YUKAWA INSTITUTE FOR THEORETICAL PHYSICS

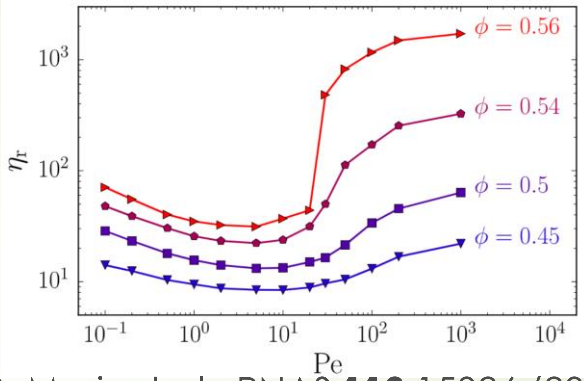
国立大学法人 東京農工大学
機械システム工学専攻・学科
Mechanical Systems Engineering

Discontinuous shear thickening of a moderately dense inertial suspension composed of frictionless soft-core particles

Satoshi Takada (TUAT)
Kazuhiro Hara (TUAT)
Hisao Hayakawa (YITP, Kyoto Univ.)

Shear thickening

- Shear thickening:
  viscosity: $\eta_s(\varphi) = \sigma(\varphi)/\dot{\gamma} \nearrow$
  against the shear rate $\dot{\gamma}$.



R. Mari, et al., PNAS **112**,15326 (2015)

- Discontinuous shear thickening (DST) is fascinating, where the viscosity discontinuously increases.

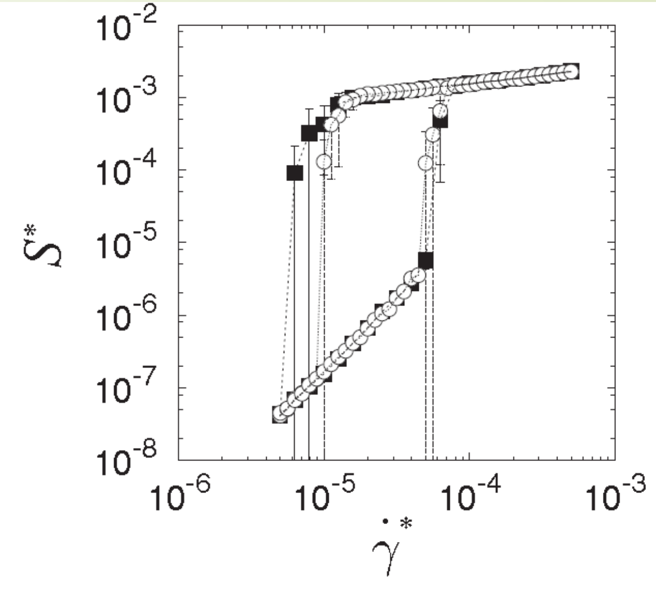
⇒ DST is studied in many contexts and setups.

Shear thickening

DST for dense systems (simulations)

- Mutual friction is important.

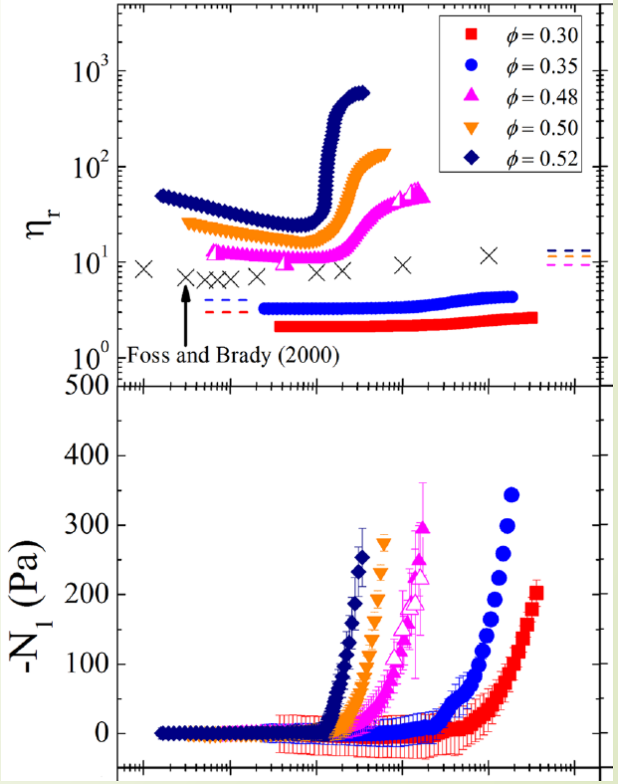
  M.Otsuki & H. Hayakawa, Phys. Rev. E **83**, 051301 (2011)
  R. Seto, et al., Phys. Rev. Lett. **111**, 218301 (2013)



DST for colloidal systems (experiments)

- Normal stress difference

  is also important.
  C. D. Cwalina & N.J. Wagner, J. Rheol. **58**, 949 (2014)

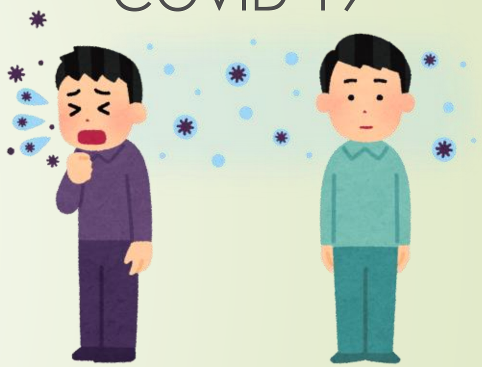


Gas-solid suspensions

- Inertial suspensions
  (a model of aerosols)
- Particle size: $1 - 70\ \mu m$
- Homogeneity is kept.
- Not dense system
  (theoretical treatment is available)
- Ex.) spray, COVID-19, nursing bed,⋯

