MAGNETIZATION STUDY OF UNiSi AND ITS HYDRIDE*

J. Šebek, A.V. Andreev

Institute of Physics ASCR
Na Slovance 2, 18221 Prague 8, The Czech Republic

F. Honda, A.V. Kolomiets, L. Havela, and V. Sechovský

Department of Electronic Structures, Charles University
Ke Karlovu 5, 12116 Prague 2, The Czech Republic

(Received July 10, 2002)

We have grown a single crystal of UNiSi and measured the magnetization and susceptibility as functions of temperature and magnetic field applied along the principal crystallographic axes. Magnetic ordering with a relatively weak easy-plane anisotropy is indicated below 90 K. In addition, we present results of investigation of hydrogen absorption on the crystal structure and magnetic properties of UNiSi.

PACS numbers: 75.30.–m, 75.50.Cc, 61.10.–i

1. Introduction

UNiSi is one of uranium intermetallics UTX (T is a late transition metal, X is Si or Ge), which crystallize in the orthorhombic TiNiSi-type structure. Three magnetic phase transitions were deduced from magnetization and specific-heat data obtained on polycrystalline samples. In the ground state, UNiSi exhibits a spontaneous magnetization, which means a ferromagnetic phase or a phase with a ferromagnetic component. Specific-heat data indicate an electronic specific-heat coefficient $\gamma = 134$ mJ/(mol*K^2) [1]. Here we present the results of first single-crystal magnetization study of UNiSi. In addition, we report on the influence of hydrogen absorption on the crystal structure and magnetic properties of this compound.

* Presented at the International Conference on Strongly Correlated Electron Systems, (SCES 02), Cracow, Poland, July 10–13, 2002.

2. Experimental

A single-crystal ingot of UNiSi was pulled out from the stoichiometric melt by a modified Czochralski method in a tetra-arc furnace. The product was found very high brittle and containing many cracks. Therefore, it spontaneously crumbled to rather small grains within short time. Nevertheless, we succeeded to extract a single crystalline sample large enough (35 mg) for the magnetization and magnetic susceptibility measurements. The X-ray powder diffraction of neighboring parts of ingot confirmed the single-phase composition with the TiNiSi-type structure and lattice parameters in good agreement with literature. The magnetization (magnetic susceptibility) along the principal axes was measured in the SQUID magnetometer (Quantum Design) in fields up to 5 T in the temperature range 5–300 K.

3. Results

Figure 1 shows the magnetization curves measured at 5 K in fields along the principal axes. Contrary to other UTX compounds, UNiSi exhibits rather weak magnetic anisotropy, which is of the easy-plane type. The magnetization data along the b- and c-axis, respectively, are nearly identical. The virgin magnetization curves show an S-shape in low fields. Above 1 T, the b- and c-axis magnetization further increases with magnetic field and reach more than 0.7 μ_B/f.u. in 5 T while showing only a moderate tendency to saturation. The hysteresis of magnetization curves leads to a remanent magnetization of 0.12 μ_B/f.u. (at 5 K), which indicates a ferromagnetic component in the magnetic structure. The S-shape and hysteresis in easy $b-c$ plane disappear above 10 K. The a-axis magnetization increases with field almost linearly up to a considerably lower value of 0.3 μ_B/f.u. in 5 T. The temperature dependence of the b- and c-axis magnetization, respectively (see also Fig. 1), measured in low fields indicate magnetic history effects below 90 K. In the literature, three magnetic phase transitions are stated for UNiSi, at 10, 18 and 87 K [1]. We confirmed that there are some anomalies at all temperatures, but conclusion whether the phase transitions indeed occur will be done after neutron-diffraction study of the single crystal. In the paramagnetic range, the magnetic susceptibility (Fig. 2) obeys the Curie–Weiss law (above ~140 K) with $\mu_{\mathrm{eff}} = 3.05\ \mu_B$ and $\Theta_p = -17$ K (a-axis), -24 K (b-axis) and -50 K (c-axis). Preliminary specific-heat data show no distinct peak-like anomaly that is usual in case of a magnetic phase transition, nevertheless some features can be recognized in the temperature derivative of specific heat around 85 K and below 20 K. The C/T *vs.* T^2 is linear below 10 K and points, neglecting the magnetic contribution, to $\gamma = 166$ mJmol^{-1}K^{-2}. All these results point to weak itinerant $5f$-electron magnetism.

