Journal of Statistical Physics, Vol. 117, Nos. 1/2, October 2004 (© 2004)

Dynamical Arrest, Tracer Diffusion and Kinetically Constrained Lattice Gases

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Received February 11, 2004; accepted April 22, 2004

We analyze the tagged particle diffusion for kinetically constrained models for glassy systems. We present a method, focusing on the Kob–Andersen model as an example, which allows to prove lower and upper bounds for the self-diffusion coefficient D_S. This method leads to the exact density dependence of D_S, at high density, for models with finite defects and to prove diffusivity, $D_S > 0$, at any finite density for highly cooperative models. A more general outcome is that under very general assumptions one can exclude that a dynamical transition, like the one predicted by the Mode-Coupling-Theory of glasses, takes place at a finite temperature/chemical potential for systems of interacting particle on a lattice.

KEY WORDS: Tagged particle diffusion; kinetically constrained lattice gases; glassy dynamics.

1. INTRODUCTION

Many physical systems, in particular glass forming liquids, display a very slow dynamics at low temperature/high density.(1) The laboratory glass transition corresponds to the temperature/density at which the structural relaxation timescale becomes larger than the experimental one (e.g. one hour). At this point the glass-forming liquid falls out of equilibrium and becomes an amorphous rigid material called glass. Thus, the laboratory glass transition is nothing else than a dynamical crossover and not a true dynamical transition. However, a natural question is whether this dramatic

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0022-4715/04/1000-0027/0

increasing of the structural relaxation time is due to an underlying dynamical transition that takes place at lower temperature/higher density (but that is unreachable experimentally). Indeed different analytical approaches, in particular the Mode-Coupling-Theory (MCT) of the glass transition[2] predict a dynamical arrest at a finite temperature and chemical potential at which the structural relaxation timescale diverges and the diffusion coefficient of a tagged particle, called self-diffusion coefficient D_S, vanishes. The interpretation of this transition is based on the cage effect: particles are trapped in the cage formed by their neighbors (however see also ref. 3). For particles interacting with a smooth potential it is widely accepted that this MCT transition describes at most a dynamical crossover. Instead in the case of potentials with a hard core part, in particular hard sphere systems, there is no agreement. Experiments on colloids,[4] that can be indeed modeled by hard sphere systems, are very often interpreted as if a real dynamical transition took place. However, in these systems the microscopic timescale is much larger than for the other glass forming liquids (approximatively nine order of magnitude), thus it could be argued that one is just looking to the very first part of the increasing of the relaxation timescale, here MCT indeed applies but the point at which the dynamical MCT crossover takes place is shifted toward unreachable experimental timescales.

What can one say theoretically about the existence of this type of transition called sometimes dynamical arrest? First let us focus on the case of interacting particle systems on a lattice. If the potential between the particle is short-range, the Hamiltonian is not singular and the only constraint is the hard core one, i.e. maximum one particle per site, then it has been proved by Spohn[5,6] that on long time and length scales the tagged particle performs a simple Brownian motion with a self-diffusion coefficient that is positive at any finite temperature and any finite chemical potential (these models are called in the mathematical literature symmetric exclusion processes with speed change and spin exchange dynamics; in the following we will refer simply to them as RLG, i.e. reversible lattice gases). Actually, one needs also the system to be ergodic but physically this is just due to the fact that if there is a very large correlation length and/or a very large correlation time than, in principle, the tagged particle will enter in the Brownian motion regime on larger length and time scales. So at a critical point the tagged particle could take an infinite time to enter in the Brownian motion regime. However, the lower bound on the self-diffusion coefficient proved in refs. 5 and 6 is valid regardless of the existence of a critical point. Thus, even if a phase transition would take place at a temperature T_c and a chemical potential μ_c one gets $\lim_{T\to T_c;\mu\to\mu_c} D_S(T,\mu) > 0$.

However, the lattice models that are known to display a whole phenomenology analogous to glass forming liquids and for which the existence of a dynamical arrest at finite temperature/chemical potential has been suggested, are the ones in which further hard constraints besides hard core, are imposed. Among the most studied ones there are the kinetically constrained lattice gases (KCLG) for which the jump rates of particles are different from zero not only if the constraint of having maximum one particle per site is verified but *also* if some additional constraint is verified, hence the name kinetically constrained (see ref. 7 for a recent review). We will comment on the extension of our work to statically constrained models, like the Lattice Glass Models introduced in ref. 8, and more general cases in the conclusion.

Note that the presence of additional hard constraints can change the physical mechanism behind tagged particle diffusion quite a lot. As explained in ref. 7 the KCLG are characterized at high density by the existence of "defects" that are the analog of vacancies for the RLG. A rather good understanding has been reached for models in which defects, that can freely move in an otherwise completely filled lattice, consist in a finite number, independent of the particle density, of vacancies. Instead, the cases in which the motion of "defects" involve a number of vacancies that diverges when the particle density approaches one, were much less understood. Indeed in the literature one can find numerical simulations suggesting a dynamical transition at a density less than one.[7] Recently, in collaboration with Fisher,[9] we have analyzed one of the most studied model of this type, the Kob-Andersen model[10] proving that no dynamical transition takes place whatsoever and unveiling what is the mechanism inducing the slow dynamics. Our results and techniques apply also to the other highly cooperative KCLG.[11]

In this paper, under very general assumptions, we show how one can generalize the proof of diffusivity, $D_S > 0$ of refs. 5 and 6 to KCLG (this has been already done for simple KCLG with finite size defects in ref. 12) Our aim is twofold. First, we want to show that under very general assumptions (see Sections 2 and 7) a dynamical arrest cannot take place at finite temperature/chemical potential even if there are hard constraints other than the hard core one. Second, we want to present a rigorous technique and a method that allows to obtain upper and lower bounds on the self-diffusion coefficient for KCLG (or more general interacting particle systems). The usual techniques applied to KCLG in order to obtain predictions on D_S, as diagrammatic resummation or approximate closure of exact equations (see refs. 7 and 13), are completely out of control in the high density regime. Although some of them work well in the intermediate density regime compared to the results of numerical simulations, they

fail in general at high density and, often, predict a spurious dynamical transition. Thus, in this context it is particularly important to have rigorous methods that allow one to obtain solid analytical predictions. We will focus on the Kob–Andersen model as an example but our method can be applied to also other KCLG or more general interacting particle systems.

Let us finally comment on the continuum case. We are not aware of any proof of diffusivity at low temperature/high density in the case of Hamiltonian dynamics. Instead in the case of interacting Brownian particles, even with hard core (a good model for colloidal systems), it has been recently shown that the self diffusion coefficient is positive at any finite temperature/chemical potential under very general assumptions.[14] The proof works for hard spheres, Lennard–Jones potential, etc ... and it is, as our proof, a generalization of the Spohn's proof[5,6] for RLG.

We think that the technique we will make use of is not well known in the community working on KCLG therefore we have written this article in a detailed and self-contained way. The expert reader may skip Section 4 and quickly go through Section 5.

We introduce in detail the KCLG in Section 2 and we explain in detail the KA model in Section 3. In Section 4, we recall some probabilistic techniques that have been used to prove bounds on the self-diffusion coefficient for the RLG.[6] In Section 5, focusing on the KA model on a triangular lattice as an example, we explain how one can obtain the high density behavior of the self-diffusion coefficient for KCLG with "defects" formed by a finite number of vacancies. This section is useful to introduce the notation and the method before facing the difficult case of highly cooperative KCLG. In Section 6, focusing on the KA model on a square lattice, we explain how one can obtain strictly positive lower bounds in the case of highly cooperative KCLG. Finally, in Section 7 we present a final discussion of our results.

2. KINETICALLY CONSTRAINED LATTICE GASES

In the last twenty years there has been a growing interest in kinetically constrained lattice gases. These were introduced as models for supercooled liquids close to the glass transition[15] and nowadays they are also studied as paradigm for general glassy systems.[7] KCLG are (apparently) similar to RLG. As discussed before there is however an important difference in the choice of the jump rates of particles that are different from zero not only if the constraint of having maximum one particle per site is verified but *also* if some additional constraint is verified, hence the name kinetically constrained. This choice of jump rates was originally devised in order to mimic the cage effect, that might be at the heart of the glassy

behavior and the slow dynamics of glass forming liquids. Indeed a molecule in a dense liquid is typically trapped in a cage created by surrounding particles (see ref. 16 for a visual experimental example) and this takes place in the regime of temperature and density at which the dynamics slows down dramatically.

