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Magnetic relaxation phenomena in a CuMn spin glass

C. Djurberg, K. Jonason[a], and P. Nordblad

Department of Materials Science, Uppsala University, Box 534, 751 21 Uppsala, Sweden

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Abstract. Experiments on the temperature and time dependence of the response function and the field cooled magnetisation of a Cu(Mn) spin glass at temperatures below the zero field spin glass temperature are used to explore the non-equilibrium nature of the underlying spin configuration. The results imply that a certain spin configuration is imprinted on the system as the temperature is decreased at a constant cooling rate. The cooling rate governs the magnitude of the FC magnetisation ($M_{\mathrm{FC}}(H,T)$). Any intermittent halt at a constant temperature, T_{i}, imprints an extended spin configuration, a process that is reflected *e.g.* in a downward relaxation of M_{FC}. On continued cooling at the same rate, the magnitude of $M_{\mathrm{FC}}(T)$ remains at a lower level than that of a continuous cooling curve. These results are put into the context of the corresponding behaviour of the response function as observed in measurements of the relaxation of the zero field cooled magnetisation.

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

The non-equilibrium character of the slow relaxation of the magnetisation of $3d$ spin glasses below the zero field phase transition temperature has been extensively studied by dc-magnetisation [1,2] and ac-susceptibility [3,4] experiments as well as MC-simulations [5]. Different models to describe the spin glass phase and the observed ageing phenomenon have been suggested [6,7] and discussed in connection with the empirical data. In this paper we report results from dc-magnetic relaxation experiments of the field cooled (FC) and the zero field cooled (ZFC) magnetisation. The field cooled magnetisation is found to approach a reversible magnetisation level ($M_{\mathrm{FCrev}}(T)$) if the sample is continuously cooled and re-heated at one and the same rate while remaining at temperatures below T_{g}. On an intermittent stop at constant temperature, the field cooled magnetisation relaxes downward and on continued cooling the magnetisation remains at a lower level than in a continuous cooling process. On re-heating the sample the lower magnetisation level is maintained only up to the temperature of the intermittent halt, whereas on further heating above the halt temperature the reference level of a continuous process is progressively regained.

We also show that the wait time dependence of the response function at constant temperature is governed by the cooling/heating rate and is independent of any long time equilibration at sufficiently lower or higher temperatures. However, for temperatures that are only slightly different, the response function is affected by a wait time at the nearby temperature; a region of overlap is observed.

This behaviour suggests that there is a certain favorable magnetisation associated with the spin configuration that the FC spin glass attains in a cooling process at a specific rate. This spin configuration and its magnetisation remains essentially unperturbed if the system is re-heated at the same rate. If the sample is kept at a constant temperature, the spin configuration is free to re-configure unrestrictedly on large length scales and the FC-magnetisation decreases. This extended spin structure becomes imprinted and remains "frozen" in when the sample is further cooled, and is only washed out when the temperature reaches well above the temperature for the intermittent halt.

2 Experimental

The sample is a bulk piece of a Cu(Mn13.5at%). The experiments were performed in a non-commercial SQUID magnetometer specially designed for low field dynamic magnetic susceptibility studies [8]. Two basic experimental procedures are employed in the study.

(i) Measurements of the zero field cooled magnetisation (the response function). The sample is cooled in zero field and subjected to certain thermal sequences after which a weak magnetic field is applied and the relaxation of the magnetisation is recorded at constant temperature, *i.e.* the temperature and history dependent response function is measured.

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[a] e-mail: `Kristian.Jonason@material.uu.se`

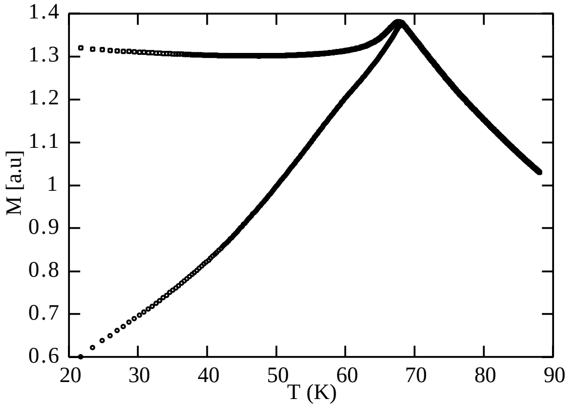

Fig. 1. FC- and ZFC-magnetisation *vs.* temperature of Cu(Mn13.5at%), $H = 1$ Oe.

