Equilibrating Glassy Systems with Parallel Tempering

Walter Kob*, Claudio Brangian*, Torsten Stühn*, and Ryoichi Yamamoto†

* Institute of Physics, Johannes Gutenberg-University, Staudinger Weg 7, D–55099 Mainz, Germany
† Department of Physics, Kyoto University, Kyoto 606-8502, Japan

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Abstract

We discuss the efficiency of the so-called parallel tempering method to equilibrate glassy systems also at low temperatures. The main focus is on two structural glass models, SiO₂ and a Lennard-Jones system, but we also investigate a fully connected 10 state Potts-glass. By calculating the mean squared displacement of a tagged particle and the spin-autocorrelation function, we find that for these three glass-formers the parallel tempering method is indeed able to generate, at low temperatures, new independent configurations at a rate which is O(100) times faster than more traditional algorithms, such as molecular dynamics and single spin flip Monte Carlo dynamics. In addition we find that this speedup increases with decreasing temperature. The reliability of the results is checked by calculating the distribution of the energy at various temperatures and by showing that these can be mapped onto each other by the reweighting technique.

1 Introduction

Despite the progress made in recent years [1, 2], our understanding of the dynamics of deeply supercooled liquids and the properties of glasses is still far from satisfactory [3]. Although the so-called mode-coupling theory (MCT) [4] seems to give a very reliable description of this dynamics around the MCT-temperature $T_c$, the relaxation dynamics significantly below $T_c$, i.e. close to the experimental glass transition temperature $T_g$, still waits for a deeper theoretical understanding. The problem is that in this temperature range the mechanism leading to the relaxation of the system seems to be governed by the so-called hopping processes, i.e. the system relaxes in an activated way, and that these processes are not readily describable by MCT in its ideal version. (Although the extended version of MCT [5], which to some extend takes into account these hopping processes, is able to make precise predictions for the dynamics at intermediate time scales, it looses its predictive power at long times, i.e. the $\alpha$–relaxation regime.) Although experiments are very useful to give information on particle averaged properties of supercooled liquids and glasses, such as the viscosity or the intermediate scattering function, they are
less well adapted to study the system on a very local scale, e.g. on the level of a single particle. This is, however, exactly the information which is needed if one wants to come to an understanding of the relaxation mechanism in these systems if one wants to go beyond the use of simple models, such as a particle moving around in a frozen environment or similar simple pictures. One method which in the past has been proven to be extremely useful to reveal the necessary details of structure and dynamics of complex systems are computer simulations \[5\]. Since such simulations allow to access at any instant the full microscopic information, they are also ideally suited to learn more about the relaxation mechanisms in supercooled systems and glasses \[1\]. One problem of such simulations is, however, that the time and length scales accessible are rather limited (typically 100 ns and box sizes of 100Å\(^3\)) and this has prevented in the past to study, e.g., the equilibrium dynamics of supercooled liquids close to \(T_g\), where the typical time scales are on the order of 10-1000 s, i.e. 15-17 decades longer than the typical time step of used in a simulation which is on the order of 1/100 of the inverse of the Debye frequency, i.e. 1-10 fs. One possibility to avoid this problem to some extend is to use a sophisticated Monte Carlo algorithm that allows to equilibrate the system even at those low temperatures at which standard molecular dynamics or Monte Carlo algorithms fail to reach equilibration. In recent years various authors have proposed different Monte Carlo algorithms that should be suited to reach the equilibrium state even at low temperatures \[1, 8, 14, 11, 12\] and have also been successfully applied to various systems \[13, 14, 15, 16, 17\] (see, however, Ref. \[18\] for discouraging results). Here we will discuss the application of the so-called parallel tempering (PT) algorithm, proposed by Hukushima et al. \[11\], a method that is similar in spirit to the replica exchange algorithm proposed earlier by Swendsen and Wang \[12\] to the case of structural glasses and to the Potts-glass. Although for structural glasses with a small number of particles \((N \leq 36)\) the PT method has been successfully tested by Coluzzi and Parisi \[19\] we are here interested in using larger systems \((N = 336\) and \(N = 1000)\). In the next sections we will discuss the details of the PT algorithm, then introduce the models we investigated and subsequently discuss the results.

