Harmonic susceptibilities of an alloy of $Ni_{77}Mn_{23}$

1998 J. Phys.: Condens. Matter 10 191

(http://iopscience.iop.org/0953-8984/10/1/022)

J. Phys.: Condens. Matter **10** (1998) 191–203. Printed in the UK PII: S0953-8984(98)77530-7

Harmonic susceptibilities of an alloy of $Ni_{77}Mn_{23}$

A Gencer†, I Ercan‡ and B Özçelik†§

† Department of Physics, Faculty of Sciences, Ankara University, 06100-Tandoğan, Ankara, Turkey

‡ Ankara Nuclear Research and Training Centre, 06105, Saray, Ankara, Turkey

Received 2 September 1996, in final form 16 June 1997

Abstract. Low-field harmonic susceptibilities of a $Ni_{77}Mn_{23}$ alloy have been measured in the temperature range 15–325 K at three different frequencies. The measurements exhibit both frequency and amplitude dependencies. Observed dependencies are compared with the existing theories of linear and nonlinear susceptibilities with reference to short- and long-range interactions. Although the sample exhibits some features similar to the ordinary spin-glass systems at low temperatures, the Curie temperature T_C was found to be larger than expected. However, the sample exhibits some interesting properties in terms of spin frustration below T_f at some frequency and amplitude. The coexistence of spin-glass with ferromagnetic states at low temperatures signifies the nondivergent behaviour of the high-order harmonic susceptibilities in the vicinity of T_f. The critical exponents $\gamma = 3.2 \pm 0.2$ and $\beta = 1.68 \pm 0.4$ suggests that the third harmonic susceptibility is essentially different from the observed divergent behaviour in spin glasses.

1. Introduction

It is widely recognized that the $Ni_{77}Mn_{23}$ alloy is a re-entrant spin-glass (RSG) system. Re-entrant systems exhibit two successive phase transitions with respect to temperature [1–3]. Despite their dominant paramagnetic (PM) behaviour at temperatures above the Curie point, T_C, they enter into a new ferromagnetic (FM) phase below T_C (onset temperature of long-range order). However, they exhibit quite interesting physical phenomena upon further cooling by entering into a second phase at the so-called freezing temperature (T_f), where the system is believed to have some common properties with pure spin-glass (SG) systems [4]. The magnetic behaviour of such systems is of fundamental importance in terms of understanding the mixed interactions between the d-state electrons as a function of temperature [5].

Harmonic susceptibilities ($\chi_n = \chi_n' - i\chi_n''$, where $n = 1, 2, 3, \ldots$ etc) have generally been used to study the magnetic behaviour in such systems [6–8]. Intrinsic to the AC susceptibility, the in-phase component, χ', of the fundamental susceptibility $\chi_1 = \chi_1' - i\chi''$ is mostly associated with flux entry into the sample (proportional to the total magnetic moment at the peak value of the AC field) while the out-of-phase component χ'' is mostly proportional to the magnetic moment trapped within the sample at the zero-field excursion of the AC field. In metallic SGs, strong frequency dependence is observed in the fundamental susceptibility due to eddy currents [8–11]. The effects of eddy currents can be assumed to be negligible at sufficiently low frequencies provided that flux entry is not limited to

§ Present address: Department of Physics, Faculty of Arts–Sciences, Çukurova University, Adana, Turkey.

the skin depth of the sample. However, determination of the freezing temperature T_f at which the cusp in AC susceptibility occur reveals a value different from the results of DC experiments.

In addition to the linear susceptibility (fundamental susceptibility, $n = 1$) measurements, a large number of investigations have been performed by nonlinear susceptibility ($n = 3$, third-harmonic susceptibility) measurements to study their magnetic behaviour at temperatures close to T_f [12–15]. Such studies were also performed near T_C in random-spin systems [16]. The higher-harmonic susceptibilities give details of local magnetic behaviour.

It is generally accepted that the out-of-phase component, $\chi_3''(T)$ resembles that of $-\chi_1''(T)$ and that the magnitude of $\chi_3''(T)$ is proportional to the measuring-field amplitude [8]. It is also known that the in-phase component, $\chi_3'(T)$, is very small in magnitude compared with that of $\chi_3''(T)$. In a reversible FM system, the magnetization versus the field (M–H) would be linear. The linearity could be an indication of reversible domain-wall motion for sufficiently small fields. In such a state, odd harmonic (nonlinear) susceptibilities have zero values [17]. Nonzero values of $\chi_3'(T)$ could be attributed to the irreversible domain-wall motion. The re-entrant systems are in fact metastable systems, in which domain growth is prevented from taking on the magnetic behaviour of the pure SG systems [18]. In a previous communication [14], we focused on the relaxational behaviour of the re-entrant system of $Ni_{81}Mn_{19}$ around T_f.

