Glassy behavior induced by geometrical frustration in a hard-core lattice-gas model

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Abstract. – We introduce a hard-core lattice-gas model on generalized Bethe lattices and investigate its compaction behavior analytically and numerically. If compactified slowly, the system undergoes a first-order crystallization transition. If compactified much faster, the system stays in a meta-stable liquid state and undergoes a glass transition under further compaction. We show that this behavior is induced by geometrical frustration which appears due to the existence of short loops in the generalized Bethe lattices. We also compare our results to numerical simulations of a three-dimensional analog of the model.

The last decades have seen a great interest in understanding the structural glass transition [1, 2]. The latter is given by a dramatic dynamical slowing-down which prevents the systems from reaching the equilibrium state at experimental time scales. For glass-forming liquids, e.g., the glass transition is usually identified by the temperature where the viscosity exceeds \(10^{13}\) poise. A central role in theoretical approaches is played by fully connected spin-glass models with multi-spin interactions [2, 3]. These models in fact show a purely dynamical transition towards a glassy state followed by a thermodynamic transition at lower temperature. The dynamical transition, even if not strictly present in real materials due to activated processes, is believed to present some similarities to the structural glass transition.

The analogies between fully connected spin glasses and structural glasses are, however, of purely phenomenological nature, missing a microscopic verification. The main differences for spin-glass models are that they have multi-spin interaction, which in addition are disordered (quenched disorder) and long-ranged (fully connected models). So, in particular, these models do not have any crystalline state.

Recently, some of these points could be cured by considering models based on Bethe lattices or Husimi trees, which are the analog of Bethe lattices for multi-spin interactions: On finite length scales, these lattices have no inhomogeneities. The corresponding models undergo a crystallization transition which was found to be of first order in the most interesting cases. By cooling or compactifying these models relatively fast, the system stays in the meta-stable liquid (or paramagnetic) phase. In analogy to their fully connected counterparts, they undergo a dynamical glass transition at even lower temperature, which is followed by a thermodynamic transition [4, 5]. The latter cannot be observed in simulations (or experiments) since the system does not equilibrate below the dynamical transition. These models still have, however,
the problem of having multi-spin interactions (which, in some cases, are introduced by local constraints concerning more than two particles, as seen in [5]). Similar models were also discussed in the context of compactified granular matter [6].

Here we present a lattice-gas model with pure two-particle interactions. Defined on a usual, i.e., locally tree-like Bethe lattice, the model does neither show a first-order crystallization nor a glass transition. If, however, the Bethe lattice is generalized to include short loops, a dynamical behavior reminiscent of the glass transition in liquids of hard particles or Lennard-Jones liquids [7] is found. This glass transition is induced by geometrical frustration [8] which prohibits constructing a globally dense configuration from purely local rules.

As the original Bethe lattice, also the generalized Bethe lattices studied here are of mean-field nature. In order to compare our results to more realistic models, we introduce a three-dimensional lattice gas showing the same kind of geometrical frustration. We study this model be means of Monte Carlo (MC) simulations: The behavior of the model shares many features with the mean-field case, even if they are in general much less pronounced.

The hard-sphere lattice-gas model can be defined on any graph $G = (V, E)$. Each site $i \in V$ can be occupied by at most one particle, $x_i \in \{0, 1\}$. Particles are assumed to have a hard core of radius one, i.e. neighboring sites can never be occupied simultaneously: $x_ix_j = 0$ if $\{i, j\} \in E$. Note that this defines a two-particle interaction. If the graph is a tree, i.e. if it does not contain any loops, dense particle packings can be generated from a simple local rule: Starting with one particle, all neighboring sites have to be empty due to the hard-core constraint. Particles can again be put to all second neighbors and so on. Geometrical frustration enters via loops. If, e.g., some of the second neighbors of a site are directly connected by an edge, they cannot be occupied simultaneously. The resulting arbitrariness in selecting occupied second neighbors finally leads to globally disordered packings of sub-maximal density.

