Single-file diffusion in a multilayered channel

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We demonstrate that strongly repelling Brownian particles confined in two-dimensional microchannels with multiple layers show single-file diffusion. At long times the mean-square displacement (MSD) is proportional to $t^{1/2}$, as in the one-dimensional case. On an intermediate timescale the MSD is further reduced. It scales with a minimal exponent that decreases with the number of layers in the channel. In the limit of infinite width of the channel, the MSD time evolution shows a crossover to a logarithmic time dependence.

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I. INTRODUCTION

We carried out Brownian dynamics simulations of interacting superparamagnetic colloidal particles in two-dimensional microchannels. Superparamagnetic colloidal particles at interfaces can serve as an ideal model system to study crystallization in two dimensions [1–6]. The interaction between the particles can be tuned by an external magnetic field. The interaction strength can be viewed as an inverse temperature of the system. As one increases the external magnetic field the interparticle repulsion increases and the particles start to freeze into a triangular lattice.

The two-dimensional crystal is special since it has no true long-range order, which has been proven by Mermin [7,8]. This is also true for the interaction potential used in our study [7]. Instead, the two-dimensional crystal exhibits only quasi-long-range order. Thus the displacement from perfect lattice sites increases logarithmically with distance in two dimensions. Therefore, the mean-square displacement also does not converge to a constant, but diverges on long timescales due to elastic deformation of the two-dimensional crystal.

For the system of superparamagnetic colloids the crystallization is a two-step process with an intermediate hexatic phase with a sixfold orientational order only and a solid phase with both translational and orientational order. This two-dimensional melting is well understood by the formation of topological defects (Kosterlitz-Thouless-Halperin-Nelson-Young theory [1–3,9]), whereas less is known about the dynamics in the solid phase, because of the exponentially diverging timescales.

To overcome this problem we study the transition from narrow quasi-one-dimensional channels to the infinite two-dimensional crystal as a function of channel width L_Y at constant density. Particles in restricted two-dimensional microchannels are also well studied [10–14]. In this confined system the particles form layers parallel to the walls. This layering takes place at interaction strengths similar to those of the phase transition in the unrestricted system. If the channel width L_Y is an integer multiple of the layer spacing R of the triangular lattice, then the layering is assisted by the walls and occurs at lower interaction strengths. However, in the case of a misfit such that the channel width L_Y is in between two of these stable situations, the layering is suppressed up to quite high interaction strengths [11]. As the interaction strengths increases, the crossover from n_l to $n_l + 1$ layers gets sharper [12].

It is well known that for narrow channels, with only one layer, the particles undergo single-file diffusion (SFD), due to their sequential ordering [15–31]. The long-time evolution of the mean-square displacement (MSD) is not proportional to the time, as it is in the case of normal diffusion, but proportional to $\sqrt{t}$. The long-time limit in a one-dimensional channel with interacting particles is given by

$$\langle \Delta x^2(t) \rangle = 2F\sqrt{t}, \tag{1}$$

with single-file mobility F, which is determined by the average space a particle has before it encounters another particle. The first sign of single-file diffusion has been found by Hodgkin and Keynes [15] in the permeability of potassium in nerve fibers. The exponent $1/2$ in Eq. (1) has been theoretically derived for Brownian particles by Harris [16] and for a one-dimensional lattice gas by Richards [17] and Fedders [18]. Single-file diffusion was observed by Hahn *et al.* [19] and then directly in real-space experiments in colloidal ring systems by Wei *et al.* [20]. In recent years it has been found that SFD is a more general phenomenon that occurs in many different systems [21–31].

In narrow channels with a few layers slightly below the liquid-solid phase transition the MSD shows single-file diffusion on an intermediate-timescale, but only if layering is favored by the channel width. The intermediate diffusion exponent has a nonmonotonic dependence on the width L_Y of the channel [13,25].

In this paper we concentrate on the high interaction limit above the liquid-solid phase transition. We are interested in the mean-square displacement $\langle \Delta x^2(t) \rangle$ along the channel direction. We will study how the single-file behavior with a mean-square displacement proportional to $\sqrt{t}$ in the one-dimensional case will cross over to a logarithmically diverging mean-square displacement in the unrestricted two-dimensional crystal. Therefore, the width L_Y of the channel

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is varied at constant particle density n and at high interaction strength beyond the liquid-solid phase transition. We will take advantage of the universal property of the diffusion exponent and compare the Brownian system to a simpler lattice-gas system, which is much less numerically expensive.

