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Heisenberg-like critical properties in ferromagnetic $Nd_{1-x}Pb_xMnO_3$ single crystals

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Abstract

Static magnetization for single crystals of insulating $Nd_{0.85}Pb_{0.15}MnO_3$ and marginally conducting $Nd_{0.70}Pb_{0.30}MnO_3$ has been studied around the ferromagnetic to paramagnetic transition temperature T_C. Results of measurements carried out in the critical range $|(T - T_C)/T_C| \leqslant 0.1$ are reported. Critical exponents β and γ for the thermal behaviour of magnetization and susceptibility have been obtained both by modified Arrott plots and the Kouvel–Fisher method. The exponent δ independently obtained from the critical isotherm was found to satisfy the Widom scaling relation $\delta = \gamma/\beta + 1$. For both compositions the values of exponents are consistent with those expected for isotropic magnets belonging to the Heisenberg universality class with short-range exchange in three dimensions. Correspondingly, the specific heat displays only a cusp-like anomaly at the critical temperature of these crystals which is consistent with an exponent $\alpha < 0$. The results show that the ferromagnetic ordering transition in $Nd_{1-x}Pb_xMnO_3$ in the composition range $0.15 \leqslant x \leqslant 0.40$ is continuous. This mixed-valent manganite displays the conventional properties of a Heisenberg-like ferromagnet, irrespective of the differing transport properties and in spite of low ordering temperatures $T_C = 109$ and 147.2 K for $x = 0.15$ and 0.30, respectively.

1. Introduction

In doped perovskite manganites $R_{1-x}A_xMnO_3$, with R rare-earths or lanthanides and A divalent ions [1, 2], a huge negative magnetoresistance is observed close to the ferromagnetic transition in a certain range of composition $x \approx 0.15$–0.4 [2–5]. The effect has prompted renewed interest in this family of compounds (for recent reviews, see [6]). A metal–insulator

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(MI) transition in these manganites is usually associated with the ferromagnetic (FM) to paramagnetic (PM) transition. The basic physics of the ferromagnetic metallic state in the mixed-valent manganites has been explained by the double-exchange (DE) mechanism [7–9]. Hence, the strong 'colossal' magnetoresistance (CMR) appears as a consequence of this DE interaction mechanism between localized moments, which is mediated by mobile electrons that also carry the electrical transport. This picture had to be modified, as it was found that the effective carrier–spin interaction is too weak to cause the CMR [10]. Still, the DE model is widely believed to be sufficient to explain the properties of the metallic ferromagnetic state in a class of mixed-valent manganites with high transition temperatures, epitomized by $La_{0.7}Sr_{0.3}MnO_3$ [6, 11]. At present it is not clear which mechanisms [10, 12–14] in other ferromagnetic manganites cause the marked deviations of their magnetic and transport properties from the DE model.

In order to understand the relation between the MI transition and CMR, the nature of the ferromagnetic transition in these materials must be clarified. The DE coupling has distinguishable properties compared to the ordinary direct exchange interactions in localized spin models. Since the interaction originates from the motion of conduction electrons, the effective range of the interaction may depend on the kinetics of electrons and, therefore, on temperature through the thermal fluctuations.

So, it is not clear how the interaction is renormalized near the critical point and what universality class governs the magnetic transition. Critical phenomena in the double-exchange model have been first described within mean-field theory [15]. Later, Motome and Furukawa [16] predicted, on the basis of computational studies for simplified DE models, that the FM–PM transition in CMR manganites should belong to the short-range Heisenberg universality class, and emphasized the importance of experimental studies on critical properties for understanding the magnetic coupling mechanism in FM manganites. Limitations on the computational investigations generally hinder the precise determination of critical properties for realistic models. Further computational studies on DE systems show that FM–PM transitions may become discontinuous, depending on doping level and competition with antiferromagnetic superexchange [17]. Similar features are found in phenomenological models considering phase separation between metallic–ferromagnetic and insulating–paramagnetic nanoscale regions as the underlying physical process that drives the FM–PM transition [18].

