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Properties of Cu-flux-grown UCu_2Si_2

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Abstract

Single crystals of UCu_2Si_2 grown from Cu flux have a 50 K antiferromagnetic transition below the 100 K ferromagnetic transition, this not being seen in polycrystalline material. We speculate on the possible reason for this.

Rare-earth and uranium intermetallics forming in the ThCr_2Si_2 (or BaAl_4) and the closely related CaBe_2Ge_2 structures have been extensively investigated since the discovery of heavy-fermion superconductivity in CeCu_2Si_2 by Steglich in 1979. Motivation for this study comes from the early observations by Steglich and collaborators that the properties of CeCu_2Si_2 were sensitive to Cu stoichiometry, Cu excess favouring the superconductivity.

With this in mind, crystals of UCu_2Si_2 have been grown by slow cooling of dilute solutions of molten Cu containing U and Si in the stoichiometric ratio 1:2. Crystals were etched from the Cu matrix using diluted HNO_3 . The crystals formed as millimetre-size plates. The powder x-ray diffraction pattern at room temperature showed well resolved Cu $K\alpha_1$ – α_2 splittings with the BaAl_4 structure type, the refined lattice parameters $a_0 = 3.984 \text{ \AA}$, $c_0 = 9.937 \text{ \AA}$, agreeing well with literature values. Magnetization measurements were performed, for $2 \text{ K} \leq T \leq 350 \text{ K}$, using a commercial SQUID magnetometer (Quantum Design). Heat capacity and resistivity measurements, for $0.35 \text{ K} \leq T \leq 300 \text{ K}$, were carried out in a commercial physical property measurement system (Quantum Design).

The previous experimental work that we are aware of has been carried out on polycrystalline material. The ferromagnetism observed near 100 K appears robust across all samples. Elastic neutron scattering [1] finds an ordered moment aligned along the c -axis of $1.6 \mu_B$, in agreement with the saturation magnetization found in susceptibility measurements. Also observed in several studies [2, 3] has been an antiferromagnetic transition that occurs 5 K above the ferromagnetic transition.

Our magnetic susceptibility measurements on the Cu-flux-grown single crystals are shown in figure 1, parallel and perpendicular to the plates which grow in the ab -plane. The inset shows the Curie–Weiss plot of this data. The c -axis data have a ferromagnetic Curie–Weiss intercept of 104 K, close to where spontaneous magnetic moment along the c -axis develops. The ab -plane has an antiferromagnetic intercept of -55 K . This, in turn, is close to the temperature where we observe a first-order-like drop in the c -axis magnetization. Three further aspects of

