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ELECTRONIC PROPERTIES OF  $U_2Pt_2Sn$ 

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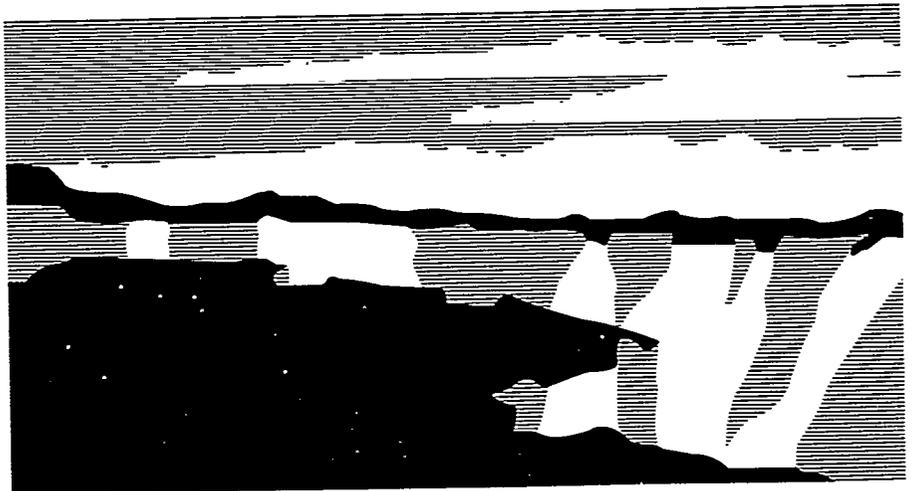
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# ELECTRONIC PROPERTIES OF $U_2Pt_2Sn$

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## Abstract

$U_2Pt_2Sn$  is crystallizing in an ordered version (space group  $P4_2/mnm$ ) of the tetragonal  $U_3Si_2$ -type of structure. Clear anomalies in the magnetic susceptibility, specific heat and electrical resistivity around 15 K indicate that  $U_2Pt_2Sn$  orders antiferromagnetically below this temperature. As expected for an antiferromagnet, the susceptibility and specific heat anomalies are shifted to lower temperatures upon application of external magnetic field. The specific-heat coefficient  $\gamma = 327$  mJ/mol f.u.  $K^2$  remains unchanged in fields up to 5T. The antiferromagnetic ground state of  $U_2Pt_2Sn$  can be concluded also from the metamagnetic transition around 22 T in the magnetization at 4.2 K and from magnetic reflections in the neutron-diffraction pattern at low temperatures.

PACS: 7525, 7530 C, 7550 C

## I. INTRODUCTION

Recently, great attention has been given to the isostructural group of  $U_2T_2X$  compounds ( $T =$  transition metal and  $X =$  In, Sn, Al or Ga)<sup>1,2,3</sup>. The development of their magnetic properties (from Pauli paramagnetism to local-moment behavior) is influenced by the hybridization of the 5f-electron states with ligand electron states. The 5f-d hybridization is demonstrated by the evolution of magnetism with respect to the transition metal component<sup>1,4,5</sup>.

$U_2Pt_2Sn$  has been first reported to adopt the tetragonal  $U_3Si_2$ -type structure (space group  $P4/mnm$ )<sup>6</sup>. More recent studies pointed out a derivative form with the space group  $P4_2/mnm$ <sup>7</sup>. Both related structures consist of two alternating plane sheets, one containing only U atoms and the other accommodating T and X atoms. The shortest interatomic U-U distance in  $U_2Pt_2Sn$  is about 350 pm, which places this compound into a critical region between localized and itinerant 5f-electron behavior in Hill's classification<sup>8</sup>.

The aim of the present contribution is to investigate the type of the crystal structure, the type of the ground state, as well as to study the magnetic, transport and thermal properties of polycrystalline  $U_2Pt_2Sn$ .

## II. EXPERIMENTAL

A polycrystalline sample of  $U_2Pt_2Sn$  has been prepared by arc-melting appropriate amounts of the constituting elements under argon atmosphere. The purity and composition homogeneity were checked by X-ray diffraction and by electron-microprobe analysis. The microprobe indicated less than 1 vol. % of UPtSn as a secondary phase. The average composition of the main phase only slightly deviates from the ideal stoichiometric composition of  $U_2Pt_2Sn$ . A slight U excess and Sn deficiency, both of the order of 1 at. %, were found.

