

Disorder and magnetic effects on the electron-transport properties of iron-chromium-boron metallic glasses

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We report measurements of the electrical resistivity and magnetoresistivity of $\text{Fe}_{80-x}\text{Cr}_x\text{B}_{20}$ ($0 \leq x \leq 32$) from 60 mK to 300 K and in magnetic fields up to 4.3 T. The results show the presence of two effects: one of nonmagnetic origin, associated with electron-electron interaction in a highly disordered system, and the other of magnetic origin, probably due to the Kondo effect and spin fluctuations. The high-field, isotropic magnetoresistivity is examined in detail and shown to be quantitatively inconsistent with the currently available models.

I. INTRODUCTION

The electrical resistivity of metallic glasses at low temperature has been the subject of considerable research and some controversy for several years. Two general features stand out: the first, known as Mooij correlation,^{1,2} is the rough correlation between the room-temperature resistivity coefficient $\alpha = \partial \ln \rho / \partial T |_{300 \text{ K}}$ and the magnitude of the resistivity, α being negative above approximately 150 $\mu\Omega \text{ cm}$ and positive below; the second is that in nearly every system, $\alpha \leq 0$ at low temperature, irrespective of its high-temperature value.³

The first glassy alloys to be made generally exhibited positive α at high temperature so that a resistance minimum was observed at low temperature, usually around 20 K. This observation, coupled with a variation with temperature that could be crudely characterized as logarithmic, led a number of authors^{4,5} to invoke the Kondo effect as the cause. However, the general insensitivity of the low-temperature resistivity coefficient to an applied magnetic field together with the observation of similar behavior in ferromagnetically ordered systems led other workers to prefer an explanation of structural origin, most notably scattering from atomic tunneling states.^{6,7} The merits and deficiencies of the two models have been reviewed by Harris and Strom-Olsen⁸ who concluded that neither of the models provides a good quantitative fit to the data.

In the past few years it has been shown that the low-temperature resistivity behavior of metallic glasses might be explained on the basis of two theories of the quantum corrections to the resistivity in disordered metals; one invoking electron-electron interaction in a disordered environment^{9,10} and the other one electron-quantum interference effects, the so-called weak-localization effects.^{11,12} In spite of the recent efforts to test these hypotheses in metallic glasses,^{13–20} the question is still open. One difficulty in distinguishing between the various models is a surprising absence of suitable systematic surveys. Specifically missing is a system where the magnetic order may be progressively suppressed without significant changes in structure. In earlier works^{21,22} we showed that these conditions are met in $\text{Fe}_{80-x}\text{Cr}_x\text{B}_{20}$ thus making this system

a most suitable candidate for such an investigation.

We examined in detail the electrical resistivity over the range 100 mK to 300 K in fields up to 4.3 T and the thermoelectric power from 4.2 to 550 K. In the present article we report the electrical resistivity; the thermopower will be presented in a forthcoming article.

II. EXPERIMENTAL DETAILS

Alloys of Fe-Cr-B were prepared in an arc furnace under titanium-gettered high-purity argon starting with Fe 99.999% purity, Cr 99.999% purity, and B 99.95% purity. The resulting pellets (typically 1–2 g) were melt-spun under 15 kPa He onto a copper wheel with tangential velocity ranging from 35 to 50 m/s. Samples of $\text{Fe}_{80-x}\text{Cr}_x\text{B}_{20}$ were made in the range $0 \leq x \leq 32$; one sample was made of composition $\text{Fe}_{48}\text{Cr}_{35}\text{B}_{17}$. Debye-Scherrer x-ray photography of the resulting ribbons (typically 1–2 mm wide by 25 μm thick) showed no detectable crystalline lines. Electron microprobe analysis of selected samples confirmed that fluctuations in composition must arise over scales smaller than the beam diameter (approximately 1 μm). Differential scanning calorimetry (DSC) was used to determine the crystallization temperature T_{cr} , the enthalpy of crystallization ΔH and, in samples where $x \leq 10$, the Curie temperature T_{C} .

The room-temperature electrical resistance was measured on long sections of ribbon (≥ 0.5 m) using a four-terminal dc technique and converted to electrical resistivity using the measured length, mass, and mass density. The accuracy on the electrical resistivity is approximately 2% (this includes an estimated systematic error of approximately 1% associated with using the reciprocal of the average cross-sectional area instead of the average of its reciprocal). Changes in resistance with temperature or magnetic field were detected to within a few parts per millions using a four-terminal ac ohm meter.²³ The strain effects due to differential thermal contraction were minimized by fixing the samples to the holder using Mylar foil, and putting varnish only on their ends. In order to minimize demagnetization and Hall effects, transverse and longitudinal magnetoresistance were measured with the magnetic field in the plane of the samples.

III. RESULTS AND DISCUSSION

A. Magnetic properties

The major physical parameters of the full series of $\text{Fe}_{80-x}\text{Cr}_x\text{B}_{20}$ metallic glasses are given in Table I. The magnetic properties may be summarized as follows: Alloys with $x \leq 28$ show a sharp ferromagnetic transition at the temperature T_C , the average moment per iron atom, μ_{Fe} , and the average hyperfine field, $\langle H_{\text{hf}} \rangle$, all falling monotonically upon addition of Cr. As we have noted earlier,^{21,22} the composition dependence of μ_{Fe} and $\langle H_{\text{hf}} \rangle$ imply that the magnetic moment on an iron atom disappears when it has more than four Cr atoms as nearest neighbors. The onset of magnetism, when the alloys are approached from the Cr-rich end, is due to the inhomogeneous formation of a moment on the iron atom. After the disappearance of ferromagnetism there is a narrow region, $28 < x < 33$, in which a spin-glass-like phase is observed below some characteristic spin-freezing temperature T_f . In this regime, Mössbauer studies²⁴ show only weak-relaxation effects below T_f , giving additional support to the view that in these alloys μ_{Fe} has been almost destroyed by Cr.

