Dynamical behavior of the Biroli-Mezard model

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Abstract. Some essential dynamical features of the Biroli-Mezard model are discussed. We observe a dynamical slowing down which for higher densities depends both on the observation time and the age of the system. We relate this aging behavior to the real space dynamics of the system, where due to some essential initial restructuring the system gains mobility. This allows us to get a better understanding of what is causing the two-time dependence of the correlation function. Finally, we quantify how heterogeneous the slow dynamics of the BM model is by investigating the dynamical susceptibility and discuss its dependence on the age of the system.

1. Introduction
By now, glasses have been under investigation for several decades [1]. Nevertheless, quite some of their fundamental mechanisms are yet to be understood. Experiments have showed that the slow dynamics of these systems possesses highly non-trivial space-time correlations [2], though the characteristic size of the dynamical heterogeneities is far from established. Hysteresis effects in heating-cooling cycles and the influence of the cooling rate when producing glassy systems clearly evidence their out-of-equilibrium status. Meanwhile, the current state of the art to describe this non-equilibrium dynamics does not go much further than phenomenological equations [3]. On the theoretical front several approaches have been explored to obtain some further insight also into why perhaps the experimental methods are not adequate enough to reveal some of the fundamental underlying physics of glasses. Mode coupling theory (MCT) was one of the first approaches which seemed relatively appropriate, but complications emerging when extending it to lower temperature regimes stimulated some alternative approaches. With the emergence of kinetically defined lattice gas models, such as the Kob-Andersen model or the triangular lattice gas, is quite extensive [5, 6, 7, 8]. These systems have a trivial equilibrium behavior. Nevertheless, the introduction of some simple kinetic rules allows to observe stretched
exponential relaxation, which in case of the triangular lattice gas clearly consists of two steps as predicted by MCT, and dynamical heterogeneities. The simple equilibrium situation which on the one hand allows for easy computer simulation, necessitates the inclusion of gravity effects or possible exchange with a particle reservoir to admit for the observation of out-of-equilibrium effects [9, 10]. The main disadvantage of these systems is the impossibility of a dynamic arrest in such finite size lattice gas models [7].

More recently, thermodynamically defined lattice gas models [11, 12, 13] have been introduced precisely in order to overcome this a priori exclusion of the existence of a thermodynamic transition, whether dynamic or static of nature. Again one observes the slow relaxation process, whether or not composed out of two steps, which is most probably related to the non-trivial spatio-temporal correlations. Their main drawback is that simulating these systems is computationally more demanding. On the other hand, as we investigate extensively during the course of this paper, they do not require the introduction of any external source to observe out-of-equilibrium effects: they are inherently out-of-equilibrium systems. Most interestingly, the mean field versions can be analyzed analytically, and reveal to have a phenomenology quite similar to that of mean field discontinuous spin glass models. So far, the analysis for finite models is restricted to simulations. The good news, however, is that at least until now all possible scenarios for interpreting the glass transition remain open for these type of models. Finally, these systems crystallize, putting simulations somewhat on the same level as experiments. So, on one hand it seems we defeat our purpose of simplifying the study by concentrating on simulations. On the other hand, we can still follow these systems rather closely and actually visualize them, so they could actually help to close the gap between simulation and experiment, while they also have the advantage of being analytically tractable in the mean field case. Of course, the connection between mean field and finite dimensional systems is not evident at all, which is why in any case we shall also closely investigate the extent to which our outcome is prone to finite size effects.

1.1. The Biroli-Mezard model
The BM model is a lattice gas model in which each particle can have at most \( l \) neighboring particles. Such a configuration can be achieved by performing a soft annealing procedure as was proposed in the original paper [11]. In case only one type of particle is present, the system crystallizes completely, which is why we shall consider mixtures of particles [14]. More precisely, all the following results are obtained for three-dimensional lattice models containing 30\% of particles with the constraint \( l = 1 \) and 70\% of particles for which \( l = 3 \). As mentioned in the introduction, the mean field version of such single-particle models has been studied in [15]. They found that the model both undergoes a static and a dynamic transition, which could eventually be preceded by a kinetic transition. Of course, this mean field picture does not guarantee the same should be valid for finite dimensional models. But at least it is clear from the definition of the BM model that there must exist a limiting density \( \rho_\infty \), most likely topology dependent, above which the BM model is no longer thermodynamically defined. Moreover, the thermodynamic definition does not only make the equilibrium dynamics non-trivial, as opposed to the KA model, its kinetically defined counterpart, it gives no guarantee for phase space to be connected. The energy of an allowed BM configuration is considered to be zero, while a prohibited configuration has infinite energy. Thus, the resulting energy landscape is simplified with respect to real systems in a sense that valleys do not contain many subvalleys, but it is certainly not trivial and could eventually be partitioned into several fully disconnected sections.

