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Relaxation properties in a diffusive model of extended objects on a triangular lattice

J.R. Šćepanović [a], Lj. Budinski-Petković [b], I. Lončarević [b], M. Petković [c], Z.M. Jakšić [a], S.B. Vrhovac [a,*]

[a] Institute of Physics Belgrade, University of Belgrade, Pregrevica 118, Zemun 11080, Belgrade, Serbia

[b] Faculty of Engineering, Trg D. Obradovića 6, Novi Sad 21000, Serbia

[c] RTRK, Novi Sad 21000, Serbia

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ABSTRACT

In a preceding paper, Šćepanović et al. [J.R. Šćepanović, I. Lončarević, Lj. Budinski-Petković, Z.M. Jakšić, S.B. Vrhovac, Phys. Rev. E 84 (2011) 031109. http://dx.doi.org/10.1103/PhysRevE.84.031109] studied the diffusive motion of k-mers on the planar triangular lattice. Among other features of this system, we observed that the suppression of rotational motion results in a subdiffusive dynamics on intermediate length and time scales. We also confirmed that systems of this kind generally exhibit heterogeneous dynamics. Here we extend this analysis to objects of various shapes that can be made by self-avoiding random walks on a triangular lattice. We start by studying the percolation properties of random sequential adsorption of extended objects on a triangular lattice. We find that for various objects of the same length, the threshold ρ_p^* of more compact shapes exceeds the ρ_p^* of elongated ones. At the lower densities of ρ_p^*, the long-time decay of the self-intermediate scattering function (SISF) is characterized by the Kohlrausch–Williams–Watts law. It is found that near the percolation threshold ρ_p^*, the decay of SISF to zero occurs via the power-law for sufficiently low wave-vectors. Our results establish that power-law divergence of the relaxation time τ as a function of density ρ occurs at a shape-dependent critical density ρ_c above the percolation threshold ρ_p^*. In the case of k-mers, the critical density ρ_c cannot be distinguished from the closest packing limit $\rho_{CPL} \lessapprox 1$. For other objects, the critical density ρ_c is usually below the jamming limit ρ_{jam}.

1. Introduction

In the past two decades, a large number of observations related to anomalous diffusion [1] have been reported in several fields of physics and related sciences. Recent attention focuses on the anomalous slow dynamics in a broad variety of systems such as colloidal gels [2], porous materials [3–6], glass forming systems [7,8], granular fluids [9,10] and biological media [11,12]. The signature of subdiffusion is that the mean square displacement of particles grows sublinearly with time, which may be related to the non-Markovian nature of the stochastic process present in these systems. The reduced mobility is often accompanied by dynamical heterogeneities [13,14] and cooperative motion [15]. A system is considered as dynamically heterogeneous if dynamically distinguishable populations of particles with different mobilities can be isolated by a computer simulation or experiment. Deviations of the van Hove correlation function from the Gaussian behavior are usually ascribed to the presence of particles that are substantially faster or slower than the average [13,14].

* Corresponding author.
E-mail address: vrhovac@ipb.ac.rs (S.B. Vrhovac).
URL: http://www.ipb.ac.rs/~vrhovac/ (S.B. Vrhovac).

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In a preceding paper [16], we studied a model of the diffusive motion of k-mers on the planar triangular lattice. In this model, the moves of a randomly chosen k-mer can be either translation along its axis or rotation when k-mer changes its orientation. The only interactions between the particles are the geometrical ones, i.e., we allow only the single-k-mer moves that do not cause double occupation at any site. In the absence of rotation, a k-mer can only move along one of the three axis bisecting the triangular lattice. In this case, the model can serve as an example of mutually intersecting arrays of single-file systems (''channels'') [17]. Prominent examples of such systems are zeolites of MFI structure type like ZSM-5 [18,19].

The analysis in Ref. [16] pertained mainly to relaxation process at densities high enough that the mean-squared displacements exhibit a subdiffusive behavior at intermediate times between the initial transient and the long-time diffusive regime. We have shown that there is a pronounced deviation of the van Hove correlation function from the Gaussian distribution, especially at high densities. We have found that the decay of self-intermediate scattering function (Eq. (6)) to zero occurs via the Kohlrausch–Williams–Watts law (Eq. (7)) for all values of the wave vector q_n and for all densities investigated. We have shown that for dimers, relaxation times display a power-law divergence at densities around the closest packing limit $\rho_{CPL} \lessapprox 1$. The time scale and the diffusion coefficient show qualitatively the expected behavior as a function of density, since the inverse of the self diffusion coefficient D_s also seems to diverge with the power law at the maximum density ρ_{CPL}. These studies have shown that our model reproduces many aspects of the dynamics of glass-forming systems, e.g., the slowing-down of the dynamics on increasing the density suggesting the existence of a transition of structural arrest at ρ_{CPL}.

