Local properties of the potential-energy landscape of a model glass: Understanding the low-temperature anomalies

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Though the existence of two-level systems (TLS) is widely accepted to explain low temperature anomalies in the sound absorption, heat capacity, thermal conductivity and other quantities, an exact description of their microscopic nature is still lacking. We performed computer simulations for a binary Lennard-Jones system, using a newly developed algorithm to locate double-well potentials (DWP) and thus two-level systems on a systematic basis. We show that the intrinsic limitations of computer simulations like finite time and finite size problems do not hamper this analysis. We discuss how the DWP are embedded in the total potential energy landscape. It turns out that most DWP are connected to the dynamics of the smaller particles and that these DWP are rather localized. However, DWP related to the larger particles are more collective.

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I. INTRODUCTION

It is well known that most kinds of disordered solids show anomalous behavior at very low temperatures as compared to their crystalline counterparts. Many of the observed features can be explained by the Standard Tunneling Model (STM)[1,2] and its generalization which is the Soft-Potential Model.[3–6] The basic idea of the STM is the existence of a broad distribution of Two Level Systems (TLS). The TLS can couple to strain and electric fields and therefore influence quantities like the heat capacity, thermal conductivity, sound absorption and dielectric response; see Ref. 7 for a review. The STM predicts a linear dependence of the heat capacity on temperature and a quadratic dependence of the thermal conductivity on temperature. The STM gives a good general agreement with experimental results down to temperatures around 100 mK.

So far it has not been possible to derive a theory of the glass transition or of the low-temperature anomalies from first principles, i.e., from the Hamiltonian of the glassy system. This means, that, except for the few cases where possible TLS have been successfully identified by means of computer simulations,[8,9] the STM and the recent developments, mentioned above, are almost purely phenomenological. In particular researchers wanted to explain why the nearly constant ratio of the density of TLS and their coupling to phonons, if compared for very different glasses, is so similar.[10] Already one decade ago this has been interpreted by Yu and Leggett[11,12] and Coppersmith[13] as an indication that the observed TLS are highly collective excitations of many underlying microscopic TLS, resulting from the interaction among TLS. In the meantime, however, it has been shown that the interaction is only relevant in the mK regime.[14–16] An alternative scenario has been proposed by Lubchenko and Wolynes.[17] They consider the glass as a mosaic of frustrated domain walls, separating individual cells. In their model the collective tunneling process finally involves $O(10^2)$ molecules which only move a fraction d_L/a of a nearest-neighbor distance $((d_L/a)^2 \approx 0.01)$. Unfortunately, the object of these theories, namely the TLS, are still somewhat obscure because experimentally it is very difficult to characterize their microscopic nature.

Formally, a TLS corresponds to a pair of local minima, or a double-well potential (DWP), on the potential energy landscape (PEL). The minima need to have an energy difference less than $k_B T$ and a small distance in configuration space because otherwise no tunneling would occur. One may wonder whether computer simulations might help to elucidate the relevant properties of TLS and thus to prove their existence. Computer simulations are strongly limited in several directions: (a) Typically rather small systems have to be used to analyze the PEL. This may give rise to significant finite size effects; (b) due to finite simulation times the cooling process is extremely fast so that the resulting glassy structure may be vastly different as compared to the experimental situation. This may strongly influence the properties of the TLS obtained by computer simulations; (c) due to possible imperfections of the search algorithm to identify TLS one may possibly miss a significant fraction of TLS.

In this contribution we will show that these problems are not relevant for the problem of locating TLS and that it is indeed possible to obtain detailed and unbiased information about the nature of TLS. A particular challenge is the systematic search of TLS. Most of the early computer simulations in this field[18–22] did not attempt to systematically find DWP. In this work we present computer simulations on a model glass former (binary Lennard-Jones) to systematically identify the TLS. A first step in this direction has been already published a decade ago.[23–25] At that time, however, it was not possible to exclude that any of the above-mentioned problems might hamper the analysis. With improved algorithms and faster computers this has become possible nowadays. Furthermore the TLS are related to the properties of the glass transition. Qualitatively, one may say that TLS probe the PEL on a very local scale whereas for the understanding of the glass transition much larger regions of the PEL are relevant. Here we would like to mention that in recent years computer simulations succeeded in extracting many important features of the PEL of supercooled liquids.[26–34]

