Hydrodynamic function and effective transport coefficients for suspensions of spherical particles II

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INNOVATIVE ECONOMY
NATIONAL COHESION STRATEGY

FNP
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EUROPEAN UNION
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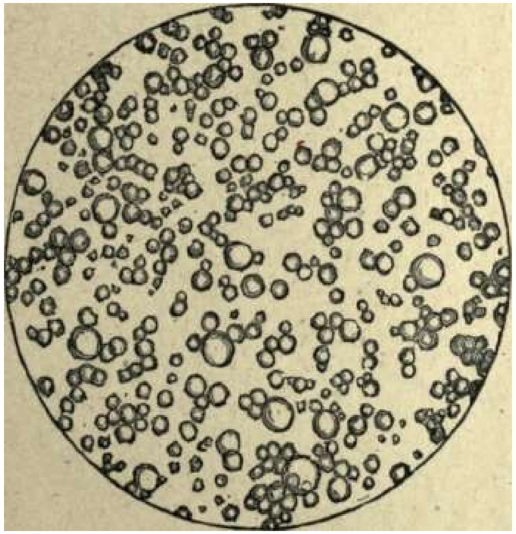
Suspensions



Macro:

Effective viscosity
Sedimentation coefficient
Hydrodynamic factor

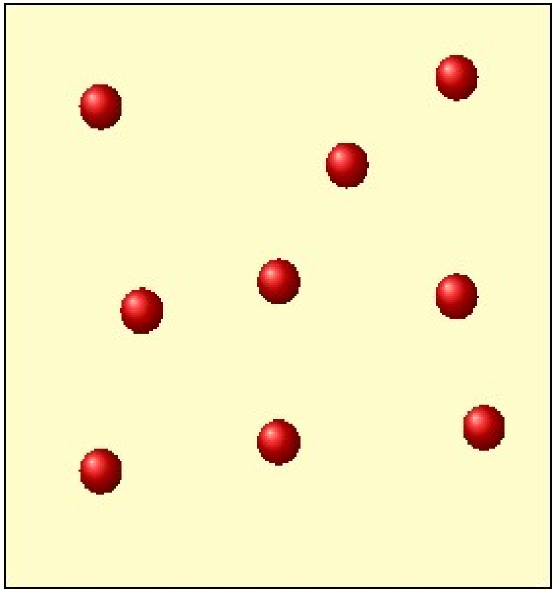
Micro:

Radius of particles
Viscosity of fluid
Number density of particles

“The simplest” system:
Suspension of spherical particles (hard spheres)
Problem: from micro to macro

Hard-sphere suspension – microscopic description

Unbounded liquid,
N particles



Force densities acting on suspension: $\mathbf{f}_0(\mathbf{r})$ (fluid), $\mathbf{f}_{\text{part}}(\mathbf{r})$ (particles)

Response of suspension: $\mathbf{V}_i, \quad \mathbf{\Omega}_i, \quad \sum_{i=1}^{N} \mathbf{f}_i(\mathbf{r})$

translational and angular velocity

surface forces

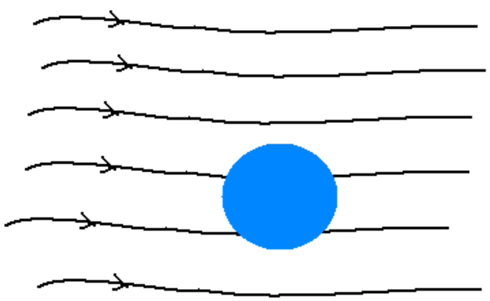
Stokes equations:

$$\begin{aligned} \nabla p(\mathbf{r}) - \eta \Delta \mathbf{v}(\mathbf{r}) &= \mathbf{f}_0(\mathbf{r}) + \sum_{i=1}^{N} \mathbf{f}_i(\mathbf{r}) \\ \nabla \cdot \mathbf{v}(\mathbf{r}) &= 0 \end{aligned}$$

Calculation of hydrodynamic interactions

Single particle in ambient flow

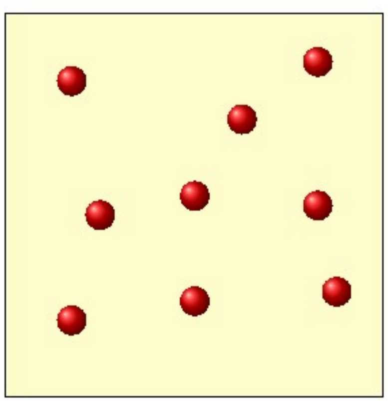
$$\mathbf{v}_0\left(\mathbf{r}\right) = \int d^3\mathbf{r}'\mathbf{G}\left(\mathbf{r}-\mathbf{r}'\right)\cdot\mathbf{f}_0\left(\mathbf{r}'\right)$$



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

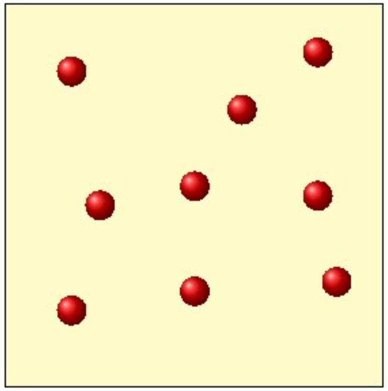
Single freely moving particle response operator

Suspension in ambient flow:



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

Scattering series



ambient flow

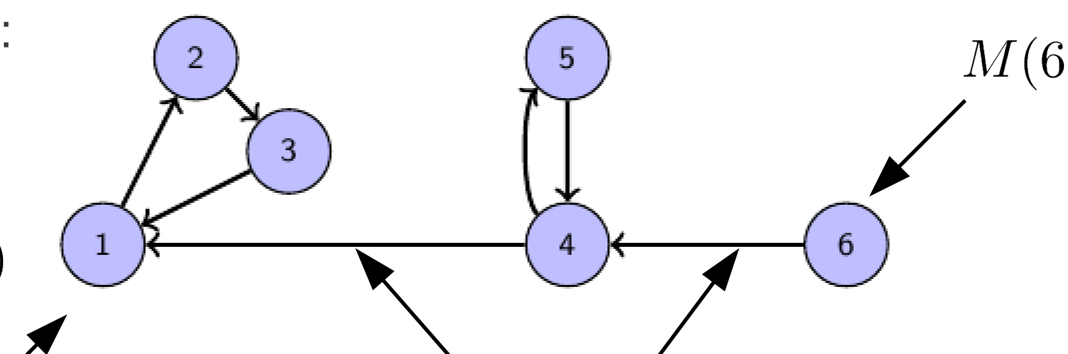
$$\mathbf{f}_i = \left(\mathbf{M}(i) + \sum_{j \neq i} \mathbf{M}(i)\mathbf{G}\mathbf{M}(j) + \sum_{j \neq i} \sum_{k \neq j} \mathbf{M}(i)\mathbf{G}\mathbf{M}(j)\mathbf{G}\mathbf{M}(k) + \dots \right) \mathbf{v}_0$$

