Crowding competes with trapping to enhance interfacial diffusion

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Received: 10 February 2022 / Accepted: 1 March 2022
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Abstract Diffusion of species on biological membranes or materials interfaces is expected to slow down with an increase in their density, but also due to their intermittent binding to functional moieties or surface-defects. These processes, known as crowding and trapping, respectively, occur simultaneously in a broad range of interfacial systems. However their combined effect on the diffusion coefficients was not studied hitherto. Here, we analytically calculate and numerically validate by Monte Carlo simulations an expression for the diffusion coefficient of a two-dimensional lattice gas in a field of immobilized traps. As expected, trapping and crowding both suppress transport but, surprisingly, the diffusion coefficient is non-monotonous. Namely, increasing gas densities increases trap occupancy while crowding is not overpowering, such that the diffusion reaches a maximum. These results should be relevant to interfacial growth phenomena, as discussed in the context of nascent adhesions in cells.

1 Introduction

Diffusive transport at interfaces is an ubiquitous process in nature. Prototypical examples involve proteins on a phospholipid membrane of a living cell [1], nanoparticles in liquid-filled solid pores [2], or molecules interacting with growing or functional materials interfaces [3]. The characteristic of this transport is that it takes place on a two-dimensional surface, which by its structure and molecular composition may be very complex. Often transport takes place at an appreciable concentration of diffusing species which furthermore interact with defects on the surface or specifically incorporated functional moieties such as proteins on the membranes [4]. These local interactions trap the diffusing species, intermittently arresting the particle, which in turn reflects on the effective diffusion coefficient.

Diffusion on interfaces has been theoretically studied as a simplified problem of a stochastic motion on a lattice [5,6]. In these models, steric hindrance between diffusing particles is typically introduced, which prevents double occupancy of the same lattice site. For a gas of diffusing particles in these conditions, long-time diffusion coefficient has been determined by calculating many-particle correlation functions [7–9], as well as by considering memory effects of diffusing particles [10]. These different approximations were recently complemented with the exact calculation of the tracer particles’ probability distribution function on a crowded lattice [11], yielding the diffusion coefficient as a function of gas density or surface coverage.

While the increased density of diffusing species rescales the diffusion constant at long times [12], interactions with defects, however, yield a rich dynamic behavior [13,14], which was extensively studied using theoretical [15–17] and computational means [18–20]. Particular efforts were focused on understanding the effect of interaction types on the diffusion of a tracer particle, which lead to the random trap model [21], Havlin–Weiss comb model [22], or the quenched trap model [23].

Computational approaches provided insight on the effects of surface heterogeneity using the bivariate trap model [24,25]. Here, the usage of lattice gas became established as they were shown to be a computationally efficient for coarse-graining and accurate in the limit of large systems [26]. Hence, kinetic Monte Carlo simulations were used to study the diffusion of a tracer on a surface with binding sites characterized by multiple energy levels [27]. It has been established that the tracer diffusion in the field of traps exhibits a sub-diffusive motion on short time scales and normal diffusion on long time scales [1,27–29]. These works provided significant impact on the role of the trap energetics on the diffusivity of a tracer [21], however, the combined effects of crowding and trapping were not addressed so far in full depth to our knowledge.

In this work, we aim to rectify this situation by providing an approximate theory for surface diffusion of crowded particles in a field of simple traps. We propose an expression for the long-time effective diffusion coefficient of the crowding particles using the scaling argument as well as the master equation approach. The model is validated by a favorable comparison with Monte Carlo (MC) simulations [30] (methodological details in Supplementary Information). In agreement with previous works [1,27–29], we find that an intermittent sub-diffusive regime is inherited to the system as soon as the traps are introduced. More surprisingly, however, we...
find that in the long time limit the diffusion coefficient may be enhanced by intermediate crowding effects for any concentration of traps.