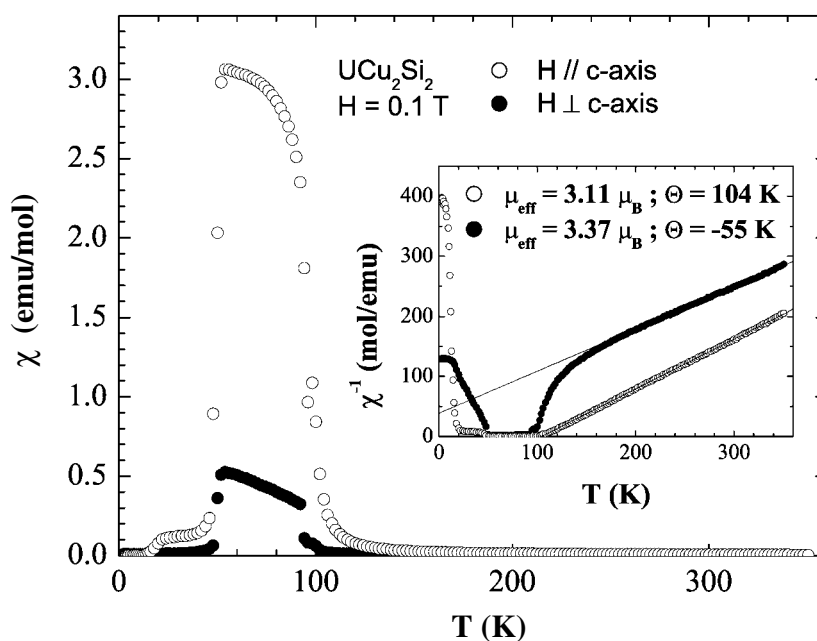


Figure 1. The magnetic susceptibility temperature of UCu_2Si_2 parallel and perpendicular to the c -axis. The inset shows Curie–Weiss plots.

the data are noted. First, there is a dip in the c -axis data as the spontaneous magnetic moment is being established. Second, the ab -plane magnetization which tracks the c -axis data at much reduced amplitude, in addition to the downward curvature in χ_{ab}^{-1} near 100 K, can be accounted for by a small misalignment of the crystal. And third, there is another relatively small feature seen in χ_c near 20 K, which remains mysterious.

The 50 K antiferromagnetic transition has not been observed in other measurements on UCu_2Si_2 . The question naturally arises as to why the ferromagnetism is so robust, the 50 K antiferromagnetism apparently delicate. The magnetization as a function of applied field (figure 2) shows from another perspective the fragile nature of this lower antiferromagnetic transition: only modest external magnetic fields applied parallel to the c -axis at 5 K are needed to induce the fully ordered ferromagnetic state, and the antiferromagnetic state is not obvious in the hysteresis loop (in the ab -plane, 5 T does not induce a metamagnetic transition at 5 K). Somewhat surprisingly, we find the same antiferromagnetic transition temperature for measurements made in 0.001 T as in 0.1 T. We note that a field energy for the saturation moment found in this compound ($1.8 \mu_B$) corresponding to a $T_N = 50$ K implies an applied field of order 40 T, far larger than 1.5 T found to induce the ferromagnetic state at 5 K.

The electrical resistivity (figure 3, ρ_{ab}) tells us more about these crystals. We can clearly see the loss of electrical resistivity below T_C . There is a hysteretic Cr-type anomaly near 70 K, which is not coinciding with the antiferromagnetic transition seen in the magnetic susceptibility. We do not have an explanation for this. However, we note that the antiferromagnetic ordering direction appears to be the c -axis. Superzone boundaries arising from this order are not expected to influence ρ_{ab} significantly. It is also evident that the electrical resistivity is very large and that the residual resistance ratio is small. It is remarkable that the resistivity of these single crystals is more than two orders of magnitude larger than the reported polycrystalline value [4]. This indicates disorder in the crystal, the most likely candidate cause for which is