More specifically, kinetically constrained lattice gases are stochastic lattice gases with hard core exclusion, i.e. systems of particles on a lattice Λ with the constraint that on each site there can be at most one particle. A configuration is therefore defined by giving for each site $x \in \Lambda$ the occupation number $\eta_x \in \{0, 1\}$, which represents an empty or occupied site, respectively. The dynamics is given by a continuous time Markov process on the configuration space $\Omega_\Lambda = \{0, 1\}^{|\Lambda|}$ which consists of a sequence of particle jumps. A particle at site x attempts to jump to a different site y with a fixed rate $c_{x,y}(\eta)$, which in general depends both on $\{x, y\}$ and on the configuration η over the entire lattice. The discretized time version of the process is the following. At time t choose at random a particle, let x be its position, and a site y. At time $t + \mathrm{d}t$, the particle has jumped from x to y with probability $c_{xy}(\eta(t))$ and not jumped with probability $1 - c_{xy}(\eta(t))$. In other words, the probability measure at time t, μ_t, can be derived by the initial measure μ_0 as

$$\mu_t(\eta) = \sum_{\eta' \in \{0,1\}^{|\Lambda|}} \exp(\mathcal{L}t)(\eta, \eta')\mu_0(\eta'), \tag{1}$$

where $\mathcal{L}$, the *generator of the Markov process*, is the operator which acts on local functions $f : \Omega_\Lambda \to \mathbb{R}$ as

$$\mathcal{L}f(\eta) = \sum_{\{x,y\} \subset \Lambda} c_{x,y}(\eta)\left(f(\eta^{xy}) - f(\eta)\right), \tag{2}$$

where we defined

$$(\eta^{xy})_z := \begin{cases} \eta_y & \text{if } z = x, \\ \eta_x & \text{if } z = y, \\ \eta_z & \text{if } z \neq x, y. \end{cases} \tag{3}$$

The simplest model is the simple symmetric exclusion process, SSEP, in which $c_{x,y}^{\mathrm{SSEP}}(\eta) = \eta_x(1 - \eta_y) + \eta_y(1 - \eta_x)$ for nearest neighbors $\{x, y\}$, $c_{x,y}(\eta) = 0$ otherwise. Therefore, only nearest neighbor jumps are allowed and there are no further kinetical constraints besides hard core. The definition *kinetically constrained* refers more properly to models in which jump

rates impose additional requirements in order for the nearest neighbor move to be allowed. In other words, the rate $c_{x,y}(\eta)$ can be zero for some choices of the configuration η and the couple $\{x, y\}$ even if $\eta_x = 1$, $\eta_y = 0$, thus preventing the jump of a particle from site x to final empty site y. From the above definition it is immediate to see that dynamics preserves the number of particles, i.e. the hyperplanes with fixed number N of particles $\Omega_{\Lambda,N} := \{\eta \in \Omega_\Lambda \ \sum_{x\in\Lambda} \eta_x = N\}$ are invariant under dynamics. Moreover, in general the rates are chosen in order to satisfy detailed balance w.r.t. the uniform measure $\nu_{\Lambda,N}$ on such hyperplanes. In other words, the condition $c_{x,y}(\eta) = c_{y,x}(\eta^{xy})$ is satisfied for any choice of η and the couple $\{x, y\}$. This implies that the generator is reversible with respect to $\nu_{\Lambda,N}$ and therefore $\nu_{\Lambda,N}$ is stationary.[3] Note that $\nu_{\Lambda,N}$ is nothing else than canonical measure with zero Hamiltonian, i.e. with this choice of the rules there are no static interactions beyond hard core and an equilibrium transition cannot occur. However, a priori it is possible that a dynamical ergodic/non-ergodic transition occurs for some choices of the rules. To our knowledge, the KCLG which have been considered so far do not display such transition (see refs. 9 and 11 for the proof that an ergodic/non-ergodic transition does not occur in some highly cooperative KCLG). On the other hand, such transition has been proved to occur for a kinetically constrained spin model, namely North-East model.[17,18] We shall discuss in the following possible forms of such transition rates. However, we emphasize since now that the degeneracy of the rates implies that $\nu_{\Lambda,N}$ is not the unique invariant measure, i.e. the system is not irreducible on $\Omega_{\Lambda,N}$ and this will have several consequences on dynamics inducing a very different behavior with respect to RLG case.

3. DEFINITION OF THE KOB–ANDERSEN MODEL

The KA model is a kinetically constrained lattice model with jump rates

$$c_{x,y}(\eta) := \begin{cases} c_{x,y}^{\mathrm{SSEP}}(\eta) & \text{if } \sum_{\substack{z\in\Lambda, z\neq y \\ d(x,z)=1}} \eta_z \leqslant m \text{ and } \sum_{\substack{z\in\Lambda, z\neq x \\ d(y,z)=1}} \eta_z \leqslant m, \\ 0 & \text{otherwise,} \end{cases} \tag{4}$$

namely a particle can move only if the hard core constraint is verified, as for the SSEP, *and* only if both before and after the move it has no

[3] Let $\mu(g,h) = \sum_{\eta\in\Omega} \mu(\eta) g(\eta) h(\eta)$. $\mathcal{L}$ is reversible with respect to μ if, for any functions f and g, the equality $\mu(g, \mathcal{L}f) = \mu(f, \mathcal{L}g)$ holds. By a direct calculation it is possible to check that detailed balance implies reversibility with respect to $\nu_{\Lambda,N}$, therefore the choice $g(\eta) = 1$ $\forall\eta$, implies $\mu(\mathcal{L}f) = 0$ $\forall f$. This, together with (1) implies that $\nu_{\Lambda,N}$ is invariant under time evolution.

more than m neighboring particles. If Λ is an hypercubic d-dimensional lattice m will take values only from 0 to $2d-1$ (different values of m define different KA models). Note that for $m=2d-1$ the simple symmetric exclusion case with Hamiltonian equal to zero is recovered. For future purposes it is useful to reformulate the rule in term of motion of vacancies. Indeed, as can be easily verified, the above dynamical rule corresponds to *vacancies* moving only if the initial and final sites have at least $s=z-m-1$ neighboring vacancies, with $z=2d$ the coordination number of the lattice. Therefore, the model is completely defined by the choice of the couple d, m or equivalently d, s.

Note that these rates satisfy detailed balance with respect to $\nu_{\Lambda,N}$, i.e. the uniform measure on the hyperplanes with fixed number of particles. However, there exist configurations that are blocked under the dynamics, $\nu_{\Lambda,N}$ is not the unique invariant measure on the hyperplane. For example, in the case $d=2$ $s=1$ with periodic boundary condition, a configuration which has a double row of sites completely filled belongs to a different ergodic component with respect to any other configuration which does not contain such structure. Indeed, one can directly check that the particles belonging to the double row can never move.

On the infinite lattice $\Lambda=\mathbb{Z}^d$ the model satisfies detailed balance with respect to the Bernoulli product measure $\mu_{\mathbb{Z}}^d, \rho$ at any density ρ. It is immediate to check that for $s \geqslant d$ the system is not ergodic at any density $\rho>0$. Indeed in this case all d-dimensional hypercubes of any size which are completely occupied by particles are blocked forever. On the other hand, in refs. 9 and 11, we have proved that for $s<d$ the model is ergodic at any density $\rho \in [0,1]$ with probability one. As a by product of the ergodicity proof we have established the following property, which will be a key ingredient for the proof in Section 6. Let p be a positive number less than one. For any fixed density $\rho<1$ there exists a finite length $\Xi(\rho)$ such that, by sorting at random with probability $\mu_{\Lambda,\rho}$ a configuration on a hypercubic d-dimensional lattice Λ of linear size $\Xi(\rho)$, with probability greater than p this configuration is such that any particle exchange inside a finite box around the origin can be performed through a suitable path of allowed moves.[4] Furthermore p can be taken arbitrary close to one taking a suitable (large but finite) $\Xi(\rho)$. In the following we let a configuration with such property be a *frameable configuration* and refer to refs. 9 and 11 for the proof (and an explanation of why we call these configurations "frameable").

[4]The length $\Xi(\rho)$ depends on the choice of d, s and the size of the box, we drop the dependence on these parameters for simplicity of notation.

4. DIFFUSION OF THE TAGGED PARTICLE

Consider a kinetically constrained model on the infinite lattice $\Lambda = \mathbb{Z}^d$ and start at time zero from the equilibrium distribution, so that the process will be stationary. Then single out one particle, the tracer, and analyze its motion. In the density regime where the process is ergodic one can repeat the arguments in refs. 6 and 19 and show that under a diffusive rescaling the position of the tracer at time t, $\vec{x}(t)$, converges to a Brownian motion with self diffusion matrix $D_S(\rho)$. More precisely, $\lim_{\epsilon \to 0} \epsilon \vec{x}(\epsilon^{-2}t) = \sqrt{2D_S}\, \vec{b}(t)$, where $\vec{b}(t)$ is the standard Brownian motion and the self-diffusion matrix $D_S(\rho)$ is given by the variational formula:[(5)5]

$$
\begin{aligned}
(\vec{l}\cdot D_S(\rho)\cdot\vec{l}) \;= \inf_f \Bigg[& \frac{1}{2} \sum_{\{y\neq 0\}\subset\Lambda} \mu_{\rho,0}\Bigg(c_{0,y}(\eta)(1-\eta(y)) \Bigg[\sum_{i=1}^{d} (\vec{l}\cdot\vec{d}) \\
& + f(\tau_{-y}\eta^{0y}) - f(\eta) \Bigg]^2 \Bigg) \\
& + \frac{1}{4} \sum_{\substack{\{x,y\}\subset\Lambda \\ x\neq 0, y\neq 0}} \mu_{\rho,0}\left(c_{x,y}(\eta)[f(\eta^{xy}) - f(\eta)]^2 \right) \Bigg], \qquad (5)
\end{aligned}
$$

where $\vec{l}$ is a unit vector in $\mathbb{Z}^d$, $(\vec{a}\cdot\vec{b})$ is Euclidean scalar product, $\tau_{-y}\eta$ is the configuration obtained translating the configuration η of y, $\mu_{\rho,0}$ is the Bernoulli measure at density ρ conditioned to the existence of a particle in the origin and the infimum is over all real-valued local functions f, i.e. functions which depend on a finite number of occupation variables. For spatially isotropic systems, as the one we will deal with, the self-diffusion matrix is usually proportional to identity, the proportionality coefficient being the so-called *self-diffusion coefficient*. For future purposes it is useful to define the two sums in (5) as $D_1(\rho, f)$ and $D_2(\rho, f)$

$$
\begin{aligned}
D_1(\rho, f) = & \frac{1}{2} \sum_{\{y\neq 0\}\subset\Lambda} \mu_{\rho,0}(c_{0,y}(\eta)(1-\eta(y))[\sum_{i=1}^{d} (\vec{l}\cdot\vec{d}) \\
& + f(\tau_{-y}\eta^{0y}) - f(\eta)]^2 \Big), \qquad (6)
\end{aligned}
$$

$$
D_2(\rho, f) = \frac{1}{4} \sum_{\substack{\{x,y\}\subset\Lambda \\ x\neq 0, y\neq 0}} \mu_{\rho,0}\left(c_{x,y}(\eta)[f(\eta^{xy}) - f(\eta)]^2 \right). \qquad (7)
$$

[5]Note that by the identity (5) one obtain that the matrix D_S is positive, therefore the square root is a well defined function.