B. Electrical resistivity

Turning now to the electrical resistivity, two strikingly contrasted features are observed: (i) the magnitude of the resistivity is almost independent of Cr concentration, and (ii) the temperature dependence of the resistivity is a strong function of Cr concentration. This may be seen by comparing $\rho_{290\text{ K}}$ with $\alpha_{290\text{ K}}$ in Table I or, more directly, by looking at the overall temperature dependence of the normalized resistance change as shown in Fig. 1 for selected alloys. Even the addition of as little as 1 at. % Cr drastically reduces $\alpha_{290\text{ K}}$ from its value in $\text{Fe}_{20}\text{B}_{20}$, while the room-temperature resistance changes by less than one percent. When the temperature dependence is examined

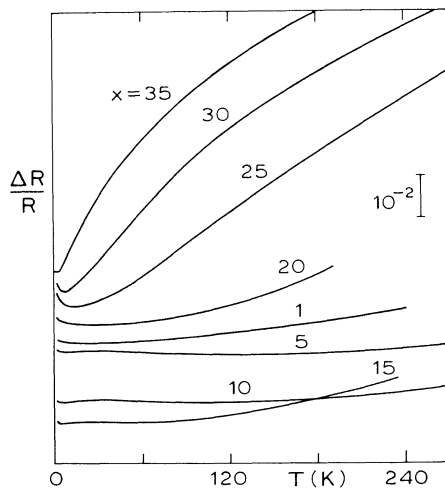


FIG. 1. Temperature dependence of the normalized resistance $\Delta R/R = [R(T) - R(4.2\text{ K})]/R(4.2\text{ K})$ of $\text{Fe}_{80-x}\text{Cr}_x\text{B}_{20}$ ($1 \leq x \leq 30$) and $\text{Fe}_{48}\text{Cr}_{35}\text{B}_{17}$ metallic glasses. For clarity, the various curves have been zero shifted.

TABLE I. Crystallization, magnetic, and electronic transport parameters of $\text{Fe}_{80-x}\text{Cr}_x\text{B}_{20}$ metallic glasses. The numbers in parentheses represent the error on the last digit(s).

x	T_{cr} (K)	ΔH (kJ/mole)	T_C (K)	T_f (K)	μ_{Fe} (μ_B)	$\langle H_{\text{hf}} \rangle$ (T)	$\rho_{290\text{ K}}$ ($\mu\Omega\text{ cm}$)	$\alpha_{290\text{ K}}$ (10^{-4} K^{-1})	$\partial \ln \rho / \partial H$ (10^{-4} T^{-1})	$10^4 \Delta \rho / \rho_0$
0	742.8	7.20	659(1)		2.00	28.6	118.8(20)	1.27	-1.98 ^a	55 ^a
1	743.2	6.82	632(1)				120.2(20)	0.60	-0.03	
2	747.0	6.92	597(1)				120.6(20)			
5	768.0	6.51	500(1)		1.80(2)	24.5	124.0(20)	0.33	2.58	23.8
10	786.5	6.57	355(5)		1.38(2)	19.0	123.4(20)	0.61	3.18	8.5
15	803.0	5.77	225.6(2)		0.99(2)	12.6	124.5(20)	1.55	4.10	3.1
20	820.2	6.69	120.2(2)		0.65(2)	7.4	126.2(20)	1.55		1.3
25	834.0	6.77	33.20(5)		0.31(2)	3.9	128.5(20)	2.00		<0.4
26	833.1	7.31	26.0(10)				125.0(20)			<0.4
27	836.7	7.22	20.5(10)				132.5(20)	1.70		<0.4
28	832.1	7.56					129.0(20)			<0.4
29				7.60						0
30	827.6			6.07						0
32				3.47						0

^aReference 38.

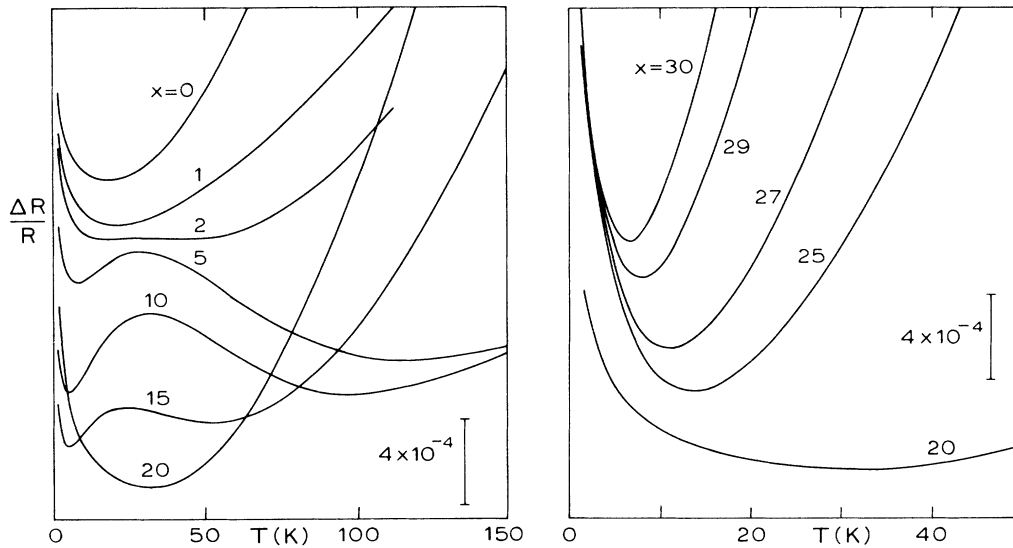


FIG. 2. Temperature dependence of the normalized resistance $\Delta R/R = [R(T) - R(4.2 \text{ K})]/R(4.2 \text{ K})$ of $\text{Fe}_{80-x}\text{Cr}_x\text{B}_{20}$ metallic glasses. A double minimum is observed in alloys containing 2, 5, 10, and 15 at. % chromium. For clarity, the various curves have been zero shifted.

