I. INTRODUCTION

The diffusion and spreading of polymers and other large molecules on solid surfaces is an interesting theoretical problem with important applications related to thin surface films. Many experimental studies have been carried out on the diffusive dynamics of polymers and smaller molecules in such systems [1–9]. However, most theoretical investigations to date mostly deal with the (tracer) diffusion of single atoms or molecules [10–15], and there have been only relatively few theoretical studies concerning the collective diffusion properties of larger molecules or polymers on surfaces [16–20]. In the case of a finite surface coverage or number density \( \rho \), difference must be made between the tracer and collective diffusion coefficients \( D_T(\rho) \) and \( D_C(\rho) \), respectively, even if the only interaction is site blocking. The existing studies have shown that while the monotonically decay of \( D_T(\rho) \) as a function of \( \rho \) can be qualitatively understood by blocking in the case of athermal polymers, the behavior of the density fluctuations and thus \( D_C(\rho) \) is nontrivial. There are strong entropic interactions present that influence \( D_C(\rho) \) and the spreading dynamics of flexible chainlike molecules on smooth surfaces in the lattice-gas limit [16,17,19,20]. In particular, for such molecules \( D_C(\rho) \) typically displays a maximum value at some intermediate densities, such that the relative magnitude of the maximum decreases for stiffer chains, or in the presence of attractive interchain interactions. The maximum is due to the competition between the compressibility and mobility of the overlay, both of which are strongly dependent on the density.

A particularly interesting case occurs if the molecules on the surface can be approximated by orientationally symmetric, circular particles. Such systems are also relevant for the diffusion of geometrically confined colloidal systems which have been studied recently [21–25] as an extension of the classical problem of dynamics of three-dimensional (3D) colloidal systems [26–34]. In the 2D case, the equilibrium properties of hard and interacting disks have been studied since the 1960’s [35–44] and are rather well understood although some open questions remain concerning, e.g., the nature of the freezing transition. The dynamical properties of 2D (colloidal) liquids have also been considered in some works [39,45,30,41,46–48], but detailed studies have not been carried out throughout the density range.

In this paper we present results of numerical and analytic studies of diffusion in a very simple model system of 2D Brownian hard spheres (disks) and rigid rodlike molecules in two dimensions. We consider the case where there are no explicit particle-surface interactions. The present study thus complements the work done previously on flexible chainlike molecules using the fluctuating bond (FB) lattice model with Monte Carlo dynamics, which corresponds to the lattice-gas limit with a strongly attractive surface potential that confines the chains on the lattice sites [16,19]. Here we focus on the behavior of the various relevant diffusion coefficients as a function of the number density of surface particles \( \rho \) in the submonolayer regime. For the hard disk case, we find that while \( D_T(\rho) \) decreases monotonically up to the freezing transition, \( D_C(\rho) \) is wholly determined by the inverse compressibility which increases rapidly on approaching freezing. We also study memory effects associated with tracer diffusion, and present theoretical estimates of \( D_T(\rho) \) from the mode-mode coupling approximation. In the case of rigid rods with a short-range repulsion we find that both \( D_T(\rho) \) and \( D_C(\rho) \) behave in a manner very similar to the case of individual disks when there is no orientational ordering.
II. THE MODEL

A. Hard disks

The model system we consider here consists of an ensemble of 2D hard disks which can be thought of being confined to a smooth and structureless surface, with no explicit particle-surface interaction potential. Since for hard disks temperature does not play any role, the only relevant parameter is the scaled particle density $\rho_s = \rho \sigma^2$, where $\rho = N/L^2$ is the number density of $N$ disks in a system of linear size $L$, and $\sigma$ is the disk diameter.

In the present work for numerical studies we use a combination of molecular dynamics (MD) and Monte Carlo (MC) simulation techniques. All dynamical quantities here have been computed with MD, while MC sampling has been used to obtain static thermodynamic averages. With MD, it is convenient to use a soft interaction potential between the particles rather than the infinite hard-sphere potential. The hard-sphere results can, however, be recovered by doing a simple rescaling of the density if we use a soft potential of the form \[ V(r) = \epsilon \left( \frac{\sigma}{r} \right)^n \],

where $r$ is the separation between the particles, and for $n$ we chose here the value of 12. The rescaling in density required in order to obtain the corresponding hard sphere results is then

\[ \bar{\rho} = \left( \frac{\epsilon}{k_B T} \right)^{2/n} \rho_s, \]

where $T$ is the temperature of the soft-sphere system. Here, we have used $\epsilon = 4.0$ and $k_B T = 0.1402$. In the simulations a cutoff in the potential at $2.5 \sigma$ was used.

