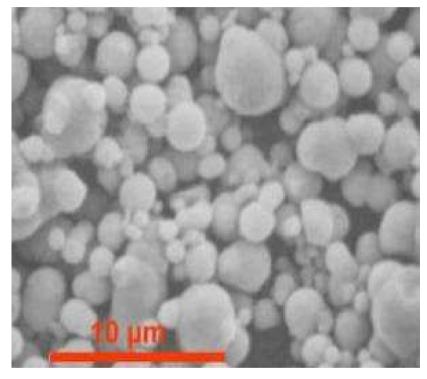
Short-time transport properties of suspensions of spherical particles

Karol Makuch



Introduction - suspensions

minute particles in liquid



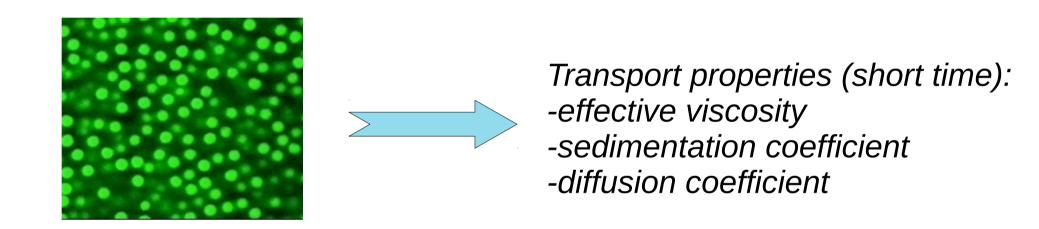
Liquid:	
-temperature	T
-viscosity	μ
-density of the fluid	$ ho_f$

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

milk, blood,...

Goal of our work

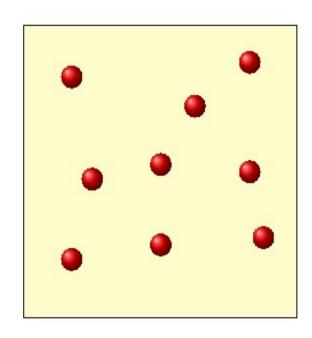
Monodisperse suspension of spherical particles



Over 100 years of research - still an open question

Hard-sphere suspension

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



Stokes equations:

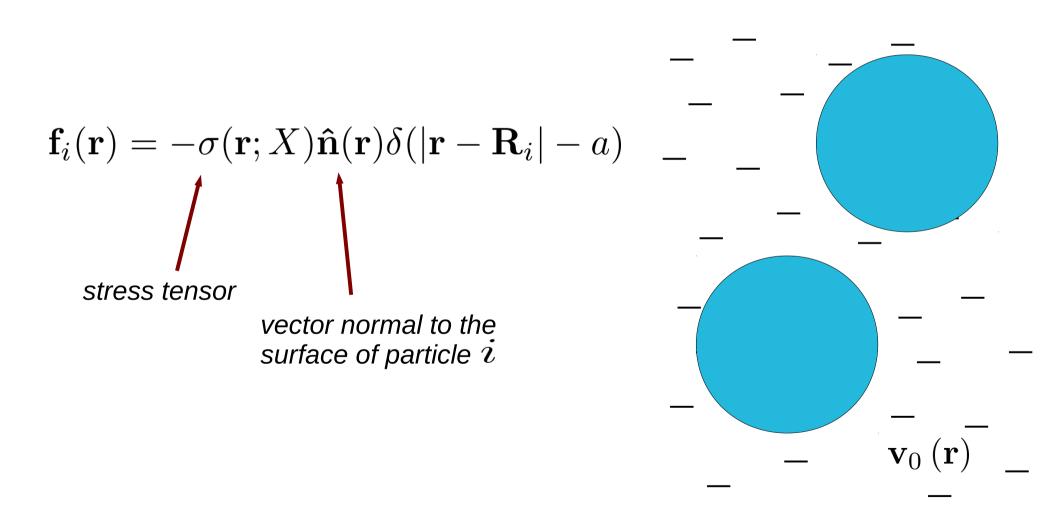
$$\nabla p(\mathbf{r}) - \eta \Delta \mathbf{v}(\mathbf{r}) = 0$$
$$\nabla \cdot \mathbf{v}(\mathbf{r}) = 0$$

Stick boundary conditions,

$${f v}({f r})
ightarrow {f v}_0({f r})$$
 for $r
ightarrow \infty$

Effective viscosity

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



Scattering series

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

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

$$i$$

$$j$$

$$k = i$$

suspension <=> dielectrics <=> other systems

Transport properties – history and scattering series

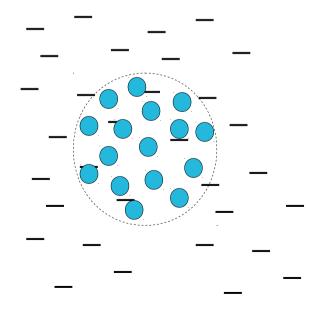


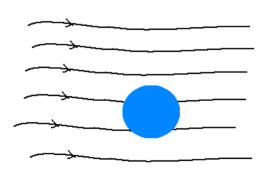
Einstein 1905 (corrected):