in finer detail, an even more complex picture emerges. This is shown in Fig. 2. The addition of 1 at. % Cr flattens the single minimum that occurs in $\text{Fe}_{20}\text{B}_{20}$ at about 17 K. The flattening is in fact the precursor to the emergence of a second minimum which is just distinguishable for $x=2$. The temperature of the second minimum at first increases with x and the minimum itself becomes more pronounced (e.g., $x=5$); at higher x , the minimum moves back towards lower temperature and gradually washes out. By $x=20$ it has merged once more with the first minimum to produce a single flat minimum at about 32 K. Thereafter this single minimum remains and moves slightly downward in temperature. An interesting corollary to this behavior is that, until $x \approx 20$, the low-temperature resistance (below approximately 4 K) is essentially unaltered by the appearance of the second minimum. This is clearly illustrated in Fig. 3 by the tem-

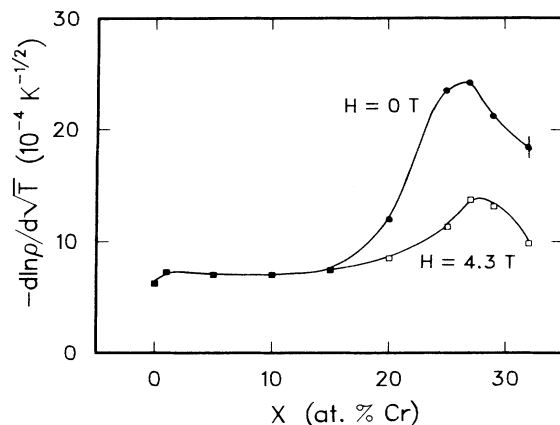


FIG. 3. Concentration dependence of the \sqrt{T} resistivity coefficient $\beta = \partial \ln \rho / \partial \sqrt{T} |_{T=1.5 \text{ K}}$ of $\text{Fe}_{80-x}\text{Cr}_x\text{B}_{20}$ metallic glasses: \bullet $H = 0 \text{ T}$; \square , $H = 4.3 \text{ T}$.

perature coefficient of the resistivity taken here as $\beta = (\partial \ln \rho / \partial \sqrt{T})_{1.5 \text{ K}}$ (for reasons which will be apparent later). Up to $x=15$, β is essentially independent of Cr concentration, but as the two minima run together it increases sharply, passes through a maximum and then diminishes. Of further significance is the effect of an applied magnetic field on β . For $\text{Fe}_{20}\text{B}_{20}$, and for all Cr compositions up to the point where β increases, a magnetic field of 4.3 T has no detectable effect. However in the region where β is enhanced (i.e., where the second minimum has remerged with the first) the magnetic field substantially reduces β , though its value still remains above the zero-field limit.

The one alloy well beyond the limit of ferromagnetism ($\text{Fe}_{48}\text{Cr}_{35}\text{B}_{17}$) shows significantly different behavior and must be considered separately: here the low-temperature coefficient is slightly enhanced by a magnetic field (Fig. 4). We indicate that the temperature dependence above the minima is generally insensitive to the magnetic field, *except* for alloys around the critical composition for ferromagnetism, where the position resistivity coefficient is reduced by the field (e.g., Fig. 4).

The above behavior suggests the following conclusions, which will be discussed in more detail below: (1) the negative-temperature coefficient at low temperatures in those alloys where it is field insensitive is of structural (disorder) origin rather than magnetic origin. (2) The second resistance minimum, which appears with the addition of Cr, is a magnetic effect; from this it follows that the low-temperature behavior in alloys with more than 15 at. % Cr has both magnetic and structural (disorder) contributions. (3) There are significant magnetic contributions to the high-temperature positive coefficient in samples close to the critical composition for ferromagnetism; these contributions must be of a different kind than those which cause the second minimum.

Before discussing these points in the light of specific

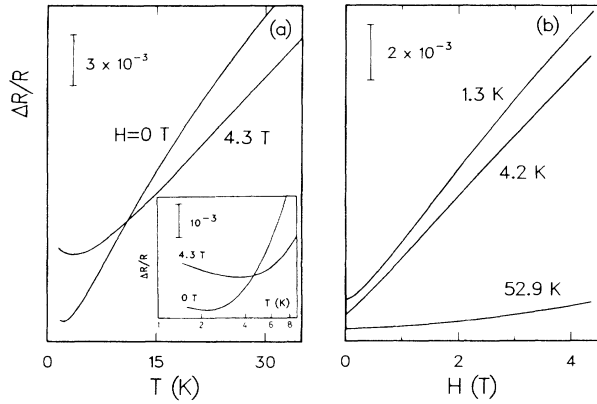


FIG. 4. (a) Temperature dependence of the normalized resistance, $\Delta R/R = [R(T) - R(4.2 \text{ K})]/R(4.2 \text{ K})$, of $\text{Fe}_{48}\text{Cr}_{35}\text{B}_{17}$ in zero and 4.3 T magnetic fields. Details of the low-temperature region are enhanced in the inset. In both cases the curves have been zero shifted. Their relative position is obtained using $([R(4.2 \text{ K}, 4.3 \text{ T}) - R(4.2 \text{ K}, 0)]/R(4.2 \text{ K}, 0)) = 9.3 \times 10^{-3}$. The transverse ($H \perp j$) magnetoresistance ($\Delta R/R = [R(H) - R(0)]/R(0)$) at 1.3, 4.2, and 52.9 K is shown in (b). For clarity, the various curves have been zero shifted.

models, we first discuss the effect that thermal expansion, remnant crystallinity, and ferromagnetic anisotropy in the resistivity (FAR) may have on the resistivity and its temperature coefficient. Although these effects would not necessarily affect qualitative conclusions, they make quantitative analysis difficult.