### 2 The parallel tempering algorithm

The PT method proposed by Hukushima et al. can be summarized as follows \[11\]: 1) We denote the Hamiltonian of interest by \(H = K(p) + E(q)\), where \(K\) and \(E\) are the kinetic and potential energy, respectively, and \(p = (p_1, p_2, \ldots, p_N)\) and \(q = (q_1, q_2, \ldots, q_N)\) are the momenta and coordinates of the particles, respectively. (If this is a spin-like Hamiltonian, we put \(K = 0\).) We now construct a new system consisting of \(M\) noninteracting subsystems, each composed of \(N\) particles, with a set of arbitrary particle configurations
\{q_1, \ldots, q_M\} and momenta \{p_1, \ldots, p_M\}. The Hamiltonian of the \(i\)-th subsystem is given by
\[ H_i(p_i, q_i) = K(p_i) + \Lambda_i E(q_i), \]
where \(\Lambda_i \in \{\lambda_1, \ldots, \lambda_M\}\) is a parameter to scale the potential. 

2) A molecular dynamics simulation is done for the total system, whose Hamiltonian is given by \(H = \sum_{i=1}^{M} H_i\), at a constant temperature \(T = \beta_0^{-1}\). In this way we obtain a canonical distribution
\[ P(q_1, \ldots, q_M; \beta_0) = \prod_{i=1}^{M} P(q_i; \Lambda_i \beta_0) \propto \exp[-\beta_0 \sum_{i=1}^{M} \Lambda_i E(q_i)] \] in configuration space. 

3) After each time interval \(\Delta t_{PT}\), we attempt to exchange the potential scaling parameter of the \(m\)-th and \(n\)-th subsystem, while \{\(q_1, \ldots, q_M\)\} and \{\(p_1, \ldots, p_M\)\} are unchanged. The acceptance of the exchange is decided in such a way that it takes care of the condition of detailed balance. Here we use the Metropolis scheme, and thus the acceptance ratio is given by
\[ w_{m,n} = \begin{cases} 1, & \Delta_{m,n} \leq 0 \\ \exp(-\Delta_{m,n}), & \Delta_{m,n} > 0 \end{cases} \]
where \(\Delta_{m,n} = \beta_0 (\Lambda_n - \Lambda_m)(E(q_m) - E(q_n))\). 

4) If steps 2) and 3) are repeated for a sufficiently long time this scheme leads to canonical distribution functions \(P(E; \beta_i)\) at a set of inverse temperatures \(\beta_i = \lambda_i \beta_0\). To make a measurement at an inverse temperature \(\beta_i\) one has to average over all those subsystems (\(i \in 1, \ldots, M\)) for which we have (temporarily) \(\beta_i = \lambda_i \beta_0\).

Although the algorithm presented is correct for general choice of the values of \(m\) and \(n\) in Eq. (2), it is advisable to exchange only neighboring subsystems in order to allow for a reasonably high acceptance rate. In addition one has also to choose the values of the coupling constants \{\(\lambda_1, \ldots, \lambda_M\)\} in such a way that neighboring subsystem have an sufficiently large overlap in their distribution of the energy.

We also note that the algorithm we just described shares many properties with the one proposed by Swendsen and Wang in 1986 [12]. These authors already realized that it is very useful to connect dynamically configurations at low temperatures to the ones at high temperatures and also gave a specific method how this can be successfully done in the case of a frustrated spin system.

3 Models and Details of the Simulations

In this paper we consider three different types of glassy systems: A realistic model for SiO2, one of the prototypical glass formers. A binary mixture of Lennard-Jones particles, i.e. an example for a simple glass former, and finally a ten state Potts-glass, an example for a spin system with a discontinuous transition from the paramagnetic phase to the spin glass phase.
SiO$_2$ is a prototype for a so-called strong glass former [20]. This means that the temperature dependence of transport quantities like the viscosity or the diffusion constant show an Arrhenius dependence. It is believed that this property is related to the fact that the structure of amorphous silica is an open tetrahedral network that is essentially independent of temperature.

