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Energy-landscape network approach to the glass transition

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Abstract

We study the energy-landscape network of Lennard-Jones clusters as a model of a glass forming system. We find the stable basins and the first-order saddles connecting them, and identify them with the network nodes and links, respectively. We analyze the network properties and model the system's evolution. Using the model, we explore the system's response to varying cooling rates, and reproduce many of the glass transition properties. We also find that the static network structure gives rise to a critical temperature where a percolation transition breaks down the space of configurations into disconnected components. Finally, we discuss the possibility of studying the system mathematically with a trap model generalized to networks.

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(Some figures in this article are in colour only in the electronic version)

1. Introduction

In recent years much effort was devoted to the understanding of supercooled liquids and structural glasses, and, in particular, the structural arrest taking place at the glass transition temperature T_g [1, 2]. The numerical investigation of the dynamics of supercooled liquids and glasses is very hard due to the presence, approaching T_g , of this very slow dynamics [3]. An appealing approach for understanding this complex dynamics is to study the properties of the system's 'energy landscape': the dynamics of the system is viewed as the motion of the 'state point', described by the $3n$ -coordinates of all particles in the multi-dimensional configuration space, or landscape, of the potential energy of the system (n is the number of particles). The landscape may be partitioned into 'basins of attraction', such that local minimization of the potential energy maps any point in a basin to the same minimum. In recent years, it has been

shown that the topological details of the basins and the paths connecting them are of great importance in determining the properties of glassy systems (e.g., [4–9]).

The representation of the landscape by its basins leads to a further simplified view of the energy landscape as a network, where the nodes are the basins and the links are the saddles connecting them. The energy-landscape network of a Lennard-Jones (LJ) system has been mapped, and some of its properties were extracted [10, 11] (for energy-landscape networks in proteins and spin systems, see [12–14]). However, the influence of the topology of the network on the dynamics of the glass transition was never studied. Here, we characterize the networks of mono- and bi-disperse LJ systems obtained by minimization of the potential energy, and use a dynamical model to study properties such as response to cooling. The integration of the landscape picture with network theory provides an interpretation of the different critical temperatures of the glass transition T_0 (Vogel–Tammann–Fulcher temperature [1, 2]) and T_g , as well as an identification of a new critical temperature T_p where a second-order phase transition separates a phase where a finite fraction of the configurations is available, and a phase with a vanishing number of accessible states.

A network model for the glass transition was introduced in [4, 5]. Here, we take advantage of more sophisticated network analysis tools such as percolation theory. In particular, our approach takes into account the heterogeneity in the number of connections of each basin (i.e., its degree k), which was recently shown to be ubiquitous in nature and crucial for the understanding of many networks' properties [15].

2. The network's static properties

We start with a detailed analysis of the static properties of the energy-landscape network. We focus on isolated small LJ systems of two types: (i) monodisperse LJ system (MLJ) with $n = 12, 14$ particles. (ii) Binary 80/20 LJ mixture (BLJ) with $n = 8 + 2$ particles. Our network reflects the landscape of potential energy (not free energy), or in other words, the entropy is not taken into account, since we assume that all basins are equivalent in terms of the number of internal states they represent. We note that the sizes of the systems we study are small compared to other systems in which molecular dynamics is run [3]. However, this is inevitable since the number of nodes increases exponentially with the number of particles and thus larger systems are computationally much harder to study [10].

To construct the energy-landscape network, we look for basins, the local minima which form the network nodes, and their transition states—first-order saddles which connect two local minima and form the network links [10]. We use the LBFGS algorithm [16] to find the basins, and the eigenmode method [17] to find the saddles. Sometimes more than one first-order saddle connect two linked basins since the landscape is a high-dimensional surface. To simplify the network, we consider only the saddle with the minimum energy barrier between two linked basins. While the BLJ system is known to be glassy [3], to avoid the crystallization process usually observed in monodisperse systems, we do not consider the state of lowest energy when setting up the network [5]. Thus, the two systems are expected to be comparable in terms of their glassy behavior.