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

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

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

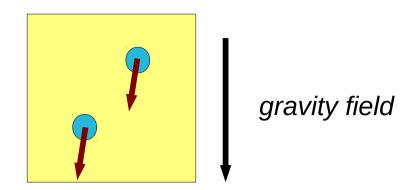




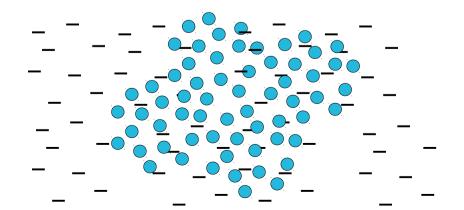
- •Finite system
- Hydrodynamic interactions neglected (no reflections, single particle)

Hydrodynamic interactions – Smoluchowski (1911)





$$\mathbf{U}_1 \leftarrow \bigcirc + \bigcirc + \bigcirc \bigcirc + \bigcirc \bigcirc \bigcirc \bigcirc$$



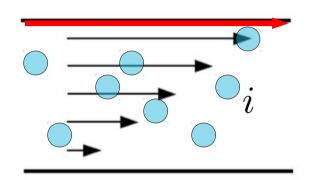
$$\int d^3r |\mathbf{G}(\mathbf{r})| = \infty$$
$$\mathbf{G}(\mathbf{r}) = \frac{1}{8\pi\eta} \frac{\mathbf{1} + \hat{\mathbf{r}}\hat{\mathbf{r}}}{r}$$

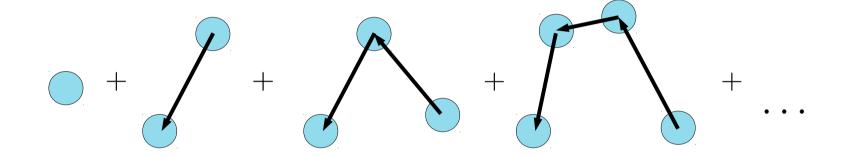
Well defined expression for effective viscosity?

Beyond diluted suspensions

Saito (1950):

-extension of Einstein work on mean-field level





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

vanishes when two particles overlap

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

Two-particle hydrodynamic interactions (1963)

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

$$+ \qquad + \qquad + \dots$$

absolute convergence

Peterson, Fixman (1963): $a_2 \approx 4.31784$

Batchelor, Green (1972): $a_2 pprox 5.2$

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

(well defined expression on two-body level)

(ad hoc renormalization)

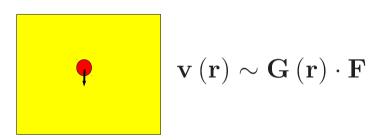
Problem with long-range HI still not solved

Hydrodynamic interactions

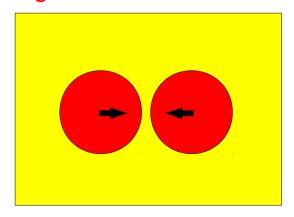
Many-body character

two-body approximation relevant for volume fractions less than about 5%

Long-range character



Strong interactions of close particles

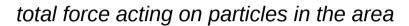


For constant velocities asymptotically infinite drag force (Jeffrey, Onishi (1984))

Effective Green function

- includes all three features of hydrodynamic interactions

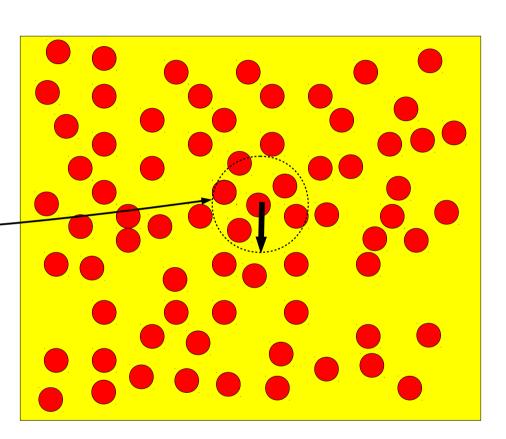
Flow caused by force acting on particles in the area



$$\mathbf{v}\left(\mathbf{r}
ight)\sim\mathbf{G}_{\mathrm{eff}}\left(\mathbf{r}
ight)\mathbf{F}$$

effective Green function (effective propagator):

