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Electronic properties of UCuSn

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Crystallographic analysis shows that UCuSn does not form in the hexagonal CaIn_2 structure as reported previously, but is an ordered ternary compound and forms in an orthorhombic structure (space group: $\text{P2}_1\text{cn}$). Bulk and neutron-diffraction measurements reveal that UCuSn orders antiferromagnetically below 62 K. At 4.2 K, high-field magnetization reveals a complex magnetization process with two metamagnetic transitions. Furthermore, bulk investigations show an additional anomaly at 25 K, but a smooth temperature dependence of various magnetic peaks down to the lowest temperature gives no evidence for a second magnetic transition. Possible scenarios responsible for the drastic changes in the electronic properties around 25 K are discussed. © 1996 American Institute of Physics. [S0021-8979(96)26208-5]

I. INTRODUCTION

The role of $5f$ -ligand hybridization in suppressing local-moment magnetism has been studied extensively in the two largest isostructural groups of UTX compounds (T = transition metal, X = p -electron metal), namely, in compounds crystallizing in the hexagonal ZrNiAl and the orthorhombic TiNiSi structures.¹ The aim to search even higher degrees of $5f$ -electron localization brings us to compounds reported to crystallize in the hexagonal CaIn_2 structure, which is formed for the very late d -metals Cu, Au, and Pd.²

In this contribution, we concentrate on UCuSn, which was thought to crystallize in this structure.²⁻⁵ For this compound, antiferromagnetic ordering at low temperatures has been reported on the basis of electrical-resistivity,^{4,5} magnetic-susceptibility,⁴ and ^{119}Sn Mössbauer studies.⁶ In addition, magnetization measurements revealed two metamagnetic transitions at about 12 and 17 T in the ascending field sweep, while in the descending magnetization curve only one transition around 12 T was found.⁴ There is also a very unusual temperature dependence of the electrical resistivity: below 60 K, $\rho(T)$ increases suddenly forming a maximum around 25 K, where it drops precipitously.^{4,5}

In order to get more insight in the electronic properties of UCuSn, we have checked and extended the study of some basic bulk properties. Here, we briefly describe also the results of additional neutron-diffraction experiments, the details of which will be published elsewhere.

II. SAMPLE PREPARATION AND CHARACTERIZATION

A polycrystalline sample of UCuSn was prepared by arc-melting stoichiometric amounts of the constituents, with no further heat treatment. The sample was powdered for the neutron diffraction experiments, which were performed on the High-Intensity Powder Diffractometer at LANSCÉ and on the C2 powder diffractometer at Chalk River. We observed a number of nuclear reflections which could not be

indexed in the hexagonal system (e.g., CaIn_2 structure type), but we were able to index and refine the whole pattern in the related orthorhombic space group $\text{P2}_1\text{cn}$. The resultant structure and parameters are given in Fig. 1 and Table I.

III. BULK PROPERTIES

We also measured the temperature dependences of the electrical resistivity, of the specific heat, and of the magnetization (at 2 and 4 T) and the results are displayed in Fig. 2.

The absolute resistivity values are intermediate between the values published in Refs. 4 and 5 and the overall shape of

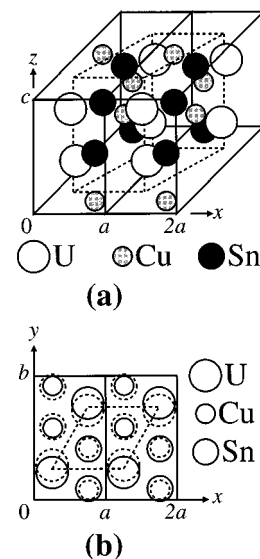


FIG. 1. Crystallographic structure of UCuSn (a) schematically drawn in a doubled cell showing the relation to the "parent hexagonal" cell. For sake of clarity, the z position was shifted by 0.25 with respect to the values given in Table I. In (b), a projection onto the a - b plane is shown, where the dashed lines represent the lower z position with respect to the solid lines. The hexagonal cell is indicated by dashed lines.

