Dynamical studies of the response function in a Spin Glass

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Experiments on the time dependence of the response function of a Ag(11 at%Mn) spin glass at a temperature below the zero field spin glass temperature are used to explore the non-equilibrium nature of the spin glass phase. It is found that the response function is only governed by the thermal history in the very neighbourhood of the actual measurement temperature. The thermal history outside this narrow region is irrelevant to the measured response. A result that implies that the thermal history during cooling (cooling rate, wait times etc.) is imprinted in the spin structure and is always retained when any higher temperature is recovered. The observations are discussed in the light of a real space droplet/domain phenomenology. The results also emphasize the importance of using controlled cooling procedures to acquire interpretable and reproducible experimental results on the non-equilibrium dynamics in spin glasses.

I. INTRODUCTION

The non-equilibrium nature of the spin glass phase has been extensively investigated since it was discovered in low frequency ac-susceptibility measurements and dc-relaxation experiments in the early 1980’s. The interpretation of the experimental results and the design of new experimental procedures emanate essentially from two different theoretical approaches: hierarchical phase space models and real space droplet/domain models. A high level of phenomenological and theoretical insight into the phenomena has by now been acquired. There remain, however, unresolved problems as to the interpretation and reproducibility of non-equilibrium results. These shortcomings do in part also prevent a useful judgement in-between the applicability of the different models to real 3d spin glasses. The recently reported memory effects, observable in low frequency ac-susceptibility experiments, not only elucidates some paradoxal features of the spin glass phase, but an in detail study of related phenomena also emphasizes the importance of: cooling/heating rates, wait times, thermalisation times etc., i.e. the detailed thermal history of the sample on the results from low frequency ac-susceptibility and dc-magnetic relaxation experiments. These complicated non-equilibrium phenomena should be considered in the perspective that an ac-experiment at only a couple of decades higher frequency, \( f \geq 100 \) Hz (observed time \( 1/\omega \leq 2 \) ms), in spite of a strong frequency dependence, simply shows an equilibrium character when measured in ordinary ac-susceptometers. The non-equilibrium processes of the system are only active on observation time scales governed by the cooling/heating rate and during a halt at constant temperature these evolve with the time the sample has been kept at constant temperature.

In this paper we report results from low field zero field cooled (zfc) relaxation experiments, i.e. measurements of the response function, at one specific temperature in the spin glass phase. The parameter we vary in a controlled way is the thermal history in the spin glass phase. The results imply that the response function is governed by: the cooling procedure in a rather narrow region just above the measurement temperature, the wait time before the response function is measured, and the cooling/heating procedure in a rather narrow region just below the measurement temperature if an additional undercooling has been carried out. It is also clearly shown that the thermal history, in the spin glass phase, at temperatures well enough separated from the measurement temperature is irrelevant to the response function at \( T_m \).

The investigation is also motivated by a lack of agreement in the detailed behaviour of the non-equilibrium dynamics, when measured on one and the same spin glass material in different magnetometers.
The spin glass temperature cusp in the zfc susceptibility around 35 K closely reflects Fig. 1. The curves are measured in a field of 40 Oe. The field cooled magnetisation is plotted vs. temperature in a spin glass temperature magnetic field on or off. Not much attention has been instructively has been used to visualise the memory phe-
tation data we show the relaxation rate, i.e. the logarithmic time perspective rather similar, cooling rates are plotted, 3 K/min open symbols and 0.5 K/min solid symbols. The wait time dependence of the data displays the characteristics of the ageing phenomenon in spin glasses (a similar ageing phenomenon is also an inherent property of other disordered and frustrated magnetic systems). The response is visually sensitive to the cooling rate, $t_w$, but the influence of $t_w$ decreases as the wait time increases. For the $t_w=10$ s curves, rapid cooling yields a maximum in $S(t)$ at a shorter observation time than slow cooling. The two curves are clearly different and do not coalesce anywhere in the measured time interval. For the longer wait times, the position of the maxima do not differ appreciably, but the magnitude is somewhat higher for the rapidly cooled curves. At short observation times, for the $t_w=10^3$ and $10^4$ s curves, there are no or weak differences in the relaxation rates, but after some time they start to deviate and there remains a cooling rate dependence all through the long time part of our observation time window.

