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Electrical resistivity and Hall-effect study of UNiAl single crystals

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We report on resistivity, susceptibility and Hall-effect measurements in magnetic fields up to 10 T and temperatures from 2 to 300 K on single-crystal platelets oriented parallel or perpendicular to the hexagonal *c* axis. While the resistivity shows little anisotropy the Hall effect and the susceptibility are highly anisotropic with a pronounced maximum for $\boldsymbol{B} \parallel \boldsymbol{c}$ and a nearly temperature independent behavior for $\boldsymbol{B} \perp \boldsymbol{c}$. The Hall data are decomposed into normal and anomalous contributions and the results are discussed in the light of the substantial anisotropy of the U–U separation parallel and perpendicular to the *c* axis.

UNiAl is one of the more than 20 ternary uranium compounds crystallizing in the hexagonal ZrNiAl-type structure. In the general formula UTX, T stands for various 3d, 4d and 5d transition elements and X is either Al, Ga, In, Sn or Sb [1]. The ZrNiAl-type structure derives from the hexagonal Fe_2P-type structure. In UNiAl, Ni occupies the phosphorus sites and U and Al are ordered in such a way in the two inequivalent Fe sublattices, that uranium occupies only every second layer perpendicular to the hexagonal *c* axis. As a result the interlayer U–U separation is nearly 0.6 Å larger than the intralayer separation. UNiAl orders antiferromagnetically below 19 K with a strongly anisotropic susceptibility [2]. For $\boldsymbol{B} \parallel \boldsymbol{c}$ the susceptibility shows a maximum at 26 K while for $\boldsymbol{B} \perp \boldsymbol{c}$ the susceptibility is nearly temperature independent. The reported electronic specific-heat coefficient $\gamma =$ 164 mJ/mol K^2 [2] classifies UNiAl as a medium heavy-electron system.

We have performed resistivity, Hall-effect and magnetization measurements in fields up to 10 T and temperatures down to 2 K on two UNiAl single crystals. The samples were thin platelets with $\boldsymbol{c}$ $\perp$ and $\parallel$ to the platelet surface, respectively. Four tungsten tips in a square arrangement were pressed onto the surface of the platelets. Because of the rectangular shape of these platelets the resistivity measurements for the anisotropic surface could not be reduced quantitatively with the Montgomery formalism [3]. However, by performing the same measurements on a copper foil with the same shape anisotropy but no anisotropy of the resistivity, we can conclude that also the resistivity of UNiAl has very little anisotropy between 300 and 100 K, but increasing anisotropy at lower temperatures. From 300 to 100 K the resistivity in the *ab* plane (ρ_{ab}) decreases by only 10 μΩcm from ≈ 180 to ≈ 170 μΩcm. Below 100 K the decrease is first a little more pronounced, then weakens to make a shallow minimum near 28 K, which is followed by a small peak at 16 K (see fig. 1 for $B = 0$ T). The residual resistivity of the sample was 159 μΩcm, which is ≈ 25 μΩcm higher than found by Brück [4]. For the resistivity along the *c* direction (ρ_c) the minimum near 28 K is a little deeper and the decrease of the resistivity with decreasing temperature below the relative maximum at 17 K is more pronounced and amounts to about 50 μΩcm. Fig. 1 shows the effect on ρ_{ab} of magnetic fields up to 10 T applied parallel to the *c* axis. The general trend is a lowering of ρ_{ab} and a shift to lower temperatures of the relative maximum and minimum. An estimate of the Néel temperature from the maximum negative slope of $\rho(T)$ gives 18.5 K for $B = 0$ and a shift of this temperature down to 12.5 K for the highest field. The former value is in excellent agreement with $T_N = 19$ K derived from a peak in the specific heat [2]. The extrapolation of the latter predicts a vanishing of T_N for $B \approx 11.5$ T.

Fig. 2 shows the Hall effect and the susceptibility measured on the very same samples for both $\boldsymbol{B} \parallel$ and $\perp$ to $\boldsymbol{c}$. We first discuss the case of $\boldsymbol{B} \parallel \boldsymbol{c}$. From the very similar strong temperature dependence of the Hall effect and the susceptibility it is evident that the Hall effect is dominated by magnetic contributions, i.e. by the anomalous part. A decomposition of the total

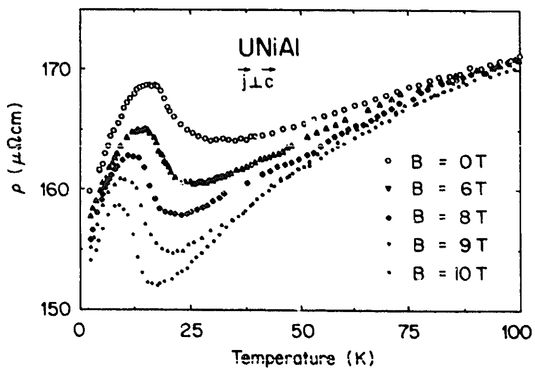

Fig. 1. Temperature dependence of the electrical resistivity in the *ab* plane of UNiAl for various fields $\boldsymbol{B} \parallel \boldsymbol{c}$.

