Eur. Phys. J. B **29**, 19–26 (2002)
DOI: 10.1140/epjb/e2002-00257-y

THE EUROPEAN PHYSICAL JOURNAL B

Aging, rejuvenation and memory effects in re-entrant ferromagnets

V. Dupuis, E. Vincent[a], M. Alba, and J. Hammann

Service de Physique de l'État Condensé, DRECAM, DSM, CEA Saclay, 91191 Gif-sur-Yvette Cedex, France

Received 6 March 2002 / Received in final form 14 June 2002
Published online 17 September 2002 –

Abstract. We have studied the slow dynamics of the ferromagnetic phases of the re-entrant $CdCr_{2x}In_{2-2x}S_4$ system for $0.85 < x \leq 1$ by means of low frequency ac susceptibility and magnetization measurements. Experimental procedures widely used in the investigation of the out-of-equilibrium dynamics of spin glasses (such as the $x = 0.85$ compound) have been applied to search for aging, rejuvenation and memory effects, and to test their dependence on the disorder introduced by dilution of the magnetic ions. Whereas the rejuvenation effect is found in all studied samples, the memory effect is clearly enhanced for increasing dilutions. The results support a description of aging in both ferromagnetic and re-entrant spin-glass phases in terms of hierarchical reconformations of domain walls pinned by the disorder.

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

1 Introduction

Disordered magnetic systems have been intensively studied in the last decades. Among them, the systems with competing magnetic interactions are of particular interest. When the contributions of ferromagnetic and antiferromagnetic interactions are comparable, conventional long range order is not stable and a spin glass behavior is obtained. If the ferromagnetic interactions are preponderant, the system displays a re-entrant behavior, with, for decreasing temperatures, a transition from the paramagnetic phase to a ferromagnetic phase, followed by a transition to a spin glass phase.

Experimentally, in some compounds the amount of ferromagnetic and antiferromagnetic interactions can be continuously varied by changing the concentration of a magnetic ion [1–3]. One such example is the $CdCr_{2x}In_{2-2x}S_4$ thiospinel [2], a frustrated magnetic insulator with ferromagnetic nearest neighbor and antiferromagnetic next nearest neighbor interactions. The $x = 0.85$ compound is a model spin glass in which aging has been extensively studied [4]. The interest in aging phenomena has been revived by experiments on the effect of temperature changes, which have revealed non trivial rejuvenation and memory effects [5], also identified later on in many disordered systems [6–9]. While these features are well accounted for in hierarchical pictures where one assumes a hierarchical organization of the metastable states of the spin glass as a function of temperature [10], their interpretation in the physical space [11] is less clear and is currently the subject of many efforts, both on the theoretical and experimental sides [12–16].

In this paper, we present a comparative study of spin-glass type phenomena, and in particular of the rejuvenation and memory phenomena, in the ferromagnetic phases of the $CdCr_{2x}In_{2-2x}S_4$ system for several concentrations x of the magnetic ions ($x > 0.85$). Our motivation for studying these ferromagnetic phases is the fact that since they are conceptually simpler than spin glasses (spins are organized in domains), the rejuvenation and memory phenomena should be more easily described in terms of geometrical spin arrangements. This argument already led us to study the ferromagnetic phase of the weakly diluted $x = 0.95$ compound [16] in which we could observe some rejuvenation and memory effects. The latter were discussed in terms of pinning and reconformations of ferromagnetic domain walls in the dilution-induced disorder. The work that we present here, involving the study of a more diluted sample ($x = 0.90$), complements the previous investigation and provides further insights on the role of disorder in these out-of-equilibrium phenomena.

The paper is organized as follows. First, we present the phase diagram of the $CdCr_{2x}In_{2-2x}S_4$ system, the general behavior of the magnetization and the low frequency ac susceptibility of the studied samples with $x > 0.85$. Then we analyse the results of isothermal ac susceptibility relaxations at various temperatures both in the ferromagnetic phases and in the re-entrant spin glass phases. This allows us, for example, to highlight the effect of disorder on isothermal aging. We describe experiments in which we

[a] e-mail: vincent@drecam.saclay.cea.fr

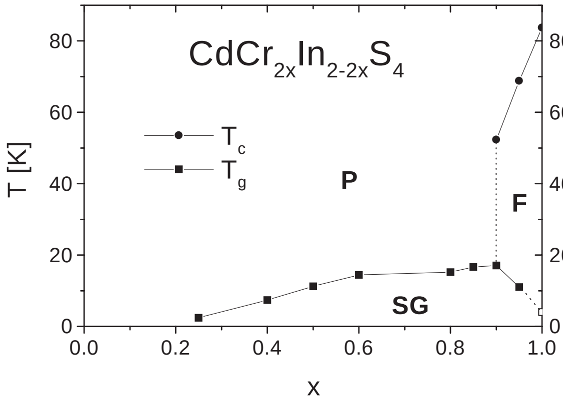

Fig. 1. Phase diagram of the $CdCr_{2x}In_{2-2x}S_4$. The points corresponding to $x < 0.85$ are extracted from [2]. For $x > 0.85$, T_c corresponds to the inflection point of the increase of the out-of-phase susceptibility $\chi''(T)$ at low frequency when cooling from high temperatures. Same convention for T_g. The open square for the $x = 1.00$ sample corresponds to the small χ'' peak observed at low temperatures (see text).

