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Critical behavior of electrical resistivity in amorphous Fe–Zr alloys

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Abstract. Electrical resistivity (ρ) of the amorphous (a-)$Fe_{100-c}Zr_c$ ($c = 8.5$, 9.5 and 10) alloys has been measured in the temperature range 77 to 300 K, which embraces the second-order magnetic phase transition at the Curie temperature point T_c. Analysis of the resistivity data particularly in the critical region reveals that these systems have a much wider range of critical region compared to other crystalline ferromagnetic materials. The value of T_c and specific heat critical exponent, α has the same values as those determined from our earlier magnetic measurements. The value of α for all the present investigated alloys are in close agreement with the values predicted for three-dimensional (3D) Heisenberg ferromagnet systems, which gives contradiction to the earlier results on similar alloys. It is observed from the analysis that the presence of quenched disorder does not have any influence on critical behavior.

Keywords. Amorphous materials; critical region; ac susceptibility; electrical resistivity; critical exponents.

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1. Introduction

The amorphous Fe–Zr metallic glasses in the composition range near 90 at. % of Fe forms an interesting disorder magnetic behavior. In some way, these alloys behave magnetically like re-entrant spin-glass systems. The phase diagram for Fe rich amorphous FeZr alloys shows that the ferromagnetic-like state occurs with the Curie temperature decreases monotonically with increasing Fe concentration from 87 to 93 at.% of Fe [1–3], which represents the one way of nearest approach to the pure amorphous iron. The critical behavior of such spin systems with quenched or frozen disorder has recently drawn interesting attention. General scaling arguments show that in this case random disorder does not change the asymptotic critical exponents (Harris criterion [4]), in the sense if $\alpha < 0$ (i.e. for $d = 3$ Heisenberg and $d = 3xy$ systems with spin dimensionality $n = 3$ and $n = 2$ respectively) quenched randomness acts as an irrelevant scaling field and hence leaves the sharpness of the transition as well as the values of the static critical exponents of the pure system unaltered and if $\alpha > 0$, a cross-over from pure to random fixed point (characterized by critical

exponents, whose values are widely different from the actual one, so that the sign of α changes. i.e $\alpha > 0$) occurs. High temperature series expansion calculations [5], renormalization group calculations [6,7], and a field theoretical calculation [8] corroborate this argument. The specific heat and magnetic measurements [9–11] on some amorphous ferromagnetism reports the theoretical point [4,6,12] of view that the addition of short-range disorder to a pure system, which undergoes a second order phase transition should not alter, if the specific heat critical exponent of the system is negative. A serious problem one has to consider, when studying the magnetic phase transitions with structural disorder is the question whether or not the topological or substantial short-range disorder is the only relevant parameter describing the structure. Principally, a thermodynamic phase transition can also exist, but the asymptotic critical behavior, which can only be observed, if the correlation length is larger than the spatial correlations of the density flucutations, which can be shifted to low reduced temperature regions.

Yamauchi, Onodora and Yamamoto [13] reported based on magnetic measurements near T_c that these systems present an anomalous critical behavior with the exponents β, γ and δ, differing subsequently from those expected for homogeneous and collinear ferromagnets. However, Kaul [14] has found experimentally and from reanalysis of experimental data (reported by Hiroyoshi *et al* [15] on magnetization for a-$Fe_{91}Zr_9$ and those of Yamauchi *et al* for $Fe_{92}Zr_8$) that the exponents β and γ, which do not significantly differ from those normally expected for a three-dimensional (3-D) Heisenberg ferromagnet in the real critical region. From these studies, it is not clear, whether these systems will exhibit a ordered 3D Heisenberg like ferromagnet behavior in asymptotic critical region or not.