It is well known that, owing to thermal fluctuations and the increasing short-range order around T_f, pronounced nonlinearities in the magnetic response appear in the magnetization of a SG. In terms of the measuring field H_{AC}, the magnetization can be expressed as an addition of odd harmonic susceptibilities as [12],

$$M(h, T) = \chi_1 H_{AC} + \chi_3 H_{AC}^3 + \chi_5 H_{AC}^5 + \cdots \tag{1}$$

where even powers of h are neglected because of the symmetry of M. In compact form, M can be given by taking a sinusoidal field, $H_{AC} = h_0 \sin(\omega t)$ as

$$M(h, T) = \sum_{n=0}^{\infty} \tilde{\chi}_{2n+1} h_0^{2n+1} \sin[(2n+1)\omega t] \tag{2}$$

where,

$$\tilde{\chi}_1 = \chi_1 + \tfrac{3}{4}\chi_3 h_0^2 + \tfrac{5}{8}\chi_5 h_0^4 + \cdots \tag{3a}$$

$$-\tilde{\chi}_3 = \tfrac{1}{4}\chi_3 + \tfrac{5}{16}\chi_5 h_0^2 + \cdots \tag{3b}$$

$$\tilde{\chi}_5 = \tfrac{1}{16}\chi_5 + \cdots \text{ etc.} \tag{3c}$$

The nonlinear susceptibilities diverge according to $\varepsilon^{-n(\gamma+\beta)+\beta}$ for $n \geqslant 1$ at temperatures in the vicinity of T_f, where ε is the reduced temperature which equals to $(T - T_f)/T_f$, γ and β are the critical exponents [19], this divergent behaviour occurs in the vicinity of T_f to indicate a phase transition from FM to SG. In other Mn-containing SGs, the critical exponents were found to be $\gamma = 3.4 \pm 0.2$ for fluorophosphate of Mn by Prejean *et al* [20], $\gamma = 3.3 \pm 0.3$ for $Cd_{1-x}Mn_xTe$ by Mauger *et al* [21], $\gamma = 2.3 \pm 0.2$ for AgMn by Levy [22]. Most other results are in the region of $3 \leqslant \gamma \leqslant 4$ [23–25].

In this work, to test whether the sample undergoes a true phase transition from FM to RSG, we have first concentrated on measurements of the harmonic susceptibilities, in the vicinity of T_f. Measurements, near the Curie temperature in the same $Ni_{77}Mn_{23}$ alloy are also important, to study the phase transition from PM to FM. Here, we present a more detailed account of different magnetic behaviours in this alloy by testing the observed temperature and amplitude dependencies in terms of various theoretical predictions.

2. Experimental

The sample investigated in this work has been prepared by using the appropriate amount of 99.999% pure Ni wire and 99.999% pure Mn flakes. They were first sealed into a quartz tube under a helium atmosphere of about $\frac{1}{3}$ atm and then melted by an rf induction furnace several times in order to obtain a homogenized alloy of $Ni_{77}Mn_{23}$ and then quenched. The quench rate is important for the elimination of the Ni_3Mn phase, which contributes to long-range interactions. It was found necessary to add some excess Mn, to minimize weight losses during the sample preparation. In addition, great care was needed to prevent reaction between the walls of quartz tube and the composition. The x-ray fluorescence (XRF) studies showed that the peak ratio between Ni and Mn equals 3.05:1. Next, the sample was shaped into a sphere of 3 mm diameter by a spark-cutting technique [26]. Finally, we applied a second heat treatment of 24 h at 900 °C before furnace cooling.

A commercial Lake Shore AC susceptometer 7130 model with a closed-cycle refrigerator for the AC-susceptibility measurements was used. The sample was kept in a helium-exchange gas for thermal equilibrium with a controllable temperature resolution better than 10 mK. The sample was moved between the centres of secondary coils in order to maximize the signal coming from the sample itself. To find the fundamental component of AC susceptibility, the lock-in amplifier (LIA) was operated incorporating an input low-pass filter to accept the fundamental harmonic of the $\mathrm{d}M/\mathrm{d}t$ signal induced by the sample and a band-pass filter for the high harmonics $(2n-1)$ to obtain magnetic signals at $(2n-1)f_0$ to filter out the signals beyond the band-pass range, while no filtering (wide-band, or flat mode) was used for AC magnetization hysteresis loops. The principle of measuring AC magnetization is based on Campbell's method [27] involving integration of the input signal $(\mathrm{d}M/\mathrm{d}t)$ coming from the sample itself, by use of a sensitive LIA.

To obtain for harmonic susceptibility measurements, automatic data acquisition was selected. The phase angle was chosen such that $\Delta M = 0$ at zero AC field excursion and χ_1'' was also zero at low temperatures for a calibration sample. All the measurements reported in this paper were taken during warming to make quantitative comparisons.

3. Results and discussion

In this section, we first present our results and then make a comparison with other work. For a uniformly magnetized sphere, the demagnetization factor (D) would be $\frac{1}{3}$ (SI). The measured AC susceptibility values were less than D^{-1} imposed by the geometry of the samples. Although demagnetizing corrections would lead to a suitable reduction in values of AC susceptibility, it does not make any significant correction of the results.

In figure 1, we have plotted the fundamental and absolute harmonic susceptibility measurements versus temperature. The transition from PM to FM followed by FM to RSG are not sharp but rather take place gradually when the temperature is lowered from above T_C and T_f respectively. In the FM state, a large number of FM domains are formed and aligned along the AC field. Magnetization is then simply the process of rearranging these domains so that their magnetic moments align parallel with the field. Whereas, in the PM state (above T_C), there also exists atomic magnetic moments which exhibit no long-range order. On the other hand, the FM state responds to AC fields by Bloch wall motion. Below 100 K or so, χ_1' starts to drop due to the spin frustration influencing the domain-wall motion [14]. In this state, antiferromagnetic (AFM) clusters are formed and the spins become viscously frozen due to the strong interaction between the FM and AFM clusters and the domain-wall motion is hindered. Therefore, one observes the low-temperature sharp decrease in $\chi_1'(T)$

