Spreading of colloid clusters in a quasi-one-dimensional channel

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The effect of hydrodynamic interactions on the spreading of clusters of colloid particles in a quasi-one-dimensional channel is analyzed both experimentally and theoretically. An n-particle cluster spreading diffusion coefficient is defined, in terms of the displacement $\Delta x(t)$ in time $t$, by $D_n = \left(\frac{1}{2nt}\sum_{i=1}^{n} \Delta x_i(t)^2\right)^{1/2}$, where the average is taken over all groups of $n$ adjacent particles. Our study focuses on the $n$-dependence of $D_n$ with some attention to the dependence of $D_n$ on colloid packing fraction. We find that the ratio of $D_n$ to the infinite dilution self-diffusion coefficient $D_S^0$ increases as $n$ increases, eventually saturating for large $n$. The observed dependence of $D_n$ on $n$ is in satisfactory agreement with the predictions obtained from both Stokesian dynamics simulations and hydrodynamic calculations using the method of reflections. © 2010 American Institute of Physics. [doi:10.1063/1.3330414]

I. INTRODUCTION

The restriction that particles cannot pass each other gives rise to many nonintuitive transport properties in dense one-dimensional (1D) systems. For example, diffusion of particles in 1D displays an asymptotic time dependence distinctively different from that in three-dimensional (3D). Whereas in 3D the asymptotic time dependence of the mean squared single particle displacement is proportional to time $t$, for the case that the particles confined to 1D are subject to a stochastic background and cannot pass one another, the mean squared single particle displacement $\langle \Delta x^2(t) \rangle$ increases as $t^{1/2}$ as $t \to \infty$.

Moreover, in 1D $\langle \Delta x^2(t) \rangle$ has different time dependencies in different time domains; for small $t$ one finds the normal time dependence for single particle diffusion, namely, $\langle \Delta x^2(t) \rangle \propto t$. Kollmann has shown that there is a profound connection between the diffusive motions in the short and long time domains in these 1D overdamped systems, specifically, that the long time behavior of the single particle mean square displacement is determined by the short time large-scale collective density fluctuations. For an infinite 1D system with overdamped motion the long time behavior of the probability density for displacement $\Delta x$ in time $t$ is

$$P_L(\Delta x, t) = \frac{1}{(4 \pi F_q^1/2)^{1/2}} \exp \left( - \frac{\Delta x^2(t)}{4 F_q^{1/2}} \right),$$

(1)

with $F_q$ the 1D mobility defined by

$$F_q = \frac{S(q)}{\rho} \left[ \frac{D_z(q)}{\pi} \right]^{1/2} \frac{1}{q^2},$$

(2)

and $q, \rho, \sigma, S(q)$, and $D_z(q)$ are the momentum transfer, linear density of particles, particle diameter, static structure factor and short time collective $q$-space diffusion coefficient, respectively.

The 1D diffusion of clusters of particles also has some interesting features. A study, by Kumar, of the diffusion of a cluster of $N$ hard rods on an infinite line, reveals that as $t \to \infty$ correlation between the displacement of the central particle in the cluster and the displacement of any other particle in the cluster decays exponentially with particle-particle separation, with a correlation length of the order of the cluster size. He also showed that correlations between displacements of the particles near one edge of the cluster with those near the other edge are much larger than with those near the center. It is a consequence of these correlations that the cluster expands symmetrically with time as $t^{1/2}$. We emphasize that all of the results quoted arise from the restriction that 1D particles cannot pass one another; generation of a large particle displacement then requires cooperative particle motion.

We also note that these analyses describe aspects of diffusive motion in a 1D system without hydrodynamic interaction between the particles.

A common experimental realization of a 1D system is a quasi-1D (q1D) colloidal suspension in a narrow channel. In such a system there are, in addition to direct colloid-colloid interactions, hydrodynamic particle-particle and particle-wall interactions carried by the solvent. Under all common experimental conditions hydrodynamic interactions between the colloid particles in the suspension are established on a short time scale compared to the time between collisions, and evidence for cooperative motion in a q1D colloidal fluid can be found at a time much smaller than the time between collisions. A crossover in the time dependence of the mean squared single particle displacement between the long time and short time behavior occurs at about the time between collisions.