The MLJ₁₄ network consists of $N = 4193$ nodes and $M = 58\,628$ links. The potential energies of the nodes are distributed approximately normally with mean $\bar{E}(T \rightarrow \infty) = -41.5$ (figure 1(a)). The energy barriers are distributed approximately exponentially $P(\Delta E) = \frac{1}{\bar{\Delta E}} e^{-\Delta E/\bar{\Delta E}}$, where $\bar{\Delta E} = 1.64$ is the average energy barrier (figure 1(b)). As observed in [10], we confirm that the MLJ₁₄ network is *scale free* [15], i.e. the degree distribution (the probability for a node to have degree k) is broad with a tail decaying as $P(k) \sim k^{-\gamma}$ with $\gamma \approx 2.7$ (figure 1(c)). The energy of a node decreases with its degree (figure 1(d)), meaning

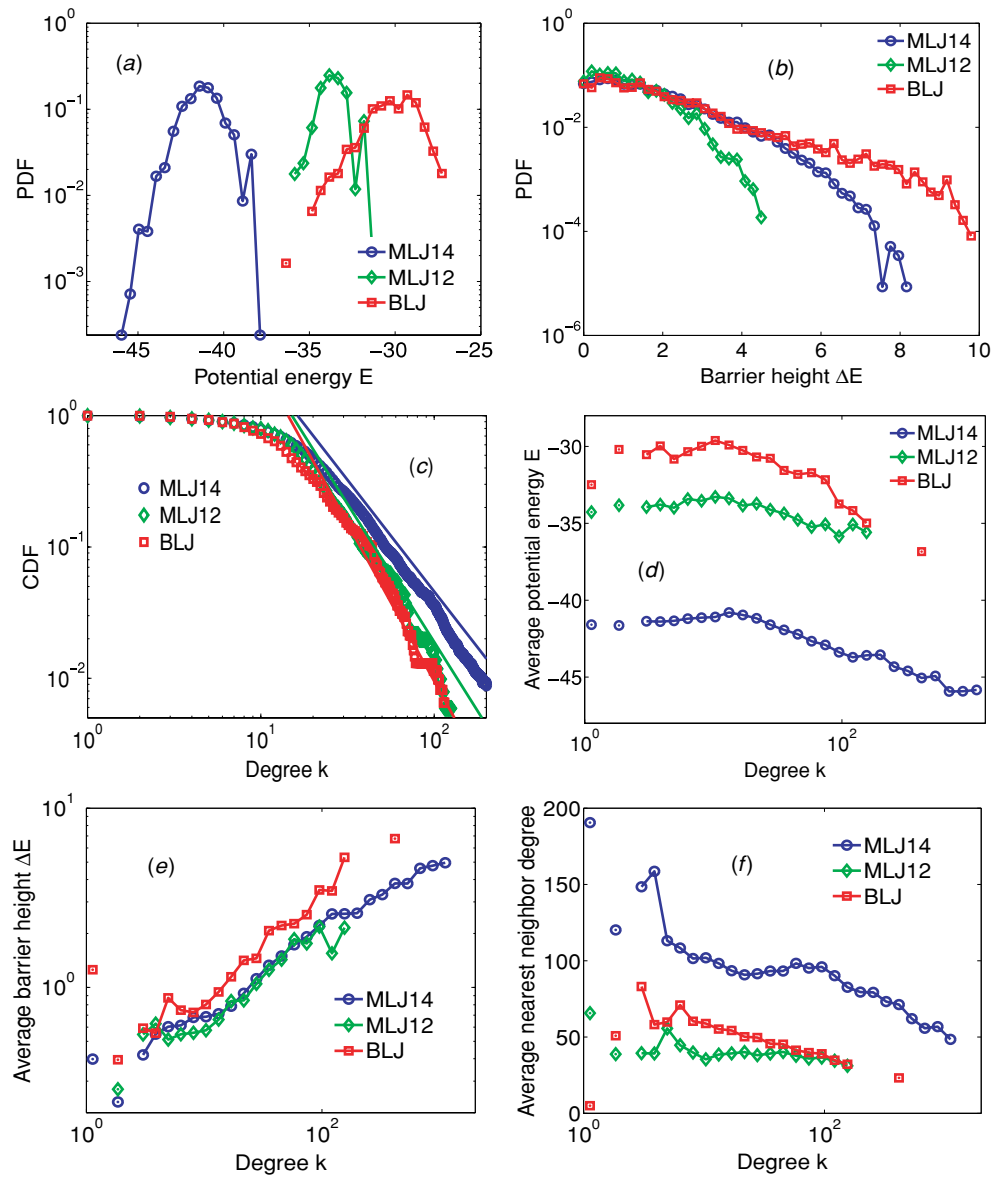


Figure 1. Properties of the Lennard-Jones energy-landscape network. Shown are the results for MLJ₁₄, MLJ₁₂ and BLJ (see the text). In all figures the data were binned and the average is plotted. (a) Distribution of potential energies E of the nodes. (b) Distribution of the heights of the energy barriers ΔE associated with the network links. (c) Cumulative distribution of node degrees k . Straight lines represent power-law decays of the form $P(k) \sim k^{-\gamma}$, with $\gamma = 2.7, 3.1, 3.4$ for MLJ₁₄, MLJ₁₂ and BLJ, respectively. (d) The average potential energy of a node E versus the degree k . (e) The average energy barrier to escape from a node ΔE versus the node degree k . (f) The average degree of node neighbors $\overline{k_{nn}}$ versus the node's degree k .

that the deepest basins can be identified with the network *hubs*, and are thus accessible to/from many other basins⁴. The average barrier height increases with the degree of the node:

⁴ Interestingly, kinetically constrained models, such as facilitated spins [31], show opposite behavior, in which the low-energy configurations are in many times frozen and have no access to any other configuration.

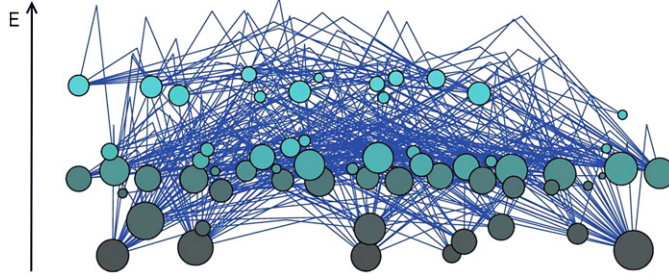


Figure 2. MLJ₁₀ network schematic. The vertical axis represents the energy, such that nodes with deeper energy are lower (and darker) in the schematic. Links' cusps correspond to the energies of the saddles, and nodes' sizes are proportional to their degree. It can be seen that highly connected nodes usually correspond to deeper basins.

$\overline{\Delta E} \sim k^\epsilon$ with $\epsilon \approx 0.43$ (figure 1(e)). The average degree of node's nearest neighbors $\overline{k_{nn}}$ slightly decreases with the node's degree (figure 1(f)), meaning hubs have many connections to low-degree nodes. We also studied 'slices' of the network in the degree (using the q -core method [18]) and energy planes. In both cases, we found that removing nodes of low degree, or high potential energy, leaves the network connected.

The MLJ₁₂ system is smaller and contains only $N = 508$ nodes and $M = 5407$ links, leading to larger fluctuations in its statistics. Yet the properties of MLJ₁₂ are qualitatively similar to MLJ₁₄. For MLJ₁₂ we find $\overline{E}(T \rightarrow \infty) = -33.87$, $\overline{\Delta E} = 1.16$, $\gamma \approx 3.1$ and $\epsilon \approx 0.41$ (figure 1). All the results reported henceforth as MLJ are for the MLJ₁₄ system, unless explicitly otherwise specified. This picture holds true also for the BLJ network, with $N = 613$ nodes and $M = 6150$ links. In the BLJ network we obtain $\overline{E}(T \rightarrow \infty) = -30.5$, $\overline{\Delta E} = 1.86$, $\gamma \approx 3.4$ and $\epsilon \approx 0.54$ (figure 1). A schematic plot of the MLJ₁₀ network is given in figure 2.