The low-field magnetization and the temperature dependencies of the dc magnetic susceptibility ( $\chi = M/H$ ) were measured between 2 and 300 K in magnetic fields up to 5 T in a Quantum Design SQUID magnetometer on a fine-powder sample with grains fixed in random orientation by diamagnetic glue.

The specific heat was measured by a standard semi-adiabatic method in the temperature region 1.3 - 50 K in fields up to 5 T.

The electrical resistivity was measured on a small bar-shaped sample by standard ac four-point method. The large error in the geometrical factor (in addition to the expected influence of internal microcracks) makes a reliable estimate of the absolute resistivity values questionable. Therefore, we display the normalized electrical resistivity

$\rho/\rho_{300K}$ .

Neutron-diffraction data at 2.6 and 24 K were obtained in the neutron powder diffractometer L6 at the Jahn-Meitner Institute using an incident neutron wavelength of 2.386 Å. For these measurements about 10 g of  $U_2Pt_2Sn$  was powdered and encapsulated in a vanadium container under He atmosphere. The data were analyzed by means of the Rietveld profile procedure<sup>9</sup> using the program Fullprof<sup>10</sup>. The neutron-scattering lengths were taken from ref.<sup>11</sup>.

## III. BULK MEASUREMENTS

The susceptibility (Fig. 1.) above 40 K is nearly field independent, whereas the  $\chi$  vs. T curves show splitting in the low-temperature region reflecting an increasing amount of a ferromagnetic component with lowering temperature. An upturn becomes progressively pronounced with decreasing magnetic field (the inset of Fig. 1). We tentatively attribute this spurious signal to the UPtSn impurity, which is ferromagnetic below 19 K<sup>12</sup>. In Fig. 1, then the susceptibility data extrapolated to infinite magnetic field are shown. Two regions on the  $1/\chi$  vs. T above 40 K can be identified: one linear above 160 K, which can be fitted by Curie-Weiss (CW) law

$$\chi = C / (T - \Theta_p) \quad (1),$$

and another strongly curved at lower temperatures which can be approximated only by a modified Curie-Weiss law (MCW)

$$\chi = C / (T - \Theta_p) + \chi_0 \quad (2).$$

In the former case, the fitting parameters are:  $C = (2.56 \pm 0.01) \cdot 10^{-5} \text{ m}^3/\text{mol f.u. K}$  ( $\Rightarrow \mu_{\text{eff}} = 2.02 \pm 0.01 \mu_B/\text{U atom}$ ) and a negative paramagnetic Curie temperature of -141.5

$\pm 1.6$  K. The MCW fit below 160 K is yielding a significantly lower value of  $C = (1.11 \pm 0.01) \cdot 10^{-5} \text{ m}^3/\text{mol f.u. K}$  ( $\mu_{\text{eff}} = 1.33 \pm 0.01 \mu_B/\text{U atom}$ ), a fairly less negative  $\Theta_p = -6.9 \pm 0.6$  K, whereas  $\chi_0 = 2.9 \cdot 10^{-8} \text{ m}^3/\text{mol f.u.}$

These fits should be taken with caution, because the polycrystalline data on highly anisotropic material as most of the  $\text{U}_2\text{T}_2\text{X}$  compounds are, may represent an admixture of very different anisotropic components of the susceptibility. These components may follow the CW law, however, the  $1/\chi$  vs.  $T$  dependence measured on polycrystal is strongly curved especially at lower temperatures. Nevertheless, the high-temperature CW fit provides a good estimate of the upper limit of  $\mu_{\text{eff}}$  (although, even this value is much smaller than the values expected for the localized  $f^{3+}$  or  $f^{4+}$  configuration). The strikingly different values of  $\Theta_p$  from the high-temperature CW fit and the low temperature MCW fit serve rough estimates of paramagnetic Curie temperatures for the hard- and easy-magnetisation direction, respectively. The  $\mu_{\text{eff}}$  value from the low temperature fit is only a fraction of the genuine effective moment and depends on the multiplicity of the easy axis. The "temperature independent parameter"  $\chi_0$  is then an arbitrary value without any deeper physical meaning.

