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Metamagnetic transitions and giant magnetoresistance in UNiGe

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Abstract

We report on the temperature dependence of the metamagnetic transitions, which occur in UNiGe in magnetic fields applied along the b and c axis. Above the ordering temperature $T_N = 50$ K, a strong magnetic and thermal history-dependence is found. Furthermore, the development of a small ferromagnetic component ($0.02\mu_B/\text{f.u.}$) at 4.2 K after releasing the field applied indicates magnetic-history effects also at low temperatures. The presumed irreversibility in the magnetization process at low temperatures is corroborated by irreproducible changes in the magnetoresistance response at 4.2 K. Nevertheless, tentative magnetic phase diagrams have been obtained for both orientations.

1. Introduction

UNiGe has been reported to crystallize in the orthorhombic TiNiSi structure [1], which is an ordered variant of the CeCu₂ structure. It was reported to order antiferromagnetically around 42 K [1–4]. Neutron-diffraction results indicate the 5 f moments to be located in the b – c plane (in the TiNiSi notation) [1, 4]. This finding is supported by the relatively weak magnetic response for magnetic fields applied along the a axis. Three metamagnetic transitions can be induced at 4.2 K in fields applied along the b (at 5, 17 and 25 T) and c axis (3 and 10 T) [5].

Recently, we have indicated a second magnetic phase transition at 50 K [6]. The neutron-diffraction studies in the temperature range 41.5–50 K revealed an incommensurate antiferromagnetic structure [7]. Furthermore,

a large reduction of the electrical resistivity was found for the forced ferromagnetic state [6].

The previous high-field investigations being restricted to 4.2 K, in the present study we present the temperature dependencies of the metamagnetic transitions for fields applied along the b and c axis. We have also reinvestigated the magnetoresistance for the same axes.

2. Experimental

The experimental results were obtained on two UNiGe single crystals, grown from the stoichiometric melt by the modified Czochralski tri-arc technique. One crystal is the same as that used in [6] and its quality has been checked by microprobe and by X-ray and neutron diffraction. The second bigger crystal, which was used for measurements of the magnetization at elevated temperatures, was checked by X-ray diffraction only.

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A slight mosaicity within the basal plane was found for this second crystal.

Magnetization and magnetoresistance measurements were performed in the Amsterdam High Field Facility in fields up to 35 T and in a SQUID magnetometer in fields up to 5 T. At 4.2 K, the measurements were performed in the 40 T magnet, in which semicontinuous fields can be generated by a controlled current output [8]. In these measurements, the sample is immersed in liquid helium in order to avoid heating of the sample during the field sweep. Magnetization measurements at higher temperatures were carried out in the 30 T magnet, which works according to the same principle as the 40 T magnet but, since the coil is operated at liquid-nitrogen temperature instead of liquid-neon temperature, the maximum field is restricted to 30 T. The sample is cooled by exchange gas and the output of a Lake-Shore temperature controller to a small heater at the sample position allows any temperature setting between 4.2 and 300 K. The temperature of the sample is monitored before and after the field pulse by a diode, which is mounted in the sample holder, so that possible heating during the field sweep can be detected. In the present experiments, the heating of the sample was found to be negligible. For the analysis of our magnetization results, it is important to note, that the field sweeps have been performed at subsequently increasing temperatures without heating the sample to temperatures above 60 K between two measurements.

3. Metamagnetic transitions

In Fig. 1, as an example the field dependence of the magnetization for field applied along the *b* and *c* axis at two temperatures is shown. The exact positions of the transitions have been determined by using field sweeps with continuously changing fields and by plotting dM/dB versus *B* (Fig. 2).

At 4.2 K, two metamagnetic transitions occur for the field applied along the *c* axis. The lower transition at about 3 T exhibits a considerable hysteresis of about 1.5 T. At higher temperatures, this hysteresis becomes smaller and the transition shifts continuously to lower fields. Low-field measurements in the SQUID magnetometer reveal this transition to disappear at 41.7 K, which is consistent with the magnetic phase transition reported in Refs. [1–6]. Between this temperature and 50 K, another weak transition evolves around 1 T, which may indicate the phase boundary of the incommensurate phase proposed in Ref. [6]. The field of the second transition (10 T at 4.2 K) is almost temperature independent up to about 60 K, where it drops precipitously. Although this temperature is higher than the 50 K transition obtained in low-field measurements, it may be

Fig. 1. Field dependence of the magnetization at 4.2 (solid lines) and 31.5 K (dotted lines) for fields applied along the *b* and *c* axis.

Fig. 2. Field dependence of the differential magnetization at various temperatures for fields applied along the *b* (right) and *c* axis (left). The arrows indicate the positions of the metamagnetic transitions.

argued that in magnetic fields the phase boundary to the paramagnetic phase is at 60 K. On the other hand, we cannot exclude short-range ordering due to strong anti-ferromagnetic correlations in this region. The situation is

complicated by the magnetic and thermal history in the sample (Fig. 3). Furthermore, a small but significant ferromagnetic component (about $0.02\mu_B/\text{f.u.}$) was found at 4.2 K after application of a magnetic field, which may reflect a slight modification of the magnetic structure at temperatures below 10 K. Further magnetization studies are necessary to clarify this behaviour.

