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Giant magnetoresistance effects in intermetallic compounds (invited)

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Giant magnetoresistance (GMR) effects are observed in several classes of bulk magnetic materials. The resistance changes at metamagnetic transitions connected with reorientation of $4f$ moments are only moderate due to the relatively weak coupling of the $4f$ and conduction electrons. Much larger GMR effects can be achieved by mechanisms involving the d states (RhFe , RCO_2), though the most spectacular resistance variations are connected with metamagnetic transitions in U -intermetallic antiferromagnets. This phenomenon can be interpreted as due to Fermi surface gapping (due to magnetic superzones) and/or due to spin-dependent scattering in analogy with magnetic multilayers.

I. INTRODUCTION

During the dramatic development in the research of magnetic multilayers, the magnetoresistance has become one of the frequently reported quantities. This originated by the discovery a large ("giant") magnetoresistance (GMR) in Fe/Cr multilayers.¹ Since then, further multilayer systems exhibiting GMR have been reported. One of the most impressive effects has been observed in Co/Cu multilayers, in which the electrical resistivity at low temperatures is reduced in magnetic field to half of its zero-field value.² The strong interest in GMR effects is closely connected with promising industrial applications in magnetoresistive reading heads and similar elements.

Magnetic multilayers are artificial superlattices in which the layers of atoms carrying magnetic moments are separated by nonmagnetic layers. The nonmagnetic layers mediate exchange interactions between the magnetic ones. In systems exhibiting GMR, these exchange interactions are, as a rule, antiferromagnetic. As a result we obtain antiparallel interlayer coupling. These exchange interactions are relatively weak and can be easily overcome by magnetic field and a ferromagnetic alignment of spins can be achieved in a relatively low field. A pronounced reduction of the electrical resistivity can be observed due to this transition, because the resistance in the ferromagnetic aligned is much smaller than in the antiferromagnetic case. The occurrence of GMR effects in multilayer systems is usually ascribed to the spin-dependent scattering.³

Magnetic-field-induced transitions in antiferromagnets have been studied in bulk materials for decades. These transitions are called metamagnetic in analogy to the metastable (metamagnetic) state which sets in above the critical field. They take place in a magnetic field sufficient to overcome the antiferromagnetic interactions and to modify the magnetic structure. The modification of the magnetic structure is frequently connected with a change of the translational sym-

metry, and is usually accompanied by a noticeable change of the electrical resistivity.

Another type of metamagnetic transition, which is also reflected in the electrical resistivity, can be found in itinerant electron materials, which are close to a magnetic instability. This class of materials can be well represented by the RCO_2 compounds (R =rare-earth metal). Here a sufficiently large magnetic field can induce magnetic moment on Co sites^{4,5} and, simultaneously, the system undergoes a transition from the paramagnetic to a magnetically ordered state.

Note that the magnetoresistance is usually defined by the expression

$$\Delta\rho/\rho = [\rho(T, H) - \rho(T, 0)]/\rho(T, 0), \quad (1)$$

where $\rho(T, H)$ and $\rho(T, 0)$ are the resistivities at a given temperature in the actual and zero magnetic field, respectively. For the transition from the antiferromagnetic to the ferromagnetically aligned state with the resistivities ρ_{AF} and ρ_F , respectively:

$$\Delta\rho/\rho = (\rho_{AF} - \rho_F)/\rho_{AF}. \quad (2)$$

In the multilayer research it is customary to use a modified expression:

$$\Delta\rho/\rho = \{\rho_{AF} - \rho_F\}/\rho_F, \quad (3)$$

which provides naturally much more spectacular values. Unless especially mentioned, we will use the representations (1) and (2).

Here, we review the magnetoresistance effects in different classes of bulk magnetic materials with a special emphasis on single-crystal data available only very recently for lower symmetry materials. We want to demonstrate that the GMR effects are frequently observed especially in $5f$ electron intermetallics and discuss the origin of these phenomena. We will discuss also required model parameters of an intermetallic system to achieve GMR effects at conditions desirable for practical applications.

