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Particle mobility and metabasin exploration in model glass-formers

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Abstract

In this work, we assess the relevance of different range particle displacements and string motions to the metabasin (MB) dynamics of a simple glass former. We find that MB transitions are triggered by the occurrence of massive rearrangements termed as "democratic clusters" or d-clusters which last on average less than 1% of the α-relaxation time. Thus, at variance from previous investigations that focused on large displacements and strings, our work points to the essential role of medium-range particle displacements: we find that d-clusters are compact objects since the local neighborhoods of the most mobile particles exhibit significant rearrangements with a clear prevalence of medium-range displacements (of around 20% of the interparticle distance). This is in contrast to the string-like movements for which we demonstrate that many of them fully develop in isolation (their local environment being mostly insensitive to them) and at times different from that of MB transitions, being thus irrelevant to the α-relaxation. Additionally, from the analysis of the MB dynamics, preliminary support to the dynamics facilitation scenario is provided.

Keywords: Glass transition; Molecular dynamics; Supercooled liquids

1. Introduction

The landscape paradigm has become the dominant description within the vast realm of complex systems [1–4]. Within this approach, the relaxation dynamics of a system can be described as the exploration it performs of its potential energy surface (PES) [1,3,4]. For simple model glass formers (binary Lennard–Jones systems) at temperatures close to the mode coupling temperature, T_c, timescale separation [1,3–5] causes the dynamics to decompose in fast vibrations around and slower transitions between local minima or basins of attraction called inherent structures (IS). Thus, the resulting relevant dynamics is called the inherent dynamics. Similar ISs are in turn arranged in superstructures called metabasins (MB), which are relevant to the α-relaxation and where the system gets confined for a relatively long time before performing a transition to another MB, an event which entails a large-scale rearrangement [1,6–8]). This underlying influence of the

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topography on the dynamics dictates the existence of different relaxation regimes [1,5]. Particularly, at temperatures close and above T_c the system is subject to a relaxation regime characterized by the prevalence of activated dynamics, which determines the emergence of complexity and diversity (including the advent of the ubiquitous non-exponential Kohlrausch relaxation law), signatures of glassy behavior. These facts are consistent with the situation in more generic contexts (complex hierarchical systems ranging from spin-glasses to biopolymers) where a variational principle has been shown to hold valid, thus suggesting the existence of universal aspects in the relaxation of complex systems [5,9–11].

Concerning the "real dynamics" counterpart of the PES exploration events, this system has been shown to present dynamical heterogeneities [12]. The topology of such heterogeneities has been determined from molecular dynamics simulations and consist mainly in string-like motions of particles (which perform long jumps of roughly one interparticle distance and that have been demonstrated to possess a ballistic nature) [5,11]. Their connection to the topographic search has already been done [5]: These motions have been shown to represent elemental activated events in the exploration of the PES within the timescale separation scenario valid at low T (that allows the system to locally equilibrate within the ISs). In this context these activated events drive the system from one IS to another. In turn, computational evidence [6–8] showed that the IS transition events involved in the exploration of a given MB consist in many back and forth jumps of particles, being thus irrelevant to the long-range diffusion of the system. Thus, MB transition events might be paramount to the long time dynamics, which might thus be pictured as a (random) walk on the MBs [6–8]. Within this context, preliminary evidence showed that the signature of the MB transition events is not given by the large jumps (and string motions) of a few particles but to the displacements of a great number of particles leading to long-range rearrangements [8]. Thus, these results point to the fact that a very few mobile particles or strings are not enough to produce the α-relaxation but that it demands the occurrence of massive "democratic" particle rearrangements [8]. However, comprehensive studies on them are still lacking and their nature has not been revealed so far. In particular, the IS transition events that lead to escape from the MBs, together with the relevance of particle motions, jump motions and string motions to them have not been elucidated. This is the main goal of the present work. To this end, in Section 2 we present the methodology and Section 3 is devoted to study the connection between different range particle mobilities and the events relevant to the MB dynamics. Finally, Section 4 presents the conclusions.

2. Metabasin dynamics

2.1. Model and computational details

We carried out a series of molecular dynamics, MD, simulations within the NVE ensemble (but qualitatively similar results can be obtained within the NPT ensemble) for a paradigm model of fragile glass former: the binary Lennard–Jones system consisting of a three-dimensional mixture of 80% of A and 20% of B particles, the size of the A particles being 10% larger than the B ones (see Refs. [5,11] for details). We also performed periodical quenches of configurations from each given MD trajectory by potential energy minimizations [5]. This procedure maps each configuration to its corresponding IS. Transitions between ISs can be identified [5] as peaks in the squared displacement, SD, but between consecutive ISs

$$\Delta R_{\mathrm{IS}}(t) = \left(\sum_{j=1}^{N}(r_j^{\mathrm{IS}}(t+\Delta t) - r_j^{\mathrm{IS}}(t))^2\right),$$

where $r_j^{\mathrm{IS}}(t)$ is the position of particle j in the IS that occurs at time t and Δt is the interval in MD steps between successive quenching minimizations. This is so since while the system is in a given basin of attraction, all minima map to the same IS. Many of these IS transitions can be shown to be due to string motions [5].

