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The zeroth law in structural glasses: an example

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Abstract

We investigate the validity of a zeroth thermodynamic law for non-equilibrium systems. In order to describe the thermodynamics of glassy systems an extra parameter has been introduced, the effective temperature, which generalizes the fluctuation-dissipation theorem to off-equilibrium systems and supposedly describes thermal fluctuations around the ageing state. We analyse two coupled systems of harmonic oscillators with Monte Carlo dynamics. We study, in detail, two types of dynamics: sequential dynamics, where the coupling between the subsystems comes only from the Hamiltonian; and parallel dynamics, where there is another source of coupling: the dynamics. We show how in the first case the effective temperatures of the two interacting subsystems are asymptotically different due to the smallness of the thermal conductivity in the ageing regime. This explains why, in structural glasses, different interacting degrees of freedom can stay at different effective temperatures, and never equilibrate.

1. Introduction

The dynamics of glassy systems has been the subject of intensive research [1, 2]. In recent decades there has been an increasing interest in trying to extend the ideas of thermodynamics to systems which are far out of equilibrium. The general problem is to quantify non-equilibrium fluctuations within a thermodynamic approach. A salient feature of systems which are in equilibrium is the fact that the equilibrium fluctuations and the linear response functions are related by the so-called fluctuation-dissipation theorem (FDT) [3], which does not hold for off-equilibrium systems. Several studies of spin-glass mean-field models have shown that a generalization of the FDT is possible through the definition of the ‘fluctuation-dissipation ratio’ (FDR) [4, 5]

$$X(t, s) = \frac{TG(t, s)}{\frac{\partial C(t, s)}{\partial s}} \quad (t \geq s) \quad (1)$$

which is equal to 1 in equilibrium. It turns out that the behaviour of the quantity $X(t, s)$ is non-trivial in the limit $t, s \rightarrow \infty$. If the lowest time s is sent to infinity the quantity $X(t, s)$

becomes a non-trivial function of the autocorrelation $C(t, s)$. This strong statement has been proved to hold in the framework of mean-field spin-glasses [4, 5]. Moreover, it has recently been recognized that the quantity X is generally related to the Parisi order parameter $P(q)$ which appears in equilibrium studies of spin-glasses, providing a natural link between the static and dynamical properties [6].

According to relation (1) the usual fluctuation-dissipation relation would be recovered if the temperature on the right-hand side of (1) were $T/X(t, s)$. This ratio is called the ‘effective temperature’ and it has been shown [7–9] that it has some of the properties of a macroscopic temperature. In fact, a proper thermometer coupled to the slow degrees of freedom can measure it. The question of the usefulness of this temperature for describing the non-equilibrium behaviour has been a subject of controversy in recent years [10]. While there is some evidence (not only theoretical but also experimental [1, 11]) that the violation of FDT gives a good temperature in the thermodynamic sense, it is unclear which properties of equilibrium temperatures are shared by the equilibrium ones.

In this paper we want to analyse how effective temperatures equalize when two systems out of equilibrium are put in contact. In other words, we want to study if a zeroth law for non-equilibrium systems exists. Let us imagine a vitrified piece of silica quenched to room temperature. Because the glass is off-equilibrium its effective temperature is higher than room temperature, but when we touch the glass it does not feel hotter than the room temperature. Thus we can think that some ‘fast’ degrees of freedom are equilibrated to the room temperature while other ‘slow’ degrees of freedom remain non-equilibrated. But why have different interacting degrees of freedom not reached thermal equilibrium for sufficiently long times? We believe that the answer lies in the fact that the conductivity in the ageing state can be extremely small, so that the two parts of the system never equilibrate [12]. However, we can achieve a deeper understanding through a detailed analysis of an illustrative example, which is simpler and provides more generic insight.