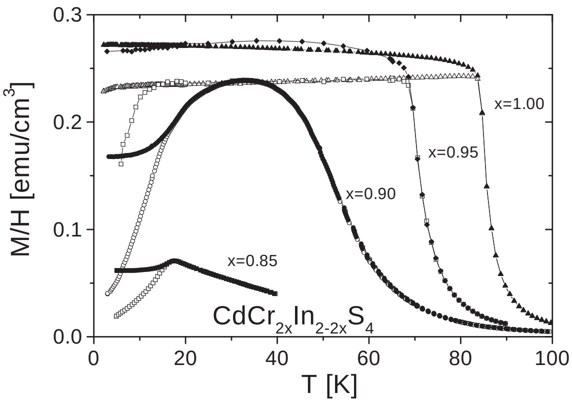

Fig. 2. ZFC (open symbols) and FC (filled symbols) magnetizations divided by the amplitude of the probing field *vs.* temperature of the $CdCr_{2x}In_{2-2x}S_4$ system, for various concentrations $0.85 \leq x \leq 1$.

have investigated the effect of temperature changes on aging. The comparison of the results for different concentrations x shows that the ability of a disordered ferromagnet to memorize aging at a given temperature is strongly dependent on the disorder strength. Whereas for weak disorder ($x = 0.95$), the memory of an isothermal aging is easily erased by a short low temperature excursion (below the temperature at which the system was aged) [16], for stronger disorder ($x = 0.90$), this memory is preserved and can only be erased by an excursion down to the re-entrant spin glass phase. Finally, we discuss the whole set of results in terms of pinning and reconformations of the ferromagnetic domain walls in their disordered environment and raise the question of the relevance of this wall dynamics for explaining the rejuvenation and memory phenomena in spin glasses.

2 Characterization of the system

The investigated samples are chromium thiospinels of formula $CdCr_{2x}In_{2-2x}S_4$ for $x = 1.00$, $x = 0.95$, $x = 0.90$ and $x = 0.85$ prepared by M. Noguès at the CNRS Meudon-Bellevue laboratory. They have been well characterized in the past by various techniques [2] including neutron diffraction [17] and display behaviors ranging from pure ferromagnet, to re-entrant ferromagnet and spin glass for decreasing x. The samples are polycrystalline powders and the measurements reported here were performed with a commercial *Cryogenic*^{Ltd} S600 SQUID magnetometer.

2.1 Structure and phase diagram

The phase diagram of the $CdCr_{2x}In_{2-2x}S_4$ is shown in Figure 1. The pure compound $CdCr_2S_4$ is a normal spinel, *i.e.* all the Cr^{3+} ions occupy octahedral (B) sites of the lattice whereas the tetrahedral (A) sites are occupied by Cd^{2+} ions only. The Cr^{3+} ions are in a $3d^3$ electronic state, leading to a magnetic moment of spin only type with $S = \frac{3}{2}$. The main magnetic interactions between a Cr^{3+} ion and its six nearest neighbors are positive [17] ($J_1/k_B = 13.25 \pm 0.12$ K) and induce ferromagnetism below $T_C = 84.8$ K. The existence of super exchange antiferromagnetic interactions [18] between the twelve third nearest neighbors ($J_3/k_B = -0.915 \pm 0.015$ K) induces frustration in the magnetic lattice that can be quantified by the weighted interaction ratio $R = \frac{z_3 \times J_3}{z_1 \times J_1} = -0.138$, to be compared to the critical ratio for the stability of ferromagnetism $R_C = -0.25$ in the spinel structure [19]. The dilution of the Cr^{3+} ions by non magnetic In^{3+} ions enhances this frustration as the antiferromagnetic couplings *via* the In^{3+} orbitals J_L are amplified by a factor $R_{In} = J_L/J_3 \sim 9.0$ [20,21]. This large enhancement explains quantitatively the absence of a percolating ferromagnetic cluster for dilutions $(1 - x)$ greater than about 15% [2]. The dilute samples ($0.15 < x < 0.85$) therefore undergo a unique phase transition towards a spin glass phase at low temperatures.

For higher concentrations in chromium ions ($0.85 < x < 1$), the sequence of phase transitions upon cooling down is as followed: a para-ferromagnetic phase transition first occurs at a critical temperature T_c, followed by the onset of spin glass type irreversibilities, at a lower critical temperature T_g. This phenomenon is usually referred to as reentrance.

