Superparamagnetic colloids confined in narrow corrugated substrates

S. Herrera-Velarde and R. Castañeda-Priego*

Instituto de Física, Universidad de Guanajuato, Loma del Bosque 103, Lomas del Campestre, 37150 León, Guanajuato, Mexico

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We report a Brownian dynamics simulation study of the structure and dynamics of superparamagnetic colloids subject to external substrate potentials and confined in narrow channels. Our study is motivated by the importance of phenomena like commensurable-incommensurable phase transitions, anomalous diffusion, and stochastic activation processes that are closely related to the system under investigation. We focus mainly on the role of the substrate in the order-disorder mechanisms that lead to a rich variety of commensurate and incommensurate phases, as well as its effect on the single-file diffusion in interacting systems and the depinning transition in one dimension.

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I. INTRODUCTION

The study of static and dynamic properties of interacting colloidal particles in the presence of external fields is an interesting and complex subject that has grown in the last few years. This subject is increasingly important because a quantitative understanding of colloidal dispersions under external fields allows elucidation of the physical properties of other complex systems that are of industrial, chemical, and biological relevance. For example, colloidal particles under external laser fields have highlighted the importance of the ordering and dynamics of atomic systems on surfaces, such as atomic monolayers [1,2], and the relevance of stochastic resonance in nonlinear bistable systems [3]. For a review of colloids under external control see, for instance, Ref. [4].

During the last few decades, the study of dynamical properties in quasi-one dimensional (quasi-1D) channels has become a fascinating topic from both theoretical and experimental points of view. This is due to the fact that the dynamics of fluids in restricted geometries is very different from that in the bulk. For example, when the channel is so narrow that mutual passage is not possible, a correlation between subsequent displacements emerges, so that the motion of individual particles requires the collective motion of many other particles in the same direction; this leads to a subdiffusive process characterized by an anomalous mean-square displacement of the form $\lim_{t\gg\tau} W(t)=2F\sqrt{t}$ [6], where F is the so-called single-file diffusion (SFD) mobility factor. Particularly, interacting particles in narrow channels and subject to external fields represent an interesting model system to study several topics, such as the basis of commensurate-incommensurate transitions, the SFD in the inhomogeneous case, and the conditions for depinning in 1D, as well as the study of nonlinear dynamics in systems with anharmonic interactions [7].

In recent years, experimental corroboration of SFD has been performed successfully by several research groups [8–10]. In particular, Lutz *et al.* [11,12] created 1D circular channels by means of scanning optical tweezers in order to avoid the presence of lateral confinement walls. This experimental setup allows a reduction of the dissipative hydrodynamic interactions, thus leading to higher particle mobility. This technique has elucidated the typical diffusion of charged colloids in a free single file—i.e., in the absence of a substrate.

Nowadays, due to several experimental and theoretical studies, the diffusion on homogeneous substrates is now understood, but the dynamical properties on corrugated surfaces are still less known, although the latter case is more relevant for modeling atomic surfaces. Furthermore, there is no experimental evidence of the way in which a modulated substrate affects the physical properties of interacting particles along the channel. Nonetheless, from a simulation point of view, the mobility of noninteracting pointlike particles on periodic substrates [13] and the diffusion of charged colloids under modulated sinusoidal substrates [14] have shown that the presence of the substrate does not invalidate the diffusive behavior $W(t)\propto\sqrt{t}$.

Although most experimental results for SFD have been performed with charged colloids, the study of the same phenomenon in similar systems permits one to gain a better understanding of the relevant physical processes that can take place, such as anomalous diffusion, hopping rate of particles in periodic potentials, and depinning transition, among others. In the present work, motivated by whether the rich scenario of novel phases and dynamical processes are dependent of the kind of interaction potential, we extend our previous study for charged colloids [14] to the case of superparamagnetic colloids. We basically study the commensurate-incommensurate phases and the subdiffusive process of colloids with dipolelike interactions in narrow corrugated channels over an extended range of substrate strengths. We should mention that our results can be corroborated in experiments of colloidal particles confined in 1D optical channels and subject to periodic light-forces [11,12]. The latter ones are commonly used for modeling periodic substrates [2].

The paper is organized as follows. In Sec. II we describe the interaction potential between colloids, the Brownian simulation algorithm, and the main quantities to characterize the structure and dynamics of the system. In Sec. III, the substrate-free case is analyzed in terms of the potential strength. In Sec. IV, the pair distribution function and the

*ramoncp@fisica.ugto.mx

mean-square displacement are studied as functions of the substrate parameters. Moreover, the depinning transition is discussed through the loss of correlation observed in the system structure and the enhancement in the particle mobility factor. In addition, the Frenkel-Kontorova model [15,16] is briefly summarized in order to compare the resulting phases near to the zero temperature with those at finite temperature. Finally, the paper is closed with a section of conclusions.

II. MODEL SYSTEM AND SIMULATION TECHNIQUES

A. Interaction between colloidal particles

A striking advantage of colloidal dispersions lies in the fact that they can be used as model systems for soft condensed matter and inherent properties can be studied simultaneously by using three different complementary methods: namely, experiments, theory, and computer simulations. In particular, superparamagnetic colloids at the air-water interface have served as excellent models to investigate fundamental properties that are related to the role of hydrodynamics and melting transition, as well as the elastic behavior and the phonon band structure in two-dimensional crystals [17,18]. In such an experimental model, an external and constant magnetic field is applied in the perpendicular direction of the interface. This leads to a long-range magnetic dipole-dipole interaction between colloids of the form [17]

$$\beta u(r) = \frac{\Gamma}{r^3}, \tag{1}$$

where r is the separation between two colloids in units of the mean interparticle distance $d \equiv \rho^{-1}$, which is basically the natural length scale of the system [17], and $\Gamma \equiv \beta(\frac{\mu_0}{4\pi})\chi_{eff}^2 B^2/d^3$ is the mean interaction energy or interparticle strength normalized with the thermal energy $\beta^{-1} \equiv k_B T$, with k_B Boltzmann's constant and T the temperature, B the applied magnetic field, and χ_{eff} the effective magnetic susceptibility of the particles. Therefore, a variation in Γ can be related with a change in B, T, or ρ, the latter one being the particle number density.

