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# Electrical resistivity through the spin-glass regime of amorphous $Fe_xNi_{80-x}P_{14}B_6$

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The electrical resistivities $\rho$ of amorphous metallic spin-glass alloys ($Fe_xNi_{80-x}P_{14}B_6$) indicate that the relative importance of the collective versus individual aspects of the spin-freezing phenomenon are *not* the same in the transport properties of metallic amorphous systems as in crystalline spin-glasses. The observed negative magnetoresistance and the concentration dependence of the low-temperature behavior of $\rho$ are indicative of a contribution of magnetic origin which, however, cannot be adequately explained on the basis of current theories.

Studies on spin-glasses have continued to be of growing interest from theoretical as well as experimental points of view. While the precise nature of the spin-freezing transition still remains a mystery, the general features of the manifestations of this "phase transition"[1] in the static and dynamic properties of crystalline metallic spin-glasses are well known. In particular, the magnetic contributions to the electrical resistivity in the spin-glass regime of metallic crystalline alloys have been fairly well explained theoretically,[2] and confirmed experimentally,[3] in terms of a competition between the RKKY (Ruderman-Kittel-Kasuya-Yosida) and "Kondo" mechanisms. Studies of the role of the RKKY interaction in the collective freezing phenomena are therefore of particular relevance.[4] One approach to this question, from the transport property point of view, would be to study the effect of attenuating the RKKY interaction according to the exponential law $e^{-(r/L)}$, where $L$ is the electron mean free path, as suggested by de Gennes.[5] A direct test can be achieved by studying amorphous metallic dilute alloys where the mean free path $L$ is of order of interatomic distances as deduced from free-electron-theory considerations. Furthermore, studies on rapidly quenched amorphous ribbons minimize the usual metallurgical complications such as segregation and the crystal defects often encountered in the production of crystalline alloys. In addition, such studies provide further insight into the origin of the low-temperature resistivity anomalies observed in amorphous transition-metal–metalloid systems. In this paper we present the first systematic study of the electrical resistance and the magnetoresistance through the spin-glass regime in amorphous $Fe_xNi_{1-x}P_{14}B_6$ alloys. These studies, along with earlier ac-susceptibility work on the same system,[6] show that the resistivity behavior in amorphous metallic alloys in the spin-glass regime is completely different from that observed for crystalline metallic spin-glasses. In particular, the resistivity in amorphous alloys exhibits a simple "Kondo-like" behavior even at temperatures two orders of magnitude lower than the spin-freezing temperatures, which is in contrast to the *positive* temperature dependence observed in crystalline metallic spin-glasses. These results indicate that the relative importance of the interaction between spins, versus that between the individual spins and conduction electrons, are different in metallic amorphous spin-glass systems at least as far as the transport properties are concerned. The observed *negative* magnetoresistance, together with the compositional dependence of the resistance as described below, cannot be explained by present theories based on electron interactions of nonmagnetic origin. A microscopic description of the magnetic origin of these anomalies is not yet available.

Samples in the present studies were cut from ribbons of the alloy system $Fe_xNi_{1-x}P_{14}B_6$ in which earlier studies[6] indicated a spin-glass regime for $x < 5$. These amorphous ribbons with $x = 0$, 1, 2, 3, 4, 5, 7, 13, 20, and 27 were prepared by the melt-spinning technique.[7] The ribbons were typically ~1 mm wide and ~30 $\mu$m thick. Resistance measurements in the temperature range 0.1–5 K were carried out in a dilution refrigerator using an ac four-probe technique. Studies at higher temperatures, to 300 K, were mostly carried out using a current-comparator bridge. Magnetoresistance studies were made in fields $B \leq 0.56$ T. The relative precision of the resistivity data is typically 1–10 ppm, while temperatures were determined to within a few millidegrees Kelvin.

The electrical resistivities of all the alloys exhibited a minimum at low temperatures similar to what has been observed in many other amorphous metal-metalloid systems.[8] The resistivity behavior of the $x = 5$ alloy near the spin-glass regime is shown in Fig. 1. The rise in the resistivity is found to increase continuously down to 0.1 K although in the figure only data above ~1 K are shown. Also shown are the ac-magnetic-susceptibility data for this same alloy as measured by Onn *et al.*,[6] in which the spin-freezing temperature $T_f$ was found to be near 9 K. There is no noticeable feature in the resistivity which can be associated with $T_f$, and this is true for all the other samples having $x < 5$. The lack of any sharp anomaly at $T_f$ itself is not surprising and can be understood in terms of the scaling laws along with known critical exponents.[9] It must be pointed out that such a resistivity behavior in the spin-glass regime has been observed before,[10] and thus this behavior is not confined to the present alloy system alone. This temperature dependence of the resistivity below $T_f$ is quite different than that observed in crystalline spin-glasses, for which the resistivity exhibits a broad maximum above $T_f$ and a *positive* temperature dependence below $T_f$.[11] The question arises as to whether the negative temperature dependence of the resistivity in the present alloys is, in fact, associated with the magnetic state of the sample, or whether it is caused by a nonmagnetic electron scattering mechanism as suggested by previous authors.[12]

