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Cross-over from entropic to thermal dynamics in glassy models

Eric M Bertin

CEA-Service de Physique de l'État Condensé, Centre d'Études de Saclay, F-91191 Gif-sur-Yvette Cedex, France

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Abstract

We study the emergence of a cross-over from entropically driven to thermally activated dynamics in different versions of the 'entropic' phase space model introduced by Barrat and Mézard, and previously considered in the zero temperature limit. We first focus on the low temperature ($T \leqslant T_g$) ageing phase of the original model and show that a short time singularity appears in the correlation function for $T_g/2 < T < T_g$, leading to dynamical ultrametricity at $T = T_g$. We then consider the finite size version of this model, showing that the long time dynamics is always thermally activated beyond a size dependent cross-over time scale. We also generalize the model, introducing a threshold energy so as to mimic a phase space composed of saddles above the threshold, and minima below. In this case, the cross-over time scale becomes much smaller than the equilibration time, and both kinds of ageing dynamics are successively found, inducing a non-trivial ageing scaling which does not reduce to the usual t/t_w (or even t/t_w^v) one.

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A major part of our understanding of the glass transition comes from mode coupling theory (MCT) for structural glasses [1, 2] and from dynamical mean-field theory for spin glasses [3, 4], which are the most sophisticated microscopic theories available in glassy physics. Apart from these theories, the energy landscape paradigm has raised considerable interest in recent years [5–8], due to the increasing power of computer simulations which now allow physicists to implement the seminal ideas of Goldstein [9], specified later by Stillinger and Weber [10]. These studies have suggested the association of the mode coupling transition temperature T_c , where MCT locates an ideal glass transition, with a threshold energy E_{th} of geometric nature, separating regions of phase space dominated by saddle points for $E > E_{th}$ from regions dominated by minima for $E < E_{th}$ [11–15]. In this framework, the ideal glass transition found in MCT and in mean-field theories is explained on the one hand by the sharp separation between saddles and minima, and on the other hand by the divergence of the energy barriers surrounding minima [16].

If one of these two conditions was not fulfilled, the ideal glass transition would be smeared out, which indeed happens in real glass formers [2]. In particular, if energy barriers remain finite, one may expect not only the behaviour of the relaxation time with temperature to be smoothed, but also correlation functions to reflect this dynamical cross-over between a time regime when saddles are mainly explored and a later regime in which the system visits minima in majority. The key point here is that visiting minima corresponds to thermally activated dynamics where the system has to overcome energy barriers, which thus depends crucially on temperature, whereas visiting saddles are associated with a rather temperature independent kind of dynamics, sometimes called 'entropic dynamics': the evolution of the system is mainly limited by the lack of escape downwards directions, when visiting a saddle with only a few negative eigenvalues. Interestingly, this dynamical transition has already been observed in Lennard–Jones supercooled liquids [17, 18], but a quantitative analysis of the associated cross-over scales has not been achieved yet.

In this paper, we study quantitatively this cross-over and its dependence upon relevant parameters in simple stochastic models of glassy dynamics. We first consider the phase space model introduced by Barrat and Mézard [19]—hereafter denoted as BM model—which has seen a recent surge of interest in the context of fluctuation dissipation relations [20, 21], and study the ageing regime of the correlation function $C(t_w, t_w + t)$ in the whole glassy phase $T < T_g$, generalizing previous results obtained in the limit $T \to 0$. We find a temperature independent long time behaviour $(t \gg t_w)$, as well as a short time singularity $(t \ll t_w)$ in the temperature range $T_g/2 < T < T_g$, which leads to dynamical ultrametricity [22] for $T = T_g$, as in the trap model [23]. We then study the correlation function in the *finite size* BM model, both in equilibrium and in the ageing regime, which allows us to demonstrate clearly the existence of a dynamical cross-over from entropic to activated dynamics. Finally, the BM model is generalized by introducing a threshold energy so as to mimic a phase space structure composed of saddles and minima. We show that the change of ageing dynamics (from entropic to thermally activated) in this model leads to a non-trivial scaling behaviour, and that the cross-over time scale can be much smaller than the equilibration time.

1. The Barrat-Mézard model

One of the first attempts to describe ageing dynamics with simple stochastic phase space models was the trap model proposed by Bouchaud [24], with the aim of suggesting a possible general scenario for glassiness, which emphasizes activation effects. A few years later, Barrat and Mézard [19] proposed a somewhat similar model, but where thermal activation was replaced by an 'entropic activation', i.e. by the fact that at low temperature fewer and fewer lower energy states could be reached when the system is already in a low energy state. In a continuous energy formulation, corresponding to an infinite number of states, the BM model is defined as a Markovian process where a 'particle' (which usually corresponds to the representative point of a system in its phase space) is allowed to jump from one energy state to any other one. These energy states are distributed according to an *a priori* distribution $\rho(E)$, and transition rates W(E'|E) from energy E to energy E' correspond to the Glauber choice:

$$W(E'|E) = \frac{\Gamma_0 \rho(E')}{1 + e^{(E'-E)/T}} \tag{1}$$

where Γ_0 is a microscopic frequency scale, which we shall set to unity in the following. The dynamics of the BM model is then described by the probability P(E, t) of having energy E at

time t, which satisfies the following master equation:

$$\frac{\partial P}{\partial t} = \int_{-\infty}^{0} dE'[W(E|E')P(E',t) - W(E'|E)P(E,t)] \tag{2}$$

where for simplicity—but without loss of generality since we are interested in the low energy dynamics—we have restricted $\rho(E)$ to be non-zero only for E < 0.

