Diffusion between evolving interfaces

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Abstract
Diffusion in an evolving environment is studied by continuous-time Monte Carlo simulations. Diffusion is modeled by continuous-time random walkers on a lattice, in a dynamic environment provided by bubbles between two one-dimensional interfaces driven symmetrically towards each other. For one-dimensional random walkers constrained by the interfaces, the bubble size distribution dominates diffusion. For two-dimensional random walkers, it is also controlled by the topography and dynamics of the interfaces. The results of the one-dimensional case are recovered in the limit where the interfaces are strongly driven. Even with simple hard-core repulsion between the interfaces and the particles, diffusion is found to depend strongly on the details of the dynamical rules of particles close to the interfaces.

1. Introduction
Diffusion phenomena are ubiquitous in nature, with familiar examples ranging from heat conduction to osmosis. Often diffusion occurs in a random or nonideal environment as it does, for example, in the presence of mobile or immobile (in the timescale of diffusion) lattice imperfections. Due to the complexity of real materials, transport has been considered within simplified theoretical frameworks, often utilizing the random-walk picture to describe some assumed underlying microscopy. Novel biophysical applications can be expected to emerge for transport restricted by soft (and fluctuating) interfaces forming narrow or even nanoscale channels [1]. In particular, the crossover from bulk-dominated to boundary-dominated diffusion is of considerable theoretical and experimental interest.

The study of random walks in a random environment (RWRE) has a long history and, since the results [2, 3] from the 1970s, as reviewed in [4], a vast amount of information has been accumulated. This randomness has been considered to manifest itself as non-homogeneous transition rates [5–7]. Spatially, (frozen) transition rates can sometimes be described by random walkers as in the Sinai model [8]. In general, theoretical studies have mostly been limited to models where the environment, including the possible geometric constraints, is either stationary or fast compared with the jump rate of the walkers. The mathematical problem of the random walk in a random environment has been considered in [9]. The asymptotics of diffusion in a continuum under a random forcing in the presence of damping were analyzed in [10] and diffusion in restricted geometries with homogeneous transition rates was considered in [11]. A two-species zero-range process [12, 13] with suitably chosen transition rates leads to dynamics which can be considered as a diffusing particle in an evolving environment [14]. However, in existing studies the focus has not been on (undriven) diffusion. In [15] Sanel et al considered a situation where particles are immersed in a background fluid inside a narrow channel. From the point of view of a single particle in the dilute limit, this could be interpreted as diffusion in a dynamic environment. In that work the focus was on the transition from single file dynamics to Fickian diffusion. The existing studies on particle dynamics in the presence of interfaces are mainly for particles immersed in a driven liquid [16]. A related problem, the influence of geometry fluctuations on lateral diffusion in biological systems, was studied very recently in [17], where it is noted that geometry fluctuations at a finite scale can affect diffusion at all scales.

In this paper, we consider diffusion in a dynamic restricting environment, inside open evolving ‘bubbles’ between two interfaces. For this, we combine two most simple models, the solid-on-solid (SOS) model of interfaces and the continuous-time random walk on a lattice, models that are widely used and known to describe well interface fluctuations and particle diffusion. To be more specific, we study diffusion on a lattice in the environment produced by the dynamics of the BCSOS2 model introduced in [18], containing two non-intersecting interfaces driven against each other. Thus, due to the interface dynamics, the actual transition rates of a diffusing particle become dependent on time and position, when the
jumps of the particle are possible only inside the ‘bubbles’ between the interfaces. Diffusion in the hydrodynamic limit will then depend on the dynamics of the bubbles, e.g. through their growth and merging. We shall concentrate on cases where the particles do not affect the motion of the interfaces so that the dynamics of the two interfaces, e.g. the bubble size distributions and their correlations, are in principle known [18] and thus, in addition to simulations, analytical arguments can be developed for various limits, which is the particular strength of the model. We use our model as a testing ground for various ideas describing different regimes of behavior of the diffusion coefficient. In addition, we study the possible consequences of various choices of microscopic dynamics on the interaction of the interfaces and the diffusing particle. To reduce the dimension of the parameter space, the models we combine are relatively simple. However, most of our results are expected not to be dependent on the details of the model, but are characteristic of systems where the size distribution of the bubbles and their dynamics (at the bubble scale) inside a material or at an interface between two materials become the rate-limiting factor for diffusion (at the hydrodynamic scale).

This paper is organized as follows. In section 2 we define the evolving environment provided by the two interfaces and the dynamic rules of the walker (particle) in detail. After that, in section 3, we describe the algorithms needed for efficient simulation of the combined dynamics and the sampling of the main quantities. An experimentally oriented reader could first skip sections 2 and 3 and proceed to section 3.4, where we briefly describe the parameters and their physical interpretation. Our results for the combined interface and particle dynamics are presented in sections 4 and 5. A concluding discussion of our results is given in section 6.

