Journal of NUCLEAR SCIENCE and TECHNOLOGY, Supplement 3, p. 184–186 (November 2002)

Magnetic field induced irreversibility in Hall resistivity and specific heat of UNiAl

O. SYSHCHENKO[1*], V. Sechovský[1], H. Sato[2], and Y. Kobayashi[2]
[1]*Dep. of Electronic Structures, Charles University, 121 16 Praha 2, The Czech Republic*
[2]*Tokyo Metropolitan University, Hachioji-shi, Tokyo 192-03, Japan*

We report on our systematic study of evolution of magnetic-history dependent Hall resistivity and specific heat of UNiAl with respect to magnetic field and temperature. It is known[1)] that "virgin" curves expressing the field dependencies of the bulk properties of UNiAl measured from the ZFC state clearly differ from these observed on a sample, which has once appeared in the high-field state. The different shapes of $M(B)$, $\rho(B)$ and $\rho_H(B)$ curves are strongly suggestive of field induced changes in the spectrum of spin fluctuations in UNiAl. This is corroborated by the effect of "field annealing" on the low temperature gamma-value derived from our present specific-heat measurements. The magnetic-field induced irreversibility of Hall resistivity is the strongest at the lowest temperature of measurements (1.7 K), decay with increasing temperature and vanish around 7 K. We have also observed that exposing UNiAl is not a necessary condition to observe irreversibility in lower fields. Based on the results of our measurements, a magnetic phase diagram is constructed and discussed in the light of results of recent neutron-diffraction measurements.

KEYWORDS: UNiAl, antiferromagnetism, metamagnetic transition, Hall resistivity, specific heat, irreversibility

I. Introduction

UNiAl is an itinerant 5*f*-electron antiferromagnet ($T_N \approx 19$ K) with spin-fluctuation effects that are reflected in a reduced magnitude of the U moments and a high value of the linear coefficient of the specific heat $\gamma \approx 165$ mJ/molK^2 [1)]. Owing to a huge uniaxial magnetocrystalline anisotropy, a considerable magnetic-field influence on the electronic properties of this material is only observed if a magnetic field is applied along the *c*-axis of the hexagonal ZrNiAl-type structure. At temperatures lower than 7 K, UNiAl undergoes a first-order metamagnetic transition ($B_c \approx 11.4$ T) between the low-field antiferromagnetic state and a high-field ferromagnetic ordering of U moments that is accompanied by a sharp anomaly in the electron transport and lattice properties[1)]. According to previous reports[2,3)], the field dependencies of the magnetization, magnetoresistivity, Hall resistivity, magnetostriction and elastic constants, if measured starting from the zero-field-cooled (ZFC) state, clearly differ from data observed on a sample that has once appeared in the high-field ferromagnetic state. This effect is sample independent; so far it is observable on all available single-crystal samples. Here we present the results of Hall resistivity ρ_H and specific heat-measurements as a function of temperature and magnetic field performed on a UNiAl single crystal with emphasis on the magnetic-history phenomena.

II. Experimental

The single crystal has been grown in a tri-arc furnace by Czochralski method[4)]. The Hall resistivity and specific heat measurements were performed using a Physical Property Measurement System (PPMS - Quantum Design). The ρ_H was measured on a UNiAl basal-plane-cut plate sample by an AC five-point technique. The magnetic field up to 12 T was applied along the *c*-axis. The sample was first zero field-cooled (ZFC) to the temperature of measurement (in the range 2.5 – 10 K); then the field was ramped up to 12 T and down to 0 T (1st ramp up and down) and again up to 12 T and down to 0 T (2nd ramp up and down). The first ramp-up data will be denoted as $\rho_{H,ZFC}(B)$ and the latter as $\rho_{H,FC}(B)$. To investigate the onset field of irreversible phenomena also, various magnetoresistivity measurements with ramping the field up to a certain field value $B \leq B_c$ and back to 0 T were performed.

The C_p vs. T data were collected in zero field after ZFC or FC down to 0.6 K (FC = cooling in a field of 14 T parallel to *c*). The field dependencies of specific heat were measured at 3.2 K in the following sequence: ZFC, measured at selected fields increasing from 0 to 14 T ($C_{p,ZFC}$ vs. B data) followed by decreasing field from 14 to 0 T ($C_{p,FC}$ vs. B data) data. The C_p vs. T measurements were done both for the ZFC and FC samples down to 0.6 K.