1.1. Zero temperature results

Contrary to the trap model, and to any model driven by thermal activation, the temperature can be set to zero without freezing completely the dynamics, leading to an evolution somewhat similar to the steepest descent dynamics in a high dimensional continuous energy landscape. A notable property of this model in the zero temperature limit, which has been studied in details in [19], is that the dynamics, expressed in terms of sojourn times instead of energies, becomes completely independent of the functional form of the distribution $\rho(E)$; in particular, $\rho(E)$ can be bounded or not. Defining the average sojourn time $\tau(E)$ at energy E by the relation

$$\frac{1}{\tau(E)} \equiv \int_{-\infty}^{0} dE' W(E'|E) \tag{3}$$

which reduces for T=0 to $\tau(E)^{-1}=\int_{-\infty}^E \mathrm{d}E'\rho(E')$, one finds that the *a priori* distribution $\psi_0(\tau)$ derived from $\rho(E)$ is given by $\psi_0(\tau)=\tau^{-2}\theta(\tau-1)$, independently of $\rho(E)$. As a result the dynamical distribution $p(\tau,t)$, which has been computed in [19], does not depend on the shape of $\rho(E)$. It was found that $p(\tau,t)$ cannot reach a steady state, but exhibits for large t a scaling form $p(\tau,t)=t^{-1}\phi_0(\tau/t)$, typical of the ageing behaviour, with $\phi_0(u)$ given by

$$\phi_0(u) = \frac{1}{u^2} e^{-1/u}.$$
 (4)

Defining the correlation function $C(t_w, t_w + t)$ as the probability of not moving between t_w and $t_w + t$, which can be written as

$$C(t_w, t_w + t) \equiv \int_{-\infty}^{0} dE P(E, t_w) e^{-t/\tau(E)}$$
 (5)

one finds for T = 0 and large t_w :

$$C(t_w, t_w + t) = \frac{t_w}{t_w + t}. (6)$$

Note that in the 'short time' regime defined by $1 \ll t \ll t_w$, the correlation function $C(t_w, t_w + t)$ behaves linearly, in the sense that $1 - C \sim t/t_w$; this will be important in the discussion of the finite temperature results.

1.2. Ageing at finite temperature

If one chooses an exponential density of states $\rho(E) = \beta_g \exp(\beta_g E)\theta(-E)$, as is usually done in the trap model, then the Boltzmann equilibrium distribution becomes non-normalizable for $T \leqslant T_g \equiv 1/\beta_g$. In this case, the dynamical distribution drifts towards deeper and deeper energies, leading to the ageing phenomenon, as can be shown for instance from the behaviour of the correlation function. In this section, we present the new results we have obtained in the ageing regime for the finite temperature case, namely for $0 < T < T_g$.

As was done for the trap model in [25], one can then look for a scaling solution for the dynamical energy distribution. A natural scaling variable would be the ratio of the typical time $\tau(E)$ the system spends in a state with energy E—as defined in equation (3)—to the time t elapsed after the quench. Computing $\tau(E)$ for $0 < T < T_{\rm g}$ and large |E| yields

$$\tau(E) = \frac{\sin \pi \mu}{\pi \mu} e^{-E/T_g} \tag{7}$$

where $\mu \equiv T/T_g$. We choose a scaling variable u proportional to $\tau(E)/t$, defining it through $u^{-1} = t \, \mathrm{e}^{E/T_g}$. This leads to a distribution $P(E,t) = \beta_g u \phi(u)$, where $\phi(u)$ is normalized by $\int_0^\infty \mathrm{d}u \, \phi(u) = 1$. Once these substitutions are made in equation (2), the following equation is obtained, using the integration variable $v = t^{-1} \, \mathrm{e}^{-E'/T_g}$:

$$u^{2}\phi'(u) + (u - J)\phi(u) = -\frac{1}{u} \int_{0}^{\infty} dv \frac{\phi(v)}{1 + (v/u)^{1/\mu}}$$
 (8)

where $J \equiv \pi \mu/\sin \pi \mu$. The asymptotic behaviour of the distribution P(E, t) can be extracted rather easily from this equation. Let us analyse for instance the large u behaviour of $\phi(u)$ —or large |E| behaviour of P(E, t). In this case, the integral on the rhs of equation (8) reduces to

$$\int_0^\infty dv \frac{\phi(v)}{1 + (v/u)^{1/\mu}} \simeq \int_0^\infty dv \,\phi(v) = 1 \qquad u \gg 1$$
 (9)

so that equation (8) reads, neglecting also J with respect to u and calling $\phi_l(u)$ the asymptotic large u expression of $\phi(u)$,

$$u^{2}\phi'_{l}(u) + u\phi_{l}(u) = -\frac{1}{u}.$$
(10)

The general solution of this equation reads

$$\phi_l(u) = \frac{A}{u} + \frac{1}{u^2}. (11)$$

The constraint that $\phi(u)$ must be normalizable requires that A=0, so that $\phi_l(u)=u^{-2}$, in agreement with the results obtained in the zero temperature limit—see equation (4). Interestingly, we find here the *exact* asymptotic expression for $\phi(u)$, including the prefactor—equal to 1—although we performed only an asymptotic analysis on a linear equation. Note also that taking into account the leading correction to the approximation made in equation (9) leads to $\phi_l(u) - u^{-2} \sim u^{-3}$. Neglecting these corrections, one thus finds that P(E,t) is equal to $\beta_g t \, \mathrm{e}^{E/T_g}$ —i.e. proportional to $\rho(E)$ —for $\mathrm{e}^{|E|/T_g} \gg t$.

In the opposite case $u \ll 1$, the equation governing the asymptotic expression $\phi_s(u)$ of $\phi(u)$ reads

$$u^{2}\phi'_{s}(u) - J\phi_{s}(u) = -\int_{0}^{\infty} dz \frac{\phi_{s}(uz)}{1 + z^{1/\mu}}.$$
 (12)

Searching for a power law solution $\phi_s(u) = au^{\alpha}$, the above equation is satisfied only if

$$J = \int_0^\infty dz \frac{z^\alpha}{1 + z^{1/\mu}} \tag{13}$$

since the first term $u^2\phi'_s(u)$ can be neglected in this case. Replacing by the respective values of both sides, one has

$$\frac{\pi\mu}{\sin\pi\mu} = \frac{\pi\mu}{\sin(1+\alpha)\pi\mu}.$$
 (14)

The convergence of the integral appearing in equation (13) requires that $0 < (1 + \alpha)\mu < 1$. In this range, equation (14) admits two solutions: $\alpha = 0$ and $\alpha = 1/\mu - 2$. Since the zero

temperature limit of the model is regular, the finite temperature solution must converge towards the zero temperature distribution given by equation (4), which decays faster than any power law for $u \to 0$, so that one expects $\alpha \to \infty$ for $\mu \to 0$. The correct solution for $\phi(u)$ is thus

$$\phi(u) \sim u^{1/\mu - 2} \qquad u \ll 1.$$
 (15)

So the energy distribution behaves at small |E|—i.e. for $e^{|E|/T_g} \ll t$ —as

$$P(E,t) \sim t^{1-1/\mu} e^{(1-1/\mu)E/T_g} \sim t^{1-1/\mu} \rho(E) e^{-E/T}$$
 (16)

which means that small energies are almost equilibrated.

