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Incommensurate antiferromagnetic phase in UNiGe

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By specific-heat, magnetization, electrical resistivity, and neutron-diffraction measurements on a single crystal we have confirmed that UNiGe orders antiferromagnetically below 50.5 K into an incommensurate phase with $\mathbf{q}=(0,1/2,-1/2)\pm(0,\delta,\delta)$, $\delta\sim 0.15$. δ decreases continuously with decreasing temperature to ~ 0.123 at 41.5 K, where the incommensurate phase vanishes in a first-order phase transition and a commensurate antiferromagnetic structure with $\mathbf{q}=(0,1/2,1/2)$ sets in and remains stable down to the lowest temperatures. If a magnetic field sufficient to induce a metamagnetic transition (1–5 T) is applied along the c axis, both antiferromagnetic phases are transformed to an uncompensated AF phase with $\mathbf{q}=(0,1/3,1/3)$ yielding a nonzero magnetization $M\approx 1/3\times M_S$. The latter structure is destroyed and a complete alignment of U moments is achieved in fields above 10 T. The strikingly different B - T diagrams observed for a magnetic field applied along different crystallographic directions reflect strongly anisotropic exchange interactions.

I. INTRODUCTION

UNiGe belongs to the isostructural group of the UTX compounds (T =transition metal, $X=p$ metal), which crystallize in the orthorhombic TiNiSi-type structure. The nearest-neighbor uranium atoms in this structure form zigzag chains along the a axis. The coordination of U atoms is intimately connected with the anisotropy of bonding of $5f$ orbitals, which has serious consequences for the symmetry of the $5f$ -electron magnetism.¹ Specifically, in UNiGe and other isostructural UTX compounds, the easy-plane magnetocrystalline anisotropy with the hard-magnetization direction along the a axis is observed as a rule.^{1,2} This is manifest in the low-temperature magnetization, which is small and linearly dependent on the magnetic field up to 35 T applied along the a axis. For the other two field directions (along b and c) almost saturated magnetization M_S due to aligned U moments of $1.45\ \mu_B$ is attained above metamagnetic transitions.² Note that the magnetization curves at 4.2 K display two metamagnetic transitions at 17 and 25 T in the field applied along b (and at 3 and 10 T in $B\parallel c$). In both field geometries, the magnetization observed above the first transition amounts to approximately $1/3\times M_S^2$.

For some time, UNiGe was believed to order magnetically around 42 K,^{2–7} although some indications of another transition around 50 K could be seen in the specific-heat data of Kawamata *et al.*⁷ Moreover, controversial conclusions about the magnetic structure at low temperatures could be found in the literature.^{6,8} This unsatisfactory situation motivated us to perform an extensive study of a well-defined single crystal of UNiGe, which was governed by a Czochralski technique in a tri-arc furnace at the University of Amsterdam. Besides measurements of bulk properties (magnetization, electrical resistivity, and specific heat) over wide temperature and external magnetic-field intervals, we have

performed extensive neutron-diffraction experiments. Results and experimental details of bulk measurements were published elsewhere,^{2,9} along with preliminary neutron data indicating the existence of the incommensurate antiferromagnetic phase (IAFP) below 50 K¹¹. In this paper we concentrate on both the temperature and magnetic-field stability of the IAFP in the complex magnetic phase diagram of UNiGe.

II. RESULTS AND DISCUSSION

The specific heat of UNiGe exhibits a sharp peak at 41.5 K and a weaker maximum around 50 K (see Fig. 1). The first-order magnetic phase transition at 41.5 K is also clearly reflected in the magnetization and resistivity.^{2,9} Closer inspection of magnetization and electrical resistivity results, however, also reveals around 50 K slight (but well noticeable

FIG. 1. Temperature dependence of the specific heat of UNiGe.

