Journal of Alloys and Compounds, 207/208 (1994) 249–253
JALCOM 3099

Magnetoresistance in actinide and lanthanide intermetallics

L. Havela and V. Sechovsky
Department of Metal Physics, Charles University, Ke Karlovu 5, 121 16 Prague 2 (Czech Republic)

K. Prokes, H. Nakotte, E. Brück and F.R. de Boer
Van der Waals-Zeeman Laboratory, University of Amsterdam, Valckenierstraat 65, 1018 XE Amsterdam (Netherlands)

Abstract

Magnetoresistance studies in narrow 5f band materials show that a giant drop of electrical resistance accompanies frequently field-induced reorientation of uranium magnetic moments in antiferromagnets. We show that a strong spin-dependent scattering, originating in the strong 5f hybridization with conduction electron states, is the most plausible origin of this effect, the magnitude of which by far exceeds similar effects found in magnetic multilayers and in bulk 4f antiferromagnets.

1. Introduction

Electrical transport in intermetallic compounds of light actinides is at a rather low level of understanding. In general, this originates in the complex behaviour of the 5f electron states, which are more extended in space than the 4f states in lanthanides. In most cases, the 5f states can be described in terms of a narrow band of strongly correlated electron states. The strong interaction between the 5f and conduction electron states, which is due to their hybridization, can be described as a resonant scattering, which strongly affects the transport properties. Inspecting the values of the electrical resistivity in U-intermetallics, a group of materials with small U–U separation (mostly compounds with high U-content) is found to behave in a standard way, *i.e.* similar to transition metal compounds. Their resistivity ρ is usually increasing with T and reaches values not exceeding 200 $\mu\Omega$ cm, which is supposed to be the upper limit in ordinary metallic systems. Typical examples of such "broad-5f-band" materials are Laves phase compounds UT_2 [1,2]. However, in compounds classified as "narrow-5f-band systems" a more exotic resistance behaviour is observed frequently. In these cases, a much higher ρ value (sometimes exceeding 500 $\mu\Omega$ cm and a rather flat high temperature $\rho(T)$ dependence are found as a rule. In compounds displaying magnetic ordering, a steep decrease in ρ in the low temperature range is seen frequently. This decrease is related to the critical temperature of magnetic ordering T_c. One can think about the resistance in such systems as dominated by an enormous spin-disorder contribution of hundreds of $\mu\Omega$ cm, which is gradually removed below T_c. The nearly flat high temperature part of the $\rho(T)$ dependence apparently originates from the fact that the electron-phonon scattering is much weaker than the scattering of conduction electrons by the 5f moments. In the case of such a strong scattering, Matthiessen's rule (the additivity of resistivity contributions of different scattering mechanisms) does not hold.

For ferromagnetic compounds, the values of the residual resistivity ρ are rather small (of the order of 10 $\mu\Omega$ cm), which is expected as spin-disorder resistivity is removed due to the ideal periodicity of magnetic moments in the low temperature limit. The initial increase in ρ with T follows an approximate quadratic dependence, which can be explained by electron-electron and electron-magnon scattering. However, for antiferromagnetic (AF) 5f materials, one observes much larger ρ values, which was originally interpreted as a sign of poor sample quality. This observation was nevertheless confirmed in numerous studies of high-quality single crystals. Moreover, the resistance measurements on anisotropic materials reveal that the anomalously large ρ are found for the current direction along the antiferromagnetic coupling of the moments. The aim of the present paper is to describe the resistance behaviour in 5f antiferromagnets and to compare the results with available experimental data on 4f materials. We concentrate on those compounds that can be driven to a state with ferromagnetically aligned moments by available magnetic fields. The accompanying magnetoresistance effects in actinide materials are found to

SSDI 0925-8388(94)03099-I

exceed even the so-called "giant magnetoresistance effect", observed in magnetic multilayers. However, in both types of materials, a similar mechanism of the resistance drop, connected with the moment reorientation, can be envisaged.

2. Experimental results

This contribution concerns the electrical resistance of UTX compounds, which are formed in different types of crystal structures depending on the choice of the transition metal T and the non-transition metal X. The compounds mentioned here were studied in the form of single crystals prepared by the Czochralski technique in a tri-arc furnace. The only exception is UPdIn for which single crystal whiskers grown along the hexagonal axis were used.

