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Memory interference effects in spin glasses

K. Jonason[1,2], P. Nordblad[1,a], E. Vincent[2,b], J. Hammann[2], and J.-P. Bouchaud[2]

[1] Department of Materials Science, Uppsala University, Box 534, SE-751 21 Uppsala, Sweden
[2] Service de Physique de l'État Condensé, CEA Saclay, 91191 Gif-sur-Yvette Cedex, France

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Abstract. When a spin glass is cooled down, a memory of the cooling process is imprinted in the spin structure. This memory can be disclosed in a continuous heating measurement of the ac-susceptibility. *E.g.*, if a continuous cooling process is intermittently halted during a certain aging time at one or two intermediate temperatures, the trace of the previous stop(s) is recovered when the sample is continuously re-heated [1]. However, heating the sample above the aging temperature, but keeping it below T_g, erases the memory of the thermal history at lower temperatures. We also show that a memory imprinted at a higher temperature can be erased by waiting a long enough time at a lower temperature. Predictions from two complementary spin glass descriptions, a hierarchical phase space model and a real space droplet picture are contested with these memory phenomena and interference effects.

PACS. 75.50.Lk Spin glasses and other random magnets – 75.10.Nr Spin-glass and other random models – 75.40.Gb Dynamic properties (dynamic susceptibility, spin waves, spin diffusion, dynamic scaling, etc.)

1 Introduction

The non-equilibrium character of the dynamics of $3d$ spin glasses below the zero field phase transition temperature has been extensively studied by both experimentalists and theorists [2]. Two different main tracks have been used to describe the aging and non-equilibrium dynamics that is characteristic of spin glasses. On the one hand phase space pictures [3], which originate from mean field theory [4] and prescribe a hierarchical arrangement of metastable states, and on the other hand real space droplet scaling models which have been developed from renormalisation group arguments [5,6]. Independently, a theoretical description (which we shall not discuss here) of aging effects [7] and temperature variation effects [8] has been given by the direct solution of the dynamical equations of mean-field like models.

When a spin glass is quenched from a high temperature (above T_g) to a temperature T_1 below T_g, a wait time dependence of the dynamic magnetic response is observed. This aging behaviour [9,10] corresponds to a slow evolution of the spin configuration towards equilibrium. The "magnetic aging" observed in spin glasses resembles the "physical aging" observed in the mechanical properties of glassy polymers [11,12], or the "dielectric" aging found in supercooled liquids [13] and dielectric crystals [14]. However, a more detailed comparison of aging in these various systems would show interesting differences [13], particularly with respect to the effect of the cooling rate [1].

[a] e-mail: Per.Nordblad@Angstrom.uu.se
[b] e-mail: vincent@spec.saclay.cea.fr

Remarkable influences of slight temperature variations on the aging process in spin glasses have been evidenced in a wide set of earlier experiments [3,10,15,16]. These influences were further elucidated through the memory phenomena recently reported in [1]. The results have been interpreted both from "phase space" and "real space" points of view.

In phase space models, aging is pictured as a random walk among the metastable states. At a given temperature T_1, the system samples the valleys of a fixed free-energy landscape. On the basis of the experimental observations, it has been proposed [3] that the landscape at T_1 corresponds to a specific level of a hierarchical tree. When lowering the temperature to $T_2 < T_1$, the observed restart of aging is explained as a subdivision of the free energy valleys into new ones at a lower level of the tree. The system now has to search for equilibrium in a new, unexplored landscape, and therefore acts at T_2 as if it had been quenched from a high temperature. On the other hand, the experiments show that when heating back from T_2 to T_1 the memory of the previous aging at T_1 is recovered. In the hierarchical picture, this is produced by the T_2-valleys merging back to re-build the T_1-landscape.