MBs have been detected and characterized recently for this system by a interval bisection and a time interval bracketing method and the long time dynamics of the system has been found to be well described as a random walk on the MBs [6]. In previous works [7,8], we studied the MB superstructure of the PES by means of a different method to locate MBs: the distance matrix, DM, method (a method which has the advantages of being simpler and to contain more direct graphical information). This approach led us to the discovery that

MB transitions are triggered by "democratic" events entailing the fast, concerted rearrangement of a significant amount of particles arranged in a compact cluster. These clusters, the d-clusters, are thus responsible for the MB transitions and thus, for the α-relaxation and the long time dynamics of the system.

2.2. The analysis of metabasins: the distance matrix method

This method, first introduced for liquid water [13], represents a powerful representational tool to look for MBs [7,8,13]. It consists in plotting the matrix of mean-squared distances between a set of ISs recorded along a MD trajectory of the system [8]. We have also showed that this study can also be performed at the real dynamics, that is, without recalling to the IS formalism [7]. However, now we shall use the inherent dynamics since it is better suited to study particle hoppings since the interference of thermal vibrations is removed. Thus, we calculated the ΔR_{IS} between all the ISs obtained from periodical quenches of a given MD trajectory as above indicated, that is, we constructed a distance matrix

$$\Delta R^2_{\mathrm{IS}}(t',t'') = 1/N \sum_i (r_i(t') - r_i(t''))^2,$$

where $r_i(t)$ is the position of particle i at time t. Thus $\Delta R^2_{\mathrm{IS}}(t',t'')$ gives the system averaged squared displacement of a particle in the time interval that starts at t' and ends at t''. Note that the time average of $\Delta R^2_{\mathrm{IS}}(t',t'+\Delta t)$ over t' gives the r-average of $G_s^{\mathrm{IS}}\,(r,\Delta t)$, the self-part of the van Hove correlation function for time displacement Δt evaluated for the ISs, which gives the probability of finding a particle at a given distance of the origin at a given time Δt (the time interval between successive quenched configurations). The same is true if one averages over a very large system. For this study, as always we need to deal with MBs, we must investigate small systems, since for large systems the results originated from different subsystems would obscure the conclusions [7,8,13]. Thus, we used 150 particles. However, we also obtained the same qualitative results for small subsystems immersed in a big one, thus ruling out the possibility for finite size effects. Fig. 1.a shows a typical example for a run with $T = 0.469$ and $P = 2.296$. At this T we recorded ISs in intervals of $\Delta t = 6.21$ in reduced units (the timestep for the runs is always 0.002). This time is 10% of t^* (the timescale of maximum inhomogeneous behavior which marks the end of the β and the beginning of the α-relaxation) and corresponds to 2500 MD steps. In the work of ref. [6] the exhaustive finding of all ISs was required. In our case, however, this is not a requisite and when we speak of consecutive ISs we refer to ISs separated in time 10% of t^* (and do not imply the absence of other IS transitions between them). The gray level of the squares in the DM depends on the distance between the corresponding ISs, the darker the shading indicating the lower the distance between them. From the island structure of this matrix a clear MB structure of the landscape is evident. That is, islands are made up of closely related ISs (low ΔR^2_{IS}), which are separated, from the ISs of other islands by large distances. We can estimate the typical residence time in the MBs for this T (from island sizes) as qualitatively on the order of t^*. Given the small system size we expect this to be a good estimate (however, this timescale clearly depends on system size, since for a large system different subsystems would be undergoing MB transition events at different times). Thus, intra-MB transitions (which comprise many transitions between different ISs and string motions of particles) are fast compared to the transitions between MBs, as evidenced by the large MB residence times.

In Fig. 1(b) we also show the ΔR^2_{IS} curve with respect to the first IS (that is, a non-log or linear plot of the squared displacement from the origin at the IS level) together with the average particle displacement between consecutive ISs and the number of particles whose displacement exceeds $0.2\sigma_{\mathrm{AA}}$ (with σ_{AA} the LJ parameter for the A particles). From this figure we can learn that the mark of MB dynamics in the ΔR^2_{IS} curve is given by the presence of well-defined plateaus separated by relatively sharp steps or jumps. The plateaus are indication of MB confinement while the steps correspond to MB transition events. A recent work [6] studied transitions between MBs but characterized them by the trajectories joining the lowest-energy ISs of the corresponding MBs, or by a series of IS transitions. However, here we find that most of the MB exploration events are irrelevant to the process of escape from the MB. Thus, we identify the MB transition events as the escape events themselves, which we find that are very fast compared to the MB exploration timescale. The total length of the run of Fig. 1 is approximately the α-relaxation time. Thus, we have roughly 5–10 MB transition events within the α-relaxation.

