

Diffusion in two-dimensional colloidal systems on periodic substrates

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We study the diffusive behavior of two-dimensional charged colloidal suspensions subjected to a sinusoidal substrate by means of Brownian dynamics simulations. We mainly focus on the dependence of the mean-square displacement on the substrate strength. Our findings show a variation in the particle diffusion due to a substrate-induced distortion of the dynamic cage of nearest-neighbor colloids. This mechanism leads to a transition from normal diffusion at short times to subdiffusion on intermediate time scales. However, at long times normal diffusion is recovered. We also show that the variation in the long-time self-diffusion coefficient may be associated with the freezing and re-entrant melting transitions.

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I. INTRODUCTION

The study of transport properties in restricted dimensions is crucial to understand dynamical processes that occur in confinement conditions. For example, studies of Brownian particles in narrow channels, where no mutual passage is possible, have elucidated the activation of dynamic modes responsible for a subdiffusive process known as single-file diffusion [1–4]. Also, the study of the dynamical properties of noninteracting particles on solid surfaces has explained the anomalous diffusion in crystalline substrates [5,6]. Furthermore, the dynamics of biological motors and macromolecules occurs usually under confinement, i.e., heterogeneous substrates or cell membranes [7,8].

Generally speaking, the dynamics in confinement depends on the substrate-particle interaction, i.e., the topology of the substrate, the particle-particle interaction, temperature, and particle density. Basically, both interactions define the energetic landscape where particles diffuse. In atomic systems, it is difficult to quantify such energetic landscape. However, micron-sized particles, i.e., colloids, are ideal candidates for studying diffusion of interacting particles in external potentials since the energetic landscape can be easily controlled [9]. In particular, in the last few years two-dimensional (2D) colloidal systems created artificially by confining the colloids between two glass plates or two media (e.g., air-water interface) [10,11] have served as fascinating and well-controlled model systems to study phenomena such as freezing [12], the controversial chargelike attraction [13], and many-body effective interactions [14], among others.

From both theoretical and experimental points of view, the 2D colloidal systems exposed to periodic laser fields have been the focus of extensive investigations. Since the pioneering work of Chowdhury *et al.* [15], such systems have been found to exhibit a rich phase behavior with a solidlike order that depends on both the suspension properties and the substrate parameters, e.g., strength and periodicity [16,17]. Related research also deals with topics such as strain-induced domain formation [18], laser-induced freezing (LIF), and melting [19–23].

Although the phase behavior of 2D colloids exposed to light forces is well understood, less is known about the corresponding dynamical properties. A few works reported in such direction show a rich variety of pinned and dynamic states including pinned smectic, pinned buckled, two-phase flow, and moving partially ordered structures [24,25]. Nonetheless, the dependence of the mean-square displacement (MSD) on the substrate strength has not been analyzed in detail. The MSD is a suitable quantity for studying the dynamical behavior at different time scales. Recently, the MSD of a single Brownian particle in random and quasicrystalline potentials has been investigated [26]. Interestingly, Schmiedeberg *et al.* [26] demonstrated that although quasicrystalline potentials exhibit long-range positional order the nonequilibrium Brownian motion is very similar to the motion in a random substrate.

In this work, the MSD [$W(t)$] of charged colloids in monolayers subjected to sinusoidal-like substrates with a commensurability ratio $p=1$ is measured by means of Brownian dynamics (BD) simulations. With the increasing of the substrate strength, we observe a transition from normal diffusion, $W(t) \propto t$, at short times to subdiffusion at intermediate times, $W(t) \propto t^\alpha$, described by a variable exponent α . Such transition is explained in terms of a substrate-induced distortion of the dynamic cage of nearest-neighbor colloids. At long times normal diffusion is recovered, i.e., $\alpha=1$, with a self-diffusion coefficient that varies with the substrate strength and becomes smaller than that of the free-particle diffusion coefficient. We particularly show that the variation in the MSD at long times may be associated with the freezing and the re-entrant melting transitions [15,20,22].

The paper is organized as follows. In Sec. II, we describe the two-dimensional colloidal model system and the Brownian dynamics simulation technique. We also review the structural and dynamic properties in the substrate-free case. In Sec. III, the MSD and the average energy per particle as function of the substrate strength are discussed. Finally, the paper ends with concluding remarks.

