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Towards an experimental determination of the number of metastable states in spin-glasses?

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Abstract. — We argue that the systematic deviation from perfect aging (which means that the relaxation function only depend on the ratio $\frac{t}{t_w}$, where t_w is the waiting time) observed in spin-glasses can be accounted for if the number of available metastable states is finite. A significant fraction of these metastable states will be visited after an “ergodic” time which we estimate from the experiments to be $\simeq 10^6$ seconds. This corresponds to $\simeq 10^{12}$ - 10^{14} different metastable states per independent subsystem.

1. Introduction.

The experimental investigations of the dynamics of the spin-glass phase have revealed that the observed properties are evolving with the time elapsed from the quench: this is the aging phenomenon [1, 2]. A well known example is the relaxation of the thermo-remanent magnetisation (TRM): after field-cooling the sample from above T_g , the longer the waiting time t_w before the field is cut-off, the slower the subsequent magnetisation decay, as if the spin-glass had become “stiffer” with time. Similar effects are observed in the mechanical properties of polymer glasses [3, 4]. Aging experiments also offer a unique way of “scanning” the phase space structure of complex systems [5], and a detailed understanding of these experiments will probably offer a key for this long debated problem [6-9]. Recently, one of us proposed [10] a phenomenological theory of aging based on the assumption that energy barriers are distributed exponentially, as suggested by mean field solutions of the spin-glass problem. The probability to find the system in a given metastable state with lifetime τ is found to be *extremely broad* in the spin-glass phase. As usual with broad distributions, one is only sensitive to the few largest events encountered (see e.g. [11]). In the present case, the dynamics is entirely controlled by the longest “trap” encountered after time t_w , which is of order t_w itself. Only metastable states with lifetime of the order of t_w have a finite probability to be observed; in particular

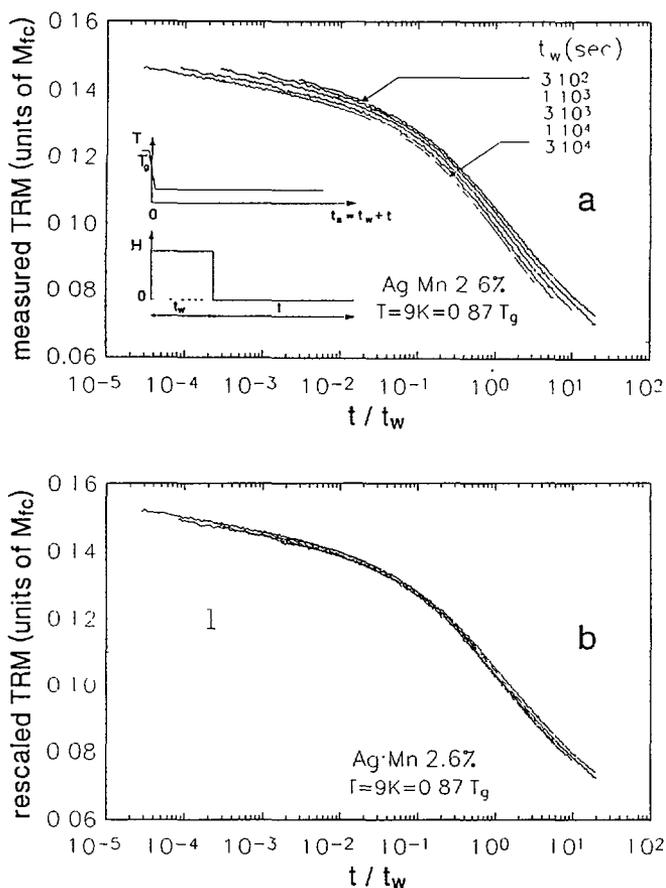


Fig. 1. — a) TRM relaxation in Ag:Mn 2.6% at 9 K, as a function of $\log(\frac{t}{t_w})$, for $t_w = 300, 1000, 3000, 10000$ and 30000 seconds. The systematic shift towards faster relaxation as t_w increases is clearly seen. Insert: sketch of the experimental TRM procedure. b) Same data as in figure 1a, but with the "interrupted aging" correction, equation (3), expanded to second order in $\frac{t+t_w}{t_{\text{erg}}}$. The error bar is an estimate of the systematic uncertainty in the vertical axis.

