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Non-linear AC susceptibility of a spin glass Pd–5.5 at.% Mn

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Abstract. We have investigated the linear and the non-linear AC susceptibilities of a Pd–Mn spin glass with 5.5 at.% Mn around its transition temperature T_f, as a function of temperature, frequency and amplitude of the AC driving field. Through linear-susceptibility measurements the transition temperature to a spin-glass phase is found to be $T_f = 3.40 \pm 0.02$ K. The third harmonic $\bar{\chi}_3$ is fitted in a limited region above T_f to a power law of the form $[(T - T_f)/T_f]^{-\gamma}$ and from this fitting the value of the critical exponent γ is found to be 2.2 ± 0.2. Since the demagnetization correction is quite small for this sample, we have also measured the fifth and seventh harmonics for $f = 234$ Hz. Since $\overline{\chi}_5/\overline{\chi}_3$ scales as $(\overline{\chi}_3)^{1+\beta/\gamma}$, by plotting $\overline{\chi}_5/\overline{\chi}_3$ versus $\overline{\chi}_3$ on a log–log scale the value of the critical exponent β is estimated and turned out to be rather small (about 0.2). We believe that this small value of β is influenced by dynamical effects and the large amplitude and frequency of the AC driving field that was necessary to obtain noise-free results.

1. Introduction

Since 1967 a great deal of experimental work has been done on Pd–Mn alloys. A theoretical description of the interactions between nearby Mn atoms in palladium was first given by Moriya [1]. Later, Rault and Burger [2] determined the Curie–Weiss temperature Θ, the Néel temperature T_N and the magnetization at 4.2 K in a magnetic field of 6 kOe for Mn concentrations up to 25 at.%. These workers argued that Pd–Mn alloys show ferromagnetism below 8 at.% Mn, and antiferromagnetism above 10 at.% Mn and that the transition from ferromagnetism to antiferromagnetism is continuous. Furthermore, it has been shown that the ferromagnetic ordering disappears when the sample absorbs hydrogen [3], and ordering changes its character. 'Giant-moment' values ($\mu_{Mn} \simeq 7.5\mu_B$) have been found by measuring the magnetization of a series of Pd–Mn alloys for Mn concentrations up to 2.45 at.% [4, 5]. From the resistivity measurements for Mn concentrations of 1.05, 2.4 and 2.91 at.%, Williams and Loram [6] and Williams *et al* [7] have suggested the existence of both ferromagnetic and antiferromagnetic coupling between Mn impurities in Pd–Mn. Zweers *et al* [8] established a transition with the characteristics of a spin glass from the temperature dependence of the linear AC susceptibility for Mn concentrations of 4, 6 and 8 at.%. At present it is well known that Pd–Mn alloys with Mn concentrations below 4 at.% are ferromagnetic, and between 4 and 10 at.% are spin glasses [8, 9].

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In addition to the linear-susceptibility measurements, a large number of investigations have recently focused upon the non-linear susceptibility of spin glasses in order to probe deeper into the nature of the spin-glass transition around the so-called freezing temperature T_f [10–14]. On the basis of the model of Edwards and Anderson [15], Suzuki [16] has proposed a phenomenological theory for spin glasses where he showed that the non-linear susceptibilities χ_{2n+1} diverge according to $\epsilon^{-n(\gamma+\beta)+\beta}$, for $n \geqslant 1$, where ϵ is the reduced temperature $(T-T_f)/T_f$. Hence, the third derivative of the magnetization, with respect to the magnetic field, diverges according to the power law $\epsilon^{-\gamma}$ at T_f and its sign is negative. In the light of these theoretical expectations we have investigated the non-linear AC susceptibilities of various kinds of samples having spin-glass, ferromagnetic and antiferromagnetic properties. The results of ferromagnetic and antiferromagnetic samples will be given elsewhere.

