

## Evidence for a magnetic contribution to the electrical resistivity in amorphous $\text{Fe}_{80}\text{B}_{20-x}\text{C}_x$ alloys

S. N. Kaul,\* W. Kettler, and M. Rosenberg

*Institut für Experimentalphysik VI, Ruhr Universität Bochum, D-4630 Bochum, West Germany*

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The electrical resistivity  $\rho$  as a function of temperature has been measured for amorphous  $\text{Fe}_{80}\text{B}_{20-x}\text{C}_x$  ( $0 \leq x \leq 10$ ) alloys in the temperature range 4.2–300 K. A detailed quantitative analysis of the resistivity data for the first time establishes that besides a dominant structural contribution  $\rho_{\text{str}}$ , there exists a significant magnetic contribution  $\rho_{\text{mag}}$  to  $\rho$  in such glasses. The values of  $\rho_{\text{mag}}$  for the investigated glassy alloys reveal that the quenched disorder does not exert any noticeable influence on the *coherent* scattering of electrons from long-wavelength magnons and that the net contribution to  $\rho$  due to the *elastic* and *incoherent* components of electron-magnon scattering is negligibly small. Furthermore, the present data analysis permits a straightforward calculation of the Debye temperature  $\Theta_D$ , the structure factor  $S_0(2k_F)$ , and  $2k_F$  from  $\rho_{\text{str}}$ ; and the values of these quantities so determined, in turn, bring to the limelight various limitations of the extended Ziman theory so far as its application to glasses containing a strong scattering transition metal (e.g., Fe in the present case) is concerned.

### I. INTRODUCTION

Immense scientific activity<sup>1–4</sup> in the field of electron transport in amorphous metallic alloys witnessed during the past decade has been basically triggered by fundamental questions such as (i) how does quenched disorder influence the electronic structure and (ii) are the band structure and the glass-forming ability of amorphous alloys related in some simple way? Of all transport coefficients, the electrical resistivity  $\rho$  has received the most attention. Despite such intense efforts, the origin of the resistivity minimum phenomenon<sup>2</sup> and the  $T^2$  dependence of  $\rho$  at low temperatures, especially for ferromagnetic glasses, remain disputed. Since the latter controversial aspect of  $\rho(T)$  is the main concern of this paper, it deserves a more elaborate description.

The temperature dependence of resistivity,  $\rho(T)$ , characteristic of a wide variety of glassy metallic alloys (including both nonmagnetic and magnetic glasses) can be summarized as follows.  $\rho(T)$  goes through a Kondo-like minimum (not observed in some nonmagnetic glasses) at low temperatures (generally below  $\sim 20$  K), varies as  $T^2$  in the intermediate-temperature region (normally  $40 \lesssim T \lesssim 100$  K) and exhibits a linear temperature dependence at high temperatures (above  $\sim 150$  K). It has been a common practice<sup>5–7</sup> to completely disregard the magnetic state of the glassy alloy under consideration and to explain the intermediate- and high-temperature resistivity data in terms of the diffraction model,<sup>8</sup> which takes into account not only the scattering of the conduction electrons by the ion cores that carry a muffin-tin potential but also the change in the shape of the structure factor,  $S(k)$ , as  $T$  is varied. The reasons for this appear to be twofold. First, in the intermediate- and high-temperature regions  $\rho$  exhibits the same temperature dependence in both magnetic and nonmagnetic glasses. Second, the diffraction

model yields both the quadratic and the linear temperature dependence of  $\rho$  at low ( $T \ll \Theta_D$ , Debye temperature) and high ( $T \gtrsim \Theta_D$ ) temperatures, respectively. However, overwhelming experimental evidence exists to demonstrate that the diffraction model does not form an adequate description of the  $T^2$  term in  $\rho(T)$  for the amorphous ferromagnetic alloys, in particular. For instance, the existence of well-defined long-wavelength spin-wave excitations which follow a normal ferromagnetic dispersion relation,  $\hbar\omega_k = Dk^2$ , has now been well established in ferromagnetic glasses by inelastic neutron scattering, magnetization, and Mössbauer measurements,<sup>9</sup> and a sharp anomaly in the temperature derivative of resistivity at the Curie temperature reminiscent of the critical resistivity behavior normally found in crystalline ferromagnets has been observed<sup>10</sup> in such glasses. These observations assert that in addition to a contribution due to the scattering of conduction electrons from the structural disorder, electron-magnon scattering (which at low temperatures gives rise to a quadratic temperature dependence for crystalline ferromagnetic  $3d$  transition metals) gives a significant contribution to  $\rho$  in magnetic glasses and hence the customary approach of completely neglecting the magnetic contribution,  $\rho_{\text{mag}}(T)$ , while analyzing the resistivity data on ferromagnetic glasses is not correct. Theoretical investigations<sup>11,12</sup> that pursue this line of thinking and use spin-wave approximation to calculate  $\rho_{\text{mag}}(T)$  for amorphous ferromagnets reveal that (i)  $\rho_{\text{mag}}(T)$  comprises two *positive* contributions; one varying as  $T^2$ , as in crystalline ferromagnets, and the other as  $T^{3/2}$ , and (ii) the contribution to  $\rho_{\text{mag}}(T)$  from the  $T^{3/2}$  term (which appears only in the amorphous case) is at least 2 orders of magnitude greater than the one arising from the  $T^2$  term. These theories, therefore, predict a  $T^{3/2}$  power law for  $\rho(T)$  at low temperatures in amorphous ferromagnets as contrasted with the  $T^2$  dependence found in these materials. This

discrepancy between theory and experiment has been taken as evidence for<sup>11</sup> or against<sup>12</sup> a magnetic origin of the observed  $T^2$  term. Despite the fact that a clear distinction between the  $T^2$  and  $T^{3/2}$  power laws in a narrow temperature range (typically from 40 to 100 K) poses an extremely difficult problem in practice, experimental evidence for the fractional exponent (1.5), and hence for a contribution of electron-magnon scattering to  $\rho$ , has recently been claimed for some amorphous Fe- and Fe-Ni-based ferromagnetic ribbons<sup>13-16</sup> and sputtered films.<sup>17</sup> But, as pointed out by Mogro-Campero *et al.*,<sup>16</sup> a power-law fit with exponent 1.5 found in some temperature region ( $T_{\min} \leq T \leq T_{\max}$ ) cannot be considered as sufficient evidence for a magnetic contribution to  $\rho$  because the temperature interval  $T_{\min} \leq T \leq T_{\max}$  may well be the transition region separating the temperature regimes, where the asymptotic  $T^2$  ( $T \leq T_{\min}$ ) and  $T$  ( $T \geq T_{\max}$ ) behaviors are observed, and this transition region could be well described by a  $T^{3/2}$  power law. So the possibility of explaining the experimental findings in terms of electron-ion potential scattering alone cannot be completely ruled out.