the microscopic time scale totally disappears from the problem [10]. Hence, quite naturally, all dynamical phenomena indeed take place on the scale of t_w . Simple arguments, building upon these ideas, show that - say in thermoremanent magnetisation relaxation experiments - the magnetisation decays as $f(\frac{t}{t_w})$, where t is the time after the switch off of the magnetic field, t_w is the "waiting time" during which the field is on, and $f(u)$ is a function which behaves as a "stretched exponential" for small u , and as a power law for large u [10, 12]. This prediction is in very good agreement with experiments over nearly five time decades for a given t_w . However, curves for different t_w fail to collapse onto the same curve when plotted versus the variable $\frac{t}{t_w}$ (see Fig. 1a): there is a systematic deviation, which suggests that the relaxation of older systems is "faster" than it should be (although of course, when plotted versus t , the older the system, the slower the relaxation). The same effect is observed both in metallic and insulating

spin-glasses [2], but also on the viscoelastic properties of polymeric glasses [3]. In order to find a unique “Master” curve, the authors of [2] proposed to take into account this systematic shift by introducing a relaxation time distribution of the form $\rho\left(\frac{\tau}{(t+t_w)^\mu}\right)$, where μ is around 0.9. This allows one to rescale all data quite satisfactorily as a function of a reduced time variable which is equal to $\frac{t}{t_w^\mu}$ in the $t \ll t_w$ limit. (A similar procedure was in fact proposed by Struik [3]). From a fundamental point of view this procedure has an intriguing consequence: on dimensional grounds, the scaling variable should really be written as $\frac{t}{\tau^{*1-\mu}t_w^\mu}$. The value of the time scale τ^* can be extracted from the experiments and is extremely large [2]: $\simeq 10^{20}$ seconds! This is surprising since only two time scales are *a priori* available: the microscopic time scale $\tau_0 \simeq 10^{-12}$ seconds (which, as explained above, should disappear in the spin-glass phase) and the experimental waiting time t_w .

The aim of this letter is to give a physical meaning to τ^* and to propose a new way to estimate it within the framework of the theory developed in [10]. We argue that the systematic deviation from a simple $\frac{t}{t_w}$ scaling can be explained *provided one takes into account the finite number of available metastable states* in real samples made of “grains” of finite size. In short, the idea is the following: a finite size system will eventually find the “deepest trap” in its phase space, which corresponds to the equilibrium state [10]. This will take a long but finite time t_{erg} , when the waiting time t_w exceeds this “ergodic” time t_{erg} , aging is “interrupted” because the phase space has been faithfully probed. Beyond this time scale, conventional stationary dynamics resume. A comparison of this *equilibrium* dynamics with the numerous available results [6], like e.g. a.c. susceptibility experiments at not too low frequency, is however beyond the scope of the present simple model.

It is reasonable to assume that the experimental samples are collections of *independent* subsystems, each of which evolving within its own complicated phase space. There will thus be a distribution of ergodic times - some shorter and some longer. The systematic deviation from a perfect $\frac{t}{t_w}$ scaling is the consequence of the partial “death” of the system, i.e. that aging is interrupted in a certain fraction of subsystems. This is what was mimicked by the previous $\frac{t}{\tau^{*1-\mu}t_w^\mu}$ scaling proposed in [2]. Here we show how one may quantify this effect, which allows us to extract from the spin-glass data a typical ergodic time of the system - which is in turn related to the number of available metastable states. In fact, this “interrupted aging” phenomenon has been observed in 2-D spin-glasses [13], in polymer glasses [14] and possibly in glassy Charge Density Wave systems [15]. All these systems however reveal important qualitative and quantitative differences. For example, the ergodic time scale in CDW appears to be much shorter than in spin-glasses.