The equation of motion of each particle in the system is given by the Langevin equation [49]:

\[ \frac{d}{dt} \bar{\rho}(t) = - \eta \bar{\rho}(t) + \bar{f}(t) + \bar{F}(t), \]

where $\bar{\rho}(t) = m \bar{v}(t)$ is the 2D momentum of the particle, $\eta$ is the friction coefficient, $t$ is time, and $\bar{f}(t)$ is the total interaction force with the other particles. The remaining random term $\bar{F}(t)$ is the driving force for the motion of the Brownian particles, and it obeys the standard fluctuation-dissipation relation [49]. In our MD simulations the equation of motion is integrated using the Velocity Verlet algorithm [49]. The system size used in most cases was $L = 100 \sigma$ with periodic boundary conditions.

The MC simulations have been carried out in such a way that a randomly chosen particle is displaced at a randomly chosen position. In this case, it is easy to carry out the simulations for hard particles by using the standard Metropolis acceptance criterion [50] according to which a move is always accepted if it does not result in an overlap between the particles. In our MC simulations the typical system size was $L = 240 \sigma$ with periodic boundary conditions. We checked for finite size effects by evaluating the results in several subsystems of the total system and then extrapolating to infinite size [40].

In 2D, it has been suggested that there is a first-order freezing transition from a fluid to a solid phase at a critical density of $\rho_{c,2} = 0.887$ [40,41], however, the most recent simulations suggest [44] that for the 2D hard disk case the freezing transition is of the Kosterlitz-Thouless-Halperin-Nelson-Young (KTHNY) type [51]. For $n = 12$ in the potential we have used in the MD simulations the freezing occurs at a higher density of $\bar{\rho}_{c,2} = 0.986$ [41]. The exact nature of the freezing transition for the $n = 12$ case in unclear [41], although simulations again suggest the KTHNY picture [42]. However, in the present work we concentrate on diffusion within the liquid phase.

B. Rigid rodlike molecules

The second system we consider here is a simple model of rigid, rodlike molecules in 2D. They are modeled by a chain of $N_c$ particles in continuum which are constrained to stay in fixed positions with respect to each other along a straight line (see Fig. 1). This constraint was implemented in the MD simulations by first computing the forces acting on all the $N_c$ separate particles comprising the molecule, and then calculating from these the torque and the translational force acting with respect to the center of mass. Following this, the molecule was rotated accordingly and its position updated. The interchain interaction was chosen to be of the form of Eq. (1), with the same parameters as for the single particles. This means that each particle in the chain interacts with all the particles in the other chains through a strongly repulsive $1/r^{12}$ potential. This effectively prevents chain overlap with the parameters used here. In the MC simulations we simply randomly displaced and rotated each rod and used the Metropolis criterion to accept or reject the new configuration.

The phase diagram of rodlike molecules is in general more complicated than that of the single particles due to the additional orientational degrees of freedom. In addition to
the liquid and solid phases there is also the possibility of a nematic phase. Recent careful simulation studies of 2D hard rods have shown that for aspect ratios of six or less the nematic phase is not realized [52]. Although our model is not in the hard potential limit, nor does the rescaling of Eq. (2) work, we did not observe any evidence of a nematic phase in our simulations below the freezing transition.

III. RESULTS FOR HARD DISKS

A. Numerical results for tracer diffusion

The tracer diffusion coefficient of individual particles $D_T$ is defined as

$$D_T = \frac{1}{2d} \lim_{t \to \infty} \left( \frac{1}{N} \sum_{i=1}^{N} \left( \vec{r}_i(t) - \vec{r}_i(0) \right)^2 \right)$$

$$= \frac{1}{d} \int_{0}^{\infty} \left( \frac{1}{N} \sum_{i=1}^{N} \vec{v}_i(t) \cdot \vec{v}_i(0) \right) dt,$$  \hspace{1cm} (4)

where $d$ is the dimension of the system and $\vec{r}_i(t)$ is the position vector of particle $i$ at time $t$, and $\vec{v}_i(t)$ its velocity. The quantity $\phi(t) = \langle \vec{v}_i(t) \cdot \vec{v}_i(0) \rangle$ is the velocity autocorrelation function associated with the tracer particle. In evaluating the diffusion coefficients during the simulations we employ the memory expansion method [53] which has been shown to be more efficient than using the expressions above.