We should point out that in contrast with charged colloids, where the potential is assumed to have a Yukawa-like form with effective parameters like the effective charge and Debye screening length [14] (which can be varied accordingly to the specific particular conditions), the interaction potential between superparamagnetic colloids depends only on the parameter Γ. Therefore, the parameter space is drastically reduced, thus facilitating a universal description of the system properties. Moreover, from an experimental point of view, the associated uncertainties in the determination of the parameters in the case of charged colloids are larger than those used to determine Γ [17]. Then, superparamagnetic colloids are excellent model systems for investigating soft matter.

B. Simulation techniques

To obtain information of the suspension structure, we calculate the pair distribution function $g(x)$ and the static structure factor $S(q_x)$. The diffusive behavior is obtained from the mean-square displacement (MSD) $W(t)$. $g(x)$ is the probability of observing a particle at a distance x from a given particle, and it is computed by averaging over equilibrium configurations through the relation [19]

$$g(x) = \frac{1}{N\rho}\left\langle \sum_{i=1}^{N-1}\sum_{j>1}^{N} \delta(x - x_{ij}) \right\rangle, \tag{2}$$

where the angular brackets $\langle \cdots \rangle$ denote a statistical (temporal or ensemble) average and N is the number of particles. $S(q_x)$ characterizes the variations in the local density as a result of the particle interactions and the coupling with the external field. Such a variations are quantified by the spatial frequency $q \equiv 2\pi/\lambda$, where λ is the corresponding wavelength. Then, the static structure factor is simulated by using the alternative relation [19]

$$S(q_x) = N^{-1}\left\langle \left(\sum_{i=1}^{N} \cos(q_{x_i} \cdot x_i)\right)^2 + \left(\sum_{i=1}^{N} \sin(q_{x_i} \cdot x_i)\right)^2 \right\rangle, \tag{3}$$

where q_x is the magnitude of the wave vector. The MSD is computed by using the expression

$$W(t) = \langle \Delta x(t)^2 \rangle = N^{-1}\sum_{i=1}^{N} \langle [x_i(t) - x_i(0)]^2 \rangle. \tag{4}$$

To simulate the structural and dynamic properties given by Eqs. (2)–(4) we use the so-called Brownian dynamics (BD) simulation method without hydrodynamic interactions. This method is based on Ermak's algorithm [20,21]

$$x_i(t + \Delta t) = x_i(t) + \beta D_0 F_i(t)\Delta t + x_i^r(t), \tag{5}$$

where $x_i(t)$ denotes the position of particle i at time t, $F_i(t)$ is the total force acting on it due to its interaction with the other particles and the substrate, D_0 is the free-particle diffusion coefficient, $x_i^r(t)$ is a random displacement sampled from a Gaussian distribution with zero mean and width $\langle x_i^r(t)^2 \rangle = 2D_0\Delta t$, and Δt is the time step.

In our simulations we move N particles (N varies between 900 and 1225 particles) of diameter σ according to Ermak's algorithm in a line of length $L/d=N$; N is properly adjusted to satisfy the continuity of the substrate on each border of the line. Periodic boundary conditions are considered only in the x direction; movement in any other direction is not allowed. Since the interparticle interaction is of long range and strongly repulsive, particles cannot feel their corresponding hard core, as we will see further below; however, its value is $\sigma \simeq 0.4077d$ [17]. The time step used in the BD simulations is $\Delta t = 2 \times 10^{-4}(\rho^2 D_0)^{-1}$. The structure is corroborated by standard Monte Carlo (MC) computer simulations [21]. A typical MC run consists of 1×10^6 steps to reach thermal equilibrium and 5×10^6 steps to perform the statistics. The maximum time reached in the BD simulations is $t_{\max} = 120(\rho^2 D_0)^{-1}$—i.e., 6×10^5 time steps. During our simulations the averages are taken over at least ten different independent stochastic realizations to reduce the statistics uncertainties and the substrate strength was initially chosen to be

zero and gradually increased in steps of $\Delta V=0.1k_BT$ to allow particle fluctuations across the substrate barriers until they reach the desired substrate strength. Nevertheless, in each step, 7×10^4 time steps are then allowed to reach properly the thermal equilibrium during the intermediate steady states. Furthermore, energy corrections, due to the long-range nature of the interparticle potential (1), are not explicitly considered in our calculations. One should recall that energy corrections are only needed when the exponent in power-law-like potentials is smaller than the system dimension [21], which is not the case in the present work. In addition, energy corrections are not needed if the length of the simulation box is sufficiently large to guarantee that the potential is, from numerical point of view, zero at distances comparable to $L/2$ [in our case it is around $\beta u(L/2)\approx10^{-4}$]. In addition, we carried out an extensive analysis of the influence of the system size. We found that our results are independent of N when $N>600$ (data not shown).

III. SUPERPARAMAGNETIC COLLOIDS IN A FREE SINGLE FILE

The structure and dynamics of paramagnetic colloids can be analyzed by varying the interaction strength Γ while keeping constant the particle number density. As we already mentioned above, this is equivalent to changing the latter one because an increase in Γ results in an increase of collision rates between particles [8].

Figures 1(a) and 1(b) show the pair distribution function $g(x)$ for different values of Γ. One immediately observes that the distribution of particles along the file behaves similar to that already well known in open systems (2D or 3D). In addition, for moderate strengths [Fig. 1(a)], $g(x)$ shows a typical fluidlike order; i.e., it decays to its corresponding ideal-gas value, $g(x)\approx1$, after a few mean interparticle distances. However, at higher strengths [Fig. 1(b)], the system becomes more structured and highly correlated at longer distances. In both cases, the characteristic length scale is basically determined by the mean interparticle distance $d=\rho^{-1}$; the peaks are successively separated by the distance d. This fact is clearly corroborated by the structure factor $S(q_x)$ shown in Fig. 1(c), where the main peak is located at the position $q_xd\approx2\pi$. Our results are corroborated with Monte Carlo simulations (solid lines) which basically reproduce the same structural information.