Turning to the correlation function $C(t_w, t_w + t)$ defined above, equation (5) can be rewritten in terms of the scaling function $\phi(u)$, using also equation (7):

$$C(t_w, t_w + t) = \int_0^\infty du \, \phi(u) \, e^{-Jt/ut_w}$$
 (17)

which shows that the correlation function exhibits a simple ageing form,

$$C(t_w, t_w + t) = C_{ag}(t/t_w). \tag{18}$$

Using the above asymptotic results for $\phi(u)$, one can find the short time and long time behaviour of the correlation function. In particular, for times $t \gg t_w$, $C(t_w, t_w + t)$ is given by

$$C(t_w + t, t_w) \simeq \frac{\sin \pi \mu}{\pi \mu} \frac{t_w}{t} \qquad t \gg t_w$$
 (19)

for any μ < 1, so that the 'tail' of the correlation function appears to be a temperature independent power law, apart from the prefactor. This point will be important for later discussions.

The short time behaviour is computed from

$$1 - C(t_w, t_w + t) = \int_0^\infty dv \, \phi(v) (1 - e^{-Jt/vt_w}). \tag{20}$$

For $t \ll t_w$, one can try to linearize the exponential in the last factor. Using equation (15), one sees that the resulting integral converges at its lower bound only if $\mu < \frac{1}{2}$; in this case, the rescaled correlation function $\mathcal{C}_{ag}(u)$ is then regular when $u \to 0$ (i.e. $1 - \mathcal{C}_{ag}(u) \sim u$), just as in the zero temperature case. In contrast, for $\frac{1}{2} < \mu < 1$, a singularity appears, and one then finds

$$1 - \mathcal{C}_{ag}\left(\frac{t}{t_w}\right) \sim \left(\frac{t}{t_w}\right)^{(1-\mu)/\mu} \qquad t \ll t_w. \tag{21}$$

This property can be interpreted as follows: the average energy $\langle E' \rangle_{\rm E}$ reached in a transition from E to E' reads

$$\langle E' \rangle_{\mathcal{E}} \equiv \tau(E) \int_{-\infty}^{0} dE' \, E' W(E'|E) = E - \frac{\pi \, \mu}{\tan \pi \, \mu} T_{\mathcal{g}}. \tag{22}$$

For $\mu < \frac{1}{2}$, $\langle E' \rangle_{\rm E} < E$ so that the dynamics resembles the zero temperature one: at each step, the energy is lowered on average. In contrast, for $\mu > \frac{1}{2}$, $\langle E' \rangle_{\rm E} > E$ and the energy is *raised* on average at each transition. So the slow drift of the average energy $\overline{E}(t)$ as a function of time results in this case from a kinetic effect: the particle stays a longer time on lower energy states. Actually, the particle typically jumps a number of times among high energy states before reaching a low energy one, then jumps back to high energies and so on. This scenario is reminiscent of what happens in the trap model, for which $\langle E' \rangle_{\rm E} = T_{\rm g}$ independently of the starting energy E, and is presumably responsible for the onset of the short time singularity. Note however that in the BM model, *both* entropic and thermal dynamics come into play for

 $1/2 < \mu < 1$, the latter being rather irrelevant for $\mu < 1/2$, but thermal activation remains in any case an auxiliary mechanism, the main one being entropic. Indeed, this can be seen from the typical sojourn time $\tau(E)$ —see equation (7)—which is always proportional to e^{-E/T_g} , and not $e^{-E/T}$ as would be the case for a purely thermal dynamics. So the present scenario is slightly different from the usual landscape picture, since the entropic channel exists here even at very low energies. We shall see in the next sections how to generalize the model in order to account for a complete vanishing of downward escape directions.

We now present numerical results in order to confirm the above analysis. computational reasons, simulations were performed with a slightly different version of the model, in which the transition rates are given by

$$\tilde{W}(E'|E) = \Gamma_0 \rho(E') \qquad E' \leqslant E$$

$$\tilde{W}(E'|E) = \Gamma_0 \rho(E') e^{-(E'-E)/T} \qquad E' > E.$$
(23)

$$\tilde{W}(E'|E) = \Gamma_0 \rho(E') e^{-(E'-E)/T}$$
 $E' > E.$ (24)

This choice is motivated by the following remarks: in order to simulate numerically the dynamics of the BM model in the continuous energy formalism (corresponding to an infinite number of states), the new energy E' is randomly chosen at each step, with a probability proportional to the transition rate $\tilde{W}(E'|E)$. To this aim, one needs to compute the integral:

$$I(E') = \tau(E) \int_{-\infty}^{E'} dE'' \, \tilde{W}(E''|E). \tag{25}$$

The value of E' is then determined by choosing a uniform random variable $u \in [0, 1]$, and by inverting the relation u = I(E') so as to get E' as a function of u. The advantage of the transition rates $\tilde{W}(E'|E)$ is that they allow us to compute analytically I(E'), as well as $E' = I^{-1}(u)$, which is not possible with the Glauber rates W(E'|E)—one would then resort to a more complex and less accurate numerical determination of $I^{-1}(u)$. These transition rates $\widetilde{W}(E'|E)$ behave qualitatively as the usual Glauber rates, so that one expects this difference not to affect the scaling and power law behaviours, but only the numerical prefactors. One first needs to check the ageing form—equation (18)—of the correlation function. This is done in figure 1, which displays the correlation function $C(t_w, t_w + t)$ for $\mu = 0.7$ and several waiting times, $t_w = 10^4$, 10^5 , 10^6 and 10^7 . The rescaling, shown in the inset, is very good, with only small deviations in the short time regime $t \ll t_w$ where finite time effects become stronger. We also test the predicted exponents for the short time and late time behaviours of the correlation, as shown in figure 2. One clearly sees the linear behaviour of 1-C for $\mu<\frac{1}{2}$, as well as the predicted singularity with exponent $(1 - \mu)/\mu$ for $\frac{1}{2} < \mu < 1$. It is however difficult to find a genuine straight line for this short time singularity since t must satisfy $1 \ll t \ll t_w$, and such a separation of scales is hard to obtain numerically (note that t_w is already as large as 10⁷). The late time regime is easier to characterize quantitatively, since the only condition is $t \gg t_w$; the corresponding power law behaviour proportional to $(t/t_w)^{-1}$, with a temperature independent exponent, is clearly shown in the inset of figure 2. As explained above, we cannot test quantitatively the prefactor predicted in equation (19) since the numerics deals with a slightly different model, but we can see that the prefactor found in simulations decreases with temperature, in qualitative agreement with the prediction.

