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# Mean-field-hydrodynamics Brownian dynamics simulations of viscosity and self-diffusion of near-hard-sphere colloidal liquids

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**Abstract.** Equations of motion for model colloidal particles are derived, which include a mean-field local density representation for the many-body-hydrodynamics (MBH) interactions. These are implemented as a modification of Ermak's 1975 Brownian dynamics free-draining algorithm, with a local density prescription for the friction coefficient. The computed long-time self-diffusion coefficients derived from this model agree well with experiment at all volume fractions. The hydrodynamics model relative and dynamic viscosities are larger than those of the free-draining algorithm, reflecting slower structural relaxation processes taking place in the new model.

## 1. Introduction

Since the development of the Brownian dynamics simulation technique by Ermak (1975) a goal has been to improve the model to include the effects of many-body-hydrodynamics (MBH) solvent-mediated forces that lead to highly correlated instantaneous trajectories of the suspended particles. A number of approaches have been proposed for including MBH in discrete-particle simulations. Many are based on the 'far-field' diffusion tensor expansion method proposed by Ermak and McCammon (1978) and extended to higher-order terms by, for example, Durlofsky *et al* (1987), Ladd (1990) and Cichocki *et al* (1994). These methods are extremely time consuming to implement, as they involve  $O(N^3)$  matrix inversions (where  $N$  is the number of colloidal particles) and involve the computation of correlated Brownian forces between different particles to satisfy the fluctuation-dissipation theorem. Consequently only relatively small systems of several hundred particles at most have been considered to date, which restricts these techniques to modelling stabilized colloidal systems. The important area of flocculation is effectively beyond the scope of this approach for the foreseeable future, as long-range (possibly fractal) structures can be important in determining many of the physical properties of these systems. Convergence of the interaction expansion at short range has also been a problem in these techniques, and near-field 'lubrication' interactions have had to be introduced in an *ad hoc* manner. The simplicity of the original Ermak algorithm has been lost and in these modifications the useful transparent link between the behaviour and characterization of a 'typical' individual particle and computed macroscopic properties has been abandoned. It is still not clear what level of implementation is required for the ('short') time-scale associated with the correlated Brownian forces. (On this time-scale the particle displacements are negligible when compared with their diameters.) Significant structural relaxation behaviour and

rheological properties are typically associated with much longer time-scales—the so-called ‘long’ time-scale. Owing to a clear separation between these two time-scales, it is possible that a detailed description of the short-time many-particle dynamics (as outlined above) is not required. Indeed it is possible that a relatively simple treatment of the short-time behaviour is sufficient to give rise to a realistic description of rheological phenomena.

In response, to remedy these deficiencies, and to explore this hypothesis, we propose a new algorithm that includes some of the effects of MBH but retains the non-tensorial form of the original Ermak algorithm. It recognizes that there are well-defined time-scales in a colloidal liquid. The ‘short’ time-scale is that of the density fluctuations of the solvent in an essentially arrested configuration of colloidal particles. The effects of phenomena originating on this time-scale are introduced into the model through a rescaling of the model’s short-time self-diffusion coefficient to that known for experimental near-hard-sphere systems. The particle friction coefficient in the free-draining model has the infinite-dilution value,  $\zeta_0$ . Here in the new model we adopt a local density approximation for the friction coefficient, so that  $\zeta > \zeta_0$ , which is consistent with this rescaling. This leads to a more realistic slowing down of structural relaxation processes (compared with Ermak algorithm) on the rheologically relevant ‘long’ time-scale.

## 2. Theory

The Ermak BD algorithm (Ermak 1975) is based on the Smoluchowski equation

$$\frac{\partial P}{\partial t} = D_0 \nabla^2 P - \frac{D_0}{k_B T} \nabla \cdot (F P) \quad (1)$$

where  $P$  is the probability distribution function that a particle in the system will undergo a displacement,  $\Delta r$ , during a time interval  $\Delta t$ .  $D_0 = k_B T / 3\pi\sigma\eta_s$  is the self-diffusion coefficient of the colloidal particle with equivalent hard-sphere diameter  $\sigma$  in a solvent of viscosity  $\eta_s$  in the limit of infinite dilution.  $k_B$  is the Boltzmann constant and  $T$  is the absolute temperature.  $F(r, t)$  is the net force on the colloidal particle from the other colloidal particles and any position-dependent external field.