II. MODEL SYSTEM, BROWNIAN DYNAMICS SIMULATION, AND SUBSTRATE-FREE CASE

A. Model system and Brownian dynamics simulation

Let us consider a two-dimensional system consisting of N particles and a particle number density $\rho=N/A$, with A being

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the total area of the system. It is well known that within the fluid phase, and in the absence of external field, colloids that interact repulsively are, in average, homogeneously distributed in the whole available area. Hence, particles are separated by a characteristic mean interparticle distance given by $d = \sqrt{\rho}$ [27].

In the BD simulation, the configuration-space trajectories are composed of successive displacements $\vec{r}_i(t + \Delta t) - \vec{r}_i(t)$ ($i = 1, \dots, N$) and generated from the stochastic equation [27]

$$\vec{r}_i(t + \Delta t) - \vec{r}_i(t) = \vartheta_B(\vec{r}^N(t))\Delta t + \delta\vec{r}_i(\Delta t), \quad (1)$$

where $\vartheta_B(\vec{r}^N)$ is a differential operator which depends on both the direct and hydrodynamic interactions [27]. The random displacements $\delta\vec{r}_i(\Delta t)$ are chosen from a multivariate Gaussian distribution function such that $\langle \delta\vec{r}_i(\Delta t) \rangle = 0$ and $\langle \delta\vec{r}_i(\Delta t) \delta\vec{r}_j(\Delta t) \rangle = 2D_{ij}\Delta t$ with D_{ij} being the diffusion tensor [27].

Neglecting hydrodynamic interactions, $D_{ij} = D_0\delta_{ij}$, Eq. (1) becomes

$$\vec{r}_i(t + \Delta t) = \vec{r}_i(t) + \beta D_0 \vec{f}_i(t) \Delta t + \delta\vec{r}_i(\Delta t), \quad (2)$$

with $\beta \equiv (k_B T)^{-1}$ being the inverse of the thermal energy and $\vec{f}_i(t) = -\vec{\nabla}_i(U^{cc} + U^{ext})$ denotes the total force acting on particle i due to the interaction with the other particles and its coupling with the substrate. D_0 is the Stokes-Einstein diffusion coefficient $D_0 = \frac{k_B T}{6\pi\eta a}$, with k_B being the Boltzmann constant, T the absolute temperature, η the solvent shear viscosity, and a the particle radius. The total potential energy due to the particle-particle interaction can be written as $U^{cc} = \sum_{j>i}^N u(r_{ij})$, where $u(r)$ represents the pair interaction potential between colloids, which is expressed here by the repulsive part of the Derjaguin-Landau-Verwey-Overbeek (DLVO)-type potential [28],

$$\beta u(r) = Z_{eff}^2 \lambda_B \left[\frac{\exp(\kappa a)}{1 + \kappa a} \right]^2 \frac{\exp(-\kappa r)}{r}, \quad (3)$$

with λ_B as the Bjerrum length, κ as the Debye screening parameter, and Z_{eff} as the effective charge [28]. The total external energy can then be written as $U^{ext} = \sum_{i=1}^N u^{ext}(\vec{r}_i)$, where $u^{ext}(\vec{r}_i)$ is the substrate potential acting on particle i , which is given by

$$u^{ext}(\vec{r}_i) = V_0 \sin\left(\frac{2\pi x_i}{a_L}\right), \quad (4)$$

with V_0 and a_L being the strength and lattice spacing of the substrate potential, respectively. Let us now define the commensurability ratio $p \equiv \sqrt{3}d/(2a_L)$. This factor allows one to describe the variety of phases in systems under the influence of periodic substrates [16].

As discussed previously, Eq. (2) gives the trajectory of each colloidal particle. The dynamics of the system is then simulated as follows. We consider a random initial configuration of $N=1600$ colloidal particles placed in a rectangular box of dimensions $L_x \times L_y$ with $L_x = \frac{\sqrt{3}}{2}L_y$ and periodic boundary conditions on each direction. Then, particles move according to Eq. (2). The system evolves from a nonequilibrium state to its corresponding equilibrium state. In order to

find such state, we check the time evolution of the energy per particle U/N , where $U = U^{cc} + U^{ext}$; in a BD simulation U varies until it reaches a constant value. In the following time steps, the system is then in thermodynamic equilibrium. Therefore, one is able to construct any function that requires the average over the particle configurations.