1.3. Dynamical ultrametricity for $T = T_g$

The study of the behaviour of the model right at the glass transition temperature $T = T_{\rm g}$ appears to be easier to solve, although the scaling form is slightly more subtle. In order to simplify the notation, we choose in this section $T_{\rm g}$ as the energy (and temperature) unit. As

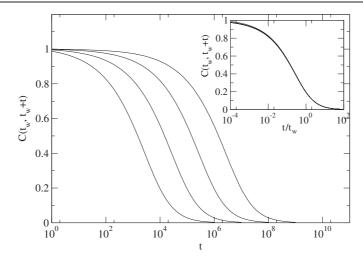


Figure 1. Plot of the ageing correlation function $C(t_w, t_w + t)$ of the BM model, for $t_w = 10^4, 10^5, 10^6$ and 10^7 (from left to right), and temperature $\mu = 0.7$. Inset: correlation plotted as a function of the rescaled time t/t_w , showing a very good collapse of the data, apart from small deviations for $t \ll t_w$ due to finite time effects.

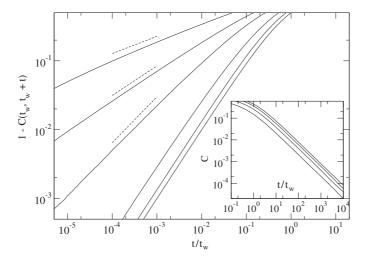


Figure 2. Short time behaviour of the correlation function; $1-C(t_w,t_w+t)$ is plotted as a function of t/t_w for several temperatures $\mu=0,0.2,0.4,0.6,0.7$ and 0.8 (from right to left), showing the onset of a short time singularity for $\frac{1}{2}<\mu<1$ with an exponent $(1-\mu)/\mu$ (dashed lines). For $\mu<\frac{1}{2},1-C$ is linear for $t\ll t_w$. Inset: long time tail of the correlation for $\mu=0.2$, 0.4, 0.6 and 0.8 (from right to left), showing a power law decay $C\sim (t/t_w)^{-1}$.

for the low temperature case ($\mu < 1$) of the BM model, one can look for a scaling variable u which links time and energy in order to transform asymptotically (for $t \to \infty$) the master equation into an ordinary differential equation. Starting again from the ratio $\tau(E)/t$, one can simplify its expression at large time and obtain the scaling variable $u = (e^E t \ln t)^{-1}$. The associated scaling function $\varphi(u)$ must be related to P(E,t) through

$$P(E,t) \simeq \frac{1}{\ln t} u \varphi(u).$$
 (26)

The normalization condition $\int_{-\infty}^{0} dE P(E, t) = 1$ translates into

$$\frac{1}{\ln t} \int_{(t \ln t)^{-1}}^{\infty} du \, \varphi(u) \to 1 \qquad t \to \infty$$
 (27)

which requires that $\varphi(u) \simeq u^{-1}$ for $u \to 0$. The master equation then becomes in the limit of large t:

$$u^{2}\varphi'(u) + (u-1)\varphi(u) = -\frac{1}{u}.$$
(28)

The condition that $\varphi(u)$ must be normalizable in the sense of equation (27) selects a unique solution, which reads

$$\varphi(u) = -\frac{1}{u}(1 - e^{-1/u}) \tag{29}$$

so that P(E, t) is given by

$$P(E,t) = \frac{1}{\ln t} [1 - \exp(-e^E t \ln t)]. \tag{30}$$

From this expression, one can compute the correlation $C(t_w, t_w + t)$ at the critical temperature:

$$C(t_w, t_w + t) = \int_{-\infty}^{0} dE \ P(E, t_w) e^{-t/\tau(E)}$$
(31)

with $\tau(E)$ given by

$$\frac{1}{\tau(E)} \equiv \int_{-\infty}^{0} \frac{e^{E'} dE'}{1 + e^{E'-E}} = e^{E} \ln(1 + e^{-E})$$
 (32)

which reduces to $|E| e^{E}$ for large |E| (E < 0). So the correlation reads, for large t_w and t,

$$C(t_w, t_w + t) = \frac{1}{\ln t_w} \int_{-\infty}^{0} dE \exp(Et e^E) [1 - \exp(-e^E t_w \ln t_w)].$$
 (33)

From this expression, one can deduce that the relevant scaling variable is no longer t/t_w , but rather $\omega = \ln t / \ln t_w$, in the sense that when taking the infinite t_w limit keeping ω fixed, $C(t_w, t_w + t)$ converges to a function $\tilde{C}(\omega)$ given by

$$\tilde{\mathcal{C}}(\omega) = 1 - \omega \quad (0 \leqslant \omega < 1) \qquad \tilde{\mathcal{C}}(\omega) = 0 \quad (\omega \geqslant 1).$$
 (34)

This is precisely the asymptotic form found in the trap model at the glass transition temperature, which was shown to satisfy dynamical ultrametricity [22, 23]. So we see that dynamical ultrametricity is indeed independent of the type of dynamics considered—entropic or thermal activation—but is rather governed by the exact balance of the Boltzmann weight and of the exponential density of states which is valid only right at the glass transition temperature. Interestingly, the differential equation—equation (28)—satisfied by the scaling function $\varphi(u)$ appears to be the same in the BM model as in the trap model—although another approach was used in [23]—leading to the same scaling functions $\varphi(.)$, even though they are derived from two different master equations. Note, however, that the scaling variables are slightly different, $u = (t \ln t e^E)^{-1}$ in the BM model, and $v = (t e^E)^{-1}$ in the trap model.

2. Finite size equilibrium

In realistic models of glasses, the structure of the global phase space is expected to be highly non-trivial, since it has to encode the finite dimensionality of the physical space. So it appears tempting, at least as a first approximation, to think of a macroscopic system as a collection

of small independent subsystems, the size of which would be of the order of the coherence length [26, 27]—see also [28] for an experimental investigation of this point. This scenario has indeed been supported by recent simulations [29] on Lennard–Jones supercooled liquids, showing that a system with 130 particles has, to a good approximation, the same dynamical behaviour as two non-interacting systems composed of 65 particles each¹.

In this context, finite size versions of the usual—trap and BM—phase space models appear to be particularly relevant, and the simplest case to study is the equilibrium one. In order to make contact with more realistic models, let us emphasize that 'states' in the present case should be associated with singular points in phase space, namely saddles or minima, so that one expects their number M to grow exponentially with the number N of physical degrees of freedom:

$$M \sim M_0 e^{\alpha N} \tag{35}$$

where α is a numerical constant. Several studies have investigated the value of α as a function of temperature when considering only minima and have found $\alpha \ll 1$ [6, 30, 31]. Note however that, bearing in mind the above picture, N should be itself a function of temperature since it depends on the coherence length $\xi(T)$ through $N \sim \xi(T)^d$.