2. Models

2.1. Model for the interface dynamics

The evolving environment for diffusion is produced by the dynamics of the BCSOS2 model that we introduced and discussed in [18]. Below we give only a brief description of the BCSOS2 model so that the dynamical rules for diffusion become well defined for the present study. The BCSOS2 model is constructed by letting two one-dimensional BCSOS interfaces [20] interact with each other.

The location or the ‘height’ of a single BCSOS interface is described by a function \( h_i(x, t) \) such that, for every site \( x = 1, \ldots, L \),

\[
h_i(x + 1, t) - h_i(x, t) = \pm 1,
\]

where without loss of generality we assume that the possible values of \( h_i \) are integers. With this restriction on local configurations of the interfaces, only two kinds of processes, adsorption \( (h_i \) locally increases) and desorption \( (h_i \) locally decreases), are available. In our continuous-time model (see section 3), the parameters \( p_i \) and \( q_i \) give the transition rates of adsorption and desorption events, respectively, for transitions allowed by the condition (1). In what follows, time and various rates are measured in units where \( p_i + q_i = 1 \).

Figure 1. A snapshot of the two interfaces \( h_1 \) and \( h_2 \) and the possible jumps of a diffusing particle with their attempt rates \( \alpha, \beta, \gamma \) and \( \nu \). Also the corresponding difference \( h_+ = h_1 - h_2 \) is shown. The jumps of the particle are possible only within the bubbles (open spaces) between the interface. The jump rate \( \nu \) is for the one-dimensional motion (see dynamics \( m = 4 \) in text) depending only on \( h_+ \). To simplify the discussion, when possible, in the text the various rate parameters are collectively referred to simply as \( \mu \) in such a way that the total rate for the particle to attempt a jump is \( \mu \).

In the BCSOS2 model there are two interfaces, \( h_1(x, t) \) and \( h_2(x, t) \), such that \( h_1(x, t) \) is an even (odd) integer for odd (even) values of \( x \), see figure 1. The coupling between the interfaces is produced by demanding that they cannot intersect:

\[
h_1(x, t) \geq h_2(x, t) \quad \text{for all } x, t.
\]

We also impose the periodic boundary conditions \( h_1(x, t) \equiv h_1(x + L, t) \) for \( i = 1, 2 \). In the full BCSOS2 model, there are then four parameters \( \{p_1, q_1, (p_2, q_2)\} \) defining the transition rates for the interfaces \( h_1 \) and \( h_2 \), respectively. To further limit the parameter space, we shall restrict the discussion to the symmetric case \( p_1 = q_2 \) and \( q_1 = p_2 \) (see [18]) so that the behavior of the BCSOS2 system is defined by one parameter, the driving parameter \( f \) defined as

\[
f \equiv p_2/q_2 - 1.
\]

For large \( f \) the interfaces are strongly driven against each other and for \( f \to 0 \) they become free. We also define the sum and difference processes defined via \( h_{\pm} = h_1 \pm h_2 \), where the sum process \( h_+ \) describes the wandering of the interfaces together and the difference process \( h_- \) is positive inside the bubbles and zero elsewhere [18]. The non-crossing condition of equation (2) is equivalent to \( h_-(x, t) \geq 0 \). The interfaces \( h_{\pm} \) are then of the RSOS type [20], obeying \( h_{\pm}(x + 1, t) - h_{\pm}(x, t) = -2, 0, +2 \). An example of \( h_1 \) and \( h_2 \) and the corresponding \( h_- \) configuration is shown in figure 1.

2.2. Models for particle diffusion

We consider a single point-sized particle diffusing between the interfaces \( h_1(x, t) \) and \( h_2(x, t) \) on a lattice \((x, y)\). The lattice point coordinates in the horizontal direction are the same
x = 1, . . . , L as for the interface model above, again with periodic boundary conditions, cf figure 1. In the ‘vertical’ direction, the lattice is infinite and the coordinates are integers y = −2, 1, 0, 1, 2, . . . and thus coincide with the possible values of h1 and h2.

We shall denote the location of the particle by (x, y). The particle does not affect the dynamics of the interfaces but, if needed, a moving interface can push the particle the distance of one or two lattice units in the vertical direction such that the location of the particle also after the change of the interface configuration satisfies the condition

\[ h_2(x, y) \leq y \leq h_1(x, y) . \]  

(4)

These are moves of the particle forced by the interface motion.

For diffusive moves of the particle, the following two rules are imposed in all cases: first, for a jump \((x, y) \rightarrow (x', y')\) to be possible, the product of the interface height differences on the departure site and the arrival site is non-zero: \(h_+(x, t)h_-(x', t) > 0\), i.e. the channel for the jump between the interfaces must be open at both ends of the jump. Second, an attempted jump arriving outside the region bounded by the interfaces is blocked. In the actual dynamics, the direction of an attempted jump is chosen without any prior knowledge of the ability of the particle to perform the jump.