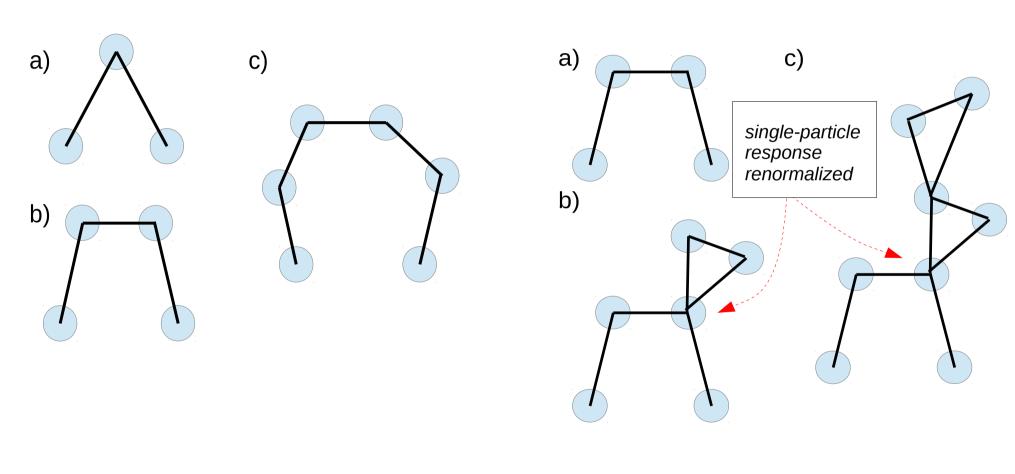
$$\mathbf{G}_{\mathrm{eff}}(\mathbf{r}) \sim rac{1}{8\pi\eta_{\mathrm{eff}}}rac{\mathbf{1}+\mathbf{\hat{r}}\mathbf{\hat{r}}}{r} = rac{\eta}{\eta_{\mathrm{eff}}}\mathbf{G}(\mathbf{r})$$
 at the distance



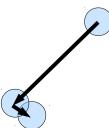


Beenakker-Mazur method (1983)

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



No correlations in position between particles in the above resummed terms



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

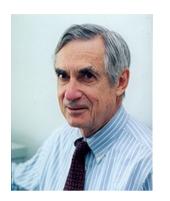
- ✓ Many-body character
- ✓ Long-range character
- X Strong interactions of close particles

No satisfactory statistical physics method including the above three features

Lubrication important!

1982 – problem of long-range HI solved







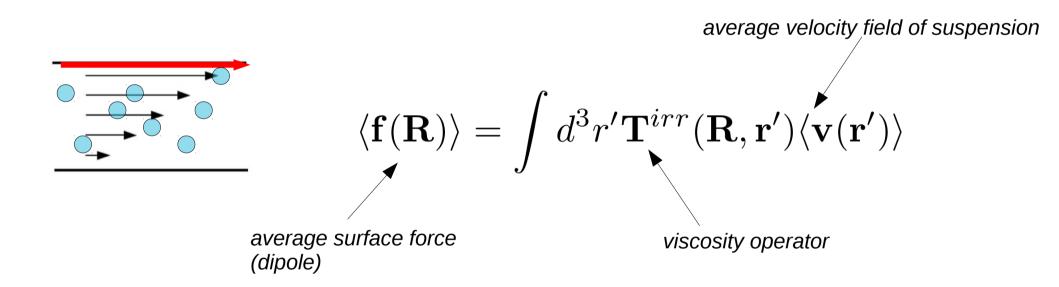
B. U. Felderhof, G. W. Ford, and E. G. D. Cohen³

Received August 24, 1981

We derive a cluster expansion for the electric susceptibility kernel of a dielectric suspension of spherically symmetric inclusions in a uniform background. This also leads to a cluster expansion for the effective dielectric constant. It is shown that the cluster integrals of any order are absolutely convergent, so that the dielectric constant is well defined and independent of the shape of the sample in the limit of a large system. We compare with virial expansions derived earlier in

Response of suspension (effective viscosity)

Viscosity by relation between pressure tensor and average flow of suspension (Landau):



Effective viscosity coefficient is given directly by the response operator T^{irr}

Felderhof, Ford, Cohen – cluster expansion (1982)

$$T^{irr} = \sum_{g=1}^{\infty} \sum_{C_1...C_g} \int dC_1 \ldots dC_g b(C_1|\ldots|C_g) S_I(C_1) \mathbf{G} \ldots \mathbf{G} S_I(C_g)$$
 Oseen tensor: $G = \frac{1}{8\pi\eta} \frac{\mathbf{1} + \hat{\mathbf{r}}\hat{\mathbf{r}}}{r}$

block distribution function (configurations of particles)