(ii) Measurements of the field cooled magnetisation. The magnetic field is applied at a temperature well above $T_g(H = 0)$ and the sample is cooled (and also re-heated) in constant field. The temperature dependence, $M_{FC}(T, t_c)$ or the relaxation $M_{FC}(T_m, t)$ of the magnetisation at constant temperature is measured. Here t_c is a characteristic time determined by the specific cooling/heating rate used in the experiment and T_m is the temperature for a relaxation measurement.

The cooling/heating rate in these measurement procedures is about 3 K/min. To acquaint with the sample the field cooled and zero field cooled magnetisation is plotted *vs.* temperature in Figure 1. The curves are measured in a field of 1 Oe and show a cusp in the ZFC susceptibility and an onset of irreversibility at about 68 K, which also closely reflects the spin glass temperature, T_g, of the sample.

3 Response function

Figure 2 shows the zero field cooled magnetisation and the corresponding relaxation rate, $S = 1/H\partial M/\partial \log t$, measured at three significantly different temperatures as a function of wait time, t_w, at constant temperature before the magnetic field is applied. The wait time dependence of the magnetic relaxation is somewhat different at the three temperatures but also shows basic similarities. At the higher temperature, the wait time causes a maximum of the relaxation rate at an observation time closely equal to t_w. At the lower temperatures, the corresponding maximum in the rate curves is somewhat broader, of lower amplitude and somewhat delayed compared to the actual t_w. It should also be mentioned here that the spin glass can be under-cooled or over-heated arbitrarily large amounts, provided the cooling/heating rate is not significantly altered; coming back to the measurement temperature yields an almost identical response to what is observed when the sample is directly brought to T_m. The governing parameter for the $t_w = 0$ and the continued wait time dependence of the response function, at the measurement temperature T_m, is the characteristic time t_c of the cooling rate. The very fact that the evolution or ageing of the response function at vastly different temperatures shows similar behaviour implies that the spin configuration that develops during cooling differs at different temperatures; the system is chaotic. To carry this point further, we show results from an experiment where the sample has been cooled to a temperature $T_m \pm \Delta T$, kept there at constant temperature a wait time 3000 s and then cooled from $T_m + \Delta T$ (or heated from $T_m - \Delta T$) to T_m, where the magnetic field is applied after a short wait time that allows thermal stability to be attained. The results of the experiment are shown in Figure 3, where the relaxation rate is plotted *vs.* log t for different magnitudes of positive ($T_m + \Delta T$), Figure 3a, and negative ($T_m - \Delta T$), Figure 3b, temperature shifts ΔT. Three different regions for the response can be distinguished:

Large ΔT: The response is indistinguishable from what is measured if the sample is cooled directly to T_m from a high temperature above $T_g(0)$, *i.e.* the time that the sample has been kept at constant temperature $T_m \pm \Delta T$ is irrelevant for the response at T_m. The spin configuration developed at $T_m \pm \Delta T$ does not map onto the configuration that develops at T_m.

Small ΔT: The response appears unaffected by the temperature shift, only the position of the maximum is pushed to shorter (negative ΔT) or longer (positive ΔT) time than the actual wait time. This behaviour can be accounted for by the temperature dependence of the dynamics and that there is an overlap between the spin configurations at the two temperatures on the time scales of our experiment. Similar results have been interpreted using length scale arguments [9] that also appear in droplet models. It should be noted that the behaviour for positive ΔT does not give the perfect overlap with the corresponding response attained at T_m that a negative ΔT gives.

