Memory and Chaos Effects in Spin Glasses

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New low frequency ac susceptibility measurements on two different spin glasses show that cooling/heating the sample at a constant rate yields an essentially reversible (but rate dependent) $\chi(T)$ curve; a downward relaxation of χ occurs during a temporary stop at constant temperature (*aging*). Two main features of our results are (i) when cooling is resumed after such a stop, χ goes back to the reversible curve (*chaos*), and (ii) upon reheating, χ perfectly traces the previous aging history (*memory*). We discuss implications of our results for a *real space* (as opposed to *phase space*) picture of spin glasses. [S0031-9007(98)07362-1]

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The dynamic properties of the spin-glass phase have been extensively studied by both experimentalists and theorists for almost two decades [1,2]. The observed properties reflect the out-of-equilibrium state of the system: The response to a field variation is logarithmically slow, and, in addition, depends on the time spent at low temperature ("aging"). Aging is fully reinitialized by heating the sample above the glass temperature T_g. It corresponds to the slow evolution of the system towards equilibrium, starting at the time of the quench below T_g. Many aspects of aging are similar to the "physical aging" phenomena that have been characterized in the mechanical properties of glassy polymers [3]. In the past few years, some interesting progress in the theoretical understanding of aging in disordered systems has been achieved [4].

From the studies of the critical behavior at T_g [5], it appears that the approach of T_g is associated with the divergence of a spin-spin correlation length, as is the case in the phase transition of classical ordered systems. In the spin-glass phase, the system is out of equilibrium: as in simple ferromagnets, it is tempting to associate aging with the progressive growth of a typical domain size towards an equilibrium infinite value. However, this simple picture cannot account for all of the experimental observations. In particular, the effect of small temperature cycles (within the spin-glass phase) is rather remarkable [6,7]:

(i) On the one hand, aging at a higher temperature barely contributes to aging at a lower temperature. Said differently (as will be discussed again below), the thermal history at sufficiently higher temperatures is irrelevant. This is at variance with a simple scenario of thermal activation over barriers, where the time spent at higher temperature would obviously help the system find its equilibrium state. Everything happens as if there were strong changes of the free-energy landscape with temperature. This point is suggestive of the "chaotic" aspect of the spin-glass phase that has been predicted from mean-field theory [8] and from scaling arguments in Refs. [9,10].

(ii) On the other hand, interesting memory effects concomitantly appear The state reached by the system at a given temperature can be retrieved after a negative temperature cycle.

In this Letter, we describe some new experiments which reveal in a rather striking way these memory and chaos effects, and we point out their implications for the construction of a real space picture of spin glasses. The results are obtained using a new experimental protocol, which has first been proposed and applied to the metallic Cu:Mn spin glass by one of us [11]. We now develop this approach in a series of measurements on the $CdCr_{1.7}In_{0.3}S_4$ insulating spin glass [12]. The universality of the out-of-equilibrium dynamics in spin glasses is evidenced by the similarity between the results on two very different realizations of spin-glass systems.