In real space droplet pictures [5,6], the aging behaviour at constant temperature is associated with a growth of spin glass ordered regions of two types (related by time-reversal symmetry). This is combined with a chaotic behaviour as a function of temperature, [17], *i.e.* the equilibrium spin configuration at one temperature is different from the equilibrium configuration at another temperature. However, there is also an overlap between

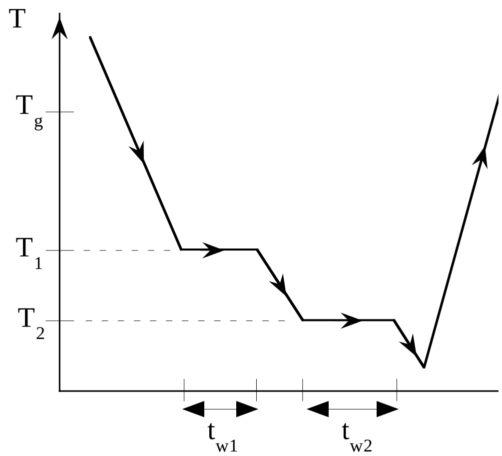

Fig. 1. The measurement procedure in the "double memory experiments" of Figures 2, 4 and 5.

the equilibrium spin structures at two different temperatures, T and $T \pm \Delta T$, on length scales shorter than the overlap length, $L_{\Delta T}$. In this picture, chaos implies that if the spin glass has been allowed to age a time, t_w, at a certain temperature, the aging process is re-initialized after a large enough temperature change. Intuitively, a growth of compact domains may not allow a memory of a high temperature spin configuration to remain imprinted in the system while the system ages at lower temperatures. However, as suggested in [1] and developed in [18], a phenomenology based upon fractal domains and droplet excitations can be able to incorporate the observed memory behaviour in a real space droplet picture. The possibility of a fractal (non-compact) geometry of the domains has been evoked in the past in various theoretical contexts [19], and also in close connection with the aging phenomena [20–22].

In this paper, we report new results on the memory phenomenon observed in low frequency ac-susceptibility of spin glasses. We first recall and demonstrate an undisturbed memory phenomenon, and then show that such a memory can be erased not only by heating the sample to a temperature above the temperature where the memory is imprinted, but also by waiting a long enough time below this temperature.

2 Experimental

The experiments were performed on the insulating spin glass $CdCr_{1.7}In_{0.3}S_4$ [23] ($T_g = 16.7$ K), in a Cryogenic Ltd S600 SQUID magnetometer at Saclay. The ac field used in the experiments had a peak magnitude of 0.3 Oe and frequency $\omega/2\pi = 0.04$ Hz. This low frequency makes the relaxation of the susceptibility at a constant temperature in the spin glass phase clearly visible (at the laboratory time scale of 10^1 to 10^5 s).

The basic experimental procedure is illustrated in Figure 1 and is as follows:

(i) cooling: the experiments are always started at 20 K, a temperature well above the spin glass temperature $T_g = 16.7$ K. The ac-susceptibility is first recorded as a function of decreasing temperature. The sample is continuously cooled, but is additionally kept at constant temperature at two intermittent temperatures T_1 and T_2 for wait times t_{w1} and t_{w2}, respectively ($T_1 < T_2 < T_g$).
(ii) heating: when the lowest temperature has been reached, the system is immediately continuously reheated and the ac-susceptibility is recorded as a function of increasing temperature.

Except at T_1 and T_2 when decreasing the temperature, the cooling and heating rates are constant (~ 0.1 K/min). At constant temperature, both components of the ac-susceptibility relax downward by about the same absolute amount. However, the relative decay of the out-of-phase is much larger than the relative decay of the in-phase component, and in the following we mainly focus on results from the out-of-phase component of the susceptibility.

3 Results

3.1 Double memory

The results of a double memory experiment are presented in Figure 2. The initial data is recorded on continuously cooling the sample including a first halt at the temperature $T_1 = 12$ K ($0.72\,T_g$) for $t_{w1} = 7$ h and a second halt at $T_2 = 9$ K ($0.54\,T_g$) for $t_{w2} = 40$ h. The cooling is then continued to $T = 5$ K, from where a new set of data is taken on increasing the temperature at a constant heating rate whithout halts. A reference curve, measured on continuous heating after cooling the sample without intermittent halts, is included in the figure.

A first important feature can be noted on the curve recorded on cooling with intermittent halts. After aging 7 h at 12 K, χ'' has relaxed downward due to aging. But when cooling resumes, the curve rises and merges with the reference curve, as if the aging at 12 K was of no influence on the state of the system at lower temperatures. This chaos-like effect (in reference to the notion of chaos in temperature introduced in [17]) points out an important difference from a simpler description of glassy systems, in which there are equivalent equilibrium states at all low temperatures and aging at any temperature implies that this equilibrium state is further approached. Here, as pictured in more detail in other experiments [1], only the last temperature interval of the cooling procedure does contribute to the approach of the equilibrium state at the final temperature.