The present paper is concerned with detailed AC susceptibility measurements of a spin-glass Pd–Mn alloy with a Mn concentration of 5.5 at.%. By using a conventional mutual-inductance technique [17, 18], we have systematically determined the temperature, frequency and AC field amplitude dependences of the linear and non-linear susceptibilities of this system.

2. Experimental methods

Generally the total non-linear susceptibility χ_n is written as

$$\chi_n(\omega) = [M(H,T) - \chi_1 H]/H \tag{1}$$

where χ_l is the linear susceptibility. When AC methods are used to determine $\chi_n(\omega)$, it is more convenient to use the series expansion

$$M(H,T) = \chi_1 H + \chi_3 H^3 + \chi_5 H^5 + \dots . \tag{2}$$

The coefficients χ_1, χ_3, χ_5, ... can be identified with the Taylor series expansion

$$M(H,T) = (1/1!)(\mathrm{d}M/\mathrm{d}H)_{H=0}H + (1/3!)(\mathrm{d}^3M/\mathrm{d}H^3)_{H=0}H^3 + \dots . \tag{3}$$

Taking $H = h_0 \sin(\omega t)$, one can write (2) in the form

$$M(h_0,T,t) = \sum_{n=0}^{\infty} \tilde{\chi}_{2n+1} h_0^{2n+1} \sin[(2n+1)\omega t] \tag{4}$$

in which

$$\begin{aligned} \tilde{\chi}_1 &= \chi_1 + \tfrac{3}{4}\chi_3 h_0^2 + \tfrac{5}{8}\chi_5 h_0^4 + \dots \\ -\tilde{\chi}_3 &= \tfrac{1}{4}\chi_3 + \tfrac{5}{16}\chi_5 h_0^2 + \dots \\ \tilde{\chi}_5 &= \tfrac{1}{16}\chi_5 + \dots \\ &\vdots \end{aligned} \tag{5}$$

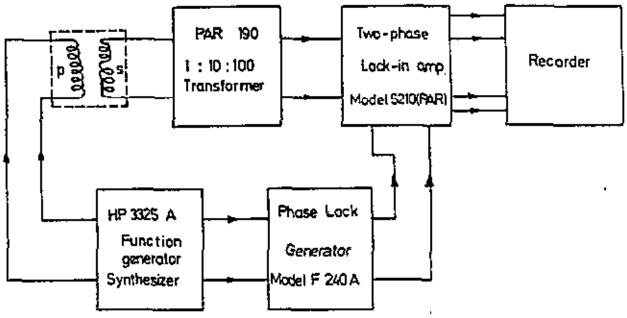

Figure 1. Experimental set-up for measurements of the linear and non-linear AC susceptibilities.

By detecting the response $M(t)$ to the AC magnetic field at the frequencies ω, 3ω, $5\omega, \ldots$, one can determine $\tilde{\chi}_1$, $\tilde{\chi}_3$, $\tilde{\chi}_5$, ..., respectively.

In order to measure the non-linear AC susceptibilities $\tilde{\chi}_{2n+1}(\omega)$, we have used the conventional mutual-inductance technique [17, 18]. The experimental set-up is shown in figure 1. The AC through the primary coil, which generates the AC field, is supplied by an HP 3325A synthesizer. The in-phase and out-of-phase components of the harmonics of χ are determined by phase-sensitivity detection of the voltage induced across the secondary-coil system when the sample is moved from the centre of one of the identical but oppositely wound secondary coils to the centre of the other. The reference signal at the required frequency $(2n + 1)\omega$ is supplied by a phase-locked generator. As no signal generator is completely free of harmonics in its output voltage, one has to correct the output signal at the frequency $(2n + 1)\omega$ for the linear response of the sample mixed into the harmonic. This is accomplished by the use of a linear reference sample, in our case a Nb metal sample. Since Nb is a superconductor below 9 K, it is a perfect diamagnet having no non-linear contribution to the susceptibility. We have established that the harmonics in the output of the synthesizer, in the frequency region $(2n+1)f = 15$–703 Hz used, have a fixed phase relation to the primary frequency. This enables us to obtain the absolute values of the in-phase and out-of-phase responses of the magnetization to the driving AC field. Maintaining such a phase relation appears to be of primary importance for spin glasses and ferromagnetic materials but has no influence on the results of the antiferromagnets [18].

