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Sensitivity to temperature perturbations of the ageing states in a re-entrant ferromagnet

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Abstract. Dynamic magnetic properties and ageing phenomena of the re-entrant ferromagnet $(Fe_{0.20}Ni_{0.80})_{75}P_{16}B_6Al_3$ are investigated by time dependent zero field cooled magnetic relaxation, $m(t)$, measurements. The influence of a temperature cycling (perturbation), $\pm\Delta T$, (prior the field application) on the relaxation rate is investigated both in the low temperature re-entrant spin glass "phase" and in the ferromagnetic phase. In the ferromagnetic phase the influence of a positive and a negative temperature cycle (of equal magnitude) on the response is almost the same (symmetric response). The result at lower temperatures, in the RSG "phase" is asymmetric, with a strongly affected response for positive, and hardly no influence on the response for negative temperature cycles. The behaviour at low temperatures is similar to what is observed in ordinary spin glasses.

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 field of random magnets provides a vast area of challenging problems. One problem of great interest is reentrant magnets where a competition between spin glass and ferro- or antiferromagnetic long range order is present. When the temperature is lowered in such a system, there is a transition from the paramagnetic to a ferro- or antiferromagnetic phase and when further lowering the temperature a transition to a re-entrant spin glass "phase" might take place. This behaviour has been seen in a number of disordered magnetic materials, both ferromagnetic and antiferromagnetic, and it also occurs in mean field theory of random magnets [1].

True re-entrance occurs in a re-entrant ferromagnet if the long range ferromagnetic order eventually disappears at a finite temperature T_{RSG} and is succeeded by a new equilibrium phase, the re-entrant spin glass phase. The problem with spin glasses is that they never reach equilibrium on laboratory (or even geological) time scales thus when the low temperature "phase" in a re-entrant system is of spin glass character, the interpretation of the experimental results becomes hazardous. In previous papers we have reported results from magnetisation and susceptibility measurements on the dynamics of the re-entrant ferromagnet $(Fe_{0.20}Ni_{0.80})_{75}P_{16}B_6Al_3$ [2–4]. Similarities but also distinct differences between the dynamics in the RSG and the FM regions have been found, *e.g.* ageing is found in both the FM and the RSG "phase" [2,3] but an established ageing state is destroyed by much weaker magnetic field perturbations in the FM than in the RSG region [4].

In this paper we report striking differences in how an established ageing state is affected by temperature cyclings (perturbations) in the RSG "phase" as compared to in the FM phase. Results which can be interpreted to support the development of a true spin glass phase at low temperatures.

2 Experimental

In slowly relaxing magnetic systems (*e.g.* spin glasses, disordered ferromagnets etc.), a non equilibrium (ageing) behaviour may be revealed from dc-magnetic relaxation experiments. In such an experiment the sample is cooled from a high temperature, T_{ref}, in the paramagnetic phase, to the measurement temperature, T_{m}, where it is kept a certain wait time, t_{w}. Thereafter a small magnetic field, h, is applied and the magnetic relaxation, $m(t)$, is recorded as a function of time. When this response is dependent on the wait time, the system displays a non equilibrium behaviour, *i.e.* it ages. In this investigation we study how the magnetic response changes when a temperature cycling, ΔT, is made prior to the field application both in the FM phase ($T_{\mathrm{c}} \approx 92$ K) and the RSG "phase" ($T_{\mathrm{RSG}} \approx 15$ K) of the re-entrant ferromagnet $(Fe_{0.20}Ni_{0.80})_{75}P_{16}B_6Al_3$.

The material $(Fe_xNi_{1-x})_{75}P_{16}B_6Al_3$ is an amorphous metal with Ruderman-Kittel-Kasuya-Yosida (RKKY) type of interactions between the magnetic ions. Its magnetic properties are mainly determined by the Fe atoms

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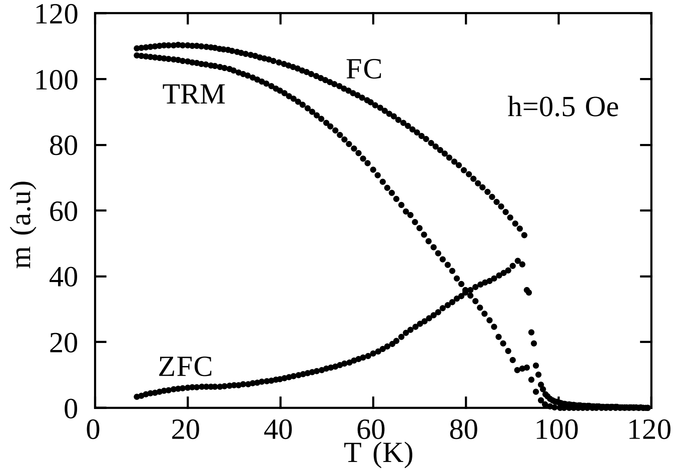

Fig. 1. The zero feld cooled (ZFC), field cooled (FC) and thermo remanent magnetisation (TRM) contours in a field of $h = 0.5$ Oe.

