

This is an Open Access document downloaded from ORCA, Cardiff University's institutional repository:<https://orca.cardiff.ac.uk/id/eprint/177683/>

This is the author's version of a work that was submitted to / accepted for publication.

Citation for final published version:

Lam, Leo S. I., Deng, Haiyao , Zhang, Wei-Bing, Nwankwo, Udoka, Xiao, Chu, Yip, Cho-Tung, Lee, Chun-Shing, Ruan, Haihui and Lam, Chi-Hang 2025. Emergent facilitation by random constraints in a facilitated random walk model of glass. *Physical Review E (statistical, nonlinear, biological, and soft matter physics)* 111 (4) , 044120. 10.1103/PhysRevE.111.044120

Publishers page: <http://dx.doi.org/10.1103/PhysRevE.111.044120>

Please note:

Changes made as a result of publishing processes such as copy-editing, formatting and page numbers may not be reflected in this version. For the definitive version of this publication, please refer to the published source. You are advised to consult the publisher's version if you wish to cite this paper.

This version is being made available in accordance with publisher policies. See <http://orca.cf.ac.uk/policies.html> for usage policies. Copyright and moral rights for publications made available in ORCA are retained by the copyright holders.



Emergent facilitation by random constraints in a facilitated random walk model of glass

Leo S.I. Lam¹, Hai-Yao Deng², Wei-Bing Zhang³, Udoka Nwankwo¹, Chu

Xiao⁴, Cho-Tung Yip⁴, Chun-Shing Lee^{4,*}, Haihui Ruan^{1,†} and Chi-Hang Lam^{5‡}

¹*Department of Mechanical Engineering, Hong Kong Polytechnic University, Hong Kong, China*

²*School of Physics and Astronomy, Cardiff University, 5 The Parade, Cardiff CF24 3AA, Wales, UK*

³*School of Physics and Electronic Sciences, Changsha University of Science and Technology, Changsha 410004, China.*

⁴*Department of Physics, Harbin Institute of Technology, Shenzhen 518055, China*

⁵*Department of Applied Physics, Hong Kong Polytechnic University, Hong Kong, China*

(Dated: December 13, 2024)

The physics of glass has been a significant topic of interest for decades. Dynamical facilitation is widely believed to be an important characteristic of glassy dynamics, but the precise mechanism is still under debate. We propose a lattice model of glass called the facilitated random walk (FRW). Each particle performs continuous time random walk in the presence of its own random local kinetic constraints. The particles do not interact energetically. Instead, they interact kinetically with a hopping rate resampling rule under which motions of a particle can randomly perturb the local kinetic constraints of other particles. This dynamic interaction is reversible, following a rate restoration rule. A step-by-step reversal of the particle motions exactly restore the previous constraints, modeling randomness quenched in the configuration space of glass. The model exhibits stretched exponential relaxation and dynamical heterogeneity typical of glasses. Despite the lack of explicit facilitation rule, the FRW shows facilitation behaviors closely analogous to those of the kinetically constrained models (KCM). The FRW is a coarse-grained version of the distinguishable particle lattice model (DPLM) and this exemplifies that compatible defect and atomistic models can complement each other on the study of glass.

I. INTRODUCTION

The understanding of the dominant modes of relaxation dynamics in glassy materials have been debated actively for decades [1–6]. Molecular dynamics (MD) simulations are instrumental in their studies as they provide complete information on the atomistic details of motions [7, 8]. Besides more realistic all-atom simulations, simplified coarse-grained models are widely believed to capture essential properties of glass. Nevertheless, due to the extremely slow dynamics of glasses, long-time relaxation dynamics in MD simulations at deep supercooling relevant to experimental conditions are still computationally inaccessible, despite the availability of equilibrium samples using advanced swap algorithms. The dominant relaxation dynamics of glass can be hidden behind vibrational and other liquid-like collective motions which diminish at deeper supercooling.

Lattice models play pivotal roles in many branches of statistical physics [9, 10]. Many phenomena can be qualitatively reproduced and certain characteristic quantities, such as scaling exponents, are in agreement with values from off-lattice simulations and experiments. Many lattice models for glass are successful in reproducing key features such as kinetic arrest and dynamical heterogeneity [11, 12]. There are however great challenges to justify assumptions in most of these models. In addition, many

glassy features may not be readily reproduced.

The n -spin facilitated model proposed by Fredrickson and Andersen (FA) [13, 14] is one of the first kinetically constrained models (KCM) [11]. It is a coarse-grained defect model of glass. An up-spin represents a defect in the form of a local region with low particle density. The flipping of a spin can only be facilitated by the presence of at least n neighboring up-spins. The dynamical facilitation picture pioneered by the FA and related models [13–17] have been gaining further support very recently [18, 19].

There have been proposal of new lattice models or variants attempting to better capture the essence of glass [20–24]. In particular, the distinguishable particle lattice model (DPLM) [20] of glass is defined by particle pair interactions and void-induced dynamics. The DPLM is an atomistic model in which each particle represents an atom or a rigidly bounded group of atoms. Voids correspond to missing particles and they are simplified representations of quasivoids found in supercooled liquids [25]. The model demonstrates emergent facilitation behaviors and has reproduced an extraordinarily wide range of kinetic and thermodynamic characteristics of glass [20, 26–35].

In this paper, we propose a facilitated random walk (FRW) model of glass. It is a coarse-grained and energetically trivialized version of the DPLM. It is a defect model with particles corresponding to voids in glass [25, 36, 37]. The FRW may open up a new class of KCM with random constraints, which can be more readily justifiable than the usual deterministic kinetic constraint rules [11]. We will explain emergent facilitation behaviors in which

* Email: anakin-cs@hotmail.com

† Email: haihui.ruan@polyu.edu.hk

‡ Email: C.H.Lam@polyu.edu.hk

dynamics are dominated by coupled particle groups with a group size depending on the constraint density. The facilitated dynamics in such mobile groups is analogous to and justifies the facilitation rule in the FA model.

The FRW model is a generalization of simpler random walk models which will first be summarized. Additional rules specific to FRW will be explained in the next section. Only one-dimensional (1D) systems will be discussed but generalization to higher dimensions is straightforward.

Continuous-time random walk (CTRW): Consider N particles on a one dimensional lattice with L sites under periodic boundary conditions. No exclusion rule is imposed such that any site i can contain $n_i = 0, 1, 2, \dots$ particles. The particle density is $\rho = N/L$, which can be varied from 0 to ∞ .

For simple CTRW with no kinetic constraint, every particle can hop to a given nearest neighboring (NN) site with a rate w_0 . With two hopping pathways to the two NN sites in 1D, the combined hopping rate of a particle to an arbitrary NN site is thus $2w_0$.