2 The model and the scaling arguments

We start with placing \( N_w \) particles, mutually interacting with a hard wall repulsive potential, on a 2D square lattice with \( N_g \) lattice sites of a length \( a_0 \). The concentration of gas particles on the lattice is \( c_w = N_w / N_g \). A random walking particle will traverse a distance \( a_0 \) during time \( \tau = a_0^2 / 4D_0 \), where \( D_0 \) is the diffusion coefficient of a particle at infinite dilution. The concentration-dependent diffusion coefficient \( D_{cr} \) (red symbols in Fig. 2) is determined as \( D_{cr}(c_w, D_0) = D_0p(c_w) \) where \( p(c_w) \) is the concentration-dependent probability of a jump, whereby the larger the concentration, the smaller the effective diffusion coefficient. It can also be seen as the factor normalizing the characteristic time to make a step in the crowded environment \( \tau_{cr} = \tau / p \). It has been calculated using different approaches, [7–9], while here we estimate it from backward correlations as in the anti-persistent random walk (APRW) model [10]. The appropriateness of this choice, which is a compromise between accuracy over the entire density range and simplicity, as demonstrated by comparison with MC simulations and with other models (see section I and Fig. 1 in Supplementary Information).

The lattice is furthermore decorated by \( N_t \) randomly placed traps, the concentration of which is denoted as \( c_t = N_t / N_g \). Despite the fact that traps and particles are on the same lattice, there is no exclusion between particles and traps, and a lattice site can be filled by both to emulate the interaction of the particle and a defect. Upon hopping onto a site with a trap, a diffusing particle binds with the probability \( P_{on} \) (Fig. 1), and unbinds with the probability \( P_{off} \). Following the detailed balance condition, the binding energy is \( \Delta E_b = -k_BT\ln(p_{on}/p_{off}) \). The latter sets the concentration of bound particles \( \langle c_b \rangle \), which can be calculated analytically from the partition function of the system [31], and its exact form is given in the SI.

To estimate the diffusion constant in the long time limit for the lattice gas in the field of traps \( D_{eff} \), we aim at coarsening the diffusion process and finding the scaling function \( f \)

\[
D_{eff}(c_w, c_t, P_{on}, P_{off}) = f(c_w, c_t, P_{on}, P_{off})D_{cr}(c_w).
\]  

(1)

Specifically, we presume that the effective diffusion coefficient can be related to the square of the average distance between two traps at which binding actually occurs \( \langle l^2 \rangle_{eff} \) and the time it takes to make this coarsened diffusion step \( \tau_{eff} \), i.e. \( 4D_{eff} = \langle l^2 \rangle_{eff} / \tau_{eff} \). Here, the average square distance between two efficient traps can be related to the effective concentration of traps \( \rho_{eff} \) as \( \langle l^2 \rangle_{eff} = a_0^2 / \rho_{eff} \). The latter can be estimated from the density of free traps \( c_t - \langle c_b \rangle \), to which a diffusing particle can bind with the probability \( P_{on} \) such that \( \rho_{eff} = (c_t - \langle c_b \rangle)P_{on} \).

The characteristic time \( \tau_{eff} \) comprises of the time that it takes to leave a bounded trap \( \tau_{off} \), and the time to diffuse to the next trap where it binds again \( \tau_{t-t} \), which is simply \( \tau_{t-t} = \langle l^2 \rangle_{eff} / 4D_{cr} = \tau_{cr} / \rho_{eff} \). To leave the trap, a particle must unbind with the probability \( P_{off} \) and make a step with the probability \( p(c_w) \), and hence, \( \tau_{off} = \tau_{cr} / [P_{off}p(c_w)] \). With \( \tau_{eff} = \tau_{off} + \tau_{t-t} \), it is straightforward to estimate the effective diffusion coefficient as

\[
D_{eff} = D_{cr}\left(1 + \frac{P_{on}}{P_{off}}\frac{c_t - \langle c_b \rangle}{p(c_w)}\right)^{-1}
\]  

(2)

3 The master equation approach

The same result can be derived by considering the master equation approach (see SI for details of the calculation). We focus on the probability of finding a particle \( P(r, t) \), at position \( r \) at time \( t \). Since the later emerges as a sum of the probability of finding a particle unbound on that site \( P_{cr}(r, t) \) and the probability of finding the particle trapped on the same site \( P_{tr}(r, t) \), we can write

\[
P(r, t) = P_{cr}(r, t) + P_{tr}(r, t).
\]  