Intermediate ΔT: On positive temperature shifts, there is a continuous shift of the position of the maximum which ends by closely merging into the $t_w = 0$ curve. On negative temperature shifts the behaviour is less pregnant and there is a dramatic broadening and levelling off of the observable maximum that continuously changes to end by perfectly merging into the $t_w = 0$ curve for $\Delta T >$ 4-5 K.

These results show that in our time window, the overlap between the attained configuration at the two temperatures is good for $\Delta T < \Delta T_0$ but gradually decreases with increasing magnitude of ΔT. At large ΔT's there is no overlap, a chaotic situation is observed. It is however in this context also important to recall results from temperature cycling experiments, *i.e.* experiments where the spin glass is aged at T_m, then subjected to a temperature cycling of magnitude ΔT, and when T_m is recovered the magnetic field is applied and the relaxation of the magnetisation is recorded. The temperature cycling experiments show that on positive temperature cycles, the spin glass appears to have a completely random initial state for large values of ΔT, whereas using negative ΔT's larger than ΔT_0 requires a substantial wait time at the lower temperature to achieve a measurably reinitialised system [10]. However, if even larger negative values of ΔT

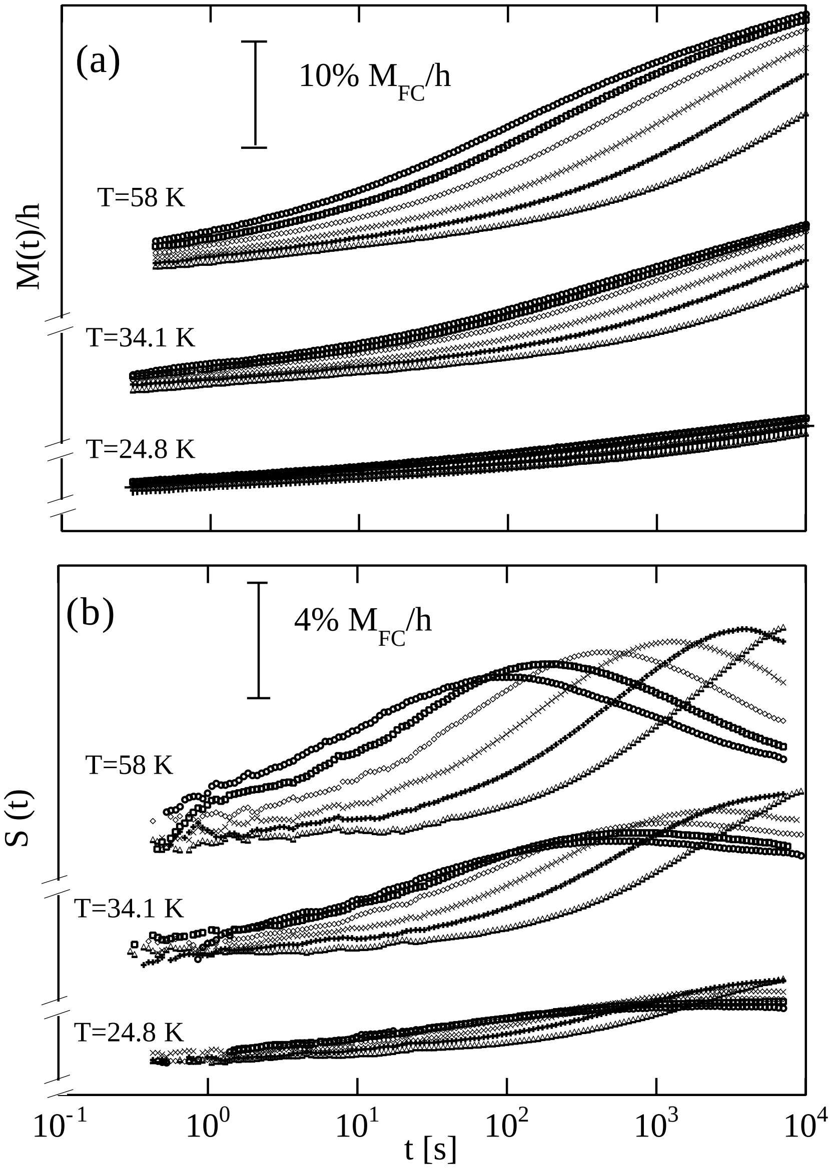

Fig. 2. (a) $M_{\rm ZFC}(T_{\rm m},t)$ measured at three different temperatures and at the wait times indicated in the figure. (b) The corresponding relaxation rate, $S = 1/H\partial M/\partial \log t$, $H = 1$ Oe.

are used, the effect of the cycling again becomes small and the spin configuration attained at $T_{\rm m}$ appears frozen.

