Recent theories of glasses as out of equilibrium systems *

Jorge Kurchan

P.M.M.H. Ecole Supérieure de Physique et Chimie Industrielles,
10, rue Vauquelin, 75231 Paris CEDEX 05, France

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Abstract

We discuss a theoretical approach to structural glasses as they happen in real situations. Older ideas based on ‘configurational entropy’, on ‘fictive temperatures’ and on Edwards’ ‘compactivity’ are sharpened and unified in an out of equilibrium context. The picture is such that it may be supported or disproved by an experimental test, which we describe.

In this note I briefly review a rather general picture of structural glasses that has emerged in the last dozen years or so, and that to my knowledge has not been summarised elsewhere from quite this point of view. To the extent that this scenario applies, one can make a theory of glasses as they happen, any reference to ideal infinitely slow annealings being superfluous.

Two sets of older ideas — one based on configurational entropy put forward by Kauzmann, Adam, Gibbs and Di Marzio\(^1\) and the other on aging, ‘fictive temperatures’\(^2\), and Edwards’ ‘compactivity’\(^3\) — are unified and developed in two ways:

- They either admit a quantitative definition, or, in certain aspects, their inherent ambiguity is uncovered.
- Models and approximations are identified within which one can calculate everything analytically, and the scenario holds strictly. This opens the way for systematic improvements of these schemes, a line that is currently pursued. Solvable cases have indicated that certain features of the older ideas had to be reconsidered, of which we shall see some instances below.

1. Aging

Consider a supercooled liquid. In order to probe its dynamics we shall use an autocorrelation function, say, \(C(t_1 - t_2) = \langle \rho_k(t_1)\rho_k(t_2) \rangle\). In Fig. 1 we show how this looks

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like when plotted in terms of \( \log(t) = \log(t_1 - t_2) \) for two different temperatures \( T_a > T_b \).

There is a fast relaxation which corresponds to rapid motion of the particles ‘inside a cage’
formed by its neighbours, much as atoms move around their lattice position in a crystal.
On much longer timescales, the actual ‘cages’ will rearrange: these are the ‘structural’ or
‘alpha’ relaxations. While the cage motion is weakly dependent on the temperature, the
structural motion slows dramatically as we cool the system.

In Fig. 1 we also show two arbitrary ways of defining an alpha-relaxation time \( t_\alpha(T) \). If,
as we decrease \( T \), we find that \( t_\alpha'(T) \) and \( t_\alpha(T) \) stay of the same order, then we have only
one slow timescale, i.e. we have in all a two-step process. For the moment it seems that
this is the situation for structural glasses. Another possibility, encountered in mean-field
spin glasses, is that on tuning the appropriate parameter any two timescales go to infinity at
different paces, the ratio \( t_\alpha/t_\alpha' \) becoming larger and larger: this is a multi-timescale situation.

This serves to underline the fact that when one talks about several timescales, the concept
becomes clear-cut only in an appropriate limit in which they separate completely. This will
turn out to be relevant in what follows.

An alternative way to describe the relaxation is by considering the integrated response
\( \chi(t_1 - t_2) \) conjugated to \( C \): in this case \( \chi(t_1 - t_2) \) is the compressibility of the mode \( k \)
with respect to a field acting continuously from \( t_2 \) to \( t_1 \). In equilibrium we have that
\( T \chi(t_1 - t_2) = C(t_1 - t_1) - C(t_1 - t_2) \), by virtue of the fluctuation-dissipation theorem
(FDT), see Fig. 2.

In cases in which we have a two-step relaxation, we can describe the slow dynamics of
the system by a plot as in Fig. 3. Such plots are in general done with the viscosity instead
of \( t_\alpha \) in the \( y \)-axis.