(3)
The time evolution of both probabilities is given by:

\[
\frac{\partial P_{\text{ct}}(r, t)}{\partial t} = \frac{\gamma}{2d} \sum_{i=1}^{d} (P_{\text{ct}}(r - e_i, t) + P_{\text{ct}}(r + e_i, t) - 2P_{\text{ct}}(r, t)) - \kappa_{\text{eff}} P_{\text{ct}}(r, t) + \kappa_{\text{off}} P_u(r, t),
\]

and

\[
\frac{\partial P_u(r, t)}{\partial t} = \kappa_{\text{on}} P_{\text{ct}}(r, t) - \kappa_{\text{off}} P_u(r, t).
\]

Here, \(\gamma = p(c_u) / \tau\) is the hopping rate of the particle and \(\kappa_{\text{on}}, \kappa_{\text{off}}\) are the effective binding and unbinding rates of any particle. By introducing Fourier transform

\[
S(k, w) = \frac{1}{\pi} \int_{-\infty}^{\infty} dt P(r, t) e^{-i(wr - k\cdot r)}
\]

of Eqs. (4) and (5) we can calculate the relation between dynamic structure factors \(S_{\text{ct}}(k, w)\) and \(S_u(k, w)\). By calculating real part of total Fourier transform \(\text{Re}(S(k, w))\) and using the relation

\[
D_{\text{eff}} = \lim_{w \to 0} \left( \lim_{k \to 0} \pi w^2 / k^2 \text{Re}(S(k, w)) \right)
\]

we obtain

\[
D_{\text{eff}} = D_{\text{ct}} \frac{\kappa_{\text{eff}}}{\kappa_{\text{on}} + \kappa_{\text{off}}}
\]

We note that for \(\kappa_{\text{eff}} = 0\) the effective diffusion constant \(D_{\text{eff}}\) reduces to \(D_{\text{ct}}\).

To understand what are the effective rates, we start from the realisation that a particle can only bind to a trap if the trap is empty. Hence, the effective binding rate is

\[
\kappa_{\text{on}} = P_u(c_t - (c_b))/\tau.
\]

Similarly, effective unbinding rate of a bounded particle depends on the availability of a free site to jump, so

\[
\kappa_{\text{off}} = P_{\text{off}} p(c_u)/\tau.
\]

Substituting these expression for effective rates in Eq. (1) yields \(D_{\text{eff}}\) equal to that in Eq. (2).

4 Results and comparison with simulations

Intuitively, one expects a monotonic decay of the effective diffusion coefficient with increasing the concentration of particles \(c_u\), which indeed is the result observed in the absence of traps \((c_t = 0\) red line and points in Fig. 2)). However, as soon as traps are introduced \((c_t > 0)\), the effective diffusion coefficient starts to non-monotonically vary with the gas density and a maximum in \(D_{\text{eff}}(c_u)\) is predicted by Eq. (2). This surprising property of lattice gas diffusion in the field of traps is confirmed by MC simulations (symbols in Fig. 2a). Quantitative agreement between theory and simulations is obtained at low \(c_u < 0.3\) (irrespective of \(c_t\)) and \(c_t < 0.3\) irrespective of \(c_u\) (see Fig. 2c). At large densities \(c_u > 0.6\) and \(c_t > 0.5\), the agreement is qualitative (deviations larger than 50, see white area in Fig. 2c), which suggest that higher-order correlations play an important role in this range of parameters.

In order to understand the mechanism underlying the non-monotonic behaviour of \(D_{\text{eff}}\), we extract the probability distribution of the actual path length traversed by walkers between two successive trapping events \(T^\text{MC}_d\) (Fig. 3). We also sample the distribution of time between two trapping events \(T^\text{MC}_d\) (inset of Fig. 3). We clearly see that at low concentrations of \(c_u\), both typical lengths of the path, and the actual time between two trapping events is in average short, which means that the particles spend most time in the traps, this being detrimental to the effective diffusion coefficient. We denote this regime as “trapping-dominated diffusion”.

The importance of trapping in this regime is confirmed by studying the diffusion of transparent particles, for which the hard core repulsion is omitted and more than one can be on the same lattice site. The normalized effective diffusion for these type of traps, this being detrimental to the effective diffusion coefficient. We denote this regime as “trapping-dominated diffusion”.

