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Electron transport in Tb- and Pr-based metallic glasses

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Electrical resistivity measurements are reported on several metallic glasses based on Pr and Tb, and Ga and various transition metals as the glass formers. In general negative temperature coefficients were observed and these are discussed in terms of the extended Ziman theory and the tunneling or localization theory. Low temperature structure in the resistivity can be understood with the coherent exchange scattering model of Asomoza *et al.*

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INTRODUCTION

One of the major puzzles in the physics of disordered solids is the anomalous resistivity behavior exhibited by many amorphous and crystalline alloys with resistivity values $\rho \gtrsim 150\mu\Omega$cm. The temperature coefficient of resistance ($\alpha = \rho^{-1}d\rho/dT$) tends to be negative for $\rho \gtrsim 150\mu\Omega$cm, and the mechanisms leading to the observed temperature dependence of $\rho(T)$ have been controversial. Briefly, the major approaches have included: (1) The extended Ziman theory [1], which is based on the temperature dependence of the structure factor $S(q,T)$; (2) The modified Kondo model which is applicable to ferromagnetically ordered metallic glasses which have some spins in essentially zero effective fields [2]; (3) The atomic tunneling model [3] in which it is assumed that conduction electrons can scatter from 'two-level systems' in analogy with the ordinary Kondo spin-flip scattering process, thus leading to a $-\ell nT$ temperature dependence of $\rho(T)$. Tsuei [4] generalized this model to include conduction electron scattering from any localized internal degree of freedom, such as phonons or magnons, which could give rise to a broad range of characteristic energies; and (4) Incipient Anderson localization in which, in highly resistive metals, phonons assist the mobility [5]. Several of these theories are discussed critically in review articles by Cote and Meisel [1] and Black [6].

In this paper we report the results of a study of the electrical resistivity of several glasses based on Pr and Tb. Specifically the systems studied are of the form $(Pr_{80}Ga_{20})_{100-x}Fe_x$ where x = 0, 10, 20, 30; $(Pr_{80}Ga_{20})_{80}T_{20}$ where T = Cr, Mn, Fe, Co, Ni, Cu, and Ga; $(Tb_{80}Ga_{20})_{90}B_{10}$; and $(Tb_{80}Ga_{20})_{100-x}Fe_x$ where x = 10, 20, 30. These rare-earth glasses are of interest because of their anomalous electrical properties, speromagnetic phase transitions, and large coercive forces as reported elsewhere [7].

EXPERIMENTAL DETAILS

The amorphous alloys were prepared by splat cooling molten alloys in a modified arc-melting apparatus. Each sample was examined with MoKα x-ray diffraction. No evidence for crystalline phases was discovered with a high-resolution Si:Li detector. The samples had dimensions typically of 1 mm x 15 mm and the four electrical leads were attached either by spot welding to 'tabs' protruding from the samples or by gluing with silver paint to printed-circuit-board leads.

RESULTS

The results of measurements of $\rho(T)$ on Tb-based glasses are shown in Fig. 1. All of the glasses exhibit negative α's and the total change in resistivity ratio, $\rho(T)/\rho(300\text{ K})$ is less than 10%. There is little, if any, structure observed at the speromagnetic ordering temperatures, T_0, which are determined from sharp peaks in a.c. susceptibility measurements [7].

