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THERMOELECTRIC POWER OF METALLIC GLASSES

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Abstract.- The thermoelectric power of a number of magnetic and nonmagnetic metallic glasses has been measured as a function of temperature between 15 K and 580 K. The temperature dependence of the thermopower for the magnetic alloys is qualitatively different from that for the nonmagnetic alloys. The data for the nonmagnetic alloys are consistent with the extended Ziman Theory for transport in liquid and amorphous metals and show positive values for thermopower, Q , when the temperature coefficient of resistivity, α , is negative and negative values for Q when α is positive. The data for the magnetic alloys are consistent with a model based on the Kondo scattering of conduction electrons by spins situated in low effective fields.

In this paper we report the results of a series of studies on the thermoelectric power of metallic glasses. These alloys can be divided into two categories: those that contain no magnetic elements and those that are ferromagnetic. We have found that these two classes of materials have distinctly different thermoelectric behavior. These differences seem to be related to the differences which have been observed in the resistivity of these glasses. In both cases the resistivity is anomalous in that the temperature coefficient of resistivity is negative

$$(\alpha \equiv \frac{1}{\rho} \frac{d\rho}{dT} < 0) \text{ over some range of temperature.}$$

However in the nonmagnetic glasses the magnitude of α decreases as the temperature is lowered to zero and α is negative over a wide range of temperature. In the ferromagnets, on the other hand, the low temperature resistivity increases approximately logarithmically as the temperature is decreased below a temperature T_{\min} where the resistivity has a minimum value. This behavior is reminiscent of the Kondo effect in crystalline alloys where the magnetic component is very dilute.

There are several theories which have been proposed to explain these resistivity anomalies.

Although they have all been able to produce a negative value for α for certain sets of parameters in the theory, and therefore fit the resistivity data, they make different predictions for the thermoelectric power. We have undertaken the present studies in order to distinguish which of the presently available theories are most applicable for describing the transport properties of metallic glasses.

One theory that has been extensively used to interpret the resistivity data of the non-magnetic glasses is the generalized Ziman theory^{1, 2} which deals with the normal potential scattering of the electrons by the disordered solid and can lead to a positive or negative value of α depending on the relative positions of $2k_F$ and k_D where k_F is the Fermi wavevector and k_D is the position of first peak in the structure factor (or one of the partial structure factors) of the glass. An alternative theory^{3, 4} proposes the existence of quantum-mechanical two-level tunneling states for some of the atoms in a disordered solid. The scattering of the conduction electrons from these states is similar to that found in the Kondo effect and may increase the resistivity dramatically at low temperature. A recent paper⁵ has resurrected the

idea of a real Kondo effect in explaining the resistivity minima in the magnetic glasses. By calculating the effective field distributions, $P(H)$, that may exist in a concentrated ferromagnetic glass, it was found that a small number of spins may still exist in essentially zero effective field. These spins may then give rise to Kondo spin-flip scattering and thus reconcile the coexistence of a Kondo effect with ferromagnetism.

The thermoelectric power predicted by these three theories will be different.⁶ The Ziman Theory predicts a thermopower, Q , which is small, linear in temperature and negative unless $k_D \sim 2k_F$ in which case it is positive. This condition is just that required by the theory in order to produce a negative value of α . Thus a negative α is accompanied by a positive Q . The other two theories, the scattering by tunneling-levels and by magnetic spins both give rise to very large anomalies--orders of magnitude larger than the Ziman theory would predict for a simple metal.

In Fig. 1 we show the thermoelectric power for several nonmagnetic alloys which have negative values of α . The data on $\text{Be}_{0.4}\text{Ti}_{0.5}\text{Zr}_{0.1}$ (Metglas 2204 of Allied Chemical Corp.) was reported by one of us earlier⁶ and subsequent measurements have confirmed these results.⁷ The $\text{Cu}_{0.5}\text{Zr}_{0.5}$ data has also been recently reported.⁸ The measurements that we show here on the $\text{Nb}_{0.5}\text{Ni}_{0.5}$ sample are consistent with the other data shown in that figure. All three curves are linear in T over a very wide range of temperatures and they all have a positive slope and a positive value of Q . This is completely consistent with the simple Ziman theory and inconsistent with the predictions of the Kondo or tunneling level models. In Fig. 2 we show the behavior of Q for another nonmagnetic glass: $\text{Pd}_{0.775}\text{Cu}_{0.06}\text{Si}_{0.165}$. In this case Q has a

a slightly different behavior from that shown for the alloys in Fig. 1. For this glass Q is predominantly negative and has a negative slope. In this glass, however, the resistivity behaves normally and has a positive value of α . Therefore the Ziman theory would predict a negative slope for Q vs. T as we have found. This then is consistent with the Ziman theory as well and further confirms the prediction that in general a positive α implies a negative Q and that a negative α implies a positive Q . (This prediction however can fail near the region where $\alpha \sim 0$ or $Q \sim 0$ since the region of positive Q depends on $S(2k_F)|v(2k_F)|^2$ being much larger than the average, up to the Fermi surface, of $S(q)|v(q)|^2$ and the region of negative α depends on when $\int_0^{2k_F} q^3 |v(q)|^2 S(q) dq$ decreases with increasing temperature). The slight curvature seen in Q may be due to other mechanisms becoming important aside from those calculated in the Ziman theory. These may be more apparent in this case than in those in Fig. 1 because the resistivity of $\text{Pd}_{0.775}\text{Cu}_{0.06}\text{Si}_{0.165}$ is more than a factor of two less than that of the other alloys studied.