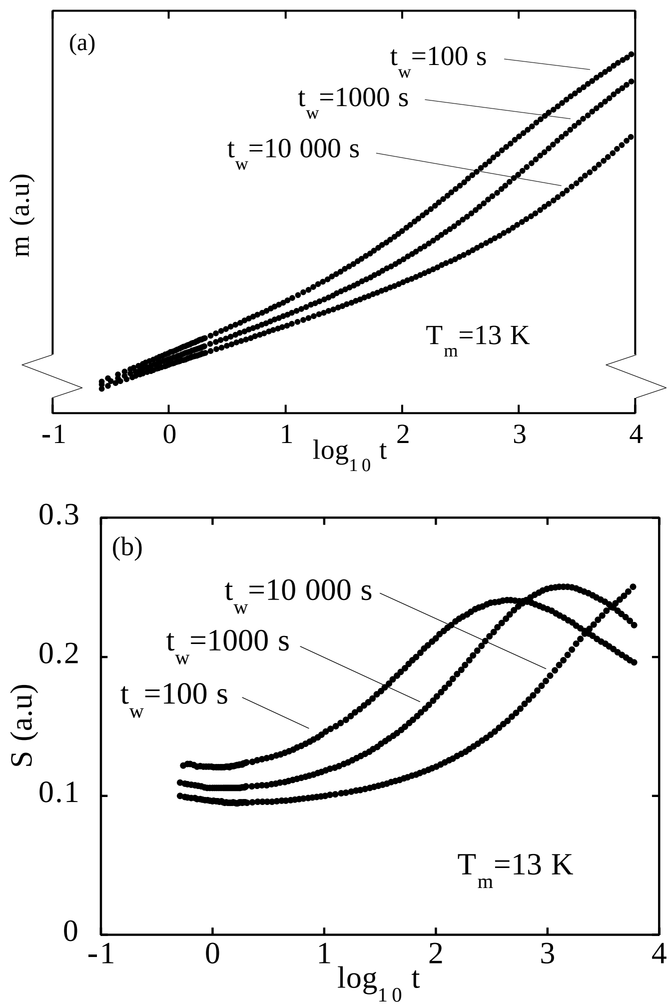

Fig. 2. $\langle m \rangle$ *vs.* $\log t$ (a) and corresponding relaxation rate $S(t) = 1/h\ \mathrm{d}m/\mathrm{d}\log t$ *vs.* $\log t$ (b), for $t_w = 10^2$, 10^3 and 10^4 s, at $T_m = 13$ K and $h = 0.2$ Oe.

since the magnetic moments of the Ni atoms are quenched due to charge transfer from the metalloids [5]. For $x = 0$, *i.e.* the pure Ni alloy, the system is non magnetic and with increasing Fe concentration long range ferromagnetic order sets in at $x = 0.17$. For $x < 0.17$ typical spin glass behaviour is found and for a range of concentrations with $x > 0.17$, re-entrant spin glass behaviour occurs at low temperatures. The sample used in this investigation, $(Fe_{0.20}Ni_{0.80})_{75}P_{16}B_6Al_3$ is in the form of a thin ribbon (cross section 0.01×1.00 mm^2 and length 4 mm). The measurements were performed in a non-commercial superconducting quantum interference device (SQUID) magnetometer [6] with the magnetic field applied along the length of the sample.

3 Results and discussion

Figure 1 shows ZFC, FC and TRM magnetisation *vs.* temperature for an applied field of 0.5 Oe. The onset of ferromagnetic long range order is signalled by a sharp increase of both the ZFC and the FC magnetisation just above the Curie temperature $T_c \approx 92$ K. A corresponding increase of the remanence is also observed, followed by a weak maximum and a continued increase at lower temperatures. A field dependent temperature, $T_{irr}(H)$, where the magnetisation becomes irreversible can be defined at the temperature where the FC and ZFC curves merge. In the ZFC magnetisation a faint maximum is observed at $T \approx 20$ K. This signals the re-entrant spin glass phase that may establish at lower temperatures.

