Journal of NUCLEAR SCIENCE and TECHNOLOGY, Supplement 3, p. 180–183 (November 2002)

On the threshold of long-range magnetic order: $UNi_{2/3}Rh_{1/3}Al$ study

V. SECHOVSKÝ[1*], O. Syshchenko[1], K. Prokeš[1], A.V. Andreev[2], M.I. Bartachevich[3] and T. Goto[3]

[1]*Dept. of Electronic Structures, Charles University, 121 16 Prague 2, The Czech Republic*
[2]*Institute of Physics, AS CR, Na Slovance 2, 182 21 Prague 8, The Czech Republic*
[3]*Institute for Solid State Physics, Tokyo University, Kashiwa 277-8581 Japan*

We present a study of magnetism of $UNi_{2/3}Rh_{1/3}Al$ single crystals in comparison to UCoAl and UNiAl. UCoAl is an itinerant *5f*–electron metamagnet with a critical field of metamagnetic transition $B_c \approx 0.6$ T applied along the *c*-axis of the hexagonal ZrNiAl-type structure. The observed pressure and alloying effects on the ground state point to competing ferromagnetic and antiferromagnetic interactions to be responsible for the non-magnetic ground state. $UNi_{2/3}Rh_{1/3}Al$, a solid solution between an antiferromagnet (UNiAl) and a ferromagnet (URhAl), also shows a nonmagnetic ground state. The present study is devoted to find whether the non-magnetic ground state of UCoAl and $UNi_{2/3}Rh_{1/3}Al$ may be due to a similar underlying microscopic mechanism. Analysis of magnetic-filed induced effects in magnetization, specific heat and electrical resistivity reveals that the physics of $UNi_{2/3}Rh_{1/3}Al$ is dominated by strong antiferromagnetic correlations persisting up to relatively high temperatures. The ground state can be understood in terms of frozen antiferromagnetically coupled U magnetic moments, Contrary, in UCoAl the ground-state U magnetic moment seems to be zero. The field-induced effects on properties of the two systems, although showing apparent analogies, are probably reflecting entirely different microscopic origin.

KEYWORDS: $UNi_{2/3}Rh_{1/3}Al$, ferromagnet, antiferromagnet, itinerant metamagnet

I. Introduction

UCoAl has a paramagnetic ground state, but already in a small magnetic field ($B_c \approx 0.6$ T at 1.6 K) it undergoes a metamagnetic transition to a ferromagnetic ordering of U moments[1,2]. The possibility of metamagnetic transition attributed to the itinerant *5f*-electron metamagnetism is indicated by a *c*-axis susceptibility maximum located at T_{max} ≈20 K. The metamagnetic transition is observed only in field parallel the *c*-axis of the UCoAl hexagonal structure (ZrNiAl-type) whereas in fields along the *c*-plane, a Pauli-paramagnetic response is measured and no metamagnetic transition is observed in fields up to 42 T[1]. The critical parameters (B_c, T_{max}) of UCoAl are sensitive to changes of external pressure and chemical surrounding of U atoms[1,2]. The pressure and alloying induced effects can be conceived within a scenario that considers the non-magnetic ground state in the UCoAl as a result of a delicate balance of ferromagnetic and antiferromagnetic exchange interactions[2]. Having this in mind, we have tried to design a "nonmagnetic" pseudoternary compound with ferromagnetic and antiferromagnetic parent materials study on each side and to investigate, whether the non-magnetic ground state of UCoAl and the new compound may be due to a similar underlying microscopic mechanism.

$UNi_{2/3}Rh_{1/3}Al$ is a solid solution of the itinerant *5f*-electron antiferromagnet (AF) UNiAl (T_N = 19.3 K) with sharp metamagnetic transition at B_c = 11.35 T (at 1.7 K) and the ferromagnetic (F) URhAl with T_C = 27 K[2]. Both parent compounds isostructural with UCoAl and also exhibit the uniaxial magnetic anisotropy. Studies on polycrystals revealed lack of magnetic ordering in $UNi_{2/3}Rh_{1/3}Al$ most probably due to a delicate balance of antiferromagnetic and ferromagnetic exchange interactions[3,4]. In this respect, $UNi_{2/3}Rh_{1/3}Al$ might be a potential isostructural analogue UCoAl. In this paper, we present first results obtained on $UNi_{2/3}Rh_{1/3}Al$ single crystals and discuss them in comparison with UCoAl results published previously. The behavior of $UNi_{2/3}Rh_{1/3}Al$ is also compared to the itinerant *5f*-electron antiferromagnet UNiAl.