3. The network dynamics

Next we turn to a characterization of the dynamics of the system. We show that application of simple assumptions about the dynamics reproduces many features of the glass transition. At high temperatures, kinetic energy permits access to most states, while for low temperatures, mutual access among basins becomes subject to considerable activation. In low temperatures near the transition (more precisely, below the so-called dynamic glass transition temperature T_d , where an exponential number of meta-stable states appears [2]), the dynamics is dominated by rare events of collective jumps among different stable positions involving many atoms. Thus, for low temperatures, we neglect the short time dynamics which is dominated by small vibrations within the basins, and model the dynamics of the system as activated jumps between connected states. We assume the transition rate between a pair of linked states follows Arrhenius law:

$$p_{ij} = \frac{1}{N-1} e^{-\Delta E_{ij}/T}, \quad (1)$$

where ΔE_{ij} is the height of the barrier separating i and j (not necessarily equal to ΔE_{ji}) and the $1/(N-1)$ factor guarantees that the rate of leaving i , equals to $\sum_{\text{all links } (i,j)} p_{ij}$ is less than 1 for any node. Note there is considerable probability for the system to remain at the current state. This will turn useful in the characterization of the dynamical slowdown.

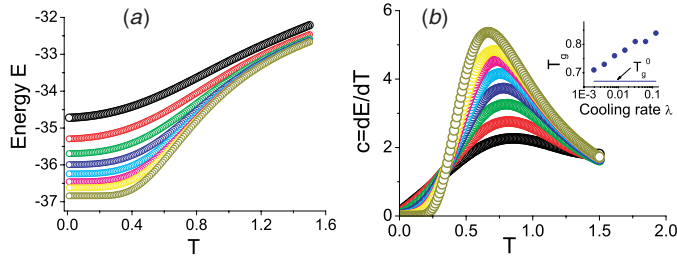


Figure 3. (a) Super-cooling in BLJ. The average energy of the system \bar{E} is plotted versus the temperature $T = T_i - \lambda t$. $T_i = 2$ and the cooling rates are (top to bottom) $\lambda = 0.01 \times \{1/8, 1/16, \dots, 1/512, 0\}$, where in each time step we iterate equation (2) once. Zero cooling rate corresponds to the equilibrium Boltzmann distribution. At $t = 0$ we assumed all states are equally probable. Similar results are found for MLJ (not shown). (b) The heat capacity $c = d\bar{E}/dT$. λ (bottom to top) is the same as in (a). Inset: the glass transition temperature T_g as a function of the cooling rate λ . The horizontal line corresponds to T_g^0 .

Experiments [1, 2] and molecular dynamics simulations [3] show that supercooling below the melting point results in a decrease in the system’s energy, up to the temperature of the glass transition T_g . At the transition, the system becomes frozen in a disordered configuration, and the rate of change of energy with respect to temperature decreases abruptly (but continuously) to a value comparable to that of a crystalline solid. We suggest that this picture, as well as the identification of the glass transition temperature T_g , can be reproduced using our simple network dynamics.

$\Phi_i(t)$, the probability of the system to be at state i at time t , evolves according to

$$\frac{d\Phi_i}{dt} = \frac{1}{N-1} \sum_{\text{all links } (i,j)} \Phi_j e^{-\Delta E_{ji}/T(t)} - \Phi_i e^{-\Delta E_{ij}/T(t)}. \quad (2)$$

We solve this set of equations numerically by iterating equation (2) once in every time step for the MLJ and BLJ networks. We use different cooling rates $T(t) = T_i - \lambda t$, where T_i is the initial temperature and λ is the cooling rate. We then calculate $\bar{E}(T) = \sum_i \Phi_i(T(t)) E_i$, where E_i is the potential energy of node i . For infinitely slow cooling, the system can be assumed to be in equilibrium, such that $\frac{d\Phi_i}{dt}$ vanishes for all i . $\bar{E}(T)$ is calculated by setting the Boltzmann distribution $\Phi_i(T) = e^{-E_i/T} / \mathcal{Z}$, where $\mathcal{Z} = \sum_i e^{-E_i/T}$. The results for BLJ, with $\lambda = 0.01 \times \{1/8, 1/16, \dots, 1/512, 0\}$, are plotted in figure 3(a). Indeed we find that our approach qualitatively reproduces the glass-forming behavior.