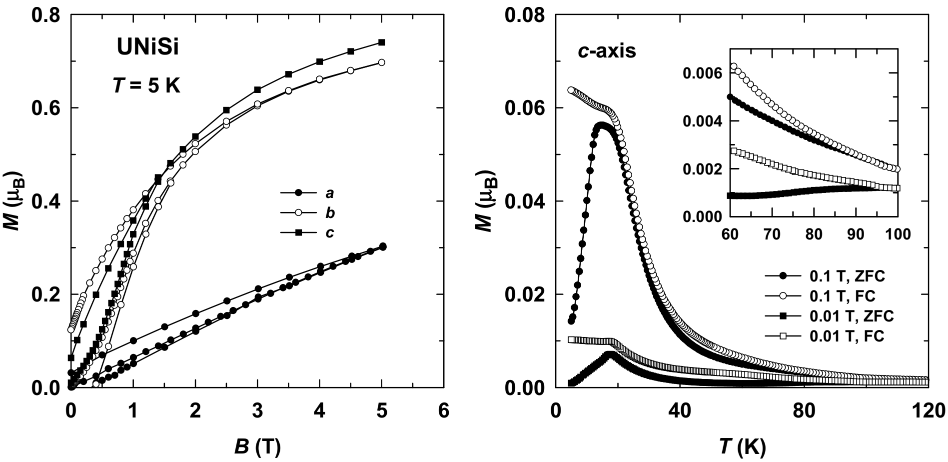

Fig. 1. Magnetization curves along the principal axes of the UNiSi crystal at 5 K (left) and temperature dependence of magnetic moment along the c axis in 0.01 T and 0.1 T (right). The inset shows enlarged detail of the M *vs.* T dependence around 80 K. The full (empty) symbols represent ZFC (FC) data.

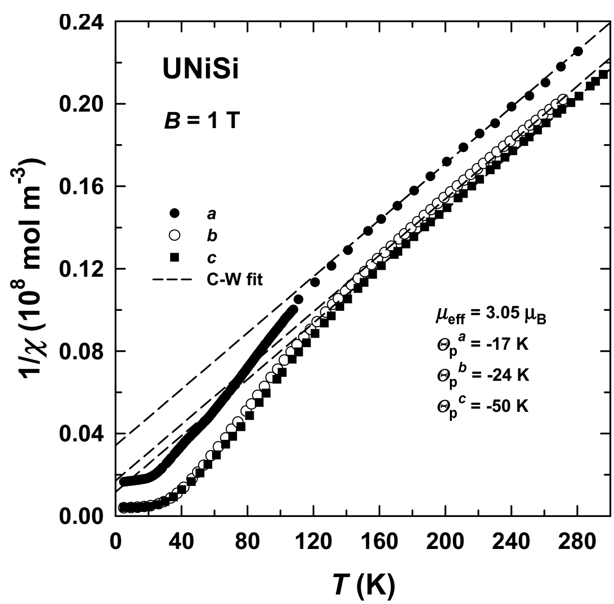

Fig. 2. Temperature dependence of inverse magnetic susceptibility measured along the principal axes in a 1 T field. The dashed lines represent the Curie–Weiss fits for each direction.

UNiSi absorbs hydrogen at rather high temperature (923 K) and hydrogen pressure (130 bar). The as-synthesized compound contains about 2 H at./f.u., but the stable composition is approximately $\mathrm{U}T\mathrm{SiH}_{1.0}$. The hydrogen absorption leads to the expansion of lattice by 8 % , which is accompanied by a symmetry change towards the hexagonal ZrBeSi-type structure. The original structure is restored when hydrogen is removed from the lattice by heating at temperatures above 923 K in vacuum. As seen in Fig. 3, hydrogenation yields ferromagnetism (or a state with ferromagnetic component) over a wide temperature range and enhancement of T_C up to 98 K. Strong magnetic history effects are found also in the hydride sample. $M(T)$ dependences indicate two possible phase transitions at T_1 and T_2. Result of a more detailed study of the hydride will be published elsewhere.

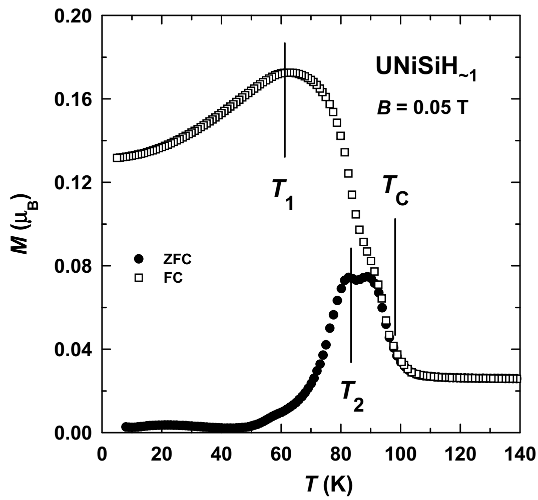

Fig. 3. Temperature dependence of magnetic moment of the UNiSiH in 0.05 T. The full (empty) symbols represent ZFC (FC) data.

This work is a part of the research program MSM113200002 that is financed by the Ministry of Education of the Czech Republic. It was also partly supported by the grant GACR 202/02/0739.

REFERENCES

[1] V. Sechovský, L. Havela, *Handbook of Magnetic Materials*, Elsevier, Amsterdam 1998, **11**, p.1. and references therein.