aerosol

COVID-19

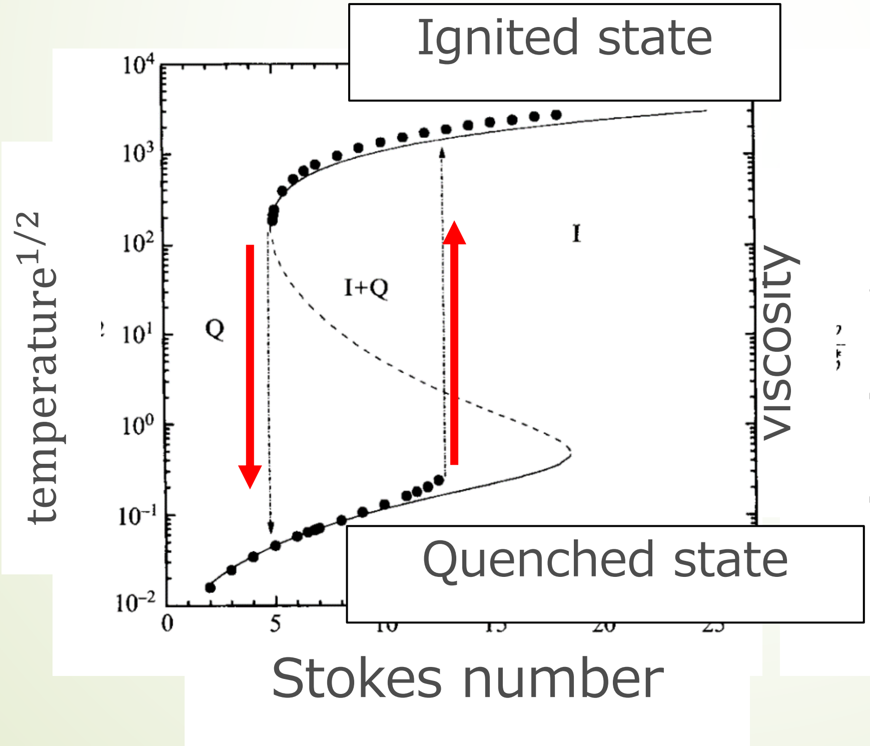


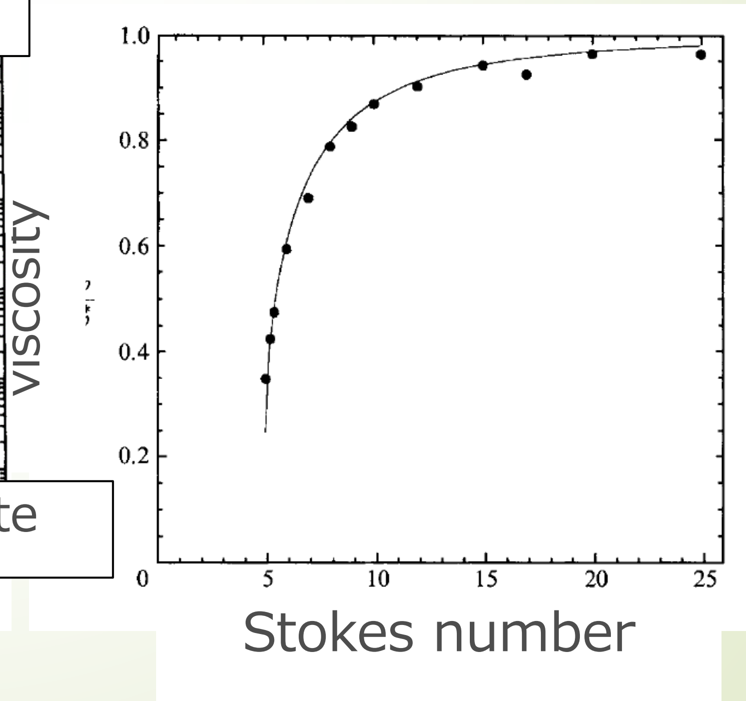
Previous (theoretical) studies for dilute systems

☞ *Tsao and Koch, JFM* ***296****, 211 (1995)*

Kinetic theoretical approach without thermal noise

⇒ DST-like (Quenched-Ingnited) transition for **temperature**, but not for **viscosity**





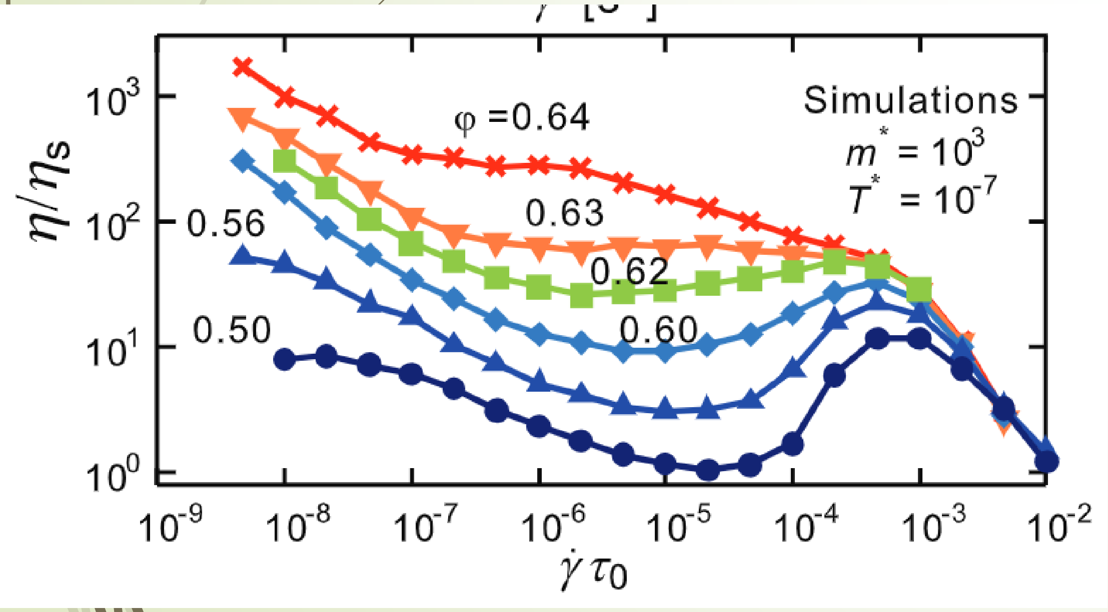
Previous study (soft-core, dense)

- Rheology of inertial suspension composed of soft-core particles

  ☞ Kawasaki, Ikeda, & Berthier, EPL (2014)

  harmonic potential: $U(r) = \frac{\varepsilon}{2}(1 - r/d)^2\Theta(1 - r/d)$

  Langevin equation: $\frac{d\boldsymbol{p}_i}{dt} = \sum_j \boldsymbol{F}_{ij} - \zeta\boldsymbol{p}_i + \boldsymbol{\xi}_i$



Complicated behaviors for moderately dense to dense systems

- Thinning → thickening → thinning for $\varphi \lesssim 0.60$
- No thickening for $\varphi \gtrsim 0.63$

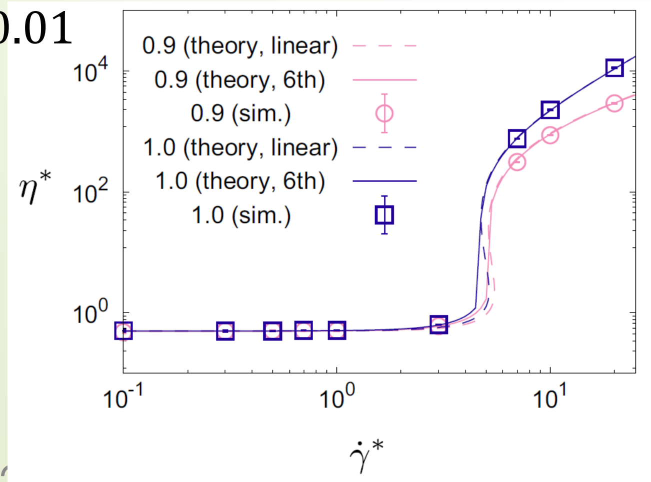
Our previous studies (hard-core)

hard-core potential:

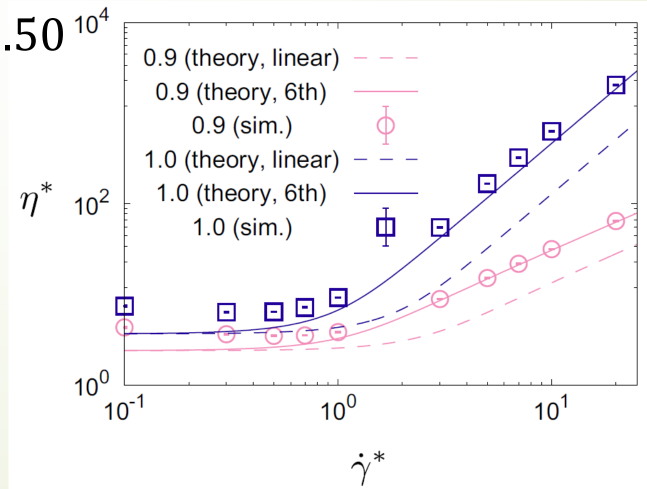
$$U(r) = \begin{cases} \infty \ (r < \sigma) \\ 0 \ \ (r \geq \sigma) \end{cases}$$

- Rheology of inertial suspension of **hard-core** particles
  ☞ Hayakawa & Takada, PTEP (2019),
  Hayakawa, Takada, & Garzó, PRE (2017),
  Takada, Hayakawa, Santos, & Garzó, PRE (2020)
- Collisional contribution is written in terms of the kinetic part,
- Discontinuous shear thickening (DST)-like behavior for dilute systems (≅ ignited-quenched transition of the kinetic temp.)
- DST-like → continuous shear thickening (CST)-like behavior at $\varphi \simeq 0.0176$.
- Kinetic theory reproduces simulation results even for $\varphi \simeq 0.50$.