Compounds crystallizing in the hexagonal ZrNiAl structure have been studied most thoroughly. This structure consists of two types of basal-plane layers, U–T and T–X, respectively, alternating along the hexagonal c-axis. The closer U–U spacing and the possibility of 5f–d hybridization within the U–T planes leads to much stronger inter-uranium magnetic coupling within the basal plane than along the c-axis. The anisotropic hybridization leads to a very strong magnetic anisotropy confining the U-moments to the c-direction. The electrical resistivity of antiferromagnetic compounds shows a qualitatively different behaviour for the current along the ab-plane, where U-moments are coupled, as a rule, ferromagnetically, compared to the resistivity for the current perpendicular to the U-layers, where the orientation of moments alternates according to a particular propagation vector. A typical example is UNiGa, which orders antiferromagnetically below 40 K [3]. Its ground state can be characterized by the sequence of $(++--+-)$ orientation of equal U magnetic moments of 1.4 μ_B [4]. In this compound, the ferromagnetic configuration is reached in the applied field of about 0.8–1 T (at 4.2 K). As shown in Fig. 1, the metamagnetic transition, which is of the first-order type, is accompanied by a drastic decrease in the resistivity of about 120 $\mu\Omega$ cm [5]. The commonly used expression $(\Delta\rho/\rho) = (\rho_{AF} - \rho_F)/\rho_F$ which characterizes the magnitude of the resistivity change, yields a value of 6.5.

Knowing the complex magnetic phase diagram of UNiGa [4], we can gain some insight into the magnetoresistance effect by inspection of the $\rho(T)$ dependencies in various magnetic fields (Fig. 2). In sufficiently high magnetic fields, in which a ferromagnetic alignment is achieved, $\rho(T)$ behaves as in a ferromagnet, with a resistance drop below the ordering temperature. Thus the anomalies in $\rho(T)$ found between 35 and 40 K in zero field, which are connected with several different

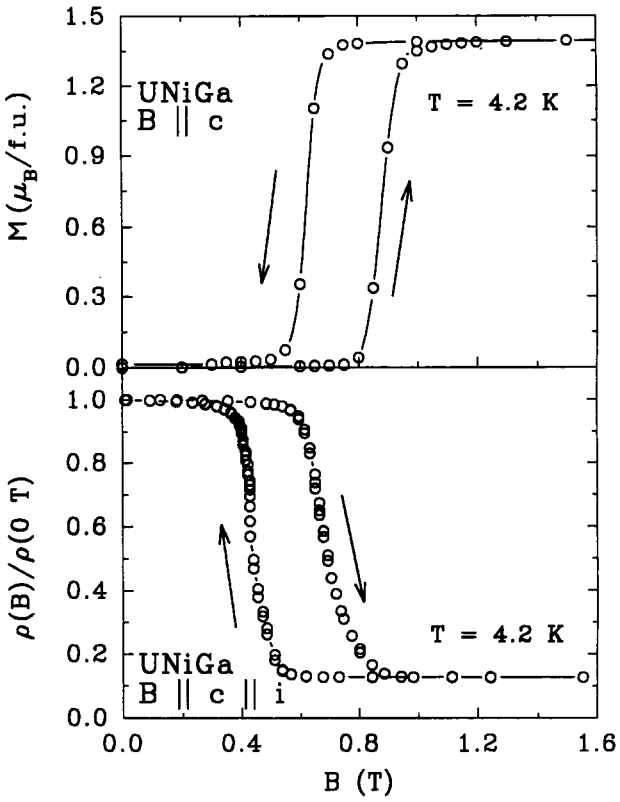

Fig. 1. Field dependence of magnetization (upper part) and relative resistance (lower part) of UNiGa with magnetic field and electrical current along the c-axis (both at T=4.2 K). The fact that the transition was found at slightly higher fields at the magnetization measurement is due to the larger demagnetization field in this case.