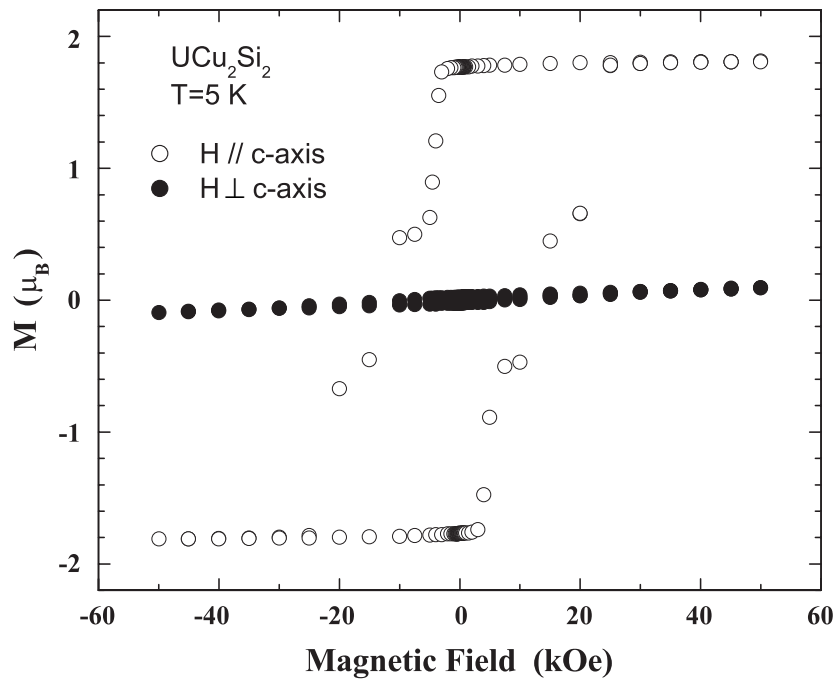


Figure 2. Magnetization parallel and perpendicular to the c -axis at 5 K.

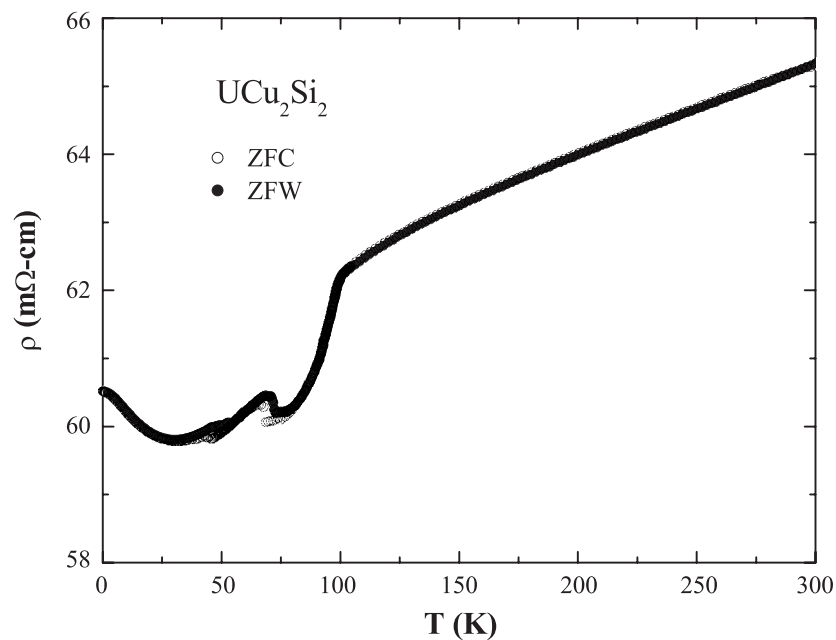


Figure 3. Electrical resistivity in the ab -plane.

chemical substitution. Studies of UIr_2Si_2 (CaBe_2Si_2 type) have found Si substituting on Ir sites at the $5a/0$ level [5]. Substitution of some Si on Cu sites is a possibility.