At this point the tradition is to embark in a detailed discussion of the form of the curve
Fig. 3. Is there a true divergence at some \( T_0 > 0 \) or not? If so, what is the nature of
the equilibrium thermodynamic state below \( T_0 \)? Here I take the point of view that these
questions, although of obvious interest, are not the most urgent as far as understanding
glasses in real life, and one can make progress without addressing them. Let us here content
ourselves with mentioning that there are some glasses (like plastics) that may have a \( T_0 > 0 \),
while others are expected to have a finite \( t_\alpha \) at all temperatures above zero. Note that
glasses from this last group, of which ‘window’ glasses are likely to be an example, are
described from the equilibrium thermodynamic point of view as being at all temperatures just supercooled liquids.

Consider now a system having at $T_a$ a timescale $t_\alpha$ of ten minutes, and at $T_b$ of an hour. What will happen if, from an equilibrium situation at $T_a$ we lower the bath’s temperature to $T_b$ in one minute? In the instant $t_w = 0$ just after the quench, while the rattling of particles inside their cages will quickly adapt to $T_b$, the cages themselves will not have the time to adapt to their new situation, as this needs structural rearrangements taking times $t_w$ much larger than an hour.

If we measure the correlations starting from different $t_w$, it is quite clear that we will observe a relaxation as in Fig. 4. The $\alpha$-relaxation time grows with $t_w$ and eventually saturates at the new value — and this can also be said about the viscosity. In other words, the two-time correlation function becomes waiting-time dependent: $C = C(t + t_w, t_w)$. In particular, if there is a true divergence in the equilibrium value of $t_\alpha(T)$ at some $T_o > T_b$, the $t_w$-dependence will stay forever. This is the aging phenomenon: quantities depend on the waiting time for a long (eventually infinite) time $t_w \gg t_\alpha(T_b)$. The situation described above will always be met in a real-life cooling procedure, as sooner or later the cooling time will be too short compared with the equilibrium $t_\alpha(T)$. Our problem is to understand the $t_w << t_\alpha(T)$ regime that ensues, whose phenomenology, we argue, does not depend crucially
on whether \( t_\alpha(T) \) is actually infinite or not.

If instead of a correlation function we look at the associated response, then we also see aging. Figure 5 is a sketch of how this would look. If we are considering a uniform compression \((k = 0)\), the vertical arrow shows the beginning of the ‘creep’ deformation, as studied in the seminal experiments by Struik.\(^4\)

We have mentioned that when the system is quenched it rapidly adapts its fast, rattling motion to the new temperature. Its structural motion instead remembers the situation in which it lost pace and fell out of equilibrium: thus the idea of using the temperature at which this happened as an extra history-encoding state variable—the ‘fictive temperature’—finds a place and a clear definition within the framework of this and the following section.

Consider a plot of the \( y \)-axis of Fig. 1 versus the \( y \) axis of Fig. 2, where time acts as a parameter. The fluctuation-dissipation theorem tells us that for temperatures \( T_a \) and \( T_b \) we obtain straight lines with gradients \(-1/T_a\) and \(-1/T_b\), respectively. What happens if we attempt to put together in the same way the aging curves Figs. 4 and 5, using both \( t \) and \( t_w \) as parameters? A sketch of this is shown in Fig. 6. Soon after the quench, all curves superpose in the range \((C > q, \chi < \chi_o)\) on a straight line with gradient \(-1/T_b\): just as we would have expected to be the case in a system thermalised to the new temperature \( T_b \).
This is not unreasonable, as it corresponds to the times ‘inside a cage’ (cfr. Figs. 4 and 5).
The surprising result appears in the slow relaxation range \((C < q, \chi > \chi_q)\) : all the curves superpose on another straight line with gradient (say) \(-1/T_{\text{eff}}\) (curve (1) of Fig. 6). Then, on a much slower timescale (many \(\alpha\)-relaxations), the straight line slowly drifts (curve (2) in Fig.6) eventually working its way to the dashed line of gradient \(-1/T_b\), at which time the system has finally re-thermalised and aging has finished.