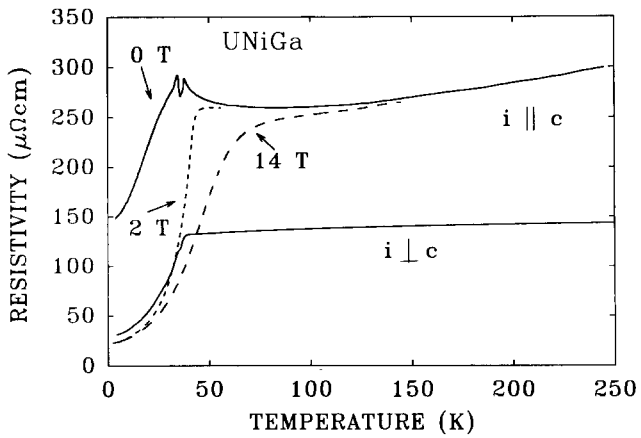

Fig. 2. Comparison of $\rho(T)$ dependences obtained for UNiGa with current along the c-axis in various magnetic fields ($B \| c$), and with current perpendicular to c in a zero field.

magnetic phases, totally disappear in the field of 2 T. Moreover, the low temperature $\rho(T)$ behaviour changes significantly. A quadratic increase is observed in both phases up to approx. 20 K, but the coefficient a is more than four times smaller in the high-field phase (5×10^{-2} $\mu\Omega$ cm K^{-2} compared to 2.4×10^{-1} $\mu\Omega$ cm

K^{-2} in the zero-field phase [5]). Therefore the absolute magnitude of the field-induced effect even increases with increasing T, and a drop of 200 $\mu\Omega$ cm is found at $T=25$ K.

Complementary information is obtained for the resistance for current i perpendicular to c, which is sensing essentially the ferromagnetic ordering of U-moments within the ab-planes. In this configuration, the $\rho(T)$ dependence resembles that of a ferromagnet already in the zero-field phase, but the field applied along c still reduces the residual resistivity from about 30 $\mu\Omega$ cm to 10 $\mu\Omega$-cm [5]. On the other hand, no field effect was observed for B applied along the ab-plane.

The second compound of this type of structure is UPdIn, where the layers of the U-moments of 1.5 μ_B are stacked along c in the sequence $(+-++-)$ [6], which gives a net magnetization of 1/5 μ_U in the ground state. An increase in the resistivity is found at low temperatures for i along c. Below the inflection point in $\rho(T)$ at 20 K, which coincides with the ordering temperature, a gradual saturation to a value of about 80 $\mu\Omega$ cm is observed. For i perpendicular to c, a more regular behaviour with a low temperature decrease of ρ appears [7]. In a field of about 4 T parallel to the c-axis, the magnetic structure transforms into the $(++-)$ stacking, and the full parallel alignment of moments is achieved at $B=16$ T. Both metamagnetic transitions are accompanied by a drop in the resistivity (Fig. 3). The major part of the magnetoresistance effect (total drop by 60 $\mu\Omega$ cm yields $\Delta\rho/\rho=3.5$) is concentrated into the latter transition.

From these two examples with $\Delta\rho \gg \rho$, it is evident that at low temperatures the magnitude of $\Delta\rho/\rho$ is strongly dependent on the residual resistivity ρ_0 in a "ferromagnetic" state, which is related essentially to crystal imperfections. Inspecting the temperature dependence of $\Delta\rho/\rho$, we note that this parameter has to decrease with increasing T even in cases where electron-phonon scattering is not of primary importance. The reason is the increase in ρ_F due to magnetic excitations, which affects $\rho(T)$ progressively as T is raised up to T_c.