The FC magnetisation remains strongly magnetised throughout the FM phase and also into the re-entrant spin glass “phase” revealing that the aligned ferromagnetic domains sustain even at low temperatures where the possible spin glass phase should appear. This may be interpreted as if the ferromagnetic phase persists also in the low temperature re-entrant spin glass “phase” which may lead to the conclusion that no true re-entrance exists in the material, but only a coexistence of spin glass and ferromagnetic order [7]. On our experimental time scale this indeed seems to be the case, but the crucial question to answer is which is the ground state. This question can however not be answered from experimental observations on a non-equilibrium system at one finite observation time. A dynamic scaling analysis intrinsically contains an extrapolation to infinite time scales and may thus allow some insight as to the nature of the low temperature phase, such an analysis on the current system has been reported elsewhere [2] and is indicative of the existence of a true re-entrant spin glass phase.

In many disordered magnetic systems the response to an applied field is dependent on the time the system has been kept at constant temperature in its “glassy” state. This was first observed in spin glasses [8] but has later also been observed in various other disordered magnetic systems [9]. Such an ageing phenomenon, may *e.g.* be disclosed by time-dependent zero-field-cooled (ZFC) magnetisation measurements at low enough fields. The sample is then cooled in zero field from a reference temperature,

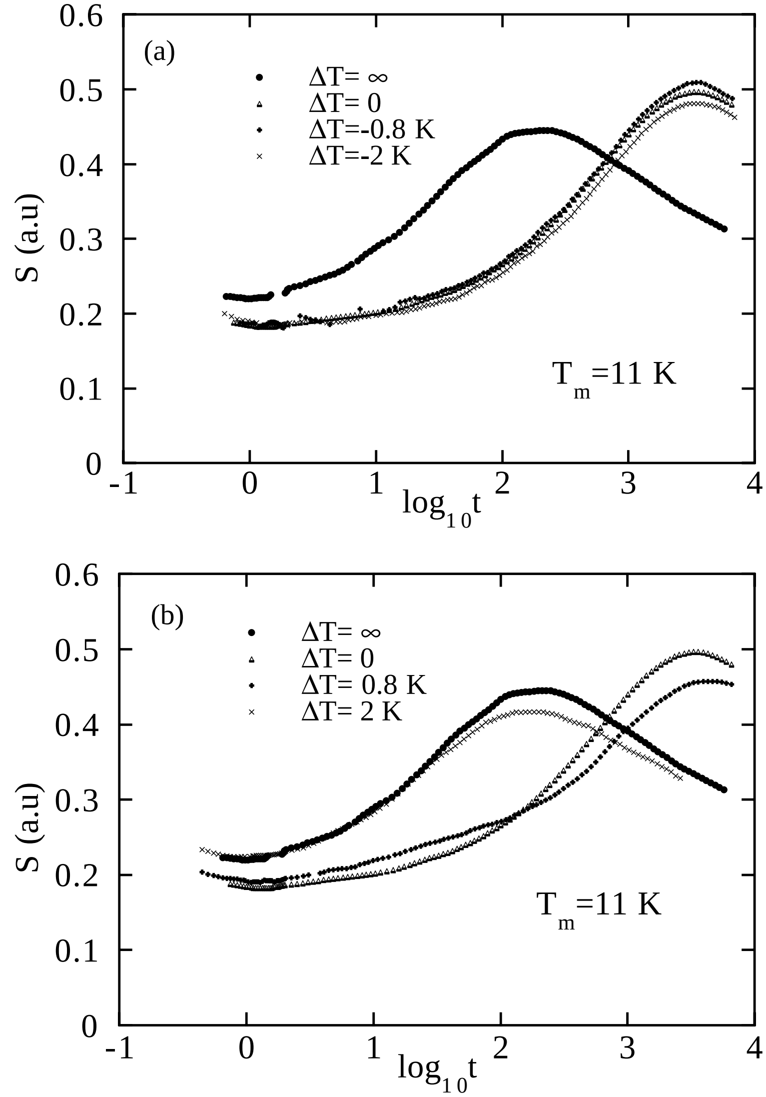

Fig. 3. The relaxation rates $S(t) = 1/h\ \mathrm{d}m/\mathrm{d}\log t$ for (a) negative ΔT's and (b) positive ΔT's at $T_\mathrm{m} = 11$ K and $h = 0.2$ Oe.