If the coordinate of colloidal particle  $i$  is  $R_i$  let  $R^N \equiv \{R_1, \dots, R_N\}$ . If the friction coefficient is made configuration dependent, i.e.,  $\zeta = \zeta(R^N)$ , equation (1) is replaced by

$$\begin{aligned} \frac{\partial P}{\partial t} &= \nabla \frac{k_B T}{\zeta} \cdot \nabla P - \nabla \cdot \left( \frac{1}{\zeta} F P \right) \\ &= \frac{k_B T}{\zeta} \nabla^2 P - \frac{1}{\zeta} \left( \frac{k_B T \nabla \zeta}{\zeta} \right) \cdot \nabla P - \frac{1}{\zeta} F \cdot \nabla P \\ &= \frac{k_B T}{\zeta} \nabla^2 P - \frac{1}{\zeta} \left( \frac{k_B T \nabla \zeta}{\zeta} + F \right) \cdot \nabla P. \end{aligned} \quad (2)$$

In the above equation and below we assume  $\nabla_i \zeta_i = 0$  and  $\nabla_i \cdot F_i = 0$  only when involved in terms resulting in  $\nabla_i P_i$ . This is an approximation and is justified on the grounds that the terms in  $\nabla_i P_i$  and  $\nabla_i^2 P_i$  dominate the diffusion equation. The missing term  $P \nabla \cdot (F \zeta)$  involves the spatial variation of  $F$ , i.e.,  $\nabla \cdot F$ . This will lead to a higher-order term in any position-update algorithm. The computer simulations suggest that this term is insignificant with typical time steps used. Simulations performed from the algorithm derived from these simplified equations generate a canonical ensemble (i.e., give the same thermodynamic quantities and distribution functions as the free-draining model),

so the simplification appears not to introduce any serious errors in practice. The solution to equation (2) is

$$P(\Delta r, \Delta t) = \left( \frac{1}{4\pi D_0 \Delta t} \right)^{3/2} \exp(-(\Delta r - (k_B T \zeta^{-1} \nabla \zeta + F(t)) \Delta t / \zeta)^2 / 4D \Delta t). \quad (3)$$

Equation (3) does not generate a canonical ensemble for the potential field associated with  $F$  (owing to the  $k_B T \zeta^{-1} \nabla \zeta$  term). Nevertheless if we define

$$F = F_0 - k_B T \zeta^{-1} \nabla \zeta \quad (4)$$

then the algorithm

$$r(t + \Delta t) = r(t) + F(t) \Delta t / \zeta(R^N) + \Delta r^B(\zeta(R^N)) \quad (5)$$

where  $\Delta r^B$  is the random displacement sampled from a Gaussian distribution of zero mean and variance,  $\langle \Delta r^2 \rangle = 6D_0 \Delta t$  will generate the canonical ensemble for the interaction field  $U_0$  where  $F_{i0} = -\nabla_i U_0$ .  $U_0$  is a sum over particles. A simulation carried out with a force field  $F$  derived from a potential field  $U$  generates a Boltzmann distribution of states appropriate to  $U_0(R^N)$ . The analytic form of  $\zeta(R)$  is not known. It is reasonable to assume that the friction coefficient will increase as more colloidal particles approach closely to an arbitrary colloidal particle, and therefore a reasonable analytic form is

$$\zeta_i(R^N) = \zeta_0 \left( 1 + C \sum_{j \neq i} \epsilon \left( \frac{\sigma}{r_{ij}} \right)^m \right) \quad (6)$$

where  $r$  is the separation between the particle centres,  $C$  is a constant,  $m$  is an arbitrary exponent,  $\zeta_0 = 3\pi\sigma\eta_s$ , and  $\epsilon$  sets the energy scale. The proposed analytic form goes to the correct limit,  $\zeta_0$ , at infinite dilution and increases strongly at high volume fraction; this arises from the crowding effects of the surrounding colloidal particles. It is reasonable to expect that at finite volume fractions the effective range of the interaction should not extend further than the first coordination shell of particles, which is achieved if  $m > 12$ . The analytic form chosen is similar to that of the direct interaction law,  $U_0$ , of interest here, and thereby allows the development of a manageable algorithm. If we assume that the thermodynamic interactions between the colloidal particles can be represented on a pairwise-additive basis with a pair potential of the generic form

