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Specific heat of UNiGe in high magnetic fields

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Specific-heat measurements were performed on a single crystal of UNiGe in magnetic fields up to 17.5 T. This compound crystallizes in the orthorhombic TiNiSi structure and undergoes two magnetic phase transitions at 41.5 and 50 K. Besides these transitions, the specific heat also shows a Schottky contribution due to the crystal-field splitting. The effects of an applied magnetic field are compared with results from previous neutron diffraction, magnetoresistance, and magnetization measurements. © 1997 American Institute of Physics. [S0021-8979(97)17508-9]

I. INTRODUCTION

Uranium intermetallic compounds are of considerable interest for a general understanding of the physics of f magnetism. A large set of experimental data is available now on the basic magnetic properties (ordering temperatures, magnetic structures, spontaneous moments, etc.) of the equiatomic UTX compounds (T =transition metal, X = p -electron element). UNiGe belongs to a large group of isostructural compounds crystallizing in the orthorhombic TiNiSi-type structure (space group $Pnma$).^{1,2} Initially, it has been thought that UNiGe has only one magnetic phase transition from the paramagnetic to the antiferromagnetic state at 41.5 K.³⁻⁵ Later, a second magnetic phase transition at 50 K had been identified in specific-heat measurements.⁶ Recently, neutron-diffraction studies have shown that the low-temperature phase of UNiGe is commensurate with $\mathbf{q}=(0,1/2,1/2)$ and that an incommensurate phase with $\mathbf{q}=(0,\delta,\delta)$ exists between 41.5 and 50 K.⁷ Furthermore, a third magnetic phase has been found which is only stable in applied fields, the $1/3$ phase which has a propagation vector $\mathbf{q}=(0,1/3,1/3)$.⁷ Simplified phase diagrams for $B\parallel b$ and $B\parallel c$ are shown in Fig. 1. At 4.2 K, three metamagnetic transitions are induced in fields applied along the b axis (at 5, 17, and 25 T), and two metamagnetic transitions along the c axis (at 3 and 10 T).⁸ Furthermore, at somewhat higher temperatures, the 17 T b -axis transition splits into two transitions.⁹ Purwanto *et al.*¹⁰ have concluded, however, that only one of the 17 T transitions and the 25 T transition are intrinsic to the b axis and that the other two transitions are due to a small misaligned crystallite. In the present article, we report on the specific heat in magnetic fields up to 17.5 T.

II. EXPERIMENTAL METHOD

The specific-heat measurements were carried out on a single crystal grown by means of the tri-arc Czochralski technique. Two pieces of UNiGe, with a total mass of about 1.22 gram, were cut by spark erosion along the three principal axes. The specific heat was measured by means of a semi-adiabatic technique in the temperature range between 400 mK and 65 K in magnetic fields up to 17.5 T¹¹ along the three principal axes.

III. RESULTS AND DISCUSSION

In zero field, the c/T curve shows anomalies at 41.5 and 50 K (Fig. 2), the temperatures where magnetic transitions occur. The specific heat was analyzed according to the expression

$$c = \gamma T + D(T, \Theta_D) + R(\Delta/T)^2 (d_0/d_1) \frac{\exp(\Delta/T)}{[1 + (d_0/d_1)\exp(\Delta/T)]^2}. \quad (1)$$

Here, γT is the electronic contribution to the specific heat and D is the Debye function (with the Debye temperature Θ_D) describing the lattice contribution to the specific heat. The last term represents a Schottky contribution to the specific heat due to the crystal-field splitting of the energy levels. Δ is the energy difference between the ground state and the first excited state and d_1/d_0 is the ratio of the degeneracies of these two states.

FIG. 1. B - T phase diagrams for (a) $B\parallel c$ axis and (b) $B\parallel b$ axis (after Purwanto *et al.*, Ref. 10). Note that there are three magnetic phases with propagation vectors $\mathbf{q}=(0,1/2,1/2)$, $(0,\delta,\delta)$, and $(0,1/3,1/3)$.

FIG. 2. Specific heat of UNiGe in zero field. The dotted line corresponds to the fitted electronic contribution. The dashed curve corresponds to fitted phonon contribution. The solid curve corresponds to the total fit, the sum of the electronic, the phonon, and the crystal field contribution.

Outside the temperature region from 35 to 52 K, the results can be best fitted using $\gamma=27.0$ mJ/mol K², $\Theta_D=260$ K, $\Delta=109$ K, and $d_1/d_0=2.0$ (see Table I). The relative importance of the three contributions can be seen in Fig. 2.