2.2 Field cooled and zero field cooled magnetizations

Figure 2 displays the Field Cooled (FC) and Zero Field Cooled (ZFC) magnetizations (normalized to the value of the applied field) *vs.* temperature for the studied samples. The ZFC measurements have been performed after cooling the samples in zero field (at ~ 80 mK/s) to 3 K, applying the field at 3 K, and recording the data upon slowly re-heating (at $\sim$3 mK/s). The FC data have been taken in the same manner (but cooling *with the field on*).

The amplitude of the small probing field was typically of ∼10 Oe in order to remain in the linear response regime. We have applied demagnetizing field factor corrections to the data to take into account the different geometries of the samples [22]. The results are presented for a spherical shape factor.

While at high temperatures, all samples are paramagnetic, two types of behavior appear at lower temperatures.

In the pure ($x = 1.00$) and the weakly diluted ($x = 0.95$) samples, a transition from the disordered paramagnetic state to a long range ferromagnetic ordered state is signalled by a sharp increase of both the ZFC and FC magnetizations above the Curie temperature T_c. This transition is confirmed by neutron diffraction measurements which evidence spontaneous magnetization, magnetic Bragg peaks and characteristic spin waves [17]. Below T_c, magnetic irreversibilities are observed in the separation of the FC and ZFC curves: the magnetization becomes history dependent. The slow increase of the FC magnetization as the temperature is decreased is accounted for by a small extra polarization of the sample when cooled in a field and can be related to the temperature dependence of the spontaneous magnetization. On the other hand, the plateau of the ZFC magnetization, at a level equal to the inverse of the demagnetizing field factor, indicates that the spins are well organized in ferromagnetic domains.

The behavior of the strongly diluted ($x = 0.90$) sample is markedly different. The increase of the FC and ZFC magnetizations upon cooling from the paramagnetic phase is less pronounced than in the less diluted samples. Neutron diffraction measurements reveal only short range ferromagnetism with no magnetic Bragg peaks nor a true divergence of the spin-spin correlations [17]. We can still define for convenience a “transition” temperature T_c at the inflection point of the magnetization increase but it is important to note that this is not a true transition to a long range ordered state. With this definition, we see that, just below T_c, no clear separation of the FC and ZFC curves is found within experimental accuracy. Nevertheless, we observe a rounded magnetization plateau at a level corresponding to the expected demagnetizing field factor which suggests that the magnetic moments in the sample are still organized in ferromagnetic domains.

At lower temperatures, while nothing special happens in the pure sample, the FC magnetization of the $x = 0.95$ and $x = 0.90$ samples decreases and tends to saturate. In parallel, we observe a sharp drop of the ZFC magnetization. This change of behavior corresponds to the transition from the ferromagnetic state to the strongly frozen re-entrant spin glass state and is to be compared with the reference spin glass behavior of the more diluted $x = 0.85$ sample also shown in Figure 2.

2.3 Low frequency ac susceptibility

To complete the previous analysis, we have measured the low frequency ac susceptibility $\chi = \chi' - \mathrm{i}\chi''$ of the samples as a function of temperature. Here again, the data has been taken while re-heating at ∼ 1–2 mK/s, after cooling to 3 K at ∼ 80 mK/s, using a probing field of weak amplitude (typically 1.7 Oe peak value) in order to remain in the linear response regime. We have used the paramagnetic phase response (assuming $\chi'' = 0$) to correct a slight frequency dependent phase shift in the detection setup. In the following, we focus on the out-of-phase component χ'' which is directly related to the dissipation processes.

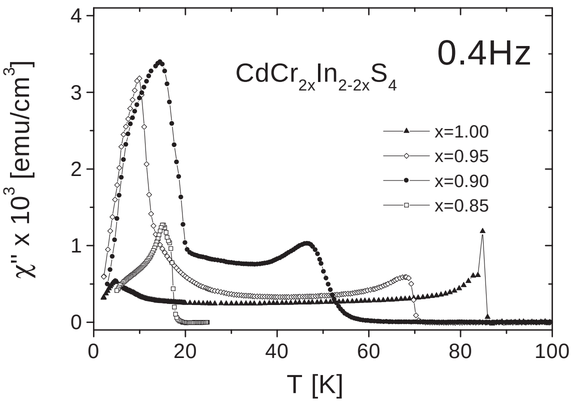

Fig. 3. Temperature dependence of the out-of-phase susceptibility χ'' of the $CdCr_{2x}In_{2-2x}S_4$ system at 0.4 Hz, for various concentrations $0.85 \leq x \leq 1$.