The parametric response-versus correlation plots like Fig.6 reveal at a glance the nature of the aging dynamics. They were introduced in the context of analytically solved models of aging\(^6\) (see Section 3), and were subsequently numerically implemented for realistic models\(^7\), for which an analytical solution is not available, with strikingly similar results\(^8\).

Let us argue that \(T_{\text{eff}}\) obtained from Fig. 6 is a true temperature for the structural motion, in the sense that it has the good thermometry and heat exchange properties. It can be shown that\(^9\):

- Any thermometer coupled to an observable in the system and tuned to respond to the timescale of structural relaxation measures exactly the \(T_{\text{eff}}\) associated with its fluctuations and response.

- A frequency filter tuned to the timescale of structural relaxation coupled to an observable in the system and to an auxiliary thermal bath of temperature \(T^*\) will transfer energy from the highest to the lowest of \((T_{\text{eff}}, T^*)\).

These two results are model-independent. In addition, within the framework of section 3 it has been shown that:

- Every pair of observables in the system gives the same fluctuation-dissipation temperatures \(T_{\text{eff}}\) at a given timescale.

These three facts characterise the FDT-related effective temperature as the only bona fide temperatures associated to the slow motion. Moreover, there is a lesson we can learn from exactly solvable models: \(T_{\text{eff}}\) turns out to be of the order of, but not quite equal to the temperature at which the system falls out of equilibrium. We have here an instance of a fact
mentioned at the beginning of the article: the old fictive temperature idea is promoted to a directly testable concept with thermodynamic (zero-th law) properties, but is also somewhat modified.

2. Ensembles and complexity

Let us discuss now what are the suggestions of Fig. 6 from the point of view of phase-space distributions. We can picture a two-timescale dynamics as a quasi-equilibrium motion for the fast evolution ‘inside the instantaneous cages’ (corresponding to the interval of correlations $C > q$) combined with a slow structural motion of the configuration of the cages. The moment the system falls out of equilibrium, the latter is not able to keep track ergodically. It is thus perfectly general that we should obtain a good equilibrium form in Fig. 6 for the motion in the interval $C > q$, and not for $C < q$.

What is not general, and begs for an explanation, is that the structural, ‘cage rearrangement’ motion behaves, from the point of view of FDT, as thermalised to another temperature, itself possibly decreasing adiabatically. This immediately raises the question of whether there is a new, hidden form of ‘ergodicity’ for the structural motion, allowing us to construct a statistical mechanics ensemble for the slow motion at each energy level.

One such construction is better introduced in the context in which it was originally formulated by Edwards. Consider a granular medium that is compactifying under gentle ‘tapping’. After each tap, the particles settle into a blocked configuration.

Now, Edwards proposed the extremely strong hypothesis that the blocked configurations reached dynamically at each density are the typical ones: at any given time, all macroscopic observables can be obtained by averaging their value over all blocked configurations of the appropriate density. This is an implicit definition of an ensemble for the dynamics. Once accepted, one immediately is led to defining an entropy (in the glass language a ‘configurational entropy’), as the logarithm of the number of blocked configurations of given volume— and energy, in the case of soft particles. Differentiating the configurational entropy with respect to the volume we obtain a ‘compactivity’, and with respect to energy an inverse configurational temperature.

This example of a granular medium is particularly simple because by looking at the system in repose some time after each tap, one concentrates on the slow rearrangements without having to deal with a superposition of simultaneous fast and slow motion.

Within the mean-field/mode-coupling scenario to be discussed in Sect. 3, it is possible to calculate averages over all the energy minima (the relevant blocked configurations for a zero temperature dynamics), as well as the their number at each energy level. The remarkable result is that the configurational temperature thus calculated à la Edwards and the FDT temperature $T_{\text{eff}}$ obtained after a deep quench are found to coincide exactly. Furthermore, the out of equilibrium macroscopic observables are correctly given by flat averages over the blocked regions.