II. RESISTANCE IN METALLIC SYSTEMS

For simplicity, the resistance in metallic systems can be discussed as due to independent scattering mechanism (i.e., supposing the validity of the Mathiessen's rule):

$$\rho = \rho_0 + \rho_{e-p} + \rho_{spd}. \quad (4)$$

Both the temperature independent term ρ_0 due to crystal structure imperfections and the electron-phonon scattering term ρ_{e-p} are present in all materials. In magnetic materials the scattering of conduction electrons on magnetic moments should be taken into account. This is reflected in the spin-disorder resistivity term ρ_{spd} . In the paramagnetic range, where the magnetic correlations are absent, electrons are scattered on entirely disordered magnetic moments. The spin-disorder resistivity originates in the exchange interactions acting on the conduction-electron spin and scales therefore with the De Gennes factor $(g-1)^2 J(J+1)$ and with the square of the coupling parameter \mathcal{J} .⁶ For a system with stable magnetic moments, ρ_{spd} is constant above the magnetic ordering temperature. In ferromagnets below T_C , ρ_{spd} decreases with temperature in a way characteristic for the type of magnetic excitation in a particular system. Finally, it should vanish in the low temperature limit. For low temperatures, a quadratic temperature dependence of ρ_{spd} can be derived^{7,8} and is frequently observed experimentally.

The situation is less clear in antiferromagnets, in which the magnetic periodicity can be different from the crystallographic one. This leads to creation of new Brillouin zone boundaries (superzone boundaries) and, consequently, gaps can appear on the Fermi surface. In this case, the effective number of charge carriers is reduced leading to initial increase of the resistivity with decreasing temperature below the Néel temperature T_N .⁹ Such effect can be found both in itinerant antiferromagnets¹⁰ and in rare-earth local-moment systems.⁹ The resistivity can be described as

$$\rho = \frac{\rho_0 + \rho_{e-p} + \rho_{spd}}{1 - gm(T)}, \quad (5)$$

where $m(T)$ is a normalized sublattice (staggered) magnetization. The truncation parameter g characterizes the effective reduction of the number of conduction electrons as a consequence of the Fermi level gapping. It is evident that this factor enhances the resistivity 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 \mathcal{J} . Therefore, the magnetoresistance effect connected with the metamagnetic transition (between the antiferromagnetic state and the state with "ferromagnetically" aligned magnetic moments) should scale approximately with ρ_{spd} , which is governed by similar parameters.

In compounds based on regular rare-earth metals, the ρ_{spd} values are, however, rather modest despite large ionic magnetic moments. The reason is apparently a weak coupling of the conduction electron spin and the ionic spin moment S . Much larger ρ_{spd} values can be expected in materials where a strong coupling can be expected. This situation is

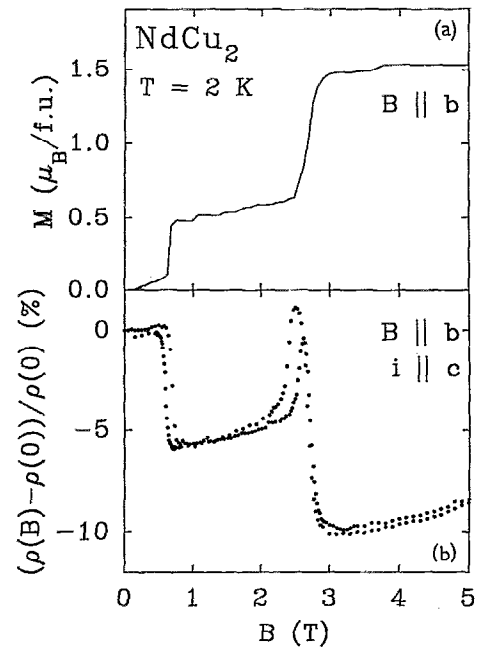


FIG. 1. (a) Magnetization and (b) transversal magnetoresistance vs magnetic field applied along the b axis measured on NdCu_2 single crystal at 2 K (data taken from Ref. 11).

likely in actinide and transition metal intermetallic compounds, as will be demonstrated on several examples.