II. Experimental

The $UNi_{2/3}Rh_{1/3}Al$ samples used for the study have been spark-erosion cut from a crystal pulled from the melt by a modified Czochralski method. First a 10-gram precursor has been melt under protective high-purity Ar atmosphere from stoichiometric amounts of elementary metals (U of 3N purity, Ni 4N, Rh and Al 5N) on a rotating copper water-cooled bottom in a tetra-arc furnace with tungsten electrodes. Then the crystal was pulled out with pulling speed 15 mm/hour using a tungsten wire as a seed. The check of crystal quality as well as its orientation for cutting has been done using the x-ray Laue method. The phase purity of the crystal and the lattice parameters were determined by a standard x-ray diffraction on a powder sample prepared from a part of the crystal.

[*] Corresponding author: Tel. (+420 2) 21 91 13 67,
Fax. (+420 2) 21 91 1617
E-mail: sech@mag.mff.cuni.cz

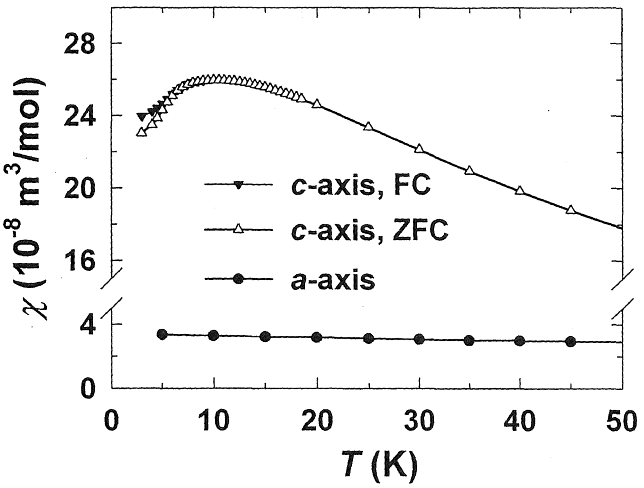

Fig. 1. The χ_c vs. T dependence for the $UNi_{2/3}Rh_{1/3}Al$ single crystal in 0.1 T applied along the *a*- and *c*-axis after cooling in zero-field-cooled (ZFC) and field-cooled (FC) regime. For the *a*-axis the FC and ZFC data sets are identical.

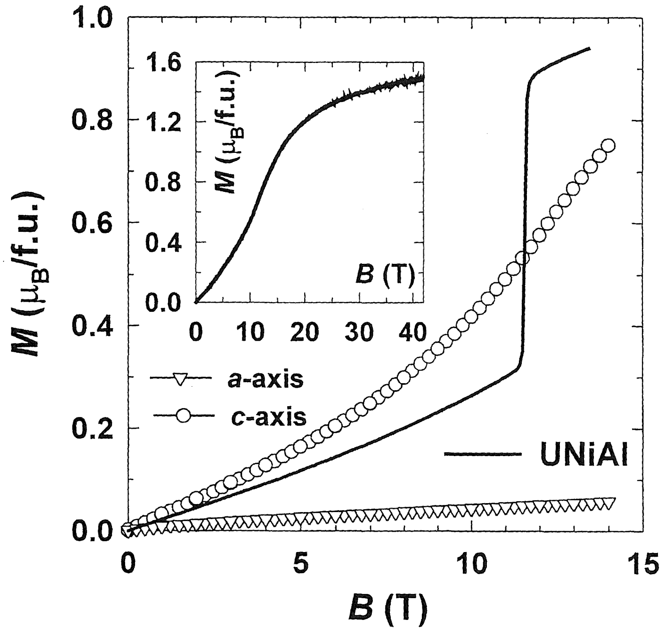

Fig. 2. The $M(B)$ curves for a $UNi_{2/3}Rh_{1/3}Al$ single crystal in fields along the *a*- and *c*-axis at 2 K in comparison with the *c*-axis curve for UNiAl. In the inset a $M(B)$ curve measured at 1.6 K in fields up to 42 T is shown.