The anomalously large magnetoresistance effects are by no means limited to U-compounds with the ZrNiAl-type of structure. A compound which was found to exhibit a similar size of magnetoresistance effect is UNiGe, which crystallizes in the orthorhombic TiNiSi structure type. Below 41.5 K, this compound orders antiferromagnetically with a propagation vector (0, 1/2, 1/2). Applying a magnetic field along the c-axis, one first induces (at about 4 T) another structure with the propagation vector (0, 1/3, 1/3) and a net magnetic moment corresponding to the stacking $(++-)$, while a parallel alignment is achieved at about 10 T [8]. In the longitudinal geometry ($i \| c$, $B \| c$), we find that ρ is

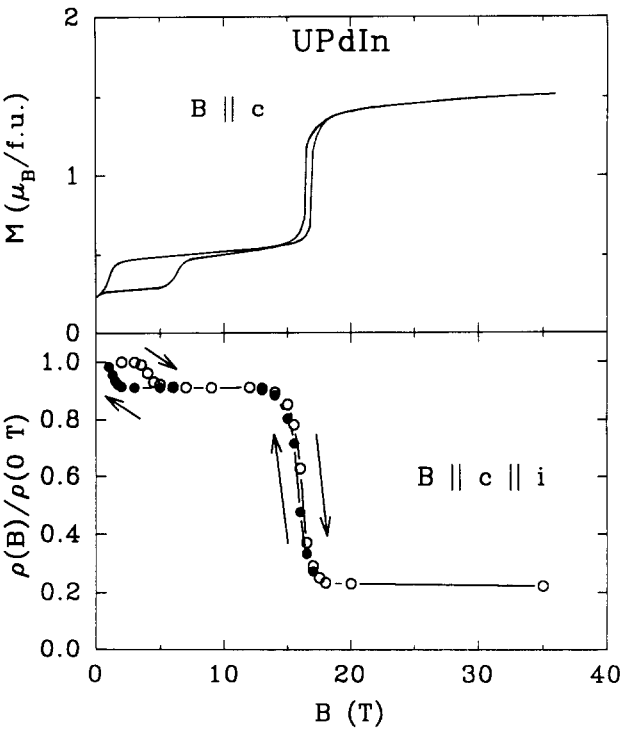

Fig. 3. Field dependence of magnetization (upper part from ref. 7) and relative electrical resistance (lower part) of UPdIn at $T=4.2$ K. Both current and field direction were along the c-axis.

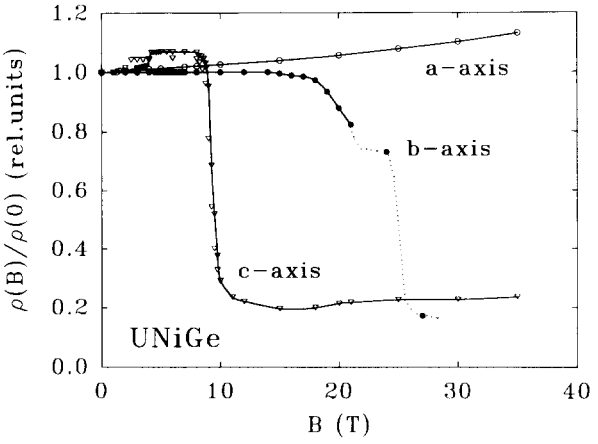

Fig. 4. Field dependence of relative resistance of UNiGe at 4.2 K. Both current and magnetic field were applied along the same crystallographic direction (longitudinal magnetoresistance). Positions of the anomalies coincide with critical fields of the metamagnetic transitions found along the c- and b-axis [8]. The a-axis is the hard magnetization axis, where no transition was observed.

reduced by a similarly large relative value as in the compounds mentioned above (Fig. 4). The absolute value of the resistivity decrease is about 80 $\mu\Omega$ cm. However, in contrast to the previous cases, an initial increase of $\rho(B)$ in the phase with the $(++-)$ stacking compared to the ground-state phase $(+-)$ is found.

Unfortunately, the necessary details of the magnetic structure of UNiGe are not yet resolved.

3. Discussion and conclusions

The examples presented above unambiguously show an additional contribution to electrical resistivity due to the antiferromagnetic coupling of magnetic moments, which can be removed by forcing the moments to orient parallel to each other. This contribution does not vanish in the low-temperature limit, and at low T it can form a substantial part of the total resistivity.

Concerning possible ways by which the antiferromagnetic ordering influences the electrical resistivity, several approaches can be found in the literature. This discussion can discard the possibility that large magnetoresistance in our compounds is due to stacking faults in the magnetic structure. As the residual resistivity in the AF state represents a substantial fraction of the total spin-disorder resistivity (more than 50% in all cases mentioned here), the concentration of such faults should be very large. This can be excluded on the basis of neutron diffraction results obtained on the same crystals, which show well defined periodicity of AF structure.