II. MODEL

We use Brownian dynamics simulation [32], which is an Euler integration of the overdamped Langevin equation. The positions $\mathbf{r}_i(t)$ of particle i are updated to a new position after the time step Δt,

$$\mathbf{r}_i(t+\Delta t) = \mathbf{r}_i(t) + \frac{D_0 \Delta t}{k_B T}\mathbf{F}_i + \sqrt{2D_0\Delta t}\mathbf{R}(t), \qquad (2)$$

with microscopic diffusion constant D_0, thermal energy $k_B T$, δ-correlated Gaussian random numbers $\mathbf{R}(t)$ with zero mean and unit variance, and the deterministic force $\mathbf{F}_i$ acting on this particle. The particles are restricted to the xy plane and confined in a channel. Periodic boundary conditions are applied in the x direction and hard-wall boundary conditions are applied in the y direction. The hard-wall boundary is realized by using the analytically known transition probability of a Brownian particle near a hard boundary as propose by Behringer and Eichhorn [33]. The length $L_X = 1000$ of the channel in the x direction is much larger than the width L_Y in the y direction to minimize finite-size effects. This is important, since it has been shown [34] for one-dimensional lattice systems with periodic boundary conditions, where the mean-square displacement of a tagged particle is computed by following the particle coordinate without renormalizing the coordinate due to periodic boundary conditions by adding or subtracting the lattice system length each time a boundary is crossed, that the exponent in Eq. (1) must cross over from 1/2 to 1 at long times due to a normal diffusion of the full particle system across the periodic boundaries. However, with sufficient channel length these timescales are larger than the simulation times.

The particles interact via the repulsive potential of parallel dipoles $V = \epsilon(\sigma/r_{ij})^3$, with interaction strength ϵ, particle diameter σ, and interparticle distance $r_{ij} = |\mathbf{r}_i - \mathbf{r}_j|$. We used a cutoff at a distance $r_c = 12.0\sigma$. This potential models the interaction of superparamagnetic colloidal particles in a perpendicular magnetic field, as they were used for experiments in two dimensions [5,6,20,35,36].

The melting in this system is described by the dimensionless control parameter [10]

$$\Gamma = \frac{\epsilon}{k_B T}\frac{\sigma^3}{R^3}, \qquad (3)$$

which is given by the ratio of potential energy to thermal energy $k_B T$. The system is in a solid phase for $\Gamma > 15$. There is also a hexatic phase for $15 > \Gamma > 12$ with only orientational order, which is discussed, e.g., in [37] for different power-law potentials. We compare the results with those of a quasi-one-dimensional lattice-gas model in channel geometry, in which the particles can only move in one dimension in their own layers parallel to the channel walls, but also interact via an exclusion process with particles on neighboring layers. The square lattice has a length of L_X in the periodic x direction and consists of n_l coupled layers in the y direction with freely moving boundary particles. At the beginning of the simulation the particles were placed equally spaced in each layer with a distance of $2d$ (d is an integer multiple of the lattice spacing), while the positions are shifted from layer to layer by d (see Fig. 1). In this manner the particles form a triangular lattice.

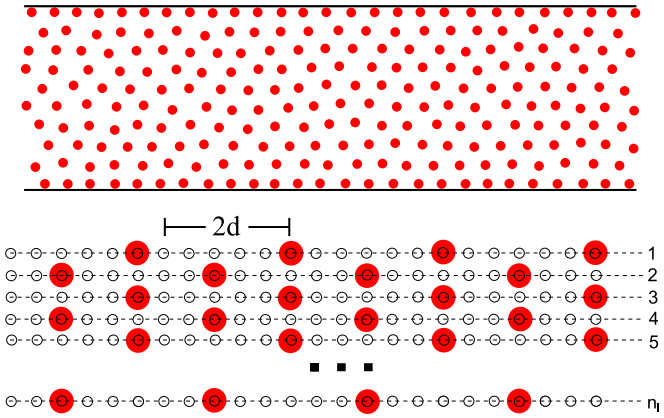

FIG. 1. Snapshot of the colloidal particles in a part of a channel of width $L_Y = 14.5$ forming $n_l = 10$ layers and a sketch of the initial condition of the lattice-gas model with particle spacing $d = 3$ and n_l layers.