With a view to resolve the above-mentioned controversy surrounding the origin of the  $T^2$  power law of  $\rho(T)$  in the intermediate-temperature range for ferromagnetic glasses, we undertook detailed electrical resistivity studies on amorphous  $\text{Fe}_{80}\text{B}_{20-x}\text{C}_x$  alloy series. The choice of this alloy system was motivated by the simple logic that one expects to detect the magnetic contribution to  $\rho$  with greater ease in alloys with a high transition-metal content than in those with a low transition-metal content. Exhaustive data analysis of the experimental results not only enables us to establish for the first time beyond doubt the existence of a magnetic contribution,  $\rho_{\text{mag}}$ , to  $\rho$  but also to make a reliable estimate of  $\rho_{\text{mag}}$  in these alloys.

## II. THEORETICAL BACKGROUND

Experimental findings that the electron transport properties of amorphous metals are usually similar<sup>18</sup> to those of the corresponding liquid metals have led various workers to use a theoretical formalism, originally proposed for simple liquid metals by Ziman<sup>19</sup> and subsequently extended by Evans *et al.*<sup>20</sup> to include liquid transition metals, to explain a number of peculiarities observed in the transport properties of metallic glasses. This theory takes into account the scattering of conduction electrons from the potential of the disordered lattice of a transition-metal system and gives the following expression<sup>20</sup> for resistivity:

$$\rho \approx \frac{30\pi^3 \hbar^3}{me^2 k_F^2 E_F \Omega} \sin^2[\eta_2(E_F)] S_T(2k_F), \quad (1)$$

where  $k_F$  and  $E_F$  are the Fermi wave vector and energy, respectively,  $\Omega$  is the atomic volume,  $\eta_2(E_F)$  is the  $d$ -partial-wave phase shift describing the scattering of the conduction electrons by the ion cores which carry a muffin-tin potential centered on each ion position,  $S_T(2k_F)$  is the temperature-dependent structure factor, and  $\hbar$ ,  $m$ , and  $e$  have their usual meaning. In this expression,  $S_T(2k_F)$  determines the temperature dependence of  $\rho$ . Several calculations<sup>8,21,22</sup> of  $S_T(k)$  within the framework of a model, now known as the diffraction model, are

available but the one<sup>8</sup> used in this work yields

$$S_T(k) \cong 1 + [S_E(k) - 1] e^{-2W_k(T)}, \quad (2)$$

where  $S_E(k)$  is the equilibrium structure factor and  $e^{-2W_k(T)}$  is the Debye-Waller factor with  $W_k(T)$ , in the Debye approximation, given by<sup>23</sup>

$$W_k(T) = W_k(0) + 4W_k(0) \left[ \frac{T}{\Theta_D} \right]^2 \int_0^{\Theta_D/T} \frac{z dz}{e^z - 1}, \quad (3a)$$

where

$$W_k(0) = 3\hbar^2 k^2 / 8Mk_B \Theta_D, \quad (3b)$$

$M$  is the atomic mass,  $k$  is the wave vector, and  $k_B$  is the Boltzmann constant. With the aid of Eqs. (1) and (2), the resistivity as a function of temperature can be expressed as<sup>8</sup>

$$\begin{aligned} \rho_{\text{str}}(T) = & \frac{30\pi^3 \hbar^3}{me^2 k_F^2 E_F \Omega} \sin^2[\eta_2(E_F)] \\ & \times \{ 1 + [S_0(2k_F) - 1] e^{-2[W_{2k_F}(T) - W_{2k_F}(0)]} \}. \end{aligned} \quad (4)$$

Note that  $\rho_{\text{str}}$  is the structural contribution to the total resistivity  $\rho$  and the parameters  $W_{2k_F}(T)$  and  $W_{2k_F}(0)$  appearing in Eq. (4) denote the values of  $W_k(T)$  and  $W_k(0)$  at  $k = 2k_F$ , i.e.,

$$W(0) = W_{2k_F}(0) = \frac{3\hbar^2 k_F^2}{2Mk_B \Theta_D}, \quad (5)$$

replaces  $W_k(0)$  in Eqs. (3a) and (3b). Dropping henceforth the subscript  $2k_F$  in  $W_{2k_F}(T)$  and  $W_{2k_F}(0)$  and str in  $\rho_{\text{str}}$  for the sake of simplicity, the temperature coefficient of the resistivity (TCR) can be calculated from Eq. (4) with the result

$$\begin{aligned} \alpha = \frac{1}{\rho} \frac{\partial \rho}{\partial T} = & 2 \left[ \frac{1 - S_T(2k_F)}{S_T(2k_F)} \right] \frac{\partial W(T)}{\partial T} \\ = & 8 \left[ \frac{1 - S_T(2k_F)}{S_T(2k_F)} \right] \frac{W(0)}{T} \\ & \times \left[ 2 \left[ \frac{T}{\Theta_D} \right]^2 \int_0^{\Theta_D/T} \frac{z}{e^z - 1} dz - \frac{1}{e^{\Theta_D/T} - 1} \right], \end{aligned} \quad (6)$$

where use has been made of Eq. (3a) to calculate  $\partial W(T)/\partial T$ . Equation (6) demonstrates that  $\partial W(T)/\partial T > 0$  at all temperatures and as such  $\alpha$  is *negative* if  $S_T(2k_F) > 1$  and *positive* if  $S_T(2k_F) < 1$ . Alternatively, a *negative*  $\alpha$  is expected only when  $2k_F$  lies in the vicinity of  $k_p$ , the  $k$  value corresponding to the first peak of  $S(k)$ ; otherwise a *positive*  $\alpha$  is expected.

In the low- and high-temperature limits, Eq. (3a)

reduces to<sup>8</sup>

$$W(T) \simeq \begin{cases} W(0) + 4W(0) \frac{\pi^2}{6} \left[ \frac{T}{\Theta_D} \right]^2, & T \ll \Theta_D \end{cases} \quad (7a)$$

$$\begin{cases} 4W(0) \left[ \frac{T}{\Theta_D} \right], & T \gtrsim \Theta_D. \end{cases} \quad (7b)$$