For moderate \(c_u\), the characteristic \(\tau^\text{MC}_d\) becomes significant while the tails of the distribution of \(T^\text{MC}_d\) are the thickest as seen by inspection of Fig. 3). This means that at these concentrations the particles spend extended time meandering through the system. In this regime, a high level of trap occupancy is achieved by the significant concentration of walkers, but the crowding effects are not sufficient to prevent the diffusion - i.e. the walkers move away from occupied traps before they interact with a free trap. This yields “crowding enhanced diffusion” - a regime in which increasing the gas density increases the diffusion coefficient.
Fig. 2 Diffusion coefficient as a function of trap density. Comparison of analytic modeling and simulations. a Effective diffusion coefficient, normalized by the diffusion coefficient at infinite dilution, as a function of the lattice gas concentration \( c_w \) for various trap densities \( c_t \) (\( c_t = 0, 0.1, 0.2, 0.3 \) and 0.4) with \( P_{\text{on}} = 0.5 \) and \( P_{\text{off}} = 0.001 \) shown in log-linear scale. Results of MC simulations are shown with symbols, and theory (Eq. 2) with solid lines. b Comparison of \( D_{\text{eff}} \) between hard core particles (symbols) and transparent particles (dashed line) from MC simulation. c The deviation of effective diffusion coefficient between our theory and simulation. The parameters we used in our calculations are mostly obtained from the ligand-receptor protein diffusion system on biological membranes (see SI for details). Diffusion coefficient \( D_0 \) is \( 1 \text{nm}^2/\text{s} \) [32]. The affinity for the trap is \( E_b = 6K_B T \), which is a typical binding energy of a ligand-receptor pair during membrane adhesion [33–35].

Fig. 3 The probability distribution of the path length of a particle between two successive trapping events \( (l_{\text{MC}}) \) is shown for \( c_w = 0.1, 0.3 \) and 0.9 for fixed \( c_t = 0.1, P_{\text{on}} = 0.5 \) and \( P_{\text{off}} = 0.001 \). In the inset, corresponding distribution of time between two successive trapping events of a particle \( (\tau_{\text{MC}}) \) is shown. Corresponding \( (l_{\text{MC}}) \) and \( (\tau_{\text{MC}}) \) is presented in the inset. The data is obtained from kinetic Monte Carlo simulations as presented in lattice units.

At high \( c_w \), nearly all traps are occupied, hence the walkers survive the longest between two trapping events, but they make significantly shorter paths than in the intermediate regime. This is because the likelihood for making a step onto a next site decreases significantly due to the high concentration of the particles, and therefore, there is enough time to actually interact with the trap. In this regime, particle-particle correlations between second order become important. We denote this regime as “the crowding dominated diffusion”.

The three regimes are clearly denoted in the diffusivity phase diagram (Fig. 4) which highlights the importance of the mean number of occupied traps \( \langle c_b \rangle \). For a given \( c_t \), the latter increases with increasing \( c_w \) until saturation, which in the case of reasonable large binding affinities presumes that either nearly all walkers are bound, or that nearly all traps are occupied, depending on their relative total number. In both cases, the particles are still diffusing and binding-unbinding kinetics are still ongoing. The boundaries between the three regimes are determined from the inflection points in \( D_{\text{eff}} / D_0 \) vs \( c_w \) for a given \( c_t \) (red dashed lines in Fig. 4), with \( D_{\text{eff}} \) maximized for the particular concentration of particles \( c_w \) as determined analytically (black dotted line) and fully supported by simulations (black symbols).