Note that this notion of "last temperature interval" depends on the observation time scale of the measurement (χ'' is only sensitive to dynamical processes with a characteristic response time of order $1/\omega$ which in these experiments corresponds to ≈ 4 s). In magnetisation relaxation experiments, the observation time corresponds to the time elapsed after the field change, and effects of aging at a higher temperature can be seen in the long-time

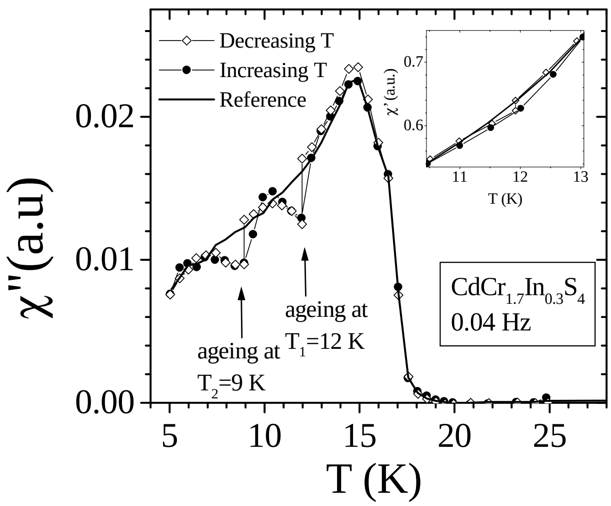

Fig. 2. Out-of-phase susceptibility *vs.* temperature. While cooling down (open diamonds), two intermittent halts are made; at $T_1 = 12$ K during $t_{\mathrm{w}1} = 7$ h, and at $T_2 = 9$ K during $t_{\mathrm{w}2} = 40$ h. The system is then reheated at a constant heating rate (full circles). The reference curve (solid line) is measured on heating the sample after cooling it without intermittent halts. The inset shows the in-phase susceptibility in the same measurement procedure, at temperatures around $T_1 = 12$ K.

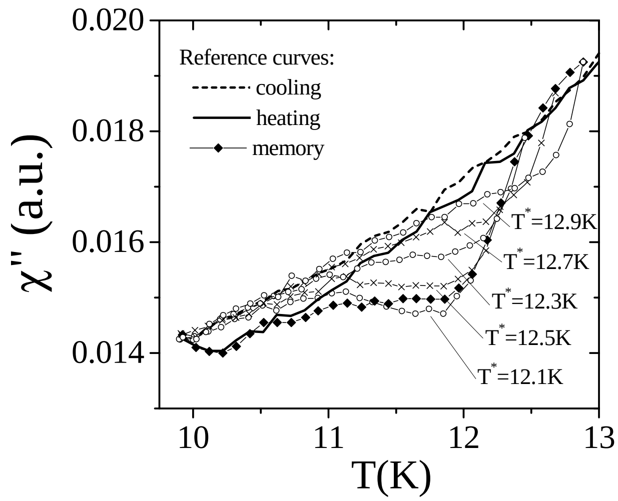

Fig. 3. Memory erasing by overheating. The memory effect at 12 K, recorded during heating with the same procedure as in Figure 2, is shown in full diamonds. Now, when reaching T^* ($T^* = 12.1$, 12.3, 12.5, 12.7 and 12.9 K), re-heating stops and the sample is cooled back. The χ'' signal (alternatively open circles and crosses for the various T^* values) during cooling shows the progressive erasing of the 12 K memory for increasing T^*. Reference curves, measured during a continuous cooling (dashed line) and heating (solid line), are also shown.

part (10^3–10^4 s) of the relaxation curves [16,24] in a correspondingly enlarged "last temperature interval" compared to that of our current ac-susceptibility experiments.

The curve recorded on re-heating in Figure 2 clearly displays the memory effect: the dips at T_1 and T_2 are recovered. The long wait time (40 h) at $T_2 = 9$ K has no apparent influence on the memory dip associated with $T_1 = 12$ K. This experiment displays a double memory where no interference effects are present, *i.e.* the two dips at T_1 and T_2 are, within our experimental accuracy, fully recovered when reheating the sample. A similar result has been obtained on a metallic Cu:Mn spin-glass sample [1,16], confirming the universality of aging dynamics in very different spin-glass realizations.

