Aging phenomena in spin glass and ferromagnetic phases: domain growth and wall dynamics

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PACS. 02.75.50.Lk – Spin glasses and other random magnets.
PACS. 03.75.10.Nr Spin-glass and other random models.
PACS. 04.75.40.Gb Dynamic properties (dynamic susceptibility, spin waves, spin diffusion, dynamic scaling, etc.).

Abstract. – We compare aging in a disordered ferromagnet and in a spin glass, by studying the different phases of a reentrant system. We have measured the relaxation of the low-frequency ac susceptibility $\chi$, in both the ferromagnetic and spin-glass phases of a $CdCr_{1.9}In_{0.1}S_4$ sample. A restart of aging processes when the temperature is lowered (‘chaos-like’ effect) is observed in both phases. The memory of previous aging at a higher temperature can be retrieved upon re-heating, but in the ferromagnetic phase it can rapidly be erased by the growth of ferromagnetic domains. We interpret the behaviour observed in the ferromagnetic phase in terms of a combination of domain growth and pinned wall reconformations, and suggest that aging in spin glasses is dominated by such wall reconformation processes.

Introduction. – In a spin glass, cooling below $T_g$ is the starting point of the slow ‘aging’ evolution. The ac susceptibility relaxes downwards with time after the initial quench (the ‘age’) (see e.g. [1] and references therein). A natural basis for the interpretation of aging is the idea of a slow domain growth of a spin-glass type ‘ordered phase’, as was developed in [2, 3]. However, several non-trivial experimental features of aging in spin glasses are hard to interpret within a simple domain growth scenario [4, 5, 6]. Namely, after aging at a given temperature, aging is reinitialized by a further temperature decrease [7], and the memory of previous aging can be retrieved upon heating back.

On the other hand, domain growth phenomena should be more directly relevant to the case of ferromagnets. Our motivation in the present work is to compare aging in these systems and in spin glasses, having in mind the discussion of a consistent picture of aging in the latter, more complex case.

$^{(1)}$ This has been called ‘chaos’, by analogy with the theoretical scenario of [3]. However, as we discuss below, this effect can be related to the temperature variation of Boltzmann weights, and a better word might be ‘rejuvenation’.

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General features of the reentrant system. –

The CdCr$_{2-x}$In$_x$S$_4$ chromium thiospinel insulator [8] has been extensively characterized by neutron diffraction measurements [9]. For $x = 0$, it is a ferromagnet ($T_c = 84.5K$) with ferromagnetic nearest neighbour interactions and antiferromagnetic next-nearest neighbour interactions. For $x \geq 0.10$, the ferromagnetic phase disappears, and a spin-glass phase appears at $\sim 20K$ [8, 10]. The $x = 0.15$ system is a well studied spin-glass compound [1, 6]. In the intermediate region $0 < x < 0.10$, the ferromagnetic phase is followed at lower temperature by a reentrant spin glass phase.

The main sample of our present study is the $x = 0.05$ compound (of the same batch as in [9]). Fig.1 shows its general magnetic features. The dc susceptibilities $\chi_{FC}$ and $\chi_{ZFC}$ have been measured along the usual field-cooled (FC) and zero-field cooled (ZFC) procedures. In the high temperature region, the dc and ac susceptibilities follow a paramagnetic behaviour. At $T_c \approx 70K$, they rise up abruptly, reaching a plateau. $\chi_{ZFC}$ and $\chi'$ at low frequency are at about the same level, $\sim 15\%$ below the $\chi_{FC}$ value. The ac out-of-phase susceptibility $\chi''$ peaks around $T_c$, and remains non-negligible in the ferromagnetic plateau region, confirming the existence of magnetic irreversibilities. At $T_g \sim 10K$, $\chi''$ shows a strong peak, while $\chi'$ and $\chi_{ZFC}$ decrease with decreasing temperature. In this low-temperature phase, spin glass features coexist with those of a regular ferromagnetic state (non zero magnetisation, Bragg peaks, spin waves [8, 9]). The dc plateau susceptibility in the ferromagnetic phase, which corresponds approximately to the expected demagnetizing factor level, indicates that the system (although disordered) is organized in ferromagnetic domains.