In this paper, we use a suspension of colloid particles confined to a q1D channel to study the influence of hydrodynamic particle-particle and particle-wall interactions on the short time diffusive behavior of clusters of particles. Our
The study augments previous reports in three ways. First, our experiments examine the diffusive behavior of clusters of various sizes that are surrounded by other particles. The extant theoretical studies analyze the behavior of clusters that are isolated on the 1D line. Second, our experiments focus on the attention on cluster diffusion in the short time limit wherein the single particle mean squared displacement satisfies \( \langle \Delta x^2(t) \rangle \sim t \). Aside from the relationship between short and long time behaviors of the mean square particle displacement derived by Kollmann, the extant theoretical studies focus the attention on the behavior of a 1D system in the limit \( t \to \infty \). Third, our studies examine the role of hydrodynamic coupling between particles and between particles and the walls in determining the cluster diffusion in the short time regime. The extant theoretical studies analyze systems without hydrodynamic coupling.

There are many possible ways of characterizing cluster motion. Here we consider the \( n \)-dependence of the \( n \)-cluster spreading diffusion coefficient \( D_n \) (see below for the definition) and the dependence of \( D_n \) on colloid packing fraction \( \eta \). This diffusion coefficient measures the rate of spreading of \( n \) adjacent particles (see Fig. 1 for a top view of a seven-particle cluster); by definition, in the absence of hydrodynamic interactions \( D_n \) becomes equal to the single particle self-diffusion coefficient \( D_S(\eta=0)=D_S^0 \) so departure of the ratio \( D_n/D_S^0 \) from unity is directly attributable to hydrodynamic interactions. We find that at fixed packing fraction \( D_n/D_S^0 \) increases as \( n \) increases, saturating at a value of \( D_n/D_S^0=1.19 \) when the packing fraction is \( \eta=0.17 \), and \( D_n/D_S^0=1.42 \) when the packing fraction is \( \eta=0.61 \). As to the packing fraction dependence of \( D_n \), we find that for \( n \gg 1 \) the cluster spreading diffusion coefficient increases as the colloid packing fraction is increased even though \( D_n(\eta) \) decreases as the packing fraction increases.

The theoretical analysis reported in this paper models the experimental system, which is a colloid suspension in a square open top channel, as a colloid suspension confined in a cylindrical capillary; this model has been shown to provide a good description of the center of mass and relative pair diffusion coefficients and of the concentration dependence of the single particle diffusion coefficient in the same q1D system used for our studies.\textsuperscript{12-15} We find, for the model system, that the values for \( D_n/D_S^0 \) predicted by both Stokesian dynamics (SD) simulations and hydrodynamic calculations based on the method of reflections are in satisfactory agreement with experiment.

II. EXPERIMENTAL DETAILS

Since the procedures used in our study have been reported elsewhere,\textsuperscript{12} here we only briefly sketch the description of the q1D experimental system. It consists of a water suspension of silica spheres (diameter \( \sigma=1.58 \pm 0.04 \mu m \), confined in a straight or a circular q1D channel printed on a polydimethylsiloxane substrate. The straight channel is 3 ± 0.3 \( \mu m \) wide, 3 ± 0.3 \( \mu m \) deep, and 2 mm long; the circular channel is 3 ± 0.3 \( \mu m \) wide, 3 ± 0.3 \( \mu m \) deep, and has a circumference of 220 \( \mu m \). A 100 \( \mu m \) thick drop of suspension is enclosed between the polymer mold and a cover slip so that the top of the channel is open to a layer of fluid. Digital video microscopy was used to determine the time-dependent colloid particle trajectories within the focal plane with time resolution 0.033s. In earlier studies we showed that the colloid particles in the q1D system are tightly confined to the centerline of the channel and float slightly above the bottom of the channel. We have measured the particle displacements along the axis of the channel at different concentrations, characterized by the 1D packing fraction \( \eta=Na/L \), where \( L \) is the length of the channel. Our results show no difference between the time-dependent particle displacements determined for the straight and circular channels.