II. TECHNICAL

A. Computational details

As a model glass former we chose a binary mixture Lennard-Jones system with 80% A-particles and 20% B-particles (BMLJ).[35–38] It is supposed to represent nickel-phosphorous (80% ^{62}Ni; 20% ^{31}P)[39] but with a 20% higher particle density, this system was first used by Kob and Anderson. The used potential is of the type

$$V_{\alpha\beta} = 4 \cdot \epsilon_{\alpha\beta}[(\sigma_{\alpha\beta}/r)^{12} - (\sigma_{\alpha\beta}/r)^6] + (a + b \cdot r), \quad (1)$$

with $\sigma_{AB} = 0.8\sigma_{AA}$, $\sigma_{BB} = 0.88\sigma_{AA}$, $\epsilon_{AB} = 1.5\epsilon_{AA}$, $\epsilon_{BB} = 0.5\epsilon_{AA}$, $m_B = 0.5m_A$. Periodic boundary conditions were used and the linear function $a + b \cdot r$ was added to ensure continuous energies and forces at the cutoff $r_c = 1.8$. The units of length, mass and energy are σ_{AA}, m_A, ϵ_{AA}, the time step within these units was set to 0.01. The simulation cell was a cube with a fixed edge length according to the number of particles and an exact particle density of $D = 1.2$. Molecular dynamics (MD) simulations, using the velocity Verlet algorithm, have been used to generate independent configurations and as part of the DWP location algorithm. For the case of Nickel-Phosphorous the energy unit corresponds to 933.9 K and σ_{AA} is 2.2 Å. We analyzed $N = 65$, 2×65, 130, 2×130, 195, 260 particle systems. The 2×65 and the 2×130 systems denote two noninteracting systems sharing one simulation box.

It is known for the same system that the dynamics above the mode-coupling temperature T_c as well as structural properties like the pair correlation function are basically independent of system size for $N \geq 65$.[29,38] Therefore one may hope that even for the very small systems finite size effects for the TLS can be neglected. It will turn out that this is indeed the case.

B. Systematic location of TLS

We have developed a new algorithm for a systematic search of TLS. Formally, the problem is to identify two nearby local minima on the potential energy landscape (PEL) of the system. In the first step a set of equilibrium configurations is generated via MD simulations at constant temperature T_{equil}. Configurations at different times are taken and used as starting configurations for subsequent minimization via the Polak-Ribiere conjugate gradient algorithm, yielding a corresponding set of local minimum energy structures, denoted *inherent* structures.[40] In the second step the goal is to locate nearby minima for these inherent structures. To search for these nearby minima we proceed as follows: (1) Reset all particle velocities with random numbers according to a normal distribution at a fixed temperature T_{search}. (2) Perform a fixed number N_{search} of MD steps in the NVE ensemble (constant number of particles, constant volume and constant energy). (3) Minimize the resulting structure. (4) Accept the new minimum if the distance d and the asymmetry Δ between both minima in configuration space (see below for an exact definition) fulfill $0 \approx d_{min} < d < d_{max}$ and $0 \approx \Delta_{min} < \Delta < \Delta_{max}$, respectively. We introduced very small minimum cutoffs, because minima cannot be distinguished below numerical precision. For the choice of the cutoff-

values; see below. (5) Repeat this procedure M times for different initial random numbers. The values of T_{search} and N_{search} are chosen to give the largest number of nearby minima per starting inherent structure.

In the final step the transition states for the generated pairs of minima are located by a modified version of the nonlocal ridge method.[38] This algorithm is very robust to identify first-order saddles between pairs of minima. In case that no such saddle exists the new minimum is dismissed. In particular this may occur if the path to the second minimum involves transitions of two (or more) independent TLS during the N_{search} MD-steps. This is the standard situation for very large systems. As a result we obtain for every starting minimum a set of minima characterized by asymmetry Δ, distance d and barrier height V. We explicitly checked that all minima have 0 and all transitions states 1 negative eigenfrequency.

This algorithm is superior to the TLS-search algorithm, used in previous works[23,25] because no indirect assumptions are made with respect to the number of particles participating in a TLS-transition or with respect to the distance d.