$\varphi = 0.01$

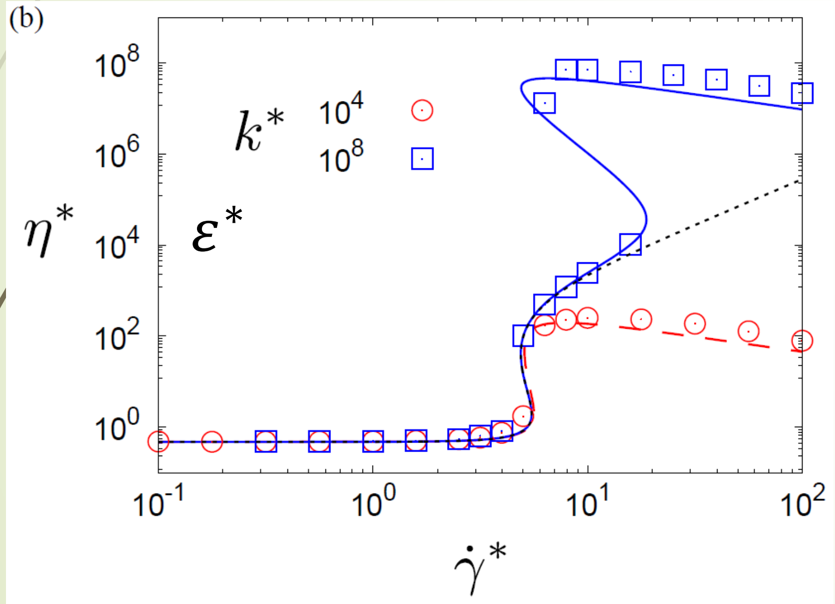


$\varphi = 0.50$



Our previous study (dilute soft-core system)

- Kinetic theory is extended to soft-core system
  ☞ S. Sugimoto and S. Takada, J. Phys. Soc. Jpn. **89**, 084803 (2020)
- System: similar to Kawasaki *et al.*, but dilute limit is considered.
- Kinetic theory is constructed.

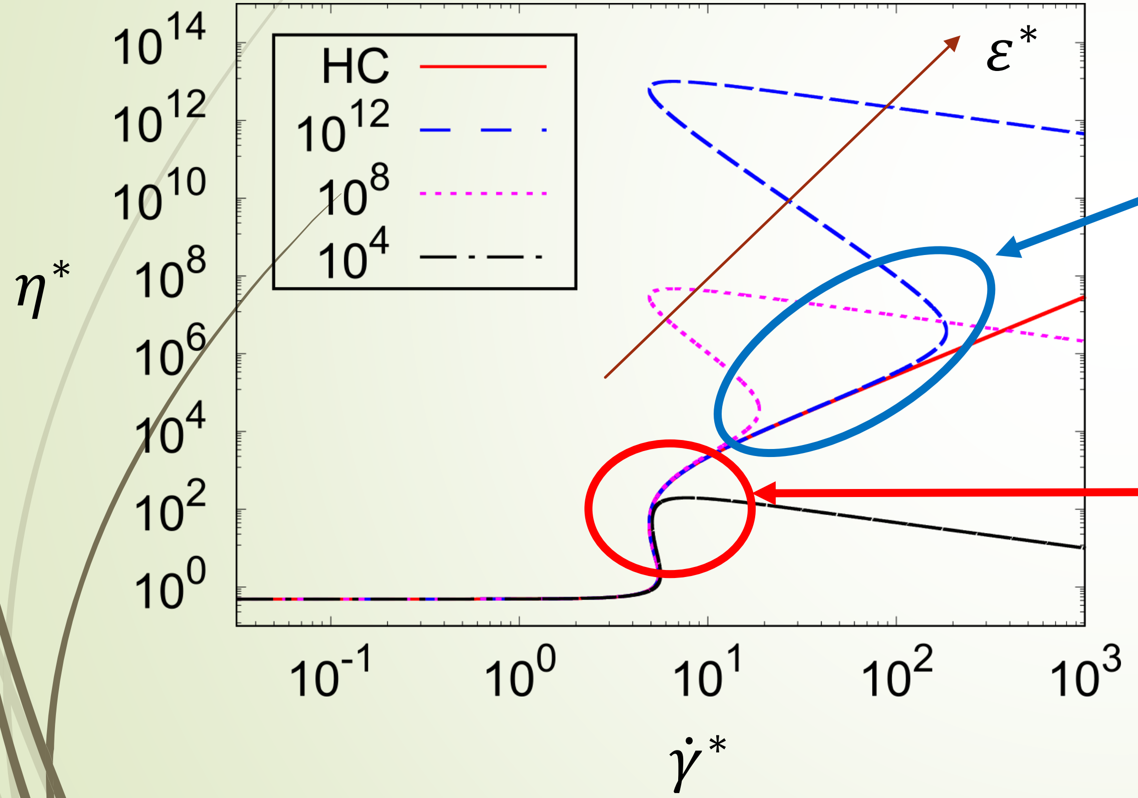


$$U(r) = \frac{\varepsilon}{2}(1 - r/d)^2 \Theta(1 - r/d)$$

- DST occurs twice when (hardness ε^*) ↗
- Good agreement with simulation
- Shear thinning for large $\dot{\gamma}^*$
- No thinning for small $\dot{\gamma}^*$

Previous study (dilute soft-core system)

- Relationship to hard-core limit?
  ⇒ Softness is inevitable at a certain shear rate.



This shear rate ↗ as φ ↗.
Softness is the origin.
Does this survive even when φ ↗ ?

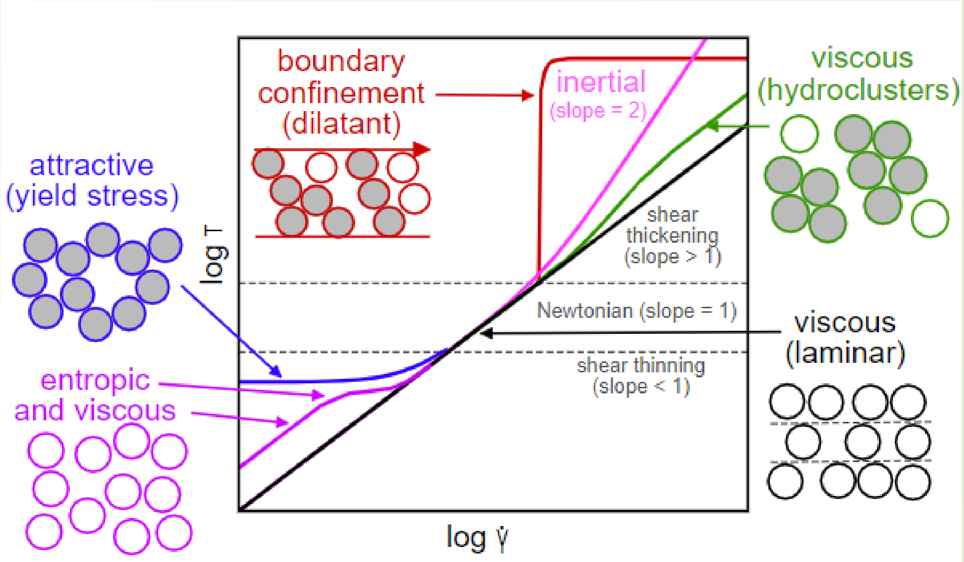
This shear rate is common.
Similar to hard-core system.
This DST will vanish as the density ↗.
(☞ Hayakawa, Takada, Garzó, PRE (2017))

$\varphi = 0.01, \xi_{\text{env}} = 1.0$

ε^* is changed.

Motivation

- What causes DST?
  - Tangential friction between particles?
    - ☞ for granular system; Otsuki & Hayakawa, PRE (2011)
  - Hydrodynamic interaction?



Our question:
Is it possible to observe DST for simple systems, especially, frictionless soft-core systems?

This question might sound crazy for people in rheology community. But this actually happens!

Theoretical oriented motivation of this study

Question:

- Can we observe DST of frictionless soft inertial suspension?
- If we can, is it possible to treat it by the kinetic theory?