The model is a set of harmonic oscillators evolving by Monte Carlo dynamics, as introduced in [13]. The importance of this model lies in the fact that at zero temperature it shows typical features of glasses such as ageing in correlation and response functions. Our interest will be in considering two coupled sets of harmonic oscillators. Thus, we can see how the main observables are affected by the coupling, in particular how the effective temperature evolves for the two sets of interacting degrees of freedom (i.e. for the two different sets of harmonic oscillators). The interaction itself can appear either through the Hamiltonian or through the Monte Carlo dynamics itself. We will discover that the effective temperature for the two sets of oscillators depends on how the coupling is implemented, and we will understand why in vitreous systems different degrees of freedom may stay at different temperatures without equilibrating at very long times. In this sense the utility of the extension of the zeroth thermodynamic law to the non-equilibrium ageing state is questioned.

This paper is organized as follows. Section 2 describes the main aspects as well as the interest of the model. Section 3 describes the two classes of couplings we consider. Section 4 discusses the results and the physical consequences of our work. The last section presents the conclusions.

2. A solvable model of a glass

As a simple model of a glass we will consider a system of uncoupled harmonic oscillators evolving with Monte Carlo dynamics, as introduced in [13] and recently revisited in [14–16]. The Hamiltonian is:

$$H = \frac{1}{2}K \sum_{i=1}^N x_i^2. \quad (2)$$

The low-temperature Monte Carlo dynamics of this system shows typical non-equilibrium features of glassy systems such as ageing in the correlation and response functions. The interest of this model is that at low temperatures the low acceptance rate generates entropic barriers that make the dynamics of the system extremely slow. The simplicity of the model makes it exactly solvable, yielding many results about the non-equilibrium behaviour.

The Monte Carlo dynamics is implemented by small movements: $x_i \rightarrow x_i + r_i/\sqrt{N}$ where the r_i are random variables Gaussian distributed with zero mean and variance Δ^2 . The move is accepted according to the Metropolis algorithm with a probability $W(\Delta E)$ which satisfies detailed balance: $W(\Delta E) = W(-\Delta E) \exp(-\beta \Delta E)$, where ΔE is the change in the Hamiltonian. The main features of this system, at zero Temperature, that we are interested in are:

- (i) *Slow decay of the energy.* The evolution equation for the energy is Markovian. The asymptotic large-time expansion of this equation shows that the energy decays logarithmically as $E(t) \sim 1/\log(t)$. It can also be seen that the acceptance ratio decays faster, according to $A(t) \sim 1/(t \log(t))$.
- (ii) *Ageing in correlation and response.* The correlation function $C(t, s)$ is defined by:

$$C(t, s) = \frac{1}{N} \sum_{i=1}^N x_i(t)x_i(s). \quad (3)$$

The response function $G(t, s)$ is calculated by applying an external field to the system. The response function is the variation of the magnetization $M(t) = \frac{1}{N} \sum_{i=1}^N x_i(t)$ of the system when the field is applied:

$$G(t, s) = \left(\frac{\delta M(t)}{\delta h(s)} \right)_{h=0} \quad t > s. \quad (4)$$

The asymptotic scaling behaviour for these two quantities is given by,

$$C(t, s) = C(s, s) \frac{D(s)}{D(t)} \quad G(t, s) = G(s, s) \frac{D(s)}{D(t)} \Theta(t - s) \quad (5)$$

with $D(t) \sim t(\log^2(t))$ and $C(s, s) = 2E(s)/K$, $G(s, s) = f(s)/K$ where $f(t)$ is a function that contains the entire dynamics and decays like $1/t$ [13]¹.

- (iii) *The effective temperature.* As stated in the introduction, the effective temperature is defined in terms of the FDR equation (1):

$$T_{\text{eff}}(t, s) = \frac{T}{X(t, s)} = \frac{\partial C(t, s)}{\partial s}. \quad (6)$$

As expected, in equilibrium $X(t, s) = 1$ and $T_{\text{eff}} = T$. Moreover, $E(s) = T/2$ and the equipartition theorem is verified. In the non-equilibrium case, the effective temperature only depends on the lowest time s for all times s and t . This is a characteristic feature of this particular model that is believed to hold for structural glass models in the asymptotic limit $s \rightarrow \infty$.