We then calculate the heat capacity $c = d\bar{E}/dT$ (figure 3(b)). We associate the temperature for which the heat capacity is maximal with the glass transition temperature T_g . We note that while this association is plausible, it cannot be made rigorous. As expected [1–3, 19], T_g decreases as the cooling rate becomes slower, approaching its equilibrium value $T_g^0 = 0.67(\pm 0.01)$ for BLJ (figure 3(b), inset). This value of T_g is a little higher than the glass transition temperature in a large BLJ system (≈ 0.45) [3]. For MLJ, the picture is similar, with $T_g^0 = 0.47(\pm 0.01)$.

Although in our model, microscopic transition rates follow Arrhenius law, we show below that the global relaxation times deviate from Arrhenius behavior at low temperatures, suggesting that LJ glass forming systems are fragile [1]. A global relaxation time is not naturally defined for the network. However, we note that as the system evolves in time, it explores the phase space in a random fashion, according to the transition probabilities given in equation (1). Thus, we associate the global relaxation time with the time it takes a random

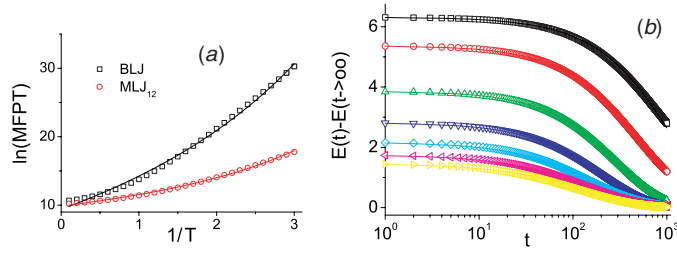


Figure 4. Dynamical properties of the energy-landscape network. (a) The mean first passage time (averaged over all sources and destinations) as a function of the inverse temperature $1/T$, for MLJ_{12} and BLJ. A super-Arrhenius behavior is observed (the slope of the curve increases with $1/T$), suggesting that the system is fragile. The lines are fits to Vogel—Tammann—Fulcher law. (b) For several temperatures in BLJ (top to bottom: $T = \{2.1, 1.8, \dots, 0.3\}$), the evolution in time of the average energy $\bar{E}(T)$ was calculated. The y-axis shows $\bar{E}(t) - \bar{E}(t \rightarrow \infty)$ (symbols), such that all curves approach zero. Curves were fitted with a stretched exponential $\bar{E}(t) - \bar{E}(t \rightarrow \infty) \propto \exp[-(t/\tau)^\beta]$ (lines). $\beta \approx 0.8$ and τ is between $[50, 700]$, and increasing with $1/T$. The picture is similar for MLJ (not shown).

walker with transition probabilities as in equation (1) starting at node i , to arrive to node j (the first passage time [20]), where i and j are randomly chosen, uniformly out of all nodes⁵. Given the network and the energy barrier heights, the average first passage time can be calculated analytically [20]. In figure 4(a), we plot the mean first passage time as a function of the inverse temperature for BLJ and MLJ. A super-Arrhenius behavior is evident, classifying these systems as a fragile glass [1]. The data seem to fit to Vogel—Tammann—Fulcher law $\tau \propto \exp[A/(T - T_0)]$ with $T_0 \approx 0.1$. However, the precise value of T_0 highly depends on the simulation details.

Time-dependent quantities can also be studied with the network. For example, the evolution of the average energy of the system at a fixed temperature can be calculated. We use equation (1) and assume that initially all states are equally probable. The results are presented in figure 4(b). For short times (up to about 10^3 time steps) the decay fits to a stretched exponential, $\bar{E}(t) - \bar{E}(t \rightarrow \infty) = A \exp[-(t/\tau)^\beta]$ with $\beta \approx 0.8 < 1$ [2]. For longer times (not shown), the decay is exponential. As in [5], the very fast relaxation, corresponding to transitions within a basin, is not represented in our model.