FIG. 2. Magnetic phase diagram of UNiGe in B parallel to the c axis, as determined from magnetization and neutron-diffraction measurements. (I) incommensurate antiferromagnetic phase, $\mathbf{q}=(0,1/2,-1/2)\pm(0,\delta,\delta)$, (II) commensurate antiferromagnetic phase, $\mathbf{q}=(0,1/2,1/2)$, (III) uncompensated antiferromagnetic phase, $\mathbf{q}=\pm(0,1/3,1/3)$, (IV) phase with "ferromagnetically" aligned U moments, and (V) paramagnetic phase.

in $\partial M/\partial T$ and $\partial \rho/\partial T$) anomalies, which corroborate the conclusion about the magnetic origin of this phase transition.

In order to obtain better knowledge of magnetic phases and transitions in UNiGe we performed neutron-diffraction experiments on the same single crystal at BENSC (on E2 and E4) and LANSCE (on SCD). The obtained magnetic phase diagram shown in Fig. 2 contains essential information from studies in magnetic fields applied along the c axis.

A. Zero magnetic field, $T \leq 41.5$ K

All observed magnetic reflections can be indexed as $h,k/2,1/2$ (with $k,1$ odd), suggesting the AF structure with $\mathbf{q}=(0,1/2,1/2)$ in agreement with Ref. 6. The U moments are locked in the b - c plane. The temperature dependence of the intensities of the magnetic reflections indicates that the U moment decreases slowly with increasing temperature.¹¹ At 40 K, the ordered U moments retain about 90% of the low-temperature value. The magnetic intensities then decrease abruptly at the 41.5 K first-order phase transition, where the low-temperature phase vanishes.

B. Zero magnetic field, $T \geq 41.5$ K

A crucial point of our research has been to indicate an IAFP, which propagates within the b - c plane. For this purpose experiments on the flat cone diffractometer E2 in Berlin and the single-crystal diffractometer SCD with an area detector at Los Alamos were indispensable. Both types of experiments provided compatible results confirming the existence of an IAFP with $\mathbf{q}=(0,1/2,-1/2)\pm(0,\delta,\delta)$. For illustration, we display in Fig. 3 typical patterns recorded on SCD in Los Alamos at 20 and 46 K, in which the difference between the respective magnetic states is manifest. Whereas at 20 K the $0, 3/2, -1/2$ is characteristic for the commensurate AF phase stable below 41.5 K, this reflection is absent at 46 K and instead two satellites shifted by $\pm(0,0.141,0.141)$ indicate the presence of the IAFP. After identifying this phase, its stability and temperature evolution of δ were stud-

FIG. 3. Magnetic reflections in the vicinity of the $0, 3/2, -1.2$ at (a) 20 K and (b) 46 K.

ied on E4 in Berlin. The results are displayed in Fig. 4. The IAFP emerges just above 41.5 K. The characteristic reflections $0, k/2 \pm \delta, -1/2 \pm \delta$ reach a maximum intensity already around 43 K and then diminish continuously with increasing temperature. The reflections are at the limit of detectability at 50 K, but some residual intensity can be seen in the background up to approximately 53 K. The parameter δ varies from ~ 0.123 at 41.5 to 0.15 at 50 K. The transition at 41.5 K is apparently of the first-order type in contrast to the second-order transition around 50.5 K.

C. $B \leq 6$ T, $B \parallel c$, $T \leq 41.5$ K

The first metalmagnetic transition exhibits a large hysteresis.¹¹ The critical fields and the hysteresis (marked by the hatched region in Fig. 2) decrease with increasing temperature. When sweeping the field upwards, the $0,k/2,1/2$ reflections disappear rapidly around the transition. On the

FIG. 4. Temperature dependence the integral intensity and the parameter δ of the $(0,1/2,-3/2)\pm(0,\delta,\delta)$ reflection.

FIG. 5. Field scans of (a) magnetization and (b) the 0 2/3 4/3 reflection at 45 K.

other hand, $h, k/3, 1/3$ -type reflections and magnetic contribution to nuclear reflections emerge (following the magnetization dependence closely).¹¹ The metalmagnetic state connected with the uncompensated AF structure therefore has $\mathbf{q}=(0,1/3,1/3)$. This leads to the collinear arrangement of the U magnetic moments oriented along the c axis with the $+-$ stacking simultaneously along the b and c axis, which gives rise to the magnetization $M=1/3 \times M_s$ in agreement with the above-mentioned result from magnetization measurements.

