

LA-UR- 95 - 2954

ELECTRONIC PROPERTIES OF UCuSn

H. Nakotte, A. Purwanto and R.A. Robinson
LANSCE, Los Alamos National Laboratory, Los Alamos, NM 87545, U.S.A.
 K. Prokeš and F.R. de Boer
*Van der Waals-Zeeman Laboratory, University of Amsterdam, 1018 XE Amsterdam,
 The Netherlands,*
 L. Havela and V. Sechovský,
Department of Metal Physics, Charles University, 12116 Prague 2, The Czech Republic
 I. Swainson
*Chalk River Laboratories, Atomic Energy of Canada Limited, Chalk River,
 Ontario K0J1J0, Canada*

Abstract

Crystallographic analysis shows that UCuSn does not form in the hexagonal CaIn₂ structure as reported previously, but is an ordered ternary compound and forms in an orthorhombic structure (space group: P2₁cn). Bulk and neutron-diffraction measurements reveal that UCuSn orders antiferromagnetically below 60 K. At 4.2 K, high-field magnetization reveal a complex magnetization process with two metamagnetic transitions. Furthermore, bulk investigations show an additional anomaly at 25 K, but a regular 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.

PACS numbers: 75.25, 75.30 c, 75.50 c

INTRODUCTION

The role of 5*f*-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 degree of 5*f*-electron localization brings us to compounds reported to crystallize in the hexagonal CaIn₂ 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 ¹¹⁹Sn 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.

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 HighIntensity Powder Diffractometer at LANSCE at the C2 powder diffractometer at Chalk River. We observed a large number of nuclear reflections which could not be indexed in the hexagonal systems (e.g. CaIn₂ structure type), but we were able to index and refine the whole pattern in the related orthorhombic space group P2₁cn. The resultant structure and parameters are given in Fig. 1 and Table 1.

BULK PROPERTIES

We also measured the temperature dependences of the electrical resistivity, of the specific heat, and of the magnetization (in 2 T 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 all three results is in good agreement. At 60 K, antiferromagnetism sets in, and reflects itself suddenly increasing the electrical resistivity. Upon further decrease of the temperature, the electrical resistivity passes a pronounced maximum around 25 K. At both temperatures (60 and 25 K), we observe anomalies in the specific heat and the magnetization. While clear maxima evolve in C and M/H around 60 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

DISCLAIMER

Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.

ferromagnetic component ($\sim 0.02 \mu_B/\text{U-atom}$) 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 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. 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].

NEUTRON-DIFFRACTION RESULTS

The bulk results above indicate two magnetic transitions of UCuSn at about 60 and 25 K. At low temperatures, the antiferromagnetic ground state was confirmed by the occurrence of additional purely magnetic reflections below 60 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.

DISCUSSION

While the investigations of bulk properties seem to indicate a second magnetic transition around 25 K, any moment re-orientation 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. Single-crystal studies showed that the two transitions of this compound 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]. It may be that UCuSn behaves in a similar fashion, and one moment component stays fluctuating below $T_N = 60$ K, but that its fluctuations gradually slow down until 25 K, where they (unlike UPdSn) stabilize at the zero value. 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.

ACKNOWLEDGMENTS

H.N. and A.P. would like to thank the Chalk-River staff for the pleasant stay during the neutron experiments. This work was supported by the U.S.-Czechoslovak Science and Technology Joint Fund under project No. 93039. It was also supported in part by the division of Basic Energy Sciences of the U.S. Department of Energy, by the Grant Agency of the Czech Republic (project nr.: 202/93/0184) and the "Stichting voor Fundamenteel Onderzoek der Materie" (FOM).

References

- [1] V. Sechovský and L. Havela, in *Ferromagnetic Materials*, eds. E.P. Wohlfarth and K.H.J. Buschow, Amsterdam: North-Holland, Vol. 4, 1988, pp. 309-491.
- [2] T.T.M. Palstra, G.J. Nieuwenhuys, R.F.M. Vlastuin, J. van den Berg and J.A. Mydosh, *J. Magn. Magn. Mater.* **67** (1987) 331.
- [3] F.R. de Boer, E. Brück, H. Nakotte, A.V. Andreev, V. Sechovský, L. Havela, P. Nozar, C.J.M. Denissen, K.H.J. Buschow, B. Vaziri, M. Meissner, H. Maletta and P. Rogl, *Physica B* **176** (1992) 275.
- [4] H. Fujii, H. Kawanaka, T. Takabatake, E. Sugiura, K. Sugiyama and M. Date, *J. Magn. Magn. Mater.* **87** (1990) 237
- [5] V.H. Tran and R. Troc, *Int. J. Mod. Phys. B* **7** (1993) 850.
- [6] R. Kruk, K. Latka, K. Tomala, R. Kmiec, R. Troc and V.H. Tran, *J. Alloys Comp.* **219** (1995) 256.
- [7] L. Havela, V. Sechovský, H. Nakotte, E. Brück and F.R. de Boer; *J. Alloys Comp.* **213/214** (1994) 243.
- [8] R.A. Robinson, J.W. Lynn, A.C. Lawson and H. Nakotte; *J. Appl. Phys.* **75** (1994) 6589.