A number of experimental studies of critical phenomena have been previously made on ferromagnetic manganites [19–23], with some controversial results concerning the critical exponents and even the order of the magnetic transitions. For example, Morrish *et al* [20] had found a high value of $\beta = 0.495$ in a $La_{0.65}Pb_{0.44}MnO_3$ crystal. This is in good agreement with later results from Lofland *et al* [21] which yield $\beta = 0.45$ in $La_{0.7}Sr_{0.3}MnO_3$, indicating a mean-field-like behaviour at the magnetic phase transition. In contrast to this, neutron scattering experiments on $La_{0.7}Sr_{0.3}MnO_3$ give a rather low β value, 0.295 [22]. Due to the drastic difference in the physical properties between $La_{0.7}Ca_{0.3}MnO_3$ with a Curie temperature $T_C \simeq 250$ K and $La_{0.7}Sr_{0.3}MnO_3$ ($T_C \simeq 340$ K), some authors distinguish the archetypical CMR compound $La_{0.7}Ca_{0.3}MnO_3$ as a 'low-T_C' manganite. It has a higher resistivity, a sharper resistivity peak near T_C and a larger CMR than $La_{0.7}Sr_{0.3}MnO_3$ [24]. The continuous nature of the FM to PM transition has been investigated carefully for high-T_C manganites such as $La_{0.7}Sr_{0.3}MnO_3$ ($T_C = 355$ K) [19]. On the other hand, an unexpected dependence of the order of the ferromagnetic transition on doping has been reported for $La_{1-x}Ca_xMnO_3$. The transition in $La_{0.8}Ca_{0.2}MnO_3$ is continuous [25], while that for $La_{0.7}Ca_{0.3}MnO_3$ was found to be of first order in various studies [26–28]. Quenched disorder in the Mn sublattice of $La_{0.67}Ca_{0.33}MnO_3$ introduced by Ga doping leads to a rounded magnetic phase transition characterized by the critical properties of a Heisenberg-like ferromagnet [29]. This result implies a conventional

behaviour of the magnetic Mn sublattice in this manganite which is, however, impaired by additional effects in the undoped $La_{0.67}Ca_{0.33}MnO_3$. There are still other conflicting results about the ferromagnetic transition in the series of compounds $La_{1-x}Ca_xMnO_3$. From static magnetization studies, a tricritcal behaviour was found for $x = 0.40$ [30], while critical exponents close to those of the Heisenberg universality class have been found for $x = 0.20$ [25]. Contrary to this, Rivadulla *et al* [31] propose a heterogeneous magnetic ordering owing to intrinsic random fields or random anisotropies for the ranges $x > 0.4$ and $x < 0.25$ with a continuous phase transition. Finally, Souza *et al* [32] find a continuous transition from a thermodynamic study on $La_{0.7}Ca_{0.3}MnO_3$ with a large positive exponent, $\alpha \simeq 0.93$, for the singular behaviour of the specific heat.

In order to test whether first-order or unconventional magnetic transitions can be observed also in other 'low-T_C' manganites, we have recently investigated the critical properties of a single crystal of $Nd_{0.6}Pb_{0.4}MnO_3$ with an MI transition T_{MI} close to $T_C \simeq 156$ K and a magnetoresistance ratio of 85% [33, 34]. This study demonstrated that the PM–FM transition in $Nd_{0.6}Pb_{0.4}MnO_3$ is continuous, with critical exponents belonging to the three-dimensional Heisenberg universality class with short-range interactions. Thus, the magnetic phase transition in this mixed-valent manganite is that of a conventional isotropic magnetic system.

Here we report precise estimations of critical exponents for low-T_C $Nd_{1-x}Pb_xMnO_3$ single crystals with $x = 0.15$ and 0.3 from magnetization data in the critical region. The crystals with these concentrations are interesting because of their differing transport properties [35]. The sample with $x = 0.15$ displays insulating behaviour, i.e. a negative temperature coefficient of resistivity down to low temperature. On the other hand, the sample with $x = 0.3$ shows an insulator to metal transition around T_C but it crosses over towards insulating behaviour at lower temperatures. Therefore, these two samples probe a region of the phase diagram where the connection, implied by the DE mechanism, between metallic conductivity behaviour and ferromagnetism is violated. Moreover, for low doping ($x < 0.2$) a possible coupling of the ferromagnetic ordering to antiferromagnetic fluctuations through a competition between ferromagnetic double-exchange and antiferromagnetic superexchange, or by coupling to charge/orbital-ordering modes is possible, which could cause fluctuation-induced first-order transitions [36, 37].