Single particle response operator

Green function for Stokes equations

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)

block structure:

$$S_I(C_1) \text{———} S_I(C_2) \text{———} S_I(C_3)$$

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

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

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

Response operator for suspension in ambient flow

$$T = \sum_{b=1}^{\infty} \sum_{C_1 \ldots C_b} \int dC_1 \ldots dC_b \; n\left(C_1 \ldots C_b\right) S_I(C_1) G \ldots G S_I(C_b)$$

s-particle distribution functions

Response of suspension

(effective viscosity)

average velocity field of suspension

$$\langle f \rangle (\mathbf{R}) = \int d\mathbf{r}' T^{irr} (\mathbf{R}, \mathbf{R}') \langle v \rangle (\mathbf{R}')$$

average surface dipole force

$$\langle v(\mathbf{R}) \rangle = v_0(\mathbf{R}) + \int d\mathbf{r}' G(\mathbf{R}, \mathbf{R}') \langle f(\mathbf{R}') \rangle$$

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

Relation between T and T^{irr} operators:

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

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

Derivation of microscopic expression for Tirr

$$T = \sum_{b=1}^{\infty} \sum_{C_1 \ldots C_b} \int dC_1 \ldots dC_b \; n\left(C_1 \ldots C_b\right) S_I(C_1) G \ldots G S_I(C_b) \qquad T = T^{irr}\left(1 - G T^{irr}\right)^{-1}$$

$$S_I(C_1) \text{———} S_I(C_2) \text{———} S_I(C_3) \text{———} \ \ldots \ \text{———} S_I(C_b)$$

$$n\left(C_1 C_2 C_3 \ldots C_b\right)$$

Diagrammatic approach...

$$S_I(C_1) \text{———} S_I(C_2) \text{———} S_I(C_3) \text{———} \ \dots \ \text{———} S_I(C_b)$$

$$n\left(C_1 C_2 C_3 \dots C_b\right)$$

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

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

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

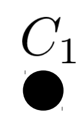
$$\begin{aligned} n(C_1 C_2 C_3) \quad = \quad & g(C_1)g(C_2)g(C_3) + g(C_1|C_2)g(C_3) \\ & + g(C_1|C_3)g(C_2) + g(C_1)g(C_2|C_3) \\ & + g(C_2|C_2|C_3) \end{aligned}$$

$$S_I(C_1) \text{———} S_I(C_2) \text{———} S_I(C_3) \text{———} \;\ldots\; \text{———} S_I(C_b)$$

$$n\left(C_1 C_2 C_3 \ldots C_b\right)$$

Diagrammatic representation of correlation functions:

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

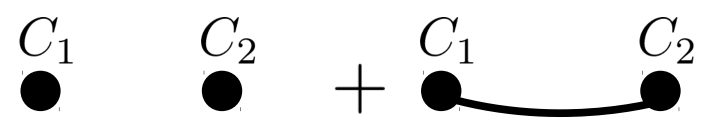
$$n(C_1) =$$ 

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

$$n\left(C_1 C_2 C_3 \ldots C_b\right)$$

Diagrammatic representation of correlation functions:

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

$n(C_1 C_2) =$ 

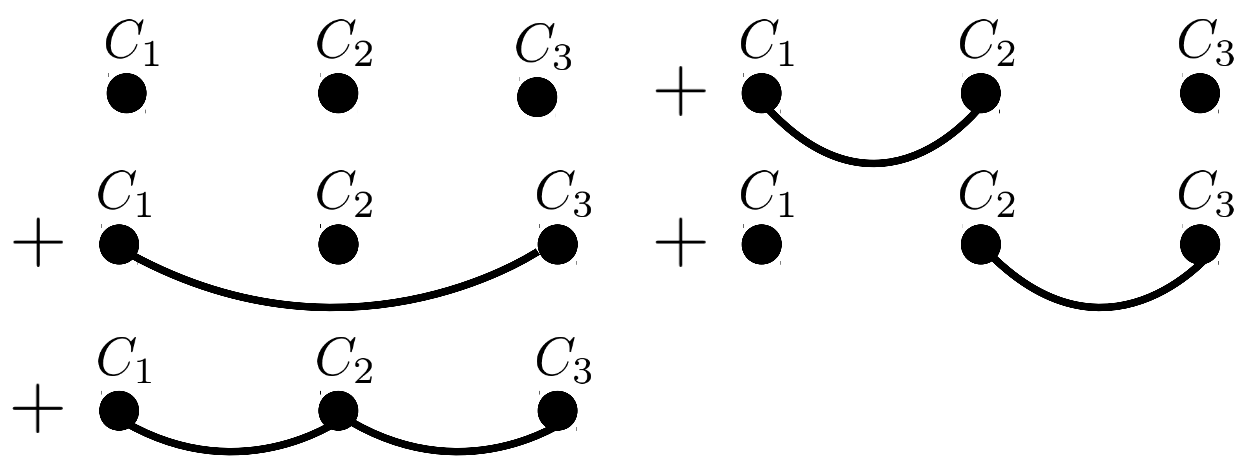
$$S_I(C_1) — S_I(C_2) — S_I(C_3) — \;\ldots\; — S_I(C_b)$$