The magnetization, specific heat and electrical resistivity as a function of magnetic field and temperature were measured in the temperature range 2-300 K in a Physical Properties Measuring System PPMS-14 (Quantum Design) with a superconducting coil providing fields up to 14 T. The high-field magnetization at 4.2 K was measured by an induction method in pulsed magnetic fields up to 40 T with a pulse duration of 20 ms.

III. Results and discussions

The uniaxial anisotropy was found also in $UNi_{2/3}Rh_{1/3}Al$ exhibiting the characteristic magnetic response in the *c*-axis whereas the basal-plane signal is Pauli paramagnetic. It is clearly demonstrated by the χ vs. T curves in **Fig. 1** as well as in the low-T magnetization data in **Fig. 2**. Consistently, the low-T magnetization is small and increases linearly with field amounting only ≈ 0.05 μ_B/f.u at 14 T. Comparison of *c*-axis magnetization data for $UNi_{2/3}Rh_{1/3}Al$ and UNiAl indicates the difference in physics of the two materials. In UNiAl one observes a sharp first-order type of MT between the low-field antiferromagnetic and high-field ferromagnetic state. The broad S-like anomaly in $UNi_{2/3}Rh_{1/3}Al$ (this

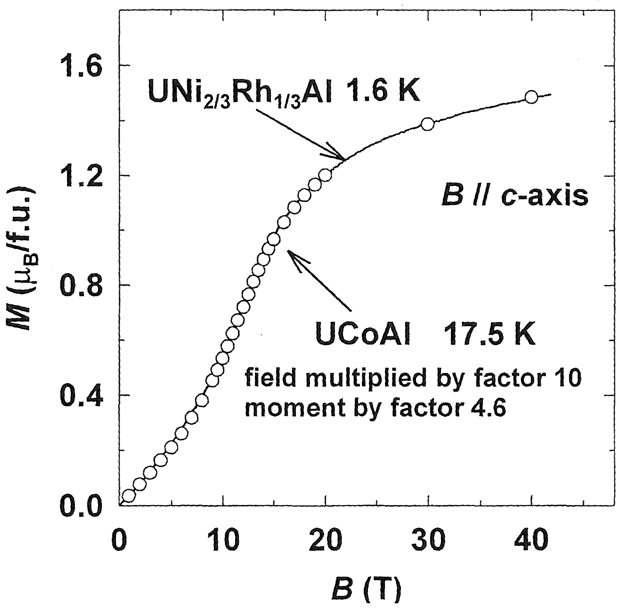

Fig. 3. The $M(B)$ curves for a $UNi_{2/3}Rh_{1/3}Al$ single crystal in fields the *c*-axis at 1.6 K (low and left scale) and on UCoAl at 17.5 K (up and right scale). The magnetic field is multiplied by 10 in case of UCoAl and the UCoAl magnetization is multiplied by 4.6.

feature becomes gradually smeared out with increasing T and disappears above 30 K) can be tentatively attributed to suppression of antiferromagnetic correlations, which probably characterize the low-field state. The idea of antiferromagnetic correlations is consistent also with the $\chi_c(T)$ maximum at 10 K. Splitting of FC and ZFC $\chi_c(T)$ curves points to thermomagnetic history phenomena below 10 K may that might indicate freezing of U moments at low temperatures.