Beyond the direct euclidian distance d between configurations (or correspondingly the distance d_i moved by particle i), we use mass weighted distances d_{mw} between two configurations:

$$d_{mw}^2(\vec{r}_1, \vec{r}_2) = \sum_i^N (d_{i,x}^2 + d_{i,y}^2 + d_{i,z}^2) \cdot \frac{m_i}{\bar{m}}. \quad (2)$$

Furthermore one can define the mass weighted reaction path approximation between two minima via

$$d_{mwrp} = d_{mw}(\vec{r}_1, \vec{r}_{trans.state}) + d_{mw}(\vec{r}_{trans.state}, \vec{r}_2), \quad (3)$$

where $\vec{r}_1, \vec{r}_2, \vec{r}_{trans.state}$ are the particle positions of the two minima and the transition state. d_{rp} is defined similarly as d_{mwrp}, but without mass-weighting. The mass-weighted reaction path is introduced because it enters the WKB-term to calculate the tunneling matrix element. For comparison with literature it is necessary to have also direct Euclidian distances. In the subsequent analysis we have attempted to employ for every type of analysis the appropriate distance. In any event, due to the similarity of all definitions, none of our results would change on a qualitative level if different distances had been used.

C. Double-well potentials vs TLS

As the ultimate goal one is looking for pairs of minima, i.e. double-well potentials (DWP), in the high-dimensional potential energy landscape with $\Delta/k_B < 1$ K, corresponding to 0.001 in LJ-units. They are expected to be relevant for the low-temperature anomalies around 1 K and thus act as TLS. Qualitatively, one would expect that many more DWP exist with asymmetries of the order of the glass transition temperature rather than 1 K. Restricting the search to DWP with such a small asymmetry is somewhat problematic because TLS are a very rare species and thus it is very difficult to find them numerically. Therefore we use as a standard choice a relatively large asymmetry of $\Delta_{max} = 0.5$. From the resulting
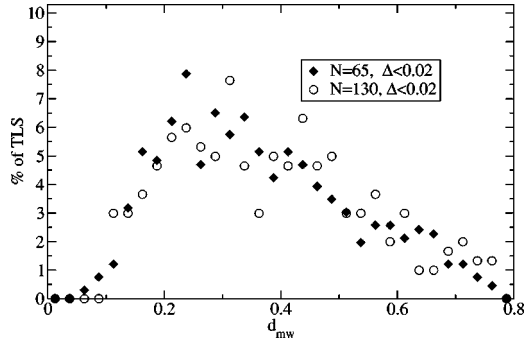
FIG. 1. The histogram of the distribution of distances for all DWP with $\Delta < 0.02$ (20 K for NiP).

data set (6522 DWP for $N=65$, 2911 for $N=130$ and 428 for $N=260$) we may easily analyze subsets of almost symmetric DWP which are relevant for the low-temperature properties. As shown in previous work also the range of very asymmetric DWP contains important information about the properties of TLS. In particular the choice $\Delta_{max}/k_B = 20$ K (0.02 in LJ-units) is small enough as compared to the glass transition temperature but large enough so that we get sufficient statistics. We always choose $d_{mwrp,max} = 0.8$ for reasons mentioned below. For the selection of the starting minima we choose $T_{equil} = 0.5$ which is slightly above the critical mode-coupling temperature of $T_c = 0.45$.[35,38]

III. RESULTS

A. Completeness of the search and finite size effects

From previous work it is known that the different parameters Δ, d, and V are strongly correlated. For example one finds in agreement with intuition that DWP with small d typically have a rather small asymmetry. Thus one would expect that most DWP which may act as TLS at low temperature are restricted to some region of d-values. Therefore we have checked whether the DWP with $\Delta < 0.02$ indeed display this restriction in d. For $N=65$ we have found 301 DWP in this range. The result is shown in Fig. 1. It turns out that the d-distribution shows a peak around $d=0.3$ and strongly decreases for larger d. For $N=130$ we get very similar results. In addition to this the tunneling probability decreases exponentially with d and thus also the relevance of TLS with large values of d. Therefore the choice of $d_{max} = 0.8$ is justified by the decreasing probability to find TLS with small asymmetry and large distances and also by the decreasing relevance for those TLS. In a next step we elucidate the quality of our search algorithm and check whether our search of TLS is complete within our specified parameter range. This property is essential to estimate the absolute number of TLS from our simulations. For this purpose we have analyzed how often the different minima around a starting minimum, i.e., how often DWP, are found during the $M=400$ search attempts per configuration. In case that most DWP are only found once or twice it is very likely that many minima are overseen. In contrast, if almost all DWP are found quite frequently it is unlikely that many DWP are missing. This conclusion is based on the assumption that the
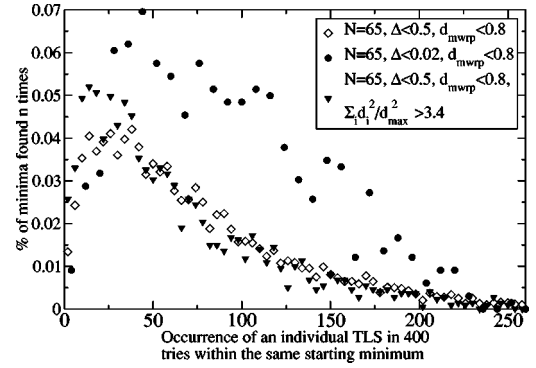