Concentrating on the position of the maximum in the relaxation rate and identifying this with an effective age of the system, $t_{eff}$, this parameter is apparently governed by the cooling rate and the wait time. When the wait time is short, $t_{eff}$ is governed by the cooling rate, whereas for longer wait times, it is dominated by the wait time, $t_w$. The position of the maximum in the relaxation rate is closely equal to the wait time for $t_w=10^3$ and $10^4$ s while for the $t_w=10$ s the maximum is delayed about a decade in time. Similar tendencies as to the evolution of the effective age with increasing wait time have earlier been reported in connection with a specific method, ‘field quenching’, to achieve a well defined initial state for ageing experiments.

To further investigate how the thermal sequence on approaching the measurement temperature influences the response function, temperature shift experiments under controlled cooling and heating rates were performed. In the negative temperature shift experiment, the system is kept a certain wait time at a shift temperature, $T_s$, above $T_m$. Thereafter the system is cooled to the measurement temperature where the field is applied immediately after the sample has become thermally stabilised. The positive temperature shift experiment follows a similar procedure, but the temperature $T_s$ is below $T_m$. Fig. 3 shows the relaxation rate when the system has been subjected to (a) negative and (b) positive temperature shifts. The wait time at $T_s$ is 1000 s and $T_m=27$ K. The cooling and heating rates are 0.5 K/min. Two reference curves measured after the sample has been cooled at 0.5 K/min directly to $T_m$ are included in the figure. One corresponds to a wait time $t_w=1000$ s and the other is measured without any wait time prior to the field application ($t_w \approx 0$). From Fig. 3(a) it is seen that for $T_s > 30$ K the response is indistinguishable from what is measured if the sample

![Graph: The relaxation rate, $S(t) = 1/h \partial m/\partial \ln t$ vs. time, measured at $T_m = 27$ K for three different wait times, $t_w$, as indicated in the figure, using two different cooling rates 3K/min. and 0.5 K/min, $h=1$ Oe.](image)
is cooled directly to \( T_m \). The time the sample has been kept at \( T_s \) is irrelevant for the response at \( T_m \). When the shift temperature \( T_s \) is lower than 30 K, the time spent at the higher temperature starts to influence the response at \( T_m \). The first deviation from the reference (\( t_w \approx 0 \)) curve occurs at long observation times, the maximum in the relaxation rate decreases in magnitude and on further decreasing \( T_s \) it shifts towards longer times, and finally when \( T_s \) approaches \( T_m \), the relaxation rate approaches the \( t_w = 1000 \, \text{s} \) reference curve. The important implication of this behaviour is that only the thermal history in a limited temperature region just above \( T_m \) governs the response function.

The results from positive temperature shifts shown in Fig. 3(b) give a somewhat more complicated result. A first observation is that when the system has been cooled to and aged at temperatures well below \( T_m \), the measured curves are identical, but different from the \( t_w \approx 0 \) reference curve, the maximum in the relaxation rate occurs at a shorter time than for the reference curve. Thus, the response function is somewhat different after only cooling to the measurement temperature at one specific cooling rate, compared to after substantially undercooling the sample and re-heat it with a similar heating rate. The curves representing \( T_s = 15 \) and 20 K are indistinguishable from each other, but when \( T_s \) is increased further, the relaxation rate gets suppressed and the maximum shifts towards longer times to finally coalesce with the reference \( t_w = 1000 \, \text{s} \) curve when \( T_s \approx T_m \). Fig. 4 shows relaxation rate curves from experiments where the sample is undercooled to \( T_s \), and either kept there 1000 s (as in Fig. 3(b)) or immediately re-heated to \( T_m \), where the relaxation is measured using \( t_w \approx 0 \). The cooling/heating rates are again 0.5 K/min. The figure shows that the wait time at the lowest temperature does not affect the results, identical curves are measured whether the sample is kept at 20 K for 1000 s or 0 s. For a shift temperature, \( T_s = 24.5 \) K, closer to \( T_m \), a clear difference is observed between the two curves with different wait times. The implication from these data is again that the thermal history far enough away from the measurement temperature is irrelevant to the response function at \( T_m \), the response is in the experimental time window fully governed by the previous cooling/heating history in the very neighbourhood of \( T_m \).