The sample used in this work was obtained from a master alloy on which specific-heat measurements were performed [19]. It was prepared by induction melting in an argon atmosphere and was formed by spark erosion into a perfect sphere of radius 0.17 cm and of mass 0.2829 g. It was then annealed at 1000 °C for 220 h and analysed chemically by means of spectrophotometry to determine the exact concentration.

3. Results and discussion

In this section, the temperature dependence of the measured in-phase and out-of-phase components of the linear and non-linear (third, fifth and seventh harmonics) susceptibilities of the Pd–5.5 at.% Mn alloy is given for different frequencies. The temperature range is from 1.2 to 4.2 K. The AC field amplitude is 0.7 Oe for the linear-susceptibility measurement and 7 Oe for the non-linear one. We should mention here that the magnitudes of the higher harmonics are much smaller than that of the linear component. Thus, in order to measure higher harmonics, one may need higher values of the driving AC field. Indeed, in this work an AC field amplitude of 0.7 Oe was enough for the linear response, but it was difficult to obtain clear signals for the non-linear responses (especially for the fifth and seventh harmonics) with the same field. Hence we had to increase the amplitude to 7 Oe. However, in order to investigate the amplitude dependence of the third harmonic, a field with an amplitude of 0.5 Oe is also used.

We should emphasize the important point that the measuring technique gives only $\tilde{\chi}_1 h_0$ for the calibration sample of Nb since, as mentioned in section 2, Nb is a perfect diamagnet in the temperature range used and has no non-linear contribution to the susceptibility. Here h_0 is the amplitude of the driving AC field. Therefore, the calibrated linear-susceptibility measurement obtained for Pd–Mn gives directly $\tilde{\chi}_1$ normalized to 1 Oe, but the calibrated third-, fifth- and seventh-harmonic measurements give $\tilde{\chi}_3 h_0^2$, $\tilde{\chi}_5 h_0^4$ and $\tilde{\chi}_7 h_0^6$, respectively. The results reported below are all normalized to 1 Oe. That is, the measured results for the third, fifth and seventh harmonics are divided by h_0^2, h_0^4 and h_0^6, respectively.

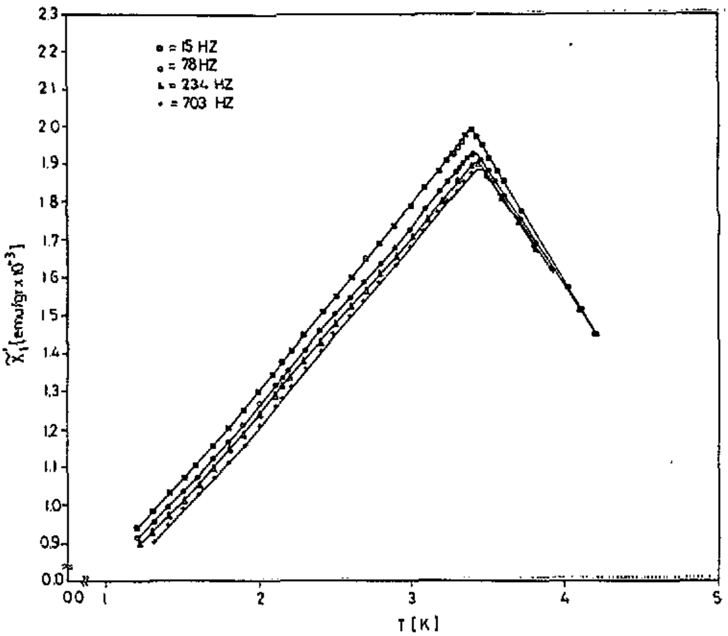

Figure 2. Temperature dependence of the in-phase component of $\tilde{\chi}_1$, measured at $h_0 = 0.7$ Oe, for different frequencies.