CTRW with quenched constraints: We now impose a distinct set of local kinetic constraints for each particle by randomly blocking some of the hopping pathways. Specifically, particle k hops from site i to a NN site j with a rate w_{ijk} , given by $w_{ijk} = w_0$ with an unblocking probability q and $w_{ijk} = 0$ otherwise. We put $w_{ijk} = w_{jik}$ in order to follow detailed balance. Otherwise, each w_{ijk} is an independent and quenched random variable sampled at the beginning of a simulation. For $q = 1$, simple CTRW is restored. For $q < 1$, every particle is locally trapped within a finite well of its own. With neither exclusion principle nor energetic interaction, particle motions are independent of each other and are trivially solvable. As the randomness in the constraints is quenched in the real space, the disorder in this model is, however, not appropriate for glass.

The rest of the paper is organized as follows. In Sec. II, we will generalize the simple models above to the FRW model. Section III explains its exact equilibrium statistics and the kinetic Monte Carlo simulation algorithm used. In Sec. IV, we demonstrate standard glassy behaviors of FRW including stretched exponential relaxations. Section V explains intuitively the emergent facilitation mechanism and the resulting ergodic property. We then conclude in Sec. VI with further discussions.

II. MODEL

The FRW model is a coarse-grained defect model of glass, similar to the FA model [13]. Each site represents a mesoscopic region with, for example, dozens of particles. A particle in FRW physically represents a defect in the form of a void [25]. Atoms or molecular units in the glass are not explicitly simulated. All results are averaged over at least two simulations.

Specifically, the model in 1D is constructed based on

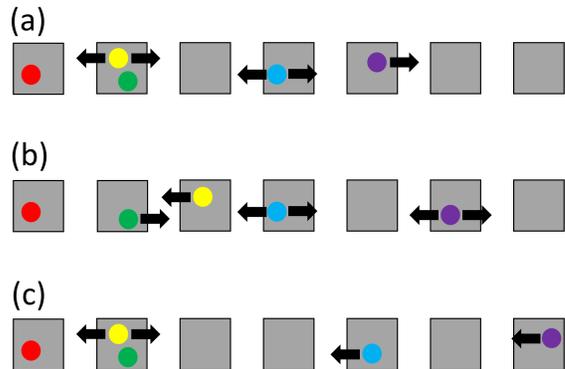


FIG. 1. A schematic showing an example of system evolution in FRW for a lattice of $L = 7$ sites (grey squares) with $N = 5$ particles (colored circles). An arrow, occurring with an unblocking probability $q = 0.5$, indicates that the particle is able to hop at rate w_0 in the corresponding direction. Starting from the initial configuration (a), the hop of the yellow particle has led to a rate resampling of the right-hopping rate of the green particle (b). In (c), the reversed hop of the yellow particle has restored the previous right-hopping rate of the green particle. In (a)-(c), note that all hops are reversible due to detailed balance.

CTRW with random constraints in the absence of particle exclusion. Similar to the descriptions in Sec. I, we consider a lattice size L , lattice constant $a = 1$, total number of particles N , density $\rho = N/L$, and occupancy $n_i = 0, 1, 2, \dots$ at site i . As before, the random constraints are such that the hopping rate $w_{ijk}(t)$ of particle k from site i to a NN site j depends on the unblocking probability q and follows

$$w_{ijk}(t) = \begin{cases} w_0 & \text{probability } q \\ 0 & \text{otherwise.} \end{cases} \quad (1)$$

with $w_0 = 1$ and they obey the detailed balance condition

$$w_{ijk}(t) = w_{jik}(t). \quad (2)$$

For the FRW model, we emphasize however that the hopping rate $w_{ijk}(t)$ is *not* quenched but can be reversibly resampled, i.e. resampled or restored, repeatedly throughout a simulation. The rules for this time dependence are the defining characteristics of FRW and are defined as follows:

- (i) **Rate resampling:** If particle k hops from site i to j , we will resample the hopping rate $w_{ijl}(t) = w_{jil}(t)$ across bond ij for all other particles $l \neq k$ using Eq. (1).
- (ii) **Rate restoration:** After particle k has hopped from site i to j , if it performs a reversed hop from j to i , we will restore the previous values of $w_{ijl}(t) = w_{jil}(t)$ across bond ij for all particles $l \neq k$.

Figure 1 shows an example of particle configuration and evolution.

The resampling rule introduces a form of dynamical interactions, in the absence of any energetic interaction. It turns the FRW into a correlated-particle model and enables rich dynamical behaviors. Physically, it models the mechanism that the motion of particle k , representing a defect, perturbs the local packing of the glass atoms at the sites. This alters the energetically favorable local hopping pathways of every other defect l , although the effect is felt only when l arrives at the sites.

On the other hand, the rate restoration rule implements randomness quenched in the configuration space but not in the real space. It physically represents that when defect k reverses its previous hop, the local atomic packing of the glass is reversed so that the previous local energetically favorable pathways of every other defect l are restored.

In general, the restoration rule can be applied to any arbitrarily long sequence of hops. If the hops are reversed step-by-step in the reversed order, the rates will be step-by-step restored. Nevertheless, in a large system, groups of particles that are well separated at all times should not affect each other. More precisely, the restoration rule applies to a whole correlated sequence of hops. It does not require reversing any other uncorrelated hop at a distance which has no impact on the rates of hops in the sequence.

III. EQUILIBRIUM STATES AND SIMULATION ALGORITHM

The FRW is a kinetic model with trivial energetics. The detailed balance condition (Eq. (2)) implies that all accessible states have identical occurrence probability in an equilibrium ensemble, implying the same system energy at all states. Furthermore, the model is ergodic in the large system size limit, as will be explained in Sec. V. As a result, the equilibrium states of FRW is trivial and can be described exactly as follows. At equilibrium, all particle arrangements are possible and are equally likely. To initialize an equilibrium system, we simply need to put each particle randomly and independently on the lattice.

We perform FRW simulations using a rejection free Monte Carlo algorithm. During a simulation, we continuously keep track of all unblocked hopping directions with rate w_0 of all particles. At every time step, we randomly choose one of these hops with a uniform probability. To choose efficiently, we search a binary tree which dynamically represents all available hops. Then, the hop is performed and the simulation time advances by $(Mw_0)^{-1}$ where M is the total number of unblocked hopping directions of all particles.

A potentially challenging part of the algorithm is related to the hopping rate restoration rule, which recalls previous rates after an arbitrarily long hopping sequence is step-by-step reversed. To enable such restoration,

one could employ a memory intensive algorithm that involves saving the history of all previous states and hopping rates into the computer memory. However, this method requires a nontrivial data structure design and prohibitively large amount of memory for long runs.

To solve this rate restoration problem, we have developed a reversible pseudo random number based algorithm so that all required rates are calculated from random number sequences. The random number generators thus encode and effectively store all possible rates, only the current system state needs to be directly stored in the computer memory.

The idea of the rate restoration algorithm is as follows. The FRW system state is primarily characterized by the positions i_k of every particle k . For bookkeeping purpose, we introduce fictitious internal states Ψ_k and Φ_{ij} of particle k and bond ij . These internal states are represented by 64-bits random numbers so that no two states will be accidentally equal in practical simulations. A hop involves updating Ψ_k and Φ_{ij} essentially to the next numbers the pseudo random number sequences. At a reversed hop, previous states are obtained by updating to the previous numbers in the sequences, which are readily calculable because we adopt reversible generators. The hopping rate $w_{ijk}(t)$ is defined as a function of the instantaneous states Ψ_k and Φ_{ij} so that previous rates can be readily restored once the previous states Ψ_k and Φ_{ij} are restored. Using this memory-less algorithm, we are able to run the simulation for long time without any memory concerns while maintaining exactly the rate restoration property. This algorithm is explained in detail in Appendix A and III.

