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Quasi-archetypal spin-glass freezing at the paramagnetic–ferromagnetic boundary in re-entrant spin glasses

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Abstract. The differential AC magnetoresistance and differential AC susceptibility of three amorphous FeNi re-entrant spin glasses have been examined as a function of temperature at the so-called paramagnetic–ferromagnetic transition under static magnetic fields up to 70 Oe. It is shown that the static field tends to nullify the effect of the ferromagnetic component and the results indicate the presence of spin-glass freezing in both the magnetoresistance and susceptibility which takes place simultaneously with the establishment of the ferromagnetic phase at the Curie temperature. The composite nature of the transition appears to be a general feature of re-entrant spin glasses.

1. Introduction

The re-entrant spin glasses (SGs) occupy an interesting compositional range between archetypal pure SGs with short-range exchange interactions and long-range interactions characteristic of the ferromagnetic range. In this range there is competition between the SG couplings and ferromagnetic (FM) couplings. Within the mean-field theory, if the parameter J defined by

$$J^2/N = [J_{ij}^2]_{av} - [J_{ij}]_{av}^2$$

is larger than J_0 defined by

$$J_0/N = [J_{ij}]_{av}$$

where N is the number of moments and J_{ij} the exchange interaction between moments i and j, then an SG obtain on the other hand if $J_0 > J$ a ferromagnetic state exists which can degenerate into an SG state at lower temperatures. However, in many practical cases the FM state is not clearly indicated and distinction between FM and superparamagnetism (SP) is often difficult; field induced ferromagnetism can occur and unambiguous interpretation of Arrot plots becomes a problem. Ideally, experiments need to be performed in very low fields. For this reason, among others, we have been involved in developing a very low-field magnetoresistance apparatus [1] to assist in the study of re-entrant SGs; this is essentially an AC magnetoresistance (ACMR) technique and has been used to supplement AC susceptibility (ACS) measurements which, of course, also involve the application of very low magnetic fields. ACMR studies are particularly useful here for, as we have shown in a number of publications [2, 3] the sign of the ACMR is sensitive to the type of magnetic coupling which obtains; in particular, archetypal SG coupling always results in negative MR

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while in the FM state it is always positive. In contrast, the ACS is always positive, a situation which in general gives no clear indication for the separation of FM and SG couplings.

In real re-entrant SGs there is also the possibility of co-existence of FM/SP and SG couplings below the Curie temperature. In an earlier paper [2] we have suggested that the so-called PM–FM transition in re-entrant SGs is also accompanied by pure SG freezing as well. The evidence for this was based on our very low-field MR measurements where, as the temperature approached the Curie temperature T_C, there was a curious sign change from positive to negative which continued over a few degrees and then decayed to zero as T_C was reached. In our studies over many years, no exceptions to the rule that SG freezing gives rise to negative MR have been found. Although not definitive, we have regarded, as a logical first step, the occurrence of negative MR as a strong indicator that SG coupling is taking place in these re-entrant SGs. At the time there was nothing in the magnetization or ACS to support this proposal; both these properties acquired very large values at T_C associated with the FM or SP states which set in as T was lowered. In this paper we will show that the ACS does in fact reveal evidence of SG freezing but in previous studies its effect has been swamped by the overpowering influence of the FM or SP phase.

During the course of our earlier investigations, it also became clear that the FM/SP component was extremely sensitive to any applied external DC magnetic field and that only a few Oersteds could reduce the ACS by as much as two orders of magnitude to rather low values. Thus it appeared as if the FM/SP component could be readily saturated by such small fields, thereby nullifying its contribution to the differential ACS (DACS) and the differential ACMR (DACMR). This suggested that any SG component, which is much less sensitive to external static fields, might be revealed unhindered by the normally dominant effect of the FM/SP component. In this paper we report the results of such a study on the DACMR and DACS on three samples of amorphous FeNi alloys all of which exhibit re-entrant behaviour, one of them being close in composition to the pure SG boundary.

2. Experimental details

Three samples of amorphous $(Fe_{1-x}Ni_x)_{77}Si_{10}B_{13}$ with values of x of 0.875, 0.85 and 0.80 have been employed in this investigation all of which exhibit re-entrant behaviour with nominal Curie temperatures of 98, 150 and 230 K respectively. The magnetic phase diagram [4] shows that the sample with $x = 0.875$ is near the pure SG–re-entrant SG boundary while $x = 0.85$ and 0.80 are well within the re-entrant regime.