At zero temperature when slow motion sets in, the system never reaches the ground state. In this ageing regime the effective temperature verifies, in the limit $s \rightarrow \infty$:

$$T_{\text{eff}}(s) = 2E(s) + \frac{2}{f(s)} \frac{\partial E(s)}{\partial s} \longrightarrow 2E(s) \quad (7)$$

Equation (7) shows how the equipartition theorem can be extended to the glassy regime.

¹ In the original paper [13] the logarithmic corrections were estimated to be $\log^{\frac{1}{2}}(t)$. The correct exponent for the logarithmic corrections was later evaluated by Nieuwenhuizen [15].

3. Two coupled systems of oscillators

Now we consider the case in which we couple two systems of harmonic oscillators. In this case it is possible to compute analytically how one system affects the other. The Hamiltonian is

$$H = \frac{K_1}{2} \sum_{i=1}^N x_i^2 + \frac{K_2}{2} \sum_{i=1}^N y_i^2 - \epsilon \sum_{i=1}^N x_i y_i \quad (8)$$

where we take $K_1 K_2 > \epsilon^2$, otherwise the system has no bounded ground state. We define the following extensive quantities

$$E_1 = \frac{K_1}{2} \sum_{i=1}^N x_i^2 \quad E_2 = \frac{K_2}{2} \sum_{i=1}^N y_i^2 \quad Q = \sum_{i=1}^N x_i y_i \quad (9)$$

where E_1 and E_2 are the energies of the bare systems while Q is the overlap between them. In this case we also consider Monte Carlo dynamics, using the Metropolis algorithm for the transition probability. Although the random changes in the degrees of freedom x_i, y_i are defined in the same way as explained in the previous section for the case of a single system, there are different ways to implement the dynamics depending on the updating procedure of the variables x_i, y_i . We will analyse two different types of dynamics:

- (i) *Uncoupled or sequential dynamics.* In this case the two sets of variables x and y are updated sequentially. First the x_i variables are updated and the move is accepted according to the total change of energy $\Delta E = \Delta E_1 - \epsilon \Delta Q$. Next, the variables y_i are changed and the move accepted according to the energy change $\Delta E = \Delta E_2 - \epsilon \Delta Q$. This procedure is then iterated. In this case, the dynamics does not affect the two sets of variables simultaneously but each set is updated independently from the other. The only coupling between the two sets of oscillators comes from the explicit coupling term ϵQ in the Hamiltonian. For $\epsilon = 0$ the dynamics is trivial because the dynamical evolution is that of two independent sets of harmonic oscillators, and everything reduces to the original model described in section 2.
- (ii) *Coupled or parallel dynamics.* In this case the x_i, y_i variables are updated in parallel according to the rule $x_i \rightarrow x_i + r_i/\sqrt{N}$, $y_i \rightarrow y_i + s_i/\sqrt{N}$ where r_i and s_i are random variables Gaussian with zero mean and variance Δ_1 and Δ_2 , respectively. The transition probability for that move $W(\Delta E)$ is determined by the change in the total energy $\Delta E = \Delta E_1 + \Delta E_2 - \epsilon \Delta Q$ introducing, on top of the explicit coupling term ϵQ in the Hamiltonian, an additional coupling between the whole set of oscillators through the parallel updating dynamics. In contrast to the uncoupled case, the $\epsilon = 0$ case is interesting here because it will show how this kind of dynamical coupling strongly influences the glassy behaviour. In fact, in the limiting case $\epsilon = 0$, there will be some changes which increase the energy of one of the two systems, this change being accepted because the total energy will decrease. Because of that, and in spite of the fact that there is no direct coupling in the Hamiltonian, the dynamics turns out to be strongly coupled.

We will focus on the behaviour of two-times quantities such as correlation and response functions and the corresponding effective temperatures. These quantities will refer to three classes of systems: the set of oscillators described by the x variables, the set of oscillators described by the y variables and the whole set of x and y variables. In the remainder of the paper the subindex 1 will refer to quantities describing the set x of oscillators, the subindex 2 will refer to quantities describing the set y of oscillators and the subindex T will refer to quantities describing the whole set of oscillators x plus y . The main set of correlation and response functions that we are interested in are:

- *Correlations.* The correlation function for the sets x and y ,

$$C_1(t, s) = \frac{1}{N} \sum_{i=1}^N x_i(t)x_i(s) \quad C_2(t, s) = \frac{1}{N} \sum_{i=1}^N y_i(t)y_i(s) \quad (10)$$

as well as the global correlation $C_T(t, s) = \frac{1}{2}(C_1(t, s) + C_2(t, s))$.