In this procedure, we record the ac susceptibility of the sample as a function of temperature. The ac field has a low frequency of $\omega/2\pi = 0.04$ Hz, to allow the relaxation of the susceptibility due to aging to be clearly visible on the scale of several hours. The peak amplitude of the ac field is 0.3 Oe, which is low enough not to affect the properties of the system. We cool the system from above $T_g = 16.7$ down to 5 K at a constant cooling rate of 0.1 K/min, and then heat it back continuously at the same rate. This yields two slightly different curves; the one obtained upon heating (a bit below the other one) is chosen as the *reference curve* and is shown as a solid line in Fig. 1. We then repeat the experiment, but now stop during cooling at an intermediate temperature $T_1 = 12$ K during a certain waiting time $t_{w1} = 7$ h. During t_{w1}, due to aging, both χ' and χ'' relax downwards, by about the same amount; for χ'', however, the relative amount is much larger, which makes the effect more visible, and in the following we concentrate mainly on the out-of-phase component. After the aging stage at T_1, the cooling procedure resumes, and one observes that χ'' and χ' merge back with the reference curve only a few Kelvin below T_1.
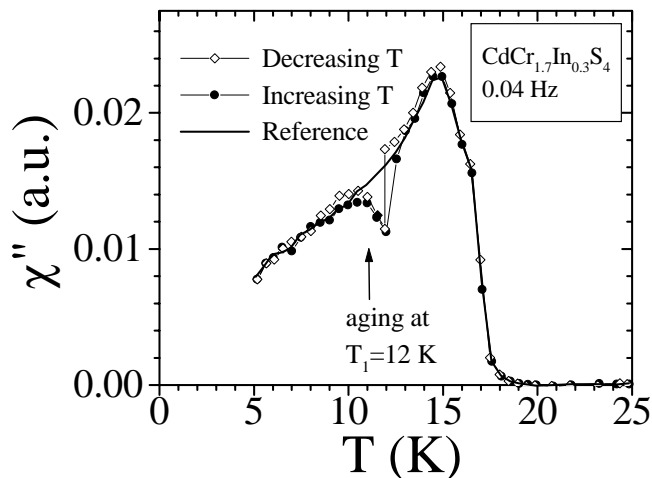
FIG. 1. Out-of-phase susceptibility χ'' of the CdCr$_{1.7}$In$_{0.3}$S$_4$ spin glass. The solid line is measured upon heating the sample at a constant rate of 0.1 K/min (reference curve). Open diamonds: the measurement is done during cooling at this same rate, except that the cooling procedure has been stopped at 12 K during 7 h to allow for aging. Cooling then resumes down to 5 K: χ'' is not influenced and goes back to the reference curve (chaos). Solid circles: after this cooling procedure, the data is taken while reheating at the previous constant rate, exhibiting memory of the aging stage at 12 K.

Thus, aging at $T_1 = 12$ K has not influenced the result at lower temperatures (*"chaos" effect*).

The surprise is that when the sample is reheated at a constant heating rate (i.e., no further stops on the way up), we find that the trace of the previous stop (the dip in χ'') is exactly recovered (see Fig. 1). The memory of what happened at $T_1 = 12$ K has not been erased by the further cooling stage, *even though χ'' at lower temperatures lies on the reference curve*. The system can actually retrieve information from several stops if they are sufficiently separated in temperature. In Fig. 2, we show a "double memory experiment," in which two aging evolutions, one at $T_1 = 12$ K and the other at $T_2 = 9$ K, are retrieved [13]. In the inset of Fig. 2, the result of a similar experiment on a Cu:Mn sample is shown [11].

As discussed above, the cooling rate dependence of the dynamics in spin glasses is largely governed by the chaos effect. For example, it has been shown that there is no difference in the aging behavior if the spin glass has been directly quenched from above T_g or if it has been subjected to a very long waiting pause immediately below T_g [7]. However, the influence of the cooling rate was not quantitatively characterized in systematic measurements, and this point is of a particular interest for the comparison between spin glasses and other glassy systems. We have therefore performed the following experiment. We cool the sample progressively and continuously (in fact, by steps of 0.5 K) from above T_g to 12 K = $0.72T_g$, using three very different cooling rates. The result is shown in Fig. 3. The initial values of χ' and χ'' are indeed
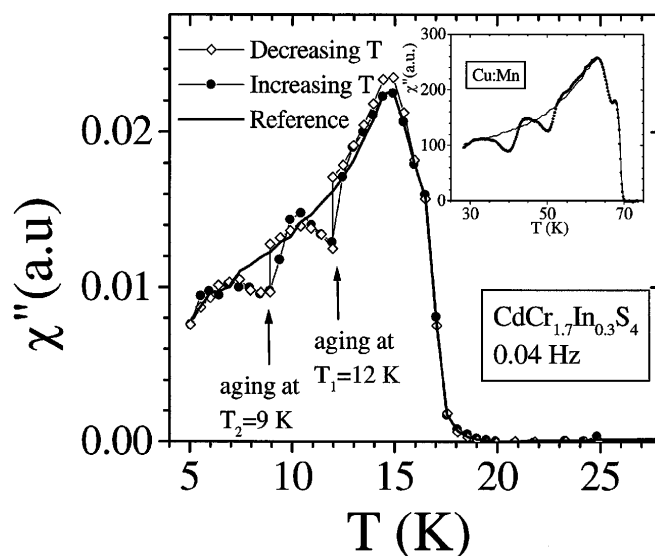