In each step one of the N particles is randomly selected and attempts to move with equal probability to an adjacent lattice site either to the left or to the right. Hence the diffusion constant of the lattice gas is $\nu = 0.5$. The particles are prevented from moving if the targeted lattice site itself is occupied or one of the two sites on the adjacent layers is occupied. Due to the alternating starting condition, a particle will never attempt to move onto an occupied site. Thus the particle is caged by the four surrounding particles on the adjacent layers. In this manner, the particles keep their mutual order.

This model is equivalent to the two-dimensional cage model of Centres and Bustingorry [38]. In their model each particle is caged by the four surrounding particles and can move within this cage in the x and y directions. However, the motion in both dimensions is independent, since the accessible region for a move in the x direction only depends on the x positions of the four surrounding particles. The same is true for the motion in the y direction. Therefore, their model can be separated in two independent systems with one-dimensional dynamics but two-dimensional interactions. This is the model presented here.

This lattice-gas system can be seen as a model for diffusion in an ideal two-dimensional solid, where the particles keep their mutual order. In a realistic solid there is also a diffusion due to interchange of particles, which is closely related to the diffusion of defects. However, in our model the interchange of particles is suppressed on the timescales we study, due to rather long-range interactions and high interaction strengths.

III. RESULTS

All results of the Brownian dynamics simulation are given in units of particle diameter σ, thermal energy $k_B T$, and diffusion times $\tau_D = \frac{\sigma^2}{D_0}$. Simulations have been performed at fixed

particle density $n = 0.4$ in channels of length $L_X = 1000$ and variable width $1 \leqslant L_Y \leqslant 33.8$ with up to $N = 13\,520$ particles. The MSD has been calculated from 60×10^6 simulation steps and averaged over 50 systems with different random seeds and initial conditions. For the purpose of equilibration, the system starts with random particles positions and the interaction strength is slowly increased. We study the colloidal systems in the high interaction limit with $\epsilon = 200$. This corresponds to a value of the dimensionless interaction parameter of $\Gamma = 62.8$, which is far in the solid phase for the unrestricted two-dimensional system. The interesting nonlinear behavior near the critical Γ has already been discussed in Ref. [25].

In the two-dimensional system the particles form a triangular lattice. In channels the positional order perpendicular to wall is stabilized at the expense of the positional order along the channel direction. The particles form layers parallel to the wall with a spacing of $R = (\sqrt{3}/2n)^{0.5} \approx 1.47$. As one can see in Fig. 1, the density on the boundary layer is slightly higher, thus the stable formation of n_l layers is slightly shifted compared to the ideal case where the expected number of layers is $L_Y/R + 1$. With a higher number of layers this shift becomes smaller.

The crossover from the one-dimensional channel with only one layer to the two-dimensional channel with two layers is a bit vague, mostly because the definition of the density is different in one and two dimensions [39]. Eventually the particle will form two layers, where the particles tend to be alternately ordered at one of the two walls of the channel. The strict alternate order is not stable, since there is no long-range order in one dimension. Strictly speaking, this argument should be true for all finite width L_Y and all number of layers, but already three and four layers of particles are quite stable.

A. Diffusion along the channel

Regarding the diffusion along the channel direction there is no distinct difference between a system with two layers and a system with one layer. The particles perform single-file diffusion as long as the particles cannot bypass each other. However, what happens if one further increases the width L_Y of the channel at constant particle density? How does the crossover from the single-file diffusion in an ordered one-dimensional system to the diffusion in a two-dimensional crystal take place?

We are interested in the diffusion due to elastic lattice deformation, therefore we look at a system with strong repelling particles, where the interchange between two or more particles is suppressed. In the lattice-gas model, interchange of the particle order is strictly forbidden.

Figure 2(a) shows the evolution of the mean-square displacement of the colloidal particles along the x axis for different channel widths L_Y, at which the particles form stable layers. In the smallest channel of width $L_Y = 3.8$ the particles form three layers and show a similar evolution of the mean-square displacement as in the case of single-file diffusion. If the number of layers is increased, the mean-square displacement decreases. It shows an intermediate subdiffusion with an even smaller exponent below the single-file exponent 0.5, but the long-time evolution develops again with the single-file exponent. The single-file mobility F of this long-time evolution decreases as the number of layers increases. From these data it is not clear whether the single-file mobility vanishes in the limit of an infinite number of layers.