The SiO$_2$ model we use has been proposed by van Beest et al. (BKS) [21] on the basis of ab initio calculations. In this system two ions of type $\alpha$ and $\beta$ ($\alpha, \beta \in \{\text{Si, O}\}$) that are a distance $r$ apart interact via the following potential:

$$\phi(r) = \frac{q_\alpha q_\beta e^2}{r} + A_{\alpha\beta} \exp(-B_{\alpha\beta}r) - \frac{C_{\alpha\beta}}{r^6},$$  

where the values of the parameters $q_\alpha$, $A_{\alpha\beta}$, $B_{\alpha\beta}$, and $C_{\alpha\beta}$ can be found in Ref. [21]. Previous computer simulations have shown that the BKS model gives a very good description of the static and dynamic properties of real silica [22, 23, 24, 25, 26, 27]. However, so far these tests could be done only at relatively high temperatures, i.e. $T \geq 2750$K, since below this temperature the relaxation time exceeds the time scale accessible to normal molecular dynamics simulations (in this case 20 ns). This temperature has to be compared with the experimental value of $T_g$, 1450K, i.e. so far it has been possible to equilibrate the system only at temperatures two times higher than $T_g$.

In the following we will present results in which we used the PT algorithm to equilibrate the system at low temperatures. Between the attempted exchanges between two subsystems we propagated the particles in the isokinetic ensemble at constant volume. The system size was 336 ions and the time step was 1.6 fs. The results presented below are for $M = 32$ subsystems, the number of time steps for equilibration and production was each $4 \cdot 10^6$, and the value of $\Delta t_{PT}$ was 1000 time steps. More details on this simulation can be found in Ref. [28].

The second structural glass is a binary (80:20) mixture of Lennard-Jones particles. If we denote the majority species by A and the minority species by B the interaction between two particles are given by $\phi_{\alpha\beta}(r) = 4\epsilon_{\alpha\beta}[(\sigma_{\alpha\beta}/r)^{12} - (\sigma_{\alpha\beta}/r)^6]$, where $r$ is the distance between particles $i$ and $j$. The interaction parameters are $\alpha, \beta \in \{A, B\}$, $\epsilon_{AA} = 1$, $\epsilon_{AB} = 1.5$, $\epsilon_{BB} = 0.5$, $\sigma_{AA} = 1$, $\sigma_{AB} = 0.8$, and $\sigma_{BB} = 0.88$. In the following we will measure length and energy in units of $\sigma_{AA}$ and $\epsilon_{AA}$, respectively, (setting $k_B = 1$) and time in units of $(m\sigma_{AA}^2/48\epsilon_{AA})^{1/2}$, where $m$ is the mass of the particles. In the simulation we used a cubic box, of length 9.4, with periodic boundary conditions, the total number of particles was 1000, and the time step was 0.01732. The number of subsystems was 16, the number of time steps for equilibration and production was each $5 \cdot 10^6$, and the value of $\Delta t_{PT}$ was 1000 time steps. More details on the simulation can be found in Ref. [17]. In the past many properties of this model have been investigated [3, 29, 30, 31, 32, 33, 34] and it has been found that its relaxation dynamics becomes very slow at around $T = 0.45 - 0.43$, i.e. its starts
to exceed the time scales accessible to normal molecular dynamics simulations, which is on the order of $10^8$ time steps. In contrast to the Arrhenius-like increase of the relaxation times, as it is found in SiO$_2$, this system shows at low temperatures an increase which can be fitted well with a power-law, and thus this is considered to be a fragile glass former [20]. The structure of the system resembles the one of randomly closed packed hard spheres and is thus very different from the open network in silica. Last not least it has to be mentioned that the simplicity of the interaction of this model allows to obtain results for this system which have a significantly higher statistical accuracy than the ones for the silica model discussed above, since in the latter one has to calculate numerically expensive long range interactions. This is the reason why in the results discussed below the data for the silica system is quite a bit more noisy than the one for the Lennard-Jones system.

The Potts-glass we consider is an example of a spin glass which shows a discontinuous transition from the paramagnetic phase to a spin glass phase if the number of states is larger than 4 [35]. It has been suggested that such type of models show a qualitatively similar dynamics as structural glasses [36, 37], and therefore it is of interest to understand their properties in more detail. Here we use the version in which each spin $\sigma_i$ can take one of $q = 10$ different states ($\sigma_i \in \{1, 2, \ldots, 10\}$) and each spin interacts with every other one with an interaction $J_{ij}$. Thus the Hamiltonian is:

$$H = -\frac{1}{2} \sum_i \sum_{j \neq i}^N J_{ij} \cdot (q\delta_{\sigma_i, \sigma_j} - 1) \quad (4)$$