The time step, Δt , in Eq. (2) is chosen not too big in order to ensure that the force acting on the particle is approximately constant during its duration, but it must be not too small to guarantee that we can use a description on the diffusive regime. In view of this, we have used a time step of $\Delta t^* \equiv \frac{D_0}{a^2} \Delta t = 0.0002$.

B. Substrate-free case

Typically, in experiments of 2D colloidal systems to be exposed to light forces, the particle bulk density is usually chosen between 5% and 10% below the spontaneous crystallization [20]. Following the same protocol we should find the density at which our 2D system crystallizes (according to the values of the interparticle potential parameters chosen for the study). Then, once the crystallization density is obtained, the substrate periodicity is adjusted to fix the commensurability ratio $p=1$. The potential parameters were taken from Ref. [18], i.e., $Z_{eff}=5400$, $\lambda_B=0.72$ nm, $\kappa^{-1}=550$ nm, and $a=1.4$ μ m.

Nowadays, there are several empirical criteria for two-dimensional freezing. One of them states that the amplitude of the first maximum of the liquid structure factor, $S(q)$, reaches a value of 5.5 [29]. Also, a dynamical criterion for two-dimensional freezing (valid in both three and two dimensions) was introduced by Löwen [30]. It says that the ratio of the long- and short-time diffusion coefficients is 0.1 along the freezing line. We follow both criteria to find the crystallization density.

Physical quantities such as $S(q)$ and $W(t)$ can be measured either through experimental techniques, such as dynamic light scattering, or numerically, such as BD simulations (see, e.g., [27]). The static structure factor is simulated here by using the relation [31]

$$S(q) = N^{-1} \left\langle \left(\sum_{i=1}^N \cos(\vec{q} \cdot \vec{r}_i) \right)^2 + \left(\sum_{i=1}^N \sin(\vec{q} \cdot \vec{r}_i) \right)^2 \right\rangle, \quad (5)$$

where the angular brackets $\langle \dots \rangle$ denote an ensemble or temporal average and q is the magnitude of the wave vector \vec{q} . We average over 20 different angles of observation to improve the statistics for the calculation of $S(q)$. The MSD is computed according to the relation [27]

$$W(t) \equiv \langle \Delta r(t)^2 \rangle = N^{-1} \sum_{i=1}^N \langle [\vec{r}_i(t) - \vec{r}_i(0)]^2 \rangle. \quad (6)$$

This equation can be rewritten as $W(t) = W_x(t) + W_y(t)$, where $W_x(t)$ and $W_y(t)$ denote the MSD in the directions x and y , respectively.

Using Eq. (3) with the potential parameters mentioned previously, we simulate the structure of a 2D suspension of charged colloidal particles. Figure 1 shows $S(q)$ for different

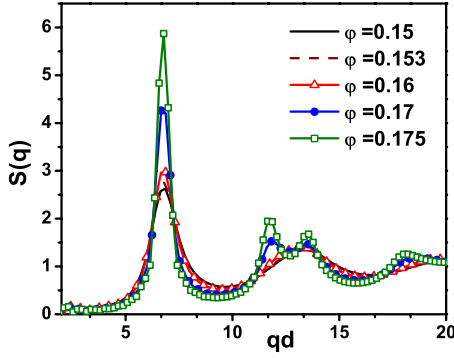


FIG. 1. (Color online) Static structure factor $S(q)$ obtained from BD simulations for different packing fractions, φ .

packing fractions, $\varphi \equiv \pi a^2 \rho$. One can observe that the position of the main peak does not change appreciably and it is located at the position $qd \approx 2\pi$; this clearly indicates that the mean distance between colloids is given by d regardless of the packing fraction value. Furthermore, the height of the main peak is increasing up to a value of ≈ 5.87 at $\varphi = 0.175$.