The low-field MR was measured with a unique apparatus, developed in this laboratory and described in an earlier paper [1], where the excitation field was a pulsed square wave. For the DACMR employed here, the apparatus was modified for a sinusoidal excitation field, details of which have been reported in a further publication [5]. The samples were in the form of ribbons 60 mm long, 2 mm wide and approximately 0.05 mm thick with the magnetic field directed along the long axis. In this configuration the demagnetizing factor was negligible.

The ACS was measured by a conventional susceptometer which was also capable of measurement in the presence of an external static DC field produced by a surrounding solenoid. For these measurements the samples were 10 mm long but their width was reduced to 0.4 mm to ensure a low demagnetizing factor and four pieces were mounted in each case in the sample holder.

In both the MR and ACS measurements the errors associated with induced AC currents from the excitation coils into the surrounding DC solenoids were eliminated by a method described by Barnard [6] and all measurements were performed at a frequency of 511 Hz.

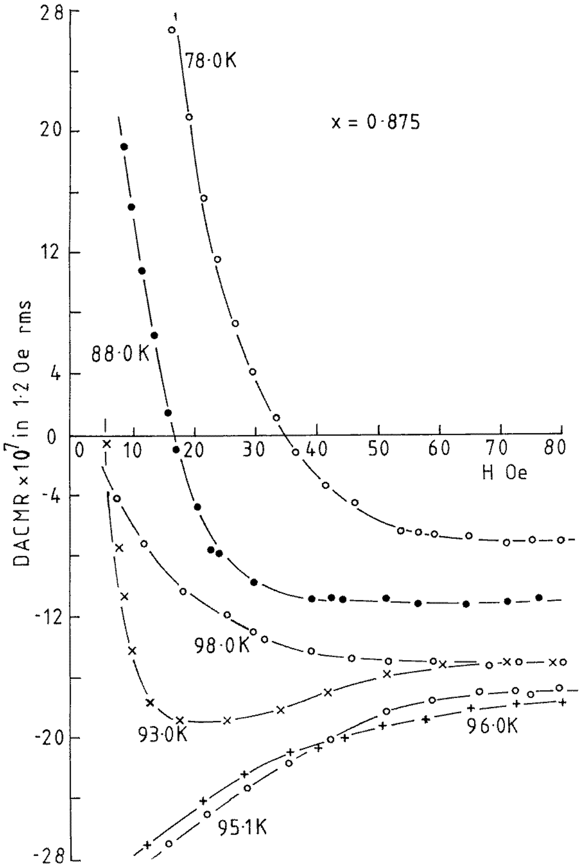

Figure 1. The DACMR of the morphous sample $(Fe_{1-x}Ni_x)_{77}Si_{10}B_{13}$ ($x = 0.875$) as a function of static magnetic field at various temperatures below the Curie temperature. The oscillating magnetic field was 1.2 Oe.

3. Results

In figure 1 is shown the variation of the DACMR of the sample with $x = 0.875$ in an oscillating field of 1.2 Oe as a function of static magnetic field at various temperatures above 78 K. At 78 K and in small static fields the DACMR is positive as a result of the dominant effect of the FM component, but with increasing static field the DACMR changes sign to become negative and saturation sets in beyond 60 Oe. At higher temperatures the positive DACMR contribution decreases and for $T > 96$ K DACMR is negative in all static fields. At all temperatures between 77 and 100 K saturation of the DACMR occurs for static fields greater than 60 Oe. As mentioned in the introduction, it is clear that these low static fields tend to saturate the FM component, leaving a magnetoresistance associated with what we believe is solely a SG contribution.