We have extensively tested the reliability of our software implementation of these algorithms. In particular, we have checked that the known equilibrium states are arrived at and maintained throughout the kinetic Monte Carlo simulations. Furthermore, a highly non-trivial test is the quantitative verification of an exact mobility threshold for two-particle systems, as will be reported elsewhere.

IV. GLASSY CHARACTERISTICS

We now explain FRW simulation results, demonstrating typical glassy characteristics.

Dynamical facilitation: Position-time graphs in Fig. 2 illustrate particle motions in equilibrium FRW systems for an unblocking probability $q = 0.5$ under different particle density ρ . In Fig. 2(a) with a low density $\rho = 0.05$, particles are mostly isolated and they are all permanently trapped within small traps. Coupled pairs of particles have movements confined to slightly wider traps. We will explain in Sec. V that such permanent trapping is only a finite size effect.

In Fig. 2(b) where the density is doubled to $\rho = 0.1$, a mobile group, usually consisting of 3 particles, traverses over much of the system and can indeed travel unbound-

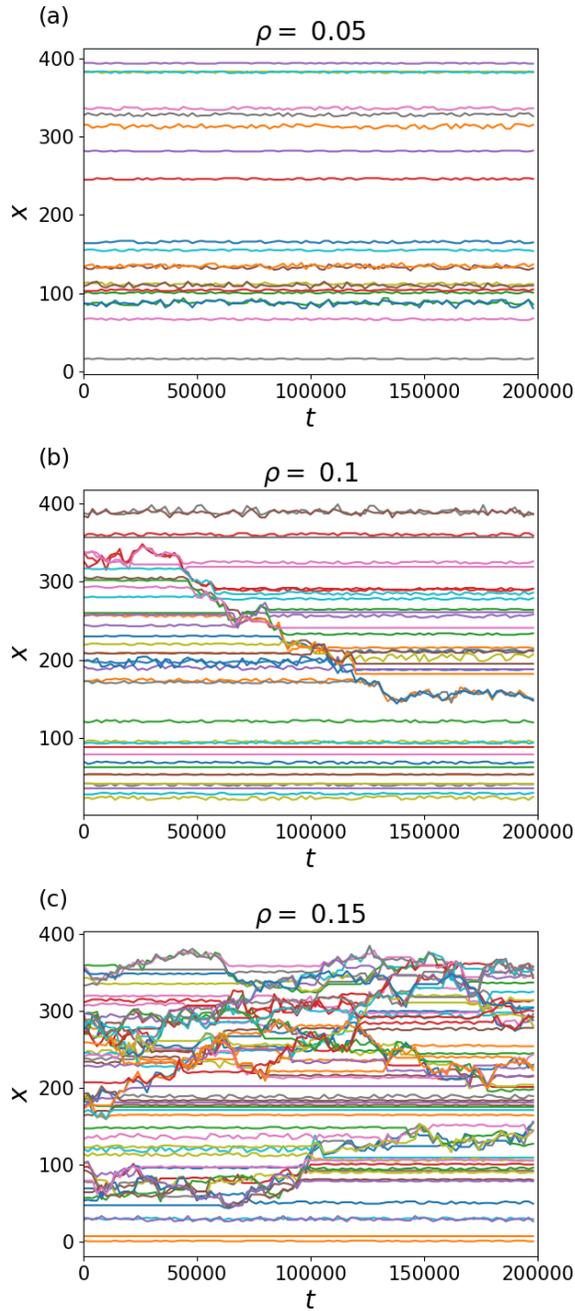


FIG. 2. Position-time graphs of particles for lattice size $L = 400$, unblocking probability $q = 0.5$ and density $\rho = 0.05$ (a), 0.1 (b) and 0.15 (c). Mobile groups in (b) and (c) each consist of 3 particles most of the time. The lack of any mobile group in (a) is only a finite size effect.

edly at long time. As the mobile group moves, it picks up, drops off or exchanges particles while keeping at least 3 particles at all times. When not part of the mobile group, particles are generally trapped at their respective locations. Importantly, the drastically higher mobility observed when particles are coupled together demonstrates an emergent facilitation phenomenon which will be intu-

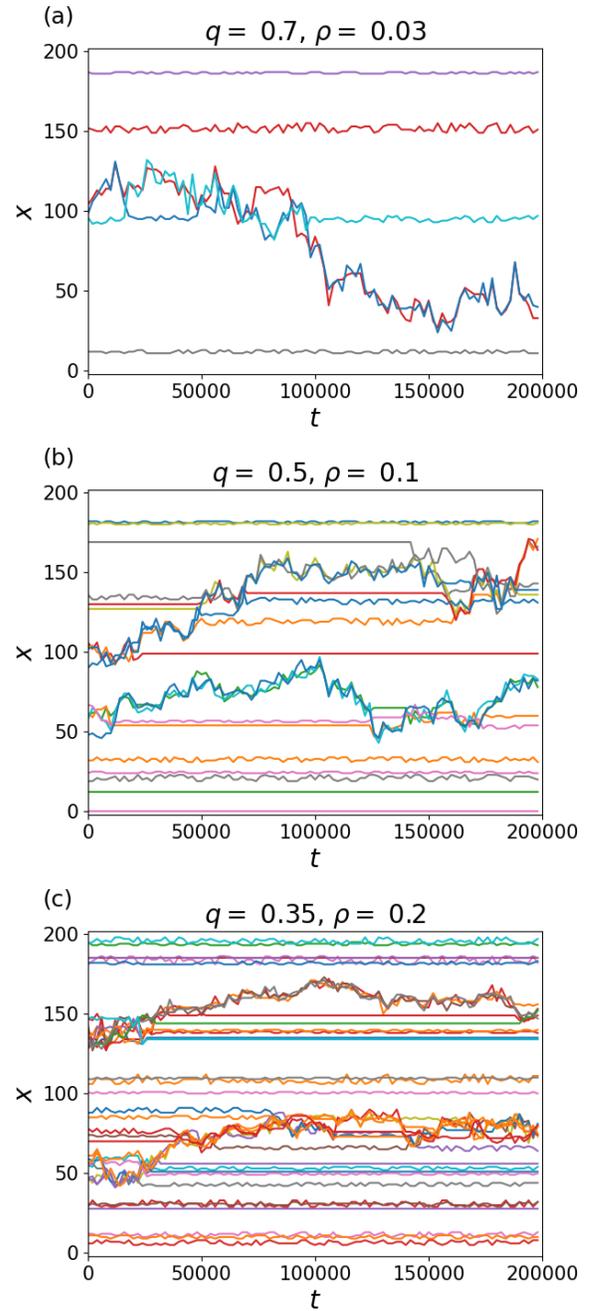


FIG. 3. Position-time graphs of particles for lattice size $L = 200$, unblocking probability $q = 0.7$ (a), 0.5 (b) and 0.35 (c). To reveal relatively isolated mobile groups, we take density $\rho = 0.03$ (a), 0.1 (b) and 0.2 (c). Each mobile group usually consists of 2, 3, and 4 particles in (a), (b) and (c) respectively.

itively explained in Sec. V.

