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f-BAND NARROWING IN URANIUM INTERMETALLICS[†]

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Although the discovery of heavy fermion behavior in uranium compounds has attracted a great deal of attention, relatively little work has been done which is sufficiently systematic to allow an assessment of the relationship of such behavior to more common phenomena, such as mixed valence, narrow-band effects, etc. In this paper we report bulk property measurements for a number of alloys which form a part of such a systematic study. The approach has been to take relatively simple and well-understood materials and alter their behavior by alloying to produce heavy fermion or Kondo behavior in a controlled way.

Extensive studies have shown that the f electrons in URh₃ are band-like. In particular, detailed de Haas - van Alphen studies have provided Fermi surface data which are in very good agreement with band-structure calculations.¹ In analogy with previous work² on the compounds CePd₃B_x, we have prepared and investigated the alloy system URh₃B_x. X-ray studies show that the metal ion lattice retains the AuCu₃ structure with an increase in lattice

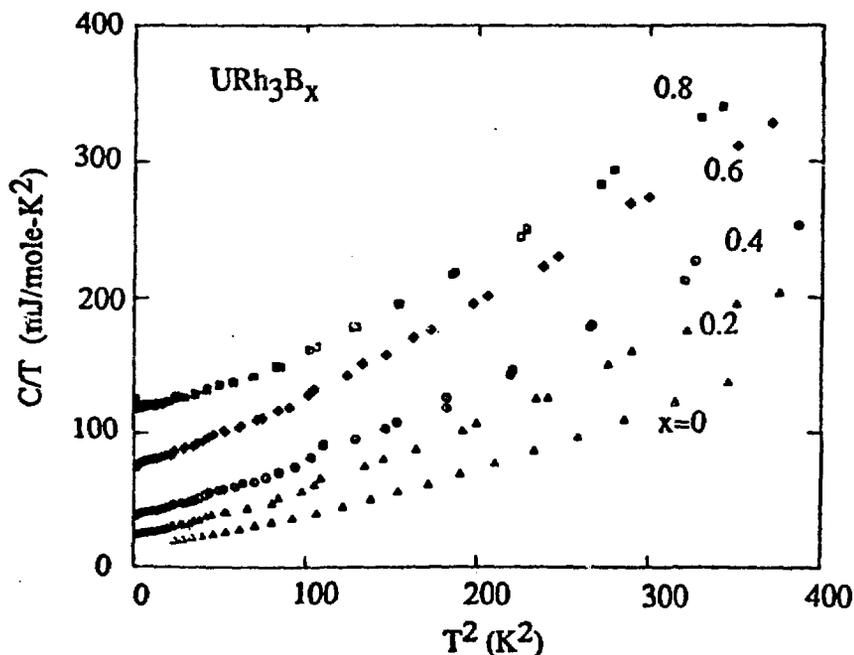


Figure 1. Heat capacity data for URh₃B_x alloys.

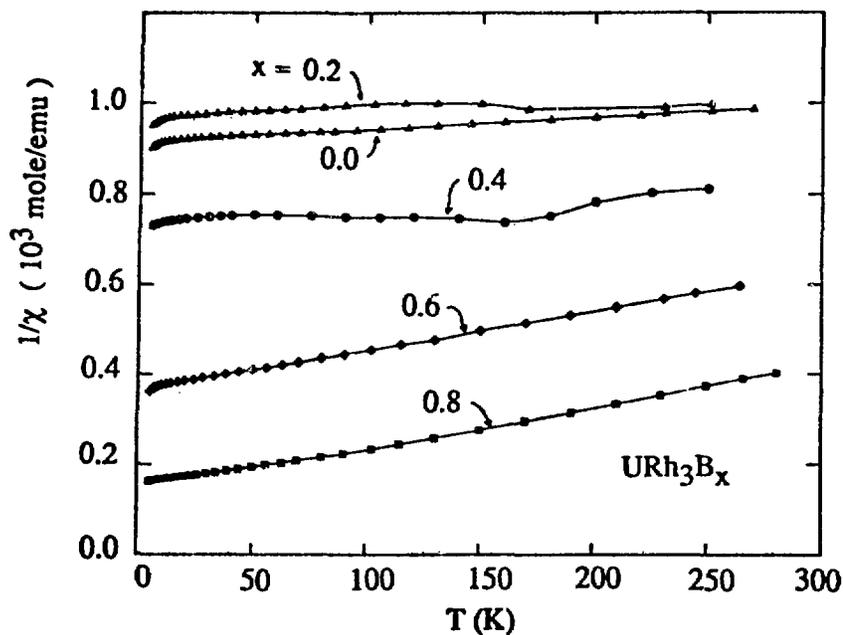


Figure 2. Inverse magnetic susceptibility data for URh_3B_x alloys. The solid lines do not represent theoretical fits to data.