We find that the critical exponents for the $Nd_{1-x}Pb_xMnO_3$ system with $x = 0.15, 0.3$ also belong to the three-dimensional Heisenberg universality class with short-range interactions. Hence the PM–FM phase transition for these systems remains that of a conventional magnet, in spite of the fact that the transport properties are markedly different for these compositions as compared to the metallic system $Nd_{0.6}Pb_{0.4}MnO_3$.

In the next section, the experimental study is described. Section 3 introduces the method of the static-scaling analysis. Section 4 gives the results for the magnetization data along with data for the specific heat measured for the same crystals.

2. Experimental details

The single crystals used for the measurement were grown by a flux method [38, 39]. Crystals with various Pb concentrations were grown. Among these crystals, $Nd_{0.85}Pb_{0.15}MnO_3$ and $Nd_{0.7}Pb_{0.3}MnO_3$ were chosen for the present investigation. Extensive magnetization data $M(T, H)$ were collected on well characterized $Nd_{1-x}Pb_xMnO_3$ single crystals of dimensions $3.5 \times 2.3 \times 1$ mm^3 ($x = 0.15$) and $2.15 \times 1.875 \times 0.9$ mm^3 ($x = 0.3$) in external static magnetic fields H up to 48 kOe. Using a SQUID magnetometer, the measurements were carried out in the temperature ranges 100 K $< T <$ 129 K for $x = 0.15$ ($T_C = 109$ K) and

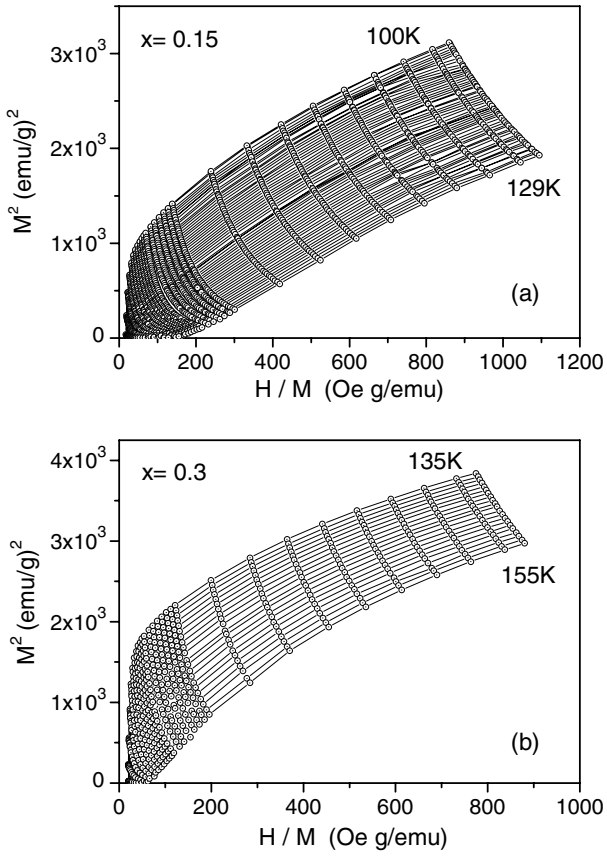

Figure 1. Arrott plot of $Nd_{1-x}Pb_xMnO_3$ for (a) $x = 0.15$, (b) $x = 0.3$.

135 K $< T <$ 155 K for $x = 0.3$ ($T_C = 148.5$ K) encompassing the critical region near the PM–FM phase transition. The data were measured in temperature steps of 0.5 K with the magnetic field direction along the long edge of the crystals, which corresponds to a high-symmetry direction. The magnetization versus field data are corrected by a demagnetization factor that has been determined by a standard procedure from low-field DC-susceptibility measurements. The specific heat measurements down to 2 K were done with a PPMS system (Quantum Design).