To clarify the long-ranged correlation between colloids for high interaction strengths, we also analyze the envelope of the $g(x)$. It is well known that an envelope which decays algebraically can be associated with high ordering in contrast with the exponential decay which is directly related with a fluidlike structure. Figure 2 depicts some pair correlation functions already shown in Fig. 1 together with their corresponding envelope. This figure is plotted logarithmically; this representation is suitable for studying the decay of the pair correlation function. We observe that for small Γ, the envelope can be best described by a single exponential (see solid lines for $\Gamma<4$). This clearly demonstrates the short-ranged nature of correlation between particles. However, for $\Gamma>4$ the envelope is best fitted by using an algebraic fit.

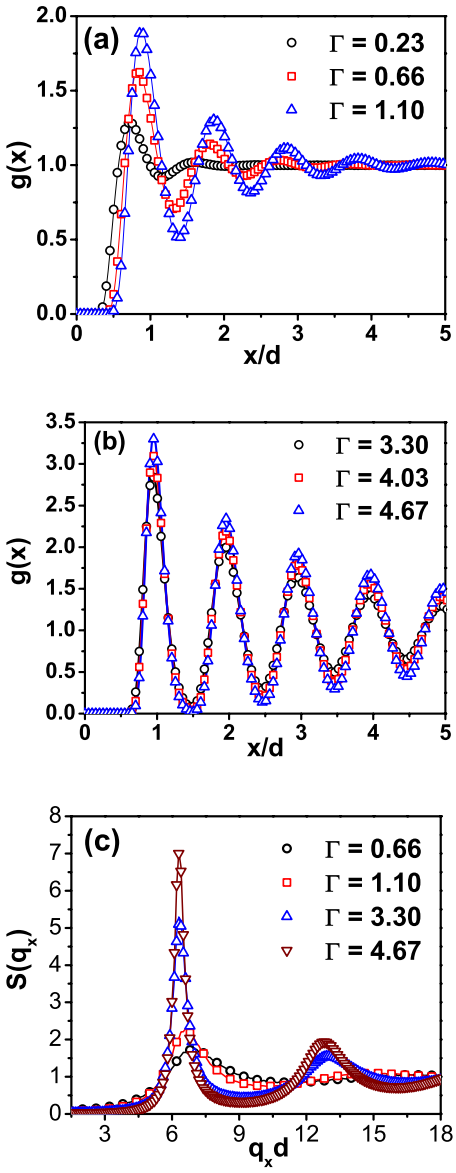

FIG. 1. (Color online) Pair distribution function $g(x)$ for (a) moderate and (b) high interaction strengths Γ. (c) Static structure factor $S(q_x)$ (symbols) obtained from BD simulations. The structure is corroborated by means of MC simulations (solid lines).

This behavior evidences the long-ranged ordering of the colloids along of the channel. Such interesting transformation from the liquid-state microstructure to quasicrystal (periodic) microstructure will be studied in detail elsewhere.

Diffusion properties, characterized by the reduced mean-square displacement $W^*(t)\equiv W(t)/d^2$ and the reduced single-file mobility factor $F^*\equiv F\rho/D_0^{1/2}$, are shown in Figs. 3(a) and 3(b), respectively. They are in qualitative agreement with previous experimental results [8,11,12,22]. Particularly, we observe that at sufficiently short times, $t<10^{-3}(\rho^2D_0)^{-1}$, where the displacement of individuals particles is governed by the interaction with the solvent, the diffusion is normal—i.e., $W(t)\propto t$. By increasing the time, the presence of adjacent particles becomes more important until eventually a crossover occurs at times $t>10^{-1}(\rho^2D_0)^{-1}$ in which the dynamics becomes subdiffusive with an anomalous exponent α

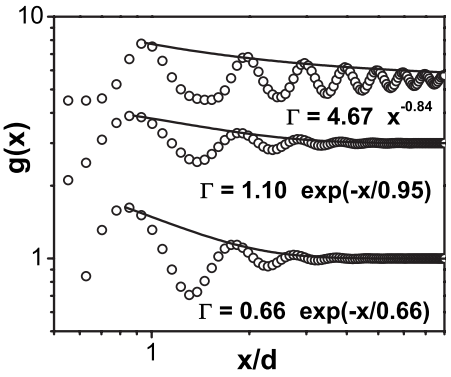

FIG. 2. Pair distribution function $g(x)$. The curves are displaced in a vertical direction for clarity. From bottom to top the particle strengths are $\Gamma=0.66$, 1.1, and 4.67. The envelopes are indicated by solid lines.

$=1/2$—i.e., $W(t)\propto t^{1/2}$. The crossover time from normal diffusion to subdiffusion occurs at earlier times as Γ is increased. This behavior is due to the fact that by increasing the interparticle interaction strength the direct particle-particle interaction becomes more important at shorter times and the energetic barrier imposed by the other neighbor colloidal particles reduces the particle acceleration [8,14]. Moreover, the particle mobility decays with increasing Γ in an exponential-like form [Fig. 3(b)] in agreement with experiments [5,8,11,12], theory [5] and simulations [13,14].

The results depicted in Figs. 1 and 3 are very similar to those reported previously in Figs. 1 and 2, respectively, of

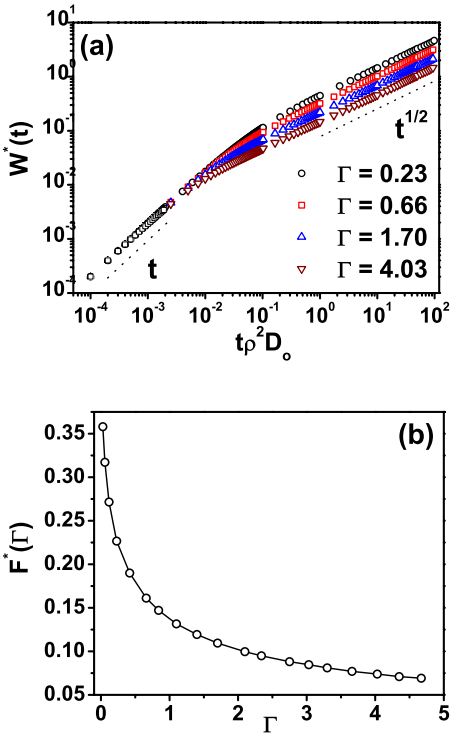

FIG. 3. (Color online) (a) Reduced mean-square displacement $W^*(t)\equiv W(t)/d^2$ for different particle interaction strengths Γ. (b) Reduced mobility factor $F^*\equiv F\rho/D_0^{1/2}$ as a function of Γ extracted from the fit to the simulation data according to relation $W(t)=2F\sqrt{t}$. The solid line is just a guide for the eye.