The resulting form of the variational expression (5) can be generalized also to the case of Brownian interacting particles[20] and is indeed quite remarkable.[5] First, it implies that the self-diffusion coefficient is a monotonic increasing function of the microscopic rates. Second, it disentangles partially the contribution to D_S coming from the motion of the tracer from the one (indirectly) due to the motion of the environment. In fact this expression has been obtained changing the reference system in such a way that the tracer is always in the origin, thus $c_{0,y}$ is the rate of jump of the tracer whereas $c_{x,y}$ with $x, y \neq 0$ is the rate of jump for the environment. As a consequence, by keeping only D_1 in the variational expression, one obtains the self-diffusion coefficent of the tracer when the environment is frozen. This provides a simple lower bound for low enough density. However, even for the SSEP, for densities higher than the site percolation transition, the tracer does not diffuse in this random environment and one is forced to take into account that the environment is not frozen. From the mathematical point of view, inequalities on D_2 allow one to prove that $D_S > 0$ at $\rho < 1$[6] for the RLG case in any dimension greater than one. Since this proof uses as a key ingredient the fact that jumps from occupied to neighboring empty sites are always allowed, it cannot be trivially extended to kinetically constrained models. We emphasize that this is not only a technical difficulty. Indeed, due to the presence of kinetic constraints, the physical mechanism behind the diffusion of the tagged particle may be rather different as we shall show in Section 5.

In Section 5, we will prove that $D_S > 0$ at any $\rho < 1$ for KA model on a triangular lattice. For this choice of the lattice there exist "defects", namely clusters of two vacancies that can freely move through an otherwise totally filled lattice and such that the tagged particle can move whenever such "defects" passes by. Therefore, these defects play the same role of the vacancies for the RLG case and the above mentioned proof of[6] (see also ref. 5) can be generalized. In the following, we sketch this result in some detail since it can be extended to other models with finite "defects" (different models with finite defects where already considered in ref. 12) and it is useful to introduce the notation and the method before analyzing the more difficult case of KA model on hypercubic lattices. Indeed, in this case it is not possible to construct finite cluster of vacancies that can freely move into an otherwise totally filled lattice and additional work is required to prove diffusivity. As explained in ref. 9, in this case the mechanism behind diffusion is based on the existence of "quasi-defects", namely cluster of vacancies which can move into typical regions of the system and allow the motion of a tagged particle (via a particular path of elementary moves) when they pass by. The size of such defects is density dependent and diverges for $\rho \to 1$, which makes the motion *highly cooperative*.

5. A MODEL WITH FINITE SIZE DEFECTS: KA $S=1$ ON A TRIANGULAR LATTICE

Consider the KA model with $s=1$ on a triangular lattice Λ represented in Fig. 1. More precisely, the set of sites in Λ is the union of sites in a square lattice Λ_1 and in its dual Λ_2, i.e. the lattice obtained by displacing Λ_1 of $(e_1+e_2)/2$ with $e_1=(1,0)$, $e_2=(0,1)$. Furthermore, two sites $\{x, y\} \in \Lambda$ are nearest neighbors if $x-y=\pm e_1$ or $x-y=\pm(e_1+e_2)/2$ or else $x-y=\pm(-e_1+e_2)/2$. As already noticed, rates (4) reformulated in terms of vacancies correspond to the rule that a vacancy can move only if the initial and final sites have at least s neighboring vacancies, where s equals one in the case we are considering. In the triangular lattice two neighboring sites always share a common third neighbor. Therefore, any of two nearest neighbor vacancies can move to the common third neighbor. In other words, a set of two neighboring vacancies, which we call "a defect", can be freely moved into an otherwise totally filled lattice, as can be immediately checked (see Fig. 2).

5.1. Heuristic Arguments and Upper Bound on the Self-Diffusion Coefficient

There is a simple heuristic argument based on the independent motion of these defects that leads to the correct density dependence of the self-diffusion coefficient at high density. Call ρ_{d} the density of defects and τ_{d} the timescale on which defects move. The self-diffusion coefficient D_{s} of

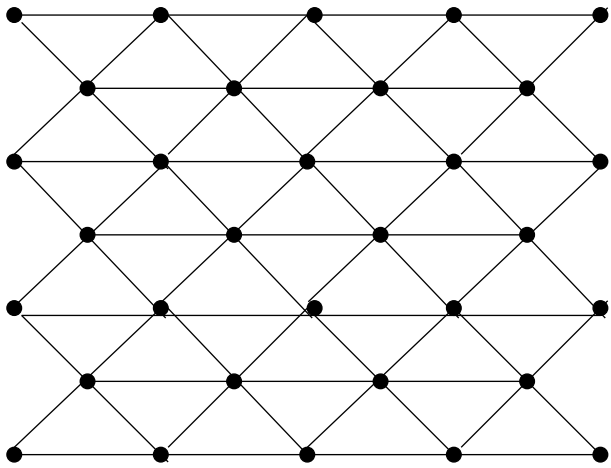

Fig. 1. The triangular lattice.

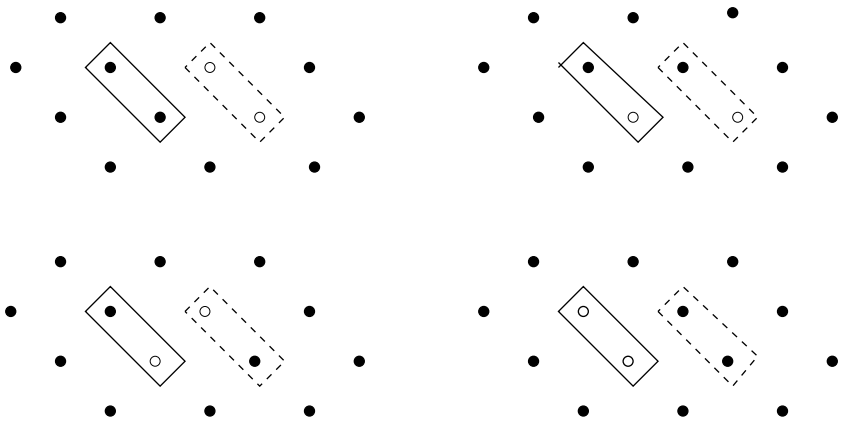

Fig. 2. Sequence of moves which allow the displacement of a couple of neighboring vacancies. Circles denotes empty sites, filled dots stand for occupied sites.

a tagged particle is expected to be proportional to the inverse of the time τ_p on which each particle moves of one step. On the timescale τ_d the number of particles that have jumped is of the order $V\rho_d$ where V is the total number of sites. Thus, we find:

$$\frac{\tau_p}{\tau_d} V\rho_d \propto V\rho. \tag{8}$$

As a consequence, at density close to one, we get $D_S \propto \rho_d/\tau_d$. Note that we have assumed that the size of the defects do not change, in particular do not diverge in the limit $\rho \to 1$, otherwise the reasoning has to be changed slightly. Since for the KA model on the triangular lattice we are focusing on defects that are formed by two vacancies we find, in the limit $\rho \to 1$, $\rho_d \propto (1-\rho)^2$ and $\tau_d \propto O(1)$, hence, $D_S \propto (1-\rho)^2$ (similarly in the SSEP case a defect is just a vacancy and one gets $D_S \propto (1-\rho)$).

In the following, we will show how, using the existence of these defects, it is possible to generalize the proof of the RLG case[6] and show that indeed $D_S \propto (1-\rho)^2$ in the high density limit. In principle our procedure is generalizable to all cases with defects having a size which does not diverge in the limit $\rho \to 1$. To obtain $D_S \propto (1-\rho)^2$ we shall prove that when ρ is close enough to one D_S is bounded from above and below, respectively by $K_U(1-\rho)^2$ and $K_L(1-\rho)^2$, where $K_{U,L}$, are two positive constants (of course $K_U \geqslant K_L$).

The proof of the upper bound is very easy: it consists just in choosing an appropriate test function $f(\eta)$ and evaluating the term in the parenthesis in the variational formula (5). Indeed consider the test function $f_0(\eta) =$

η_0, which for each configuration is equal to the occupation number in the origin. Plugging it into (5) we find a term which is proportional to the probability to have a vacancy on a fixed site and at least another vacancy close to it (this is nothing else that the probability to have a defect). Thus, we obtain $D_S \leqslant K_U(1-\rho)^2$, where K_U is a suitable positive constant.

5.2. Lower Bound on the Self-Diffusion Coefficient

Now let us focus on the lower bound which is more involved. Instead of dealing directly with the variational formula (5), we define a proper auxiliary model and proceed in two steps. First, we establish that $D_S^{aux} > 0$; second, we prove that $D_S \geqslant cD_S^{aux}$ with c a positive constant. More precisely, we will introduce an auxiliary process and prove that the diffusion coefficient in direction e_1 is positive, i.e. $e_1 D_s^{aux} e_1 > 0$ and $e_1 D_S e_1 \geqslant e_1 D_S^{aux} e_1$. In an analogous way one can then introduce an auxiliary process to show the same inequalities in direction $(e_2 + e_1)$, which completes the proof of the positivity of the self-diffusion matrix.

