LA-UR- 95-2834

Title: GIANT MAGNETORESISTANCE EFFECTS IN 5f-MATERIALS

Author(s): L. Havela, V. Sechovsky, K. Prokes, H. Nakotte, H. Fujii, and A. Lacerda

Submitted to: Conference for Strongly Correlated Electron Systems, Goa, India, 26-30 Sep 95; to be published in Physica B

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

Los Alamos
NATIONAL LABORATORY

MASTER

DISCLAIMER

Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.

GIANT MAGNETORESISTANCE EFFECTS IN 5f-MATERIALS

L. Havela[1], V. Sechovský[1], K. Prokeš[1,2], H. Nakotte[3], H. Fujii[3] and A. Lacerda[5]

[1]Department of Metal Physics, Charles University, 121 16 Prague 2, The Czech Republic

[2]Van der Waals-Zeeman Institute, University of Amsterdam, 1018 XE Amsterdam, The Netherlands

[3]LANSCE, Los Alamos National Laboratory, NM 87545, Los Alamos, USA

[4]Faculty of Integrated Arts and Sciences, Hiroshima University, Higashi-Hiroshima 724, Japan

[5]NHMFL, Los Alamos National laboratory, NM 87545, Los Alamos, USA

Abstract

Very large magnetoresistance effects related to reorientation of magnetic moments were observed in a number of U-intermetallics. The resemblance to magnetic multilayers is a basis of discussion of possible mechanisms of these phenomena, in background of which is probably the strong hybridization of *5f-* and conduction-electron states. A clear cut evidence of relative contributions of varied scattering rate on one side and carrier concentration on the other side can be presumably obtained from experiments on samples with controlled disorder

Introduction

Large changes of the electrical resistance of materials under external magnetic field, called Giant Magnetoresistance (GMR) Effects, are known mainly in the context of artificially prepared sandwich and multilayer systems, which are nowadays envisaged as suitable materials for magnetic recording heads. On the other hand, even much larger magnetoresistance effects can be encountered in certain intermetallic compounds. If we exclude those materials, in which a metal-insulator transition can be triggered by magnetic field, we find that a systematic occurrence of GMR effects is connected with a reorientation of 5f-magnetic moments in actinide intermetallic compounds. As the moment reorientation is clearly the driving force also for GMR effects in multilayers, it is interesting to compare phenomena and ideas for their interpretation in both fields of research. Multilayer systems are conceived as semi-classical objects in most of cases, and the higher resistivity in the AF-coupled state is taken as the consequence of magnetic scattering at layer boundaries in conditions of large interfacial roughness. On the other hand, the high-resistance state in bulk antiferromagnets is normally attributed to superzone boundary formation due to an additional (magnetic) periodicity, i.e. in fact the reduction of carrier concentration (Fermi level gapping).

Although the progress in preparation techniques of single crystals of actinide (especially uranium) intermetallic compounds brings an opportunity for a systematic research of transport properties of these materials. Up to now, no coherent theory is able to account for the full range of diverse transport phenomena due to the complexity of the physics of strongly correlated systems. For U-compounds, one has to consider not only effects connected with magnetic ordering, but also instability of magnetic moment itself. Fortunately, the most typical bulk actinide materials displaying large GMR effects are those, which can be characterised as local 5f-moment systems.

Our aim is to discuss effects observed in actinide intermetallics on the background of various approaches being developed mainly in the context of magnetic multilayers. Such analysis can help us to specify plausible mechanisms, which come into consideration for actinide intermetallics, and leads to suggestions how to distinguish between them.

Experimental findings

Here we concentrate on materials, in which a large reduction of the resistivity is connected with magnetic field induced suppression of an antiferromagnetic (AF) coupling in representatives of certain classes of U-compounds.

a) UNiGa belongs to the UTX compounds, which crystallize in the hexagonal ZrNiAl-type structure. It consists of U-Ni and Ni-Ga basal-plane layers alternating along the c-axis, forming thus a naturally layered structure resembling magnetic multilayers. The strong bonding of the $5f$-wavefunctions within the basal plane and the considerable $5f$ orbital moments lead to a huge magnetic anisotropy confining the U-moments in the c-direction and to a ferromagnetic coupling of moments within a basal plane. Therefore, magnetic structures consisting of ferromagnetic basal-plane layers are observed below the Néel temperature $T_N = 39.5$ K [1,2]. The stacking found at low temperatures is antiferromagnetic of the type $+ + - + - -$ with equal U-moments $\mu_U = 1.4\ \mu_B$.