In all the experiments, what is measured is the logarithmic derivative of the resistance with respect to temperature at constant pressure, $\alpha_R \equiv \partial \ln R / \partial T|_P$, whereas the quantity of interest theoretically is $\alpha_V \equiv \partial \ln \rho / \partial T|_V$. These two quantities are related by the expression

$$\alpha_V = \alpha_R + \left[1 - 3 \left(\frac{\partial \ln \rho}{\partial \ln V} \right) \right] \alpha_L, \quad (1)$$

where $\alpha_L \equiv \partial \ln L / \partial T|_P$ is the thermal expansion coefficient and $\partial \ln \rho / \partial \ln V|_P$ is the volume dependence of the resistivity. Unfortunately, neither α_L nor $\partial \ln \rho / \partial \ln V|_P$ have been measured for $\text{Fe}_{80-x}\text{Cr}_x\text{B}_{20}$ alloys. However both Fe-B and $(\text{Fe,Cr})_{85}\text{B}_{15}$ show invar characteristics below T_C where α_L lies in the approximate range $\pm 1 \times 10^{-5} \text{ K}^{-1}$, which for most metallic glasses $\partial \ln \rho / \partial \ln V$ lies in the range ± 1 . If we take these values as representative, α_V and α_R differ by a quantity of the order of $\pm 2 \times 10^{-5} \text{ K}^{-1}$ at and above 150 K. Although this quantity cannot readily be neglected in any detailed quantitative analysis (for example, it could introduce a small difference between the position of the minima). Furthermore, the temperature dependence of α_L when $T \ll T_C$, is too small to give rise to the local resistance maximum observed near 30 K.

The influence of remnant crystallinity may be approximated by assuming that a small fraction f of the sample consists of a uniform distribution of spherical crystallites.

In this case the measured α is given by²⁵

$$\alpha = \alpha_a + \frac{9f[\rho_{\text{cr}}(\alpha_{\text{cr}} - \alpha_a)/\rho_a]}{[1 + 2(\rho_{\text{cr}}/\rho_a)^2]} + O(f^2), \quad (2)$$

where the subscripts a and cr refer to the amorphous and crystalline phases, respectively. At high temperatures α_{cr} lies between $5 \times 10^{-4} \text{ K}^{-1}$ and 10^{-3} K^{-1} over the whole range and $\rho_{\text{cr}}/\rho_a \approx 0.5$. Our x-ray data indicates $f \leq 0.02$. From these figures we estimate that the cast-in crystallite contribution to α is less than $2 \times 10^{-5} \text{ K}^{-1}$.

Finally, it is well known that ferromagnetic metals show an excess resistivity due both to domain-wall scattering and to the distortion of the current lines at domain walls. The former effect is insignificant in the present case because the electron mean free path is of the order of 10 Å or less. However, the latter effect (as well as the ferromagnetic anisotropy in the resistivity²⁶) can change the reduced resistivity by up to 5×10^{-3} so that the temperature dependence of the resistivity in zero applied field can depend critically on the magnetic history of the sample. To avoid these difficulties, we applied a small field ($< 0.02 \text{ T}$) to saturate the magnetization. This field is, of course, too small to influence the scattering mechanisms. From the above discussion we conclude that extraneous effects are not of major importance in the data presented.

We now turn to a detailed discussion of the behavior of α . As outlined in the introduction, the fact that in almost all metallic glasses α is negative at sufficiently low temperatures has been long known. In magnetic alloys this behavior was first ascribed to Kondo scattering. In a crystalline ferromagnet the exchange field freezes out the spin degrees of freedom, but in an amorphous ferromagnet the exchange field is spread over some range. It was argued (from fitting to Mössbauer data) that some spins would see zero-exchange field and so would be able to produce an observable Kondo effect.^{4,5} The insensitivity of ρ to an external field was interpreted as meaning that the distribution of fields, $P(H)$, close to zero was flat⁵ so that shifting it either way by an external field had no effect on the number of *free* spins. This explanation of course was limited to ferromagnetic glasses with an appropriate mixture of positive and negative exchange interactions. It cannot explain the low-temperature negative temperature coefficients of resistivity (TCR) in nonmagnetic systems, even though the effect in these alloys is of similar magnitude.³ In any event, we believe that the behavior of the low-temperature resistivity on the addition of small amounts of Cr is strong evidence against this interpretation in $\text{Fe}_{80}\text{B}_{20}$ or in those alloys where there is no significant field effect. From the Mössbauer data^{22,24} it is clearly seen that the hyperfine field is extremely sensitive to Cr content; from which it follows that the number of spins in zero internal field should also be very sensitive to Cr content. However, as stated earlier, the low-temperature form of the resistivity is virtually independent of Cr composition until it exceeds 15 at. %. In these alloys therefore, we do not believe that the negative TCR at low temperatures can be due to the Kondo effect. We also believe that we can eliminate scattering from two-

level systems (TLS's) because we see no detectable change in α upon annealing, whereas it is known that the number of TLS's is drastically reduced by annealing.²⁷

The clue to the origin of the low-temperature behavior of ρ is provided by its temperature dependence. As was first pointed out by Rapp *et al.* for some Fe-based glasses²⁸ and later by Cochrane and Strom-Olsen³ for a wide variety of alloys, there is an excellent one-parameter fit of the data to \sqrt{T} below about 2 K. Cochrane and Strom-Olsen³ then showed that most metallic glasses followed the empirical rule

$$\sigma(T) - \sigma(0) = (500 \pm 100) \sqrt{T} (\Omega \text{m})^{-1}.$$

The fact that Fe-Cr-B (up to 15 at. % Cr) follows this rule is further evidence against magnetic effects as the cause of the low-temperature behavior since the rule applies equally for nonmagnetic alloys.

The origin of the \sqrt{T} behavior may be found in the influence of electron-electron interactions in a disordered environment—the so-called Coulombic interaction effect. In highly disordered environments the motion of conduction electrons is diffusive rather than ballistic so that the conductivity should be written in terms of a diffusion constant as

$$\sigma = D(e^2(dN/d\mu)), \quad (3)$$

where $dN/d\mu$ is the thermodynamic electronic density of states.

Under these conditions the effective electron-electron screening is altered from the value for a free-electron gas calculated by Fermi-liquid theory and this leads to a correction to the conductivity.¹⁰ The principal contribution arises from the so-called diffusion channel which predicts (in the absence of spin-orbit interaction) an additional term to the conductivity

$$\begin{aligned} \sigma_D(H, T) = & \frac{1.3}{\sqrt{2}} \left[\frac{e^2}{4\pi^2\hbar} \right] \left[\frac{k_B T}{\hbar D} \right]^{1/2} \\ & \times \left[\frac{4}{3} + \lambda^{j=1}(F) \left[\frac{3}{2} + \frac{g_3(h)}{1.3} \right] \right], \quad (4) \end{aligned}$$

where

$$\lambda^{j=1}(F) = \left(\frac{32}{3}\right) [1 + 3F/4 - (1 + F/2)^{3/2}] F^{-1}$$

measures the strength of the electron-electron interaction. The function $g_3(h)$, where $h = g\mu_B H/k_B T$, has the asymptotic form $0.056h^2$ when $h \ll 1$ and $(\sqrt{h} - 1.3)$ when $h \gg 1$. Equation (4) predicts a \sqrt{T} dependence to the resistivity in zero field.

