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Comparison of giant magnetoresistance in multilayer systems and uranium compounds

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The enormous magnetoresistance effect at low temperatures in various uranium-based intermetallic compounds, which exceeds by far the effect found in magnetic-multilayer systems, is a consequence of the strong $5f$ conduction electron hybridization. The consequences of spin-dependent scattering for the giant magnetoresistance (GMR) effect in uranium intermetallics will be discussed and compared with the behavior in multilayer systems. Possible changes of the model parameters to achieve giant magnetoresistance (GMR) effects at higher temperatures, which are desirable for practical applications, are considered.

I. INTRODUCTION AND MOTIVATION

The occurrence of a large reduction of the electrical resistance upon application of a magnetic field in some magnetic-multilayer systems has attracted much attention in view of the possible applications (e.g., magnetic field sensor devices). As this reduction is much larger than the usual magnetoresistance effect in nonmagnetic and magnetic metals, one usually speaks about "giant magnetoresistance" (GMR) in this context. The GMR effect was originally discovered in Fe/Cr multilayers.[1] Since then, several other magnetic multilayer systems exhibiting the GMR effect have been found, which may be as large as $\Delta\rho/\rho=(\rho_{AF}-\rho_F)/\rho_F=65\%$ at room temperature and may exceed 110% at low temperatures (e.g., in Co/Cu multilayers[2]). The strong dependency on the layer thickness and interface mixing (roughness, etc.) reported (for a review see Ref. 3) request several special preparative conditions for multilayer "growth" like ultrahigh vacuum, good layer thickness control, etc. This is a serious drawback with respect to possible applications, because at present such conditions can be achieved only in rather specialized laboratories, so that applications seem to be far from practice.

On the other hand, there exists a second class of materials exhibiting the GMR effect, which was hardly noticed by research groups actually dealing with the GMR in multilayers. Recently, we have reported on the GMR in various uranium-based compounds, which crystallize in layered-like structures.[4–7] By driving these compounds through a metamagnetic transition towards an induced ferromagnetic alignment, a drastic reduction of the electrical resistivity is found, which exceeds that of multilayer systems by far and is as large as $\Delta\rho/\rho=650\%$ at 4.2 K in the case of UNiGa.[4,8] Although the GMR in the uranium compounds studied up to now is only present at low temperatures and (in most cases) relatively high magnetic fields, a deeper understanding of the mechanism responsible of the GMR effects in these materials could yield a considerable impetus in materials research.

In this contribution, we review the magnetoresistance effects observed in different UTX compounds (T=transition metal and X=p electron metal), which have been studied extensively on single crystals. The GMR effect found in UNiGa,[4,8] UNiAl,[5,8] UPdIn,[6,8] and UNiGe[7,8] will be compared with the effect in multilayer systems. The mechanism responsible for GMR in both types of systems and their main differences will be discussed.

II. EXPERIMENTAL RESULTS

Within an extensive research program on UTX compounds, we have studied the compounds crystallizing in the hexagonal ZrNiAl structure in most detail. The structure is built up by alternating U–T and T–X basal plane layers. For the compounds crystallizing in this structure, we find the uranium magnetic moments coupled ferromagnetically within the U–T planes, while the weaker interplane coupling is ferro- or antiferromagnetic leading to ordering with magnetic moments pointing in the c direction. In magnetic multilayers as well, ferromagnetically ordered layers are found, but in contrast to the uranium compounds here the magnetic moments are confined to the plane.