We now focus our attention to the reduced MSD, $W^*(t) \equiv W(t)/a^2$, shown in Fig. 2. We can observe that for short times, over small distances as compared to the particle size, the MSD is not affected by the direct-interaction forces since the configuration of the dynamic cage of nearest-neighbor particles has not changed appreciably during the short time t . The MSD shows a linear diffusion law with a short-time self-diffusion coefficient D_S equal to the Stokesian diffusion coefficient D_0 of an isolated Brownian particle. This is due to the neglecting of hydrodynamic interactions, which in principle act instantaneously and could modify the value of D_S even at short times. At intermediate times, the motion of the particle becomes retarded by direct interaction with the cage of neighboring particles, since the cage becomes distorted from its equilibrium structure. The distortion implies a transition from linear to a sublinear time dependence of $W(t)$. At long times, the particle has experienced many collisions with other particles and undergone a long-distance random walk which again leads to a linear behavior, characterized by the long-time self-diffusion coefficient, D_L .

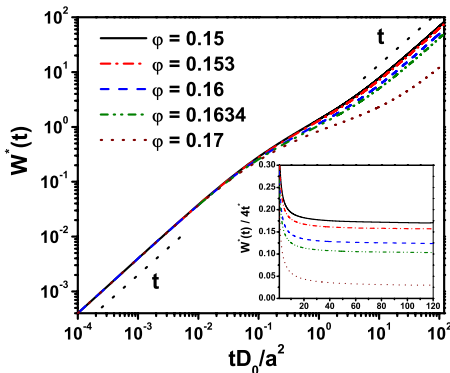


FIG. 2. (Color online) Reduced mean-square displacement, $W^*(t) \equiv W(t)/a^2$, for different packing fractions with $V_0 = 0$ and potential parameters taken from Ref. [18]. Inset: ratio $W^*(t)/4t^*$ which at long times reduces to D_L/D_S .

In general, for homogeneous systems, a linear increase in the MSD at short and long times is observed. This behavior can be summarized as follows:

$$W(t) = \begin{cases} 2DD_S t, & \tau_B \ll t < \tau_S \\ 2DD_L t, & t \gg \tau_S, \end{cases} \quad (7)$$

where τ_B is the typical relaxation time of the particle velocity due to the solvent friction, $\tau_S = a^2/D_0$ is the structural relaxation time, and D indicates the dimension of the system. It has also been observed that $D_L \leq D_S$, which can be attributed to the hindrance of the particle motion by the direct forces, regardless whether these forces are repulsive or attractive [27].

From the inset of Fig. 2, one can easily observe that at long times the ratio $W^*(t)/4t^*$ (with $t^* = tD_0/a^2$) results equivalent to the ratio $\frac{D_L}{D_S}$ and takes values smaller than 0.1 for packing fractions $\varphi > 0.1634$. Therefore, by following both the static and dynamic criteria for two-dimensional freezing, one can state that our homogeneous system crystallizes around the packing fractions $0.1634 < \varphi_c < 0.175$. Therefore, we fix the density of our reference system 10% below the value $\varphi = 0.17$, then obtain a packing fraction of ≈ 0.153 . This value is used in our simulations with substrate, i.e., $V_0 \neq 0$.

III. MEAN-SQUARE DISPLACEMENT: SUBSTRATE EFFECTS

The reduced MSD as function of $V_0/k_B T$ is depicted in Fig. 3. We observe that by increasing $V_0/k_B T$, a transition from normal short-time diffusion to subdiffusion at intermediate time scales appears. The short-time diffusion ($t^* < 10^{-1}$) only depends on the collision of colloids with the solvent, which means that $D_S = D_0$ regardless of the value of the substrate strength. At intermediate times, the particle dynamics is strongly affected by both the cage generated by nearest-neighbor particles and the *effective* cage induced by the substrate. Interestingly, the crossover time from normal diffusion to subdiffusion occurs at earlier times as $V_0/k_B T$ is increased. This is due to the fact that increasing the substrate strength the direct particle-substrate interaction becomes more important inclusive at shorter times. For small V_0 ($\leq 3k_B T$) [see Fig. 3(a)], the transition from normal diffusion to subdiffusion occurs continuously, i.e., diffusion decreases by increasing V_0 . The reduction in the particle diffusion is due to the particles must spend a longer time for crossing several substrate minima before reaching the diffusive motion. This causes that particles diffuse (or oscillate) around the position of each substrate minimum for a long period of time. This means that the time needed to surmount the energetic barrier increases with the substrate strength. For high V_0 ($\geq 4k_B T$) [see Fig. 3(b)] the MSD seems to collapse onto a master curve, although we observe variations (see inset). At long times the MSD recovers the linear increase as the one described in Eq. (7). Nonetheless, the corresponding long-time self-diffusion coefficient D_L varies strongly with V_0 . Here one can appreciate that an increase in V_0 is not necessarily related with a decrease in the particle

