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Specific heat of some trans-uranium intermetallic compounds – search for heavy fermions

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Abstract

We measured the low temperature specific heat of single-phase samples of $PuRh_2$, $PuGa_3$, and $NpCd_{11}$. These materials are candidates for heavy fermion behavior based on their structures and, in the case of $PuRh_2$, on other existing measurements such as magnetic susceptibility. We find rather large γ values for $PuGa_3$ (225 mJ mol^{-1} K^{-2}) and $PuRh_2$ (145 mJ mol^{-1} K^{-2}), while that for $NpCd_{11}$ (10 mJ mol^{-1} K^{-2}) is low enough to suggest that another channel of behavior (*e.g.* magnetism) uses up the available f-electron entropy.

1. Introduction

The magnetic susceptibility of $PuRh_2$ at 2.5 K has been reported [1] to be 4.2×10^{-3} emu mol^{-1}, with a slight rounded peak at 10 K, $\chi_{max}\approx4.4\times10^{-3}$ emu mol^{-1}. The heavy fermion compound UPt_3 has approximately similar behavior, with $\chi_{max}=7.2\times10^{-3}$ emu mol^{-1} at 19 K for a polycrystalline specimen [2]. UPt_3 occurs in the hexagonal DO19 structure, while $PuRh_2$ occurs in the cubic C15 structure. UPt_3 shows [3, 4] evidence of spin fluctuations in the specific heat (an upturn at low T in the C/T data which obeys $C\sim T^3$ ln$T$) and a large γ ($T\rightarrow0$) of 450 mJ mol^{-1} K^{-2}. Despite the difference in structure, $PuRh_2$ is a reasonable candidate for further investigation for possible heavy fermion and/or spin fluctuation behavior, particularly since two other examples of spin fluctuation behavior occur in the C15 structure, namely UAl_2 [5] and $TiBe_2$ [6].

$PuGa_3$ occurs in the DO19 structure. Based on arguments of structure, any 4f or 5f intermetallic compound which occurs in the DO19 structure is an interesting candidate for heavy fermion behavior, since both UPt_3 (γ ($T\rightarrow0$)=450 mJ mol^{-1} K^{-2}) and $CeAl_3$ (γ ($T\rightarrow0$)=1600 mJ mol^{-1} K^{-2}) possess the DO19 structure.

UCd_{11} is known [7] to be a heavy fermion antiferromagnet, $T_N=5.0$ K, and γ ($T>5$ K)=840 mJ mol^{-1} K^{-2}. Usually, Np analog compounds are magnetic at higher ordering temperatures than their U counterparts (*e.g.* $NpBe_{13}$ orders antiferromagnetically at 3.4 K while UBe_{13} instead becomes diamagnetic–superconducting at 0.9 K). However, if the expected magnetic order in $NpCd_{11}$ is not too strongly localized in character, some of the remaining unrealized magnetic entropy (*i.e.* that fraction of $R\ln(2s+1)$ not involved in the transition) could be expected to contribute to a large electronic density of states and high γ at lower temperatures.

2. Results and discussion

The specific heats of $NpCd_{11}$, $PuRh_2$, and $PuGa_3$ are shown in Figs. 1, 2 and 3 respectively. Specific heat data from 12 to 59 K for $NpCd_{11}$ (not shown) indicate that C/T remains essentially constant at 3.8 J mol^{-1} K^{-2} between 32 and 59 K, with no sign of any major anomaly in the specific heat. At low temperatures, C/T approaches a low value, 10 mJ mol^{-1} K^{-2}. This result was verified for two different preparations of $NpCd_{11}$, both having X-ray patterns indicative of pure $NpCd_{11}$ in the same structure as UCd_{11}. Based on data from other U and Np homologous intermetallic compounds, where it is generally true that the Np compounds order at a higher temperature than the corresponding homolog compound, $NpCd_{11}$ is expected to order magnetically above 59 K. Magnetic susceptibility data to verify this are necessary.

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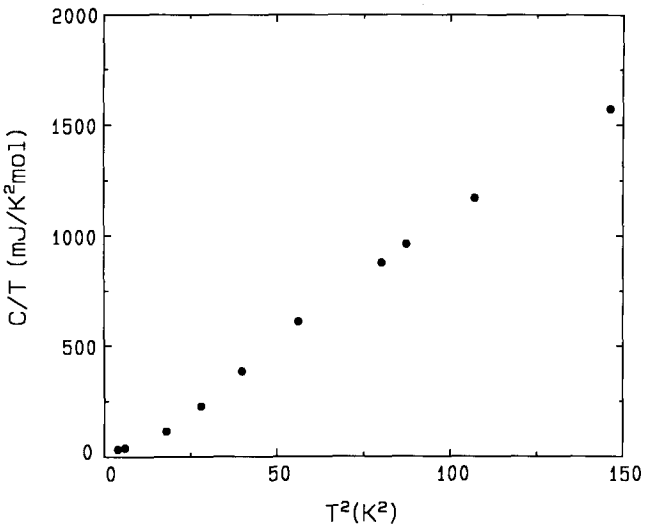

Fig. 1. Low temperature specific heat C divided by temperature T *vs.* T^2 for $NpCd_{11}$. As discussed in the text, these data plus data to 59 K (not shown) show no evidence of a magnetic anomaly.

