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Magnetic phases in UNiGe

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Abstract

Antiferromagnetic ordering in UNiGe below 50 K has been confirmed by specific-heat, magnetization, electrical resistivity and neutron-diffraction studies. Between 41.5 and 50 K, an incommensurate AF phase with $q=(0,1/2,-1/2)\pm\delta(0,1,1)$, $\delta=0.141$ (at 46 K) appears, whereas a commensurate AF structure with $q=(0,1/2,1/2)$ sets on below 41.5 K. In magnetic fields (parallel to the *c*-axis) sufficient for a metamagnetic transition both the structures are transformed to a phase with $q=(0,1/3,1/3)$ yielding a non-zero magnetization $M\approx\frac{1}{3}M_s$.

UNiGe is one of 14 UT(Si,Ge) compounds which have been reported to adopt the orthorhombic $CeCu_2$-type structure (space group *Imma*) [1] or its ordered ternary version of the TiNiSi type (*Pnma*) [2]. Note that the lattice constants *a*, *b*, *c* in the TiNiSi notation correspond to *b*, *a*, *c* in the $CeCu_2$-type lattice.

In a number of papers on UNiGe polycrystals a magnetic phase transition to antiferromagnetic ordering around 40–44 K has been reported [1, 3–5]. On the basis of powder neutron diffraction results, Murasik *et al.* [6] claimed that UNiGe crystallizes in the $CeCu_2$-type structure with Ni and Ge atoms being randomly distributed on copper sites. An antiferromagnetic structure with a propagation vector $q=(1/2,0,1/2)$ and uranium magnetic moments ($\mu=(1.37\pm0.07)\mu_B$ at 13 K) oriented along the *a* axis was proposed below $T=43.3$ K. The possibility of a first-order magnetic phase transition was considered. In contradiction, Kawamata *et al.* [7] claim that they observed on a UNiGe single crystal at 10 K only magnetic reflections which coincide with nuclear reflections, which they naturally interpreted in terms of the identical size of the magnetic and chemical unit cell. This not very satisfactory situation about UNiGe has motivated our further studies on single crystals reported previously [8]. The original measurements of the magnetization curves at 4.2 K (up to 38 T) and temperature dependence of magnetic susceptibility were completed by an extended study of the magnetization, specific heat and electrical resistivity as a function of temperature and magnetic field. Moreover, we performed neutron diffraction experiments on a powder sample and on the single crystal.

The temperature dependence of the specific heat shown in Fig. 1 exhibits a sharp peak at 41.5 K and a broad anomaly around 50 K in a good agreement with data by Kawamata *et al.* [7]. The peak at 41.5 K corresponds to the first-order magnetic phase transition mentioned above. Anomalies at this temperature were observed also in the temperature dependence of magnetic susceptibility and electrical resistivity. The powder neutron diffraction pattern at 3 K consists of pure nuclear reflections consistent with the TiNiSi-type structure and magnetic reflections pointing to an antiferromagnetic structure of U moments $\mu\approx1.4\mu_B$ (within the *b*–*c* plane) with a propagation vector $q=(0,1/2,1/2)$. This pattern remains generally unchanged up to 41.5 K. Temperature scans of several magnetic reflections made on the single crystal reveal only a weak decrease in uranium magnetic moment with increasing temperature (Fig. 2). At 40 K, the moment amounts still to about 90% of its low temperature value. The magnetic state between 41.5 and 50 K is not yet fully understood. The slight anomalies around 50 K observed also in the temperature dependence of both the susceptibility and