The interactions $J_{ij}$ are drawn from a gaussian probability distribution with mean $(3 - q)/(N - 1)$ and variance $(N - 1)/2$ [35]. It has been show that in the thermodynamic limit this model has a dynamical singularity at $T_d = 1.14$, slightly above the static singularity at $T_c = 1.13$ [38]. We have considered system sizes $N$ between 32 and 2560 [35] but here we will discuss only results for $N = 320$. In principle one has of course to average all the results obtained over the quenched disorder, i.e. the interactions $J_{ij}$. For the present case we have not done this (i.e. we consider only one realization of the disorder) but we have tested that the results presented below are independent of the choice of $J_{ij}$. The number of subsystems was $M = 16$ and the exchange time $\Delta t_{PT}$ was 10 Monte Carlo steps per spin. Below we will compare the results of the relaxation dynamics with the PT algorithm with the one of standard single spin flip Monte Carlo scheme. In the latter case we used the Metropolis criterion to accept or reject a move. More details on this investigation can be found in Ref. [35]
4 Results

In this section we discuss the results, i.e. we compare the efficiency of the PT algorithm with the more conventional methods (standard molecular dynamics and Metropolis Monte Carlo) to propagate the system through configuration space. First we present our findings for the SiO$_2$ system, then for the Lennard-Jones system, and finally for the Potts-glass. In all cases we first used the PT algorithm for a sufficiently long time to allow all subsystems to equilibrate. This equilibration was tested by comparing quantities like the energy or the specific heat obtained from such runs with the results from similar runs done with a standard method (molecular dynamics, single flip Monte Carlo).

From the setup of the PT algorithm it follows that each subsystem makes a random walk in temperature space. A rough estimate for the time needed until a new low-temperature configuration is produced can be obtained by looking at this random walk. For the case of SiO$_2$ a typical example is shown in Fig. 1, where we plot the temperature of one subsystem, or rather the value of the coupling constant $\Lambda_i$ (see (1)), as a function of time. The lowest temperature ($p = 1$) is 2750K and the highest one ($p = 32$) is 3922K. From this figure we recognize that it takes the system a bit more than 1000ps to generate a new independent configuration, i.e. to go from $p = 1$ to $p = M = 32$ and back. As we will see below this is significantly shorter than the time it takes in a standard molecular dynamics simulation. We also mention that the computational cost is of course also $M$ times (=number of subsystems) higher.

![Figure 1](image-url)

Figure 1: Time dependence of the coupling constant of one subsystem for the case of the SiO$_2$ system.
than in a conventional scheme. However, it has to be remembered that we also obtain $M$ independent configurations and not just one (this holds for each temperature!). Therefore if one needs many independent configurations, as it is usually the case in order to obtain reliable averages, the PT algorithm will pay off.

From this run we can calculate the distribution of the potential energy at the various temperatures. These distributions are shown in Fig. 2. (In order to avoid overcrowding of the figure we show only every second temperature and the highest one.) We see that in order to obtain a sufficiently high acceptance ratio it is necessary to have a good overlap of neighboring distributions. A smaller overlap will lead to a smaller acceptance probability and hence the random walk will take longer. On the other hand a smaller value of $M$ will allow the random walk to go faster from low temperatures to high temperatures. It is presently not clear what the optimal choice is since this will depend on the details of the system. More results on this can be found in Ref. [28].

At the beginning of this section we have mentioned that one possibility to test whether or not the system has reached equilibrium is to compare various observables with the ones obtained with a conventional simulation method. This approach is, however, only possible for those temperatures at which it is feasible to equilibrate the system also with one of the latter methods. A different possibility is to use the data for the energy distribution for the different subsystems and to test whether it is possible to reweight neighboring distributions to one common temperature [10]. If we denote the distribution
of the energy at the different coupling constants $T_i = 1/(\beta_0 \lambda_i)$ by

$$P_i(E) = P(E; T_i)$$

we should have the identity

$$P_i(E; T_j) = \frac{P_i(E) \exp[(\lambda_i - \lambda_j)\beta_0 E]}{\int dE' P_i(E') \exp[(\lambda_i - \lambda_j)\beta_0 E']}
$$

for all pairs $i$ and $j$. Note that in general this identity holds only if the different subsystems are in equilibrium and hence it can be used to check whether the total system is in equilibrium or not.