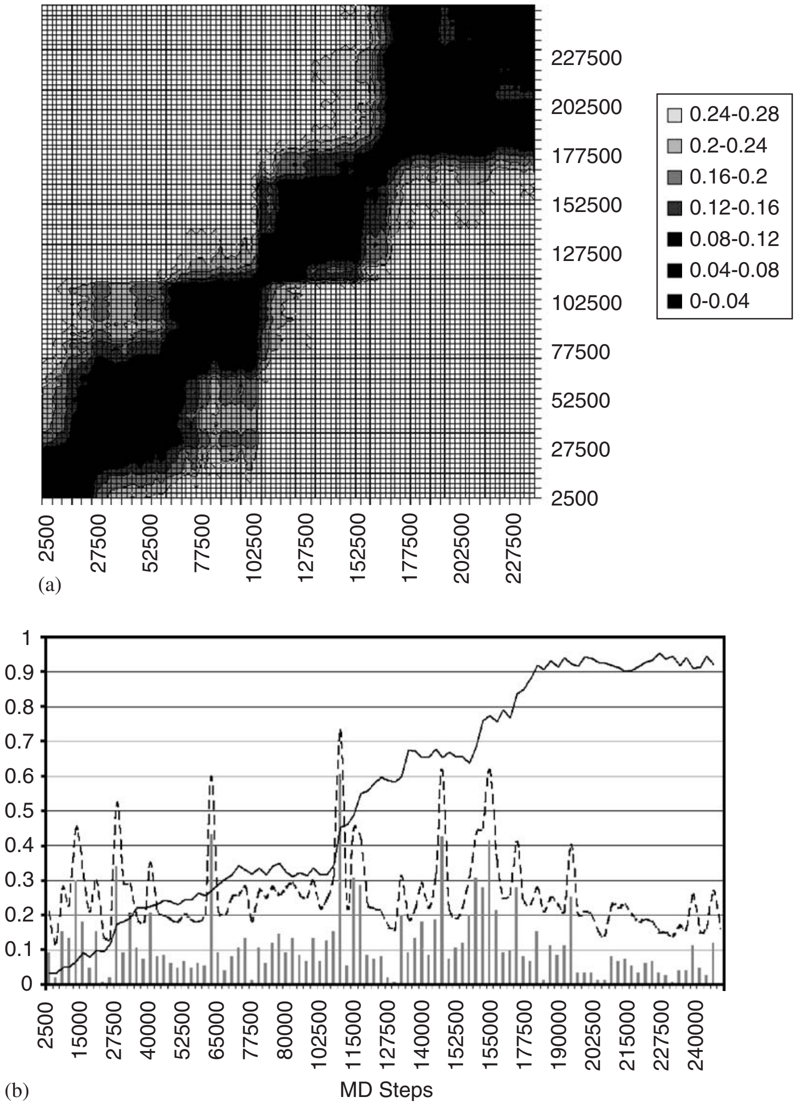


Fig. 1. (a) A typical distance matrix, ΔR^2_{IS}, for the system at $T = 0.469$. The time interval between successive recorded ISs is time is $\Delta t = 6.21$ in reduced units. This time is 10% of t^* and corresponds to 2500 MD steps (the time units we shall use from now on). A total of 100 configurations are recorded, thus giving a total run time of 250 000 MD steps. (b) Solid line: Averaged squared displacement plot; dashed line: Average particle displacement between successive ISs. Bars: Number of particles whose displacement is greater than $0.2\sigma_{\mathrm{AA}}$ between successive ISs.

From Fig. 1(b) we can also see that these MB transition events are regions with one or more peaks in the average displacement plot indicating an enhanced mobility leading to long-range rearrangements. In this sense, the most striking point arises from the inspection of the curve for the number of particles that show displacements greater than $0.2\sigma_{\mathrm{AA}}$ between consecutive ISs. Thus, Fig. 1(b) shows the connection between overall "mobility" and number of particles moving. From it we can learn that in the MB transition regions a great fraction of the 150 particles show displacements greater than $0.2\sigma_{\mathrm{AA}}$ between consecutive ISs. Thus, the fast increase in the squared displacement plot is not related to a few particles that perform large displacements but is due to the motion of a great number of particles. The choice of $0.2\sigma_{\mathrm{AA}}$ is arbitrary (the reason for it shall be given later on), but similar results were obtained when we used different threshold values.

To summarize, these results clearly confirm the MB superstructure of the PES for model glass formers (binary Lennard–Jones systems) and demonstrate the seminal relevance of MB transition events to the long time dynamics of the system. We characterized such events and demonstrated their relation to particle mobility: contrary to the situation at the IS level where a few large jumps of particles (usually involving string like movements) are influential while the rest of particles do not contribute appreciably, MB transition events demand sharp "democratic" large-scale rearrangements involving modest medium-range displacements of a great portion of the sample. These large-scale compact d-clusters could be potential candidates for the cooperatively rearranging regions proposed long ago by Adam & Gibbs.