When applying the magnetic field of 1 T along the c-axis, a ferromagnetic state is achieved via a spin-flip metamagnetic transition (Fig.1). Neutron diffraction experiment has revealed that the magnetic moment per U-atom does not change throughout this transition. As demonstrated also in Fig.1, the metamagnetic transition is accompanied by a large drop of the resistivity ρ (for current along the c-axis) amounting to 87 % at $T = 4.2$ K [3,4].

As seen from the Fig.2, for current parallel to the basal plane with ferromagnetically coupled moments, the temperature dependence of resistivity $\rho(T)$ mimics that of a ferromagnet with a dramatic drop at the ordering temperature and a gradual saturation in the low temperature limit [3]. The flat part of $\rho(T)$ above T_N manifestats that the contribution of the electron-phonon scattering, which is linearly increasing in this temperature range, plays only a minor role comparing to the spin disorder scattering (which is of the order of 100 $\mu\Omega$cm). Qualitatively different behaviour of $\rho(T)$ is observed for the current along the c-axis, i.e. sensing the antiferromagnetic coupling of U-moments. Below 80 K, the resistivity increases with decreasing temperature and the increase becomes gradually more pronounced when approaching the ordering temperature. In the range 35-40 K there are several anomalies connected with magnetic phase transitions, and finally ρ decreases at low temperatures, but the residual resistivity for this geometry is much higher (140 $\mu\Omega$cm) than in the former case. Fig 2 gives a general overview how the high resistivity is removed in the ferromagnetic state. It proves also unambiguously that the negative dρ/dT, which develops gradually below about 80 K, is due to incipient antiferromagnetic correlations [2,5].

The analysis of the low temperature data shows that the quadratic dependence $\rho = \rho_0 + aT^2$ accounts well for the resistivity in both AF and ferromagnetic (F) state for both principal current directions. Fig.3 shows, that the transition from AF to F state reduces ρ_0 from 145.9 μΩcm to 22.9 μΩcm, both for $i \,/\!/\, c$. Simultaneously, the quadratic coefficient a is reduced about 4 times from 0.214 μΩcm/K^2 to 0.053 μΩcm/K^2. The reason why the initial slope of the resistivity in AF state is much larger than in F state can be found e.g. in different nature of magnetic excitations in both states, or by different conditions of *e-e* scattering, but such behaviour is expected also from the gapping model discussed later on.

b) UNiGe does not show a distinct layered crystal structure, but magnetoresistance effects are spectacular, as well. It crystallizes in the orthorhombic structure of the TiNiSi type. In this structure type, the nearest U neighbours form a zig-zag chain along the *a*-axis, and therefore U magnetic moments are expected to order in the *b-c* plane [5]. High-field magnetization measurements (Fig. 4) show two metamagnetic transitions with magnetic field oriented along *b*- or *c*-axis, and the *a*-axis is clearly a hard magnetization direction. UNiGe is antiferromagnetic below $T_N = 50$ K (incommensurate structure), but another magnetic phase transition occurs at 41.5 K, and a commensurate magnetic structure with the propagation vector $q = (0,1/2,1/2)$ was found in the ground state [6,7]. If magnetic field is applied along the *c*-axis, UNiGe undegoes first (around 4 T) a transition into a phase with $q = (0,1/3,1/3)$ and a non-zero magnetization $M = 1/3M_s$. The subsequent transition at $B = 10$ T brings the system to a ferromagnetically aligned state. Magnetic moments of about 1.4 μB/U were supposed to be within the *bc*-plane (at least in the ground state), but recent neutron diffraction experiments are suggestive of an alternating canting out of this plane [7].

The metamagnetic transitions are accompanied by significant changes of ρ (Fig. 4). In magnetic field parallel to the *c*-axis, the first transition leads to an increase of the resistivity by several percent around 4 T is followed at 10 T by a drastic drop of ρ to about 20 % of its zero-field value.

Fig.5 shows how the ρ(*T*) behaviour changes with magnetic field. The low temperature behaviour cannot be fitted to a T^2 dependence (or any other power law) only. One has to take into account an additional exponential term accounting for electron-magnon scattering in conditions of an enetrgy gap in the magnon spectrum [5]. In zero field, the width of the gap Δ was found to be about 40 K, and thus the low-*T* data are severely affected by such term, found also in the specific heat data. In the high field state, the magnetic excitations are gradually suppressed by increasing field, which is manifest as increase of Δ to about 100 K in $B = 18$ T.