$$n\,(C_1 C_2 C_3 \ldots C_b)$$

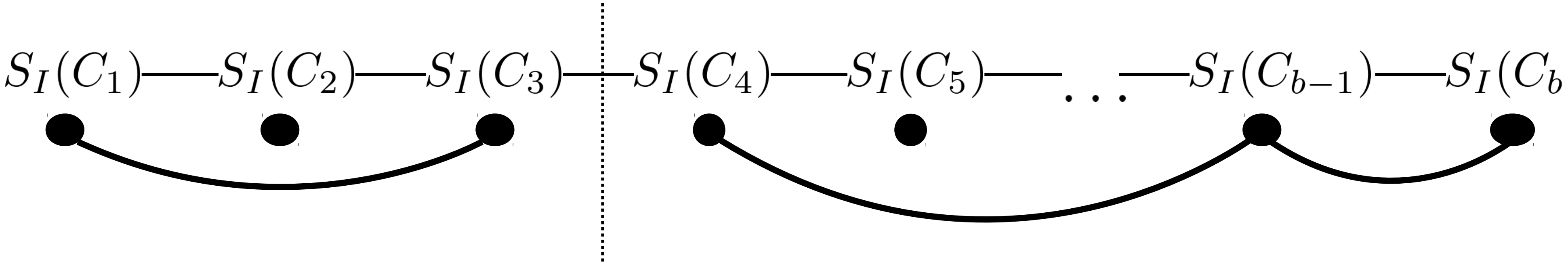
Diagrammatic representation of correlation functions:

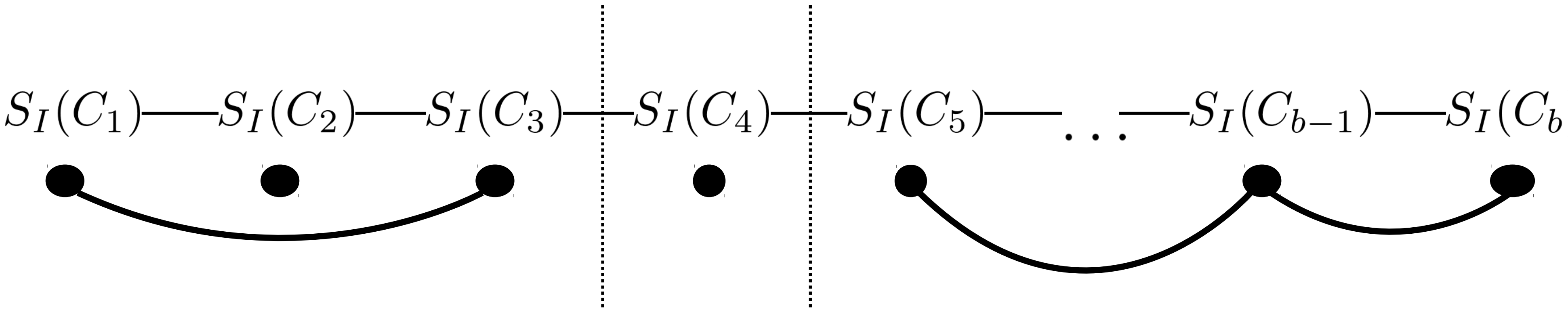
$$\begin{aligned} n(C_1C_2C_3) \quad &= \quad g(C_1)g(C_2)g(C_3) + g(C_1|C_2)g(C_3) \\ & \quad + g(C_1|C_3)g(C_2) + g(C_1)g(C_2|C_3) \\ & \quad + g(C_1|C_2|C_3) \end{aligned}$$

$$n(C_1C_2C_3) =$$

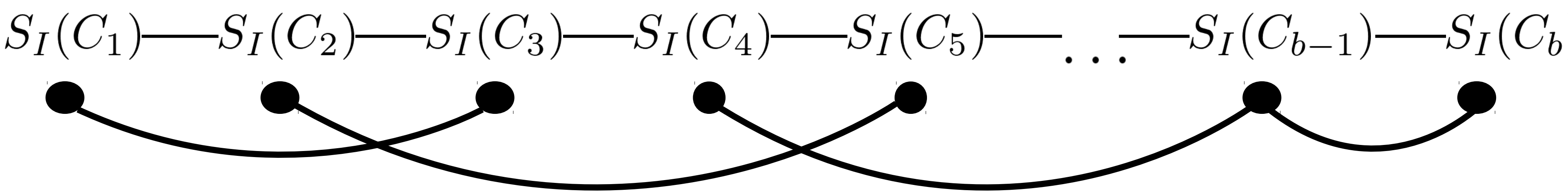


Example of reducible diagrams:





Example of irreducible diagram:



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

Cluster expansion (1982):

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

Block distribution functions (in diagrammatic language):

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

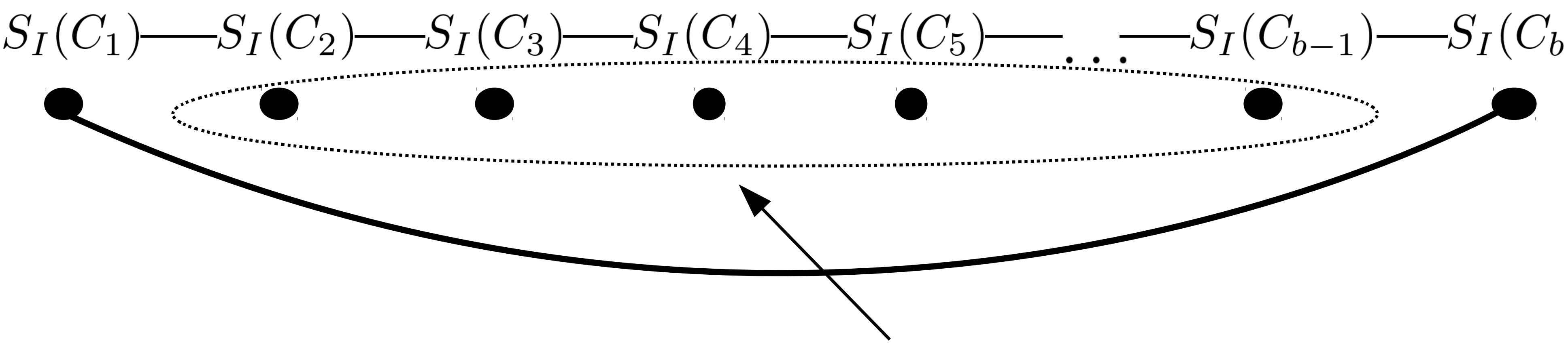
Block distribution functions (recurrence formula):

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

$$\begin{aligned} b(C_1| \ldots |C_k|C_{k+1}| \ldots |C_g) \quad &= \quad b(C_1| \ldots |C_k C_{k+1}| \ldots |C_g) \\ &\quad -b(C_1| \ldots |C_k) b(C_{k+1}| \ldots |C_g) \end{aligned}$$