Figure 2 shows the temperature dependence of the in-phase component of the linear susceptibility at different frequencies. There is a rather small shift in freezing temperature, i.e. the maximum in $\tilde{\chi}_1'$ moves towards low temperatures with decreasing frequency. For instance it is about 3.40 K for 15 Hz and about 3.45 K for 703 Hz. Therefore, $\Delta T_f/[T_f\,\Delta(\log f)]$ is found to be 8.8×10^{-3}. This value is somewhat smaller than the value of 15×10^{-3} reported for Pd–5.2 at.% Mn [20] for the frequency region from 3.7 to 234 Hz. However, it is about twice the values reported for Cu–Mn [21], Ag–Mn [22] and Au–Mn [23]. Although the magnitude of $\tilde{\chi}_1'$ is not frequency dependent above the freezing temperature, there is an obvious frequency dependence around and below it; here the magnitude decreases with increasing frequency. Such behaviour is in agreement with those of the other well known metallic spin glasses such as Cu–Mn [21] and Ag–Mn [14]. We should point out here that the spin-glass freezing temperature of our sample is estimated to be 3.40 ± 0.02 K from the peak of the linear-susceptibility result for the lowest frequency shown in figure 2. This value is in good agreement with the trend obtained by other workers [8] on the same system.

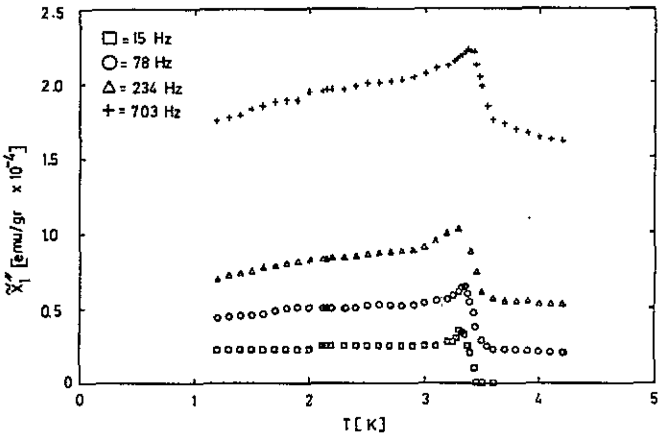

Figure 3. Temperature dependence of the out-of-phase component of $\tilde{\chi}_1$, measured at $h_0 = 0.7$ Oe, for different frequencies.

The magnitude of the out-of-phase component of the linear susceptibility shown in figure 3 has a frequency dependence both above and below the spin-glass freezing temperature. The figure indicates that the out-of-phase component seems to be dominated by the finite conductivity of the specimen in the entire temperature range. In contrast with the magnitude of the in-phase component the magnitude of the out-of-phase component decreases with decreasing frequency (compare figures 2 and 3). Furthermore, the transition temperatures at different frequencies for the out-of-phase component are slightly lower than the corresponding values of those for the in-phase component. For instance, for 15 Hz it is about 3.3 K for the out-of-phase component whereas it is about 3.4 K for the in-phase component.

In order to explore further the behaviour of the Pd–Mn system (in particular, around the spin-glass transition temperature), we have also performed non-linear-susceptibility measurements. In figures 4 and 5 the temperature dependences of the in-phase and out-of-phase components, respectively, of the third harmonic are shown for different frequencies. The frequency values as given in the figures of the third harmonic are three times the frequencies applied to the primary coil. The in-phase component $\tilde{\chi}'_3$ does not have any frequency dependence above 3.7 K, but below this temperature it becomes frequency dependent and around the spin-glass freezing temperature its dependence is striking. As in the case of the in-phase component of the linear susceptibility, T_f shifts to lower temperatures and the magnitudes of the peaks observed at transition increase with decreasing frequency. The peaks are much sharper than those for the linear case. Hence, the transitions to the spin-glass state are well defined with respect to temperature. As can be seen from figure 5, a similar

