

Memory function for collective diffusion of interacting Brownian particles

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Abstract. – The problem of memory contribution to the collective diffusion coefficient of interacting Brownian particles is considered. A well-defined theoretical expression for this contribution, free of divergent integrals, is derived. Its value is then estimated for hard sphere suspensions numerically by means of extensive computer simulations.

A good physical model for a broad class of suspensions is the system of interacting Brownian particles. Despite a great number of important contributions to the field (see [1] for a review), there are still many areas in which theoretical results are scarce. One of these is the problem of memory contribution to the collective-diffusion coefficient in colloidal suspensions.

We consider N identical spherical particles performing Brownian motion in an incompressible viscous fluid at temperature T . On the time scale characteristic for light scattering experiments the evolution of the configuration space distribution function $P(\mathbf{X}, t)$ is described by the generalized Smoluchowski equation [1]

$$\begin{aligned} \frac{\partial}{\partial t} P(\mathbf{X}, t) &= \mathcal{D}(\mathbf{X}, t) P(\mathbf{X}, t), \\ \mathcal{D}(\mathbf{X}, t) &\equiv \sum_{i,j=1}^N \frac{\partial}{\partial \mathbf{R}_i} \cdot \mathbf{D}_{ij}(\mathbf{X}) \cdot \left[\frac{\partial}{\partial \mathbf{R}_j} + \beta \frac{\partial \Phi(\mathbf{X})}{\partial \mathbf{R}_i} \right], \end{aligned} \quad (1)$$

where $\mathbf{X} = (\mathbf{R}_1, \mathbf{R}_2, \dots, \mathbf{R}_N)$, \mathbf{R}_i being the position of the i -th particle and $\beta = 1/k_B T$. The potential $\Phi(\mathbf{X})$ incorporates both an external force field and direct pair interactions. Next, $\mathbf{D}(\mathbf{X})$ is the diffusion matrix connected with the mobility matrix $\boldsymbol{\mu}$ by the generalized Einstein relation $\mathbf{D}_{ij} = k_B T \boldsymbol{\mu}_{ij}$. According to the definition of the mobility matrix, the contribution of force \mathbf{F}_j acting on particle j to the velocity of particle i is given by $\boldsymbol{\mu}_{ij} \mathbf{F}_j$. In general, due to hydrodynamic interactions, $\boldsymbol{\mu}$ depends on the configuration \mathbf{X} and is non-diagonal in particle indices. Discussions of topics related to the mobility matrix can be found in the monograph [2].

Light scattering experiments give us access to the intermediate scattering function

$$F(\mathbf{k}, t) = \lim_{\infty} \frac{1}{N} \left\langle \sum_{i=1}^N \sum_{j=1}^N e^{i\mathbf{k} \cdot (\mathbf{R}_i(0) - \mathbf{R}_j(t))} \right\rangle, \quad (2)$$

where \mathbf{k} is the wave vector, \lim_{∞} stands for the thermodynamic limit and brackets stand for the average over equilibrium distribution. Finally, $\mathbf{R}_i(t)$ is the position of the i -th particle at time t . The Laplace transform of the intermediate scattering function defined as $F(\mathbf{k}, z) = \int_0^{\infty} e^{-zt} F(\mathbf{k}, t) dt$ can be written in the frame of the Zwanzig-Mori projection operator formalism [3, 4] as

$$F(\mathbf{k}, z) = \frac{S(\mathbf{k})}{z + D(\mathbf{k}, z)k^2}, \quad (3)$$

where $S(\mathbf{k})$ is the static structure factor and $D(\mathbf{k}, z)$ is the generalized diffusion function of the following structure:

$$D(\mathbf{k}, z) = \frac{1}{k^2} \Omega(\mathbf{k})(1 - M(\mathbf{k}, z)), \quad (4)$$

with the first cumulant $\Omega(\mathbf{k})$ and the Laplace transform of the memory function $M(\mathbf{k}, z)$. The collective-diffusion coefficients are given by the following limits:

$$D_c^l = \lim_{k \rightarrow 0} \lim_{z \rightarrow 0} D(\mathbf{k}, z), \quad D_c^s = \lim_{k \rightarrow 0} \frac{\Omega(\mathbf{k})}{k^2}, \quad (5)$$

where D_c^l and D_c^s stand for the long- and short-time diffusion coefficient, respectively. Their difference is caused by the memory effects in the system, which come from the relaxation of the distribution function. The dimensionless factor Δ measuring the strength of these effects is given by [5]

$$\Delta = \frac{D_c^s - D_c^l}{D_c^s} = \lim_{k \rightarrow 0} M(\mathbf{k}, z = 0). \quad (6)$$