That for the PT run such a reweighting does indeed lead to a nice collapse of the different $P_i(E; T_j)$ onto one master curve is shown in Fig. 3, where we plot these functions for the case $j = 16$, which corresponds to $T = 3273$K, for $i = 4, \ldots, 15$ and $i = 17, \ldots, 27$. Thus from this plot we have evidence that the system has indeed equilibrated within the time span of the simulation.

Having checked that the algorithm does indeed allow to equilibrate the system even at low temperatures it is of course important to see how efficiently this is done. One possibility to measure this is to calculate the mean squared displacement (MSD) of a tagged particle,

$$\langle \Delta r^2(t) \rangle = \langle |r_j(t) - r_j(0)|^2 \rangle$$

Figure 3: Reweighted probability distributions for the energy of the SiO$_2$ system for the temperature interval given in the figure. See main text for more details.
Figure 4: Time dependence of the mean squared displacement for Si at different temperatures. The dashed lines are from PT runs and correspond to temperatures 3922K, 3585K, 3235K, 3019K, and 2750K (top to bottom). The solid lines are from conventional molecular dynamics runs and correspond to temperatures 6100K, 4700K, 4000K, 3580K, 3250K, and 3000K (top to bottom).

The time dependence of $\Delta \langle r^2(t) \rangle$ is shown in Fig. 4 for the case of silicon for various temperatures (dashed lines). Also included in the graph are the MSD obtained from a standard microcanonical run of the same system at similar temperatures (solid lines). From this figure we recognize that at low temperatures and long times the MSD from the PT is larger by about a factor to 100, thus demonstrating that this type of dynamics is significantly faster than the conventional one.

The silica system we just considered was a glass former whose structure is given by an open tetrahedral network and whose dynamics at low temperatures shows an Arrhenius dependence. The Lennard-Jones system we consider next has very different properties in that its structure is rather similar to a dense packing of hard spheres and its dynamics shows a temperature dependence which is stronger than an Arrhenius law. Therefore it is of interest to see whether the PT method also works for this kind of system, which is done now.

Note that the MSD from the PT simulation was calculated by starting at configurations that were not yet in equilibrium. Thus the equilibrium MSD will be slightly different for times smaller than the equilibration time. We have checked, however, that at long times the shown curves are identical to the one in equilibrium.
In Fig. 5 we show the distribution of the energy for all 16 subsystems. From the curves we see that in this case it is possible to obtain much better data than for the silica system, since the numerical demand is quite a bit smaller in the former type of system (due to the short range nature of the interactions). That also in this case the PT dynamics is indeed able to equilibrate the system is demonstrated in Fig. 6a where we show the same distribution functions reweighted to the temperature $T = 0.506$. From this figure we see that all neighboring distributions collapse nicely onto the one for $T = 0.506$, thus giving evidence that the system is indeed in equilibrium. In order to check whether this type of test is indeed sufficiently sensitive to detect whether or not the system is in equilibrium we have also made a run with standard molecular dynamics (at constant “temperature” 0.45) using as starting configuration the same as we used for the PT at that temperature. The length of this simulation was the same as the one for the PT and from earlier simulations it is known [29] that this time is not sufficient to equilibrate the system via conventional molecular dynamics. From Fig. 6b one sees that the curves stemming from the low temperatures do not fall on the master curve obtained from the higher temperatures. Thus we conclude that this way to analyze the data is indeed able to detect whether or not the system is in equilibrium and hence we have good evidence that Fig. 6a shows that the PT method has equilibrated the system.

Also in this case we judge the efficiency of the PT method by calculating the time dependence of the mean squared displacement of the A particles and
Figure 6: Probability distribution for the energy of the Lennard-Jones system for the temperatures given in the figure reweighted to $T = 0.506$. a) data from PT. b) data from conventional molecular dynamics.

compare it with the one obtained from standard molecular dynamics simulations. In Fig. 7 we show the two curves for the lowest temperature, $T = 0.45$. It is recognized immediately that also for the Lennard-Jones system the PT algorithm leads to a much faster propagation of the system through configuration space in that at long times the MSD from the PT is around 100 times larger than the one from the standard molecular dynamics.