Considering M energy states randomly chosen from an exponential distribution $\rho(E)$, the equilibrium distribution concentrates onto the lowest energy states, so that the correlation function decays on a time scale corresponding to the typical largest sojourn time τ_{max} : $C_{\text{eq}}(t,M) = \mathcal{C}(t/\tau_{\text{max}})$. The value of τ_{max} can be estimated using a scaling argument: in a finite size model, one can imagine that all the sojourn times are chosen at random according to the probability distribution $\psi(\tau)$ associated with the *infinite* size model. In the exponential trap model, $\psi_{\text{TM}}(\tau) = \mu/\tau^{1+\mu}$ and this picture is indeed exact, since the relation $\tau = \mathrm{e}^{E/T}$ holds locally, at the trap level: one then has $\tau_{\text{max}} \sim M^{1/\mu}$. In contrast, in the BM model, the relation given in equation (7) is valid only in the infinite size limit, and should be only an approximation in the finite M case. Still, one can hope that this approximation is able to give the correct scaling of τ_{max} with M: $\psi(\tau) \sim 1/\tau^2$ then yields $\tau_{\text{max}} \sim M$.

An interesting question naturally arises about the finite size equilibrium dynamics in the low temperature phase. Given that for $T \leqslant T_{\rm g}$, the continuous energy equilibrium distribution $P_{\rm eq}(E) \propto \rho(E)\,{\rm e}^{-E/T}$ becomes non-normalizable, a cut-off scale necessarily appears in order to regularize the distribution $P_{\rm eq}(E)$. One way to introduce such a cut-off is precisely to consider a finite number of states. Otherwise, the spontaneous cut-off is generated by the dynamics itself, since it is given by the time t_w elapsed after the quench. So one can wonder whether or not the dynamics is qualitatively the same whatever the nature of the cut-off; we shall see that the answer depends on the model considered.

In the exponential trap model in the low temperature phase $\mu < 1$, with a finite number M of traps, the correlation $C_{\rm eq}^{\rm TM}(t,M)$ is thus a function $C_{\rm TM}(t/M^{1/\mu})$ of the rescaled time $t/M^{1/\mu}$, which can be computed in the asymptotic regimes $t \ll M^{1/\mu}$ and $t \gg M^{1/\mu}$, leading to the interesting result that the behaviour is similar to that found in the ageing case, replacing the cut-off t_w by $M^{1/\mu}$. However, the prefactors are different from that found in the ageing case [32]. Namely, one finds

$$1 - C_{\text{eq}}^{\text{TM}}(t, M) \simeq \kappa_s \left(\frac{t}{M^{1/\mu}}\right)^{1-\mu} \qquad t \ll M^{1/\mu}$$
(36)

Note that this decomposition into a large number of 'elementary correlated cells' should not be confused with the usual procedure invoked to justify self-averageing properties in disordered systems, where each cell is still assumed to be very large.

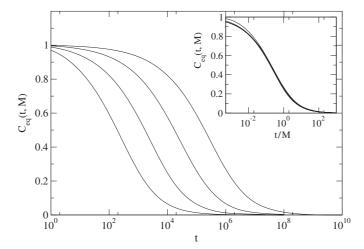


Figure 3. Plot of the finite size equilibrium correlation function $C_{\rm eq}(t,M)$ of the BM model, for $M=10^3$, 10^4 , 10^5 and 10^6 , and temperature $\mu=0.7$. Inset: correlation plotted as a function of the rescaled time t/M, showing a good collapse of the data, except for short times where the $M=10^3$ curve does not rescale completely.

$$C_{\rm eq}^{\rm TM}(t,M) \simeq \kappa_l \left(\frac{t}{M^{1/\mu}}\right)^{-\mu} \qquad t \gg M^{1/\mu}$$
 (37)

where the prefactors κ_s and κ_l are given by

$$\kappa_s = \frac{1}{1-\mu} \Gamma(\mu) \Gamma(1/\mu) \Gamma(1-\mu)^{-1/\mu}$$
(38)

$$\kappa_l = \sqrt{2}^{1-2\mu} \mu \sqrt{\pi} \Gamma\left(\frac{1}{2} + \mu\right). \tag{39}$$

This shows that in the trap model, the asymptotic behaviour of the rescaled correlation function for $\mu < 1$ is the same for t, much smaller or much greater that the cut-off time scale, whatever the origin of this scale, namely static (number of sites) or dynamic (waiting time). It would be interesting to see if this property also holds for the BM model.

To this aim, we first check numerically the proposed scaling in t/M for the BM model, using in this case the natural version of the model with Glauber rates, since for finite size systems, the continuous energy formalism is no longer valid. The numerical algorithm we use is then more complex, and does not simplify for a particular choice of the transition rates. Figure 3 displays $C_{\rm eq}(t,M)$ for several values of M, namely $M=10^3$, 10^4 , 10^5 and 10^6 ; the inset shows that the data collapse well when plotted as a function of t/M, with again small deviations in the short time regime which requires larger system sizes to reach the asymptotic scaling form.

Turning to the asymptotic behaviour of the correlation, one sees in figure 4 that the late time behaviour of $C_{\rm eq}(t,M)$ is no longer temperature independent, as in the ageing case, but depends strongly on temperature, and the slope is precisely equal, to numerical precision, to $-\mu$. This is strong evidence for a thermally driven dynamics at large times, which corresponds to the fact that the particle can only escape from the deepest traps through thermal activation. In contrast, the short time singularity is not modified with respect to the ageing case, as can be seen in the inset of figure 4. This was expected as the short time dynamics is dominated

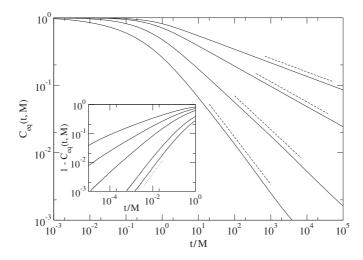


Figure 4. Asymptotic large time behaviour $(t \gg M)$ of the correlation $C_{\rm eq}(t,M)$ with $M=10^3$ for temperatures $\mu=0.2,0.3,0.5$ and 0.7 (from top to bottom), showing a temperature dependent power law tail $C_{\rm eq}(t,M) \sim (t/M)^{-\mu}$ (dashed lines), typical of activated dynamics. Inset: short time behaviour $(t \ll M)$ of the correlation, with $M=10^6$, $\mu=0.3,0.4,0.6,0.7$ and 0.8, showing the onset of the same short time singularity as in the ageing regime (see figure 2; dotted line: slope -1).

by 'intermediate' energy states—i.e. the highest states among the low energy ones onto which the equilibrium measure concentrates—from which one can escape downwards.