A detailed study of particle mobility related to the events described for MB dynamics has not been performed so far, and this is the aim of next section. The former results indicate that the α-relaxation is not directly related to strings but to massive rearrangements associated with the elastic relaxation of the sample [7]. However, as we shall see in Section 3, since such MB transition regions do present some very mobile particles and even string motions, it is not easy to distinguish what is cause or effect. Nevertheless, when we study certain pairs of consecutive ISs (always separated a $10\%t^*$ time interval) but within the plateaus or MBs (as identified in the squared displacement plot of Fig. 1(b)) we find a few particles with great displacements and string motions. However, contrary to the situation for consecutive ISs in MB transition regions where a great percentage of the particles exhibit mobilities of around $0.2\sigma_{AA}$, these mobile particles do not need a massive rearrangement of the sample. That is, most of the particles are not affected by these large displacements and only perform negligibly small displacements. Moreover, when we consider ISs separated by larger times (times close to t^* but both within a plateau) we also find some large displacements and string movements but still only a few amount of particles show displacements greater than $0.2\sigma_{AA}$ (that is, the mobility distribution is not very different from the one depicted by the solid line in Fig. 2). These points will be addressed in detail in Section 3.

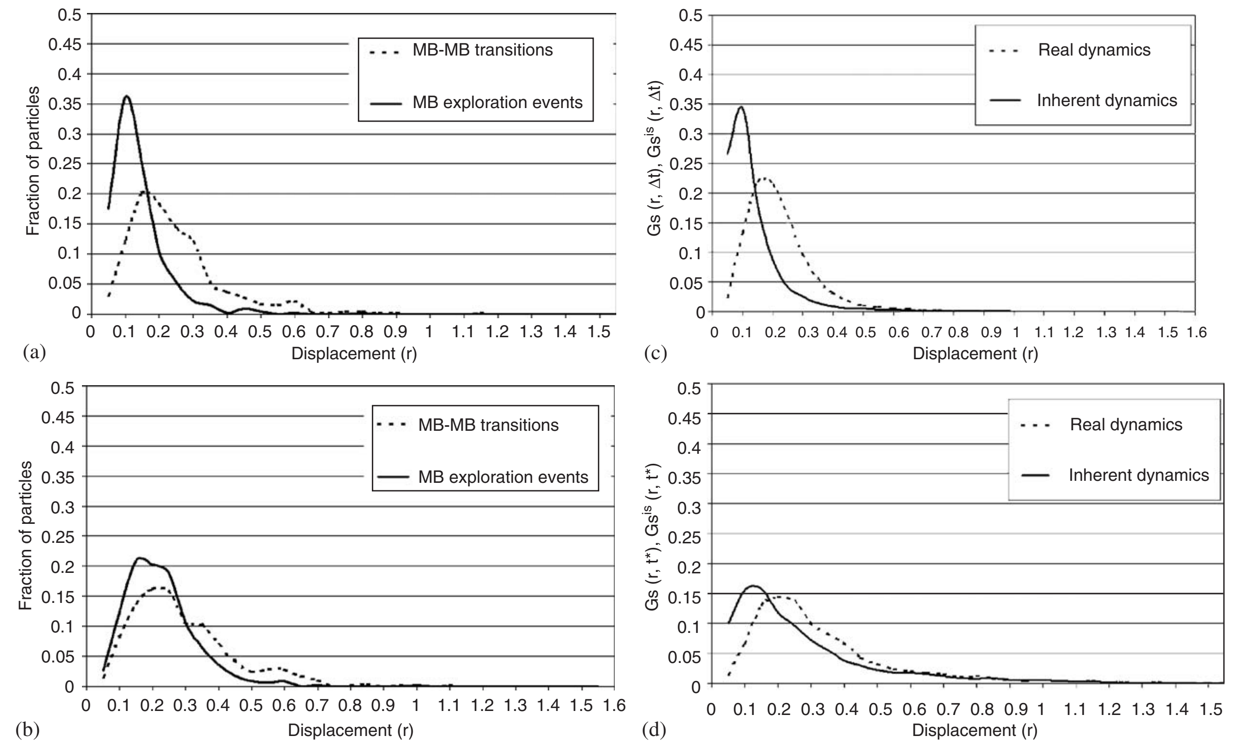


Fig. 2. Distribution of particle displacements for Δt time intervals (between successive ISs). (a) Solid line: For Δt time intervals corresponding to MB exploration events (the distribution implies an average over typical events of the run of Fig. 1). Dashed line: For time intervals corresponding to MB–MB transition events (the results also represent averages over the MB transitions of Fig. 1). (b) Idem to (a) but for the real dynamics (that is, by using directly the instantaneous configurations instead of the ISs). (c) Plot of the self-part of the van Hove function, $G_s^{IS}(r, \Delta t)$ (for the IS dynamics), and $G_s(r, \Delta t)$ (for the real dynamics). (d) Plot of $G_s^{IS}(r, t^*)$ and $G_s(r, t^*)$.