Example of scattering sequence (many-body):

$$M(1)GM(3)GM(2)GM(1) \times$$

 $G \times M(4)GM(5)GM(4) \times G \times M(6)$

short range hydrodynamic interactions (strong interactions of close particles)

long range hydrodynamic interactions

$$S_I(123)GS_I(45)GS_I(6)$$

$$C_1 \equiv 123$$
 $C_2 \equiv 45$ $C_3 \equiv 6$

Felderhof, Ford, Cohen – microscopic explanation of Clausius-Mossotti (Saito) formula (1983)

The following definition

$$\mathbf{T}_{CM}^{irr} = \mathbf{T}^{irr} \left(1 + [h\mathbf{G}] \mathbf{T}^{irr} \right)^{-1}$$

and approximate closure relation

$$\mathbf{T}_{CM}^{irr} \approx n_1 \mathbf{\hat{M}}$$

lead to Saito formula

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

Our approach – renormalization of the propagator

Cluster expansion (1982):

 $T^{irr} = \sum_{g=1}^{\infty} \sum_{C_1...C_g} \int dC_1...dC_g b(C_1|...|C_g) S_I(C_1) \mathbf{G}...\mathbf{G}S_I(C_g)$

block distribution function (configurations of particles)

Oseen tensor (pure liquid):

short-range hydrodynamic interaction

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

Ring expansion (2015):

$$T^{irr} = \sum_{g=1}^{\infty} \sum_{C_1 \dots C_g} \int dC_1 \dots dC_g H(C_1|\dots|C_g) S_I(C_1) \mathbf{G}_{\text{eff}} \dots \mathbf{G}_{\text{eff}} S_I(C_g)$$

block correlation function (configurations of particles); H=b for g=1,2, H different from b for g>2. Effective Green function:

$$\mathbf{G}_{\mathrm{eff}}(\mathbf{r}) \sim rac{\eta}{\eta_{\mathrm{eff}}} \mathbf{G}(\mathbf{r})$$

convergence

Generalization of Clausius-Mossotti approximation

Constructing approximate method by carrying over approximation from cluster expansion to ring expansion with the following modification:

$$G \Longrightarrow G_{\text{eff}}$$

Clausius-Mossotti (Saito) approximation

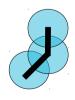


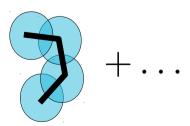
Generalized Clausius-Mossotti approximation

(two-body hydrodynamic interactions incomplete – the same as in Beenakker and Mazur scheme)









Volume fraction, two-body correlation function \Longrightarrow

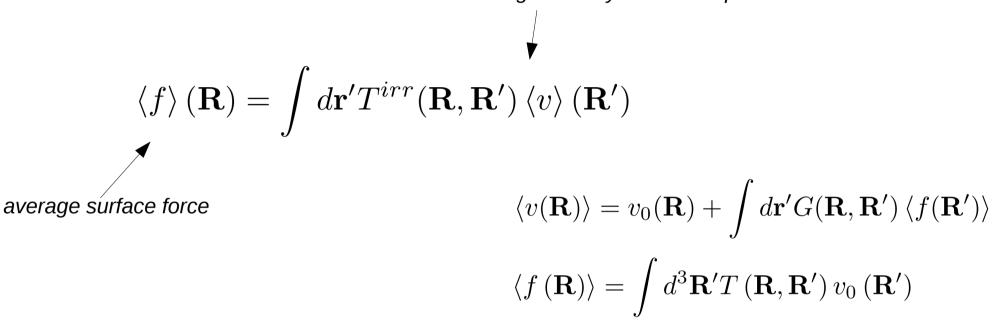


transport properties

Felderhof, Ford and Cohen cluster expansion (1982)

(effective viscosity)

average velocity field of suspension



Relation between T and T^{irr} operators:

$$T = T^{irr} \left(1 - GT^{irr} \right)^{-1}$$

Effective viscosity coefficient is given directly by the response operator $\,T^{\imath r r}$

Felderhof, Ford and Cohen cluster expansion (1982)

$$T = \sum_{b=1}^{\infty} \sum_{C_1...C_b} \int dC_1...dC_b \ n(C_1...C_b) S_I(C_1)G...GS_I(C_b)$$

$$T = T^{irr} \left(1 - GT^{irr} \right)^{-1}$$

$$S_I(C_1)$$
— $S_I(C_2)$ — $S_I(C_3)$ —... — $S_I(C_b)$

 $n\left(C_1C_2C_3\dots C_b\right)$

Diagrammatic approach...