$$U_0(r, n) = \epsilon(\sigma/r)^n \quad (7)$$

so that  $F(n) = -\nabla U_0(n, R^N)$  then

$$-k_B T \zeta^{-1} \nabla \zeta = \zeta_0 C k_B T \zeta^{-1} F(m). \quad (8)$$

The position-update algorithm becomes

$$r(t + \Delta t) = r(t) + (F(n, t) + \zeta_0 C k_B T \zeta(R^N)^{-1} F(m, t)) \Delta t / \zeta(R^N) + \Delta r^B(\zeta(R^N)). \quad (9)$$

If we let  $m = n$  then

$$r(t + \Delta t) = r(t) + (1 + \zeta_0 C k_B T \zeta(R^N)^{-1}) F(n, t) \Delta t / \zeta(R^N) + \Delta r^B(\zeta(R^N)). \quad (10)$$

The important point about this algorithm is that it generates a canonical ensemble for the potential field defined by particles interacting through equation (7). The value of  $C$  is adjusted at each volume fraction so that the mean square displacements arising from the Brownian force are consistent with the experimentally derived value for the short-time diffusion coefficient,  $D_s$ , i.e.,

$$\langle (\Delta r^B)^2 \rangle = 6k_B T \langle \zeta^{-1}(R^N) \rangle \Delta t = 6D_s \Delta t \quad (11)$$

where (Lionberger and Russel 1994)

$$\frac{D_s}{D_0} = (1.00 - 1.56\phi)(1.00 - 0.27\phi) \quad (12)$$

is a good approximation. The long-time self-diffusion coefficient,  $D_L$ , fitted to the experimental data of Medina-Noyola (1988) and Goodwin and Ottewill (1991) between  $0.04 < \phi < 0.5$  is

$$\frac{D_L}{D_0} = 0.8(1.00 - 1.15\phi)(1.00 - 2.00\phi). \quad (13)$$

### 3. Simulation details

We consider a cubic simulation cell interacting with an inverse power potential given in equation (7) with the values,  $\epsilon = k_B T$  and  $n, m = 36$  truncated at  $r_{ij} > 1.3\sigma$ , and  $n, m = 18$  truncated at  $r_{ij} > 2.2\sigma$ , which is the case of the parameter  $n$  for our previous studies (see, e.g., Heyes and Mitchell 1994). The number of molecules in the BD cell,  $N$ , is 108 or 256. The reduced number density  $\rho = N\sigma^3/V$ , where  $V$  is the volume of the simulation cell. The volume fraction,  $\phi = \pi\rho/6$ , is more commonly used in the colloid literature, and we therefore use this quantity instead of  $\rho$ . Calculated thermodynamic and structural properties are quoted in particle-based reduced units, e.g.,  $\sigma$  for length and  $\epsilon$  for energy. The time step was typically  $1.0 \times 10^{-4} a^2/D_0$  where  $a$  is the radius of the colloidal particle. Production simulations were for in excess of  $100a^2/D_0$ .

For the inverse power potentials considered here, the interaction energy, pressure and mechanical properties are trivially related (Heyes and Mitchell 1994). The average interaction energy per particle,  $u$ , is given by

$$u = \frac{1}{2N} \sum_{i=1}^N \sum_{j \neq i}^N \langle U_{ij}(r_{ij}) \rangle. \quad (14)$$

The infinite-frequency shear-rigidity modulus in the zero-strain-amplitude limit,  $G_\infty = G'(\omega \rightarrow \infty)$ , is given by

$$G_\infty = (n^2 - 3n)\rho \langle u \rangle / 15 \quad (15)$$

using the formula of Zwanzig and Mountain (1965). The stress tensor,  $\sigma$ , in this model is given by

$$\sigma = \frac{\rho}{N} \sum_{i=1}^{N-1} \sum_{j=i+1}^N \langle r_{ij} r_{ij} / r_{ij} \rangle V'_{ij}. \quad (16)$$