A complete understanding of the critical phenomena in amorphous systems, therefore demands a systematic experimental study on materials having varying degrees and types of quenched disorder. The specific heat measurement is the most informative experimental tool for this kind of study. Because, it has very small critical exponent and hence it is most sensitive to the smearing of the phase transition. But the main requirement for this study is that a large amount of sample is needed to perform accurate specific heat measurement in the critical region. An alternative, but indirect method to study the critical behaviors of specific heat makes use of theoretically predicted [16–19] and experimentally [20–23] verified fact that the magnetic contribution to the temperature derivative of electrical resistivity, $\mathrm{d}\rho_m/\mathrm{d}T$, since thermal derivative of resistivity is directly proportional to the specific heat of the system $[\mathrm{d}\rho_m(T)/\mathrm{d}T \propto C_m(T)]$ in the critical region. The electrical resistivity measurement can be preformed even on a very small sample with closed interval of temperature and it could be given same types of information about the nature of magnetic phase diagram as the specific heat measurements. We have chosen here a set of binary alloy samples of $Fe_{100-c}Zr_c$ ($c = 8.5, 9.5$ and 10) for critical behavior analysis using the highly precise electrical resistivity data in order to resolve the controversy, whether or not the iron rich amorphous Fe–Zr alloys exhibit a normal phase transition to long-range ferromagnetic order at T_c and to confirm whether the presence of disorder has any influence on critical behavior of resistivity or not?

2. Experimental details

Amorphous (a-)$Fe_{100-c}Zr_c$ $(c = 8.5, 9.5, 10)$ alloy ribbons of 1–3 mm width and 15–30 μm thickness was prepared by melt-spun quenching technique from high purity elements.

The amorphous nature of the samples is confirmed by X-ray diffraction studies using Cu-K_α radiation. Highly precise electrical resistivity measurements were performed in these alloys in the temperature range from 77 to 300 K using a standard dc four-probe technique in the temperature interval of approximately 0.7 K and 0.1 K in the transition region with a resolution better than one part in 10^5 using Keithly nanovoltmeter system. The direction of the constant current passing through the sample was reversed at each temperature, in order to correct the measured voltage for any thermo-emf generated due to some temperature-gradient across the sample. The sample temperatures were determined with an accuracy of 10 mK by precisely measuring the resistance of a Pt sensor and calibrated silicon diode thermometer.

3. Results and discussion

From the measured electrical resistivity data as a function temperature (ρ vs. T), the temperature derivative of resistivity ($\mathrm{d}\rho/\mathrm{d}T$) has been derived as follows: A set of three adjacent experimental points with coordinates (ρ_1, T_1), (ρ_2, T_2) and (ρ_3, T_3). The slopes $(\Delta\rho/\Delta T) = (\rho_2 - \rho_1)/(T_2 - T_1)$ and $(\Delta\rho/\Delta T)' = (\rho_3 - \rho_2)/(T_3 - T_2)$ are computed and then their average value $[(\Delta\rho/\Delta T) + (\Delta\rho/\Delta T)']/2$ is taken to represent the value of $(\mathrm{d}\rho/\mathrm{d}T)$ at the middle temperature $T = (T_1 + T_3)/2$. The values of temperature derivative of resistivity at different temperatures so obtained, are normalized to the resistivity value at Curie temperature $\rho(T_\mathrm{c})$, because the absolute value of $\mathrm{d}\rho/\mathrm{d}T$ is inaccurate to within 7–10 %, mainly due to the measurement in thickness of the ribbon sample, since the thickness of the sample is in the order of μm. Figure 1 depict the temperature derivative of resistivity normalized with respect to resistivity at Curie temperature, is plotted as a function of temperature in the temperature range 77 to 300 K for a-Fe$_{100-c}$Zr$_c$ with $c = 8.5, 9.5$ and for $c = 10$ respectively. The temperature derivative of resistivity is positive and its value increases as Zr concentration increases almost by one order at room temperature. But it decreases continuously with temperature and assumes negative values. At about 187 K, 208 K, 228 K, there is a clearly defined kink, where the slope changes rapidly for $c = 8.5, 9.5$ and 10 respectively. The anomaly at these particular temperatures corresponds well to the temperature, where the paramagnetic to ferromagnetic transition is reported from bulk magnetization (BM) and ac susceptibilty (ACS) measurements [14,27]. Below this point, the temperature derivative of resistivity decreases till 77 K. The total fractional change in the thermal derivative of resistivity from 77 to 300 K is almost equal for all the investigated samples. It is very clear from the figures that the sharpness of the transition is not clear as observed in crystalline ferromagnets.