In the case of 2D hard disks, the only relevant parameter that $D_T$ depends on is the scaled density $\bar{\rho}$. Previously, the tracer diffusion coefficient has been studied as a function of density for a few special values of $\bar{\rho}$ [54,41]. $D_T(\bar{\rho})$ has been found to be a monotonically decreasing function of density from its initial Brownian value of $D_T(0) = k_BT/m \eta$ ($5.14 \times 10^{-3}$ in our units), as could be expected from simple blocking or backscattering arguments [55]. In particular, at $\bar{\rho}_c$, Löwen has proposed a dynamical freezing criterion which states that the ratio $D_T(\bar{\rho}_c)/D_T(0)$ attains a universal value of 0.086±0.01 at freezing.

Our MD simulation results for $D_T(\bar{\rho})$ are listed in Table I and shown in Fig. 2. We find very good agreement with the previous numerical results, as can be seen from Fig. 2. The tracer diffusion coefficient $D_T(\bar{\rho})$ is a decreasing function of density within the liquid phase, and shows a more rapid decrease close to the freezing transition. We have tested Löwen’s freezing criterion at $\bar{\rho}_c = 0.986$ and find that $D_T(\bar{\rho}_c)/D_T(0) = 0.0759\pm0.0004$ which is in very good agreement with the overall Löwen criterion, but somewhat smaller than the value of 0.099±0.003 reported by Löwen for the 1/r potential [41]. We note that from our data it seems that $D_T(\bar{\rho})$ approaches zero continuously at $\bar{\rho}_c$, which would indicate a continuous freezing transition for the case of the 1/r potential [41]. However, we have not attempted a systematic finite-size study in the vicinity of $\bar{\rho}_c$ to determine the nature of the freezing transition.

B. Velocity autocorrelation functions

As can be seen from the definition of $D_T$ in Eq. (4), the velocity autocorrelation function $\phi(t)$ is the fundamental quantity in diffusion. The time dependence of $\phi(t)$ has been studied in dense 3D Brownian liquids [55–57]. It was found that the deviations from exponential behavior were small, but increased with increasing density and in particular near the freezing transition [55]. More recent studies [56,29] have demonstrated that the temporal decay of $\phi(t)$ does not seem to follow simple exponential behavior, but rather a stretched exponential form. Recently, the temporal behavior of $\phi(t)$ (and its associated memory function to be defined below) have been studied in detail in a variety of strongly interacting 2D dissipative systems [53,20,58,59], including a dense, viscous hard disk fluid [46]. It has been demonstrated that in many cases, $\phi(t)$ displays an intermediate time power law decay $\propto t^{-\alpha}$, where the value of the effective exponent $\alpha$ can be related to interaction and ordering effects in the system [58]. In particular, if there are no ordered phases present, the value of $\alpha$ is typically about or larger than two for strongly repulsive and less than two for attractive systems.

In the present case we have studied this issue through the temporal behavior of $\phi(t)$ and the corresponding memory function $M(t)$ defined through [20]

$$\frac{d\phi(t)}{dt} = i\Omega_0 \phi(t) - \int_{0}^{t} M(t-s) \phi(s) ds,$$ \hspace{1cm} (5)

where $i\Omega_0$ is the so-called frequency variable which vanishes in continuum. We find that for smaller densities, $\phi(t)$

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**TABLE I.** Numerical values of the normalized tracer diffusion coefficients for 2D hard disks from MD simulations

<table>
<thead>
<tr>
<th>$\bar{\rho}$</th>
<th>$D_T(\bar{\rho})/D_T(0)$</th>
<th>$\bar{\rho}$</th>
<th>$D_T(\bar{\rho})/D_T(0)$</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.000</td>
<td>1.000</td>
<td>0.000</td>
<td>1.000</td>
</tr>
<tr>
<td>0.110</td>
<td>0.829(1)</td>
<td>0.220</td>
<td>0.6900(8)</td>
</tr>
<tr>
<td>0.329</td>
<td>0.5762(5)</td>
<td>0.549</td>
<td>0.3996(9)</td>
</tr>
</tbody>
</table>