III. EXAMPLES OF GMR IN BULK MATERIALS

As an example of one of the largest magnetoresistance effects in regular-rare-earth intermetallic compounds, we show in Fig. 1 the magnetoresistance of NdCu_2 which is antiferromagnetic below 6.3 K.^{11,12} By applying the magnetic field along the b axis two metamagnetic transitions can be induced leading to a step-wise increase of the magnetization in ≈ 0.6 and ≈ 2.8 T. Both transitions are associated with pronounced changes of the resistance leading to $\Delta\rho/\rho \approx -10\%$ in 3 T. The increase of $\Delta\rho/\rho$ with the approaching critical field can be attributed to the enhanced scattering due to intersite fluctuations.^{13,14}

The large magnetoresistance changes in NdCu_2 are confined to very low temperatures because of the low value of T_N . Therefore, SmMn_2Ge_2 , which is antiferromagnetic between ≈ 100 and 150 K and ferromagnetic outside this range,^{15,16} might be more attractive for applications. The transition from the antiferromagnetic state to a low-resistance ferro-state can be induced by moderate fields (below 1 T), but the reduction of the resistivity does not exceed 10%.

A similar situation can be found in $\text{Ce}(\text{Fe}_{1-x}\text{Co}_x)_2$, with $\sim 10\%$ cobalt substitution. This material first becomes ferromagnetic below ~ 180 K, and with further lowering temperature a transition to the low-temperature antiferromagnetic state appears at ≈ 80 K.^{17,18} This transition manifests in a dramatic increase of the electrical resistivity.¹⁹ The ferromagnetic (and presumably the low-resistance) state can be then recovered by application of a sufficient magnetic field.¹⁹ Similar loss of ferromagnetism at low temperatures can also

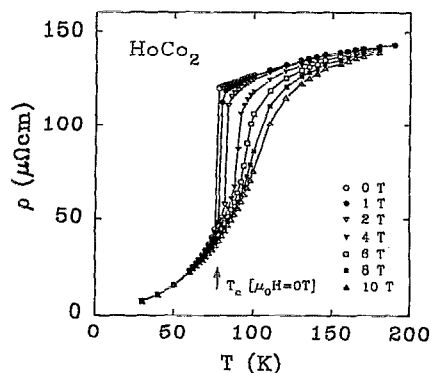


FIG. 2. Temperature dependence of the electrical resistivity in HoCo_2 in several magnetic fields (see Ref. 4).

be observed for small substitutions of Fe (in CeFe_2) by Al,²⁰ Si,²¹ Ru, Ir, and Os,²² which could also lead to large magnetoresistance.

Significantly larger effects can be observed in materials, where the external magnetic field assists in the stabilization of magnetic moments. This situation can be followed, e.g., in RCO_2 compounds. For some rare earths, the Co moments are formed from the highly susceptible matrix of the 3d states by the concerted action of ordered rare earth moments at the Curie temperature T_C , which thus becomes a first order transition. Above T_C , the large resistivity is affected not only by disordered rare-earth moments, but the strong spin-fluctuations in the Co 3d-electron subsystem contribute, as well. The electron-spin fluctuation scattering is removed in the transition by a sudden drop of a considerable absolute value (e.g., $\approx 80 \mu\Omega \text{ cm}$ in DyCo_2 and HoCo_2).^{4,5,23} The effect of exchange fields of the 4f subsystem on the 3d one can be naturally assisted by the external magnetic field, which shifts the transition towards higher temperatures. Thus, for a particular temperature from a limited range above T_C , the resistivity can be reduced drastically. A typical example of HoCo_2 can be seen in Fig. 2. Qualitatively similar behavior also occurs in DyCo_2 , ErCo_2 , and TmCo_2 .^{4,5,23} Although present in compounds containing rare earths, the effect is clearly due to the onset of 3d magnetism in this case.