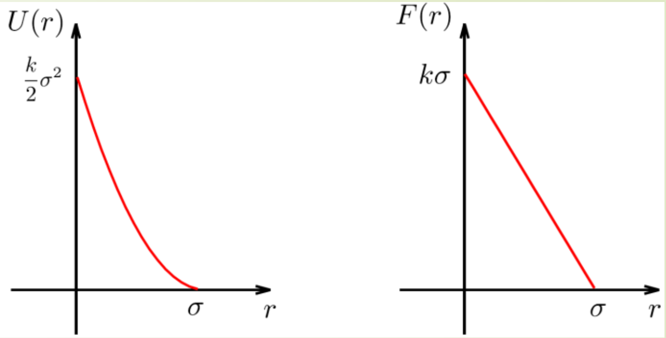
Approach:

- Molecular dynamics (MD) simulation
- Kinetic theory of inertial suspension

	Hard-core	Soft-core
Dilute	Hayakawa & Takada PTEP (2019)	Sugimoto & Takada JPSJ (2020)
Moderately dense	Hayakawa, Takada, & Garzó PRE (2017), Takada, Hayakawa, Santos, & Garzó PRE (2020)	This study

Simulation model and setup

$\varepsilon = k\sigma^2$



- Monodisperse particles (mass: m, diameter: σ)
- Interaction = harmonic potential

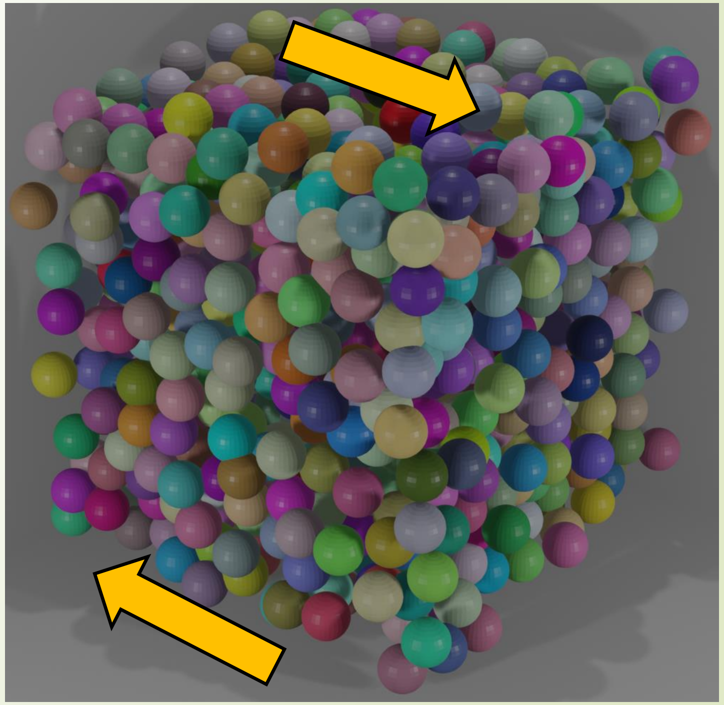
$$\boldsymbol{F}(\boldsymbol{r}) = -\frac{\partial U(r)}{\partial \boldsymbol{r}}, U(r) = \frac{\varepsilon}{2}\left(1 - \frac{r}{\sigma}\right)^2 \Theta\left(1 - \frac{r}{\sigma}\right)$$

- Equation of motion = Langevin eq.

$$\frac{d\boldsymbol{p}_i}{dt} = \sum_j \boldsymbol{F}(\boldsymbol{r}_{ij}) - \zeta \boldsymbol{p}_i + m\boldsymbol{\xi}_i$$

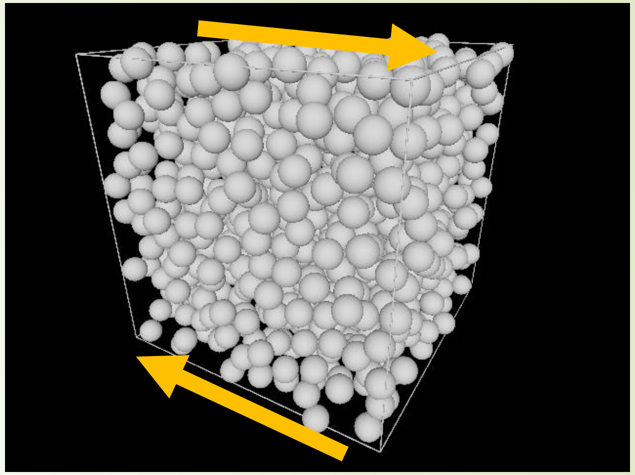
$\boldsymbol{p}_i \equiv m(\boldsymbol{v}_i - \dot{\gamma} y_i \hat{\boldsymbol{e}}_x)$: peculiar momentum

- Shear is applied.
  Sllod + Lees-Edwards b.c.



Langevin model for suspensions

Langevin equation: $\frac{d\boldsymbol{p}_i}{dt} = \underbrace{\sum_j \boldsymbol{F}_{ij}}_{①} \underbrace{- \zeta \boldsymbol{p}_i}_{②} + \underbrace{m\boldsymbol{\xi}_i}_{③}$



$\boldsymbol{p}_i \equiv m(\boldsymbol{v}_i - \dot{\gamma} y_i \boldsymbol{e}_x) = m\boldsymbol{V}_i$: peculiar momentum

① interparticle force between particles

② drag term (Stokesian)

③ thermal noise term satisfying

$$\langle \boldsymbol{\xi}_i(t) \rangle = 0,$$

$$\langle \xi_{i,\alpha}(t) \xi_{j,\beta}(t') \rangle = \frac{2\zeta T_{\mathrm{env}}}{m} \delta_{ij} \delta_{\alpha\beta} \delta(t - t')$$

Control parameters

① Packing fraction: φ

② Shear rate: $\dot{\gamma} \Rightarrow \dot{\gamma}^* \equiv \dot{\gamma}/\zeta$

③ Particle hardness: $\varepsilon \Rightarrow \varepsilon^* \equiv \frac{\varepsilon}{m\sigma^2\zeta^2}$

④ Env. temp.: $T_{\mathrm{env}} \Rightarrow \xi_{\mathrm{env}} \equiv \sqrt{\frac{T_{\mathrm{env}}}{m} \frac{1}{\zeta\sigma}}$

Langevin model for suspensions

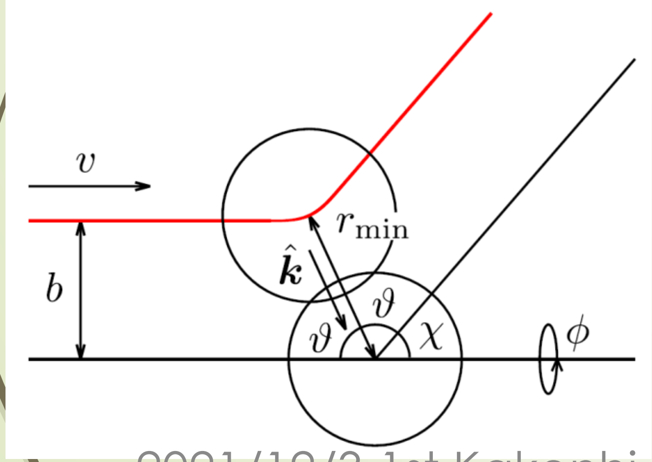
Langevin equation: $\frac{d\boldsymbol{p}_i}{dt} = \sum_j \boldsymbol{F}_{ij} - \zeta \boldsymbol{p}_i + m\boldsymbol{\xi}_i$