D. $B \leq 6$ T, $B \parallel c$, $T \geq 41.5$ K

The IAFP is stable in magnetic fields up to about 1 T, where it starts to transform gradually to phase III. This field correlates well with that of the metalmagnetic transition shown in Fig. 5(a). The $0, k/3, 1/3$ reflections representing the phase III persist up to ~ 51 K, where a first-order transition (in contrast to the second-order transition in zero field) to the high-temperature paramagnetic phase takes place as shown in Fig. 5(b).

When the magnetic field is applied along the b axis a two-step metalmagnetic process appears. However, the critical fields of the metalmagnetic transitions are considerably higher, indicating pronounced anisotropy of the exchange interactions.

To analyze magnetic phases in U intermetallics, models considering the relation of U coordination and the type of

anisotropy can be employed. The experimental findings in UNiGe corroborate the empirical rules¹ relating the symmetry of the bonding of the $5f$ orbitals in a particular structure to the type of magnetocrystalline anisotropy. The strong bonding axis (a axis in UNiGe and structure-related UTX compounds) determines the hard-magnetization direction whereas the magnetic moments are locked perpendicular to the hard direction (in the b - c plane in UNiGe). The exchange interaction along the strong bonding axis (plane) is usually strong and ferromagnetic, whereas the considerably weaker interaction(s) in the perpendicular direction(s) mediate the coupling between the ferromagnetic chains (planes). These interactions are frequently frustrated and a sequence of incommensurate and commensurate phases can be observed¹² when temperature is decreased.

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- ¹ V. Sechovský, L. Havela, H. Nakotte, F.R. de Boer, and E. Brück, *J. Alloys Comp.* (in press).
- ² L. Havela, V. Sechovský, F. R. de Boer, E. Brück, and H. Nakotte, *Physica B* **177**, 159 (1992).
- ³ R. Troc and V. H. Tran, *J. Magn. Magn. Mater.* **73**, 38 (1988).
- ⁴ K. H. J. Buschow, E. Brück, R. G. van Wierst, F. R. de Boer, L. Havela, V. Sechovský, P. Nozar, E. Sugiura, M. Ono, M. Date, and A. Yamagishi, *J. Appl. Phys.* **67**, 5215 (1990).
- ⁵ S. Kawamata, K. Ishimoto, H. Iwasaki, N. Kobayashi, Y. Yamaguchi, T. Komatsubara, G. Kido, T. Mitsugashira, and Y. Muto, *J. Magn. Magn. Mater.* **90-91**, 513 (1990).
- ⁶ A. Murasik, P. Fischer, R. Troc, and V. H. Tran, *J. Phys. Condens. Matter* **3**, 1841 (1991).
- ⁷ S. Kawamata, H. Iwasaki, and N. Kobayashi, *J. Magn. Magn. Mater.* **104-107**, 55 (1990).
- ⁸ S. Kawamata, K. Ishimoto, Y. Yamaguchi, and T. Komatsubara, *J. Magn. Magn. Mater.* **104-107**, 51 (1990).
- ⁹ K. Prokeš, H. Nakotte, E. Brück, F. R. de Boer, L. Havela, V. Sechovský, P. Svoboda, and H. Maletta, *IEEE Trans. Magn.* (in press).
- ¹⁰ F. R. de Boer, K. Prokeš, H. Nakotte, E. Brück, P. Svoboda, V. Sechovský, L. Havela, and H. Maletta, *Physica B* (in press).
- ¹¹ V. Sechovský, L. Havela, A. Purwanto, A. C. Larson, R. A. Robinson, K. Prokeš, E. Brück, F. R. de Boer, P. Svoboda, H. Maletta, and M. Winkelmann, *J. Alloys Comp.* (in press).
- ¹² D. Gignoux and D. Schmitt, *Phys. Rev. B* **48**, 12682 (1994).