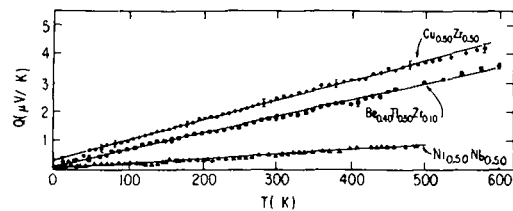


Fig. 1.--Thermoelectric power of the nonmagnetic amorphous alloys $\text{Be}_{0.4}\text{Ti}_{0.5}\text{Zr}_{0.1}$ (Metglas 2204) and $\text{Cu}_{0.5}\text{Zr}_{0.5}$ and $\text{Nb}_{0.5}\text{Ni}_{0.5}$.

In Figs. 3 and 4 we show the thermoelectric power of several magnetic metallic glasses.⁸ We see immediately that they have different behavior from that seen for the nonmagnetic samples in that they have considerable structure in their temperature dependence. This is consistent with the theory of Grest and Nagel⁵ based on magnetic Kondo

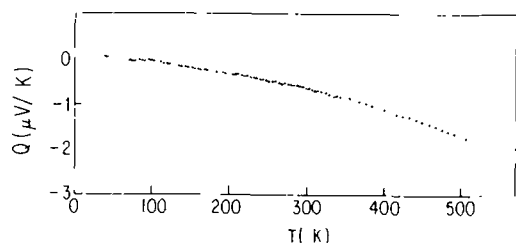


Fig. 2.--Thermoelectric power of the nonmagnetic alloy $\text{Pd}_{0.775}\text{Cu}_{0.06}\text{Si}_{0.165}$.

scattering. The reason that the structures observed in Figs. 3 and 4 are small compared to what is seen in crystalline Kondo systems (i.e., peaks in the thermoelectric power as great as $40 \mu\text{V}/\text{K}$) is that the thermoelectric power of the Kondo effects is weighted, using the Nordheim-Gorter rule, by its contribution to the total resistivity which is small. The presence of Cr in the Fe Ni Cr P B alloy shown in Fig. 3 seems to make the structure observed in Q disappear. This may be due to the Cr contributing a Kondo effect of its own with opposite sign to that of Fe, as is sometimes seen in crystals. Therefore the Cr tends to cancel the Kondo effect seen in the sample without Cr. A more detailed analysis of this data is described elsewhere.⁸ For the present purposes it is sufficient to note that the behavior of the magnetic samples is different from that of the nonmagnetic ones and that the structure seen in the data is sensitive to the magnetic elements contained in the sample.

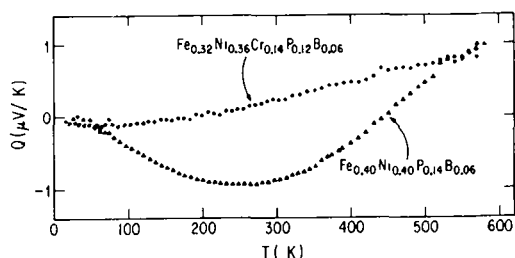


Fig. 3.--Thermoelectric power of the amorphous magnetic alloys $\text{Fe}_{0.40}\text{Ni}_{0.40}\text{P}_{0.14}\text{B}_{0.06}$ (Metglas 2826) and $\text{Fe}_{0.32}\text{Ni}_{0.36}\text{Cr}_{0.14}\text{P}_{0.12}\text{B}_{0.06}$ (Metglas 2826A).

In conclusion we have shown that the thermoelectric power has different behavior in magnetic than in nonmagnetic metallic glass alloys. In the nonmagnetic glasses the behavior is very simple: Q is small and very linear in T . If α is negative then Q for the samples studied is positive and for the one sample we have reported where α is positive Q is negative. This is consistent with the Ziman theory for electronic transport and not consistent

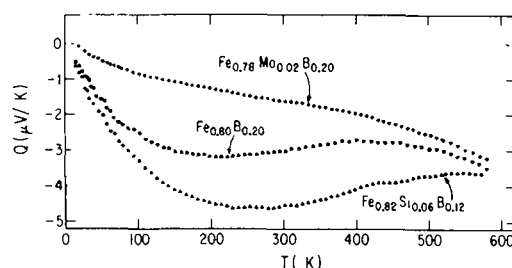


Fig. 4.--Thermoelectric power of the amorphous magnetic alloys $\text{Fe}_{0.8}\text{B}_{0.2}$ (Metglas 2605), $\text{Fe}_{0.78}\text{Mo}_{0.02}\text{B}_{0.20}$ (Metglas 2605A) and $\text{Fe}_{0.82}\text{Si}_{0.06}\text{B}_{0.12}$ (Metglas 2605S).

with the Kondo or tunneling level models. Moreover this is the first time that such simple, behavior has been found in a class of alloys. These are the only alloys for which the textbook predictions of a linear thermoelectric power seems to be completely valid. In the magnetic glasses we find that there is structure in the temperature dependence of Q . The tentative explanation is that this is due to a real Kondo effect caused by the conduction electrons scattering from the few spins that exist in the tail of the $P(H)$ distribution near zero effective field. This explanation would help account for the extreme sensitivity of the resistivity and thermoelectric power to the type of magnetic constituents in the sample.

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