$$S_I(C_1)$$
— $S_I(C_2)$ — $S_I(C_3)$ —... — $S_I(C_b)$

$$n(C_1C_2C_3...C_b)$$

Definition of correlation functions g (between groups of particles):

$$n(C_1) = g(C_1)$$

$$n(C_1C_2) = g(C_1)g(C_2) + g(C_1|C_2)$$

$$n(C_1C_2C_3) = g(C_1)g(C_2)g(C_3) + g(C_1|C_2)g(C_3) + g(C_1|C_3)g(C_2) + g(C_1)g(C_2|C_3) + g(C_2|C_2|C_3)$$

$$S_I(C_1)$$
 $S_I(C_2)$ $S_I(C_3)$ \ldots $S_I(C_b)$ $S_I(C_1)$

Diagrammatic representation of correlation functions:

$$n(C_1) = g(C_1)$$

$$n(C_1) = {lackbox{}^{C_1}}$$

$$S_I(C_1)$$
— $S_I(C_2)$ — $S_I(C_3)$ —... — $S_I(C_b)$

$$n(C_1C_2C_3...C_b)$$

Diagrammatic representation of correlation functions:

$$n(C_1C_2) = g(C_1)g(C_2) + g(C_1|C_2)$$

$$n(C_1C_2) = {}^{C_1} {}^{C_2} {}^{C_2} + {}^{C_1} {}^{C_2}$$

$$S_I(C_1)$$
— $S_I(C_2)$ — $S_I(C_3)$ —... — $S_I(C_b)$

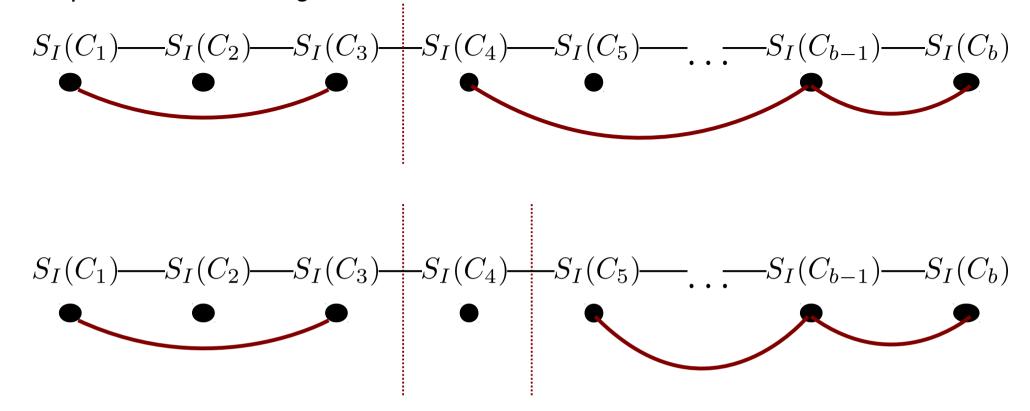
$$n(C_1C_2C_3...C_b)$$

Diagrammatic representation of correlation functions:

$$n(C_1C_2C_3) = g(C_1)g(C_2)g(C_3) + g(C_1|C_2)g(C_3) + g(C_1|C_3)g(C_2) + g(C_1)g(C_2|C_3) + g(C_1|C_2|C_3)$$

$$+g(C_1|C_2|C_3)$$

Example of reducible diagrams:



Example of irreducible diagram:

$$S_I(C_1)$$
— $S_I(C_2)$ — $S_I(C_3)$ — $S_I(C_4)$ — $S_I(C_5)$ —...— $S_I(C_{b-1})$ — $S_I(C_b)$

$$T = T^{irr} + T^{irr}GT$$

Felderhof, Ford and Cohen cluster expansion (1982)

$$T^{irr} = \sum_{g=1}^{\infty} \sum_{C_1...C_g} \int dC_1...dC_g b(C_1|...|C_g) S_I(C_1) G...GS_I(C_g)$$

Block distribution functions (in diagrammatic language):

$$b(C_1|\ldots|C_g) = \text{ All terms from } n(C_1\ldots C_g) \text{ giving irreducible diagrams for sequence } C_1|\ldots|C_g$$

Block distribution functions (recurrence formula):

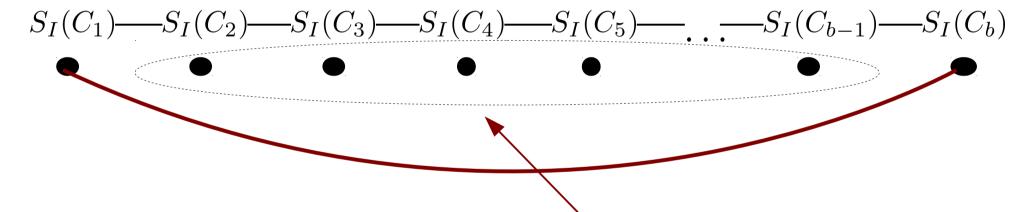
$$b(C) = n(C)$$

$$b(C_1| \dots |C_k|C_{k+1}| \dots |C_g) = b(C_1| \dots |C_kC_{k+1}| \dots |C_g)$$