FIG. 2. The histogram of how often a nearby minimum was found for the 65 particle system after $M=400$ attempts per starting minimum. The triangles correspond to data limited to participation ratios above average (1/3 of all found) and are included here for later use.

finding probabilities of the DWP are continuous, i.e. there is not a second class of invisible DWP, strictly separated from the DWP found in our simulations. For $N=65$ particles the distribution of TLS counts is shown in Fig. 2 for both $\Delta_{max} = 0.5$ and $\Delta_{max} = 0.02$. Already for the first choice it turns out that the distribution has its maximum at a count above 15 and the likeliness for lower counts decreases rapidly. Thus most minima are indeed found with a high probability and the search is therefore almost complete. For the DWP with $\Delta < 0.02$ this effect is even more pronounced. This comparison also shows that DWP with smaller asymmetry are found more easily than DWP with larger asymmetry. Furthermore we analyzed whether for the system size of $N=65$ finite size effects are present. As already mentioned above, general finite size effects are absent for the dynamics above T_c. It turns out that all properties of DWP (partly presented in this work, partly in a subsequent publication) were identical in the observed N-range between 65 and 260 within statistical noise. There is, however, one exception. Within the parameter range, specified above, we found on average 0.65 DWP per starting minimum for $N=65$ and 0.94 DWP per starting minimum for $N=130$. Naively one would have expected twice the number of DWP for $N=130$. This means that on the per-particle basis the number of TLS' is roughly 30% too small for $N=130$ as compared to $N=65$; see Table I for a quick overview. This effect is even more pronounced for $N=260$. Two reasons are possible: either we have found a significant finite size effect (which may appear surprising because all properties of the TLS are identical) or this effect may be caused by the algorithm.

TABLE I. Summary of the found number of DWP for the used asymmetry restrictions and particle numbers.

		All DWP are restricted to $d < 0.8$			
N	Δ	Analyzed minima	DWP	DWP per particle	M
65	<0.5	10009	6522	0.01	400
130	<0.5	3100	2911	0.0072	800
65	<0.02	10009	628	0.00097	400
65	<0.001	10009	43	6.3×10^{-5}	400

To test the second possibility we performed simulations with two noninteracting systems A and B sharing a box. For determining minima the relative positions of the two independent systems were preserved and the energies added. In the combined system the absolute number of TLS is by construction twice as high as in the elementary system. Interestingly, using the same simulation parameters as above we also observed a relative decrease of circa 30% when analyzing the combined system 2.65. Thus it is very likely that the apparent finite size effect, described above, is solely caused by the algorithm. This effect can be rationalized. If a minimum, corresponding to a TLS, is found in configuration A with a probability of 0.9 per attempt and in configuration B another TLS is found with a probability of 0.1 per attempt, they change to $0.81 = 0.9 \cdot (1 - 0.1)$ and $0.01 = 0.1 \cdot (1 - 0.9)$, respectively, when both configurations are considered together and if combined transitions are ignored (see above). This effect gives rise to a sharp decrease in the probability to detect elementary transitions for larger systems. Therefore the best procedure is to analyze rather small systems which, however, are large enough to be void of relevant finite size effects. In analogy to our results in Ref. 38 the system size of $N = 65$ seems to be a very good compromise and, according to Fig. 2, allows one to obtain the absolute number of TLS.