The explicit expression for Δ may be obtained not only from the Zwanzig-Mori formalism, but also by solving the problem of linear reaction of the system to the external force field $\mathbf{E}(\mathbf{r}, t)$ acting on the particles [6] (sedimentation problem). In the frame of linear response one gets for the particle current $\mathbf{j}(\mathbf{r})$ induced in the system

$$\langle \mathbf{j}(\mathbf{r}) \rangle_t = \int d\mathbf{r}' \int_{-\infty}^t dt' \mathbf{X}(\mathbf{r} - \mathbf{r}', t - t') \mathbf{E}(\mathbf{r}', t'). \quad (7)$$

The symbol $\langle \rangle_t$ stands for the average over the time-dependent, non-equilibrium probability distribution. The response kernel \mathbf{X} can be written in the form

$$\mathbf{X}(\mathbf{r}, t) = \mathbf{X}_{\text{ins}}(\mathbf{r})\delta(t) + \mathbf{X}_{\text{ret}}(\mathbf{r}, t), \quad (8)$$

where the kernel $\mathbf{X}_{\text{ins}}(\mathbf{r})$ describes the instantaneous response of the system, whereas $\mathbf{X}_{\text{ret}}(\mathbf{r}, t)$ the retarded one. For interacting Brownian particles these read [6]

$$\mathbf{X}_{\text{ins}}(\mathbf{r} - \mathbf{r}') = \left\langle \sum_{i,j=1}^N \delta(\mathbf{r} - \mathbf{R}_i) \boldsymbol{\mu}_{ij} \delta(\mathbf{r}' - \mathbf{R}_j) \right\rangle, \quad (9)$$

$$\begin{aligned} \mathbf{X}_{\text{ret}}(\mathbf{r} - \mathbf{r}', t) = & -\beta^{-1} \left\langle \sum_{i,j,k,l=1}^N \delta(\mathbf{r} - \mathbf{R}_i) [(\nabla_k + \beta \mathbf{F}_k) \boldsymbol{\mu}_{ki}] \cdot \right. \\ & \left. \cdot e^{\mathcal{L}t} [(\nabla_l + \beta \mathbf{F}_l) \cdot \boldsymbol{\mu}_{lj}] \delta(\mathbf{r}' - \mathbf{R}_j) \right\rangle. \end{aligned} \quad (10)$$

Here \mathcal{L} is the adjoint Smoluchowski operator, obeying $\mathcal{D}P_{\text{eq}}(\mathbf{X})\dots = P_{\text{eq}}(\mathbf{X})\mathcal{L}\dots$, and \mathbf{F}_j is the total interparticle force acting on particle j . In the response operator language one gets for the memory function

$$M(k, t) = -\frac{\hat{X}_{\text{ret}}(k, t)}{\hat{X}_{\text{ins}}(k)}, \quad (11)$$

where $\hat{X}_{\text{ins}}(k)$ and $\hat{X}_{\text{ret}}(k, t)$ are the traces of Fourier transforms of kernels $\mathbf{X}_{\text{ins}}(\mathbf{r})$ and $\mathbf{X}_{\text{ret}}(\mathbf{r}, t)$, respectively.

It has been noticed (see, *e.g.*, ref. [7]) that the memory factor Δ identically vanishes either when hydrodynamic interactions are neglected (*i.e.* when $\mu_{ij} = \mu_0 \delta_{ij}$, where μ_0 is the single-particle mobility) or the two-body approximation for the matrix $\boldsymbol{\mu}$ is used. Ackerson [7] has suggested that if at least three-body hydrodynamic interactions are taken into account, the factor Δ should have non-zero value. Also Pusey points out in ref. [1] that “it seems that there should be a non-zero memory term in the collective diffusion”, however “the extant experimental evidence indicates” that the term “is relatively small”. There are, alas, no papers in the literature taking up this problem. The aim of the present work is to fill a gap, to provide theoretical results for the memory factor Δ and to obtain numerical estimates of its value for hard-sphere suspensions.

The main theoretical difficulty one encounters when analyzing the problem is caused by the long-range character of hydrodynamic interactions: elements of mobility matrix decay with interparticle distance R as $R^{-\gamma}$ with $\gamma = 1, 2, 3$. This results in the fact that the response kernels (9) and (10) are non-local (*e.g.*, they depend on the shape of the container enclosing the suspension) and are not integrable. Hence various functions characterizing the suspension (like the memory function $M(k, z)$) are not continuous at $\mathbf{k} = 0$ [8]. This in turn means that one has to perform a cumbersome $\mathbf{k} \rightarrow 0$ limit in order to get Δ from eq. (6) and cannot just put $\mathbf{k} = 0$ there.