$$-b(C_1| \dots |C_k)b(C_{k+1}| \dots |C_g)$$

Additional resummation of FFC expansion (2015)

$$T^{irr} = \sum_{g=1}^{\infty} \sum_{C_1...C_g} \int dC_1 ... dC_g b(C_1|...|C_g) S_I(C_1) G ... GS_I(C_g)$$



all correlations are possible here, they yield

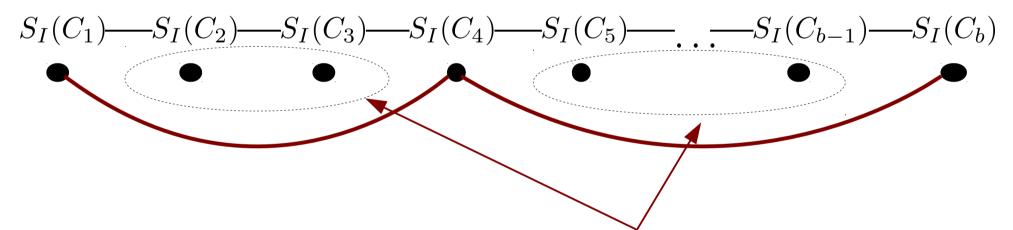
$$n(C_2 \dots C_{b-1})$$

and after resummation of scattering sequences and integration:

$$G_{eff} = G + GTG$$

Additional resummation of FFC expansion (2015)

$$T^{irr} = \sum_{g=1}^{\infty} \sum_{C_1...C_g} \int dC_1 ... dC_g b(C_1|...|C_g) S_I(C_1) G ... GS_I(C_g)$$



all correlations are possible here, they yield

$$n(C_2C_3)n(C_5\dots C_{b-1})$$

and after resummation of scattering sequences and integration:

$$G_{eff} = G + GTG$$

$$T^{irr} = \sum_{b=1}^{\infty} \sum_{C_1...C_b} \int dC_1...dC_b H(C_1|...|C_b) S_I(C_1) G_{\text{eff}}...G_{\text{eff}} S_I(C_b)$$

Block correlation functions (in diagrammatic language):

$$H(C_1|\ldots|C_g) =$$
 All chains from $n(C_1\ldots C_g)$

Chains – all terms from $n(C_1 \dots C_g)$ which connect all points (also through intersection), e.g.

$$H(C_1) = {\overset{C_1}{\bullet}}$$

$$H(C_1|C_2) = C_1 C_2$$

$$H(C_1|C_2|C_3) = C_1 C_2 C_3$$

$$T^{irr} = \sum_{b=1}^{\infty} \sum_{C_1...C_b} \int dC_1...dC_b H(C_1|...|C_b) S_I(C_1) G_{\text{eff}}...G_{\text{eff}} S_I(C_b)$$

Block correlation functions (in diagrammatic language):

$$H(C_1|\ldots|C_g)=$$
 All chains from $n(C_1\ldots C_g)$

Chains – all terms from $n(C_1 \dots C_g)$ which connect all points (also through intersection), e.g.

$$H(C_1|C_2|C_3|C_4) = \begin{pmatrix} C_1 & C_2 & C_3 & C_4 \\ & & & & & \\ & & & & & \\ & & & & & \\ & & & & & \\ & & & & & \\ & & & & & \\ & & & & & \\ & & & & & \\ & & & & & \\ & & & & & \\ & & & & & \\ & & & & \\ & & & & & \\ & & & & & \\ & & & & & \\ & & & & & \\ & & & & & \\ & & & & & \\ & & & & & \\ & & & & & \\ & & & & & \\ & & & & & \\ & & & & & \\ & & & & & \\ & & & & \\ & & & & & \\ & & & & \\ & & & & \\ & & & & \\ & & & & \\ & & & & \\ & & & & \\ & & & & \\ & & & & \\ & & & & \\$$

$$T^{irr} = \sum_{b=1}^{\infty} \sum_{C_1...C_b} \int dC_1...dC_b H(C_1|...|C_b) S_I(C_1) G_{\text{eff}}...G_{\text{eff}} S_I(C_b)$$

Block correlation functions (in diagrammatic language):

$$H(C_1|\ldots|C_g) =$$
 All chains from $n(C_1\ldots C_g)$

Block correlation functions (recurrence formula):

$$b(C_1|...|C_b) = \sum_{r=1}^{b-1} \sum_{1=i_1 < i_2 < ... < i_{r+1} = b} H(C_{i_1}|...|C_{i_{r+1}}) \times n(\{C_{i_1}...C_{i_2}\} \setminus \{C_{i_1}C_{i_2}\}) ... n(\{C_{i_r}...C_{i_{r+1}}\} \setminus \{C_{i_r}C_{i_{r+1}}\})$$