To proceed we counted the number of DWP with $\Delta < 0.02$. It turns out that one DWP exists per 1000 particles, i.e. 0.063 DWP per independent configurations divided by N. For even smaller asymmetry $\Delta / k_B < 1$ K (≈ 0.001) we observe one DWP, i.e. TLS, per 15000 particles. Using the density of NiP one ends up with 5×10^{47} J^{-1} m^{-3} TLS. Actually, the parameter P, which can be determined experimentally and gives an experimental number of TLS, is roughly smaller by one decade because the contribution of the individual TLS are weighted by a factor $\Delta_0^2 / (\Delta_0^2 + \Delta^2) < 1$ where Δ_0 is the tunneling matrix element. More specifically we obtain $P \approx 1.3 \times 10^{46}$ J^{-1} m^{-3} (see Ref. 41) which is close to the value of $P \approx 1.5 \times 10^{46}$ J^{-1} m^{-3}, obtained in Ref. 23. Most of the remaining difference with, e.g. $P = 8 \times 10^{44}$ J^{-1} m^{-3} for silicate can be directly explained by the fact that most glasses have larger elementary units like the SiO$_4$ tetrahedra for silicate. This means that the number of DWP per volume is even smaller, yielding an estimate very close to the experimental value of silicate.

B. Embedding of TLS in the potential energy landscape

Now we discuss how the TLS are embedded into the overall PEL. As already discussed above, computer simulations suffer from finite simulation times. As a consequence the starting inherent structures will have a relatively high energy as compared to inherent structures the system would have reached if equilibration at lower temperature T_{equil} close to the calorimetric glass transition were possible. In order to elucidate this aspect closer we would briefly like to summarize the previous results for this LJ-system.[38] (1) Inherent structure energies range between −304 and −287. The low-energy cutoff, however, is not due to the bottom of the PEL. Rather it indicates that the number of minima with even lower energy is so small that they were not detected during
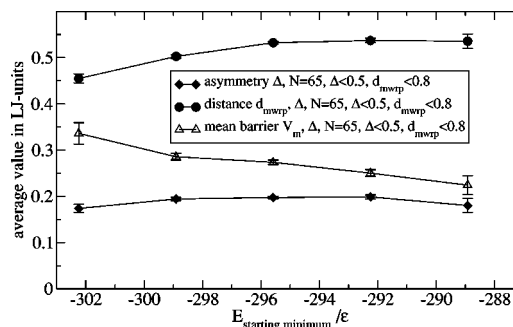

FIG. 3. The dependence of the DWP parameters on the energy of the starting minimum. The lines are a guide to the eye.

the simulations. (2) More strictly, the distribution of energies follows the left part of a Gaussian distribution. This implies that the number of states with higher energy is always exponentially larger than the number of states with somewhat lower energy. (3) It can be estimated that the bottom of the potential energy landscape is around −306. This number can be estimated from a determination of the total configurational entropy.[42,43] Thus even for very small cooling rates energies would be larger than −306. (4) The dynamics around and below T_c can be interpreted as jumps between different traps, denoted metabasins. They contain a number of individual inherent structures.

As our simulations cover a broad range of minimum energies we checked to which degree the properties of the DWP depend on the energy of the starting inherent structure. Figure 3 shows the dependence of the three DWP parameters on this energy. Interestingly, the dependencies are rather small. In particular there is no indication to believe that DWP in the relevant low-temperature energy range between −306 and −302 are very different. This is in agreement with the observed independence of the inherent structure dynamics observed by Vogel *et al.*[33] Of course, on a strict quantitative level minor variations should be taken into account.

Furthermore we analyzed whether the number of DWP per starting configuration depends on its potential energy; see Fig. 4. In order to work with a larger data set we have included asymmetries up to 0.8. Furthermore we have analyzed the DWP for different subsets with respect to the distance d_{mwrp}. It turns out that the dependence on energy is very weak. Even in the most extreme case ($d_{mwrp} < 0.8$) the number of DWP changes only by a factor of 2 when com-
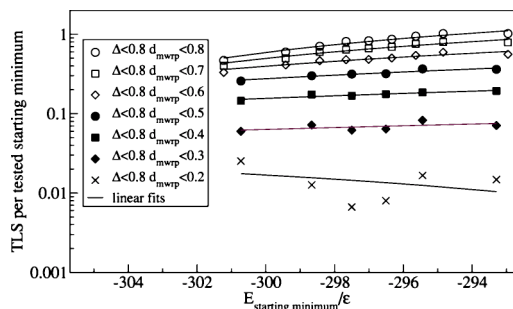