Figure 3 shows χ'' at 0.4 Hz *vs.* temperature for all the studied samples. The onset of ferromagnetism is signalled by a sharp increase in χ'' when cooling from high temperatures. This increase in the dissipation indicates that the characteristic relaxation times of the system have reached a value comparable to the macroscopic measurement time (equal here to the inverse of the ac field frequency). This is the standard scenario expected at a second order phase transition where the average relaxation time diverges. We can define a transition temperature T_c at the inflection point of the χ''-increase. We find that T_c slightly decreases for decreasing frequencies, in agreement with the expectation from dynamic scaling arguments. Below T_c, χ'' keeps a non zero value, roughly temperature independent in the whole ferromagnetic region meaning that there are still dissipation processes at the time scale of the experiments in the ferromagnetic phase of the studied samples, even in the pure one. The stronger the dilution, the higher this plateau value of χ''. The primary effect of an increase in the disorder strength is therefore an increase of the level of low frequency dissipation.

At lower temperatures, sharp peaks with a critical frequency dependence [23,24] are observed in the out-of-phase susceptibility χ'' of the diluted samples ($x = 0.95$ and $x = 0.90$) and signal the transition from the ferromagnetic state to the re-entrant spin glass state. The corresponding transition temperatures estimated from the inflection points at low frequency (0.4 Hz) are about the same as the ones at which strong irreversibilities are observed in FC-ZFC measurements. Curiously, a small peak in χ'' is also observed at low temperatures in the pure $x = 1.00$ sample where no existence of a re-entrant spin glass phase has ever been reported. However, while

Table 1. Amplitude of χ'' relaxation at 0.4 Hz after slow cooling (∼1 mK/s), in absolute and relative values, for the $x = 0.95$ and the $x = 0.90$ $CdCr_{2x}In_{2-2x}S_4$ samples after a waiting time $t_w = 15\,000$ s.

T [K]	$\Delta\chi'' \times 10^5$ [emu/cm^3]	$\Delta\chi''/\chi'' \times 100$
$x = 0.95$[a]		
67 ($0.95T_c$)	2.4	4.7
40 ($0.57T_c$)	1.0	3.1
8 ($0.80T_g$)	26.2	11.6
5 ($0.50T_g$)	8.6	5.3
$x = 0.90$[b]		
42 ($0.84T_c$)	3.4	3.8
30 ($0.60T_c$)	1.1	1.6
17 ($0.95T_g$)	16.0	6.7
10 ($0.56T_g$)	29.0	12.9

[a] $T_c = 70$ K and $T_g = 10$ K.
[b] $T_c = 50$ K and $T_g = 18$ K.

the frequency dependence of the peaks observed in the diluted samples is characteristic of critical slowing down (in the frequency range explored [0.04 Hz–8 Hz]), we have found that the position of the peak in the pure sample has an Arrhenius type frequency dependence, evocative of a dynamical freezing of the spins. Even if the origin of this peak is at this moment not completely clear, it does not seem to correspond to a phase transition as in the diluted samples.

3 Slow dynamics and aging at constant temperature

We have investigated the isothermal slow dynamics and aging of the $CdCr_{2x}In_{2-2x}S_4$ samples in the following way. The samples were cooled from their paramagnetic phase down to various successive aging temperatures T_m, in both the ferromagnetic and the re-entrant spin glass phases (with a cooling rate of typically 1 mK/s). At each fixed aging temperature, the time evolution of the ac susceptibility was recorded for three different frequencies: 0.04 Hz, 0.4 Hz and 4 Hz.

In all cases, we observe a slow time decay (aging) of the ac susceptibility $\chi(\omega, t)$ towards an asymptotic frequency dependent value $\chi_0(\omega)$ (stationary susceptibility). The relaxation is more important in relative value on the out-of-phase component than on the in-phase component and in the following we focus on χ''.

The influence of temperature on aging is illustrated in Table 1, in which we report, for the $x = 0.95$ and $x = 0.90$ samples, the amplitude of the χ'' relaxations (in absolute and relative values) at fixed frequency, observed at various temperatures and for a given waiting time $t_w = 15\,000$ s. The trend is the same for both samples. The relaxations are always weaker in the ferromagnetic phase than in the re-entrant spin glass phase. Moreover, in the ferromagnetic phase, the relaxation is mainly important near T_c and tends to disappear in the experimental background noise at lower temperatures.

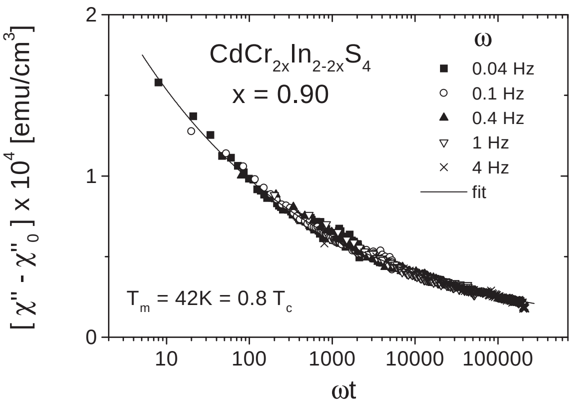

Fig. 4. Scaling plot of the χ'' relaxation curves of the $x = 0.90$ sample after a quench (∼80 mK/s) to $T_m = 42$ K as a function of ωt and for frequencies of the ac field ranging from 0.04 Hz to 4 Hz. The asymptotic stationary part χ''_0 has been subtracted. All the curves lie on an unique master curve which can be nicely fitted by a power law with a small exponent −0.2.