Summarising these results on the properties of the response function, we find: the response function and its wait time dependence is unique for each temperature and cooling/heating rate. When the wait time becomes much longer than the time scale, $t_{\rm c}$, of the cooling/heating rate, the response function $p(t,T,t_{\rm w})$ is determined only by T and $t_{\rm w}$. If the sample is kept intermittently at a constant temperature $T_{\rm i}$ a time $t_{\rm i}$, and then further cooled and aged at a substantially lower temperature, the response function when the temperature $T_{\rm i}$ is recovered is closely equal to only $p(t,T_{\rm i},t_{\rm i})$. On the contrary, if the temperature is increased substantially above $T_{\rm i}$, the response when $T_{\rm i}$ is recovered is the same as cooling the sample directly to $T_{\rm i}$. The system can carry the information from numerous intermittent stops provided they are sufficiently separated in temperature and that the temperatures are only recovered after heating from lower temperatures. This memory behaviour is also anticipated to be observable in the temperature dependence of the ac-susceptibility [11]. This is illustrated in Figure 4 by a plot of the temperature dependence of the out of phase component of the low frequency, $f = 0.51$ Hz, ac-susceptibility of our spin glass sample. Three curves are displayed, which all are measured on heating at one and the same constant heating rate after using three different cooling procedures. The curve marked ref. in the figure shows the result when the sample has been cooled continuously to a low temperature. The curve marked single shows the result when the sample was intermittently halted for 6 hours at $T_1 = 40$ K during cooling. The curve marked double shows the result when the sample was intermittently halted for 6 hours first at $T_1 = 50$ K and then at $T_1 = 40$ K during cooling.