As a result, a cross-over appears in the equilibrium *finite size* correlation function between an entropic behaviour at short times and an activated one at long times. A natural question at this stage would be to see if this cross-over is a peculiarity of the equilibrium case, or if it can be found also in dynamical regimes.

3. Cross-over in the ageing regime

Thermally, activated phenomena are known to play an important role in glassy physics, and numerous experimental studies have focused on the effect of temperature on glasses (see e.g. [33] for structural glasses and [34] for spin glasses). Theoretical works also support this view, since the ideal glass transition predicted by MCT has been shown to be smeared out in real glasses by activated processes [1, 2]. However, in structural glasses, these activated processes are usually assumed to be relevant only on time scales close to the equilibration time $\tau(T)$ —which becomes very large below the glass transition temperature—and not in the ageing regime which corresponds to shorter time scales. Indeed, recent simulations on Lennard–Jones liquids seem to validate a mean-field-like scenario with a well-defined effective temperature [35], at odds with the behaviour of thermally driven models such as the trap model [36]—except in some very special cases [20].

One might thus think that the cross-over from entropic to thermal dynamics observed in numerical simulations [17, 18] is only due to the small size of the systems considered, and that the associated time scale should be very large for macroscopic systems, as shown in the mathematical analysis of the dynamics of the random energy model [37, 38]. Still, the arguments presented above suggest that for a finite dimensional system, this time scale should be governed by the coherence length rather than the global size of the system.

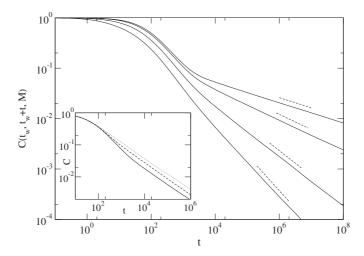


Figure 5. Correlation $C(t_w, t_w + t, M)$ in the out of equilibrium regime of the finite size model, for $M = 10^3$, $t_w = 10^2$ and $\mu = 0.2, 0.3, 0.5$ and 0.7. One sees a cross-over from a rather temperature independent (entropic) regime to a temperature dependent (activated) one with exponent $-\mu$ (dashed lines). Inset: effect of a finite connectivity Z, for $M = 10^3$, $t_w = 10^2$ and $\mu = 0.5$; dotted line: Z = 100, dashed line: Z = 300 and full line: fully connected case. Reducing the connectivity decreases the cross-over time scale.

One can also wonder whether the cross-over time scale could be possibly much smaller than the equilibration time under some conditions.

In this section, we study the onset of a dynamical cross-over from entropic to activated behaviour in the ageing regime of the correlation function. To this aim, we consider the finite size BM model as well as a generalization including a threshold energy below which all states are isolated minima, and show that the dynamical cross-over time is equal to the equilibration time only for the fully connected model, and becomes much smaller as soon as a non-trivial phase space structure is taken into account.

3.1. Ageing in a finite size system

A natural generalization of the analysis performed in the previous section is to study the ageing regime $1 \ll t_w \ll M$ of the finite size BM model, with the aim of showing the existence of a cross-over in the correlation $C(t_w, t_w + t, M)$ from the usual BM ageing form for $t \ll M$ to an activated behaviour for $t \gg M$. So the correlation function is now characterized by a couple of time scales, t_w and M, instead of only one of these.

Numerical results for the correlation function $C(t_w, t_w + t, M)$ are shown in figure 5 for $M = 10^3$, $t_w = 10^2$ and various temperatures ($\mu = 0.2, 0.3, 0.5$ and 0.7). Apart from a global shift, the 'early parts' of the curves (i.e. times t such that $t_w \leq t \leq M$) are almost temperature independent, which is characteristic of an entropic kind of dynamics, as we have seen in the previous sections. In contrast, the late ($t \geq M$) power law decay, with an exponent $-\mu$ (dashed lines) is the fingerprint of thermally activated dynamics.

As we have only considered up to now fully connected models, one can wonder whether or not a finite connectivity $Z \ll M$ would strongly modify the previous results. Although a detailed study of the BM model with finite connectivity is beyond the scope of the present work, we comment here on the results of preliminary simulations on random graphs with finite average connectivity. In particular, decreasing the connectivity does not affect much

the overall decay time of the correlation function, in equilibrium as well as in the ageing regime. Still, out-of-equilibrium numerical data presented in the inset of figure 5 for $M=10^3$ and different values of Z indicate that reducing the connectivity tends to decrease the cross-over time scale beyond which thermal activation becomes dominant. This could have been expected, since a finite connectivity generates a lot of minima, from which one can escape only through thermal activation.

From the above results, one may suggest the following scenario for the behaviour of realistic glass formers. One the one hand, one expects as discussed above that a large system can be considered as a collection of (roughly) independent small subsystems [29], the size of which should be of the order of the coherence length, so that finite size phenomenological models should indeed be relevant to describe real glasses. On the other hand, we have just seen that even with an entropic kind of dynamics, a regime dominated by thermal activation appears beyond a cross-over time scale (which diverges with the size of the system). However, a finite connectivity of the network of saddles and minima can strongly reduce this cross-over time scale, so that it can be much shorter than the equilibration time scale and might be accessible in numerical simulations or experimental results. In the (long time) activated regime, one thus expects generically the dynamics of a realistic glass former to be similar to that of the trap model. One may wonder at this stage whether only a small number of minima should be considered to describe these activated phenomena, instead of a large number as is usually done in the trap model. In the low temperature phase of the fully connected (finite size) BM model, activated phenomena are indeed due to a small number of deepest states. In contrast, when taking into account a finite connectivity, numerous local minima appear, and all these minima give a contribution to the dynamics in the ageing regime. The latter case is probably similar to what happens in real glass formers.

Although a quantitative study of the finite size BM model in the out-of-equilibrium regime would be of great interest, it would require a larger scale separation than the one we obtained, which is hard to reach for computational reasons. We thus propose another version of the BM model, in which one can safely take the limit $M \to \infty$ without eliminating the dynamical cross-over we are presently interested in. This can be done by taking into account a non-trivial structure of phase space.