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**FIG. 2.** The normalized tracer diffusion coefficient for 2D hard disks as obtained from MCA calculations and from the MD simulations. The open circles have been obtained using Eq. (10) and the lines are spline fit guides to the eye.
shows rather small deviations from exponential behavior indicating weak memory effects. However, at larger densities there is an intermediate power law type of behavior which can be well fitted by [58,59]
\begin{equation}
\phi(t) = \frac{\phi(0)}{1 + At^x},
\end{equation}
which leads to an algebraic decay \( \phi(t) \sim t^{-x} \) for \( At^x > 1 \). In Fig. 3 we show the normalized velocity autocorrelation function as a function of time, and the inset shows the corresponding memory function. The topmost data set in the figure has been taken at the onset of the freezing transition at \( \tilde{\rho} = 0.986 \). Fitting an effective power law to \( \phi(t) \) gives \( x = 1.2 \) which is close to the values reported in the literature [46,58]. Due to the relatively short range of the effective power law, it is difficult to pin down the value of \( x \) accurately, however.

### IV. ANALYTIC APPROXIMATIONS FOR DIFFUSION COEFFICIENTS

#### A. Tracer diffusion

There exists a variety of theoretical approaches in order to quantitatively understand the density dependence of \( D_T \) for colloidal systems, with and without hydrodynamic interactions [26–28,30,32,34]. The case of a 2D Brownian liquid considered here has also been tackled recently [45]. One of the most commonly used approaches for calculation of transport coefficients is based on the mode-mode coupling approximation (MCA) [60]. Recently, the MCA was applied to the 3D hard Brownian sphere case by Indrani and Ramaswamy (IR) [30]. Here, we generalize their theory to the 2D case. Within the MCA, \( D_T \) is given by
\begin{equation}
D_T = \frac{D_0}{1 + \Sigma_0},
\end{equation}
where the self-energy \( \Sigma_0 = \Sigma(k = 0, z = 0) \) is obtained from the equation
\begin{equation}
\Sigma(k, z) = \frac{1}{8 \pi^2 \rho} \int_0^\infty e^{-z} v(q) C_s([k - \hat{q}], t) C_s(q, t) dt d^2q.
\end{equation}
The interaction vertex is given by \( v(q) = q^2 [1 - S^{-1}(q)]^2 \), where \( S(k) \) is the static structure factor. The Laplace transforms of the self-density and collective-density correlation functions are given by
\begin{equation}
C_s(k, z) = \frac{S(k)}{k^2 S^{-1}(k)} \left( z + \frac{1}{1 + \Sigma(k, z)} \right),
\end{equation}
and
\begin{equation}
C_c(k, z) = \frac{S(k)}{k^2 S^{-1}(k)} \left( z + \frac{1}{1 + \Sigma(k, z)} \right).
\end{equation}
Equation (8) is a 3D integral equation, which has to be solved in a self-consistent manner to obtain \( \Sigma(k, z) \). The integration in \( q \) space is a convolution and can be done using Fourier transforms. Fourier transform is also employed in making the inverse Laplace transform numerically to get \( C_c(k, t) \) and \( C_s(k, t) \) from Eqs. (9) and (10).

We have solved \( \Sigma(k, z) \) iteratively by first setting \( \Sigma(k, z) = 0 \) for all \( k \) and \( z \) and inserting this into Eqs. (9) and (10), and integrating Eq. (8). The resulting new \( \Sigma(k, z) \) is then fed into the correlation functions Eqs. (9) and (10). The iteration is continued until \( \Sigma(k, z) \) converges.

The only input into this calculation is the static structure factor \( S(k) \). For this we have used the approximate expression by Ripoll and Tejero [61] which is based on the Percus-Yevick equation [62]. We have tested their approximation by comparing it to a direct numerical calculation of \( S(k) \), and find that it reproduces \( S(k) \) well except very close to the freezing transition. Our results for the self-consistent iteration of the MCA are shown in Fig. 2 with open circles. At lower densities, the MCA overestimates \( D_T \) but then rapidly seems to approach zero at about \( \tilde{\rho} = 0.8 \). Thus, it is not in very good quantitative agreement with \( D_T \).