There are also purely d systems with a large magnetoresistance. One case known for many years is the equiatomic ordered FeRh compound.²⁴ It is antiferromagnetic below 340 K, where it undergoes a transition to the ferromagnetic state. At lower temperatures, the ferro-state can be achieved in the magnetic field, which increases from 0 T at 340 K to about 30 T at 4.2 K (the critical field is 4 T at room temperature). As the resistivity increases strongly at 340 K due to the $F \rightarrow AF$ transition, the suppression of antiferromagnetism in the field leads to a dramatic reduction of the resistivity. As the residual resistivity in the ferromagnetic state can be rather small, the relative drop of its value is largest—by the factor of 20—in the low temperature limit ($\Delta\rho \approx 20 \mu\Omega \text{ cm}$). Recent calculations²⁵ yield the electron structure in both phases. The Fe atoms display local moments of $\approx 3\mu_B$, while the Rh local moments are zero in the antiferromagnetic configuration and $1\mu_B$ for the ferromagnetic configuration. This means that the Rh magnetic moment is induced by the

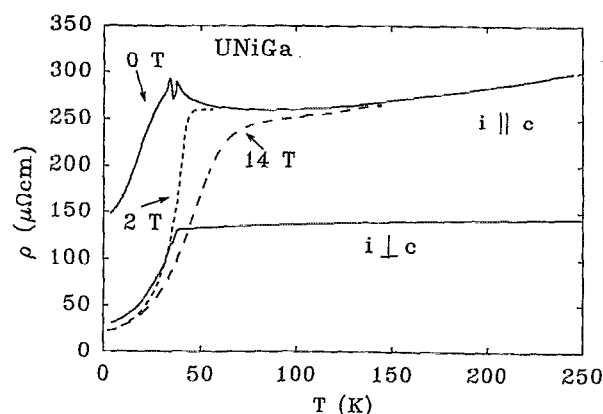


FIG. 3. Temperature dependence of the electrical resistivity in the UNiGa single crystal for $i \parallel c$ and $i \perp c$. For $i \parallel c$ also results measured in 2 and 14 T ($B \parallel c$) are displayed.

$AF \rightarrow F$ transition similar to the onset of the Co magnetic moment at the ordering temperature in some RCO_2 compounds. Then both the spin-flip transition of Fe magnetic moments and the onset of Rh magnetism can be accounted for by the large magnetoresistance change in FeRh . Large magnetoresistance changes accompanying the magnetic-phase transitions in the transition metal sublattice have also been observed in the $\text{La}(\text{Fe}_x\text{Al}_{1-x})_{13}$ system.²⁶

Much larger MR effects occur in systems with uranium, a most thoroughly studied light actinide. We will review some experimental data on UTX compounds, which are formed in several types of crystal structures. The compounds crystallizing in the hexagonal ZrNiAl structure have been studied most thoroughly. This structure consists of U-T and T-X basal-plane layers alternating along the c axis. The stronger U-U coupling within the plane leads to a very strong magnetic anisotropy confining the U moments in the c direction. The magnetic interactions along c are much weaker and in some cases antiferromagnetic with various propagation vectors. The electrical resistivity in antiferromagnetic compounds shows qualitatively different behavior for the current along the basal plane and along the c axis (see Fig. 3). A typical example is UNiGa ,²⁷ which orders antiferromagnetically below 40 K. The ground state can be characterized by the sequence of $(++--)$ orientation of equal U magnetic moments of $1.4\mu_B$. The ferromagnetic configuration is reached in ~ 0.8 –1 T (at 4.2 K). As shown in Fig. 4, the first-order metamagnetic transition is accompanied by a drastic decrease of the resistivity of about $120 \mu\Omega \text{ cm}$ ($\Delta\rho/\rho = 86\%$).²⁸ Knowing the complex magnetic phase diagram of UNiGa , one can gain some insight into the magnetoresistance effect by inspection of the $\rho(T)$ dependencies in various magnetic fields (Fig. 3). In high magnetic fields sufficient to suppress the antiferromagnetic correlations, $\rho(T)$ 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, which are connected with several different antiferromagnetic phases existing in zero field, totally disappear in the field of 2 T. The slower increase of the resistivity in the ferromagnetic phase means that the largest