One approach considers the change of the periodicity of the scattering centers, which leads to new Brillouin zone boundaries. A substantial reconstruction of the Fermi surface may occur, if a new Brillouin zone boundary cuts the Fermi surface into two parts, and an electron energy gap may be created along the new periodicity direction. This mechanism has been applied as an explanation for the increase in ρ in the AF state of rare earths [9]. However, it is not very likely responsible for the major part of the magnetoresistance effects discussed above, because in this case we should also expect drastic changes in the γ-coefficient of the low-temperature specific heat, which reflects the density of electron states at E_F. This is not the case in UNiGa and γ was found to be the same in both F and AF states [4].

Another mechanism, which can lead to a large negative magnetoresistance, is the Kondo effect (*e.g.* for $CeAl_3$ [10]), where the magnetic field can lead to a decoupling of conduction electrons from Kondo centres similar to the effect of the increase in T above the Kondo temperature. The case of UNiGa, where the magnetic moments determined by neutron diffraction are practically the same in the F and AF states (1.4 μ_B/U [4]), again shows that the magnetoresistance effects should be explained without the Kondo effect or any other mechanism related to a 5f-moment instability.

As the gapping of a portion of the Fermi surface is not of primary importance, we concentrate on a spin-dependent scattering mechanisms. In this concept, the electrons with different spin orientation are supposed to experience different potentials and have a different k-space distribution. The origin of the spin-dependent scattering can be understood if we consider the scattering of electrons by a magnetic ion in a state with magnetic quantum number m_s. The electron in spin state s experiences a potential consisting of a non-magnetic part $V(r)$ and the magnetic part given by $-2J(r)\boldsymbol{s}\cdot\boldsymbol{S}$, where J is the exchange interaction parameter. Thus for electrons with $s=+1/2$ (spin-up electrons), the total potential is $V(r)-m_sJ(r)$. Supposing only elastic collisions (without spin-flip scattering), the scattering probability is given by the square of the matrix elements of the total potential between initial and final conduction-electron states. Thus for spin-up electrons, the scattering probability is given by $(\mathscr{V}^2+m_s^2\mathscr{J}^2-2m_s\mathscr{J}\mathscr{V})$, where $\mathscr{J}$ and $\mathscr{V}$ are the particular matrix elements of $J(r)$ and $V(r)$. Similarly, for spin-down electrons one obtains $(\mathscr{V}^2+m_s^2\mathscr{J}^2+2m_s\mathscr{J}\mathscr{V})$. Thus the asymmetry of scattering arises from the interference term. The origin of the spin dependent scattering is even more apparent in band magnetics, where a net magnetic moment arises as due to splitting of spin-up and spin-down sub-bands in the vicinity of E_F (magnetic 3d metals, or *e.g.* U-intermetallics). In such materials, one gets the asymmetry of scattering originating from differences in the density of final states (d or f) for the respective spin orientations, which, in a resonant scattering picture, means different probabilities of resonant scattering for spin-up and spin-down conduction electrons. Moreover, we can suppose that in U-intermetallics the strong hybridization of the conduction-electron states with the spin polarized 5f states may lead to a different s- and p-electron density at E_F for each sub-band.

To understand how the magnetoresistance effect can originate from the spin-dependent scattering, we can benefit from the concepts developed for the description of transport in magnetic superlattices. The magnetoresistance effect in various sandwich structures and superlattices is related to the alignment of direction of magnetization in adjacent magnetic layers [11]. Although the "giant magnetoresistance effect" in multilayers is much smaller than the effects described above, its phenomenology is very similar to that of highly anisotropic layered U-intermetallics, in which the electron mean free path is, similar to multilayers, larger than the characteristic layer spacing (at least in the field aligned state).