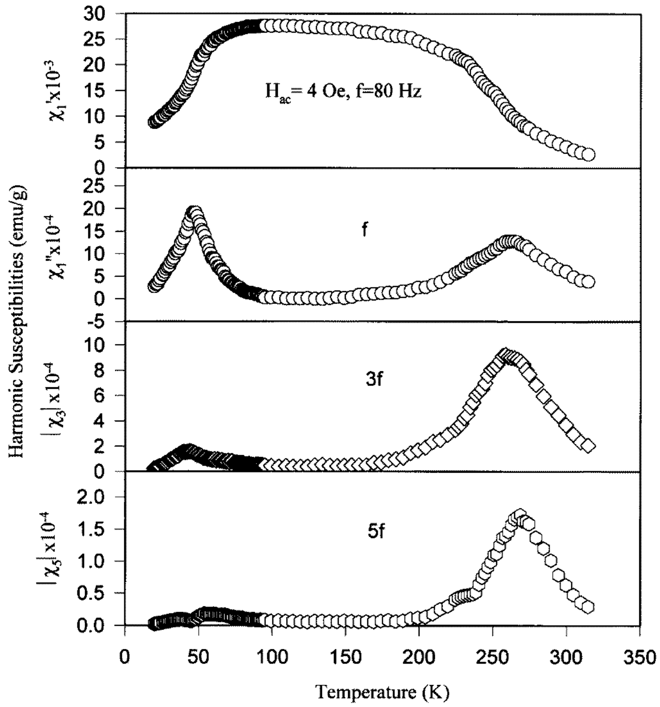

Figure 1. Temperature-dependent variation of the fundamental, absolute third and fifth harmonic susceptibilities, exhibiting two successive transitions with respect to T.

and an accompanying peak in the $\chi_1''(T)$. The out-of-phase component, $\chi_1''(T)$ also exhibits a high-temperature peak to mark the relevant transition from PM to FM. Note that the high-temperature peak is smaller in size than the low-temperature peak. The peak widths are about 25–30 K at the FM–RSG and 50–60 K at the PM–FM transitions, respectively. Similar wide peaks were also observed in the superparamagnetic alloy $Cu_{97}Co_3$ by Bitoh *et al* [6], while a 1–2 K wide peak was observed in $Au_{96}Fe_4$ at the PM–SG transition and at the PM–FM transition of $Au_{82}Fe_{18}$ alloy by Bitoh *et al* [7] and Ni by Shirane *et al* [8]. The sample behaves spuriously as though it is a superparamagnetic alloy near T_C.

The absolute third-harmonic susceptibility $|\chi_3|$ mimics the qualitative shape of $\chi_1''(T)$ as shown in figure 1. Nonlinear odd-harmonic susceptibilities give qualitative features of the interactions between the spins. The upper peaks are larger in magnitude than the lower ones both in the third and fifth harmonics. At high temperatures mostly the short-range interactions are present, while at low temperatures the interactions are of long-range order in spite of some spin frustration. Therefore, the system is not in a pure SG state, but coexists with a less dominant FM character below a less well-defined temperature T_f. In the thermodynamic sense, it is not clear whether the transition is a true phase transition. If there exists a peak in the high-order susceptibilities, this should be associated with the change of interactions.

Figure 2 shows the temperature dependencies of the fundamental susceptibility at the frequency 80 Hz with the AC field amplitudes $h_0 = 1$, 2 and 4 Oe. Note that there exists a small shift in $\chi_1'(T)$ to lower temperatures with increasing field amplitude. As the field amplitude increases, the spin frustration occurs at slightly lower temperatures. This implies that the field energy causing spins to be aligned in the field direction overcomes the

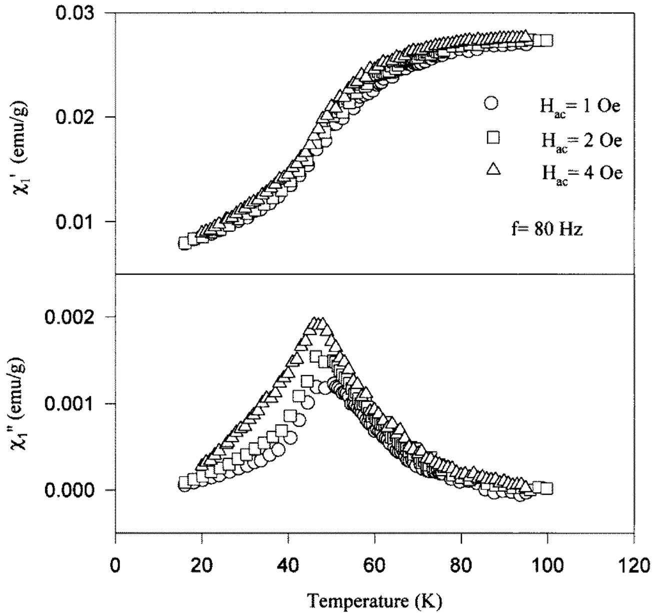

Figure 2. AC field dependence of the fundamental susceptibility, both in-phase and out-of-phase components. As the field amplitude increases, the peak in $\chi_1''(T)$ becomes broadened and shifted to to lower temperatures, where irreversible domain-wall motion is hindered.

Dzyaloshinsky–Moria (DM) anisotropy type of interaction which is a function of temperature but is independent of the field amplitude.

In figures 3 and 4, we have plotted the absolute third and fifth harmonics at 80 Hz with $h_0 = 1$, 2 and 4 Oe. Note that the familiar peak shape is present for all small fields. For 1 Oe, there exists a dominant peak with diminishing tail at lower temperatures (figure 3). One can immediately see that the magnitude of the peaks exhibits amplitude dependencies. This is understandable as the $|\chi_3|$ is nearly proportional to h_0^2. In addition, as the field amplitude is increased, the small tail becomes dominant to become a single peak in $|\chi_3|$. The small tail is more apparent in $|\chi_5|$ (figure 4). This might suggest that $|\chi_5|$ includes the local interactions more effectively in comparison with $|\chi_3|$. Experimentally, $|\chi_5|$ is measured at 5f and the fundamental response is easily filtered out. Therefore, the observed signal can then be related mostly to the short-range interactions. For long-range interactions which mostly occur in the FM state above 100 K or so, the high-order harmonic susceptibilities yield zero until the temperature T_C.