The variation of the DACMR in 25 and 70 Oe as a function of temperature for $x = 0.875$ is shown in figure 2. The negative saturation values in 70 Oe are typical of a conventional SG with a freezing temperature of 97 K. In 25 Oe the negative peak occurs at the slightly lower temperature of 95.5 K with a more pronounced peak. It seems likely that two effects

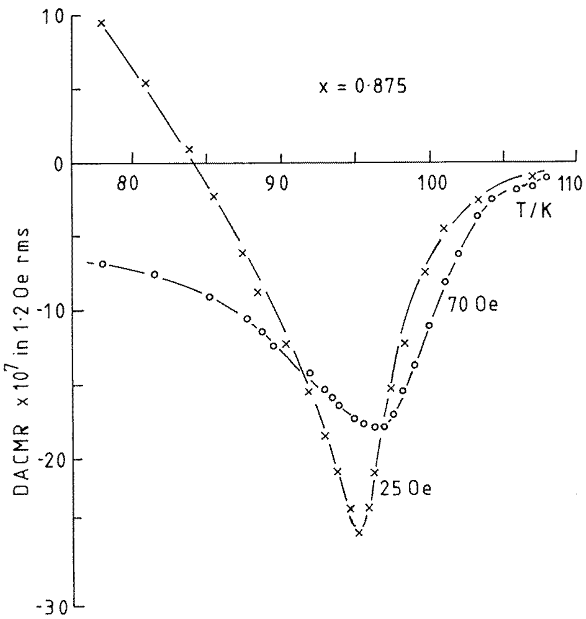

Figure 2. The DACMR of the sample with $x = 0.875$ as a function of temperature in static fields of 25 and 70 Oe.

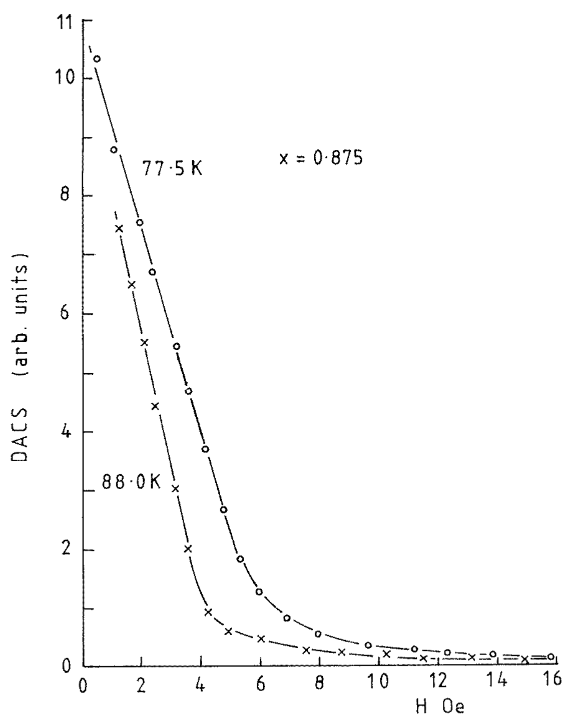

Figure 3. The DACS of the sample with $x = 0.875$ as a function of static field at 77.5 and 88 K.

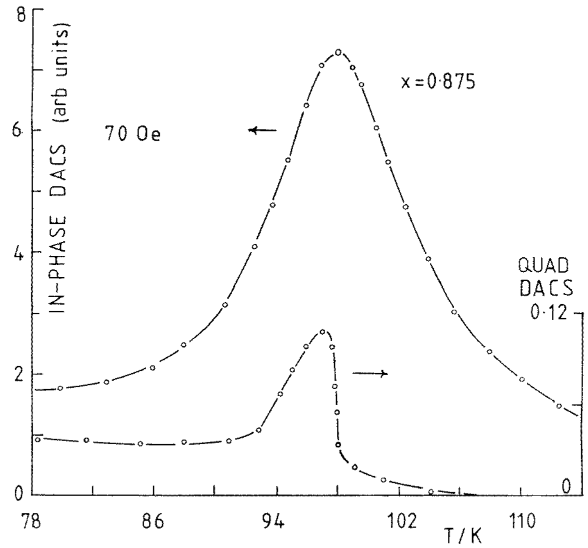

Figure 4. The temperature variation of the in-phase and quadrature DACS for the sample with $x = 0.875$ under a static field of 70 Oe.

are occurring here: firstly the lower static field allows a greater positive FM contribution to obtain but this, acting alone, would produce *lower* absolute values of the DACMR in this region instead of the *larger* values observed. However, just as in the ACS where the SG peaks are attenuated and rounded by larger static fields, so this is likely with the DACMR and gives rise to a larger and more pronounced peak in 25 Oe than that in 70 Oe. It would appear that this effect around 95–98 K is much greater than that of the FM contribution but clearly the FM contribution dominates below 84 K where the DACMR is entirely positive.