Further analysis of Tirr (2011)

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



all correlations are possible here, they yield

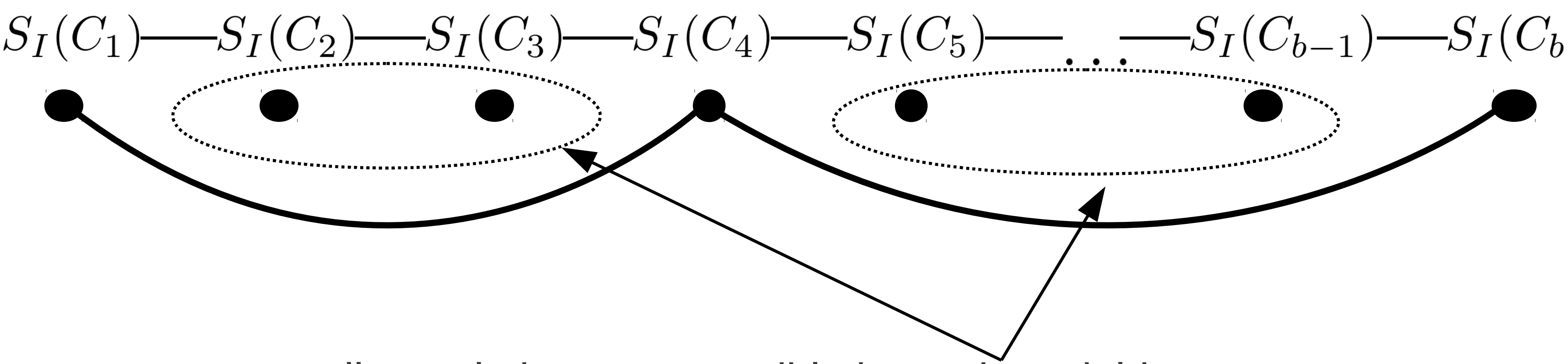
$$n(C_2 \ldots C_{b-1})$$

and after resummation of scattering sequences and integration:

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

Further analysis of Tirr (2011)

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



all correlations are possible here, they yield

$$n(C_2 C_3) n(C_5 \dots C_{b-1})$$

and after resummation of scattering sequences and integration:

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

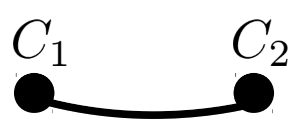
$$T^{irr} = \sum_{b=1}^{\infty} \sum_{C_1 \ldots C_b} \int dC_1 \ldots dC_b H(C_1 | \ldots | C_b) S_I(C_1) G_{\text{eff}} \ldots G_{\text{eff}} S_I(C_b)$$

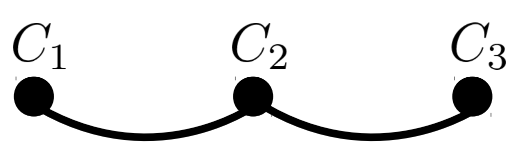
Block correlation functions (in diagrammatic language):

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

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

$H(C_1) =$ 

$H(C_1 | C_2) =$ 

$H(C_1 | C_2 | C_3) =$ 

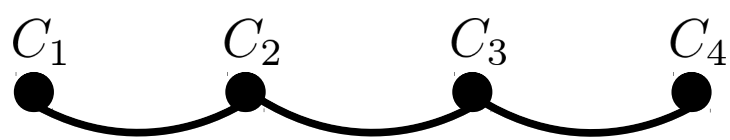
$$T^{irr} = \sum_{b=1}^{\infty} \sum_{C_1 \ldots C_b} \int dC_1 \ldots dC_b H(C_1| \ldots |C_b) S_I(C_1) G_{\text{eff}} \ldots G_{\text{eff}} S_I(C_b)$$

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 $n(C_1 \ldots C_g)$ which connect all points (also through intersection), e.g.

$H(C_1|C_2|C_3|C_4) =$



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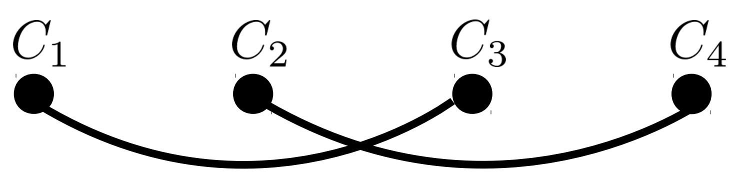


$$T^{irr} = \sum_{b=1}^{\infty} \sum_{C_1 \ldots C_b} \int dC_1 \ldots dC_b H(C_1| \ldots |C_b) S_I(C_1) G_{\rm eff} \ldots G_{\rm eff} S_I(C_b)$$

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):