To investigate the influence of geometrical frustration, let us consider the following generalized Bethe lattices: They are constructed from basic units consisting of $p$-cliques, i.e. fully connected subgraphs of $p$ vertices. From these basic units, a graph is constructed by joining $K + 1$ cliques in every vertex, see fig. 1. A usual Bethe lattice is obtained for $p = 2$, where the basic cliques are just simple edges, and the graph is locally tree-like. For $p > 2$, the graph contains triangles, i.e. local loops, giving rise to geometrical frustration.

The thermodynamic properties of the model are given by the grand-canonical partition function

$$\Xi = \sum_{\{x_i\} \in \{0, 1\}^N} e^{\mu \sum_i x_i} \prod_{\{i,j\} \in E} (1 - x_ix_j).$$

(1)

The last product includes the hard-core constraint, it equals one for allowed packings, and zero if any two neighboring sites are occupied simultaneously. The chemical potential $\mu$ is coupled to the total particle number and can be used to tune the particle density. In our system, we study the compaction of the model which can be achieved by increasing $\mu$. In particular, densest packings are reached in the limit $\mu \to \infty$.

The model can be solved most easily using the cavity method [9] which, in the liquid as well as in the crystalline state, reduces to the Bethe-Peierls iterative method. Note, however, that the latter is modified with respect to the presence of short loops. On a coarse-grained level, where every local clique is considered as one object, the (hyper-)graph becomes locally tree-like and therefore solvable by an iterative procedure. In the following, we use this coarse-grained tree-like structure and speak about trees, branches, etc. only in this sense. The cliques are numbered by $\nu = 1, \ldots, (K+1)N/p$. We closely follow the presentation and notation of [9], cf. also [5].
Fig. 1 – Part of the generalized Bethe lattice. The unit cell is equivalent to a clique of \( p \) vertices (here \( p = 4 \)). Each site is contained in \( K + 1 \) unit cells (here \( K = 3 \)).

Fig. 2 – Density \( \rho \) as a function of the chemical potential \( \mu \), for \( p = 4 \), \( K = 3 \). The full lines give analytical results for the liquid and the crystalline phases. The spinodal and the crystallization points, as well as the dynamic and static glass transition, are marked by vertical lines (from left to right). Results are compared with numerical compaction curves for random generalized Bethe lattices of size \( N = 999 \), averaged over 100 graphs. Here \( \delta \mu = 0.2 \) was used. Inset: compaction rate dependence for high values of \( \mu \) for \( n_{\text{MC}} \) ranging from 5000 (top) to 10 (bottom).

Let us consider a branch of the graph which is rooted in vertex \( i \). One of the cliques containing vertex \( i \) is not part of the branch, we denote it by \( \nu \). We first calculate the partition function \( \Xi_0(i, \nu) \) of this branch where \( i \) is fixed to be empty (\( x_i = 0 \)) or occupied (\( x_i = 1 \)). Let us denote the other \( K \) cliques containing \( i \) by \( \nu_1, \ldots, \nu_K \), and the further vertices contained in clique \( \nu_k \) by \( j_{1,k}, \ldots, j_{p-1,k}, k = 1, \ldots, K \). Due to the hard-sphere constraint, every clique is allowed to carry at most one particle, we thus find

\[
\Xi_0(i, \nu) = \prod_{k=1}^{K} \left[ \prod_{a=1}^{p-1} \Xi_0(j_{a,k}, \nu_k) + \sum_{a=1}^{p-1} \Xi_1(j_{a,k}, \nu_k) \prod_{b \neq a} \Xi_0(j_{b,k}, \nu_k) \right],
\]

\[
\Xi_1(i, \nu) = e^\mu \prod_{k=1}^{K} \left[ \prod_{a=1}^{p-1} \Xi_0(j_{a,k}, \nu_k) \right].
\]

Introducing cavity fields \( \mu h_\nu^\nu = \ln(\Xi_1(i, \nu)/\Xi_0(i, \nu)) \), these iterate according to

\[
h_\nu^\nu = 1 + \sum_{k=1}^{K} u(h_{j_{1,k}}^\nu, \ldots, h_{j_{p-1,k}}^\nu),
\]

\[
u(h_1, \ldots, h_{p-1}) = -\frac{1}{\mu} \ln \left( 1 + \sum_{a=1}^{p-1} e^{\mu h_a^\nu} \right),
\]