3. Particle mobility and metabasin dynamics

In the previous section, we identified the regimes where the system locally explores the MBs and the sharp events that mark the transition form one MB to another, the democratic events. However, the nature of the particle movements that characterize such events has not been elucidated. In particular, the relevance and interplay between particle movements of different range in each regime needs to be assessed. The distribution of particle mobilities in the different kind of events mentioned is shown in Fig. 2(a). The solid line depicts the distribution function of particle displacements for pairs of consecutive ISs chosen within the MBs (which we shall call MB exploration events). The dashed line displays the same distribution function but at MB–MB transitions. In both cases we averaged over different pairs of consecutive ISs of the run of Fig. 1 on the corresponding kind of region (MB explorations and MB–MB transitions). We note that the distribution function for the MB–MB transitions is calculated for the pairs of consecutive ISs at MB–MB transitions (that ones with a high number of particles with displacements greater than $0.2\sigma_{AA}$ in Fig. 1(b)). If we incorporate to the calculation pairs of ISs close but not exactly at the MB transitions the curve changes significantly, a fact that points to the sharpness of the democratic event. Fig. 2(b) shows the situation at the real dynamics for the same cases shown in Fig. 2(a) (that is, by using the instantaneous configurations instead of the corresponding IS structures obtained by their minimization). We also show in Fig. 2(c) the function $G_s^{IS}(r, t, t+\Delta t)$ averaged over all the pairs of consecutive ISs, that is, $G_s^{IS}(r, \Delta t)$, the Inherent Dynamics counterpart (calculated over the ISs of the trajectory) of the self-part of the van Hove function at $t = \Delta t = 10\% t^*$ (the van Hove function for the real dynamics is also provided for comparison). Finally, Fig. 2(d) displays the situation for $G_s\,(r, t^*)$ and $G_s^{IS}\,(r, t^*)$, the self-part of the van Hove functions at $t = t^*$.

By comparing the different figures for the Inherent Dynamics and the real dynamics we can see that they are qualitatively similar but that the quenching process notably makes the functions sharpen by eliminating the thermal vibration effects (more importantly for the caging regime of the dynamics). Thus, at variance from our previous work [7], in this work we shall focus at the IS dynamics in order to study the role of different range particle mobilities to MB dynamics, since the real dynamics does not allow to obtain clear results. From Fig. 2(a) we can see that MB transitions display an enhanced particle mobility with respect to the events that locally explore MBs. Additionally, the fact that the solid line of Fig. 2(a) is very similar to Fig. 2(c) indicates that the contribution of local MB exploration events dominate the van Hove function and thus, that MB transitions are fast, rare events. These curves also demonstrate that MB transitions are due to the democratic motions of many particles that perform medium-range displacements and not to the presence of a few very mobile ones. Thus, the distribution for the MB–MB transitions (dashed line of Fig. 2(a)) is much broader than that of the MB exploration events (Fig. 2(a) solid line) and than the van Hove function (Fig. 2(c)). This curve shows a maximum close to $0.2\sigma_{AA}$ (for this reason we used 0.2 as threshold or cutoff in Fig. 1(b)). Thus, modest albeit significant displacements dominate the distribution. In any case, the number of particles performing large displacements (at this temperature the mobility condition in the real dynamics is around $0.65\sigma_{AA}$) is low. Moreover, the contribution of these very mobile particles to the global value of the squared displacement between consecutive ISs (that is, the jumps in Fig. 1(b)) is always poor. Thus, MB transition events are not marked by the large displacements of a few particles. Rather, the signature of MB transitions is the occurrence of "democratic" massive rearrangements entailing the medium-range displacements of a great portion of the particles. Each of these particle displacements would be judged irrelevant if considered isolated and in terms of their length. In turn, the solid line of Fig. 2 (and also Fig. 2(c)) display sharp distributions with most of the particles performing very small displacements between 0.05 and $0.1\,\sigma_{AA}$ and with only a few amount of particles exhibiting displacements close to or greater than $0.2\sigma_{AA}$ (some large displacements are also present but with low probability).