After subtraction of the lattice contribution, the magnetic entropy at 65 K is estimated to be somewhat smaller than R . Using the expression

$$S_{\text{mag}} = R \ln(2J+1), \quad (2)$$

we obtain a J value of 0.81 which is much smaller than the values for the free U^{3+} ($J=9/2$) and U^{4+} ($J=4$) configurations.

In Fig. 3, we have plotted the field dependence of the specific heat for fields applied along the c axis. These results are consistent with the B - T phase boundaries reported from previous neutron diffraction, magnetization, and magnetoresistance studies.¹⁰ The anomaly at 41.5 K marks the transition between the commensurate and the incommensurate phase, while the transition at 50 K is between the incommensurate and the paramagnetic phase. The antiferromagnetic peak at 41.5 K vanishes around 2 T, while the peak at 50 K still persists above 10 T. It should be noted that our specific-heat results do not show an anomaly corresponding with the transition from the $1/3$ phase to the commensurate phase, rather irreversible heat effects in the intermediate field range

TABLE I. Characteristic parameters for the measured specific heat of a UNiGe single crystal in different magnetic fields (B) applied along the different principal axes.

B (T)	axis	γ (mJ/mol K ²)	Δ (K)	d_1/d_0	S_{mag} (J/mol K)
0	--	27.0 ± 0.50	109 (2)	2.0	8.01
4	b	26.5 ± 1.00	107 (2)	2.0	7.81
8	c	34.0 ± 1.00	111 (1)	2.0	7.32
12	a	26.8 ± 0.50	112 (1)	2.0	7.43
16	b	38.2 ± 0.50	109 (2)	2.0	7.68

FIG. 3. Temperature dependence of the specific heat of UNiGe measured in various magnetic fields applied parallel to the c axis.

were observed. Along the c axis, the linear electronic term in the specific heat (γ) showed an increase with field between 4 and 12 T and tended to saturate above 12 T.

In Fig. 4, we show the results for fields applied along the b axis. The peak at 41.5 K disappears between 2 and 4 T, while the 50 K peak is still present at 16 T. Again, these findings are consistent with the previously reported B - T phase diagram. The linear electronic term (γ) showed an increase with field between 4 and 8 T, saturated between 8 and 12 T, and again increased above 12 T.

FIG. 4. Specific heat of UNiGe around the antiferromagnetic phase transitions in magnetic fields of 0, 2, 4, 8, 12, and 16 T ($B||b$). The first transition at 41.5 K disappears between 2 and 4 T.

FIG. 5. Temperature dependence of the specific heat of UNiGe in various magnetic fields between 0 and 16 T ($B \parallel a$) in the representation c/T vs T .

For $B \parallel a$, no field dependence of the specific heat is expected as the a axis is known to be the hard magnetization axis.⁸ However, the peak at 41.5 K is found to have a weak temperature dependence up to 12 T, above which the peak smears out. The anomaly at 50 K on the other hand, becomes more pronounced with increasing field (see Fig. 5). At present, it is not clear whether this behavior is due to some slight disorientation of the crystal, or whether it is a signature of the x component of the magnetic moment which has recently been determined in a polarized-neutron-diffraction experiment.¹²

Comparing UNiGe with UNiAl (UNiAl crystallizes in the hexagonal ZrNiAl structure with very similar U–U interatomic distance but uniaxial anisotropy¹³) when applying magnetic field, one finds that the antiferromagnetic transition of UNiAl shifts to a lower temperature while that of UNiGe disappears. This main difference suggests that the U magnetic moments are more localized in UNiGe than in UNiAl.

IV. SUMMARY

We have performed specific-heat measurements on a UNiGe single crystal in magnetic fields up to 17.5 T. The

antiferromagnetic transition at 41.5 K is reflected in the specific heat and disappears between 1 and 2 T when the field is applied along the c axis. It disappears between 2 and 4 T when the field is applied along the b axis. At 50 K, a small anomaly is found which is expected on the basis of the phase diagram. The low-temperature specific-heat coefficient γ determined at 1 K amounts to 27.0 mJ/mol K² in zero field and is affected by the application of an external magnetic field up to 16 T along the b or the c axes. The crystal-field splitting between the ground state and the first excited state hardly shows an increase in magnetic fields. All the results obtained on the UNiGe single crystal suggest that the U magnetic moments are more localized than in UNiAl.

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