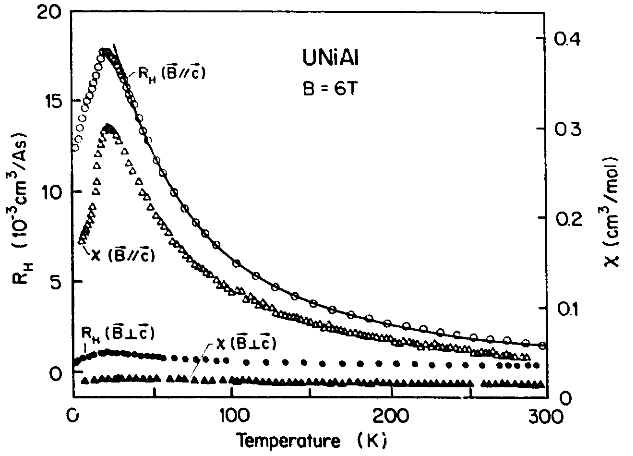

Fig. 2. Temperature dependence of the susceptibility (right scale) and the Hall effect (left scale) of UNiAl for $\boldsymbol{B}\parallel$ and $\perp$ to $\boldsymbol{c}$. The full line is a fit with eq. (1) (see text).

Hall effect for $B = 6$ T into normal and anomalous contributions using the empirical ansatz

$$R_H(T) = R_0 + \chi(T)R_s \quad (1)$$

is shown in fig. 3. We obtain a straight line intersecting the ordinate at the value $R_0 = -1.33 \times 10^{-3}$ cm^3/A s. In a one-band model this corresponds to a conduction-electron concentration of 0.77 per formula unit. The slope of the straight line gives for R_s the positive value 2.17 cm^3/A s. Thus, as commonly observed in uranium compounds [5–8] the total Hall effect is positive at room temperature and below, due to the dominance of the extraordinary Hall contribution. Fig. 2 shows by full line the fit of the total Hall effect with the parameters derived from fig. 3. We see

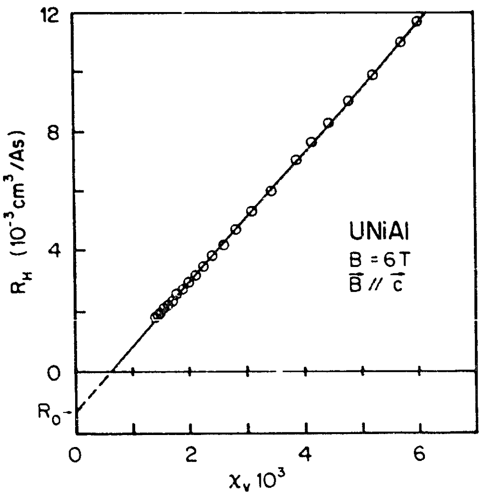

Fig. 3. Decomposition of the total Hall effect for $\boldsymbol{B}\parallel\boldsymbol{c}$ assuming temperature independent normal and anomalous coefficients R_0 and R_s.

that the experimental data start to deviate from our fit which assumes temperature-independent values for R_0 and R_s below 50 K. For fields of 4 and 10 T very similar Hall curves have been obtained and indeed $\rho_H = R_H B$ is linear in B for fields up to 10 T. For $\boldsymbol{B}\parallel\boldsymbol{c}$, we have observed the appearance of small differences in R_H for the current flowing either parallel or perpendicular to the $\boldsymbol{a}$ axis in the ordered phase. This may indicate a magnetic-order induced distortion within the $\boldsymbol{ab}$ plane lifting its isotropy. Recent neutron diffraction measurements in zero field [9] report a complex magnetic structure with a wave vector $\boldsymbol{k}$ = (0.1, 0.1,.0.5).

The pronounced sensitivity of the Hall effect to the magnetization becomes also evident comparing the susceptibility and the Hall effect for $\boldsymbol{B}\perp\boldsymbol{c}$ (fig. 2). While the former appears to be temperature independent on the scale shown, the latter displays a small, but clearly resolved maximum near 20 K. Assuming then also a Curie–Weiss term for $\chi_\perp$, we have tried a decomposition of the Hall effect into a normal and an anomalous part. With a paramagnetic Curie temperature of -30 K we obtain reasonable fits for both the current flowing parallel to $\boldsymbol{a}$ and to $\boldsymbol{c}$. However, contrary to $\boldsymbol{B}\parallel\boldsymbol{c}$, the normal Hall coefficient R_0 is now positive, indicating a strongly anisotropic Fermi surface.

We relate the strong anisotropy of the magnetotransport properties of UNiAl to the substantial difference of the U–U nearest-neighbor separation parallel and perpendicular to the basal plane. In the basal plane the nearest-neighbor U–U separation is 3.5 Å. This puts UNiAl on a Hill plot in the class of itinerant antiferromagnets like UN or nonordering Pauli or van Vleck paramagnets like UC, UAl_2 or U_2PtC_2. In the $\boldsymbol{c}$ direction, the U–U distance of 4.1 Å is similar to the nearest-neighbor distance in UAs, UCu_2Si_2, UPt_3 and URu_2Si_2 which do order and so does UNiAl. However, we do not observe signs of a single Kondo-impurity effect or a coherence effect like in the heavy-fermion systems UPt_3 [6] and URu_2Si_2 [7]. Thus, we suggest to describe the f states as itinerant within the $\boldsymbol{ab}$ plane and as localized along the $\boldsymbol{c}$ axis. The higher resistivity within the $\boldsymbol{ab}$ plane is attributed to a stronger hybridization of the conduction electrons with the delocalized f electrons. On the other hand, the smaller exchange within the $\boldsymbol{ab}$ plane should reduce the sensitivity to magnetic order, explaining the smaller reduction of the resistivity ρ_{ab} in the ordered phase.

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