Beenakker-Mazur expansion
for suspensions of repulsive particles

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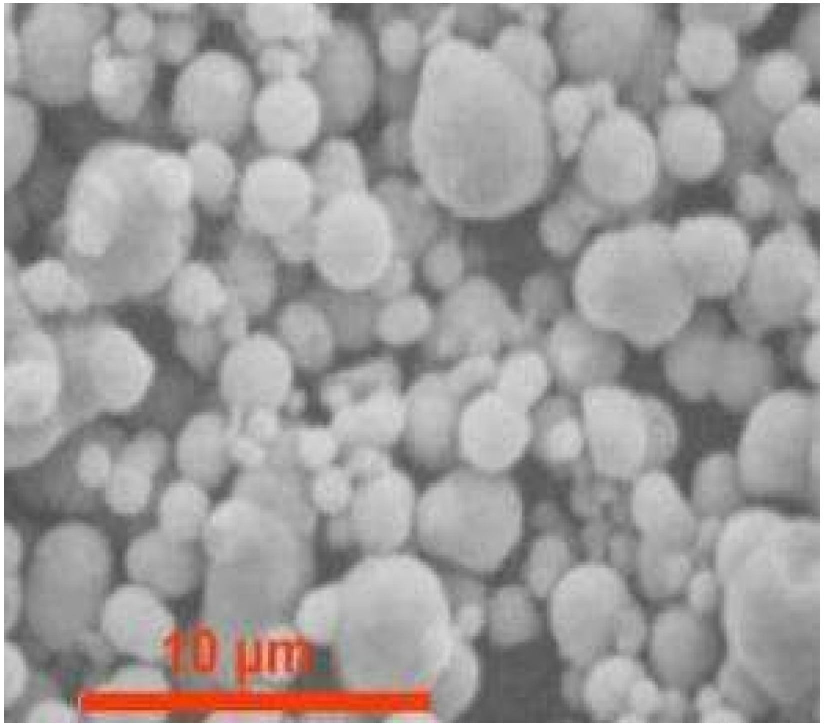
Introduction - suspensions

minute particles in liquid



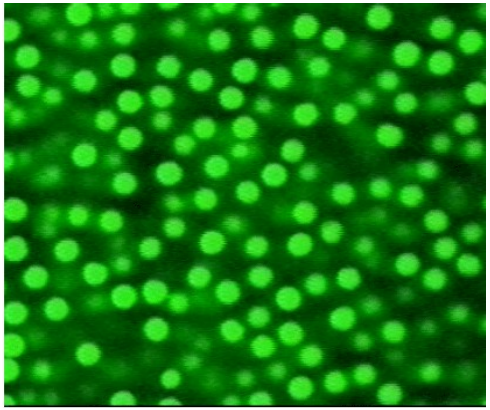
Liquid:
-temperature T
-viscosity μ
-density of the fluid ρ_f

Particles:
-radius a
-density of material ρ_p
-volume fraction ϕ

milk, blood,...

Aim of our work

Monodisperse suspension of spherical particles



Transport properties (short time):
-effective viscosity
-sedimentation coefficient
-diffusion coefficient

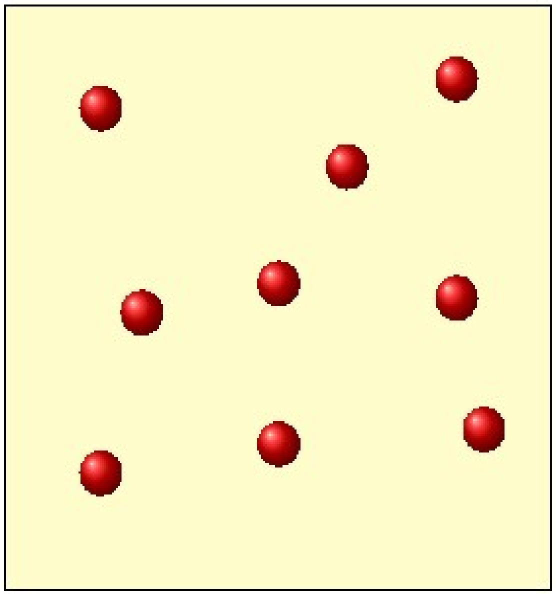
Over 100 years of research - still an open question
The most comprehensive method nowadays: Beenakker-Mazur method

To assess Beenakker-Mazur method in case of e.g. rotational self-diffusion or effective viscosity coefficient for suspension of repulsive particles by comparison with numerical simulations

Comments on polidispersity or nonspherical particles

Hard-sphere suspension

Unbounded liquid,
N particles in configuration $X \equiv \mathbf{R}_1, \ldots, \mathbf{R}_N$



Stokes equations:

$$\begin{aligned} \nabla p\left(\mathbf{r}\right) - \eta\Delta\mathbf{v}\left(\mathbf{r}\right) &= 0 \\ \nabla \cdot \mathbf{v}\left(\mathbf{r}\right) &= 0 \end{aligned}$$

Stick boundary conditions,

$\mathbf{v}(\mathbf{r}) \rightarrow \mathbf{v}_0(\mathbf{r})$ *for* $r \rightarrow \infty$

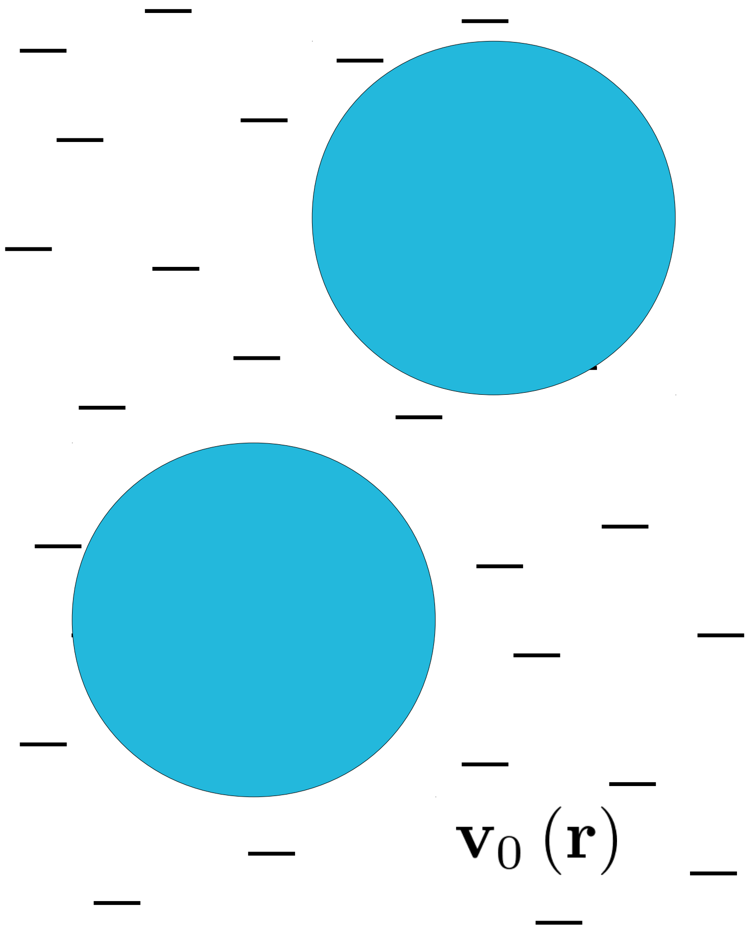
Effective viscosity

Landau: effective viscosity related to force on the surface of particles

$$\mathbf{f}_i(\mathbf{r}) = -\sigma(\mathbf{r}; X)\hat{\mathbf{n}}(\mathbf{r})\delta(|\mathbf{r} - \mathbf{R}_i| - a)$$