- *Response functions.* The response function for the sets x and y are defined in the following way. Consider two external fields h_1 and h_2 conjugate, respectively, to the magnetizations $M_1 = \frac{1}{N} \sum_{i=1}^N x_i$ and $M_2 = \frac{1}{N} \sum_{i=1}^N y_i$, such that:

$$H = \frac{K_1}{2} \sum_{i=1}^N x_i^2 + \frac{K_2}{2} \sum_{i=1}^N y_i^2 - \sum_i (h_1 x_i + h_2 y_i) - \epsilon \sum_i x_i y_i. \quad (11)$$

The response functions $G_1(t, s)$, $G_2(t, s)$ measure the changes in the magnetizations $M_1(t)$ and $M_2(t)$ induced by their respective conjugate fields h_1 and h_2 applied at time s . These are defined by:

$$G_{1,2}(t, s) = \left(\frac{\delta M_{1,2}(t)}{\delta h_{1,2}(s)} \right)_{h_{1,2}=0}. \quad (12)$$

Apart from these two response functions we may define the global response function $G_T(t, s)$ as the change in the global magnetization $M_T = \frac{1}{2}(M_1 + M_2)$ induced by a field conjugate to the total magnetization:

$$G_T(t, s) = \left(\frac{\delta M_T(t)}{\delta h(s)} \right)_{h=0} = \frac{1}{2}(G_1(t, s) + G_2(t, s)). \quad (13)$$

- *Effective temperatures.* From the correlation and response functions we may define three effective temperatures: T_{eff}^1 for system 1, T_{eff}^2 for system 2 and T_{eff}^T for the global system. These are defined as follows:

$$T_{\text{eff}}^1 = \left(\frac{\frac{\partial C_1(t,s)}{\partial s}}{G_1(t, s)} \right) \quad T_{\text{eff}}^2 = \left(\frac{\frac{\partial C_2(t,s)}{\partial s}}{G_2(t, s)} \right) \quad T_{\text{eff}}^T = \left(\frac{\frac{\partial C_T(t,s)}{\partial s}}{G_T(t, s)} \right). \quad (14)$$

We will analyse in detail the three effective temperatures for the coupled and the uncoupled cases and show when, depending on the dynamics, these effective temperatures equalize.

4. Results

In this section we will summarize and analyse the results obtained in [14], where the interested reader will find all the technical details. It can be shown that the equilibrium results are the expected ones. Independently of the dynamics, the effective temperatures are just the temperature of the bath

$$T_{\text{eff}}^1 = (2E_1 - \epsilon Q)/N = T \quad T_{\text{eff}}^2 = (2E_2 - \epsilon Q)/N = T \quad (15)$$

where, in equilibrium, the energies of the subsystems are:

$$E_1^{\text{eq}} = E_2^{\text{eq}} = \frac{K_1 K_2 T}{2(K_1 K_2 - \epsilon^2)} N \quad Q^{\text{eq}} = \frac{2\epsilon E_1^{\text{eq}}}{K_1 K_2} N. \quad (16)$$

Now we analyse the results for the effective temperatures for the two different dynamics defined in the off-equilibrium regime.