Concerning the frequency dependence of the relaxation, we observe that the lower the frequency ω, the greater the relaxation amplitude observed in the experimental time window. This result is in agreement with a previous investigation of the $x = 0.95$ sample [16], where a qualitative ωt scaling (characteristic of spin glasses [4]) of the non stationary part of the susceptibility was found both in the re-entrant spin glass and in the ferromagnetic phases. Here, we have studied this scaling behavior of the isothermal ac susceptibility relaxation in a more quantitative way. We have recorded the relaxation of the ac susceptibility of the $x = 0.90$ sample following a quench (∼80 mK/s) to a temperature $T_m = 42$ K in the ferromagnetic phase for 8 frequencies ω of the ac field in the range [0.04 Hz–4 Hz]. Figure 4 shows the resulting relaxation curves for the out-of-phase component χ'' as a function of the reduced variable ωt. As we can see in the figure, the scaling works pretty well over nearly five decades. All the relaxation curves, after subtraction of a constant which corresponds to the stationary part $\chi''_0(\omega)$, lie on a unique master curve which is a function of ωt and can be nicely fitted by a power law with an exponent of order −0.2 (same value as commonly observed in spin glasses [4]).

4 Rejuvenation and memory effects

In conventional spin glasses, such as the $x = 0.85$ compound, it is known that decreasing the temperature from T to $T - \Delta T$ in the spin glass phase, after having isothermally aged the sample at T, strongly restarts the dissipation processes. This is the rejuvenation effect [5], the system seems to forget its previous equilibration stage at T. For a sufficiently large ΔT, the system rejuvenates completely and the relaxation of the ac susceptibility at $T - \Delta T$ is identical to that obtained after a direct quench from the paramagnetic phase. The other striking feature happens when the system is re-heated to the initial temperature T after a given low temperature history. No

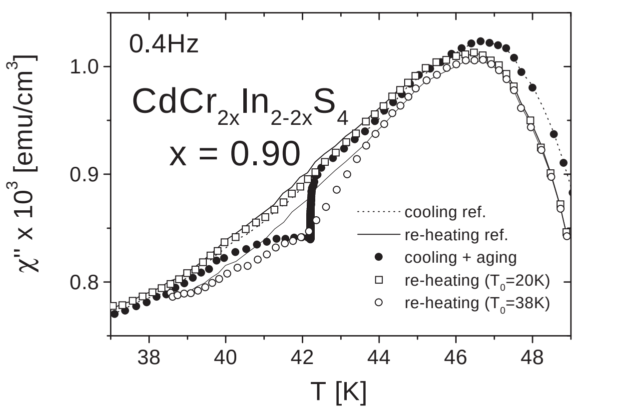

Fig. 5. Aging, rejuvenation and memory effects on χ'' *vs.* temperature in a single stop experiment at $T_m = 42$ K (see details about the procedure in the text). Aging is evidenced by the downward relaxation of χ'' while the rejuvenation effect corresponds to the increase of χ'' back to the reference when cooling is resumed down to T_0. Upon re-heating, a clear memory dip centered around T_m is seen for $T_0 = 38$ K. In contrast, for $T_0 = 20$ K, no memory is found.

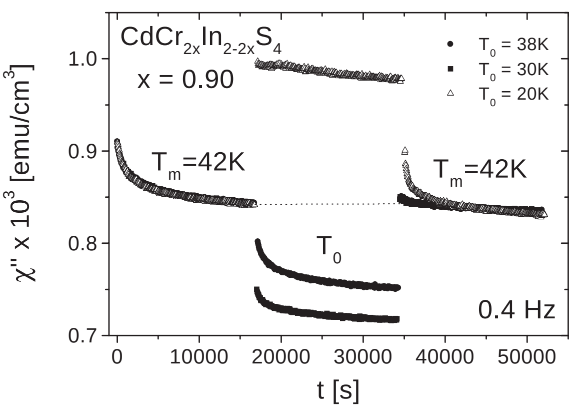

Fig. 6. Aging, rejuvenation and memory effect on χ'' *vs.* time in temperature cycling experiments. After the quench from high temperature down to $T_m = 42$ K, χ'' is relaxing downwards with time. Cooling down after 17 000 s to T_0, induces a rejuvenation effect evidenced here by the restart of relaxation processes for $T_0 = 38$ K and $T_0 = 30$ K, not for $T_0 = 20$ K. When re-heating to T_m, the relaxation of χ'' is the continuation (memory) of the one that occured before the cycle for $T_0 = 38$ K and $T_0 = 30$ K. In contrast, for $T_0 = 20$ K a strong restart of aging processes, analogous to a quench from high temperature is seen meaning that no memory is found is this case.

restart of aging is found and the relaxation is the exact continuation of the one that occurred before going to low temperatures. This is the memory effect [5], aging at $T - \Delta T$ had no influence on aging at T. These effects have been interpreted as an evidence for a hierarchical organization (tree-like for example) as a function of temperature of the metastable states of the spin glass [10].