stress tensor

vector normal to the surface of particle i

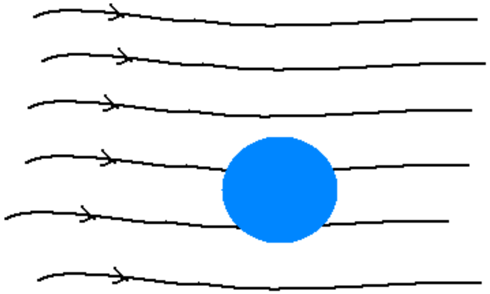


Single particle

Lamb (1895) $\mathbf{v}^{+}_{lm\sigma}(\mathbf{r})$

$$l = 1, 2, \ldots, \infty$$
$$m = -l, \ldots, l$$
$$\sigma = 0, 1, 2$$

Single particle in ambient flow $\mathbf{v}_0(\mathbf{r})$



$$\mathbf{f}_1(\mathbf{r}) = \int d\mathbf{r}' \mathbf{M}(\mathbf{r} - \mathbf{R}_1, \mathbf{r}' - \mathbf{R}_1)\, \mathbf{v}_0(\mathbf{r}')$$

Single particle operator
(Felderhof 1976)

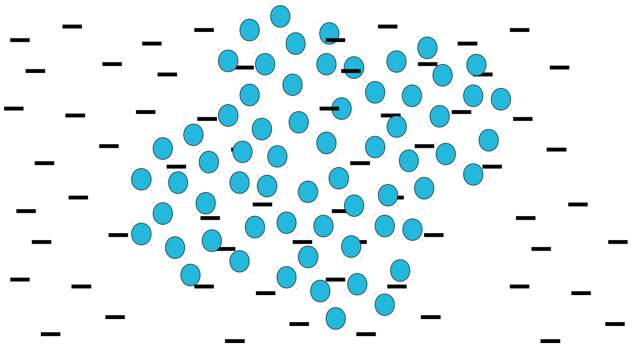
Surface force density
(Cox Brenner (1967); Mazur, Bedeaux (1974))

$$\mathbf{v}(\mathbf{r}) = \mathbf{v}_0(\mathbf{r}) + \int d^3\mathbf{r}' \mathbf{G}(\mathbf{r} - \mathbf{r}') \cdot \mathbf{f}_1(\mathbf{r}')$$

Oseen tensor:

$$\mathbf{G}(\mathbf{r}) = \frac{1}{8\pi\eta} \frac{\mathbf{1} + \hat{\mathbf{r}}\hat{\mathbf{r}}}{r}$$

Suspension



Ambient flow for the particle i in suspension:

$$\mathbf{v}_i(\mathbf{r}) = \mathbf{v}_0(\mathbf{r}) + \sum_{j \neq i} \int d^3\mathbf{r}' \mathbf{G}(\mathbf{r} - \mathbf{r}') \cdot \mathbf{f}_j(\mathbf{r}')$$

Single particle problem with modified ambient flow

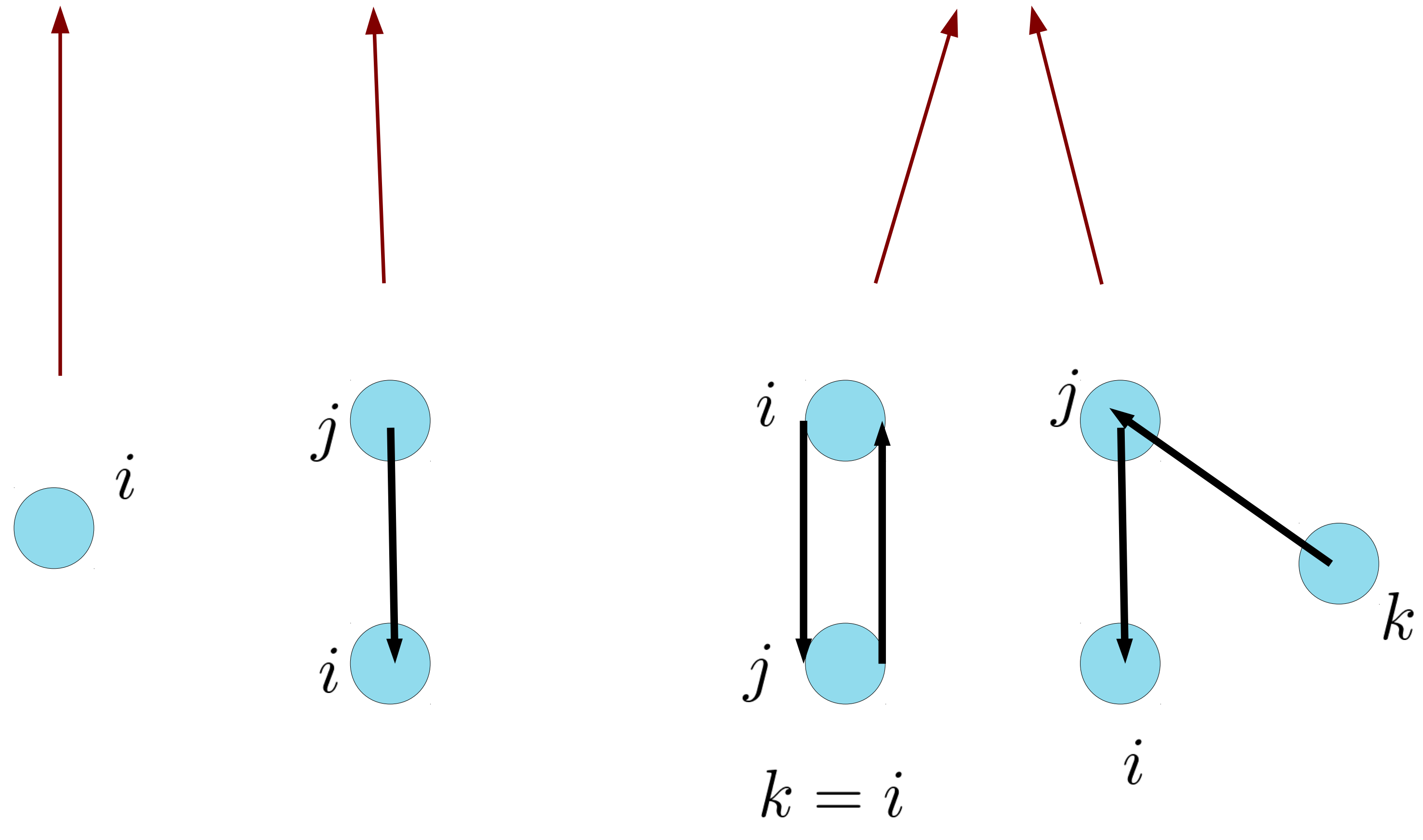
$$\mathbf{f}_i = \mathbf{M}(i) \left(\mathbf{v}_0 + \sum_{i \neq j} \mathbf{G}\mathbf{f}_j \right)$$