A charge transported in a multilayer is considered as being carried independently by spin-up and spin-down channels (spin-flip scattering is neglected). Assuming the probability of scattering of a spin-up conduction electron to be much smaller on spin-up local

moments than on spin-down moments, the charge is transmitted predominantly through the spin-up channel. If, however, the sign of the magnetic moments alternates, transport of spin-up electrons is intercepted by spin-down moments and the total resistance increases substantially. In analogy, in the band magnetism one has to adopt the picture of one spin direction being a majority one in one atomic layer and a minority one in neighbour magnetic layers in the AF state.

Up to now the largest magnetoresistance effects were found in 5f materials (more precisely in light actinide materials). One of the reasons is undoubtedly the strong exchange interaction between the 5f and conduction electrons, which is induced by the hybridization. This interaction is much stronger than in comparable 4f systems because of the larger spatial extent of the 5f wave functions. This may also be the origin of the much larger spin-disorder scattering observed in actinides comparing to lanthanides, where values one order of magnitude smaller are typical [12]. Consistently, one finds resistance changes connected with moment alignment much smaller in 4f systems (*e.g.* [13]) than in 5f systems although the spin moments reach larger values in most lanthanides than in U-compounds.

Nevertheless, the model presented above should be taken with caution in actinides. Its validity is limited to a situation in which spin-flip processes are relatively unimportant. In actinides, the strong spin-orbit interaction (typically about 1 eV) implies that conservation of s over a longer electron path is questionable. A stronger mixing of spin-up and spin-down conduction electron states should then lead to a reduction in the magnetoresistance effect.

Acknowledgments

We would like to thank P.F. de Châtel for a fruitful discussion. This work is part of the research program of the "Stichting voor Fundamenteel Onderzoek der Materie (FOM)", and was also supported by Charles University by Grant no. 312. Part of the work of L.H. and V.S. was supported by the Alexander von Humboldt Foundation.

References

1 V. Sechovsky, Z. Smetana, G. Hilscher, E. Gratz and H. Sassik, *Physica B, 102* (1980) 277.
2 L. Havela, J. Hrebik and J. Sternberk, *Acta Phys. Slov., 31* (1981) 145.
3 L. Havela, V. Sechovsky, L. Jirman, F.R. de Boer and E. Brück, *J. Appl. Phys., 69* (1991) 4813.
4 V. Sechovský, L. Havela, F.R. de Boer, E. Brück, T. Suzuki, S. Ikeda, S. Nishigori and T. Fujita, *Physica B, 186–188* (1993) 775.
5 V. Sechovsky, L. Havela, L. Jirman, W. Ye, T. Takabatake, H. Fujii, E. Brück, F.R. de Boer and H. Nakotte, *J. Appl. Phys., 70* (1991) 5794.
6 E. Sugiura, K. Sugiyama, H. Kawanaka, T. Takabatake, H. Fujii and M. Date, *J. Magn. Magn. Mater., 90–91* (1990) 65.
7 H. Fujii, H. Kawanaka, M. Nagasawa, T. Takabatake, Y. Aoki, T. Suzuki, T. Fujita, E. Sugiura, K. Sugiyama and M. Date, *J. Magn. Magn. Mater., 90–91* (1990) 507.
8 V. Sechovsky, L. Havela, K. Prokes, H. Nakotte, E. Brück, F.R. de Boer, P. Svoboda and H. Maletta, *Proc. 23rd Journées des Actinides*, Schwarzwald 1993, 05.3.
9 R.J. Elliot and F.A. Wedgwood, *Proc. Phys. Soc., 81* (1963) 846.
10 G.M. Roesler and P.M. Tedrow, *Phys. Rev. B, 45* (1992) 12893.
11 M.N. Baibich, J.M. Broto, A. Fert, F. Nguyen Van Dau, F. Petroff, P. Eitenne, G. Creuzet, A. Friederich and J. Chazelas, *Phys. Rev. Lett., 61* (1988) 2472.
12 E. Gratz and M.J. Zuckermann, in K.A. Gschneidner, Jr. and L. Eyring (eds.), *Handbook on the Physics and Chemistry of Rare Earths*, Vol. 5, North-Holland, Amsterdam, 1982, pp. 117–216.
13 P. Svoboda, M. Divis, A.V. Andreev, N.V. Baranov, M.I. Bartashevich and P.E. Markin, *J. Magn. Magn. Mater., 104–107* (1992) 1329.