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

Comparison of ring expansion with cluster expansion

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

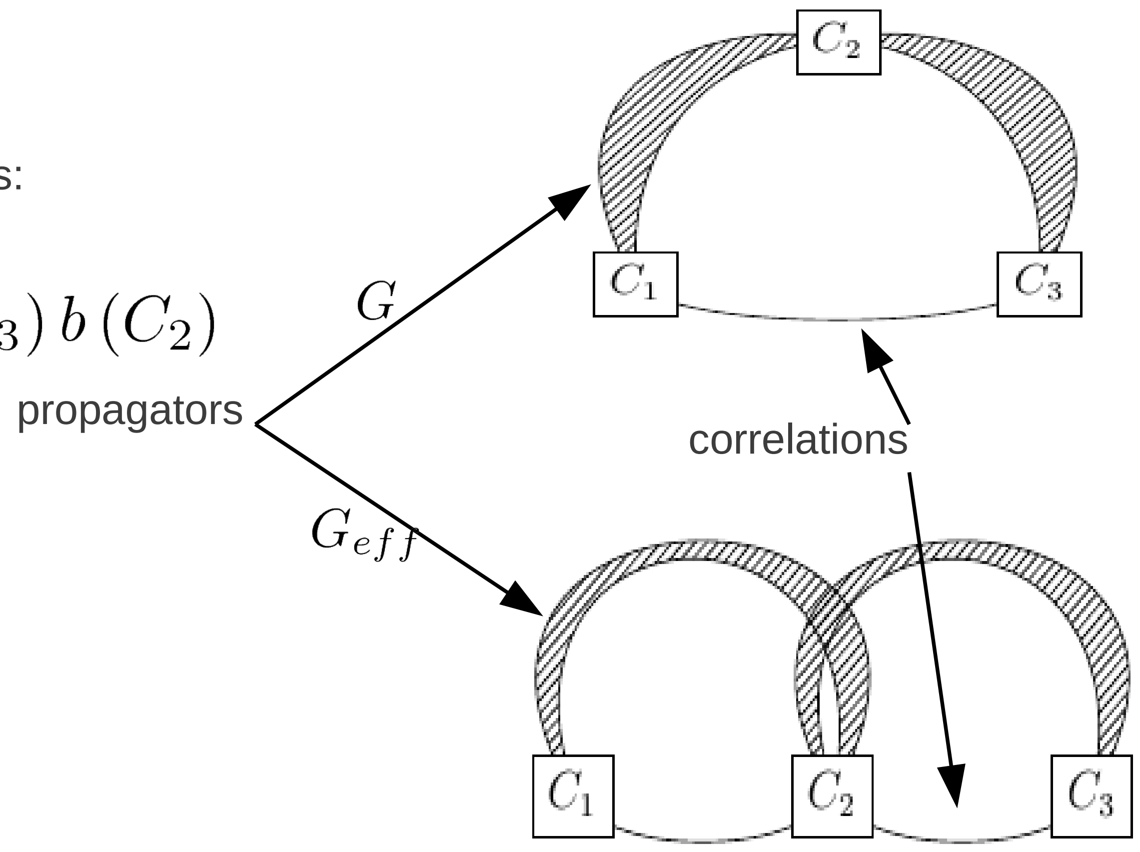
Ring expansion (2011)

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

When middle group goes away from others:

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

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



Two important differences:
-propagator
-volume of integration

Two approximation schemes

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

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

Clausius-Mossotti approximation $\Longrightarrow$ Generalized Clausius-Mossotti approximation

(two-body hydrodynamic interactions incomplete – the same as in δγ scheme (1983))

One-ring approximation (fully takes into account two-body hydrodynamic interactions)

Input:

-volume fraction

-two-body correlation function (PY); (three-particle correlation function by two body correlation function (Kirkwood))

-two-body hydrodynamic interactions

Repeating structures in T^{irr}

$$T^{irr} = T^{irr}_{CM} \left(1 - [hG]\, T^{irr}_{CM}\right)^{-1}$$

Clausius-Mossotti operator

Clausius-Mossotti approximation: $T^{irr}_{CM} \approx nM$

$$T^{irr} = T^{irr}_{RCM} \left(1 - [hG_{\text{eff}}]\, T^{irr}_{RCM}\right)^{-1}$$

renormalized Clausius-Mossotti operator

Generalized Clausius-Mossotti approximation: $T^{irr}_{RCM} \approx nB$

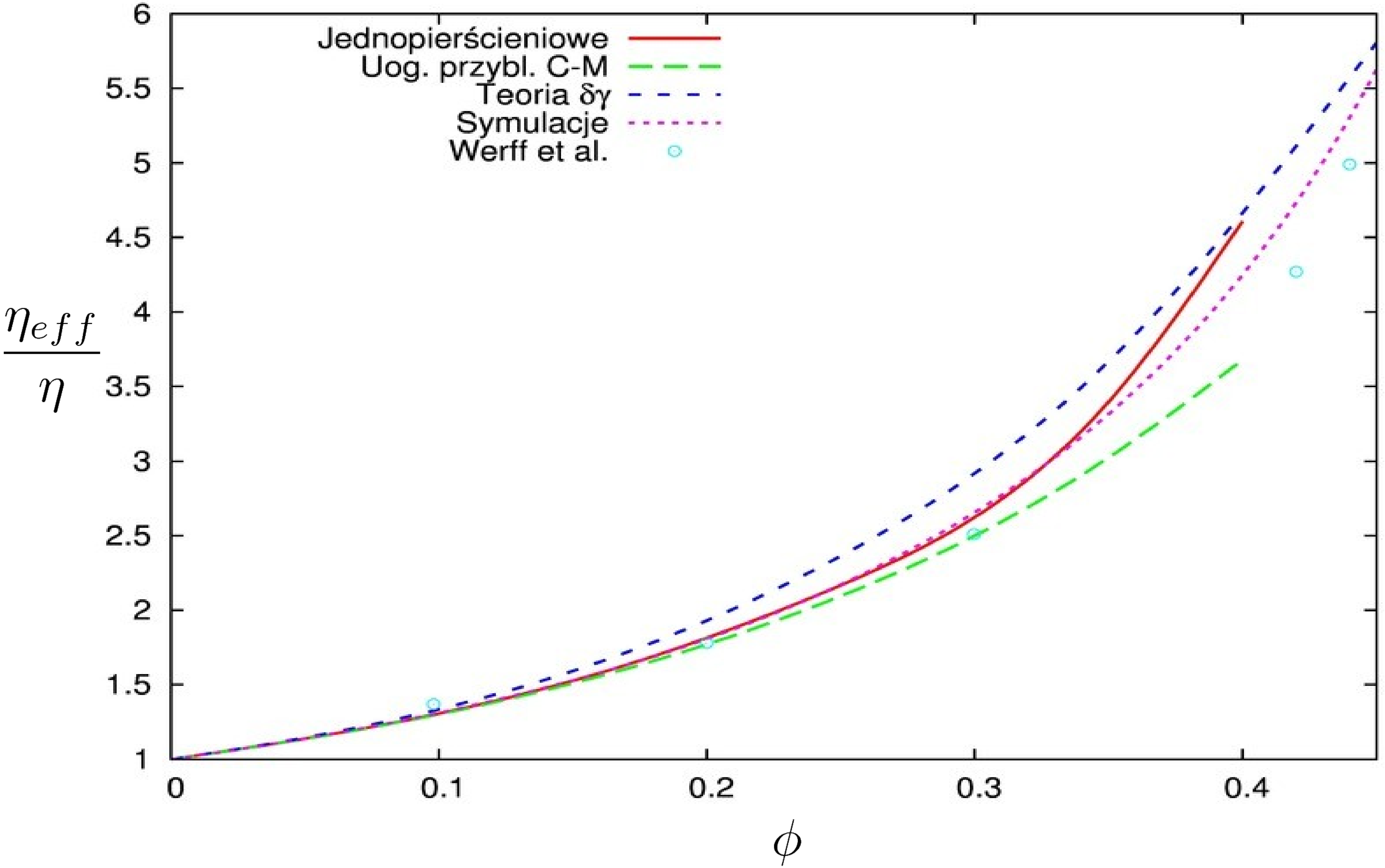
Approximate method formulated in terms of approximation for T^{irr}_{RCM}