Enskog kinetic equation for the inertial suspension

$$\left(\frac{\partial}{\partial t} - \dot{\gamma} V_y \frac{\partial}{\partial V_x}\right) f(\boldsymbol{V}, t) = \zeta \frac{\partial}{\partial \boldsymbol{V}} \cdot \left(\left[\boldsymbol{V} + \frac{T_{\rm env}}{m}\frac{\partial}{\partial \boldsymbol{V}}\right] f(\boldsymbol{V}, t)\right) + J_{\rm E}(\boldsymbol{V}|f, f)$$

shear — drag from the solvent — particle interaction

Collision integral: $J_{\rm E}[\boldsymbol{V}|f, f] = \int d\boldsymbol{V}_2 \int d\widehat{\boldsymbol{k}} \sigma_{\rm s}(\chi, V_{12}) V_{12}$
$\times \left[f^{(2)}\left(\boldsymbol{r}, \boldsymbol{r} + r_{\rm min}\widehat{\boldsymbol{k}}, \boldsymbol{V}_1'', \boldsymbol{V}_2'', t\right) - f^{(2)}\left(\boldsymbol{r}, \boldsymbol{r} - r_{\rm min}\widehat{\boldsymbol{k}}, \boldsymbol{V}_1, \boldsymbol{V}_2, t\right)\right]$

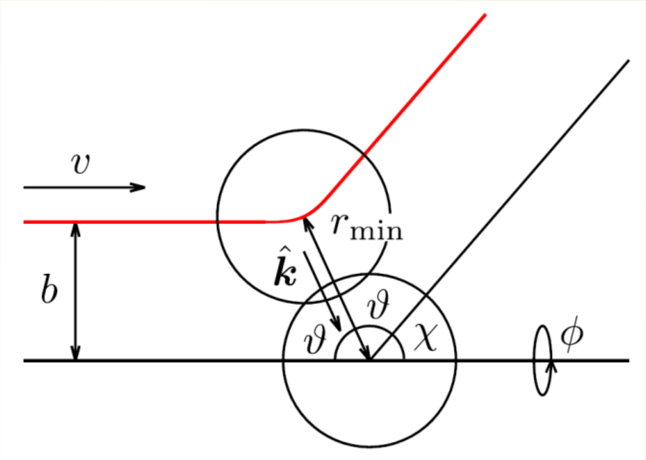


$\sigma_{\rm s}(\chi, V_{12})$: collision cross section determined from the scattering problem

$f^{(2)}(\boldsymbol{r}_1, \boldsymbol{r}_2, \boldsymbol{V}_1, \boldsymbol{V}_2, t) \simeq g_0 f(r_1, V_1, t) f(r_2, V_2, t)$: decoupling arrox.

Softness of particles

- Scattering analysis
  ⇒ Appearance of softness



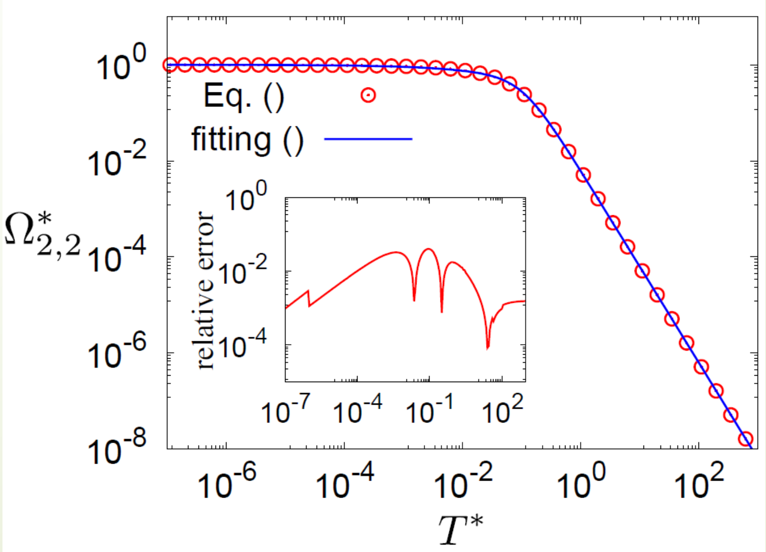
Omega integral:

$$\Omega^*_{2,2}(T^*) \equiv \int_0^\infty dy\, y^7 e^{-y^2} \int_0^1 db^* b^* \sin^2 \chi\left(b^*, 2y\sqrt{T^*}\right)$$

- The ratio of the coll. freq. of soft particles to that of hard-core particles

$$\Omega^*_{2,2} = \nu_{\text{soft}}/\nu_{\text{HC}}$$

- Low T: hard-core like ($\Omega^*_{2,2} \simeq 1$)
- High T: softer and softer ($\Omega^*_{2,2} \to 0$)



Enskog kinetic equation for the inertial suspension

$$\left(\frac{\partial}{\partial t} - \dot{\gamma} V_y \frac{\partial}{\partial V_x}\right) f(\boldsymbol{V}, t) = \zeta \frac{\partial}{\partial \boldsymbol{V}} \cdot \left(\left[\boldsymbol{V} + \frac{T_{\rm env}}{m}\frac{\partial}{\partial \boldsymbol{V}}\right] f(\boldsymbol{V}, t)\right) + J_{\rm E}(\boldsymbol{V}|f,f)$$

Evolution equation for the kinetic stress:

$$\frac{\partial}{\partial t} P_{\alpha\beta}^k + \dot{\gamma}\left(\delta_{\alpha x} P_{y\beta}^k + \delta_{\beta x} P_{y\alpha}^k\right) = -2\zeta\left(P_{\alpha\beta}^k - nT_{\rm env}\delta_{\alpha\beta}\right) - \Lambda_{\alpha\beta}$$

Kinetic stress: $P_{\alpha\beta}^k = m\int d\boldsymbol{V} V_\alpha V_\beta f(\boldsymbol{V}, t)$

Moment of the collision integral: $\Lambda_{\alpha\beta} = -m\int d\boldsymbol{V} V_\alpha V_\beta J_{\rm E}(V|f,f)$

This equation is NOT closed!

Closure: Grad's moment method

$$f(\boldsymbol{V}) = f_{\rm M}(\boldsymbol{V})\left(1 + \frac{m}{2T}\left(\frac{P_{\alpha\beta}^k}{nT} - \delta_{\alpha\beta}\right) V_\alpha V_\beta\right)$$

Maxwellian distribution

$$f_{\rm M}(\boldsymbol{V}) = n\left(\frac{m}{2\pi T}\right)^{\frac{3}{2}} \exp\left(-\frac{mV^2}{2T}\right)$$

Further two assumptions:

- **Replacement** (for any $F(\hat{\boldsymbol{k}}, r_{min})$)

$$\int d\boldsymbol{\Omega}\sigma_{\mathrm{s}}(\chi, V_{12})V_{12}\, F(\hat{\boldsymbol{k}}, r_{\mathrm{min}}) \to \Omega_{2,2}^{*}\, d^2 \int d\hat{\boldsymbol{k}}\, \Theta(\boldsymbol{V}_{12}\cdot\hat{\boldsymbol{k}})(\boldsymbol{V}_{12}\cdot\hat{\boldsymbol{k}})F(\hat{\boldsymbol{k}}, d)$$

$\sigma_{\mathrm{s}}(\chi, V_{12})V_{12}$: collision cylinder of soft particles
$\Rightarrow d^2(\boldsymbol{V}_{12}\cdot\hat{\boldsymbol{k}})$: that of hard-core particles + $\Omega_{2,2}^{*}$ (softness)
Same procedure as hard-core system

- Linear approximation with respect to $\dot{\boldsymbol{\gamma}}^{*}$

We can obtain a set of equations for

① $\theta \equiv \frac{T}{T_{\mathrm{env}}}$, ② $\Delta\theta \equiv \frac{P_{xx}^{k}-P_{yy}^{k}}{nT_{\mathrm{env}}}$, ③ $\delta\theta \equiv \frac{P_{xx}^{k}-P_{zz}^{k}}{nT_{\mathrm{env}}}$, ④ $\Pi_{\alpha\beta}^{*} \equiv \frac{P_{\alpha\beta}^{k}}{nT_{\mathrm{env}}} - \theta\delta_{\alpha\beta}$.

(①: dim.less. temp., ②,③: dim.less. anisotropic temp., ④ dim.less. shear stress)

※ The contact part of the stress is written in terms of the kinetic part.