These models teach us something else: for quenches to non-zero temperature, it is not the energy minima that have to be computed in order to reproduce the dynamic result, but rather the number of metastable states. This immediately poses a problem: while in the
mean-field models a metastable state is unambiguously defined as a region in phase space from which the system never escapes, for realistic, finite-dimensional models at $T > 0$ all metastable states have a finite lifetime. In other words, there is no absolute separation between cage and structural motion and there is an essential ambiguity in the concept of metastable state. This ambiguity carries over to the concept of configurational entropy, and hence to the configurational temperature, as the number of states we count depends on the minimal lifetime we expect from a state to call it a ‘state’.

This is a rather rapidly developing and not fully settled field, several alternatives have been suggested but many questions remain open. One strategy that is easy to implement numerically is to identify as a state the Gibbs-Boltzmann distribution restricted to the set of configurations in the basin of attraction of each energy minimum — the ‘inherent structure’ construction. Though this identification is not without problems of principle, the results in connection with the aging dynamics are encouraging. Recently, a more direct comparison between compaction results and Edwards ensembles in a schematic three dimensional model has yielded very good agreement.

Another strategy is to use a direct definition of ‘states with a given lifetime’. Using a construction of Gaveau and Schulman, one can compute averages over states having lifetimes of any preassigned value. Yet another possibility is to directly construct a phenomenological two-temperature thermodynamics, without invoking a statistical mechanical ensemble.

Let us conclude by remarking that the role played by configurational entropy here is stronger than in the Kauzmann picture of an ideal glass transition, since it involves the out of equilibrium dynamics, and not only an infinitely slow cooling. This has an advantage with respect to robustness, since the present scenario would be relevant in situations in which a thermodynamic glass transition either does not exist or is unobservable at experimentally accessible times.

At any rate, let us insist again, these ideas of generalised ensembles can only be taken seriously to the extent that the existence of FDT temperatures, independent of the observable at each timescale, is verified experimentally.

3. Methods and Models

Within a set of approximations and models the scenario discussed above holds rigorously. While these approximations are far from perfect, they have suggested tests that have been performed on realistic systems, with a considerable degree of success.

The paradigmatic and best studied model of supercooled liquids with two-step relaxations is the mode-coupling theory (MCT). These are approximate equations for the two-point dynamical correlation functions, which can be obtained through addition of a partial (though infinite) set of terms in the perturbative expansion of the exact dynamics.

An apparently different point of view was developed by Kauzmann, and Adam, Gibbs and DiMarzio, who first pictured the total entropy as composed of ‘rapid’ and ‘configurational’ degrees of freedom, and the ‘ideal glass’ transition as the temperature at which the system runs out of configurational entropy.
In a remarkably insightful set of papers, Kirkpatrick, Thirumalai and Wolynes showed that both approaches can be seen as aspects of the same theory. To do this, they first noted that MCT is the exact solution of the two-time correlations of the high temperature phase of a family of fully connected models having spin glass-like Hamiltonians but with multi-spin interactions. (This is why we referred above to the ‘mean-field/mode-coupling’ models). They then showed that the partition function of these models ignores completely the dynamic transition at $T_{MCT}$, but has an equilibrium transition at a lower temperature $T_K$. They explained this fact by showing that between $T_{MCT}$ and $T_K$ the phase-space is composed of exponentially many, mutually inaccessible states that the Gibbs measure confuses into a single, large, deceivingly liquid-like state. The equilibrium transition at $T_K$ happens because at just that temperature the number of states contributing to the measure becomes of order one: precisely Kauzmann’s idea with the logarithm of the number of states playing the role of configurational entropy. The paradigm of this form of transition is Derrida’s Random Energy Model.

A natural question that arises immediately is, what does a system satisfying exactly mode coupling equations for $T > T_{MCT}$ do if quenched below the dynamic glass transition $T < T_{MCT}$? The answer is that it ages: the correlation functions depend on the waiting time forever. Furthermore, the FDT plot of Fig. 6 converges to two a strict two-temperature behaviour, with a $T_{eff}$ of the order of (but not equal to) the transition temperature.