Figure 2(c) corresponding to an even higher density $\rho = 0.15$ shows more abundant mobile groups. Each group also contains at least 3 particles. Group sizes fluctuate due the close proximity to many other mobile or trapped particles.

The dominant size of mobile groups in FRW decreases

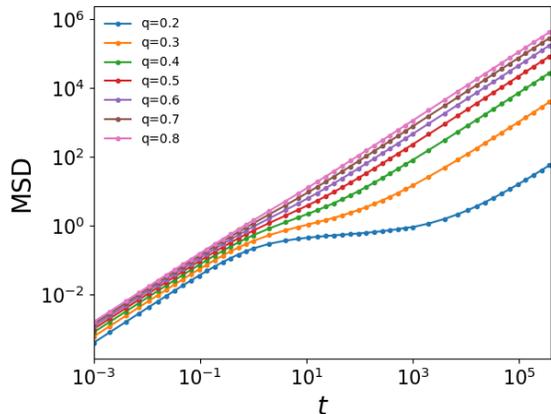


FIG. 4. Particle MSD against time t in log-log scales for various unblocking probability q and density $\rho = 0.8$.

with the unblocking probability q . To show this, we have selected values of q where the typical mobile group sizes are 2, 3 and 4 as plotted in Fig. 3(a), (b) and (c) respectively. A different ρ is used in each case to give only one or two mobile groups for clarity. This dependence is explained in Sec. V.

Plateaus in Mean Square Displacement: We calculate the particle mean square displacement (MSD) defined as

$$MSD = \langle |x_k(t) - x_k(0)|^2 \rangle \quad (3)$$

where $x_k(t)$ denotes the position of particle k at time t . Figure 4 shows the MSD against time for different q and $\rho = 0.8$. At long time, the slopes of the lines in the log-log plot is close to unity, indicating the diffusive regime. Subdiffusive plateaus, characteristic of glass, appear at intermediate time at low q . They indicate temporary trapping of particles either isolated or in small immobile groups as described above. At long time, most momentarily trapped particles have been repeatedly picked up and displaced by the larger mobile groups, resulting at the diffusive regime. We observe that plateaus are more pronounced at small q . This reveals stronger particle trapping and more glassy at small q . More extensive results also show that a smaller ρ also increases the glassiness.

Self-intermediate scattering function: We have measured the self-intermediate scattering function (SISF) defined as

$$F_s(k, t) = \langle e^{ik \cdot (x_k(t) - x_k(0))} \rangle \quad (4)$$

for $k = (2\pi/\lambda)$ with $\lambda = 10$ [20]. Results are shown in Fig. 5(a) for different q and $\rho = 0.8$. As expected of glassy systems, our data is well approximated by the Kohlrausch-Williams-Watts (KWW) stretched exponential function $A \exp[-(t/\tau)^\beta]$, where the amplitude $A \simeq 1$, τ is the structural relaxation time and β is the stretching

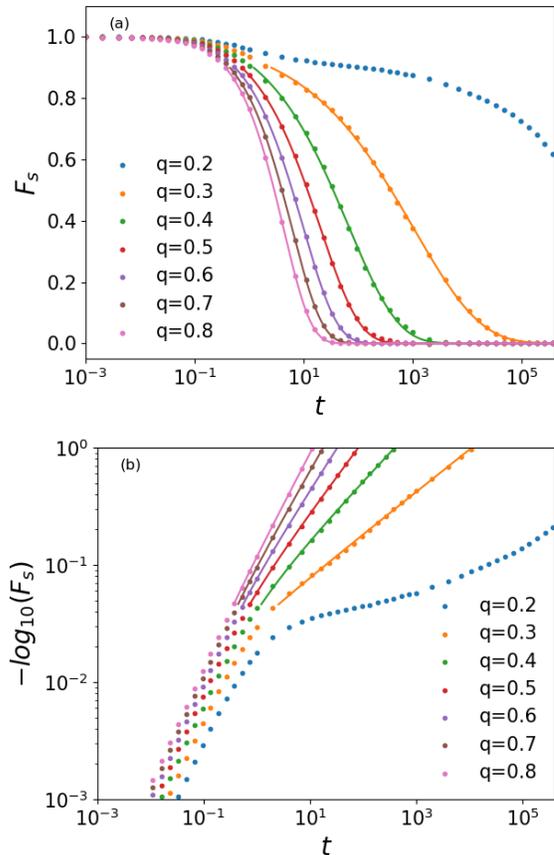


FIG. 5. (a) Self-intermediate scattering function F_s against time t (symbols) in linear-log scales for various unblocking probabilities q with density $\rho = 0.8$. (b) Same data as in (a) plotted as $-\log(F_s)$ versus t in log-log scale. (a)-(b) Data is fitted to the Kohlrausch-Williams-Watts (KWW) stretched exponential function for $F_s < 0.9$ (lines). Data for $q = 0.2$ is not fitted as a full relaxation is not reached.

exponent. Fig. 5(b) displays a log-log plot of $-\log(F_s)$ against t . The approximately linear regions at large t demonstrate the applicability of the KWW form.

Using fitted values of the KWW form in Fig. 5(a), we plot β against q in Fig. 6. We observe that β decreases from 0.86 to 0.37 as q decreases from 0.8 to 0.3. This range coincides well with typical values of glass.

Diffusion coefficient: We measure the MSD for various unblocking probability q and density ρ , and calculate the particle diffusion coefficient as

$$D = \frac{1}{2d} \lim_{t \rightarrow \infty} \frac{MSD}{t}. \quad (5)$$

We use the data regime where the slope in the log-log plot of MSD versus time is larger than 0.96, which we deem sufficiently close to 1. Fig. 7(a) shows D against q in a semi-log plot with different ρ . As q is expected to increase with temperature, we also plot D against $1/q$ as shown in Fig. 7(b). The near linear relation resembles an Arrhenius dependence of D on q .

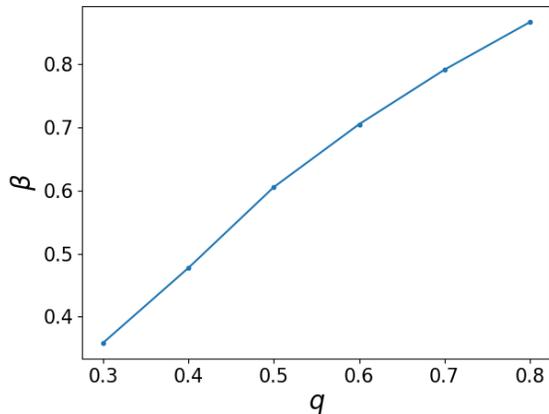


FIG. 6. Stretching exponent β against q from KWW fits of the SISF in Fig. 5(a).