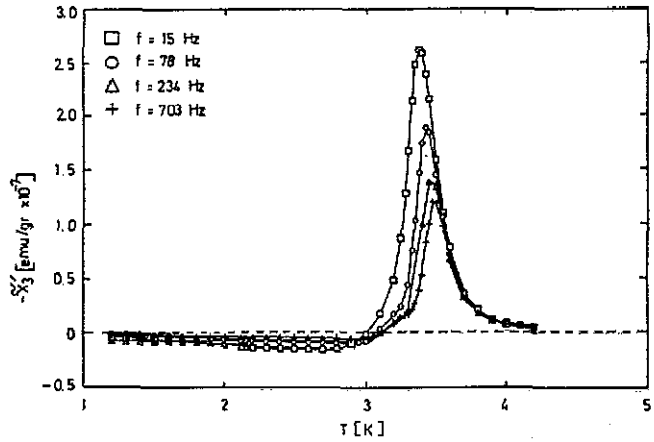

Figure 4. Temperature dependence of the in-phase component of $\tilde{\chi}_3$, measured at $h_0 = 7$ Oe but normalized to 1 Oe, i.e. the measured values were divided by 7^2, for different frequencies.

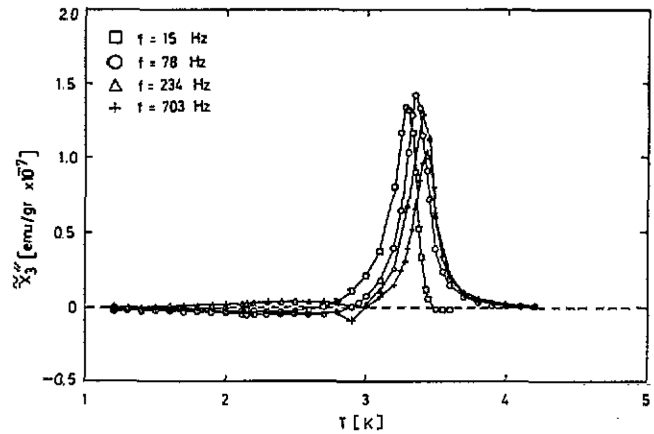

Figure 5. Temperature dependence of the out-of-phase component of $\tilde{\chi}_3$, measured at $h_0 = 7$ Oe but normalized to 1 Oe, i.e. the measured values were divided by 7^2, for different frequencies.

behaviour is also observed for the out-of-phase component $\tilde{\chi}_3''$. However, for this component the magnitudes of the peaks seem to be frequency independent below 234 Hz. Furthermore, we should mention here that the small negative $\tilde{\chi}_3''$-values appearing well below T_f are unphysical. These data points illustrate the experimental error limits and need not be seriously considered.

The absolute values of the third harmonic $\overline{\chi}_3$ obtained by using the square root of the sum of the squares for the in-phase and out-of-phase components are plotted in figure 6 against the temperature for different frequencies. The general behaviour of $\overline{\chi}_3$ is the same as that of $\tilde{\chi}_3'$, as is expected, and the frequency dependence is clearly seen. Figure 7 shows $\overline{\chi}_3$ versus reduced temperature $\epsilon = (T - T_f)/T_f$ on a log–log scale for the data obtained at 15 Hz in the temperature range from 4.2 to 3.43 K (i.e. for the data obtained for temperatures approaching T_f from above). The value of the critical exponent γ obtained from the initial slope given by the line