$$T = nB + A$$

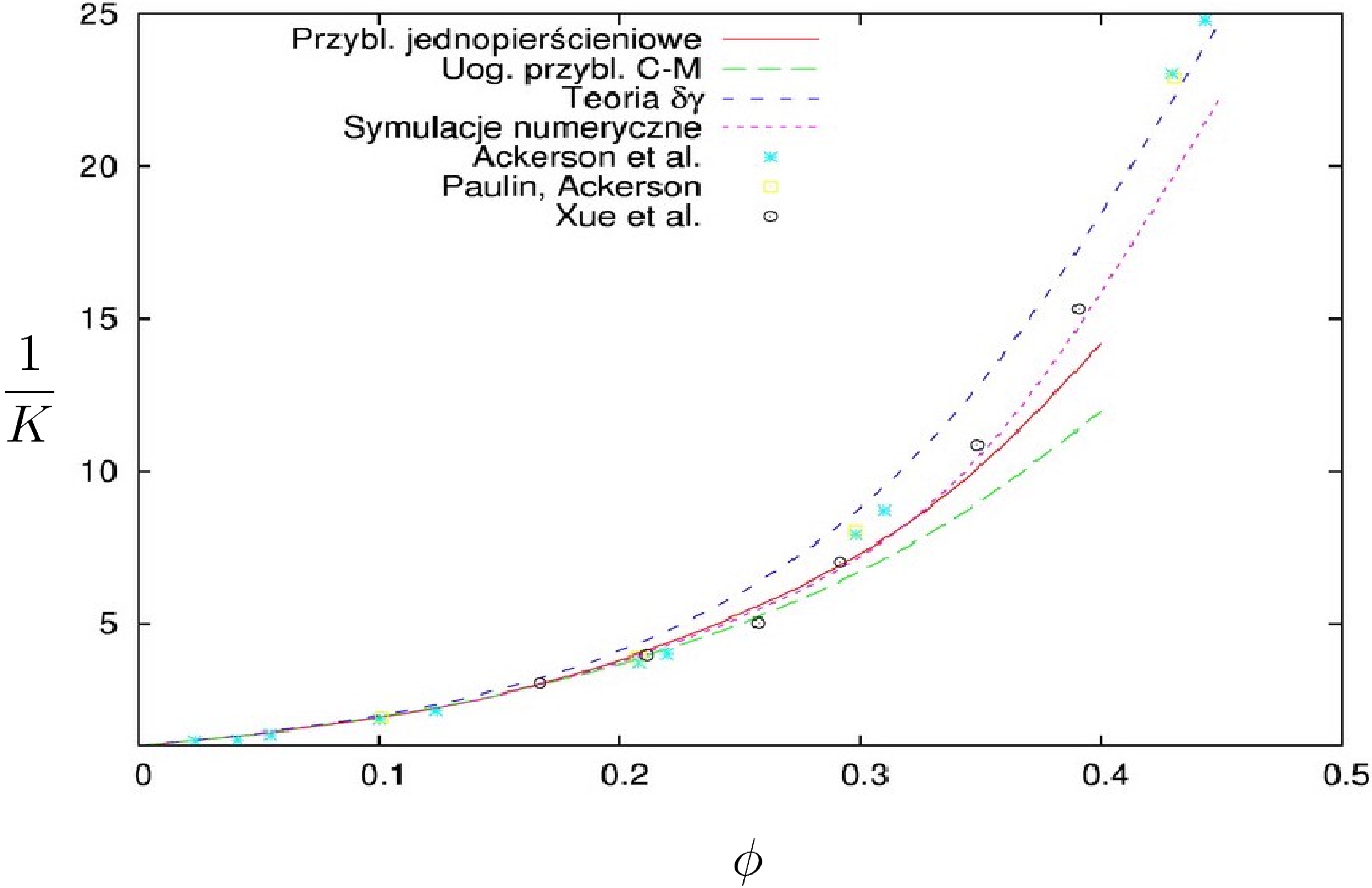
sequences that starts and ends on the same particle

$$nB = nM + \int d\mathbf{R}' A(\mathbf{R}, \mathbf{R}') G(\mathbf{R}', \mathbf{R}) M$$