V. FACILITATION MECHANISM AND ERGODICITY

Isolated particles are often trapped as show in Fig. 2(a) for example. The maximum distance an isolated particle can travel from its initial position in either direction is bounded and follows a discrete exponential distribution with a mean

$$s_{trap} = \frac{1}{1-q} - 1. \quad (6)$$

Including the initial site and noting the two possible directions of travel, the average trap size W_{trap} of an isolated particle is

$$W_{trap} = \frac{2}{1-q} - 1 \quad (7)$$

which is finite for any $q < 1$.

Therefore, isolated particles are either completely immobile or could only repetitively move within the trap. These motions are responsible for an initial rise in the MSD when they start to explore their traps. However, trap boundaries begin to limit their motions at longer time. This causes particles to hop repetitively within the trap, resulting in the MSD plateaus as shown in Fig. 4 for small q , corresponding to narrow traps.

In contrast, particles that are close to each other form mobile groups. For example, a mobile group consisting of 2 particles emerges at $q = 0.7$ as shown Fig. 3(a). At this value of q , all isolated particles are trapped, while coupled pairs are mostly mobile. The higher mobility of pairs compared to isolated particles clearly demonstrates facilitation in FRW.

The facilitation mechanism can be intuitively understood as follows. Assume that particle k is bounded at time t_1 by a barrier bond ij at an average distance s_{trap} , i.e. $w_{ijk}(t_1) = 0$. The rate is quenched only until it is resampled by having the barrier broken. This is done by

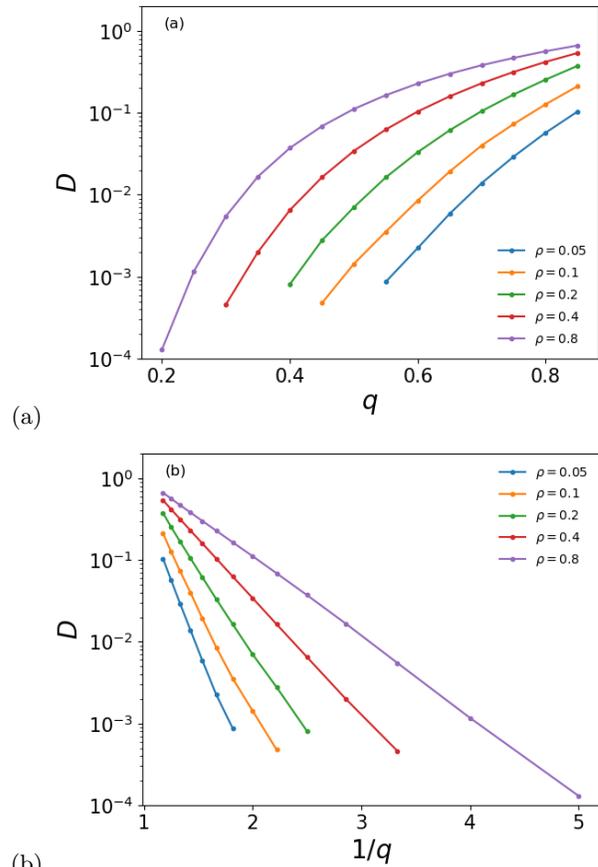


FIG. 7. (a) Diffusion coefficient D against unblocking probability q (a) and $1/q$ (b) in semi-log scales for various density ρ .

having another particle l hop across the bond ij at time $t_2 > t_1$, which triggers a resampling with an unblocking probability q . If this results at $w_{ijk}(t_2) = w_0$, the barrier is lifted and particle k is untrapped. Conversely, particle k can also untrap particle l . In general, if the individual traps of particles k and l overlap spatially, the pair may be able to continuously untrap each other, resulting at a mobile pair as observed in Fig. 3(a).

As q decreases, particle pairs become trapped permanently due to the more abundant constraints as observed in Fig. 3(b), although the trap size of a pair is in general wider than that of a single particle. Nevertheless, adding a third particle enhances untrapping and hence facilitation, resulting in a mobile triplets. Mobile triplets thus dominate motions as observed in Fig. 3(b). Similarly, as seen in Fig. 3(c), at an even smaller q , even triplets are immobile but there are mobile groups each of 4 coupled particles.

For any unblocking probability q , our results support that there is a dominant mobile group size of m^* coupled particles. Smaller groups are immobile while larger groups are less abundant and play a lesser role in the dynamics. As q decreases so that constraints are more

numerous, m^* increases because stronger facilitation with more frequent rate resampling is needed for mobility. There is an associated sequence of mobility transitions as larger and large groups become immobile. A quantitative study of these mobility transitions will be reported elsewhere.

Mobile groups, once exist in a FRW system, survive permanently. They may merge and split as observed in Figs. 2 and 3 but do not vanish completely. This is because if such a system could turn into a trapped state, a trapped state can also evolve into a mobile one by the time reversal symmetry guaranteed by the detailed balance conditions. This contradicts the assumption of a trapped state and hence cannot happen.

Ergodicity of large FRW systems is assumed when deriving equilibrium statistics in Sec. III and this will now be justified self-consistently. We have explained in Sec. III that particles are randomly distributed with uniform probability at equilibrium, assuming ergodicity. Due to the random distribution, for any $q > 0$ and $\rho > 0$, a few sites with m^* particles must exist initially in a sufficiently large system, where m^* is the dominant mobile group size. These groups are mostly mobile and thus some of them must be able to traverse over the whole lattice at long time, untrapping, exchanging with and displacing all particles repeatedly. This justifies the ergodicity assumption.

VI. DISCUSSIONS

Emergent facilitation: Neither the facilitation process nor the dominant mobile group size in FRW is imposed directly by any rule in the model definition. The motions of particle l can facilitate the motions of particle k by breaking barriers as explained in Sec. V. However, it can equally likely inhibit the motions by installing new barriers. Importantly, as all hops are reversible, inhibitions are only temporary and particles can wait until new barriers are reversed before advancing again. They impose no new permanent confinements and thus only a minor slowdown. In contrast, facilitation successively opens up newer configurations, resulting in mobility. Therefore, facilitation is the emergent predominant impact of rate resampling, rather than inhibition.

The FRW model is energetically trivial, like most KCM [11]. However, we are not claiming that energetic interactions are unimportant in glass. Kinetic constraints, i.e. $w_{ijk} = 0$, occurring with probability $1 - q$ according to Eq. (1) model energetically unfavorable particle hops. The FRW assumes that once all significant energy barriers with $w_{ijk} = 0$ are identified, the constrained dynamics along the energy valleys can essentially be considered, as a first approximation, a flat potential energy landscape (PEL). This is sufficient for reproducing kinetic slowdown in glass. However, thermodynamic quantities such as heat capacity cannot be studied in the current energetically trivial form of the FRW model, a situation

similar to that of typical KCMs [38, 39].