An estimate of  $W(0)$ , using the values  $\Theta_D \cong 300$  K,  $M \cong 8 \times 10^{-23}$  g and  $E_F \cong 10$  eV, typical of metallic glasses, in the relation  $W(0) = 3\hbar^2 k_F^2 / 2Mk_B \Theta_D = (3E_F / k_B \Theta_D) (m/M)$ , shows that  $W(0) \cong 0.013$ . Bearing in mind that  $W(0) \ll 1$ , Eqs. (4) and (7) can be combined and the exponential function in the modified version of Eq. (4) expanded in power series to yield the asymptotic temperature dependence of  $\rho$  as

$$\rho(T) \simeq \begin{cases} C \left\{ S_0(2k_F) + \left[ \frac{2\pi}{\Theta_D} \right]^2 \frac{W(0)}{3} [1 - S_0(2k_F)] T^2 \right\}, & T \ll \Theta_D \end{cases} \quad (8a)$$

$$\begin{cases} C \left\{ S_0(2k_F) - 2W(0)[1 - S_0(2k_F)] + \frac{8W(0)}{\Theta_D} [1 - S_0(2k_F)] T \right\}, & T \gtrsim \Theta_D \end{cases} \quad (8b)$$

with

$$C = \frac{30\pi^3 \hbar^3}{me^2 k_F^2 E_F \Omega} \sin^2[\eta_2(E_F)].$$

In order to facilitate a direct comparison between theory and experiment, a different form of Eq. (4) has been used in the present work, i.e.,

$$\begin{aligned} r(T) &= \frac{\rho(T)}{\rho(T_0)} = a_1 + a_2 e^{-2[W(T) - W(0)]} \\ &= a_1 + a_2 \exp \left[ -8W(0) \left[ \frac{T}{\Theta_D} \right]^2 \int_0^{\Theta_D/T} \frac{z}{e^z - 1} dz \right] \\ &\equiv f_1(T), \end{aligned} \quad (9)$$

where

$$a_1 = \{ 1 + [S_0(2k_F) - 1] e^{-2[W(T_0) - W(0)]} \}^{-1}, \quad (10a)$$

$$a_2 = a_1 [S_0(2k_F) - 1], \quad (10b)$$

or

$$S_0(2k_F) = 1 + (a_2/a_1),$$

and  $T_0 = 273.15$  K is the ice temperature.

In view of the experimental evidence already presented in the Introduction, electron-magnon scattering is expected to significantly contribute to  $\rho$  in ferromagnetic metallic glasses at low temperatures. Within the framework of the spin-disorder model, Richter *et al.*<sup>12</sup> have calculated  $\rho_{\text{mag}}(T)$  at low temperatures for an amorphous Heisenberg ferromagnet in the spin-wave approximation with the result

$$\frac{\rho_{\text{mag}}(T)}{\rho_{\text{mag}}(0)} = 1 + \frac{\Omega_c}{(2\pi)^2 S} \Gamma\left(\frac{3}{2}\right) \xi\left(\frac{3}{2}\right) \left[ \frac{k_B T}{D} \right]^{3/2} + \frac{1}{J_s(2k_F)} \left[ \frac{\pi^2}{3S} \left[ \frac{k_B T}{D} \right]^2 - \int_0^{2k_F} dk \frac{k^3 S(k) (\hbar\omega_k / k_B T)}{(e^{\hbar\omega_k / k_B T} - 1)(1 - e^{-\hbar\omega_k / k_B T})} \right] \quad (11a)$$

and

$$\rho_{\text{mag}}(0) = \frac{2\Omega_c}{3\hbar^3} \left[ \left[ \frac{\Omega}{N_e e} \right] m S J_{sd} \right]^2 J_s(2k_F), \quad (11b)$$

where  $\Omega_c$  and  $\Omega$  are the atomic and sample volumes, respectively,  $S$  is the spin of the local magnetic moment,  $N_e$  denotes the number of electrons,  $D$  is the spin-wave stiffness constant given by the magnon dispersion relation  $\hbar\omega_k = Dk^2$ ,  $\Gamma$  and  $\xi$  are the gamma and Riemann zeta functions, respectively,  $J_{sd}$  is the  $s$ - $d$  exchange coup-

ling constant and  $J_s(2k_F) = \int_0^{2k_F} k^3 S(k) dk$ . In Eq. (11a), the second term is an outcome of a partial cancellation of two competing  $T^{3/2}$  terms; one arises from *incoherent* (momentum nonconserving) electron-magnon scattering and *increases* with increasing temperature, and the other originates from the *elastic scattering* of conduction electrons from the randomly oriented temperature-dependent local moments and *decreases* as the temperature is increased, whereas the third term is a *coherent* spin-wave term. The fourth (integral) term, which arises because of disorder, forms only a minute correction to the  $T^2$  term

indicating thereby that the coherent scattering of electrons by long-wavelength magnons is not significantly altered by the quenched disorder. Another important feature pertaining to Eq. (11) is that  $\rho_{\text{mag}}(0)$  and the  $T^{3/2}$  term both vanish in the case of crystalline ferromagnets whereas they possess finite values for amorphous ferromagnets. With a view to find out the relative importance of the  $T^{3/2}$  and  $T^2$  terms, Richter *et al.*<sup>12</sup> have estimated the order of magnitude of these terms assuming  $a=3 \text{ \AA}$ ,  $\Omega_c=a^3$ ,  $S=1$ ,  $D=185 \text{ meV \AA}^2$  and  $k_F=1.36 \text{ \AA}^{-1}$  as

$$\frac{\rho_{\text{mag}}(T)}{\rho_{\text{mag}}(0)} = 1 + 1.6 \times 10^{-5} T^{3/2} + 2.4 \times 10^{-7} T^2, \quad (12)$$

where the temperature is measured in K. Equation (12) indicates that  $\rho_{\text{mag}}(T)$  in amorphous ferromagnets should vary as  $T^{3/2}$  at low temperatures as contrasted with the  $T^2$  variation of  $\rho_{\text{mag}}(T)$  in the crystalline case.

### III. EXPERIMENTAL DETAILS

Amorphous  $\text{Fe}_{80}\text{B}_{20-x}\text{C}_x$  alloy ribbons ( $\sim 1 \text{ mm}$  wide and  $30 \text{ }\mu\text{m}$  thick) were prepared by rapid quenching from the melt onto a rotating copper drum and procured from the General Electric Company, New York. While copper wires of  $0.13 \text{ mm}$  diameter attached to the ends of a  $20\text{-mm}$ -long ribbon by means of silver paste formed the current contacts, the voltage contacts ( $\sim 5 \text{ mm}$  apart) were made by bonding very thin aluminum wires of  $25 \text{ }\mu\text{m}$  diameter onto the sample using the ultrasonic method. The sample was then pasted to  $1 \text{ mm}$  thick glass or sapphire wafer with Apiezon-N grease, which serves two purposes. First, it ensures a good thermal contact of the sample with the resistance thermometers, which find themselves placed underneath the wafer. Second, it allows for the changes in the sample dimensions during thermal cycling and thereby avoids stress-induced effects, which could otherwise significantly affect the results in stress-sensitive materials like metallic glasses.

Electrical resistivity was measured at  $\sim 0.5 \text{ K}$  intervals ( $\leq 0.1 \text{ K}$  intervals for  $T \leq 50 \text{ K}$ ) in the temperature range  $4.2$  to  $300 \text{ K}$  employing a four-probe dc method. The experimental setup used for these measurements could resolve a few ppm change in resistivity with ease. The sample temperature below and above  $50 \text{ K}$  was monitored by calibrated germanium and platinum-resistance thermometers, respectively.