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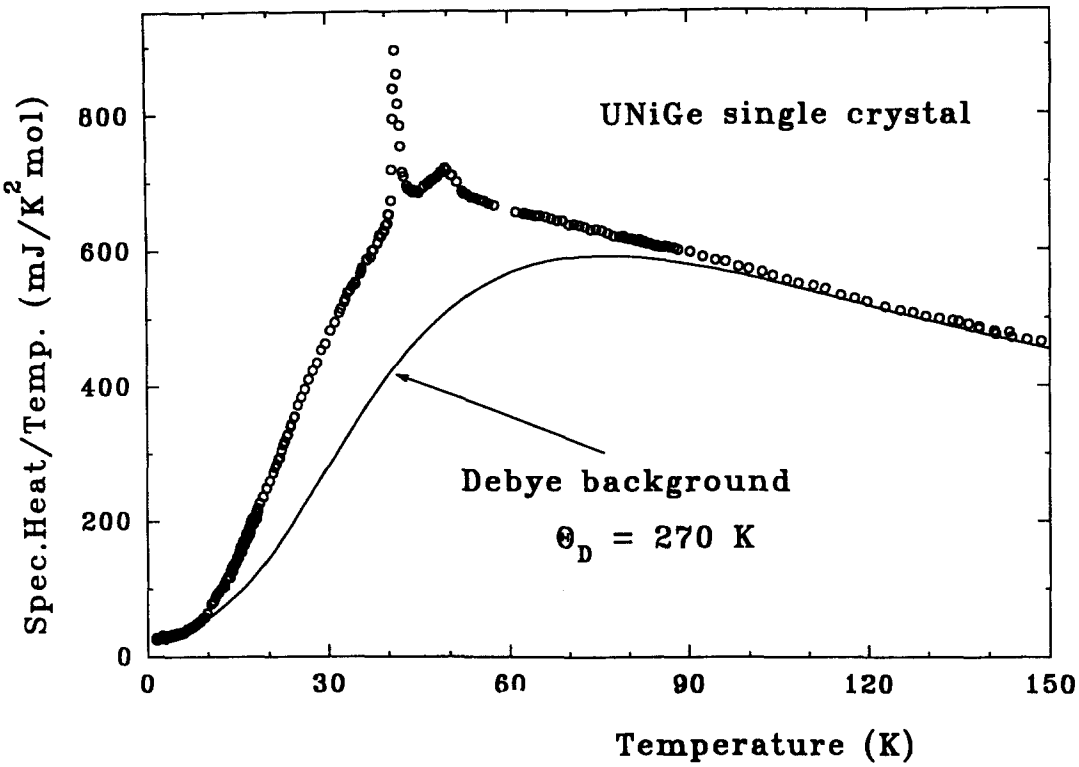

Fig. 1. *C/T vs. T* plots displaying the specific heat behaviour of UNiGe.

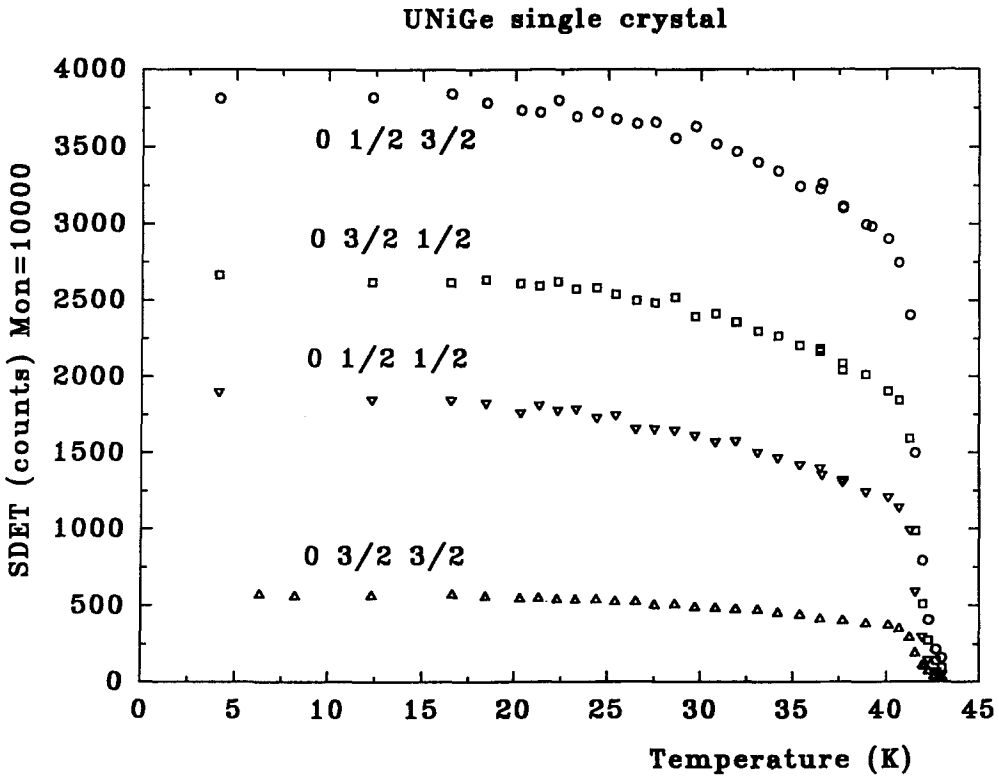

Fig. 2. Temperature dependence of several 0,*k*/2,*l*/2 reflections of UNiGe.