This experimental procedure has been recently reproduced in extensive simulations of the $3d$ Edwards-Anderson model. Although weaker and more spread out in temperature, similar effects of a restart of aging upon cooling and of a memory effect upon heating have been found [25].

The memory phenomenon is also observable in the in-phase susceptibility, χ'. In the inset of Figure 2, χ' is plotted in the region around $T_1 = 12$ K. A relaxation due to aging is visible, and it is also clear that when cooling resumes after aging, the χ' curve rather rapidly merges with the reference curve (chaos-like effect). Upon re-heating, the memory effect can be distinguished, the memory curve clearly departs from the reference in the 11–13 K range. The relative weakness of the deviation compared to the large χ' value can be understood in the following way. Aging in spin glasses mainly affects processes with relaxation times of the order of the age of the system; processes with shorter relaxation times are already equilibrated, and processes with longer relaxation times are not active. χ' measures the integrated response of all short time processes up to the observation time $1/\omega$, whereas χ'' only probes processes with relaxation times of order $1/\omega$. The relative influence of aging is thus smaller in χ' than in χ''.

3.2 Memory erasing by heating

The memory of aging at T_1 remains imprinted in the system during additional aging stages at sufficiently lower temperatures, and is recovered when heating back to T_1. We have investigated what remains of this T_1-memory after heating up to $T^* > T_1$ (keeping of course $T^* < T_{\mathrm{g}}$). The experiments were performed using only one intermittent stop at $T_1 = 12$ K for $t_{\mathrm{w}1} = 3$ h, and continuing the cooling to about 10 K. Then χ'' was recorded upon heating the sample to T^* and immediately re-cooling it to 10 K. The results are displayed in Figure 3. For higher and higher T^*, the memory dip at T_1 becomes weaker and weaker, finally fading out at $T^* \sim 13$ K. The additional shallow dips observed in a limited temperature region just below T^* and just above 10 K, the two temperatures where the temperature change is reversed, are due to the finite heating/cooling rate and to the overlap within this temperature range between the state created on heating (cooling) and the desireable state on re-cooling (re-heating) the sample [18].

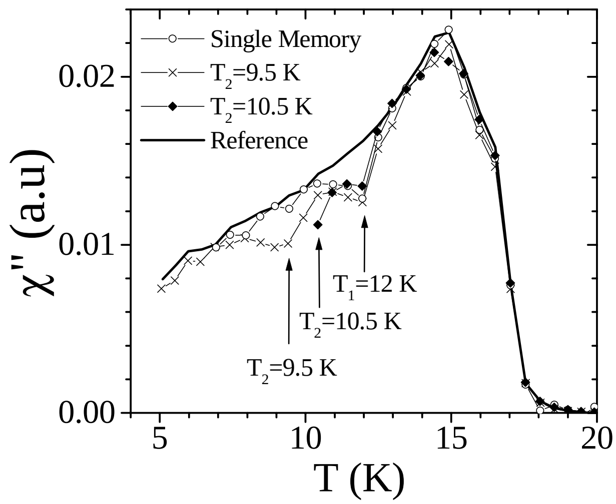

Fig. 4. Effect on the memory at $T_1 = 12$ K ($t_{w1} = 3$ h) of aging at slightly lower temperatures T_2 (9.5 K and 10.5 K) during $t_{w2} = 6$ h. All data shown was taken on reheating after the various histories. In addition to the reference curve (solid line), a "single memory" curve (open circles) is presented; it shows the 12 K memory when no additional aging at T_2 is performed. For $T_2 = 9.5$ K (crosses), there is almost no effect; for $T_2 = 10.5$ K, the 12 K memory is partly erased.