FIG. 4. The dependence of the number of DWP per starting minimum on the energy of the starting minimum.
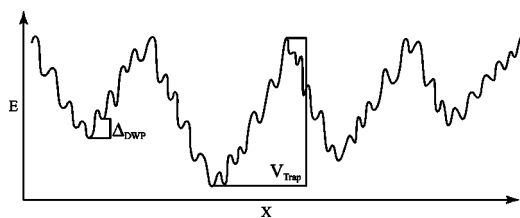
FIG. 5. One dimensional sketch of the potential energy landscape. The asymmetry Δ_{DWP} and the barrier height for the DWP vary between 0 and $2T_c$. The barrier height for the traps V_{Trap} on the other hand varies between 1 and $20T_c$.
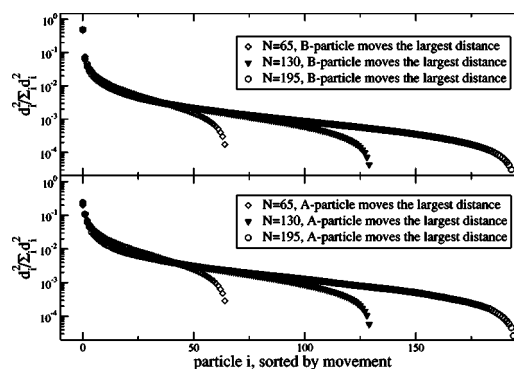

FIG. 6. The contribution of each particle to the total translational motion in a DWP. The DWP with an A-particle as a central particle (bottom) and a B-particle as a central particle (top) show different behavior.

paring different energies. This small variation nearly vanishes when restricting ourselves to DWP with small distances. In summary, we may conclude that on a local scale the properties of the potential energy landscape do not depend on the height in the landscape. Evidently this property must break down when taking into account pairs of minima with a larger spatial separation. Because the number of minima for, e.g., energies around −295 is by a factor of 10^5 larger than the number of minima for energies around −300 (see Ref. 42) it is evident that for larger d many more pairs of minima with similar energy can be found for −295 as compared to −300.

In Fig. 5 we attempt to sketch the PEL of the LJ-system. The most prominent features are the individual traps. In contrast, the DWP analyzed in this work correspond to the little wiggles within the traps. Typical energy scales have been given in the figure caption showing the separation of transitions between adjacent inherent structures, already relevant at low temperatures, and between traps, relevant above the glass transition.

C. Microscopic nature of DWP

Furthermore we have studied the microscopic nature of the DWP in great detail. One of the most elementary questions one may ask is how many particles are involved in the translation between the two minima of a DWP? To establish an accurate picture of the microscopic nature of the TLS we analyzed the number and types of particles participating in the transition from one minimum to the other. As a matter of fact all particles move when going from one minimum to the other. In Fig. 6 we show the distribution of distances moved by the different particles during the transition between both minima of a DWP. The distances are sorted according to their actual size. The curvature for the last particles is caused by the minimization of the distance of the two minima in a finite system. The curves for different systems show only small deviations for the particles which move a larger distance. It can be seen that the first particle, which moves the biggest distance, moves much farther than the second particle if it is a B-particle (small); this is less pronounced if it is an A-particle. As the particle which moves the biggest distance can usually be clearly distinguished from the other particles we denote it as *central*. With this definition we find that in over 90% of all DWP the central particle is a B-particle, although the concentration of B-particles is only

20%. DWP with central B-particles thus dominate DWP with a central A-particle.

From these data one may define a participation ratio to estimate the number of particles involved in the transition between two minima. In the case that n particles are moving some fixed distance and the other particles do not participate at all, one would obtain a participation ratio of n. In the present case, of course, one has a broad distribution and thus different definitions of the participation ratio yield different values, as shown in Table II. The value for the total system varies between 2.1 and 9.5. Therefore it is more informative to consider the total distribution of translations instead of a single number; see Fig. 6. We note in passing that many authors use the definition $d^4/\Sigma_i d_i^4$ to characterize the participation ratio. Unfortunately, for this definition it is not possible to draw a distribution, sorted by particle displacement, as shown in Fig. 6. Therefore we have used $1/\langle d_{max}^2/d^2 \rangle$ for this purpose.