3.2. Effect of a threshold energy

Coming back to the paradigm of the energy landscape with a sharply defined separation in energy between saddles and minima [11–15], one could try to mimic this phase space structure by introducing by hand a threshold energy E_{th} acting as a kinetic constraint, such that direct transitions between states with energies E and E' are forbidden if E and E' are both below $E_{\rm th}$, and otherwise unchanged with respect to the usual BM model. In other words, states with energy $E > E_{th}$ can be considered as saddles, from which all other states are accessible, whereas states with energy $E < E_{th}$ represent isolated minima from which the system can escape only by jumping to a saddle, before eventually reaching another (possibly deeper) minimum. Such a definition could be applied to a finite size model, and the equilibration time in this case would be a function of M. In Lennard–Jones liquids, this threshold E_{th} has been found to be proportional to the number N of degrees of freedom but, as argued above, N should be actually bounded by $\xi(T)^d$, so that E_{th} is expected to remain finite even for macroscopic systems. Conversely, M should also remain finite, but assuming that the smallest energy E_{\min} found in the system lies much below E_{th} , one can still take the limit $M \to \infty$ keeping E_{th} fixed, so as to describe the dynamics on time scales much smaller than the finite size equilibration time.

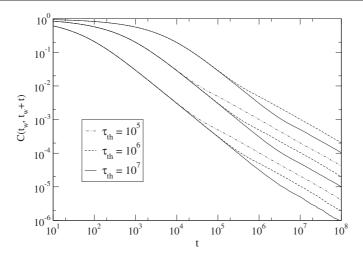


Figure 6. Plot of the correlation function $C(t_w, t_w + t)$ of the ThBM, in the ageing case ($\mu = 0.7$) and in the regime $t \gg t_w$ (long time) and $t_w \ll \tau_{\text{th}}$. Both τ_{th} and t_w are varied: $\tau_{\text{th}} = 10^5$ ($t_w = 10^2, 10^3, 10^4$); t_w increases from left to right. A cross-over appears between an entropic regime, with exponent -1, and an activated one, with exponent $-\mu$.

Figure 6 displays the results of numerical simulations of the correlation function in this generalized model, denoted hereafter as ThBM ('threshold Barrat–Mézard') model, for several values of t_w and $E_{\rm th}$, focusing on the long time behaviour ($t\gg t_w$). The natural time versus energy relation in the BM model allows us to define a time scale $\tau_{\rm th}$ associated with $E_{\rm th}$ through $\tau_{\rm th}=\exp(|E_{\rm th}|/T_{\rm g})$ —we neglect here the prefactor appearing in equation (7)—and curves are thus labelled by $\tau_{\rm th}$ in figure 6. The simulations show the onset of a cross-over between two different power law behaviours, the first one with an exponent -1 corresponding to the usual long time behaviour of the BM model (entropic dynamics), and the other one with an exponent $-\mu$ typical of the long time tail of the correlation in the trap model (thermally activated dynamics).

Let us now present a scaling argument in order to gain a better understanding of this cross-over. We first focus on the most interesting case for which the typical energies reached at t_w , of the order of $-T_g \ln t_w$, remain far enough above $E_{\rm th}$.

We assume, and we have checked numerically, that the tail $(E \text{ much below } -T_g \ln t_w)$ of the dynamical energy distribution $P(E,t_w)$ keeps the usual form $P(E,t_w) \propto \mathrm{e}^{E/T_g}$ found both in the BM and in the trap model, for $E > E_{\mathrm{th}}$ as well as for $E < E_{\mathrm{th}}$. One thus expects the following asymptotic behaviour for the dynamical distribution $p(\tau,t_w)$ of sojourn times:

$$p(\tau, t_w) \sim \frac{1}{t_w} \left(\frac{t_w}{\tau}\right)^2 \qquad \qquad \tau \ll \tau_{\text{th}}$$
 (40)

$$p(\tau, t_w) \sim \frac{A}{t_w} \left(\frac{t_w}{\tau}\right)^{1+\mu} \qquad \tau \gg \tau_{\text{th}}$$
 (41)

these expressions correspond to the large τ tail of the dynamical distribution in the usual BM model and in the trap model respectively. The rescaling factor A is determined by matching the two asymptotic expressions for $\tau = \tau_{\text{th}}$, which yields $A = (t_w/\tau_{\text{th}})^{1-\mu}$. Computing the

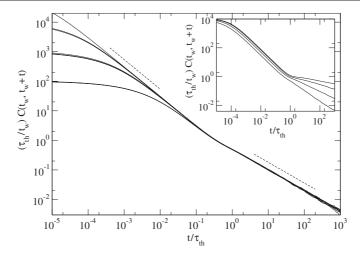


Figure 7. Same curves as in figure 6, rescaled in their late part in order to show the cross-over scaling, by plotting $(\tau_{th}/t_w)C(t_w,t_w+t)$ versus t/τ_{th} (see text). Dashed lines indicate slopes -1 and $-\mu$. Inset: rescaled correlation function for $\mu=0.2,0.3,0.5$ and 0.7 (from top to bottom), $t_w=10^3$ and $\tau_{th}=10^7$, showing the strong temperature dependence for $t\gg\tau_{th}$, whereas the slope for $t_w\ll t\ll\tau_{th}$ is independent of μ .

correlation for $t \ll \tau_{\text{th}}$, one recovers as previously $C(t_w, t_w + t) \sim t_w/t$, whereas for $t \gg \tau_{\text{th}}$, one now has

$$C(t_w, t_w + t) \sim \frac{t_w}{\tau_{th}} \left(\frac{\tau_{th}}{t}\right)^{\mu}. \tag{42}$$

Summarizing the above results, one can introduce a scaling function $f(\cdot)$ such that $C(t_w, t_w + t)$ reads

$$C(t_w, t_w + t) = \frac{t_w}{\tau_{th}} f\left(\frac{t}{\tau_{th}}\right) \tag{43}$$

with the following asymptotic behaviour for f(z):

$$f(z) \sim \frac{1}{z} \quad (z \ll 1) \qquad f(z) \sim \frac{1}{z^{\mu}} \quad (z \gg 1).$$
 (44)

In order to test this prediction, we have plotted in figure 7 the previous curves using the rescaled variables $(\tau_{th}/t_w)C(t_w,t_w+t)$ and t/τ_{th} . The resulting collapse of the late part of the curves (i.e. for $t\gg t_w$) is very good, confirming the validity of the above scaling analysis. Note that the whole curves cannot be rescaled in a simple way since the early part of the correlation still scales in t/t_w as usual (rescaling not shown), so that two different scaling regimes $t\sim t_w$ and $t\sim \tau_{th}$ have to be considered separately. Figure 7 shows that in the limit of a large separation of scales between t_w and τ_{th} , the plot would reduce to two straight lines with different slopes -1 and $-\mu$. The temperature dependence is clearly shown in the inset of figure 7, showing as expected that the slope is independent of μ for $t_w \ll t \ll \tau_{th}$ (and equal to -1), whereas it is strongly temperature dependent (and equal to $-\mu$) for $t\gg \tau_{th}$.

