ELSEVIER

Physica B 230–232 (1997) 108–110

PHYSICA B

Thermodynamic study of the magnetic phase transition in UNi_4B

S.A.M. Mentink[a,*], H. Amitsuka[b,c], A. de Visser[b], Z. Slanič[d], D.P. Belanger[d], J.J. Neumeier[e], J.D. Thompson[e], A.A. Menovsky[b,f], J.A. Mydosh[f], T.E. Mason[a]

[a]*Department of Physics, University of Toronto, 60 Saint George Street, Toronto, Ont., Canada M5S 1A7*
[b]*Van der Waals–Zeeman Laboratory, University of Amsterdam, The Netherlands*
[c]*Department of Physics, Hokkaido University, Sapporo 060, Japan*
[d]*Department of Physics, University of California, Santa Cruz, CA 95064, USA*
[e]*Los Alamos National Laboratory, Los Alamos, NM 87545, USA*
[f]*Kamerlingh Onnes Laboratory, Leiden University, The Netherlands*

Abstract

We investigate the magnetic phase transition in UNi_4B by specific heat, $c(T)$, thermal expansion and resistivity under pressure. These experiments reveal that the hexagonal symmetry is not broken at $T_N = 20$ K, and predicts a hydrostatic pressure dependence of T_N of -29 mK/kbar, close to the result from resistivity. The anomalous upturn of c/T to 470 mJ/mol K^2 at 0.4 K is reduced to 160 mJ/mol K^2 in 16 T $\parallel$ b-axis. The specific heat critical exponent $\alpha = -0.15(2)$ suggests 3D Heisenberg universality in zero field.

Keywords: UNi_4B; Specific heat; Thermal expansion; Magnetic phase transition; Phase diagram

The understanding of the mechanism that stabilizes the partially ordered magnetic structure of UNi_4B is still incomplete [1,2]. The crystallographic superstructure creates two U-atoms of different site symmetry, which are either magnetically ordered if twofold symmetric, or paramagnetic if sixfold symmetric. The antiferromagnetic (AF) interactions in this metallic compound are long range, and therefore in principle frustrated in the nearly triangular magnetic sublattice. Such frustration, in combination with proximity to a magnetic–nonmagnetic transition and strong anisotropy has been shown theoretically to lead to possible stable "mixed phases", with sites of ordered moments and zero-moment sites [3]. They are realized in, e.g., $DyMn_2$ [3] and CeSb [4]. Here we present a detailed study of thermal expansion and specific heat to determine anisotropy and critical exponent. Hydrostatic pressure was applied to investigate the proximity to a nonmagnetic state.

All experiments were performed on single crystals. The specific heat was measured in magnetic fields up to 16 T $\parallel$ b axis ($\perp$ a axis in the basal plane). Fig. 1 shows c/T from 0.4 to 25 K in 0 and 16 T, which is above 8 T, where a transition to a spin-reorientation phase occurs [5]. In zero field, the AF phase transition is λ-shaped. Below 7 K, c/T increases down to the lowest measured $T = 0.35$ K; at 0.4 K, $c/T = 470$ mJ/mol K^2. No further phase transition was observed down to 1 mK [5], and the lattice contribution at this temperature is negligible [1]. c/T follows the same power law $\alpha T^{-0.29}$ found earlier [1] down to 0.65 K, but increases faster at lower T. The presence of this large entropy at low T implies the existence of residual magnetic moments on the nonorderning U-sites. Upon applying a field (at $T = 0.4$ K), c/T first increases slightly,

---

* Corresponding author.

PII S0921-4526(96)00561-3

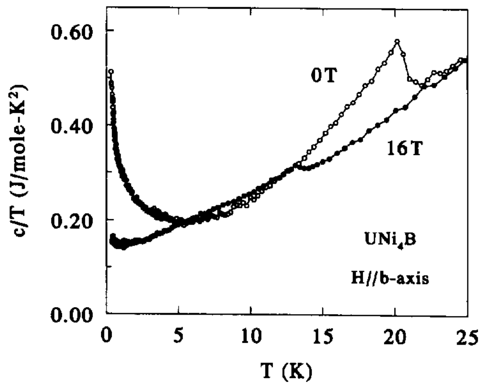

Fig. 1. Specific heat divided by temperature of UNi₄B, measured in zero field (○) and 16 T ∥ b-axis (●).

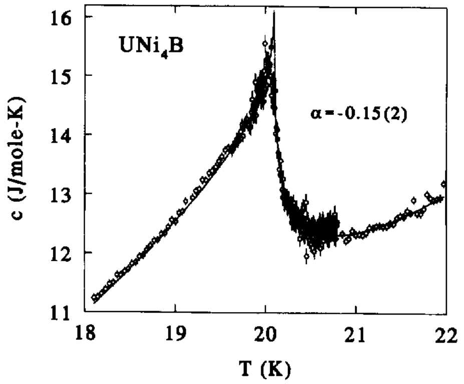

Fig. 2. High-resolution specific heat data of the antiferromagnetic transition in UNi₄B. The solid line is a fit to Eq. (1), yielding $\alpha = -0.15 \pm 0.02$.

with a maximum near 2.5 T, then falls off, with a kink at 8 T, before slowly decreasing to 160 mJ/mol K^2 at 16 T [6]. From the decrease with field we infer that the Kondo effect, acting on the disordered moments, is most likely responsible for the large γ. A qualitative change is observed in the 16 T data. The anomaly at $T_N = 13.1$ K is very small and the T dependence of c/T below T_N changes from αT^2 to αT, indicative of a reduction of dimensionality of the spin waves from three to two. In units of $R\ln 2$, the total entropy at 25 K equals 1.55 for $B = 0$ and 1.36 for B 16 T, indicating a shift of the entropy towards higher temperature.