We can obtain an upper limit for D from Eq. (4) by taking the limit $\lambda^{j=1} = 0$; a reasonable assumption because spin splitting and spin scattering significantly reduce the temperature contribution from this Hartree term. This gives $D \sim 3 \times 10^{-5} \text{ m}^2/\text{s}$, a value that may be compared to $D \sim 30 \times 10^{-5} \text{ m}^2/\text{s}$ estimated from Eq. (3) by assuming that conduction is by s electrons alone, with one s electron per Fe or Cr atom. However, it has been pointed out by Ballentine and coworkers^{29,30} that in liquid and amorphous transition metals, the d electrons play a significant, almost equal, role in the conductivity. In fact, if we use a

value for the density of states obtained from specific-heat measurements in $\text{Fe}_{80}\text{B}_{20}$,³¹ together with the value 0.5 for the electron-phonon mass-enhancement coefficient, we estimate $D \sim 2.9 \times 10^{-5} \text{ m}^2/\text{s}$ from Eq. (3). We therefore conclude that Eq. (4) can give a satisfactory account of the \sqrt{T} dependence of ρ in all alloys up to 15 at. % Cr, provided we include the d electrons as current carriers.³²

We can also explain the absence of any field dependence to the \sqrt{T} slope. In a simple case where there is no magnetic ordering, spin-flip scattering and spin-orbit scattering, Eq. (4) predicts that in a large field the \sqrt{T} slope is greater, by a factor of about $(1 + 9\lambda^{j=1}/24)/(1 + 9\lambda^{j=1}/8)$, than in zero field. However, as indicated earlier, the presence of spin splitting and spin scattering puts us in the limit $h \gg 1$ even in the absence of an external magnetic field. In fact, in the magnetically ordered state, the external field is added to the large internal exchange field (which we estimate here to be about 400 T, using the value 0.05 eV for the s - d exchange integral). Hence, in the present system the applied field has no effect on the coefficient β .

For samples containing more than 15 at. % Cr, there is clear evidence of an additional mechanism contributing to the low-temperature resistivity, which, from its sensitivity to the applied field, must be of magnetic origin.³³ This mechanism must also be responsible for the second minimum seen in samples with lower Cr content. Although we cannot offer a quantitative explanation we believe that this mechanism may in fact be Kondo scattering, as originally proposed for metallic glasses,^{4,5} but which we rejected earlier as a *general* explanation for a negative TCR at low temperatures. If the internal-field distribution is $P(H_{\text{int}})$ and if $\rho_K(H, T)$ is the Kondo resistivity at field H and temperature T , then the contribution of the Kondo effect to ρ is

$$\rho_K(T) \sim \int P(H_{\text{int}}) \rho_K(H_{\text{int}}, T) dH_{\text{int}}. \quad (5)$$

Unless $P(H)$ has a long tail (≥ 5 T) extending to negative values [which is not the case for Fe-Cr-B (Refs. 22 and 24)], then $\rho_K(T)$ will be affected by an external field that is greater than the larger of $k_B T_K/g\mu_B$ or $k_B T/g\mu_B$. We know that $\langle H_{\text{hf}} \rangle$ moves toward zero upon addition of Cr. To account for the data we must also assume that the Kondo temperature, T_K , is reduced upon addition of Cr. At low concentrations of Cr, T_K is high and at the same time there are very few sites available for Kondo scattering: this produces the barely resolved second minimum, one which is saturated at low temperatures and is insensitive to the applied field. The exact location of the second minimum is of course the result of an interplay between several effects, including the change of structure factor, but these are not of fundamental importance. As the concentration of Cr is raised, the number of sites in a small field increases strongly and so produces the pronounced minimum observed in the 5 and 10 at. % Cr samples. At the same time T_K must decrease. By 20 at. % Cr, $k_B T_K$ falls within the range of available fields (i.e., $g\mu_B H \sim k_B T_K$) and the external field can affect the temperature dependence of the resistance. Finally, the size of this contribution must decrease as the magnetism in the system is suppressed.

Before turning our attention to the magnetoresistance, we examine the behavior of the one alloy that does not show any magnetic transition, $\text{Fe}_{48}\text{Cr}_{35}\text{B}_{17}$. In this alloy we believe there are strong contributions due to spin fluctuations and this accounts for the different form of the data. A large contribution to ρ due to spin fluctuations (ρ_{sf}) has recently been observed in Fe-Zr metallic glasses.³⁴ In these alloys the background contribution could be subtracted and ρ_{sf} found to be in good agreement with the model of Kaiser and Doniach³⁵ (i.e., a $\sim T^2$ dependence for $T \leq T_{\text{sf}}$ with a crossover to a T dependence above T_{sf} , where T_{sf} is an effective spin-fluctuation temperature). In $\text{Fe}_{80-x}\text{Cr}_x\text{B}_{20}$ alloys the rapid increase in the positive contribution to ρ observed for compositions close to the onset of magnetism ($x \leq 25$) is in qualitative agreement with the latter model. The effect of an external magnetic field on ρ_{sf} has been discussed by Schulz³⁶ and it appears as a field dependence of T_{sf} . The latter can increase or decrease with the applied field so that the temperature dependence can be enhanced or reduced. The observed reduction in the positive-resistivity coefficient in the presence of a magnetic field suggests that if spin fluctuations are the cause of the additional temperature dependence, T_{sf} must be increased by the magnetic field. This is best seen in the nonmagnetic alloy $\text{Fe}_{48}\text{Cr}_{35}\text{B}_{17}$, where the negative-resistivity coefficient is enhanced by the field and the approximate T^2 behavior extended to higher temperatures. It must be noted, however, that the observed large positive magnetoresistance must have a different origin.

