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Specific heat and materials analysis on $U_{1-x}Th_xPt_3$ for $0 \leqslant x \leqslant 0.05$

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Abstract

UPt_3 exhibits anomalous, possibly time-fluctuating antiferromagnetic (AFM) order below 6 K. Th substitution induces conventional AFM order with the same magnetic structure. Recent μSR studies on $U_{1-x}Th_xPt_3$ for $0 \leqslant x \leqslant 0.05$ showed that the transition into the conventional AFM state was sharp for $x = 0.05$, but broadened for $x \leqslant 0.02$ (Phys. Rev. B 84 (2003) 224421), indicative of a crossover behavior. We present X-ray diffraction and transmission electron microscopy (TEM) results that show no significant material inhomogeneity in those samples. However, specific heat measurements corroborate the μSR measurements and show signs of an increase near 7 K for $x > 0.01$. This supports the conjecture that Th impurities slow down the fluctuating AFM, rendering them observable on the timescale of thermodynamic measurements.

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Keywords: UPt_3; Heavy-fermion; Antiferromagnetism; TEM

The anomalous "small-moment" AFM state (SMAF) of UPt_3, from which unconventional superconductivity emerges, is intriguing. It is observed only via neutron and magnetic X-ray scattering, with a tiny staggered moment ($\sim 0.02\mu_B$) and an onset temperature of ~6 K [1]. Sufficiently sensitive local probes (μSR and NMR) fail to detect entry into the SMAF, leading to the speculation that the SMAF is time-fluctuating. Th substitution ($U_{1-x}Th_xPt_3$) induces conventional "large-moment" AFM (LMAF), with a maximum staggered moment $\sim 0.6\mu_B$ and $T_N \sim 6$–7 K for

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$x = 0.05$ [2]. Pd substitution for Pt produces similar effects [3]. Recent μSR measurements showed that Th substituted samples with $x \leqslant 0.02$ had a broad transition, starting near 7 K, into the LMAF phase, in contrast to the sharp transitions for $x = 0.05$ and also for Pd-doped samples [4]. It was conjectured that the Th impurities slowed down the fluctuations associated with SMAF, rendering them observable on μSR timescales.

In this work, we present materials analysis and specific heat measurements on $U_{1-x}Th_xPt_3$ for $0 \leqslant x \leqslant 0.05$ in order to (a) test for a chemical origin for the broadening (e.g., a second phase) and (b) to search for a thermodynamic signature of the broadened transition.

The preparation of the polycrystalline samples was described in Ref. [3]. Metallography and X-ray measurements coupled with a Rietveld refinement show no second phases in any samples. The *a* and *c* lattice parameters agree with reported values for pure UPt_3 [2], and are found to change by $< 7 \times 10^{-3}$ Å between 0 and 5% Th doping.

Three samples (0, 2 and 5% Th) were prepared for transmission electron microscopy (TEM) characterization by focused ion beam milling slices extracted in situ to ~100 nm final thickness using a 30 KeV Ga+ ion beam. They were imaged in a conventional TEM operating at 300 KeV. All three alloys showed the presence of intragranular nanoparticles. In Fig. 1, the regular dark field image shows 8 such particles; weak-beam dark field imaging (inset) shows they are typically thin plates, ~100 nm long and < 10 nm thick. No particles were found at any grain boundaries. The particles varied somewhat in the longest dimension (100–300 nm) between different alloys and different foils extracted from a given sample. The range of interparticle spacings was 250–1000 nm.

The particles' small size and small volume fraction (< 0.1%) precluded crystallographic identification. They may result from precipitation of $(U,Th)Pt_y$ with $y = 2$ or 5 due to slight non-stoichiometry. Zone axis diffraction patterns confirmed that the host phase was hexagonal $(U,Th)Pt_3$.

Specific heat (Fig. 2) was measured between 10 K and 0.5 K (except for $x = 0.05$, measured to 2 K). Not shown is the data for $x = 0$, which is nearly identical to the $x = 0.005$ data for $T > 1$ K, and exhibits a double superconducting transition. Our data agree well with earlier results [5].