As mentioned before, one can study the free energy minima of the corresponding model: flat averages over metastable states of the appropriate energy give the correct out of equilibrium values of macroscopic observables. Furthermore, the configurational temperature obtained as the logarithmic derivative of the number of states with respect to the free energy turns out to give exactly the dynamically defined $1/T_{eff}$. Hence, within these models Edwards’ ‘compactivity’ ideas are also strictly realised.

Now, it is a well known fact that the mode coupling transition becomes a crossover in real life. This also has an explanation when the nature of the models is considered. The transition at $T_{MCT}$ relies on the existence of fully stable excited states, but these will eventually decay in any finite-dimensional model through nucleation. On the other hand, while the equilibrium transition at $T_K$ may or may not in a realistic model become a crossover as well, there is no a priori reason for this to be the case.

There are several strategies starting from these considerations. Given the limitations of the models considered, it is first mandatory to check directly with a simulation of a realistic model (say, a Lennard-Jones liquid) whether a two-temperature aging scenario remains valid in the presence of activated processes that may eventually lead the system to thermalisation. The evidence seems to be positive. A much harder numerical check concerns the nature of the equilibrium phase at low temperatures, using such small samples as one can thermalise.

From the analytic point of view, there are various different approaches being pursued. Perhaps the most basic is to try to understand whether there is a generic reason that there should be only one FDT temperature for all observables per widely separated timescale — independently from the approximation scheme in which this scenario was first (perhaps accidentally) found.

On a more practical line, there are several schemes for improving the approximations for the dynamics by considering diagrammatic resumations, although dealing with activated processes is notoriously hard. One can also construct better approximations for the equilib-
rium calculations. This approach, advocated by Mezard and Parisi, may be motivated by the following consideration: Clearly, an equilibrium calculation is relevant at such late times that energy, volume, and all macroscopic observables can be considered to have relaxed to their thermodynamic equilibrium value. If a glassy system has a transition below which $t_\alpha$ diverges strictly, even if all quantities have reached essentially their equilibrium value their $T_{eff}$ might still never reach the bath's temperature: It turns out that in such cases we can calculate the limiting $t \to \infty$ form of Fig. 6 using equilibrium techniques, and the result is directly related to Parisi's order parameter. (Obviously, within this approach, 'strong' glasses having $T_o = 0$ are described as liquids). This rather surprising connection between equilibrium and the last stages of out of equilibrium relaxation involves certain general assumptions, and in the specific case of structural glasses it requires the exclusion from the equilibrium calculation of all configurations having a degree of crystallisation.

4. Experiments

As remarked in several places above, the main observable feature of the present scenario for glasses is the existence of well separated timescales and a single fluctuation-dissipation temperature per timescale, shared by all observables. For example, the relation between spontaneous diffusion and mobility in a given time interval should be the same for different kinds of particles within the glass, or between rotational and translational degrees of freedom of the same particle. This relation defines a temperature that only coincides with the bath’s temperature for the fastest relaxations.

The fluctuation-dissipation experiments we have so far are comparisons of the dielectric susceptibility and polarisation noise in low-temperature glycerol and in Laponite gel, respectively. They show clearly that the low frequency ratio between noise amplitude and susceptibility stays different from the bath temperature after very long times. The analysis of the data is somewhat complicated by the fact that, for experimental reasons, these results are given in the frequency domain, and it is difficult to unravel the fast and slow timescales.

Ideally, one should go on to measure the time correlation between several fluctuating quantities and compare these with the associated responses — and check that the proportionality factor is the same at equal timescales for all observables. If, and when, this is not the case, the scenario discussed in this paper does not apply.

The measurements are always complicated by the waiting-time dependence of correlations and responses. An approach that may be useful in practice is to study glasses that are gently driven, for example by shearing, and to use the surprising fact that in such conditions aging can be stopped without changing the response versus correlation curve.
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