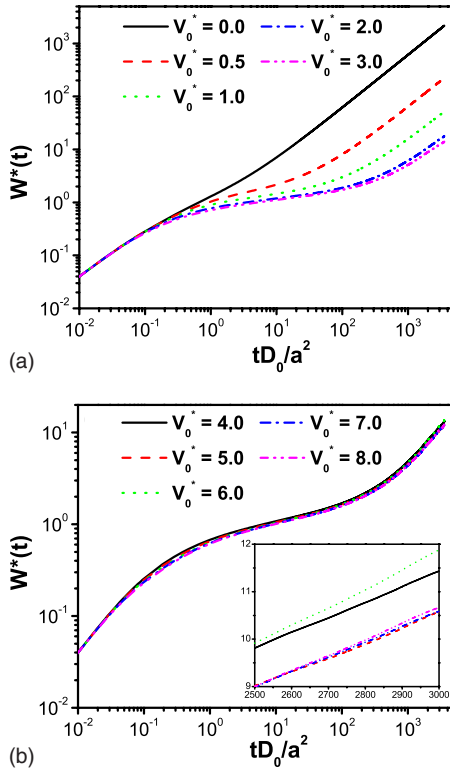


FIG. 3. (Color online) Reduced mean-square displacement, $W^*(t)$, for (a) small substrate strengths: $0 \leq V_0/k_B T \leq 3$ and for (b) large substrate strengths: $4 \leq V_0/k_B T \leq 8$. Inset shows the long-time dynamics.

diffusion, i.e., particles can diffuse faster in some cases. For example, it is clearly evident that for $V_0 = 5k_B T$ the diffusion becomes slower than the case $V_0 = 6k_B T$ [see inset of Fig. 3(b)]. This variation may be associated with the so-called LIF and laser-induced melting (LIM) phenomena [15,19,20]; we will come back to this point later. For higher substrate strengths, $V_0 > 9k_B T$, the diffusion decreases again (see below).

To understand the effect of the substrate on the particle dynamics, we also study both the MSDs on the direction parallel, $W_x(t)$, and perpendicular, $W_y(t)$, to the substrate. In the homogeneous case, $V_0 = 0$, we checked that $W_x(t) = W_y(t) = \frac{1}{2}W(t)$, i.e., colloids diffuse, in average, in the same fashion in any direction. $W_x(t)$ is shown in Fig. 4(a) for small substrate strengths, $V_0/k_B T = 0, \dots, 3$, and in Fig. 4(b) for high substrate strengths, $V_0/k_B T = 4, \dots, 8$. In both cases, at sufficiently short times, where the individual particles do not feel the direct interactions with the other colloids, normal diffusion occurs and the MSD is found to be $W_x(t) \propto t$. At intermediate times the diffusion process becomes subdiffusive and for $V_0 < 3k_B T$ normal diffusion is recovered at sufficiently long times. However, for high substrate strengths a well-defined plateau is observed and normal diffusion seems to be completely suppressed in the x direction. This result can be interpreted as a kind of freezing or glasslike transition along the parallel direction. Looking now at the height of the plateau, $\gamma^2 \equiv W_x(t^* \approx 10)$, one can estimate the cage size induced by the substrate. This means that γ allows one to quantify the linear dimensions of the cage (or the line length)