Ref. [14]. In general, both systems show a transition from fluidlike order to long-ranged ordering and a linear diffusion at short times (basically in the same interval of time). Furthermore, at longtimes, the non-Fickian behavior is always observed no matter the nature of the interaction potential. However, the transport coefficient or mobility factor is slightly smaller for the superparamagnetic colloids than in the charged case; this leads to a slower particle diffusion. Then, the resemblances between both systems confirm that a variation in Γ (which can be due to a change in the temperature), at fixed density, produces the same effect in both static and dynamic properties that a variation in the particle concentration keeping constant the interaction potential.

IV. SUPERPARAMAGNETIC COLLOIDS IN PERIODIC SUBSTRATES

Systems which possess two or more competing length scales are very common in nature. The competition between different length scales leads to a large variety of structures and commensurate-incommensurate (C-I) phase transitions. These systems have received intense attention during the last few years (see, e.g., Refs. [7,23,24]).

The simplest model system with competing length scales was proposed by Frenkel-Kontorova (FK). This model has been extensively discussed, and we here only mention its main features, which are necessary for the basis of our discussion. The FK model is a one-dimensional model of a system of particles treated as a harmonic chain with equilibrium lattice spacing a adsorbed on a periodic substrate with a periodic lattice of period b. It has been used successfully for the description of a vast number of different condensed matter systems and phenomena. It provides a simple and realistic description of C-I transitions when thermal fluctuations are unimportant as they are basically at zero temperature [15,16]; i.e., the system energy is only characterized by the potential energy. Then, by analyzing the energetic landscape of the system one can understand the origin of commensurable and incommensurable phases described in the FK model [7,23,24].

The FK model shows commensurate structures with the average spacing between adsorbate particle a rational multiple of b; this means $\tilde{a}=(p/q)b$ with p and q relatively prime integers consisting of periodically repeated unit cells of length pb containing q adsorbate atoms for every p substrate minima. For an interesting discussion of the FK phase diagram, see Ref. [16].

Interestingly, the FK model and our model system possess similarites. It is therefore expected to find structural properties similar to those already found in the FK model [16]. However, we should emphasize that, on the one hand, we are considering a system where the adsorbed particles interact continuously through the potential given by Eq. (1); each particle interacts with its nearest neighbors as well as with further neighbors due to the long-ranged nature of the interaction potential; i.e., we consider explicitly anharmmonic interactions and, on the other hand, thermal fluctuations will play an important role, as we will see further below, and then cannot be completely neglected. Then, the energy potential can be written as

$$U = \sum_{i<j} u(x_{ij}) + \sum_{i} V_{ext}(x_i;a_L), \tag{6}$$

where the first term takes into account the pair interaction which is given by (1), while the second term describes the particle interaction with the periodic substrate,

$$V_{ext}(x;a_L) = V_0 \sin\left(\frac{2\pi x}{a_L}\right), \tag{7}$$

V_0 being the substrate strength, x the particle position, and a_L the substrate periodicity.

A. Structure

In our previous work [14], we demonstrated that a one-dimensional colloidal system with anharmonic interactions and subject to periodic external fields provides new structural phases. Nonetheless, different interparticle potentials, in general, lead to new phases. Particularly, Ref. [14] uses a treatment with short-range interactions (Yukawa-like). In this work, we deal with long-range interactions with a dipolelike form.

Before to discuss our results, it is convenient to remark that in the absence of a substrate ($V_0=0$) we found that colloids form a highly ordered fluid with a characteristic length scale given by the mean particle distance $d=\rho^{-1}$. From d and the substrate periodicity a_L, we define the so-called commensurability ratio $p \equiv d/a_L$ [14]. This factor allows us to characterize the resulting type of commensurate, or incommensurate, structure. We should notice, however, that a variation in p will be related with a change in a_L.

Figure 4(a) shows the correlation function (symbols) for the case $a_L=\sigma$ and $\Gamma \equiv 4.08$, which corresponds to $p \approx 2.45$, for three different substrate strengths $V_0=1.6k_BT$, $2.8k_BT$, and $4.4k_BT$. According with the FK model, this system is close to a commensurable phase with $\tilde{a}=\frac{5}{2}a_L$. This means that we expect a state with repeated periodic unit cells of length $5a_L$ that contains two colloids every five substrate minima. In Fig. 4(a) we also plot the positions of the peaks (solid lines) corresponding to the case $V_0=0$ (multiples of $d \approx 2.4\sigma$). One clearly observes that the substrate induces changes in the local structure of the suspension, in which the maxima position is shifted with respect to the case without substrate. This implies that, on the one hand, the potential will always modulate the chain even in the case where it is not strong enough to force the commensurability and, on the other hand, substrate-induced correlations at separations smaller than d are due to there being almost three substrate periods for each mean separation. Both effects become stronger with the substrate strength; as V_0 increases the tendency of adsorbed particles to seek potential minima to also increase. Furthermore, the competition between colloid-colloid and colloid-substrate interactions leads to a distortion of the neighbors layer which is not present in the substrate-free case. This deformation affects the distribution of colloids along the channel, thus leading to a rich variety of adsorbate phases. In consequence, one can notice that the nearest neighbor is closer to any central particle than in the case without substrate whereas the second neighbor becomes closer to the first one, but it is further from the third one. Such interesting phenomena can better be visualized in the static structure factor $S(q_x)$. The latter one gives relevant information about the length scales of the system by looking at the position of its peaks. In Fig. 4(b) we show the $S(q_x)$ for the systems of Fig. 4(a). One can distinguish three peculiar peaks at the positions $q_x\sigma \approx 2.6$, 2π, and 8.8. These peak positions are related with the position of the first peak of the $g(x)$, the substrate periodicity, and the separation between the first and second neighbors, respectively. Then, the peaks at the positions $q_x\sigma \approx 2.6$ and $q_x\sigma \approx 8.8$ reveal that two adsorbed particles can be further and closer, respectively, than the distance between two consecutive substrate minima.