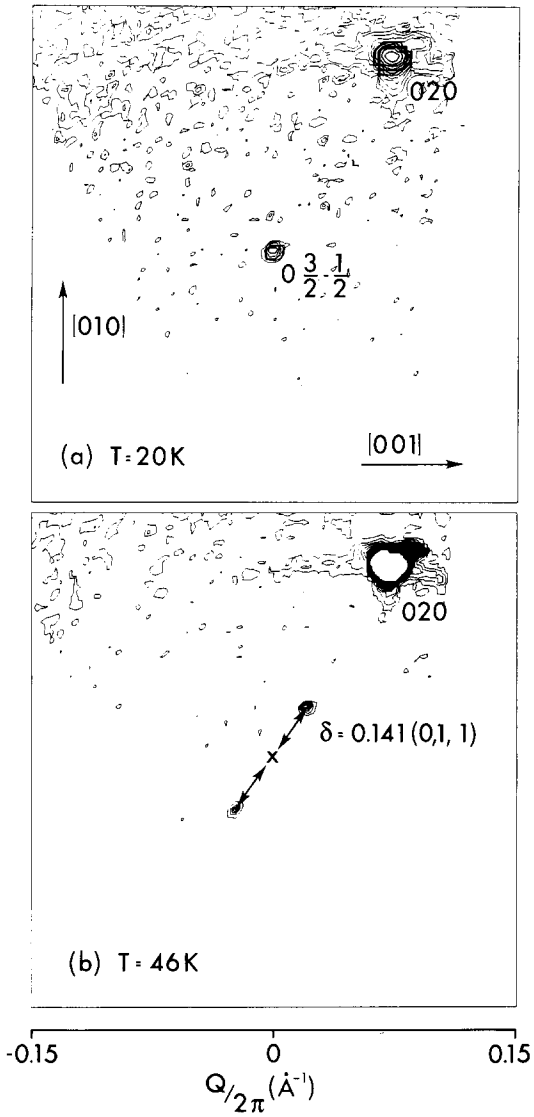

Fig. 3. Magnetic reflections in the vicinity of 0,3/2,1/2 in UNiGe: (a) pattern from the commensurate magnetic phase at 20 K; (b) incommensurate satellites at 46 K.

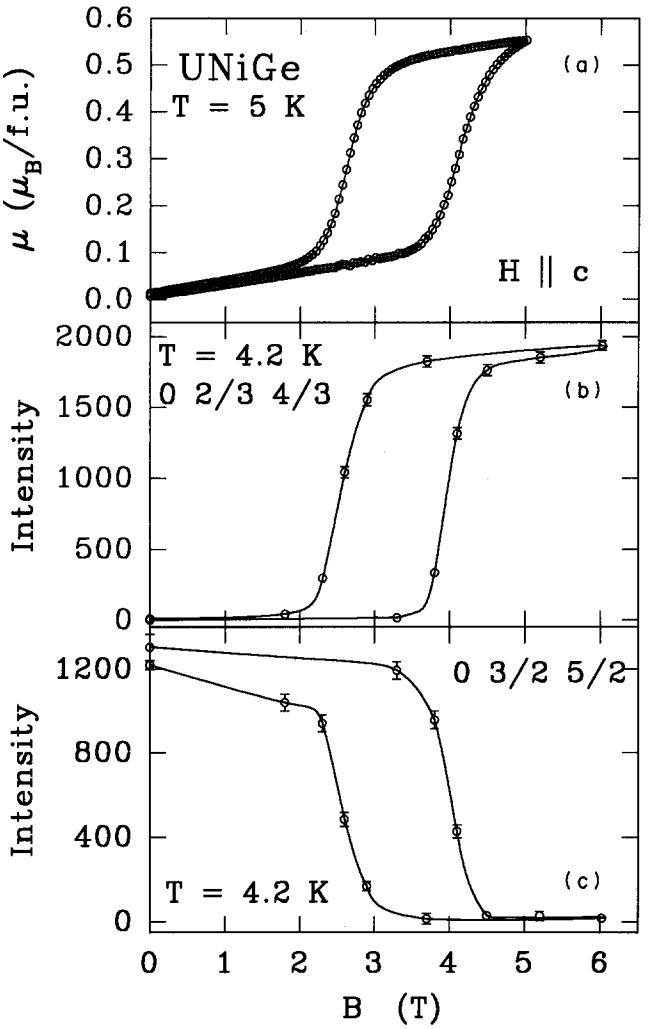

Fig. 4. Field dependence of (a) the magnetization, and (b) the 0,2/3,4/3 and (c) 0,3/2,5/2 reflections of UNiGe in the magnetic field applied along the c axis.

the resistivity are indicative of a magnetic phase transition from the paramagnetic state to a sort of magnetic ordering. Preliminary neutron diffraction studies by SCD at LANSCE and E3 at BENSC revealed between 41.5 and 50 K an incommensurate magnetic structure with a temperature-dependent propagation vector in the b–c plane. The propagation vector $\boldsymbol{q}=(0,1/2,-1/2)\pm\boldsymbol{\delta}$, where $\boldsymbol{\delta}=0.141(0,1,1)$ at 46 K, as shown in Fig. 3. Further neutron diffraction studies are under way.