4. Percolation

In addition to dynamical properties, the network topology gives rise to a static critical temperature T_p , where the phase space of configurations breaks into disconnected components. This is revealed by percolation theory applied to the energy-landscape network [21]. Percolation theory is a powerful framework for the study of transport in disordered systems. In its simplest form, it is engaged in the study of conduction in a lattice in which only a fraction p of the sites, or bonds, are conducting [22–24]. This problem is relevant in various contexts in which critical phenomena take place, from superconductors and gelation to forest fires and oil searching. The theory predicts the value of a critical fraction p_c above which the bulk

⁵ The relaxation time increases with the inverse temperature in a super-Arrhenius form even if the initial sites are weighted by their equilibrium occupation probability $\exp[-E_i/T]$.

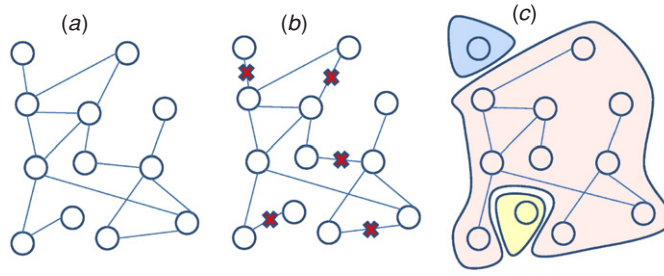


Figure 5. A schematic of network percolation. (a) The original network. (b) A fraction $q = 1/3$ (5/15) of the links are removed from the network. (c) The network after removal consists of one large cluster of 10 nodes and two small clusters of one node each.

sample is conducting, as well as the size, dimension, total conductance, diffusion coefficients and other properties of the percolation clusters.

In recent years, percolation theory has been successfully applied to networks to derive criteria for network stability. In a percolation process over a network, a fraction $q = 1 - p$ of the network links is removed [25, 26]. A percolation transition occurs when a critical fraction $q_c = 1 - p_c$ of the links is removed such that the network disintegrates. The critical point where the network breaks down is identified by a vanishing size of the largest connected cluster as well as a divergence in the size of the second largest cluster [22] (figure 5).

Pictorially, the evolution of the connectivity of the energy landscape as the temperature is lowered resembles a percolation process. At high enough temperatures, the system has sufficient thermal energy to cross most energy barriers. Thus, connected basins are accessible from each other and the network is intact. At low temperatures, links which are associated with a barrier of height $\Delta E \gg T$ can practically not be crossed and thus can be considered as absent. Thus, as the temperature is lowered, the network becomes less and less connected, until reaching the percolation threshold where it fully disintegrates. At that point, the system is frozen in an isolated region of the landscape, whose size is a zero fraction of the entire phase space. A percolation transition of the phase space has been predicted long ago for spin glasses [21]. Here, we use the network representation of LJ clusters to show explicitly how the percolation transition is realized.

Since the probability for a link to be ‘active’ decreases with a decreasing temperature, we suggest that links are excluded with probability $1 - e^{-\Delta E/T}$, where ΔE is the link’s barrier energy. This way, for high T , all links remain and the network is connected, while for low T many links are removed. We then measure (figure 6) the size of the largest and second largest cluster (where we define a cluster as a set of nodes mutually accessible from each other) for MLJ and BLJ. A percolation transition is evident at $T_p = 0.26 \pm 0.01$ for MLJ and $T_p = 0.47 \pm 0.01$ for BLJ, indicating a second-order phase transition between a phase where many configurations are available and a phase with a vanishing number of accessible states.