Now we start to test whether the transition in the low temperature region obeys the existing theories, related to the nonlinear harmonic susceptibilities with $n = 3$ and 5. In figure 5(a), we have plotted the absolute third-harmonic values versus the reduced temperature ($\varepsilon = (T - T_f)/T_f$) on a log–log scale for the frequency of 125 Hz, AC field amplitude of 10 Oe and $T > T_f \cong 60$ K. As can be seen, the curve bends away from the linear line and becomes rounded. The rounding of the curves on approaching T_f is due to inhomogeneities in the sample and most importantly is indicative of the dynamic effects which cause the spin system to lose its equilibrium. The value of the critical exponent γ, which is characteristic of static phase transition to a SG state, is estimated as 3.2 ± 0.2 from the initial slope of the given line shown in the figure 5(a). This asymptotic value is in agreement with the results of the static measurements for Cu–Mn [28, 29] and AC

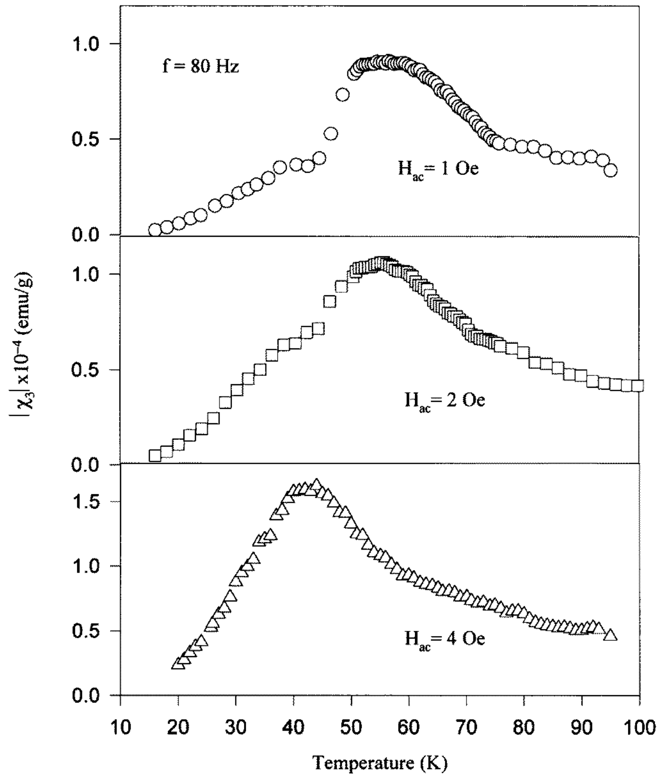

Figure 3. AC field dependence of the absolute third harmonic susceptibilities. As the field increases spin frustration starts at lower temperatures. The tail also diminishes for $H_{AC} = 4$ Oe.

measurements for Pd–Mn [12, 30], for fluorophosphate of Mn [20] and for $Cd_{1-x}Mn_xTe$ [21]. Other estimates range as $3 \leqslant \gamma \leqslant 4$ [23–25]. However, our result is larger than the values reported for other Mn containing SGs [12, 22, 31]. The lower value in [31] is presumably due to the improper determination in a large magnetic field exceeding the crossover magnetic field.

In order to deduce the value of the other critical exponent β related to the order parameter a plot of χ_5/χ_3 versus χ_3 is made on a double logarithmic scale. This should give a straight line when approaching T_f from above as first suggested by Suzuki [19]. The slope of the line is $1 + \beta/\gamma$. Figure 5(*b*) shows that such a plot yields very distributed points. Since it is difficult to draw a line using such scattered data, we were not be able to extract a sensible β value. By plotting χ_5/χ_3 against χ_3 on a log–log scale, we have tried only to estimate β. The slope of this plot gives a value of 1.68 ± 0.4. This is rather large for systems exhibiting a real SG behaviour, where one would expect β between 0.5 and 1 [22, 28, 31, 32]. We believe that the reason we obtain such a high value of β, with poor accuracy, may be attributed to several different mechanisms. Among these are the dynamic effects of intrinsic behaviour due to metallic losses which give rise to eddy-current losses to yield strongly frequency-dependent behaviour [8, 9, 33]. This contributes a nondivergent behaviour of the odd-harmonic susceptibilities. It is clearly advantageous to measure the magnetic response of the sample at sufficiently low frequencies to eliminate the dynamic effects, which are intrinsic to metallic SGs.