Figure captions:

Fig. 1: Crystallographic structure of UCuSn (a) schematically drawn in a doubled cell showing the relation to a distorted hexagonal cell. For sake of clarity, the z position was shifted by 0.25 with respect to the values given in table 1. 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.

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

Fig. 3: Field dependence of the magnetization of UCuSn at 4.2 K measured on 'free powder' and 'fixed powder' (see text) represented as solid and dotted lines, respectively.

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.

Table:

Table 1: Refined structural parameters for UCuSn at 78 K

Space Group: P2₁cn

	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)

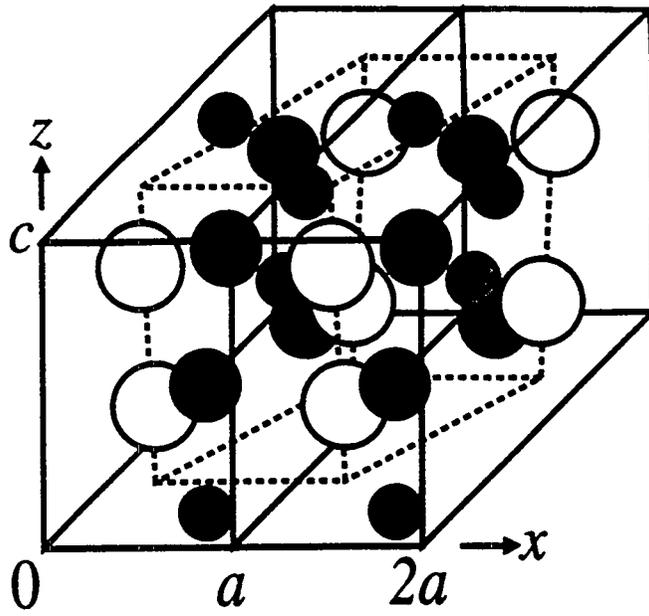
c (Å) = 7.2203(2)

R factors

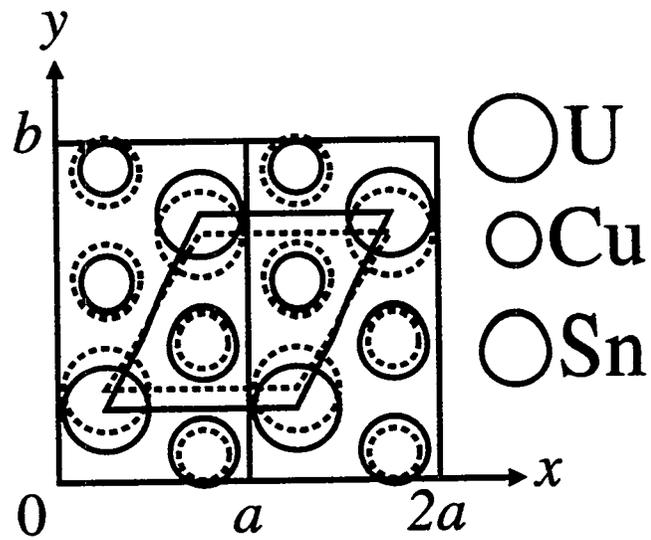
R_{wp} = 4.29%

R_p = 3.09%

reduced χ^2 = 3.88



(a)



(b)