4.1. Sequential case

The first quantity we have to focus on is the energy of each of the two subsystems. In this case we can find an asymptotic solution for the dynamical equations [14]. To first order in logarithmic corrections $1/\log(t)$ we find in the limit $\epsilon \approx 0$:

$$E_1 = \frac{K_1^2 K_2 \Delta_1^2}{16(K_1 K_2 - \epsilon^2) \log(t)} N \quad E_2 = \frac{K_1 K_2^2 \Delta_2^2}{16(K_1 K_2 - \epsilon^2) \log(t)} N. \quad (17)$$

Solving the dynamic equations for the correlations and responses one can find an analytic expression for the effective temperatures, considering the two-times t, s both large but with $t - s \ll s$. For a weak coupling (i.e. $\epsilon \approx 0$) the values of the effective temperatures are, in the limit $s \rightarrow \infty$:

$$T_{\text{eff}}^1 \approx (2E_1(s))/N + \mathcal{O}(\epsilon^2) \approx \frac{K_1^2 K_2 \Delta_1^2}{8(K_1 K_2 - \epsilon^2) \log(t)} \quad (18)$$

$$T_{\text{eff}}^2 \approx (2E_2(s))/N + \mathcal{O}(\epsilon^2) \approx \frac{K_1 K_2^2 \Delta_2^2}{8(K_1 K_2 - \epsilon^2) \log(t)}. \quad (19)$$

This yields in the $s \rightarrow \infty$ limit a non-vanishing relative difference $T_{\text{eff}}^1/T_{\text{eff}}^2 - 1$. This is a consequence of the fact that the two energies are different in the long-time regime (see equation (17)). For each subsystem the effective temperature verifies the equipartition theorem for long times. This means that, at any time, each subsystem can be considered as if it were at ‘quasi-equilibrium’ at its corresponding effective temperature. The fact that the two effective temperatures are different implies that there are some degrees of freedom hotter than others. One can then imagine that there is always some kind of heat transfer or current flow going from the ‘hot degrees’ of freedom to the ‘cold’ ones. Then, one may ask why the effective temperatures do not asymptotically equalize. The reason is that the off-equilibrium conductivity may vanish with time fast enough for the heat transfer not to be able to compensate for the temperature difference². In this situation, if we compute the total effective temperature for the whole system we see that in the off-equilibrium regime this temperature does not coincide with the sum of the energies of the systems. This is the same situation we found in equilibrium. If we have two systems in local equilibrium at different temperatures and put them in contact, the global system never verifies FDT unless the two temperatures are the same. In our case, we have two systems which are in ‘quasi-equilibrium’ at two different effective temperatures, so the T_{eff}^T would never be the sum of the two energies unless the two effective temperatures $T_{\text{eff}}^1, T_{\text{eff}}^2$ were the same. In other words, two systems that are thermodynamically stable at different temperatures are not globally stable when put in contact.

4.2. Parallel dynamics

As in the case without coupling, the interesting limit for the dynamics is the case where the temperature of the bath is zero. The energies and the overlap then decay to zero logarithmically:

$$E_1 = \frac{K_1 K_2 J}{8(K_1 K_2 - \epsilon^2) \log(t)} N \quad E_2 = \frac{K_1 K_2 J}{8(K_1 K_2 - \epsilon^2) \log(t)} N \quad (20)$$

$$Q = \frac{\epsilon J}{4(K_1 K_2 - \epsilon^2) \log(t)} N \quad \text{with} \quad J = \frac{K_1 \Delta_1^2}{2} + \frac{K_2 \Delta_2^2}{2}.$$

For the effective temperature we may write, up to order ϵ^2 and in the limit $s \rightarrow \infty$ (with t/s finite)

$$T_{\text{eff}}^1 = (2E_1(s) - \epsilon Q(s))/N \quad T_{\text{eff}}^2 = (2E_2(s) - \epsilon Q(s))/N \quad (21)$$

² A detailed study can be found in [12].

and because the asymptotic values of $E_1(s)$ and $E_2(s)$ are the same the effective temperatures for the subsystems are also the same in the long-time limit. Note that the case with dynamic coupling (i.e. parallel dynamics) is qualitatively different from the case without dynamic coupling (sequential dynamics), because now all the degrees of freedom are at the same effective temperature in the long-time limit. As a result, if we consider the global system, the total effective temperature defined in (14) is, in the limit $s \rightarrow \infty$ with t/s finite

$$T_{\text{eff}}^T = T_{\text{eff}}^1 = T_{\text{eff}}^2 = (2E'(s) - \epsilon Q(s))/N \quad (22)$$