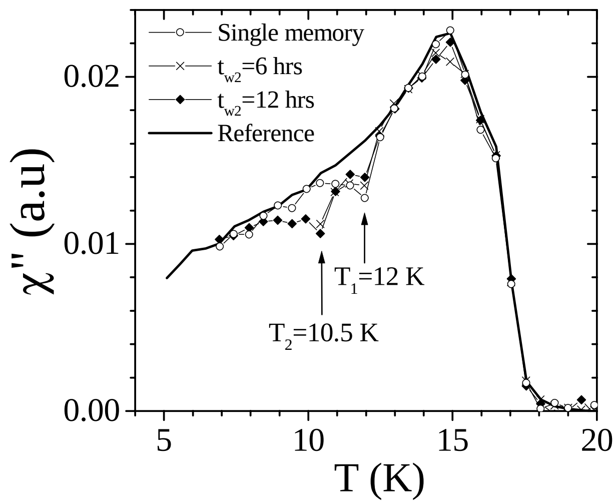

Fig. 5. Effect on the memory at $T_1 = 12$ K ($t_{w1} = 3$ h) of aging at $T_2 = 10.5$ K during $t_{w2} = 6$ (crosses) and 12 h (full diamonds). Same conventions as in Figure 4.

3.3 Memory interference

The memory is also affected by aging at a lower temperature, provided this temperature is close enough to T_1 or the time spent there is long enough. In order to systematically investigate this interference effect, we have performed double memory experiments in which we have varied the parameters T_2 and t_{w2} of the aging stage at the lower temperature, but kept the initial aging temperature $T_1 = 12$ K and wait time $t_{w1} = 3$ h fixed.

Figure 4 shows the results using a fixed value of $t_{w2} = 6$ h but two different values of T_2 (9.5 and 10.5 K). Two reference heating curves are added for comparison; one is recorded after a cooling procedure where no halts are made, and the other after cooling with only a single halt at $T_1 = 12$ K for 3 h ("single memory"). This latter curve is a reference for a pure memory effect at 12 K. The obtained 12 K dip is about the same for the pure memory and the double memory curve with $T_2 = 9.5$ K. However, in the experiment performed with $T_2 = 10.5$ K, the memory of the dip achieved at $T_1 = 12$ K has become more shallow. Thus, for a temperature difference $\Delta T = 1.5$ K, the memory of aging gets partly re-initialised, while for $\Delta T = 2.5$ K no re-initialisation is observed.

Figure 5 shows the results using a fixed value of $T_2 = 10.5$ K but two different values of the wait time at T_2 $t_{w2} = 6$ h and $t_{w2} = 12$ h. The reference curves are the same as in Figure 4. The longer the time spent at T_2, the larger is the part of the memory dip at T_1 that has been erased.

4 Discussion

4.1 Memory and chaos effects in phase space pictures

Some effects related to these memory and memory interference phenomena have been explored in the past through various experimental procedures, as well in ac as in dc (magnetisation relaxation following a field change) measurements [3,10,15]. The hierarchical phase space picture [3] has been developed as a guideline that accounts for the various results of the experiments. Although this hierarchical picture deals with metastable states as a function of temperature, it is obviously reminiscent of the hierarchical organization of the pure states as a function of their overlap in the Parisi solution of the mean field spin glass [4]. We want to recall that some more quantitative analyses of the experiments [26] have shown that the barrier growth for decreasing temperatures should be associated with a divergence of some barriers at any temperature below T_g. Thus, what can be observed of the organization of the metastable states might well be applicable to the pure states themselves. From a different point of view, another link between the hierarchical picture and mean field results has been proposed in a tree version of Bouchaud's trap model [21].

The restart of aging when the temperature is again decreased after having been halted at some value indicates that the free-energy landscape has been strongly perturbed. The metastable states have been reshuffled, but not in any random manner, because the memory effect implies a return to the previously formed landscape (with the initial population distribution) when the temperature is raised back. The restart of aging then corresponds to the growth of the barriers and to the birth of other ones, which subdivide the previous valleys into new ones where the system again starts some *ab initio* aging. This hierarchical ramification is easily reversed to produce the memory effect when the temperature is increased back.

The full memory effect seen in the experiment of Figure 2 requires a large enough temperature separation $\Delta T = T_1 - T_2$, as is shown from the memory interference effects displayed in Figures 4 and 5. If one forgets the restart of aging at the lower temperature, the memory effect can be given a simple explanation: the slowing down related to thermal activation is freezing all further evolution of the system. However, the memory effect takes place while important relaxations occur at lower temperatures, and it is clear that there must be some smaller limit of ΔT below which the T_2 evolution is of influence on the T_1 memory. From other measurements [3,10], it has been shown that in the limit of small enough ΔT's (of order 0.1–0.5 K), the time spent at T_2 contributes essentially additively to the aging at T_1, as an effective supplementary aging time. In that situation of small ΔT, the landscape at T_2 is not very different from that at T_1 (large "overlap"); the same barriers are relevant to the aging processes, although being crossed more slowly at T_2.