C. Isotropic magnetoresistivity

The field and temperature dependence of the magnetoresistance of alloys containing 15 at. % Cr or less is qualitatively similar. A typical plot of the transverse magnetoresistance ($\mathbf{H} \perp \mathbf{j}$) is shown in Fig. 5 for the alloy $\text{Fe}_{75}\text{Cr}_5\text{B}_{20}$ at selected temperatures. The large and anisotropic contribution observed at low fields will be the ob-

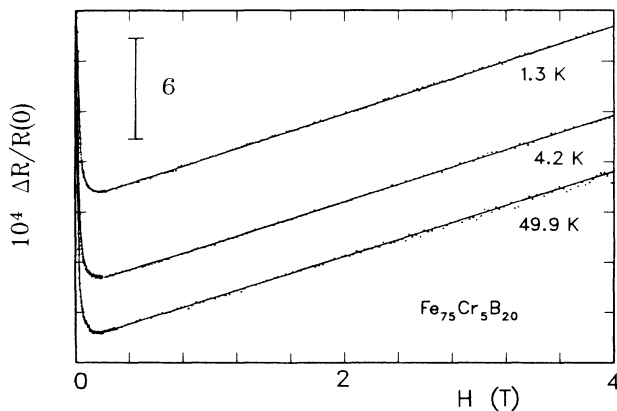


FIG. 5. Normalized transverse magnetoresistance of $\text{Fe}_{75}\text{Cr}_5\text{B}_{20}$ at various temperatures. In order to minimize demagnetization effects, the magnetic field was applied in the plane of the sample. For clarity, the curves have been zero shifted.

ject of a forthcoming article. However, for completeness, we have tabulated values of the ferromagnetic anisotropy in the resistivity, $\Delta\rho/\rho_0$, at 4.2 K (Table I). Beyond technical saturation, the magnetoresistance is isotropic, varies linearly with the applied field, and (at least up to 20 K) is independent of temperature. For this reason we have compiled only values of the forced magnetoresistance, $\partial \ln R / \partial H |_{P,T}$, at 4.2 K (Table I). It is negative in $\text{Fe}_{80}\text{B}_{20}$ but changes sign upon addition of as little as 1 at. % Cr, a trend similar to that observed in $\text{Fe}_{78}\text{Mo}_2\text{B}_{20}$ where $\partial \ln R / \partial H |_{P,4.2\text{K}} \simeq 1.1 \times 10^{-4} \text{ T}^{-1}$.³⁷

A qualitatively different behavior is observed for the magnetoresistance of alloys with $x \geq 20$ (Fig. 6) as well as for the one alloy well beyond the composition for magnetic order $\text{Fe}_{48}\text{Cr}_{35}\text{B}_{17}$ [Fig. 4(b)]. In these alloys, the magnetoresistance varies nonlinearly with field and, as pointed out in Sec. III B, shows a strong temperature dependence. There, the magnetoresistance seems to consist of a positive term showing a weak temperature dependence and a negative term with a strong temperature dependence.

Before we discuss the magnetoresistivity in the light of specific models, we estimate the magnitude of two extraneous contributions. One of them is associated with the forced volume magnetorestriction $\partial \ln V / \partial H |_{P,T}$, the other with the ferromagnetic anisotropy in the resistivity and high-field magnetic susceptibility χ_{hf} .

Ferromagnetic metallic glasses usually exhibit an isotropic dilation which, beyond technical saturation, is proportional to the applied magnetic field. The theoretical quantity of interest, namely the forced magnetoresistivity at constant volume $\partial \ln \rho / \partial H |_{V,T}$ is related to the mea-

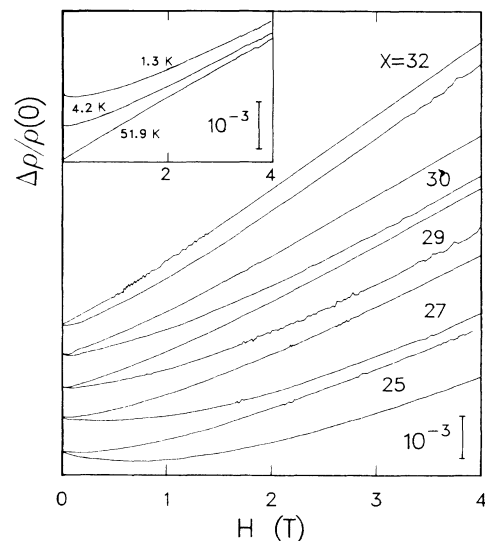


FIG. 6. Normalized transverse magnetoresistance ($\Delta R/R = [R(H) - R(0)]/R(0)$) of $\text{Fe}_{80-x}\text{Cr}_x\text{B}_{20}$ metallic glasses. The various curves have been zero shifted and grouped in pairs with common origin. The lower curve of a pair corresponds to $T = 1.3$ K and the upper curve to $T = 4.2$ K. The transverse magnetoresistance of the alloy $\text{Fe}_{60}\text{Cr}_{20}\text{B}_{20}$ is shown in the inset.

sured quantity, $\partial \ln R / \partial H |_{P,T}$, by

$$\left(\frac{\partial \ln \rho}{\partial H} \right)_{V,T} = \left(\frac{\partial \ln R}{\partial H} \right)_{P,T} + \left[\frac{1}{3} - \frac{\partial \ln \rho}{\partial \ln V} \right]_T \left(\frac{\partial \ln V}{\partial H} \right)_{P,T}. \quad (6)$$

Usually, the term on the extreme right in Eq. (6) is neglected. However, the situation can be quite different in invar alloys such as Fe-B and Fe-Cr-B.³⁸ Unfortunately, because $\partial \ln V / \partial H$ and $\partial \ln \rho / \partial V$ have been measured for only a few Fe-Cr-B glassy alloys, we can only give a rough estimate of the contribution from this term. Using measured values of $\partial \ln V / \partial H$ (Ref. 39) and the representative estimate $\partial \ln \rho / \partial V \sim 0.95$ (Fe₈₀B₂₀ at room temperature), we find the magnetovolume contribution to the force magnetoresistivity to be approximately $-2.5 \times 10^{-5} \text{ T}^{-1}$ in Fe₈₀B₂₀ and $-4.8 \times 10^{-6} \text{ T}^{-1}$ in Fe₇₄Cr₁₁B₁₅. Within the framework of Wohlfarth theory of itinerant ferromagnetism when $T \ll T_C$, this contribution is expected to show only a weak temperature dependence. It follows that even at the lowest temperature, the difference between $(\partial \ln \rho / \partial H |_{V,T})$ and $\partial \ln R / \partial H |_{P,T}$ is less than approximately 10% (except near $x=1$). For this reason, the magnetovolume correction will not be considered in later discussions.