here 120 K, to a measurement temperature (T_m), and kept there a wait time (t_w). Thereafter a dc field (h) is applied and the magnetisation is recorded as a function of time. In Figure 2a m is plotted *vs.* log t at $T_\mathrm{m} = 13$ K, $h = 0.2$ Oe, $t_\mathrm{w} = 10^2, 10^3$, and 10^4 s. The corresponding relaxation rate, $S(t)$, is plotted in Figure 2b. At this temperature the system is in its re-entrant spin glass "phase" [2]. A strong wait time dependence is observed. The m *vs.* log t curve has an inflection point where $\log t \approx \log t_\mathrm{w}$ and the relaxation rate attains a corresponding maximum. A rather similar ageing behaviour is also observed in the ferromagnetic phase [3]. The fact that ageing exists in both the FM and the RSG regions may spontaneously be interpreted to suggest that the two phases are dynamically equivalent. However, if there exists a true phase transition, the dynamic behaviour is expected to show some significant distinctions in-between the two phases. One striking change when passing from the RSG to the FM region is an enormous decrease of the magnitude of the magnetic field perturbation that is needed to reinitialise an established ageing state [4]. Here we discuss how an established ageing state is reinitialised due to a controlled temperature perturbation, and again find striking differences in-between the behaviour in the RSG and the FM phase.

Disordered magnetic systems that display an ageing behaviour are affected by temperature cyclings or shifts after the initial wait time [10]. This influence is clearly seen in the magnetic relaxation curves and confirms that the observed ageing effect originates from the chaotic nature of the underlying spin configuration. To examine the differences in the ferromagnetic and in the re-entrant spin glass phases temperature cycling experiments were performed at three different temperatures, $T_\mathrm{m} = 67$ K, $T_\mathrm{m} = 40$ K and $T_\mathrm{m} = 11$ K. The experimental procedure is as follows: first the system is cooled in zero field from the reference temperature, $T_\mathrm{ref} = 120$ K, to the measurement temperature. After a wait time, $t_\mathrm{w} = 3000$ s, the system is subjected to a temperature cycling of magnitude, ΔT, a time