Finally we investigate the efficiency of the PT algorithm for the third type
of system, the fully connected Potts-glass. We have done two PT simulations with $T = 0.9$ and $T = 0.7$ as the lowest temperatures. Also for this system we have checked that the distribution functions for the energy, $P_i(E)$, obtained in the different subsystems can be reweighted onto one master curve, which is again evidence that the PT algorithm does indeed equilibrate the system.

In order to see how fast this is done we have calculated the normalized spin-autocorrelation function $C(t)$:

$$C(t) = \frac{1}{N(1 - 1/q)} \sum_i^N (\delta\sigma_i(t)\sigma_i(0) - 1/q).$$

The time dependence of this quantity is shown in Fig. 8 at the two mentioned temperatures. Using configurations which have been equilibrated with the PT algorithm as starting configurations, we also did conventional single spin flip Monte Carlo simulations at the same temperature. The resulting autocorrelation functions are included in the figure also. From the different curves we recognize that at the higher temperature the PT method leads to a relaxation which is more than one decade faster than the one of the standard Monte Carlo procedure. This factor has increased to more than 100 at the lower temperature and we see that at this temperature the equilibration of the system with the standard Monte Carlo method becomes hardly feasible. We have also checked that this type of speedup is typical in that it does not depend on the realization of the disorder, i.e. the bonds $J_{ij}$ in the Hamiltonian (4) [39].
Thus we conclude that also for this glass model the PT algorithm leads to a much quicker equilibration of the system than a standard simulation method.

5 Summary

In this article we have tested to what extent the parallel tempering algorithm is useful to equilibrate glassy systems. Whereas in the past it has been shown that this method works well for lattice systems, such as Ising spin systems [11, 42], we now focused on off-lattice models. In particular we investigated a model for SiO$_2$, a network glass-former whose transport properties at low temperatures show an Arrhenius dependence, and a binary Lennard-Jones system, a glass former which is structurally similar to a hard sphere system and whose temperature dependence of the relaxation times shows at low temperatures a strong deviation from the Arrhenius law. We have found that for both types of glass-formers the PT method leads to a significant ($O(10^2)$) acceleration of the relaxation dynamics and that this factor increases even more with increasing temperature. These results are confirmed by our simulation of a fully connected Potts-glass, a frustrated system which is believed to share many similarities with structural glasses.

It must be expected that there exists a temperature $T_{g,PT}$ below which also the PT method is not able to equilibrate the system, thus hindering one to investigate the equilibrium dynamics below this effective glass transition tem-
perature. What the value of $T_{g, PT}$ is, how it depends on the type of system, the system size, or the PT exchange time $t_{PT}$, is presently not clear and thus has to be investigated in more detail. However, already now we see that this “new” algorithm (its main features dates back to 1986! [12]) will allow that computer simulations probe the static and dynamics properties of glassy systems at temperatures which are well below the ones that have been accessible so far.

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References

[1] See, e.g., Proceedings of the “Third International Discussion Meeting”, Vigo 1997, J. Non-Cryst. Solids 235-237 (1998).

[2] W. Götze, J. Phys.: Condens. Matter 10A, 1 (1999).

[3] K. Binder, J. Baschnagel, W. Kob, and W. Paul, Physics World, Dec. 1999, p. 54.

[4] W. Götze, p. 287 in Liquids, Freezing and the Glass Transition Eds.: J. P. Hansen, D. Levesque and J. Zinn-Justin, Les Houches. Session LI, 1989, (North-Holland, Amsterdam, 1991); W. Götze and L. Sjögren, Rep. Prog. Phys. 55, 241 (1992).

[5] K. Binder and G. Ciccotti (Eds.), Monte Carlo and Molecular Dynamics of Condensed Matter Systems (Italian Physical Society, Bologna, 1996).

[6] W. Kob, J. Phys.: Condens. Matter 11, R85 (1999).

[7] B. A. Berg and T. Neuhaus, Phys. Lett. B, 267, 249 (1991); Phys. Rev. Lett., 68, 9 (1992).

[8] J. Lee, Phys. Rev. Lett., 71, 211 (1993).

[9] A. P. Lyubartsev, A. A. Martsinovski, S. V. Shevkunov, and P.N. Vorontsov-Velyaminov, J. Chem. Phys., 96 1776 (1992).

[10] E. Marinari and G. Parisi, Europhys. Lett., 19, 451 (1992).