3. Static scaling for magnetization data

According to the scaling hypothesis, a second-order phase transition near the Curie point T_C is characterized by a set of interrelated critical exponents, α, β, γ, δ etc, and a magnetic equation of state [40]. The temperature dependence of the spontaneous magnetization $M_S(T) = \lim_{H\to 0} M$ just below T_C is governed by exponent β through the relation

$$M_S(T) = M_0(-\epsilon)^{\beta}, \qquad \epsilon < 0, \tag{1}$$

and that of the inverse initial susceptibility $\chi_0^{-1}(T) = \lim_{H\to 0}(H/M)$ just above T_C by γ through

$$\chi_0^{-1}(T) = (h_0/M_0)\epsilon^{\gamma}, \qquad \epsilon > 0 \tag{2}$$

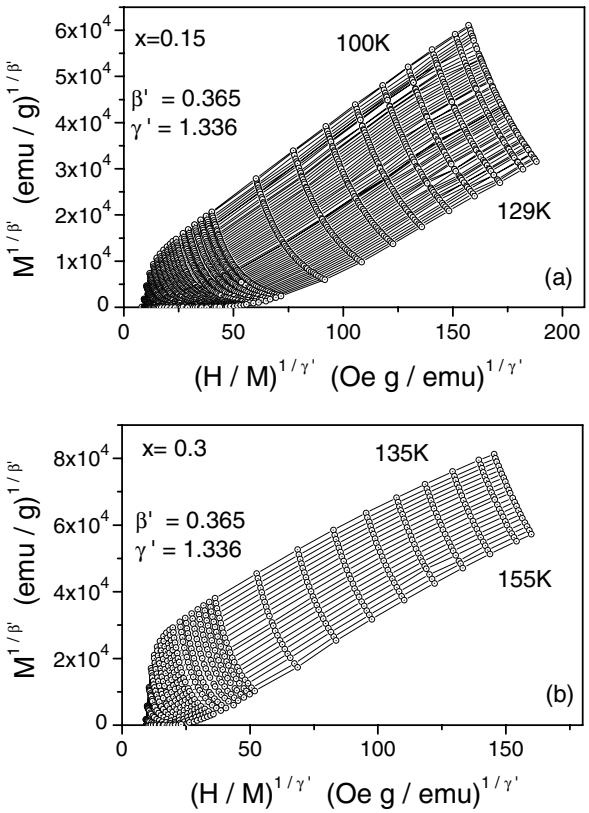

Figure 2. Modified Arrott plot of $Nd_{1-x}Pb_xMnO_3$ for (a) $x = 0.15$, (b) $x = 0.3$ with critical exponents of 3D Heisenberg universality class.

and the exponent δ relates M and H at T_C as

$$M = DH^{1/\delta}, \qquad \epsilon = 0 \tag{3}$$

where $\epsilon = (T - T_C)/T_C$ and M_0, h_0/M_0 and D are the critical amplitudes.

The magnetic equation of state in the critical region is written as

$$M(H, \epsilon) = \epsilon^{\beta} f_{\pm}(H/\epsilon^{\beta+\gamma}), \tag{4}$$

where f_+ for $T > T_C$ and f_- for $T < T_C$, respectively, are regular functions. Equation (4) implies that M/ϵ^{β}, as a function of $H/\epsilon^{\beta+\gamma}$, falls on two universal curves, one for temperatures above T_C and the other for temperatures below T_C.

4. Results and discussions

Figure 1 shows the Arrott plot, M^2 versus H/M, constructed from the raw M–H isotherms of the two samples after correcting the external magnetic field for demagnetization effects. According to mean-field theory, near T_C these curves should show a series of straight lines for different temperatures and the line at $T = T_C$ should pass through the origin. In the present case the curves in the Arrott plot are nonlinear and concave. This shows that the mean-field theory does not describe the critical behaviour for this system. Therefore, the values of $M_S(T)$ and $\chi_0^{-1}(T)$ were determined using a modified Arrott plot [41], in which $M^{1/\beta'}$ is plotted versus