It is important to stress the difference between the kinetically defined KA model and the thermodynamically defined BM model. While in the former the slow dynamics can be discussed mostly in terms of the presence of local vacancies [16], the local dynamics of the BM model depends both on the local vacancies and neighboring particles. This is better explained in figure 1 for a two-dimensional model with particles constrained by \( l = 2 \). From that figure
it is clear that even though the dynamics in both models becomes necessarily cooperative for sufficiently high densities, they are very different in origin.

Figure 1. In case of the KA model the encircled particle can move both up or to the right. The same particle in the BM model will remain immobile until either its lower neighbor first moves to the left, or the particles in grey scale (its eventual future neighbors) first loose one of their neighbors.

In the remaining of this paper we will focus on a three-dimensional realization of the BM model with the above mentioned mixture. Both density and the linear length of the cubical sample are varied and the resulting differences are discussed appropriately. It was already suggested in [11] that for this three-dimensional mixture a dynamic transition should occur around \( \rho_c \sim 0.565 \), which is why we did not perform simulations on systems denser than these. The linear size \( L \) of the systems varies from 10 to 30 times the lattice constant.

2. Ageing dynamics

A short numerical analysis of the dynamics of the BM model was already given in the original paper. It was clear the BM model relaxes in a non-exponential way as is typical for glasses, and it was mentioned the system ages. In this section we investigate the nature of this ageing behavior further by considering the two-time correlation function

\[
C(t, t_w) = \frac{1}{V \rho (1 - \rho)} \sum_i \left[ n_i(t) n_i(t_w) - \rho^2 \right].
\]

Here \( V \) is the volume of the system, ie, the total number of sites, and \( n_i \) is the occupation number of site \( i \), either at the waiting time \( t_w \) (the time elapsed since the preparation of the system), or at time \( t \), ie, the actual age of the system (the total simulation time).

Figure 2. Two-time correlation functions \( C(t, t_w) \) as a function of the logarithm of the elapsed time \( t - t_w \) (since the waiting time \( t_w \)), for a system of linear size \( L = 30 \) and particle density \( \rho = 0.542 \). During the initial time window, the upper curves correspond to shorter waiting times, while at later elapsed times they are the lower ones.

In figure 2 we present our results for this correlation function in case of the largest system investigated with density \( \rho = 0.542 \). Clearly the system relaxes in a non-exponential way. Roughly speaking, we can state it initially decays faster. As the observation time \( t - t_w \) increases, the system decorrelates more and more slowly. The initial regime could be identified with the so-called local \( \beta \)-relaxation of MCT. MCT predicts the existence of a plateau separating an
earlier from a later $\beta$-regime, which is not present in the case of the BM model. The subsequent slower decay would then be the $\alpha$-relaxation.

Although no analytical predictions in terms of both the age of the system and the observation time are yet available, one would generally not expect the two different types of ageing regimes observed here. For short elapsed times $t - t_w$, older systems decay faster than their younger counterparts. At later times this effect is clearly overcome and the more standard form of ageing is recovered for which older systems forget their original configuration more slowly. In the following section we investigate the origin of the initial acceleration in older systems by directly visualizing the system in real space. Whatever might be the cause, it is clear from the above picture that the initial effect is overcome at longer observation times. We indicate this time at which the crossover occurs by $t_A$. This crossover time $t_A$ occurs at later times for denser and larger systems.