An upper limit for the forced magnetoresistivity due to the interplay between $(\Delta \rho / \rho_0)$ and (χ_{hf}) is given by $\chi_{\text{hf}} \Delta \rho / (\rho_0 M_s)$. The correction due to this term is estimated to be smaller than $3 \times 10^{-8} \text{ T}^{-1}$ in Fe₈₀B₂₀ and 10^{-6} T^{-1} in Fe₆₀Cr₂₀B₂₀ and, consequently, can be safely neglected.

There are various possible contributions to the magnetoresistance including weak localization, Coulombic interaction, and scattering from magnetic ions. Weak localization provides a reasonable description of the magnetoresistivity in many nonmagnetic metallic glasses.^{13,18-20} For this reason, we first examine the contribution that arises from this effect. Limiting values of this contribution are given by the approximate high-field expression [i.e., $B \gg \hbar / (4eD\tau_\phi)$, where τ_ϕ is the phase coherence time]⁴⁰

$$-3.4 \times 10^{-4} \sqrt{B} \leq \frac{\Delta \rho}{\rho} \leq 1.7 \times 10^{-4} \sqrt{B}. \quad (7)$$

Here, we have used $\rho \simeq 120 \mu\Omega \text{ cm}$ and $B = \mu_0 | (H + M_s) |$. The lower and upper bounds correspond to $B \gg H_{\text{so}}$ and $B \ll H_{\text{so}}$, respectively, [where $H_{\text{so}} = \hbar / (4eD\tau_{\text{so}})$ and τ_{so}^{-1} is the spin-orbit scattering rate]. Although the limits of Eq. (7) are comparable to the measured values, the model fails to accurately describe the data. Specifically: (1) even for Fe₈₀B₂₀ (where $\mu_0 M_s \simeq 1.8 \text{ T}$) the linear-field dependence of the magnetoresistivity is incompatible with Eq. (7); (2) the sign change of the forced magnetoresistivity upon addition of 2 at. % Cr cannot be attributed to changes in τ_{so} ; (3) the absence of a temperature dependence of the forced magnetoresistivity cannot be explained.⁴¹

For the alloys with $x \geq 20$, the change of sign of the magnetoresistivity with increasing field is opposite to that predicted by weak localization,⁴⁰ and the localization con-

tribution at 4 T and 4.2 K is at most 20% of the observed signal in Fe₅₅Cr₂₅B₂₀ and 10% in Fe₄₈Cr₃₂B₂₀. Once the magnitude of H_ϕ is considered it becomes clear that the temperature dependence of the magnetoresistance of these alloys is also inconsistent with the model.³³

We therefore conclude that weak localization must be negligible over the entire composition range. In alloys with $x \leq 15$, such behavior is consistent with the theory, provided we assume $H_\phi \geq 1 \text{ T}$ even down to the lowest temperatures. We suggest that the required additional (and weakly temperature-dependent) dephasing mechanism could be provided by "free spins" which we believe occur in all alloys at the level of at least a few parts per million (of course in alloys with $x > 15$ they occur in much larger number).

Since Coulombic interaction effects provide a good description of the temperature dependence of ρ it seems reasonable to discuss its contribution to the magnetoresistance. Unfortunately, as we now show, it too cannot explain the data quantitatively, except perhaps near the critical composition for ferromagnetism. As indicated earlier, in a ferromagnetic metal, the magnetic field in Eq. (4) can be replaced by $H + H_{\text{int}}$. The effective field H_{int} felt by s -like conduction electrons can be as large as 500 T while that associated with the d -like electrons should be even larger. It follows that at low temperatures ($k_B T / g\mu_B \ll H_{\text{int}}$) the magnetoresistivity from Eq. (4) can be approximated by

$$\frac{\Delta \rho}{\rho^2} \simeq - \left[\frac{e^2}{4\pi^2 \hbar} \right] \lambda^{j=1(F)} \left[\frac{g\mu_B (H + H_{\text{int}})}{2\hbar D} \right]^{1/2}. \quad (8)$$

Accordingly, when $H, k_B T / g\mu_B \ll H_{\text{int}}$, the resistivity shows a linear-field dependence, $\partial \ln \rho / \partial H$ shows a weak temperature dependence and its sign is determined by H_{int} . These features are in fact qualitatively similar to the observed magnetoresistance of the alloys with $x \leq 15$. However, to account for the absence of a temperature dependence in Fe₇₅Cr₅B₂₀, the condition $|H_{\text{int}}| \geq 50 \text{ T}$ must be satisfied. Using $D \simeq 3 \times 10^{-5} \text{ m}^2/\text{s}$ and $|\lambda^{j=1(F)} \simeq \frac{8}{3}|$, gives $|\partial \ln \rho / \partial H| \leq 2.4 \times 10^{-5} \text{ T}^{-1}$. This is more than one order of magnitude smaller than the range over which it varies in these alloys. Finally, the sign change of the forced magnetoresistivity upon addition of Cr is inconsistent with the model, as it was for weak localization.