Figure 2(b) shows the time-dependent diffusion exponent

$$\alpha(t) = \left.\frac{d \log \langle \Delta x^2(t') \rangle}{d \log t'}\right|_{t'=t}, \tag{4}$$

which is defined as the logarithmic derivative of the MSD. The time-dependent exponent reveals that the long-time evolution is single-file-like for all channels with 24 or fewer layers. To go to even wider channels it would be necessary to not only increase the width, but also to increase the simulation time and the length of the channel, in order to avoid finite-size effects. Due to computer time restrictions, it is necessary to switch to the simpler lattice-gas model.

In the lattice-gas system the number of layers is varied up to a maximum number of $n_l = 600$ layers. The channels have a length of $L = 2500$ lattice sites per layer. The average particle spacing d is also varied between 2 and 6. The MSD has been averaged over 10^8 time steps and 50 different random seeds. With a proper rescaling of the time $t^* = t\frac{d^2}{\nu}$ and space $x^* = x\frac{1}{\sqrt{d(d-1)}}$, the MSD is nearly independent of d and mainly depends on the number of layers n_l. In the following only dimensionless (reduced) quantities are used and the asterisks are omitted.

Figure 2(c) shows the mean-square displacement of the lattice gas with a different number of layers n_l and different average particle spacings d. For $n_l = 2$ layers this model is equivalent to the single-file lattice gas, also known as a simple exclusion process in one dimension. The long-time limit of the mean-square displacement is analytically known to be $\langle \Delta x^2(t) \rangle = 2(\frac{t}{\pi})^{0.5}$ [34]. For a higher number of layers we see the same behavior as in the colloidal system; however, now we can extend the study to wider channels.

Here we show that the mean-square displacement converges for an infinite number of layers to a logarithmic time dependence. It can be expressed in a minimal form

$$\langle \Delta x^2(t) \rangle_{n_l \to \infty} = C \ln\left(1 + \frac{2t}{C}\right), \tag{5}$$

which is designed to fit the short-time linear diffusion and has one additional fit parameter C that describes the long-time logarithmic behavior. The fits to the MSD of $n_l = 300$ give slightly different values for different layer spacing d around $C \approx 0.25$. In Fig. 2(c) this curve is plotted with $C = 0.25$ as the limit $n_l = \infty$. The MSD for $n_l = 600$ does not deviate from this limit for times $t < 10^5$. This fit does not give the exact time evolution, but shows that the MSD increases logarithmically in time in the limit $L_Y \to \infty$.

Figure 2(d) shows the time-dependent diffusion exponent of the lattice gas. It shows the same behavior as the MSD of the Brownian particle in the limit of high interaction strengths. All curves up to $n_l = 20$ layers converge for long times to the single-file exponent 0.5 indicated by the horizontal line. For wider channels the simulation time is too short to reach this limit, since the timescales diverge exponentially. For the largest channel with $n_l = 600$ layers the time-dependent diffusion exponent is strictly monotonically falling in the interval of the simulation time.

