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A comparison of relaxation dynamics in the spin-glass and re-entrant phases of Ni–Mn

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Abstract. The decay of the low-field thermoremanent magnetization has been measured over four decades of observation time, as a function of temperature and wait time, in both the re-entrant phase of a strongly bond-disordered Ni–23.5 at.% Mn ferromagnet and within the ordered phase of an Ni–25 at.% Mn spin glass. The relaxation isotherms exhibit virtually identical systematics in the two phases; on a logarithmic time perspective, the isotherms are characterized by an inflection point which shifts from longer to shorter observation times with increasing temperature and which is sensitive to the length of time that the system is held at a constant temperature prior to removing the field. Moreover, both sets of isotherms are accurately described, over the entire experimental time window, by a common relaxation function consisting of the superposition of a stretched exponential and a constant, namely $M_R(t) = M_0 + M_1 \exp[-(t/\tau)^{1-n}]$, with best-fit parameters n and τ typical of canonical spin-glass behaviour. This study represents the first systematic comparison of relaxation dynamics in the re-entrant and spin-glass phases and provides compelling evidence for the equivalence of the two ordered states.

1. Introduction

Over a restricted portion of their magnetic phase diagrams, magnetic systems with random exchange due to some form of quenched structural disorder appear to exhibit sequential magnetic transitions [1], in which the system evolves with decreasing temperature from a paramagnet to a ferromagnet and then to a ground-state configuration with spin-glass-like characteristics. This 're-entrant' behaviour is accompanied by a distinctive experimental signature [1]; upon cooling, the zero-field dynamic response first increases rapidly, then passes through a typical ferromagnetic Hopkinson maximum, followed by a weakly temperature-dependent 'plateau', and then abruptly decreases as the ferromagnetic phase collapses into the 're-entrant' phase. While mean-field vector spin models [2] for bond-disordered magnets suggest a possible correlation between the re-entrant transition and the onset of transverse spin-glass freezing, no compelling experimental evidence for genuine critical behaviour at the re-entrant phase boundary currently exists, and the microscopic details of the ordered spin configuration within the re-entrant phase remain unresolved. Furthermore, while the viscous properties of spin glasses have received considerable experimental and theoretical attention comparatively little effort has been focused on the characterization of analogous relaxation phenomena in random ferromagnets, particularly in their re-entrant configuration.

Spin glasses exhibit highly unusual relaxation dynamics [3] which indicate that, in the ordered phase, true equilibrium is essentially unattainable within normal laboratory time scales. If the system is cooled rapidly through T_{SG} to a temperature $T < T_{SG}$ and a time t_w is allowed to elapse at constant temperature before the system is probed, then, for observation times t such that $\ln t \ll \ln t_w$, quasi-equilibrium relaxation occurs through local spin rearrangements within effectively equilibrium domains while, for $\ln t \gg \ln t_w$, the relaxation is dominated by non-equilibrium processes related to domain growth. According to the phenomenological theory of Fisher and Huse [4], both types of dynamics are governed by activation over energy barriers which grow with increasing length scale, leading to an extremely slow logarithmic decay of the temporal spin correlations in both regimes, but characterized by different exponents. Experimentally, the essential features of remanent relaxation in spin glasses can be *approximately* [5] modelled (although not without some theoretical justification [6]) by a stretched-exponential time dependence $m(t) \sim \exp[-(t/\tau)^{1-n}]$ with $n < 1$ and with $\tau \simeq t_w$, so that, on a logarithmic time perspective, the crossover between equilibrium and non-equilibrium relaxation coincides with an inflection point in the relaxation curve at $\log\, t_{\rm infl} = \log\, \tau \simeq \log\, t_w$ and hence with a maximum in the relaxation rate $S(t) \equiv -\partial m/\partial(\ln t)$. In this paper, we present a comparison of thermoremanent relaxation in the spin-glass and re-entrant phases of Ni–Mn, and we show that, within the limitations of an analysis based on the approximate stretched-exponential representation, these two phases exhibit essentially *identical* relaxation dynamics.