Fig. 1. – $ac$ and $dc$ susceptibilities of the $CdCr_{1.9}In_{0.1}S_4$ sample. Both in-phase ($\chi'$) and out-of-phase ($\chi''$) components of the $ac$ susceptibility at frequency $0.04Hz$ are shown (open symbols). The $dc$ susceptibility (ZFC and FC, full symbols) has been measured in a field of 10 Oe.

Fig. 2. – Hysteretic behaviour of $\chi''$ in the vicinity of the ferromagnetic transition, upon cooling and re-heating at the different rates of $0.17K/min$ (full symbols) or $0.05K/min$ (open symbols). In this case, one can attribute the decrease of $\chi''$ to the decrease of the dissipative domain wall contribution. No hysteresis is visible on $\chi'$ (inset).
In Fig. 2, we show $\chi''(T)$ measurements in the vicinity of the ferromagnetic transition. The curves have been taken upon cooling from the paramagnetic phase down to 65K and heating back, for two different cooling/ heating rates. $\chi''$ is clearly hysteretic (when heating back it is always lower than during cooling), and sensitive to the rate of temperature change (at a given temperature, $\chi''$ is lower for slower rates). This effect is much larger than in spin-glasses, in which the cooling rate is to a large extent irrelevant (see [3]). But it is similar to the behaviour observed e.g. in a dipolar glass [11], where thermally activated domain growth is, as in the present case, the natural scenario. In Fig. 2, the obtained heating curve is the same for both cooling/heating rates. The hysteresis seen in Fig. 2 is essentially determined during the time spent around $T_c$, where domain walls are indeed expected to be weakly pinned. Finally, no significant hysteresis is seen on $\chi'$ (inset of Fig. 2), probably because $\chi'$, dominated by a volume response of the domains, is much less sensitive than $\chi''$ to domain wall dynamics.

Comparing aging phenomena in the spin-glass and ferromagnetic phases.

We have applied to this system the procedure which allowed the characterization of the so-called ‘memory and chaos effects’ in spin glasses [4]. The ac susceptibility has been measured at the three frequencies of 0.04, 0.4 and 4 Hz in the same run. We use the data in the paramagnetic regime (assuming $\chi'' = 0$) for checking and correcting slight frequency-dependent phase shifts in the detection setup.

We first determine ‘reference curves’ (solid lines in Fig. 3-4): starting from the paramagnetic phase, the temperature is continuously decreased down to 3K at a rate of 0.1K/min, and then raised back to 100K at this same rate. In a second experiment, we study aging in the following way. At 6 temperatures $T_i = 67, 40, 20, 13, 8$ and 5K, we have interrupted the cooling, stabilized the temperature, and let the system age at constant $T_i$ during 9 hours. When reaching 3K, we continuously re-heated up to 100K (the whole measurement lasts about 3 days).

At each temperature $T_i$, $\chi''$ slowly relaxes downwards with time. In the spin-glass temperature region (Fig. 3), the $\chi''$ relaxation is the strongest in absolute as well as relative value (amounting, for 0.04Hz, resp. to 9, 20 and 10% at 5, 8, and 13K). The corresponding $\chi'$ relaxation, although systematic, remains lower than 1%. In the ferromagnetic region (Fig. 4), a $\chi''$ relaxation is also clearly observed (although weaker than at low temperature, amounting for 0.04Hz to resp. 4 and 5% at 40 and 67K). Fig. 5 shows the frequency dependence of the aging part of $\chi''$. In both ferromagnetic and spin-glass phases, the same systematic trend of a stronger relaxation at lower frequency is found. Quantitatively, the curves can be rescaled onto a unique curve as a function of the scaling variable $\omega(t + t_0)$ ($t_0$ being an off-set time which takes into account the fact that the cooling procedure is not instantaneous). This scaling is equivalent to the (approximate) $t/t_w$ scaling of dc experiments, which is typically seen in spin-glasses (see discussions in [3]).