In order to reach a better understanding of these non trivial rejuvenation and memory effects in terms of geometrical spin arrangements, we have previously investigated the pure and weakly disordered ferromagnetic phase of the $x = 1.00$ and $x = 0.95$ compounds [16]. We found clear rejuvenation effects coexisting with a very weak memory of aging, which was easily erased by an excursion at lower temperature. Here, we report similar investigations of the rejuvenation and memory phenomena in the short range ferromagnetic phase of the $x = 0.90$ sample. Our results shed light on the role of disorder in the rejuvenation and memory phenomenology.

In a first experiment, we have studied the rejuvenation and memory phenomena in the temperature dependence of the ac susceptibility. Figure 5 shows the out of phase susceptibility χ'' of the $x = 0.90$ sample at 0.4 Hz *vs.* temperature. All along the experiment a sweeping rate $|\mathrm{d}T/\mathrm{d}t|$ of $\sim$3 mK/s was used. The sample was cooled down to a measurement temperature $T_m = 42$ K where it was isothermally aged during 8 h (full symbols). After this aging stage, the cooling was resumed down to a final temperature T_0 (two cases, $T_0 = 38$ K and $T_0 = 20$ K, were studied) and finally, the sample was slowly re-heated from T_0 to the paramagnetic phase (open symbols). For comparison, a reference curve, recorded with the same protocol but without the aging stage, is also plotted on this graph (dotted and solid lines for cooling and re-heating).

Aging is clearly visible in the downward relaxation with time of χ'' at T_m. When cooling is resumed after the aging stage at T_m, a rejuvenation effect is evidenced by the increase of χ'' back to the reference cooling curve. Considering now the re-heating process, we see that in the experiment with a short excursion down to $T_0 = 38$ K, a memory of the aging at T_m is found as χ'' departs from the heating reference curve on approaching T_m and displays a characteristic dip around T_m before merging back with the reference curve. In contrast, in the experiment with a longer excursion down to $T_0 = 20$ K, the re-heating curve remains superimposed onto the re-heating reference curve and no memory dip is found. Let us mention that in another experiment made with the same protocol but with $T_0 = 30$ K, we observed a memory effect identical to the one observed with $T_0 = 38$ K.

In a second experiment, we have studied the rejuvenation and memory phenomena as a function of time. Figure 6 shows the out of phase susceptibility χ'' at 0.4 Hz *vs.* observation time measured with a negative temperature cycling protocol: the sample was quenched from high temperatures down to the measurement temperature $T_m = 42$ K, aged there during $t_1 = 17\,000$ s, cooled then to T_0, aged at this temperature another $t_2 = 17\,000$ s then heated back to T_m and finally aged there again for $t_3 = 17\,000$ s. Three values were used, $T_0 = 38$ K, 30 K and 20 K.

Just after the quench, χ'' relaxes downwards with time (it is the same kind of relaxation as in the exp. of Fig. 5). Cooling from T_m to T_0 induces a restart of aging for $T_0 = 38$ K and $T_0 = 30$ K: it is the rejuvenation effect observed here as a function of time. Heating back to T_m reveals the memory of the previous aging stage at T_m: except for a short transient regime, the relaxation at T_m is the exact continuation of the one that occurred before the cycle. However, in the experiment going down to $T_0 = 20$ K (that is, in the border region between the ferromagnetic and spin-glass phases), the relaxation at $T_0 = 20$ K does not show any upturn signing up a rejuvenation effect. We do not have a reference curve of a relaxation at 20 K after a direct quench for comparison; further studies in this temperature region would be of interest. A strong restart of aging is observed when coming back to T_m from $T_0 = 20$ K after the negative cycle (in agreement with the absence of memory in the experiments of Fig. 5). We have checked that this restart of aging is not a long transient which would yield to a memory at a longer observation time as the data points cannot be superposed onto a reference isothermal relaxation curve.

In summary, the experiments reported above show that the rejuvenation effect is a characteristic feature of disordered ferromagnetic phases, as was already observed in [16,25]. It is not influenced by the amount of disorder. In contrast, the ability of a ferromagnet to store and retrieve a memory of a previous aging strongly depends on the disorder strength. Whereas the memory is weak and rapidly erased by a low temperature excursion for the weakly disordered $x = 0.95$ sample, it is much more robust in this more disordered $x = 0.90$ sample. For the latter, the memory of aging is not much affected by a low temperature excursion below T_m as long as the lowest temperature reached during the experiment remains sufficiently above the re-entrant spin glass transition temperature.