III. Results and discussion

In **Fig. 1a**, the $\rho_H(B)$ curves obtained at several temperatures are displayed. At 2.5 K the MT is accompanied by a sharp ρ_H drop at B_c. The $\rho_{H,ZFC}(B)$ and $\rho_{H,FC}(B)$ curves in fields below B_c show a large difference. The latter result in the field-induced irreversibility reported also for other electronic properties of UNiAl[2,3)]; 2nd ramp up and down data are identical with the $\rho_{H,FC}(B)$ curve. With increasing temperature the value of B_c gradually decreases and the ρ_H drop at MT becomes reduced. For $T \geq 7$ K, the anomaly becomes modified and smeared out.

* Corresponding author: Tel. (+420 2) 21 91 13 52,
Fax. (+420 2) 21 91 13 51
E-mail: syshchen@mag.mff.cuni.cz

Simultaneously, the difference between the $\rho_{H,ZFC}(B)$ and $\rho_{H,FC}(B)$ curves, becomes rapidly reduced with increasing temperature and vanishes at $T^* = 7$ K that is in agreement with our magnetoresistivity data[5]. The T^* is considered to be a characteristic temperature for UNiAl defining the irreversibility line and the temperature at which MT loses its first order character[1].

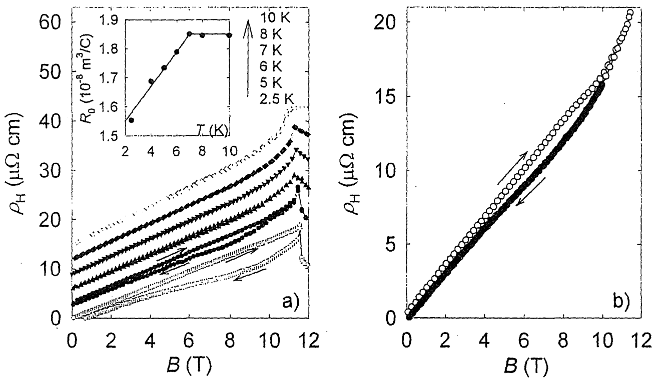

Fig. 1 a) The ρ_H *vs.* $\mu_0 H$ plot for UNiAl at different temperatures. The arrows indicate directions of the field sweeps. (Note that for the sake of clarity the curves at different temperatures are shifted by 5 $\mu\Omega$ cm.) Inset: Temperature dependence of the normal Hall coefficient (R_0) extracted from the linear part of $\rho_{H,ZFC}$(H) curves.
b) Hall resistivity ρ_H for UNiAl at 4 K in fields up to H_c.

At 2.5 K, the $\rho_{H,ZFC}$ varies linearly with field for $B < B_c$, although the magnetization is a non-linear function of magnetic field[3]. This result suggests that the normal Hall effect[6] dominates the $\rho_{H,ZFC}$ behavior at low temperatures

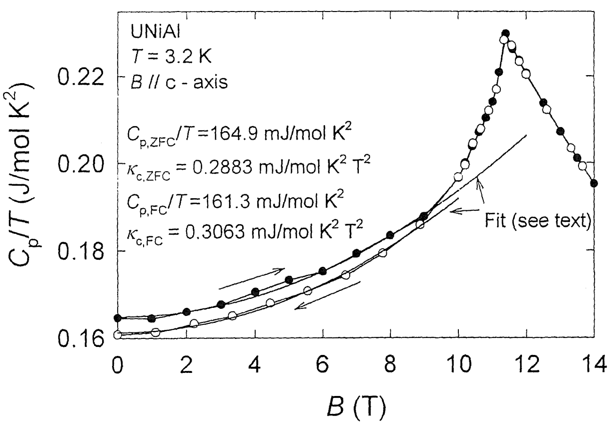

Fig. 2 Specific heat C_p/T as a function of magnetic field B. The lines represent fits (see text).

(the $\rho_{H,FC}$ vs. H dependence, on the other hand, is strongly nonlinear, especially at low temperatures and magnetic fields higher than 4 T). The value of R_0 (the normal Hall effect coefficient[6]) that is defined as the slope of $\rho_{H,ZFC}(B)$ curve is $1.55{*}10^{-8}$ m^3/C at 2.5 K. The R_0 value increases linearly with temperature up to $1.92{*}10^{-8}$ m^3/C at $T^* = 7$ K but then remains temperature independent at least up to 10 K (see inset of **Fig. 1a**).