Finally, one may ask the question whether the participation ratio depends on the properties of the DWP. Most naturally, one might think of a relation to the distance between both minima because the distance directly appears in all definitions of the participation ratio. In Fig. 7 the dependence of the participation ratio (second definition) on the distance is shown. As expected for DWP with larger distances more particles contribute to the translation. Actually, in previous work (Ref. 44, p. 483) we have shown for a very similar LJ-system that the slope might decrease for a larger distance; still the participation ratio increases with distance.

D. Spatial distribution of DWP

In the STM it is assumed that the TLS are randomly distributed in space. To check this hypothesis we first calculate

TABLE II. Partition ratios for different definitions and for A- and B-particles as central particles. No mass-weighting has been used for the distances.

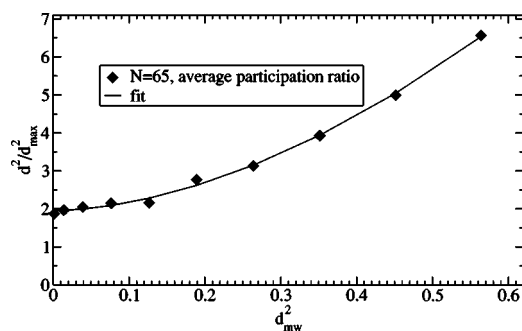| Central particle	$1/\langle d_{max}^2/d^2 \rangle$	$\langle d^2/d_{max}^2 \rangle$	$\langle \Sigma_i d_i/d_{max} \rangle$	$\langle d^4/\Sigma_i d_i^4 \rangle$
average	2.14	3.29	9.50	7.65
B	2.04	3.01	8.97	6.95
A	4.95	6.72	16.11	16.43

FIG. 7. The participation ratio does significantly dependent on the distance of the configurations.

the probability to find two DWP which have the same central particle. We find that, for both $N=65$ and $N=130$ an enhanced probability that a single particle is central for two different DWP. To elucidate this aspect further we made a simplified analysis for the 65 particle system. Instead of considering all pairs of central particles we considered only B-B-pairs and only those pairs, which originate from configurations with exactly two observed DWP. For independent DWP one would expect that in $1/13=7.7\%$ of all cases the central particle is identical. Rather we found $(17\pm2)\%$. Thus there is indeed a significantly increased probability that two DWP are spatially correlated, which can also be interpreted as an increased probability for triple well potentials. We find that when omitting DWP with the same central particle the other DWP are randomly distributed in the system. This was checked by computing the average distance of the remaining central particles in cases where more than 1 DWP is present in the system. It turns out that this distance is within statistical uncertainty identical to the distance of randomly chosen particles. The results for the 130 particles system show the same behavior. We do not consider the observed phenomena to be a severe deviation from the STM, because the abundance of the triple well potentials is rather small.

IV. DISCUSSION AND SUMMARY

We have presented a new reliable algorithm to systematically locate DWP in a model glass former. It turned out that the intrinsic limitations of computer simulations do not hamper the quality of our results. Thus we can indeed get information about TLS, relevant for understanding the low-temperature anomalies.

It turns out that the number of TLS, directly obtained from our data, is compatible with the number of TLS observed experimentally. This conclusion had been already drawn from our previous work. This time, however, we can exclude that possible systematic artifacts hamper our analysis. Thus it is likely that TLS, responsible for the low-temperature anomalies, result from elementary noninteracting two-state systems rather than from collective excitations.

As a by-product of our simulation we realized that the DWP are not randomly distributed in space. Rather there is a strong tendency that two DWP are located at the same central particle, i.e. form a triple well potential. This suggests

that some structural features of the glass favor the formation of DWP. A possible candidate is the coordination sphere around the central particle. To check whether these triple well potentials are relevant at temperatures in the Kelvin regime we also checked if the asymmetries of the two connected double wells correlate. No such correlation could be found for our set of triple well potentials. Therefore it is very unlikely that all three minima have energies within a very narrow energy range of a few Kelvin. Thus at low temperatures nearly all triple well potentials will at best serve as two-state TLS. This statistical argument as well as the fact that the number of triple well potentials is rather small (about 1.3% of all DWP in the range of asymmetries and distances used in our work) shows that the observed phenomena mainly indicate structural features but are otherwise irrelevant for the low temperature anomalies.