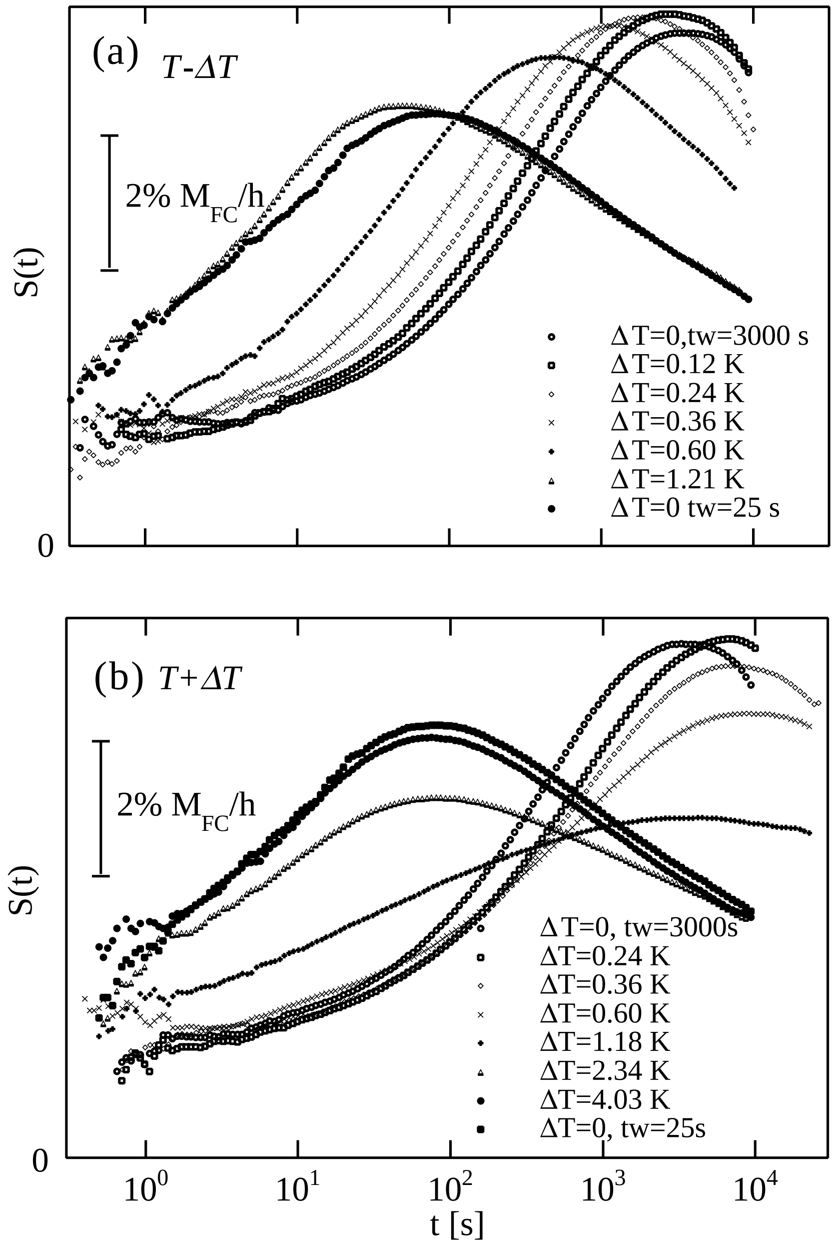

Fig. 3. The relaxation rate of the zero field cooled magnetisation measured after keeping the sample at constant temperature at $T_{\mathrm{m}} \pm \Delta T$ a wait time $t_{\mathrm{w}} = 3000$ s, and then shift the temperature to $T_{\mathrm{m}} = 58$ K and measure the relaxation of the magnetisation in an applied field, $H = 1$ Oe: (a) shifts $T_{\mathrm{m}} - \Delta T$ to T_{m}, (b) shifts $T_{\mathrm{m}} + \Delta T$ to T_{m}.

The result is in line with the discussion above. It is possible to retrieve information from not only one stop but also two stops and more if they are well separated in temperature. The dip(s) on re-heating the sample is a consequence of the above discussed memory of the halt(s) at T_{i} during cooling. The memory is erased when the temperature reaches well above T_{i}. Further experimental results illustrating this behaviour have recently been published [12].

4 Relaxation in constant field

The field cooled magnetisation of spin glasses is a quantity that falls out of equilibrium at the irreversibility temperature, *i.e.* the temperature where the ZFC and FC magnetisation curves merge. At lower temperature the FC magnetisation becomes cooling rate dependent and relaxes if the sample is kept at constant temperature. Some time dependent features of the FC-magnetisation have earlier been reported for a three dimensional metallic spin glass [13] and for two dimensional spin glasses [14] recently followed by a more extensive study on an insulating three-dimensional Ising system [15]. To connect to the experiments described above we have performed measurements of the relaxation of the FC magnetisation after cooling from a high temperature at one and the same cooling rate and recording the relaxation at constant temperature.

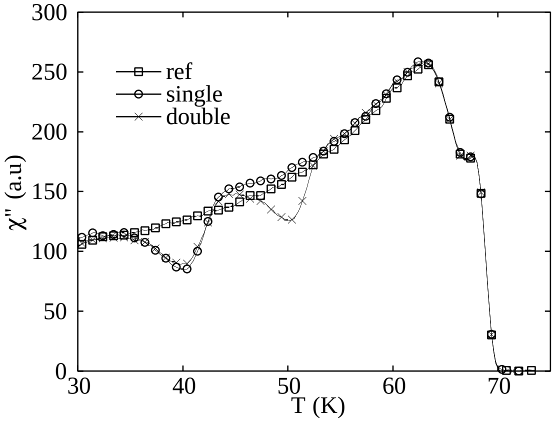

Fig. 4. $\chi''(T,\omega,t)$ *vs.* T. Single memory experiment: while cooling, the sample was kept at $T_1 = 40$ K for 6 hours. Double memory experiment: The sample was kept at $T_1 = 50$ K (6 hours) and $T_2 = 40$ K (6 hours). Displayed in the figure are the resulting curves achieved when the system is continuously reheated after the different cooling procedures. The reference curve is measured on heating after the system had been continuously cooled at a constant cooling rate. The continuous cooling/heating rate is about 0.5 K/min in all three experiments.