The magnetic phase transition at  $T_N = 15$  K is reflected by the peak in the  $C_p/T$  vs  $T$  curve (Fig. 2.a). The peak is shifted by 0.3 K in 5 T as expected for an antiferromagnet. The linear coefficient of the specific heat  $\gamma = 327.0 \pm 0.3 \text{ mJ/mol f.u. K}^2$  follows from the linear extrapolation of  $C_p/T$  vs.  $T^2$  plot in the region 1.3 - 8.5 K to zero Kelvin. This value which accounts for two U atoms in the formula unit is not affected by magnetic field to 5 T. The value of 163 mJ/mol  $\text{K}^2$  calculated per one U atom is certainly enhanced with respect to normal non-magnetic metals.

The onset of antiferromagnetic ordering at  $T_N = 15$  K is reflected also in the electrical resistivity (Fig. 2.b). The electrical resistivity is slightly increasing with lowering temperature and passing a shallow maximum around 35 K. This behavior is commonly observed in strongly hybridized U-based materials. After some precursor behavior around  $T_N$  the resistivity drops at lower temperatures. No quadratic temperature dependence is observed down to 4.2 K.

The magnetization curves of  $\text{U}_2\text{Pt}_2\text{Sn}$  at 4.2 K show a metamagnetic transition at about 22 T<sup>13</sup>. This result corroborates the conclusion about the antiferromagnetic ground state. The magnetization for the field-aligned (random-oriented) powder sample yields a value of 1.49 (1.23)  $\mu_B$  /f.u. in 56 T. The magnetization curve is far from saturation even in this very high magnetic field. We cannot exclude that another metamagnetic transition may occur in higher fields and therefore, do not dare making any conclusion about the type of anisotropy from comparison of present data obtained on the field-aligned and random oriented samples.

The low-field magnetization exhibits a small hysteresis loop developing below 20 K, which is believed to be due to the ferromagnetic UPtSn impurity.

#### IV. NEUTRON DIFFRACTION RESULTS

As two possible crystal structures for  $\text{U}_2\text{Pt}_2\text{Sn}$  have been reported in the literature, the determination of the exact crystal structure was our first task in the neutron-diffraction experiment. Two tetragonal crystallographic structures were considered: the ideal tetragonal  $\text{U}_3\text{Si}_2$ -type of structure (space group  $P4/mbm$ ) and its variant, in which U atoms are forced by relatively large Pt atoms to move from their original positions forming

thus zig-zag chains along the c-axis (space group  $P4_2/mnm$ ). The pattern recorded (Fig. 3.) above the transition temperature, at 24 K, can be fully indexed by using the latter type of structure. The structural parameters are summarized in Table. I. Reflections of the very weak UPtSn secondary phase cannot be detected.

In the spectrum recorded at 2.6 K, new - purely magnetic reflections and additional magnetic contributions on some of the nuclear Bragg reflections (see the difference spectrum in Fig. 4.) can be resolved. This clearly indicates that the magnetic periodicity is different from the crystallographic one, which is compatible with the antiferromagnetic ordering suggested from bulk experiments. The magnetic structure refinement is difficult from the available powder pattern. In this stage, no magnetic unit cell which would index all observed magnetic peaks could be deduced. Single-crystal data are desirable to choose a proper model out of many, which are roughly satisfying the powder data.

## V. CONCLUSIONS

By means of powder neutron diffraction, we have shown that  $U_2Pt_2Sn$  crystallizes in an ordered variant (space group  $P4_2/mnm$ ) of the tetragonal  $U_3Si_2$ -type of structure and that it orders antiferromagnetically at low temperatures ( $T_N = 15$  K). The results obtained by means of the magnetic-susceptibility, the specific-heat and the electrical-resistivity suggests that the magnetism in  $U_2Pt_2Sn$  is governed by to certain extent delocalized 5f-electron states in U atoms. However, among the  $U_2T_2X$  compounds studied, this material can be considered as an example of a system with rather well-defined U magnetic moments. In order to elucidate the magnetic structure and specific features of magnetic anisotropy, single-crystalline  $U_2Pt_2Sn$  are strongly desirable.