But this is not the case in Figures 4 and 5, where intermediate values of ΔT have been chosen. The memory interference effect demonstrated in Figures 4 and 5 is in agreement with earlier ac and dc experiments which used negative temperature cycling procedures [3,15] and intermediate magnitudes of ΔT. Such experiments were performed so that the sample first was aged at T_1 a wait time t_{w1}, and thereafter cooled to T_2 and kept there a substantial wait time t_{w2}, after which it was re-heated to T_1, where the relaxation of the ac or dc signal was recorded. The results of these experiments are that a partial reinitialisation of the system has occurred, but that simultaneously a memory of the original aging at T_1 remains. In Figures 4 and 5, the partial loss of the T_1 memory dip corresponds to such a partial reinitialisation.

In this case of an intermediate value of $\Delta T \sim 1$ K, there are indeed differences between the landscapes at both temperatures. Still, they are hierarchically related, since a memory effect is found. But the memory loss of Figures 4 and 5 suggests that the free-energies of the bottom of the valleys are different at T_1 and T_2, meaning that the thermodynamic equilibrium phase is different from one temperature to another. As discussed previously [1], the restart of aging when the temperature is lowered is suggestive of chaos between the equilibrium correlations at different temperatures. The conclusion from the current results is thus that the free-energies of the metastable states vary chaotically with temperature, which reinforces the idea of a "chaotic nature of the spin glass phase" [17].

4.2 Towards an understanding of memory and chaos effects in real space

While these phase space pictures allow a good description of many aspects of the experimental results, a correct real space picture would form the basis for a microscopic understanding of the physics behind the phenomena. As aging proceeds, χ" decreases, which means a decrease of the number of dynamical processes that have a time scale of order $1/\omega$. Aging corresponds to an overall shift of a maximum in the spectrum of relaxation times towards longer times, as was understood from the early observations of aging in magnetisation relaxation experiments [27]. Thinking of the dynamics in terms of groups of spins which are simultaneously flipped, longer response times are naturally associated with larger groups of spins. In such "droplet" [5] and "domain" [6] pictures, aging corresponds to the progressive increase of a typical size of spin glass domains. Difficulties are encountered in this real space description with "memory and chaos" effects [1]. On the one hand, the restart of aging processes when the temperature is lowered indicates the growth of domains of different types at different temperatures. On the other hand, the memory of previous aging at a higher temperature can be retrieved; thus, the low temperature growth of domains of a given type does not irreversibly destroy the spin structures that have developed at a higher temperature.

However, a heuristic interpretation of aging in spin glasses in terms of droplet excitations and growth of spin glass equilibrium domains, along the lines suggested in [1] and developed in [18], is perhaps able to include the memory phenomena discussed in this paper. The carrying idea of this phenomenology is that at each temperature there exists an equilibrium spin glass configuration that is two fold degenerate by spin reversal symmetry. The simple picture of Fisher-Huse [5], where only compact domains are considered, is hard to reconcile with the memory effect reported here [1]. As suggested in various theoretical work [19–22], we assume that the initial spin configuration results in an interpenetrating network of fractal "up" and "down" domains of all sizes separated by rough domain walls. We furthermore propose to modify somewhat the original interpretation of the "overlap length" $L_{\Delta T}$. The standard picture states that the equilibrium configurations corresponding to two nearby temperatures T and $T \pm \Delta T$, are completely different as soon as one looks at a scale larger than $L(\Delta T)$. However, in a non-equilibrium situation, we believe that some fractal large scale (larger than $L_{\Delta T}$) "skeletons", carrying robust correlations, can survive to the change of temperature, and are responsible for the memory effects. An assumption of this sort is, we think, needed to account for the existence of domains of all sizes within the initial condition. If the initial spin configuration was purely random as compared to the equilibrium one, then the problem would be tantamount to that of percolation far from the critical point, where only small domains exist.