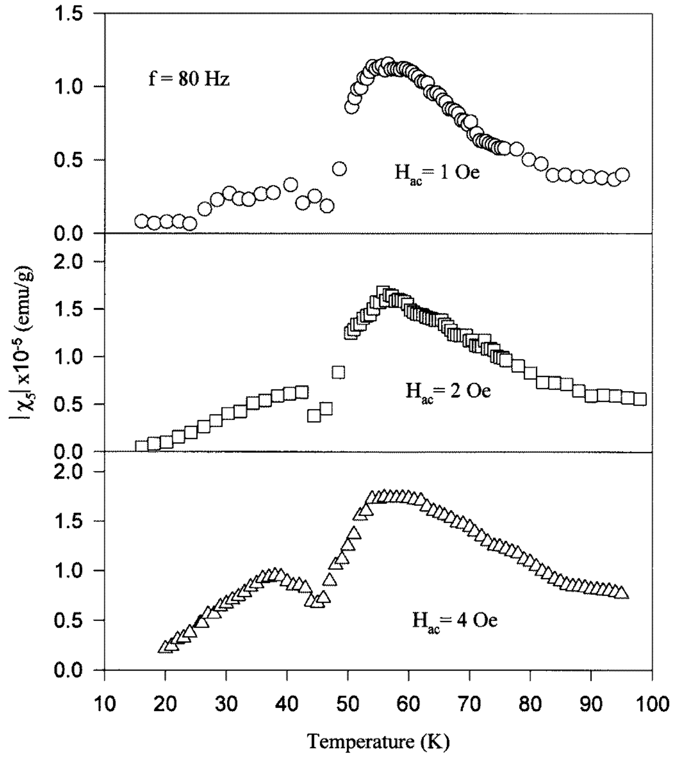

Figure 4. AC field dependence of the absolute fifth harmonic susceptibilities. Note that there exists a double-peak feature, not present in the third harmonic susceptibility.

In figure 6, we have plotted the frequency dependent behaviour of the third-harmonic susceptibility for 5, 80 and 125 Hz at 1 Oe (rms) in the temperature range of 20–60 K. Note that there is a significant frequency dependence associated with flux entry involving relaxation of the spins during one AC period of the field. The frustrated spins are being viscously frozen and hence making a contribution to the frequency dependence. However, there are still some free spins that contribute to the magnetic response, which follow up the AC field modulation almost instantly. However, the hindrance of domain-wall motion is dominant at low temperatures. Therefore we can conclude that even below a less well-defined temperature of T_f, there is still some long-range order between the d-state electrons of Ni and Ni–Mn atoms. We earlier pointed out [14] that the additional DC bias field would weaken the long-range order even at temperatures below T_f. Therefore, we have applied an additional DC field of 19.1 Oe (maximum available field in the magnetometer) to check whether the remaining interactions in the presence of an additional DC field are present, the dramatic effect of the DC field together with higher-harmonic susceptibility results are given in figure 7. Note that the additional DC field has no significant effect at high temperatures around and above 100 K. Below the peak temperature which loosely corresponds to the regime of the freezing temperature T_f, there exists a deflection and $\chi_1'(T)$ drops to slightly lower values. The same effect is also seen in $\chi_1''(T)$ and in the absolute higher harmonic susceptibilities (third and fifth). The field affects the formation of the peak. However, the spin dynamics are effective below the peak temperature and must be eliminated to test the observed dependencies with the theoretical predictions. These

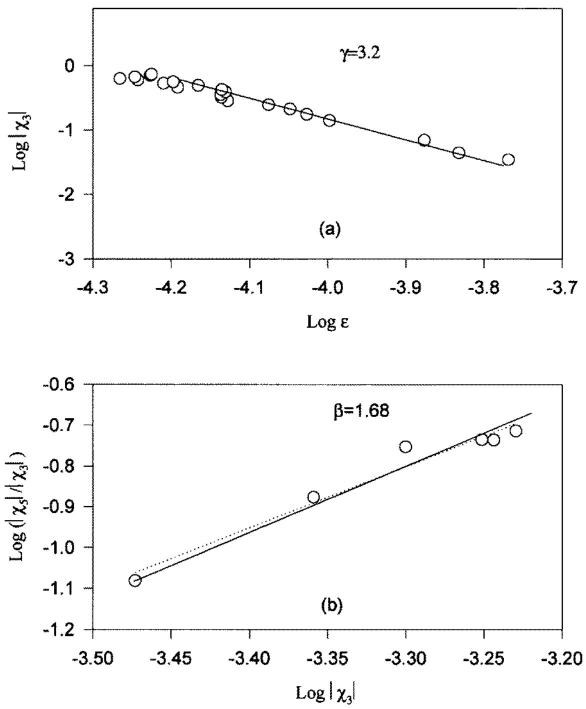

Figure 5. (a) Extraction of the critical exponent γ from the log–log plot of third harmonic versus the reduced temperature ε (b) and of the parameter β from the plot of $\log(\chi_5/\chi_3)$ versus $\log(\chi_3)$.

predictions are only valid around and above T_f if there exists a distinct divergent peak in the nonlinear higher-harmonic susceptibilities. Thus, the measurements of higher-harmonic susceptibilities reveal the coexistence of the SG character with the less dominant FM state, both manifest mixed interactions.

We next focus our attention on the high-temperature regime of the susceptibility. This regime appears to be more difficult to characterize. Normally the Curie temperature (T_C) can be found by extrapolation of the χ'^{-1} versus T plot. However, the gradual change forces determination by a more sophisticated plot of Kouvel–Fisher [34] as shown in figure 8(*a*). Then T_C can be determined from a function $F(T) = \chi_1'^{-1}(\mathrm{d}\chi_1'^{-1}/\mathrm{d}T)^{-1}$ versus T plot. If the temperature dependence of χ_1' above T_C is dominated by a power-law scaling as $\chi_1'^{-1}\alpha(T - T_C)^{\eta}$, where η is an exponent determining the sharpness of transition, the function will become $F(T) = (T - T_C)/\eta$ at temperatures slightly above T_C. A plot of $F(T)$ against T in the close vicinity of T_C would give a straight line if the change in $\chi_1'(T)$ is sufficiently sharp. Note that at temperatures above and around 210 K, it does not appear to be a single straight line whose slope lies around 1. The $F(T)$ only becomes zero below 180 K. Between 210 and 180 K, the FM long-range order dominantly prevails in the sample with less effective short-range interactions. That is, there is no genuine long-range order presumably due to the presence Ni_3Mn phase as suggested by Kunkel *et al* [35]. The long-range order appears to be complete below 180 K until very low temperatures of around 100 K. In addition, the absolute harmonic susceptibilities were given in figures 8(*b*) and (*c*) in the presence of an additional DC bias field. A cusp-like dependence is expected in the odd higher-harmonic susceptibilities of a magnetic system at the vicinity of phase transition