## ACKNOWLEDGMENTS

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### Figure Caption

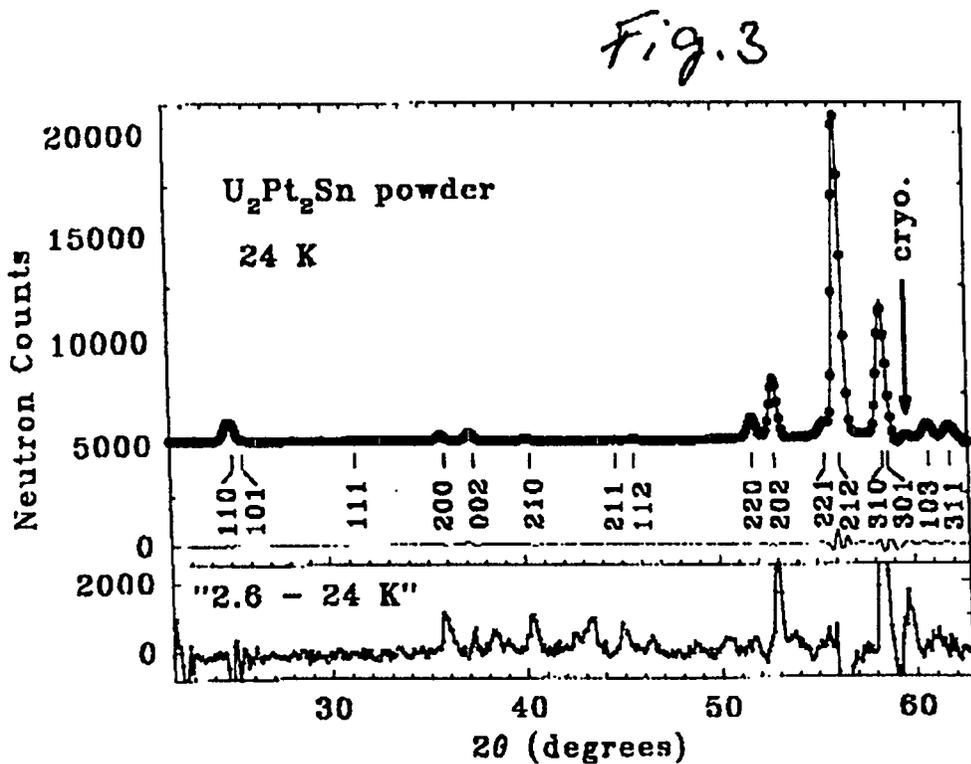
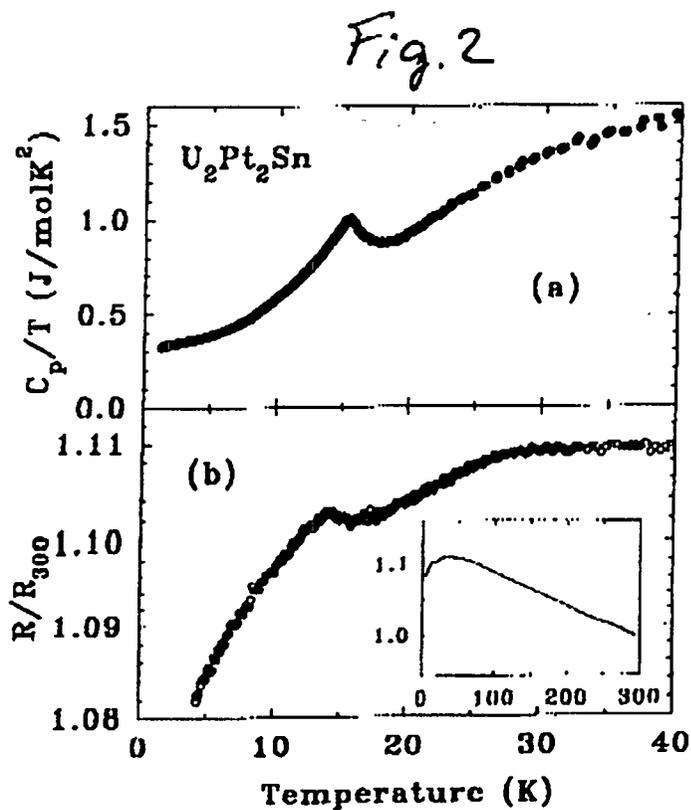
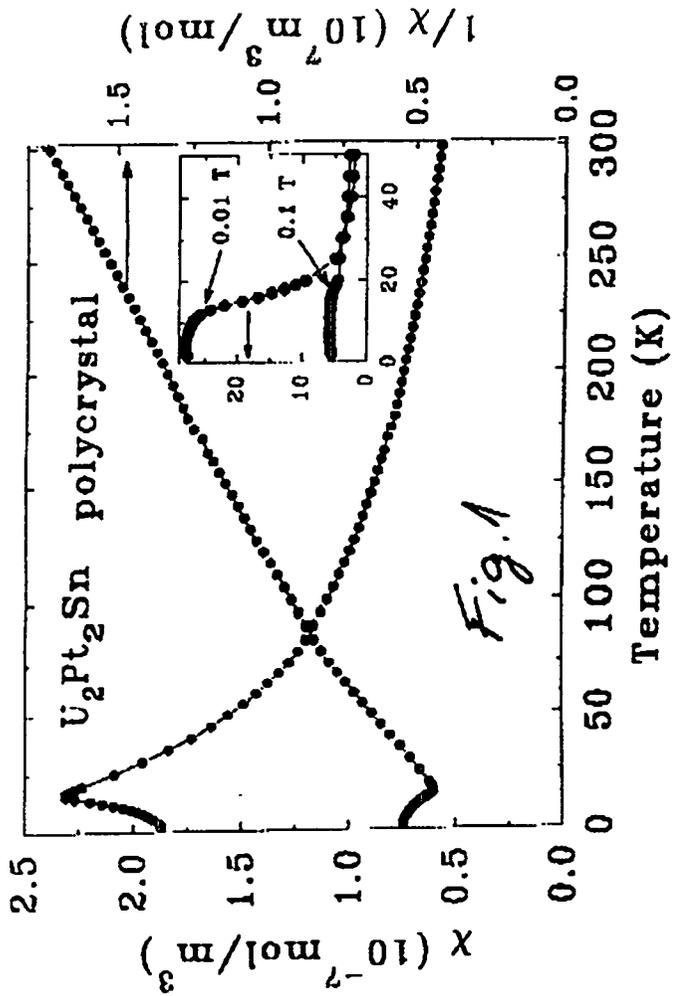
- Fig.1.:** Temperature dependence of the magnetic susceptibility of  $U_2Pt_2Sn$  extrapolated to infinite magnetic field together with its reciprocal value. In the inset the data collected in 0.01 and 0.1 T are shown.
- Fig.2.:** Temperature dependence of the specific heat of  $U_2Pt_2Sn$  in the representation  $C_p/T$  vs. T (a) and the electrical resistivity (b) in zero magnetic field.
- Fig.3.:** The neutron-diffraction spectrum of  $U_2Pt_2Sn$  recorded at 24 K together with the best fit. The full line at the bottom represents the difference between the experimental data and the fit. The 2.6 - 24 K spectrum is displayed, as well.

### Table Caption

TABLE I. The refined structural parameters of  $U_2Pt_2Sn$  at 24 K.

TABLE I.

Space group:	$P4_2/mnm, Z = 2$			T = 24 K
U1	4f	$x_{U1} \ x_{U1} \ 0$	$x_{U1} = 0.3414 (7)$	
U2	4g	$x_{U2} \ -x_{U2} \ 0$	$x_{U2} = 0.2044 (30)$	
Pt	8j	$x_{Pt} \ x_{Pt} \ z_{Pt}$	$x_{Pt} = 0.1302 (4)$	$z_{Pt} = 0.2208 (8)$
Sn	4d	$0 \ 1/2 \ 1/4$		
<b>Lattice parameters:</b>			<b>R factors:</b>	
$a = 770.496 \pm 0.285$ pm			$R_p = 5.43 \%$	
$c = 740.760 \pm 0.296$ pm			$R_{wp} = 8.16 \%$	
			$R_B = 1.95 \%$	
			$\chi^2 = 3.30$	



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