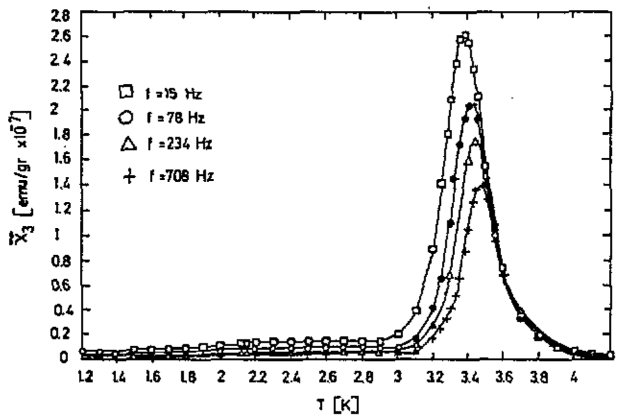

Figure 6. Temperature dependence of the absolute value of $\tilde{\chi}_3$, i.e. $\overline{\chi}_3 \equiv \sqrt{(\chi_3')^2 + (\chi_3'')^2}$ obtained from the data in figures 4 and 5, for different frequencies.

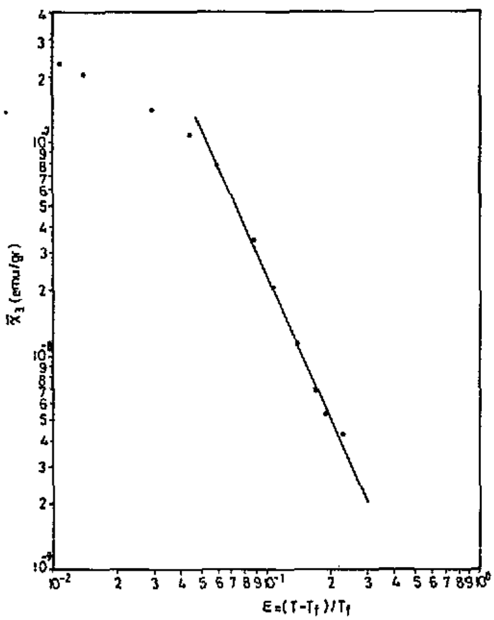

Figure 7. For $3f$ = 15 Hz, the absolute value of $\tilde{\chi}_3$, i.e. $\overline{\chi}_3$, versus $\log \epsilon$ for the data in high-temperature regime between 3.434 and 4.2 K. On the horizontal axis, ϵ is the reduced temperature $(T - T_f)/T_f$, where $T_f = 3.4$ K.

shown in the figure is found to be 2.2 ± 0.2. This asymptotic value is in agreement with the result of the static measurements for Pd–Mn [24]. The corresponding values for AgMn and $Eu_{0.4}Sr_{0.6}S$ are 2.3 ± 0.2 [14] and 2.34 ± 0.20 [18], respectively. A problem here is the limited range of reduced temperatures, less than a decade, where the power-law fit is valid. The curve for $\overline{\chi}_3(\epsilon)$ quickly bends away from the line (see figure 7) and this behaviour indicates the importance of relaxational or dynamical effects which at 15 Hz cause the spin system to lose its equilibrium. So the question of a true phase transition becomes clouded by the non-equilibrium dynamics, and only in the high-temperature regime does there seem to be the start of critical behaviour as represented by the power law with its critical exponent τ.

In order to investigate the AC field amplitude dependence of the third harmonic, a field with an amplitude of 0.5 Oe was also used. The results indicate that the magnitude of $\tilde{\chi}_3$ strongly increases with decreasing field amplitude.

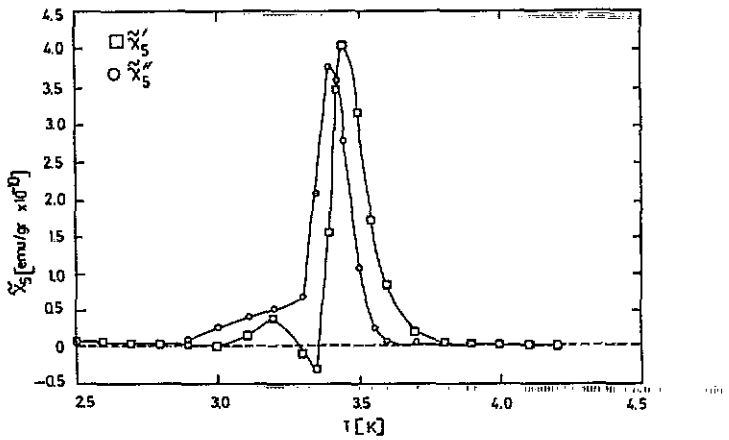

Figure 8. Temperature dependences of the in-phase and out-of-phase components of $\tilde{\chi}_5$, measured at $h_0 = 7$ Oe but normalized to 1 Oe, i.e. the measured values were divided by 7^4, for $5f = 234$ Hz.