5.2.1. Construction of the Auxiliary Process

Let us introduce some notation. Consider the following subsets of Λ:

$$R_x^1 := \left\{x + e_1, \quad x + \frac{e_1 + e_2}{2}\right\},$$
$$R_x^2 := \left\{(x - e_1, \quad x + \frac{-e_1 + e_2}{2}\right\}, \tag{9}$$

R_x^i are the two couples of neighboring sites of x represented in Fig. 3.

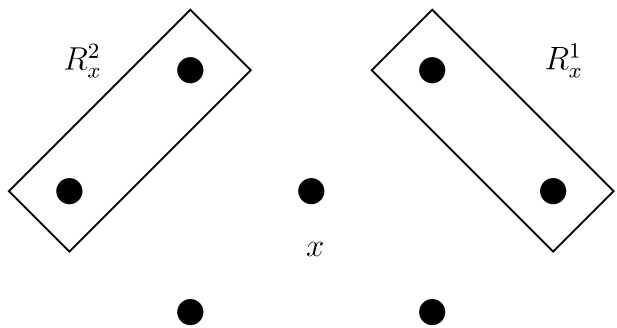

Fig. 3. Sites inside the closed line correspond, from left to right, to sets R_x^1 and R_x^2.

We next define $\eta^{1,2,x}$ as the configuration obtained from η by exchanging the occupation numbers in R_x^1 with the corresponding ones in R_x^2:

$$\left(\eta^{1,2,x}\right)_z := \begin{cases} \eta_{x-e_1} & \text{if } z = x+e_1, \\ \eta_{x+e_1} & \text{if } z = x-e_1, \\ \eta_{x+\frac{e_1+e_2}{2}} & \text{if } z = x+\frac{e_1+e_2}{2}, \\ \eta_{x+\frac{-e_1+e_2}{2}} & \text{if } z = x+\frac{-e_1+e_2}{2}, \\ \eta_z & \text{if } z \notin R_x^1 \cup R_x^2. \end{cases} \tag{10}$$

Finally, we introduce the events:

$$\mathcal{A}_x^i := \left\{\eta \in \Omega : \eta_z = 0 \,\forall z \in R_x^i\right\}, \quad \mathcal{A}_x := \mathcal{A}_x^1 \cup \mathcal{A}_x^2, \tag{11}$$

which contain configurations having the two neighboring sites in R_x^i empty, namely configuration with a defect in R_x^i.

Let us denote by x the position of the tagged particle and define the auxiliary process as follows. At time $t=0$ the tagged particle is in the origin and there is a "defect" in a neighboring couple of sites, namely $x(0)=0$ and $\eta(0) \in \mathcal{A}_x$. (i.e. $\eta_{e_1}(0) = \eta_{e_2/2+e_1/2}(0) = 0$ or $\eta_{-e_1}(0) = \eta_{-e_1/2+e_2/2}(0) = 0$). Then the process evolves as follows:

(i) The tagged particles can jump from x to $x+e_1$. The rate of the jump is one if $\eta \in \mathcal{A}_x^1$ (i.e. $\eta_{x+e_1}=0$ and $\eta_{x-e_1/2+e_2/2}=0$).
(ii) The tagged particles can jump from x to $x-e_1$. The rate of the jump is one if $\eta \in \mathcal{A}_x^2$ (i.e. $\eta_{x-e_1}=0$ and $\eta_{x-e_1/2+e_2/2}=0$).
(iii) Configuration η is exchanged to $\eta^{1,2,x}$ (i.e. the occupation variables of sites $x+e_1$, $x+e_1/2+e_2/2$ are exchanged with the occupation variable in $x-e_1$ and $x-e_1/2+e_2/2$, respectively). The rate of the exchange is one.
(iv) All other moves are not allowed.

With this definition of the rules it is immediate to check that, since the configuration at time zero is such that $\eta(0) \in \mathcal{A}_{x(0)}$, at any subsequent time $t>0$ the condition $\eta(t) \in \mathcal{A}_{x(t)}$ will hold. Moreover, since the defect can be moved from R_x^1 to R_x^2 and viceversa (move (iii) above), the jump of the tagged particle to any of the two neighbors in direction e_1 can always be performed via a sequence of at most two moves. Therefore the self-diffusion coefficient in direction e_1 is strictly greater than zero at any density $\rho<1$, namely $e_1 D_S^{\text{aux}} e_1 > 0$. This can be rigorously proved in the same way as for RLG (see ref. 5).

5.2.2. Proof of the Inequality between D_S and D_S^{aux}

Let us now turn to the second step, namely establishing inequality $D_{\mathrm{S}} > cD_{\mathrm{S}}^{\mathrm{aux}}$, with $c > 0$. Since move (iii) for the auxiliary process is not allowed for KA, some work is required to establish such inequality. The basic idea is to show that all the moves allowed for the auxiliary process can be performed through a proper finite sequence of elementary nearest neighbors jumps which are allowed for the original model. If this is true then it is natural to expect that the above inequality among the diffusion coefficients can be rigorously established. Let us outline the proof. Consider the second term, D_2, of the variational formula (5). The above choice of the rates yields for the auxiliary process:

$$D_2^{\mathrm{aux}}(f,\rho) = \frac{1}{4\mu_{\rho,0}(\mathcal{A}_x)} < \mathbb{I}_{\mathcal{A}_x}(\eta)\left(f(\eta^{1,2,x}) - f(\eta)\right)^2 >_0 . \tag{12}$$

As already mentioned any couple of neighboring vacancies can be freely moved through the lattice using elementary moves allowed by KA, see Fig. 2. In particular, since the configuration which enters in (12) have a couple of vacancies either in the set R_x^1 or R_x^2 (or both), by using the basic moves in Fig. 2. it is possible to construct a path of elementary nearest neighbor exchanges which have unit rate for KA model and connect η to $\eta^{1,2,x}$. Moreover, such path is independent on the configuration outside R_x^1 and R_x^2. In other words, by recalling definition (3) and letting the exchange operator $T_{x,y}$ be

$$T_{x,y}\eta = \eta^{x,y} \tag{13}$$

the following equivalence holds:

$$\eta^{1,2,x} = \prod_{j=1}^{n} T_{x_{j+1},x_j}\eta, \tag{14}$$

where n is the length of the above defined path and $\eta \to \eta^{x_{j+1},x_j}$ is the elementary exchange which constitute the jth move of the path. Therefore, term $f(\eta^{1,2,x}) - f(\eta)$ in (12) can be rewritten as

$$f(\eta^{1,2,x}) - f(\eta) = \sum_{i=1}^{n}\left(f(T_{x_{i+1},x_i}\eta_i) - f(\eta_i)\right), \tag{15}$$

where we have defined $\eta_1, \ldots, \eta_n$ as $\eta_1 \equiv \eta$, $\eta_i \equiv \eta_{i-1}^{x_i,x_{i-1}}$ and, to obtain the right hand side, we have subtracted and added each term $f(T_{x_{i+1},x_i}\eta_i)$ to

the left hand side. This procedure is usually called in the mathematical literature "telescoping a sum" and the right hand side a telescopic sum.

Then, by using (15) and Cauchy-Scharwz inequality we obtain:

$$\Big(f(\eta^{1,2,x}) - f(\eta)\Big)^2 \leqslant n \sum_{j=1}^{n} \big(f(T_{x_{j+1},x_j}\eta_j) - f(\eta_j)\big)^2 . \tag{16}$$

Since the path $\eta_1, \ldots, \eta_n$ has been chosen in order that the exchanges in the right hand side are all elementary exchanges allowed for KA process, i.e. with unit rate, we can rewrite the above inequality by introducing in the right hand side the corresponding jump rates:

$$\Big(f(\eta^{1,2,x}) - f(\eta)\Big)^2 \leqslant n \sum_{j=1}^{n} c_{x_{j+1},x_j}(\eta_j) \big(f(T_{x_j,x_{j+1}}\eta_j) - f(\eta_j)\big)^2 . \tag{17}$$

Inserting the above inequality in (12), using the change of variables $\eta_j \to \eta$ and the invariance of equilibrium measure under exchange of occupation numbers we find:

$$\begin{aligned} D_2^{\mathrm{aux}}(f,\rho) &= \frac{1}{4\mu_{\rho,0}(\mathcal{A}^x)} < \mathbb{I}_{\mathcal{A}_x}(\eta) \Big(f(\eta^{1,2,x}) - f(\eta)\Big)^2 >_0 \\ &\leqslant \frac{1}{4\mu_{\rho,0}(\mathcal{A}^x)}\ n\ \mathcal{N} \sum_{\substack{\{x,y\}\subset\Lambda \\ d(x,y)=1}} < c_{x,y}(\eta) \big(f(\eta^{x,y}) - f(\eta)\big)^2 >_0 , \end{aligned} \tag{18}$$

where we let $\mathcal{N}$ be the maximal number of times a single exchange has been used in the path ($\mathcal{N} := \max_{\{ij\}} \mathcal{N}_{ij}$ where the maximum is taken over all the couples of neighboring $\{i, j\}$ and $\mathcal{N}_{ij}$ is the number of times operator T^{ij} appears in expression (14)). We stress that $\mathcal{N}$ and n are independent on the choice of the configuration, since the path is fixed once for all and does not depend on the value of the configuration outside $\mathcal{A}_x$. From the above inequality and recalling the definition (5) we conclude that:

$$D_2(\rho, f) \geqslant \frac{\mu_{\rho,0}(\mathcal{A}^x)}{n\mathcal{N}} D_2^{\mathrm{aux}}(\rho, f) \tag{19}$$

for any ρ and f. On the other hand, since the rates of the moves for the tagged particle in the auxiliary process are always smaller or equal to the correspondent rates for KA (see moves (i) and (iii)), we get:

$$D_1(\rho, f) \geqslant \mu_{\rho,0}(\mathcal{A}^x) D_1^{\mathrm{aux}}(\rho, f) \tag{20}$$

for any ρ and f. Therefore,

$$D_S(\rho) \geqslant \frac{\mu_{\rho,0}(\mathcal{A}^x)}{n\mathcal{N}} \; D_S^{\text{aux}}(\rho) \geqslant c\mu_{\rho,0}(\mathcal{A}^x) \; D_S^{\text{aux}}(\rho) \tag{21}$$

with c a strictly positive constant and $\mu_{\rho,0}(\mathcal{A}^x) = (1 - \rho^4 - 4\rho^3(1-\rho) - 4\rho^2(1-\rho)^2)$, which is also strictly positive at any density $\rho < 1$. In particular, in the high density limit, $\mu_{\rho,0}(\mathcal{A}^x) \propto (1-\rho)^2$, hence, we finally obtain $D_S \geqslant K_L(1-\rho)^2$ with K_L a positive constant.