Available evidence indicates that the negative temperature dependence of the resistivity in these alloys is indeed of magnetic origin. Figure 2 shows the temperature dependence of the normalized resistivity, $[R(T) - R_{\min}]/R_{\min}$, as a function of the reduced temperature, $T/T_{\min}$, for four of the samples studied. As is further illustrated in Fig. 3, the magnitude of the "rise in resistivity" at low temperatures is strongly dependent on the Fe concentration and has a broad maximum around $x = 5$, the border between spin-glass and mictomagnetic behavior. Similar information is conveyed by the concentration dependence of $T_{\min}$, which also exhibits a maximum around $x = 5$. It would be difficult to explain these compositional dependencies using a model of nonmagnetic electron scattering. Figure 4 shows the observed magnetoresistance $[R(H) - R(0)]/R(0)$, again for the sample $x = 5$ alloy. The magnetoresistance is *negative* and of the same order of magnitude as observed in comparable crystalline alloys. Furthermore, the magnetoresistance is reproducible and independent of

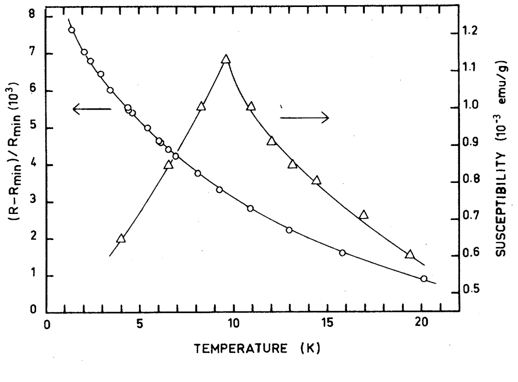

FIG. 1. Typical temperature dependence of the ac susceptibility (from Ref. 3) and the resistivity of amorphous $Fe_xNi_{80-x}P_{14}B_6$ alloys in the spin-glass regime, shown here for the alloy with $x = 5$. The resistivity continues to increase with decreasing $T$ to at least 0.1 K.

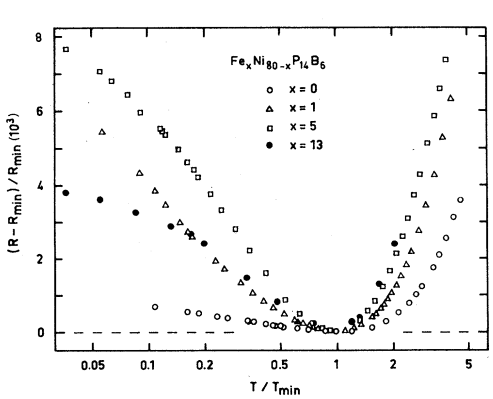

FIG. 2. Temperature dependence of the electrical resistivity (normalized to $R_{\min}$ at $T_{\min}$) of four of the amorphous $Fe_xNi_{80-x}P_{14}B_6$ alloys studied.

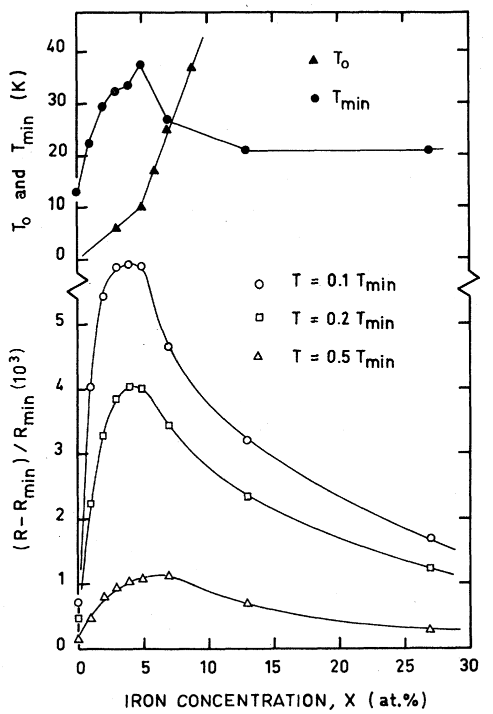

FIG. 3. The rise in the resistivity $[R(T)-R_{\text{min}}]/R_{\text{min}}$, plotted at various values of $T/T_{\text{min}}$, as a function of concentration for amorphous $\text{Fe}_x\text{Ni}_{80-x}\text{P}_{14}\text{B}_6$ alloys. In the upper part of the figure are shown temperatures at which the resistance minimum occurs. Also shown are the spin-glass or mictomagnetic transition temperatures, $T_0$, for these alloys as taken from Ref. 3.