The magnetization curves at 4.2 K display metamagnetic transitions in fields $\boldsymbol{B}$ parallel to $\boldsymbol{b}$ (at 17 and 25 T) and $\boldsymbol{B}$ parallel to $\boldsymbol{c}$ (at 3 and 10 T) reflecting changes in antiferromagnetic structure towards the parallel alignment of moments in high fields ($M=1.45\mu_B$ per formula unit). A weak linear magnetic response to field $\boldsymbol{B}$ parallel to $\boldsymbol{a}$ ($M=0.23\mu_B$ per formula unit) manifests a huge magnetic anisotropy. In order to study the microscopic origin of different magnetic states between the transitions we have started neutron diffraction experiments on the UNiGe crystal in magnetic fields. So far we have measured the behaviour in fields up to 6 T along the c axis. The phase above 3 T (amounting to $M=\frac{1}{3}M_s$) is consistent with the propagation vector $\boldsymbol{q}=(0,1/3,1/3)$, which yields $+\,+\,-$ stacking of uranium magnetic moments along both the b and the c directions. The pronounced hysteresis of the metamagnetic transition seen on the magnetization curve was reproduced when measuring $h,k/2,l/2$ and $h,k/3,l/3$ reflections with increasing and decreasing magnetic field as shown in Fig. 4. The first transition in fields along the b axis cannot be reached by static magnetic fields available at neutron beams. Considering the dramatic difference in the magnetoresistance response on the first metamagnetic transition in fields along the c and b axes displayed in Fig. 5, we can conclude that $\boldsymbol{q}$ of the

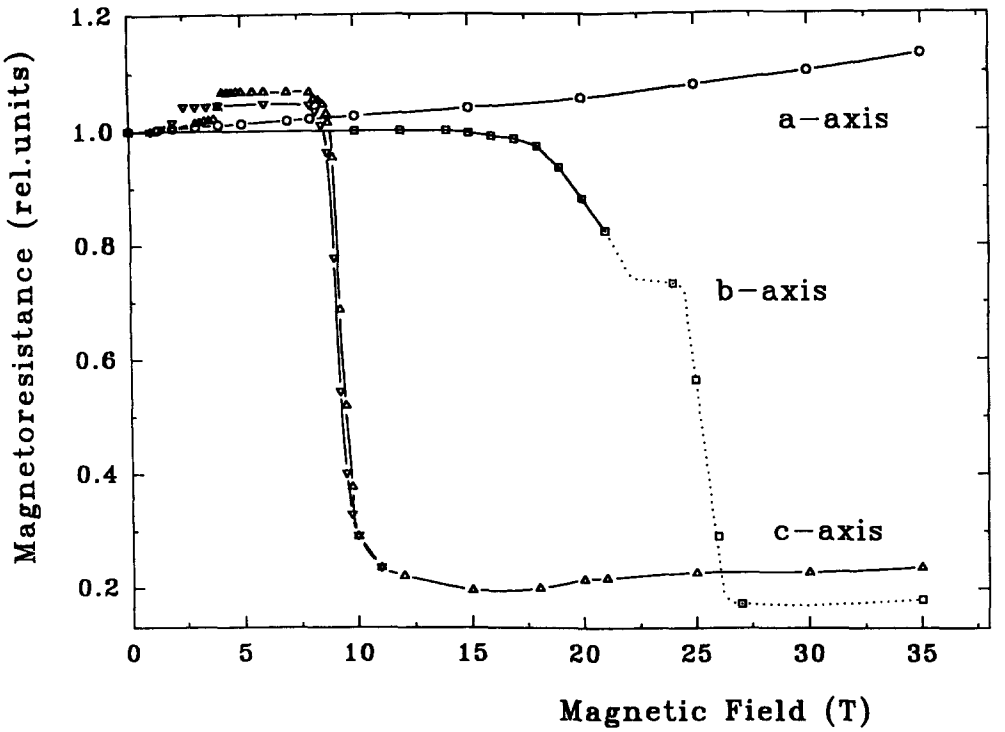

Fig. 5. Longitudinal magnetoresistance in UNiGe in the magnetic field applied along the *a*, *b* and *c* axes.

magnetic structure above 17 T (***B*** parallel to ***c***) cannot be equal to (0,1/3,1/3).

The up to now observed features of magnetic structures in UNiGe confirm the conclusions, which we drew previously from bulk experiments, suggesting that the strong bonding of 5f orbitals along the *a* axis lead to a huge magnetic anisotropy characterized by uranium magnetic moments perpendicular to this direction, which seems to be a general feature in UTX compounds with the TiNiSi-type structure.

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