6. SELF-DIFFUSION COEFFICIENT FOR HIGHLY COOPERATIVE KCLG

In the following, we analyze the $s = 1$ KA model on a square lattice. This is a case in which it is impossible to identify defects with a density independent size that can freely moves inside the lattice. Thus, the key ingredient for the diffusivity proof discussed in Section 5.2.2 does not hold. This is not a simple technical difficulty but it is deeply related to the fact that diffusion takes place in a different way in this case. As it has been found in ref. 9 by analytical and numerical arguments the diffusion takes place thanks to the cooperative motion of a number of vacancies that diverges approaching unit density. This leads to an extremely rapid decreasing of the self-diffusion coefficient that, however, remains always positive if the density is less than one.

In the following, we shall prove this statement using a procedure which is very general and that can be applied to the other highly cooperative KA models as well as to many other interacting lattice particle systems. In this highly cooperative case, at variance with what done in section 5.2.2, we will not discuss the upper bound on D_S. Mainly because we do not expect that the lower bound we establish does give the right density dependence (see refs. 9 and 11).

The strategy of the proof is similar to the one discussed in Section 5.2.2.: we introduce a proper auxiliary process such that $D_S > cD_S^{\text{aux}}$ with $c > 0$ and then we prove that $D_S^{\text{aux}} > 0$. The key physical ingredient that allows us to find that such an auxiliary process exists is that (refs. 9 and 11 and Section 3) all the particle exchanges inside a finite box around the origin can be performed with a very high probability p through a suitable path in configuration space that involves particles at most at distance $\Xi(\rho)$ from the origin. We call the restricted configuration in the sublattice Λ_Ξ of size Ξ around the origin *frameable* (see Section 3). Furthermore p can be chosen arbitrary close to one taking a

suitably large $\Xi(\rho)$. Using this property we construct an auxiliary process that maps onto a random walk in a random environment and has $D_S^{aux} > 0$.

Let us explain the idea in more detail before focusing on technical details. Consider a configuration on the infinite lattice $\mathbb{Z}^2$ sorted at random with Bernoulli measure at density ρ and focus on a sub-lattice Λ_Ξ of linear size $\Xi(\rho)$. As recalled in Section 3, we know that the restriction of the configuration to Λ_Ξ is frameable with probability almost one. Therefore, in the initial configuration the tagged particle is with very high probability inside a frameable region of size Ξ. Moreover, if one divides the infinite lattice in sub-lattices of linear size Ξ, there exists with unit probability a percolating cluster of sub-lattices such that the initial configuration restricted to each sub-lattice is frameable.

Thus, roughly speaking, if we define an auxiliary process such that: (1) the tagged particle can move if it is inside a frameable square, (2) during the dynamics frameable sublattices remain frameable and the tagged particle remains always inside the percolating cluster then we can reconstruct any move of such process through a finite sequence of moves allowed by KA (indeed any nearest neighbor move in a frameable configuration of linear size Ξ can be performed through a sequence of elementary moves allowed by KA rules and the length of such path is at most of order Ξ^2)and prove inequality $D_S > cD_S^{aux}$. Furthermore since in the auxiliary process the particle moves on a percolating giant cluster then $D_S^{aux} > 0$.

6.1. Construction of the Auxiliary Diffusive Process for the KA $s = 1$ on a Square Lattice

Let us introduce some notation. Consider the following subsets of $\mathbb{Z}^2$:

$$
\begin{aligned}
R_x^{(\pm 1)} &:= \big\{(x_1 \pm e_1, x_2), (x_1 \pm e_1, x_2 + e_2)\big\},\\
R_x^{(\pm 2)} &:= \big\{(x_1 \pm e_1, x_2), (x_1 \pm e_1, x_2 - e_2)\big\},\\
R_x^{(\pm 3)} &:= \big\{(x_1, x_2 \pm e_2), (x_1 + e_1, x_2 \pm e_2)\big\},\\
R_x^{(\pm 4)} &:= \big\{(x_1, x_2 \pm e_2), (x_1 - e_1, x_2 \pm e_2)\big\},\\
Q_x^{(\pm 1)} &:= \big\{(x_1, x_2 + e_2) \cup R_x^{(\pm 1)}\big\},\\
Q_x^{(\pm 2)} &:= \big\{(x_1, x_2 - e_2) \cup R_x^{(\pm 2)}\big\},
\end{aligned}
\tag{22}
$$

R_x^i are the eight possible couples of neighboring sites $\{y, z\}$ such that $y, z \neq x$ and $|y - x| = 1$ or $|z - x| = 1$ (see Fig.4), in other words one among y and z is neighboring site to x and the couple does not contain x; Q_x^i are

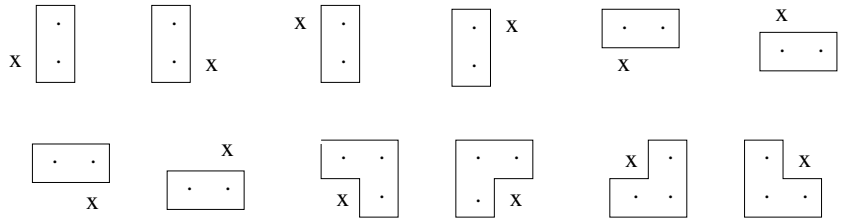

Fig. 4. Sites inside the closed line correspond, from left to right an up down, to sets $R_x^{+1}, R_x^{-1}, R_x^{+2}, R_x^{-2}, R_x^{+3}, R_x^{-3}, R_x^{+4}, R_x^{-4}, Q_x^{+1}, Q_x^{-1}, Q_x^{+2}, Q_x^{-2}$.

the four possible choices of three sites that, together with x, form a two by two square (see Fig. 4). We define $\eta^{R_x^{+i}, R_x^{-i}}$, for $i \in \{\pm 1, \ldots, \pm 4\}$ as the configuration obtained from η by exchanging the occupation numbers in R_x^{+i} with the corresponding ones in R_x^{-i}:

$$\left(\eta^{R_x^{+i}, R_x^{-i}}\right)_z := \begin{cases} \eta_{x \mp 2e_1} & \text{if} \quad z \in R_x^{\pm i}, \\ \eta_z & \text{if} \quad z \notin R_x^{+i} \cup R_x^{-i}. \end{cases} \tag{23}$$

Then, for $i \in \{\pm 1, \pm 2\}$, we introduce the events:

$$\mathcal{A}_x^i := \left\{\eta \in \Omega : \eta_z = 0 \, \forall z \in Q_x^i\right\}, \tag{24}$$

i.e. configurations having all the sites of set Q_x^i empty and,

$$\mathcal{B}_x^i := \left\{\eta \in \Omega : \eta_z = 1 \ \ \forall z \in R_x^i, \ \eta \in \mathcal{F}_x^i\right\} \tag{25}$$

for $i \in \{\pm 1, \pm 2, \pm 3, \pm 4\}$, where:

$$\mathcal{F}_x^i := \left\{\eta \in \Omega : \exists \Lambda \in \mathcal{S} : R_x^i \subset \Lambda, \ \ \delta(R_x^i, \partial \Lambda) \geqslant 3, \ \ \eta|_\Lambda \in \mathcal{F}_\Lambda\right\}, \tag{26}$$

and $\mathcal{S}$ is the set of squares in $\mathbb{Z}^2$ of linear size at most Ξ. Here, $\partial \Lambda$ is the boundary of square sublattice Λ, $\eta|_\Lambda$ is the restriction of a configuration to Λ, $\mathcal{F}_\Lambda$ is the set of frameable configuration in Λ and $\delta(A, B)$ is the minimum over the Euclidean distance of all the couples $\{x, y\}$ with $x \in A$ and $y \in B$. In other words, $\mathcal{B}_x^i$ is the set of configurations in which a pair of sites adjacent to x (region R_x^i) is filled and is internal to a frameable square of linear size at most Ξ. Let $\eta(0)$ be the configuration and $x(0)$ the position of the tagged particle at time zero, we define $\bar{\eta}(0)$ as

$$\bar{\eta}(0)_z = \begin{cases} 1 \ \text{ if } z \in Q_x^i \quad \forall i, \\ \eta_z \ \text{ otherwise.} \end{cases} \tag{27}$$

The dynamics of the auxiliary process is chosen as follows:

(i) The tagged particle can move from x to $x+e_1$. The jump has rate one if $\eta \in \mathcal{A}_x^{+1}$ and $\bar{\eta}(0) \in \mathcal{B}_x^{+1}$ or $\eta \in \mathcal{A}_x^{+2}$ and $\bar{\eta}(0)\mathcal{B}_x^{+2}$, zero otherwise.
(ii) The tagged particle can move from x to $x-e_1$. The jump has rate one if $\eta \in \mathcal{A}_x^{-1}$ and $\bar{\eta}(0) \in \mathcal{B}_x^{-1}$ or $\eta \in \mathcal{A}_x^{-2}$ and $\bar{\eta}(0) \in \mathcal{B}_x^{-2}$, zero otherwise.
(iii) The tagged particle can move from x to $x+e_2$. The jump has rate one if $\eta \in \mathcal{A}_x^{+1}$ and $\bar{\eta}(0) \in \mathcal{B}_x^{+1}$ or $\eta \in \mathcal{A}_x^{-1}$ and $\bar{\eta}(0) \in \mathcal{B}_x^{-1}$, zero otherwise.
(iv) The tagged particle can move from x to $x-e_2$. The jump has rate one if $\eta \in \mathcal{A}_x^{-2}$ and $\bar{\eta}(0) \in \mathcal{B}_x^{-2}$ or $\eta \in \mathcal{A}_x^{+2}$ and $\bar{\eta}(0) \in \mathcal{B}_x^{+2}$, zero otherwise.
(v) Configuration η can be transformed in $\eta^{R_x^{+1}, R_x^{-1}}$, namely the exchange of occupation variables in R_x^{+1} and R_x^{-1} can be performed. The move has rate one if $\eta \in \mathcal{A}_x^{+1}$ and $\bar{\eta}(0) \in \mathcal{B}_x^{-1}$ or $\eta \in \mathcal{A}_x^{-1}$ and $\bar{\eta}(0) \in \mathcal{B}_x^{+1}$, zero otherwise.
(vi) Configuration η can be transformed in $\eta^{R_x^{+2}, R_x^{-2}}$, namely the exchange of occupation variables in R_x^{+2} and R_x^{-2} can be performed. The move has rate one if $\eta \in \mathcal{A}_x^{+2}$ and $\bar{\eta}(0) \in \mathcal{B}_x^{-2}$ or $\eta \in \mathcal{A}_x^{-2}$ and $\bar{\eta}(0) \in \mathcal{B}_x^{+2}$, zero otherwise.
(vii) Configuration η can be transformed in $\eta^{R_x^{+3}, R_x^{-3}}$, namely the exchange of occupation variables in R_x^{+3} and R_x^{-3} can be performed. The move has rate one if $\eta \in \mathcal{A}_x^{+1}$ and $\bar{\eta}(0) \in \mathcal{B}_x^{-3}$ or $\eta \in \mathcal{A}_x^{+2}$ and $\bar{\eta}(0) \in \mathcal{B}_x^{+3}$, zero otherwise.
(viii) Configuration η can be transformed in $\eta^{R_x^{+4}, R_x^{-4}}$, namely the exchange of occupation variables in R_x^{+4} and R_x^{-4} can be performed. The move has rate one if $\eta \in \mathcal{A}_x^{-1}$ and $\bar{\eta}(0) \in \mathcal{B}_x^{-4}$ or $\eta \in \mathcal{A}_x^{-2}$ and $\bar{\eta}(0) \in \mathcal{B}_x^{+4}$, zero otherwise.
(ix) All other moves are not allowed.

In the following we will show that the above choice of the rates is such that the auxiliary process has a positive diffusion coefficient and any move can be reconstructed by a finite sequence of elementary moves allowed by KA, hence, $D_S > cD_{aux} > 0$.

Consider an initial configuration such that the tagged particle is inside a frameable square of size Ξ and such that all the sites in at least one of the sets $Q_{x(0)}^{(\pm 1)}, Q_{x(0)}^{(\pm 2)}$ (see Fig. 4) are empty, where $x(0)$ is the position of the tagged particle (i.e. $\eta(0) \in \mathcal{A}_x^i$ for some (i)). Then, both conditions will hold at any subsequent time. Indeed, moves (i)–(iv) are such that the tagged particle remains always inside the empty two by two square. On the other hand, moves (v)–(viii) are devised in order that the

only vacancies that are moved during the process are those which belong at time zero to this two by two square, therefore sublattices of size Ξ that are frameable at time zero remain frameable at later times.[6] The fact that moves for the auxiliary process occur always inside frameable regions of size at most Ξ implies that any move can be performed through a finite sequence of elementary moves allowed by KA. Indeed, by the properties of frameable configurations, any move inside a configuration of size Ξ can be performed by a sequence of order $O(\Xi^2)$ moves with positive rate for KA dynamics.[9,11] By using path arguments analogous to those used in Section 5 for the triangular case, it is then possible to establish inequality $D_{\mathrm{S}} \geqslant c D_{\mathrm{S}}^{\mathrm{aux}}$, with c positive. Let us shortly recall how this argument works and emphasize an important difference occurring in this case. For the triangular case we have defined an auxiliary process such that any move of the latter can be performed by a finite path of at most n nearest neighbor moves allowed for the considered model. Such path does not depend on the choice of the configuration. Then, we have rewritten each term of the variational formula (5) of $D_{\mathrm{S}}^{\mathrm{aux}}$ as a telescopic sum on the exchanges along this path. Finally, by using Cauchy-Schwartz inequality, the fact that each possible move is used at most twice in the path and by performing an exchange of variables, we concluded that $c D_{\mathrm{S}}^{\mathrm{aux}} \leqslant D_{\mathrm{S}}$ with $c > 0$. In this case we can proceed analogously, with the length n of the paths at most Ξ^2. However, the path now depends on the whole configuration inside the box of size Ξ since the sequence of allowed moves that one has to do in order to perform a pair exchange depends on the positions of the vacancies, see refs. 9 and 11. This yields in the inequality among $D_{\mathrm{S}}^{\mathrm{aux}}$ and D_{S} an overall factor $\mathcal{N} = 2^{\Xi^2}$ besides the factor due to $n = \Xi^2$. Let us explain in some detail this statement.

With a path argument analogous to the one done before, we rewrite each term in $D_{\mathrm{S}}^{\mathrm{aux}}$ corresponding to the exchange of particles in R_x^i, R_x^{-i} as a telescopic sum over allowed exchanges for KA, namely:

$$\left(f(\eta^{R_x^{+i}, R_x^{-i}}) - f(\eta) \right)^2 \leqslant \Xi^2 \sum_{j=0}^{n} c_{x_{j-1}, x_j}(\eta_j) \left(f(T_{x_{j-1}, x_j} \eta_j) - f(\eta_j) \right)^2, \tag{28}$$

where $c_{x,y}$ are the jump rate of KA model, $\eta_0 = \eta, \eta_1, \ldots, \eta_n = \eta^{R_x^{+i}, R_x^{-i}}$ is the path of allowed elementary moves which connects η to $\eta^{R_x^{+i}, R_x^{-i}}$ and such that $\eta_i = \eta_{i-1}^{x_{i-1}, x_i}$ for a couple of nearest neighbors $\{x_i, x_{i-1}\}$. To obtain above inequality we used Cauchy-Schwartz inequality and the

[6] More precisely, sublattices that are frameable in $\bar{\eta}(0)$ are frameable also for $\eta(t)$ at any t.

fact that $n \leqslant \Xi^2$. In order to obtain from left and right hand side the terms which appear in the variational formula (5) for the auxiliary process and for KA, respectively, we should average inequality (28) over the Bernoulli measure conditioned to have a particle in zero. As we already emphasized, the sequence $x_0, \ldots, x_n$ of sites in which the exchange is done, depends on the positions of vacancies in configuration η. Therefore, if we do the change of variable $\eta_i \to \eta$ in (28) and use the invariance of measure under exchange of variables, many different terms on the left can give rise to the same term on the right. Actually, the crucial thing to know is the following. To each configuration η for which the exchange is allowed by the auxiliary process, associate the correspondent path $\eta_0, \ldots, \eta_n$ in configuration space.[7] Then, for each elementary nearest neighbor exchange e, denote by $\mathcal{N}_e$ the number of different configurations η that use such exchange and let $\mathcal{N} \equiv max_e \, \mathcal{N}_e$. Therefore, $\mathcal{N}$ is the overall factor coming from possible overcounting of configuration when going from the mean of the left hand side of (28) to the terms in the variational formula (5) for D_S. Physically $\mathcal{N}$ takes into account the most severe dynamical bottleneck in phase space (see ref. 9). Moreover, since each path is composed of moves internal to the frameable region of size $\ell \leqslant \Xi$ which contains the tagged particle, $\mathcal{N}$ is for sure less or equal to the total number of configurations inside a square of size Ξ, thus $\mathcal{N} \leqslant 2^{\Xi^2}$. Therefore, we finally obtain, closely enough to unit density:

$$D_S \geqslant c(1-\rho)^3 \quad \mu_{\rho,\Xi}(\mathcal{F}) \quad \frac{1}{\Xi^4 2^{\Xi^2}} \quad D_S^{\text{aux}}, \tag{29}$$

where c is a positive constant. The term $(1-\rho^3)\mu_{\rho,\Xi}(\mathcal{F})$ comes from the condition that the configuration at time zero should have the tagged particle with three vacancies around and be inside a frameable square of size at most Ξ and D_S^{aux} is the diffusion coefficient of the auxiliary process subject to this condition. This ends the first part of the proof. In Section 6.2 we shall show that indeed $D_S^{\text{aux}} > 0$.

[7] Of course there could be different sequences to perform the same move. However, one can always give a prescription associating one of them for any choice of η and any give exchange.