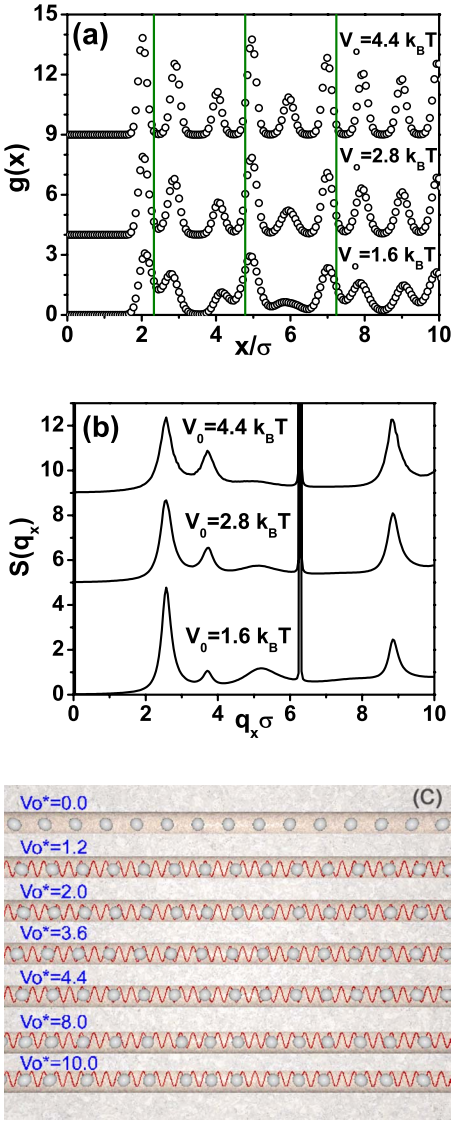

FIG. 4. (Color online) (a) Pair distribution functions and (b) static structure factors for the case $p=2.45$ with $\Gamma=4.08$ for three different substrate strengths. The perpendicular solid lines in (a) represent the position of each maximum for the case $V_0=0$ and the curves are shifted for clarity. (c) Average configuration of the particles along the channels. The sinusoidal term of Eq. (7) is plotted (dashed lines) on each channel to understand the particle ordering respect to the substrate minima.

These kinds of particle distributions, also observed in the FK model (see, e.g., [16]) and recently in charged colloidal systems [14], are usually called discommensurations because they break the commensurate registry of the adsorbate and substrate lattices. However, the height of the peak at $q_x\sigma = 2\pi$ indicates that there are regions on the substrate where the particles are located on the substrate minima. Figure 4(c) shows the snapshots of the average particle positions. Note that a commensurable phase is found for higher values of V_0 ($\geq 10k_BT$). However, the C phase that we observe is slightly different than that predicted by the FK model. In this case, we find a chaotic phase that is "pinned" to the potential. This means that the colloids are distributed in a random way among the substrate minima. This chaotic phase is similar to the commensurable phase, although the average period $\tilde{a}$ is, in general, incommensurate with the substrate period a_L. For a more comprehensive discussion about chaotic phases, see, for instance, Ref. [25].

We have also chosen the case $a_L = 2.4\sigma$. The purpose of this choice is twofold. First, it allows us to elucidate the features when $p=1$, comparing its properties with the case of harmonic interactions—i.e., FK model; in this case, the FK model predicts a commensurate structure with $\tilde{a}=a_L$. Second, it is related with the diffusion in the ground state $p=1$ and allows one to compare the dynamical properties with the cases $p>1$ and $p<1$, as we discuss below. Then, Fig. 5(a) shows the distribution function for this case; the values of V_0 are the same as in the previous case. We observe that the peak positions match with the substrate minima and basically they are located at the same position as in the case without substrate no matter the strength of V_0. This is easy to understand if one recalls that $d \approx a_L$, so that the natural length scale of particles matches with the substrate periodicity. For high values of V_0 it is expected that one adsorbate colloid sits on each substrate minimum. This scenario can be better appreciated in the static structure factor $S(q_x)$ [see Fig. 5(b)], which shows three main peaks at very-well-defined positions. The main peak is at the position $q_x\sigma \approx 2.6$, and the other peak positions are multiples of the main peak position, so they are at $q_x\sigma \approx 5.2$ and $q_x\sigma \approx 7.8$. This confirms that each particle is absorbed on every potential minimum. To visualize this picture, Fig. 5(c) shows the snapshots of the average particle position. For $V_0 = 8k_BT$ the commensurate structure can be nicely observed.

We now discuss the case $p<1$. This case is depicted in Fig. 6 for $a_L = 3\sigma$ or $p \approx 0.82$. Immediately, one observes that $g(x)$ acquires a less pronounced structure as V_0 increases; the height of the maxima and minima decrease. This result, in contrast with the previous cases, suggests a kind of *loss* of correlation among the particles and a possible scenario where particles become depinned from the sinusoidal substrate—i.e., a depinning transition. This behavior is confirmed by the structure factor in Fig. 6(b). It shows practically one peak at the position $q_x\sigma \approx 2.69$ which is related to the mean interparticle separation, confirming then that the local order at short and large separations has notably changed. In addition, by looking at the height of the $S(q_x)$ at the position $q_x\sigma = 2\pi/3$ (and integer multiples of it) one can appreciate that for $V_0 < 2.8k_BT$ there are no particles on the substrate minima. However, for higher values of V_0 some particles begin to be located around the potential minima, but no commensurable phase is observed. This is another piece of evidence of the depinning effect; it is also observed in Fig. 6(c) where it is shown that the particles are distributed in a random way along the substrate and only a few particles sit on the potential minima. Nevertheless, we also find that at $V_0 \approx 10k_BT$ the system is close to a commensurable phase and the pinning transition is recovered for $V_0 > 15k_BT$ (data not shown) where a commensurable phase with $p=9/11$ is observed—i.e., 9 colloids every 11 substrate minima. We should point out that the loss of correlation was also observed in the case of short-range interactions [14]; nonetheless, the distribution of particles along the channel in such case is completely