The problem can be solved by the following renormalization procedure. First, one must introduce the average velocity of suspension $\langle \mathbf{v}(\mathbf{r}) \rangle_t$. Here $\mathbf{v}(\mathbf{r})$ is equal to the fluid velocity wherever \mathbf{r} is inside the fluid, and coincides with the rigid-body motion wherever \mathbf{r} lies inside the particle. Note that $\langle \mathbf{v}(\mathbf{r}) \rangle_t$ is generally non-zero, even when viewed from the laboratory frame (cf. the intrinsic convection problem [9]). However, using the incompressibility of the fluid one can prove that when the container is at rest, the integral of $\langle \mathbf{v}(\mathbf{r}) \rangle_t$ over the total suspension volume V vanishes. In the Fourier-transform language this can be expressed as

$$\langle \hat{\mathbf{v}}(\mathbf{k} = 0) \rangle_t = 0. \quad (12)$$

The above must be taken into account when the limit $k \rightarrow 0$ in eq. (6) is being carried out.

In the next step of the procedure the linear-response scheme (7) is generalized by introducing additional disturbance — an imposed flow field $\mathbf{v}_0(\mathbf{r})$, which satisfies hydrodynamic equations in the absence of particles. As a consequence, one must add a term of the form $\int d\mathbf{r}' \int_{-\infty}^t dt' \mathbf{Y}(\mathbf{r} - \mathbf{r}', t - t') \mathbf{v}_0(\mathbf{r}', t')$ to the right-hand side of eq. (7). Finally, the imposed flow $\mathbf{v}_0(\mathbf{r})$ is expressed in terms of $\langle \mathbf{v}(\mathbf{r}) \rangle_t$ and henceforth eliminated from the response equation

which then takes the form

$$\begin{aligned} \langle \mathbf{j}(\mathbf{r}) \rangle_t &= \int_{-\infty}^t dt' \int d\mathbf{r}' \mathbf{X}^{\text{irr}}(\mathbf{r} - \mathbf{r}', t - t') \mathbf{E}(\mathbf{r}', t') + \\ &+ \int_{-\infty}^t dt' \int d\mathbf{r}' \mathbf{Y}^{\text{irr}}(\mathbf{r} - \mathbf{r}', t - t') \langle \mathbf{v}(\mathbf{r}') \rangle_{t'}. \end{aligned} \quad (13)$$

It turns out that the “irreducible” kernels $\mathbf{X}^{\text{irr}}(\mathbf{r})$ and $\mathbf{Y}^{\text{irr}}(\mathbf{r})$ are devoid of all infinite-range terms and therefore their Fourier transforms have well-defined $\mathbf{k} = 0$ value. All the geometrical effects are now accounted for in the average suspension velocity $\langle \mathbf{v}(\mathbf{r}) \rangle_t$.

In the case of instantaneous response the above renormalization procedure has been carried out in refs. [10–13]. However, generalization of the scheme to incorporate effects of relaxation of the distribution function (described by the retarded-response kernel) is not trivial, as the evolution operator \mathcal{L} in eq. (10) is long-ranged itself. To carry it out, we have performed the scattering expansion [14] of all operators including the evolution operator \mathcal{L} . A special diagrammatic technique [15], has been developed to facilitate operations on subsequent terms in the expansion. In particular, the diagrams corresponding to long-range, non-integrable terms in the expansion turn out to have common topological features, which makes it easy to resum them and to incorporate them into the average velocity field $\langle \mathbf{v}(\mathbf{r}) \rangle_t$. As a final result, theoretical expressions for the irreducible retarded-response kernels \mathbf{X}^{irr} and \mathbf{Y}^{irr} are obtained. The details of the derivation will be given in a subsequent regular article. It is worth mentioning that a similar renormalization scheme have been carried out in ref. [16] for frequency-dependent viscosity.