The present model does not compute the MBH contribution to the stress, as this is determined during the short-time regime, characteristics of which are incorporated in the model rather than being derived properties of the model. The current model still only calculates the *thermodynamic contribution to the stress (as for the original Ermak algorithm)*, but has a more realistic prescription for the time evolution of the colloid assembly (which *does* have the effects of MBH implicit in it). The new model therefore improves the representation of the system as it evolves over the long-time regime, with consequent improvement in the thermodynamic contribution to the viscosity. The Newtonian viscosity and linear response dynamic moduli are obtained using the shear-stress autocorrelation function,  $C_s(t)$ , defined as

$$C_s(t) = \frac{N}{\rho k_B T} \langle \sigma_{xy}(0) \sigma_{xy}(t) \rangle \quad (17)$$

where  $\langle \dots \rangle$  indicates an average over time origins.

If we assume that the hydrodynamic contribution to the viscosity is equal to a constant value at all frequencies (or alternatively shear rates), i.e.,  $\eta_\infty$ , then the Green-Kubo formula in the present model gives the difference between the Newtonian viscosity  $\eta_0$  (the zero-shear-rate limit) and  $\eta_\infty$ , the infinite-frequency viscosity (or alternatively the infinite-shear-rate viscosity). The Newtonian viscosity,  $\eta_0$ , is then related to  $C_s(t)$  through

$$\eta_0 = \eta_\infty + \int_0^\infty C_s(t) dt. \quad (18)$$

We have for the dynamic moduli

$$G^*(\omega) = i\omega \int_0^\infty C_s(t)e^{-i\omega t} dt \quad (19)$$

where the complex shear modulus  $G^*(\omega) = G'(\omega) + iG''(\omega)$  (the storage and loss moduli respectively). Also the complex viscosity is given by  $\eta^*(\omega) = G^*(\omega)/i\omega$ . In order to obtain  $\eta_\infty = \eta'(\omega) = G''/\omega$  as  $\omega \rightarrow \infty$ , the system would have to be modelled realistically on the time-scale of the density fluctuations of the solvent, using for example a lattice gas treatment (Ladd 1993). The computation of the system on this time-scale is beyond the scope of the current model. Therefore all viscosities reported below are implicitly assumed to be relative to the reference 'hydrodynamic' viscosity,  $\eta_\infty$ , and are also normalized by the solvent viscosity,  $\eta_s$ . The model also does not compute the single-particle contribution to the viscosity, given by Einstein's expression  $\eta = \eta_s(1 + 2.5\phi)$  (Russel *et al* 1989) which therefore has to be included on comparison with experimental data.

The self-diffusion coefficients were obtained from the force autocorrelation function, generalizing the treatment of Åkesson and Jönsson (1985):

$$D(t) = D_0 - \frac{1}{3} \int_0^t (1 - t'/t) \langle (F(0)/\zeta(0)) \cdot (F(t')/\zeta(t')) \rangle dt'. \quad (20)$$