2. Experimental details

Two alloys of Ni–Mn, one a spin glass containing nominally 25 at.% Mn and the other a re-entrant ferromagnet containing nominally 23.5 at.% Mn, were prepared by arc melting appropriate amounts of 99.997% pure Ni foil and 99.99% pure Mn flakes on the water-cooled copper hearth of an argon arc furnace using a tungsten electrode. Each alloy was repeatedly inverted and remelted in order to achieve homogeneous consistency. A portion of each of the original ingots was cold rolled into a thin sheet, and two long thin needle-shaped samples with dimensions 9.7 mm × 0.3 mm × 0.2 mm for Ni–25 at.% Mn and 10.1 mm × 0.2 mm × 0.2 mm for Ni–23.5 at.% Mn, were prepared from the sheets. Each needle was encapsulated in a quartz tube in an argon atmosphere, annealed for 3 d at 900 °C and then quenched rapidly into water, in order to generate an atomically disordered state with a minimum of strongly ferromagnetic Ni_3Mn phase [7]. The static magnetization and remanent relaxation measurements were performed with a variable-temperature variable-frequency SQUID susceptometer, which has been described in detail elsewhere in the literature [8].

3. Data analysis and discussion

The two alloys investigated here are located close to, but on either side of, the multicritical point ($c_m \simeq 24$ at.% Mn), and both have been magnetically characterized, in earlier studies on samples of nominally the same concentrations as these, by measurements of the dynamic response at an excitation frequency ν of 16 Hz. The Ni–23.5 at.% Mn sample is a re-entrant ferromagnet [9], with triple-peaked isofields consisting of a ferromagnetic critical peak, with a field-dependent amplitude and temperature governed by critical exponents γ and δ which exhibit field- and temperature-dependent systematics indicative of substantial bond disorder, and two re-entrant peaks, with significant irreversibility apparent in the lower temperature component, while the Ni–25 at.% Mn sample is a concentrated spin glass [8], with no evidence of ferromagnetic critical peaks and with susceptibility isofields consisting of a single peak which is systematically suppressed in amplitude and shifted downwards in temperature with increasing field.

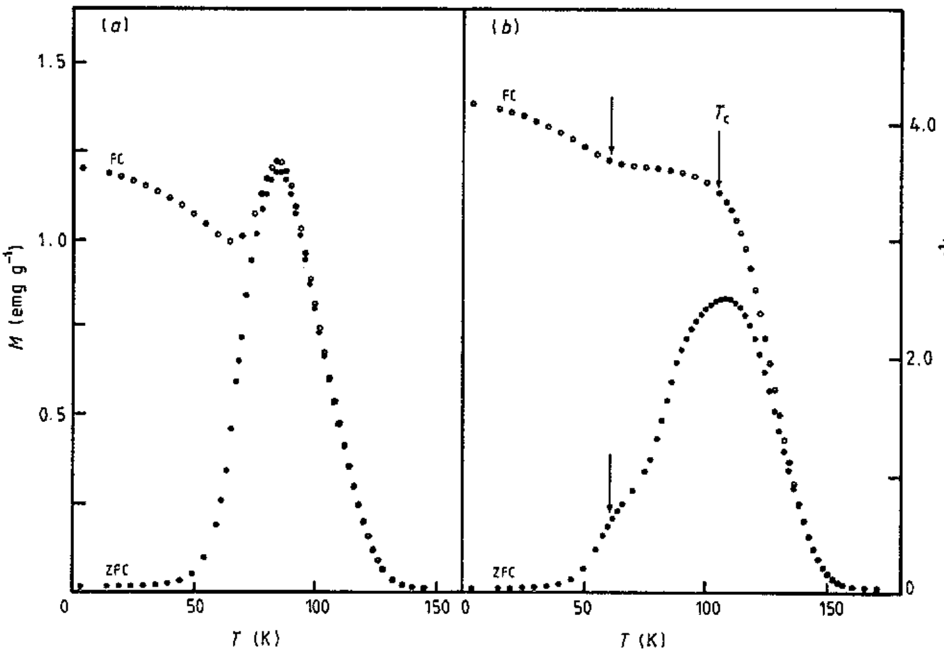

Figure 1. (*a*) Temperature dependence of the FC and ZFC magnetization of Ni–25 at.% Mn measured in an applied field H_a = 1.0 Oe. (*b*) Temperature of the FC and ZFC magnetization of Ni–23.5 at.% Mn measured in H_a = 1.0 Oe. The vertical arrows near T = 60 K mark the appropriate location of the re-entrant transition.