In the long-wavelength limit, which is of interest for us, the Fourier transform of eq. (13) takes a slightly simpler form as $\lim_{\mathbf{k} \rightarrow 0} \hat{\mathbf{Y}}^{\text{irr}}(\mathbf{k}, t) = n\delta(t)\mathbf{I}$, where \mathbf{I} is the identity matrix. Thus for $\mathbf{k} \rightarrow 0$ one has

$$\langle \hat{\mathbf{j}}(\mathbf{k}) \rangle_t - n \langle \hat{\mathbf{v}}(\mathbf{k}) \rangle_t = \int_{-\infty}^t dt' \hat{\mathbf{X}}^{\text{irr}}(\mathbf{k}, t - t') \hat{\mathbf{E}}(\mathbf{k}, t'), \quad (14)$$

where, as before, the hat denotes a Fourier transform of a given quantity. Now, this equation together with the condition (12) allows us to derive a well-defined expression for the limit $M(t) = \lim_{\mathbf{k} \rightarrow 0} M(\mathbf{k}, t)$. Namely, in the same way in which the formula (11) has been obtained from eq. (7) one gets

$$M(t) = - \frac{\hat{X}_{\text{ret}}^{\text{irr}}(k=0, t)}{\hat{X}_{\text{ins}}^{\text{irr}}(k=0)}. \quad (15)$$

The quantities $\hat{X}_{\text{ins}}^{\text{irr}}$ and $\hat{X}_{\text{ret}}^{\text{irr}}$ are defined as traces of the instantaneous and retarded part of the kernel $\hat{\mathbf{X}}^{\text{irr}}$, respectively. The above formula leads also to a well-defined expression for the memory factor since, according to eq. (6),

$$\Delta = \int_0^{\infty} M(t) dt. \quad (16)$$

The next aim is to estimate values of the factor above numerically. It is reasonable to divide this task into two stages: calculations of the initial value of the memory function $M(t=0)$ and calculations of the mean relaxation time $\tau_M = M(t=0)^{-1} \int_0^{\infty} M(t) dt$. The reason why we have singled out the initial value of the memory function is that it can be estimated by means of equilibrium averaging only, which can be done with much greater

TABLE I – The estimates of the factor Δ for hard-sphere suspension of volume fraction ϕ obtained from initial values of the memory function $M(t=0)$ calculated by Monte Carlo averaging and the relaxation time $\tau_c = a^2 S(0)/D_s^*$.

ϕ	$D_0^{-1} a^2 M(t=0)$	$D_0 a^{-2} \tau_c$	Δ_{est}
0.01	$(1.55 \pm 0.05) \cdot 10^{-4}$	0.94	0.01%
0.1	$(1.7 \pm 0.1) \cdot 10^{-2}$	0.55	1%
0.2	$(9.2 \pm 0.3) \cdot 10^{-2}$	0.32	3%
0.3	0.24 ± 0.015	0.20	5%
0.4	0.54 ± 0.03	0.13	7%
0.45	0.67 ± 0.02	0.10	7%

accuracy than the calculations of τ_M , which require Brownian dynamic simulations. Therefore, as was noted by Zwanzig and Ailawadi [17], such a two-step procedure increases considerably the accuracy of the numerically obtained memory function. Additionally, even without the Brownian dynamics simulations, one can estimate the value of the memory factor Δ , provided one assumes that the mean relaxation time τ_M would be similar to characteristic times of other relaxation processes in the system.

There are two main numerical problems that one encounters when calculating the response kernels. The first is the computation of the mobility matrix $\boldsymbol{\mu}$ for a given configuration of particles. It is achieved by means of numerical implementation of the multipole expansion method [18]. The second, even more complex problem, is the calculation of the divergence of the mobility matrix $\nabla \cdot \boldsymbol{\mu}$. Although in principle this can be done by numerical differentiation of $\boldsymbol{\mu}$, such a scheme is not only extremely time- and memory-consuming but also inaccurate. Instead, we have devised a scheme of calculation of the divergence of the mobility matrix in an analytical way with the use of the above-mentioned multipole expansion method [19]. Such a scheme can be helpful not only in the present problem but also in any numerical studies on Brownian suspensions whenever the divergence of mobility is needed, for example in Brownian dynamics simulations.

Numerical calculations have been performed for hard-sphere suspensions. The direct pair interactions are then described by the hard-core potential.

For small particle concentrations the value of $M(t=0)$ can be assessed by means of the virial expansion. The first non-vanishing term in the expansion corresponds to the three-particle contribution. For the hard-sphere system one gets, from eq. (15),

$$\frac{a^2}{D_0} M(t=0) = m_3 \phi^2 + O(\phi^3), \quad (17)$$

with

$$m_3 = \frac{9\eta^2}{8a^2} \int d\mathbf{R}_2 d\mathbf{R}_3 \left(\sum_{i,j=1}^3 \nabla_j \cdot \boldsymbol{\mu}_{ij} \right)^2 W(1,2,3). \quad (18)$$

In the equations above ϕ is the volume fraction, a is the sphere radius and $D_0 = k_B T (6\pi\eta a)^{-1}$ with η standing for the fluid viscosity. The initial value of the memory function is rescaled by a^2/D_0 in order to make it dimensionless. Finally, $W(1,2,3)$ is unity for non-overlapping configurations of the spheres and vanishes otherwise. By means of the numerical integration in the three-body configuration space (analogous to that presented in [20]) we have obtained $m_3 = 1.42 \pm 0.02$.