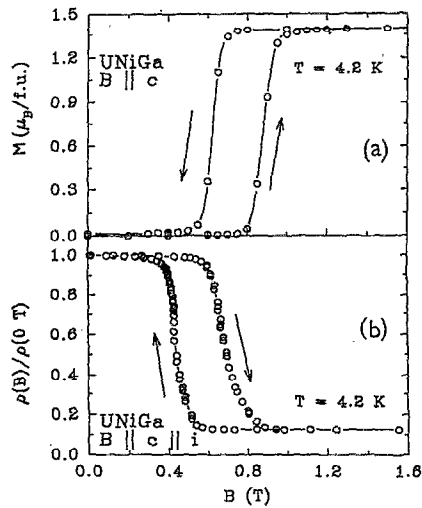


FIG. 4. (a) Magnetization and (b) relative electrical resistivity for $i \parallel c$ vs magnetic field applied along the c axis measured on UNiGa single crystal at 4.2 K.

drop $\Delta\rho \approx 200 \mu\Omega \text{ cm}$ is found at 25 K. Around this temperature a two-step metamagnetic process is observed (see Fig. 5). For current perpendicular to the c axis, which is sensing essentially the ferromagnetic ordering of U moments within the basal-plane sheets, 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 ~ 30 to $\sim 10 \mu\Omega \text{ cm}$.

The second compound of this type of structure is UPdIn, where the layers of the U moments of $1.5\mu_B$ are stacked along c in the sequence $(++--+-)$, which gives a net ground-state magnetization of $1/5\mu_U$. As can be seen in Fig. 6, an increase of the resistivity is found at low temperatures for current along the c axis.²⁹ Below the inflection point in $\rho(T)$ at 20 K, which coincides with the ordering tempera-

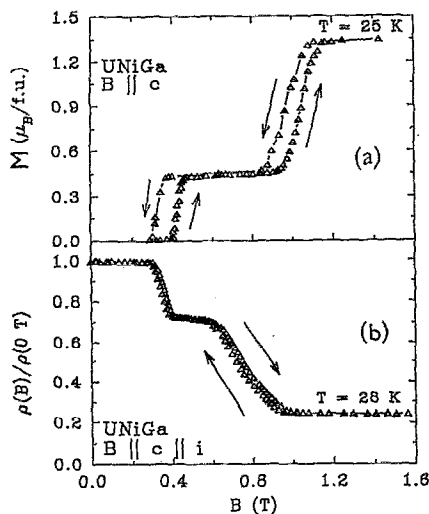


FIG. 5. (a) Magnetization and (b) relative electrical resistivity for $i \parallel c$ vs magnetic field applied along the c axis measured on UNiGa single crystal at 25 and 28 K, respectively.

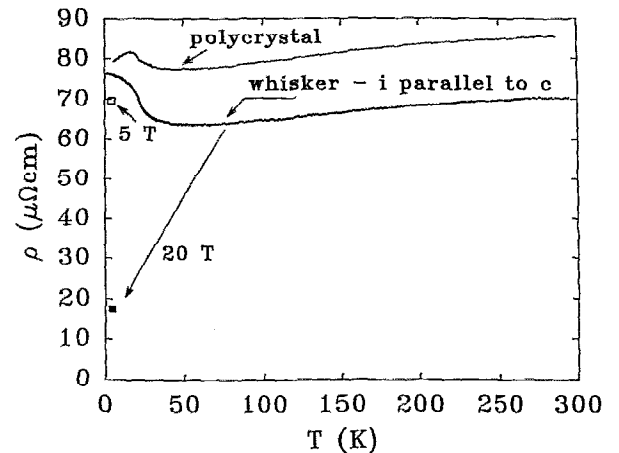


FIG. 6. Temperature dependence of the electrical resistivity of a UPdIn polycrystal and a single-crystalline whisker for $i \parallel c$. The two additional points indicate the value to which the resistance of the whisker drops in applied fields above the two metamagnetic transitions seen in Fig. 7.