Discussion

A standard scheme used to explain the magnetoresistance is the two-current model, which assumes two parallel currents, one for each spin direction. It describes certainly well the situation, in which the spin diffusion length, i.e. the distance over which a conduction electron spin does not change its sign, is considerably larger than the electron mean free path λ. This means that an electron is scattered several times conserving its spin. Scattering mechanisms, which do not conserve spins, are those, which lead to changing of the spin of local magnetic moments, as e.g. electron-magnon scattering. Probability of such scattering events is low at low temperatures, but increases with population of magnetic excitations at $T > 0$.

Assuming thus $1/\rho = 1/\rho\uparrow + 1/\rho\downarrow$, where $\rho\uparrow$ and $\rho\downarrow$ are the resistivities pertinent to a respective spin channel. In an asymmetric (as e.g. ferromagnetic) case

$$\rho_F = \rho\uparrow \rho\downarrow / (\rho\uparrow + \rho\downarrow) \qquad (1).$$

A symmetric (antiferromagnetic) case, in which $\rho\uparrow = \rho\downarrow$, leads to

$$\rho_{AF} = (\rho\uparrow + \rho\downarrow)/4 \qquad (2).$$

Having defined $\Delta\rho/\rho = (\rho_{AF} - \rho_F)/\rho_{AF}$, we obtain [8]

$$\Delta\rho/\rho = (\rho\uparrow - \rho\downarrow)^2/(\rho\uparrow + \rho\downarrow)^2 \qquad (3).$$

A multilayer can be represented as a sequence of alternating ferromagnetic and paramagnetic layers, which have to be thin enough comparing to the mean free path of conduction electrons. Then electron travelling perpendicular to such layers can sense a mutual orientation of magnetization in adjacent ferromagnetic layers. Provided large asymmetry between $\rho\uparrow$ and $\rho\downarrow$, current flows mainly through the low-resistance spin channel in a ferromagnetically aligned system. However, if the magnetization direction of adjacent layers alternates (as in the zero-field state), each electron irrespectively to its spin passes through sequence of low- and high-resistance areas and according to eq.2 the total resistance is higher than in eq.1 given the same $\rho\uparrow$ and $\rho\downarrow$.

In multilayers, microscopic reasons for the asymmetry in $\rho\uparrow$ and $\rho\downarrow$ can be found primarily in the spin dependence of the mean free path λ, which can be expressed as $\lambda = v_F \tau$, where v_F is the Fermi velocity and τ the relaxation time, and both v_F and τ can be spin-dependent. The origin of 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 $-2J(r)s\,S$,

where J is the exchange interaction parameter. Thus for electrons with $s = +1/2$ (spin-up electrons), the total potential is $V(r)-m_s J(r)$.

Supposing only elastic collisions (without spin-flip scattering), the scattering probability is given by a 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 $(v^2 + m_s^2 j^2 - 2m_s j v)$, where j and v are the particular matrix elements of $J(r)$ and $V(r)$. Similarly, for spin-down electrons one obtains $(v^2 + m_s^2 j^2 + 2m_s j v)$. Thus the asymmetry of scattering arises from the interference term.

Two metals forming the multilayer have in general different non-magnetic potential. In special cases this difference can be, for one spin direction, compensated by the magnetic terms. Clearly such channel will have very low resistivity and we encounter distinct GMR effects. The scattering probability can be, however, strongly affected by the density of final states, which may be strongly spin dependent. One can imagine that a spin-up electron has a higher scattering probability at the interface between non-magnetic and magnetic layer, if the density of empty states the same spin orientation is dominant at E_F. Presumably in some cases this effect can even outweight a weaker scattering potential in such situation. Finally, we should also consider an effect of spin polarisation of conduction electrons, which can produce a strong asymmetry in the conduction electron density at E_F. Using the two current model, the contiguous spin channel with a high density leads to a low resistivity in the ferromagnetic state, whereas the alternating polarisation in an AF state leads to a restriction of states in the k-space, which can carry the current. In such a situation it is not the mean free path, which is affected, but the effective carrier concentration.