The specific heat as a function of applied magnetic field B at $T = 3.2$ K in C_p/T vs. B representation is shown in **Fig. 2**. The present data in the ZFC case are in good agreement with earlier results reported by Brück et al.[7]. A smooth progressive increase of $C_{p,ZFC}/T$ is seen up to its maximum $C_{p,ZFC}/T$ value of 235 mJ/mol K^2 found at B_c = 11.3 T. Above the transition field B_c, $C_{p,ZFC}/T$ decreases down to 195.3 mJ/mol K^2 at 14 T. For high-field ($H > 9$ T) the $C_{p,FC}/T$ data are identical. The two curves, however, split below 9 T and the "residual ($H = 0$ T) value" of $(C_{p,ZFC}/T)_0 = 164.9$ mJ/mol K^2 is clearly higher than $(C_{p,FC}/T)_0$, which amounts to 160.8 mJ/mol K^2. It should be noted that a measurement with increasing field starting from this state (FC) produces data identical to the data obtained with decreasing field. The $C_{p,ZFC}/T$ vs. B dependence can be recovered only by heating the sample to temperatures higher than T_N and cooling back in zero field.

The initial increase of both, the $C_{p,ZFC}/T$ vs. B and $C_{p,FC}/T$ vs. B, can be fitted to a quadratic $C_pT = (C_p/T)_0 + \kappa_c B^2$ dependence. The value of $\kappa_{c,FC} = 0.306$ J/mol K^2 T^2 is somewhat higher than $\kappa_{c,ZFC}$ = 0.288 mJ/mol K^2 T^2, which is found to be in good agreement with previous studies[7].

The temperature dependencies of the specific heat for the ZFC and FC samples are shown in **Fig. 3** as C_p/T vs. T^2. One may distinguish three temperature regions - for T > 7.5 K the data follow a straight line that extrapolates to a γ value of 142 mJ/ mol K^2. The FC and ZFC curve split below about 7.5 K and down to 3.5 K. We observe two different straight lines pointing to gamma values $\gamma_{ZFC} \approx$ 160 mJ/ mol K^2 and $\gamma_{FC} \approx$ 155 mJ/ mol K^2, respectively. At the lowest temperatures for both data sets one observes an upturn, which is most pronounced for the field cooled sample. The difference in γ may be tentatively attributed to different spin fluctuations involved in the ZFC and FC case.

Note that the splitting appears at the temperature ($T \approx 7$ K) that coincides with the characteristic temperature T^* that was extracted from our magnetoresistivity[5] and present Hall resistivity data.

As shown in[7] the high γ value in UNiAl may be attributed to on-site and inter-site spin fluctuations. The on-site spin fluctuations are related to hybridization of U *5f*-states with valence states of ligands and *5f*-*5f* hopping processes between U neighbors. The intersite magnetic fluctuations that can be excited in an antiferromagnet like UNiAl by applying a magnetic field are probably a source for increasing C_p/T values with magnetic field up to B_c. After passing the metamagnetic transition, the specific heat values decrease with increasing field as expected for a ferromagnet. The origin of the complex C_p/T behavior below 2 K is unclear at this stage. Magnetic fluctuations with low characteristic energies[7] together with a nuclear contribution to the specific heat may be considered to be responsible for such behavior.

The *B-T* magnetic phase diagram shown in **Fig. 4** displays in one plot the temperature dependence of the critical MT field H_1, identified with the inflection point on a magnetic isotherm at MT and the field dependence of T_N determined

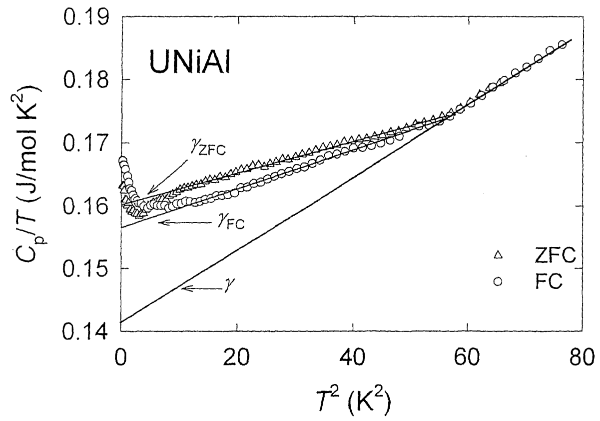

Fig. 3 Low-temperature part of the zero field specific heat of UNiAl for the ZFC sample and the FC (14 T) sample.

