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Characterization of UPd_3 stabilized in the DO19 structure

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Abstract

A new DO19 compound $U(Pd_{0.8}Au_{0.2})_3$ was prepared. Replacing Pt in the DO19 alloy UPt_3 by Pd normally leads to a transition to the DO24 structure. This is prevented when 20% Pd is replaced by Au. Previous experiments have shown that spin fluctuations in UPt_3 are suppressed and the large specific heat γ value ($C/T(1.2\ K) = 420\ mJ\ mol^{-1}\ K^{-2}$) is decreased by half when alloying with 20% Pd. For UPd_3 stabilized in the DO19 structure with 20% Au, it is found that the electronic contribution to the specific heat decreases further, with a γ value of only 40 mJ (U-mol $K^2)^{-1}$.

Keywords: DO19 structure; Heavy fermions

1. Introduction

Heavy fermion research into new materials with the hexagonal DO19 structure is of interest because some of the protagonists of heavy fermions, such as $CeAl_3$ and UPt_3, are DO19 systems. Leaving the structure unchanged, investigations on new DO19 compounds may offer a better understanding of how parameters other than the structure determine the formation of the heavy fermion ground state.

The substitution of small amounts (less than 10%) of Pt by Pd in UPt_3 leads to complex behavior. C/T increases and an anomaly develops, which corresponds to antiferromagnetic order [1,2]. Further doping of 20% Pd on the Pt sites in UPt_3 suppresses the characteristic upturn in C/T of UPt_3, usually described in terms of spin fluctuation phenomena. The DO19 alloy $U(Pt_{0.8}Pd_{0.2})_3$ shows "normal metal behavior", $C = \gamma T + \beta T^3$, with a γ value of 235 mJ (U-mol $K^2)^{-1}$ [3].

The investigation of $U(Pt_xPd_{1-x})_3$ with $x > 0.67$ is difficult, because further alloying of Pd, which is smaller than Pt, in h.c.p. UPt_3 changes the structure. The atomic radii of Pd and Pt are 1.376 Å and 1.387 Å respectively [4]. $U(Pt_xPd_{1-x})_3$ remains in the DO19 structure for $x > 0.67$; for $0.42 < x < 0.67$ the system is characterized by a ten layer stacking sequence and for $x < 0.42$ the material occurs in the d.h.c.p. DO24 structure [5].

Searching for possibilities to obtain further information on the influence of the replacement of Pt by mainly Pd in UPt_3, we discovered that the DO19 structure could be stabilized by alloying Au in UPd_3. In this paper, we present the properties of $U(Pt_{0.8}Pd_{0.2})_3$, in particular its low temperature specific heat.

2. Experimental details, results and discussion

The sample was prepared by arc melting under a purified argon atmosphere. The purity of the elements U and Pd was 99.95% and of Au was 99.99%. Part of the button was wrapped in Ta foil, sealed in an evacuated quartz glass tube and annealed at 1000°C for 4 weeks to homogenize the sample.

X-ray powder diffraction measurements ($2\theta < 135°$) were performed at room temperature with a Siemens D5000 diffractometer in Bragg-Brentano geometry. The diffraction pattern cannot be identified with the UPd_3 structure, as expected. The existence of diffraction lines with Miller indices (2 0 3), (2 0 5), (4 0 3), (4 0 5), (4 2 3) and (4 2 5) differentiates the DO24 from the DO19 pattern. These lines were detected in our reference sample of UPd_3. In UPd_3, these peaks possess intensities between 14% and 40% of the maximum peak (2 2 8), and should be clearly visible and distinguishable from other intensity maxima in the

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DO24 structure even if 20% of the Pd atoms are replaced by Au. In the diffractometer pattern of $U(Pd_{0.8}Au_{0.2})_3$, these lines cannot be found. However, the pattern of the latter sample can consistently be indexed by a DO19 structure without any resolvable second phases. Thus we assume that replacing 20% Pd by larger Au atoms (atomic radius, 1.429 Å [4]) restores the UPt_3 structure.

This conclusion is supported by another observation. The transition from the DO19 to the DO24 structure in $U(Pd_{1-x}Pt_x)_3$ is accompanied by a characteristic jump in the c/a ratio as described in Ref. [5]. Alloying Pd in UPt_3, while retaining the UPt_3 structure, leads to variations in the lattice constants (less than 0.4% in c, less than 0.1% in a) with a c/a ratio of approximately 0.85. However, on alloying UPd_3 with Pt, the lattice parameter c remains nearly constant; a increases with Pt content by about 0.45%. The counterpart to c in h.c.p. DO19 is $c/2$ in the d.h.c.p. DO24 lattice due to the different stacking of atomic layers. Therefore comparing $c/2$ with the lattice constant a yields a ratio of 0.835 or less, which is smaller than that of UPt_3, reflecting the different stacking sequences of the two structures. For UPd_3, we obtained the lattice constants $a = 5.769(2)$ Å and $c = 9.640(5)$ Å in good agreement with McEwen et al. [5], and for $U(Pd_{0.8}Au_{0.2})_3$, we obtained $a = 5.738(8)$ Å and $c = 4.987(8)$ Å corresponding to a c/a ratio of 0.87. Thus c/a increases significantly in comparison with UPd_3, pointing the way to UPt_3, but with a smaller a value and a larger c value than UPt_3 ($a = 5.75$ Å, $c = 4.899$ Å [5]).

Samples with 10% and 40% Au on the Pd sites have also been prepared. $U(Pd_{0.9}Au_{0.1})_3$ cannot be assigned to a DO19 or DO24 lattice. The diffraction pattern is consistently indexed on the assumption that both structures occur in the sample. In $U(Pd_{0.6}Au_{0.4})_3$, unresolved lines due to second phases appear. Therefore it seems that the range of stability of the DO19 structure for $U(Pd_{1-x}Au_x)_3$ is rather narrow.