TABLE II – The values of the factor Δ and the mean relaxation time τ_M for the hard-sphere suspension of volume fraction ϕ obtained from the equilibrium Monte Carlo averaging and Brownian dynamics technique.

ϕ	$D_0 a^{-2} \tau_M$	Δ
0.2	0.126 ± 0.025	0.01 ± 0.003
0.3	0.120 ± 0.015	0.03 ± 0.01
0.4	0.09 ± 0.02	0.05 ± 0.015

Virial expansion results can be used for very dilute suspensions only. For larger concentrations we have calculated $M(t=0)$ by means of the Monte Carlo averaging for the system with periodic boundary conditions. The use of such conditions simplifies considerably the expression for the memory function. In fact, when deriving the mobility matrix for a periodic system one must add the constrain that the net suspension velocity in the whole sample vanishes [21–23], as otherwise the divergences in the fluid velocity field would appear. In that way eq. (12) is automatically satisfied, which, in turn, implies that we can relax the irreducibility condition in eq. (15) and the expression for $M(t)$ in case of hard spheres takes the simple form

$$M(t) = \frac{\left\langle \sum_{i,j,k,l=1}^N (\nabla_j \cdot \boldsymbol{\mu}_{ij}(0)) \cdot (\nabla_l \cdot \boldsymbol{\mu}_{kl}(t)) \right\rangle_{\text{per}}}{\beta \left\langle \sum_{i,j=1}^N \text{Tr } \boldsymbol{\mu}_{ij} \right\rangle_{\text{per}}}, \quad (19)$$

where $\nabla \cdot \boldsymbol{\mu}(t)$ is the divergence of the mobility matrix at time t , the symbol $\langle \rangle_{\text{per}}$ stands for the average over hard-sphere configurations with periodic boundary conditions.

To account for the finite-size effects in the periodic sample, one analyzes the dependence of $M(t=0, N)$ on the number of spheres in the periodic cell N . It turns out that the dependence of the data on N can be described by the function $A + BN^{-1/3}$. By fitting the data for $N = 30, 50, 60, 70$ and 100 to the dependence above we have obtained the asymptotic values $M(t=0, N = \infty)$ for a wide range of volume fractions (see table I).

Now we turn to the estimation of the mean relaxation time τ_M . This is obtained by means of Brownian dynamics simulations [24, 25] in which one constructs the configuration space trajectories of the system which are realizations of the stochastic process described by eq. (1). In this way one can assess the time dependence of the memory function $M(t)$. The simulations have been performed for three values of the volume fraction: $\phi = 0.2, 0.3$ and 0.4 for different numbers of spheres in a periodic cell (up to 100). By combining the results for τ_M with the values of $M(t=0)$, we calculate the values of the memory factor Δ . The results obtained in this way are given in table II. One concludes from inspecting the data that the memory effect is expected to be quite small but it is growing with the volume fraction.

As was mentioned before, some estimation of the mean relaxation time of the memory function can be obtained from the hypothesis that it would not be much different from characteristic times of other relaxation times for the collective processes in a suspension. These may be estimated with the use of the semi-phenomenological reasoning due to Medina-Noyola [26]. He argued, namely, that the effect of hydrodynamic interactions on the dynamics of a suspension can be taken into account by replacing in the formulae the one-particle diffusion coefficient D_0 , which describes the diffusion in the absence of hydrodynamic interactions, by the short-time self-diffusion coefficient D_s^s . As the characteristic time for the collective phenomena in the absence of hydrodynamic interactions reads simply $\tau_c^0 = a^2 S(0)/D_0$, the reasoning above would give the characteristic time for the collective processes for systems with hydrodynamic interactions to be $\tau_c = a^2 S(0)/D_s^s$. Assuming that the relaxation time of our memory function

τ_M is not very different from τ_c and taking the values of D_s^s from the numerical simulations of Ladd [27], we get the estimations of Δ , which together with the values of τ_c are presented in table I. We see that these estimations are reasonable.

The obtained results agree qualitatively with the experimental data (cf. [1]), quantitative comparison being difficult because of large experimental errors. It is to be hoped that the present results would stimulate experimentalists to perform more precise measurements of the collective diffusion coefficients.

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