III. REPRESENTATION OF THE EXPERIMENTAL DATA

We focus our attention on the diffusive spreading of the particles in a cluster of \( n \) particles. In this paper we define an \( n \)-cluster to be any \( n \) consecutive particles along the line irrespective of their separations and irrespective of the existence of other particles to the left or right of the selected set. The \( n \)-particle cluster spreading diffusion coefficient, defined in terms of \( i=1, \ldots,n \) adjacent particles that have individual particle displacements \( \Delta x_i(t) \) in time \( t \) for small \( t(t \ll t_{\text{collision}}) \), is

\[
D_n = \frac{1}{2nt} \left( \sum_{i=1}^{n} \Delta x_i(t) \right)^2 .
\]

The average is taken over all groups of \( n \) adjacent particles along the axis of the channel. Note that this \( n \)-particle cluster diffusion coefficient is not the same as the cluster center of mass diffusion coefficient,

\[
D_{cm} = \frac{1}{2t} \left( \frac{1}{n} \sum_{i=1}^{n} \Delta x_i(t) \right)^2 .
\]

After some simple manipulations we find that

\[
D_n = \frac{1}{2nt} \left( \sum_{i=1}^{n} \Delta x_i(t) \right)^2 = \frac{1}{n} \sum_{i,j=1}^{n} D_{ij} ,
\]

with \( D_{ij}=(\Delta x_i \Delta x_j)/2t \). From Eq. (5) we see that if hydrodynamic interactions are turned off \( D_{ij}=D_S^0 \), \( D_{ij}=0 \) for \( j \neq 1 \), and \( D_{ij}=D_S^0 \). We display in Fig. 2 the experimentally determined \( n \)-dependence of \( D_n \) for two packing fractions. The three important features of these data are the increase of \( D_n \) with \( n \), its saturation at large \( n \), and the increase of \( D_n \) with packing fraction. The limiting values of \( D_n/D_S^0 \) are \( \lim_{\eta \to \infty}(D_n/D_S^0) =1.42 \) when \( \eta=0.61 \) and \( \lim_{\eta \to \infty}(D_n/D_S^0)=1.19 \) when \( \eta=0.17 \).
FIG. 2. The n-dependence of the n-particle cluster spreading diffusion coefficient for two q1D packing fractions. The experimental data at each packing fraction are fit to the functional form $D_n/D_n^0 = A(1 - B/n)$, with parameters A and B discussed in the text.

IV. THEORETICAL CALCULATION OF THE n-PARTICLE CLUSTER DIFFUSION COEFFICIENT

A. General remarks

Computation of the contribution of hydrodynamic interactions to the terms in the sum in Eq. (5) is complicated by the fact that the n particles in a cluster have different numbers of neighbors; in any given cluster of size $n > 5$ there is at least one particle in the middle, which has nearest and next nearest neighbors on both sides, and particles at the ends that have either only a nearest or nearest and next nearest neighbor on one side. Thus, summing over the terms in Eq. (5) requires consideration of end corrections. Because in a q1D system hydrodynamic screening cuts off the effective interaction on the scale length of the channel width, which is somewhat more than a particle diameter, we need not consider end effects beyond two particle diameters.

We have previously computed the hydrodynamic interactions between two and three particles and between these particles and the wall in a q1D colloid suspension; these calculations provide the information necessary for computation of $D_n$ for $n=2$ and 3. Rather than extend those calculations to n-particle clusters we introduce an approximation based on summing nearest neighbor and next nearest neighbor interactions in an n-particle cluster. This approximation takes advantage of the screening of the hydrodynamic interactions and uses the notion of effective end particles. The number of effective end particles is not necessarily an integer since it incorporates the differences between end and next-to-end hydrodynamic interactions. The number of effective end particles is determined by fitting the experimental data with a form that we now derive.

For an n-particle cluster (see Fig. 1 for a seven-cluster example), the cluster spreading diffusion coefficient can be written as

$$D_n = \frac{1}{n} \sum_{i,j=1}^{n} D_{ij} = \frac{1}{n} \sum_{i=1}^{n} \left( \sum_{j=1}^{n} D_{ij} \right).$$