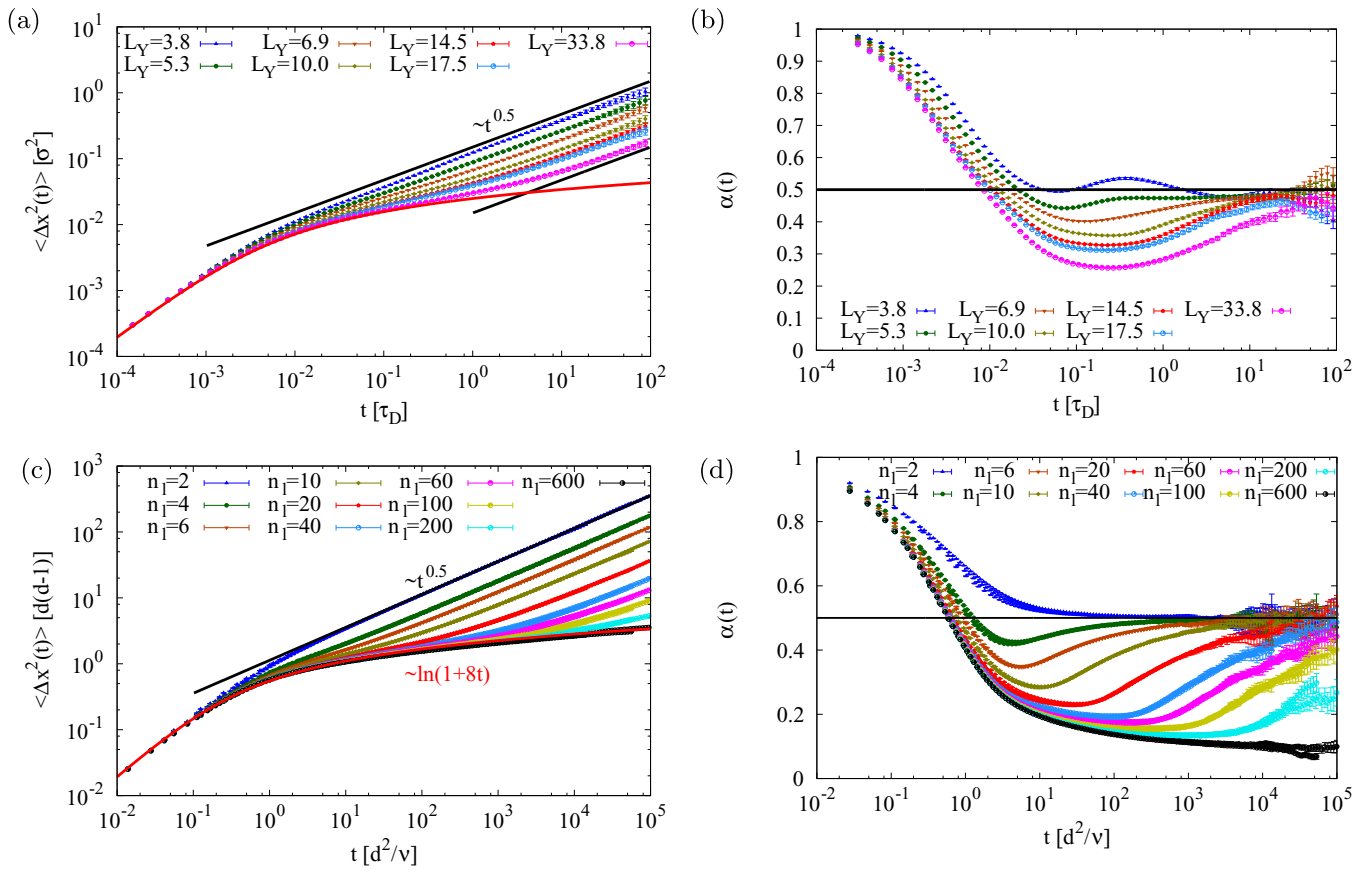

FIG. 2. (a) Mean-square displacement and (b) time-dependent exponent $\alpha(t)$ for the colloidal channel of width L_Y, corresponding to $n_l = 3, 4, 5, 7, 10, 12, 24$ layers, and (c) MSD in a master plot with all average particle spacings $d = 2, 3, 6$ and (d) time-dependent exponent $\alpha(t)$ in the corresponding lattice-gas system with n_l layers.

The diffusion exponent reveals another universal behavior. It seems that the minimum of the time-dependent exponent $\alpha(t)$ not only is independent of d in the lattice-gas system, but is also similar to the high interaction limit of the colloidal system for corresponding channel widths. Figure 3 shows the minimal diffusion exponent $\alpha_{\min}$, which is the minimal value of the diffusion exponent $\alpha(t)$. It is plotted as a function of the number of layers n_l for the lattice gas and as a function of the reduced width L_Y/R for the colloidal system. For the colloidal system the minimal exponent converges for large Γ to the limiting results of the lattice-gas system. For intermediate Γ it shows an oscillating behavior as a function of channel width (as in Ref. [25]).