Fig. 2(a) also justifies our choice made in Section 2 of a mobility of $0.2\sigma_{AA}$ as a cutoff for the particles of the d-clusters: at such value the distribution function for the MB transition events exceeds the function for the MB exploration events. From Fig. 2(c) we can also see that the value of the Inherent dynamics van Hove function above $r = 0.2\sigma_{AA}$ at the time interval $\Delta t = 10\% t^*$ is low, a point that gives additional justification for our prior election of such value to characterize medium-range displacements (modest albeit significant displacements). Additionally, the existence of certain similarity between the distribution function given by the dashed line of Fig. 2(a) (for MB transition events that lasts $10\% t^*$) and 2d (the van Hove function at t^* that is, for a time interval 10 times longer) indicates that MB transition events dominate the global mobility of the system. That is, during the full exploration of a MB (whose mean residence time is around t^* [7,8]) a few particles are very mobile but there is not a big number of particles with mobilities larger than $0.2\sigma_{AA}$. Thus, the main contribution to the mobility exhibited by the van Hove function evaluated at t^* comes out from the MB transitions (since on average one MB transition occurs within any given t^* time interval).

We note that Fig. 2(a), dashed line, presents a shoulder at displacements a bit larger than $0.2\sigma_{AA}$. This might indeed be evidence of the presence of a close second peak. Since the d-clusters involve 20–40% of the particles we think that we can divide the system at the MB–MB transitions in roughly two kinds of particles: ''mobile'' and ''immobile'' if they belong to the region of the sample where the d-cluster occurs or not, respectively. The displacement distribution for the first group presents a peak close to but above $0.2\sigma_{AA}$, while the second group presents a behavior close to that prescribed by the van Hove function (the same behavior exhibited by all the particles at time intervals within the MBs or islands) since they are not mainly affected by the d-cluster. Thus, this group shows a maximum between 0.1 and $0.15\sigma_{AA}$. Thus, the overall behavior displayed by Fig. 2(a), dashed line, represents a convolution of both peaks.

We now analyze the role of different range motions to the MB exploration and transition events. First, we classify particles according to their mobility. We shall say that ''mobile'' particles (or particles with medium range mobility) are those whose mobility in a Δt time span (that is, $10\% t^*$) in the inherent dynamic is greater than $0.2\sigma_{AA}$ (the election of this value has been justified above). We shall also classify as ''very mobile'' particles those whose displacement in such conditions is greater than $0.5\sigma_{AA}$. We remember that for this system (at the same temperature, pressure and density), previous studies [5,12] defined mobile particles as that performing displacements greater than around $0.65\sigma_{AA}$ for the real dynamics and after a t^* time interval (10 times greater than the one we are considering here) but that in previous works [5] we have found displacements of such length to occur in the real dynamics much faster than that (at ballistic times). However, from Fig. 2 we can note that the van Hove functions for the Inherent dynamics are shifted to the left with respect to the ones of the real dynamics (since the thermal motion is removed by the quenching), so $0.5\sigma_{AA}$ seems to be a good choice for very mobile particles at the inherent dynamics. These very mobile particles can thus be related to the very mobile particles and strings of previous studies of the real dynamics. This election is also based on the observation of the development of a small peak in Fig. 2(a) (dashed line) for the MB transitions at approximately $r = 0.5\sigma_{AA}$.

Integrating Fig. 2 for the cases of interest we can learn that the percentage of mobile particles is roughly 10% while around 1% of the particles are very mobile, always for consecutive ISs within the MBs (MB exploration events). However, for consecutive ISs in MB transitions the percentages are approximately 45% and 5%, respectively. Thus, both values are small for MB exploration events while only the percentage of mobile particles is big for MB transitions. This again speaks of an enhanced mobility at MB transitions and points to the relevance of medium-range displacements to these events.

Both for Δt time intervals (between pairs of consecutive ISs) within the MB transitions and in a small set of Δt time intervals within the MBs we found a few very mobile particles and even short strings. That very mobile particles and even strings be found within MBs where the number of mobile particles (mobility greater than $0.2\sigma_{AA}$) is low, points to the fact that these events do not necessarily require the concurrent rearrangement of the local environment. This point is evident from a study of the neighborhood of such particles. Thus, we looked at particles within a distance of $1.5\sigma_{AA}$ of very mobile particles (mobility greater than $0.5\sigma_{AA}$) and studied their mobility in such time interval. This cutoff implies looking at the nearest neighbors since it lies within the minimum after the first peak of the radial distribution function of both kind of particles, A and B. For first neighbors of very mobile particles within MB transitions we found that approximately 65% of them were mobile (mobility greater than $0.2\sigma_{AA}$) while for very mobile particles within the MB this percentage dropped to around 25%. This included isolated particles and also some very mobile particles participating in short strings. This fact means that very mobile particles and strings do not require by themselves an important local rearrangement of their neighborhood for MB exploration events. This situation is contrary to that in MB transition events where compact d-clusters of mobile particles emerge. Again, the existence of a few very mobile particles and strings is not enough to trigger MB transitions and thus, the α-relaxation of the sample. In Fig. 3 we show examples of short strings found both in MB transitions and within MBs (always in Δt time