Fig. 3 displays the $M_c(B)$ data in relative scales for $UNi_{2/3}Rh_{1/3}Al$ at 1.6 K and UCoAl at 17.5 K. The two curves almost coincide that reveals main difference between the low-T states in the two compounds. Up to our opinion, the road S-shape $M_c(B)$ anomaly is not connected with real magnetic phase transition to ferromagnetic state but only with suppression of antiferromagnetic correlations. That

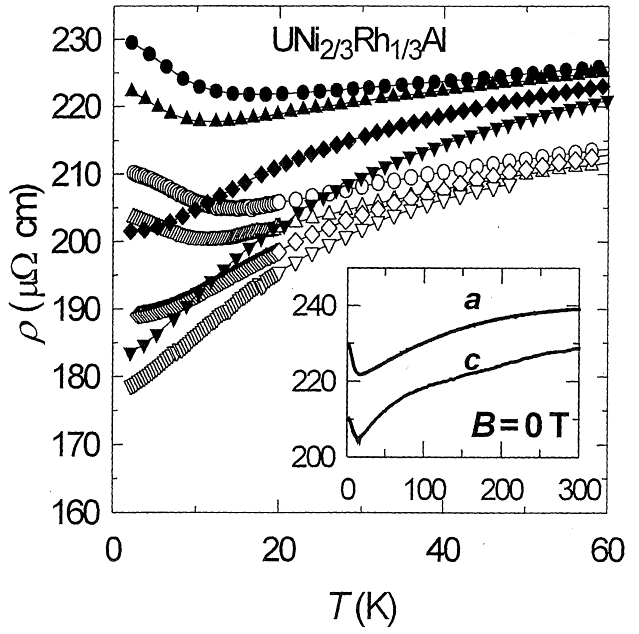

Fig. 4. The $\rho(T)$ curves measured for current along the a- (closed symbols) and c-axis (open symbols) in magnetic fields applied along c.

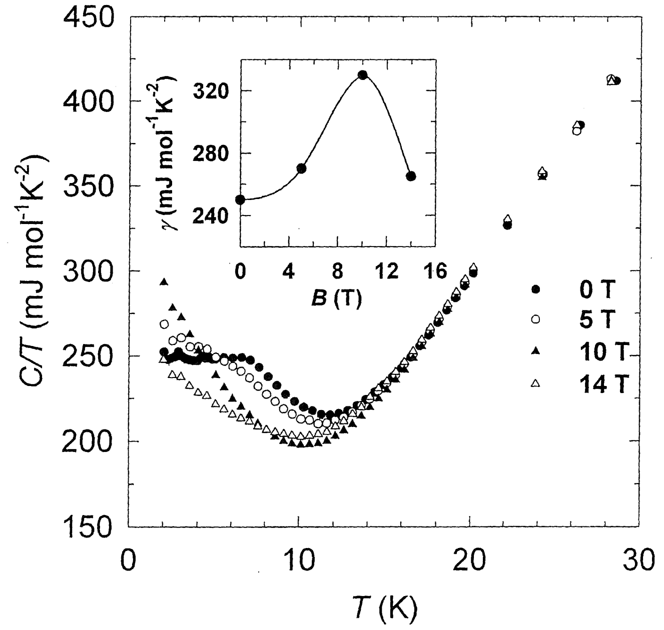

Fig. 6. The C/T vs. T dependence for a $UNi_{2/3}Rh_{1/3}Al$ single crystal in various fields applied along the c-axis. The inset shows the field dependence of the 2 K C/T values.

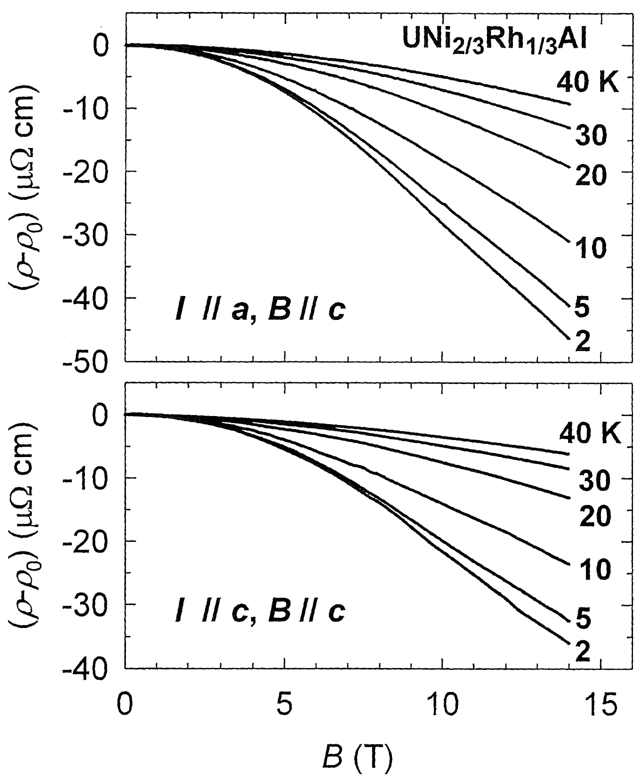

Fig. 5. The $\rho(T)$ curves measured at various temperatures for current along a (closed symbols) and c (open symbols) in fields applied along c.

