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LOW-TEMPERATURE ANOMALIES IN THE SPECIFIC HEAT OF HEAVY-FERMION UPt_3

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High-precision specific-heat measurements (1.3 K $< T <$ 20 K) on UPt_3 yield two small, but distinct, anomalies at 6.2 K, where the c/T curves shows a kink, and at 7.3 K, where a small peak is observed. Samples of different origin gave identical results. The possible connection with antiferromagnetic order yielding small moments ($T_N = 5$ K), as recently probed by neutron experiments, is discussed.

A new common feature of heavy-fermion compounds seems to be the occurrence of small ordered moments developing out of antiferromagnetic fluctuations at low temperatures. At present, it has been illustrated by various microprobe techniques (μSR, NMR and neutron scattering) that the heavy-fermion systems URu_2Si_2, $CeAl_3$, $CeRu_2Si_2$ and UPt_3 (might) undergo such phase transitions. In the case of UPt_3, a first indication of magnetic ordering was deduced from μSR experiments [1], in which below 4 K very slow spin fluctuations, corresponding to an ordered moment of $10^{-3}\mu_B$, were observed. In subsequent neutron-diffraction work Aeppli et al. [2] showed that UPt_3 can be an antiferromagnet with an ordered moment of $(0.02 \pm 0.01)\mu_B$/U-atom and a Néel temperature $T_N = 5$ K. However, similtaneously Frings et al. [3] performed neutron-diffraction studies on two samples of different sources, and found that antiferromagnetic (AF) order was present in one of the samples only ($T_N = 5$ K, $\mu = 0.01\mu_B$/U-atom). The detection of AF order in UPt_3 is a consequence of preceding studies on the alloyed systems $U(Pt_{1-x}Pd_x)_3$ [4] and $U_{1-y}Th_yPt_3$ [5] in which AF order appears (with a maximum T_N of ~ 6 K) for $0.02 \leq x \leq 0.10$ and $0.02 \leq y \leq 0.10$, respectively. The AF order, of the spin-density wave type, was hinted [4,5] by sharp anomalies in the specific heat and resistivity and confirmed by neutron-diffraction experiments on $U(Pt_{0.95}Pd_{0.05})_3$ [6] and $U_{0.95}Th_{0.05}Pt_3$ [7]. The magnetic structure consists of a doubling of the unit cell in the hexagonal plane. The ordered moment ($(0.6 \pm 0.2)\mu_B$/U-atom) points along the *b*-axis. In search for AF fluctuations in pure UPt_3 along the *b*-axis, Aeppli et al. and Frings et al. observed the forementioned weak magnetic Bragg scattering below $T_N = 5$ K.

For a better understanding of this probably itinerant type of AF magnetism, it would be desirable if the anomaly at T_N could be observed in thermal properties, e.g. the specific heat. Previous $c(T)$ data [8] allowed not for a confirmation of AF order due to the relative precision of somewhat less than 1%. Therefore, we decided to perform high-precision (relative error 0.1%) $c(T)$ measurements. Since the AF order was known to be sample dependent [3], we studied three samples: the magnetic and non-magnetic crystals used in the neutron work of Frings et al., and a polycrystalline sample from a third source. The magnetic crystal has been prepared by the float-zone method by E. Bucher, the non-magnetic crystal was grown by the Czochralski method by A.A. Menovsky and the polycrystalline sample was prepared in an induction furnace by L. Taillefer. In the following these samples will be addressed as #1, #2 and #3, respectively. General information (purity, T_c, electronic mean free path, etc.) of this type of samples can be found in the papers of Aeppli et al. [2], de Visser et al. [8] and Taillefer et al. [9], respectively. A more detailed characterization of samples #1, #2 and #3 is in progress, and will be published elsewhere.

Specific-heat measurements have been performed employing the adiabatic technique, the sample being cooled with a mechanical heat switch. In order to minimize thermal losses, the shield surrounding the sample was kept at the same temperature as the sample. Temperatures were read with an Allen Bradley resistor (68 Ω at 300 K) that was calibrated "in situ" just after the

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$c(T)$ measurements, against the vapour pressures of ^{3}He (1.2–3.3 K), ^{4}He (2.2–4.5 K) and para H_2 (13.8–21.1 K) and a gas thermometer (4–30 K). It is emphasized that the temperature calibration has been made for each sample run, keeping the AB resistor at liquid helium temperatures. The mass of the samples amounted to ~ 15 g.

The experimental results are shown in fig. 1 for sample #1. We also show the electronic specific heat and the phonon contribution [10]. The shape of the $c_e(T)$-curve suggests that two overlapping contributions are present: one centered near 10 K and the other above 20 K [11]. However, care should be taken, since above 17.3 K data for c_{ph} have been extrapolated from the original figure in ref. [10], thus increasing the error bar on c_e. The emphasis of the present paper is on the structure in c/T near 7 K. It is shown in detail in fig. 2 for samples #1, #2 and #3.