Taking advantage of the hydrodynamic screening in the q1D system, for large n it is a good approximation to assume that the pair distribution function $g_2(x)$ for two particles with separation x in the cluster is the same as the pair distribution function for the bulk q1D system for which there is an analytical form giving the functional dependence on x and the density of the system. Then for a particle in the middle of the cluster (labeled 1) we have

$$\sum_{j=1}^{n} D_{1j} = D_{11} + 2\eta \int_{1}^{\infty} g_2(x)D_{1j}(x)dx,$$

and for a particle at the end (labeled 2) we have

$$\sum_{j=1}^{n} D_{2j} = D_{22} + \eta \int_{1}^{\infty} g_2(x)D_{2j}(x)dx.$$

If we assume that there are $m$ effective particles at the ends of a large cluster, and that $m$ is independent of $n$, Eq. (6) can be reduced to

$$D_n = \frac{1}{n} \left[ nD_{11} + (n-m)2\eta \int_{1}^{\infty} g_2(x)D_{1j}(x)dx + m\eta \int_{1}^{\infty} g_2(x)D_{1j}(x)dx \right]$$

$$= D_{11} + \left( 1 - \frac{m}{2n} \right) 2\eta \int_{1}^{\infty} g_2(x)D_{1j}(x)dx.$$

From Eq. (9) we see that the n-particle cluster spreading diffusion coefficient, normalized by the infinite diffusion limit of the self-diffusion coefficient, will approach its large n limit rather slowly, namely, as

$$\frac{D_n}{D_S^0} = A(1 - B/n),$$

where $A = \sum_{j=1}^{n} D_{1j}/D_S^0 = \lim_{n \to \infty} (D_n/D_S^0)$ and $B = m(A - (D_S(\eta)/D_S^0))/2A$ [note that $D_S(\eta) = D_{11}$]. Because of the end effects, the diffusive spreading of the particles in the cluster increases as the size of the cluster increases, despite the fact that the hydrodynamic interaction in our q1D system is screened on the length scale of the channel width.

B. Hydrodynamic calculations

We now turn to the hydrodynamic calculation of the n-cluster spreading diffusion coefficient using the so-called method of reflections. Before describing this analysis it is important to remind the reader that the experimental studies were carried out in an open square channel. We have argued in an earlier paper that the principal effects of the hydrodynamic coupling between particles in the open square channel are correctly captured when one calculates those effects in a closed cylindrical channel, since there must be a surface with zero tangential fluid velocity that connects the channel lips in the experiment. Although the shape of that surface remains unknown to us, the use of a closed cylindrical capillary in place of the open square channel retains the essential boundary conditions of the experimental system. Specifically, in our previous theoretical work the q1D colloid system was confined in a cylindrical capillary with radius $R_0$ and length $L \gg R_0$, inside of which is a viscous fluid contain-
TABLE I. Values of $A$, $B$, and $m$ obtained from the fit of the experimental and simulation data to Eq. (4). The values of $\lim_{n \to \infty} (D_n/D_S^n)$ are obtained from Eq. (5).

<table>
<thead>
<tr>
<th>$\eta$</th>
<th>$a/R_0$</th>
<th>$A$</th>
<th>$B$</th>
<th>$m$</th>
<th>$\lim_{n \to \infty} (D_n/D_S^n)$</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.17</td>
<td>0.15</td>
<td>1.19</td>
<td>0.183</td>
<td>2</td>
<td>1.13</td>
</tr>
<tr>
<td>0.61</td>
<td>0.15</td>
<td>1.42</td>
<td>0.8</td>
<td>3</td>
<td>1.57</td>
</tr>
<tr>
<td>0.4</td>
<td>0.216</td>
<td>1.21</td>
<td>0.23</td>
<td>2.2</td>
<td>1.22</td>
</tr>
</tbody>
</table>

For $\eta=0.17$, $a/R_0=0.15$, $\lim_{n \to \infty} (D_n/D_S^n)=1.13$ when $\eta=0.17$ and $\lim_{n \to \infty} (D_n/D_S^n)=1.57$ when $\eta=0.61$. These values for $\lim_{n \to \infty} (D_n/D_S^n)$ are to be compared to the values obtained from fitting the experimental data (see Fig. 2 for the fit) to the functional form in Eq. (10), and find $A = \lim_{n \to \infty} (D_n/D_S^n)=1.19$ for $\eta=0.17$ and $A = \lim_{n \to \infty} (D_n/D_S^n)=1.42$ for $\eta=0.61$. The results are summarized in Table I.