from the evolution of the T_N-related specific-heat anomaly with magnetic fields. The two curves $H_l(T)$ and T_N(H) coincide at temperatures up to approximately T^*. In this temperature range ($T \leq T^*$) a sharp peak in the specific heat occurs at T_N, the MT is of the first order and the field-induced irreversibility is observed. At higher temperatures ($T > T^*$) the two curves split and the magnetic phase transition at T_N is of second-order type[1], the MT becomes smeared out and the irreversibility is lost.

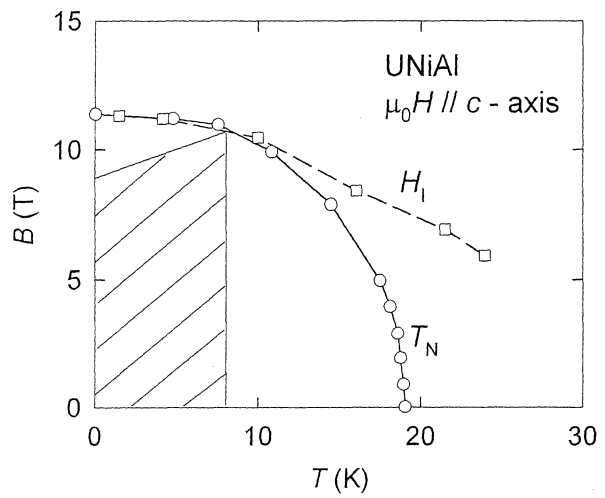

Fig. 4 Tentative *B-T* phase diagram for UNiAl.

As mentioned above, the irreversibility has been reported for UNiAl at low temperatures after being exposed to magnetic fields larger than B_c[2,3] although recent neutron scattering results[8] suggest that irreversibility occurs on microscopic scale below B_c. Our present Hall resistivity data (see **Fig. 1b**) and magnetoresistance results[5] also clearly show that fields somewhat lower than B_c are sufficient to induce the irreversibility. The hatched region in the magnetic phase diagram represents the phase space where the irreversibility is observed.

Neutron-diffraction measurements on UNiAl in[8,9] revealed that the magnetic-history effects in UNiAl have a microscopic origin. The neutron-diffraction data point out an irreversible change of the basal plane propagation vector component of the magnetic structure. The "microscopic" phase diagram determined from the neutron experiment[9] is analogous to the "macroscopic" one determined from our bulk measurements.

The irreversibility in UNiAl is especially pronounced in the fields-induced irreversible change of shapes of $\rho_H(H)$, $M(H)$, $\rho(H)$[2]. This finding is strongly suggestive of field-induced changes in the spectrum of spin fluctuations, which play a substantial role in the physics of UNiAl. This is also corroborated by our low-temperature specific-heat data.

Acknowledgment

This research has been supported by the Ministry of Education of the Czech Republic (project # MSM113200002 the Grant Agency of the Czech Republic and grant # 202/99/0184).

References

1) V. Sechovský and L. Havela, in: Handbook on Magnetic Materials, ed. K.H.J. Buschow, (Amsterdam, North Holland, 1998), Vol. 11., p. 1.
2) V. Sechovský *et al.*, Physica **B 281&282** (2000) 208.
3) V. Sechovský *et al.*, J.Appl.Phys **89 (2)** (2001) 7639.
4) K. Prokeš, Thesis, University of Amsterdam (1997).
5) O. Syshchenko *et al.*, Physica B (2001) – In press.
6) L. Berger and G. Bergmann, in: The Hall Effect and its Applications, ed. C.L. Chien and C.R. Westgate (Plenum, New York, 1980), p. 55.
7) E. Brück, Thesis, University of Amsterdam (1991).
8) K. Prokeš *et al.*, Physica B **259 – 261** (1999) 246.
9) K. Prokeš *et al.*, Proc. 31iémes Journées des Actinides, Saint Malo 2001, O.24.