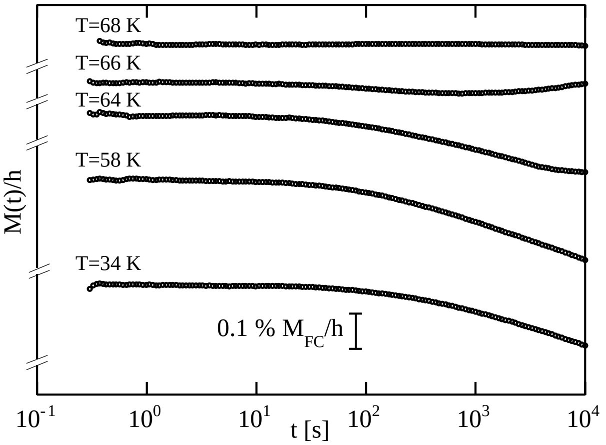

Fig. 5. $M_{\mathrm{FC}}(T_{\mathrm{m}}, t)$ *vs.* log t measured at some different temperatures as indicated in the figure.

Some curves recorded at different temperatures are shown in Figure 5. It is worth to note that in our experimental time window, there is an upward relaxation at high temperatures near T_{g}, an initial downward relaxation followed by a turning upwards on longer time scales at a somewhat lower temperature and only a downward relaxation at lower temperatures. We choose to further investigate how the system responds to a temperature cycling at the same measurement temperature as the response function was studied. In Figure 6a the results after keeping the sample at 58 K and record the relaxation of the magnetisation during 10^4 s, then subject the sample to a temperature cycle of magnitude ΔT, and when T_{m} is recovered record the continued relaxation of the magnetisation. The sample is heated to the higher temperature and immediately re-cooled to the measurement temperature. The original magnitude and relaxation of the magnetisation is regained if a large ΔT is used. For $\Delta T < \Delta T_0$ the relaxation continues apparently undisturbed when T_{m} is recovered and for intermediate values of ΔT a continuously increasing regained magnitude of M is observed. (The ΔT_0 observed in these experiments has about the same value as the ΔT_0 obtained in the cycling experiments for the response function recapitulated above.) Figure 6b shows corresponding results using negative cycling. The relaxation of the magnetisation is first recorded during 3000 s at T_{m}, then the sample is kept at the lower temperature for 10^4 s and the relaxation is recorded. Thereafter the sample is heated to T_{m} and the continued relaxation is recorded. In the figure the relaxation is plotted at T_{m} also for $\Delta T = 0$ to have a reference curve for the whole measured time interval. Some features are noticeable. For small values of ΔT, the relaxation during the temperature cycle results in a decreased magnetisation compared to only remaining at T_{m}. For larger values T_{m} there is a slightly increased magnetisation. It should also be stressed that if the sample is subjected to only a temperature cycling ΔT to a lower temperature and heated back at the rate t_{c}, the magnetisation retains the magnetisation attained after 3000 s and continues to relax apparently unaffected by the temperature cycling. The overall picture is that the FC magnetisation is to some extent affected by the cycling, but maybe most significantly, in spite of the rather long wait time at the lower temperature, the magnetisation on recovering T_{m} is rather closely equal to the magnetisation level reached when only keeping the sample at constant temperature. Also seen from the figure is that the relaxation during the period at the lower temperature is in fact as large as if the sample had been immediately cooled to that temperature without stopping at T_{m}, however with a significantly decreased initial magnitude. In summary it is found that the field cooled magnetisation is essentially governed by:

(i) A closely reversible magnetisation, $M_{\mathrm{FCrev}}(T, t_{\mathrm{c}})$ is approached if the same cooling and heating rate is used.