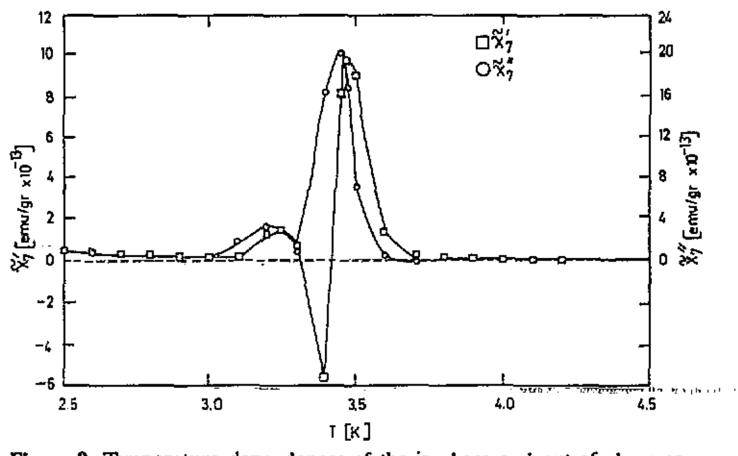

Figure 9. Temperature dependences of the in-phase and out-of-phase components of $\tilde{\chi}_7$, measured at $h_0 = 7$ Oe but normalized to 1 Oe, i.e. the measured values were divided by 7^6, for $7f = 234$ Hz.

Since the demagnetization correction is negligible for our sample, it is possible to measure the higher-order harmonics, such as $\tilde{\chi}_5$ and $\tilde{\chi}_7$. Thus, fifth- and seventh-harmonic measurements are also carried out on the Pd–Mn sample used in this work. Figures 8 and 9 show the in-phase and out-of-phase components of $\tilde{\chi}_5$ and $\tilde{\chi}_7$, respectively. By comparing figures 4, 5, 8 and 9, one can conclude that susceptibility upturns around T_f become faster with increasing harmonics, which is in agreement with the conclusion of Levy [14]. The in-phase components of both the fifth and the seventh harmonics exhibit negative values in a very small temperature interval below T_f. We believe that this behaviour is caused by some kind of dynamical effects. Because, as indicated in the introduction, the non-linear susceptibilities $\tilde{\chi}_{2n+1}$, should diverge according to $\epsilon^{-n(\gamma+\beta)+\beta}$, for $n \geqslant 1$ [16], $\overline{\chi}_5/\overline{\chi}_3$ and $\overline{\chi}_7/\overline{\chi}_5$ scale as $\epsilon^{-(\gamma+\beta)}$, or alternatively as $(\overline{\chi}_3)^{1+\beta/\gamma}$, just above T_f. Accordingly, one can obtain the critical exponent β by plotting $\overline{\chi}_5/\overline{\chi}_3$ or $\overline{\chi}_7/\overline{\chi}_5$ against $\overline{\chi}_3$ on a log–log scale. This should be a straight line when approaching T_f from above [14, 25]. The slope of the line is $1+\beta/\gamma$. Figure 10 shows $\overline{\chi}_5/\overline{\chi}_3$ versus $\overline{\chi}_3$, in the temperature range from 3.8 to 3.45 K for a frequency of 234 Hz. By using the value of $\gamma = 2.2$, β is found to be about 0.2. This value is clearly smaller than those β-values reported for other spin glasses which are in the range 0.5–1 [10, 11, 14, 25]. We believe that the reason for obtaining smaller values of β is due to the AC field amplitude dependences of the non-linear susceptibilities. As we have already mentioned above, our measured third harmonic strongly depends on the amplitude of the driving AC field. With increasing field amplitude, the responses seem to be suppressed around T_f. It is physically expected that this suppression becomes stronger with increasing harmonics, i.e. $\overline{\chi}_5$ is suppressed more than $\overline{\chi}_3$, etc. This may cause the $\overline{\chi}_5/\overline{\chi}_3$ or $\overline{\chi}_7/\overline{\chi}_5$ ratios to be smaller than expected. In our calculations we have used the results obtained in an AC field of 7 Oe amplitude and 234 Hz frequency. If we could have used fields with smaller amplitudes, we might have found higher β-values. However, as we have emphasized in the first paragraph of this section, the decreasing AC field amplitude causes the ratio of real signal to noise to decrease unavoidably. Therefore, especially for measurements of the fifth and seventh harmonics, smaller fields gave unsatisfactory results.