The allowed excitations in this system are droplets of correlated "up" or "down" regions of spins of all sizes. Within this model, the magnetisation of the sample in response to a weak magnetic field is caused by polarisation of droplets, and the out-of-phase component of the susceptibility directly reflects the number of droplet excitations in the sample with a relaxation time equal to $1/\omega$. The size of spin glass domains is in the following denoted R and the size of a droplet excitation L. As a function of time, the size of the excited droplets grows as $L(T, t_a)$.

The effect of a droplet excitation on the spin configuration is different depending on the size and position of the droplet and the age of the spin glass system. A small droplet excitation $L \ll R$ most probably is just an excitation within an equilibrium spin glass configuration, yielding no measurable change of the spin system. An excitation of size $L \approx R$ may (i) remove the circumventing domain wall separating an up domain from a down domain, (ii) slightly displace an existing domain wall or (iii) just occur within an equilibrium spin configuration.

After a few decades in time, the result of the numerous dispersed droplet excitations of sizes $L \leq L(T, t_a)$ is that most domains of size $R \ll L(T, t_a)$ are removed, whereas most larger domains remain essentially unaffected, only having experienced numerous slight domain wall displacements. In other words, in this picture, the structure of the large domains is unaffected by the dynamics. If the temperature now is changed to a temperature where the overlap length, $L_{\Delta T}$, is smaller than the typical droplet size $L(T_1, t_a)$ active at the original temperature T_1, the new initial condition still leads to an interpenetrating network of up and down domains of all sizes relevant to the new temperature (chaos implies that the domain spin structure is different from the equilibrium configuration at T_1). A similar process as discussed above now creates a spin configuration of equilibrium structure on small length scales, where only small domains (up to a size $L(T_2, t_a)$) are erased, leaving large ones essentially unaffected. Returning to the original temperature, there will exist a new domain pattern on small length scales corresponding to droplets with short relaxation time and there will additionally remain an essentially unperturbed domain pattern on large length scales originating from the initial wait time at this temperature. The small length scale domains are rapidly washed out and a domain pattern equivalent to the original one is rapidly recovered.

These dynamic features of the spin structure are reflected in the susceptibility experiments discussed above. The out of phase component gives a measure of the number of droplets that have a relaxation time equal to the observation time of the experiment, $1/\omega$, the decrease of the magnitude of the susceptibility with time implies that the number of droplets of relaxation time $1/\omega$ decays toward an equilibrium value obtained when "all" domains of this size are extinguished and all subsequent droplet excitations of this size occur within spin glass ordered regions. The fact that a dip occurs when the temperature is recovered mirrors that the long length scale spin configuration is maintained during an aging period at lower temperatures, where only droplet excitations on much smaller length scales are active. The memory interference effects are within this picture immediate consequences of that the droplet excitations at the nearby temperature T_2 are allowed to reach the length scales of the original domain growth at T_1 leading to an enhanced number of droplet excitations of the size of this reconstructed domain pattern. When heating above the temperature for the aging process, Figure 3, and outside the region of overlapping states, the equilibration processes at the high temperature reach longer length scales than at the aging temperature, and the memory of the equilibration at T_1 is rapidly washed out. On the other hand, in the experiments where the sample is cooled (Figs. 4 and 5) below T_1, the processes require longer aging time to reach the length scales of the aging at T_1 and the intereference effects become larger with increased time and higher temperature.

5 Conclusions

When cooling a spin glass to a low temperature in the spin glass phase, a memory of the specific cooling sequence is imprinted in the spin configuration and this memory can be recalled when the system is continuously re-heated at a constant heating rate [1]. *E.g.* in a "double memory experiment" two intermittent halts, one at T_1 a time $t_{\mathrm{w}1}$ and another at T_2 a time $t_{\mathrm{w}2}$ are made while cooling the sample. Depending on the parameters T_2 and $t_{\mathrm{w}2}$ it is possible to partly reinitialise (erase) or fully keep the memory of the halt at the higher temperature T_1.

These memory and memory interference effects can on the one hand be incorporated in hierarchical models for the configurational energies at different temperatures including a chaotic nature of the spin glass phase. On the other hand, a preliminary phenomenological real space picture has been proposed to account for the observed phenomena. In our mind, much remains to be done on the theoretical side to put this "fractal" droplet picture on a firmer footing.

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