ture, a gradual saturation to $\rho_0 \approx 80 \mu\Omega \text{ cm}$ is observed. For current perpendicular to the c axis, a more regular behavior with a low temperature decrease of the resistivity appears. In a field of 4 T along the c axis, the magnetic structure transforms into the $(++--)$ stacking, and the full parallel alignment of moments is achieved in 16 T. Both metamagnetic transitions (Fig. 8) are accompanied by a drop in the resistivity. The major part of the magnetoresistance effect (total drop of $60 \mu\Omega \text{ cm}$) is concentrated into the latter transition. From these two examples with $\Delta\rho \gg \rho_0$, it is evident that the magnitude of $\Delta\rho/\rho$ at low temperatures is strongly dependent on the residual resistivity ρ_0 in a "ferromagnetic" state, which is essentially related to crystal imperfections. Inspecting the temperature dependence of $\Delta\rho/\rho$, we note that this parameter should decrease at high T even in cases where electron-phonon scattering is not of primary importance. The reason is the increase of the resistivity due to magnetic ex-

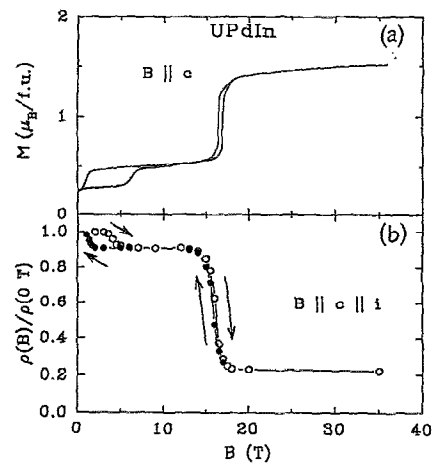


FIG. 7. (a) Magnetization and (b) relative electrical resistivity for $i \parallel c$ vs magnetic field applied along the c axis measured on UPdIn single crystal at 4.2 K.

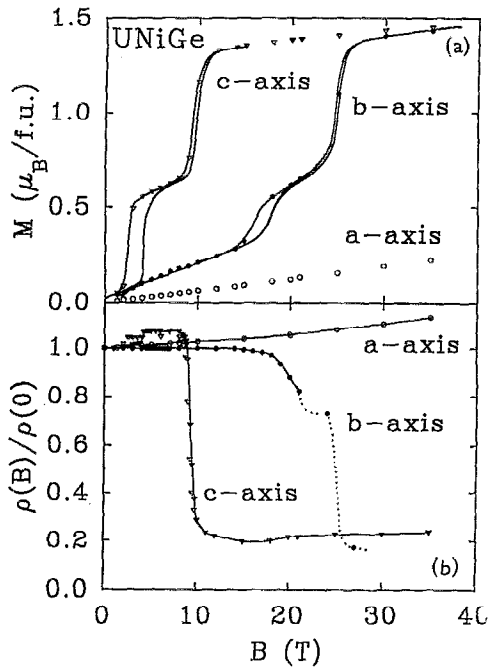


FIG. 8. (a) Magnetization and (b) relative electrical resistivity for $i||c$ vs magnetic field applied along the three principal crystallographic axes measured on UNiGe single crystal at 4.2 K.

citations, which affects $\rho(T)$ progressively with raising temperature up to the ordering temperature.