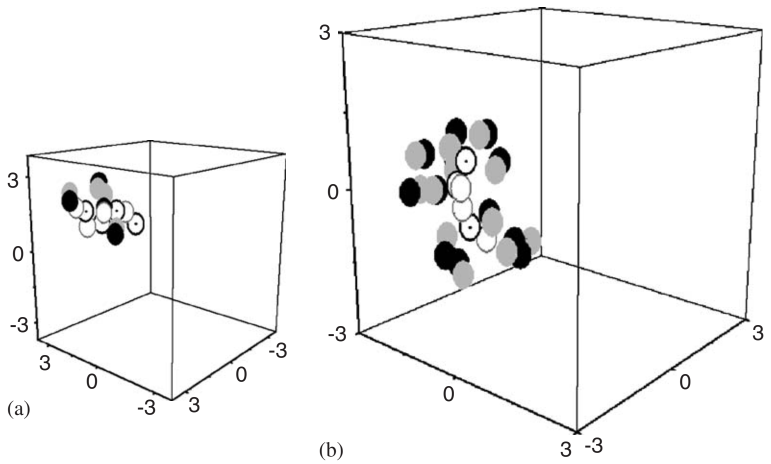
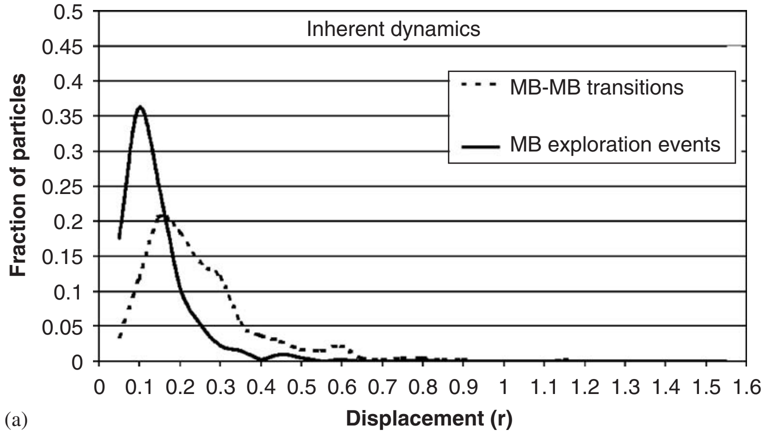


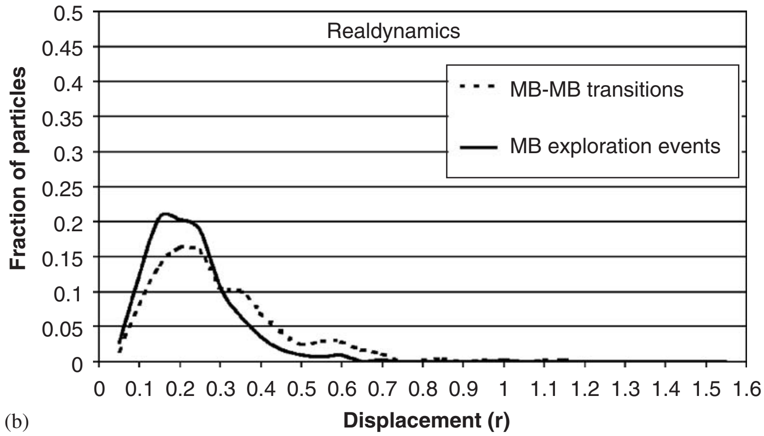
Fig. 3. (a) An example of a short string (of very mobile particles) at a MB exploration event, displaying the displacements of the first neighbors of the particles comprised. The very mobile particles are indicated with white spheres with a central dot (initial position) and white spheres (final position). Their first neighbors which are mobile are indicated with black spheres (initial position) and gray spheres (final position). (b) Idem to (a) but for a string within a MB–MB transition event. Note the enhancement in the number of mobile first neighbors.

intervals), together with the displacements of the mobile particles (mobility greater than $0.2\sigma_{AA}$) of their neighborhood.

