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Search for the Kondo effect of Pr ions doped in non-magnetic matrices

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Abstract. The results of electrical resistivity ρ measurements in the temperature interval 1.5–300 K on the intermetallic compounds YCu_2Si_2, YNi_2Si_2, YCu_2Ge_2, YNi_2Sn_2, YPd_2Si_2, YPt_2Si_2, YRu_2, YPt_2, YPd_3 and $ScAl_2$ containing small amounts of Pr as well as on $Sc_{0.95}Pr_{0.05}$ are reported in order to look for the Kondo behaviour of Pr ions. The logarithmic variation in ρ, however, is not apparent in the temperature range of investigation in any of the alloys, although these alloys are potential candidates for exhibiting Kondo behaviour.

One of the consequences of the proximity of the Ce 4f level to the Fermi level E_F is the observation of the Kondo effect in Ce-containing metals [1]. However, there is no intense activity to look for similar effects among Pr-based metals, barring a few successful claims about Pr-doped Pd metal [2], $Zr_{1-x}Pr_xB_{12}$ [3] and $PrSn_3$ [4], although it is now believed that there is a tendency for the Pr 4f orbital also to exhibit a certain degree of extended character. With this motivation, we present the results of our investigation on the electrical resistivity behaviour of Pr in the dilute limit (less than 20 at.% Pr) in several potentially good candidates (see below for details) of non-magnetic matrices to look for the Kondo effect.

The samples $Y_{1-x}Pr_xCu_2Si_2$ ($x = 0, 0.05, 0.2$), $Y_{1-x}Pr_xNi_2Si_2$ ($x = 0, 0.05, 0.1$), $Y_{1-x}Pr_xCu_2Ge_2$ ($x = 0, 0.05, 0.1$), $Y_{1-x}Pr_xPd_2Si_2$ ($x = 0, 0.05, 0.1$), $Y_{1-x}Pr_xPt_2Si_2$ ($x = 0, 0.05, 0.1$), $Y_{1-x}Pr_xNi_2Sn_2$ ($x = 0, 0.05, 0.1$), $Y_{1-x}Pr_xRu_2$ ($x = 0, 0.05, 0.1$), $Y_{1-x}Pr_xPt_2$ ($x = 0, 0.05, 0.1$), $Y_{1-x}Pr_xPd_3$ ($x = 0, 0.05, 0.1$), $Sc_{0.95}Pr_{0.05}Al_2$ and $Sc_{0.95}Pr_{0.05}$ were prepared by arc melting stoichiometric amounts of constituent elements. The pseudoternary alloys were homogenized at 800 °C in evacuated sealed quartz tubes for 1 week. Powder x-ray diffraction patterns suggest that all the ternary compounds form in the $BaAl_4$-type structure [5], although a few unidentified lines were noticed for tin-containing specimens. The pseudobinary samples form, as expected [6], in the Laves phase structures. Electrical resistivity, ρ, measurements were performed in the temperature interval 1.5–300 K by a conventional four-probe method, employing a nanovoltmeter.

The results of the electrical resistivity ρ measurements on all these specimens are shown in figures 1–3. The temperature-dependent resistivity behaviour of Pr-doped specimens follows the same trend as that of the corresponding non-magnetic analogue, although a difference in the slope of $d\rho/dt$ is noticed in some cases, possibly because of uncertainties in the measurements of sample dimensions. Clearly, the logarithmic variation in ρ with temperature is not apparent in any of the alloys,

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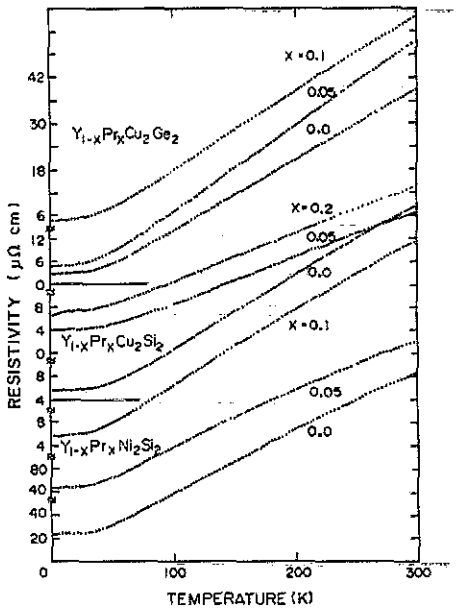

Figure 1. Electrical resistivity as a function of temperature (1.5–300 K) for the alloys $Y_{1-x}Pr_xCu_2Si_2$, $Y_{1-x}Pr_xNi_2Si_2$ and $Y_{1-x}Pr_xCu_2Ge_2$.

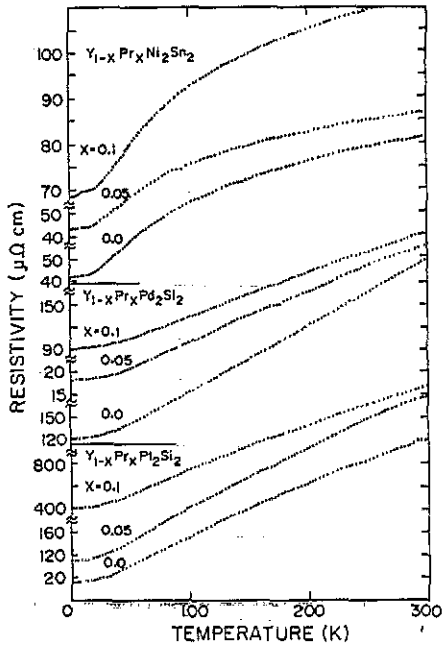

Figure 2. Electrical resistivity as a function of temperature (1.5–300 K) for the alloys $Y_{1-x}Pr_xPd_2Si_2$, $Y_{1-x}Pr_xPt_2Si_2$ and $Y_{1-x}Pr_xNi_2Sn_2$.

thereby suggesting that there is no evidence for the Kondo effect in these alloys.