(ii) If the system is halted at a constant temperature, T_{i}, a time t_{i}, the FC-magnetisation decays an amount, $\Delta M(T_{\mathrm{i}}, t_{\mathrm{i}})$ and if the cooling is continued at the original cooling rate, the magnetisation remains at the lower magnetisation value:

$$M_{\mathrm{FC}}(T > T_{\mathrm{i}}, t_{\mathrm{c}}) = M_{\mathrm{FCrev}}(T, t_{\mathrm{c}}) \tag{1}$$

$$M_{\mathrm{FC}}(T < T_{\mathrm{i}}, t_{\mathrm{c}}) = M_{\mathrm{FCrev}}(T, t_{\mathrm{c}}) - \Delta M(T_{\mathrm{i}}, t_{\mathrm{i}}). \tag{2}$$

(iii) On re-heating the FC sample, the magnetisation rather adequately follows the same track as during cooling, including an increase at the temperature, T_{i}, where the sample was kept at constant temperature (see Eq. (2)).

These observations are summarised in the schematic drawing of Figure 7 where a cooling curve including intermittent halts at two different temperatures and the subsequent continuous heating curve are illustrated. It is worth to notice that the halt temperatures should be significantly separated for the behaviour to be resolved.

Many experiments on relaxation in spin glasses are made using the relaxation of the thermo remanent