Considering the time window right of $t_A$ we thus recover the system ages. In case of the BM model, this can be interpreted in terms of its non-trivial energy landscape. When the system is created it finds itself in a certain region of phase space and as time passes it explores more of this available energy landscape. A young system clearly has very little information on the structure of phase space. Older systems on the other hand, having explored much more of it, can get increasingly trapped in certain regions of it. Of course, for the BM model the trapping only consists in entering a region with fewer escaping directions. It is just horizontally trapped, not vertically. This simplified version of the energy landscape has the advantage of being easier to simulate. At the same time, it allows the system to escape a certain region of phase space more easily. In any case, it seems the knowledge of the surrounding phase space causes older systems to decorrelate more slowly.

![Figure 3. Two-time correlation functions $C(t, t_w)$ as a function of the logarithm of the elapsed time $t-t_w$ (since the waiting time $t_w = 16777216$) for systems of different size and fixed particle density $\rho = 0.535$.](image)

For the systems of linear size $L = 30$ we find the initial configuration, ie, the configuration of the system at $t_w$, is never fully forgotten during the length of our simulations. Smaller, and less dense systems do decorrelate completely during our run. This is presented in figure 3.

Finally we mention we have tried to apply some of the usual two-time scaling laws to our results of the correlation function. Unfortunately, the presence of the unexpected initial acceleration of older systems does not allow us to present any rigorous results on this account.

3. Crystallization
As is clear from the previous section, the BM model reveals some typical glassy behavior, such as the non-exponential relaxation and standard ageing. On the other hand, it does suffer some unexpected acceleration during an initial time window. To clarify this point further and to understand better in general what is structurally occurring, we compute the dynamic structure
factor

\[ S_k(t) = \frac{\langle \rho_k^* \rho_k \rangle}{N}, \]  

(2)

where \( \rho_k \equiv \sum_{i=1}^{N} \exp(ikr_i) \). At the time we have just created the desired BM configuration, the structure factor is typical of a glassy system. As time passes, this remains the case in most directions of the wavevector \( k \). However, for some specific values of \( k \), namely directions close to the body diagonal, the dynamic structure factor does grow during a limited time interval. We present the structure factor in these relevant directions in figure 4(a) for the smallest system investigated. Similar pictures are obtained for systems of different densities and sizes. Thus the system is crystallizing during a limited intermediate time interval. Nevertheless, during that same time interval we already recover the system is out-of-equilibrium. To get a better understanding of what this growing of the structure factor actually entails, we visualize an exemplary configuration, obtained at the end of the simulation in figure 4(b). No small nuclei are recovered, but rather the whole system has stratified in a direction close to the body diagonal. Several dense planes are separated from each other by usually one less dense plane. The fact that this restructuring process is limited to a restricted time interval and the fact that the eventual result is not so regular at all, reassures us the kinetics of the system is not fully dominated by crystallization. However, it is important to consider the role this restructuring plays in the overall dynamical observations of the BM system. The difficulty in distinguishing between the dynamics of a highly disordered polycrystal and a locally stable glassy phase was already addressed in [17].

By computing the average local density \( \rho_{nn}(t) \) of each particle, ie, the number of neighboring occupied sites each particle has on average, one gets an idea of the local environment and the possible effect of the overall restructuring on the local dynamics. The result for \( \rho_{nn}(t) \) for the same system as in figure 4(a) is presented in figure 5. We observe that due to the global restructuring the local environment of the particles becomes less dense. Also included in the same figure is the density \( \rho_{mobile}(t) \) of mobile particles at a given time \( t \) during the simulation. As is to be expected, the fact that the direct neighborhood of the particles becomes less dense during a certain time interval, results in a higher percentage of particles which will be allowed to move at the next time step.

In summary, the growth of the dynamic structure factor corresponds to an overall restructuring of the system, though it is restricted to a limited time interval. On a local level, it results in a less dense environment around each particle, making them in turn more mobile.
Turning back to our findings on the two-time correlation function, we are now able to interpret what is going on during the initial observation time window. As we start observing, older systems will already have restructured and have a higher overall mobility with respect to their younger counterparts. From the decay of the correlation function it is clear that this gain in mobility allows these older systems to initially decay more quickly. At later elapsed times, however, the higher local mobility of older systems is no longer enough to help them relax faster. More even, the subsequent dominating relaxation mechanism must be much stronger than the initial mobility increase, since at large elapsed times we only recover the standard ageing behavior.