We now put forward the logic behind looking for the Kondo effect in the above alloys. In order to choose suitable alloys for our purpose, we are guided by knowledge of the magnetic behaviour in the compounds of other rare earths such as Ce, Eu and Yb. The guidelines are as follows.

(i) The dilute limit of Pr compounds, for which the magnetic ordering temperatures T_N which are much higher than those expected on the basis of de Gennes scaling. It is well established for the Ce systems that both T_N and T_K (the Kondo temperature) increase with increasing 4f–conduction band coupling strength J [1] and hence an anomalously high T_N (breakdown of de Gennes scaling) naturally means the enhancement of the Kondo interaction. This has been found to be the case for several Ce systems [7, 8]. $PrCu_2Si_2$, $PrCu_2Ge_2$ and $PrNi_2Si_2$ belong to this class of compounds, as it is well known that the Pr sublattices order (about 21 K, about 15 K and about 20 K, respectively) antiferromagnetically at temperatures larger than known for the corresponding Gd analogue (about 15 K, about 12 K and about 14 K, respectively).

(ii) The dilute limit of the Pr compounds, the corresponding Ce, Eu and Yb analogues of which show strongly mixed-valence or Kondo behaviour. It is quite well known that Eu and Yb normally tend to be divalent and, in a chemical environment of strong electron affinity or positive chemical pressure, Eu and Yb acquire a fluctuating valence or even trivalent state. For instance, $EuPd_2Si_2$ [9] and $YbPd_2Si_2$ [10] are mixed-valence compounds and hence anomalies in $Y_{1-x}Pr_xPd_2Si_2$ series are expected. Although Eu in $EuAl_2$ is divalent, Eu doped in $ScAl_2$ has a strongly mixed valency

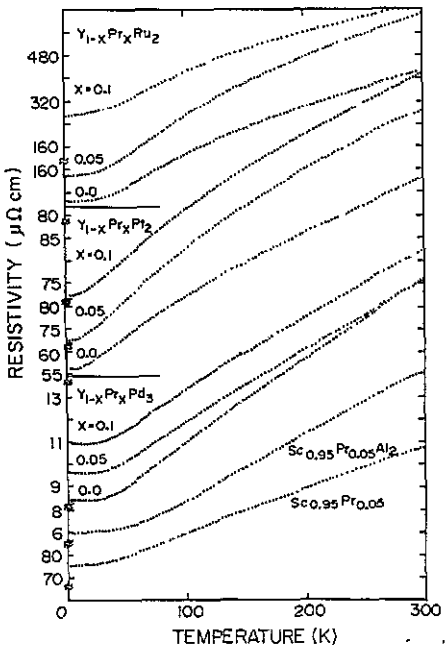

Figure 3. Electrical resistivity as a function of temperature (1.5–300 K) for the alloys $Y_{1-x}Pr_xRu_2$, $Y_{1-x}Pr_xPt_2$, $Y_{1-x}Pr_xPd_3$, $Sc_{0.95}Pr_{0.05}Al_2$ and $Sc_{0.95}Pr_{0.05}$.

nature [11] owing to a large positive chemical pressure; this explains the choice of $Sc_{0.95}Pr_{0.05}Al_2$; for the same reason, the study of $Sc_{0.95}Pr_{0.05}$ is undertaken, as Ce in $Sc_{0.95}Ce_{0.05}$ was shown to exhibit a mixed-valence-like behaviour [12].

(iii) The dilute limit of Pr compounds, in which valence band photoemission spectra show a peak near the Fermi level E_F in addition to the feature well below (about 2.5 eV) E_F, similar to that known for Ce-based Kondo lattices. Some groups attribute this Fermi level peak in Ce systems to Kondo resonance [13]. Binary Pr compounds [14] belong to this category and hence the Kondo effect is expected in $Y_{1-x}Pr_xRu_2$ and $Y_{1-x}Pr_xPd_3$. These alloys also obey the guideline (ii) as $CeRu_2$ and $CePd_3$ are well known to be strong mixed-valence compounds and Eu in $EuPd_3$ is trivalent [6]; thus there is a tendency for strong 4f–conduction band coupling in this chemical environment.

(iv) Pr alloys in which the valence orbitals of neighbouring ions in the lattice are spatially diffused so that these orbitals can undergo effective hybridization with the Pr 4f orbital. Thus Sn- and Pt-containing specimens may be suited to our purpose.

Thus, we have chosen a large set of dilute Pr alloys based on the above ideas to search for the Pr Kondo effect. To our surprise, the logarithmic variation in ρ due to the Kondo effect is not seen in the experimental data of any of these alloys. It may appear that the magnetic resistivity of $Y_{1-x}Pr_xCu_2Si_2$ and $Y_{1-x}Pr_xPd_2Si_2$ obtained after subtracting the lattice resistivity of the corresponding Y analogue decreases with increasing temperature. However, we do not wish to attach any significance to this observation, as there is a non-negligible error in the slope of the ρ against T plot for most of the alloys due to microcracks in the specimens. It is of interest to perform magnetoresistance and thermopower measurements to confirm the absence of the Kondo effect in the above alloys. If the presence of the Kondo effect is shown

by these future studies, it might be tempting to propose that the depopulation of the closely spaced crystal-field levels for Pr, in some cases to a singlet ground state, dampens the resistivity increase with decreasing temperature. If the absence of Kondo effect is conclusively established, then it may imply that the anomalous magnetism in cases like $PrCu_2Si_2$ is orbitally driven [15] and not spin driven.

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