FIG. 2. Same as in Fig. 1 (CdCr$_{1.7}$In$_{0.3}$S$_4$ insulating sample), but with two stops during cooling, which allow the spin glass to age 7 h at 12 K and then 40 h at 9 K. Both aging memories are retrieved independently when heating back (solid circles). The inset shows a similar "double memory" experiment performed on the Cu:Mn metallic spin glass [11].

different: Slower cooling yields a smaller initial value of the susceptibility, a value that is closer to "equilibrium." A small horizontal shift of the curves along the time scale allows the superposition of the three of them; the curves obtained after a slower cooling are somewhat "older."
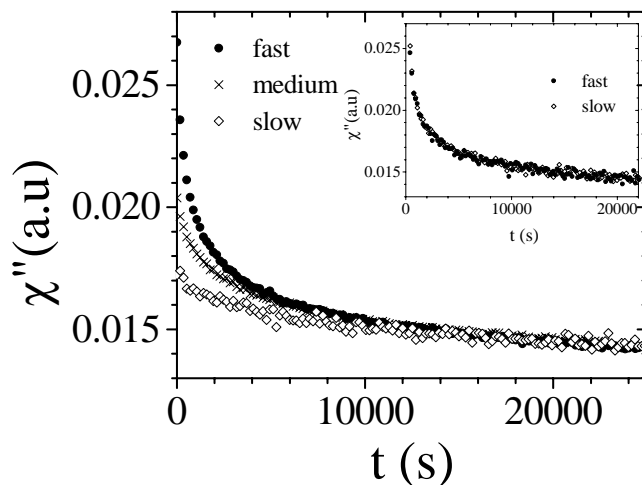

FIG. 3. χ'' relaxation at 12 K as a function of time: effect of the cooling rate on aging. The CdCr$_{1.7}$In$_{0.3}$S$_4$ sample has been cooled from above $T_g = 17$ to 12 K at very different speeds: 2.6 K/min (solid circles), 0.08 K/min (crosses), 0.015 K/min (open diamonds). In the inset, another procedure is used which shows that this cooling rate effect is due only to the last temperature interval: constant rate of 0.8 K/min (solid circles) or 0.08 K/min (open diamonds) from 17 to 14 K, but in both cases rapid quench from 14 to 12 K.

However, all curves are clearly converging towards the same asymptotic value. This behavior contrasts with that observed in systems where thermally activated domain growth is important, for example the dielectric relaxation of the dipole glass $K_{1-x}Li_xTaO_3$ [14]. There, it is found that different cooling rates lead the system to very different apparent asymptotic values of the dielectric constant.

One can show, furthermore, that the cooling rate effect seen in Fig. 3 is due entirely to the last temperature interval, and not at all to the time spent at higher temperatures. We again use different cooling rates from above $T_g = 16.7$ to 14 K, but then we rapidly cool from 14 to 12 K, where the relaxation is measured. In this procedure, despite very different average cooling rates, the last two Kelvin are always crossed at the same speed. The result [in Fig. 3 (inset)] is unambiguous: the obtained relaxation is the same in all cases, for χ' as well as for χ''. Thus, in a spin glass, the only influence of the cooling rate on the aging state is due to the very last temperature interval before reaching the measurement temperature, while, due to chaos effects, the time spent at higher temperatures does not contribute. Again, this strongly contrasts with the case of $K_{1-x}Li_xTaO_3$ alluded to above, where it is the time spent at temperatures near the glass transition that mostly determines the final state after the quench [14].