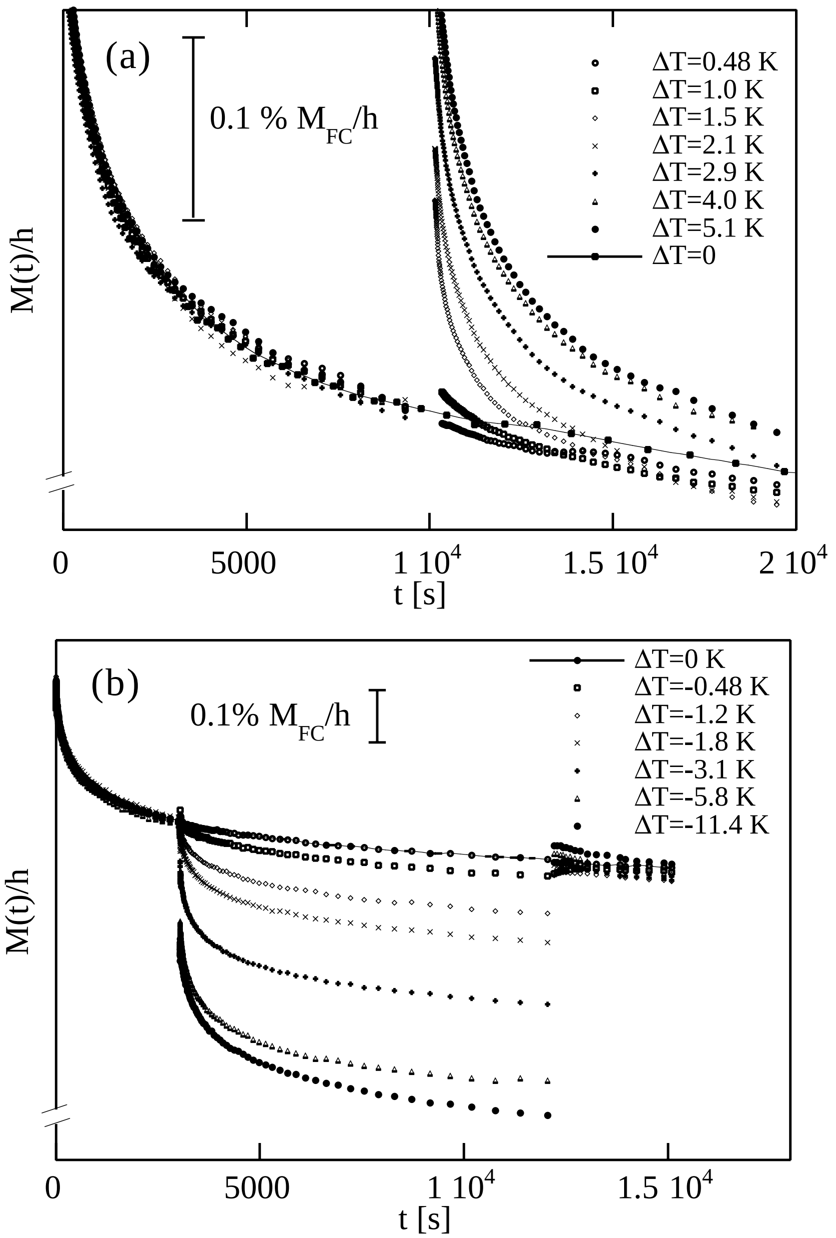

Fig. 6. $M_{\rm FC}$ *vs.* time at $T_{\rm m} = 58$ K and $H = 10$ Oe. (a) After 10^4 s the temperature is cycled ΔT and on recovering $T_{\rm m}$ the recording is continued. (b) The relaxation at $T_{\rm m}$ is recorded 3000 s, then the sample is cooled to $T_{\rm m}$-ΔT, allowed to relax during 10^4 s, as shown in the figure, and then heated back to $T_{\rm m}$ where the continued relaxation is recorded an additional 3000 s.

magnetisation (TRM) as a probe, *i.e.* the relaxation observed after cooling the sample in a constant field to a measurement temperature, cutting the applied field and then recording the relaxation of the magnetisation. The magnitude of the TRM measured at a temperature well below an intermittent halt will have a different magnitude than if immediately cooled to the measurement temperature. However, the relaxation rate will remain identical in the two procedures. For a $t_{\rm w} = 0$ example we can write:

$$M_{\rm TRM}(t, t_{\rm c}) = M_{\rm FCrev}(T, t_{\rm c}) + M_{\rm ZFC}(t, t_{\rm c}) \qquad (3)$$

$$M_{\rm TRM}(t, t_{\rm c}, t_{\rm i}) = (M_{\rm FCrev}(t, t_{\rm c}) - \Delta M) + M_{\rm ZFC}(t, t_{\rm c}) \qquad (4)$$

for the two cases. That the termoremanent magnetisation carries information on $M_{\rm FC}$ distinguishes it from the zero field cooled relaxation that always only sees the response function.

5 Conclusions

From measurements of the response function and the FC magnetisation we derive the following characteristics of the spin glass magnetisation and dynamics on our experimental time scales. An energetically favourable

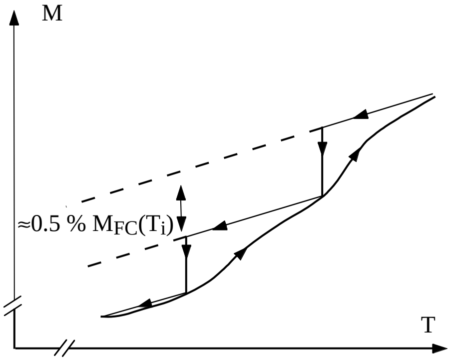

Fig. 7. A schematic drawing of M_{FC} *vs.* T where two intermittent halts are made at two well separated temperatures during cooling. The curve on continuously re-heating the sample is also indicated.

non-equilibrium spin configuration is imprinted on a spin glass when cooling to a low temperature. This configuration is essentially preserved when heating the sample at an identical heating rate, the FC magnetisation is governed by the characteristic time t_{c} and the response function measured at constant temperature is independent of the thermal history. If the sample is kept at a constant temperature, the spin configuration rearranges on large length scales, which implies a decay of the FC magnetisation and an altered response function. This configuration is imprinted on the system and is "frozen" in on lowering the temperature. When the same temperature is recovered on re-heating, the sample appears as if the thermal history at low temperature had not occurred. These results verify that the spin glass phase is chaotic, that there is an overlap between the equilibrium spin configurations developed at two slightly different temperatures and also show that a spin configuration attained at a high temperature is "imprinted" on the system on lowering the temperature.

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