For diffusion on the square lattice there are a few natural choices for the possible particle jumps: \((x, y) \rightarrow (x', y')\).

Dynamics \(m = 1\): the most obvious case are the nearest-neighbor jumps such that the product of the interface height differences on the departure site and the arrival site is non-zero: \(h_+(x, t)h_-(x', t) > 0\), i.e. the channel for the jump between the interfaces must be open at both ends of the jump. Second, an attempted jump arriving outside the region bounded by the interfaces is blocked. In the actual dynamics, the direction of an attempted jump is chosen without any prior knowledge of the ability of the particle to perform the jump.

For diffusion on the square lattice there are a few natural choices for the possible particle jumps: \((x, y) \rightarrow (x', y')\).

Dynamics \(m = 2\): the particle jumps diagonally, i.e. \((x, y) \rightarrow (x, y\pm 1)\) independently, with the attempt rate \(\gamma\). The total attempt rate is then \(\mu = 4\gamma\). This process is expected to be efficient on tilted sections of the interfaces like the rightmost part of the snapshot configuration in figure 1.

Dynamics \(m = 3\): this is a combination of the jumps available in dynamics 1 and 2. In this work we chose \(\alpha = \beta = \gamma\) so that \(\mu = 8\alpha\).

In addition to the three models above, which we shall call two-dimensional particle dynamics, we consider simplified dynamical rules, which will be called one-dimensional.

Dynamics \(m = 4\): in this model only the \(x\) coordinate of the particle matters and the particle is allowed to perform the jump \((x, y) \rightarrow (x', y')\) whenever the channel is open, i.e. \(h_+(x, t)h_-(x', t) > 0\). Then the effect of the interface dynamics on the possibility of the particular jump is fully determined by \(h_+(x, t)\) and diffusion is most directly controlled by the bubbles. In this case, in addition to the jump of the particle in the horizontal direction, there is, when needed, a move in the vertical direction over one lattice unit to keep the particle between the interfaces such that \(h_2(x, y) \leq y \leq h_1(x, t)\). The attempt rate of the jump in this case is denoted by \(\nu\). The effect of forced moves on waiting times of the particle in the continuous-time dynamics is discussed in more detail in section 3.2.

3. Numerical methods

3.1. Interface dynamics

For the dynamics of the interfaces, which is the most time-consuming part of the numerics, we used the so-called \(N\)-fold [21] algorithm in our continuous-time Monte Carlo simulations. In the \(N\)-fold algorithm the possible transitions are divided into \(N\) classes according to their probabilities. After finding those classes, one finds all lattice points \(x, y\) which belong to a certain class \(j\). The next step is to calculate the set of time-dependent variables \(Q = \sum_{j=1}^{N} n_j P_j\), where \(n_j\) is the number of those lattice points which belong to the class \(j\) and \(P_j\) is the probability associated with \(j\). The class \(j\) of the event, which will occur, is next determined by finding \(j\) such that \(Q_{j-1} \leq R < Q_j\), where \(R\) is a random number with uniform distribution in the interval \([0, Q_N]\). After finding the class, one randomly chooses a location \((x, y)\) within this class.

The waiting time for something to happen in the system consisting of the two interfaces then is \(\Delta t = t_1 - t_0\) times: first, the latest instant of real time \(t_0\) after which the particle tries to jump is drawn from the exponential distribution \(\Delta t = -\ln(R_1)/\mu\), where \(R_0\) is a uniformly distributed random number in the interval \([0, 1]\).

In the combined model containing both the interfaces and the particle, the particle does not affect the dynamics of the interfaces, which will evolve as described in the first paragraph above. To include the diffusing particle, we need two waiting times: the waiting time \(\Delta t\) of the interface system and the waiting time \(\Delta t_p\) of the particle. We then keep track of two times: first, the latest instant of real time \(t_0\), when there was a move in the interface system. Second, the latest instant of time \(t_0\), when there was either an attempt of the particle to jump or a forced move of the particle. Then, if \(t_1 + \Delta t_1 < t_0 + \Delta t_p\), the next event in the system will be a move of the interface, otherwise the next event will be an attempt to move the particle.

After this, there still are in the continuous-time dynamics two obvious ways to handle the forced moves (in dynamics \(m = 1, 2, 3\)): (A) the clock of the particle remains intact in a forced move or (B) its waiting time \(\Delta t_p\) for the particle is updated after it. We shall consider both choices since they produce quite different results and both can be physically justifiable from some microscopy. With this choice the dynamics of the combined model becomes defined.
3.3. Sampling of main quantities

The sampling of the observables was done with a constant time interval after reaching the steady state from an initial configuration consisting of two completely disordered interfaces at a fixed distance from each other. With the N-fold method, reaching the steady state for interface configurations turns out not to be very difficult but especially for sampling of diffusion quite long runs were required.