Figure 3 shows the effect of a static magnetic field on the DACS. The effect here is much greater than in the case of the DACMR where at 77.5 and 88 K fields as low at 16 Oe reduced the susceptibility by two orders of magnitude. In figure 4 is shown the temperature variation in 70 Oe of the DACS for $x = 0.875$ where it is apparent that a typical peak characteristic of an SG is observed with a freezing temperature, 98 K, almost identical to that observed in the DACMR. There was also a small discontinuity in the quadrature DACS also typical of a conventional SG. Thus the anomaly observed in the DACMR which we have reported on earlier as a SG transition is now observed in the DACS with characteristics similar to those of a conventional SG. However, in contrast to ordinary SGs [7], we could detect no frequency dependence of the DACS when measured over the frequency range 20–1600 Hz. It may be that the presence of the static field supresses the frequency dependence.

These results were obtained on a sample close to the accepted compositional boundary between pure SG and re-entrant SG behaviour. Further experiments were conducted with two other alloys ($x = 0.85$ and $x = 0.80$), which lie deeper into the re-entrant regime and where it might be expected that any SG freezing might be absent or, at least, greatly reduced. In figure 5 for the DACMR, where the results for all three alloys are shown, it is clear that the negative DACMR is just as prominent for $x = 0.85$ and 0.80 as for $x = 0.875$. Likewise, in figure 6, the DACS for $x = 0.85$ and 0.80 shows exactly the same phenomena as $x = 0.875$ with freezing temperatures of 150 and 232 K respectively. Also plotted in figure 6 is the temperature variation of the susceptibility for $x = 0.80$ in zero static field

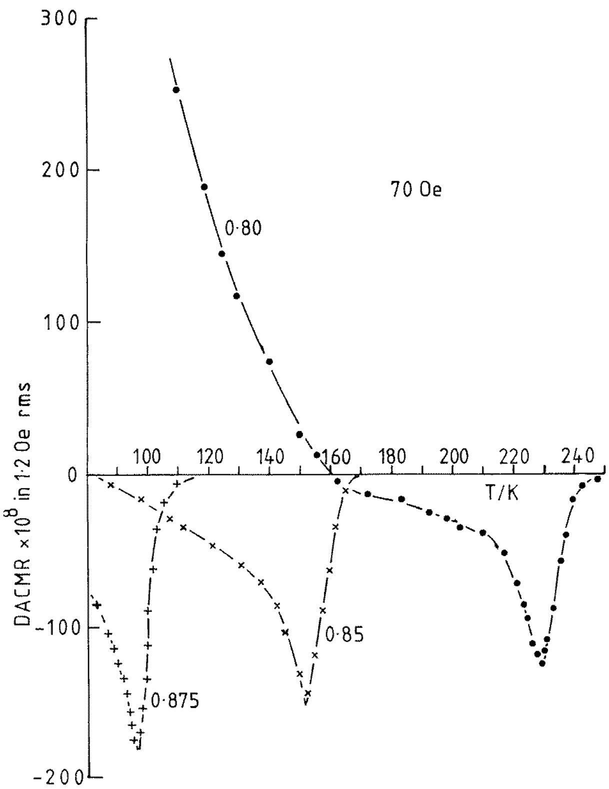

Figure 5. The DACMR in 70 Oe for three samples of amorphous $(Fe_{1-x}Ni_x)_{77}Si_{10}B_{13}$ with x values of 0.875, 0.85 and 0.80.

and thus where the FM component is present. Here values some 200 times greater than those in 70 Oe are observed and thus completely mask the SG component.

4. Discussion

The results presented in figures 1–6 clearly show new phenomena occuring in both the DACMR and DACS once the overpowering effect of the FM component has been nullified by the application of a modest static field. There can be no doubt that the ACS peaks are closely associated with those of the negative DACMR and appear to be completely consistent with those of a conventional SG. Furthermore, it is apparent that this SG freezing occurs at temperatures which coincide with the Curie temperature for the formation of the FM state. Thus it appears that as the temperature is reduced in the paramagnetic phase towards the ordering temperature, there occur parallel moment couplings which result in the formation of the FM phase, but at the same time conditions exist for frustration for some moments which couple into the random frozen state of a conventional SG. It should be pointed out that this phenomenon is not just associated with the amorphous Fe–Ni system under discussion here; there are already indications that similar effects take place, for example, in crystalline FeNiMn alloys [3], where we have seen the negative peak in the DACMR and the incipient onset of a peak in the DACS where some measurements were made in small static magnetic fields. We have also reported on the similar existence of negative MR in the