Large error in the measurement of the ribbon thickness and width, arising mainly due to an erratic variation of these quantities over the entire length of the ribbon, results in a large inaccuracy ( $\sim 10\%$ ) in the absolute values of  $\rho$ . In order to get rid of such uncertainties, the values of  $\rho$  at different temperatures have been normalized to the resistivity value at  $T_0=273.15 \text{ K}$ , which is the ice point.

### IV. RESULTS AND ANALYSIS

The temperature dependence of the normalized resistivity ratio  $r(T)=\rho(T)/\rho(T_0)$  in the temperature range  $4.2$  to  $300 \text{ K}$  for amorphous  $\text{Fe}_{80}\text{B}_{20-x}\text{C}_x$  alloys with  $x=0, 2, 4$ , and  $10$  is shown in Fig. 1. It is noticed from this figure that the total variation of  $r(T)$  within the in-

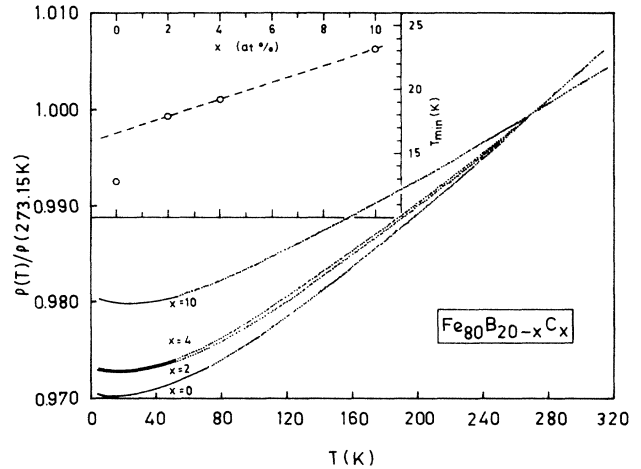


FIG. 1. Temperature dependence of the normalized resistivity ratio for amorphous  $\text{Fe}_{80}\text{B}_{20-x}\text{C}_x$  alloys. The inset displays the concentration dependence of  $T_{\text{min}}$ .

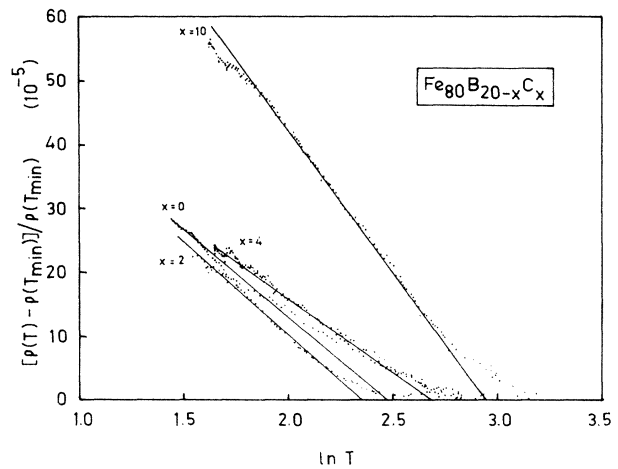


FIG. 2.  $[\rho(T) - \rho(T_{\text{min}})]/\rho(T_{\text{min}})$  vs  $\ln T$  for  $a\text{-Fe}_{80}\text{B}_{20-x}\text{C}_x$  alloys. The continuous straight lines through the data points serve as a guide to the eye.

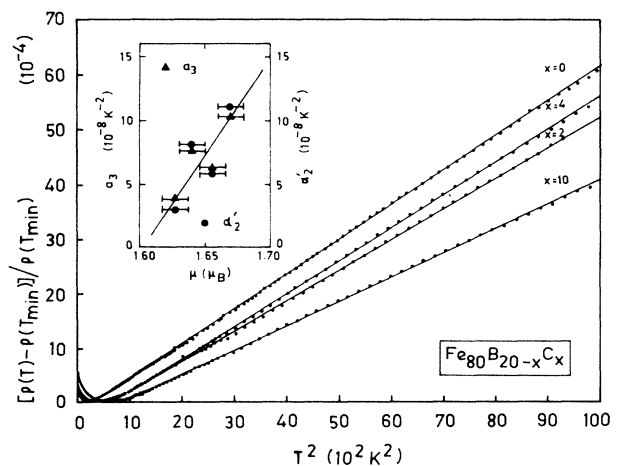


FIG. 3.  $[\rho(T) - \rho(T_{\text{min}})]/\rho(T_{\text{min}})$  vs  $T^2$  for  $a\text{-Fe}_{80}\text{B}_{20-x}\text{C}_x$  alloys. Straight lines through the data points are intended to serve as a guide to the eye. In the inset, the coefficients  $a_3$  and  $a_2$  are shown plotted against magnetic moment  $\mu$ . The least-squares-fit straight line through the data points is also shown.

TABLE I. Details of the low-temperature least-squares fits, based on Eqs. (13) and (14) of the text, for amorphous  $\text{Fe}_{80}\text{B}_{20-x}\text{C}_x$  alloys. Numbers in the parentheses denote the estimated uncertainty in the significant figure.

$x$ (at. %)	$\alpha_0$	$\alpha_2$ ( $10^{-7} \text{ K}^{-2}$ )	Fit range $\Delta T$ (K)	$\chi^2$ ( $10^{-10}$ )	$\beta_0$	$\beta$ ( $10^{-4} \ln \text{K}$ )	Fit range $\Delta T$ (K)	$\chi^2$ ( $10^{-11}$ )
0	0.970	6.13(1)	50–100	3.70	0.971	–3.28(2)	4.0–8.0	1.13
2	0.972	5.52(1)	50–100	2.67	0.973	–1.81(2)	5.0–12.0	1.15
4	0.973	5.85(2)	50–100	7.11	0.973	–2.21(2)	4.5–12.0	2.83
10	0.979	4.41(2)	50–100	5.23	0.981	–4.31(2)	6.0–15.5	2.23

vestigated temperature range *decreases* with increasing  $x$  and does not, in any case, exceed 4% of the value of  $r$  at 300 K. At low temperatures (10–25 K),  $\rho$  as a function of temperature goes through a well-defined *minimum* at a temperature  $T_{\min}$ , which increases roughly linearly with  $x$  (inset in Fig. 1).

Figures 2 and 3 serve to demonstrate that resistivity exhibits a variation with temperature which can be well described by the relations

$$r(T) = \beta_0 + \beta \ln T, \quad T < T_{\min} \quad (13)$$

and

$$r(T) = \alpha_0 + \alpha_2 T^2, \quad T > T_{\min} \quad (14)$$

in the temperature ranges  $4 \lesssim T \lesssim 12$  K and  $50 \leq T \leq 100$  K, respectively. Least-squares fits to the resistivity data based on Eqs. (13) and (14) in the specified temperature ranges give the values for the coefficients  $\beta_0$ ,  $\beta$ ,  $\alpha_0$ , and  $\alpha_2$  listed in Table I.