where $E' = E_1 = E_2$ and Q are given by (20). Now the dynamic coupling equalizes the energies of the subsystems and we can define an effective temperature for the global system using FDT. The situation is the same as in equilibrium systems. If we have two systems in equilibrium at a certain temperature T , FDT not only holds for each subsystem but also holds for the whole system, with the temperature of the bath T . Even if we specialize to the case in which the coupling constant vanishes, $\epsilon = 0$, then the systems are still coupled through the dynamics and we obtain, qualitatively, the same results: $T_{\text{eff}}^T = T_{\text{eff}}^1 = T_{\text{eff}}^2 = (2E'(s))/N$ with $E'(s) \approx \frac{J}{8 \log(s)} N$. We conclude that the dynamic coupling does not allow the presence of more than one effective temperature in the whole system because even in the absence of an explicit coupling in the Hamiltonian, the dynamics itself causes the energies to equalize in the long-time regime.

5. Conclusions

In this paper we have focused our attention on the concept of the effective temperature defined through the FDR (1). The effective temperature, a parameter defined as an extension of FDT to the off-equilibrium regime, has been introduced in the context of glass theory in order to understand the physics behind the dynamic behaviour of these out-of-equilibrium systems. In this paper we hope to have clarified some aspects of the physical meaning of this effective temperature. We have studied two types of coupling between two subsystems of oscillators, both in an ageing state, finding that the way we couple them is crucial for the validity of the zeroth law in the off-equilibrium regime. The two cases we studied are the dynamically uncoupled or sequential case and the dynamically coupled or parallel case. In short, for the sequential case the coupling between the variables of the two subsystems in the dynamics arises only through the Hamiltonian term ϵQ . For the parallel case, the variables of the two subsystems are simultaneously updated leading to further interaction between the two subsystems (on top of the ϵQ coupling term in the energy).

For the dynamically uncoupled or sequential case the two subsystems asymptotically reach different effective temperatures which never equalize. So we can divide the system into two parts, each part characterized by its own effective temperature. The reason for this behaviour is that the off-equilibrium thermal conductivity decays too quickly to allow for an asymptotic equalization of the two effective temperatures. In fact, it decays as [12]:

$$J \approx \frac{1}{t \log^2(t)} \left(\frac{1}{T_{\text{eff}}^1} - \frac{1}{T_{\text{eff}}^2} \right). \quad (23)$$

Physically this means that, as time goes on, although the effective temperatures of the systems are different the conductivity is not large enough and they cannot equilibrate. This seems to be the reason for the existence of different temperatures in real glasses, in which there are *fast* degrees of freedom and *slow* ones at different effective temperatures that never equalize due to the smallness of the conductivity. Our conclusion is that the zeroth law is probably valid but hardly effective due to the very small conductivity between the two subsystems in the ageing state.

We have seen that for the dynamically coupled or parallel case, the two effective temperatures equalize and the two subsystems are in a sort of thermal equilibrium in the ageing state. As a result, all degrees of freedom have the same temperature. In this case, the direct coupling of the two subsystems through the parallel dynamics makes the conductivity much larger than in the sequential case and so in this case a zeroth law is effective in the ageing state. In fact, these results are also valid when we consider the special case $\epsilon = 0$, which shows that the dynamics itself is enough to equalize the effective temperatures.

Dynamics in real structural glasses involves small-scale motions of atoms, and coupling between the different degrees of freedom occurs at the level of the energy or Hamiltonian and never at the level of the dynamics. Thus, out of the two types of coupling that we have considered, only the first one is realistic.

The results of this paper explain, then, why different degrees of freedom in structural glasses can stay at different effective temperatures forever. The off-equilibrium conductivity or heat transfer between the different degrees of freedom is small enough for the equalization of the effective temperatures associated with the different degrees never to occur. This explains why when we touch a piece of glass we feel it at room temperature. To conclude, although a zeroth law for non-equilibrium glassy systems may hold, it is hardly effective because of the small energy transfer occurring between degrees of freedom at different effective temperatures. It would be very interesting to pursue this investigation further by studying other solvable examples and showing that what we have exemplified here is generally valid for structural glasses as well as for other glassy systems.

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