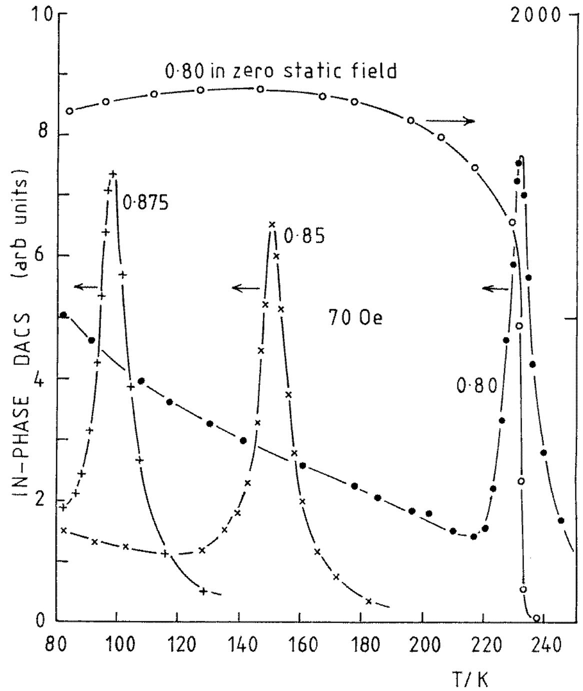

Figure 6. The variation of the DACS in 70 Oe for three samples of $(Fe_{1-x}Ni_x)_{77}Si_{10}B_{13}$ with x values of 0.875, 0.85 and 0.80. Also shown is the variation of the ACS in zero static field for the sample with $x = 0.80$. The results for the three samples have been normalized to the same sample weight.

Au–Fe re-entrant system [2] at the Curie temperature, which strongly suggests that a SG peak in the DACS would be observed if measurements in a static magnetic were performed. Thus simultaneous FM and SG couplings appear to be quite a general feature of so-called re-entrant SGs at the conventional Curie temperature.

The effect of the static magnetic field is considerably greater in the case of the ACS than in the ACMR as is shown in figures 1 and 3. If it is assumed that the MR, $\Delta\rho$, within the FM phase is proportional to the magnetization M squared, i.e.

$$\Delta\rho = AM(H)^2$$

where A is a constant then

$$\frac{\partial\Delta\rho(H)}{\partial H} = 2M(H)\frac{\partial M(H)}{\partial H} \tag{1}$$

where $\partial M(H)/\partial H$ is the in-phase differential susceptibility at $M(H)$. Since $M(H)$ is an increasing function of H and $\partial M(H)/\partial H$ a strongly decreasing function (figure 3), it is evident that $\partial\rho(H)/\partial H$ will decrease less strongly than $\partial M(H)/\partial H$. In the limit of high static field, the magnetization M of the FM phase (equation (1)) will saturate and the FM differential susceptibility $\partial M(H)/\partial H$ will become zero. Equation (1) then shows that $\partial\Delta\rho(H)/\partial H$, the DACMR of the FM phase, becomes zero also. As a result only any SG component will then be available for measurement. Whilst this component is also attenuated by the static field [8], the effect is not strong compared with the FM phase and is clearly still detectable as our results show.

The occurrence of a Curie temperature for the FM component and a freezing temperature for the SG component occuring at the same temperature is an interesting phenomenon and suggests the two are inter-related, possibly one effect triggering the other. One could visualize, as the Curie temperature is approached, a process whereby a set of randomly distributed paramagnetic moments first results in parallel coupling between favourably positioned moments; the increase in the local magnetic field modifies the RKKY polarization of the conduction electrons, resulting in other moments experiencing competing interactions or frustration effects, thereby simultaneously locking into a frozen SG phase at the same temperature.

The results presented in this paper concern the magnetic phenomena taking place at the so-called PM–FM boundary in re-entrant SG systems where we believe SG freezing occurs as well. A further, and possibly more complex, problem concerns the processes taking place at still lower temperatures, where the collapse of the FM phase to a SG phase is conventionally thought to take place. Further results on the DACS and DACMR at this transition will be reported in a later paper.

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