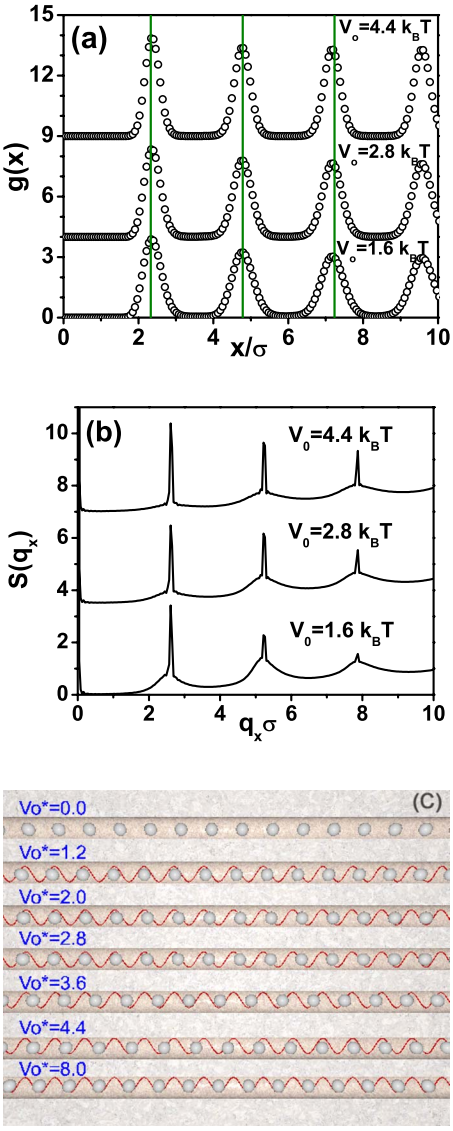

FIG. 5. (Color online) (a) Pair distribution functions and (b) static structure factors for the case $p=1$ with $\Gamma=4.08$ for three different substrate strengths. The perpendicular solid lines in (a) represent the position of each maximum for the case $V_0=0$ and the curves are shifted for clarity. (c) Average configuration of the particles along the channels. The sinusoidal term of Eq. (7) is plotted (dashed lines) on each channel.

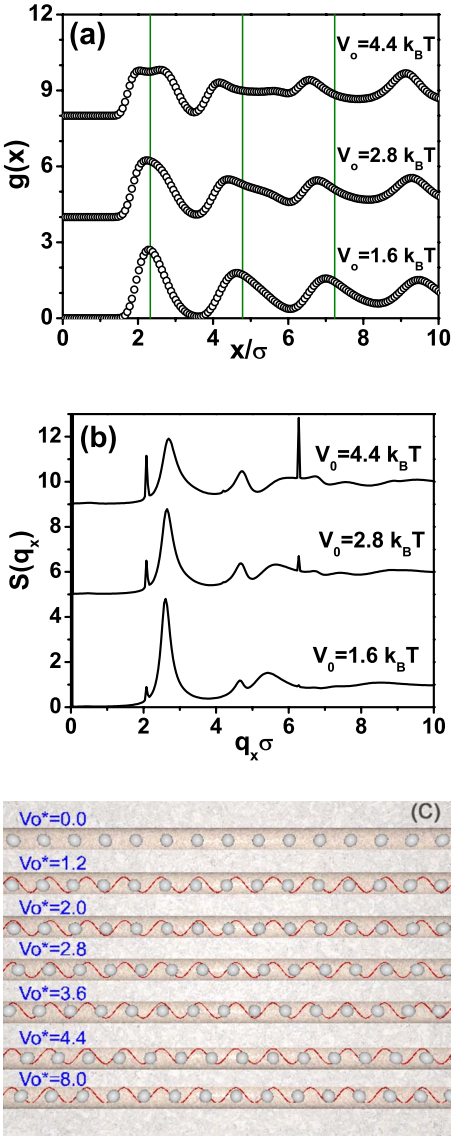

FIG. 6. (Color online) (a) Pair distribution functions and (b) static structure factors for the case $p=0.82$ with $\Gamma=4.08$ for three different substrate strengths. The perpendicular solid lines in (a) represent the position of each maximum for the case $V_0=0$ and the curves are shifted for clarity. (c) Average configuration of the particles along the channels. The sinusoidal term of Eq. (7) is plotted (dashed lines) on each channel.

different (see Ref. [14]). We come back to the discussion of the depinning transition below.

So far, we have analyzed the variation of the structure by changing the substrate parameters while keeping Γ constant. We clearly appreciate a similar behavior already discussed in our previous work with charged colloids [14], where the pinning-depinnig transition is also addressed and observed. This scenario indicates that the structure in one-dimensional systems with purely repulsive interactions and subject to external periodic potentials exhibits a *universal behavior*. This important characteristic gives us a unique opportunity for the structural manipulation by only changing the substrate parameters.

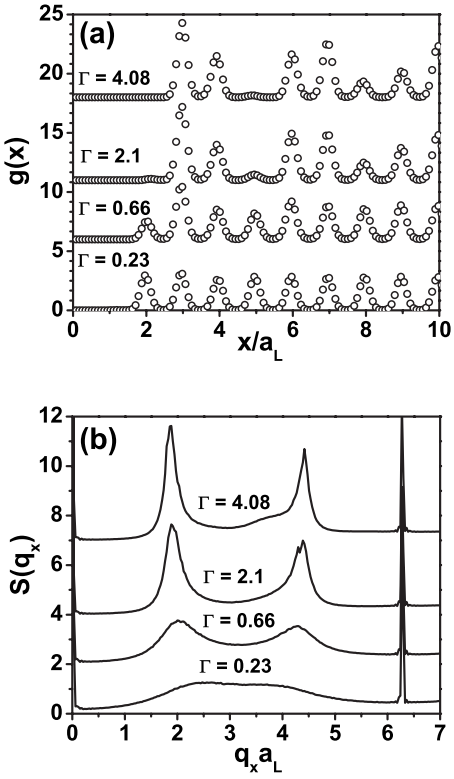

FIG. 7. (a) Pair distribution functions and (b) static structure factors for different values of Γ with $a_L=5.5$ μm and $V_0=3.6k_BT$. The curves are shifted for clarity.