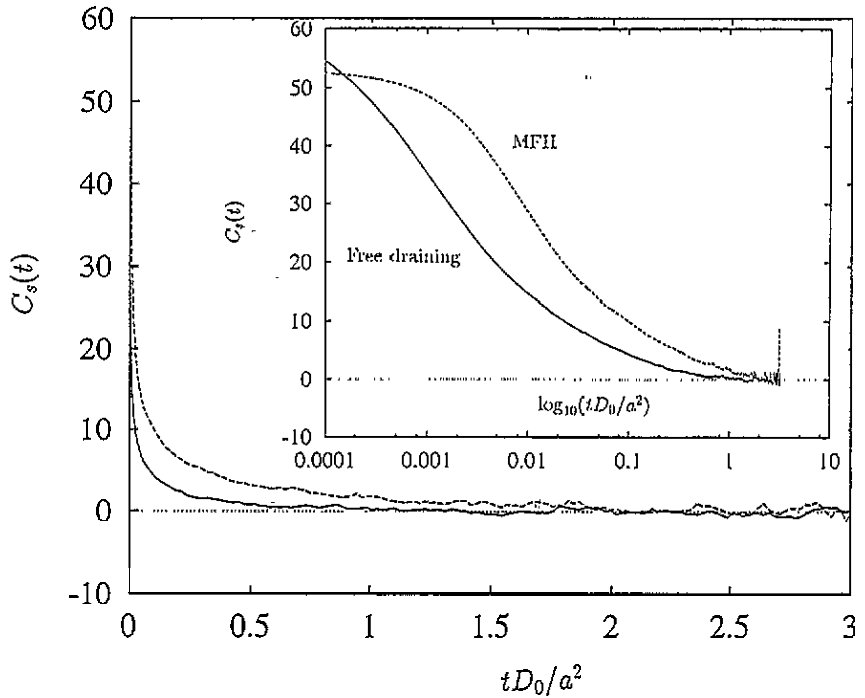
All correlation functions were computed using a fast-Fourier-transform (FFT) method (Futrelle and McGinty 1971). (The method of calculating diffusion coefficients from particle displacements cannot be cast into a working FFT expression.) Correlations were made over  $\sim 40\,000$  time steps, owing to the small time step used (the correlation functions were typically for a time of  $3.2a^2/D_0$ ). The Brownian force term in the equations of motion necessitates a very small time step, by molecular dynamics standards. The conventional multiplicative method in time (see Fincham and Heyes 1985) becomes prohibitively time consuming in this case.

#### 4. Results and discussion

The computed moduli and viscosities are given in table 1 for a selection of modelled states. Two types of simulation were performed. Mean-field-hydrodynamics computations were carried out using either equation (9) or (10). Ermak's free-draining model was also used, by setting  $\zeta = \zeta_0$  and neglecting the  $C$ -term in equation (9) or (10). The table confirms that the computed infinite-frequency dynamic moduli,  $G_\infty$ , are independent of the two algorithms considered. This is because  $G_\infty$  is a static property of the system and is therefore uniquely defined by the canonical ensemble for a given interaction potential field. If the term in  $C$  in equation (10) is omitted and yet the friction coefficient is still computed using equation (6), the static properties differ from those associated with the interaction potential, and are functions of  $C$ . The radial distribution function is also distorted, with excessive particle overlap.

**Table 1.** The infinite-frequency shear-rigidity modulus  $G_\infty$  obtained using equation (15). The parameter  $C$  is only relevant in the MBH simulations. Key:  $n$  is the exponent in the interaction potential of equation (7) and  $m$  is the exponent in the hydrodynamic interaction of equation (6). FD refers to the Ermak Brownian dynamics algorithm of equation (9) without the term in  $C$  and setting  $\zeta = \zeta_0$ . MFH refers to the mean-field-hydrodynamics version of equation (9).  $\Delta\eta = \eta/\eta_s - 1 - 2.5\phi$ . (The latter two terms account for the single-particle contribution to the viscosity.) The standard errors in the last digit are given in brackets.

$\phi$	$n$	$m$	$C$	$N$	$G_{FD}$	$G_{MFH}$	$\Delta\eta_{FD}$	$\Delta\eta_{MFH}$
0.150	36	36	6.38	256	1.84(5)	1.87(5)	0.077(3)	0.17(3)
0.3403	36	36	3.80	108	18.63(5)	18.62(5)	0.81(2)	2.39(3)
0.3403	36	18	1.33	256	18.67(5)	18.61(4)	0.91(2)	1.99(5)
0.3403	36	36	3.80	256	18.67(5)	18.76(5)	0.91(2)	2.55(5)
0.400	36	36	3.20	256	33.08(5)	33.13(5)	1.77(2)	5.69(5)
0.450	18	18	1.07	256	23.81(5)	23.79(5)	2.65(4)	12.5(2)
0.450	36	18	1.31	256	52.45(5)	52.59(5)	4.41(5)	14.1(3)
0.450	36	36	2.73	256	52.45(5)	52.68(5)	4.41(5)	14.4(3)



**Figure 1.** The shear-stress autocorrelation function  $C_s(t)$  for the state point  $N = 256$ ,  $\phi = 0.450$  and  $n = m = 36$  obtained using the two equations of motion. Key: the solid line is from the Ermak free-draining algorithm, which is given by equation (10) with  $C = 0$  and  $\zeta = \zeta_0$ ; the dashed line is for mean-field hydrodynamics (MFH) and was obtained using equation (10) with  $C \neq 0$ . The corresponding plot of  $\log_{10}(tD_0/a^2)$  is given in the inset.