For the magnetic field applied along the b axis, three metamagnetic transitions are found at 4.2 K in high magnetic fields. The 25 T transition is only marginally affected at temperatures up to 60 K where it drops rapidly, similar to the 10 T transition for the field along the c axis. Also for the b -axis orientation, the phase boundary of the incommensurate phase between 41.7 and 50 K can be monitored. However, contrary to the findings for the c axis, for the b axis a third weak transition is found at rather low fields besides the transition at 17 T at 4.2 K. The 17 T transition is slightly broader than the one in Refs. [5,6], reflecting that the present crystal is less perfect. It can clearly be monitored that this transition surprisingly splits into two above 15 K. Above 41.7 K, both branches cannot be detected any more. The third transition, which is hardly visible in Figs. 1 and 2, was extensively studied in the SQUID magnetometer. A negligible hysteresis and a continuous decrease with temperature were found. Also this transition disappears at 41.7 K. Below 10 K, also for the b -axis a small ferromagnetic component was found after application of a magnetic field. At 4.2 K, the occurrence of a low-field transition at about 2.2 T in the virgin curve (compared

Fig. 3. Magnetic phase diagram for $B//c$ -axis. The open symbols represent the data obtained in field-up sweeps, while the full symbols represent the results for field-down sweeps. In the hatched areas a complicated magnetic history-dependent behaviour is expected (see text). The lines are guides to the eye.

Fig. 4. Magnetic phase diagram for $B//b$ -axis. The open symbols represent the data obtained in field-up sweeps, while the full symbols represent the results for field-down sweeps. In the hatched areas a complicated magnetic history-dependent behaviour is expected (see text). The lines are guides to the eye.

to about 5 T after a field has been applied) may thereby indicate an irreversible magnetization process at low temperatures.

Tentative phase diagrams for the b and the c axis orientation are presented in Figs. 3 and 4, respectively. By comparing the two phase diagrams, it may be argued that the phases 4 (c -axis) and 6 (b -axis) have the same or at least similar magnetic structures. This would suggest, that application of a magnetic field along the c axis leads to a direct transformation of the low-field structure to the structure III, while in the b -axis orientation the same structure is achieved in a more complex way. In the hatched areas in Figs. 3 and 4, a strong magnetic and thermal-history dependence of the sample is expected.

4. Giant magnetoresistance

The field dependence of the electrical resistivity was measured by step-like field pulses, in which the field was kept constant for at least 100 ms in order to avoid eddy-current effects. The DC four point method and a current of about 50 mA were used. The magnetoresistance effect was determined by using a calibration pulse with zero current (100% reduction) or half of the current (50% reduction) after the field pulse. However, one should be aware that this procedure does not account for irreversible field-induced changes of the zero-field resistivity. In Fig. 5, the magnetoresistance results are shown for the b and c axis.

Fig. 5. Field dependence of the electrical resistivity normalized to the 'zero-field' value for $i//B//c$ -axis and $i//B//b$ -axis. The open symbols represent the data obtained in field-up sweeps, while the full symbols represent the results for field-down sweeps. Note that the 'zero-field' value is dependent on the magnetic history and therefore affects the values of the relative reduction. For $i//B//b$ -axis, the result obtained in Ref. [6] is displayed as a dashed line. All other lines are guides to the eye.

Results for field and current applied along the c axis have been published earlier [6]. The two metamagnetic transitions in the magnetization are accompanied by drastic changes of the electrical resistivity. While a step-like increase of about 8% of the resistivity is found at 3 T, there is a drastic drop of the resistivity to about 20% of its zero-field value for the fields above 10 T, where a forced ferromagnetic alignment is achieved. The hysteresis of the magnetoresistance in the 3 T transition can be related to the hysteresis in the magnetization. It should be noted, that in the intermediate phase the resistivity values for decreasing field are slightly lower than for increasing field. As a slight change in the electrical resistivity has been monitored after application of fields higher than 10 T, we may argue that this small difference is related to possible irreversible changes in the magnetic structure discussed above.

In addition to the c axis orientation, we have performed a complete measurement for current and magnetic field parallel to the b axis, of which only preliminary

data were given in Ref. [6]. The second magnetoresistance set presented here confirms the occurrence of three magnetic transitions at 4.2 K. Pronounced changes in the electrical resistivity are seen at about 5, 17 and 25 T. However, it was not possible to determine the value of the magnetoresistance effect unambiguously, because also for the b -axis orientation an irreversible change of the zero-field resistance after application of fields higher than 25 T was found. Furthermore, the increase of the electrical resistivity at about 5 T was not observed in the previous study [6]. In order to get some more insight in this disagreement, we have checked the magnetoresistance effect on the second crystal, which is of a slightly poorer quality. These measurements show the increase of the electrical resistivity at about 5 T sometimes to be present and sometimes absent, which naturally affects also the values of the magnetoresistance effect for higher applied field. Again, this behaviour is very likely to be connected with the magnetic history of the sample. We estimate the reduction of the electrical resistivity in the forced ferromagnetic state (about 25 T) to amount to at least 58% with respect to the 'zero-field' value. This value is much smaller than the 80% reported previously [6] on the same crystal, where possible changes of the 'zero-field' resistance were not considered. In order to clarify this unsatisfactory situation more elaborate magnetoresistance studies in static fields will be performed in the near future.

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