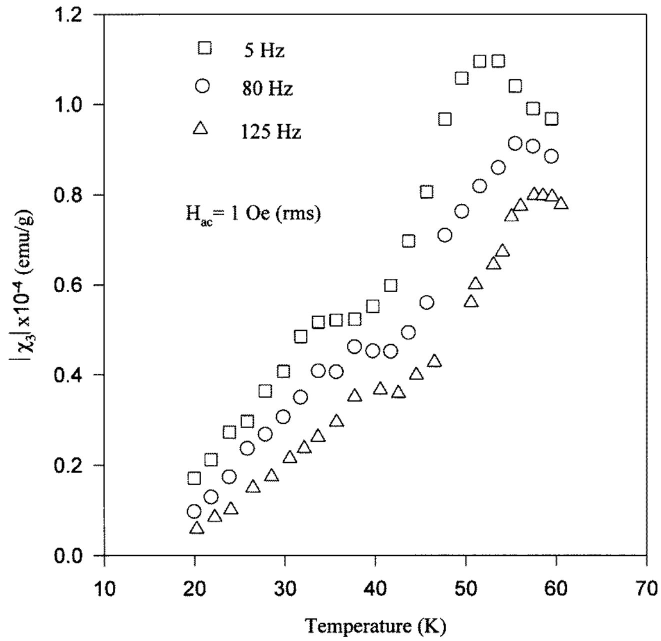

Figure 6. Frequency dependence of the absolute third-harmonic susceptibility at lower temperatures. As the frequency increases the peak shifts to higher temperatures.

or change of interactions. As seen in the figure, the odd high harmonics (third and fifth) give a cusp-like dependence with temperature. The long-range order appears to be complete at low temperatures below 180 K. This temperature is not in agreement with the work of Kouvel [36, 37]. There exists a shift of almost 20 K towards high temperatures. This may be associated with the fact that the sample exhibits a superparamagnetic transition, although $\chi_1'(T)$ implies that spin clusters appear to resemble FM percolation on a large scale. Above 180 K, there are some mixed interactions as evident from the nonzero value of $|\chi_3|$. The linear behaviour in M would loosely correspond to the long-range order which only exists in the FM state (around 100 K and up to 180 K). On the other hand, the even high-harmonic susceptibilities are always zero when there is symmetry in the M–H curve. Note that the M–H curves of the sample are always symmetric as shown in figure 9, which will be dealt with later. However, the symmetry may break down at the vicinity of a phase transition and the even harmonics can also be used to determine the characteristics of the transition. On the other hand, the signal to be measured for the even harmonics at temperatures beyond the critical behaviour would be weak due to the symmetry in M.

The AC magnetization measurements were first carried out as a function of temperature at an AC field of 10.0 Oe (rms). In figure 9, AC magnetization versus applied field is plotted at different temperatures of around 20, 25, 48, 60, 140 and 316 K. At 316 K, the hysteresis loop is S shaped and is consistent with the results of figure 1. The magnetic response is weak and appears to be dominated by short-range interactions with direct exchange. The magnetization becomes saturated at about 12 Oe before the AC field reaches its peak value H_m. This suggests that the magnetization during one complete cycle is nonsinusoidal and therefore nonlinear. As the temperature is lowered to around 140 K (below the Curie point), the magnetization becomes larger and AC loss per cycle decreases, because the AC field amplitude becomes very much less than the field required for saturation. In this case, long-

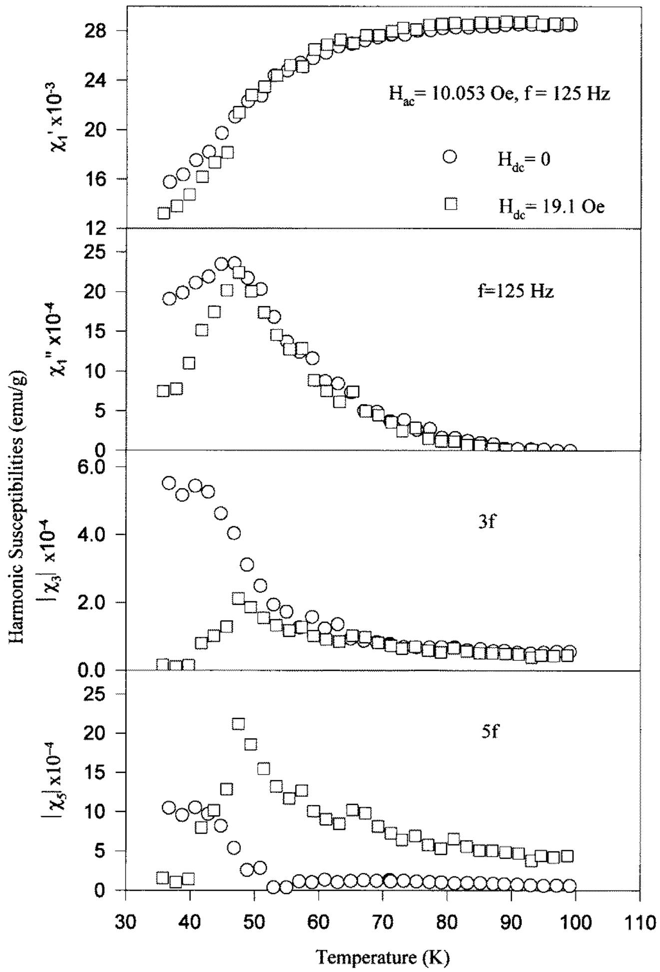

Figure 7. Effect of additional DC field on the fundamental, third and fifth harmonic susceptibilities. (H_{AC} = 10.05 Oe, H_{DC} = 19.1 Oe, f = 125 Hz). The DC field has a dramatic effect on the nonlinear susceptibilities. The peak formation has been observed with the application of DC field.