Figure 1 shows the temperature dependence of the static magnetization of both samples, measured under both field-cooled (FC) and zero-field-cooled (ZFC) conditions, in a static applied field H_a = 1.0 Oe. The spin-glass sample in figure 1(*a*) exhibits a single, essentially reversible peak, below which the FC and ZFC curves diverge and time effects become appreciable. By contrast, the re-entrant sample in figure 1(*b*) shows evidence of significant irreversible behaviour over a temperature interval which *includes* the peak in the ZFC magnetization and persists up to $T \simeq$ 130 K; the crossover between the ferromagnetic and re-entrant phases is visible in the FC magnetization as a rapid change in curvature in the vicinity of $T \simeq$ 60 K and, in the ZFC magnetization, as a weak shoulder below the principal maximum (vertical arrows in figure 1(*b*)).

Our first goal was to characterize the viscous properties of the spin-glass state, and figure 2(*a*) shows several representative thermoremanent relaxation isotherms for the Ni–25 at.% Mn spin glass plotted on a logarithmic time scale. Each isotherm was obtained by following an identical experimental procedure; the sample was first

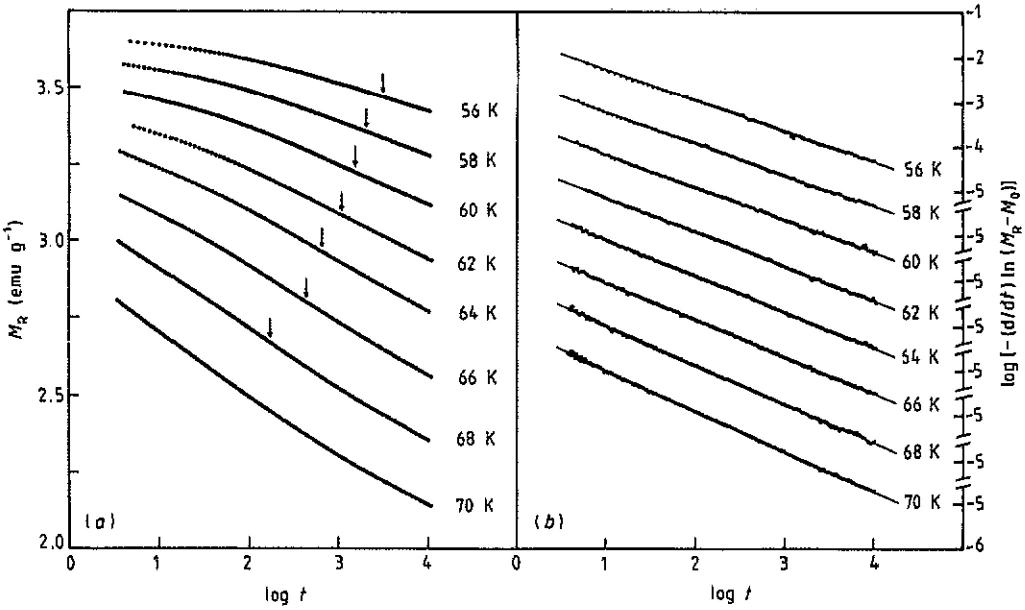

Figure 2. (*a*) A set of relaxation isotherms for Ni–25 at.% Mn all corresponding to a wait time t_w = 60 s. The vertical arrows mark the locations of the inflection points. (*b*) Double-logarithmic plots of $-(d/dt)\ln(M_R - M_0)$ as a function of t for isotherms in (*a*).