\( i.e. \) they result from a linear superposition of the local chemical potential \( \mu \) and fields propagated along the cliques; and they take values \( h_\nu^\nu \in [1 - K - \ln(p - 1 + e^{-\mu})/\mu, 1] \). The
true effective field acting on a vertex is given by the contributions propagated from all neighbors, i.e. from all $K+1$ cliques containing the vertex, $H_i = 1 + \sum_{k=1}^{K+1} u(h_{j_1,k}^{(k)}, \ldots, h_{j_p,k}^{(k)}) \in [-K - \ln(p - 1 + e^{-\mu})/\mu, 1]$. Note that the r.h.s. of this equation still contains the fields $h_j^{(k)}$ coming from single branches only. The distribution of these fields serves as an order parameter and allows to identify liquid, crystalline and glassy states. Furthermore, its knowledge allows to calculate the grand-canonical potential [9], with $\mathcal{E}$ denoting the set of all $p$-cliques in the graph, $-\ln \Xi = -K \sum_i \ln(1 + e^{\mu H_i}) + \sum_{\nu \in \mathcal{E}} \ln(1 + \sum_{i \in \nu} e^{\mu h_i^{(\nu)})}$, as well as the average particle density $\rho$ per site, $N \rho = \sum_i x_i = \sum_i e^{\mu H_i} / (1 + e^{\mu H_i})$.

The liquid phase, which is expected to be globally stable for small chemical potential $\mu$, is characterized by its translational invariance. The effective fields $h^{(\nu)}_i$ and consequently $H_i$ do not depend on the site index $i$ and are thus given by the homogeneous fixed point of eq. (3).

For $K \geq 2$, this translational invariance breaks down in the crystalline phase: On each clique, there is exactly one site of high average density, all the other $(p - 1)$ vertices have homogeneous lower density. There are thus two fields $h^{(1)} < h^{(2)}$, which are a solution of

\[
\begin{align*}
    h^{(1)} &= 1 + Ku(h^{(2)}, h^{(1)}, \ldots, h^{(1)}), \\
    h^{(2)} &= 1 + Ku(h^{(1)}, \ldots, h^{(1)}).
\end{align*}
\]  

For $p = 2$, i.e. usual Bethe lattices, the crystalline solution appears continuously due to a local instability of the liquid solution, the solidification is thus of second order, and the model easily crystallizes even if compactified relatively fast [10].

In the remaining part of the paper we therefore concentrate on the more interesting case $p \geq 3$, i.e. on graphs showing geometrical frustration due to short loops. There the crystalline solution appears discontinuously at a spinodal point $\mu_s$. Still, its grand-canonical potential is larger than the one of the liquid phase. The two potentials cross at the crystallization transition $\mu_c > \mu_s$, which thus describes a first-order transition being accompanied by a discontinuous density jump. Sending $\mu \to \infty$, the crystalline system quickly approaches the maximal density $1/p$. For the example $p = 4, K = 3$, which is presented in fig. 2, the transition points are given by $\mu_s \simeq 1.73$ and $\mu_c \simeq 2.18$.

The liquid phase stays, however, meta-stable even above $\mu_c$ describing thus a super-cooled liquid. If we compactify the system rather quickly, it initially stays liquid. However, at higher density, the system even falls out of this local equilibrium: An exponential number of glassy states of smaller density (and higher grand-canonical potential) appear discontinuously at a dynamical transition $\mu_d$ and trap the system. At $\mu_{rsb}$ a sub-exponential number of glassy states finally reach a lower grand-canonical potential than the liquid state. In the equivalent replica approach, this corresponds to one-step replica symmetry breaking. For $p = 4$ and $K = 3$ we find $\mu_d \simeq 5.7$ and $\mu_{rsb} \simeq 6.1$, i.e. they are located far beyond the crystallization transition; at $\mu_d$ there are about $\exp[0.0088N]$ metastable states, but the clustering phenomenon becomes more pronounced for larger $p$ and $K$.