In contrast to static properties, the viscosities,  $\eta$ , are quite different in the two models. The MFH values are up to three to four times larger than the free-draining values. This is because the shear-stress autocorrelation function decays more slowly for the MFH system

than for the free-draining model (see figure 1 for an example). The viscosities we calculate are proportional to the area under this curve. The free-draining-simulation viscosities are quite sensitive to the value of  $n$  in the interaction potential (they increase with  $n$ ). This sensitivity diminishes somewhat on implementation of the MFH algorithm. The values of the viscosity then become comparatively insensitive to  $n$  and  $m$ . On fixing the short-time self-diffusion coefficient of the model to the experimental value, the values of  $C$  and  $m$  tend to compensate for variations in the other parameter resulting in an insensitivity in the long-time self-diffusion coefficient (see below) and the viscosity to the  $C$ - and  $m$ -values chosen.

Table 2. As for table 1, except that the relaxation times defined by  $(\eta_0 - \eta'(\omega \rightarrow \infty))/G_\infty$  are given, from simulation and experiment (Shikata and Pearson (1994), equation (A4), for  $G_\infty$ ; Lionberger and Russel (1994) for  $\eta'(\omega \rightarrow \infty)$ ; and van der Werff *et al* (1989) for  $\eta_0(\phi)$ ).  $\tau$  is in units of  $\eta_s \sigma^3 / k_B T$ . Note that  $\tau = \Delta\eta / G_\infty$ .

$\phi$	$n$	$m$	$N$	$\tau_{FD}$	$\tau_{MFH}$	$\tau_{exp}$
0.150	36	36	256	0.042	0.091	0.200
0.3403	36	36	108	0.043	0.128	0.128
0.3403	36	18	256	0.049	0.106	0.128
0.3403	36	36	256	0.048	0.136	0.128
0.400	36	36	256	0.054	0.172	0.120
0.450	18	18	256	0.111	0.525	0.123
0.450	36	18	256	0.084	0.268	0.123
0.450	36	36	256	0.084	0.273	0.123

The shear-stress relaxation time,  $\tau = (\eta_0 - \eta'(\omega \rightarrow \infty))/G_\infty = \Delta\eta/G_\infty$ , is the area under the normalized autocorrelation function curve. Table 2 gives  $\tau$ -values computed by simulation and derived from experiment. The relaxation times are larger in the MBH case as is intuitively reasonable. One expects short-range many-body hydrodynamics to slow down local structural relaxation through the increase in the local friction coefficient  $\zeta_i(\mathbf{R})$ , which is defined in equation (6). There is still great uncertainty concerning the correct values of the viscosity and infinite-shear-rate modulus of high-volume-fraction near-hard-sphere dispersions (because it is not easy to obtain an accurate value for the volume fraction). Nevertheless, the fitted literature values, also shown in table 2, are close to the computed values, typically lying between the free-draining and current MFH values (being somewhat closer to the latter). The frequency-dependent viscosity  $|\eta^*(\omega)|/\eta_s$  shows a relative enhancement in intensity at all frequencies, but especially at low frequencies, compared with the free-draining model (see figure 2 for an example), again reflecting slower structural relaxation in the latter case.