In multilayers, the quantum mechanical approach has been applied only recently (e.g. [9]). It is shown that the coherence of wave function over a superlattice leads to trapping of certain electronic states which do not then contribute to conduction.

Considering all possible reasons for strong asymmetry in $\rho\uparrow$ and $\rho\downarrow$, first we want to realise which mechanisms can be generally translated into the area of bulk magnetic materials of various types. First, we have to dismiss notions like interface roughness scattering, which was assumed to be a crucial ingredient in multilayers. Crystals of intermetallic compounds are normally characterised by a much higher perfection due to weaker intermixing of ions in particular sites, and display clean metamagnetic transitions. Furthermore, it is not usual to consider a compound, even if crystallizes in a layered crystal structure, as a semiclassical assembly of layers of different type (magnetic and nonmagnetic). It has indeed not much sense

in case of the mean free path λ much larger than the characteristic length of the layering. In such case the chemical layering is imperceptible. The effect of magnetic ordering comes as the the quantum effects of periodicity (as the superstructure gaps) and as a scattering on magnetic excitations. In a standard situation the former effect should be dominant at least in the low-T range, where the spin disorder term decays. However, in the case of very strong coupling, i.e. if λ approaches the interatomic distances, the spin dependent scattering on individual AF-coupled layers or atoms can clearly become important, and generally the system can resemble a situation with real-space inhomogeneities. The strength of the spin disorder scattering, which is shown above for the two examples, leads indeed frequently to spin-disorder resistivity ρ_{spd} of hundreds μΩcm.

In many cases we observe that the $\Delta\rho$ values depend on number of $+ -$ interfaces along the electron path. The simple idea, that each such interface works as an effective scattering center, can be however rejected, because we see on the example of UNiGe, that certain reduction of number of such interfaces can even enhance ρ. Moreover, simple quantum mechanical arguments show absence of scattering on any structure with full periodicity. Thus we have to turn attention also to models for bulk antiferromagnets.

In the most simple model [10] the temperature dependence of resistivity in the local-moment antiferromagnetic state can be described by

$$\rho = \frac{\rho_0 + \rho_{e\text{-}p} + \rho_{spd}}{1 - g\, m(T)} \qquad (5)$$

where $m(T)$ is a normalised sublattice (staggered) magnetization, ρ_0 the residual resistivity and $\rho_{e\text{-}p}$ the contribution of electron-phonon scattering. The truncation parameter g characterises the effective reduction of the number of conduction electrons as a consequence of the Fermi level gapping. It is evident that this factor enhances ρ even in the low temperature limit.

The difference between ρ_F (resistance in the ferromagnetic state) and ρ_{AF} (resistance in the antiferromagnetic state) depends on the width of the gap in the electron energy spectrum, which scales with the exchange coupling parameter $\jmath$ between the local moments and conduction electrons. This model is very sensitive to the mutual position of the new Brillouin zone and the Fermi level, and can thus explain the case of UNiGe, in which ρ increases at the transition between two AF structures, although the number of $+ -$ interfaces decreases.

One of characteristic features of actinide intermetallics is a rather strong hybridization of $5f$ states carrying magnetic moments with conduction electron states. Thus we have good

reasons both for a strong spin-dependent scattering and for dramatic gapping effects in these materials. If we wish to distinguish between these two plausible mechanisms, we have to employ methods which can indicate changes of the effective number of charge carriers (Hall effect), or detect dramatic changes of the Fermi surface (e.g. thermoelectric power). Moreover, electron energy gaps should be observable by various spectroscopy techniques. Certain hints could be also obtained by resistivity studies of samples with various purity or perfection, which affects ρ_0. In case of dominating gapping effects one expects at $T \to 0$

$$\rho_{AF} = \frac{\rho_0}{1 - g\, m(T)} \qquad (6).$$

If we vary the residual resistivity in the ferromagnetic state ρ_0, then substituting $\rho_0 + \varepsilon$ for ρ_0, we obtain ρ_{AF} changed by $\varepsilon / [1 - gm(T)]$. This means that $\Delta\rho$ should scale with ρ_0. Similar scaling can be, to our opinion, expected in case where a spin polarisation of conduction electron states and consequent k-space restrictions would play a dominant role. The difference is probably in the fact, that also those AF - F transitions, which are *not* connected with the change of symmetry (enlargement of the unit cell), would lead to a strong GMR effects, too.