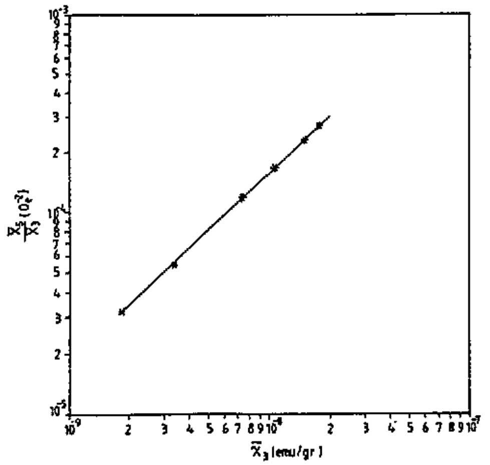

Figure 10. $\overline{\chi}_5/\overline{\chi}_3$ against $\overline{\chi}_3$ on a log–log scale, for the temperature range 3.8–3.45 K. The $\overline{\chi}_3$ and $\overline{\chi}_5$ data are taken from figure 6 ($3f = 234$ Hz) and figure 8 ($5f = 234$ Hz), respectively.

4. Conclusions and summary

We have carried out a systematic investigation of the linear and non-linear AC susceptibilities $\tilde{\chi}_{2n+1}$ ($n = 0, 1, 2, 3$) of the spin glass Pd–Mn with 5.5 at.% Mn. We find an incipient divergence in $\tilde{\chi}_{2n+1}$ for $n \geqslant 1$. Thus, our results confirm that the non-linear susceptibility is indicative of the spin-glass phase transition as predicted in the phenomenological and scaling theories [16]. By measuring the third harmonic $\tilde{\chi}_3$, which diverges as $\epsilon^{-\gamma}$ the value of γ has been found to be 2.2 ± 0.2 from the data just above T_f, which is in reasonable agreement with the values determined for other spin glasses. However, the relaxational or dynamical effects seem to cause the spin system to lose its equilibrium as T_f is approached. Hence, the theoretically expected phase transition becomes inaccessible to experiment because of the non-equilibrium dynamics and, therefore, the critical behaviour represented by $\epsilon^{-\gamma}$ is valid only in a limited reduced-temperature range, less than a decade, in the high-temperature regime.

In addition, since the demagnetization effect for our sample is not very important, we were also able to measure susceptibility peaks for the fifth and seventh harmonics at $f = 234$ Hz and an AC field amplitude of 7 Oe. Since the theory [16] indicates that $\overline{\chi}_5/\overline{\chi}_3$ scales as $(\overline{\chi}_3)^{1+\beta/\gamma}$, by plotting $\overline{\chi}_5/\overline{\chi}_3$ against $\overline{\chi}_3$, one can also obtain the value of β. Our data give $\beta = 0.2$ which is much smaller than the value expected (about 0.7). This is probably caused by the above dynamical effects and the high amplitude and high frequency of the AC driving field which had to be used to obtain noise-free data especially for the fifth and seventh harmonics.

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