Comparison of ring expansion with cluster expansion

Felderhof, Ford, Cohen:
$$T^{irr} = \sum_{b=1}^{\infty} \sum_{C_1 \dots C_b} \int dC_1 \dots dC_b b(C_1|\dots|C_b) S_I(C_1) G \dots GS_I(C_b)$$

Ring expansion (2015)

$$T^{irr} = \sum_{b=1}^{\infty} \sum_{C_1...C_b} \int dC_1...dC_b H(C_1|...|C_b) S_I(C_1) G_{\text{eff}}...G_{\text{eff}} S_I(C_b)$$

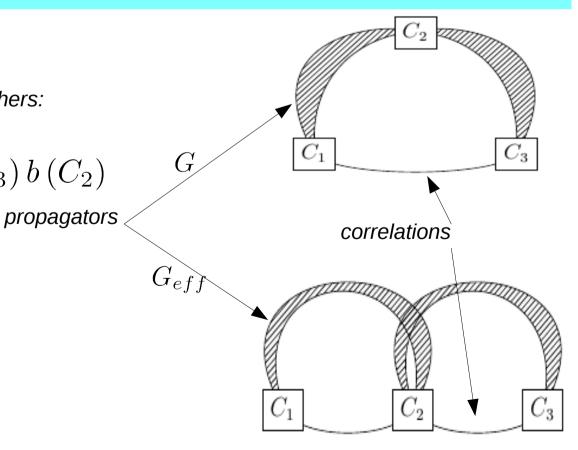
When the middle group goes away from others:

$$b(C_1|C_2|C_3) \longrightarrow b(C_1|C_3)b(C_2)$$

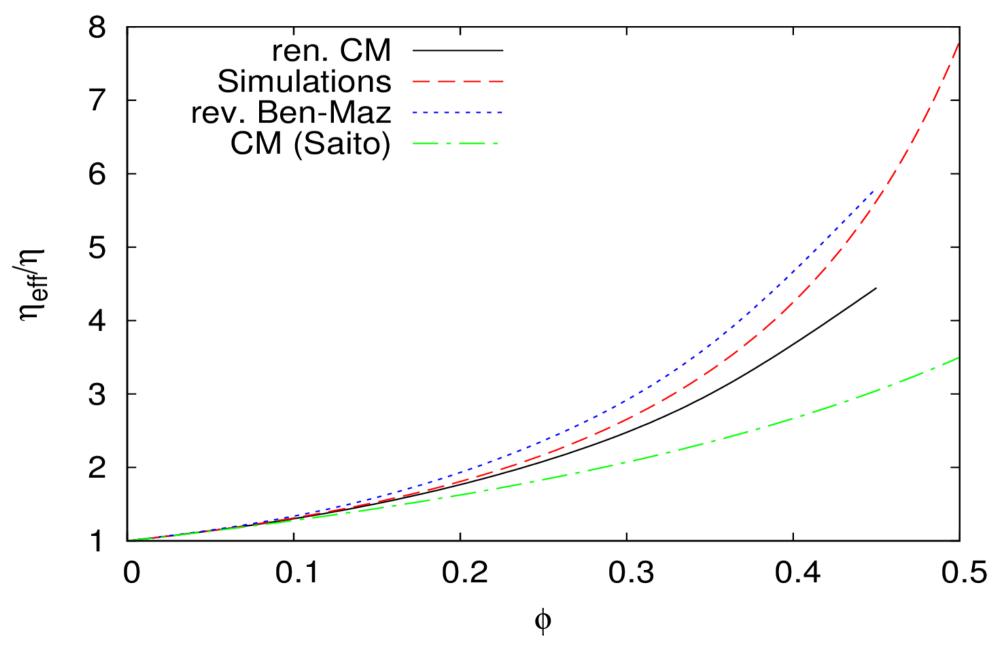
$$H\left(C_1|C_2|C_3\right)\longrightarrow 0$$

Two important differences:

- -propagator
- -volume of integration



Effective viscosity



K.M. Phys. Rev. E, accepted

Summary

- •Long-range, many-body hydrodynamic interactions and strong interactions of close particles are important in suspensions (still an open problem)
- •Rigorous ring expansion can grasp all of the above features (opposite to δy)
- •Generalized Clausius-Mossotti approximation (two-body hydrodynamic interactions not fully taken; comparable to $\delta \gamma$ scheme)



Under suspervision (in years 2005-2011) of Bogdan Cichocki, Univeristy of Warsaw

Outlook

•one-ring approximation (full two-body hydrodynamic interactions, much better accuracy than hitherto theoretical methods in comparison to numerical simulations for volume fraction less than 35%)