The most important point concerns the effect on aging of temperature changes. At all temperatures $T_i$ (hence in both phases), we find that when cooling is resumed after aging $\chi''$ merges back (even increasing in some cases) with the reference curve, although this reference is a non-equilibrium curve. This happens here exactly like in spin glasses [4, 13], in a similar fashion as in some other systems [12, 13]. Aging processes have to restart at lower temperatures, as if the aging evolution was not cumulative with that at a higher temperature (‘chaos-like’ or ‘rejuvenation’ effect), in contrast with the continuous hysteresis phenomenon displayed in Fig. 2. Another important feature is seen in the $\chi''(T)$ curve taken upon re-heating steadily from 3K. When reaching each of the temperatures $T_i$ in the spin-glass region (5, 8 and 13K), $\chi''(T)$ departs from the reference curve and traces back a dip which displays the memory of the past relaxation at $T_i$. Such memory effects have been characterized in details in spin glasses [3, 13]. In the ferromagnetic phase (20, 40 and 67K), in contrast, no memory effect is found with this procedure (Fig. 4). This result is in agreement with previous ac
Fig. 3. – Aging effects in the spin-glass region. The solid lines show a reference behaviour for continuous cooling and re-heating (the re-heating curve is slightly lower than the cooling curve). Open diamonds: when cooling is stopped at different temperatures $T_i = 13, 8$ and $5\,\text{K}$ during $9\,\text{h}$, $\chi''$ relaxes due to aging, but when cooling resumes $\chi''$ merges with the reference curve. Solid circles: when re-heating after cooling with stops for aging, the memory of aging is retrieved.

Fig. 4. – Aging effects for $T_i = 66.5\,\text{K}$ (beginning of the ferromagnetic phase). Same experiment and conventions as in Fig. 3, but here no memory effect can be seen.

and dc temperature cycling experiments performed on another reentrant system \cite{14}, in which however disorder effects seem to be stronger than here (larger FC-ZFC separation, widely rounded ferromagnetic $\chi_{FC}$ plateau, a behaviour which is very similar to that of the more diluted $x = 0.90$ thiospinel \cite{8}). In \cite{14}, it is observed that the effect of positive and negative temperature cycles is similar in the ferromagnetic phase (rejuvenation-type upon cooling as well as heating), while it is strongly asymmetric in the spin-glass phase (rejuvenation upon cooling, memory upon heating). Hence, rejuvenation effects are found in these disordered ferromagnets like in spin glasses, but memory effects are not readily seen in the ferromagnetic phase, where domain growth processes are important. In the following, we discuss wall dynamics as a possible source of interplay between spin-glass like and domain growth processes, and show that memory effects can be obtained in the ferromagnetic phase provided that the low-temperature cycling is short enough.

Ferromagnetic aging: a combination of domain growth and spin-glass like dynamics. –

In an ideal ferromagnet, domain growth only involves microscopic time scales. In the presence of pinning disorder (here dilution of $Cr$ by $In$, or structural defects), a domain wall has many metastable configurations between which it makes thermally activated hops. This gives rise to slow dynamics at the laboratory time scale. Actually, the problem of pinned elastic interfaces has similarities with that of spin-glasses. The frustration comes from the elastic energy associated to a deformation of the wall, which limits the possible excursions between the favourable pinning sites. Several theoretical arguments \cite{15} suggest that the energy landscape of a pinned domain wall is hierarchical, with small length scale $\ell$ reconformations corresponding
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\[ \chi''(\omega, t) - \chi''(\omega) \quad T = 40K \]

\[ \chi''(\omega, t) - \chi''(\omega) \quad T = 8K \]

\[ \chi''(\omega, t) - \chi''(\omega) \quad (\text{em} \text{u/cm}^3) \]

\[ \omega (t + t_0) \]