While performing the previous experiments on the $x = 0.90$ sample, we observed a surprising behavior of the ac susceptibility which may be related to the mechanism responsible for the erasure of the memory. When recording the cooling/re-heating reference curves of Figure 5 for $T_0 = 20$ K, we found that in the region [20 K,35 K], the cooling curve was always below the re-heating curve. This means that the state which progressively develops in the short range ferromagnetic phase while cooling from the paramagnetic phase is less dissipative than the one obtained when re-heating from the low temperature re-entrant spin glass phase. This rather unusual phenomenon is displayed in Figure 7. We have investigated the aging properties of these two states which we call for convenience the cooled state and the re-heated state. Figure 7 shows χ'' *vs.* temperature at 0.4 Hz measured along the following procedure: slow cooling from high temperatures to 3 K with an aging stop at 30 K during 4 h (full circles), slow cooling to 3 K and re-heating with an aging stop at 30 K for 6 h (open circles) during the re-heating.

In the figure, the hysteresis between cooling and re-heating is clearly evidenced. If we consider now the relaxations, we find a larger relaxation amplitude in the re-heated state than in the cooled state. This extra dissipation gained in the vicinity of the re-entrant spin glass transition indicate some incompatibility between the short range ferromagnetic correlations that develop while cooling the sample from the paramagnetic phase and those that develop in the vicinity of the re-entrant spin glass phase. The region where we observe a large hysteresis between cooling and re-heating may be a region where

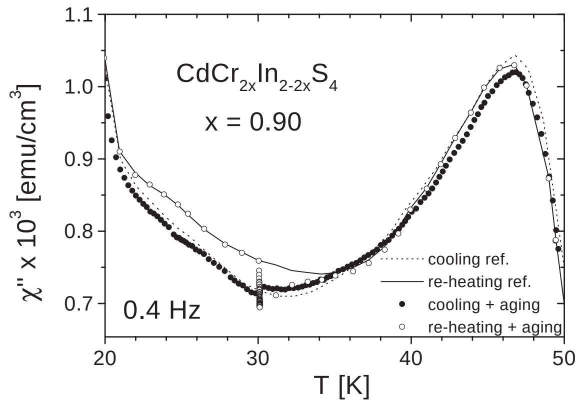

Fig. 7. Relaxation of χ'' during a waiting time t_w at $T_m = 30$ K for the $x = 0.90$ sample. Filled circles: the relaxation is made during the cooling, $t_w = 4$ h. Open circles: the relaxation is made during the re-heating after continuous cooling down to $T_0 = 20$ K, $t_w = 6$ h. The dotted and solid lines correspond to cooling and re-heating reference curves (the temperature sweeping rates are of order 1−4 mK/s). The (downwards) relaxation amplitude is larger in the re-heated state than in the cooled state. The cooling and re-heating reference curves display the same trend as the re-heating curve is clearly above the cooling one in the region [20 K–35 K].

the two types of correlations coexist in proportions that depend on the thermal history. In the cooled state, this region corresponds mainly to a ferromagnetic pattern whereas in the re-heated state, it corresponds mainly to a spin glass pattern. We believe that the development of these spin glass correlations at low temperature is responsible for the erasure of the memory of any previous aging in the short range ferromagnetic state.

5 Discussion

The series of experiments presented in this paper illustrates how the static and dynamic properties of a ferromagnet, the $CdCr_2S_4$ are influenced by the introduction of site disorder. In a non-disordered ferromagnet, the susceptibility is mainly related to the domain pattern in the sample *via* the demagnetizing field, and the out-of-phase ac susceptibility gives more specific informations on the stiffness and the density of the domain walls. In this case, dynamic properties simply reflect the growth of the domains with time, which mainly proceeds by wall displacements. This phenomenon which is only related to the surface tension of the walls is a fast process.

The introduction of pinning disorder, such as site dilution or structural defects, changes drastically this picture [26]. The frustration which arises from the competition between the elastic energy associated with the wall deformation (which tends to favor a smooth interface) and the pinning energy associated with the disorder (which favors a rough interface) results in a drastic slowing down of dynamical processes. A domain wall has now many configurations between which it can make thermally activated hops. The wall displacements hindered by the frustration then proceed by reconformations of some parts of the wall which roughly correspond to jumps from one pinning site to another. In the presence of a driving magnetic field which forces the domain growth (a field ramp for exemple), this phenomenon is commonly known as the Barkhausen noise. In contrast, without any driving field, the overall domain growth can be extremely slow and one can observe the slow reconformations of the domain walls in a static pinning disorder without any notable growth of the domains.