Surprisingly, no anomaly is found at $T_N = 5$ K, but instead two small, but distinct, anomalies are observed at 6.2 K, where the c/T curve shows a kink, and at 7.3 K, where a small peak (with a magnitude of 1% of c/T) is observed. As follows from fig. 2, the results for the three samples are very alike (although the structure at 6.2 K for sample #3 is somewhat more pronounced). Careful measurements on copper samples exclude that these anomalies are due to the addenda (about 10% of the total heat capacity). This is furthermore confirmed by the absence of these anomalies in data on samples of other compounds with much smaller heat capacities. As mentioned above, the thermometer was calibrated for each sample run.

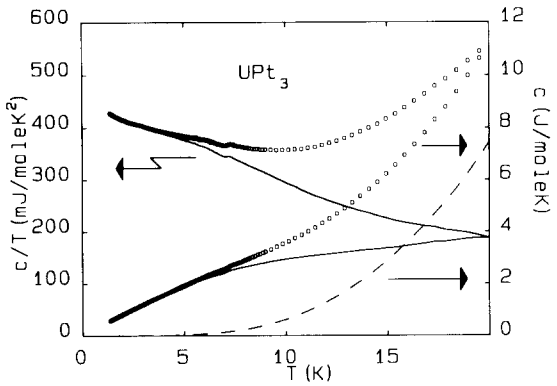

Fig. 1. Specific heat of UPt_3 sample #1 (○). The solid and dashed lines represent c_e and c_{ph} [10], respectively.

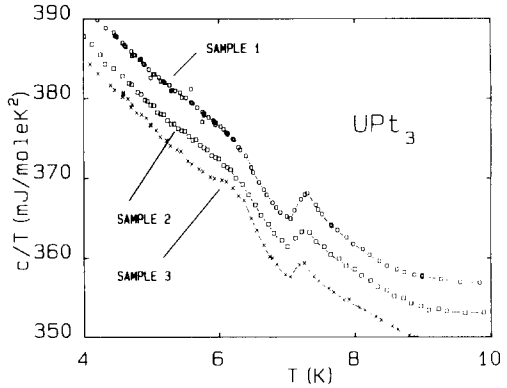

Fig. 2. c/T vs. T of UPt_3 sample #1 (○), sample #2 (□) and sample #3 (×). For sake of clarity, the latter curve (×) has been shifted by -5 mJ/molK^2. Dashed lines serve as a guide to the eye.

Analytic functions $T(R)$ were made in three temperature regions: $T < 3.5$ K, 3 K $< T <$ 10 K and 9 K < T, excluding possible irregularities near 7 K due to non-matching of the functions. Seen the above considerations, we conclude that the anomalies are intrinsic.

Next we discuss whether the observed anomalies might be caused by contamination of the samples. In particular Pb, Pd and Th should be considered. Small amounts of Pb ($T_c = 7.2$ K) might give rise to the anomaly at 7.3 K. Pd and Th doping leads to antiferromagnetism with large anomalies in $c(T)$ near 6 K (see above). Since, at present, no studies of the impurity contents for our samples are available, we refer to ref. [2] in which upperbounds of 10 ppm by weight for Pb (probably due to radioactive decay) and 1 ppm by weight for Pd and Th, are given. Simple calculations show that such concentrations are a factor 10^4 and 10^2 too small, respectively, in order to account for the measured anomalies at 6.2 and 7.3 K. Pb, Pd or Th impurities can thus be discarded as the cause of the anomalies.

At present we cannot offer a satisfactory explanation for the structure in c/T. The shape of the anomalies is rather puzzling (there is no clearcut way of subtracting a background), but indicates that the involved entropy is very small. As the anomalies are observed in the magnetic *and* the non-magnetic sample, their relation with the AF order is questionable. However, the possibility remains that the magnetic Bragg peaks were

not detected in sample #2, due to sensitivity or sample quality (mosaic) variations. In that case, the kink at 6.2K might reflect the AF transition, with a precursor effect at 7.3 K. Preliminary $c(T)$ measurements in a magnetic field of 0.1T reveal that the small peak at 7.3 K disappears, while the kink at 6.2 K remains.

AF order at 5 K in UPt_3 is unlikely according to the phase diagram of $U(Pt_{1-x}Pd_x)_3$. For $x = 0.05$ $T_N = 5.8$ K, and drops to $T_N = 3.6$ K for $x = 0.02$. No AF order has been observed for a 1% compound [12]. On the other hand, one cannot exclude that the antiferromagnetism in pure UPt_3 is of a different nature than in the doped compounds, and might be rapidly depressed on alloying with Pd, as is the case for the superconductivity [4]. This would suggest that AF order in pure UPt_3 is related to delicate Fermi surface nesting, that is easily altered by alloying. It is interesting to note that the anomalies remind one of the two phase transitions observed in dhcp UPd_3 at 5 and 7 K [13]. The possibility that crystal defects in hcp UPt_3 (for instance dhcp stacking faults) give rise to small ordered moments asks for further investigations. We finally note that crystal defects were found to be present in UPt_3 crystals, as, for instance, can be concluded from the non-resolution limited diffraction peaks [2].

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