Such mechanism could be somewhat preferred inspecting the specific heat results on UNiGa [11] which show practically no change of γ with the crossover to the F-state, which excludes any significant gapping in this case.

In a case, if there was no gapping and all the effect of magnetic moments reorientation provides an additional unspecified magnetic scattering without changing the carrier concentration, the ε value should be only an additive parameter in both states. In the framework of the two-current formalism, these relations can be modified by different situation in each spin channel, but in a strongly asymmetric case, in which one channel is dominant, the relations are still approximately valid. For such type of study, one has to evaluate absolute values of $\Delta\rho$ and ρ_0 for various ρ_0. The most precise way would be to use one sample with fixed geometry of contacts and subject it to irradiation by heavy particles.

Certain hint can be, however, obtained by inspection of $\rho(T)$ dependences in both high- and low-ρ states, which yield also parameters important for any practical application. Magnetic excitations, populated at elevated temperatures, are not only effective scatterers, but they also lead to flipping of conduction electron spins, which is equivalent to mixing of spin-up and spin-down currents and the total conductivity is hampered relatively much more in the ferromagnetic state. If there were not for the channel mixing, ρ_F and ρ_{AF} should keep a mutual proportionality within the gapping model, unless the gap is suppressed or the staggered

magnetization decays close to the ordering temperature. Such class of behaviour is expected for any model in which the effective concentration of conduction electrons is the parameter primarily affected by moments reorientation, an expression similar to (6) should be valid, if we neglect effects like different types of excitations in F and AF state at the same temperature.

Thus situation can be quite different in materials with planar anisotropy, where influence of low-energy excitations of the magnon type can be expected, and in strongly anisotropic uniaxial materials, where no collective excitations exist with energies below the anisotropy gap (which can be minimally 200 K in most of UTX compounds with the ZrNiAl structure [5]). In this case $\rho_F(T)$ will not be affected much by the channel mixing at low T.

Comparison of the two model compounds shows that in uniaxial UNiGa, $\Delta\rho(T)/\rho(T)_{AF}$ is practically constant up to 15 K. Magnetization curves obtained on UNiGe show that this system is close to a planar anisotropy. If we compare ρ-values in fields 0 and 10 T, we see that $\Delta\rho(T)/\rho(T)_{AF}$ decreases systematically with increasing T. This can be consistently explained considering a magnetic inelastic scattering on magnon-like excitations. Such excitations are, however, strongly suppressed already in $B = 18$ T, and $\Delta\rho(T)$ and $\rho(T)_{AF}$ restore their proportionality up to about 20 K.

Conclusions

The occurrence of sizeable magnetoresistance effects in good quality single crystals makes the role of interfacial roughness effects questionable, but the two current model (separate spin-up and spin-down currents with negligible mixing) developed for description of multilayers can be adapted for defect-free crystals, as well. In such model we obtain the high-resistivity state probably due to the Fermi surface gapping or due to k-space restrictions in the AF state. The latter mechanism works apparently even in the situation, where no gapping of the Fermi surface occurs, i.e. if the total carrier density does not change in the AF state.

Acknowledgements

References

[1] V. Sechovský, L. Havela, P. Svoboda, A.V. Andreev, P. Burlet, K. Prokes, H. Nakotte,
    F.R. de Boer, E. Brück, R.A. Robinson and H. Maletta
    J.Magn.Magn.Mater. 140-144 (1995) 1379

[2] K. Prokeš, E. Brück, F. R. de Boer, M. Mihálik, A. Menovsky, P. Burlet, J.M. Mignot, L. Havela and V. Sechovský, J.Appl.Phys (in press)

[3] V. Sechovský, L. Havela, L. Jirman, W. Ye, T. Takabatake, H. Fujii, E. Brück, F.R. de Boer, and H. Nakotte, J.App.Phys. 79 (1991) 5794

[4] L.Havela, V.Sechovský, L. Jirman, F.R. de Boer and E. Brück J.Appl.Phys. 69 (1991) 4813

[5] L. Havela, V. Sechovský, H. Nakotte, E. Brück and F.R. de Boer IEEE Trans. Mag. 30 (1994) 1130

[6] V. Sechovský, L. Havela, A. Purwanto, Allen C. Larson, R.A. Robinson, K. Prokeš, H. Nakotte, E. Brück, F.R. de Boer, P. Svoboda, H. Maletta and M. Winkelmann, J.Alloys.Comp. 293.294 (1994) 536