warmed to a reference temperature T_R = 140 K, well within the paramagnetic regime, in order to eliminate any remanent magnetization and then cooled in a static field H_c = 1.0 Oe to a predetermined measuring temperature T_m within the ordered phase (the cooling time t_c was essentially independent of T_m, and consistently in the range t_c = 975 ± 45 s); after a 'wait time' t_w = 60 s had elapsed at constant temperature T_m, the field was abruptly removed, and the decay of the thermoremanent magnetization was recorded over nearly four decades of observation time: $1\ \mathrm{s} < t \leqslant 10^4$ s. The most prominent feature of the relaxation curves in figure 2(*a*), when viewed on a logarithmic time perspective, is the inflection point, which propagates from longer to shorter times with increasing temperature, leading to a gradual and systematic change in curvature from concave downwards at low temperatures to concave upwards at high temperatures. (The locations of the inflection points in figure 2(*a*) were established by plotting the local slopes of the isotherms, or relaxation rate, $S(t) \equiv -\partial M_R/\partial(\ln t)$, on a logarithmic time scale, thus generating well defined maxima centred at $\log t_{\mathrm{infl}}$.) Another significant feature of all the relaxation isotherms in figure 2(*a*) is their dependence on the wait time t_w for which the system is held at a constant temperature prior to changing the field. As a typical illustration of this behaviour, figure 3 shows a comparison of three relaxation curves, all measured at the same temperature T_m = 66 K, but for different wait times t_w = 60, 360 and 1800 s; the non-equilibrium character of the relaxation is readily apparent in this figure as a shift in the location of the inflection point towards longer observation times with increasing wait time. In order to generate the most suitable functional description of the relaxation isotherms in figure 2(*a*), the data were compared with some of the more common theoretical representations of spin-glass relaxation, individually and in combination; the simplest formulation which was capable of accurately reproducing all the structural features of the experimental data, over the entire observational time window, consisted of the superposition of a stretched exponential and a constant term: $M_R(t) = M_0 + M_1 \exp[-(t/\tau)^{1-n}]$. Figure 2(*b*) shows that double-logarithmic plots of $-(d/dt)[\ln(M_R - M_0)]$ as a function of t yield excellent straight lines, and an inspection of table 1, which lists the values of the corresponding best-fit parameters, shows that the exponents n and characteristic times τ exhibit canonical spin-glass behaviour [10].

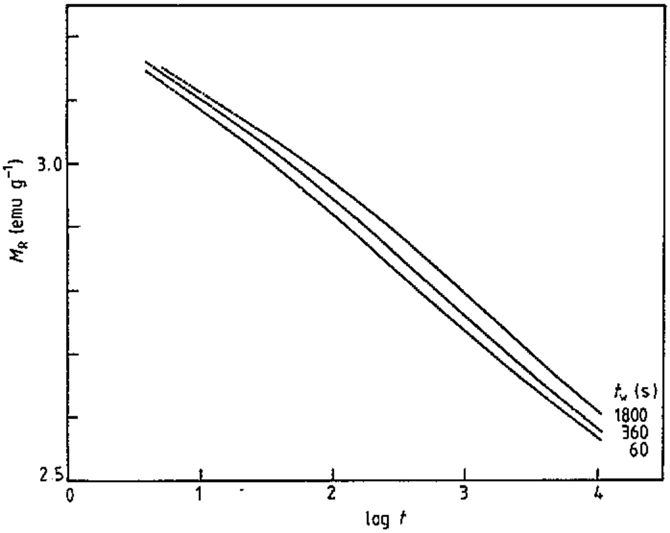

Figure 3. An illustration of the influence of the wait time t_w on a typical relaxation isotherm T_m = 66 K for Ni–25 at.% Mn.

Table 1. Best-fit parameters for Ni–25 at.% Mn for t_w = 60 s.

T(K)	M_0(emu g^{-1})	n	τ(s)
56	3.35 ± 0.01	0.68 ± 0.02	(2.7 ± 0.7) × 10^3
58	3.19 ± 0.01	0.71 ± 0.01	(2.1 ± 0.3) × 10^3
60	3.00 ± 0.02	0.74 ± 0.02	(1.7 ± 0.6) × 10^3
62	2.78 ± 0.02	0.77 ± 0.01	(1.5 ± 0.3) × 10^3
64	2.58 ± 0.02	0.80 ± 0.01	(8.7 ± 1.0) × 10^2
66	2.36 ± 0.02	0.82 ± 0.01	(4.0 ± 0.2) × 10^2
68	2.10 ± 0.04	0.87 ± 0.01	67 ± 14
70	1.83 ± 0.04	0.91 ± 0.01	1.4 ± 1.0

Having established a quantitative picture of spin-glass relaxation in Ni–Mn, we now turn our attention to the re-entrant Ni–Mn ferromagnet, and figure 4(*a*) summarizes the relaxation dynamics at four representative temperatures T_m within the re-entrant phase ($T \leqslant 60$ K) of the Ni–23.5 at.% Mn sample. The measurement procedure was identical with that employed for the spin-glass sample, although the tendency for residual relaxation effects to persist well above the peak in the ZFC magnetization necessitated the choice of a somewhat higher reference temperature T_R = 170 K, for the re-entrant ferromagnet (with, however, comparable cooling times t_c =