Fig. 5. – Scaling of the \( \chi'' \)-relaxation curves at \( T_i = 8K \) and \( T_i = 40K \) for different frequencies 0.04, 0.4 and 4Hz. The curves have been shifted vertically in order to compensate for the different equilibrium values, and an off-set time \( t_0 \) has been added to account for the fact that the cooling procedure is not instantaneous. We have also shown for comparison the fit proposed in [16, 3]:

\[ \chi''(\omega, t) = \chi''(\omega) + \left[ \omega (t + t_0) \right]^{-b}, \quad b = 0.2 \text{ for both temperatures}. \]

Fig. 6. – Partial memory effect in the ferromagnetic phase. The procedure is similar to that of Fig. 3-4, except that the low-temperature excursion was limited to either \( T_d = 64K \) (full circles), 60K (open squares), 50K (full triangles) or 30K (crosses). For a short enough low-temperature history (e.g. \( T_d = 64K \), full circles), the re-heating curves show a partial memory effect, which progressively faints out when significant ferromagnetic domain growth processes can occur (i.e. for decreasing \( T_d \) values).

The main figure concerns the \( \text{CdCr}_1\text{In}_{0.1}\text{S}_4 \) compound; in the inset, we show the same effect on the non-diluted \( \text{CdCr}_2\text{S}_4 \) ferromagnet (same conventions as in main figure, with \( T_d = 78, 75, 65K \)).

to small energy barriers \( E(\ell) \sim T\ell^b \).

In the spin glass phase, the rejuvenation upon cooling and the memory when heating back have been ascribed to a hierarchical organization of the metastable states, which are progressively uncovered as the temperature is decreased [4, 11, 14] (see also [17]). In brief, when the temperature is reduced, the system remains in a deep well (allowing for the ‘memory effect’), while new subwells appear, inducing new aging processes (rejuvenation effect). For a pinned domain wall, the hierarchy of reconformation length scales [15] is an appealing transposition in ‘real space’ of the spin-glass hierarchy of metastable states, in the following sense. As time elapses at fixed temperature, wall reconformations occur at longer and longer length scales (aging), while shorter length processes \( \ell < \ell^* \), with \( E(\ell^*) = kT \), are fluctuating in equilibrium. When the temperature is lowered, the long wavelength modes become frozen (thus corresponding to the deep well, which allows the memory), and the ‘glass length’ \( \ell^* \) decreases to \( \ell'^* \). The modes such that \( \ell'^* < \ell < \ell^* \) are aging (rejuvenation) \[ [16, 15] \]. These reconformations are expected to contribute to \( \chi'' \) as a function of \( \omega t \) \[ [15, 18] \] as indeed observed here in both phases.

In the ferromagnetic phase (and particularly close to \( T_c \)), in parallel with these reconformation processes, the average domain size grows with time. Thus, the impurities with which
a domain wall interacts are changing. Obviously, this net motion of the domain walls cannot preserve the memory of the reconformations. Domain growth effects are directly visible in the overall decrease of $\chi''$ between cooling and heating, a strong effect close to the transition at 70K (Fig.2 and 4), also present (slighter, but systematic) in the spin-glass phase (Fig.3). The absence of memory in Fig.4 is observed after a long period spent at lower temperatures, during which it is likely that the positions of the domain walls have indeed significantly changed.

