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DOI

[10.1016/0304-8853\(94\)00606-7](https://doi.org/10.1016/0304-8853(94)00606-7)

Publication date

1995

Published in

Journal of Magnetism and Magnetic Materials

[Link to publication](#)

Citation for published version (APA):

Kindo, K., Fukushima, T., Kumada, T., de Boer, F. R., Nakotte, H., Prokes, K., Havela, L., Sechovsky, V., Seret, A., Winand, J. M., Spirlet, J. C., & Rebizant, J. (1995). Electronic properties of U₂Ni₂Sn. *Journal of Magnetism and Magnetic Materials*, 140-144, 1369-1370. [https://doi.org/10.1016/0304-8853\(94\)00606-7](https://doi.org/10.1016/0304-8853(94)00606-7)

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Electronic properties of U_2Ni_2Sn

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Abstract

We report on the bulk electronic properties of U_2Ni_2Sn , which crystallizes in the tetragonal U_3Si_2 structure. Pronounced anomalies in the temperature dependencies of magnetic susceptibility, specific heat and electrical resistivity indicate magnetic ordering below $T_N = 26$ K. An antiferromagnetic ground state is corroborated by the occurrence of three metamagnetic transitions at about 30, 39 and 51 T in the field dependence of the magnetization. The lack of any saturation tendency in the highest field applied (57 T) may suggest further transition(s) to occur in even higher fields.

Recently, a new large isostructural group of uranium compounds consisting up to now of 18 compounds with stoichiometry U_2T_2X ($T = Fe, Ru, Co, Rh, Ir, Ni, Pd, Pt$ and $X = In, Sn, Al$ and Ga) has been discovered [1,2]. Intensive investigations of magnetic properties of these compounds reveal that they are governed by the hybridization of the 5f-electron states with neighbouring 5f states, with d states of transition element, with p states of constituting X-element and by hybridization with the conduction electrons. As the strength of 5f–5f hybridization is relatively weak with respect to 5f–d and 5f–p hybridization [3], the magnetic properties are influenced mainly by 5f–d hybridization and one can follow the development of the magnetic properties (from Pauli paramagnetism in U_2Co_2In to heavy-fermion behaviour in U_2Pt_2In) with respect to constituting T element in the In or the Sn containing compounds [4,5].

The ternary stannide U_2Ni_2Sn , as one member of this group, was prepared by arc-melting appropriate amounts of the constituting elements. It adopts the ordered version of the tetragonal U_3Si_2 -type of structure (Fig. 1) (space group $P4/mbm$, $Z = 2$). The uranium atoms occupy 4h positions ($x, x + 0.5, 0.5$) with $x = 0.1743$, Ni takes 4g positions ($y, y + 0.5, 0$) with $y = 0.3756$ and Sn occupies 2a positions (0, 0, 0). The lattice parameters are $a = 726.3$ pm and $c = 369.1$ pm. Each U atom has one nearest U

neighbour at a distance of 358.1 pm, two next-nearest U neighbours along the c-axis at a distance of $c = 369.1$ pm and four third-nearest U atoms within the basal plane at a distance of 379.4 pm. There are two Ni atoms at a distance of 277.2 pm and four others 284.8 pm apart. Finally, each U atom has four Sn neighbours at a distance of 327.1 pm. The structure, cell parameters and atomic positions were determined on a small single crystal by means of an Enraf–Nonius four-circle diffractometer [2].

U_2Ni_2Sn was investigated by means of electrical-resistivity (4.2–300 K), magnetic-susceptibility (4.2–300 K) and specific-heat (1.3–50 K) measurements. All results indicate that this compound exhibits antiferromagnetic ordering below $T_N = 26.0$ K [4,5] (Fig. 2). Above 150 K, the temperature dependence of the magnetic susceptibility was fitted to a Curie–Weiss law, resulting in $\mu_{eff} = 3.11\mu_B$

Fig. 1. Schematic representation of the structure of U_2Ni_2Sn .

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and a negative paramagnetic Curie temperature of -219 K. The specific heat reveals an enhanced electronic contribution. The coefficient γ deduced by extrapolation of C_p/T to zero Kelvin amounts to 172 mJ/mol K² f.u. The electrical resistivity measured on a bar-shaped sample exhibits a negative slope at high temperatures, which is common to hybridized U compounds, and decreases upon approaching the transition at $T_N = 26$ K.

The magnetization curves of U_2Ni_2Sn were measured on two kinds of samples: **a**, on powder free to orient itself in the external magnetic field, and **b**, on powder fixed by frozen alcohol and thus representing an ideal polycrystal. As the magnetization curve for free powder taken in the High-Field Installation at the University of Amsterdam in semicontinuous field sweeps up to 35 T showed only one metamagnetic transition at around 30 T, the magnetization measurements were extended at the High-Field Facility at Osaka University in short pulsed fields up to 57 T. Three metamagnetic transitions at 30 , 39 and 51 T are detected at 4.2 K reaching for free powder a magnetization of $1.3\mu_B$ /f.u. at 57 T. (Fig. 3). For powder fixed by frozen alcohol no metamagnetic transitions are visible. The results from both high-field experiments are consistent and yield the same values for the magnetization. The lack of any tendency to saturation at the highest fields suggests further transitions to occur even at higher fields.

A neutron-diffraction study [6] has shown that the magnetic unit cell of U_2Ni_2Sn is twice as large as the chemical one and that the U magnetic moments are perpendicular to the c -axis, forming a collinear antiferromagnet.

Fig. 2. Temperature dependence of the magnetic susceptibility (a), the specific heat (b), and the electrical resistivity (c) of U_2Ni_2Sn .

Fig. 3. Field dependence of the magnetization of U_2Ni_2Sn for powder free to orient in the external field (Δ) and for powder fixed by frozen alcohol ($\circ$). The lines represent continuous field sweeps.

netic structure. The size of the U moment is reported to be $1.05\mu_B$ at 1.5 K which is significantly higher than the value obtained in the magnetization measurements.

The fact that in the magnetization curve for free powder three metamagnetic transitions are observed without any sign of saturation at the highest fields suggests that the magnetization processes in U_2Ni_2Sn are very complex. All results show that the magnetic properties are affected by the $5f$ ligand hybridization. For final conclusions more experimental results are necessary.

Acknowledgements: This work is part of the research program of the 'Stichting voor Fundamenteel Onderzoek der Materie (FOM)', by the Grant Agency of The Czech Republic (grant no. 202/93/0184). Support to A.S. and J.M.W. given in the framework of the EC funded training program HC&M is acknowledged.

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