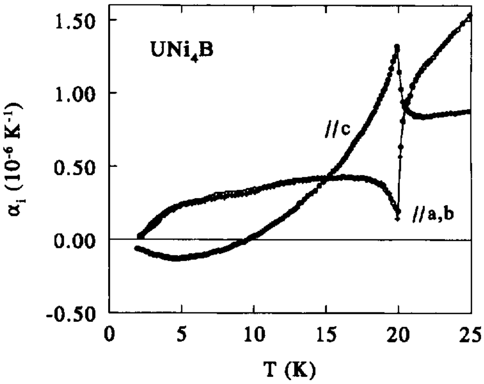

Fig. 3. Thermal expansion of UNi₄B parallel to the a (○), b (+), and c (●) axes in zero magnetic field.

Such a change in character of the phase transition will be reflected in the critical behaviour. A high-resolution $c(T)$ measurement on a carefully annealed sample in $H = 0$ is shown in Fig. 2. By fitting the data to within 50 mK above and below $T_N = 20.10$ K (fixed in the fit) to the expression:

$$c/R = \frac{A_{+,-}}{\alpha}|t|^{-\alpha} + B + CT + DT^2, \qquad (1)$$

where $t = (T - T_N)/T_N$ and R the gas constant, we find the critical exponent $\alpha = -0.15 \pm 0.02$ and the amplitude ratio $A_+/A_- = 1.78 \pm 0.10$. These values are expected for 3D Heisenberg universality [7,8], and in accord with the order parameter exponent $\beta = 0.38(2)$ [1]. We expect a change of universality when crossing the spin-reorientation field of 8 T [6].

Fig. 3 shows the thermal expansion coefficients, α_i, for three crystal directions, a, b and c, in zero field. At T_N, α_i show λ-type anomalies with opposite direction. Since $\alpha_a = \alpha_b$, the crystal symmetry remains hexagonal through T_N. Using the Ehrenfest relation:

$$\left(\frac{dT_N}{dp_i}\right)_{B,p'} = \frac{V_m \Delta\alpha_i}{\Delta(c/T)}, \qquad (2)$$

we calculate, with $V_m = 4.45 \times 10^{-5}$ m^3/mol, $dT_N/dp_c = +14.9(6)$ mK/kbar and $dT_N/dp_{a,b} = 22.1(8)$ mK/kbar. The hydrostatic pressure derivative of T_N then equals $dT_N/dp_c = -29(2)$ mK/kbar.

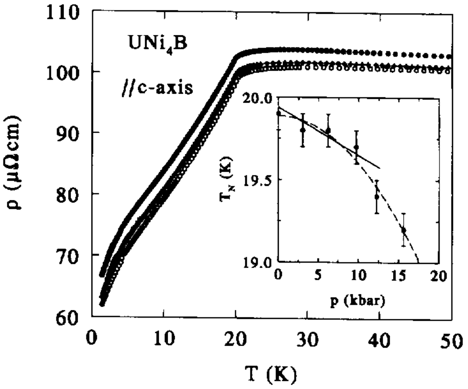

Fig. 4. Resistivity of UNi$_4$B ∥ c-axis under hydrostatic pressures of 0 (○), 9.7 (+) and 15.6 kbar (●). The inset shows T_N versus p, with the solid line the initial pressure dependence as calculated from Eq. (2). The dashed line is a guide to the eye.

The increase of c/T at low T is reflected by the change of sign of α_c, implying an expanding c-axis below 10 K.

Fig. 4 displays the resistivity along the c-axis, measured in $H = 0$ at various hydrostatic pressures up to 15.6 kbar. T_N is determined within 0.1 K by the maximum of $d\rho_c/dT$ (not shown), and decreases from 19.9 K at $p = 0$ to 19.2 K at $p = 15.6$ kbar. The observed initial reduction is in accord with the predicted value of -29 mK/kbar.

In conclusion, UNi$_4$B exhibits a large electronic specific heat coefficient, which still increases at 0.4 K. Its field dependence suggests Kondo-like screening of the disordered U-moments. The Néel transition in zero field is of 3D Heisenberg universality, but changes character towards 2D behaviour above 8 T. UNi$_4$B fulfils the requirements for, and can be classified as, a mixed-phase material, where the uranium atoms at the nonordering sites carry a residual magnetic moment. A theoretical model using these ideas was presented at this conference by Lacroix et al. [9].

This work was partially supported by CIAR and NSERC of Canada and the Dutch Foundation FOM. Work at Santa Cruz was supported by the DOE under grant No. DE-FG03-87ER45324. Work at Los Alamos was performed under the auspices of the US Department of Energy.

References

[1] S.A.M. Mentink et al., Phys. Rev. Lett. 73 (1994) 1031.
[2] S.A.M. Mentink et al., in Proc. Int. Conf. on Strongly Correlated Electron Systems, Goa, India (1995).
[3] M.D. Nuñez Regueiro and C. Lacroix, Phys. Rev. B 50 (1994) 16063.
[4] R. Ballou et al., Phys. Rev. Lett. 66 (1991) 1910.
[5] S.A.M. Mentink et al., Phys. Rev. B 51 (1995) 11567.
[6] A full account of this work will be published elsewhere.
[7] J.C. Guillou and J. Zinn-Justin, Phys. Rev. Lett. 39 (1997) 95.
[8] V. Privman et al., in: Phase Transitions and Critical Phenomena, Vol. 14., eds. C. Domb and J.L. Lebowitz (Academic Press, New York, 1989).
[9] C. Lacroix et al., these Proceedings, Physica B 230–232 (1997).