6.2. Lower Bound for the Self-Diffusion Coefficient of the Auxiliary Process

Let us now prove that $D_S^{\text{aux}} > 0$, i.e. that diffusivity holds for the auxiliary process. In this case, the mechanism which guarantees diffusivity is different from the one discussed in Section 5. Analogously to the KA on a triangular lattice (Section 5), the auxiliary process we have just introduced is such that if the tagged particle is at time zero in a two by two square of vacancies (i.e. $\eta(0) \in \mathcal{A}^i_{x(0)}$ for some i) and inside a larger frameable square of size at most Ξ, both conditions will be always fulfilled at later times. However, now it is not true that the tagged particle can always be moved in a chosen direction e_i through a proper path. For example, if we want to move it in direction e_1 this is possible only if $\eta \in \mathcal{A}_x^{+1}$ or $\eta \in \mathcal{A}_x^{+2}$. Otherwise, if $\eta \in \mathcal{A}_x^{-1}$ or $\eta \in \mathcal{A}_x^{-2}$, the move is allowed only if before one makes the exchange $\eta \to \eta^{R_x^{+1}, R_x - 1}$ or $\eta \to \eta^{R_x^{+2}, R_x - 2}$, respectively. However, these exchanges of rectangles (which are the analogous of exchange $\eta \to \eta^{x-e_i, x+e_i}$ for RLG) are not always allowed. Indeed (see rules (v)–(viii)) they have positive rate only if in the initial configuration the rectangular regions R_x^{-1}, respectively, R_x^{-2}, do not contain vacancies and are inside a frameable square of size at most Ξ. Note that the rate of such exchanges (i.e. the rate of the exchange $\eta \to \eta^{R_x^{+1}, R_x - 1}$ and $\eta \to \eta^{R_x^{+2}, R_x - 2}$ conditioned to the fact that $\eta \in \mathcal{A}_x^{-1}$ or $\eta \in \mathcal{A}_x^{-2}$, respectively) does not depend on the configuration η, but is fixed once for all by the choice of the initial configuration $\eta(0)$. In other words, the choice of the initial configuration fixes the *good* rectangles that can be exchanged. This observation will allow us to map the motion of the two by two square of three vacancies plus tagged particle to a random walk in a random environment corresponding to the cluster of good rectangles. We emphasize that this cluster does not change during dynamics, therefore the randomness of the environment is *quenched*. Note that the probability p_g for a given rectangle to be good is greater than ρ^2 (the probability that both the sites inside the rectangle are occupied) multiplied for the probability that it is inside a frameable region of size at most Ξ which is almost one (this is the key physical ingredient). Therefore, in the high density regime p_g is well above the threshold of conventional site percolation. This implies that with unit probability the initial configuration has a percolating cluster of good rectangles. By using that above the percolation threshold random walk on random environment has a positive diffusion coefficient[20] we will therefore obtain that the diffusion coefficient of the auxiliary process is strictly positive. In the following, we will sketch the proof of the above argument in some detail.

Let $\eta_{(0,0)} = \eta(0)$ be the initial configuration. Let us define the following sequence of configurations $\eta_{(m,n)}$ for $(m,n) \in \mathbb{Z}^2$:

$$\eta_{(m+1,n)} \begin{cases} \eta_{(m,n)}^{x,x+e_1} & \text{if } \eta_{(m,n)} \in \mathcal{A}_x^{+1}, \quad \bar{\eta}(0) \in \mathcal{B}_x^{+1}, \\ \eta_{(m,n)}^{x,x+e_1} & \text{if } \eta_{(m,n)} \in \mathcal{A}_x^{+2}, \quad \bar{\eta}(0) \in \mathcal{B}_x^{+2}, \\ \eta_{(m,n)}^{R_x^{+1},R_x^{-1}} & \text{if } \eta_{(m,n)} \in \mathcal{A}_x^{-1}, \quad \bar{\eta}(0) \in \mathcal{B}_x^{+1}, \\ \eta_{(m,n)}^{R_x^{+2},R_x^{-2}} & \text{if } \eta_{(m,n)} \in \mathcal{A}_x^{-2}, \quad \bar{\eta}(0) \in \mathcal{B}_x^{+1}, \end{cases} \tag{30}$$

$$\eta_{(m,n+1)} \begin{cases} \eta_{(m,n)}^{x,x+e_2} & \text{if } \eta_{(m,n)} \in \mathcal{A}_x^{+1}, \quad \bar{\eta}(0) \in \mathcal{B}_x^{+3}, \\ \eta_{(m,n)}^{x,x+e_2} & \text{if } \eta_{(m,n)} \in \mathcal{A}_x^{-1}, \quad \bar{\eta}(0) \in \mathcal{B}_x^{+4}, \\ \eta_{(m,n)}^{R_x^{+3},R_x^{-3}} & \text{if } \eta_{(m,n)} \in \mathcal{A}_x^{+2}, \quad \bar{\eta}(0) \in \mathcal{B}_x^{+3}, \\ \eta_{(m,n)}^{R_x^{+4},R_x^{-4}} & \text{if } \eta_{(m,n)} \in \mathcal{A}_x^{-2}, \quad \bar{\eta}(0) \in \mathcal{B}_x^{+4}, \end{cases} \tag{31}$$

$$\eta_{(m-1,n)} \begin{cases} \eta_{(m,n)}^{x,x-e_1} & \text{if } \eta_{(m,n)} \in \mathcal{A}_x^{-1}, \quad \bar{\eta}(0) \in \mathcal{B}_x^{-1}, \\ \eta_{(m,n)}^{x,x-e_1} & \text{if } \eta_{(m,n)} \in \mathcal{A}_x^{-2}, \quad \bar{\eta}(0) \in \mathcal{B}_x^{-2}, \\ \eta_{(m,n)}^{R_x^{+1},R_x^{-1}} & \text{if } \eta_{(m,n)} \in \mathcal{A}_x^{+1}, \quad \bar{\eta}(0) \in \mathcal{B}^{-1}, \\ \eta_{(m,n)}^{R_x^{+2},R_x^{-2}} & \text{if } \eta_{(m,n)} \in \mathcal{A}_x^{+2}, \quad \bar{\eta}(0) \in \mathcal{B}^{-2}, \end{cases} \tag{32}$$

$$\eta^{(m,n-1)} \begin{cases} \eta_{(m,n)}^{x,x-e_2} & \text{if } \eta_{(m,n)} \in \mathcal{A}_x^{+2}, \quad \bar{\eta}(0) \in \mathcal{B}_x^{-3}, \\ \eta_{(m,n)}^{x,x-e_2} & \text{if } \eta_{(m,n)} \in \mathcal{A}_x^{-2}, \quad \bar{\eta}(0) \in \mathcal{B}_x^{-4}, \\ \eta_{(m,n)}^{R_x^{+3},R_x^{-3}} & \text{if } \eta_{(m,n)} \in \mathcal{A}_x^{+1}, \quad \bar{\eta}(0) \in \mathcal{B}^{-3}, \\ \eta_{(m,n)}^{R_x^{+4},R_x^{-4}} & \text{if } \eta_{(m,n)} \in \mathcal{A}_x^{-1}, \quad \bar{\eta}(0) \in \mathcal{B}^{-4}, \end{cases} \tag{33}$$

where $x = x(m,n)$ is the position of the tagged particle in configuration $\eta_{(m,n)}$ (we drop the dependence of x on m and n to get a more readable notation). Note that given $\eta_{(m,n)}$ and $x(m,n)$ for a couple (m,n), using the above definitions one can reconstruct the whole sequence.

Let us define a Markov process on $\mathbb{Z}^2$ with generator G acting on functions $f:(m,n)\to\mathbb{R}$ as

$$\begin{aligned} Gf(m,n) := & \sum_{i=\pm 1} \lambda_i^1(m,n)\,(f(m+i,n)-f(m,n)) \\ & + \sum_{i=\pm 1} \lambda_i^2(m,n)\,(f(m,n+i)-f(m,n))\,, \end{aligned} \tag{34}$$

namely a two-dimensional random walk with rates $\lambda^i_{\pm 1}$ for the jump in direction $\pm e_i$. We can now chose these rates in order that $\eta_{(m(t),n(t))} = \eta(t)$ for the auxiliary process. More precisely, we chose the rates in order that for any function $f(\eta)$ the expectation value over the probability μ_t evoluted with the generator of the auxiliary process coincides with the expectation over the measure on $(m(t),n(t))$ generated by (34). By considering the dynamics of the auxiliary process and definition (34), one can directly check that the choice of $\lambda^i_{\pm 1}$ which satisfies above requirement is the following:

$$\begin{aligned} \lambda^1_{+1}(m,n) = & \mathbb{1}_{\mathcal{A}_x^{+1}}(\eta_{(m,n)})\mathbb{1}_{\mathcal{B}_x^{+1}\cap\mathcal{B}_x^{+2}}(\eta_{(0,0)}) \\ & + \mathbb{1}_{\mathcal{A}_x^{+2}}(\eta_{(m,n)})\mathbb{1}_{\mathcal{B}_x^{+1}\cap\mathcal{B}_x^{+2}}(\eta_{(0,0)}) \\ & + \mathbb{1}_{\mathcal{A}_x^{-1}}(\eta_{(m,n)})\mathbb{1}_{\mathcal{B}_x^{+1}}(\eta_{(0,0)}) \\ & + \mathbb{1}_{\mathcal{A}_x^{-2}}(\eta_{(m,n)})\mathbb{1}_{\mathcal{B}_x^{+2}}(\eta_{(0,0)}), \end{aligned} \tag{35}$$