C. SD simulations

We have also carried out SD simulations of the influence of hydrodynamic interactions on the motions of the particles in a q1D colloid suspension. Following the work of Brady and co-workers,17,18 the influence of hydrodynamic interactions is incorporated in our SD simulation via the values of the diffusion coefficients. Starting with the Langevin equation, the particle displacement $\Delta x_i(t)$ is determined from

$$\Delta x_i(t) = \sum_j \frac{\partial D_{ij}}{\partial x_j} \Delta t + \sum_j \frac{D_{ij} F_j}{kT} \Delta t + R_i(\Delta t),$$

in which $R_i(\Delta t)$ is a random displacement with a Gaussian distribution that has 0 average and a variance-covariance $\langle R(\Delta t) R(\Delta t) \rangle = 2D_i \Delta t$. The particle displacements satisfy the multivariate Gaussian distribution function whose moments are specified by

$$\langle \Delta x_i(\Delta t) \Delta x_j(\Delta t) \rangle = 2D_{ij} \Delta t.$$

In Eqs. (12)–(14) $F_i$ is the sum of interparticle and external forces acting on particle $i$, and all variables are to be evaluated at the beginning of the time step. The particle-particle and particle-wall hydrodynamic interactions are incorporated in the calculation of the diffusion coefficient $D_{ij}$ which is carried out analytically by the method of reflections. With these values of $D_{ij}$ we generate displacements for the particles.

In our SD simulations we used $N=80$ particles of unit diameter on a straight line of length 200, with periodic boundary conditions; the packing fraction was taken to be 0.4 and the ratio of particle radius to cylinder radius was taken to be $a/R_0=0.216$. The bare particle-particle interaction was represented by the continuous but nearly hard rod potential $U(x) = C(x-1/2)^2$, with $C=2 \times 10^{-19}$ and $\gamma=64$. After equilibration, 12 000 time steps ($\Delta t = 1.25 \times 10^{-4}$) were taken and particle configurations at each time step were recorded for evaluation. Values for the set of $N$ displacements $\{R_i\}$ were generated using a multivariate normal generator applying the Rotational Method described in Ref. 19. This method utilizes the fact that the $k \times k$ variance-covariance matrix $C$ is positive definite and symmetric. So if we generate $k$ independent univariate random variables $Y_k = (Y_1, Y_2, \ldots, Y_k)$, we can obtain a random sample of X $= Y P^T$ which has a $k$-dimensional normal distribution with 0 mean and variance-covariance matrix $C$, where $P^T C P = I$.

In Fig. 3 we display the results of the SD simulations for the $n$-particle cluster spreading diffusion coefficient, normalized by the infinite dilution self-diffusion coefficient, as a function of $n$. We have run our SD simulation 200 times, and each SD run recorded 80 particle trajectories over 12 000 time steps which results in an overall statistical error of our numerical simulation of less than 1%. The data points with $6 \leq n \leq 20$ are fit to the functional form $y = A (1-B/n)$ with $A=1.21$ and $B=0.23$. These values of $A$ and $B$ correspond to $A = \lim_{n \to \infty} (D_n/D_S^n)=1.21$ and $m = 2AB/(A-[D_n(0.4)/D_S^n])=2.2$ (we used the experimental values $D_0=0.105$ $\mu m^2/s$ and $D_S^n=0.11$ $\mu m^2/s$, see Ref. 14). The computed value of $\lim_{n \to \infty} (D_n/D_S^n)$ is to be compared to the value obtained from Eq. (11), namely, $\lim_{n \to \infty} (D_n/D_S^n)=1.22$ (see Table I). The value of $m$ found from the fitting $m=2.2$ implies that there is just one effective particle at each end of the cluster (particles second or third from the end contribute little to this end effect). From the data displayed we see that the interpolation formula fits the simulation re-
results very well, even at small $n$ where our approximation becomes less accurate. The concordance of the results for $D_n$ validates the approximations used in the calculations based on the method of reflections.