Fig. 1

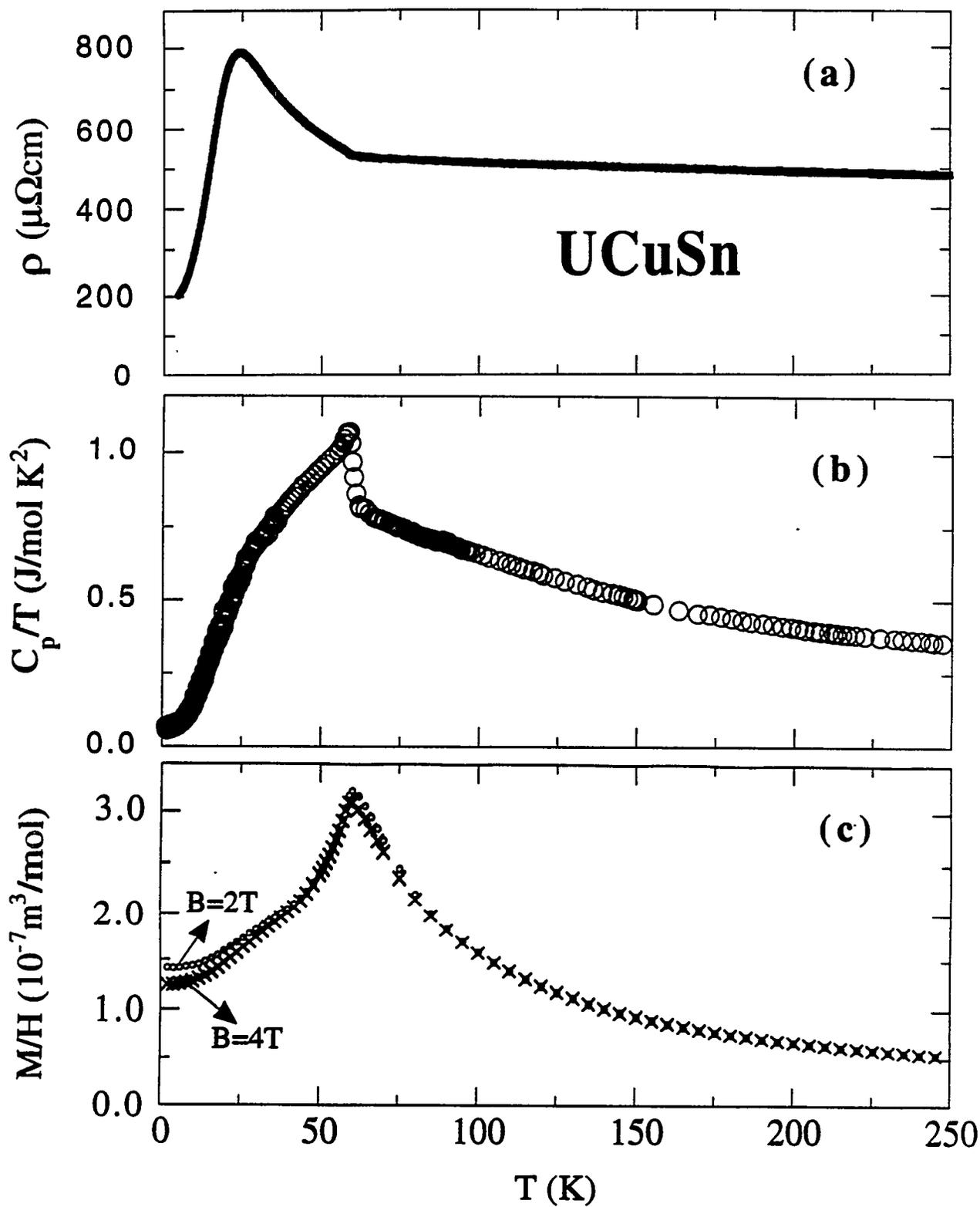


Fig. 2

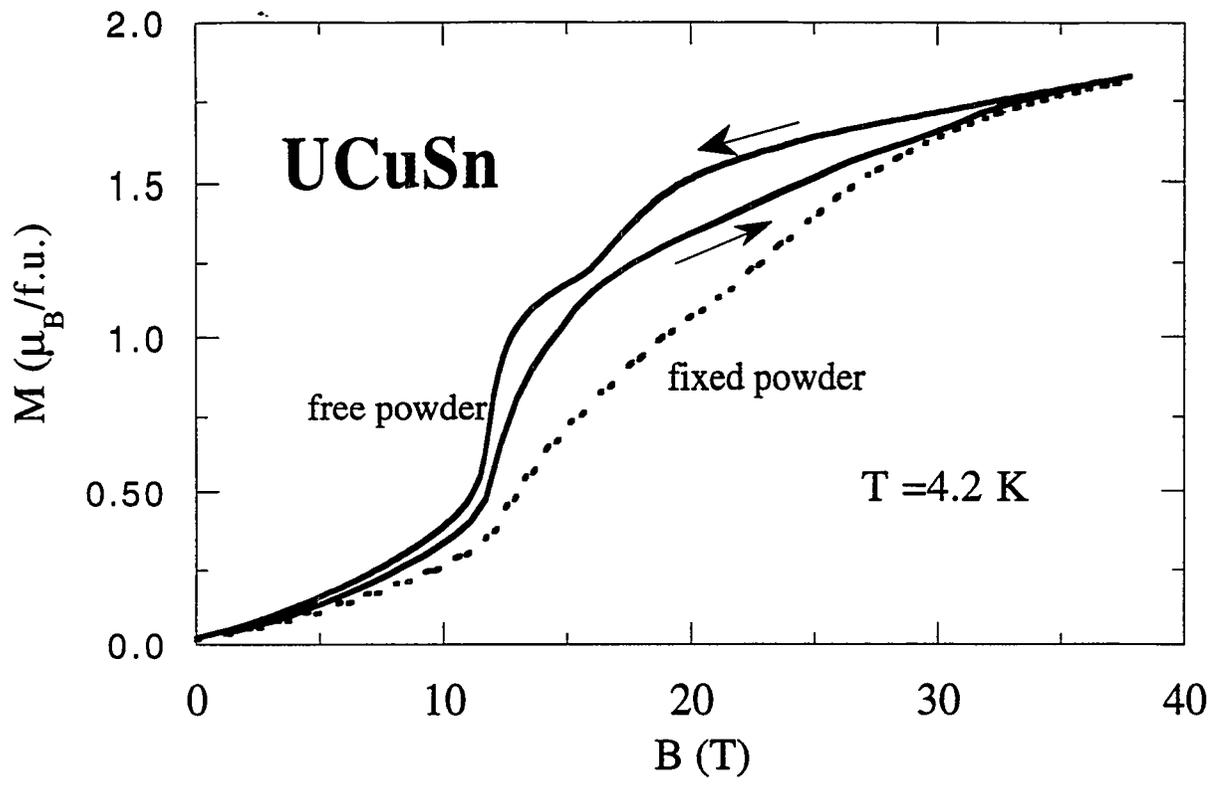


Fig 3