To state the difference between the local environments of very mobile particles at MB transitions and MB exploration events quantitatively, we studied the distribution of displacements of the nearest neighbors of such particles in both kinds of events. In Fig. 4 we show the results for the MB exploration events and MB transitions indicated in Fig. 2. Again, the results are similar for the IS and real dynamics (with the curves for the real dynamics being broader and displaced to the right and the results for the IS dynamics being much clearer). A first point that comes out from the great similarity between Figs. 2(a),(b) and 4(a),(b) is that nothing special occurs around the very mobile particles (we note that in Fig. 2 we calculated the global particle displacements distribution while in Fig. 4 we restrict the calculation to the first neighbors of very mobile particles). That is, the distribution of particle displacements around very mobile particles (of their first neighbors) is practically identical to the global distribution of particle mobility. To be around a very mobile particle (which do not represent a significant percentage of the particles neither at MB exploration events nor at MB transitions, as can be seen from Fig. 2) represents no difference than being anywhere else in the sample in which regards to mobility. This means that very mobile particles play no special role at promoting local rearrangements. Again, as in Fig. 2, focusing on the IS dynamics we can see that the situation is notably very different for MB transitions and MB exploration events since for the last ones the distribution is narrow with a peak at a value far below $0.2\sigma_{AA}$ while for the former ones the distribution is much broader and the peak is displaced to the right indicating the presence of many mobile particles. Thus, very mobile particles (and strings) in MB–MB transition events imply the concurrent rearrangement of their local neighborhood including many mobile particles (in other words, this is the signature of being within a d-cluster, around a very mobile particle or anywhere else). This point reveals the compactness of the d-clusters responsible for the MB–MB transitions. Here again, as in Fig. 2, not many very mobile particles are found in the neighborhood of other very mobile ones, but displacements of many ranges are observed with a clear maximum around 0.15–$0.2\sigma_{AA}$. Thus, no prevalence of very mobile particles is observed in MB–MB transitions but medium range displacements dominate particle mobility. In turn, very mobile particles (and strings) in MB exploration events do not imply major changes in their local neighborhood and they both appear and die in isolation, without generating d-clusters of mobility. We also note that most of the mobile particles in MB transitions (whose total number for each MB transition is depicted in Fig. 1(b)) are first neighbors of very mobile particles. The percentage amounts to approximately 80% (while the percentage drops notably for MB exploration events), a fact that also demonstrates the compactness of d-clusters.



(a)



(b)

Fig. 4. (a) Distribution of displacements of the first neighbors of very mobile particles in MB exploration events in the IS dynamics (solid line) and MB–MB transition events (dashed line) in the IS dynamics (the distributions imply averages over typical events of the run of Fig. 1). (b) Idem to (a) but for the real dynamics.

Finally, we studied the three-dimensional evolution of mobility during the MB dynamics. In Fig. 5 we plot the 3D positions of very mobile particles for a set of plateau—MB transition–plateau, where the plateaus represent the full exploration of the MBs. That is, the mobility in the plateaus is now considered during all the time they last (the fulltime the system is confined within the local MB) and not in $\Delta t = 10\% t^*$ time intervals. We have noticed that the mobility developed by the system in the timescale Δt of a MB transition is similar to the total mobility during a whole plateau (around 10 times larger timescales). Very mobile particles within consecutive plateaus are confined each to certain regions of the sample while the ones of the MB transitions seem to be more extended and to link both plateaus. An interesting proposal [14] is the dynamic facilitation scenario where an space–time history of mobile regions should emerge since mobile regions are facilitated by the prior mobility of a neighboring one. Our preliminary results of Fig. 5 seem to be in favor of such scenario. However, here we are only studying a small system. In a big system, different subsystems or regions of the sample undergo MB transitions (developing d-clusters) at different times, making possible a richer behavior than the one that can emerge in a small system. In any case, from Fig. 5 we can learn that successive mobile regions (plateau or MB transitions) are close in space and that many very mobile particles of a given region are first neighbors of particles, which have been very mobile in the previous one.

4. Conclusions

The main result of this work is to evidence the basic connection of medium-range particle displacements and the α-relaxation of a simple glass former. These results demonstrate that MB transition events are signed by a

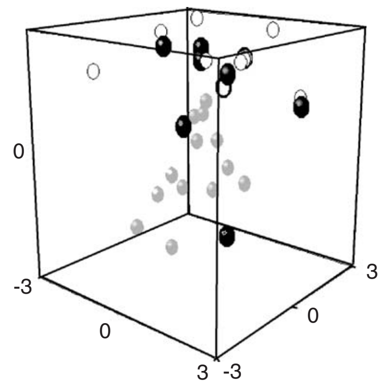


Fig. 5. 3D positions of very mobile particles for a set of plateau—MB transition—plateau. The particles are drawn at their initial position in the corresponding time interval. Each plateau and transition is indicated by the times in MD steps of its first and last IS. The color of the particles of the plateaus and MB transitions are as follows: white spheres denote particles in the plateau that goes from 30 000 to 65 000, black spheres indicate particles belonging to the MB–MB transition within [65 000–67 500] and gray spheres indicate particles corresponding to the plateau that goes from 67 500 to 110 000.

great enhancement of medium-range mobility of particles arranged in compact d-clusters. We show that these events dominate the α-relaxation since it does not suffice to have a few very mobile particles in order to produce relevant relaxation events. We also characterized the local environment of very mobile particles and strings within both MB exploration events and MB transitions: While in the latter their local neighborhood exhibits a great number of medium-range displacements, in the former they occur without being mainly "felt" by the rest of the sample. Thus, very mobile particles and strings within MB exploration events are born and die in isolation, without any connection to α-relaxation events. Finally, the inspection of the time evolution of mobility reveals that certain space proximity seems to hold for successive mobile regions, thus providing preliminary support to the dynamical facilitation picture.

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