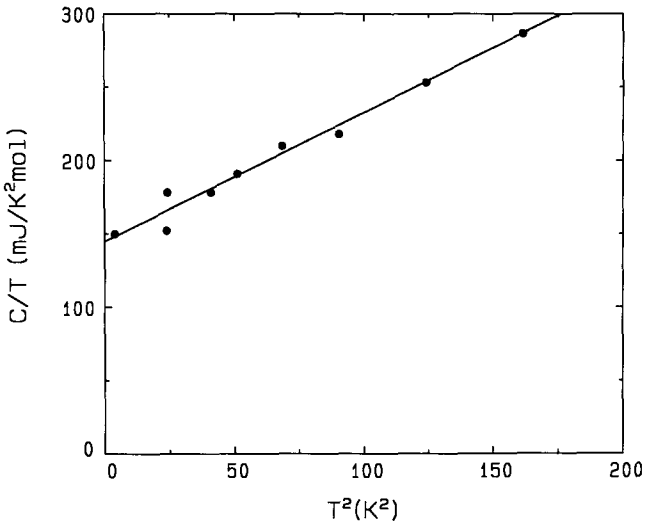

Fig. 2. Low temperature specific heat C divided by temperature T *vs.* T^2 for $PuRh_2$. The data show no sign of a $T^3 \ln T$ upturn at the lowest temperatures.

The low temperature specific heat data for $PuRh_2$ give the result that $\gamma\ (T \to 0) = 145 \pm 10$ mJ mol^{-1} K^{-2} and $\theta_D = 190 \pm 10$ K. There is no sign of any upturn in the low temperature specific heat indicative of spin fluctuations. The size of the γ value qualifies $PuRh_2$ as a middle-heavy fermion system without, however, the temperature dependence in the low temperature γ (equivalent to C/T as $T \to 0$) that is characteristic [2] of most heavy fermion systems.

The low temperature (only measured down to 4.9 K owing to the self-heating and the larger-sized sample used) data for $PuGa_3$, Fig. 3, show similar behavior to

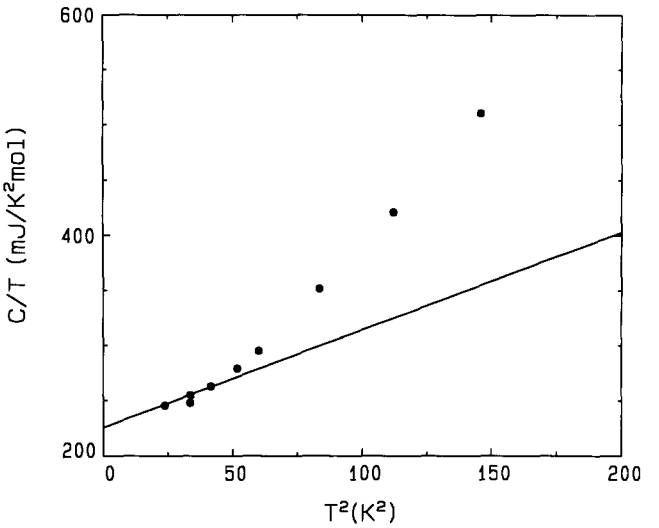

Fig. 3. Low temperature specific heat C divided by temperature T *vs.* T^2 for $PuGa_3$. Owing to self-heating which limited the lowest temperature of measurement to 4.9 K, the extrapolation of the C/T data to $T=0$ is more uncertain than for the $PuRh_2$ data.

those for $PuRh_2$, with $\gamma\ (T \to 0) = 225 \pm 25$ mJ mol^{-1} K^{-2} and $\theta_D = 210 \pm 10$ K.

Thus, although our measurements have discovered two more "middle-weight" fermion systems, they serve chiefly as a further indication that a successful search for heavy fermion behavior needs input in addition to structural, compositional, or magnetic similarity. The details of the 4 or 5f hybridization with the electrons of the non-f atoms are clearly of critical importance for heavy fermion behavior, as proven by, for example, work [8] on $CeCu_{6-x}Ag_x$. Also, the progression from U to Np to Pu in a given intermetallic compound, even if the structure remains constant, entails enormous changes in the magnetic behavior, as seen [9] in the series UBe_{13}, $NpBe_{13}$, and $PuBe_{13}$.

Acknowledgments

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