The purpose of this paper is to extend the analysis in Ref. [16] to objects of various shapes that can be made by self-avoiding random walks on a triangular lattice [20]. We address the following questions: (1) How does particle shape modify slow dynamics in our diffusive lattice model? (2) How do various objects promote or suppress kinetic arrest of the system depending on degree of shape anisotropy? Our approach can provide some answers to such questions because the slowing-down of the dynamics in the model can be understood as a consequence of steric effects, that make certain moves of particles impossible owing to an effective high local density.

Now, we sketch the main features of our model. The system is initialized by the random sequential adsorption (RSA) model [21–24]. In two dimension (2D), RSA is a process in which the objects of a specified shape are randomly and sequentially deposited onto a substrate. Excluded volume, or particle–particle interaction, is incorporated by rejection of deposition overlap, while the particle–substrate interaction is modeled by the irreversibility of deposition. The deposition process ceases when all unoccupied spaces are smaller than the size of an adsorbed particle. The system is then jammed in a nonequilibrium disordered state for which the limiting (jamming) coverage ρ_{jam} is less than the corresponding density of closest packing ρ_{CPL}. However, when adsorbed particles are allowed to diffuse, jammed configurations can be relaxed by particle movement (RSAD models). Jamming density in RSAD models is higher and in some cases closest packing density is reached [24]. Random closest packing states are characterized by the absence of mobile particles. At a predetermined density $\rho_0 < \rho_{CPL}$, the deposition is terminated and random diffusive dynamics is initiated in our system. In general, objects have both translational and rotational motion. However as we focus on the subdiffusive behavior, we suppress the rotational motion of objects.

As a first step toward answering the above questions, we analyze the percolation properties of random sequential adsorption of extended objects on a triangular lattice. Dynamical behavior of model at large length scales is studied via the mean square displacement of a particle (MSD, Eq. (3)). We evaluate the relaxation times by studying the time decay of the self-part of the intermediate scattering function (SISF, Eq. (6)). In particular, we analyze the relaxation time dependence both on the wave-vector (length scale) and on the density of the system. Most of our attention is focused on the influence of shape on the temporal behavior of MSD and SISF.

In Section 2 we introduce our model and give some details of our simulations. We present the simulation results and discussions in Section 3. Finally, Section 4 contains some additional comments and final remarks.

2. Definition of the model and the simulation method

We study the random diffusion of extended objects on a planar triangular lattice with fixed density, where the interparticle interaction is limited to hard-core exclusion up to the nearest neighbors. The initial state of the system is prepared through the random sequential adsorption (RSA) model where by deposition of particles we reach the desired value of the density. The depositing objects are made by directed self-avoiding random walks on the lattice. On a triangular lattice objects with a symmetry axis of first, second, third and sixth order can be formed. Rotational symmetry of order n_s, also called n_s-fold rotational symmetry, with respect to a particular axis perpendicular to the triangular lattice, means that rotation by an angle of $2\pi/n_s$ does not change the object. We performed numerical simulations for all such shapes of length $\ell = 1$, 2 and 3, covering two, three and four lattice sites, respectively. All these objects are shown in Table 1. On a triangular lattice it would also be interesting to examine the behavior of a hexagon shown in Table 1.

We start with an initially empty triangular lattice. At each Monte Carlo step a lattice site is selected at random. If the selected site is unoccupied, deposition of the object is tried in one of the six orientations. We fix both the direction and the beginning of the walk that makes the shape at the selected site and search whether all successive ℓ sites are unoccupied. If so, we occupy these $\ell + 1$ sites and place the object. If the attempt fails, a new site and a new direction are selected at random. The coverage of the surface is increased by inserting the objects randomly up to the chosen coverage fraction ρ_0. In this way we are able to prepare the system in disordered initial state with a statistically reproducible density ρ_0.