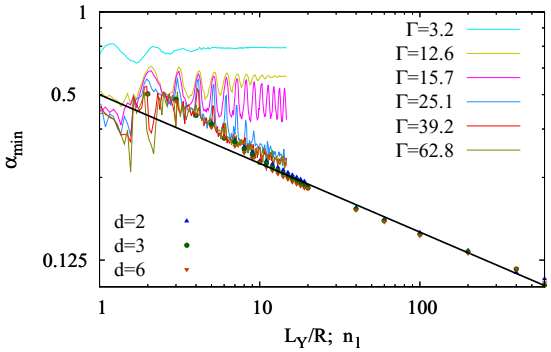

FIG. 3. Minimal diffusion exponent $\alpha_{\min}$ of the Brownian system as a function of reduced width L_Y/R for different interaction strength Γ and minimal diffusion exponent of the lattice-gas system as a function of the number of layers n_l with different mean particle separation d.

For layered systems with multiple ordered layers the minimal diffusion exponent is a universal quantity that only depends on the number of layers n_l. For a high number of layers the minimal exponent seems to follow a power-law decay that can be well described by

$$\alpha_{\min} = 0.5 n_l^{-0.25}. \tag{6}$$

Hence the minimal diffusion exponent vanishes in the limit of the two-dimensional system ($n_l \to \infty$). This implies the vanishing of the long-time single-file mobility F.

B. Diffusion in the direction orthogonal to the wall

Figure 4(a) shows the mean-square displacement orthogonal to the channel direction for the colloid system for different channel widths L_Y. The motion of the particles in the y direction is confined to their layers. Therefore, the mean-square displacement in the orthogonal direction stays finite. It converges for long timescales to a plateau. The plateau value increases with the number of layers in the channel. However, for the limit of an infinite number of layers the MSD in the y direction is the same as the MSD in the x direction,

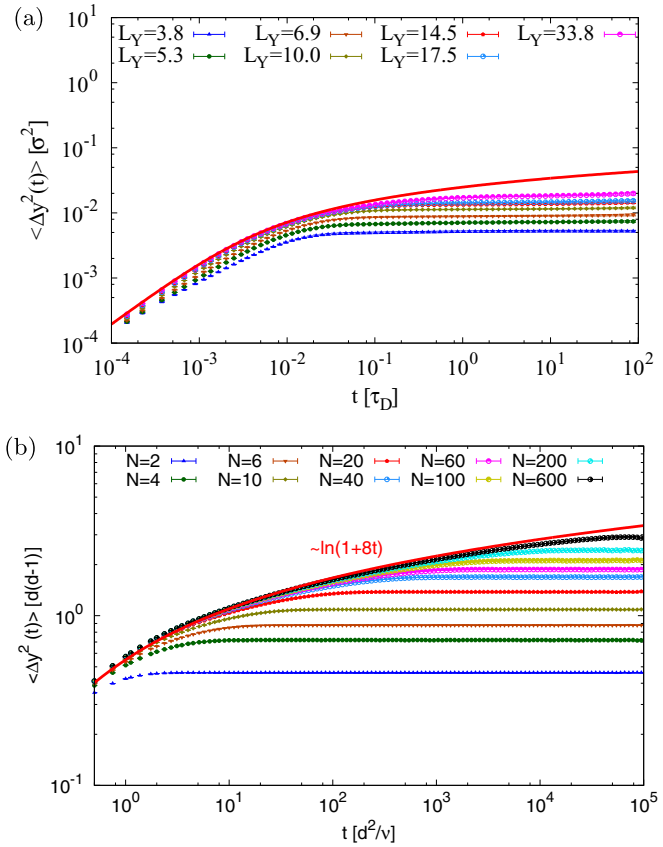

FIG. 4. (a) Mean-square displacement in the direction orthogonal to the walls for the colloidal channel of width L_Y and (b) MSD for the corresponding lattice-gas model with many layers and a finite length $L_Y = Nd$ with closed ends (only for average particle spacing $d = 2$).

since in the limit of infinite channel widths the influence of boundary conditions vanishes. Indeed, the MSD in the y direction approaches the same logarithmic scaling as in the x direction. However, the maximal channel width $L_Y = 33.8$ with only 24 layers is too small to reach the limit $L_Y \to \infty$. Again, the lattice-gas model helps to analyze the corresponding diffusion behavior in much larger systems. Figure 4(b) shows the mean-square displacement of the corresponding lattice-gas model with many layers, but only a finite length of $L_Y = Nd$ and closed ends. Here N corresponds to the number of layers in the Brownian particle system. Since the motion is limited to the finite length scale, the MSD stays also finite as in the colloidal system. With increasing channel lengths the long-time plateau rises and occurs at larger timescales. In the limit $L_Y \to \infty$ the mean-square displacement converges from below to the same logarithmic scaling as the system with periodic boundaries [Fig. 2(d)].