Solution in the form of the following scattering series (hydrodynamic interactions)

$$\mathbf{f}_i = \left(\mathbf{M}(i) + \sum_{j \neq i} \mathbf{M}(i)\mathbf{G}\mathbf{M}(j) + \sum_{j \neq i} \sum_{k \neq j} \mathbf{M}(i)\mathbf{G}\mathbf{M}(j)\mathbf{G}\mathbf{M}(k) + \ldots \right) \mathbf{v}_0$$

Scattering series

$$\mathbf{f}_i = \left(\mathbf{M}(i) + \sum_{j \neq i} \mathbf{M}(i)\mathbf{G}\mathbf{M}(j) + \sum_{j \neq i} \sum_{k \neq j} \mathbf{M}(i)\mathbf{G}\mathbf{M}(j)\mathbf{G}\mathbf{M}(k) + \ldots \right) \mathbf{v}_0$$



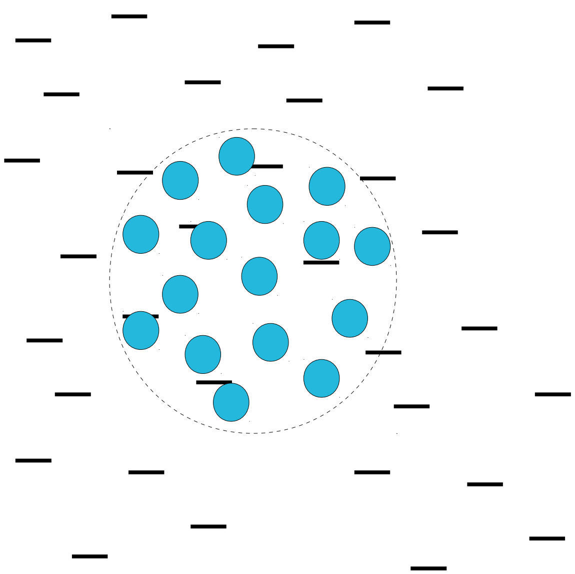
Transport properties – history and scattering series

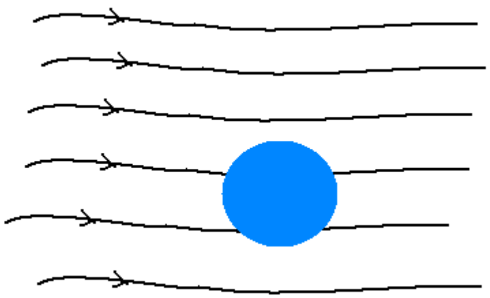
Einstein 1905 (corrected):

$$\eta_{eff} = \eta(1 + \frac{5}{2}\phi)$$

$$\phi = \frac{4}{3}\pi a^3 n$$

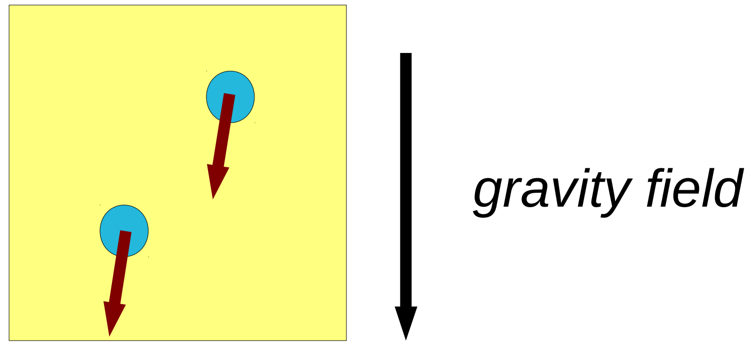
Single particle in ambient (shear) flow $\mathbf{v}_0(\mathbf{r})$

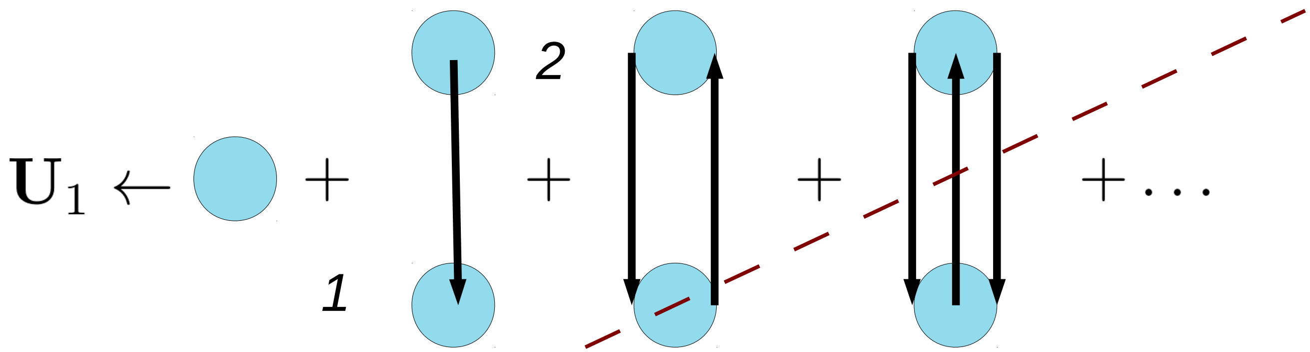


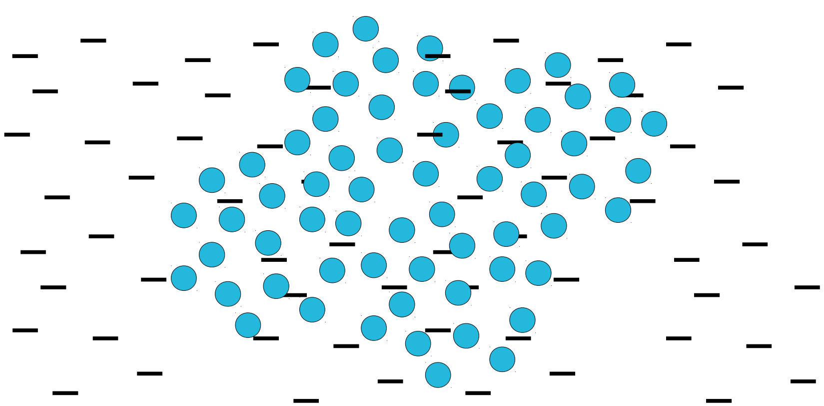


Hydrodynamic interactions neglected (no reflections, single particle)