To illustrate the effects due to the variation in Γ, we now fix the substrate periodicity $a_L=5.5$ μm [2] and substrate strength $V_0=3.6k_BT$. Figure 7(a) shows the distribution functions for several interaction strengths. We find that for small values of Γ (<1.1) the particles are sit on each substrate minimum because the pair-potential contribution is smaller than V_0. In fact, for $\Gamma<0.23$ each $g(x)$ collapses in the same curve (data not shown). In contrast, for higher interaction strengths, the pair interaction between particles becomes as relevant as the colloid-substrate potential and therefore the competition between both interactions lead to several incommensurable phases. Furthermore, the structure factor in Fig. 7(b) also indicates, as in the previous cases, that by increasing Γ the distortion in the neighbors layer becomes higher; i.e., colloids are nonuniformly distributed along the substrate and the particle ordering cannot be described by a unique length scale and, on average, two colloids are separated by distances larger than the distance between two consecutive substrate minima (see the two peaks before the position $q_xa_L=2\pi$). This structural behavior is qualitatively similar to that reported in [14], although the latter one is due to a variation in the particle concentration.

B. Dynamics

In general, particle transport or diffusion in one-dimensional channels under periodic substrates has been reported to be slower than that in the homogenous case (substrate free), but its magnitude depends on the energetic

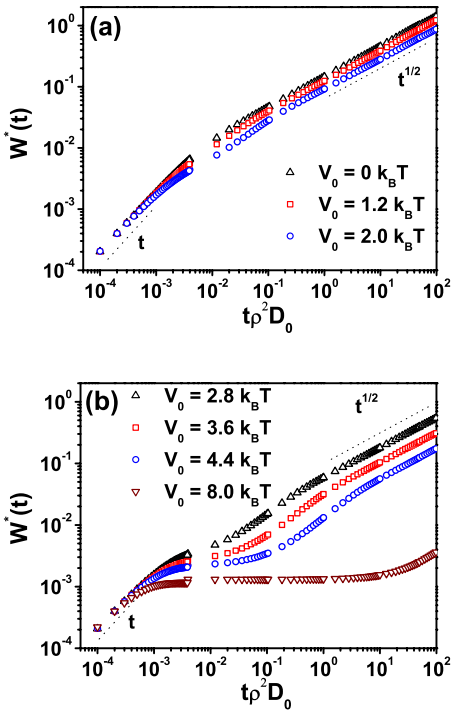

FIG. 8. (Color online) Reduced MSDs for different substrate strengths with parameters $p=2.45$ and $\Gamma=4.08$.

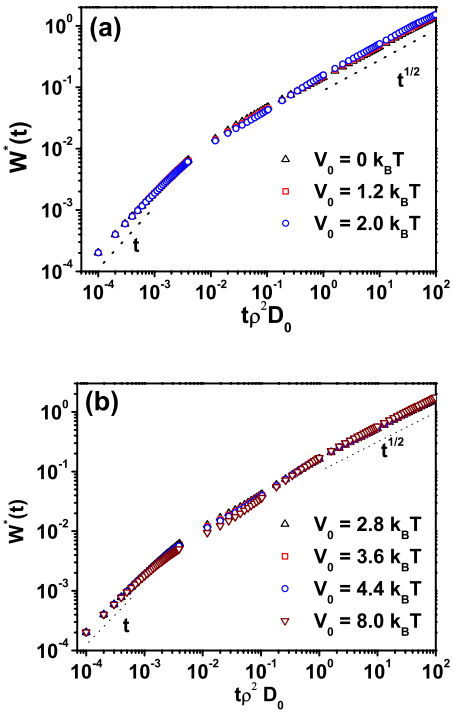

FIG. 9. (Color online) Reduced MSDs for different substrate strengths with parameters $p=0.82$ and $\Gamma=4.08$.

balance between the particle-particle and particle-substrate interactions [13,14]. In particular, we now extend our previous study [14] to the case with long-range interparticle potentials and stronger substrate strengths.

The reduced mean-square displacements $W^*(t)$ for $p>1$ are shown in Figs. 8(a) and 8(b). We appreciate that at short times ($t<10^{-3}/\rho^2 D_0$) normal diffusion occurs due to the particles and do not feel the direct interactions with the other colloids—i.e., $W(t)\propto t$. Nonetheless, at long times the dynamics process becomes subdiffusive and is described by the non-Fickian relation $W(t)\propto t^{1/2}$. For small V_0, the transition from normal diffusion to subdiffusion occurs continuously, however, for high V_0 the diffusion of particles is severely affected by the substrate; at intermediate times particles diffuse slower than at short and long times. The reduction in the particle diffusion is due to the energetic barrier imposed by the substrate. This causes the particles diffusion (or oscillate) around each substrate minimum for a long period of time before reaching diffusive collective motion. Therefore, the time required to surmount the energetic barrier increases with the substrate strength. In particular, for $V_0=8k_BT$, we observe that in the interval $10^{-3}<t\rho^2D_0<10^1$ the reduced MSD exhibits a plateau which indicates that the system is probably trapped in a glassylike state; however, at longer times $t\rho^2D_0>10^1$, the MSD increases again, indicating that the non-Fickian behavior can probably be reached at much longer times. For larger substrate strengths, we have also found pinned or locked states in which particles cannot escape from the substrate minimum and thus the diffusion or mobility goes quickly to zero (data not shown). Additionally, the mobility factor decreases when V_0 increases (see open circles in Fig. 10).

The reduced MSDs for $p=1$ are quantitatively similar to the case previously discussed (data not shown); both show a comparable behavior at all time regimes. However, as illustrated in Fig. 10 (open squares), the mobility factor is, in general, smaller. This is due to the system being in a commensurable state and hence particles quickly tend to seek the adjoining potential minimum; they oscillate around it for a long time. Interestingly, we have shown that for $p\geq 1$ the mobility factor decreases with V_0 no matter the kind of interparticle potential [14]. Nonetheless, the diffusion of superparamgnetic colloids is slower than the case of charged colloids (see also Fig. 9 in [14]).