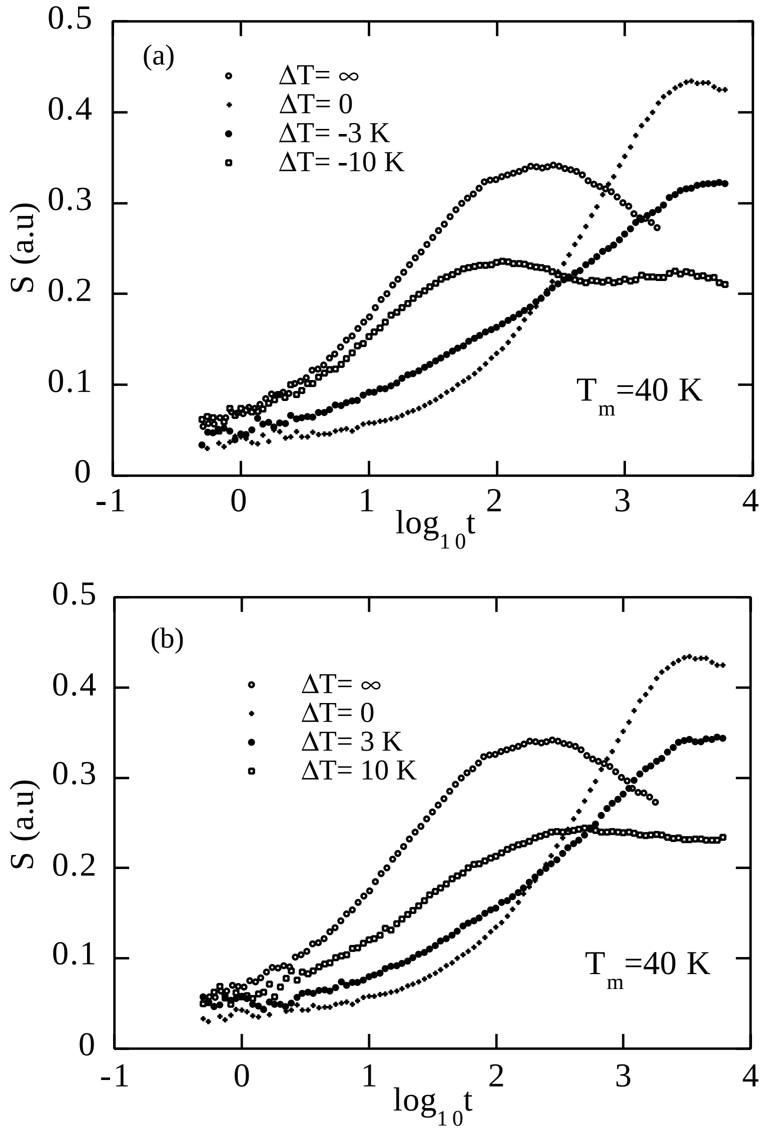

Fig. 4. The relaxation rates $S(t) = 1/h \; \mathrm{d}m/\mathrm{d}\log t$ for (a) negative ΔT's and (b) positive ΔT's at $T_\mathrm{m} = 40$ K and $h = 0.02$ Oe.

$t_{\Delta T} = 10$ s spent at the cycling temperature. The time for heating and cooling is not counted for in this measure. When T_m is recovered the probing field h is applied and the relaxation of the magnetisation is recorded as a function of time.

Figure 3a shows the relaxation rate $S(t)$ measured at $T_\mathrm{m} = 11$ K with $h = 0.2$ Oe for two different magnitudes of negative temperature cyclings, $\Delta T = -0.8$ K and -2 K. The curve marked $\Delta T = \infty$ corresponds to a ΔT that takes the system above T_c. This is the "youngest" curve that is possible to get by cooling directly to T_m and recording the relaxation without wait time. The achieved maximum at $t \approx 100$ s is assigned to an effective wait time governed by the cooling rate and the time allowed for temperature stabilisation when T_m is recovered. The $\Delta T = 0$ curve is the relaxation rate of an ordinary ZFC relaxation curve measured with $t_\mathrm{w} = 3000$ s. The two negative temperature cycles have a very weak influence on the relaxation rate. The magnitude of the maximum at $t \approx 3000$ s is slightly altered compared to the $\Delta T = 0$ K curve. In Figure 3b the result for two positive temperature cyclings with the same magnitude as in the previous figure is presented. The difference is striking. The maximum in $S(t)$ at $t \approx 3000$ s decreases and for $\Delta T = 2$ K the system becomes totally reinitialised, the maximum occurs at a shorter time, almost coinciding with the $\Delta T = \infty$ behaviour. This means that a positive temperature cycle of 2 K, has almost the same effect as cooling the system directly to $T_\mathrm{m} = 11$ K. A similarly asymmetric response for positive and negative ΔT's is observed in ordinary spin glasses [10].

In Figures 4 and 5 the relaxation rate is displayed for $T_\mathrm{m} = 40$ K and $T_\mathrm{m} = 67$ K, in the ferromagnetic phase, for negative (a) and positive (b) ΔT's. The magnitude of the temperature cyclings are chosen so $\Delta T/T_\mathrm{m}$ is comparable at the three different measurement temperatures