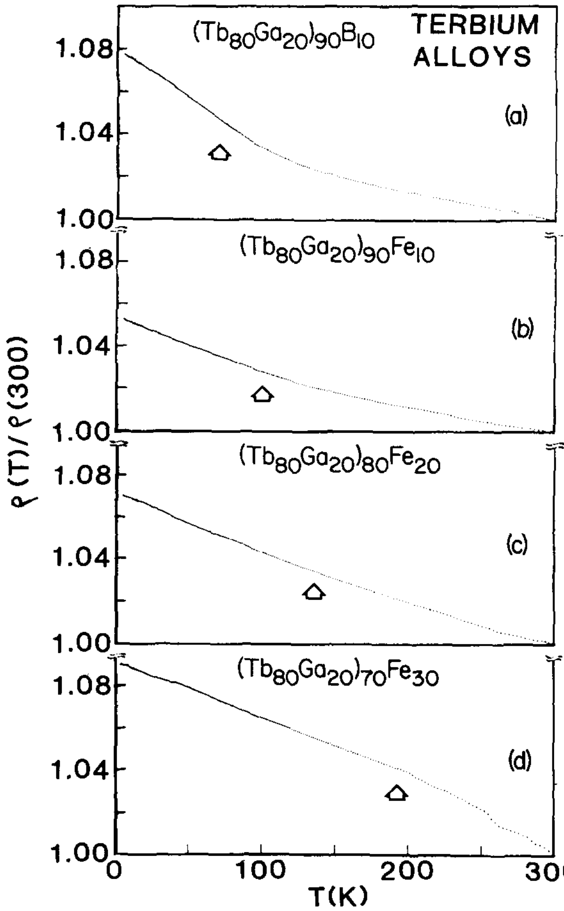

Fig. 1. Resistivity ratios for Tb-based glasses. Arrows indicate magnetic ordering temperatures.

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Figure 2 exhibits the resistivity behavior of Pr-based glasses containing Ga and Ga with Cr. Especially to be noted is that Cr leads to a positive α and saturation at high temperatures, and at $T \approx 30$ K there is a smooth resistance minimum. The other Pr-based glasses all have $\alpha < 0$ at high temperatures. The Pr glasses containing Ga only (Fig. 2) and Ga and B exhibit sharp upturns at speromagnetic ordering temperatures indicated by arrows. On the other hand the glasses containing Fe and Cr do not show sharp structure at T_0.

As a further test of the effect of the transition metal on the behavior of $\rho(T)$, measurements were made on Pr-based glasses containing the 3d elements from Mn to Cu, as shown in Fig. 3. There is little qualitative difference in $\rho(T)$ among these samples so the presence or absence of a local moment on the 3d element does not crucially affect $\rho(T)$. Table 1 summarizes some properties of selected glasses.

DISCUSSION

Except for the glass containing Cr, all the samples exhibited negative α's. Since the extended Ziman theory has been employed successfully to rationalize $\rho(T)$ for rare-earth liquid metals [8], it is natural to ask whether it also is consistent with our high-temperature results on rare-earth-based glasses. In the simplest form of the theory [1]

$$\rho(T) \propto 1 + S(2k_F, T), \qquad (1)$$

where it is assumed that the Fermi wavevector is given by $k_F = q_p/2$, where q_p is the position of the first peak in the structure factor. Thus the temperature dependence of $\rho(T)$ follows that of $S(q_p,T)$ if

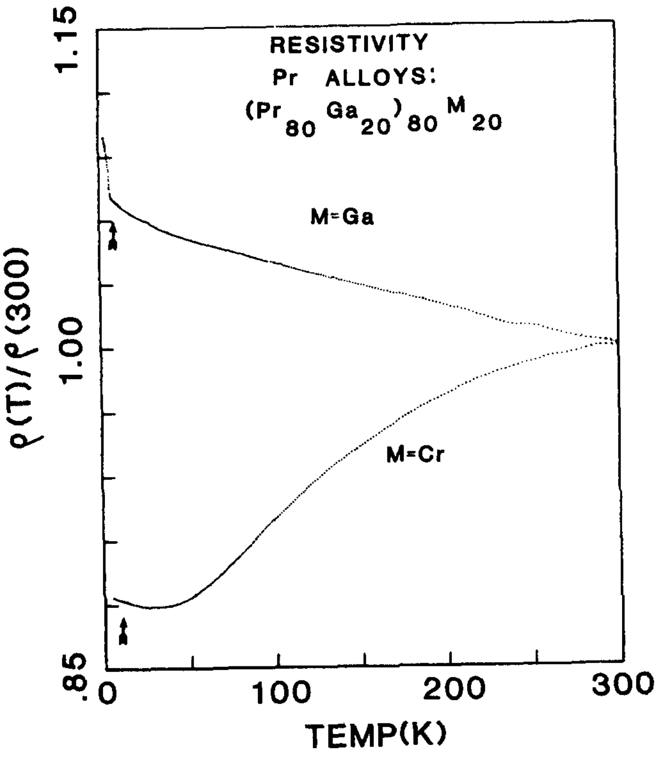

Fig. 2. Resistivity ratios for Pr-based glasses. Arrows indicate magnetic ordering temperatures.