The magnetic susceptibility χ was determined by a Quantum Design Squid magnetometer. The χ value at 1.8 K of 19 memu U-mol^{-1} is two to three times larger than the susceptibility of UPt_3 [6] and comparable with that of UPd_3 [7]. $\chi(T)$ does not show any hint of magnetic ordering up to 400 K and behaves Curie–Weiss-like above 100 K. The magnetization at 2 K is linear in fields up to 70000 G.

The specific heat was measured by a relaxation method within an error of ±3%. Fig. 1 shows the specific heat for $U(Pd_{0.8}Au_{0.2})_3$, UPt_3 and $U(Pt_{0.8}Pd_{0.2})_3$. Compared with the other compounds, C/T decreases further in $U(Pd_{0.8}Au_{0.2})_3$. Instead of an upturn in C/T as for UPt_3, which is suppressed in palladium-poor $U(Pt_{0.8}Pd_{0.2})_3$, a downturn in palladium-rich $U(Pd_{0.8}Au_{0.2})_3$ is seen. It extrapolates to a γ value of 40 ± 5 mJ (U-mol K^2)$^{-1}$.

An estimation of the electronic contribution to the

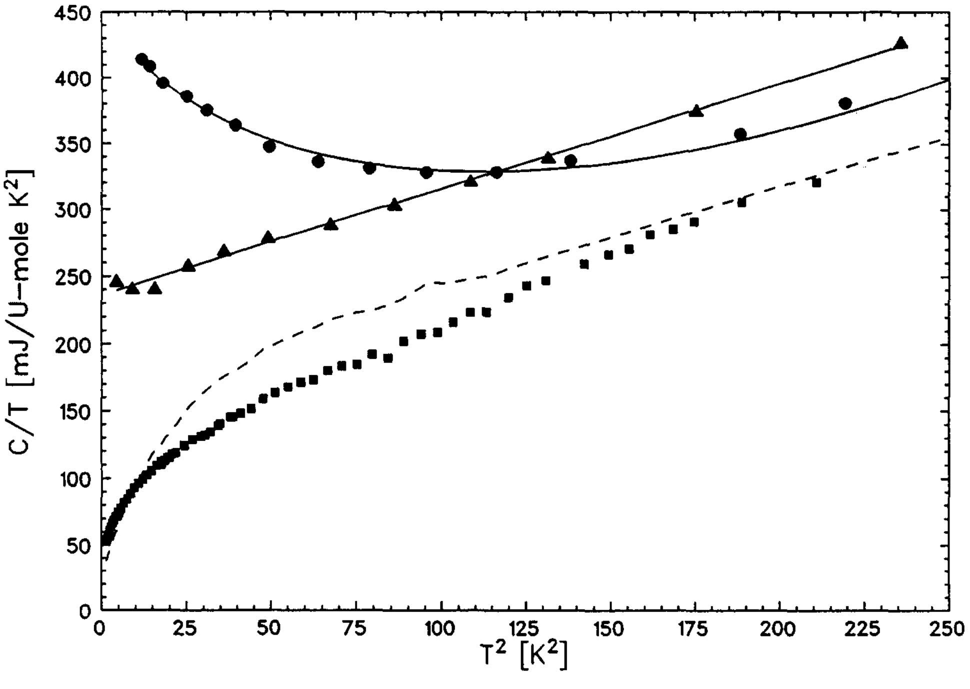

Fig. 1. Specific heat of $U(Pd_{0.8}Au_{0.2})_3$ (■), $U(Pt_{0.8}Pd_{0.2})_3$ (▲) and UPt_3 (●). The data of the last two were taken from Ref. [3]. The full lines represent fits of $\gamma T + \beta T^3$ ($U(Pt_{0.8}Pd_{0.2})_3$) and $\gamma T + \beta T^3 + \delta T \ln T^3$ (UPt_3) respectively. The broken line corresponds to $U(Pd_{0.8}Pt_{0.2})_3$.

entropy below 15 K shows that in $U(Pd_{0.8}Au_{0.2})_3$ only 30% of that in UPt_3 occurs. To compare the alloys, the phonon contribution $C \sim \beta_{ph}T^3$ has been subtracted for both. For UPt_3, $\beta_{ph} = 0.85$ mJ (U-mol $K^4)^{-1}$ [8] has been taken. β_{ph} for $U(Pd_{0.8}Au_{0.2})_3$ has been determined from the specific heat data. The question arises as to why the entropy decreases. Will it be revealed at higher temperatures or are degrees of freedom lost in the lattice where Pt has been substituted completely by Pd and Au?

It is tempting to ascribe the decrease in the specific heat to Pd, but we must be careful since the role gold plays in the compound is not clear. However, the specific heat of another Pd-rich sample $U(Pd_{0.8}Pt_{0.2})_3$ shows a similar behavior (see Fig. 1, broken line). This compound occurs in the DO24 structure. The anomalies seen in UPd_3 [9] vanish due to alloying with Pt and the heat capacity has roughly the same magnitude as that of $U(Pd_{0.8}Au_{0.2})_3$.

Thus a trend from heavy fermion behavior to local moment behavior is observed in the Pd-rich DO19 lattice $U(Pd_{0.8}Au_{0.2})_3$. This is reminiscent of the properties of UPt_3 ("heavy fermion") compared with UPd_3 ("local moment"), and it must be considered whether the differences between the two compounds can be traced to the different crystal structures as argued in Ref. [10].

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