The original calculation of IR has been criticized by Fuchs [31] who pointed out that Eq. (10) should actually be replaced by
\begin{equation}
C_c(k, z) = \frac{S(k)}{k^2 S^{-1}(k)} \left( z + \frac{1}{1 + k^2 \Sigma_c(k, z)} \right),
\end{equation}
where \( \Sigma_c(k, z) \) is finite for \( k \rightarrow 0 \). We have tried to improve the MCA results by simply setting \( \Sigma_c(k, z) = \Sigma(k, z) \). In Fig. 2 we show results for this case, but find that \( D_T \) from the MCA decays even more rapidly. We also tried some other approximations for \( \Sigma_c(k, z) \), but always found that using Eq. (11) did not improve the quantitative agreement with simulation data over the whole range of densities. Thus, we must...
conclude that despite the fact that memory effects in the present case are not very strong, the MCA does not give quantitatively good results for $D_T$ [63].

**B. Collective diffusion**

In addition to tracer diffusion of single particles, an interesting question concerns the behavior of collective density fluctuations in colloidal systems [26]. The collective diffusion coefficient $D_C(\rho)$ characterizing these fluctuations can be defined by means of the diffusion equation

$$\frac{\partial \rho(\vec{r},t)}{\partial t} = \nabla \cdot [D_C(\rho) \nabla \rho(\vec{r},t)].$$

(12)

Another equivalent way is through the Green-Kubo equation

$$D_c = \xi D_{cm} = \xi \lim_{t \to \infty} \frac{1}{2dNt} \langle |\vec{R}(t)|^2 \rangle,$$

(13)

where $\xi = \langle N \rangle / [\langle N^2 \rangle - \langle N \rangle^2]$ is the thermodynamic factor (proportional to the inverse of the compressibility $\kappa_T$), and $\vec{R}(t) = \sum_{i=1}^{N} [\vec{r}_i(t) - \vec{r}_i(0)]$ is the center-of-mass (c.m.) displacement [10].

In the case of Brownian hard spheres, it is an exact result that the c.m. mobility $D_{cm}$ is independent of the density $\rho$ [26]. This is because the interparticle interactions preserve the c.m. momentum, and thus $D_{cm}(\rho) = D_{cm}(0) = D_T(0)$. This means that the density dependence of $D_C(\rho)$ is solely determined by the static thermodynamic factor $\xi$, and there are no memory effects in $D_C$. The quantity $\xi$ can be conveniently obtained from the static structure factor $S(k)$ [10].

The static structure factor can also be obtained from the equation of state. For the present case of a 2D hard disk system there exists several approximate equations of state in the literature. A particularly simple analytic form for hard convex particles has been derived by Boublik [64] as

$$\xi = \frac{1 + 2\rho(\gamma - 1)}{(1 - \rho)^{\gamma/2}} + \frac{2\rho[1 + \rho(\gamma - 1)]}{(1 - \rho)^3},$$

(14)

where the aspect ratio $\gamma = \pi R_c^2 / A_c$, for convex particles of area $A_c$ and perimeter $2\pi R_c$. In Fig. 4 we show the results for $D_C(\rho)$ from Eq. (14) with $\gamma = 1$ as compared to our direct MC simulations of $\xi$. Except for densities very close to the freezing transition, the simple formula of Eq. (14) predicts the behavior of $D_C$ very well. We have also examined a number of viral expansions for the equation of state of 2D hard disks, and find that the one given by van Rensburg [65] gives the best agreement with our numerical data at the highest densities in the vicinity of the freezing transition.

**V. RESULTS FOR RODLIKE MOLECULES**

As explained in Sec. II, the second case that we have examined is that of rigid rods consisting of six 2D disks bound together. The definitions of the diffusion coefficients $D_T$ and $D_C$ remain the same, with the coordinates and velocities now referring to the c.m. of each rod. In addition, since the rods possess a rotational degree of freedom, one can also define the angular (tracer) diffusion coefficient $D_R$ by

$$D_R = \frac{1}{4} \lim_{t \to \infty} \frac{1}{t} \left\{ \frac{1}{N/N_c} \sum_{i=1}^{N/N_c} |\theta_i(t) - \theta_i(0)|^2 \right\},$$

(15)

where $\theta_i$ is the angle of rotation of rod $i$ with respect to a fixed axis [66]. The memory expansion method applied to calculate $D_T$ and $D_C$ can also be used for the angular diffusion coefficient in a straightforward manner.