$$\begin{aligned} \lambda^1_{-1}(m,n) = & \mathbb{1}_{\mathcal{A}_x^{-1}}(\eta_{(m,n)})\mathbb{1}_{\mathcal{B}_x^{-1}\cap\mathcal{B}_x^{-2}}(\eta_{(0,0)}) \\ & + \mathbb{1}_{\mathcal{A}_x^{-2}}(\eta_{(m,n)})\mathbb{1}_{\mathcal{B}_x^{-1}\cap\mathcal{B}_x^{-2}}(\eta_{(0,0)}) \\ & + \mathbb{1}_{\mathcal{A}_x^{+1}}(\eta_{(m,n)})\mathbb{1}_{\mathcal{B}_x^{-1}}(\eta_{(0,0)}) \\ & + \mathbb{1}_{\mathcal{A}_x^{+2}}(\eta_{(m,n)})\mathbb{1}_{\mathcal{B}_x^{-2}}(\eta_{(0,0)}), \end{aligned} \tag{36}$$

$$\begin{aligned} \lambda^2_{+1}(m,n) = & \mathbb{1}_{\mathcal{A}_x^{2}}(\eta_{(m,n)})\mathbb{1}_{\mathcal{B}_x^{+3}\cap\mathcal{B}_x^{+4}}(\eta_{(0,0)}) \\ & + \mathbb{1}_{\mathcal{A}_x^{-1}}(\eta_{(m,n)})\mathbb{1}_{\mathcal{B}_x^{+3}\cap\mathcal{B}_x^{+4}}(\eta_{(0,0)}) \\ & + \mathbb{1}_{\mathcal{A}_x^{2}}(\eta_{(m,n)})\mathbb{1}_{\mathcal{B}_x^{+3}}(\eta_{(0,0)}) \\ & + \mathbb{1}_{\mathcal{A}_x^{-2}}(\eta_{(m,n)})\mathbb{1}_{\mathcal{B}_x^{+4}}(\eta_{(0,0)}), \end{aligned} \tag{37}$$

$$\begin{aligned} \lambda^2_{-1}(m,n) = & \mathbb{1}_{\mathcal{A}_x^{2}}(\eta_{(m,n)})\mathbb{1}_{\mathcal{B}_x^{-3}\cap\mathcal{B}_x^{-4}}(\eta_{(0,0)}) \\ & + \mathbb{1}_{\mathcal{A}_x^{-2}}(\eta_{(m,n)})\mathbb{1}_{\mathcal{B}_x^{-3}\cap\mathcal{B}_x^{-4}}(\eta_{(0,0)}) \\ & + \mathbb{1}_{\mathcal{A}_x^{+1}}(\eta_{(m,n)})\mathbb{1}_{\mathcal{B}_x^{-3}}(\eta_{(0,0)}) \\ & + \mathbb{1}_{\mathcal{A}_x^{-1}}(\eta_{(m,n)})\mathbb{1}_{\mathcal{B}_x^{-4}}(\eta_{(0,0)}). \end{aligned} \tag{38}$$

Let us define also

$$\begin{aligned}
\bar{\lambda}^1_{+1}(m,n) &= \mathbb{1}_{\mathcal{B}_x^{+1}\cap\mathcal{B}_x^{+2}}(\eta_{(0,0)}),\\
\bar{\lambda}^1_{-1}(m,n) &= \mathbb{1}_{\mathcal{B}_x^{-1}\cap\mathcal{B}_x^{-2}}(\eta_{(0,0)}),\\
\bar{\lambda}^1_{+1}(m,n) &= \mathbb{1}_{\mathcal{B}_x^{+3}\cap\mathcal{B}_x^{+4}}(\eta_{(0,0)}),\\
\bar{\lambda}^1_{+1}(m,n) &= \mathbb{1}_{\mathcal{B}_x^{-3}\cap\mathcal{B}_x^{-4}}(\eta_{(0,0)}),
\end{aligned} \tag{39}$$

and

$$\begin{aligned}
\bar{G}f(m,n) := &\sum_{i=\pm1}\bar{\lambda}^1_i(m,n)\,(f(m+i,n)-f(m,n))\\
&+\sum_{i=\pm1}\bar{\lambda}^2_i(m,n)\,(f(m,n+i)-f(m,n))\,.
\end{aligned} \tag{40}$$

From definitions (35) and (39) it is immediate to check that $\lambda^i_{\pm1}\geqslant\bar{\lambda}^i_{\pm1}$. Because the variational formula (5) implies that the self-diffusion coefficient is a monotonic increasing function of the jump rates we find that D_S for the random walk process with rates $\lambda^i_{\pm1}$ is certainly larger than the one for the process with rates $\bar{\lambda}^i_{\pm1}$. By recalling definition (25) and results on crossover length Ξ, we know that for sufficiently high density the probability w.r.t. Bernoulli measure of events:

$$\begin{aligned}
&\mathcal{B}_x^{+1}\cap\mathcal{B}_x^{+2},\\
&\mathcal{B}_x^{-1}\cap\mathcal{B}_x^{-2},\\
&\mathcal{B}_x^{+3}\cap\mathcal{B}_x^{+4},\\
&\mathcal{B}_x^{-3}\cap\mathcal{B}_x^{-4},
\end{aligned}$$

is almost one, and therefore greater then threshold probability for conventional percolation on the square lattice. Hence, by using the result in ref. 20 which establishes a central limit theorem for random walk in random environment when bond probability is greater than percolation threshold we find that D_S for the process with rates $\bar{\lambda}^i_{\pm1}$ (and therefore D_S for the process with rates $\lambda^i_{\pm1}$) is strictly positive.[8] Moreover, since when m goes

[8]Note that the percolation problem we consider is a site percolation problem in which the site probability is correlated over a *finite* distance equal to $2\Xi(\rho)$ whereas in ref. 20 it was analyzed only the case with independent bond probability. However, we expect that their result can be generalized to our case. Furthermore from the physical point of view there is no doubt that D_S will be positive in our case of correlated site disorder whenever a giant cluster exists (except at the critical point).

to $m+2$ (n to $n+2$) the first (second) coordinate of the tagged particle position increases at least of one unit, we obtain that

$$\mu_t \left(x_1(t)^2 + x_2(t)^2 \right) \geqslant \frac{1}{4} E \left(m(t)^2 + n(t)^2 \right) > ct, \tag{41}$$

where μ_t is the evoluted of initial measure $\mu_{\rho,0}$ under the auxiliary process and c a positive constant. This allows us to conclude that $D_{\mathrm{S}}^{\mathrm{aux}} > 0$ at any $\rho < 1$ which, together with inequality (29), implies $D_{\mathrm{S}} > 0$ for KA model.

7. CONCLUSION

In this work, we present a general procedure, focusing on the KA model as an example, that allows one to prove lower and upper bounds on interacting particle systems on a lattice in the case of vanishing rates. This is a generalization of the Spohn's proof[5,6] for RLG. In particular, focusing on KA $s=1$ model on a triangular lattice as an example, we show how to obtain the exact density dependence of D_{S} in the high density limit for KCLG with finite size defects (see also ref. 12). Whereas for highly cooperative KCLG, i.e. when the size of the defects diverges approaching unit density, our procedure allows to prove diffusivity, $D_{\mathrm{S}} > 0$, at any density smaller than one as we have shown for the KA $s=1$ model on a square lattice.

As we have stressed previously our method is completely general and can be applied to general (short-range) interacting particle systems on a lattice, including systems with a non-trivial equilibrium measure as for example the statically constrained models introduced in ref. 8. In order to be successful, it just needs one crucial key physical property. One has to know that, for a random equilibrium configuration, all the particle exchanges inside a finite box around a fixed site, say the origin, can be performed with a very high probability p through a suitable path in configuration space (allowed by the kinetic rules) that involves particles at most at distance Ξ from the origin. Furthermore, one needs also that p can be chosen arbitrary close to one taking a suitably large Ξ.

This property is certainly valid if, given two equilibrium configurations taken at random inside a box of size L, one can show that there exists with probability p' at least a path in configuration space, allowed by the kinetic rules, that connects the two configurations and that $\lim_{L\to\infty} p' = 1$.[9] Note that is just equivalent to say that the system

[9]The boundary condition for the box has to be chosen such that it is the worst possible in order to find the path connecting two configurations. For example for the KA one would choose boundary conditions that are equivalent to embed the box in a completely filled lattice.

at hand is irreducible in the thermodynamic limit. More correctly, in the thermodynamic limit one irreducible component Ω covers all the configuration space, i.e. an equilibrium configuration taken at random belongs to Ω with probability one. Therefore, our results, combined with the ones obtained for Brownian interacting particle systems,[14] strongly suggest that the only case in which a dynamical arrest (at which D_S vanishes at a finite temperature/chemical potential) might happen is only when a irreducible–reducible transition takes place (for an infinite system) and that one has to identify the two types of transition.

To our knowledge, none of the short-range interacting particle systems which have been considered so far (on a lattice as well as in the continuum) has been proved to display such reducibility transition in dimension larger than one.

Finally, we want to stress again that the non-vanishing of the self-diffusion coefficient D_S does not imply at all that the structural relaxation time scale τ_α cannot diverge. In particular, as discussed previously, at a second order phase transition the structural relaxation time scale diverges whereas the self-diffusion coefficient stays finite. Thus, the decoupling between D_S and $1/\tau_\alpha$ is *a necessary condition for the existence of an ideal glass transition* taking place at finite temperature and chemical potential. In experiments on fragile liquids[21] a decoupling is indeed observed between D_S and $1/\tau_\alpha$ (more correctly between D_S and the viscosity η) but not as strong as one would have very close to a phase transition, i.e. $D_S \tau_\alpha \propto \tau_\alpha$.

ACKNOWLEDGMENTS

It is pleasure to thank Daniel S. Fisher for all the discussions that we had on the subject of this work and, more generally, on glassy systems. We are also grateful to Lorenzo Bertini for very useful discussions.

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