In order to bring out the functional dependence of  $\rho$  on  $T$  for  $T > 100$  K, the temperature derivative of  $r(T)$  has been plotted as a function of temperature in Fig. 4. A cursory glance at this figure indicates that in the temperature interval  $120 \leq T \leq 300$  K  $r(T)$  obeys the simple relation:

$$r(T) = \alpha'_0 + \alpha'_1 T + \alpha'_2 T^2. \quad (15)$$

However, a close scrutiny of the  $dr(T)/dT [=r'(T)]$  curves reveals that they present a slight but finite curvature up to a temperature as high as  $\approx 200$  K and beyond this temperature  $r'(T) \propto T$ . This inference is corroborated by the finding that the parameters  $\alpha'_0$ ,  $\alpha'_1$ , and  $\alpha'_2$  in the least-squares-fit computer program are found to vary when the resistivity data are fitted to Eq. (15) over a temperature range, defined by  $T^* \leq T \leq 300$  K, which is progressively narrowed down by increasing  $T^*$  from an initial value of 120 K to a final value of 200 K; the values of the coefficients remain essentially unaltered when the temperature range of the fit is further narrowed down by increasing  $T^*$  beyond 200 K. The values of the coefficients  $\alpha'_0$ ,  $\alpha'_1$ , and  $\alpha'_2$  for the two extreme fitting ranges, i.e.,  $120 \leq T \leq 300$  K and  $200 \leq T \leq 300$  K, are given in Table II.

## V. DISCUSSION

To begin with, we completely ignore the magnetic contribution to  $\rho$  and attempt a *quantitative* explanation of the results in terms of the diffraction model. According to this model, the fractional change of resistivity between 0 and 300 K,  $\Delta\rho/\rho$ , the normalized resistivity at 300 K,  $r(300 \text{ K})$ , and the temperature coefficient of resistivity,  $\alpha'_1$ , are given by the expressions

$$\frac{\Delta\rho}{\rho} = \frac{\rho(300 \text{ K}) - \rho(0 \text{ K})}{\rho(0 \text{ K})} = \frac{2[1 - S_0(2k_F)]}{S_0(2k_F)} \mathcal{W}(0) \left[ \frac{4 \times 300}{\Theta_D} - 1 \right], \quad (16)$$

$$r(300 \text{ K}) = \frac{S_0(2k_F) + 2\mathcal{W}(0)[1 - S_0(2k_F)][(4 \times 300/\Theta_D) - 1]}{S_0(2k_F) + 2\mathcal{W}(0)[1 - S_0(2k_F)][(4T_0/\Theta_D) - 1]}, \quad (17)$$

TABLE II. Details of the high-temperature least-squares fits, based on Eq. (15) of the text, for amorphous  $\text{Fe}_{80}\text{B}_{20-x}\text{C}_x$  alloys. Numbers in the parentheses denote the estimated uncertainty in the least significant figure.

$x$ (at. %)	$\alpha'_0$	$\alpha'_1$ ( $10^{-4} \text{ K}^{-1}$ )	$\alpha'_2$ ( $10^{-8} \text{ K}^{-2}$ )	Fit range $\Delta T$ (K)	$\chi^2$ ( $10^{-10}$ )	$\Theta_D$ (K)
0	0.965	1.029(3)	9.71(7)	120–300	5.04	328(4)
	0.964	1.106(12)	8.15(25)	200–300	3.59	342(4)
2	0.968	0.909(4)	10.02(9)	120–300	11.50	331(5)
	0.968	0.854(11)	11.15(21)	200–300	4.35	319(5)
4	0.968	0.897(5)	9.57(11)	120–300	14.93	301(7)
	0.971	1.083(17)	5.83(33)	200–300	9.71	338(7)
10	0.976	0.725(3)	5.55(8)	120–300	8.42	310(5)
	0.974	0.858(11)	3.00(21)	200–300	4.54	349(5)

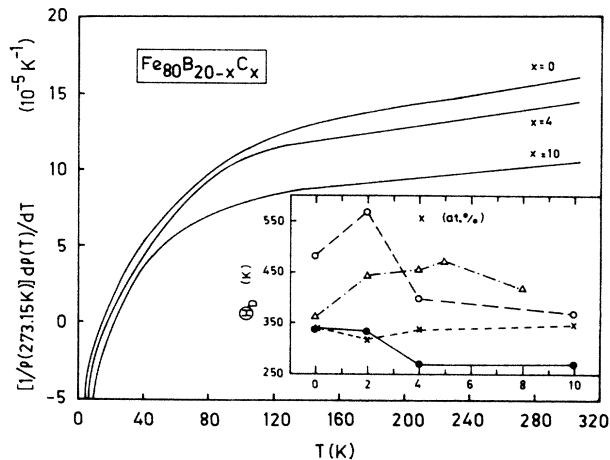


FIG. 4. The temperature derivative of the normalized resistivity ratio as a function of temperature for  $a$ - $\text{Fe}_{80}\text{B}_{20-x}\text{C}_x$  alloys. The inset depicts the concentration dependence of the Debye temperature;  $\Theta_D$  values obtained from  $f_1(T)$  fits, open circles;  $\Theta_D$  values extracted from  $f_2(T)$  fits, closed circles; values deduced from the asymptotic fits [Eq. (20) of the text], crosses; values determined from the specific-heat measurements and taken from Ref. 36, open triangles.

and

$$\alpha'_1 = \frac{8W(0)[1 - S_0(2k_F)]/\Theta_D}{S_0(2k_F) + 2W(0)[1 - S_0(2k_F)][(4T_0/\Theta_D) - 1]}, \quad (18)$$

which derive their origin from Eq. (8). These relations demonstrate that a direct comparison between theory and experiment cannot be made without prior knowledge of  $W(0)$ ,  $\Theta_D$ , and  $S_0(2k_F)$ . In order to arrive at the numerical estimates of these quantities,  $r(T)$  data have been fitted to Eq. (9), which gives the complete functional form of  $\rho(T)$ , over the temperature range  $50 \leq T \leq 300$  K by using a nonlinear least-squares-fit computer program, which treats  $a_1$ ,  $a_2$ ,  $W(0)$ , and  $\Theta_D$  as free parameters and is based on the Marquardt's maximum-likelihood algorithm.<sup>24</sup> The parameter values so obtained and the corresponding values of  $k_F$  and  $S_0(2k_F)$  deduced from them using Eqs. (5) and (10b), respectively, are presented in Table III. Theoretical values of  $\Delta\rho/\rho$ ,  $r(300$  K), and  $\alpha'_1$  are then deduced by using the values of  $W(0)$ ,  $\Theta_D$ , and  $S_0(2k_F)$  from Table III in Eqs. (16)–(18) and compared with the observed values in Table IV. From Tables III and IV one notices that (i) a close agreement between the calculated and observed values of  $\Delta\rho/\rho$  and  $r(300$  K) can be achieved but not without paying the heavy penalty in terms of the *unphysically large*  $k_F$  values, and (ii) even after paying such a high price, a large discrepancy between the experimental and theoretical values of  $\alpha'_1$  remains. While the former observation provides an experimental confirmation for the previous theoretical result<sup>25</sup> that for metallic glasses containing a strong-scattering transition metal (Fe or Co), the extended Ziman-Faber theory (on which the diffraction model is based) repro-

TABLE III. Parameter values for amorphous  $\text{Fe}_{80}\text{B}_{20-x}\text{C}_x$  alloys obtained by fitting  $r(T)$  data to the expressions  $r(T) = f_1(T)$  [Eq. (9)] and  $r(T) = f_1(T) + a_3T^2 \equiv f_2(T)$  [Eq. (22)] in the temperature interval  $50 \leq T \leq 300$  K. While the values within the square brackets pertain to the least-squares fits based on the relation  $r(T) = f_2(T)$ , the numbers in the parentheses denote estimated uncertainty in the least significant figure.