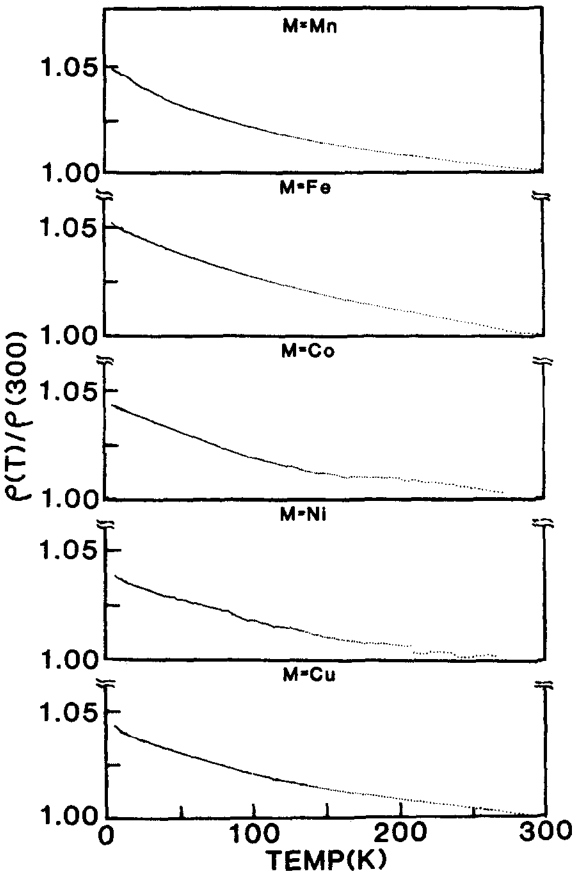

Fig. 3. Resistivity ratios for $(Pr_{80}Ga_{20})_{80}M_{20}$ for glasses with M = Mn, Fe, Co, Ni and Cu.

TABLE 1

Properties of Selected Metallic Glasses

Composition	q_p (Å^{-1})	$2k_F$ (Å^{-1})	$\rho(300)$ μΩcm (±10%)	T_0(K)
$Pr_{80}Ga_{20}$	2.24	2.18	213	11
$(Pr_{80}Ga_{20})_{90}Fe_{10}$	2.25	2.27	230	13
$(Pr_{80}Ga_{20})_{80}Fe_{20}$	2.32	2.29	222	12
$(Pr_{80}Ga_{20})_{70}Fe_{30}$	2.28	2.33		10
$(Pr_{80}Ga_{20})_{80}Cr_{20}$	2.26		183	9.5
$(Pr_{80}Ga_{20})_{80}Mn_{20}$	2.28	2.33	216	21
$(Pr_{80}Ga_{20})_{80}Co_{20}$	2.30	2.25	229	13
$(Pr_{80}Ga_{20})_{80}Ni_{20}$	2.35	2.25	220	6.2
$(Pr_{80}Ga_{20})_{80}Cu_{20}$	2.29	2.27	196	8
$(Tb_{80}Ga_{20})_{90}B_{10}$	2.29	2.44	277	69
$(Tb_{80}Ga_{20})_{90}Fe_{10}$	2.31	2.33		99
$(Tb_{80}Ga_{20})_{80}Fe_{20}$	2.35	2.35	269	139
$(Tb_{80}Ga_{20})_{70}Fe_{30}$	2.39	2.44		∿200

$2k_F$ matches q_p. Table 1 shows the comparison of $2k_F$ and the experimentally measured q_p; here it was assumed that Fe, Ni, and Co contributed 1.2, 1.0, and 1.0 electrons to the Fermi sphere, respectively [9], Ga and B contributed 3 electrons, and Pr and Tb contributed 1 and 1.3 electrons, respectively [8]. No value of $2k_F$ is given for the Cr glass because the effective valence, Z, for Cr is not known. If one assumes $Z = 0$, then $2k_F = 2.0$ Å^{-1} which is about 10% different from q_p. With this one possible exception, however, the Table shows that q_p and $2k_F$ are generally very close to each other, which suggests that the extended Ziman theory is applicable to these glasses. On the other hand, for $T > \theta$, where θ is the Debye temperature, S(q,T) should decrease linearly in T. The data show that this generally is not the case, thus casting some doubt on the relevance of the Ziman theory to these glasses.