We have therefore performed a slightly different experiment, with the aim of controlling the effect of domain growth after the aging relaxation. After aging 3.5h at $T_i = 66.6K$, the sample is cooled to a temperature $T_d$ which remains close to $T_i$, and then re-heated. In order to still accelerate the procedure, we have suppressed the measurement at the lowest frequency of 0.04Hz. Fig.6 shows that when the low-temperature excursion is small enough (like $T_d = 64K$), a partial memory of aging is revealed. The curves are somewhat noisy, but the effect is systematic as a function of $T_d$: for lower and lower $T_d$ values (60, 50 and 30K), the dip progressively disappears. The same experiment, performed in the middle of the ferromagnetic plateau ($T_i = 40K, T_d = 30$ and 35K), shows the same qualitative trend (although with weaker relaxation). Furthermore, we have recently started the investigation of the non-diluted CdCr$_2$S$_4$ ferromagnet. The first results (inset of Fig.6) do again show the same ‘rejuvenation and partial memory’ effects. In this non-diluted system, there are also competing interactions. Further studies on ferromagnets with non-frustrated interactions are needed in order to clarify the respective contributions of the frustration of interactions on the one hand, and of pinning mechanisms on defects of various origins on the other hand.

Finally, let us note that a similar systematic shift of the dip position with $T_d$ is observed in both samples of Fig.6. This is reminiscent of the way in which the memory is erased by a slight re-heating in the $x = 0.85$ thiospinel spin glass (see 2nd ref. of [6]), and is yet unexplained.

**Conclusions.**

Within the domain-growth description of spin-glasses, the way to interpret the rejuvenation effect (and the weak cooling rate dependence of the measured quantities) is to invoke the idea of the chaotic evolution of the spin-glass order with temperature. However, as emphasized in [6], this is difficult to reconcile with the simultaneous memory effect discussed above. Furthermore, such chaos should be absent in the ferromagnetic phase (or indeed only concern the domain wall conformations). Let us finally note that until now numerical simulations have failed in identifying this type of ‘chaos’.

On the other hand, the coexistence of memory and rejuvenation has readily been interpreted in a hierarchical phase space picture, which finds in the present case a natural real space interpretation in terms of wall reconformations. Comparing the spin-glass and ferromagnetic situations suggests that the dynamics in spin glasses is dominated by wall reconformation processes and not by domain growth, which would erase the memory, and lead to strong cooling rate dependence. The extrapolation of this idea to the case of spin glasses raises incentive questions about the nature of ‘domains’ and ‘walls’ in this case. Many ideas on the non-trivial geometry of the spin-glass domains have indeed been proposed over the years. A possibility is that the ordered phase of a spin-glass contains a large number of pinned, zero tension walls, which reconform in their disordered landscape without any overall tendency to coarsen. The growth of a spin-glass correlation length, reported both numerically and experimentally, could then be understood as the progressive increase of a typical length scale for wall reconformations at fixed temperature, while the effect of temperature variations would involve a hierarchy of different length scales.
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REFERENCES

[1] E. Vincent, J. Hammann, M. Ocio, J.-P. Bouchaud, L.F. Cugliandolo, in Complex Behaviour of Glassy Systems, Springer Verlag Lecture Notes in Physics Vol.492, M. Rubi Editor, 1997, pp.184-219, and refs. therein.

[2] D. S. Fisher and D. A. Huse, Phys. Rev. B 38, 373 and 386 (1988); G. J. M Koper and H. J. Hilhorst, J. Phys. France 49, 429 (1988).

[3] For a review of different theoretical models leading to aging, in particular mean-field models, see: J.-P. Bouchaud, L.F. Cugliandolo, J. Kurchan, M. M´ezard, in ‘Spin-glasses and Random Fields’, A. P. Young edt. (World Scientific, 1998)

[4] Ph. Refregier, E. Vincent, J. Hammann and M. Ocio, J. Phys. France 48, 1533 (1987); E. Vincent, J.-P. Bouchaud, J. Hammann and F. Lefloch, Phil. Mag. B 71, 489 (1995).

[5] P. Granberg, L. Lundgren and P. Nordblad, J. Magn. Magn. Mater. 92, 228 (1990); P. Granberg, L. Sandlund, P. Nordblad, P. Svedlindh and L. Lundgren, Phys. Rev. B 38, 7097 (1988).