A typical example of uranium compounds is UNiGa, which orders antiferromagnetically at 40 K. Its antiferromagnetic ground state is characterized by a sequence of (++−−+−) orientations of equal magnetic moments. At low temperatures, UNiGa aligns ferromagnetically upon application of a magnetic field of about 1 T along the c axis. In the configuration $i\|B\|c$ axis, the metamagnetic transition toward the induced ferromagnetic alignment is accompanied by a drastic decrease of the resistivity, which exceeds 120 $\mu\Omega$ cm at 4.2 K (Fig. 1). This means a change of the resistivity $\Delta\rho/\rho$ equal to 650%. The effect in the perpendicular configuration $i\perp B\|c$ axis $\Delta\rho/\rho$ is smaller, but it still amounts to 200%. In both configurations, we observe a considerable hysteresis of about 0.25 T at 4.2 K (above 10 K there is no hysteresis observed), which reflects the hysteresis seen in the magnetization curves. No metamagnetic transition is observed for the

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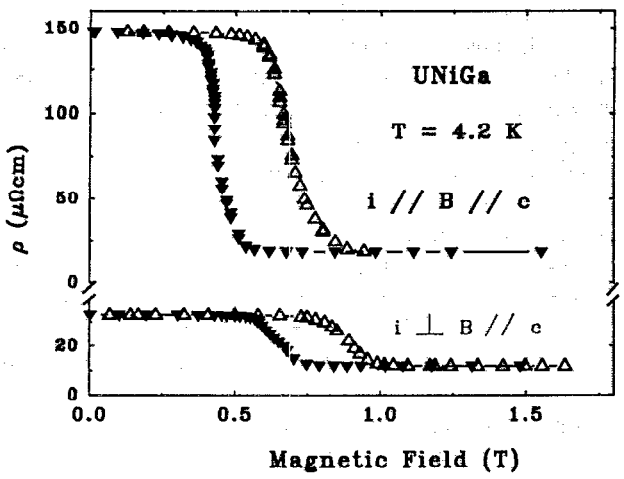

FIG. 1. Field dependence of the electrical resistivity of UNiGa at 4.2 K in the configuration $i\|B\|c$ axis and $i\perp B\|c$ axis. The open symbols represent the result obtained with increasing field, while the full symbols represent the one obtained with decreasing field. The lines are guides to the eye.

$B\perp c$ axis. As a consequence, there is also no comparable field effect in the electrical resistivity.

Another antiferromagnet with the ZrNiAl structure is UNiAl (Fig. 2), which orders antiferromagnetically at 18 K. At 4.2 K, the ferromagnetic alignment in this compound is achieved in fields above 11 T. Again, we find the metamagnetic transition to be accompanied by a drastic reduction of the electrical resistivity for fields applied along the c axis ($\Delta\rho/\rho$=425% for the $i\|B\|c$ axis and $\Delta\rho/\rho$=330% for the $i\perp B\|c$ axis). In UNiAl, the slow saturation tendency in the ferromagnetically aligned phase may originate from spin fluctuations present in this compound. In contrast to the previous compound, we do not observe any hysteresis.

Giant magnetoresistance effects in uranium intermetallics are not limited to pure antiferromagnets, which is illustrated by UPdIn, where an uncompensated antiferromagnetic structure with the stacking sequence $(++-+-)$ is found

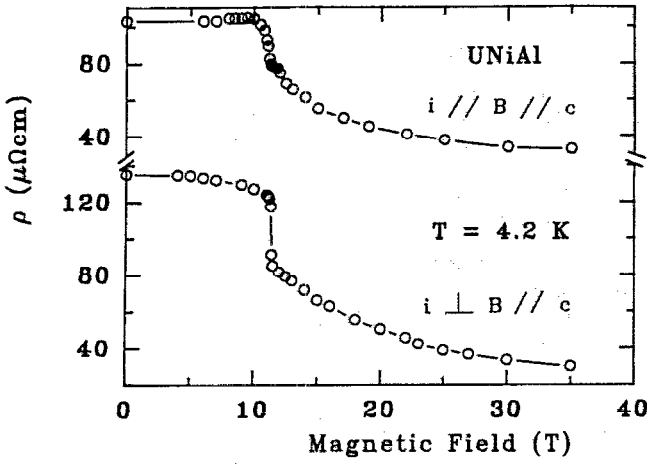

FIG. 2. Field dependence of the electrical resistivity of UNiAl at 4.2 K in the configuration $i\|B\|c$ axis and $i\perp B\|c$ axis. The symbols represent the data points measured in fields, which were kept constant for at least 150 ms. The lines are guides to the eye.