2. The model.

In order to be self-contained, let us first recall the results of reference [10]. An exponential distribution of barrier heights (suggested by the mean-field models of spin-glasses [16, 7] induces a power-law distribution for the lifetime of metastable states:

$$\psi(\tau) = \frac{\tau_0^x}{\tau^{1+x}} \quad (\tau \gg \tau_0) \quad (1)$$

where τ_0 is a microscopic time scale ($\sim 10^{-12}$ s). $x(T)$ is a temperature dependent parameter describing the structure of phase space: the characteristic free-energy barriers are of the order

of $\frac{T}{x(T)}$ [7], and might thus be temperature dependent. x is between 0 and 1 in the spin-glass phase, and thus, from equation (1), one finds that the average exploration time is infinite. The probability $P(\tau)$ to find the system in a metastable state of lifetime τ is proportional to $\tau\psi(\tau)$ which, for $x < 1$, is non-normalisable [9, 10, 17]. For a finite observation time t_w , however, the distribution is cut-off above $\tau \simeq t_w$ - hence leading to a t_w dependent normalisation. The resulting $P(\tau)$ is then found to be independent of τ_0 : $P(\tau) \propto \frac{t_w^{x-1}}{\tau^x}$ (for $\tau < t_w$). These only ingredients are sufficient to induce aging effects [18]. For $x > 1$, on the other hand, the average exploration time is finite and conventional stationary relaxation occurs.

There might however be physical mechanisms which limit the possible values of τ . For example if, as discussed above, the total number of metastable states of a given subsystem is finite, $= \mathcal{S}$, the "ergodic time" is of the order of the longest τ drawn from distribution (1): $t_{\text{erg}}(\mathcal{S}) \simeq \tau_0 \mathcal{S}^{\frac{1}{x}}$. The relaxation of the magnetisation $m(t, t_w, t_{\text{erg}}(\mathcal{S}))$ after a given waiting time t_w is found to be, for $t_w \ll t_{\text{erg}}(\mathcal{S})$, $f(\frac{t}{t_w})$ with $f(u) = \exp - \left[\frac{\gamma_I}{1-x} u^{1-x} \right]$ for $u \ll 1$, and $f(u) \propto u^{-\gamma_{II}}$ for $u \gg 1$ [10]. γ_I is a number quantifying the tendency of the system to loose its magnetisation rather than continue to evolve within magnetized states. γ_{II} is proportional to γ_I , with an x dependent coefficient (which was, for simplicity, taken to be 1 in [10] - see below). For longer times $t_w \gg t_{\text{erg}}(\mathcal{S})$, $m(t, t_w, t_{\text{erg}}(\mathcal{S}))$ becomes a function of $\frac{t}{t_{\text{erg}}(\mathcal{S})}$. This function is in fact very nearly the same f [10] (and the small difference does not affect the following conclusions: only its *monotonic* character is of importance). Hence, the magnetisation of a given subsystem can be written in a compact form as:

$$m(t, t_w, t_{\text{erg}}(\mathcal{S})) = m_0(\mathcal{S}) f \left(\frac{t}{t^* \left[\frac{t+t_w}{t_{\text{erg}}(\mathcal{S})} \right]} \right) \quad (2)$$

with $t^*[\] = t_w$ for $t + t_w \ll t_{\text{erg}}(\mathcal{S})$ and $t^*[\] = t_{\text{erg}}(\mathcal{S})$ for $t + t_w \gg t_{\text{erg}}(\mathcal{S})$. In words, equation (2) simply means that once equilibrium is achieved ($t + t_w \gg t_{\text{erg}}(\mathcal{S})$), the magnetisation is much smaller than its "short" time extrapolation, $f(\frac{t}{t_w})$.

Now, assuming that the number of metastable states varies from one subsystem to another according to a distribution $\mathcal{P}(\frac{\mathcal{S}}{\mathcal{S}_0})$ (\mathcal{S}_0 is thus a "typical" number of metastable states per subsystem), the total (observed) magnetisation $M(t, t_w)$ is obtained as the average over \mathcal{P} of equation (2). With these only assumptions, one finally finds:

$$\frac{M(t, t_w)}{M_0} = f\left(\frac{t}{t_w}\right) \left(1 - \mathcal{F} \left[\left(\frac{t+t_w}{t_{\text{erg}}^0} \right)^x \right] \right) \quad (3)$$

where $t_{\text{erg}}^0 \equiv \tau_0 \mathcal{S}_0^{\frac{1}{x}}$ is the "ergodic time" of the system, and $\mathcal{F}[\]$ is an increasing function which depends on the choice of $m_0(\mathcal{S})$ and \mathcal{P} . Before taking a specific example, it should be noted that equation (3) qualitatively agrees with the experimental trend: older systems indeed relax "faster" when plotted versus $\frac{t}{t_w}$