constant up to $x=0.8$, and a neutron diffraction measurement³ has shown that B sits in the interstitial body-centered position of this lattice. Both the expansion of the lattice as x increases and the bonding of B to Rh ions should result in a dehybridization of the U f -electrons, and a consequent narrowing of the hybridization width. Heat capacity and susceptibility data are shown for these alloys in Figs. 1 and 2. The addition of B causes a substantial increase in the electronic density of states, as evidenced by the increase in the electronic heat capacity coefficient from $\gamma=14.4$ mJ/mole- K^2 for $x=0$ to 119 mJ/mole- K^2 for $x=0.8$. Further evidence for strong band-narrowing effects is seen in the development from a temperature independent susceptibility for $x=0$ to a well-defined Curie-Weiss behavior for large x values. Bandstructure calculations⁴ for URh_3 and URh_3B explicitly

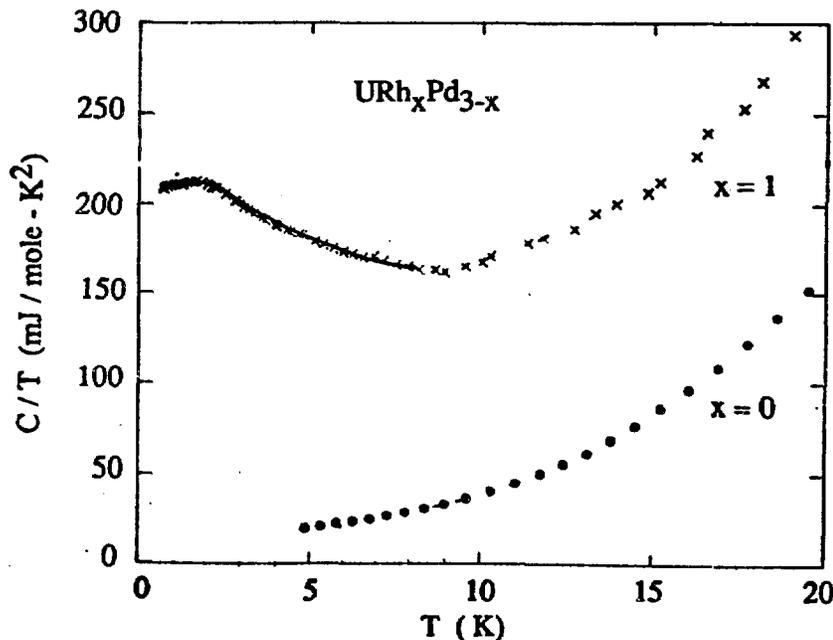


Figure 3. Heat capacity data for URh_xPd_{3-x} alloys. The solid line for $URh_{1.0}Pd_{2.0}$ is given by $C/T = \gamma + AT^2 + BT^2 \ln(T/16.5)$, with $\gamma = 0.221$ J/mole- K^2 , $A = 2.49 \times 10^{-4}$ J/mole- K^4 and $B = 1.61 \times 10^{-2}$ J/mole- K^2 .

show the change from a wide f-d hybridized band to a narrow band with predominantly f character, and can reproduce the increase in density of states with mass enhancement factors m^*/m of 1.4 for $x=0$ and ~ 3 for $x=1$.

Similar studies have been carried out for pseudobinary alloys $URh_{3-x}Pd_x$, i.e. between a compound having itinerant f-electrons and one having localized f-electrons. X-ray studies show that the $AuCu_3$ structure is retained for $x < 2.7$. In that structure, substitution of Pd for Rh again causes band-narrowing. This can be seen in the susceptibility which varies with x in a way very similar to that shown in Fig. 2. The heat capacity (Fig. 3) increases by about an order of magnitude as x varies from 0 to 2. In addition, the compounds for $x > 2$ give a large increase in C/T at temperatures below approximately 10K, indicative of many-body enhancement effects, and a peak in the heat capacity at approximately 2 K. The solid line shows the results of fitting the electronic specific heat with a $T^2 \ln T$ dependence, as would be obtained for spin-fluctuation effects. Band-structure calculations are being carried out for these alloys to assess the relative importance of single-electron and many-body effects.

For Pd concentrations greater than 2.8, the $UPd_{3-x}Rh_x$ alloys take the $TiNi_3$ structure appropriate to UPd_3 . In those cases, the bulk property measurements show relatively normal metallic behavior due to the localized nature of the f electrons, but other alloys having this same structure type are dramatically different. Figs. 4 and 5 show heat capacity and resistivity data for $UPt_{3-x}Ir_x$. For $1.9 < x < 2.8$, x-ray studies show that these materials have the $TiNi_3$ structure and so, by analogy with UPd_3 , may be expected to have f-electrons nearer to the localized limit than those discussed above. In this case, the heat capacity data show low temperature γ values approaching 600 mJ/mole-K^2 . For the alloy $UPt_{2.6}Ir_{0.4}$, which shows the largest γ value yet obtained in this series, the resistivity also shows a minimum as can be obtained from the Kondo effect. Therefore, it appears to be reasonable to describe this as a concentrated Kondo alloy, which is in keeping with the supposition that the f electrons are almost localized. No coherence effects have been observed down to 1.4 K, and measurements of the resistivity are being extended to lower temperatures.