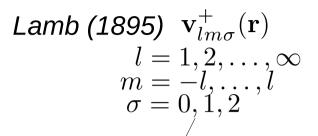
•straightforward generalization for different suspensions of spherical particles (droplets, spherical polymers) with different distributions (charged particles)

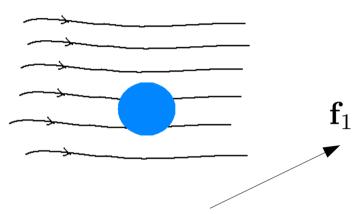
polidisperse suspensions

suspension of nonspherical particles (e.g. double sphere)

Single particle

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





$$\mathbf{f}_{1}(\mathbf{r}) = \int d\mathbf{r}' \mathbf{M} \left(\mathbf{r} - \mathbf{R}_{1}, \mathbf{r}' - \mathbf{R}_{1}\right) \mathbf{v}_{0}(\mathbf{r}')$$

Single particle operator (Felderhof 1976)

Surface force density
(Cox Brenner (1967): M

(Cox Brenner (1967); Mazur, Bedeaux (1974))

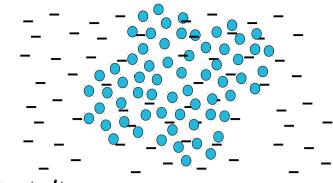
$$\mathbf{v}(\mathbf{r}) = \mathbf{v}_0(\mathbf{r}) + \int d^3 \mathbf{r}' \mathbf{G}(\mathbf{r} - \mathbf{r}') \cdot \mathbf{f}_1(\mathbf{r}')$$

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

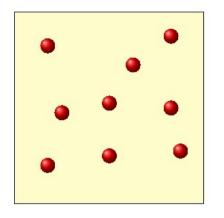
$$\mathbf{f}_i = \mathbf{M}(i) \left(\mathbf{v}_0 + \sum_{i \neq j} \mathbf{G} \mathbf{f}_j \right)$$

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

ambient flow



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

Single particle response operator

Example of scattering sequence (many-body): $M(1)GM(3)GM(2)GM(1) \times \\ G \times M(4)GM(5)GM(4) \times G \times M(6)$ $Short\ range\ hydrodynamic\ interactions\ (strong\ interactions\ of\ close\ particles)$

long range hydrodynamic interactions (nodal line)

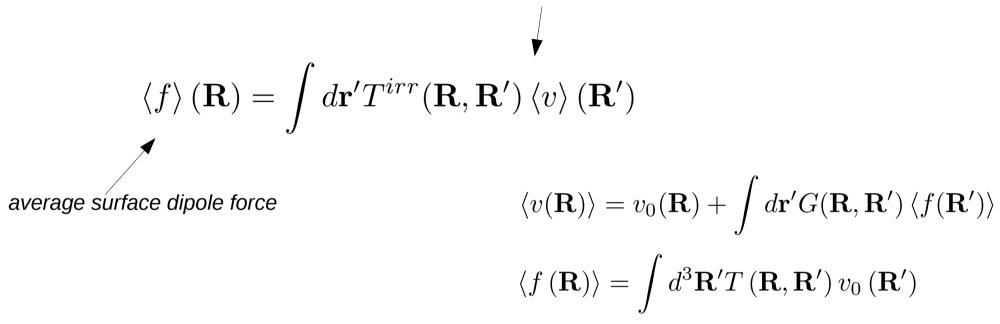
Green function for Stokes equations

block structure:
$$S_I(C_1)$$
— $S_I(C_2)$ — $S_I(C_3)$
 $C_1 \equiv 123$ $C_2 \equiv 45$ $C_3 \equiv 6$

Response of suspension

(effective viscosity)

average velocity field of suspension



Relation between T and T^{irr} operators:

$$T = T^{irr} \left(1 - GT^{irr} \right)^{-1}$$

Effective viscosity coefficient is given directly by the response operator $\,T^{\imath r r}$

Macroscopic description

Average force density:

$$\langle f(\mathbf{R}) \rangle \equiv \left\langle \sum_{i} f_{i} \delta(\mathbf{R} - i) \right\rangle$$

Average over probability distribution for configurations of particles, thermodynamic limit

$$\langle f(\mathbf{R}) \rangle = \int d^3 \mathbf{R}' T(\mathbf{R}, \mathbf{R}') v_0(\mathbf{R}')$$

Response operator for suspension in ambient flow

$$T = \sum_{b=1}^{\infty} \sum_{C_1 \dots C_b} \int dC_1 \dots dC_b \, n \, (C_1 \dots C_b) \, S_I(C_1) G \dots GS_I(C_b)$$

s-particle distribution functions