$x$ (at. %)	$a_1$	$a_2$	$W(0)$	$\Theta_D$ (K)	$S_0(2k_F)$	$k_F$ ( $\text{\AA}^{-1}$ )	$\chi^2$ ( $10^{-9}$ )	$a_3$ ( $10^{-8} \text{ K}^{-2}$ )	$d$ ( $\text{g}/\text{cm}^3$ )	$\mu/\text{alloy}$ atom at 77 K ( $\mu_B$ )
0	1.270(4) [1.269(9)]	-0.300(4) [-0.299(9)]	0.0363(7) [0.0183(6)]	486(3) [341(2)]	0.764(14) [0.764(32)]	3.37(4) [2.00(3)]	9.311 [0.452]		7.39 <sup>a</sup>	1.640(10) <sup>b</sup>
2	1.116(2) [1.080(3)]	-0.144(2) [-0.108(3)]	0.093(3) [0.044(2)]	571(7) [334(8)]	0.871(17) [0.900(28)]	5.85(10) [3.08(8)]	21.979 [3.365]	[7.62(11)]	7.41 <sup>a</sup>	1.670(10) <sup>a</sup>
4	1.534(5) [1.533(22)]	-0.562(5) [-0.561(22)]	0.0133(2) [0.0068(3)]	400(4) [271(5)]	0.634(9) [0.634(41)]	1.85(16) [1.09(3)]	8.926 [1.734]	[10.36(24)]	7.44 <sup>a</sup>	1.656(10) <sup>a</sup>
10	1.699(10) [1.699(16)]	-0.720(10) [-0.720(16)]	0.0069(1) [0.0040(2)]	370(3) [270(5)]	0.576(15) [0.576(24)]	1.28(1) [0.84(2)]	3.705 [1.172]	[6.36(20)]	7.51 <sup>a</sup>	1.627(10) <sup>a</sup>

<sup>a</sup>Data taken from Ref. 34.

<sup>b</sup>Reference 35; value at 4.2 K.



ferent temperatures, employing the values of  $\alpha'_1$ ,  $\alpha'_2$  and  $\alpha_2$  given in Tables I and II, reveals that  $\rho_{\text{str}}$  dominates (greater by at least an order of magnitude) over  $\rho_{\text{mag}}$  in the entire temperature range from 50 to 300 K and hence the magnetic contribution to  $\rho$  can be easily missed unless great care is exercised while analyzing the results. With the terms  $\alpha_2 T^2$ ,  $\alpha'_1 T$ , and  $\alpha'_2 T^2$  of the low- and high-temperature fits, Eqs. (14) and (15), identified as  $[\rho_{\text{str}}(T) + \rho_{\text{mag}}(T)]$ ,  $\rho_{\text{str}}(T)$  and  $\rho_{\text{mag}}(T)$ , respectively, Eqs. (6)–(8), (14) and (15) can be combined to give  $\Theta_D$  as

$$\Theta_D \simeq \frac{\pi^2}{6} \frac{\alpha'_1}{\alpha_2 - \alpha'_2}. \quad (20)$$

Values of  $\Theta_D$  deduced from this relation using the values of  $\alpha_2$ ,  $\alpha'_1$ , and  $\alpha'_2$  obtained from the low- and high-temperature fits (Tables I and II) are listed in Table II.

Apart from the fits in the asymptotic temperature regions [Eqs. (14) and (15)], two types of fits to the experimental data in the temperature range 50 to 300 K, both based on Eq. (19) with  $\rho_{\text{str}}(T)$  and  $\rho_{\text{mag}}(T)$  given by Eqs. (4) and (11), respectively, have been attempted by using a nonlinear least-squares-fit computer program of the kind mentioned earlier. Before going into the details of these fits, we make a rough estimate of  $\rho_{\text{mag}}(0)$  with the intention of finding out how small or big this quantity is for

amorphous ferromagnets. With the aid of the well-known free-electron expressions, namely,  $E_F = \hbar^2 k_F^2 / 2m$  and  $k_F^3 = 3\pi^2 N_e / \Omega$ , Eq. (11b) can be rewritten in the form

$$\rho_{\text{mag}}(0) = \frac{3\pi\hbar\Omega_c S^2}{16e^2 k_F^2} \left[ \frac{J_{sd}}{E_F} \right]^2 J_s(2k_F). \quad (21)$$

Substitution in this equation of the typical values  $\Omega_c = a^3 = (3)^3 \text{ \AA}^3$ ,  $S = 1$ ,  $k_F = 1.36 \text{ \AA}^{-1}$ ,  $J_s(2k_F) = 3.0 \text{ \AA}^{-4}$ ,  $J_{sd} \simeq 0.1 \text{ eV}$ , and  $E_F \simeq 10 \text{ eV}$  yields  $\rho_{\text{mag}}(0) \simeq 0.1 \mu\Omega \text{ cm}$ . This value should be compared with that of  $\rho_{\text{str}}(0)$  which for amorphous ferromagnets could be as large as  $100 \mu\Omega \text{ cm}$ . This comparison and the remark made in Sec. II about the magnitude of the fourth term in Eq. (11a) permit us to neglect the first and fourth terms in this equation [Eq. (11a)] while attempting the above-mentioned fits. In the first type of fit, we drop the second term in Eq. (11a) also and fit the  $r(T)$  data to the following relation:

$$r(T) = f_1(T) + a_3 T^2 \equiv f_2(T), \quad (22)$$

where  $f_1(T)$  is given by Eq. (9). The values of the fit parameters  $a_1$ ,  $a_2$ ,  $a_3$ ,  $W(0)$ , and  $\Theta_D$  so determined are used in Eqs. (10b) and (5) to calculate  $S_0(2k_F)$  and  $k_F$ , respectively, and in the following expressions

$$\frac{\Delta\rho}{\rho} = \frac{2[1 - S_0(2k_F)]}{S_0(2k_F)} W(0) \left[ \frac{4 \times 300}{\Theta_D} - 1 \right] + \frac{a_3 \times (300)^2}{S_0(2k_F)}, \quad (23)$$