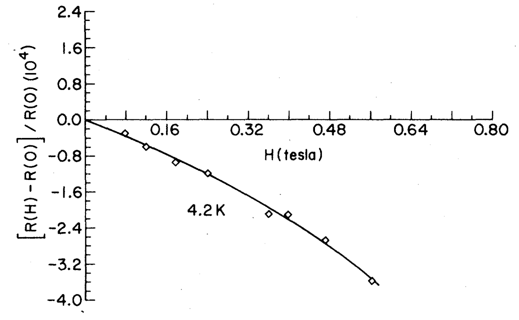

FIG. 4. Field dependence of the magnetoresistance for an amorphous $\text{Fe}_5\text{Ni}_{75}\text{P}_{14}\text{B}_6$ alloy well below its spin-freezing temperature of $\simeq 9$ K.

the orientation of the applied field relative to the current.[13] At temperatures well above $T_f$ the magnetoresistance was zero within experimental accuracy. A similar field dependence for the resistance is observed for other samples having $x < 5$, and for temperatures lower than $T_f$. Again, this behavior strongly suggests a magnetic origin to the resistivity rise at low temperatures.

Among the mechanisms introduced to account for the resistivity anomaly at low temperatures in amorphous alloys, two models have been widely discussed. One model, mentioned above, employs electron scattering from the localized two-level excitations in amorphous materials.[12] This model cannot explain the compositional and magnetic field dependence observed in the present alloys. The second model is based on spin fluctuations at the weak exchange-field sites that occur because of the randomness of the structure.[14,15] This mechanism, which is similar to that responsible for the Kondo phenomena, would appear to be applicable to the present results. However, the magnitude of the susceptibility below $T_f$ is found to be a minimum around $x=5$, while the resistivity rise (Fig. 3) is a maximum. Furthermore, the coefficient of the linear term in temperature for the magnetic contribution to the specific heat is found to continuously decrease as the value of $x$ is increased.[16] Hence, the density of weak-field sites would appear to require a different $x$ dependence for different measured properties. In brief, while the resistance rise and other low-temperature features of these alloys are convincingly of magnetic origin in the spin-glass regime, a microscopic explanation of these phenomena appears not to be available.

In a qualitative sense, two basic energy scales have been invoked in describing the low-temperature behavior of metallic crystalline spin-glasses,[2] namely, the rms RKKY interaction strength, $\Delta_c$, between the spins, and the Kondo interaction between conduction electrons and individual spins as characterized by $T_K$. The Kondo term causes the negative temperature dependence of the resistivity, while the maximum above $T_f$, followed by a positive temperature dependence at lower temperatures is produced by the decrease in spin-flip scattering associated with spin-freezing through the collective $\Delta_c$ interaction. Broadly speaking, the properties of a "spin-glass" or a "Kondo system" are therefore determined by the ratio $\Delta_c/T_K$. Clearly, the competition that is taking place between these mechanisms in metallic amorphous spin-glass, where the electron mean free path is not more than a couple of atomic distances, is of considerable interest and needs to be explored theoretically.

It may be useful to point out that the behavior of the electrical resistivity for $x > 5$, i.e., in the mictomagnetic regime, is rather similar to that below $x \simeq 5$. The resistivity has the same general temperature dependence, lacking any distinct features associated with the magnetic transitions. Also, the magnetoresistance is negative below the magnetic transition temperatures. Only the rather subtle behavior depicted in Fig. 3 separates the spin-glass regime insofar as the electrical resistivity is concerned.

In summary, we find the electrical resistivity of amorphous $Fe_xNi_{80-x}P_{14}B_6$ alloys behaves quite differently in the spin-glass regime than do crystalline spin-glasses, yet the pertinent electron scattering mechanisms are of magnetic origin. As far as we know this is the only physical property that behaves differently between the amorphous and crystalline states of metallic spin-glasses. No theory adequately accounts for the low-temperature magnetic behavior of these glassy alloy systems.

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