The $x = 0.05$ data show a sharp transition near 7 K, in agreement with the μSR measurements [4]. For $x = 0.02$ the size of the anomaly is smaller and broader than for 0.05, as is true for Pd substituted samples. We also find a weak structure near 6.5 K for $x = 0.02$. This is highlighted in Fig. 3 by plotting the temperature derivative, and the anomaly clearly occurs at the same temperature

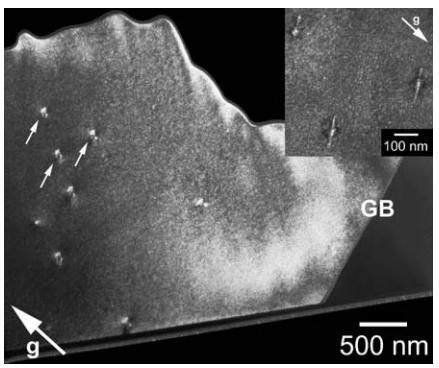

Fig. 1. TEM dark field images of a 5% Th specimen taken under 2-beam conditions (GB = grain boundary). Inset: weak-beam image of the three particles marked by arrows better reveals their true shapes and sizes.

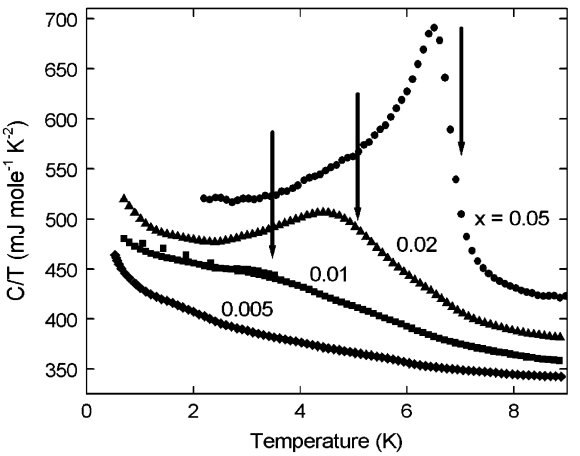

Fig. 2. Specific heat results for four samples of $U_{1-x}Th_xPt_3$. Arrows mark the Néel temperatures from Ref. [4]. Note the bump near 6.5 K for $x = 0.02$. Curves are offset for clarity.

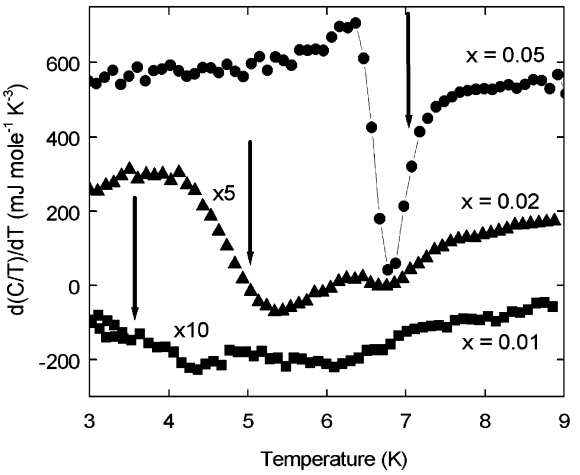

Fig. 3. Derivative of the data in Fig. 2. The minimum near 6 K for $x = 0.01$ is related to the thermometer calibration. Arrows as in Fig. 2. Curves are offset for clarity.

as for $x = 0.05$. This structure was observed for two independently fabricated samples, and in two different calorimeters. Indeed, the $x = 0.02$ data in Ref. [5] appear to exhibit this anomaly. If it were caused by chemical inhomogeneity, ~1% of the $x = 0.02$ sample would need to have a local Th concentration of 0.05. This small volume fraction could not have caused the results of Ref. [4].

The nanoparticles observed by TEM, and any associated local strains, are present for $x = 0$ (no magnetism), 0.02 (broad transition), and 0.05 (sharp transition), and are highly unlikely to cause the broadening reported in Ref. [4] for $0.005 \leqslant x \leqslant 0.02$. While we cannot rule out that the additional specific heat contribution for $x = 0.02$ is caused by chemical inhomogeneity, there is no apparent reason why 5 at% phase should preferentially form in a sample with $x = 0.02$ stoichiometry.

We note that any magnetic fluctuations will contribute to the specific heat, and if their characteristic energy (and frequency) is reduced, this contribution at low temperatures increases. Thus the additional contribution to the $x = 0.2$ specific heat is also consistent with a slowing down of the fluctuations associated with the onset of SMAF near 7 K.

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