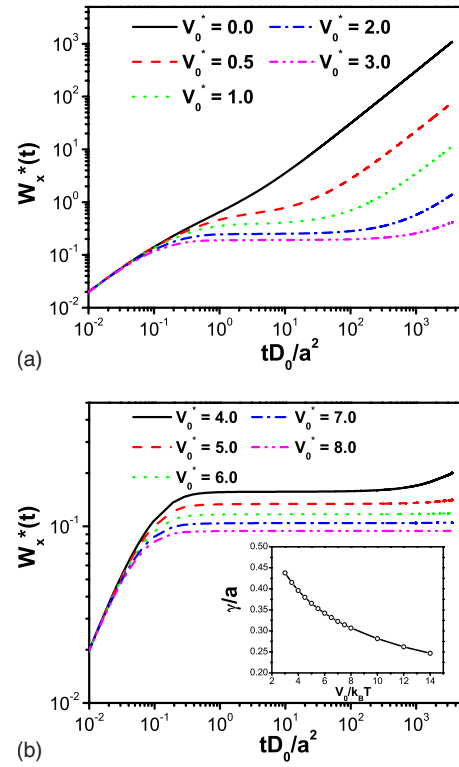


FIG. 4. (Color online) Reduced mean-square displacement in the x direction, $W_x^*(t)$, for (a) small substrate strengths: $0 \leq V_0/k_B T \leq 3$ and for (b) large substrate strengths: $4 \leq V_0/k_B T \leq 8$. Inset: square root of the plateau height, $\gamma/a \equiv \sqrt{W_x^*(t^* = 10)}$, as a function of $V_0/k_B T$. Symbols are obtained from the simulations and the line is a fit of the form $\gamma \propto \exp(-V_0/V_c)$.

where the particle position fluctuates around the substrate minimum in the x direction. A simple exponential decay fit, $\gamma \propto \exp(-V_0/V_c)$, can be used to reproduce the whole behavior of γ [see inset of Fig. 4(b)]. We find that $V_c \approx 5.25k_B T$, which would correspond to the substrate strength at which diffusion in x is completely suppressed. Therefore, the variation observed in the MSD of Fig. 3 cannot be explicitly attributed to $W_x(t)$.

Although the substrate affects the particle diffusion along the x direction, colloids can still diffuse in the perpendicular direction, i.e., y direction. Figure 5 shows $W_y(t)$, which evidently reflects the variation in the MSD of Fig. 3 for large substrate strengths. The only way to explain diffusion along the y direction is through thermal fluctuations, which are not completely suppressed by the substrate and therefore promote diffusion along the perpendicular direction.

So far, one can conclude that the mean-square displacement of interacting particles in a periodic substrate behaves as $W(t) \propto t^\alpha$. Both fitting and derivative procedures to the MSD of Fig. 3 permit to reveal the behavior of the exponent α as a function of $V_0/k_B T$ at different time scales. At short times evidently $\alpha = 1$. Figure 6 shows that at long times $\alpha \approx 1$. This means that normal diffusion is recovered and the linear law (7) can accurately be used to describe both self-diffusion coefficients at short and long times. However, at intermediate times the dynamics becomes subdiffusive with an exponent α which decreases with V_0 . Nonetheless, a well-

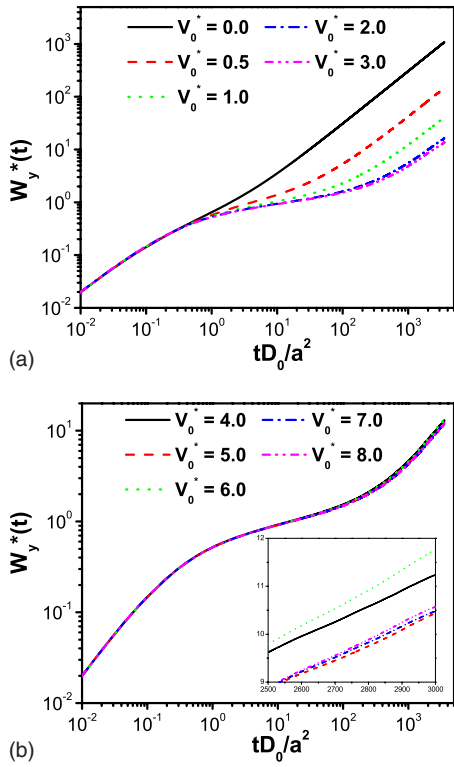


FIG. 5. (Color online) Reduced mean-square displacement in the y direction, $W_y^*(t)$, for (a) small substrate strengths: $0 \leq V_0/k_B T \leq 3$ and for (b) large substrate strengths: $4 \leq V_0/k_B T \leq 8$. Inset: variation in $W_y^*(t)$ at long times.

defined plateau for $V_0 > 5k_B T$ is observed. Such plateau indicates that the particle subdiffusion no longer depends on the substrate strength and, as we will see further below, this effect is due to the colloid-colloid interaction.