900 ± 60 s). A comparison of figure 4(*a*) with figure 2(*a*) shows that the systematics of the thermoremanent decay within the re-entrant phase of the ferromagnet are virtually identical with those of the 'pure' spin glass, with the re-entrant isotherms also characterized by inflection points which shift from longer to shorter observation times with increasing temperature, and which exhibit a similar sensitivity to the wait time t_w (compare the T_m = 55 K isotherms in figure 4(*a*) for t_w = 60 s and t_w = 7200 s), indicative, as before, of non-equilibrium processes. (By contrast, there is no evidence for such non-equilibrium behaviour within the ferromagnetic phase, 60 K $< T \leqslant$ 104 K, where a preliminary analysis [8] suggests a weak power-law decay with wait-time-independent parameters.) Furthermore, as the double-logarithmic plots in figure 4(*b*) reveal, this resemblance transcends mere qualitative comparisons based on superficial features, to include the actual functional representation of the remanent relaxation; thus, the superposition of a stretched exponential and a constant also provides an excellent description of all the re-entrant relaxation isotherms, and the values of the best-fit parameters listed in table 2 are essentially indistinguishable from their spin-glass counterparts in table 1.

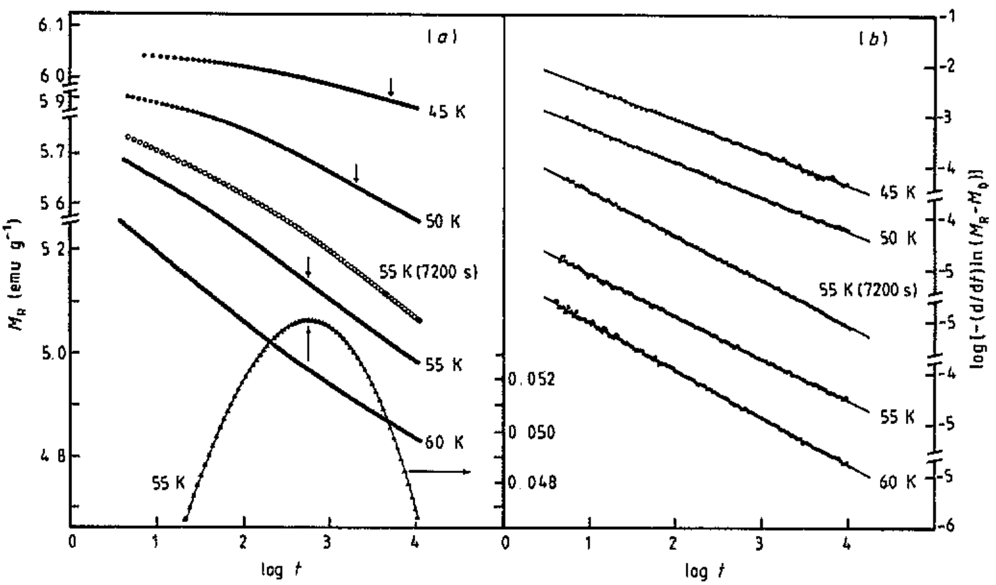

Figure 4. (*a*) A set of relaxation isotherms measured within the re-entrant phase of the Ni–23.5 at.% Mn ferromagnet. The full circles correspond to a wait time t_w = 60 s, and the open circles to t_w = 7200 s; the vertical arrows mark the inflection points. The full triangles show the relaxation rate $S = -\partial M_R/\partial \ln t$ for the T_m = 55 K isotherm for t_w = 60 s. (*b*) Double-logarithmic plots of $-(d/dt)\ln(M_R - M_0)$ as a function of t for the isotherms in (*a*).

The preceding comparison of thermoremanent relaxation dynamics offers novel and compelling evidence for the equivalence of the re-entrant and spin-glass phases; under comparable experimental conditions, the thermoremanent decay in both phases is dominated by non-equilibrium age-dependent processes and is describable by a common relaxation function which has been linked, theoretically, to a mesoscopic picture of growth and fragmentation of spin-glass domains [6]. Nevertheless, some specific features of the current study require further comment.