Typically of the order of $10^3$ independent runs were performed for the interfaces in such a way that there were $10^2$ particles diffusing (independently of each other) between the interfaces, the linear size of the simulation cell being $L = 100$ (for the finite size scaling studies mentioned in the text larger systems were used). The reason for this procedure is that the dynamics of the interfaces even with the N-fold algorithm is computationally the most time-consuming part of the simulation, so we used each sequence of interface configurations to produce many particle trajectories, the total statistics thus being of the order of $10^5$.

To characterize the statistical properties of the interfaces $h_1$ and $h_2$, we use their roughness or width [20] defined as

$$W(f) = \sqrt{\langle |h_i(x, t) - \bar{h}_i(t)|^2 \rangle}.$$  \hspace{1cm} (5)

Here $i = 1, 2$ and $\bar{h}_i(t)$ is the spatially averaged height of the interface configuration at time $t$, and the angle brackets denote ensemble average, i.e., average over independent simulations. The kink density $\bar{k}(f)$ is the density of those locations $x$, for which $h_i(x - 1, t) \neq h_i(x + 1, t)$ for each of the interfaces $i = 1, 2$ separately. This definition follows from the fact that a BCSOS interface even in the flat state has an intrinsic roughness, because $|h_i(x, t) - h_i(x \pm 1, t)| = 1$. In the notations for these quantities we suppress the dependence on system size $L$.

The bubble size distribution per site was sampled at the same instant of time as the width of the interfaces. It gives the probability that a randomly chosen location $x$ belongs to a bubble of size $\bar{\ell}$, which is the length of the bubble in the $x$ direction. We shall be interested in bubble size distributions $P_f(\ell, f)$, normalized so that only bubbles with $j \leq \ell < L$ are taken into account, $\sum_{\ell=j}^{L-1} P_f(\ell, f) = 1$, so that they are related by

$$P_f(\ell, f) = P_0(\ell, f) \left[ 1 - \sum_{\ell=0}^{j-1} P_0(\ell, f) \right]^{-1}.$$  \hspace{1cm} (6)

In this $\ell = 0$ means that $h_i(x, t) = 0$ for site $x$. In the configurations where the interfaces did not touch each other at all, the bubble size was recorded as a count in bin $\ell = L$. For certain purposes we also sample the bubble size distributions normalized for $j \leq \ell \leq L$, which we denote by $P_f^*(\ell, f)$, so that $\sum_{\ell=j}^{L} P_f^*(\ell, f) = 1$. In addition, we compute the (physical) average bubble size $\bar{\ell}(f)$ and also $\bar{\ell}^*(f)$ defined as

$$\bar{\ell}(f) = \sum_{\ell=0}^{L-1} \ell P_0(\ell, f) \quad \bar{\ell}^*(f) = \sum_{\ell=0}^{L-1} \ell P_0^*(\ell, f).$$  \hspace{1cm} (7)

The mean waiting time for the change of the bubble size we shall denote by $\bar{\tau}(\ell, f)$.

The diffusion coefficients, denoted here by $D_{obs}$, were determined in the long-time regime $\langle (\Delta x(t))^2 \rangle > \bar{\ell}$, as a slope of the mean square displacement via [22]

$$\langle (\Delta x(t))^2 \rangle = 2D_{obs} t.$$  \hspace{1cm} (8)

where $\Delta x(t) = x_f(t_0 + t) - x_f(t_0)$ is the particle displacement during a time interval of length $t$. In the simulations, to reach the hydrodynamic regime not only for interfaces but for diffusion as well, we run the dynamics long enough so that the square root of $\langle (\Delta x(t))^2 \rangle$ is much larger than the bubble size. Particle diffusion in the $y$ direction, on the other hand, is in the long-time limit simply controlled by interface wandering and the effective size-dependent diffusion coefficient related to that.

3.4. Overview of the parameters and their meaning

Our full model for diffusion restricted by fluctuating interfaces thus contains the following variable parameters:

(i) The driving parameter $f$, defined in equation (3) of section 2.1, drives the interfaces towards each other. For interfaces to be coupled in the steady state we must have $f > 0$. For increasing $f$, the channel for diffusion becomes narrower and the lengths of the locally open paths, i.e. the bubble sizes, get smaller and the timescale of fluctuations gets longer. For $f \leq 0$ the interfaces would be driven apart from each other and diffusion between them would become an unrestricted random walk.