The properties of the TLS reflect local properties of the PEL. In connection with our previous work on global features of the PEL it is possible to have a view on the PEL, encompassing the transport dynamics above and the local dynamics below the glass transition. It becomes evident that the TLS are part of the individual traps in which the PEL can be decomposed.

We would like to mention two discrepancies with other work. One is the magnitude of the absolute movement of a central B- or A-particle, which is found to be quite large: $d_{B,rp}^2=0.13$ and $d_{A,rp}^2=0.06$ in LJ units. These values are about an order of magnitude larger than the estimation given by Lubchenko and Wolyness[17] in their frustrated domain wall model. The other discrepancy concerns the participation ratios. The observed participation ratios, obtained from averaging over all DWP, are much lower than those observed by other groups investigating the PEL, namely by Oligschleger and Schober in soft sphere glasses and LJ-systems[31,32] or Vogel et al.[33] in the same system as used in this work. In Refs. 31–33 the authors have analyzed the transitions between adjacent minima as resulting from a molecular dynamics trajectory at a given temperature and found of the order of 20 particles. Comparing this with our average value of around 3 this seems to be a major difference, as already stated by Oligschleger and Schober[31] in relation to our earlier work[23] where similarly small values have been reported. As shown in Fig. 2 this difference is not due to the fact that our algorithm is not able to identify DWP with large participation ratios. For a closer discussion of this discrepancy one has to take care of the actual definition, used to characterize the participation ratio. In the reported work the two latter definitions in Table II have been used. Thus the reported values have to be compared to our participation ratios of about 7–10. There is, however, still a remaining difference of a factor of 2–3.

This difference can be rationalized by the different methods of locating inherent structures. In the present work we have attempted to localize all inherent structures within a certain distance to the original minimum. This approach was motivated by the observation that DWP with small asymmetries typically correspond to nearby minima (see Fig. 1) so that DWP with large distances between the minima are typically irrelevant for the understanding of the low-temperature anomalies. As a direct consequence the typical distances be-

tween the minima are significantly smaller found by the present method as compared to the results reported in Refs. 31–33 which allude more to the properties of the glass transition. This can be quantified via the configurational distance $d^C = \Sigma_i \, (d_{i,x}^2 + d_{i,y}^2 + d_{i,z}^2)^{0.5}$. Comparing our value with the value reported in Ref. 33 we find a difference of a factor of three.

Following the results, reported in Fig. 7, this difference directly translates into differences of the participation ratio. Extrapolating the data shown in Fig. 7 to distances around unity one ends up with an increase of the participation ratio by a factor of 2–3 which is exactly the factor, which was missing above. Thus our present results are fully consistent with the results discussed in previous work. We just mention in passing that the situation may be even more complicated since the DWP, obtained by Schober *et al.* and Vogel *et al.*, have been obtained from MD trajectories. Thus there exists an implicit weighting by the probability to find these DWP. In contrast, in our approach a systematic determination of all DWP within a specific parameter range has been conducted. This may hamper the comparison of DWP, obtained by the two different methods, even further.

Having in mind the dominance of the small B-particles in the transition between two minima it becomes obvious that the first two definitions of the participation ratio better reflect the dominance of the single-particle character of DWP transitions. Thus we feel it is more intuitive to speak of 3 rather than of 7–10 particles which dominate the translation between DWP relevant for the low-temperature anomalies. In any event, from a theoretical point of view it it is the whole distribution in Fig. 6 which fully characterizes the nature of the translational dynamics between minima in the low-temperature regime.

Our results imply that experimental findings for materials with a smaller minority components (like the analyzed LJ-system) should be dominated by B-type TLS. More generally, one can ask whether the results obtained for our LJ-system are also relevant for other systems. Actually, we already saw that the DWP with central A-particles involve more cooperative processes. Our preliminary results for silica indicate that the participation ratios are similar to those of the A-particles. In general one might speculate that for TLS, which mainly contain very similar molecules, the transition is more collective (like in pure silica) and behave like the A-particles in the present case, while for TLS consisting of small molecules in a matrix the transition is similar to the more localized process, as seen for the B-particles.

Now after validating the methods the next step is to get a closer insight into the microscopic properties of the DWP and to perform a direct comparison with experiments on the low-temperature anomalies. Work along this line will be published elsewhere.

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