Actually, the dynamics of pinned elastic interfaces has many similarities with that of spin-glasses. Theoretical arguments [26] suggest that their energy landscape is hierarchical with small length scale l reconformations corresponding to hoping of small energy barriers $E(l) \sim \Upsilon l^{\theta}$. In spin glasses, the rejuvenation effect upon cooling and the memory when re-heating are usually ascribed to a hierarchical organization of the metastable states [10]. When the temperature is lowered, the system remains in a deep well (memory effect), while new subwells appear, inducing new aging processes (rejuvenation effect). A similar hierarchical scheme is expected for the dynamics of a pinned domain wall. As time elapses at fixed temperature, the wall equilibrates on longer and longer length scales (aging), while shorter length scale processes are fluctuating in equilibrium. When the temperature is lowered, the conformation of the wall on large length scale freezes (allowing for the memory upon re-heating) and the equilibration now restarts on shorter length scales, which are no longer in equilibrium because their Boltzmann weight has changed (rejuvenation). In parallel with these hierarchical reconformation processes of the walls, the average domain size may grow with time. In that case, the disorder seen by the domain wall is changing with time and obviously, this net motion tends to erase the memory of the reconformations.

The above scenario accounts well for the observed behavior in the pure $x = 1.00$ and weakly diluted $x = 0.95$ samples [16], and also that of some disordered dielectric crystals [27]. In both $x = 1.00$ and $x = 0.95$ samples, aging and rejuvenation are clearly found in parallel with a weak memory of aging which is erased by domain growth during a long excursion to lower temperatures. The case of the more disordered $x = 0.90$ sample, described here in details, is interesting since it shows that the memory effect is stabilized by the disorder and supports the scenario described above. The stronger the pinning of the domain walls, the slower their overall displacement, the better the memory of the domain wall reconformations. This memory can nevertheless be erased by a low temperature excursion close to the reentrant spin glass transition, which indicates that the spin glass order establishing at low temperature is incompatible with the short range ferromagnetic order developing above the re-entrant spin glass transition temperature.

It is tempting to extend this scenario of hierarchical wall reconformations in a pinning disorder to the case of spin glasses, because it provides us with a simple explanation for the rejuvenation and memory phenomena in the real space of spins without invoking temperature chaos arguments [11,14] (see [13,28]). In a spin glass, the nature of these walls is not as clear as in a ferromagnet, and remains an open question. They may be related to the non trivial sponge-like excitations recently discussed from the results of 3d spin-glass simulations [29]. It would be interesting to test the effect of temperature changes on such excitations.

Finally, let us mention that our results are consistent with other investigations on the re-entrant ferromagnet $(Fe_{0.20}Ni_{0.80})_{75}P_{16}Al_3$ and also on the disordered manganite $Y_{0.7}Ca_{0.3}MnO_3$ [25]. The former sample resembles our $x = 0.90$ compound regarding the temperature dependence of the susceptibility (rounded plateau in the ferromagnetic region); the absence of memory effect is probably due to the proximity of the re-entrant spin glass phase transition.

6 Conclusion

In this article, we have studied the slow dynamics of the ferromagnetic phases of the $CdCr_{2x}In_{2-2x}S_4$ system for $x = 1.00$, $x = 0.95$ and $x = 0.90$ by means of ac susceptibility and magnetization measurements. We found that increasing the disorder strength induces a slowing down of the ac response of the ferromagnetic phases resulting in an increase of the low frequency dissipation. Using relaxation procedures developed for the study of the out of equilibrium dynamics of spin glasses, we have found an aging behavior of the low frequency ac susceptibility both in the ferromagnetic phases and in the re-entrant spin glass phases, with the same qualitative features as in conventional spin glasses.

We have searched for rejuvenation and memory phenomena in the ferromagnetic phases of the studied samples. Whereas the weakly disordered $x = 0.95$ compound displays only a weak memory of aging (rapidly erased by an excursion at lower temperatures), the more disordered $x = 0.90$ compound shows a more robust memory of aging which can only be erased by the growth of spin glass correlations in the vicinity of the re-entrant spin glass transition. Our results support a scenario in which, aging, rejuvenation and memory are described in terms of hierarchical reconformations of elastic walls in a random pinning disorder. In this picture, the memory is contained in the large length scale conformation of the walls and is erased by the growth of the domains. Our results show that an increase in the disorder strength tends to reduce the domain growth and allows to observe clear rejuvenation and memory effects. The extension of this wall reconformation scenario to the case of spin glasses raises interesting questions such as the nature of the domains and the walls.

We are grateful to X. Colson for his active participation to this work. We also wish to thank J.-P. Bouchaud, F. Bert, D. Hérisson, M. Ocio for stimulating discussions during this work, L. Berthier and M. Noguès for important remarks on the manuscript, and L. Le Pape for his technical assistance.

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