In Fig. 5, results for (a) negative and (b) positive temperature shifts using two different cooling rates 3 K/min and 0.5 K/min (as in Fig. 3) are displayed. The cooling/heating rate from \( T_s \) to \( T_m = 27 \) K is always the same, 0.5 K/min. For negative temperature shifts (Fig. 5 (a)) there is no influence of the different cooling rates for \( T_s > 29 \) K. The rapid and slow cooling give the same response at the measurement temperature. For \( T_s = 28 \).

**FIG. 3.** The relaxation rate, \( S(t) \) vs. time, employing a halt lasting 1000 s in the cooling procedure at different temperatures (a) \( T_s = 34, 32, 30, 29, 28 \) and 27.5 K above \( T_m = 27 \) K and at different temperatures (b) \( T_s = 15, 20, 24, 25, 26 \) and 26.5 K below \( T_m = 27 \) K (b). In both (a) and (b) two reference curves are included that are measured after immediately cooling the sample to \( T_m \) at a cooling rate of 0.5 K/min. and using \( t_w \approx 0 \) and 1000 s, \( h = 1 \, \text{Oe} \).

**FIG. 4.** The relaxation rate \( S(t) \) vs. time, after cooling the sample to a low temperature \( T_s = 20 \) and 24.5 K, halt at this temperature for 0 s or 1000 s and then approach \( T_m = 27 \) K at a heating rate of 0.5 K/min. where the field is immediately applied. A reference curve \( t_w \approx 0 \) measured after cooling the sample directly to \( T_m \) without any halt is also plotted, \( h = 1 \, \text{Oe} \).
K influences of the different cooling rates start to become observable at observation times, \( t > 10^3 \) s. At shorter observation times no sign of the different cooling rates can be resolved. When \( T_s \) gets even closer to \( T_m \), the influence from the differences in cooling rates appears earlier, but still there is a region at shorter observation times where the response is independent of the cooling rate.

For positive temperature shifts, Fig. 5 (b), the result is different. There are clear cooling rate effects for all different temperatures \( T_s \). The curve representing rapid cooling always has a larger magnitude and a sharper maximum that occurs at shorter observation times than the corresponding slowly cooled curve. This result implies that the cooling rate in the region just above \( T_m \) always remains one of the governing parameters for the response after undercooling the sample, i.e. a memory of this cooling process becomes imprinted in the spin structure and is conserved in spite of the re-structuring that occurs at lower temperatures.

**IV. DISCUSSION**

Our current results on the non-equilibrium response function show that the cooling rate significantly influences the measured response function, especially the response at short wait times is dominated by the specific cooling rate employed. Also, in experiments where the sample has been undercooled below the measurement temperature, the cooling rate above the measurement temperature remains one of the governing parameters for the non-equilibrium response. These findings have practical importance for the design of experimental procedures and when making detailed comparisons between results obtained in different experimental set-ups and in different laboratories on similar spin glass materials.

We have, using different experimental procedures, elucidated an apparently paradoxical property of the non-equilibrium spin structure in spin glasses: the spin structure records the cooling history, this thermal history (cooling/heating rate, wait times etc.) remains imprinted in the configuration, and the fragment of the thermal history confined in a rather narrow region close to any higher temperature \( T_m \) within the spin glass phase, can be recovered in a relaxation experiment at \( T_m \). A continuous memory recording occurs on cooling, in spite of the fact that the spin structure is subjected to substantial reconfiguration at all temperatures below \( T_g \) as is observed from the ever present ageing behaviour; and although the short time response appears equilibrated at all temperatures in the spin glass phase.