5 Discussion and conclusions

In summary, we discussed the simultaneous effect of crowding and trapping on surface diffusion. Using scaling arguments and from the master equation approach, we show that the diffusion is directly related to the density of free traps and not to the absolute density of traps, the occupancy of which is defined by the density of the gas and affinity of the gas for the traps. We find the so-called trapping-dominated diffusion as long as there are more traps than walkers in the system allowing significant interactions of the two, impeding the capacity of walkers to explore the system. When the number of traps and walkers are comparable crowding-enhanced
Contour plot of the mean concentration of bound particles $\langle c_b \rangle$ in the $c_w$-$c_f$ plane. The intersection points between solid black lines and red dashed lines represent the inflection points, calculated from our theory. Two red dashed inflection lines divide the plot into three regions, namely 
trapping-dominated diffusion region (TDD), crowding enhanced diffusion region (CED) and crowding-dominated diffusion region (CDD). The black dashed line and points represent the critical concentration of crowding, at which the effective diffusion coefficient is maximized. The lines have been calculated from our theory, whereas the discrete data points have been obtained from MC simulation using $P_{on} = 0.5$ and $P_{off} = 0.001$

diffusion takes place, where the diffusivity increases with the density of walkers. In this regime, a large number of traps are occupied but there is a significant fraction of particles still able to diffuse in an environment that is not overly crowded, which optimizes the diffusivity constant. Finally, if the number of walkers dominates, than crowding becomes significant even at relatively low $c_w$. The traps are, by and large occupied, but the particles have a smaller likelihood to move to the next site as it is likely already occupied. Consequently the system displays crowding-dominated diffusion where the effective diffusion constant continuously decays with the density of walkers. These results are confirmed by scaling arguments, analytical modeling and kMC simulations.

This mechanism is distinct from recent observation of enhanced diffusion in active systems. The latter was characterized in several theoretical studies, including in diffusion of rod-shaped active particles [36], tracer diffusion inside active particles bath [37] and external force driven tracer diffusion [38]. In these systems diffusion is enhanced either by the energetics of active particles or by external force driven dynamics. However, in our minimalistic model of lattice gas in the presence of traps crowding-enhanced diffusion relies solely on the interplay between crowding and trapping as part of equilibrium thermodynamics.

In various systems such as proteins on membranes interacting with the cell environment, colloidal films or granular particles in trapping potentials, or in diffusion on solid interfaces with defects, the simultaneous effects of crowding and trapping are common. Our theoretical model provides the key tools to analyze such systems. However, to further broaden its applicability or framework could be extended to account for lateral “cis” interactions between traps and walkers. Modeling crowded membranes, may also require the inclusion of multitude of walkers and trap types. This would impose stronger correlations.

From the experimental point of view, recent progress in label-free single-molecule techniques such as iSCAT, and development of more stable dies used in STED microscopy, can provide detailed analysis of the discussed systems. One prominent example is a measured 4-fold decrease in diffusivity of integrins diffusing on the cell membrane and interacting with RGD fragments on the surface forming focal adhesion [39], which is consistent with our crowding-dominated regime. Following the characteristics of nascent adhesions [4,40], our model would predict trapping-dominated regime (see SI Fig.4a), with a crossover in diffusivity taking place during maturation of adhesions. While this prediction is yet to be confirmed, an interesting consequence of such a change in diffusivity is the change in the effective unbinding probability of integrins (see SI Fig.4b). This can have an impact on the system’s entropy production, and the sensing efficiency of the ensemble. This intriguing effect will be investigated more thoroughly in the future.

Few recent experimental studies, furthermore, suggested that crowding can favorably affect diffusion. One example is the facilitated diffusion of DNA-recognizing protein during specific target search over a long DNA strand [41–43], which is a system that shows similar features as our model. Non-monotonous behavior was also observed in simulations of a tracer diffusing in a field of particles crowding the environment, as a function of the density of crowder and the depth of the minimum of the tracer-particle interaction potential, which is a result consistent with our findings albeit in three dimensions [44,45]. Moreover, in three dimensions both experimental and theoretical studies on polymer transport in a crowded medium suggested that crowding enhances long-term diffusivity [46,47]. It would be therefore interesting to extend our work from surface to volume diffusion and account for more complex behavior and properties of traps.

Supplementary Information The online version contains supplementary material available at https://doi.org/10.1140/epjp/s13360-022-02571-4.

Acknowledgements We acknowledge the funding by ERC StG 2013-337283 of the European Research Council, the German Science Foundation Program SFB 1411 Design of Particulate Systems and the Institute of Ruđer Bošković support funds.

Funding Open Access funding enabled and organized by Projekt DEAL.
Data availability  The datasets generated during and analysed during the current study are available from the corresponding author on reasonable request.

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