There have been quite a number of approaches, inspired by Parisi's solution of mean-field models, in which aging can be described in terms of a random walk in the space of the metastable states [15–17]. The memory and chaos effects have been described in terms of a *hierarchical organization of the metastable states as a function of temperature* [7], a picture in which the growth of the free-energy barriers when the temperature decreases could be characterized quantitatively [18]. Although these phase space pictures are very helpful (and have been used very early in the context of spin glasses and glasses), it is obvious that they need to be linked with real space pictures, where the experimental signal can be attributed to certain clusters of spins which flip collectively. Macroscopically, aging means that the system becomes "stiffer" with time, in the sense that the response to a field variation becomes slower and slower with increasing age. Slower response means larger free-energy barriers, and, correspondingly, a larger number of spins to be simultaneously flipped: One is thus naturally led to think in terms of growing "domains" (or "droplets") of correlated spins. The simplest picture based on this idea has been proposed by Fisher and Huse [19] and Koper and Hilhorst [20] in slightly different terms. It is based on the postulate that, at any given temperature below the spin-glass transition, there is only one phase (and its spin reversed counterpart) to be considered, much as in a standard ferromagnet. The difference is of course that all spins do not point in one direction, but arrange in a random (but fixed) way imposed by the interplay between the disordered nature of the interactions and the temperature. One can, however, by convention call one of the phases "up" and the other one "down"; again, as in a ferromagnet, the typical domain size is expected to grow with time, albeit logarithmically slowly since the domain walls are pinned by the disorder.

In principle, this picture should lead to very strong cooling rate effects, which are, as stated above, not those that are observed experimentally in spin glasses (see Fig. 3). However, if one assumes (as suggested by mean-field and scaling arguments) *chaos with temperature*, in the sense that the phase growing at temperature T_1 is not at all the "correct" equilibrium phase for another temperature T_2, the cooling rate dependence can indeed be small since the time spent at a higher temperature does not bring the system any closer to equilibrium. This must be contrasted with random fieldlike systems, where the equilibrium state is the same in the whole low temperature phase, and where cooling rate effects are strong.

The chaotic dependence of the phase with temperature allows one to argue why aging is restarted when the temperature is lowered: The configuration reached after aging at T_1 is, from the point of view of the "T_2 phase," completely random. Correspondingly, new T_2 domains have to grow. The problem, however, comes from the observed memory effect: it shows that the T_2 domains must indeed grow somewhere, but *without destroying the preexisting T_1 domains*. The only possibility is that the T_2 domains *do not nucleate everywhere*, but only around certain favorable nucleation centers, *coexisting* with the previous backbone of T_1 domains. This is, however, in contradiction with the idea that, at any temperature, only one phase (and thus two types of domains) is enough to describe the dynamics of the system completely, since we already require the coexistence of *two* types of domains (T_1 and T_2 domains). The same argument shows that, at the first temperature T_1, the system must actually be in a mixture of all of the different phases encountered between T_g and T_1. Intuitively, it seems clear that, if the system is so fragile to temperature changes, then by the same token it is hard to imagine that other nearby "phases" will not nucleate simultaneously with the nominal phase. In other words, the question is whether, at long times, the "defects" are only domain walls between two well-identified phases (as in the coarsening/droplet picture), or whether these defects are more complicated (branched) objects.

We believe that the memory effect discussed above is incompatible with a picture of the *out-of-equilibrium* dynamics based on one type of domains only. Obviously, this does not exclude the possibility that the *equilibrium* phase is unique; from an experimental point of view, however, the question is not relevant, since the system is never in equilibrium. A similar conclusion has been suggested by "second noise spectrum" experiments [21], and by recent out-of-equilibrium numerical simulations [22].

The construction of a consistent "hierarchical droplet" picture appears completely open and beyond the scope of the present paper. A possibility (discussed in Refs. [4,16]) is that correlations at a given temperature establish themselves progressively, but only over non-compact (i.e., fractal) clusters of spins, which are large enough to be frozen at that temperature, but leave the surrounding sea of spins relatively free. As the temperature is lowered, smaller clusters begin to freeze (and thus provide a new aging signal), while larger clusters are completely blocked (thus leading to the memory effect). Let us note that the idea of "droplets within droplets" has previously been discussed in [23,24] without (to our knowledge) getting to the stage of a more quantitative model. The present experiments, which suggest the coexistence of many different time scales (and thus, presumably, many length scales) may force one to take the idea of fractal droplets more seriously.

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