An important quantity in colloidal dynamics is the ratio D_L/D_S . In homogeneous systems, such ratio allows quantifying the effect of the interparticle interactions on the system transport properties [27]. Moreover, it has also been used to introduce a dynamical criterion for describing the liquid-freezing transition in colloidal suspensions [30]. Following similar ideas, here we try to establish a connection between the long-time self-diffusion coefficient and both LIF and

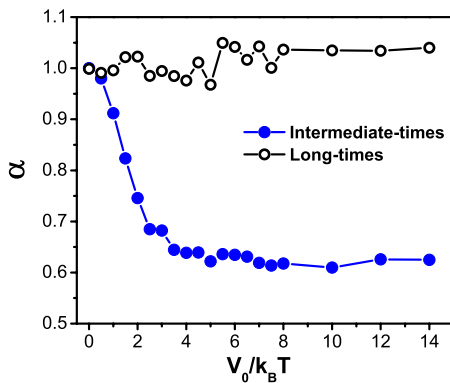


FIG. 6. (Color online) Behavior of the exponent α , which fits the MSD of Fig. 3, as a function of V_0 at intermediate and long times. Lines are just a guide for the eyes.

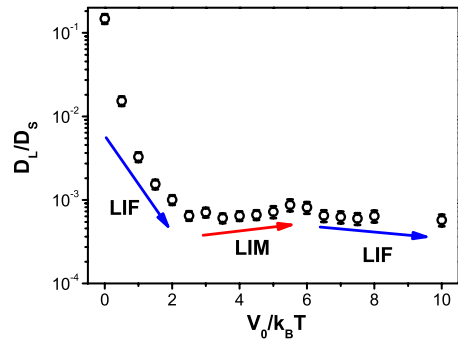


FIG. 7. (Color online) Long-time self-diffusion coefficient for different substrate strengths, $V_0/k_B T$. Blue arrows indicate a possible LIF, whereas red arrow describes the region of a possible LIM.

LIM transitions. Figure 7 depicts the ratio D_L/D_S for different substrate strengths. Immediately, we can observe that D_L is 2 or 3 orders of magnitude smaller than D_S . Therefore, if one assumes that the Löwen criterion [30] also holds in this case, the system should be evidently in a freezinglike state. Nonetheless, the variations in D_L can be a strong evidence of the LIF and LIM phenomena [15,19,20]. To clarify this point, we should mention that LIF takes place at relatively small substrate strengths [22], where it is expected a decrease in D_L with V_0 , as the one observed in $0 < V_0/k_B T < 2$. Similarly, the LIM transition is observed at higher values of V_0 [22]. In such transition, a remelting of the crystal back into a modulated liquid phase occurs. Then, in terms of a diffusion process, a modulated liquid should be characterized by an increase, not necessarily continuous, in the particle diffusion, as the one depicted in the interval $3 < V_0/k_B T < 6$. Hence, the variation in the long-time self-diffusion coefficient seems to describe qualitatively the expecting dynamical features of both LIF and LIM transitions. In this sense, our findings are twofold. First, they allow us to verify the phase behavior of the system and, second, to establish a dynamical-like criterion, based on the variations in D_L , to describe the effect of the substrate on the system phase since a static criterion based on the decay of the pair-correlation function, $g(y)$, is already well established [22].

The subdiffusive process at intermediate times can be explained by using energetic arguments. We should emphasize that for substrate strengths $V_0 > 1.5k_B T$, the colloids are trapped on the potential minima along the direction parallel to the substrate. They are basically separated by a distance close to a_L forming well-spaced strips of colloids. Figure 8 allows us to visualize the structural changes in the system as function of V_0 . Although the snapshots suggest a crystal-like structure, a simple Voronoi analysis shows that the systems are not completely in the hexagonal crystal phase (data not shown). Clearly, each strip cannot be independent; otherwise, colloids must diffuse as in a one-dimensional channel. In such case, the MSD would scale as $W(t) \propto t^\alpha$ with $\alpha = 1/2$ [3,4]. However, Fig. 6 demonstrates that at intermediate times $\alpha \neq 1/2$. This behavior should be related with the fact that the pair interaction between colloids of different strips is not completely negligible; each strip can interact with its nearest neighbor.