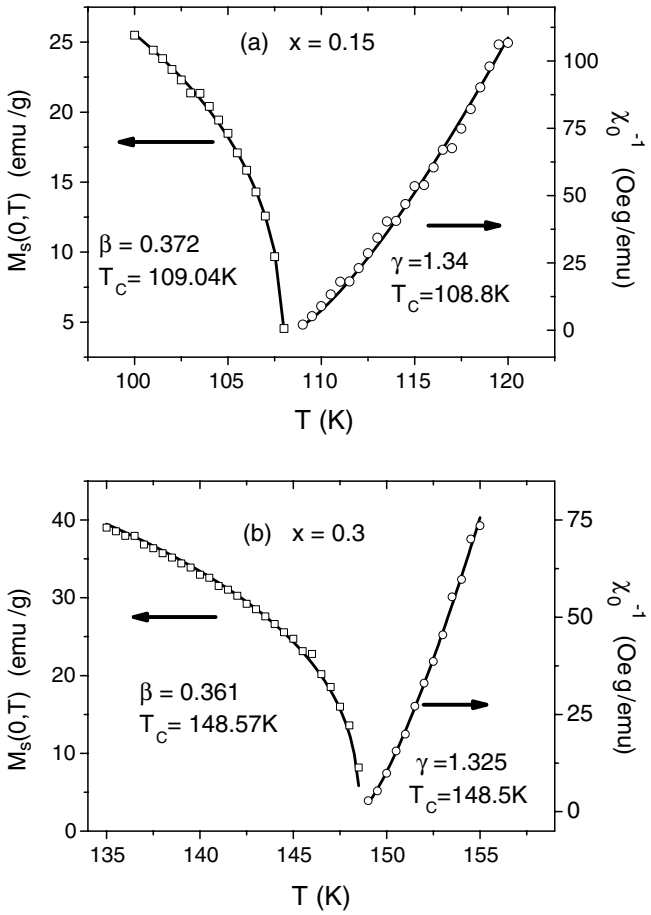

Figure 3. Plots of $M_S(T)$ and $\chi_0^{-1}(T)$ of $Nd_{1-x}Pb_xMnO_3$ for (a) $x = 0.15$, (b) $x = 0.3$.

$(H/M)^{1/\gamma'}$ (see figure 2). As trial values, we have chosen $\beta' = 0.365$ and $\gamma' = 1.336$, the critical exponents of the 3D Heisenberg model [42]. As this plot results in nearly straight lines (for sufficiently high fields) a linear extrapolation from fields above 2 kOe to the intercepts with the axes $M^{1/\beta'}$ and $(H/M)^{1/\gamma'}$ gives the values of spontaneous magnetization $M_S(T)$ and inverse susceptibility $\chi_0^{-1}(T)$, respectively. These values as functions of temperature are plotted in figure 3. The continuous curves in figure 3 show the independent power law fits to $M_S(T)$ and $\chi_0^{-1}(T)$ according to equations (1) and (2), respectively. Table 1 lists the results of the fits for the two compositions. These results show that our trial values are already close to the correct critical exponents. In particular, the two fits for $M_S(T)$ and $\chi_0^{-1}(T)$ yield consistent values for T_C. It is to be noted that the origin for the deviations from a straight line in the modified Arrott plots at low field are generally not fully understood [43]. In any case, the shape anisotropy of the samples has no discernible effect on these low-field deviations [44]. In fact, the curves passing through the origins (after extrapolation of the straight line region) in figure 2 belong to $T = 109.0(5)$ K for $x = 0.15$ and 148.5(5) K for $x = 0.3$, which are near to the respective T_C values obtained from the fits shown in figure 3. Alternatively, the values of T_C, β and γ have also been obtained by the Kouvel–Fisher (KF) method [45]. According to this method, plots of $M_S(dM_S/dT)^{-1}$ versus T and $\chi^{-1}(d\chi^{-1}/dT)^{-1}$ versus T should yield