The long-time self-diffusion coefficients,  $D_L$ , are computed from  $D(t)$  as  $t \rightarrow \infty$ . Table 3 presents the  $D_L$ -values for the free-draining (FD) and MFH models. Considering first the FD model, we have at all volume fractions  $D_s = D_0$ , the self-diffusion coefficient of the colloidal particle at infinite dilution. Although  $D_L/D_0 < 1$ , the values are quite close to the experimental values of  $D_s/D_0$ . The  $D_L$ -values computed using MFH are statistically indistinguishable from the experimental values. This suggests that the present simple implementation of short-time many-body hydrodynamics has realistic consequences for the long-time dynamics of the colloidal assembly. Structural relaxations have been slowed down by the inclusion of a  $\zeta < \zeta_0$ , which can be viewed as a decrease in the effective temperature of the Brownian particles. In fact, the values of  $D_L^{MFH}/D_L^{FD}$  are quite close to the computed value  $\zeta_0/\langle\zeta\rangle$ . In this model much of the enhancement of the



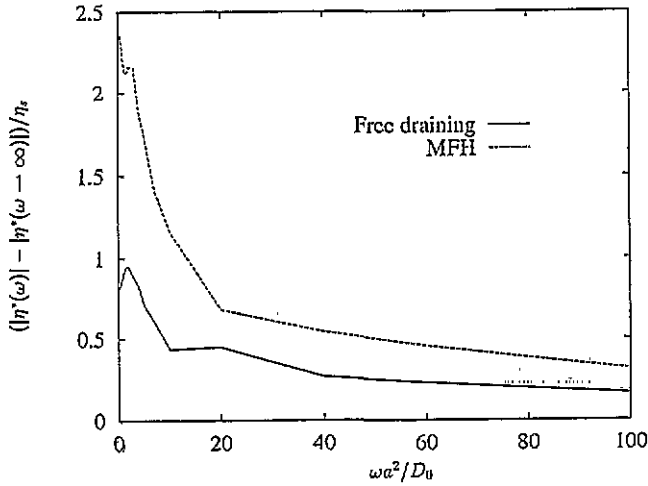


Figure 2. The absolute complex viscosity for the  $N = 256$ ,  $\phi = 0.3403$  and  $n = m = 36$  state; the key is as for figure 1.

Table 3. The long-time self-diffusion coefficients,  $D_L$ , from the long-time limit of equation (20) as  $t \rightarrow \sim 3a^2/D_0$ . The notation is as for table 1. The standard errors in the last digit are given in brackets. The experimental values of  $D_L$  come from equation (13).

$\phi$	$n$	$m$	$N$	$\langle \zeta \rangle / \zeta_0$	$D_L^{FD} / D_0$	$D_L^{MBH} / D_0$	$D_L^{exp} / D_0$
0.150	36	36	256	2.05(1)	0.735(2)	0.485(2)	0.463(5)
0.3403	36	36	108	3.75(1)	0.416(4)	0.155(3)	0.156(5)
0.3403	36	18	256	2.64(1)	0.415(4)	0.168(4)	0.156(5)
0.3403	36	36	256	3.77(1)	0.415(4)	0.147(3)	0.156(5)
0.400	36	36	256	4.50(1)	0.239(4)	0.074(3)	0.086(4)
0.450	18	18	256	4.30(1)	0.233(4)	0.041(3)	0.039(2)
0.450	36	18	256	4.10(1)	0.205(5)	0.040(3)	0.039(2)
0.450	36	36	256	5.22(1)	0.205(5)	0.041(3)	0.039(2)

viscosity can be attributed to this decrease in the self-diffusion coefficients. As noted for the viscosity, the value of the long-time self-diffusion coefficient is insensitive to the value of  $m$ , as the value of  $C$  changes to compensate in order to produce the same short-time self-diffusion coefficient.

## 5. Conclusions

We propose equations of motion that include a mean-field representation of many-body hydrodynamics, MBH. These have been implemented in a Brownian dynamics algorithm. The model is semi-empirical, in that it includes the known short-time-scale properties of near-hard-sphere colloidal liquids, and is therefore best referred to as mean-field hydrodynamics, MFH. It incorporates short-time hydrodynamics effects in the form of a local density approximation for the friction coefficient on each model colloidal particle, without requirement for a computationally expensive tensorial treatment of the system. The parameters in the mean-field model are adjusted to achieve agreement with the experimental short-time self-diffusion coefficients.

The computed viscosities of the MFH systems are typically three to four times larger than

those of the corresponding free-draining level simulations, as a result of slower structural (and therefore stress) relaxations in the systems. The colloidal assemblies are retarded in their structural evolution by virtue of the changes that we have made to the equations of motion. We find excellent agreement between experimental and computed long-time self-diffusion coefficients.

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