Hydrodynamic interactions – Smoluchowski (1911)







$$\int d^3r\,|\mathbf{G}(\mathbf{r})| = \infty$$

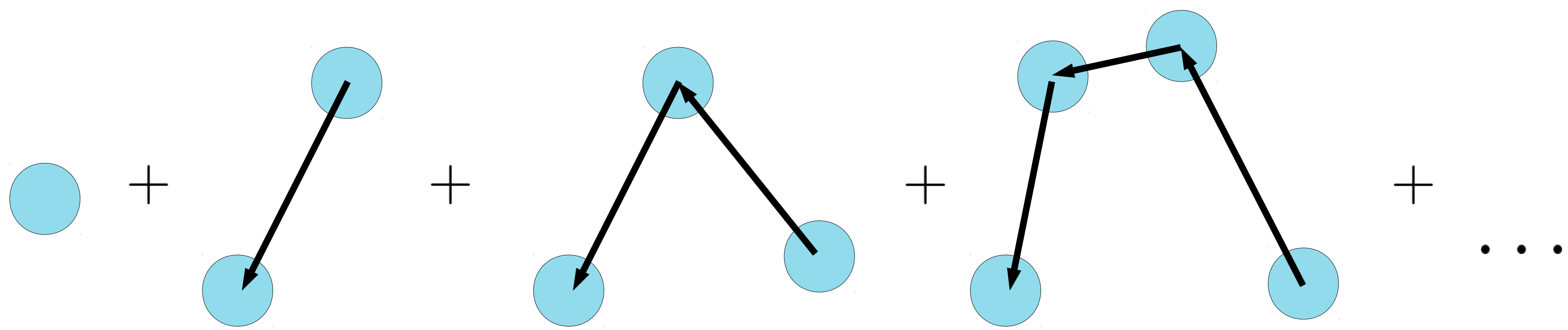
$$\mathbf{G}(\mathbf{r}) = \frac{1}{8\pi\eta}\,\frac{\mathbf{1} + \hat{\mathbf{r}}\hat{\mathbf{r}}}{r}$$

Well defined expression for effective viscosity?
Problem solved by Felderhof, Ford and Cohen (1982)

Beyond diluted suspensions

Saito (1950):

-extension of Einstein work on a mean-field level



$$\mathbf{M}(i)\mathbf{G}\mathbf{M}(j) \to W(\mathbf{R}_i - \mathbf{R}_j)\mathbf{M}(i)\mathbf{G}\mathbf{M}(j)$$

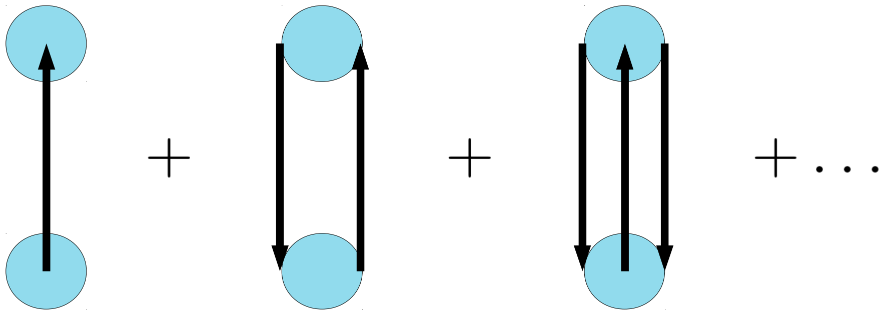
vanishes when two particles overlap

Saito formula:

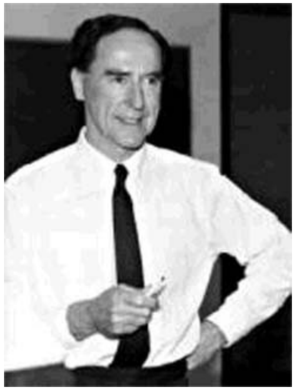
$$\frac{\eta_{eff}}{\eta} = \frac{1 + \frac{3}{2}\phi}{1 - \phi}$$

Two-particle hydrodynamic interactions (1972)

$$\frac{\eta_{eff}}{\eta} = 1 + \frac{5}{2}\phi + a_2\phi^2 + \ldots$$



(strong hydrodynamic interactions of close particles - lubrication)



absolute convergence

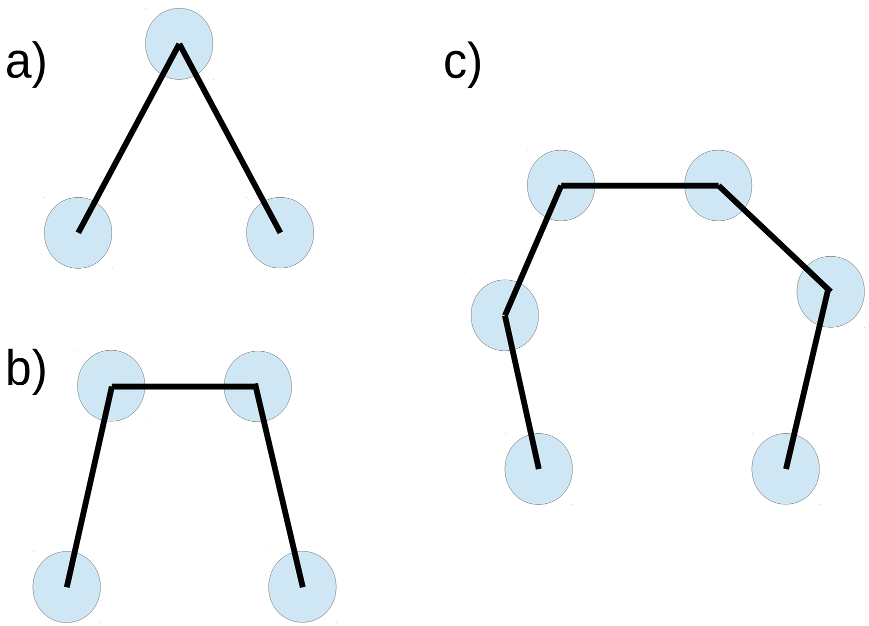
Batchelor, Green (1972): $a_2 \approx 5.2$