Employing the droplet scaling model and the concepts chaos and overlaplength, the observations of non-equilibrium and ageing behaviour observed after cooling the sample to a temperature in the spin glass phase can be accounted for as an immediate consequence of the growth of the size of correlated spin glass domains at constant temperature. Interpretations of ageing in these terms have been extensively discussed in numerous articles and are recently reviewed in ref. A somewhat modified version of the droplet scaling model that has been constructed to account for the memory behaviour is extensively discussed in ref. 7.

The continuous memory recording of the thermal history that is exemplified by the current experimental results does however require a comment on the paradoxal possibility that reconfiguration on different length scales are fully separable, i.e. that reconfiguration of the spin structure on short length scales allows a simultaneous stability of the old configuration on larger length scales. The droplet model prescribes one unique equilibrium spin glass state with time reversal symmetry and that domains of spin glass ordered regions grow unrestrictedly with time at constant temperature. If the temperature is altered, the equilibrium configuration also alters due to chaos, but there is an overlap on short length scales.
between the equilibrium configurations, the length scale of which rapidly decreases with increasing separation between the two temperatures. The experimental results show that a memory of the spin structure that has developed at high temperatures remains imprinted in the non-equilibrium structure at lower temperatures (but also that it is rapidly erased if the temperature is increased above the original aging temperature). Such a re-stored spin structure requires that all reconfiguration at lower temperatures must occur only on small length scales and in dispersed regions. The bulk of the numerous droplet excitations of different sizes that do occur may not cause irreversible changes of the spin structure on large length scales, but are to be excited within already equilibrated regions of spin glass order.

How can this rather abstract picture of the dynamic spin structure be related to our current experimental observations? The processes that cause the increase of the magnetisation in a zfc magnetic relaxation experiment is a polarisation of spontaneous droplet excitations. The measured quantity, the zfc magnetisation, gives an integrated value of polarisation of all droplet excitations with relaxation time shorter than the observation time and the measured relaxation corresponds to droplets with relaxation time of the order of the observation time, \( t \). In an ac-susceptibility experiment, the in-phase component gives the integrated value of all droplet excitations with relaxation time shorter than the observation time, \( t = 1/\omega \), and the out-of phase component measures the actual number of droplets of relaxation time equal to \( 1/\omega \). The non-equilibrium characteristics imply that the distribution of droplet excitations changes with the time spent at constant temperature and that there is an excess of droplet excitations of a size that corresponds to a relaxation time of the order of the wait time. On shorter time scales an equilibrium distribution has been attained, reflected by the equilibrium response always obtained in ac-susceptibility experiments at higher frequencies, and in zfc measurements at long but different wait times by the fact that a similar (equilibrium) response is approached at the shortest observation times. The implication of the cooling rate dependence is that the actual distribution of active droplets is governed by the cooling rate and the wait time at constant temperature, and that this distribution in turn is governed by the underlying spin configuration. The phenomenon that the sample retains a distribution of droplets that is governed by the cooling rate and any previous wait time at the measurement temperature, then implies that a closely equivalent spin configuration to the original one is also retained when the temperature is recovered.

V. CONCLUSIONS

We have shown that non-equilibrium dynamics measured at a specific temperature in spin glasses is primarily governed by the thermal history close to this temperature during the cooling sequence. If the sample has been undercooled, the response is also partly affected by the heating rate towards the measurement temperature. The behaviour may be incorporated in a real space picture of a random spin configuration containing fractal spin glass domains of sizes that increase through spontaneous droplet excitations.

The results emphasise the importance of well controlled experimental procedures when studying non-equilibrium dynamics of spin glasses.

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