A set of dynamic equations:

$$-\frac{2\dot\gamma^*}{3}\mathcal{C}\Pi^*_{xy} = 2(\theta-1),$$
$$-2\dot\gamma^*\Pi^*_{xy} = (2+\nu^*_{\rm soft})\Delta\theta,$$
$$-2\dot\gamma^*\mathcal{E}\Pi^*_{xy} = (2+\nu^*_{\rm soft})\delta\theta,$$
$$(2+\nu^*_{\rm soft})\Pi^*_{xy} = \dot\gamma^*\left(\frac{2}{3}\mathcal{D}\Delta\theta - \frac{1}{3}\mathcal{E}\delta\theta - \mathcal{C}\theta\right),$$

$$\mathcal{C} \equiv 1 + \frac{8}{5}\varphi g_0 \Omega^*_{2,2},$$
$$\mathcal{D} \equiv 1 - \frac{4}{35}\varphi g_0 \Omega^*_{2,2},$$
$$\mathcal{E} \equiv 1 - \frac{16}{35}\varphi g_0 \Omega^*_{2,2}.$$

Then, the rheology is described by

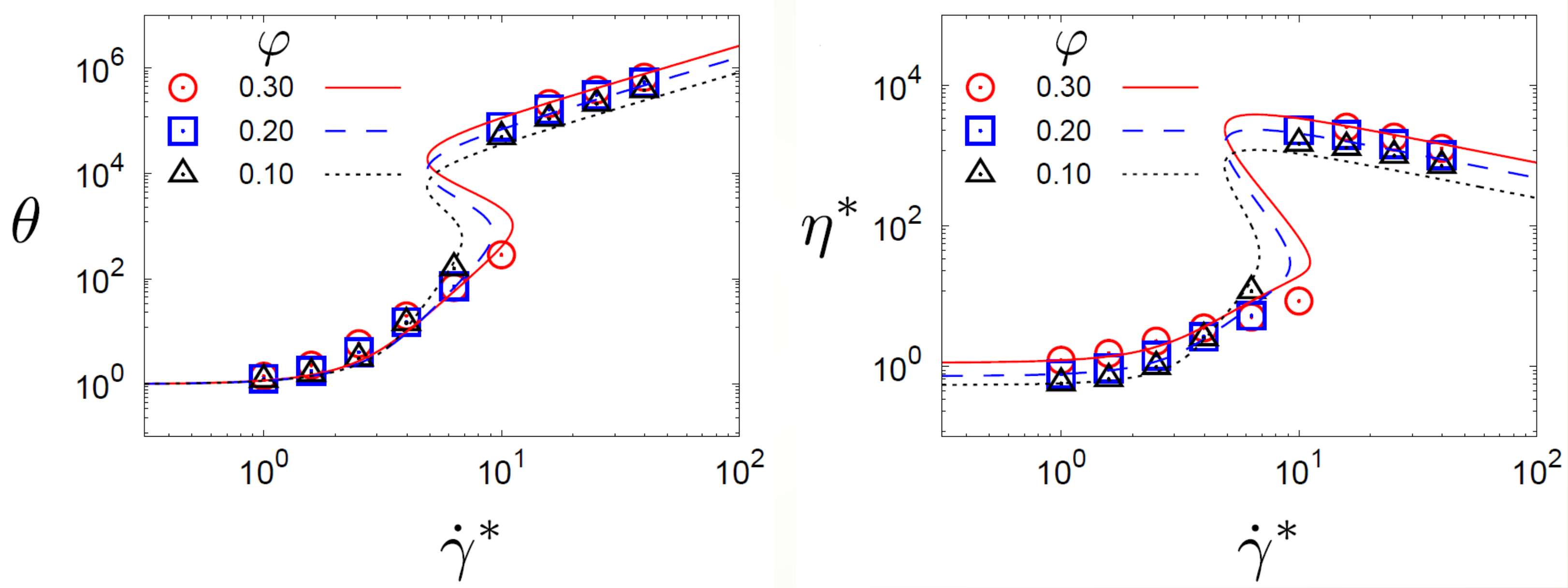
$$\dot\gamma^* = \sqrt{\frac{-3(1-\theta^{-1})(2+\nu^*_{\rm soft})}{\mathcal{C}\mathcal{F}}},$$
$$\Pi^*_{xy} = -\frac{3}{\dot\gamma^*\mathcal{C}}(\theta-1),$$
$$\Delta\theta = \frac{3}{\mathcal{C}}\frac{2(\theta-1)}{2+\nu^*_{\rm soft}},$$
$$\delta\theta = \frac{3\mathcal{E}}{\mathcal{C}}\frac{2(\theta-1)}{2+\nu^*_{\rm soft}}.$$

$$\mathcal{F} \equiv \frac{2}{3}\mathcal{D}\frac{\Delta\theta}{\theta} - \frac{1}{3}\mathcal{E}\frac{\delta\theta}{\theta} - \mathcal{C} = \frac{2\mathcal{D}-\mathcal{E}^2}{\mathcal{C}}\frac{2(1-\theta^{-1})}{2+\nu^*_{\rm soft}} - \mathcal{C}.$$

All quantities are written
as a function of the temperature.

Flow curves

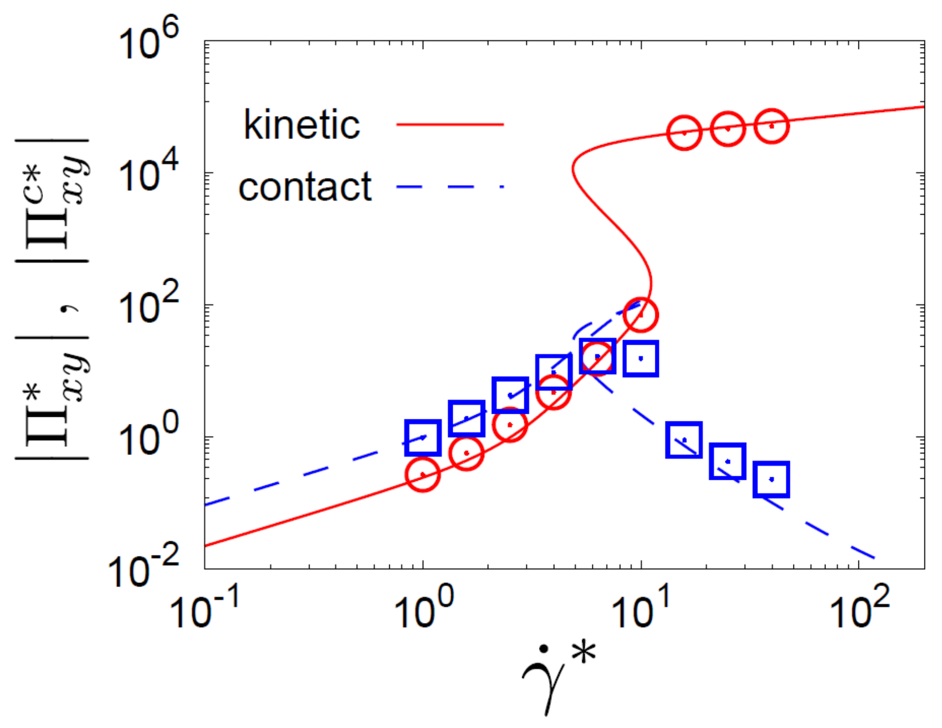
- Temperature and viscosity against the shear rate
  $\varphi = 0.10, 0.20, 0.30; \varepsilon^* = 10^4, \xi_{\mathrm{env}} = 1.0$



- DST-like behavior survives even for the finite density! (softness induced DST)
- Shear thinning in the high shear regime
- Good agreement with the simulation