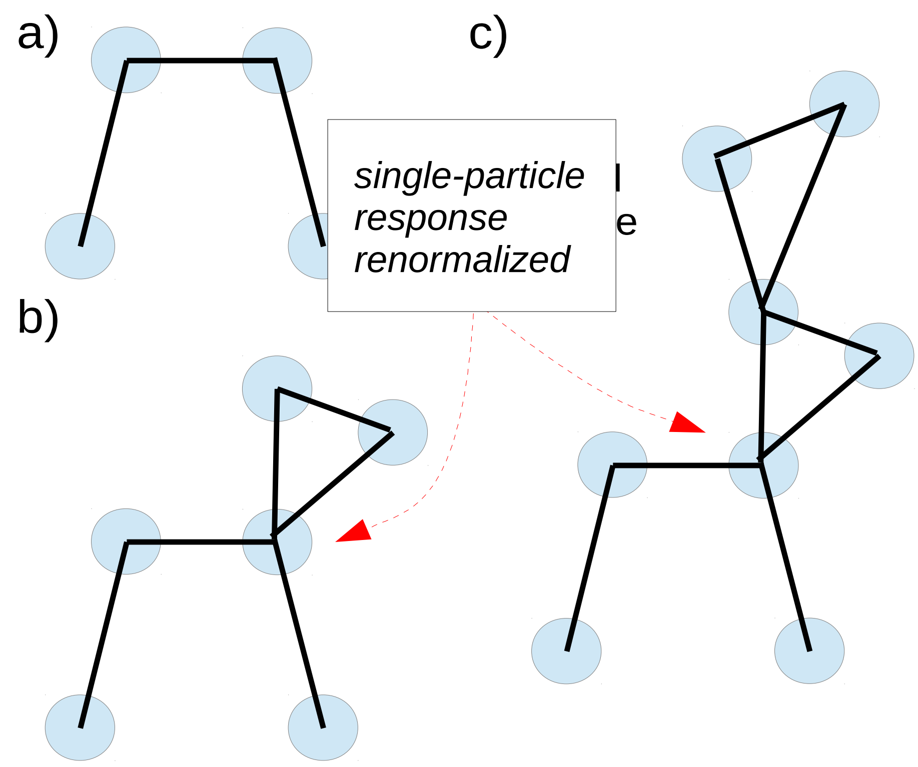
$$\int d^3r \left|\mathbf{G}(\mathbf{r})\right| = \infty$$

(ad hoc renormalization)

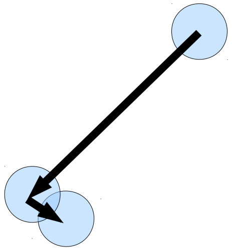
Beenakker-Mazur method (1983)

Idea of the method – resummation of certain class of hydrodynamic interactions – 'ring-selfcorrelations'





No correlations in position between particles in the above resummed terms

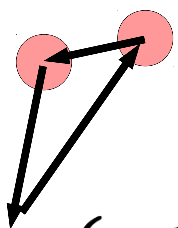


Resummation of ring-self correlations

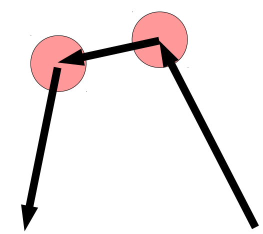
Beenakker and Mazur introduce a kernel of a single particle operator

$$\mathcal{M}(\mathbf{R},\mathbf{R}';X) = \delta(\mathbf{R}-\mathbf{R}')\sum_{i=1}^{N} M(\mathbf{R}_i)\delta(\mathbf{R}-\mathbf{R}_i)$$

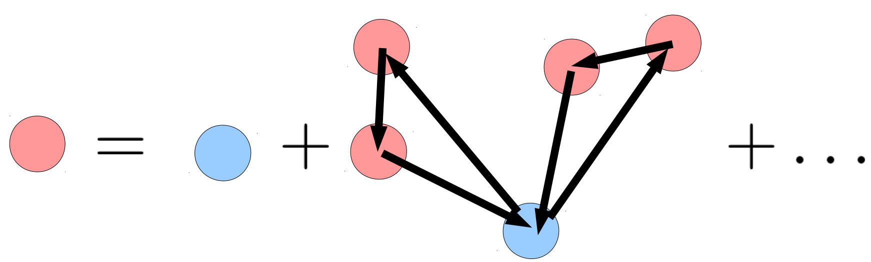
Ring-selfcorrelations:



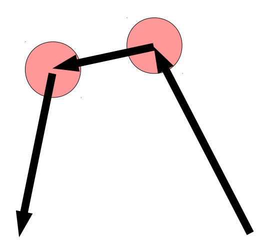
$$G^s_{\langle\mathcal{M}_R\rangle}(\mathbf{R},\mathbf{R}') = \begin{cases} G_{\langle\mathcal{M}_R\rangle}(\mathbf{R},\mathbf{R}') & \text{for } \mathbf{R}=\mathbf{R}' \\ 0 & \text{for } \mathbf{R}\neq\mathbf{R}' \end{cases}$$



$$\mathcal{M}_R = \mathcal{M}\left(1 - G^s_{\langle\mathcal{M}_R\rangle}\mathcal{M}\right)^{-1}$$



$$G_{\langle\mathcal{M}_R\rangle} = \tilde{G}\left(1 - \langle\mathcal{M}_R\rangle\,\tilde{G}\right)^{-1}$$



Beenakker-Mazur method (1983)

Beenakker-Mazur represented the scattering series

$$\mathbf{M}(i) + \sum_{j \neq i} \mathbf{M}(i)\mathbf{G}\mathbf{M}(j) + \sum_{j \neq i} \sum_{k \neq j} \mathbf{M}(i)\mathbf{G}\mathbf{M}(j)\mathbf{G}\mathbf{M}(k) + \ldots$$

by the following equivalent form (expansion in renormalized fluctuations)

$$\mathcal{M} + \mathcal{M} G_{\langle \mathcal{M}_R \rangle} \left[1 - \left(\mathcal{M}_R - \langle \mathcal{M}_R \rangle \right) \tilde{G}_{\langle \mathcal{M}_R \rangle} \right]^{-1} \mathcal{M}_R$$

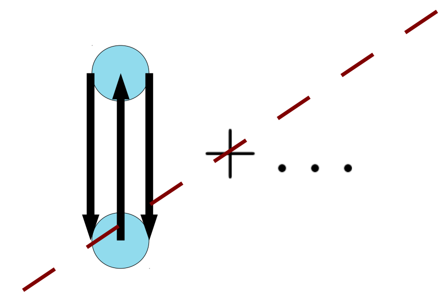
Delta gamma scheme (Beenakker Mazur method): the above expression up to second order in fluctuations, averaged over configurations of particles

$$\left\langle \mathcal{M} + \mathcal{M} G_{\langle \mathcal{M}_R \rangle} \mathcal{M}_R + \mathcal{M} G_{\langle \mathcal{M}_R \rangle} \left(\mathcal{M}_R - \langle \mathcal{M}_R \rangle \right) \tilde{G}_{\langle \mathcal{M}_R \rangle} \mathcal{M}_R \right\rangle$$

Beenakker and Mazur scheme

Beenakker and Mazur scheme – expansion in density fluctuations (1983). The most comprehensive statistical physics theory for short times properties of suspension nowadays

- ✓ *Many-body character*
- ✓ *Long-range character*
- ✗ *Strong interactions of close particles*



No satisfactory statistical physics method including the above three features (still an open problem)

Propagator does not depend on correlations (rdf)

$$\mathcal{M} + \mathcal{M} G_{\langle \mathcal{M}_R \rangle} \left[1 - \left(\mathcal{M}_R - \langle \mathcal{M}_R \rangle \right) \tilde{G}_{\langle \mathcal{M}_R \rangle} \right]^{-1} \mathcal{M}_R$$

How interactions (e.g. electrostatic) influence results of Beenakker-Mazur method?