The interesting feature is that the temperature derivative of resistivity of the system at this particular temperature is directly proportional to the specific heat of the system. In case of ferromagnetic materials, as the temperature approaches T_c the charge carriers interact with electron spins undergoing the critical spin-fluctuations. So, in ferromagnets for the spherical Fermi surface, the resistivity can be written in the form of

$$\rho \propto \int_0^{2k_\mathrm{F}} q^3 \Gamma(q, T) \mathrm{d}q. \tag{1}$$

k_F is the Fermi wave vector and $\Gamma(q, \eta) = c/(k^2 + q^2)$, where $k = k_0 \eta^{1/2}$ and the reduced temperature $\eta = [(T - T_\mathrm{c})/T_\mathrm{c}]$. The thermal derivative of the resistivity as a function of reduced temperature will be

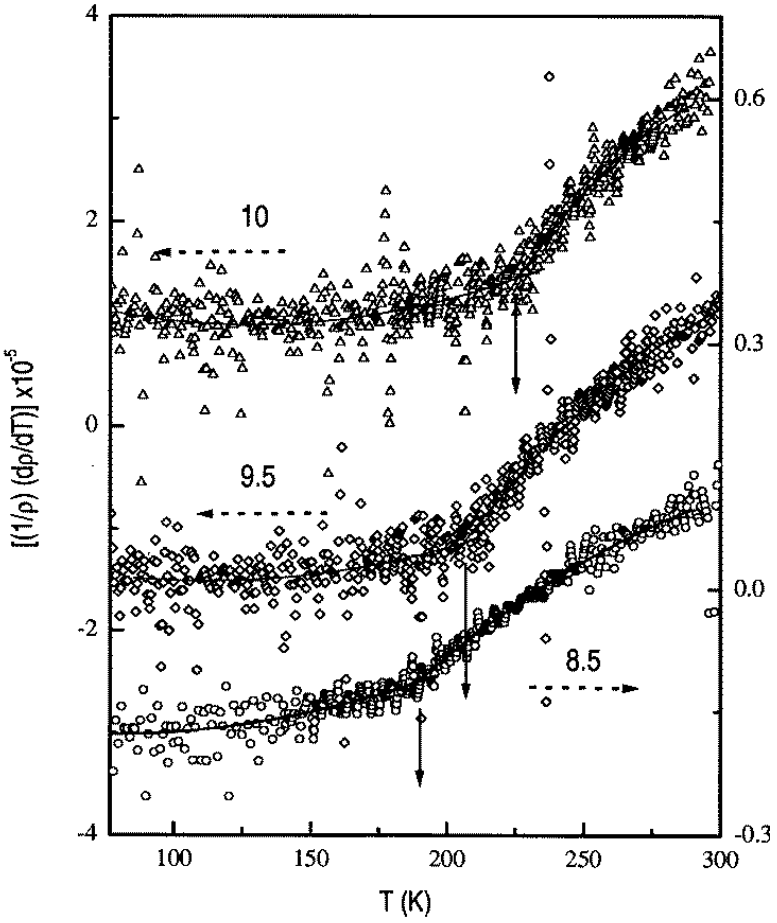

Figure 1. The thermal derivative of resistivity normalized with respect to resistivity value at T_c, $\rho(T_c)$ $[C(T) = (1/\rho(T_c))(\mathrm{d}\rho/\mathrm{d}T)]$, is plotted as a function of temperature for a-$Fe_{100-c}Zr_c$ with $c = 8.5$ (o), 9.5 ($\diamond$) and 10 (Δ). The line passing through the data is a best fit to eqs (4) and (5) for temperature below and above T_c with the exponents obtained from fitting results.

$$\left(\frac{\mathrm{d}\rho}{\mathrm{d}\eta}\right) = \left(\frac{\mathrm{d}\rho}{\mathrm{d}T}\right) \propto \eta^{-\alpha} \propto C_p, \tag{2}$$

where C_p is the specific heat, which is given by Fisher–Langer [17] relation

$$\left(\frac{\mathrm{d}\Gamma_{as}}{\mathrm{d}T}\right) \propto \frac{\mathrm{d}U}{\mathrm{d}T} = C_p. \tag{3}$$