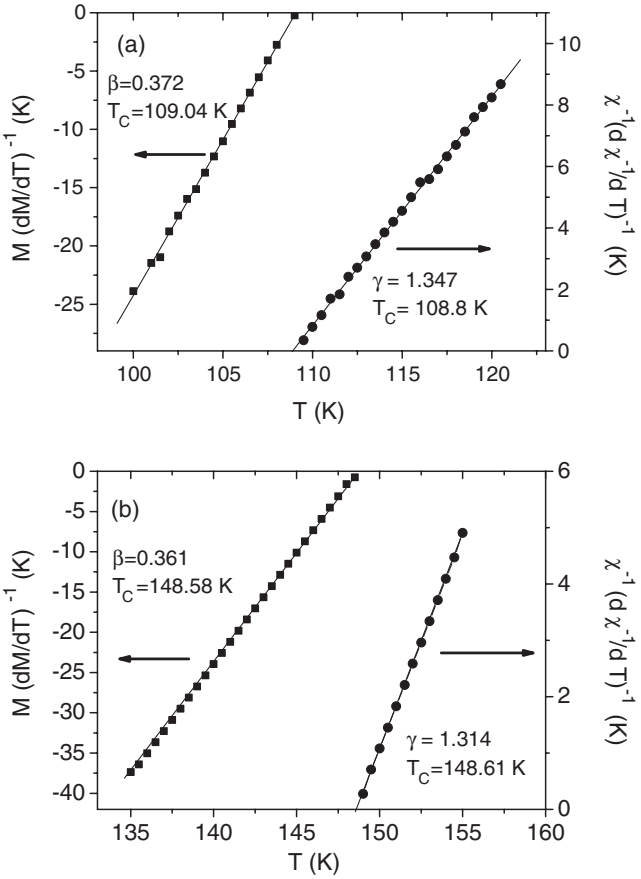

Figure 4. Kouvel–Fisher plots of $Nd_{1-x}Pb_xMnO_3$ for (a) $x = 0.15$, (b) $x = 0.3$.

Table 1. Values of critical exponents (β, γ) of $Nd_{1-x}Pb_xMnO_3$ ($x = 0.15, 0.3$) using the modified Arrott (MA) and Kouvel–Fisher (KF) methods, and value for exponent δ determined from the critical isotherm.

Composition	Method	T_C, β	T_C, γ	δ
$x = 0.15$	MA	109.04(4) K; 0.372(1)	108.8(1) K; 1.34(3)	4.67(3)
	KF	109.04(1) K; 0.372(4)	108.8(2) K; 1.347(1)	
$x = 0.30$	MA	148.57(8) K; 0.361(13)	148.5(3) K; 1.325(1)	4.62(4)
	KF	148.58(2) K; 0.361(5)	148.61(3) K; 1.314(2)	

straight lines with slopes $1/\beta$ and $1/\gamma$, respectively. When extrapolated to the ordinate, these straight lines should intercept the T axes equally at the Curie temperature. These plots are shown in figure 4 for both compositions x. The straight lines obtained from a least-square fit to these data give the values of β and γ, which are shown in table 1.

The value of δ has been directly obtained by plotting the critical isotherm. Figure 5 shows $M(T_C, H)$ versus H on a log–log scale. According to equation (3), this should be a straight

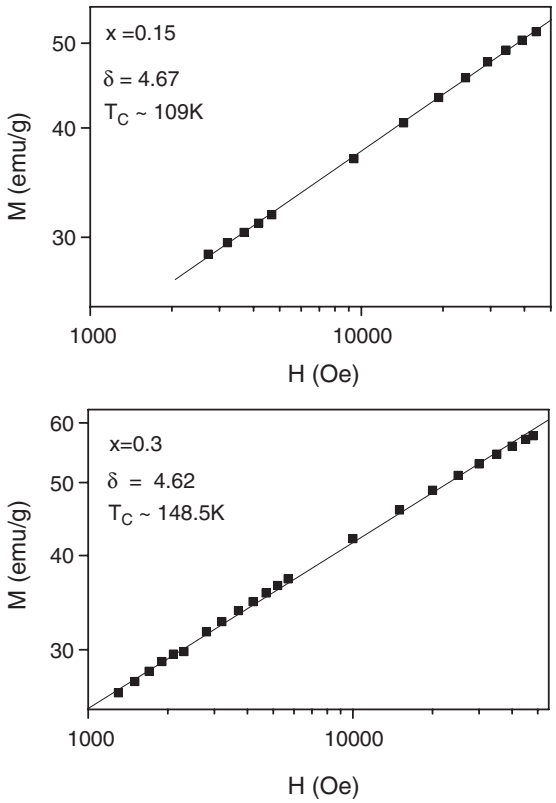

Figure 5. $M_S(T = T_c, H)$ versus H plots of $Nd_{1-x}Pb_xMnO_3$ for $x = 0.15$ and 0.3 in log–log scale.

line with a slope $1/\delta$. From the linear fit we obtain $\delta = 4.67(3)$ for $x = 0.15$ and $\delta = 4.62(4)$ for $x = 0.3$.

The critical exponents β, γ, and δ are related through the Widom scaling relation [46]

$$\delta = 1 + \gamma/\beta. \tag{5}$$

Using the above determined values of β and γ from figure 3, the critical exponents obtained in this study obey the Widom scaling relation remarkably well for both compositions studied. In order to check whether our data in the critical region obey the magnetic equation of state equation (4), M/ϵ^{β} as a function of $H/\epsilon^{\beta+\gamma}$ is plotted in figure 6 for $x = 0.15$ and 0.3. It can be clearly seen that all the points fall on two curves, one for $T < T_C$ and the other for $T > T_C$. Thus the obtained values of the critical exponents and T_C are reliable and in agreement with the scaling hypothesis.