3. Analysis of the experimental data.

The simplest assumption is that m_0 and \mathcal{P} behave regularly when their arguments are small. In this case, one finds, for $t + t_w \ll t_{\text{erg}}^0$ $\mathcal{F}(u) = Au + Bu^2 + \dots$, where A is a constant which

Table I. — Parameters x , γ_I , γ_{II} , t_{erg}^0 and “complexity” Ω for the Ag:Mn 2.6% sample [2]. x , γ_I were extracted by fitting the “initial” part of the TRM relaxation ($t \leq 0.3t_w$) to a stretched exponential, while γ_{II} is obtained from the long time part ($t \geq 2t_w$) of the relaxations. Note that $\Omega(T = 10 \text{ K})$ could not be estimated because of our lack of precision in the vertical positioning of the relaxation curves. This is a problem close to T_g since the magnetisation falls rapidly to small values.

$T(\text{K})$	x	γ_I	γ_{II}	$t_{\text{erg}}^0 (10^6 \text{ s.})$	Ω
8	0.65 ± 0.05	0.083 ± 0.010	0.090 ± 0.003	4 ± 2	12.1 ± 0.9
9	0.65 ± 0.05	0.0124 ± 0.015	0.127 ± 0.003	2 ± 1	11.9 ± 0.9
9.5	0.68 ± 0.05	0.153 ± 0.020	0.155 ± 0.005	0.8 ± 0.3	12.2 ± 0.9
10	0.75 ± 0.05	0.221 ± 0.030	0.265 ± 0.005	-	-

Table II. — Same as in table I, but for the $\text{CdCr}_{1.7}\text{In}_{0.3}\text{S}_4$ sample [2]. Here, the initial part of the relaxation was extended to $t \leq 0.5t_w$. The value of the parameters given here are compatible with those obtained in reference [10], where the additional assumption $\gamma_I \equiv \gamma_{II}$ was made.

$T(\text{K})$	x	γ_I	γ_{II}	$t_{\text{erg}}^0 (10^6 \text{ s.})$	Ω
10	0.76 ± 0.05	0.059 ± 0.008	0.058 ± 0.003	5 ± 2	14.2 ± 0.9
12	0.76 ± 0.05	0.100 ± 0.01	0.100 ± 0.003	3 ± 1	14.0 ± 0.9
14	0.73 ± 0.05	0.200 ± 0.02	0.182 ± 0.010	0.6 ± 0.2	13.0 ± 0.9

can be reabsorbed in the definition of t_{erg}^0 [19]. We show in figure 1b the same data as in figure 1a, but with the “interrupted aging” correction, equation (3). The only free parameters are the ergodic time of the system $t_{\text{erg}}^0(\mathcal{S})$ and the value of B (the value of x is fixed by fitting the “youngest” curve - see [19]). We have however imposed the same value of $B = -3$ (i.e. the same functional form for \mathcal{P}) for all temperatures and samples. The collapse of the curves is quite good in all cases (see Fig. 1b). Hence, and this is the main result of this paper, *our interrupted aging scenario is able to mimic a “ $\frac{t}{t_w^\mu}$ ” scaling with $\mu < 1$.*

The corresponding values of the parameters x , γ_I , γ_{II} , t_{erg}^0 , and of the “complexity” $\Omega \equiv x \log_{10}(\frac{t_{\text{erg}}^0}{\tau_0})$ are given in tables I and II, for different temperatures and two different samples: Ag:Mn 2.6% (AgMn), which is metallic and $\text{CdCr}_{1.7}\text{In}_{0.3}\text{S}_4$ (CrIn) which is an insulating spin glass. It is interesting to see that both samples lead to similar conclusions:

a) The ergodic time t_{erg}^0 is of the order of 10^6 seconds, and thus, within our interpretation, the number of metastable states per “subsystem” is found to be $\simeq 10^{12} - 10^{14}$ (see Tabs. I, II). This number is however not easy to discuss since we do not know precisely what these “subsystems” are. They may be, for example, magnetically disconnected regions (like grains) the size of which being determined by the sample preparation, and thus is temperature independent. It could also be that the phase space of 3d spin-glasses is broken into mutually inaccessible regions (“true” ergodicity breaking). In this case, \mathcal{S} would denote the number of metastable states within one such ergodic component. This number would then be an intrinsic property of 3d spin-glasses. We however think that numerical simulations [20] and/or experiments on mesoscopic spin-glasses [21] are really needed to understand how the ergodic time t_{erg}^0 is related to the number of spins contained in a subsystem.