In order to analyse the experimental data, the usual power law for the divergence of $\mathrm{d}\rho/\mathrm{d}T$ in the critical region is given by Geldert *et al* [24], which is another form of eq. (2),

$$C(T) = \left[\frac{1}{\rho(T_{\rm c})}\frac{{\rm d}\rho}{{\rm d}T}\right] = \frac{A^+}{\alpha}\left((-\eta)^{-\alpha} - 1\right) + B^+ \quad T < T_{\rm c}, \tag{4}$$

$$C'(T) = \left[\frac{1}{\rho(T_{\rm c})}\frac{{\rm d}\rho}{{\rm d}T}\right] = \frac{A^-}{\alpha'}\left(\eta^{-\alpha'} - 1\right) + B^- \quad T > T_{\rm c}, \tag{5}$$

where A and B are constants, α and α' are specific heat critical exponents below and above Curie temperature. The parameter values given in table 1, have been arrived at by adopting the following fitting procedure: (i) the Curie temperature, $T_{\rm c}$ (taken from thermal derivative to the resistivity at which ${\rm d}\rho/{\rm d}T$ begins to increase rapidly with increasing temperature) is kept fixed and the values of the remaining parameters are varied so, as to achieve the best fit to the experimental data in a given temperature range, (ii) the values determined by the above method in a given temperature interval for the parameters A, B and α and the assumed value of $T_{\rm c}$, are taken as the initial values for these parameters while attempting a theoretical fit to the experimental data in terms of eqs (4) and (5) by varying all the four parameters A, B, α and $T_{\rm c}$. The temperature range defined by $\eta_{\rm min} \leq \eta \leq \eta_{\rm max}$ is varied by varying $\eta_{\rm max}$ in order to find out the extent to which the best values of the fitting parameters and the quality of the fit, as determined by the mean-square error, which is defined as:

$$\chi^2 = \left(\frac{1}{N - N_{\rm para}}\right)\sum_1^N \frac{\left[C_{\rm theo} - C_{\rm exp}\right]^2}{\sigma^2}, \tag{6}$$

where N is number of data within the selected temperature region, $N_{\rm para}$ is the number of parameters, which are used in eqs (4) and (5) for fitting. As is usually the case, the values of the fitting parameters are found to depend on the range of η over which the data are fitted. The parameters listed in table 1 give the best fit to the data as inferred from the least value of the mean squared error in the specified temperature range and do not vary significantly (i.e. the variation of α falls well within the error limits given for these parameters in table 1) as the fit range is progressively narrowed down by decreasing $\eta_{\rm max}$ towards $\eta_{\rm min}$. Inclusion of experimental data outside the specified temperature range

Table 1. Values of different parameters used to fit the experimental data to eqs (4) and (5) of the text. Relevant parameter values deduced from the magnetic data are included for comparison.

c	A (10^{-4} K^{-1})	B (10^{-4} K^{-1})	$T_{\rm c}^M$ (K)	$T_{\rm c}^\rho$	$\eta = (T - T_{\rm c}^\rho)/T_{\rm c}^\rho$ fit range	α^ρ [α']	α^M
8.5	5.45(12)	3.32(4)	186.5(3)	187.4(8)	0.01 to 0.12 [−0.10 to −0.005]	−0.22(9) [−0.33(10)]	–
9.5	5.25(9)	3.56(4)	207.6(2)	208.2(6)	0.02 to 0.10 [−0.12 to −0.01]	−0.18(14) [−0.22(8)]	–
10	4.98(14)	3.78(4)	227.6(2)	228.5(9)	0.005 to 0.13 [−0.13 to −0.009]	−0.13(7) [−0.20(6)]	−0.18(6)$^+$

Abbreviations: $T_{\rm c}^M$ ($T_{\rm c}^\rho$) and α^M (α^ρ) are the $T_{\rm c}$ and α values deduced from the magnetization (resistivity) data. $^+$ values of α calculated from the relation $\alpha = 2(1-\beta)-\gamma$ using the values of β and γ determined in ref. [26,27].