(ii) The second parameter is the generic jump attempt frequency $\mu$ of the diffusing particle. The possible jump directions (horizontal, vertical or diagonal, see also figure 1) are controlled by the parameter $m = 1, 2, 3, 4$ described in section 2.2. The simplest choice is the one-dimensional jumps in the model with $m = 4$, which is a good starting point for analytical arguments for the scaling of diffusion with the model parameters.

(iii) The particle jumps are restricted by the dynamic environment provided by the interfaces. The technical details of this are given in section 2.2, but there are no additional parameters involved. However, there remain two physically reasonable ways to realize the occasions, when the interfaces would possibly move the particle. This is described in section 3.2: in scheme (A) the particle is considered in such cases to move together with an interface with its diffusive ‘clock’ left intact and in (B) its ‘clock’ is updated after the interface move. The physical meaning of these choices is considered in section 6.

4. Results for interface dynamics

In this section we first complement the study of [18] to characterize the time evolution of the coupled interfaces themselves (without a diffusing particle) in the stationary state. In the observed dynamics, a few timescales of interest can be monitored to gain insight also into the behavior of
diffusion between the interfaces to be discussed in section 5. The first timescale is the average time \( T_{\text{int}} \) elapsed between consecutive changes in the interface configuration. The second one, denoted by \( T_{\text{lat}} \), is the average timescale over which a single location in the \( x \) direction stays within a bubble \( \ell > 0 \) in the \( x \) direction. Also the third one describes the behavior of the interface system only: it is the average waiting time \( T_{\text{bub}}^{(j)} \) for a change in the bubble size:

\[
T_{\text{bub}}^{(j)}(f) = \sum_{\ell=j}^{L-1} P_{\ell}(\ell, f) \bar{T}(\ell, f),
\]

where \( P_{\ell}(\ell, f) \) is the normalized bubble size distribution and \( \bar{T}(\ell, f) \) is the corresponding average waiting time for something to happen for a bubble of size \( \ell \), see section 3.3. To obtain more detailed information, we computed \( T_{\text{bub}}^{(j)} \) for \( j = 0, 1, 2 \) because the bubbles of size \( \ell = 0, 1, 2 \) control diffusion for large \( f \).

In figure 2 we present the timescales \( T_{\text{int}}, T_{\text{lat}} \) and \( T_{\text{bub}}^{(j)} \) describing the interface dynamics. The time \( T_{\text{int}} \) is determined by the interface configuration and its behavior is not monotonic as can be seen from figure 3, where a shallow minimum is observed. On the other hand, the roughness \( W \) as a function of \( f \), shown in the same figure, displays a dip at \( f_w \approx 0.15 \). The non-monotonic behavior is explained by the entropic effects through the reduced configuration space available for the interfaces [18]. The finite size scaling of the dip position, \( f_w(L) \), was studied in [18]: we only mention here the result \( f_w \sim L^{-1/3} \) for large \( L \). In passing we note that such a deroughening due to interactions between interfaces has been experimentally observed in another context, see the articles in [19]. It is evident from figure 2 that the point \( f = f_w \) is also the crossing point for the dynamical properties of the interfaces. For \( f > f_w \), \( T_{\text{int}} \) increases rapidly and displays the asymptotic behavior \( T_{\text{int}} \sim f \) for large \( f \), because the rate-limiting factor is the time required to create new bubbles, which is proportional to \( 1/q_2 \sim (1 - q_2)/q_2 - 1 \equiv f \) for \( q_2 \ll 0 \) in this limit.

In figure 2 we also observe that \( T_{\text{int}} \sim 1/f \) for \( f \ll f_w \). This is controlled by the timescale of (finite) interfaces wandering apart from each other and then back together (effectively biased diffusion), which is proportional to \( 1/(p_2 - q_2) = 1/(2p_2 - 1) \sim 1/f \). For large \( f \) we have \( T_{\text{int}} \sim 1/2 \) because the smallest bubbles \( \ell = 1 \) in this limit disappear with the rate \( 2q_1 \rightarrow 2 \). For increasing \( f \), the timescale \( T_{\text{bub}}^{(0)} \) increases without limit with the waiting time for a bubble to appear as \( T_{\text{bub}}^{(0)} \sim f \), but \( T_{\text{bub}}^{(1)} \rightarrow 1/2 \) by the same argument as for \( T_{\text{int}} \). Also the behavior of \( T_{\text{bub}}^{(2)} \) has the same asymptotics, as can easily be seen by inspecting the one and only possible two-step process, where a bubble of size \( \ell = 2 \), the rate-limiting factor being the shrinking of the bubble from either end.