TABLE I. Refined structural parameters for UCuSn at 300 K.

Space group: P2 ₁ cn			
	x	y	z
U (4a)	0.25 ^(a)	0.2788(1)	0.4982(2)
Cu (4a)	0.2597(19)	0.9168(2)	0.6958(1)
Sn (4a)	0.2586(20)	0.5835(4)	0.7762(1)

(a) fixed to define the origin

Lattice parameters

a (Å)=4.5391(2)

b (Å)=7.8397(2)

c (Å)=7.2203(2)

R factors

R_{wp} =4.29%

R_p =3.09%

reduced χ^2 =3.88

all three results is in good agreement. In our sample, antiferromagnetism sets in around 62 K, which is reflected by a sudden increase of the electrical resistivity. Upon further decrease of the temperature, the electrical resistivity passes a pronounced maximum around 25 K. At both temperatures (62 and 25 K), we observe anomalies in the specific heat and the magnetization. While clear maxima evolve in C and M/H around 62 K, the low-temperature anomaly is far less pronounced. Around 25–30 K, only slight changes in the derivatives are observed. In addition, we find that a small, but significant ferromagnetic component ($\sim 0.02\mu_B/U$ atom) evolves below 30 K. At present, we are not sure whether this is intrinsic or due to a small amount of a ferromagnetic impurity phase.

High-field-magnetization studies were performed at the High-Field Facility in Amsterdam on powder, both free to be

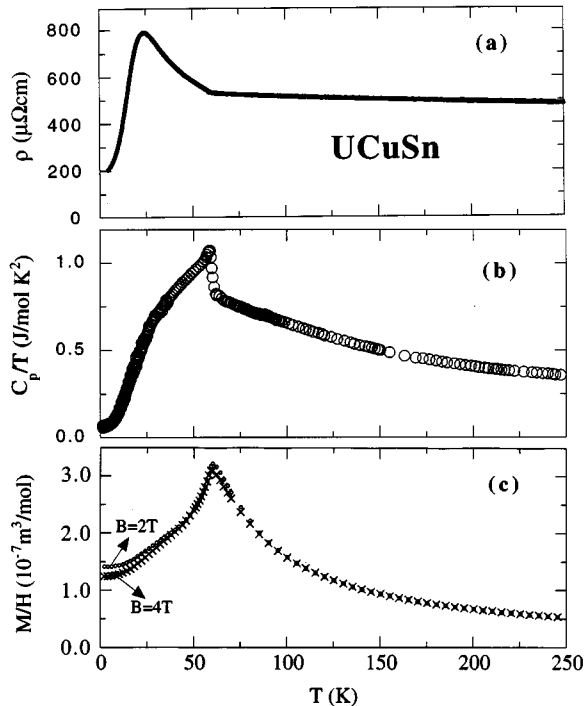


FIG. 2. Temperature dependences for UCuSn of (a) the electrical resistivity, (b) the specific heat, and (c) the magnetization (as M/H for $H=2$ and 4).

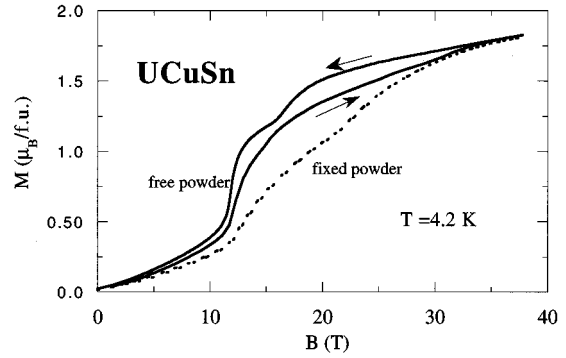


FIG. 3. Field dependence of the magnetization of UCuSn at 4.2 K measured on ‘free powder’ and ‘fixed powder’ represented as solid and dotted lines, respectively.