Technically, this can be established using the cavity approach which generalizes the Bethe-Peierls solution to more than one state [9]. The main difference to the liquid solution is that, in different pure states, there are different cavity fields $h^{(\nu)}_i$. They can be characterized by their histograms $P_i^{(\nu)}(h)$ which have to be determined self-consistently using the cavity equations, for technical details see the analogous case discussed in [9]. With $P_i^{(\nu)}(h)$ being Dirac peaks in the liquid phase, the dynamical transition point $\mu_d$ can be identified by the discontinuous appearance of a non-trivial solution for $P_i^{(\nu)}(h)$ which immediately covers the full interval of allowed fields. The static transition $\mu_{rsb}$ follows from the point where the replica-symmetry-broken grand-canonical potential becomes dominant with respect to the liquid one [11].
For comparison, we have performed numerical simulations obtained on fixed-connectivity random graphs built from 4-cliques. From an analytical point of view, they share the liquid and the glassy state with the generalized Bethe lattices discussed above. Only the crystalline state is inconsistent with the random character of the graph entering via disordered loops of length $O(N)$, and it cannot be observed in the simulations. The simulation works as follows. For each MC step, a site $i$ is selected randomly. With probability $p = 0.5$ a MOVE (M) step is performed, and with probability $1 - p$ an EXCHANGE (EX) step:

M: If site $i$ is empty and has exactly one occupied neighbor, the particle is moved to $i$. In all other cases, the configuration remains unchanged.

EX: If the site is occupied, the particle is removed with probability $\exp[-\mu]$. If the site is empty, and all neighboring sites are empty, a particle is placed on $i$.

Note that in this way detailed balance is fulfilled, and the equilibrium distribution is the one corresponding to eq. (1). All simulations start with $\mu = 0$. Then $\mu$ is increased in steps of $\delta \mu = 0.1$. For every value of the chemical potential, we perform $n_{MC}N$ Monte Carlo steps.

In fig. 2, the analytic findings are compared with numerical results. Note that in principle the value of $\mu_d$ can be inferred from the point where the numerical curve deviates from the liquid one in the limit of infinitesimal slow compaction ($n_{MC} \to \infty$). Even if our data turned out to be incompatible with every tested form of fits (as a function of $n_{MC}$), this point appears to be consistent with the analytical prediction which is marked by a vertical line in the figure.

As already mentioned, also the generalized Bethe lattices are of mean-field nature. Whereas a similar glassy behavior was found also numerically in a two-dimensional model of cross-shaped particles [12], it is useful to test our theoretical predictions at least qualitatively in a finite-dimensional model which is closer to our mean-field model. In particular, it should be characterized by the same kind of geometrical frustration. We thus consider a three-dimensional lattice with $N = L^3$ sites. It is generated from a tetrahedron, i.e. a 4-clique, which is translated along its edges. Again each site is contained in $K + 1 = 4$ tetrahedra. We impose periodic boundary conditions. On top of this lattice, we again consider our lattice gas of hard spheres of radius one, and we numerically study its behavior by means of MC simulations up to size $N = 100^3$.

As in its mean-field counterpart, the system is found to be in a liquid phase for small $\mu$: All particles are free to move around, they are not localized, and the time-averaged single-site density is homogeneous. For slow compaction, we observe a discontinuous crystallization transition at $\mu \simeq 2$, cf. fig. 3. At this point, the symmetry between sites breaks down, every tetrahedron has one site of higher-than-average density, and three of lower density. The average density exhibits a discontinuity and quickly approaches 0.25, i.e. its theoretically maximal value, if we further increase $\mu$. The first-order character of this transition can be numerically tested by decreasing $\mu$ again. Doing this we observe a hysteresis, cf. fig. 3, the system stays in the crystalline state even for $\mu < 2$.

If, starting from the liquid state at low $\mu$, we increase $\mu$ quite fast, crystallization is avoided. The system stays at a density lower than the crystalline one. This phenomenon appears in analogy to the super-cooled, meta-stable liquid observed in the mean-field model. At higher $\mu$, the system starts to freeze into a glassy state, being characterized by a density which is much smaller than 0.25. This asymptotic density depends heavily on the compaction rate.