The last timescale shown in figure 2 is the average waiting time for a change involving a bubble of size \( \ell \geq 2 \) to occur in a given lattice site, which we denote by \( T_{\text{mob}} \), since it is related to configuration changes that change the effective mobility of a particle by increasing or decreasing its possible range of motion. This differs from \( T_{\text{bub}}^{(2)} \) in that, for example, for a particle sitting at site \( x \), where the sites belong to a bubble with \( \ell \leq 1 \), i.e., for a particle stuck in a locally closed configuration, in averaging \( T_{\text{mob}} \) we count the time for the particle to be mobile again, i.e., for the site to be in a bubble with \( \ell \geq 2 \) again. For a bubble with \( \ell > 2 \), on the other hand, each change of its size will result in a greater or smaller effective mobility of the diffusing particle. For \( f \gg f_w \) we have \( T_{\text{mob}} \sim f^2 \), since the rate-limiting process contributing to it in this limit is a two-step process, where a bubble of size two becomes created starting from a configuration where there are no bubbles.

To characterize the properties of the interface configurations in more detail, we present in figure 4 the mean bubble size \( \bar{\ell} \) and the kink density \( \bar{k} \). The non-monotonicity of the differently normalized (see the caption) mean bubble size \( \bar{\ell} \) is a consequence of the fact that for finite \( L \) the maximum size of a bubble is limited. Below the roughness dip, for \( f < f_w \), the probability of completely open bubbles \( (\ell \geq L) \), neglected in the computation of \( \bar{\ell} \), rapidly increases for decreasing \( f \), while the number of such bubbles is essentially zero for \( f > f_w \). In the vicinity of the dip, for \( f \approx f_w \), we observe \( \bar{\ell} \sim f^{-4/3} \).
5. Results for particle diffusion

For a continuous-time unbiased random walk the one-dimensional mean square displacement is of the form [22]

\[ \langle (\Delta x(t))^2 \rangle = \langle N(t) \rangle \sigma_{2x}^2, \tag{10} \]

where \( \langle N(t) \rangle \) denotes the average number of jumps in time \( t \) and \( \sigma_{2x} \) is the variance of the displacement of individual jumps. In what follows we shall use equation (10) to justify a theoretical model for diffusion between the interfaces for \( f > f_w \). For the different models of particle dynamics in the presence of the interfaces, i.e. the models \( m = 1, \ldots, 4 \) defined in section 2.2, we shall report our results as a function of the drive parameter \( f \) and the total jump rate \( \mu \) as the dimensionless ratios

\[ D_m(f, \mu) = \frac{D_{m1}(f, \mu)}{D_{free}(\mu)}, \tag{11} \]

where \( D_{m1}(f, \mu) \) is the observed diffusion coefficient according to equation (8) and \( D_{free}(\mu) \) is that corresponding to the same intrinsic jump rates without the interfaces. These ratios then give the effect of the interfaces on diffusion, while \( \mu \) is the total attempt rate of jumps for the given model in units of the total attempt rate of local interface configuration changes. For a free random walk with \( \sigma_{2x}^2 = 1 \), combining equations (17) and (10) gives the diffusion coefficient for free motion in the \( x \) direction, for example, as \( D_{free}^{(1)} = \alpha = \mu/4 \) and \( D_{free}^{(4)} = \nu = \mu/2 \). We shall begin our discussion with the simplified dynamics \( m = 4 \), because for it the generic features of diffusion between the interfaces become more transparent.

### 5.1. Diffusion for the one-dimensional particle dynamics (model 4)

In the case of slow particles the interface configuration will change many times between the jump attempts of the particle. In this case, the success ratio of the jump attempts can be evaluated from the bubble size distribution \( P_0(\ell, f) \) and it is

\[ g(f) = \sum_{\ell=2}^{\ell=L-1} P_0(\ell, f) \frac{\ell - 1}{\ell} + P_0(L, f), \tag{12} \]

because only within bubbles with \( \ell \geq 2 \) can the particles move and with probability \( (\ell - 1)/\ell \) is an attempted jump possible since the particle cannot jump out of the bubble so that two of the \( 2\ell \) attempts are blocked by the bubble edges. The mean-field prediction for the diffusion coefficient is then

\[ D_{mf}(f, \mu) = g(f) D_{free}(\mu). \tag{13} \]

The simulation results for the model \( m = 4 \) together with this mean-field approximation are presented in figure 5. For \( \mu \ll 1/T_{int} \), i.e. for slow diffusion, the curves \( D_s(f, \mu) \) follow the mean-field prediction of equation (13).