Check for rotational self-diffusion coefficient...

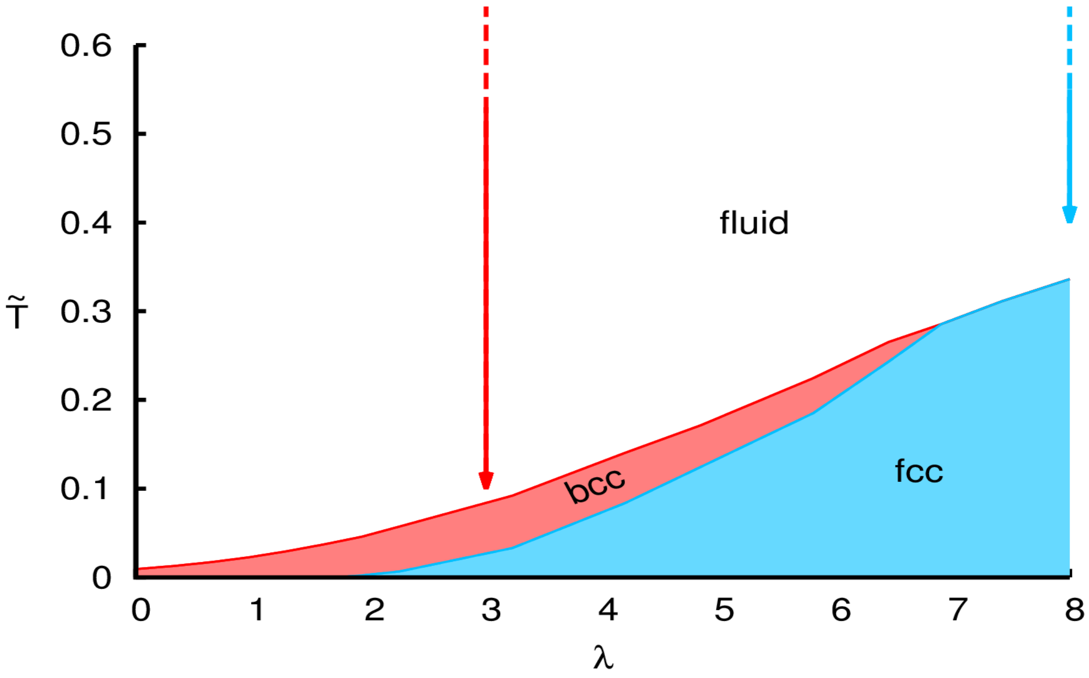
Yukawa-hard core repulsive potential

screening length

$$\frac{u(r)}{k_B T} = \begin{cases} \frac{1}{\tilde{T}} e^{\lambda} e^{-\lambda r/\langle r \rangle} \frac{\langle r \rangle}{r} & \text{for } r > 2a \\ \infty & r < 2a \end{cases}$$

For constant λ *, the limit of hard sphere is for* $\tilde{T} \to \infty$

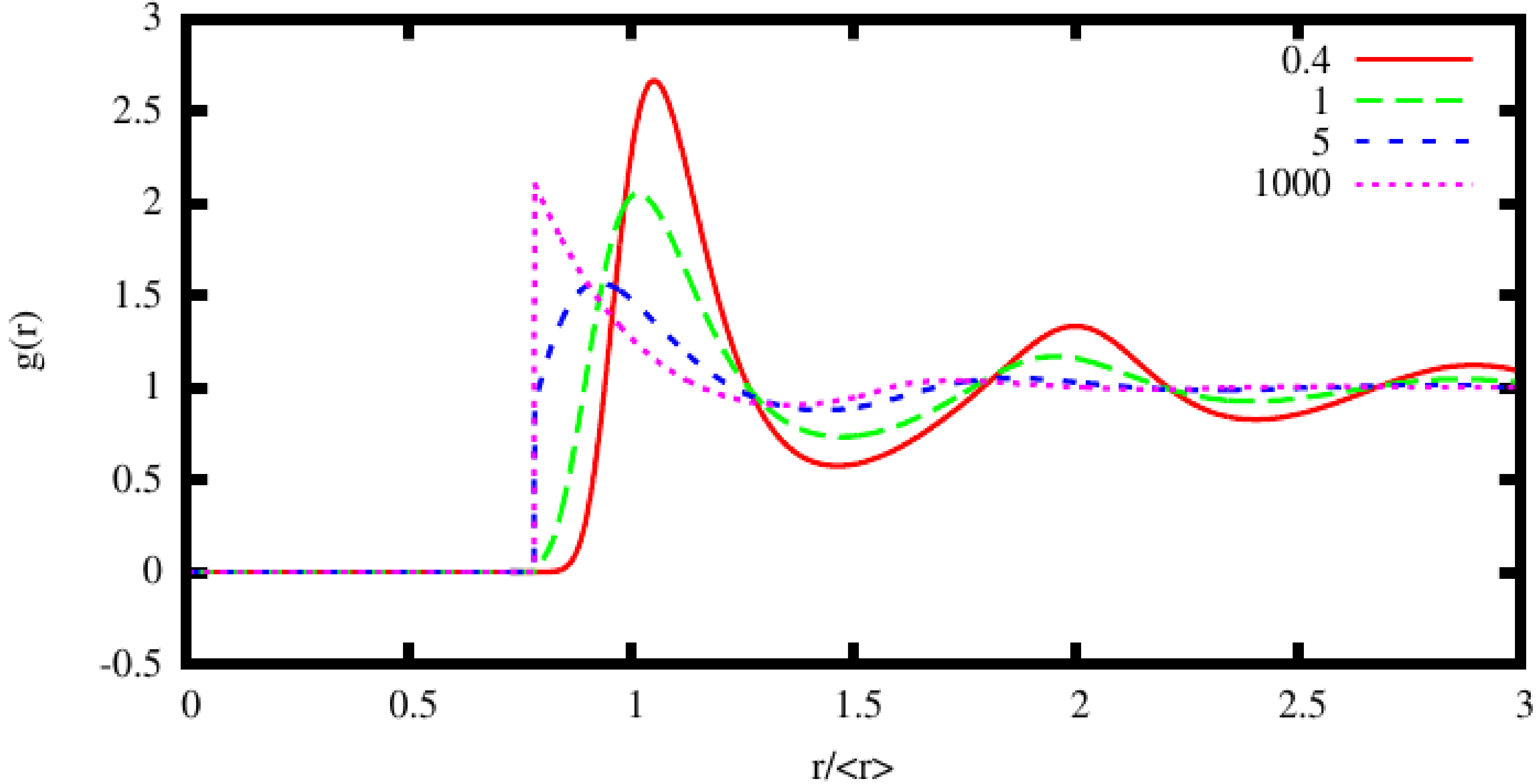
Equilibrium phase diagram:



Radial distribution function

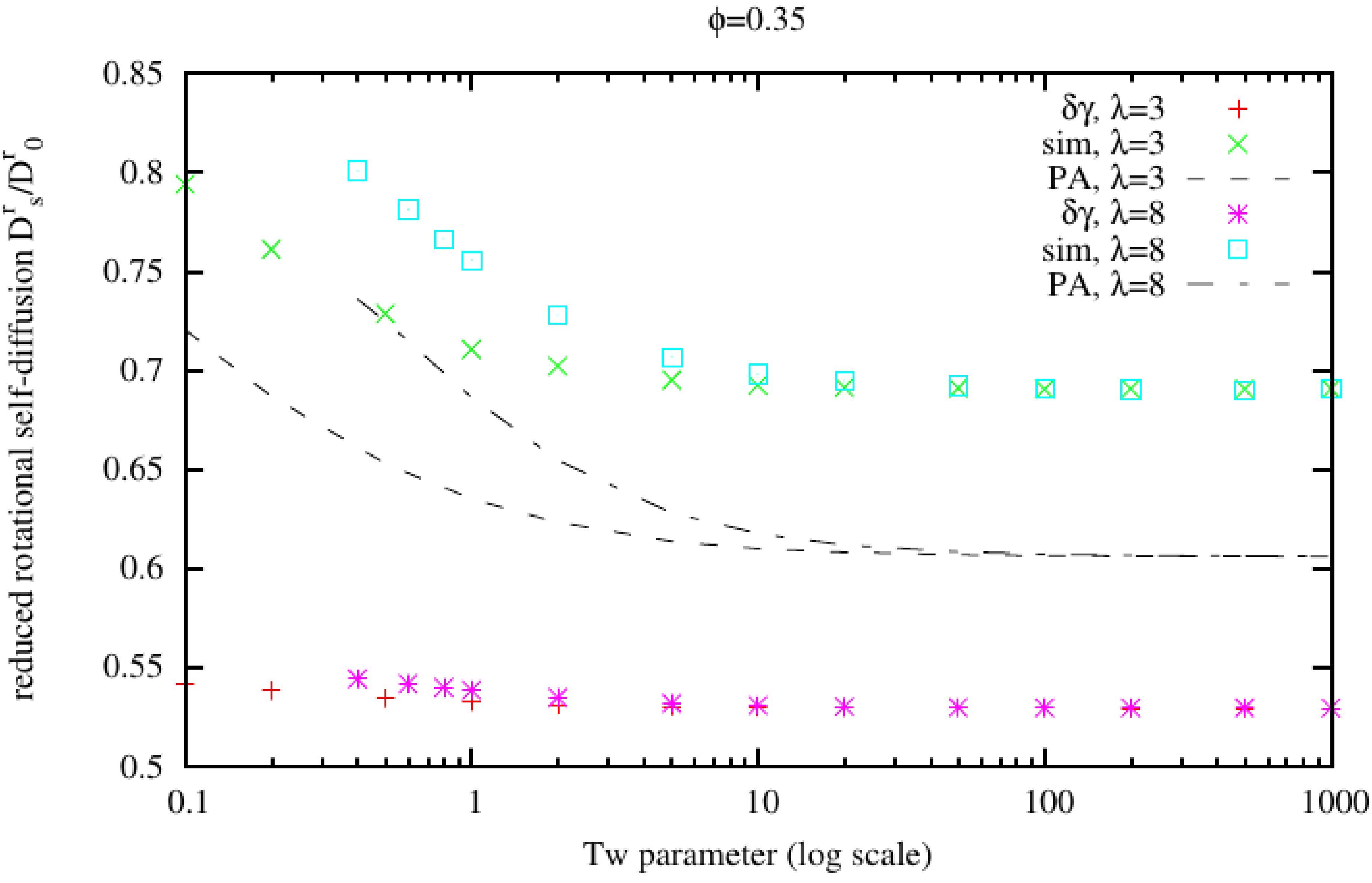
Radial distribution function calculated by Rogers-Young scheme
(results similar to Monte Carlo calculations)