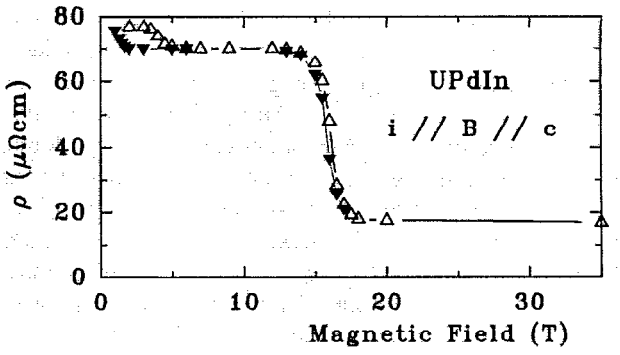

FIG. 3. Field dependence of the electrical resistivity of UPdIn at 4.2 K in the configuration $i\|B\|c$ axis. The open symbols represent the data points measured in fields, which were kept constant for at least 150 ms. Full symbols represent the measurement obtained after a preceding 20-T pulse (decreasing field). The lines are guides to the eye.

below 8.5 K. At 4.2 K, the application of a magnetic field ($B\|c$ axis) yields a transformation of the magnetic structure of UPdIn to a structure with a stacking sequence $(++-)$ in a field of about 3 T, which displays a hysteresis of about 1 T. The full ferromagnetic alignment is achieved above 17 T. Again, we find both metamagnetic transitions to be accompanied by a reduction of the electrical resistivity (Fig. 3). The total effect $\Delta\rho/\rho$ amounts to 350%.

The example of the antiferromagnet UNiGe, which crystallizes in the orthorhombic TiNiSi structure, shows that GMR effects are not restricted to a ZrNiAl structure-type of compounds. At 4.2 K, the destruction of the antiferromagnetic structure of UNiGe is reflected by two metamagnetic transitions at 3 and 9 T for the $B\|c$ axis (Fig. 4). For this compound, the total effect $\Delta\rho/\rho$ is equal to 290%. However, in contrast to the previous example an increased electrical resistivity is found for the intermediate phase between 3 and 9 T.

III. DISCUSSION AND CONCLUSIONS

In the ZrNiAl-type of compounds, the closer U–U distance and the stronger $5f$-d hybridization within the U–T layers lead to a much stronger magnetic coupling of the ura-

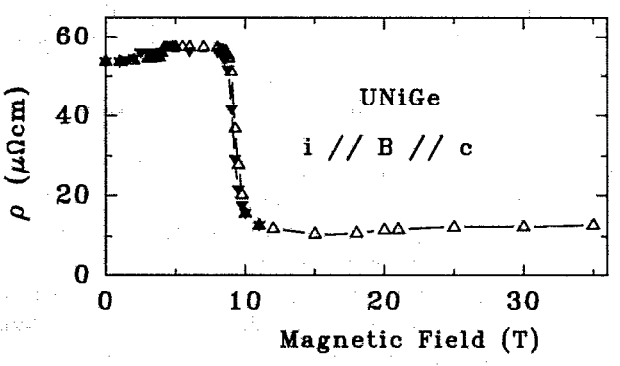

FIG. 4. Field dependence of the electrical resistivity of UNiGe at 4.2 K in the configuration $i\|B\|c$ axis. The symbols represent the results obtained with increasing (open triangles) and decreasing fields (full triangles), which were kept constant for at least 100 ms. The lines are guides to the eye.