As an alternative approach we have fitted several of our $\rho(T)/\rho(300\ K) \equiv r(T)$ curves to the expression based on tunneling [3] or localization [4]:

$$r(T) = r_0 - c\ \ell n\ (T^2 + \Delta^2), \qquad (2)$$

where Δ is the energy characteric of the localized internal degree of freedom. Fig. 4 shows two examples of such fits; the parameters are: $r_0 = 1.106$, $c = 0.009$, $\Delta = 32.9$ K for the Pr-Cu glass; and $r_0 = 1.193$, $c = 0.017$, and $\Delta = 30.2$ K for the Tb-B glass. A similar fit was obtained for $(Pr_{80}Ga_{20})_{80}Fe_{20}$ with $r_0 = 1.128$, $c = 0.011$, and $\Delta = 35.4$ K. We also have found Eq. (2) to fit r(T) well for several other glasses of the form $(R_{80}Ga_{20})_{80}T_{20}$ where R = La, Nd, Er, and T = Fe.

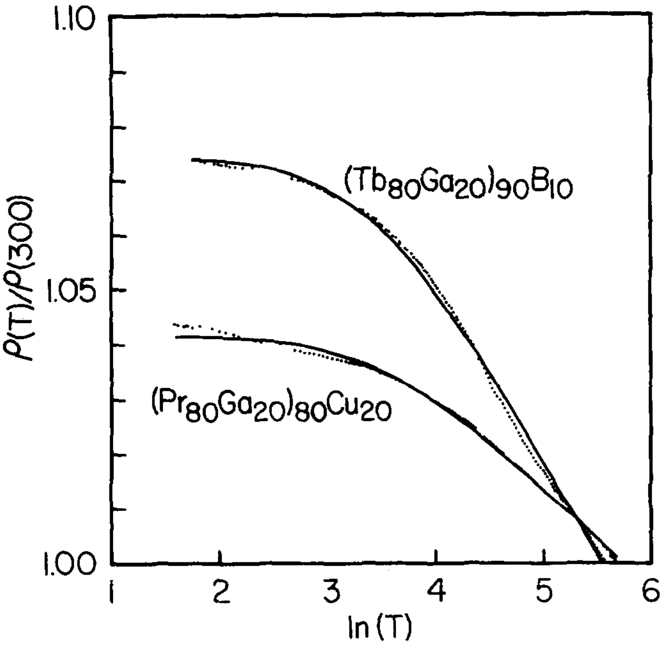

Fig. 4. Fits to the tunneling/localization expression. The deviation of the fit from the data at low T for the Pr glass may be due to magnetic ordering.

Mueller, et al. [10] have obtained excellent fits of their data on a-$La_{66}Al_{34}$ to Eq. (2). Given a wide variety of glass resistivities which are represented by Eq. (2), it is clear that the $-\ell nT$ behavior is not a Kondo effect due to local moments on the Pr or Tb atoms, nor on the Fe atoms. The ubiquitous character of $\rho(T)$ behavior as expressed by Eq. (2) suggests that the idea of incipient localization or scattering from localized excitations may have merit. But at present it is difficult to confirm definitively the origin of negative α's in these glasses.

The sharp low-temperature upturns in $\rho(T)$ for the Pr glasses without 3d elements (Fe or Cr) in them are similar to those we have observed previously in Er-based glasses [11]. These data can be understood in terms of the theory of Asomoza et al. [12], who derived sharp structure in $\rho(T)$ in metallic glasses at magnetic ordering temperatures. A detailed comparison of our results with this theory will be presented elsewhere [13]. The low temperature upturn in $\rho(T)$ seen in the Pr-Ga-Cr glass is likely due to the modified Kondo mechanism as discussed by Grest and Nagel [2].

In summary, the sign of α in the present resistivity results is consistent with the extended Ziman theory if reasonable conduction electron contributions are assumed. However $\rho(T)$ is not linear in T for $T > \theta$ as required by the theory. The data were shown to fit the tunneling or localization model fairly well. Evidence was presented on the effect of magnetic ordering on $\rho(T)$ and this can be understood with the coherent exchange model.

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