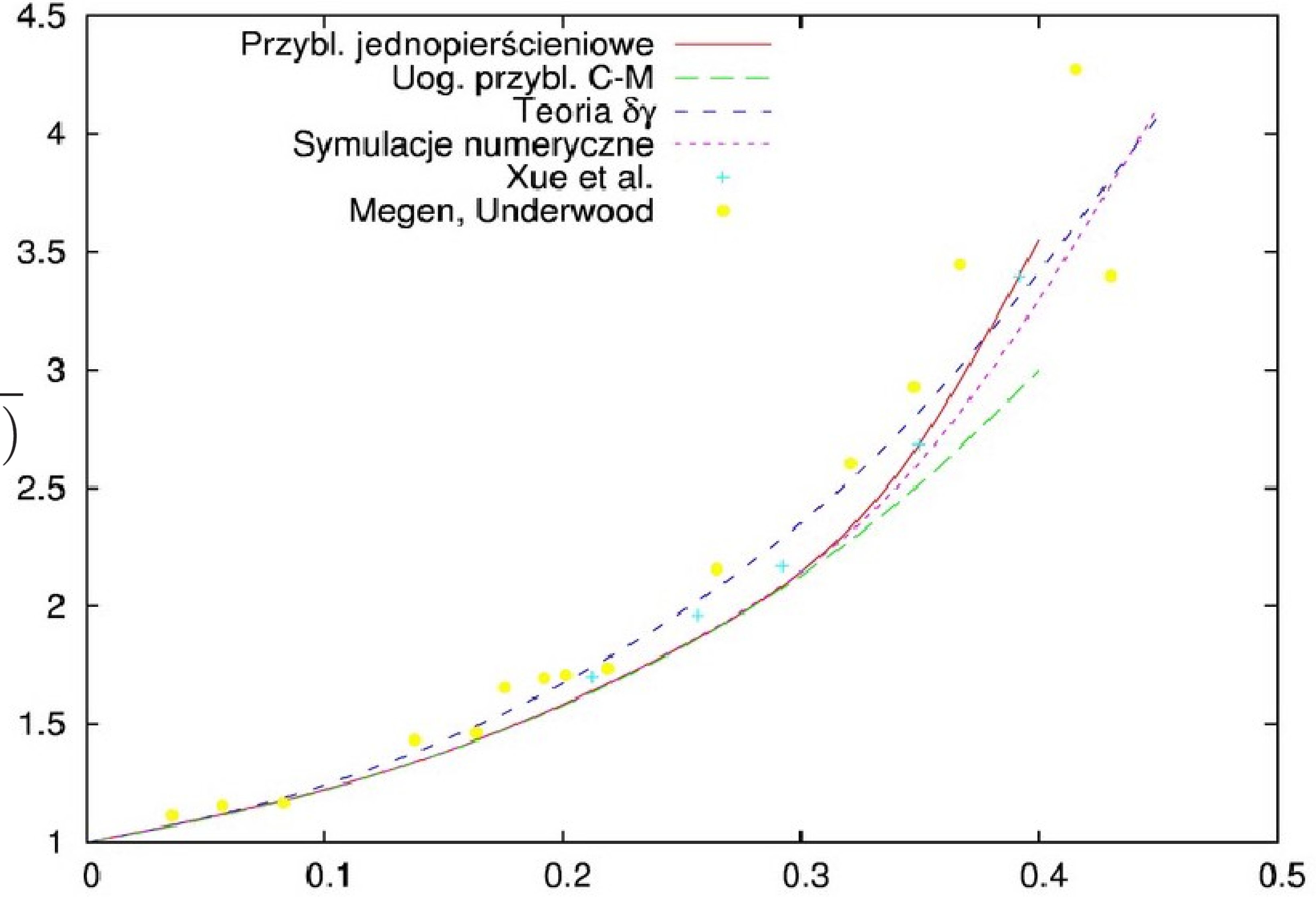
Effective viscosity



Inverse of sedimentation coefficient $K = H(0)$



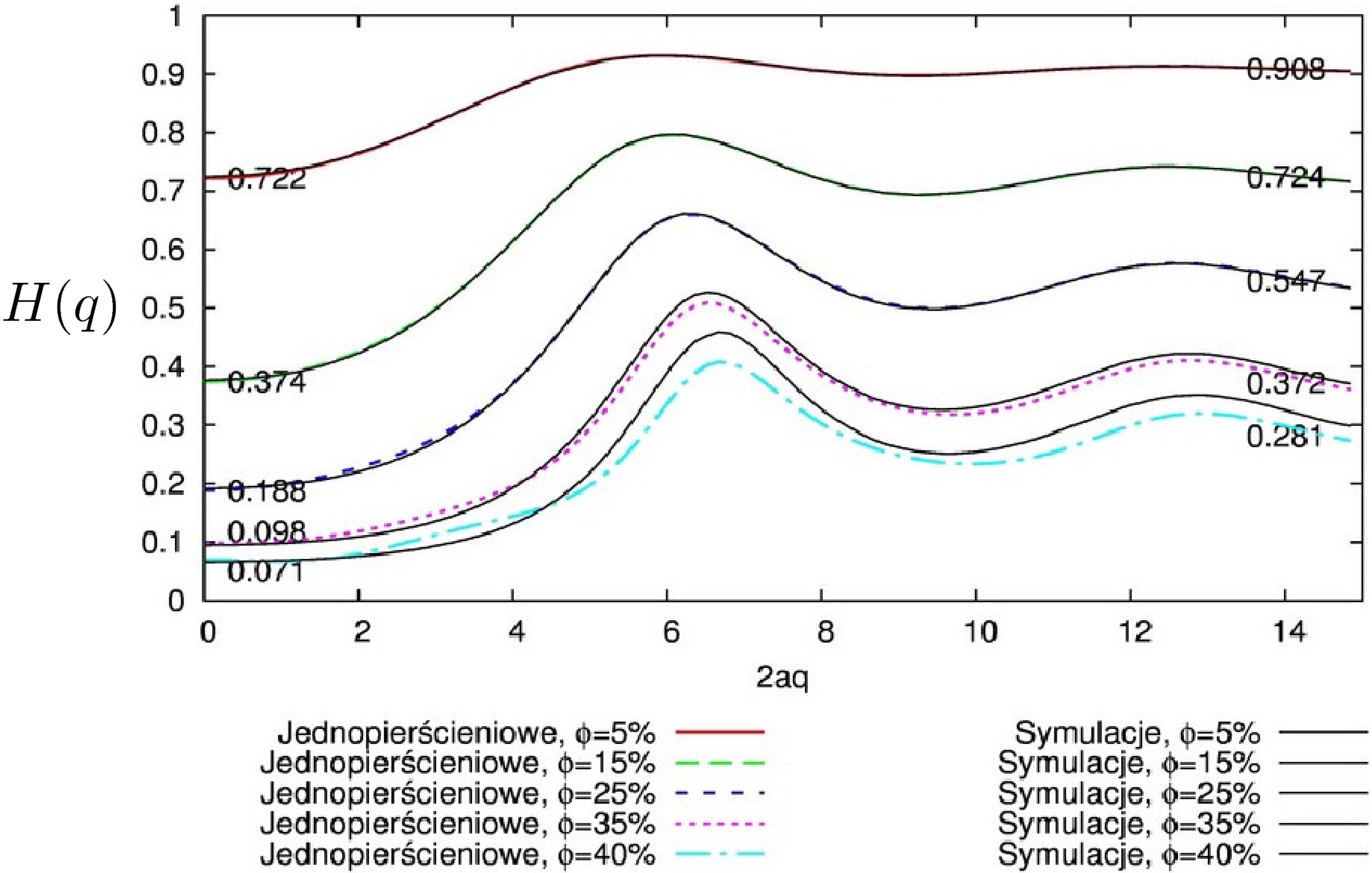
Inverse of mobility of single particle in suspension $H(\infty)$



$$\frac{1}{H(\infty)}$$

$$\phi$$

Hydrodynamic factor – one ring approximation



Summary and possibilities

- Long-range, many-body hydrodynamic interactions and strong interactions of close particles are important in suspensions
- Ring expansion for transport coefficients – can grasp all of three above features
- Two approximation schemes for transport coefficients:
  - generalized Clausius-Mossotti approximation (two-body hydrodynamic interactions not fully taken; comparable to $\delta\gamma$ scheme),
  - 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%)
- Simple generalization for different suspensions of spherical particles (droplets, spherical polymers) with different distributions (charged particles)