happens in $UNi_{2/3}Rh_{1/3}Al$ at 1.6 K in contrast to UCoAl in which a sharp metamagnetic transition to ferromagnetic state is observed at 1.6 K[1]. This conclusion is corroborated also by comparison of $\chi_c(T)$ curves for the two compounds. In the first case only a poorly pronounced maximum is observed (χ_{min} at the lowest T is more than 90% of χ_{max} value whereas $\chi < 0.3\ \chi_{max}$ for UCoAl). Splitting of FC and ZFC $\chi_c(T)$ curves for $UNi_{2/3}Rh_{1/3}Al$ points to freezing of U moments.

The c-axis-resistivity at low T and in magnetic fields (see **Fig. 4**) also corroborates influence of antiferromagnetic correlations in $UNi_{2/3}Rh_{1/3}Al$. The $\rho_c(T)$ curve reaches a minimum at 10 K and shows an increase with further decreasing temperature. This feature is gradually suppressed in fields yielding a large negative magnetoresistance (see **Fig. 5**) in correlation with evolution of the S-like $M_c(B)$ anomaly. The $\rho(T)$ curves for UCoAl, on the other hand, show no visible anomaly although they cannot be approximated by a quadratic law that was interpreted in terms of non-Fermi liquid behavior. The quadratic $\rho(T)$ dependence is recovered above in fields above metamagnetic transition[5].

As can be seen in **Fig. 6**, the C/T data are nearly constant below 8 K (≈ 250 mJ/molK^2) and are strongly modified in magnetic fields showing a maximum of the 2 K value of C/T in fields around 10 T.

The low temperature susceptibility, resistivity and specific heat of $UNi_{2/3}Rh_{1/3}Al$[4] and UCoAl[5] by no means exhibit behavior expected for a non-magnetic Fermi liquid. The anomalous low-T scaling of these parameters can be attributed to proximity of magnetic ordering and competing exchange correlations. Fermi-liquid features seem to be restored in both materials in the high-field state (above metamagnetic transition transition or the S-shape anomaly). Nevertheless, physics of the two discussed materials seems to be rather different. This is most probably due to substitutional disorder in the Ni,Rh sublattice and strong role of antiferromagnetic correlations in $UNi_{2/3}Rh_{1/3}Al$.

Acknowledgment

This work is a part of the research program MSM113200002 that is financed by the Ministry of Education of the Czech Republic. It has been partially supported also by the Grant Agency of the Czech Republic (grant # 202/99/0184).

References

1) N.V. Mushnikov, T. Goto, K. Kamishima, H. Yamada, A.V. Andreev, Y. Shiokawa, A. Iwaoand V. Sechovsky, Phys.Rev.B **59** (1999) 6877.
2) Sechovsky, V. and Havela, L. Magnetism of ternary intermetallic compounds of uranium. Buschow, K. H. J. Handbook of Magnetic Materials. [1] (1998) 1-289, Amsterdam, Elsevier Science B.V.
3) A.V. Andreev, R.V. Dremov, V. Sechovsky, Y. Uwatoko, N.V. Mushnikov, T. Goto, K. Prokes, Y. Shiokawa, Y. Homma, I.H. Hagmusaand J.C.P. Klaasse, J.Alloys Comp. **282** (1999) 64.
4) K. Prokes, I.H. Hagmusa, P. Estrela, J.C.P. Klaasse, A.V. Andreev, V. Sechovsky, E. Brückand F.R. de Boer, Physica B **281&282** (2000) 377.
5) A.V. Kolomiets, L. Havela, V. Sechovsky, L. Delongand D.B. Watkins, J.Appl.Phys. **83** (1998) 6435.