UNiGe³⁰ 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 the magnetic field along the c axis, one first induces (in approximately 4 T) another magnetic structure with the propagation vector $(0,1/3,1/3)$ and a net magnetic moment corresponding to the stacking $(+-)$. The parallel alignment of U moments is achieved above 10 T. In the longitudinal geometry ($i||c, B||c$), we find that ρ is reduced by a similarly large relative value as in the compounds mentioned above (Fig. 8). The absolute value of the resistivity decrease is about $80 \mu\Omega \text{ cm}$. However, in contrast to the previous cases, $\rho(B)$ first increases by the transition from the ground-state phase $(+-)$ to the one with the $(++-)$ stacking.

UPdGe³¹ is formed within the same structure type. Unlike UNiGe, it is ferromagnetic below $T_C=28$ K. The antiferromagnetic ordering (long wavelength modulated structure) existing between 28 and 50 K, can be suppressed by a moderate magnetic field applied along the c axis (see Fig. 9). The transition field can be tuned by temperature variations, and the maximum size of the drop of ρ can reach about $150 \mu\Omega \text{ cm}$.

IV. DISCUSSION AND CONCLUSIONS

The examples presented above show unambiguously an additional contribution to the electrical resistivity due to the antiferromagnetic coupling of magnetic moments, which can be removed by forcing the moments to orient parallel to each

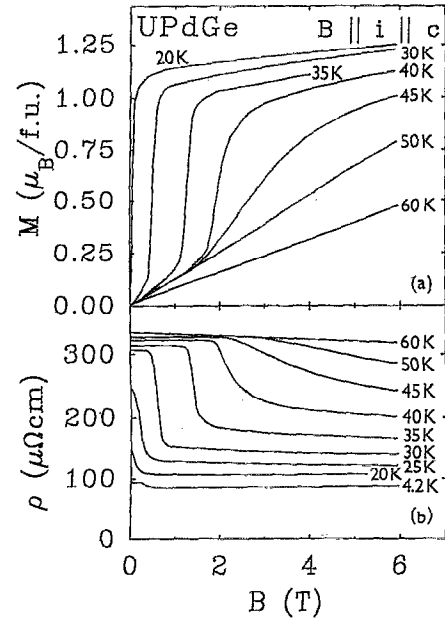


FIG. 9. (a) Magnetization and (b) electrical resistivity for $i||c$ vs magnetic field applied along the three principal crystallographic axes measured on UPdGe single crystal at different temperatures.

other. This contribution does not vanish in the low-temperature limit, and at low temperatures it can contribute a substantial part of the total resistivity.

Concerning possible mechanisms by which the antiferromagnetic ordering influences the electrical resistivity, one should also consider the spin-dependent scattering, which can play an important role besides the Fermi-surface gapping, and which is often considered in the context of multilayers. 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 different scattering amplitude for electrons with spin parallel or antiparallel to ionic magnetic moments in a local moment case. In band magnetism, the asymmetry of the scattering is given due to significantly different partial densities of states at E_F for each subband (spin-up or spin-down). In both cases, the increment of the resistivity in the AF state is dependent on a concentration of $+-$ interfaces. It is worth systematic experimental effort to check whether it is really the case, i.e., whether the larger resistivity in the AF state is due to an additional scattering mechanism, or if the explicit parameter is the modification of the effective conduction-electron concentration. In a case such as this there should be a proportionality between ρ_{AF} and ρ_F irrespectively of, e.g., the ρ_0 value. The unique proportionality constant given by the truncation factor mentioned above should be observed at least in the low- T limit, where different excitations in antiferromagnetic and "ferromagnetic" states can be neglected. The size of the magnetoresistance effect should scale more with the size of the AF unit cell in this case.

Keeping in mind an applicability potential, future effort should focus on materials with exchange interactions strong enough to guarantee ordering in the room temperature range.

As shown in the example of FeRh, this need not lead to extremely high metamagnetic fields in compounds with competing ferro- and antiferro-interactions. The orientation on artificial multilayers or cluster systems is apparently not the only promising stream of GMR research.

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