Renormalized Clausius-Mossotti operator

$$
\begin{aligned}
T^{irr}_{RCM} &= \sum_{r=0}^{\infty} T^{irr}_{RCM,r}, \\
T^{irr}_{RCM,0}\left(\mathbf{R},\mathbf{R}'\right) &= \sum_{C_1} \int dC_1 n\left(C_1\right) S_I(C_1;\mathbf{R},\mathbf{R}'), \\
T^{irr}_{RCM,1}\left(\mathbf{R},\mathbf{R}'\right) &= \sum_{C_1,C_2} \int dC_1 dC_2 \int d^3\mathbf{R}_1 d^3\mathbf{R}_2 \;\left[H(C_1|C_2) - h\left(\mathbf{R}_1,\mathbf{R}_2\right)\right] \times \\
&\times n\left(C_1\right) S_I(C_1;\mathbf{R},\mathbf{R}_1) G_{eff}\left(\mathbf{R}_1,\mathbf{R}_2\right) n\left(C_2\right) S_I(C_2;\mathbf{R}_2,\mathbf{R}'), \\
&\ldots
\end{aligned}
$$

One-ring approximation:

At most one ring in T^{irr}_{RCM}

At most two particle hydrodynamic interactions in S_I

Kirkwood approximation for three-particle distribution function:

$$n\left(123\right) \approx n^3 g\left(12\right) g\left(13\right) g\left(23\right)$$

Renormalization of two-particle interactions:

$$S_I(12) -> B S_I(12) B \qquad\qquad T^{irr} = nB + B\mathcal{T}^{irr} B$$

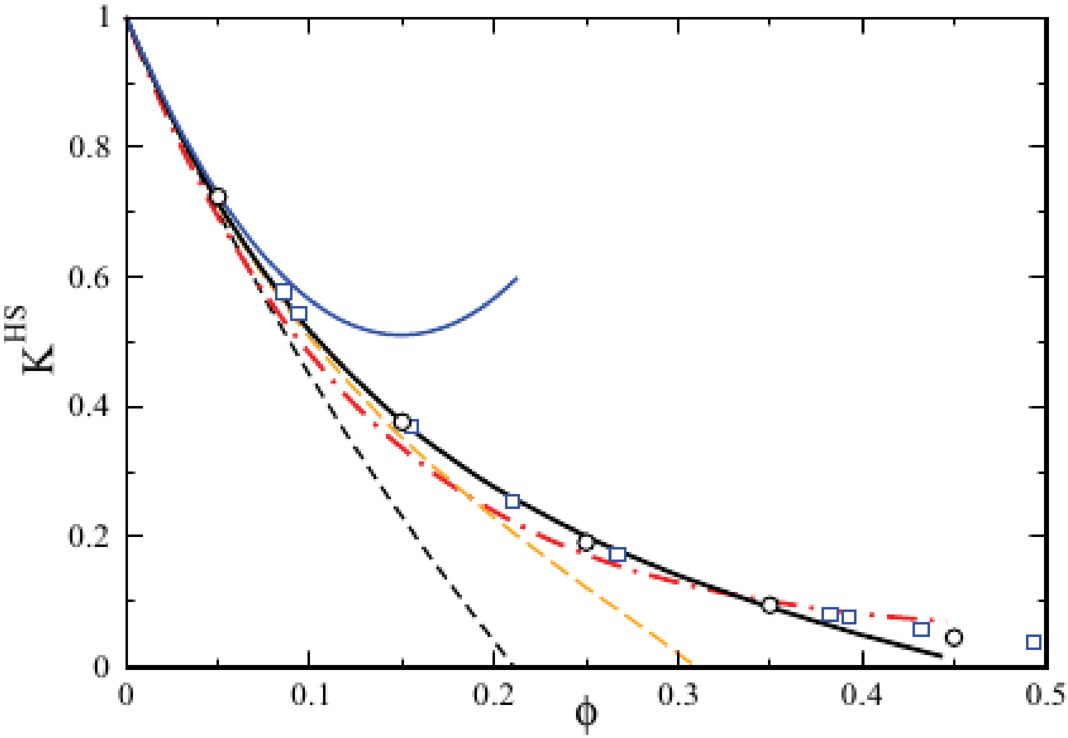


Fig. 6.3: *Reduced short-time sedimentation coefficient, K^{HS}, of neutral hard spheres. Open circles: Hydrodynamic force multipole simulation data by Abade et al. [26]. Open Squares: Lattice-Boltzmann simulation data by Segrè et al. [208]. Black dashed line: PA-scheme result. Dashed-dotted red line: uncorrected $\delta\gamma$-scheme result. Dashed orange line: self-part corrected $\delta\gamma$-scheme result, with $d_s/d_{t,0}$ taken from the PA-scheme. Solid black line: self-part corrected $\delta\gamma$-scheme result, with $d_s/d_{t,0}$ according to Eq. (4.26). Solid blue line: second-order virial result $K^{HS} = 1 - 6.546\phi + 21.918\phi^2$ [166]. The static structure factor input was obtained using the analytic Percus-Yevick solution.*

Approximate methods hitherto

There are many...

Clausius-Mossotti like (further)

“δγ scheme” (most comprehensive hitherto):

$$\eta_{eff} = c_0 + c_1 \delta\gamma + c_2 (\delta\gamma)^2 + \ldots$$

Fully taking into account two-body HI demands infinite order

Renormalization

Cluster expansion (1982):

short-range hydrodynamic interaction

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

block distribution function
(configurations of particles)

Oseen tensor (pure liquid):

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

Ring expansion (2011):

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

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

Effective propagator:

$$\mathbf{G}_{\text{eff}}(\mathbf{r}) \sim \frac{\eta}{\eta_{\text{eff}}} \mathbf{G}(\mathbf{r})$$