The diffusive process for $p<1$ is depicted in Fig. 9. One observes that for any substrate strength $W(t)$ behaves almost as a free single file ($V_0=0$), although the (small) differences can be noticed at long times. These differences are related with the nonmonotonic variation of the reduced mobility factor shown in Fig. 10 (open triangles). Surprisingly, at moderate substrate strengths the mobility factor F increases and takes values larger than the case without substrate until reach a maximum at $V_0\approx 8k_BT$. This dynamic effect implies a nonmonotonic behavior of the particle transport and, clearly, it is a piece of evidence of the depinning of the particles from the substrate and is due to the so-called noise-assisted effect in which thermal fluctuations act cooperatively leading to a higher mobility. This depinning transition can be interpreted as follows. We should notice that for $p<1$ there are more particles than substrate minima which diffuse almost *freely*. Then, those *free* particles keep dynamical coupling along the channel with the other particles which, in principle, can occupy a substrate minimum. Nevertheless, this dynamical coupling and the system noise (thermal fluctuations) cooperate to give a dynamical mode which leads to the depinning effect without affecting the subdiffusive process at long times. Additionally, this phenomenon, already seen in the

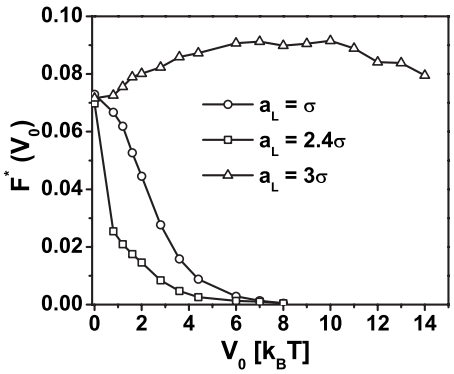

FIG. 10. Reduced mobility factor F^* as a function of the substrate strength V_0 for different substrate periodicities $a_L=\sigma$, 2.4σ, and 3σ with $\Gamma=4.08$. The line is a guide for the eye.

diffusion of a single Brownian particle drifting down a tilted washboard potential [26] and in the subdiffusion of charged colloids on periodic substrates [14], is also responsible for the loss of correlations of particles along the channel already discussed in Fig. 6. In the absence of thermal noise—i.e., in FK-like models—such an effect cannot take place. For $V_0 > 8k_BT$, the mobility factor decreases again, indicating that the substrate eventually starts to dominate the diffusive process of the particles. This means that for high V_0 the dynamical cooperative effect is not so strong to remove particles which are located on some of the substrate minima, then making more difficult the particle transport—i.e., leading to a slower diffusion.

To understand completely the diffusion properties of paramagnetic colloids on narrow corrugated substrates, we must take into account variations in Γ. In Fig. 11(a), the MSDs for the system described in Fig. 7 are shown. Interestingly, one can observe that at short and intermediate times the behavior is basically independent of Γ. In addition, the time needed to reach the diffusive collective motion seems to be the same no matter the value of the interaction strength and the asymptotic behavior at long-times scales according to the non-Fickian law $W(t) \propto t^{1/2}$. Nonetheless, the corresponding mobility factor decreases monotonically with Γ as can be noticed in Fig. 11(b). Furthermore, one can observe a similar exponential-like decay as in the homogeneous case, but we should note that the magnitude of the mobility is dramatically less. This reduction is a clear indication of the amount in which the periodic substrate can affect the transport of particles along the substrate.

Finally, we should remark that the diffusion properties of superparamagnetic colloids under one-dimensional periodic potentials share similarites with those of the charged colloids. In both cases, we notice that the diffusion decreases if $p \geq 1$ and increases when $p < 1$ and $V_0 < 10k_BT$; this result is independent of the kind of interaction potential between colloids. Thus, our simulations predict that the diffusion mechanisms can be also manipulated by only changing the substrate properties accordingly.

V. CONCLUSIONS

Colloids play a prominent role as model systems that allow highlighting the general principles and mechanisms of

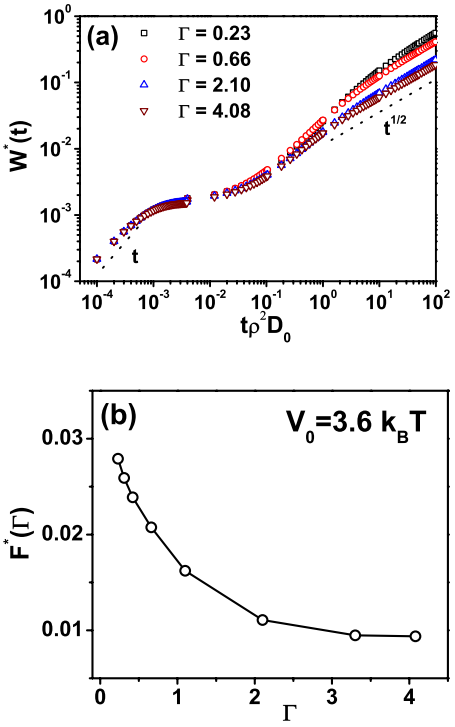

FIG. 11. (Color online) (a) Reduced mean-square displacement $W^*(t)$ for the case $a_L=5.5$ μm with $V_0=3.6k_BT$ for different interaction strengths Γ. (b) Reduced mobility factor F^*. The line is a guide for the eye.

phase transitions in systems with attractive and repulsive interactions. In particular, we have investigated both the structure and dynamics of superparamagnetic colloidal particles in narrow channels and subject to one-dimensional periodic potentials.

We observed that commensurable and incommensurable phases appear due to the competition between both the particle-particle and particle-substrate interactions and their corresponding length scales. We found that there is a deformation of the neighbors' layer which affects the distribution of colloids along the channel.

We have also shown that the depinning can directly be quantified through a loss of correlations in the structure or the enhancement of the transport coefficient. This effect seems to be purely cooperative due to the coupling between the thermal noise and the dynamical modes of the *free* particles diffusing on the substrate. Moreover, our results have allowed extending the understanding of the single-file diffusion in systems composed of interacting Brownian particles under one-dimensional modulated substrates, thus validating the non-Fickian law for systems of narrow corrugated substrates. These results can be corroborated in experiments with light forces. Experiments in such a direction can also corroborate the file depinning threshold reported here.

Here, we confirmed that both structural and dynamics properties of one-dimensional systems subject to periodic substrates possess a universal behavior when particles interact repulsively no matter the origin and/or range of the interaction. This information can be used to manipulate the system properties using external fields.

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