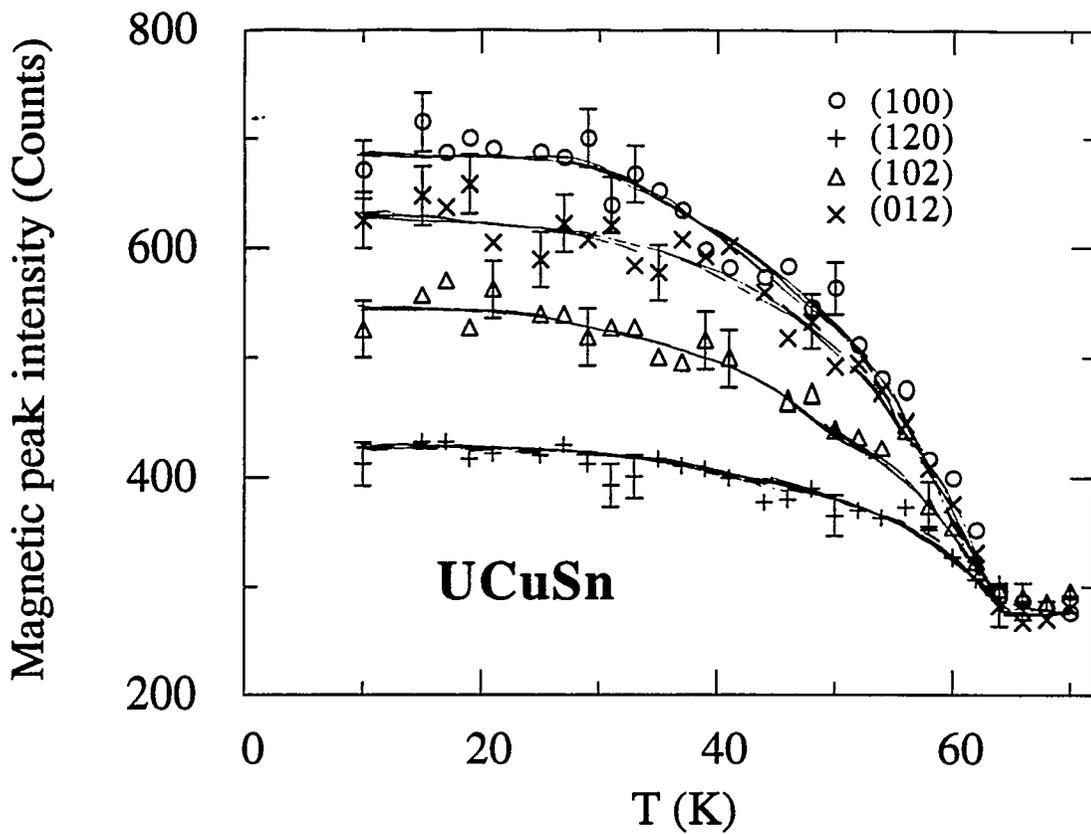


Fig. 4

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.