(listed in table 1) results not only in the different parameter values, but also in a significant increase in their error limits and mean-square error of the best fit. All computed work has been performed using the nonlinear least-square fit computer program, which is based on Marquardt's maximum likelihood algorithm. The calculated values of T_c, and α are almost same and comparable with the values calculated from the Rushbrook's scaling relation with the values of $\beta = 0.366$ and $\gamma = 1.393$, as experimentally (from BM and ACS) found by Kaul *et al* and Perumal *et al* [11,27]. The constant values of A and B observed from the fitting procedures, have less values (almost one order of magnitude) when compared to the values obtained for Ni rich samples.

The interesting features that emerge from the above data analysis are: (i) the resistivity and magnetic measurements, performed on the glassy ribbons from the same alloys yield identical results so far as the values of T_c and α are concerned, (ii) the range of the critical region for the investigated alloys is much wider, compared to crystalline ferromagnets. From our observation that the critical exponent α can be determined up to a temperature value as high as $\eta_{\max} \approx 0.12$ for $T > T_c$ and $T < T_c$, (iii) the values of α determined in this study compare favourably not only with those previously deduced for other amorphous ferromagnets from the specific heat measurements, but also with the value predicted by a 3D Heisenberg model.

It has been reported earlier by Pureur *et al* on a-$Fe_{92}Zr_8$ that the second order transition occurs due to the presence of short-range interactions and specific heat critical exponent obtained from the thermal derivative of resistivity deviates from those values expected for ordered 3D Heisenberg ferromagnet. But in our opinion, it is too precipitate to draw any such conclusions, since the data are not much capable of pinpointing exactly whether the transition is due to quenched disorder or whether it is caused by (i) thermal gradients, (ii) external stresses, (iii) large magnetostriction in these alloys, if any and (iv) the method of deriving the $\mathrm{d}\rho/\mathrm{d}T$ values from the electrical resistivity as a function of temperature curves, which could result in some sort of averaging over a wide temperature region. Very accurate measurements are taken for temperatures very close to T_c with very slow temperature drift rates. The scaling critical behavior near the ferromagnetic–paramagnetic transition of amorphous Fe–Zr rich alloys in Fe rich side is studied by Kaul [14] based on bulk magnetization measurement results that the critical exponents obtained from these studies are in close agreement with the values calculated for ordered three-dimensional Heisenberg ferromagnet. These results can be explained in terms of the infinite ferromagnetic matrix along with finite-spin-clusters model, states that even at low temperatures ($T \ll T_c$), the ferromagnetic coupling between the spins (that establish the finite clusters) is much stronger than that between the spins of the ferromagnetic matrix. The exchange interaction between the spins in the ferromagnetic matrix weakens and the ferromagnetic couplings between the spins within the spin clusters are still stronger, when the temperature increases towards T_c. Subsequently, the number of spins in the ferromagnetic matrix will decrease as the temperature increase towards $T_c (T \to T_c)$. So, only a small fraction of spins participates in the ferromagnetic to paramagnetic transition at T_c. Considering that this model is based on a bond-frustrated 3D Heisenberg model [25], the critical exponents are expected to possses 3D NN Heisenberg values. The magnetic order–disorder phase transition at the Curie point T_c is characterized by a set of critical exponents α, β, δ and γ. Based on Rushbrook's relation $[\alpha = 2(1 - \beta) - \gamma]$, the specific heat exponent is extracted from other critical exponent values, which are obtained from the BM measurement. The extracted values are not only close to our observed results, but also in good agreement to

3D Heisenberg values. Now, it is clear that T_c marks the temperature at which a transition occurs from the paramagnetic state to a state with homogeneous long-range ferromagnetic ordering, the presently studied spin systems should behave exactly the same way as crystalline ferromagnets, so far as the behavior in the critical region and at temperatures just above T_c is concerned. In order to find the sharpness of the phase transition, we have carried out very low field ACS on present studied samples. The right hand axes of figure 2 depicts the real (χ') part of ACS curve under the external field of 26 mOe for a-$Fe_{90}Zr_{10}$, where the sharp transition from paramagnetic to ferromagnetic state is observed. We have made some rough estimate of difference between the peak temperature (T_p) observed from ACS and the T_c calculated from fitting procedure, to explain the width of the transition ($\Delta T = T_c - T_p$), which indicates the amount of inhomogeneity in the present system. The earlier fact along with the observation of specific heat critical exponent in the real