range order between spins begins to constitute domains growing in size to respond as a whole to the external field, which leads to a rise in magnetization. There is then a dominant long-range order for interactions. The magnetization at the peak value of the AC field then remains unchanged with decreasing temperature down to around T_f. The magnetization at temperatures below T_f are also given in figure 9, plotted for 20, 25, 48 and 60 K. During further cooling below T_f, some AFM regions occur. Since there exists strong interactions at the interface between AFM and FM clusters, the FM clusters are blocked and they begin to become viscously frozen. The blockage of clusters with lowering temperature results in an irreversible behaviour in magnetization giving rise to a hysteric behaviour for M. These smaller AFM regions also give rise to AC losses and the hysteresis area increases (compare the difference in loop areas of the measurements performed at 20 and 60 K).

In summary, our sample $Ni_{77}Mn_{23}$ exhibits two successive transitions with respect to temperature. However, this study indicates that the observation of a plateau-like dependence

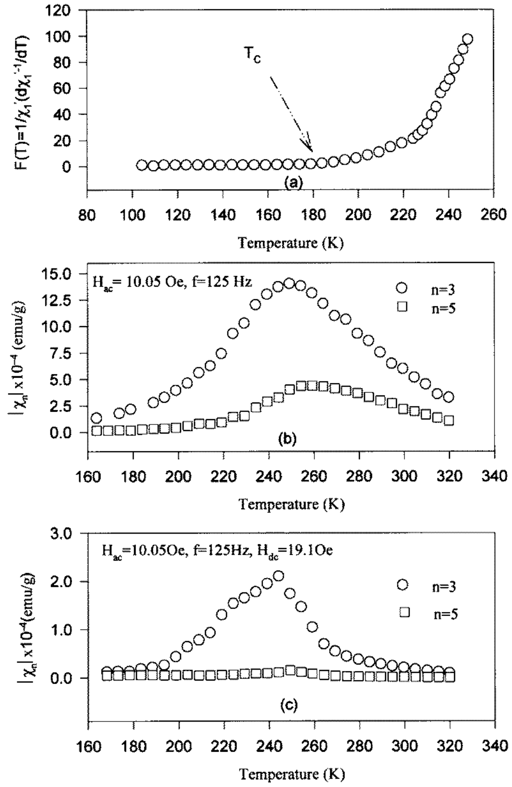

Figure 8. (a) Kouvel–Fisher plot of the fundamental susceptibility around T_C (b), the third and fifth harmonic susceptibilities in the absence and (c) in the presence of an additional DC field.

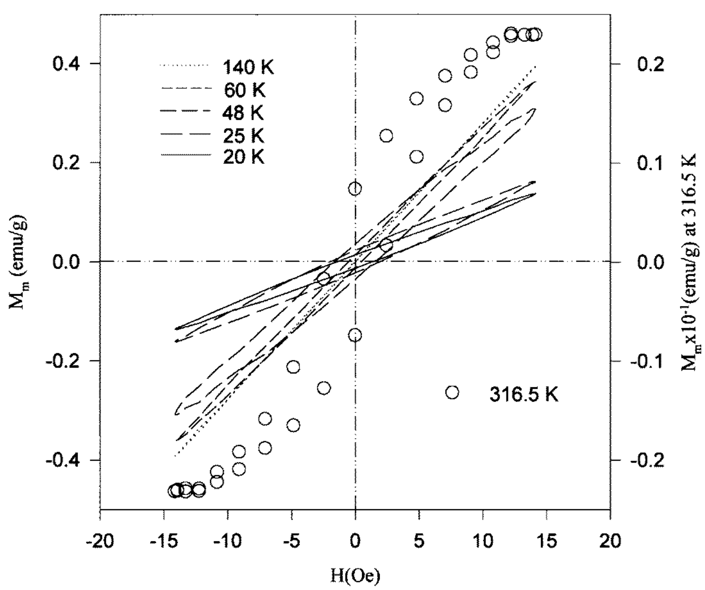

Figure 9. AC magnetization versus magnetic field at various temperatures. At high temperatures, the magnetization is S shaped. There exists an hysteresis and its magnitude is small (see the right vertical axis). As the temperature is lowered, magnetization becomes larger (see the left vertical axis), though hysteresis area becomes smaller. In the FM state, the magnetization is linear in H and complete reversible behaviour can be observable. At very low temperatures, there is again irreversible magnetization but resulting from a hindrance of domain-wall motion.

in the in-phase component and peaks in the out-of-phase component susceptibilities are insufficient to characterize these samples as re-entrant systems, even if critical peaks are observed for the criterion of FM upper transition. Higher-harmonic susceptibility measurements indicate that the gradual transition to the re-entrant SG phase is clouded by the existence of long-range interactions. Third-harmonic susceptibility which diverges theoretically as $\varepsilon^{-\gamma}$, the critical exponent, γ has been estimated to be 3.2 ± 0.2 from the data just above T_f. However, the relaxation or dynamic effects seem to cause the spin system to lose its equilibrium at temperatures approaching T_f. Hence, the theoretically expected phase transition becomes inaccessible to the experiment and the critical behaviour is only valid in a limited reduced temperature range in the close vicinity of T_f. In addition, the precise extraction of β has proved to be difficult presumably due to the nondivergent behaviour of the odd high-harmonic susceptibilities near T_f. In addition, experimental results were extremely influenced by both AC-field amplitude and measuring frequency. Here we suggest that the AC-susceptibility measurement should be performed at very low frequencies on such samples exhibiting mixed interactions. In addition, higher harmonic susceptibilities can used to determine the characteristics of the transitions more effectively.