IV. DISCUSSION

Our results suggest that the mean-square displacement in two-dimensional crystals diverges logarithmically. It is well known that the displacement from the lattice position diverges logarithmically with distance in two dimensions [7], hence it is clear that the mean-square displacement should also diverge, but the time dependence was not known. Also a logarithmic scaling of the MSD with system size has been found in two-dimensional glasses [40].

The logarithmic time scaling of the MSD in a two-dimensional colloidal crystal has not been discussed properly in the literature. In Ref. [41], a logarithmic long-time evolution of the hard-disk MSD was discussed, based on the assumption of an algebraic time decay of the positional order parameter. However, in the cited literature the decay of the translational order parameter in time has not been studied carefully.

Nelson only showed that the bond orientation order parameter decays algebraically in time in the hexatic phase, thus resulting in a logarithmic long-time evolution of the mean-square bond angle difference [42]. However, assuming a dynamical critical exponent $z = 2$ for both hexatic and crystal phases of the two-dimensional colloidal system would lead to this algebraic time decay of the positional order parameter. Hence the MSD has a logarithmic time scaling. This is also supported by the analogy to the Edwards-Wilkinson model, which shows the same scaling behavior [38,43].

Here we note that these arguments are only valid for timescales where particles do not switch their positions. Since the interchange of two particles is separated by a finite-energy barrier, there will be an associated timescale where the particles perform normal diffusion. However, on the observed timescales the mean-square displacement is given by lattice deformations and not by the diffusion of defects.

In the single-file case a particle is confined by particles in its own layer. In channels with more than two layers the particle motions are restricted by particles of more and more layers involving increasing timescales. For short times the motion of a particle is restricted by the four particles left and right on the two adjacent layers. These particles are also restricted by particle on their adjacent layers. With increasing time more and more layers get involved. The diffusion exponent drops below the single-file exponent while more layers get involved in the diffusion process. At timescales where all n_l layers are involved, no more layers can restrict the particle and the diffusion exponent increases again to the single-file exponent.

To justify why the single-file exponent is also found as the long-time limit of diffusion in channels with multiple layers, an idea of the renormalization-group theory can be followed. Quadratic patches of $n_l \times n_l$ particles can be approximated as renormalized block particles that are ordered sequentially in the channel. These block particles cannot bypass each other if the particles are ordered in a triangular lattice structure. The interaction between these blocks may be complicated, but for single-file diffusion it is only important that they cannot bypass each other. Hence the diffusion of these block particles is single-file-like. Since the single particles are caged within their block particle, there is a timescale where the MSD of the single particle is dominated by the MSD of the block particle. On this timescale the diffusion of the individual particle takes place also with the single-file exponent.

V. CONCLUSION

Channels with a finite number of layers of colloidal particles show single-file diffusion in the long-time limit. This can be reasoned by the idea of block particles spanning the

whole width of the channel. These block particles are ordered in a single-file geometry and therefore show SFD. If the single particles are caged within their block, their long-time limit of the MSD will also be single-file-like. It would not be surprising to find SFD also in ordered three-dimensional channels and in quasi-one-dimensional glasses, since it is only necessary that the particles are not allowed to rearrange within a block, spanning the whole cross section of the channel.

In the limit of the two-dimensional solid ($L_Y \to \infty$) the single-file long-time evolution is postponed to infinity. The MSD only grows logarithmically in time and the time-dependent exponent declines towards zero. This crossover implicates a minimal exponent $\alpha_{\min}$ that also decays to zero with increasing channel width. The decay is given by a universal power law that only depends on the reduced channel width L_Y/R or the number of layers n_l. With this, the single-file mobility decreases by adding more and more layers and the MSD follows for longer and longer timescales the logarithmic scaling instead of the power law of SFD. In this sense the SFD vanishes in the limit of the two-dimensional solid.

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