Repulsion decreases number of close pairs in the system

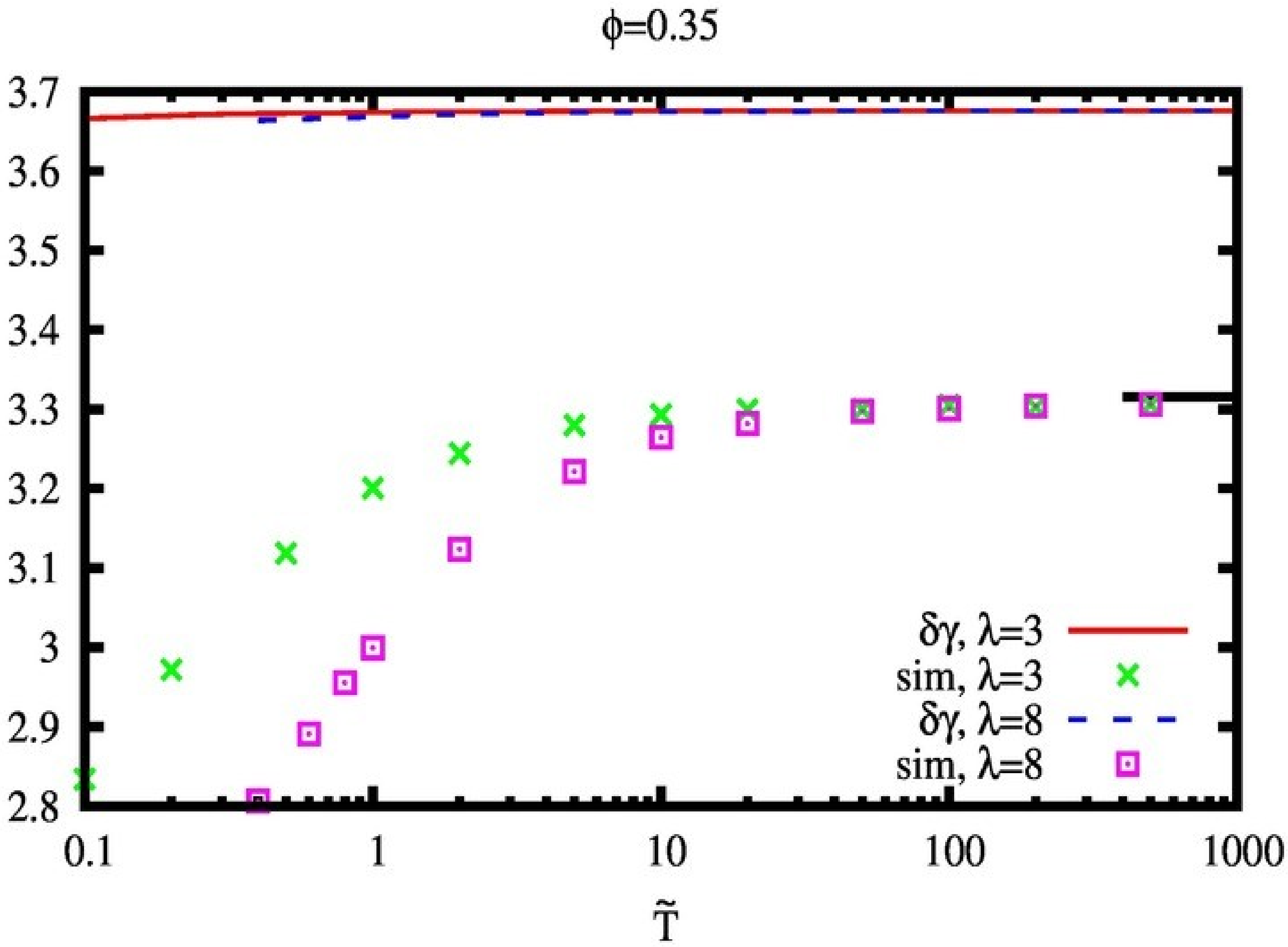


$$\phi = 0.25 \quad \lambda = 8 \quad \tilde{T} = 0.4, 1, 5, 1000$$

Rotational self-diffusion coefficient for repulsive particles by Beenakker-Mazur method



Effective viscosity coefficient for repulsive particles by Beenakker-Mazur method



Summary

- Results of BM qualitatively agrees with results of numerical simulations
- Weak dependence of BM on structure of suspension

Ongoing research in colaboration with:

Gerhard Nägele
Research Centre Jülich

Gustavo Abade
Universität Konstanz

Marco Heinen
Caltech

Important contribution: Eligiusz Wajnryb
Polish Academy of Sciences

Future perspectivies for Beenakker-Mazur scheme

In second order BM approach transport properties are given in terms of the following expansion*:*

$$\left\langle \mathcal{M} + \mathcal{M} G_{\langle \mathcal{M}_R \rangle} \mathcal{M}_R + \mathcal{M} G_{\langle \mathcal{M}_R \rangle} \left(\mathcal{M}_R - \langle \mathcal{M}_R \rangle \right) \tilde{G}_{\langle \mathcal{M}_R \rangle} \mathcal{M}_R \right\rangle$$

Straightforward generalization to*:*

-different spherical particles (permeable, mixed slip-stick b.c.)
-polidisperse systems
-nonspherical particles
-friction problem (chemical reactions?)

BM approach more sensitive to change of type of particles than to change of structure (rdf)?

Treloar, Masters (1989)

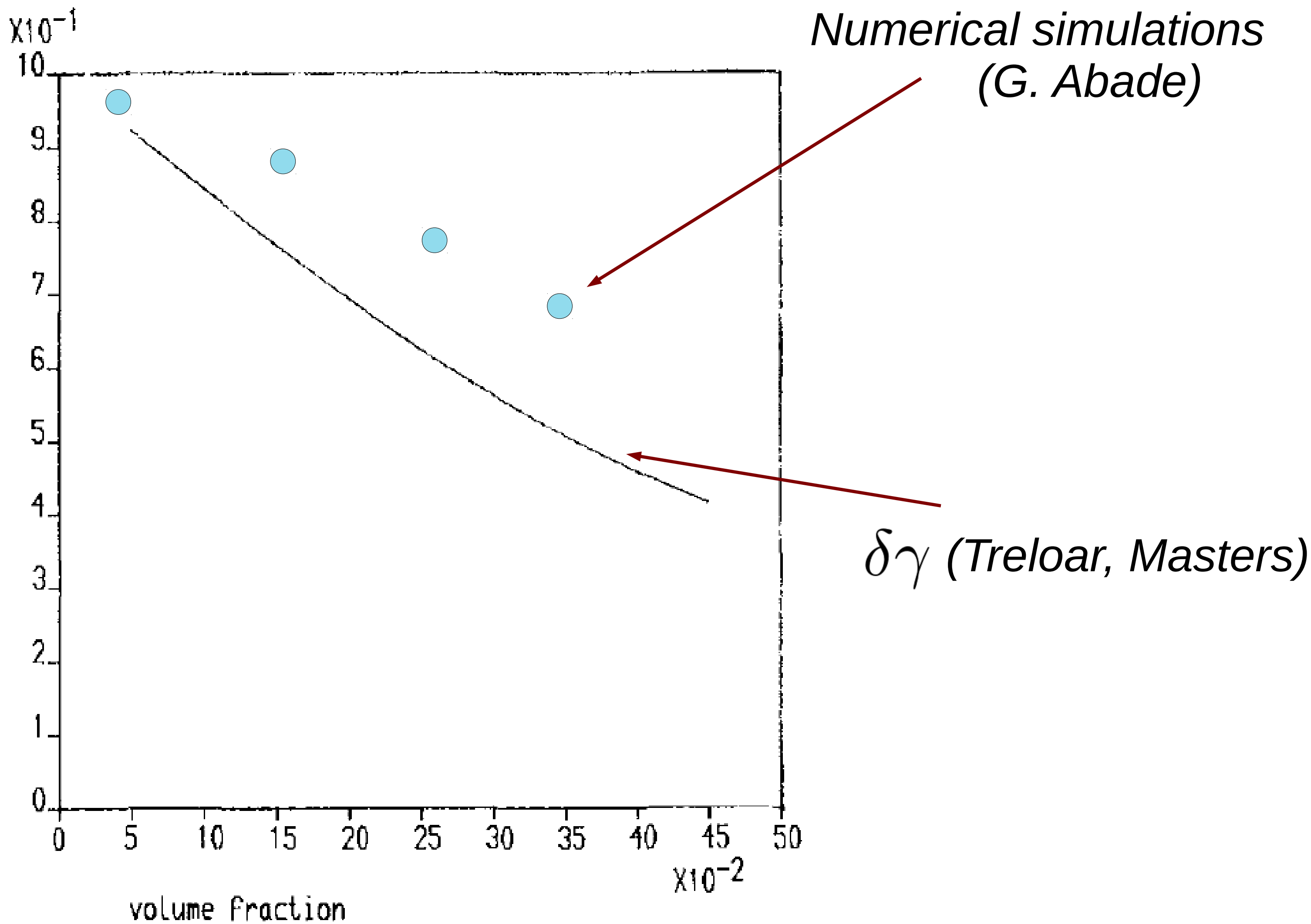


Figure 1. A plot of the normalized single particle rotational mobility, μ_S^{RR}/μ_0^{RR}, calculated to second order in the $\delta\gamma$-expansion, against volume fraction, ϕ.