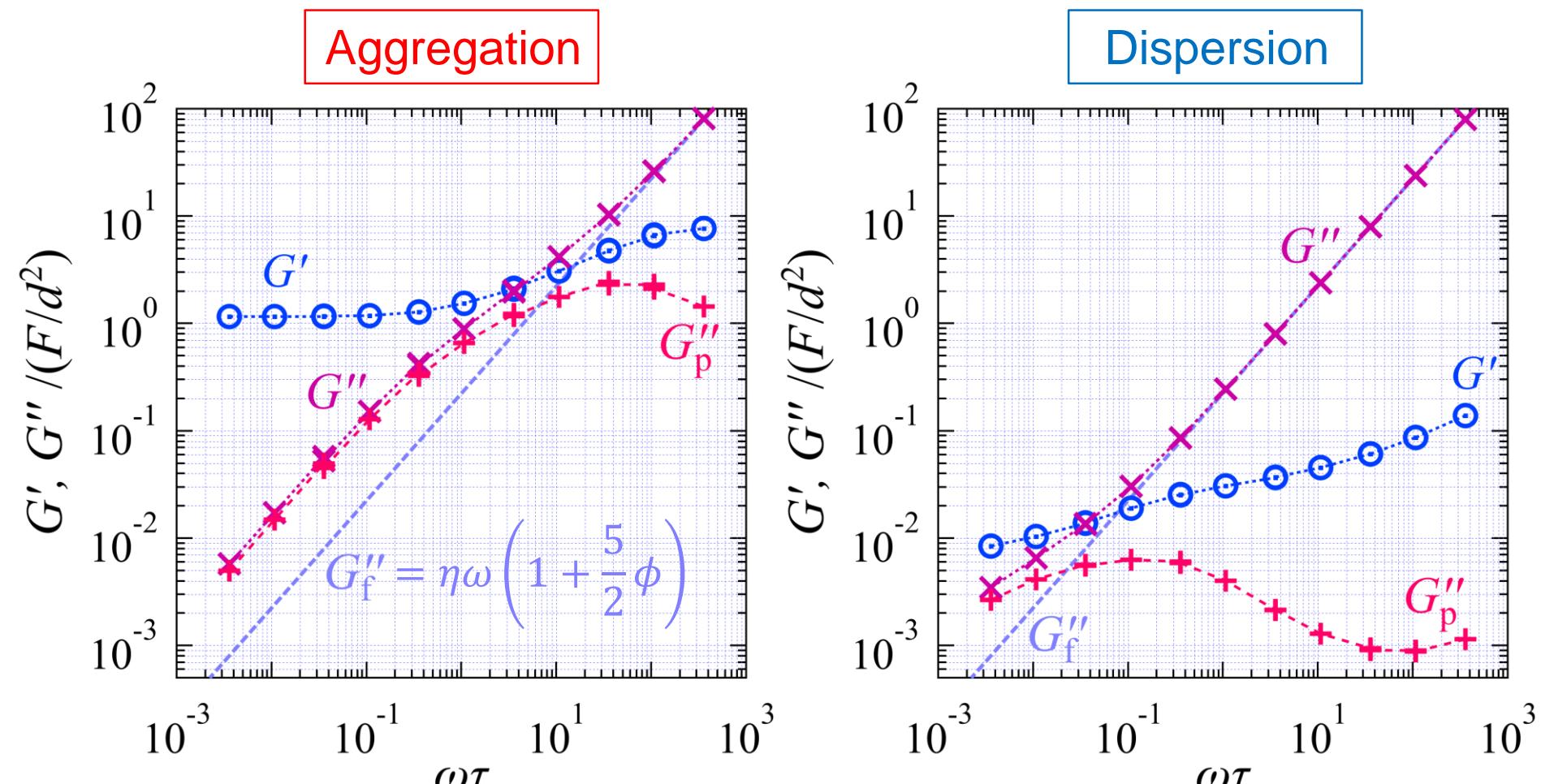
Relaxation time

$$\tau = \frac{d}{U} = \frac{3\pi\eta d^2}{F}$$

# Frequency dependence

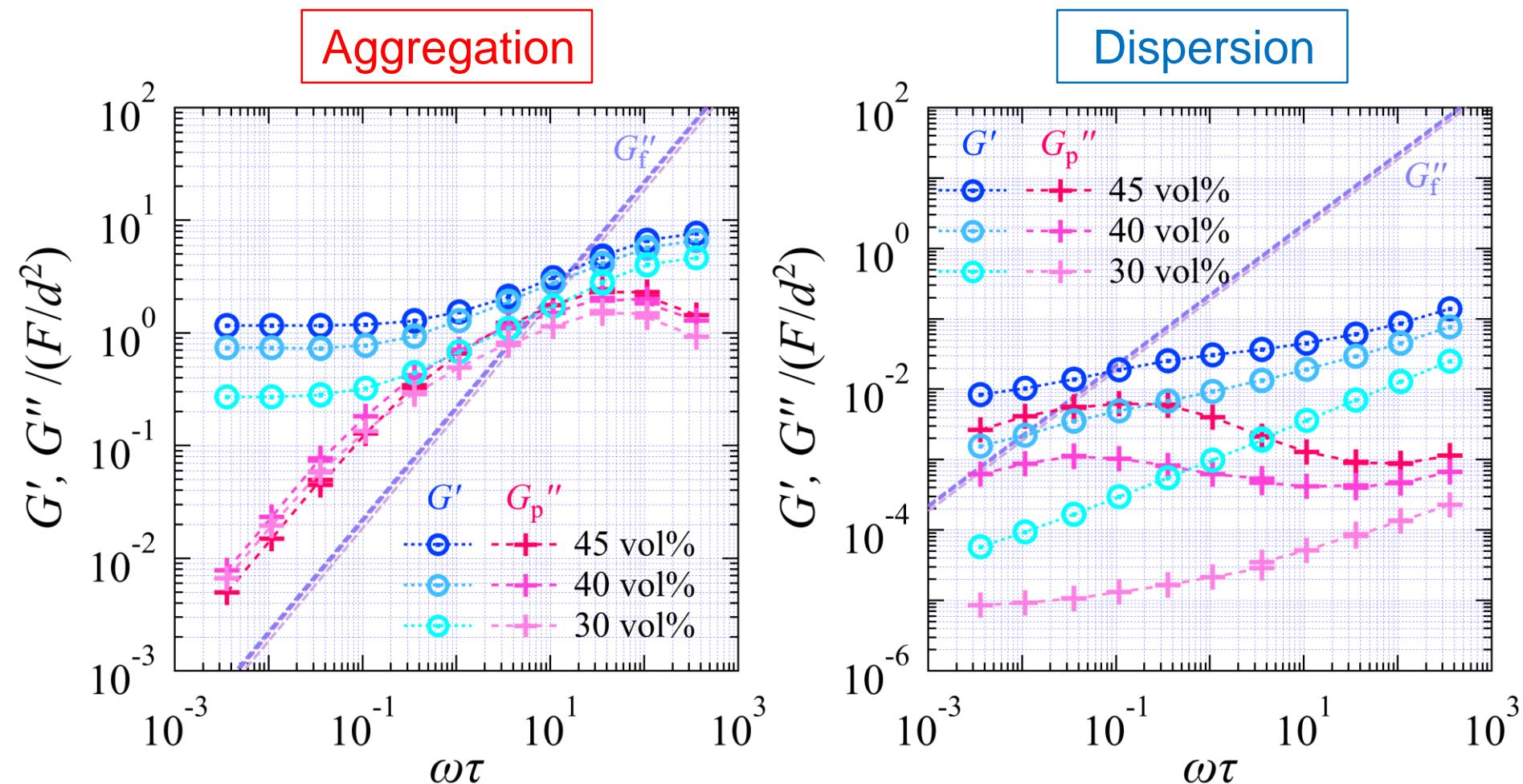


# Frequency dependence



Decomposition of loss modulus:  $G'' = G_f'' + G_p''$

# Effects of particle concentration



# Summary

- Viscoelasticity can be described by a simple model that only considers drag force and interparticle force
- Both attractive and repulsive interaction can provide elasticity
- Strain dependence reflects the structure change of particles
- Frequency dependence:
  - Aggregated particles provide a finite value of  $G'$  in low frequency limit
  - $G''$  is dominated by fluid part  $G_f''$  at high frequency
  - Non-monotonic behavior of  $G''$  would appear for concentrated slurry