In the inset of figure 5 we show a data collapse by using for \( f \) the scaling factor

\[ \sigma(\mu) = \sqrt{1 + c\mu} \tag{14} \]

with \( c = 1 \). This is an interpolation of the large \( \mu \) behavior \( \sigma(\mu) \sim \mu^{1/2} \) for \( \mu \gg 1 \), with the interface and particle timescales well separated, and the small \( \mu \) behavior \( \sigma(\mu) \approx 1 \) for \( \mu \ll 1 \), which is exactly the mean-field result discussed.
above. With this scaling, our simulation data for \( D_{ad}(f, \mu) \) for \( f < f_w \) is nicely collapsed onto the slow-particle curve defined by equation (13) and even better with the choice \( c \approx 1.1 \) (not shown), which we assign to finite size effects. However, to obtain a good data collapse for \( f > f_w \) we need a different scaling combination:

\[
D_{obs}^{(m)} \to D_{obs}^{(m)}/[1 - \exp(-\mu r_2)],
\]

corresponding to an exponential clock with \( r_2 = T_{mob}(f \to \infty) = 1/2 \). This scaling form results from the characteristic timescale of the smallest bubbles allowing diffusion of particles in this regime. This is controlled by the driving parameter \( f \) such that for \( f \gg 1 \) there are mainly bubbles of size \( \ell = 1 \), but for diffusive jumps to take place bubbles of size \( \ell = 2 \) are needed (with the time units chosen, once created, they stay open at least over one unit of time, cf section 4). This scaling is demonstrated in the log–log plot of figure 6. Because of the different rate-limiting mechanisms these scaling forms, below and above the dip region, are incompatible.

Next we consider the case of fast particles. If the particle jump rate is fast compared to the interface dynamics, an adiabatic approximation can be done as follows. The particle is then trapped inside a bubble and its location becomes uniformly distributed in the timescale of the bubble dynamics and the (effective) location of the particle can change only when the size of the bubble changes. The length of an effective particle jump via this mechanism is approximately the length of the displacement of the center-of-mass location \( b \) of the bubble. By applying equations (8) and (10) we then obtain

\[
D_{adib}(\mu, f) = \frac{1}{2} \frac{1}{T_{mob}} B^2(f),
\]

where \( T_{mob} \) is the timescale of bubble motion and \( B^2(f) \) is the mean square displacement (per jump) of the bubble obtained from

\[
B^q(f) = (|b_{new} - b_{old}|^q),
\]

with \( q = 2 \). Here \( b_{new} \) and \( b_{old} \) are the locations of a bubble before and after the change in the bubble size. This approximation is shown by the dashed line in figure 6. For large \( f \) the expected behavior \( D \sim f^{-2} \) is recovered, since the bubble jump length and thus \( B^2 \) becomes a constant for isolated bubbles and \( T_{mob} \sim f^2 \). For \( f > f_w \), the bubble motion is dominated by increasing or decreasing the bubble size by one lattice unit and, as expected, for fast particles the adiabatic approximation works well. For \( f < f_w \), merging and dissociation of bubbles becomes important. We also tested the choice \( q = 1 \), with the mean jump length squared, giving less weight to long jumps and thus giving a smaller diffusion coefficient for small \( f \) (for large \( f \) the smallest jumps dominate in any case), but the approximation is not essentially better.

The dotted curve just below the adiabatic approximation shows an approximation obtained by assuming an infinite jump rate of the particle such that, after each interface configuration change, a new location for the particle is drawn evenly distributed inside a bubble, but such that the particle displacement in the case of a completely open interface configuration is restricted by the system size. A possible way to extend this approximation for smaller \( f \) would be to utilize the known form of the bubble size distribution for large \( \ell \). This way we obtain a reduction of the diffusion coefficient that seems to work around \( f = f_w \) but apparently fails for small \( f \), cf the behavior of \( \ell^2 \) in figure 4. Another natural approximation would be to consider the size of the bubble as defining an effective mobility of the diffusing particle. Qualitatively, the behavior of \( B^2 \) and the mean squared bubble size \( \ell^2 \) are quite similar, but with incompatible limits for small and large \( f \). Unlike in equation (16) for \( B^2 \), it turns out to be difficult to assign a natural rate factor to \( \ell^2 \) to develop a reasonable approximation for the diffusion coefficient.

5.2. Diffusion for the two-dimensional particle dynamics (models 1, 2 and 3)

We next consider the effect of the ‘microscopic’ dynamics on diffusion. First, in figure 7 we show the diffusion coefficient for dynamics \( m = 1 \) with the pushes of the particle by the interfaces handled according to clock updating scheme (A) and in figure 8 according to scheme (B), see section 2.2 for details. Now it is not just the length of the bubble but also the shape of it that affects diffusion. In (A) the particle waiting time is updated after each jump attempt, while in (B) it is updated also after each push by the interfaces. The difference between (A) and (B) appears for long particle waiting times (for small \( \mu \)) in the regime where there are frequent pushes, i.e. the drive \( f \) needs to be strong enough, but not so strong that diffusion would be completely dominated by the motion of small bubbles as discussed above. The coincidence of the waiting times of the interface motion and the particle jump attempts produces at a finite value of \( f \) in (B) behavior that
looks like the suppression of diffusion sometimes observed close to phase transitions. The dip in \( D_1 \) appears close to the dip in the kink density \( \kappa \) (see the inset of figure 4), where in the local interface configurations there are more sites with possible interface configuration changes (at a kink site the interface is locally frozen) and thus more pushes. Other variants of the jump rate modification are conceivable, e.g. physically it might happen that the contact with the interfaces could boost the particle jump rates, but in the region of the parameter space, where bubble dynamics is the rate-limiting factor, it would not essentially change diffusion from what is observed in scheme (A).