Relation to the DPLM: The DPLM is a particle model of glass with void-induced dynamics [20]. Atoms are explicitly simulated, while voids are modeled effectively by empty sites. Interesting physics of glass can be reproduced at low temperatures and low densities of voids [26–35]. It can thus be conceptually simpler and computationally faster if we simulate only the voids explicitly in an effective medium of particles. This has motivated us to define the FRW as a defect model in which particles corresponds to voids in the DPLM while atoms are not simulated explicitly. In the DPLM with energetic particle pair interactions, the motion of a void alters the local particle pairings and hence the PEL experienced by other voids. This is modeled in the FRW by the hopping rate resampling rule. On the other hand, the rate restoration rule in the FRW models the recovery of the system energy in the DPLM when particle configurations are reversed. These mechanisms in the DPLM have been motivated in turn by the string interaction and repetition phenomena observed in our MD simulations [40].

As a coarse-grained version of the DPLM, the FRW can be simulated much more efficiently. In particular, it can be simulated very efficiently in 1D, which is not naturally possible for the DPLM because void-induced motions do not swap particles in 1D. Dynamics demonstrated by the FRW in general are qualitatively similar to those of the DPLM. In particular, the FRW exhibits a diffusion coefficient power law fully consistent with that in the DPLM [31], which will be explained elsewhere. Nevertheless, the present FRW does not incorporate any particle interaction energy, which dominates the thermodynamics of the DPLM. Hence, glassy phenomena reproducible by the DPLM such as heat capacity overshoot [29] and two-level system based heat capacity at low temperature [30] cannot be directly studied by this version of the FRW. Nevertheless, generalization of the FRW to include an internal energy field to thermodynamic properties is straightforward and will be studied in the future.

Another advantage of the highly simplified FRW is the possibility of an analytic description. We have reported a local random configuration tree theory of glass which has been tested on the DPLM [41, 42]. The theory in fact has been motivated by analyzing the FRW. Essential assumptions in the theory such as a tree topology of the configuration space and a bimodal distribution of hopping rates are exact in the FRW by construction, while they hold approximately for the DPLM. In addition, string-like motions are assumed to strongly perturb the local energy landscape, resulting at resampling of the rates of all other strings in a local region. The best fit to DPLM simulation data is obtained at a width $\sqrt{V} = \sqrt{12} \simeq 3.5$ of these local regions [40]. We thus suggest that a site in the FRW corresponds to a region of 3.5^d atoms wide, where d is the dimension of the space. We will apply the theory to describe the FRW quantitatively in the future for further testing and improvement of the theory.

Relation to other lattice models of glass: The

FRW and hence also the DPLM most closely resembles the FA model. In the n -spin facilitation variant of the FA model, one assumes a facilitation rule that n neighboring spins must be present before they are allowed to flip [13]. This is analogous to the FRW that m^* particles must be nearby to each other to form a mobile group. A major challenge of the KCM including the FA model is to justify the origin of the facilitation rule. We suggest that by noting $n = m^*$, the FRW provides such a justification.

Furthermore, most KCM including the FA model are highly coarse-grained defect models. From the relation between FRW and DPLM, it should be interesting to search for analogous atomistic variants of individual KCM for possible further progress.

The many interesting lattice models of glass [11, 13, 20, 21, 23, 24] should not only be applied to illustrate one or two facets of glassy properties, but ideally should be compatible with most, if not all, of the non-vibrational properties of glass. Further tests against a diverse range of glassy phenomena are urgently needed. To this end, the extensively tested DPLM [26–35] has proved to be a promising model. As the FRW may inherit many of these properties from the DPLM, its study should be of good interest for understanding glass.

In conclusion, we have proposed the FRW as a simple KCM with random constraints. It shows emergent facilitation behaviors on the motions of neighboring particles. Dynamics is then dominated by mobile groups of size dependent on the assumed constraint density, analogous to facilitation rules in the seminal FA model of glass. The FRW is a defect model and is a coarsened version of the atomistic DPLM. We believe that the study of compatible models at different coarse-graining levels allows improved understanding of their applicability and limitation for the study of glass.

ACKNOWLEDGMENTS

This work was supported by General Research Fund of Hong Kong (Grants 15210622 and 15303220).

Appendix A: System history encoded in fictitious states

The instantaneous state of a FRW system is specified by the set $\{i_k\}$ of positions of all particles k . Nevertheless, due to the quenched randomness, a system simulated up to time t can only be fully characterized by its complete history, i.e. full sets of positions $\{i_k(t)\}$ and hopping rates $\{w_{ijk}(t)\}$ for the whole duration from time 0 to t . Only then, hopping rate restoration can be possible. While a direct software implementation is possible, the data structures and the data retrieval algorithms are complex. The memory consumption is also huge.

Instead, we take the system history into account easily by effectively encoding it into expanded instantaneous

system states. In this section, we introduce the general mathematical requirements involved. In the next section, our particular implementation will be explained. To simplify the notation, we omit writing the t dependence of the variables explicitly in the following.

For any particle k , we introduce a fictitious particle internal state Ψ_k . The bond between any NN sites i and j also admits a fictitious internal state Φ_{ij} following $\Phi_{ij} = \Phi_{ji}$. The expanded instantaneous state of the system is thus specified by the sets $\{i_k\}$, $\{\Psi_k\}$ and $\{\Phi_{ij}\}$. The hopping rate w_{ijk} is deterministically calculated from

$$w_{ijk} = w(\Psi_k, \Phi_{ij}) \quad (\text{A1})$$

where the function w must be chosen to provide the required statistics followed by w_{ijk} .

After a hop of particle k from site i to site j , states are updated according to

$$\begin{aligned} \Psi'_k &= F(\xi_{ij}, \Psi_k, \Phi_{ij}) \\ \Phi'_{ij} &= G(\xi_{ij}, \Psi_k, \Phi_{ij}) \end{aligned} \quad (\text{A2})$$

where

$$\xi_{ij} = \begin{cases} 1 & \text{for site } j \text{ on the right of site } i, \\ -1 & \text{otherwise.} \end{cases} \quad (\text{A3})$$

The functions F and G must satisfy two conditions. First, to satisfy detailed balance, the new states must satisfy

$$w(\Psi_k, \Phi_{ij}) = w(\Psi'_k, \Phi'_{ij}). \quad (\text{A4})$$

Second, a reversed hop of particle k from j to i at the new states Ψ'_k and Φ'_{ij} must restore the original states,

$$\begin{aligned} \Psi_k &= F(\xi_{ji}, \Psi'_k, \Phi'_{ij}) \\ \Phi_{ij} &= G(\xi_{ji}, \Psi'_k, \Phi'_{ij}). \end{aligned} \quad (\text{A5})$$

Then, Eq. (A1) implies the restoration of also the previous rate w_{ijk} . More generally, it is easy to see that a previous hopping rate can be restored even after an arbitrary sequence of related hops have occurred and reversed by applying Eqs. (A2) and (A5) recursively. Unrelated hops occurring at a distance, which do not affect these local states under consideration, naturally play no role in the reversal.