The percolation transition at T_p is expected to take place at the final stages of the glass transition, when barriers become almost impossible to cross, and the system freezes in the glassy state. Roughly speaking, the percolation transition temperature T_p could be associated with the Kauzmann temperature T_K . At T_K , the configurational entropies of the glass and the crystal are equal (had the glass transition not intervened), and therefore the system is bound to a single, non-crystalline, ideal glass state [1, 2]. Similarly, at T_p , the system is bound to a region of vanishing size of the phase space. In a sense, this region in phase space corresponds to the

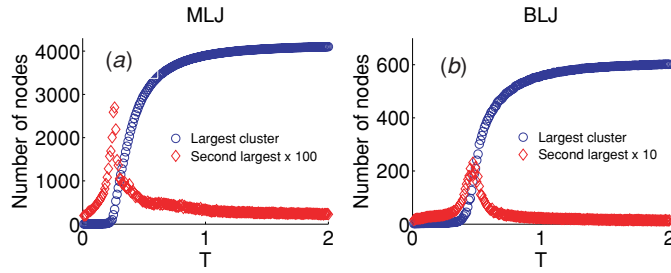


Figure 6. Percolation transition in Lennard-Jones energy landscape. (a) For MLJ system, we plot the average size of the largest and second largest cluster after the removal of each link with probability $1 - e^{-\Delta E/T}$. Clusters are strongly connected (i.e., each node can be reached from any other node in the cluster). The percolation transition takes place when the largest cluster size vanishes and the second largest cluster is maximal. (b) Same as (a) for BLJ.

ideal glass state in which the system is found at the Kauzmann temperature T_K . However, we emphasize that this correspondence is merely descriptive and cannot be made more precise.

5. Discussion

The understanding of the nature of the glass transition is a formidable task, particularly since molecular dynamics cannot approach low enough temperatures. Thus, simplified models which capture the essential properties of the phenomena are of great value. Representation of the multidimensional energy surface as a network is a particularly appealing approach, due to recently developed network analysis tools. We applied this concept here, where we studied Lennard-Jones clusters as networks of the stable basins and the links connecting them, where each link is associated with an energy barrier. We showed that the network approach qualitatively reproduces many properties of the glass transition. It is still not known whether quantitative information, such as the precise values of T_g and other temperatures can also be extracted from this kind of analysis. For that purpose, larger systems will have to be considered. The similarity, in statistical terms, between the networks of $n = 12$ and $n = 14$ encourages us to believe that similar results, at least qualitatively, will be observed in larger systems.

An alternative approach to circumvent the problem of the small system size is a mathematical model which captures the main properties of the energy-landscape network. A naive attempt would be to construct a ‘generalized trap model’ [27]. In a regular trap model the configurations of the system are fully connected, in the sense that the system can jump from any state to another. However, this is not sufficient to describe the slowing down of the dynamics, since it allows transitions which do not exist in reality [28]. In a trap model adapted to a network, each configuration is linked to precisely k other configurations, where it is particularly interesting to consider the case of a power-law distribution of degrees which is characteristic of LJ (section 2) and other systems [12]. To complete the description of the model, one can assume the distribution of energy barriers is exponential with mean ΔE (section 2). Despite the attractiveness of this simple approach, our analysis shows [29] that it leads to counterintuitive results. For example, the distribution of time τ the system remains in a configuration of degree k is a power law $P(\tau) \sim \tau^{-(1+kT/\Delta E)}$. Thus, according to model, the typical time the system stays at nodes of high degree is small, whereas it is expected that the system will spend long time at the hubs, since they are found at low potential energies (section 2). Therefore, other approaches should be sought for.

The advantage of the network approach is manifested in the application of percolation theory, which provides a natural geometrical interpretation of the structural arrest taking place at low temperatures [21]. We studied ‘bond percolation’, where we removed links in which the barrier height was high relative to the temperature, to reveal a critical temperature where the phase space breaks down into small isolated clusters. The study of glassy systems with the network approach can be further extended. For example, ageing phenomena could be studied, for either the real network or the model, by introducing more complex correlation functions. Real-space properties such as diffusion coefficients and fluctuation–dissipation relations could be studied by complementing the network with real-space information for each node. In addition, similar analysis can be pursued to other systems with complex energy landscapes such as proteins (e.g., [30]) or spin glasses [14].

Acknowledgments

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