Acknowledgments

IE is grateful to Ankara University for use of experimental facilities and to Mr E Aksu for technical expertise. We acknowledge the useful comments and discussions with Professor Arsin Aydinuraz. This work is supported by Turkish Scientific and Research Council, TÜBITAK, in part under a grant no TBAG-1343 and in part under a grant no DPT-TBAG 19.

References

[1] Hurdequint H and Kouvel J S 1986 *J. Magn. Magn. Mater.* **167** 54–7
[2] Abdul-Razzaq W and Kouvel J S 1984 *J. Appl. Phys.* **55** 1623
[3] Abdul-Razzaq W and Kouvel J S 1985 *J. Appl. Phys.* **37** 3467
[4] Huang C Y 1985 *J. Magn. Magn. Mater.* **51** 1–74
[5] Hesse J 1989 *Hyperfine Interact.* **47** 357–78
[6] Bitoh T, Ohba K, Takamatsu M, Shirane T and Chikazawa S 1993 *J. Phys. Soc. Japan* **62** 2583
[7] Bitoh T, Shirane T and Chikazawa S 1993 *J. Phys. Soc. Japan* **62** 2837
[8] Shirane T, Moriya T, Bitoh T, Sawada A, Aida H and Chikazawa S 1995 *J. Phys. Soc. Japan* **64** 951
[9] Nogués J and Rao K V 1993 *IEEE Trans. Mag.* **29** 3246
[10] Eckelt T, Böttger Ch and Hesse J 1992 *J. Magn. Magn. Mater.* **104–107** 1665
[11] Tholence J L 1993 *Solid State Commun.* **88** 917
[12] Özçelik B, Kiymaç K, Verstelle J C, van Duyneveldt A J and Mydosh J A 1992 *J. Phys.: Condens. Matter* **4** 5801–10
[13] Özçelik B, Kiymaç K, Verstelle J C, van Duyneveldt A J and Mydosh J A 1994 *J. Phys.: Condens. Matter* **6** 8309–21
[14] Gencer A and Ercan I 1997 *Int. J. Mod. Phys.* B at press
[15] Mukherjee S, Ranganathan R and Ray S B 1994 *Phys. Rev.* B **50** 1084–9
[16] Kunkel H P and Williams G 1988 *J. Magn. Magn. Mater.* **75** 98
[17] Shaulov A and Bhargava R 1991 *J. Appl. Phys.* **69** 6738
[18] Mandal K and Ghatak J K 1995 *Int. J. Mod. Phys.* B **9** 145–60
[19] Suzuki M 1977 *Prog. Theor. Phys.* **58** 1151–65
[20] Prejean J J, Carré E, Beauvillain P and Renard J P 1988 *J. Physique* C **8** 995
[21] Mauger A, Ferré J and Beauvillain P 1989 *Phys. Rev.* B **40** 862
[22] Levy L P 1988 *Phys. Rev.* B **38** 4963–73
[23] Barbara B, Malozemoff A P and Imry Y 1981 *Phys. Rev. Lett.* **47** 1852
[24] Barbara B, Malozemoff A P and Imry Y 1982 *J. Appl. Phys.* **53** 7672

[25] Berton A, Chaussy J, Odyn J, Rammal R and Tournier R 1982 *J. Physique Lett.* **40** L931
[26] Gray W J and Spedding F H 1969 *Rev. Sci. Instrum.* **40** 1427
[27] Campbell A M and Evetts J 1972 *Adv. Phys.* **21** 199
[28] Omari R, Prejean J J and Souletre J 1983 *J. Physique* **44** 1069–83
[29] Chikazawa S, Matsuyama H, Sandberg C J and Miyako Y 1982 *J. Phys. Soc. Japan* **51** 1037
Chikazawa S, Matsuyama H, Sandberg C J and Miyako Y 1981 *J. Phys. Soc. Japan* **50** 2884
[30] Williams G 1989 *J. Magn. Magn. Mater.* **81** 239–41
[31] Bouchiat H 1986 *J. Physique* **47** 71–88
[32] Yeung I, Roshka R M and Wiliams G 1987 *J. Magn. Magn. Mater.* **68** 39
[33] Berkowitz A E and Kneller E 1969 *Magnetism and Metallurgy* vol 1 (New York: Academic) p 238
[34] Kouvel J S and Fisher M E 1964 *Phys. Rev.* **136** A1626
[35] Kunkel H, Roshko R M, Ruan W and Williams G 1991 *Phil. Mag.* B **63** 1213
[36] Kouvel J S, Abdul-Razaq W and Zyq K h 1987 *Phys. Rev.* B **35** 1768–75
[37] Mirebeau I, Itoh S, Mitsuda S, Watanabe T, Endoh Y, Hennion M and Calmettes P 1991 *Phys. Rev.* B **44** 5120