nium magnetic moments within the basal plane than along the c axis. For these compounds, the hybridization-induced exchange yields a strong ferromagnetic coupling within the plane, while the coupling along the c axis is much weaker. For TiNiSi-type of compounds, a similar situation is found, in this case with strong coupling along the a axis. In magnetic multilayers, one also finds strong ferromagnetic intralayer coupling and "weak" interlayer coupling, although in these systems the direction of the moments is confined within the layers. On the other hand, similar to the situation in magnetic multilayers, in the uranium antiferromagnets the weak antiferromagnetic coupling can be overcome by relatively low external magnetic fields, which is a necessary prerequisite of the GMR effect. The occurrence of GMR effects in multilayer systems is usually ascribed to spin-dependent scattering (for a review see Ref. 9). This raises the question, whether the GMR effect in uranium intermetallics also may be explained by this mechanism. In an antiferromagnetically ordered compound, spin-dependent scattering is thought to occur at defects. In particular, magnetic atoms occupying sites in the nonmagnetic layers may be effective scatterers (this is the analog of surface roughness in multilayer structures). However, in view of the expected number of such atoms and the estimated value of spin-disorder scattering per atom make this an unlikely mechanism to account for the observed effects. Therefore, the origin of such a spin-dependent scattering term in the electrical resistivity remains unclear. A possible mechanism would be electron–electron interactions. In order to understand why the GMR effect is larger in uranium compounds than in the multilayers, we have to consider the differences of both systems in this context. The most obvious difference is the "layer" thickness and quality (determined by the crystal structure). In contrast to multilayers, in uranium intermetallics we are dealing with a perfect sequence of monolayers. Therefore, we cannot invoke the picture of subband conductivities, where crossing an interface is said to lead to a switching between majority and minority bands, resulting in resistance. Instead, the influence of the antiferromagnetic order on the band structure has to be considered.

In this picture, the electrical resistivity in the antiferromagnetic state in the uranium intermetallics is determined by a term involving impurity scattering and by the presence of new Brillouin zone boundaries, which eliminate parts of the Fermi surface. In the ferromagnetically aligned state, the electrical resistivity of the uranium compounds is thought to be mainly determined by the impurity term, which might be reduced with respect to its value in the antiferromagnetic state, due to the alignment of impurity spins. An analogous reduction must occur in multilayers, so that this cannot explain the large GMR effect in uranium compounds. A drastic change in the topology of the Fermi surface, in the effective number of carriers and in the Fermi velocities would be necessary to account for the spectacular effects observed. If this explanation is correct, one may doubt whether the large GMR effects discussed above can ever be achieved in multilayer systems.

With respect to possible applications, the compounds discussed in this contribution incur some problems. Quite apart from the problem of introducing uranium in the household or office environment, these compounds have low ordering temperatures and (in some cases) high magnetic fields are necessary to obtain ferromagnetic alignment. However, it may be expected that upon substitution and variation of the composition these parameters can be modified easily. A second difficulty may be that the production of good-quality single crystals is sometimes even more difficult than the production of multilayers. However, to obtain the GMR effect there is no need for single crystals, which is indicated already by the large reduction of the electrical resistivity for current both parallel and perpendicular to the magnetic field. Palstra *et al.*[10] have indeed reported a reduction of the electrical resistivity of UNiGa polycrystal by 60%, which means $\Delta\rho/\rho=150\%$. Furthermore, it should be noted that neither uranium nor a layered-like structure appear to be really necessary ingredients for the occurrence of GMR effects, which was demonstrated by Schinkel *et al.* on FeRh alloys, where the electrical resistivity drops from 22 to 2 $\mu\Omega$ cm upon application of a magnetic field.[11]

In conclusion, the occurrence of GMR effect in uranium compounds emphasizes that there exist alternatives for application with respect to magnetic multilayers. At this stage, we try to get a deeper insight in the mechanism responsible for GMR effects in uranium systems, which could guide the search for compounds suitable for applications.

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[8] Note, that in Refs. 4–7 there is generally used $\Delta\rho/\rho=(\rho_{AF}-\rho_F)/\rho_{AF}$, instead of the expression $\Delta\rho/\rho=(\rho_{AF}-\rho_F)/\rho_F$ commonly used in the multilayer research.
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