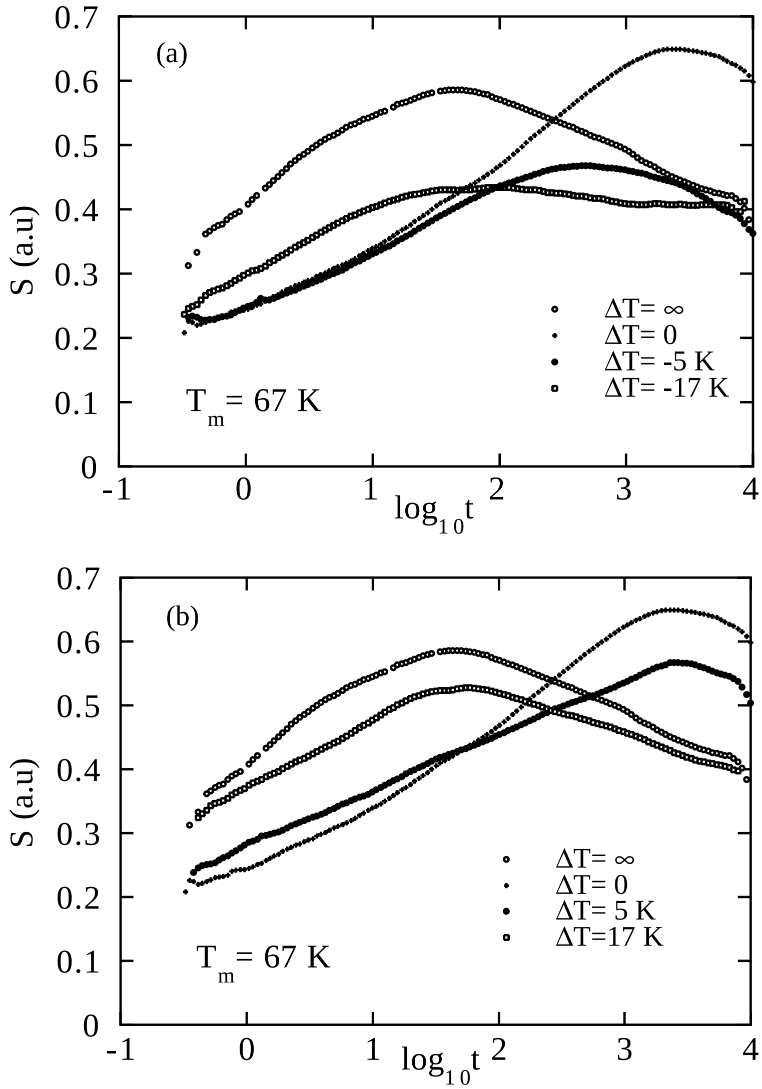

Fig. 5. The relaxation rates $S(t) = 1/h\ \mathrm{d}m/\mathrm{d}\log\ t$ for (a) negative ΔT's and (b) positive ΔT's at $T_\mathrm{m} = 67$ K and $h = 0.02$ Oe.

and for $T_\mathrm{m} = 11$ K and $T_\mathrm{m} = 67$ K $(T_\mathrm{m} - T_\mathrm{RSG})/T_\mathrm{RSG}$ and $(T_\mathrm{m} - T_\mathrm{c})/T_\mathrm{c}$ are equally large. Here a probing field of $h = 0.02$ Oe is used which is low enough to probe the intrinsic dynamics of the system [4]. Both the positive and negative temperature cycles reinitialise the system. The response is symmetric, positive and negative ΔT's of equal magnitude have qualitatively the same effect on the relaxation rate in the ferromagnetic phase. The response is somewhat less symmetric at the higher measurement temperature and for $\Delta T = 17$ K the system gets strongly reinitialised. The large positive ΔT takes the system to a temperature in the vicinity of $T_\mathrm{c} \approx 92$ K, namely $T_\mathrm{m}+\Delta T = 84$ K where the dynamical processes start to get critical. A more effective reinitialisation is then achieved.

This is in contrast with the response in the RSG phase, where the result on the relaxation rate is highly asymmetric for different signs of the ΔT's. Another notable difference in the two phases is that it is easier to reinitialise the system with a positive ΔT in the RSG phase than in the FM phase. The RSG phase is stable against negative ΔT's but extremely sensitive to positive ΔT's. The ferromagnetic phase is equally stable for positive and negative ΔT's and is more difficult to reinitialise completely if measuring at temperatures where there is no influence of the complex behaviour close to T_c.

4 Conclusions

We have studied some dynamic and static properties of the re-entrant ferromagnet $(Fe_{0.20}Ni_{0.80})_{75}P_{16}B_6Al_3$. The ZFC-FC protocol results in an irreversibility temperature, T_irr, that coincides with the onset of long range ferromagnetic order, $T_\mathrm{c} \approx 92$ K. The FC magnetisation remains highly magnetised throughout the FM phase and into the RSG phase revealing that ferromagnetic domains are present even at low temperatures at our experimental time scales.

The relaxation rate, $S(t)$, displays distinct differences for the FM and the RSG regions when the system is exposed to a temperature cycle prior the field application. In the RSG "phase" the system behaves like an ordinary SG with an asymmetric result for negative and positive ΔT's. The underlying spin configuration is very unstable against positive ΔT's but looks stable against negative ΔT's, if the duration of the cycle is constant and "short", $t_{\Delta T} \approx 10$ s. In the FM region, the results for $\pm\, \Delta T$'s are symmetric. The ferromagnetic phase is equally stable to positive and negative ΔT's. The FM phase is more difficult to completely reinitialise than the "RSG" phase if a positive ΔT is used. The fact that there is a re-initialisation (only) when large enough temperature perturbations are used, support that there is chaos between the states attained at different temperatures and that there is an overlap between states. One conclusion to be drawn from these results is that the spin glass phase has barrier heights for domain growth that increases with decreasing temperature, whereas in the ferromagnetic phase the domain growth is governed by thermal activation over barriers of constant height.

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