In the opposite regime $t_w \gg \tau_{\text{th}}$, the system is trapped into deep minima so that the behaviour becomes similar to that of the trap model, up to an energy shift. However, the particle necessarily visits states with energy $E > E_{\text{th}}$ in order to escape from minima, and then relaxes towards another minima. So a cross-over between entropic and activated dynamics is also expected in this case for $t \sim \tau_{\text{th}}$, which now falls in the short time regime $(1 \ll t \ll t_w)$.

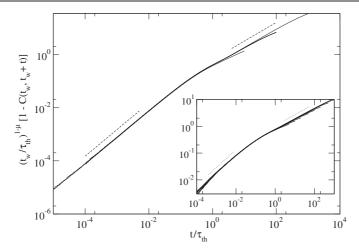


Figure 8. Short time behaviour of $C(t_w, t_w + t)$ for $\mu = 0.3$ and $t_w \gg \tau_{\text{th}}$, plotted as $(t_w/\tau_{\text{th}})^{1-\mu}[1 - C(t_w, t_w + t)]$ versus t/τ_{th} (see equation (45)), showing a cross-over from a linear (entropic) behaviour for $t \ll \tau_{\text{th}}$ to an activated one, with exponent $1 - \mu$, for $\tau_{\text{th}} \ll t \ll t_w$; $\tau_{\text{th}} = 10^3$ ($t_w = 10^5, 10^6$) and 10^4 ($t_w = 10^6, 10^7$). Inset: same plot with $\mu = 0.6$, such that the entropic regime $1 \ll t \ll \tau_{\text{th}}$ corresponds to a power law with exponent $(1 - \mu)/\mu$; $\tau_{\text{th}} = 5 \times 10^3, 10^4, 2 \times 10^4$ and $t_w = 10^5, 10^6, 10^7$.

In order to analyse this cross-over, one can derive a scaling relation along the same lines as for equation (43), which now reads:

$$1 - C(t_w, t_w + t) = \left(\frac{\tau_{\text{th}}}{t_w}\right)^{1-\mu} g\left(\frac{t}{\tau_{\text{th}}}\right)$$
(45)

where the scaling function g(z) has the following asymptotic behaviour:

$$g(z) \sim z^{\gamma} \quad (z \ll 1) \qquad g(z) \sim z^{1-\mu} \quad (z \gg 1)$$
 (46)

with $\gamma = \min[1, (1 - \mu)/\mu]$, as for the usual Barrat–Mézard model. Figure 8 displays the numerical results for different t_w and τ_{th} , with $\mu = 0.3$ and 0.6, and shows a good collapse of the data, which clearly supports the existence of a cross-over for $t \sim \tau_{th}$.

In summary, a non-fully connected phase space structure generates a large number of local minima, so that the dynamical cross-over time scale associated with the end of the entropic regime is strongly reduced with respect to the fully connected case, suggesting that this cross-over may be relevant in realistic glasses on experimental time scales, even below the glass transition temperature.

Note finally that one may wonder whether a finite number of states or the introduction of an energy threshold might help solve the question raised recently [20, 21, 36] concerning the fact that trap models do not exhibit straight ('one-step') fluctuation—dissipation plots in their ageing regime. Actually, as we have just seen, a threshold energy (introduced by hand, or by considering finite size systems) essentially leads to a cross-over between a behaviour characteristic of the usual BM model and another one typical of the trap model. It is now well established that both models (BM and trap) present a non-linear fluctuation—dissipation relation [20, 21, 36]—to be more specific, this has been shown only for $T \ll T_{\rm g}$ in the BM model [21]. As a result, one expects to observe non-linear fluctuation—dissipation relations in the ageing regime of the finite size BM model and of the ThBM introduced above.

4. Conclusion

In this paper, we have shown that a cross-over from entropic to activated behaviour arises naturally in glassy models once a finite coherence length is taken into account, allowing us to conceptually decompose a large system into a collection of independent small subsystems [26, 27]. In this framework, one is then naturally led to consider *finite size* phase space models to describe the local dynamics within an 'elementary correlated cell'.

Interestingly, calculations in the trap model show that the finite size M replaces in a rough sense the time cut-off t_w appearing in the ageing regime, without affecting the asymptotic power law short and late time behaviours of the correlation. In contrast, in the BM model, the finite size equilibrium behaviour differs significantly from the ageing one: the short time singularity remains the same, but the long time one is no longer governed by an entropic mechanism, but corresponds to a thermally activated dynamics since escape from the deepest states is only possible through jumps to higher states, if the number of states is finite. Generalizing this analysis to the ageing regime of finite size systems, where both t_w and M come into play $(t_w \ll M)$, the previous cross-over still exists for $t \sim M$ as in equilibrium, and can even be found earlier if a finite connectivity is taken into account. Although such a cross-over has already been observed in numerical simulations [17, 18], a quantitative analysis of the associated time scales would be very valuable, in order to see how it varies with temperature and with the total size of the system. In particular, if a decomposition into small correlated cells holds, one expects the cross-over time scale to saturate to a finite value for large system sizes.

Following the phase space decomposition into saddles and minima, we have then studied a generalization of the BM model in which the cross-over time scale is not induced by the finite size of the system, but instead by a threshold energy $E_{\rm th}$ which separates saddles for $E > E_{\rm th}$ from minima for $E < E_{\rm th}$. Assuming that the equilibration time is large enough, we find that the correlation function keeps ageing even beyond the cross-over time scale $\tau_{\rm th}$, but with a behaviour characteristic of activation dynamics. Interestingly, the transition from one type of ageing dynamics to the other leads to an unusual scaling, due to the fact that two time scales t_w and $\tau_{\rm th}$ are now involved instead of one. Moreover, the presence of numerous local minima in phase space leads to a cross-over time scale much smaller than the equilibration time, contrary to what happens in the fully connected case.

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