Kinetic and contact parts of shear stress

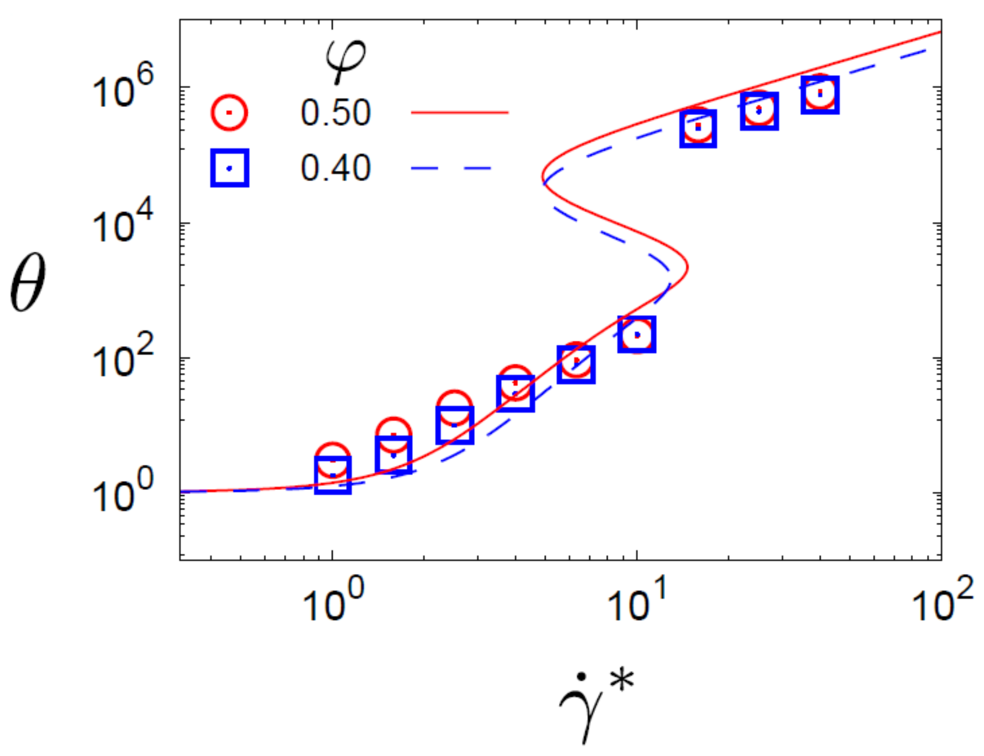
- $\varphi = 0.30, \varepsilon^* = 10^4, \xi_{\mathrm{env}} = 1.0$

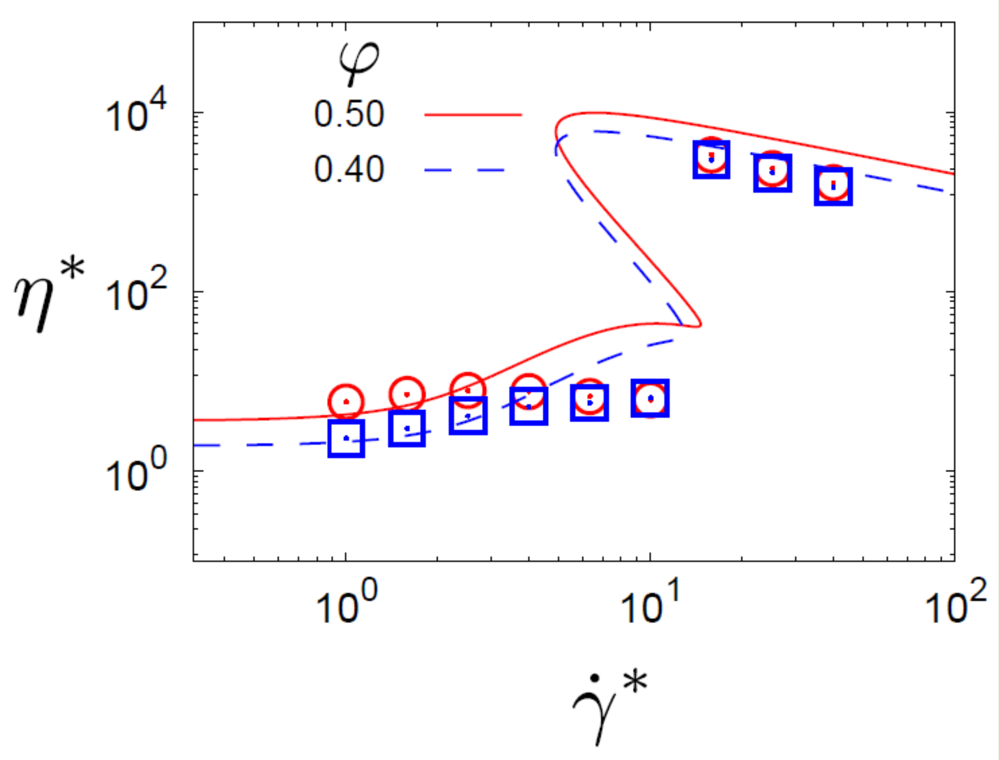


- Both contributions are well reproduced.
- Kinetic (contact) part increases (decreases) after DST.

Flow curves for denser situations

- $\varphi = 0.40, 0.50, ; \varepsilon^* = 10^4, \xi_{\text{env}} = 1.0$

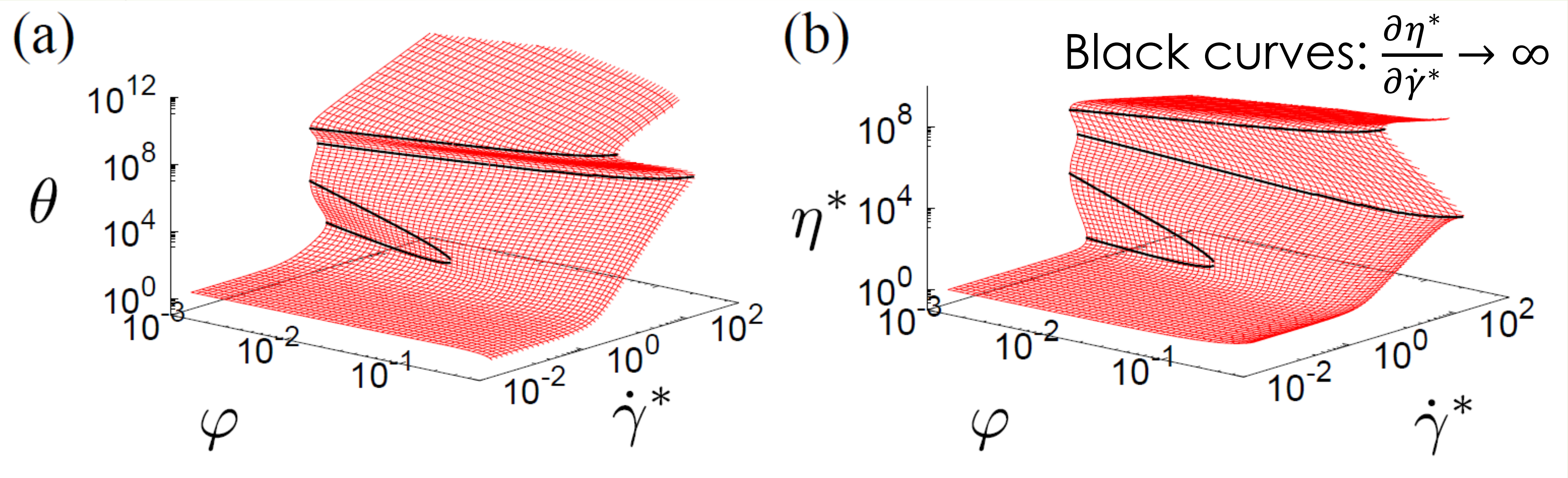




- Still good agreement with the simulation
  ⇒ Kinetic theory is available in the wide range of φ.