As rate restoration is possible by straightforward calculations based on the instantaneous system state, memorization of previous rates and the system history is not required. The dynamics of FRW with the expanded states is thus reduced to simple Markovian processes for which standard kinetic Monte Carlo approaches apply.

Appendix B: Rate restoration by reversible random numbers

We now explain our particular implementation of the expanded FRW states and their dynamics. To enable exact restoration of hopping rates, we adopt integer rather

than float-point mathematics. As pseudo random rates with controllable statistics are required, we take note of the approach of congruential random number generators and consider only integers $0, 1, \dots, \mathcal{M} - 1$ following modulo- \mathcal{M} arithmetics, i.e. $i \equiv (i \bmod \mathcal{M})$. A natural choice is $\mathcal{M} = 2^{32}$, leading to 32-bits unsigned integer data type intrinsically available in most computers. This \mathcal{M} is also sufficiently large to enable excellent pseudo random number properties.

Our algorithm requires non-trivial matrix inversions, implying that the states Ψ_k and Φ_{ij} cannot be simple integers. Instead, the next simplest choice is two-component matrices, i.e.

$$\Psi_k = \begin{pmatrix} \psi_k^{(1)} \\ \psi_k^{(2)} \end{pmatrix}, \quad \Phi_{ij} = \begin{pmatrix} \phi_{ij}^{(1)} \\ \phi_{ij}^{(2)} \end{pmatrix} \quad (\text{B1})$$

where the elements $\psi_k^{(1)}, \psi_k^{(2)}, \phi_{ij}^{(1)}$ and $\phi_{ij}^{(2)}$ are 32-bits unsigned integers. Each Ψ_k or Φ_{ij} is thus a 64-bit state.

Equation (A2), concerning a hop of particle k from site i to site j , is implemented as

$$\begin{aligned} \Psi'_k &= [U^{\xi_{ij}}(\Psi_k^T \Phi_{ij})]^T \Psi_k \\ \Phi'_{ij} &= U^{-\xi_{ij}}(\Psi_k^T \Phi_{ij}) \Phi_{ij} \end{aligned} \quad (\text{B2})$$

where the superscript T denotes transpose. A simple choice of $U(n)$ we have taken is

$$U(n) = \begin{pmatrix} n+1 & n \\ 1 & 1 \end{pmatrix} \quad (\text{B3})$$

with an inverse

$$U^{-1}(n) = \begin{pmatrix} 1 & -n \\ -1 & n+1 \end{pmatrix}. \quad (\text{B4})$$

Modulo- \mathcal{M} unsigned integer arithmetic is assumed in Eqs. (B2)-(B4) so that $-1 \equiv \mathcal{M} - 1$. The equations constitute congruential random number generators so that

the elements $\psi_k^{(1)}, \psi_k^{(2)}, \phi_{ij}^{(1)}$ and $\phi_{ij}^{(2)}$ generated by iterating Eq. (B2) in FRW simulations follow a uniform distribution in $0, 1, \dots, \mathcal{M} - 1$. This uniform distribution is readily verified in our simulations.

Equation (B2) implicitly defines our choices of the functions F and G . For any hop, the states Ψ_k and Φ_{ij} are updated via $U(n)$ and $U^{-1}(n)$ or $U^{-1}(n)$ and $U(n)$ respectively, so that the product $\Psi_k^T \Phi_{ij}$ is invariant after a hop, i.e.

$$\Psi_k'^T \Phi'_{ij} = \Psi_k^T \Phi_{ij}. \quad (\text{B5})$$

Similarly, forward and backward hops involve $U(n)$ and $U^{-1}(n)$ with the same n which cancel out each other upon multiplication. Hence, states reversal, i.e. Eq. (A5), is properly followed.

We define the rate function w in Eq. (A1) as

$$w(\Psi_k, \Phi_{ij}) = w_0 f(\Psi_k^T \Phi_{ij}). \quad (\text{B6})$$

where

$$f(n) = \theta(q - S(n)/\mathcal{M}) \quad (\text{B7})$$

with θ being the Heaviside step function and $S(n)$ to be explained below. As $\Psi_k^T \Phi_{ij}$ is invariant during the hop (see Eq. (B5)), the detailed balance condition (A4) is satisfied.

The function $S(n)$ can be the identity function, but we take a bitwise rotation operation by 16 bits, so as to enhance randomization. Then, analogous to congruential generators, $\Psi_k^T \Phi_{ij}$ and hence $S(\Psi_k^T \Phi_{ij})$ are also uniform random numbers in $0, 1, \dots, \mathcal{M} - 1$ so that $S(\Psi_k^T \Phi_{ij})/\mathcal{M}$ is a uniform random number in $[0, 1]$. Therefore, $S(\Psi_k^T \Phi_{ij}) > q$ occur with a probability q , implying that w_{ijk} follows the required statistics according to Eqs. (A1), (B6), and (B7).

-
- [1] G. Biroli and J. P. Garrahan, "Perspective: The glass transition," *J. Chem. Phys.* **138**, 12A301 (2013).
- [2] F. Arceri, F.çois P. Landes, L. Berthier, and G. Biroli, "Glasses and aging: A statistical mechanics perspective," arXiv:2006.09725 (2020).
- [3] W. H. Wang, "Dynamic relaxations and relaxation-property relationships in metallic glasses," *Progress in Materials Science* **106**, 100561 (2019).
- [4] C. Rodriguez-Tinoco, M. Gonzalez-Silveira, M. A. Ramos, and J. Rodriguez-Viejo, "Ultrastable glasses: new perspectives for an old problem," *La Rivista del Nuovo Cimento* **45**, 325 (2022).
- [5] A. Zaccone, "Theory of disordered solids," Cham: Springer **1015**, 179 (2023).
- [6] J. C. Dyre, "Solid-that-flows picture of glass-forming liquids," *J. Phys. Chem. Letters* **15**, 1603 (2024).
- [7] J.-L. Barrat, Jörg Baschnagel, and A. Lyulin, "Molecular dynamics simulations of glassy polymers," *Soft Matter* **6**, 3430 (2010).
- [8] J.-L. Barrat and L. Berthier, "Computer simulations of the glass transition and glassy materials," *Comptes Rendus. Physique* **24**, 57 (2023).
- [9] M. Plischke and B. Bergersen, *Equilibrium statistical physics* (World Scientific, 1994).
- [10] A-L Barabási and H. E. Stanley, *Fractal concepts in surface growth* (Cambridge university press, 1995).
- [11] J. P. Garrahan, P. Sollich, and C. Toninelli, "Kinetically constrained models," in *Dynamical Heterogeneities in Glasses, Colloids and Granular Media*, edited by L. Berthier, G. Biroli, J.-P. Bouchaud, L. Cipelletti, and W. van Saarloosand (Oxford University Press, 2011).
- [12] F. Ritort and P. Sollich, "Glassy dynamics of kinetically constrained models," *Adv. Phys.* **52**, 219 (2003).
- [13] G. H. Fredrickson and H. C. Andersen, "Kinetic ising

