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LETTER TO THE EDITOR

Thermopower studies of (Ce, U)NiSn

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Abstract. Data are presented for the effect of U substitutions for Ce on the thermopower of the so-called 'Kondo insulator' CeNiSn. These suggest that the gap that begins to open below 5 K is destroyed by 10% U, with the appearance of metallic Kondo character. This is in turn attenuated by further substitutions of U.

Since Kondo systems, both dilute alloys and Kondo lattices or heavy-fermion compounds, have small characteristic energy scales their thermoelectric powers are of particular interest, since thermopowers depend on the energy dependence of electron scattering. Where the Kondo effect, crystal-field splittings and magnetic interactions coexist the thermopower is almost too sensitive a probe, and theoretical treatments of Kondo lattices have only been successful [1] when alloying has removed the added complication of a temperature-dependent onset of coherence. Alloying can, however, significantly modify the ground state of such systems, as with the stabilization of antiferromagnetism in $CeCu_6$ by Au substitutions for Cu [2], and it is therefore of interest to examine the thermopower and its modification by alloying in low-carrier-density compounds, where hybridization of conduction electrons with f electrons of Ce or U has produced complete or partial gapping of the Fermi surface [3, 4].

CeNiSn is such a compound, although it seems likely that at low temperatures it is a low-carrier-density semi-metal rather than a semiconductor or the 'Kondo insulator'. The interaction of Kondo insulator behaviour with crystal-field excitations in this compound [5] and the possible role of impurity and non-stoichiometry [6] have recently been discussed theoretically. In previous work [7] we have shown that U substitutions for Ce rapidly modify the resistivity and by 20% substitution have stabilized antiferromagnetism. We now report the results of measurements of thermopower (S) in the same alloy samples. The measurements of the thermopower were made by a differential method relative to copper.

Since our samples were polycrystalline, it is interesting to compare our data for pure CeNiSn with those [8] for single-crystal samples of this material, which has an orthorhombic structure. Figure 1 shows three distinct maxima for pure CeNiSn: the lowest-temperature sharp peak (A) corresponds to that seen for the a- and c-axes data of [8], the highest-temperature rounded maximum (C) corresponds to a similar maximum for the b-axis data, and the maximum (B) at around 20 K to shoulders for the a- and b-axes data with a rounded maximum at a slightly lower temperature. The magnitudes of the thermopower at low and high temperatures, by comparison with the single-crystal data, suggest that some degree of preference exists for the a- and c-axes in our specimen.

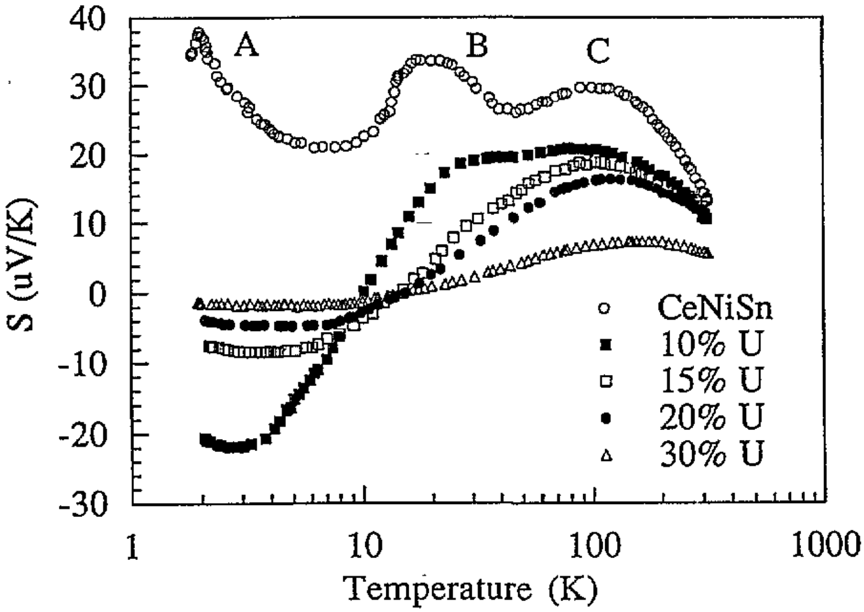

Figure 1. Semi-log plot of the thermopower for polycrstalline $(Ce_{1-x}U_x)NiSn$ with $x = 0, 0.1, 0.15, 0.2$ and 0.3.

The most striking feature of figure 1 is the complete suppression of the low-temperature peak with 10% U and the change to a large negative value in the liquid helium temperature regime. This is the regime in which the large quasi-semiconducting negative $d\rho/dT$ has been replaced (see figure 1 of [7]) by a $\rho(T)$ which has more Kondo character; this is also the region in which the electronic specific heat falls sharply. The behaviour at higher temperatures suggests that the intermediate maximum (B) of pure CeNiSn has moved upwards and broadened, thereby smearing out the 100 K maximum (C).

For larger concentrations the thermopower crosses zero at about 15 K, with steadily decreasing magnitudes with increasing U content at both higher and lower temperatures. The well defined peak in susceptibility at 6 K for the 20% U alloy which we ascribe to magnetic order is not accompanied by any significant effects in the thermopower, although there is a weak but well defined maximum in resistivity near that temperature.

The most probable interpretation of these results would be that quite small substitutions of U destroy the quasi-semiconducting character at the lowest temperature allowing a normal Kondo effect yielding a large negative thermopower to take over, as it does in many dilute alloys and in $CeCu_6$ when the coherence has been destroyed by Au substitutions for Cu [9]. Negative maxima in S have, however, been interpreted [10] in terms of a competition between Kondo compensation and RKKY interactions, but the absence of any change when magnetic order sets in in the 20% alloy seems to make this less likely. The steady reduction in thermopower as the U concentration increases suggests that non-resonant scattering is becoming dominant, but the persistence of a maximum (C) at 100 K suggests that the position of this is governed by crystal-field effects.

In summary, it seems that the transport properties of CeNiSn are only dominated by quasi-semiconducting character below about 5 K and this character is destroyed by about 10% U. The resultant Kondo character is then more slowly attenuated by increasing substitution. It will be interesting to study the low-temperature behaviours for smaller amounts of U and for other elements.

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