The discussion of the last paragraph can be better visualized in Fig. 9, where the average colloid-colloid potential

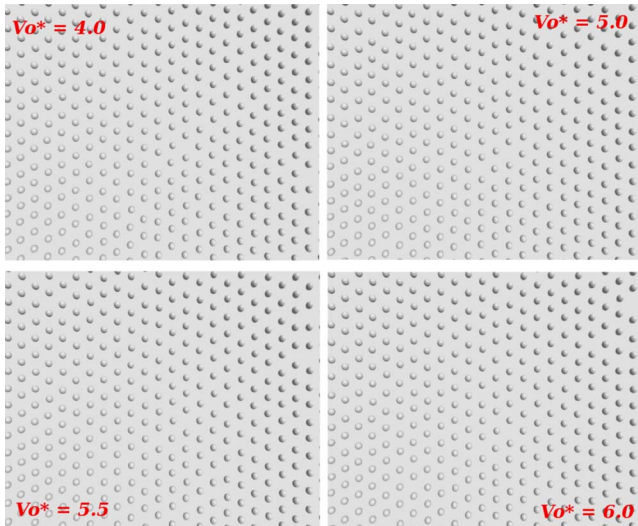


FIG. 8. (Color online) Snapshots of the two-dimensional system for different substrate strengths, $V_0/k_B T$.

energy per particle, U^{cc}/N , as function of the substrate strength is shown. We observe that such contribution decreases quickly in the interval $0 \leq V_0/k_B T \leq 2$, i.e., when the system is approaching to the freezinglike state, and shows a slower and variable behavior for $V_0 > 3k_B T$ until it reaches a constant value of $\approx 1.4k_B T$ for $V_0 > 5k_B T$. We should point out that, on one hand, the behavior of U^{cc} evidences the importance of the interparticle interaction on the subdiffusive processes that occur at intermediate time scales since α basically shows the same behavior (see Fig. 6). On the other hand, the origin of the variation in U^{cc} comes from the competition between both particle-particle and substrate-particle interactions, i.e., a distortion induced by the substrate in the configuration of the dynamic cage of nearest-neighbor colloids. Additionally, the inset shows the total energy per particle. There one can appreciate that the contribution of the substrate becomes dominant when V_0 is larger than that of the average energy per particle in the substrate-free case, i.e., $V_0 > 2k_B T$.

IV. CONCLUSIONS

We have carried out Brownian dynamics simulations to study the diffusive motion in a two-dimensional colloidal system subjected to a sinusoidal substrate. In particular, we measured the MSD for different substrate strengths. The MSD is a suitable quantity to investigate the particle dynamics at different time scales. We observed that the resulting behavior in the MSD can be explained in terms of the

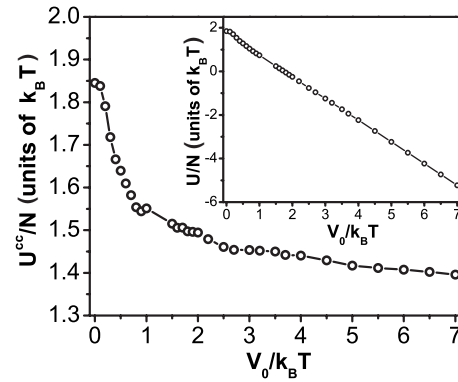


FIG. 9. Average colloid-colloid energy per particle as function of V_0 . Inset shows the average total energy per particle. The line is just a guide for the eyes.

substrate-induced distortion in the configuration of the dynamic cage of nearest-neighbor colloids.

We found that the linear increase law of the MSD can also be used to describe accurately both the short- and long-time dynamics. However, at intermediate time scales the dynamics became subdiffusive since the interparticle interaction contribution is relevant inclusive at high substrate strengths. We also observed that the variations in the long-time self-diffusion coefficient can be used to establish a dynamical-like criterion to understand the system phase behavior from a dynamical point of view. In particular, this criterion is able to explain the main features of the well-known laser-induced freezing and laser-induced melting transitions. Additionally, the diffusive behavior in the direction parallel to the substrate indicated a kind of dynamic arrest in such direction. We also found that the linear dimensions of the dynamic cage depends strongly on the substrate strength and can simply be described by an exponential-like function.

Finally, we should remark that our results can be confirmed in experiments of light forces. Moreover, the substrate model used in our work can be generalized to the case of random or periodic two-dimensional potential. This will allow us to understand fully the diffusion process of interacting Brownian particles on substrate potentials. Further investigation along these lines is in progress.

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