Flow curves (3D)

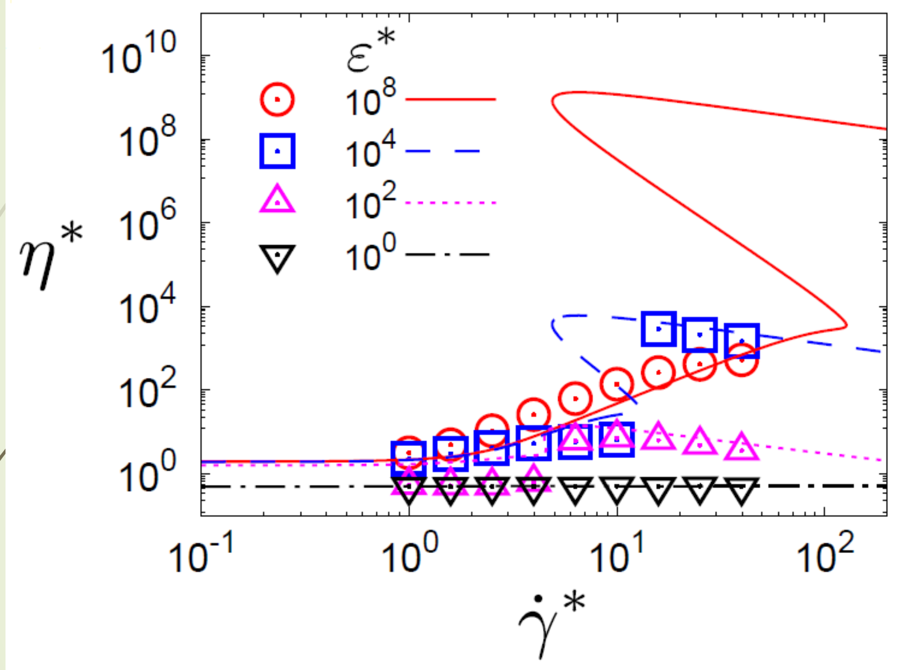
- Continuously change the packing fraction

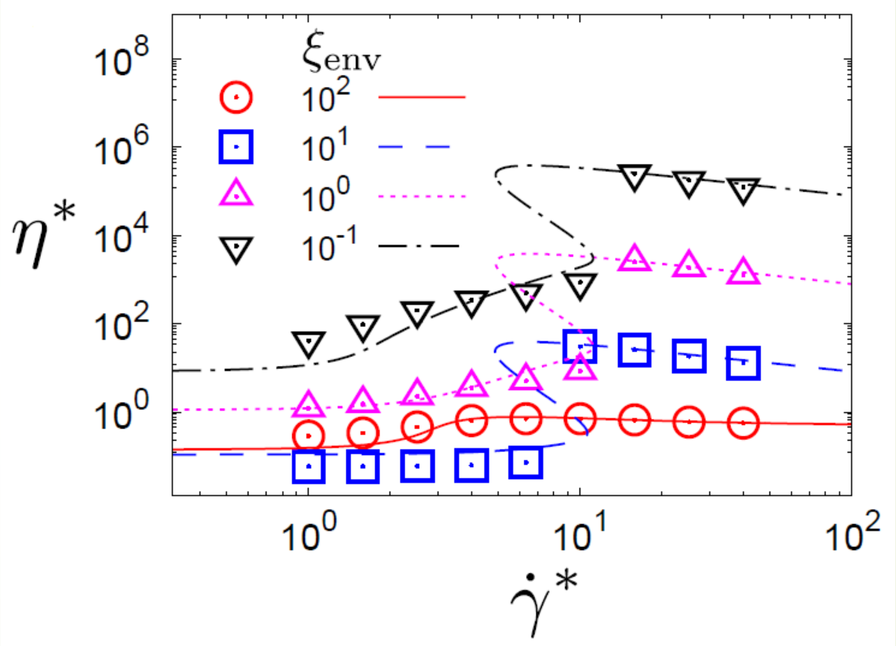


Softness induced DST exists in the wide range of φ

Comparison with simulations

- Also change $\varepsilon^* = 10^4$ and $\xi_{\rm env}$



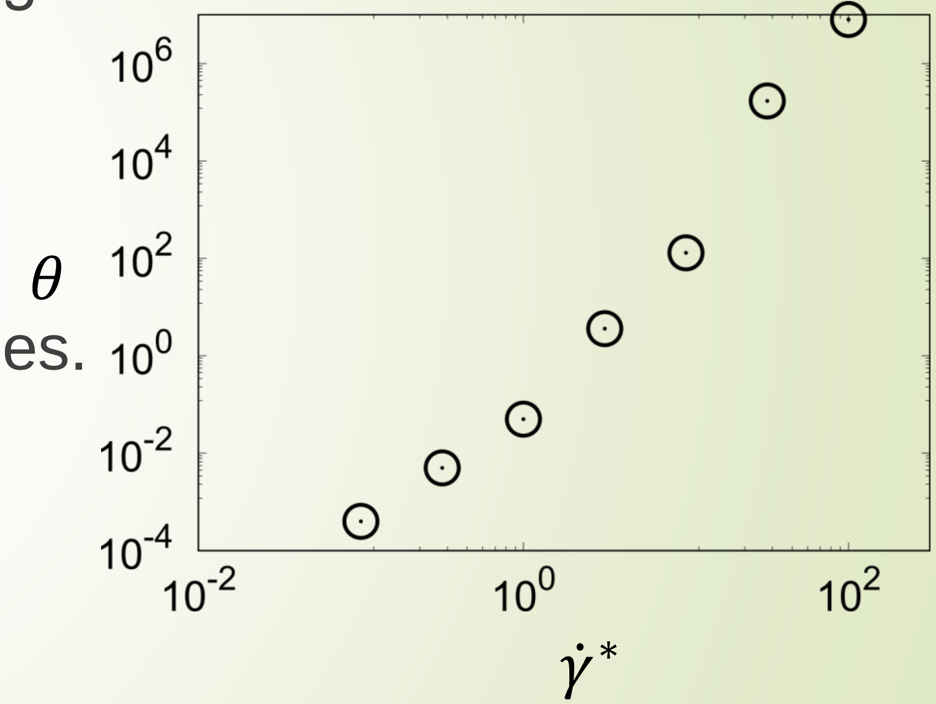


- Still good agreement for $\xi_{\rm env} \geq 1$
  ⇒ Kinetic theory is available
  in the wide range of the control parameters.

Discussion

- **Hydrodynamic interaction**
  Our current model: an oversimplified model.
  Drag from the background = Stokes's drag

  For denser systems,
  drag ⇒ the resistance matrix
  ☞ *Kim & Karrila, "Microhydrodynamics" (1991)*
  ⇒ depends on the configuration of particles.



$$\begin{bmatrix} \boldsymbol{F}_i \\ \boldsymbol{F}_j \\ \boldsymbol{\sigma}_i \\ \boldsymbol{\sigma}_j \end{bmatrix} = -\eta \begin{bmatrix} \boldsymbol{A}_{ij}^{(11)} & \boldsymbol{A}_{ij}^{(12)} & \widetilde{\boldsymbol{G}}_{ij}^{(11)} & \widetilde{\boldsymbol{G}}_{ij}^{(12)} \\ \boldsymbol{A}_{ij}^{(21)} & \boldsymbol{A}_{ij}^{(22)} & \widetilde{\boldsymbol{G}}_{ij}^{(21)} & \widetilde{\boldsymbol{G}}_{ij}^{(22)} \\ \boldsymbol{G}_{ij}^{(11)} & \boldsymbol{G}_{ij}^{(12)} & \boldsymbol{M}_{ij}^{(11)} & \boldsymbol{M}_{ij}^{(12)} \\ \boldsymbol{G}_{ij}^{(21)} & \boldsymbol{G}_{ij}^{(22)} & \boldsymbol{M}_{ij}^{(21)} & \boldsymbol{M}_{ij}^{(22)} \end{bmatrix} \begin{bmatrix} \boldsymbol{v}_i - \boldsymbol{E}\cdot\boldsymbol{x}_i \\ \boldsymbol{v}_j - \boldsymbol{E}\cdot\boldsymbol{x}_j \\ -\boldsymbol{E} \\ -\boldsymbol{E} \end{bmatrix}$$

Is it possible to observe DST?
⇒ What we should do next.

Summary

- We construct the kinetic theory of inertial suspensions of soft particles.
- Softness is characterized by $\Omega^*_{2,2}$.
- DST-like behavior survives even for finite φ.
- Good agreement with the simulation in the wide range of the control parameters

What we should do

- Hydrodynamic effect