[6] K. Jonason, E. Vincent, J. Hammann, J.-P. Bouchaud and P. Nordblad, Phys. Rev. Lett. 31, 3243 (1998); K. Jonason, P. Nordblad, E. Vincent, J. Hammann and J.-P. Bouchaud, Europ. Phys. Jour. B 13, 99 (2000).

[7] A.J. Bray and M.A. Moore, Phys. Rev. Lett.58 (1987) 57.

[8] M. Alba, J. Hammann and M. Nogu`es, J. Phys. C: Solid State Phys., 15, 5441 (1982).

[9] S. Pouget and M. Alba, Physica B 267-268, (1999) 304; S. Pouget, M. Alba, N. Fanjat and M. Nogu`es, Physica B bf 180-181, (1992) 244 ; S. Pouget and M. Alba, J. Phys.: Cond. Mat. 7, 4739 (1995).

[10] Some interesting mechanisms for the reentrance of a frustrated phase are explicited in S. Miyashita, Progr. of Theor. Phys. 69, 714 (1983) and H. Kitatani, S. Miyashita and M. Suzuki, J. Phys. Soc. Jap. 55, 865 (1986).

[11] F. Alberici, J.-P. Bouchaud, L. Cugliandolo, J. Doussineau, A. Levelut, Phys. Rev. Lett. 81 4987 (1998)

[12] P. Doussineau, T. de Lacerda-Arˆoso and A. Levelut, Europhys. Lett. 46 401(1999).

[13] L. Bellon, S. Ciliberto and C. Laroche, e-prints cond-mat/9905160 and 9906162.

[14] K. Jonason, J. Mattsson, P. Nordblad, Phys. Rev. Lett. 77, 2562 (1996); K. Jonason and P. Nordblad, Eur. Phys. J. B 10, 23 (1999).

[15] [16] L. Balents, J.-P. Bouchaud, M. M´ezard, J. Physique I 6, 1007 (1996).

[17] J.-P. Bouchaud and D.S. Dean, J. Phys. I France 5, 265 (1995), J.P. Bouchaud, e-print cond-mat/9910387.

[18] L. Cugliandolo, J. Kurchan, ‘Mean-field theory of temperature cycling in spin-glasses’, Phys. Rev. B 60 922 (1999).

[19] For recent numerical simulations of this problem, see: H. Yoshino, Phys. Rev. Lett. 81, 1483 (1998), and H. Yoshino, in preparation.

[20] A similar partial memory effect has very recently been seen by the authors of Ref. [12] in their disordered dielectric system.

[21] V. Dupuis, PhD thesis, in progress.

[22] J.Kisker, L.Santen and M. Schreckenberg, Phys. Rev. B 53, 6418 (1996); H. Takayama, H. Yoshino and T. Komori, Int. J. Mod. Phys. C 10, 647 (1999); A. Billoire and E. Marinari, preprint cond-mat/9901035.

[23] J. Villain, J. Physique France 46, 1843 (1985); J. Villain, Europhys. Lett. 2, 871 (1986); M. Feigel’man, L. Ioffe, Z. Phys. B 51 237 (1983); M. Gabay, T. Garel, J. Physique 46, 5 (1985); M. Ocio, J. Hammann and E. Vincent, J. Magn. Magn. Mater. 90-91, 329 (1990).

[24] J. Houdayer, O. C. Martin, Europhys. Lett. 49, 794 (2000).

[25] H. Rieger, Ann Rev. of Comp. Phys. II, ed. D. Stauffer (World Scientific, Singapore, 1995); E. Marinari, G. Parisi, F. Ritort and J. J. Ruiz-Lorenzo, Phys. Rev. Lett. 76, 843 (1996); T. Komori, H. Yoshino and H. Takayama, J. Phys. Soc. Jpn. 68, (1999) 3387.

[26] Y.G. Joh, R. Orbach, J. J. Wood, J. Hammann and E. Vincent, Phys. Rev. Lett. 82, 438 (1999).