4. Dynamical heterogeneities
The extent to which the dynamics of a system is heterogeneous can be measured by computing the dynamic susceptibility \[ \chi(t, t_w) = V \times \left( \langle C(t, t_w)^2 \rangle - \langle C(t, t_w) \rangle^2 \right) \tag{3} \]
A detailed one-time analysis of this quantity, where \( t_w = 0 \), is given for equilibrium systems in [19], which we will take as a reference point.

The results for samples of linear size \( L = 10 \) are given in figure 6. During a short initial time window the dynamic susceptibility grows with the elapsed time according to a power law with exponent \( \mu \sim 0.6 \). This maps fairly well to the elastic regime for which a power-law with exponent 0.5 was predicted. Moreover, one expects the dynamics of a system to be independent of the age of the system during an elastic regime as is the case for our BM mixture.

The subsequent time window corresponds to the early \( \beta \)-regime. Originating from MCT, this is generally associated to the “rattling” of particles in boxes constituted by their neighbors.
Again an approximate power-law with now a smaller exponent $\mu \sim 0.2$ is observed. As for the elastic regime, such local dynamics should be age independent as is observed. In case the correlation function revealed a plateau, so should the dynamic susceptibility, but for the BM model this is not the case. Rather, the next regime is distinguishable by its $t_w$ dependent exponent $\mu$. This late $\beta$-regime generally signals the onset of a more cooperative relaxation process. Therefore it should indeed be rather sample dependent confirmed by the steeper slope in our log-log plot. Moreover, the beginning of this late $\beta$-regime coincides with $t_A$, i.e., the time at which the standard ageing dynamics overcomes the initial preparation effect. This confirms not only the close relationship between the $\beta$-regime and the degree to which the dynamics is cooperative, but also suggests a strong link between the latter and the state of being out-of-equilibrium.

The late $\beta$-regime continues into the so-called $\alpha$-regime. The dynamic susceptibility is then expected to peak around the $\alpha$-relaxation time $\tau_\alpha$, which is the characteristic time of the structural relaxation dynamics. Though we do not have any absolute measures of these characteristic relaxation times in the BM model, the time $t^*$ at which the dynamic susceptibility peaks is smaller than the time at which the system has completely lost its memory. If $t^*$ does depend on the waiting time, it seems that at least it is not a very strong dependence. The actual value of $\chi(t, t_w)$ around $t^*$ does grow with the age of the system, confirming older systems necessarily relax in a more cooperative way.

Predictions on the final decay of the dynamic susceptibility are probably the least solid. First of all because we are only able to observe such a clear decay for the smallest systems investigated. Also, in the case of lattice gases with trivial equilibrium behavior one can analytically check the system should evolve to a state with a constant dynamic susceptibility, namely one. However, in the case of the BM model, no assumptions can be made on whether the expectation values of the correlation functions of the occupation numbers $n_i$ can be factorized, making it much harder to analytically establish a limiting value of the dynamic susceptibility. Numerically, we find the dynamic susceptibility decays towards a waiting time and density dependent value. The $t_w$-dependency is of course due to the fact that we are dealing with a system which is out-of-equilibrium. The dependency on the density of the final value of $\chi(t, t_w)$ suggests the dynamics is intrinsically heterogenous.

5. Conclusion
In this paper we analyzed the dynamics of the thermodynamically defined BM model. At first sight the local dynamics seems quite similar to the purely kinetic lattice gas models. However their dynamical behavior reveals to be much richer, though unfortunately also sometimes more problematic to deal with. In fact we did find the model has the tendency to crystallize, though for the particular mixture we investigated this is certainly not the main driving force behind its dynamics. During a later observation time window the system shows the typical glassy pathology, namely it ages. We suggest that the non trivial energy landscape of the BM model causes the correlation function to depend both on the age of the system and the observation time. Moreover, the analysis of the dynamic susceptibility suggests a strong link between cooperative relaxation and the ageing dynamics. Finally, we performed a detailed analysis of the dynamic susceptibility in order to get a better understanding of how and when the dynamics becomes heterogeneous in the BM model.

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