- model of the glass transition,” *Phys. Rev. Lett.* **53**, 1244 (1984).
- [14] G. H. Fredrickson and H. C. Andersen, “Facilitated kinetic ising models and the glass transition,” *J. Chem. Phys.* **83**, 5822 (1985).
- [15] R. G. Palmer, D. L. Stein, E. Abrahams, and P. W. Anderson, “Models of hierarchically constrained dynamics for glassy relaxation,” *Phys. Rev. Lett.* **53**, 958 (1984).
- [16] D. Chandler and J. P. Garrahan, “Dynamics on the way to forming glass: Bubbles in space-time,” *Annu. Rev. Phys. Chem.* **61**, 191 (2010).
- [17] A. S. Keys, L. O. Hedges, J. P. Garrahan, S. C. Glotzer, and D. Chandler, “Excitations are localized and relaxation is hierarchical in glass-forming liquids,” *Phys. Rev. X* **1**, 021013 (2011).
- [18] C. Scalliet, B. Guiselin, and L. Berthier, “Thirty milliseconds in the life of a supercooled liquid,” *Phys. Rev. X* **12**, 041028 (2022).
- [19] L. Costigliola, T. Hecksher, and J. C. Dyre, “Glass-forming liquids need facilitation,” *Proc. Natl. Acad. Sci.* **121**, e2408798121 (2024).
- [20] L.-H. Zhang and C.-H. Lam, “Emergent facilitation behavior in a distinguishable-particle lattice model of glass,” *Phys. Rev. B* **95**, 184202 (2017).
- [21] A. Tahaei, G. Biroli, M. Ozawa, M. Popović, and M. Wyart, “Scaling description of dynamical heterogeneity and avalanches of relaxation in glass-forming liquids,” *Phys. Rev. X* **13**, 031034 (2023).
- [22] M. Ozawa and G. Biroli, “Elasticity, facilitation, and dynamic heterogeneity in glass-forming liquids,” *Phys. Rev. Lett.* **130**, 138201 (2023).
- [23] M. R. Hasyim and K. K. Mandadapu, “Emergent facilitation and glassy dynamics in supercooled liquids,” *Proc. Natl. Acad. Sci.* **121**, e2322592121 (2024).
- [24] Y. Nishikawa and L. Berthier, “Collective relaxation dynamics in a three-dimensional lattice glass model,” *Phys. Rev. Lett.* **132**, 067101 (2024).
- [25] C.-T. Yip, M. Isobe, C.-H. Chan, S. Ren, K.-P. Wong, Q. Huo, C.-S. Lee, Y.-H. Tsang, Y. Han, and C.-H. Lam, “Direct evidence of void-induced structural relaxations in colloidal glass formers,” *Phys. Rev. Lett.* **125**, 258001 (2020).
- [26] M. Lulli, C.-S. Lee, H.-Y. Deng, C.-T. Yip, and C.-H. Lam, “Spatial heterogeneities in structural temperature cause kovacs’ expansion gap paradox in aging of glasses,” *Phys. Rev. Lett.* **124**, 095501 (2020).
- [27] C.-S. Lee, M. Lulli, L.-H. Zhang, H.-Y. Deng, and C.-H. Lam, “Fragile glasses associated with a dramatic drop of entropy under supercooling,” *Phys. Rev. Lett.* **125**, 265703 (2020).
- [28] Matteo Lulli, Chun-Shing Lee, Ling-Han Zhang, Hai-Yao Deng, and Chi-Hang Lam, “Kovacs effect in glass with material memory revealed in non-equilibrium particle interactions,” *J. Stat. Mech.* **2021**, 093303 (2021).
- [29] C.-S. Lee, H.-Y. Deng, C.-T. Yip, and C.-H. Lam, “Large heat-capacity jump in cooling-heating of fragile glass from kinetic monte carlo simulations based on a two-state picture,” *Phys. Rev. E* **104**, 024131 (2021).
- [30] X.-Y. Gao, H.-Y. Deng, C.-S. Lee, J.Q. You, and C.-H. Lam, “Emergence of two-level systems in glass formers: a kinetic monte carlo study,” *Soft Matter* **18**, 2211 (2022).
- [31] G. Gopinath, C.-S. Lee, X.-Y. Gao, X.-D. An, C.-H. Chan, C.-T. Yip, H.-Y. Deng, and C.-H. Lam, “Diffusion-coefficient power laws and defect-driven glassy dynamics in swap acceleration,” *Phys. Rev. Lett.* **129**, 168002 (2022).
- [32] X.-Y. Gao, C.-Y. Ong, C.-S. Lee, C.-T. Yip, H.-Y. Deng, and C.-H. Lam, “Kauzmann paradox: A possible crossover due to diminishing local excitations,” *Phys. Rev. B* **107**, 174206 (2023).
- [33] B. Li, C.-S. Lee, X.-Y. Gao, H.-Y. Deng, and C.-H. Lam, “The distinguishable-particle lattice model of glasses in three dimensions,” *Soft Matter* **20**, 1009 (2023).
- [34] C.-Y. Ong, C.-S. Lee, X.-Y. Gao, Q. Zhai, Z. Yu, R. Shi, H.-Y. Deng, and C.-H. Lam, “Relating fragile-to-strong transition to fragile glass via lattice model simulations,” *Phys. Rev. E* **109**, 054124 (2024).
- [35] Q. Zhai, X.-Y. Gao, C.-S. Lee, C.-Y. Ong, K. Yan, H.-Y. Deng, S. Yang, and C.-H. Lam, “Surface mobility gradient and emergent facilitation in glassy films,” *Soft Matter* **20**, 4389 (2024).
- [36] D. Cangialosi, V. M. Boucher, A. Alegria, and J. Colmenero, “Free volume holes diffusion to describe physical aging in poly (methyl methacrylate)/silica nanocomposites,” *J. Chem. Phys.* **135**, 014901 (2011).
- [37] B. Mei, B. Zhuang, Y. Lu, L. An, and Z.-G. Wang, “Local-average free volume correlates with dynamics in glass formers,” *J. Phys. Chem. Letters* **13**, 3957 (2022).
- [38] G. Biroli, J.-P. Bouchaud, and G. Tarjus, “Are defect models consistent with the entropy and specific heat of glass formers?” *J. Chem. Phys.* **123**, 044510 (2005).
- [39] A. S. Keys, J. P. Garrahan, and D. Chandler, “Calorimetric glass transition explained by hierarchical dynamic facilitation,” *Proc. Natl. Acad. Sci.* **110**, 4482 (2013).
- [40] C.-H. Lam, “Repetition and pair-interaction of string-like hopping motions in glassy polymers,” *J. Chem. Phys.* **146**, 244906 (2017).
- [41] C.-H. Lam, “Local random configuration-tree theory for string repetition and facilitated dynamics of glass,” *J. Stat. Mech.* **2018**, 023301 (2018).
- [42] H.-Y. Deng, C.-S. Lee, M. Lulli, L.-H. Zhang, and C.-H. Lam, “Configuration-tree theoretical calculation of the mean-squared displacement of particles in glass formers,” *J. Stat. Mech.* **2019**, 094014 (2019).