In Fig. 5 we show results for $D_T$ in the case of rodlike molecules. For comparison we also show in the same figure our numerical results for the single particle case. Since in the case of rods the scaling of the density cannot be used, the $x$ axis here is in terms of the density $\rho_s = \sigma R_c^2 / L^2$ for the $r^{-12}$ potential. Note that here $N$ is still the number of disks in the system and there are six disks in each rod. Remarkably enough, the results for rods and single particles are identical within the error bars for the whole range of density up to about the single particle freezing density $\rho_s \approx 0.55$. We esti-
mate the freezing transition of the rodlike molecules to occur at about this same density, although we did not investigate this systematically. This is also in agreement with the results of Bates and Frenkel [52].

In Fig. 6 we show the angular diffusion coefficient for the rodlike molecules with the \( r^{-12} \) repulsion. As expected [52], we find no evidence of an orientational transition in the present case and \( D_\theta(\rho) \) is a smoothly decreasing function of density.

Finally, regarding the collective diffusion of Brownian rodlike molecules, the c.m. mobility is again independent of density. Therefore the collective diffusion coefficient is solely determined by the thermodynamic factor. In Fig. 7 we show results of MC simulations for \( \xi \) including both the case of hard rods (filled circles) and rods with a repulsive \( r^{-12} \) potential (crosses). \( D_C(\rho) \) is again a strongly increasing function of \( \rho \). We can also use Eq. (14) to estimate the thermodynamic factor for the hard rod case by neglecting the nonconvexity of the actual molecule (see Fig. 1). This estimate is shown in Fig. 7 by a solid line.

VI. SUMMARY AND DISCUSSION

In this paper we have investigated the density dependence of the diffusion coefficients of hard disks and rodlike molecules that diffuse on a smooth surface, with no explicit particle-surface interactions present. We have found that in both systems the collective diffusion coefficient \( D_C(\rho) \) is completely determined by the thermodynamic factor and is an increasing function of the coverage in the liquid phase. We have also tested the accuracy of various analytical expressions for the thermodynamic factor. The tracer diffusion coefficient \( D_T(\rho) \) on the other hand, is a monotonically decreasing function of coverage and its density dependence within the liquid phase is almost identical in both systems. Also the behavior of the angular diffusion coefficient \( D_R(\rho) \) of the rods is similar to the tracer diffusion coefficient. The mode-mode coupling approximation proved to be inadequate in describing the tracer diffusion coefficient \( D_T(\rho) \) of 2D hard disks.

It is interesting to compare the present results to the previous studies of diffusion of flexible chainlike molecules on smooth surfaces [16,19]. These studies were done using the FB lattice model combined with Monte Carlo dynamics. This corresponds to the lattice-gas limit where the chains move by single segment fluctuations only, and the rigid rod limit cannot even be defined. The center-of-mass momentum is not conserved with MC dynamics which leads to a strong density dependence of the mobility. The qualitative behavior of \( D_C(\rho) \) for smaller densities remains the same for both cases, however, in that \( D_C(\rho) \) grows rapidly with density. On the other hand, at higher densities the behavior is quite different from the present case due to the lattice-gas nature of the FB model. Because of the density dependent mobility, for flexible molecules \( D_C(\rho) \) shows a maximum at some intermediate value of the density after which it will decrease rapidly toward the full coverage limit.

As far as a comparison with experiments is concerned, for flexible chains confined on a metal surface the experiments [3] have observed a strong increase in \( D_C(\rho) \), and the observed density profiles are in complete agreement with simulation results [19]. Regarding rigid molecules, the experiments of Mak, Koenler, and George [67] on the surface diffusion of cycloalkanes on Ru(001) have demonstrated that \( D_C(\rho) \) for coverages \( \leq 0.7 \) is an increasing function of \( \rho \) in the case of cyclopentane. However, since these molecules are rather small and the surface is strongly corrugated a quantitative comparison to the smooth surface case may not be possible. We also note that in order to compare with the present study, the particles should be confined to 2D without an external, attractive surface potential.

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