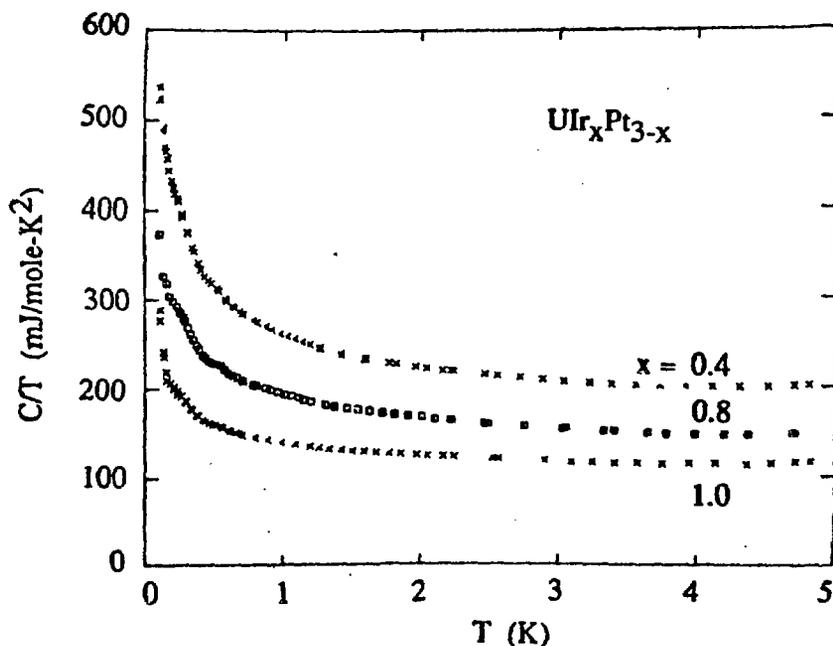


Figure 4. Low temperature heat capacity data for UIr_xPt_{3-x} alloys.

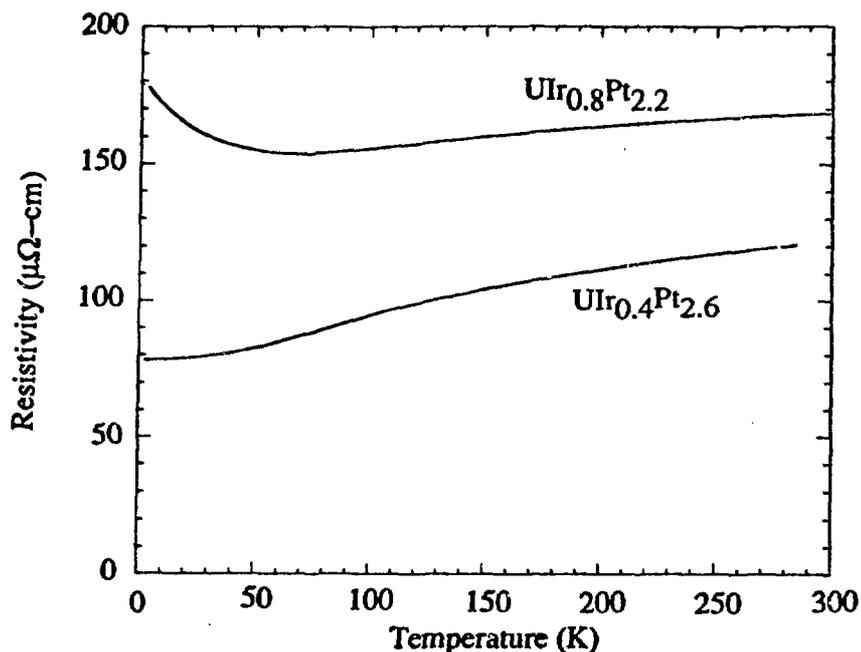


Figure 5. Resistivity data for $\text{UIr}_x\text{Pt}_{3-x}$ alloys.

In conclusion, alloying procedures have been used to vary the nature of the f electrons in U compounds. Examples have been obtained of materials showing large increases in the density of states primarily due to changes describable by a band-structure approach, materials which are derived from an itinerant f-electron state but which have many-body enhancements similar to spin-fluctuation effects, and materials with f-electrons near to localization which display extremely large mass enhancements and Kondo-like phenomena. Such approaches allow studies not only of the specific properties of heavy-fermion materials, but also of the ways in which the heavy ground-state is formed and of its relationship to better understood phenomena.

1. A. J. Arko, M. B. Brodsky, G. W. Crabtree, D. Karim, L. R. Windmiller and J. B. Ketterson, in *Plutonium 1975 and Other Actinides*, Eds. H. Blank and R. Linder (North Holland, Amsterdam, 1976) p. 291
2. R. Kuentzler, S. K. Dhar, S. K. Malik, R. Vijayaraghavan, and B. Coqblin, *Sol. St. Comm.* **50**, 145 (1984)
3. J. D. Jorgensen, private communication
4. D. D. Koelling, private communication

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