In alloys containing 20 at. % Cr or more, the observed temperature dependence is opposite to that predicted by Eq. (4). Furthermore, using the above estimate for ρ , D , and $\lambda^{j=1}$ together with $H_{\text{int}} = 0$, Eq. (4) is found to give a contribution that is nearly two orders of magnitude smaller than the observed signal. However it is possible that the contribution may be significantly enhanced by including spin fluctuations since, as pointed out by Millis and Lee,⁴² (1) the interaction parameter $|\lambda^{j=1(F)}|$ can then be much greater than 1 [in such a case $F/2 \sim (1-I)^{-1}$, where I is the Stoner parameter]; (2) the Zeeman spin splitting is much enhanced (an effect that can be accounted for by replacing the independent electron g factor by an effective g factor $g_{\text{eff}} \sim (F/2)g$). Consequently, when $F \gg 1$ the high-field limit is reached much earlier. In

particular, in the high-field limit the result of Millis and Lee⁴² is identical to Eq. (4) and may be written as

$$\frac{\Delta\rho}{\rho^2} = - \left[\frac{e^2}{4\pi^2\hbar} \right] \lambda^{j=1(F)} \left[\frac{g_{\text{eff}}\mu_B H}{2\hbar D} \right]^{1/2}. \quad (9)$$

When $F \gg 1$, $\Delta\rho/\rho^2$ can be approximated by $2.36F^2\sqrt{H}(\Omega\text{m})^{-1}$. In fact as we approach the critical composition ($x=32$) we do see an increase in the magnitude of the magnetoresistivity, though the measured behavior deviates significantly from \sqrt{H} . A value of $I \simeq 0.97$ is needed to account for the size of the magnetoresistance at 4.2 K and 4 T in the $x=32$ alloy. This is large but not unphysical. However, in this case, only if spin scattering is large,⁴³ can we explain the temperature dependence of ρ in zero field.

Calculations of the magnetoresistivity of metals containing magnetic ions have been executed in the first Born approximation (e.g., Refs. 44–47) using various levels of approximation (molecular field, etc.). In general, the suppression of spin-flip scattering by an external magnetic field gives rise to a negative contribution, while coherent spin scattering at different sites gives an additional term that may be either positive or negative.^{44–47} Consequently, the magnetoresistivity can be positive or negative. In a crystalline ferromagnet, because spin-flip scattering is frozen at $T=0$, the magnetoresistivity vanishes. Furthermore, for $T \leq T_C$ a linear-field dependence may be observed provided the applied field is less than the molecular field. The calculation of DeGennaro and Borchi⁴⁷ for metallic glasses indeed gives a negligible magnetoresistivity at $T=0$ but does not predict a linear-field dependence over any extended range.

The magnetoresistivity of our Fe-Cr-B metallic glasses is in strong disagreement with the above predictions. The linear and nearly-temperature-independent magnetoresistivity observed in alloys containing 15 at. % Cr or less is not even in qualitative agreement with the calculation of DeGennaro and Borchi; this is also true for the magnetoresistivity at $T=0$. The sign change that is observed upon the addition of approximately 2 at. % Cr is also very difficult to reconcile with such models. Furthermore, the nonlinear field dependence and change in sign with increasing field seen in alloys with $x \geq 20$ are not in qualitative agreement with the proposed models. Finally, the absence of singularity in the temperature derivative of the resistivity near T_C gives further evidence of the inadequacy of such models to describe the resistivity of Fe-Cr-B metallic glasses.

We now examine the contribution to the magnetoresistivity from the Kondo effect. This contribution, $\rho'_K(H, T)$, may be approximated by an expression similar to that of Eq. (5) (Ref. 48)

$$\rho'_K(H, T) \sim \int_{-\infty}^{+\infty} dH_{\text{int}} P(H_{\text{int}} + H) \rho_K(H_{\text{int}}, T). \quad (10)$$

The behavior of the magnetoresistivity is determined by

$P(H_{\text{int}})$ and $\rho_K(H_{\text{int}}, T)$. Even though $\rho_K(H, T)$ gives a negative magnetoresistivity, $\rho'_K(H, T)$ can have either sign. In particular, if $T, g\mu_B/k_B \ll T_K$ and $P(H_{\text{int}})$ vary slowly with field over the range $H_{\text{int}} \leq k_B T_K / g\mu_B = \Delta$, then Eq. (10) can be approximated by

$$\rho'_K(H, T) \sim \rho_K(0) \left[P(0) + \frac{\partial P}{\partial H_{\text{int}}} \Big|_{H_{\text{int}}=0} H \right] \Delta. \quad (11)$$

The magnetoresistivity varies linearly with H and its sign is determined by the rate of change of the internal-field distribution at the origin; a behavior similar to that observed in alloys with $x \leq 15$. However, to account for the absence of a temperature dependence of the forced magnetoresistivity (observed up to 50 K in $\text{Fe}_{75}\text{Cr}_3\text{B}_{20}$), T_K should be much larger than 50 K; an estimate that cannot be reconciled with the temperature dependence of the resistivity.

We suggested earlier that T_K must be less than approximately 5 K in alloys containing 20 at. % Cr or more. In this case both $P(H, T)$ and $\rho_K(H, T)$ should control the temperature and field dependence of ρ'_K . However, for $T \ll T_C$, we expect $P(H, T)$ to vary slowly with temperature compared with ρ_K , so that the magnitude of the Kondo magnetoresistivity must decrease with increasing temperature. This feature is in qualitative agreement with the behavior of the negative part to the magnetoresistivity, but quantitative comparison is not possible at present.

IV. CONCLUSION

We have separated and identified both structural and magnetic contributions to the temperature dependence of the resistivity of Fe-Cr-B metallic glasses. Not only have the contributions been identified, but a quantitative explanation has been given for the structural term and a qualitative one for the magnetic term. The structural contribution is consistent with quantum corrections to the resistivity due to electron-electron interactions. The magnetic contribution is attributed to Kondo effect and/or spin fluctuations, but further information on the electronic structure is needed before a full analysis is possible.

The magnetoresistivity is not in quantitative agreement with the above models. Specifically, no model can explain the linear and nearly-temperature-independent magnetoresistivity observed in alloys with $x \leq 15$. However, in alloys with $x \geq 20$, contributions from the Kondo effect and from quantum corrections, including the influence of spin fluctuations, might account for a significant part of the observed signal.

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