$$r(300 \text{ K}) = \frac{S_0(2k_F) + 2W(0)[1 - S_0(2k_F)][(4 \times 300 / \Theta_D) - 1] + a_3 \times (300)^2}{S_0(2k_F) + 2W(0)[1 - S_0(2k_F)][(4T_0 / \Theta_D) - 1] + a_3 T_0^2}, \quad (24)$$

$$\alpha'_1 = \frac{8W(0)[1 - S_0(2k_F)] / \Theta_D}{S_0(2k_F) + 2W(0)[1 - S_0(2k_F)][(4T_0 / \Theta_D) - 1] + a_3 T_0^2}, \quad (25)$$

to compute  $\Delta\rho/\rho$ ,  $r(300 \text{ K})$ , and  $\alpha'_1$ . Values of all the above-mentioned quantities are then compared in Tables III and IV with those deduced previously by fitting the  $r(T)$  data with the function  $f_1(T)$ . Such a comparison demonstrates that the inclusion of the magnetic contribution [the term  $a_3 T^2$  in Eq. (22)] in the expression for total resistivity does not significantly change the values of  $a_1$ ,  $a_2$  [and hence of  $S_0(2k_F)$ , except for the alloy with  $x = 2$  for which  $a_1$  and  $a_2$  do change but  $S_0(2k_F)$  remains unaltered within the error limits],  $\Delta\rho/\rho$  and  $r(300 \text{ K})$  but considerably improves the quality of the fits as inferred from the reduced (by an order of magnitude) value of the sum of squares ( $\chi^2$ ), removes the discrepancy between observed and calculated values of  $\alpha'_1$  and lowers the values of  $\Theta_D$  and  $k_F$  so as to make them fall within the physically acceptable range. Other important findings include the following: (i) in accordance with the predictions of the diffraction model, positive TCR is accompanied by  $2k_F$  values that are either well below or well above  $k_p$  [in the absence of the structural data on amorphous  $\text{Fe}_{80}\text{B}_{20-x}\text{C}_x$

alloys, we compare the determined  $2k_F$  values with  $k_p \simeq 3.1 \text{ \AA}^{-1}$ , where the first peak in the  $S(k)$  vs  $k$  curves for amorphous  $\text{Fe}_{80}\text{B}_{20}$  occurs,<sup>28</sup> since no appreciable shift in  $k_p$  is expected when B in  $\text{Fe}_{80}\text{B}_{20}$  is partially replaced by C] and  $\Theta_D$  values are now reasonably close to  $T^* \simeq 200 \text{ K}$ , the temperature above which  $\rho_{\text{str}}$  exhibits a linear temperature dependence, and (ii) the high-temperature and  $f_2(T)$  fits give roughly the same value for the coefficient of the magnetic term [the  $T^2$  term in Eqs. (15) and (22)]. Moreover, the values of this coefficient in the expression for total resistivity, i.e.,  $\beta' = \rho(T_0)a_3$  or  $\beta'' = \rho(T_0)\alpha'_2$ , for the present glassy alloys fall within the range of values obtained for the same coefficient in crystalline ferromagnetic metals and alloys.<sup>29-32</sup> For instance, the values  $\beta' = (0.91 \pm 0.03) \times 10^{-11} \Omega \text{ cm K}^{-2}$  and  $\beta'' = (0.98 \pm 0.05) \times 10^{-11} \Omega \text{ cm K}^{-2}$  [ $\rho(T_0) = 120 \pm 2 \mu\Omega \text{ cm}$ ] determined in this work for  $\text{Fe}_{80}\text{B}_{20}$  are in excellent agreement with the value  $0.96 \times 10^{-11} \Omega \text{ cm K}^{-2}$  previously reported<sup>29</sup> for crystalline Fe. This observation resurrects our earlier (Sec. II) remark that the quenched disorder does



not significantly influence the *coherent* electron-magnon scattering contribution to  $\rho$ .

In order to verify the theoretical prediction,<sup>12</sup> Eq. (12), concerning the relative importance of the  $T^{3/2}$ , and  $T^2$  terms in Eq. (11a), the second type of fit, wherein the  $r(T)$  data are fitted to the expression

$$\begin{aligned} r(T) &= f_1(T) + a_3 T^2 + a_4 T^{3/2} \\ &= f_2(T) + a_4 T^{3/2}, \end{aligned} \quad (26)$$

has been attempted. A nonlinear least-squares-fit computer program in which all six parameters  $a_1$ ,  $a_2$ ,  $a_3$ ,  $a_4$ ,  $\Theta_D$ , and  $W(0)$  are varied to achieve the best fit has been employed for this purpose. Such a fitting procedure is found to converge for some of the investigated glassy alloys only and in such cases (i) the values of all the parameters (except for  $a_4$ ) are widely different from those obtained from the previous fits, (ii)  $\Theta_D$  once again assumes unphysically large values, (iii) the coefficient  $a_4$  is *negative*, and (iv) the quality of the fits deteriorates, as inferred from the increased value of  $\chi^2$  compared with the corresponding values determined in previous fits. Efforts to fit the  $r(T)$  data in the temperature range 50 to 100 K to the relation  $r(T) = \alpha_0 + \alpha_2 T^2 + \alpha_3 T^{3/2}$  also met the same fate. Recent electrical resistivity studies<sup>33</sup> on amorphous  $\text{Fe}_{80}\text{B}_{20}$  also yield an unphysical negative value for the coefficient  $\alpha_3$  in the temperature range  $20 \leq T \leq 100$  K. The above findings strongly suggest that the  $T^{3/2}$  term in Eq. (11a), if present, is negligibly small [implying thereby that the competing contributions to  $\rho_{\text{mag}}(T)$  due to the *incoherent* and *elastic* components of electron-magnon scattering almost balance each other] compared to the  $T^2$  term. This inference is at variance with the theoretical prediction<sup>12</sup> that for amorphous ferromagnets the  $T^{3/2}$  term should dominate (greater by 2 orders of magnitude) over the  $T^2$  term in the intermediate-temperature range.