As already noted, for the calculations reported in this paper we have chosen a single packing fraction $\eta = 0.40$ as representative of the packing fraction range sampled in the experiments. The intent is to provide a theoretical interpretation of the observations and not to attempt a prediction that quantitatively reproduces the experimental data. Comparison of the predicted and measured behavior of $D_n$ is complicated by the difference between the open square channel used in the experimental studies and the cylindrical capillary used in the theoretical studies. Although we have argued that the principal effects of the hydrodynamic coupling between particles in the open square channel are correctly captured when one calculates those effects in a closed cylindrical channel, the sizes of experimental channel and the theoretical capillary are different, which makes it difficult to map the colloidal packing fraction from the square channel to the capillary.

V. PACKING FRACTION DEPENDENCE OF $D_n$

In a previous study\textsuperscript{14} we calculated the density dependence of the self-diffusion coefficient $D(\eta)$ to first order in the packing fraction for particles diffusing along the central axis of a cylindrical capillary. The coefficient $\alpha$ in the expansion $D(\eta) = D^0_\infty (1 + \alpha \eta + O(\eta^2))$ depends on the ratio $a/R_0$. Our earlier calculation is valid only for a dilute system in that it accounted for the hydrodynamic interactions between two particles separated by distance $x$ up to order $(a/x)^2$, and it assumed that the pair correlation function is independent of particle separation. We have extended our calculations of $\alpha$ to terms up to order $(a/x)^7$. The results obtained are listed in Table II. We now evaluate the packing fraction dependence of the large $n$ limit of the $n$-particle cluster spreading diffusion coefficient to first order in $\eta$.

Using Eq. (9), the large $n$ limit of the $n$-particle cluster diffusion coefficient is

$$\lim_{n \to \infty} D_n(\eta) = (1 + \alpha \eta) D^0_\infty + 2 \eta \int_1^\infty \frac{D_1(\eta = 0, x) dx + O(\eta^2)}{x}.$$  

(15)

Since $D_1(\eta)$ and $D_{12}$ are known from prior calculations, evaluation of Eq. (13) yields $\lim_{n \to \infty} D_n(\eta) = D^0_\infty + \beta \eta + O(\eta^2)$. The calculated values of $\beta$ are shown in Table II for different $a/R_0$. The results show that the $n$-particle cluster spreading diffusion coefficient increases as the packing fraction increases, signaling an enhanced rate of collective diffusion in the face of decreasing rate of single particle diffusion. Note also that as $a/R_0$ increases, $\beta$ decreases, suggesting that the influence on $D_n$ of the wall-particle hydrodynamic coupling becomes more important, and the influence on $D_n$ of the particle-particle hydrodynamic coupling becomes less important.

VI. COMMENTS

The unusual properties of diffusion of single particles and of clusters of particles in a 1D system were predicted from the results of studies of models that did not include hydrodynamic interactions. The finding, by Kumar, that as $t \to \infty$ correlations between displacements of the particles near one edge of a cluster with those near the other edge are much larger than with those near the center, so that the cluster expands symmetrically with time as $t^{1/2}$, and the finding by Kollmann, that the long time behavior of the single particle mean square displacement is determined by the short time large-scale collective density fluctuations, has prompted us to examine how hydrodynamic interaction affects the $n$-particle short time cluster spreading diffusion coefficient in a 1D colloid suspension. Given that in the absence of hydrodynamic coupling $D_n$ is equal to the one-particle self-diffusion coefficient, and at small particle separation the 1D relative pair diffusion coefficient is smaller than the one-particle self-diffusion coefficient, the naive expectation is that $D_n$ will decrease as $n$ increases at fixed packing fraction.

We find the reverse behavior, namely, that with fixed packing fraction $D_n/n^2$ increases as $n$ increases, eventually saturating at large $n$. We also find that for $n \gg 1$ $D_n$ increases as the colloidal packing fraction is increased even though $D_1(\eta)$ decreases as the packing fraction increases. Both properties are...
directly traceable to the effect of hydrodynamic interactions between the colloid particles and between the colloid particles and the walls of the q1D channel, as shown by the hydrodynamic calculations and the SD simulations reported in Sec. III. We showed previously that the packing fraction dependences of the self-diffusion and the pair diffusion coefficients of a q1D liquid are accounted for quantitatively by the screened hydrodynamic interactions between the colloid particles. Yet, despite the restriction of the length scale of the hydrodynamic coupling between a pair of particles to the width of the channel, the $n$-particle cluster spreading diffusion coefficient exhibits the effects of longer-ranged collective behavior.

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