[11] K. Hukushima and K. Nemoto, J. Phys. Soc. Jpn., 65, 1604 (1996); K. Hukushima, H. Takayama, and H. Yoshino, J. Phys. Soc. Jpn., 67, 12 (1998).
[12] R. H. Swendsen and J. S. Wang, Phys. Rev. Lett., 57, 2607 (1986).

[13] U. H. E. Hansmann, Y. Okamoto, and F. Eisenmenger, Chem. Phys. Lett., 259, 321 (1996).

[14] N. Nakajima, H. Nakamura, and A. Kidera, J. Phys. Chem. B, 101, 817 (1997).

[15] M. Achenbach, Diploma Thesis, (Johannes-Gutenberg Universität, Mainz, 1998).

[16] Y. Sugita, Y. Okamoto, Chem. Phys. Lett., in press.

[17] R. Yamamoto and W. Kob, Phys. Rev. E 61, xxxx (2000); preprint cond-mat/0001042.

[18] K. K. Bhattacharya and J. P. Sethna, Phys. Rev. E, 57, 2553 (1998).

[19] B. Coluzzi and G. Parisi, J. Phy. A 31 4349 (1998).

[20] C. A. Angell, in Relaxation in Complex Systems, Eds.: K. L. Ngai and G. B. Wright (US Dept. Commerce, Springfield, 1985).

[21] B. W. H. van Beest, G. J. Kramer, and R. A. van Santen, Phys. Rev. Lett. 64, 1955 (1991).

[22] K. Vollmayr, W. Kob, and K. Binder, Phys. Rev. B 54, 15808 (1996).

[23] T. Koslowski, W. Kob, and K. Vollmayr, Phys. Rev. B 56, 9469 (1997).

[24] J. Horbach and W. Kob, Phys. Rev. B 60, 3169 (1999).

[25] J. Horbach, W. Kob, and K. Binder, J. Phys. Chem. B 103, 4104 (1999).

[26] P. Jund and R. Jullien, Phys. Rev. B, 59, 13707 (1999).

[27] J. Horbach, W. Kob, and K. Binder, preprint cond-mat/9910507.

[28] T. Stühn and W. Kob, unpublished.

[29] W. Kob and H. C. Andersen, Phys. Rev. Lett. 73, 1376 (1994); Phys. Rev. E, 51, 4626 (1995); ibid., 52, 4134 (1995).

[30] K. Vollmayr, W. Kob, and K. Binder, J. Chem. Phys. 105, 4714 (1996).

[31] T. Gleim, W. Kob, and K. Binder, Phys. Rev. Lett. 81, 4404 (1998).

[32] F. Sciortino, W. Kob, and P. Tartaglia, Phys. Rev. Lett. 83, 3214 (1999).
[33] C. Donati, S. C. Glotzer, P. H. Poole, W. Kob, and S. J. Plimpton, Phys. Rev. E 60, 3107 (1999).

[34] W. Kob and J.-L. Barrat, Eur. Phys. J. B 13 319 (2000).

[35] D. J. Elderfield and D. Sherrington, J. Phys. C 16, L4971 (1983);
   D. J. Gross, I. Kanter, and H. Sompolinsky, Phys. Rev. Lett. 55, 304 (1985);
   G. Cwilich and T. R. Kirkpatrick, J. Phys. A 22, 4971 (1989).

[36] T. R. Kirkpatrick and D. Thirumalai, J. Phys. A 22, L149 (1989);
   D. Thirumalai and T. R. Kirkpatrick, Phys. Rev. B 38, 4881 (1988).

[37] G. Parisi, J. Phys. A, 30, L765 (1997).

[38] E. De Santis, G. Parisi, and F. Ritort, J. Phys. A: 28, 3025 (1995).

[39] C. Brangian, W. Kob, and K. Binder, unpublished.

[40] A. M. Ferrenberg and R. H. Swendsen, Phys. Rev. Lett., 61, 2635 (1988).

[41] P. Scheidler, W. Kob, J. Horbach, and K. Binder, p. 131 in Proceedings of Neutrons and Numerical Methods Grenoble, Dec. 1998, Eds. M.R. Johnson, G.J. Kearley, and H.G. Büttner, AIP conference Proceedings 479, (AIP, Woodbury, 1999).

[42] J.-S. Wang and R.H. Swendsen, Phys. Rev. B, 37, 7745 (1988); ibid. 38, 4840 (1988).