We now focus our attention on the concentration dependence of the parameters  $\alpha'_1$ ,  $\alpha'_2$ ,  $\alpha_2$ ,  $\beta$ ,  $a_3$ , and  $\Theta_D$  (Fig. 5 and the inset of Fig. 4). The results of recent magnetization<sup>34,35</sup> and specific-heat<sup>36</sup> measurements have also been included in these figures for comparison. It is evident from Fig. 5 that  $a_3$ ,  $\alpha'_2$ ,  $\beta$ , and  $\mu$  (magnetic moment per alloy atom at 4.2 and 77 K) present a strikingly similar concentration dependence which completely differs from that of  $\alpha'_1$  and  $\alpha_2$ , which among themselves, exhibit roughly the same functional dependence on  $x$ . This implies that basically two different physical mechanisms are responsible for the existence of the terms in  $\rho(T)$  with coefficients ( $a_3$ ,  $\alpha'_2$ ,  $\beta$ ) and ( $\alpha'_1$  and  $\alpha_2$ ). The marked similarity in the concentration dependence of  $a_3$ ,  $\alpha'_2$ , and  $\mu$  is further highlighted in the inset of Fig. 3 where  $a_3$  and  $\alpha'_2$  are shown to scale with  $\mu$ . This observation lends a firm support to our contention that the  $T^2$  terms in the  $f_2(T)$  and high-temperature asymptotic fits are *purely of magnetic origin*. Figure 5 would normally tempt one to make a similar statement about the logarithmic term in  $\rho(T)$  at low temperatures but a check on this temptation is provided by the observations: (a) the value of  $\beta$  depends on the temperature interval chosen for the fit and (b) our measurements do not extend to sufficiently low temperatures (mK range) where one expects to observe a *saturation*

in resistivity which is a *prerequisite* for attempting a physically meaningful distinction<sup>2</sup> between various models proposed in the literature for explaining the resistivity-minimum phenomenon in metallic glasses. A more cautious statement would be that the logarithmic up turn of resistivity at low temperatures in the glassy alloys in question *does not solely arise from the structural disorder*, as some of the existing theories claim, but the magnetic state of these alloys also plays a vital role. In view of the structural origin of the linear temperature dependence of  $\rho$  at high temperatures [the term  $\alpha'_1 T$  in Eq. (15)], the close resemblance between  $\alpha'_1(x)$  and  $\alpha_2(x)$  suggests that the quadratic temperature dependence of  $\rho(T)$  in the intermediate-temperature range results mainly from the scattering of conduction electron from the structural disorder. However, a small contribution to this  $T^2$  term [i.e., the  $\alpha_2 T^2$  term in Eq. (14)] from the *coherent* electron-magnon scattering cannot be completely ruled out. Finally, a few remarks can be made concerning the concentration dependence of  $\Theta_D$  in the present glassy alloys. One notices from the inset of Fig. 4 that the values of  $\Theta_D$  deduced from the  $f_2$  fits and Eq. (20) are in reasonable agreement with one another but they are smaller in magnitude by about 40% than those obtained from the recent specific-heat data<sup>36</sup> with the exception of  $\text{Fe}_{80}\text{B}_{20}$  for which the resistivity and specific-heat measurements both

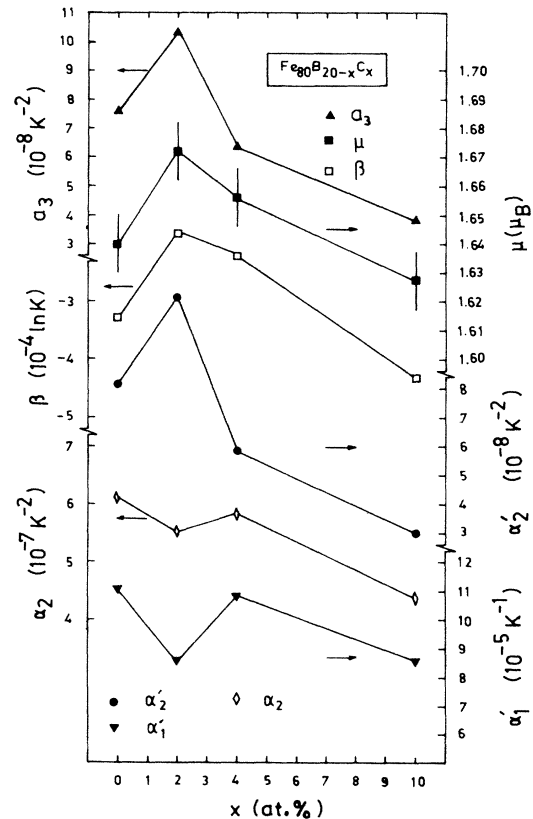


FIG. 5. Concentration dependence of the coefficients  $\alpha'_1$ ,  $\alpha'_2$ ,  $\alpha_2$ ,  $\beta$ , and  $a_3$ , and the magnetic moment  $\mu$ . Note that the values of  $\mu$  for the alloys with  $x=2, 4$ , and  $10$ , taken from Ref. 34, have been measured at  $T=77$  K whereas the value of  $\mu$  for  $\text{Fe}_{80}\text{B}_{20}$ , taken from Ref. 35, is the moment value at  $T=4.2$  K.

yield values which are fairly close to each other. Such a discrepancy between different sets of  $\Theta_D$  values, in our opinion, arises from the fact that the values of  $\Theta_D$  determined in Ref. 36 do not represent the values of  $\Theta_D$  at 0 K, which are more relevant when a comparison with the  $\Theta_D$  values deduced from the resistivity data is sought, but the average values over the temperature interval  $1.5 \leq T \leq 10$  K. It is evident from Fig. 1 of Ref. 36 that  $\Theta_D(T)$  approaches a limiting value  $\Theta_D(0)$  only for  $T \lesssim 7$  K ( $T \lesssim 10$  K for  $\text{Fe}_{80}\text{B}_{20}$ ) and beyond  $T \simeq 7$  K  $\Theta_D(T)$  increases with increasing temperature. The average value of  $\Theta_D$  over an extended temperature range is, therefore, inevitably higher than  $\Theta_D(0)$ .

## VI. CONCLUSIONS

A quantitative comparison between the theoretical predictions and the present experimental results permits us to draw the following conclusions.

- (i) Both the electron-ion potential scattering and electron-magnon scattering contribute to resistivity.
- (ii) The structural contribution dominates over the mag-

netic contribution in the entire temperature range of the present investigation.

(iii) Quenched disorder does not have any appreciable effect on the *coherent* electron-magnon scattering contribution to  $\rho$ .

(iv) The competing contributions to  $\rho_{\text{mag}}(T)$  arising from the *incoherent* and *elastic* parts of the electron-magnon scattering are of the same magnitude roughly and hence the  $T^{3/2}$  term in the expression for  $\rho_{\text{mag}}(T)$  is negligibly small compared to the  $T^2$  term.

(v) The magnetic state of the metallic glasses under consideration cannot be ignored while seeking an interpretation of the logarithmic increase of  $\rho$  for  $T < T_{\text{min}}$  in the light of the existing theories.

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\*Present address: School of Physics, University of Hyderabad, Central University P.O., Hyderabad 500 134, Adhra Pradesh, India.

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Note that the high-temperature expression of  $\rho(T)$  in this reference does not contain the term  $-2W(0)[1-S_0(2k_F)]$ ; compare Eq. (6) of this reference with Eq. (8b) of the present paper. This omission basically results from the use of a

wrong high-temperature expression of  $W(T)$  given in Ref. 8 while deriving the expression of  $\rho(T)$  for  $T \gtrsim \Theta_D$  and leads to wrong theoretical values of  $\rho$  at room temperature in this reference.

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