To support the results of the scaling analysis on static magnetization data, we have measured the specific heat for our crystals $Nd_{1-x}Pb_xMnO_3$ ($x = 0.15, 0.3$) (figure 7). Broad cusps of the specific heat observed at the respective ferromagnetic transition temperatures are consistent with Heisenberg-like behaviour with a negative critical exponent for the specific heat ($\alpha < 0$). The results further corroborate the continuous nature of the magnetic phase transition.

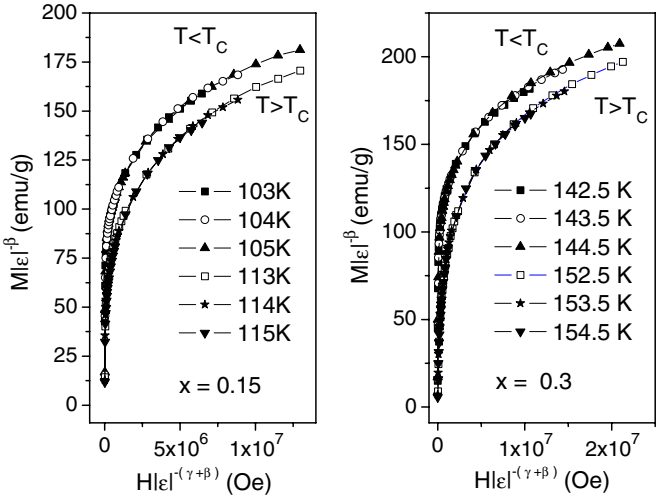

Figure 6. Scaled isotherms of $Nd_{1-x}Pb_xMnO_3$ for $x = 0.15$ and 0.3 below and above the transition temperature using β and γ as defined in the text.

(This figure is in colour only in the electronic version)

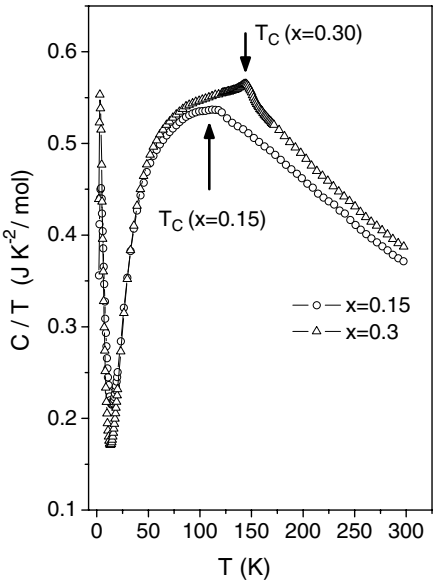

Figure 7. C/T versus T plots of $Nd_{1-x}Pb_xMnO_3$ for $x = 0.15$ and 0.3 indicating a continuous transition at T_C.

5. Conclusions

In summary, we have studied the critical behaviour of $Nd_{1-x}Pb_xMnO_3$ in the temperature region around T_C through static magnetization measurements and determined the values of critical temperature T_C and the exponents β, γ and δ, which provide a consistent description for a continuous PM–FM transition according to scaling laws. It has to be noted that the

width of the chosen temperature interval around T_C has been systematically curtailed to check that this scaling description is stable within reasonable bounds. This means that the reduced temperature range of this study, $10^{-3} < |\epsilon| \leqslant 10^{-1}$, lies within the asymptotic range of the critical behaviour for these magnets, while cross-over effects due to secondary magnetic effects, such as dipolar couplings and magnetic anisotropies, do not appreciably affect the scaling.

Clearly, the values of critical exponents found for $Nd_{1-x}Pb_xMnO_3$ agree with those of a 3D Heisenberg-like ferromagnet with short-range interactions. This is further supported by specific heat measurements. The magnetic transitions in the investigated crystals conform to the behaviour of conventional isotropic 3D magnets. Therefore, although the transition temperatures in $Nd_{1-x}Pb_xMnO_3$ are low, their behaviour is distinct from the anomalies found for $La_{1-x}Ca_xMnO_3$. The magnetic properties in the system $Nd_{1-x}Pb_xMnO_3$ appear to be closer to those expected for mixed-valent double-exchange materials.

Acknowledgments

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