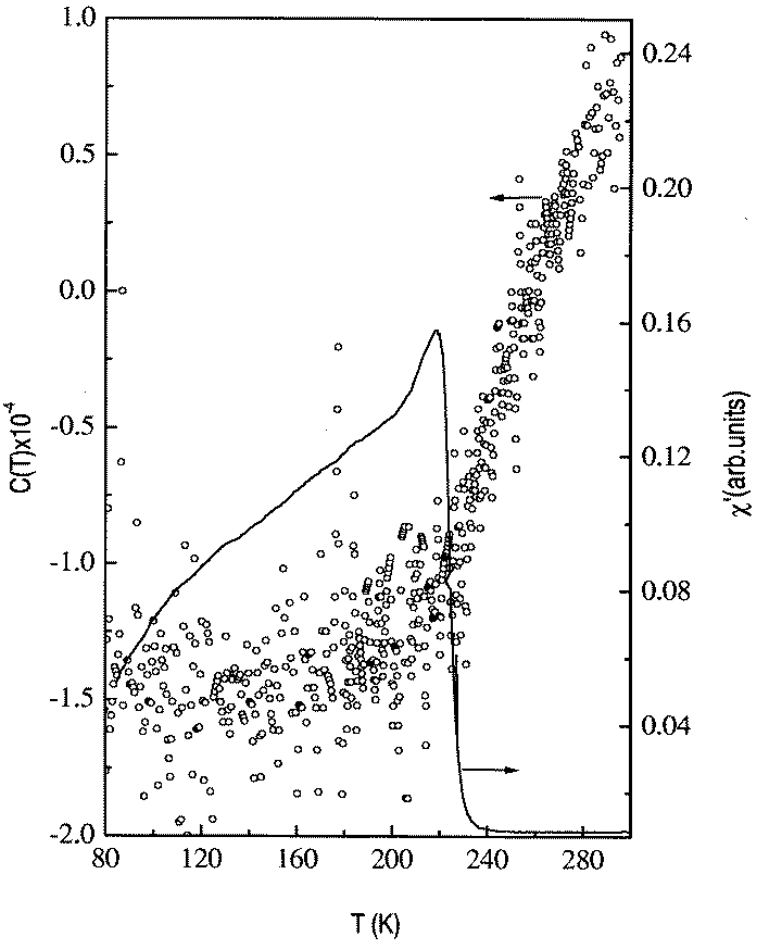

Figure 2. The $C(T)$ is plotted as a function of temperature (left hand axes) and the real part of ac susceptibility as a function of temperature is plotted (right hand axes) for amorphous $Fe_{90}Zr_{10}$ alloy.

critical region further strengthens our results that the quenched disordered has no way to alter the critical behavior of the present investigated systems.

4. Conclusions

The critical behavior of the amorphous $Fe_{100-c}Zr_c$ with $c = 8.5, 9.5$ and 10 is studied by measurement of thermal derivative of the resistivity $\mathrm{d}\rho/\mathrm{d}T$. The temperature at which the clear kink exists in $\mathrm{d}\rho/\mathrm{d}T$ is similar to the results of the magnetic measurements, where the existence of a very sharp Hopkinson maximum and the sharp onset of the spontaneous magnetization in the low-field susceptibility measurements. These two aspects suggest that the T_c is temperature at which transition occurs from paramagnetic to long-range ferromagnetic. The specific heat critical exponent is found to be negative for the present system and the value of α is in close agreement with the value predicted for ordered Heisenberg ferromagnet, which shows that the sharpness of the ferromagnetic–paramagnetic transition is not affected by the quenched disorder, which is also been supported from our ACS measurement, where the width of the transition, ΔT, is very small. At the same time, it remains unanswerable to us why the value of α is not changed till $\eta = \pm 0.12$, where the other critical exponent values, calculated from the ACS data analysis are varying considerably, beyond $\eta = \pm 0.05$ [26,27].

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