Table 2. Best-fit parameters for Ni–23.5 at.% Mn

T(K)	t_w(s)	M_0(emu g^{-1})	n	τ(s)
45	60	5.90 ± 0.01	0.65 ± 0.02	(5.0 ± 1.4) × 10^3
50	60	5.60 ± 0.01	0.67 ± 0.01	(1.9 ± 0.1) × 10^3
55	60	5.15 ± 0.01	0.82 ± 0.01	(6.0 ± 0.4) × 10^2
55	7200	4.64 ± 0.13	0.86 ± 0.01	(2.0 ± 0.7) × 10^5
60	60	4.60 ± 0.05	0.92 ± 0.02	0.5 ± 0.1

(i) While the location of the inflection point in the Ni–Mn relaxation isotherms in figures 2(a) and 4(a) is *approximately* coincident with the effective experimental age of the system, $t_{\rm eff} = t_w + t_c \simeq 10^3$ s, in accordance with theoretical expectations [6], it also clearly exhibits a systematic temperature-dependent shift towards progressively longer observation times as the measurement temperature T_m is decreased. (A similar trend is clearly apparent in some of the authoritative relaxation studies, by Lundgren and co-workers, on Cu–Mn, particularly in figure 4(a) of [5], where $t_{\rm infl}$ varies monotonically from $t_{\rm infl} < t_w$ at high temperatures to $t_{\rm infl} > t_w$ at low temperatures.) This ability to discriminate, experimentally, between the various isotherms on the basis of the age of the system ($t_{\rm infl}$), in spite of the constraints imposed on the current experimental procedure by the comparatively large cooling intervals $T_R - T_m \simeq 100$ K and correspondingly long cooling times $t_c \simeq 15$ min (far removed from the ideal 'temperature quench'), which effectively *suppress* the temperature dependence of t_c simply indicates that the system is essentially in equilibrium over a significant portion of the cooling interval, and that the aging process does not commence until the system has been cooled through some effective reference temperature T_R^* which lies well below T_R (for the spin glass, the onset of non-equilibrium behaviour occurs just below the principal maximum in $M(T)$ while, for the re-entrant ferromagnet, the absence of non-equilibrium effects throughout the ferromagnetic phase suggests that T_R^* is located close to the ferromagnetic–re-entrant phase boundary). Furthermore, from the tendency for $t_{\rm infl}$ to eventually *exceed all* the obvious experimental time constants, at the lower measurement temperatures, it is evident that slow cooling rates *accelerate* the aging process so that, while t_w remains an appropriate measure of the evolution of the system at constant temperature T_m (in fact, $t_w = 60$ s represents a lower bound on all the experimentally *observable* inflection points, as expected), the parameter t_c (or, more accurately, t'_c, if properly reckoned from the instant that the system passes through T_R^* until it reaches T_m) considerably *underestimates* the influence of the cooling process. (A comparable trend develops in some of the Cu–Mn relaxation data in [11] at the lowest measurement temperature $T_m = 21$ K, where longer cooling times $t_c \simeq 200$ s also yield $t_{\rm infl} > t_w + t_c$, even for comparatively long wait times $t_w \simeq 10t_c$).

(ii) A 'pure' stretched exponential is insufficient to provide a *complete* description of the Ni–Mn relaxation isotherms in either the re-entrant or the spin-glass phases but must be supplemented by a time-independent term which decreases with increasing temperature T_m. This constant component is also an ingredient of other empirical representations of remanent relaxation in systems such as Cu–Mn [11] and Au–Fe [12], where it exhibits similar temperature-dependent systematics [11]. While the presence of this component in the re-entrant Ni–Mn ferromagnet is certainly consistent with theoretical models [2] which predict that a longitudinal ferromagnetic spontaneous magnetization coexists with transverse spin-glass freezing within the re-entrant phase, its physical significance in the 'pure' spin-glass phase is less transparent. However, the sharp theoretical phase boundary which separates the ferromagnet regime ($m \neq 0, q \neq 0$) from the spin-glass regime ($m = 0, q \neq 0$) in *zero* field [2, 13], must *vanish* in a *finite* field [13], so that the distinction between the ferromagnetic and spin-glass phases is blurred, and the crossover becomes a more gradual continuous process; consequently, it is not surprising if finite-field measurements on a concentrated spin glass such as Ni–25 at.% Mn, with its extreme proximity to the multicritical point and with a substantial ferromagnetic bias to its bond distribution, exhibit some features in common with its ferromagnetic neighbours (even though its critical properties are distinctly *not* ferromagnetic).

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