There are, however, some crucial differences between figs. 2 and 3. If we consider the compaction curves in the mean-field model, cf. fig. 2, they overlap even beyond the crystallization transition, until the (local) equilibration time starts to be larger than the characteristic compaction time, and the system falls out of the meta-stable equilibrium. If we stop to increase
Fig. 3 – Density $\rho$ of the lattice model as a function of the chemical potential $\mu$ for $n_{MC} = 1, 2, 5, 25, 1000$ Monte Carlo sweeps per value of the chemical potential (from bottom to top). Every $n_{MC}$ sweeps, the chemical potential is increased by 0.1. The data are for one run of a lattice with $N = 50^3$ sites. The solid line represents the analytical result for the crystalline phase on the generalized Bethe lattice which can be considered as a simple mean-field approximation. Inset: density $\rho$ as a function of $\mu$ for slowly increasing ($n_{MC} = 500$) and subsequently fast decreasing ($n_{MC} = 5$) chemical potential.

Fig. 4 – Density $\rho$ of the lattice model as a function of the chemical potential $\mu$ for $n_{MC} = 2$, when the chemical potential is first decreased every $n_{MC}$ sweeps by 0.1 until $\mu_{stop}$ is reached. Then the system continues to evolve at fixed value of $\mu$. The inset shows the density as a function of time for smaller $\mu_{stop}$. The arrows indicate the times from where the chemical potential is kept fixed. The horizontal lines denote the equilibrium densities of the crystalline state at $\mu_{stop}$.

$\mu$ at some point below the glass transition, the density evolves towards the one of the liquid phase, i.e. the latter is a true metastable phase. This seems not to be true for the finite-dimensional lattice. The compaction rate dependence sets in directly at the crystallization transition, and there is no pronounced gap between a meta-stable liquid state and the globally stable solid one. In fig. 4 we report the following experiment: The system is compactified quite fast by increasing the chemical potential up to some value $\mu_{stop}$ which is chosen beyond the crystallization point. Then the system is allowed to evolve at fixed $\mu$. As can be seen in the inset for a system of size $N = 50^3$, the model still equilibrates towards the crystalline state (given by the horizontal lines), even if this equilibration time is much larger than the equilibration time in the liquid phase. In the main part of fig. 4, we show the system size dependence of this effect: The equilibration time grows with $N$, and probably diverges in the thermodynamic limit. This growth is not exponential in $N$, i.e. there are no extensive energy barriers to be overcome as in the generalized Bethe-lattice model.

To conclude, we have introduced a hard-core lattice-gas model with only two-particle interactions. On a generalized Bethe lattice, it exhibits a crystalline phase as well as glassy behavior if super-cooled sufficiently fast. In contrast to usual Bethe lattices, the generalized ones have short loops of odd length. These loops, and the resulting geometrical frustration, are responsible for the glassy behavior of the model which is similar to the behavior known from models with multi-spin interactions: If super-cooled, the system falls out of equilibrium at a dynamical transition which is accompanied by the appearance of an exponential number of
states. Also in our model, this transition is not of thermodynamic nature, a replica-symmetry-breaking equilibrium transition occurs only at higher chemical potential. It is quite interesting that similar phenomena can also be found in a completely different model considered in [13]: There, geometrical frustration is introduced into a field-theoretical setting in form of a small non-Abelian background gauge field. In a sense, these different approaches complement and re-enforce each other.

Furthermore, we have studied numerically a three-dimensional variant of the model exhibiting the same local parameters, like sizes of the short loops and number of neighbors. The behavior of the lattice model is close to the mean-field–type system except that the lattice variant does not have a true metastable phase which remains stable at the timescale of the simulation.

Another conclusion concerns the typical numerical hardness of the vertex-cover problem, which is equivalent to finding dense hard-sphere packings [14]: Graphs containing small loops, or geometrical frustration, are expected to be much harder to cover with a small vertex cover size than locally tree-like graphs like random graphs. This opens the interesting question as to how far local structures may influence the global hardness of a combinatorial problem.

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