[7] A. Purwanto, V. Sechovský, L. Havela, R.A. Robinson, H. Nakotte, Allen C. Larson, K. Prokeš, E. Brück and F.R. de Boer, Phys.Rev. B (in press)

[8] J. Inoue and S. Maekawa, J.Magn.Magn.Mater. 127 (1993) L249

[9] P.B. Visscher, Phys.Rev.B49 (1994) 3907

[10] R.J. Elliot and F.A. Wedgwood, Proc. Phys.Soc., 81 (1963) 846

[11] 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

Figure captions

Fig. 1. Field dependence of the magnetisation (upper) and electrical resistivity for current along the c-axis (lower) measured on UNiGa at 4.2 K in magnetic field applied along the c-axis.

Fig. 2. Temperature dependence of the electrical resistivity for current along the c-axis in $B = 0$, 2 and 14 T applied along c. and the zero-field resistivity for current perpendicular to c.

Fig. 3. The low temperature detail of $\rho(T)$ with quadratic fits mentioned in the text.

Fig. 4. Field dependence of the magnetisation M (upper) and ρ for current along the a, b, c-axis (lower) measured on UNiGe at 4.2 K in magnetic field applied along the respective currect direction.

Fig. 5. Temperature dependence of the electrical resistivity for current along the a-axis measured on UNiGe in magnetic fields of 0, 6, 10, and 18 T applied along the c-axis.
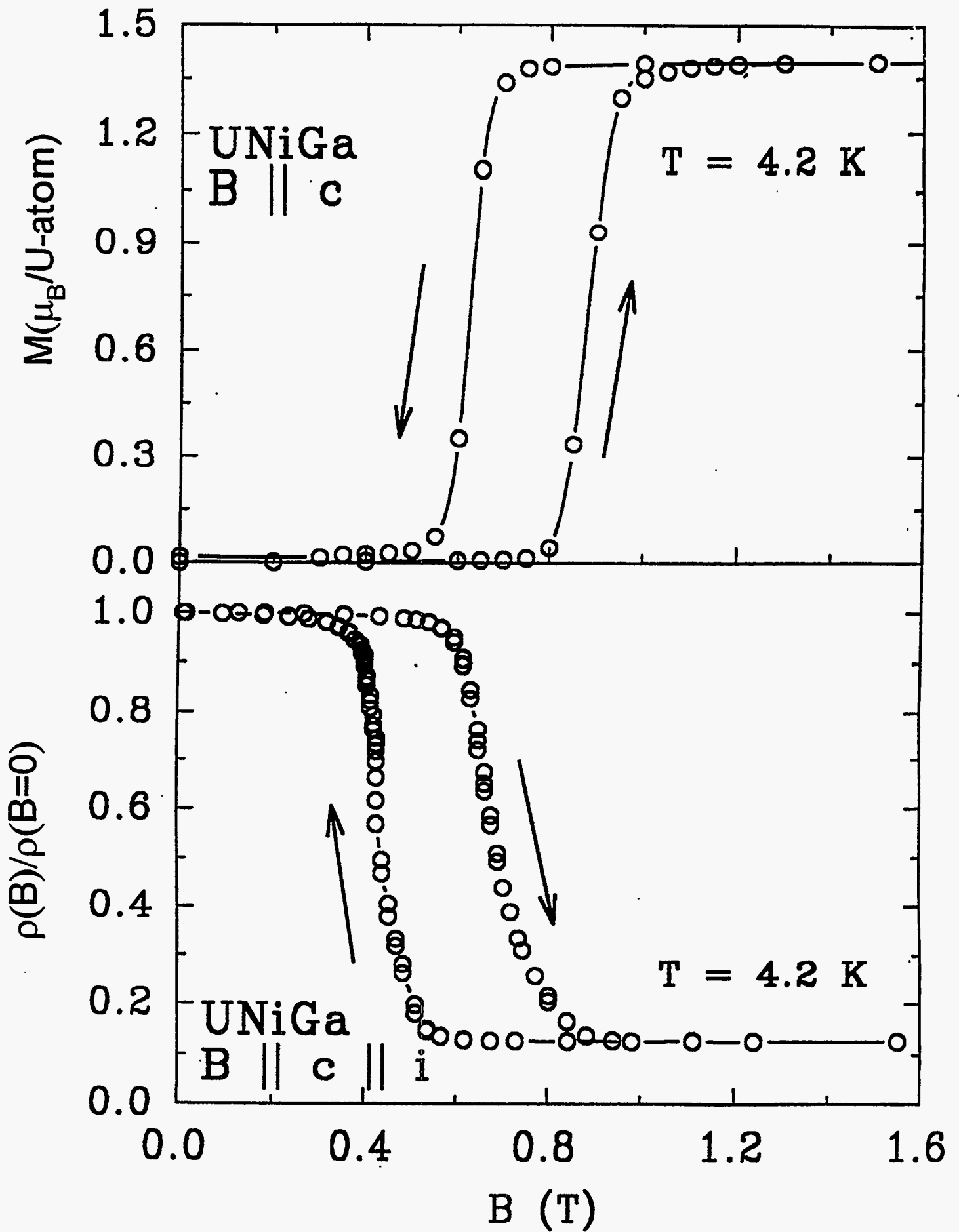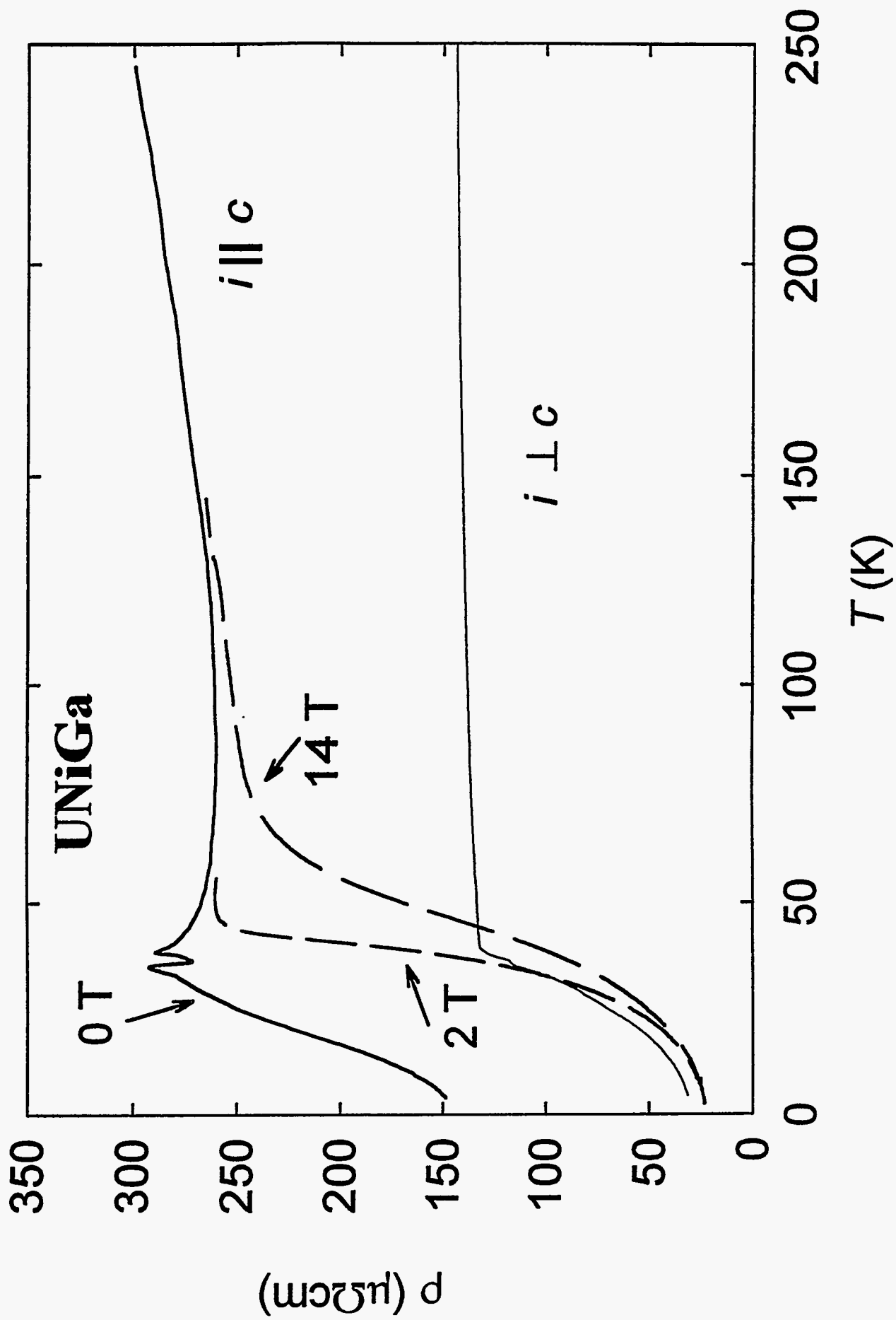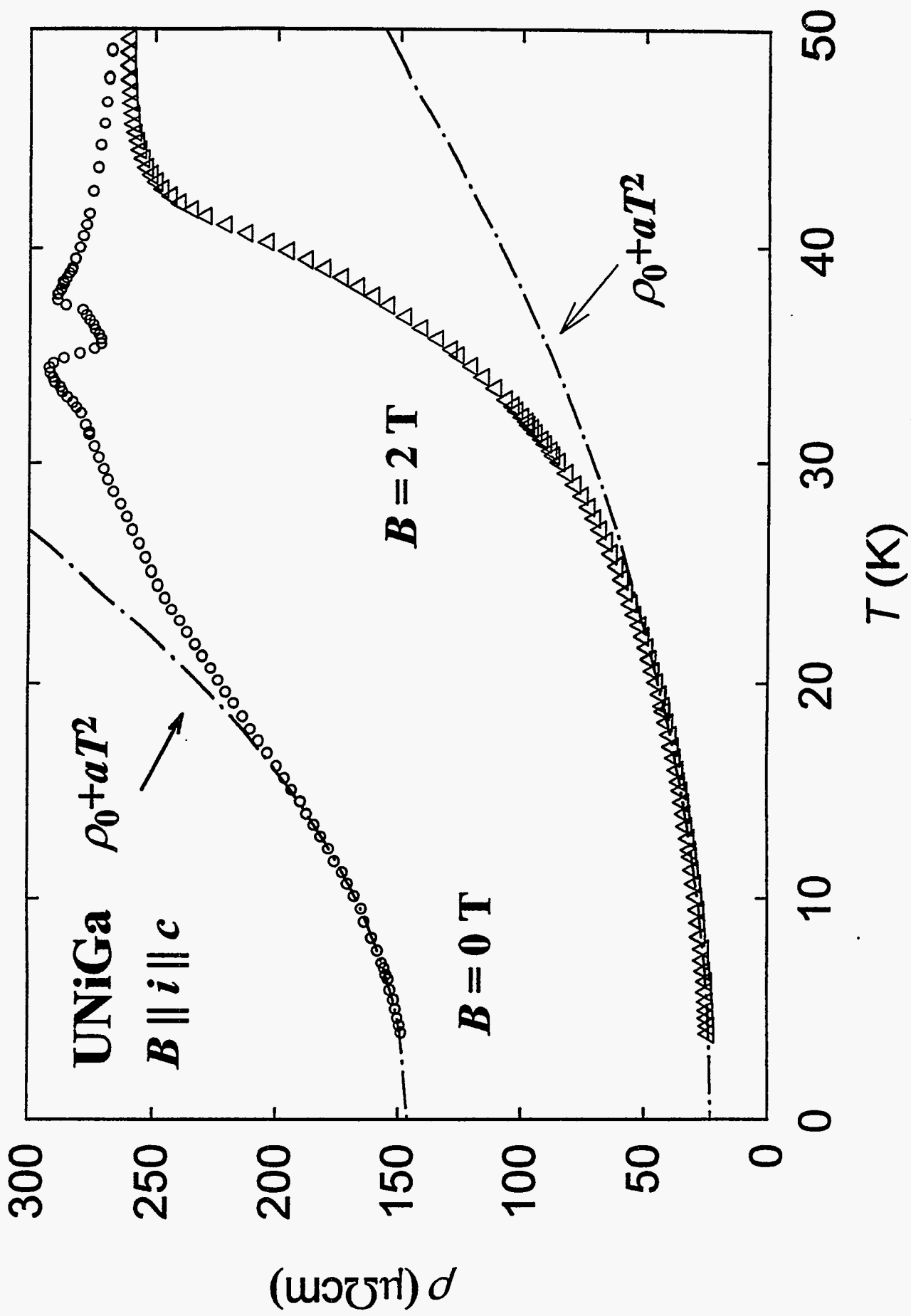
Fig. 2