oriented by the applied field (giving the response of the easy magnetization direction), and in random orientation fixed by frozen alcohol (thereby representing an “ideal” polycrystal). The former (“free powder”) result reveals a magnetic moment of $1.82\mu_B$ in 35 T, which is in excellent agreement with the results of Fujii *et al.*⁴ The antiferromagnetic ground state is corroborated by metamagnetic transitions. In contrast to the results in Ref. 4, we find metamagnetic transitions around 12 and 30 T in the ascending curve and around 11 and 15 T in the descending curve. We believe that this difference could originate in small differences in the compositions of the samples, which could also account for the slight difference in T_N . Two transitions (around 13 and 25 T) are found also in the second (fixed powder) measurement, but no hysteresis was observed. The fact, that both magnetizations are almost equal in the highest field applied, may indicate an anisotropy field only slightly higher than 30 T, which is exceptionally low among U intermetallics.⁷

IV. NEUTRON-DIFFRACTION RESULTS

The bulk results above indicate two magnetic transitions in UCuSn at about 62 and 25 K. At low temperatures, the antiferromagnetic ground state was confirmed by the occurrence of additional purely magnetic reflections below 62 K in the neutron data taken at Los Alamos. However, magnetic intensities were found also on some nuclear reflections, and our results indicate that all magnetic contributions can be indexed in the same orthorhombic unit cell.

In order to clarify the (sometimes pronounced) anomalies in the bulk properties around 25 K, we performed additional neutron-diffraction experiments on the C2 powder diffractometer at Chalk River Laboratories. Data were collected at various temperatures, and the temperature dependences of the peak intensities of some “magnetic” peaks are shown in Fig. 4. In all cases, we observe a smooth temperature dependence with no evidence for a second magnetic phase transition around 25 K and no extra magnetic peaks were observed below this temperature.

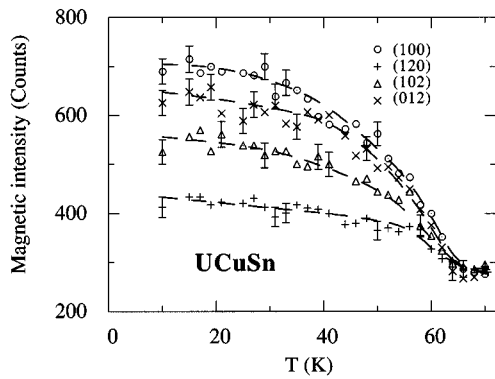


FIG. 4. Temperature dependence of the magnetic intensities of various Bragg reflections. In some cases, the intensities have been corrected for nuclear contributions. The lines are guides to the eye.

V. DISCUSSION

While the investigations of bulk properties seem to indicate a second magnetic transition around 25 K, any moment reorientation seems to be excluded by the neutron-diffraction results. Clearly, this confusing situation cannot be resolved on the basis of the present data, but we may speculate on scenarios which may account for all observations. A possible explanation may involve temperature-induced changes of the antiferromagnetic gap which ultimately could lead to significant changes in the Fermi surface and therefore strongly affects the bulk properties (without a magnetic phase transition). Such a picture has been suggested by Fujii *et al.*⁴

On the other hand, an alternative explanation arises from the comparison with UPdSn, which forms in an ordered version of the CaIn_2 structure. UPdSn exhibits two magnetic transitions which are due to ordering of the y and z components of the magnetic moment at the upper transition temperature, while the x component fluctuates until it also stabilizes at the lower transition temperature.⁸ For this compound, single-crystal studies show clear anomalies in the temperature dependence of the intensity for most magnetic

peaks, while this transition is “smeared out” in powder diffraction. As a consequence only a broad and featureless temperature dependence was found.⁹ It may be that UCuSn behaves in a similar fashion, and one moment component stays fluctuating below $T_N=62$ K, but that its fluctuations gradually slow down until 25 K. In such picture, one may anticipate an enhancement in the resistivity due to fluctuations until they die out, with little or no change in the magnetic moment.

Further high-field magnetization studies at elevated temperature are envisaged to give more insight to the 25 K transition.

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