b) As shown in figure 2, the number of metastable states of the insulating sample (CrIn) tends to increase when the temperature is lowered (it roughly doubles when the temperature is

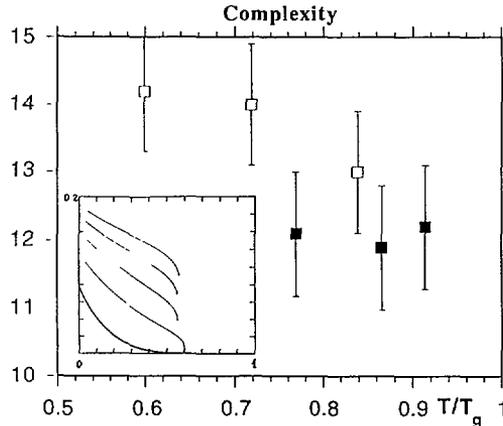


Fig. 2. — Evolution of the “complexity” $\Omega \equiv x \log_{10} \frac{t_{\text{erg}}^0}{\tau_0}$ with reduced temperature $\frac{T}{T_g}$ for both AgMn and CrIn. Note that the complexity does not seem to go to zero *continuously* at T_g . Insert: Evolution of the complexity per spin versus reduced temperature for the p -spin model, with, from bottom to top: $p = 2$ (SK model), 3,4,5 and 6 ($p = \infty$ is the Random Energy Model). Note that Ω is discontinuous at T_g as soon as $p \geq 3$. From reference [24].

lowered by 1 Kelvin). However, in contrast to the SK model [22], *this number does not seem to vanish continuously at the spin-glass transition T_g* . Such a behaviour was found in “Potts glass” models [23] and in models with $p(> 2)$ - spin interactions [16],[24] - see figure 2 (insert). The number of metastable states of the metallic sample (AgMn) seems to be roughly temperature independent, although the rather large error bars leave room for the same behaviour as in CrIn.

c) The parameter x grows towards 1 when $T \rightarrow T_g$ (at least for AgMn, but other experiments on CrIn showed the same tendency [2]). This behaviour was also found in a very clear way in reference [25], where the initial decay of the magnetisation was also fitted by a stretched exponential. Our results ($\Omega(T_g)$ finite and $x(T_g) \simeq 1$) suggest that the spin-glass transition is, in these samples, much closer to the “Random Energy Model” scenario [16], than to the “SK” scenario where $\Omega(T_g) = 0$ and $x(T_g) = 0$ [7]. We however note that x significantly departs from the REM linear behaviour $x = \frac{T}{T_g}$, which would hold if the energy barriers were temperature independent. Furthermore, the REM “replica symmetry breaking” scheme is certainly not rich enough to account for more complicated aging protocols [5] or “second spectrum” experiments [21].

d) Finally, one can see that the simplifying hypothesis $\gamma_I = \gamma_{II}$ is quite accurate. In fact, a closer look at equations (8-10) of reference [10] reveals that the ratio $\frac{\gamma_{II}}{\gamma_I}$ increases with x (and diverges for $x \rightarrow 1$), which is consistent with the experimental trend (see Tab. I).

4. Conclusion.

Let us summarize our findings: quantifying the idea that aging stops when the exploration of phase space is completed, we have proposed a way to rescale relaxations obtained for different

waiting times on a unique “Master” curve, and to understand the physical meaning of the phenomenological “ $\frac{t}{t_w^\mu}$ ” scaling. This allows to extract from the experiments a very long, temperature dependent, time scale t_{erg} beyond which aging ceases and the nature of relaxation changes. This is interesting from two standpoints: from a fundamental point of view, it allows - to our knowledge, for the first time - to estimate the number of metastable states in a complex system, and its temperature dependence. Perhaps more importantly, it allows, from a practical point of view, to extract from relatively short time measurements an ergodic time beyond which the relaxation changes qualitatively: we have in mind here the aging experiments on polymer glasses, where the mechanical properties are very similar to those discussed here [3],[4]. It could be of importance to know when these mechanical properties will appreciably change. The ideas developed here might perhaps be useful to discuss physical aging in the context of other (molecular or metallic) glasses.

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