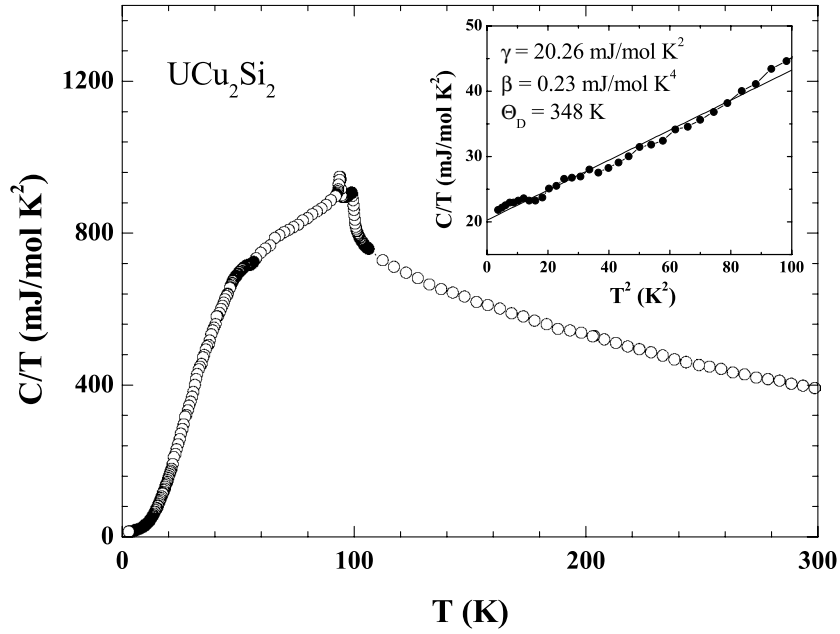


Figure 4. The temperature dependence of the specific heat divided by temperature (C/T) for UCu_2Si_2 . The inset shows the low-temperature data plotted against T^2 .

Finally, we show in figure 4 the specific heat measurement. The two specific heat anomalies near 100 K correspond nicely with the antiferromagnetic and ferromagnetic transitions which have been observed in most previous investigation. The 50 K transition is not as evident. While there is some rounding seen in this temperature range in C/T , the lack of a distinct transition can perhaps be rationalized by noting the very small expected entropy of this transition, coupled to its apparent first-order character. We point out that the extrapolated low-temperature electronic specific heat $C = \gamma = 20 \text{ mJ K}^{-2}/\text{mol U}$, a value typical of transition metal materials (see the inset in figure 4).

The properties of UCu_2Si_2 suggest that it is a local moment ferromagnet. The significant spin disorder resistivity observed is not typical of itinerant ferromagnets. Neither do Kondo effects enter in any obvious way into the physics: it seems likely that the RKKY scale strongly dominates the Kondo scale. What is surprising and unexpected is that a previously unseen low-temperature antiferromagnetic order emerges abruptly and cleanly in this apparently somewhat disordered material, and that the temperature scale for this antiferromagnetism is clearly evident in the ab -plane susceptibility. It is perhaps plausible that the short electronic mean free path in the Cu-flux-grown crystals attenuates RKKY interactions, limiting the U–U moment interactions effectively to nearest-neighbour ones.

A different possibility for explaining the difference between our single-crystal data and previous reported polycrystalline results is that the Cu-flux-grown crystals are highly stoichiometric [6]. This possibility is supported by the well resolved doublets in the x-ray pattern, suggesting low disorder in the crystals. The very large resistivity found for the crystals then implies that stoichiometric UCu_2Si_2 is semimetallic, a possibility which we are currently investigating further. We are aware of no other materials in the ThCr_2Si_2 structure which are semimetallic, but we note that the RKKY interaction is ferromagnetic at low carrier density.

It is naturally of interest to compare the behaviour of the Ce and U in the transition metal BaAl₄ disilicides. The usual framework within which the properties of these compounds are discussed is that of the competition between RKKY and Kondo energy scales. Both temperature and f-electron/conduction electron interaction J are scaled by a bandwidth scale, W . One thus expects magnetic ordering temperatures and Kondo scales to be an order of magnitude larger in U compounds compared with Ce-compound counterparts, seemingly consistent with observation.

But important questions are left open. We really know very little about U as a Kondo impurity. Experimentally, data exist for Th:U [7] and LaAl₂:U [8]. Can we be confident that the Kondo physics of crystal field-split f^2 ions maps onto that of f^1 ions? And further, are quadrupolar effects essentially irrelevant? There are as well various differences seen in the experimental record. Where are the pressure-induced superconductors in UT₂Si₂ or UT₂Ge₂ compounds? Why is there no coexisting magnetism and superconductivity in Ce 122 compounds like that found in URu₂Si₂? And why is there not a superconductivity scale found in U heavy fermions proportional to the order-of-magnitude-larger magnetic ordering temperatures?

These remarks are meant to suggest that great benefit may come through detailed studies of U intermetallics which really come to grips with the differences with Ce materials rather than simply increasing the phase space of examples.

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