The corresponding mean-field approximation of equation (13) shown in figure 7 by the full curve is presented to help the comparison between \( m = 1 \) and 4. As is evident from the comparison of figure 7 and figure 5, scaling by the factor \( \sigma(\mu) \) of equation (14) would not yield a very good data collapse for \( f \ll f_w \) in the case of two-dimensional particle dynamics. For particles fast both in the horizontal and vertical directions, the blocking of diffusion by ‘collisions’ with the interfaces is quite efficient and leads to a considerable reduction of \( D_1 \) in this regime. Note that even for small \( f \) we have \( D_1 < 1 \) for fast particles, since particle diffusion in the vertical direction is faster than that of the interfaces for any \( f > 0 \). This in part explains the spreading of the curves \( D_1(f) \) for \( f < f_w \). In the inset of figure 7 we show also the difference between the particle dynamics \( m = 1 \) and 2 with the clock updating scheme A (concerning dynamics \( m = 3 \), we find that \( D_1 < D_1 < D_2 \)). For a reasonable comparison, the jump rates for each dynamics have been chosen such that the total jump rates in the \( x \) direction match; thus we plot \( D_2(\mu/2) / D_1(\mu) \). Due to the limited accuracy of \( D_m \), the ratio \( D_2/D_1 \) becomes somewhat noisy, but a few general observations can be made. A difference first develops for \( f > f_w \), because diffusion becomes more restricted by the interfaces within bubbles of the type seen on the right (\( x = 17, \ldots, 20 \)) in the snapshot of figure 1 becoming prevalent. In such interface configurations, many of the ‘horizontal’ jumps (for \( m = 1 \)) become blocked and diffusion along the narrow channel requires also ‘vertical’ jumps, while the diagonal jumps (for \( m = 2 \)) are more effective for particle transport. However, for \( f \ll f_w \) diffusion again becomes dominated by bubble motion the way it was for \( m = 4 \), and \( D_m \sim f_w^{-2} \) for \( m = 1, 2, 3 \) for both particle clock updating schemes (A) and (B) as seen in figure 8.

6. Discussion

To summarize, we have considered one-and two-dimensional continuous-time random walkers constrained between two evolving interfaces symmetrically driven towards each other. A surprisingly complicated phenomenology appears. First, in the interface model itself there is a dip in the interface roughness at a finite value \( f_w \) of the parameter \( f \) describing the drive of the interfaces against each other [18], with different kinds of interface and bubble dynamics below and above the dip. The crossover from bubble-dominated (interface-dominated) to almost free diffusion is controlled by the relative jump rate of the particle and its interplay with the rate of the interface time evolution. In the analysis of this crossover, simple scaling arguments for the two regimes are incompatible, both physically and formally, so that the full behavior of diffusion cannot be described by a single scaling form. This can be expected to be a generic property of transport restricted by fluctuating interfaces.

Furthermore, diffusion was found to depend on the microscopic details of the interaction between the interfaces and the diffusing particles. In particular, its immediate effect on the waiting time of particle jumps was shown to be considerable, especially for dynamics physically corresponding to diffusion on a lattice in the large-friction limit, which can be realized at domain boundaries in adsorption systems. In a spatial continuum this effect would be absent, as would be the roughness dip. However, the finite size case, with the underlying microscopic structure not washed out in coarse graining, can be of interest in its own right in nanoscale
applications. Then also the size of the channel for the particles will induce a relevant length scale, in addition to the length scale related to the diffusive jumps (lattice) and the one related to the environment (bubbles).

In this work we have considered diffusion that is effectively one-dimensional even if the diffusive jumps and interactions between the tracer particles and the interfaces result from two-dimensional dynamics. The environment for the diffusing particles is then described by a chain of bubbles, whereas in higher dimensions more complicated topologies (networks) would arise, in some cases leading to a percolation problem. In such studies, like in the present one, a considerable problem is the wide gap between the ‘microscopic’ timescales related to the dynamics of the particles and the environment, and the ‘hydrodynamic’ timescale corresponding to diffusion over length scales larger than any structures in the environment experienced by the particles. We hope our work would inspire further theoretical and experimental studies of diffusion in evolving and constraining environments.

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