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Anisotropic resistivity and giant magnetoresistance in UNi_2Ge_2 and UNiGa from ab initio calculations

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Abstract

The zero-temperature resistivity in UNi_2Ge_2 and UNiGa is investigated theoretically using density-functional bandstructure theory and the linear-response approach for calculating the static resistivity for the antiferromagnetic (AF) and ferromagnetic (F) phases, from which the magnetoresistance (MR) is computed. The resistivity in the AF ground-state of UNi_2Ge_2 is calculated to be quite anisotropic, i.e. $\rho_\parallel/\rho_\perp \approx 2.1$, with $\rho_\parallel$, $\rho_\perp$ the resistivities for current parallel, respectively, perpendicular to the *c*-axis. For some Fermi surface sheets, however, extremely large resistivity anisotropies of ≈10–40 are found. A substantial anisotropic MR of $(\Delta\rho/\rho)_\parallel = -61\%$ and $(\Delta\rho/\rho)_\perp = -14\%$ is predicted. We compare the predicted MR in UNi_2Ge_2 to that of UNiGa, for which we obtain even larger MR values of $(\Delta\rho/\rho)_\parallel = -64\%$ and $(\Delta\rho/\rho)_\perp = -45\%$. The basic mechanism of the MR is, in both compounds, identified to be a superzone Fermi surface reconstruction at the metamagnetic transition. © 1998 Elsevier Science S.A.

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The uranium 5f electrons are responsible for the rich variety of unusual physical properties observed for uranium compounds. Varying from one U compound to another, the 5f electrons may be classified to be nearly localized or almost itinerant, they can give rise to Kondo-type behavior, exotic magnetic structures, and heavy-fermion superconductivity (see, e.g., [1,2]). Electrical transport measurements, i.e., Hall effect, resistivity, and magnetoresistance (MR), are widely used as tools to study U compounds, with the aim to gain information about the behavior of the conduction and 5f electrons. Quite recently, a renewed interest in the MR of U compounds arose [3]. This interest was stimulated by the discovery of the giant MR in Fe/Cr multilayers [4], which lead to many experimental and theoretical investigations of the MR phenomenon itself. Compared to the giant MR observed in transition-metal (TM) multilayers, the MR in U intermetallics is appealing for theoretical investigations for two reasons: First, the MR of several U compounds out ranges by far that of the TM multilayers. Typical examples are UniGa, UNiGe, UPdIn, and UNiSn, where magnetic field induced changes of the resistivity up to a factor of 7 were measured [5,6]. Second, for most U intermetallics the giant MR was measured on pure single crystals. The giant MR in TM multilayers, in contrast, depends on the processed superstructure quality, and therefore incoherent conduction electron scattering off rough interfaces or impurities will instantly contribute to the resistivity, however, in an unspecified amount. U intermetallics are therefore attractive materials for first-principle investigations of the origin of the giant MR phenomenon. Very recently we have given an explanation of the mechanism of the giant (sometimes called colossal) MR in UNiGa, which was shown to be largely due to characteristic Fermi surface (FS) reconstruction at the metamagnetic transition [7]. A similar mechanism has been proposed, but not yet been verified experimentally, for U_2Pd_2In [8].

Apart from the giant MR, there are other unusual transport properties in U compounds that deserve to be mentioned. One of these is the crystalline anisotropy of the electrical resistivity, i.e., the dependence of the resistivity on the crystallographic axes. In several materials, as for instance UPt_2Si_2 and UNi_2Ge_2, an extreme resistivity anisotropy has been found of $\rho_\parallel/\rho_\perp \approx 30$ for $T \to 0$ [9,10]. As yet no theoretical explanation of these measured

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resistivity anisotropies has been proposed. In the present work we report a theoretical study of both the resistivity anisotropy and MR in UNi_2Ge_2 as well as the giant MR in UNiGa on the basis of first-principles bandstructure calculations.

UNi_2Ge_2 crystallizes in the tetragonal $ThCr_2Si_2$ structure, and orders magnetically as a simple (long-range) antiferomagnet below $T_N=77$ K [11]. The physical properties of UNi_2Ge_2 have been studied by several authors [12–14]. The resistivity anisotropy was measured to be $\rho_\parallel/\rho_\perp=26$ at $T=0$ [10]. The conductance in UNi_2Ge_2 is thus quasi-two-dimensional. The resistivity furthermore displays an unusual temperature behavior: there is a sizable jump in $\rho_\parallel$ near T_N, but not in $\rho_\perp$ [10]. A similar, but smaller, jump in the resistivity has been observed in the related compound URu_2Si_2, which belongs to the group of heavy-fermion superconductors [15]. The jump at T_N is supposed to be due to a partial Fermi surface gapping, while also the reduction of spin fluctuations plays a role [16]. In addition, it is interesting that the resistivity anisotropy in UNi_2Ge_2 is different from what has been proposed to be the empirical rule in $U(TM)_2X_2$ (X=Si, Ge) compounds: $\rho_\parallel<\rho_\perp$, if the compound crystallizes in the $ThCr_2Si_2$ structure, but $\rho_\parallel>\rho_\perp$, if it crystallizes in the $CaBe_2Ge_2$ structure [9]. One of the few known exceptions of this rule is the spin-density wave compound UNi_2Si_2 [17].

The physical properties of UNiGa were recently summarized in several papers, to which we refer for details [3,5]. Here we recall that UNiGa crystallizes in the hexagonal Fe_2P structure, which exhibits a natural layer structure, consisting of layers of mainly U atoms in planes perpendicular to the *c*-axis, each of which are separated by a Ni–Ga layer. The magnetic groundstate is a complicated AF structure, in which all the U magnetic moments in one such layer are oriented either parallel or antiparallel to the *c*-axis. Two adjacent U layers are, however, only loosely exchange coupled. An anisotropic, giant MR of $(\Delta\rho/\rho)_\parallel=-87\%$ and of $(\Delta\rho/\rho)_\perp=-58\%$ was discovered in UNiGa [5]. The MR in UNiGa is thus considerably larger than those in the TM-multilayers.

To investigate the resistivity anisotropy and MR we use for the evaluation of the conductivity tensor the linear-response formalism. The static conductivity at zero temperature is given by (see, e.g., [7])

$$\sigma_{ii}=\frac{e^2}{m^2V_{uc}}\sum_{n\mathbf{k}}\tau_n|\langle n\mathbf{k}|\Pi_i|n\mathbf{k}\rangle|^2\delta(E_{n\mathbf{k}}-E_F), \quad (1)$$

where $|n\mathbf{k}\rangle$ is the electron wave function, $E_{n\mathbf{k}}$ the corresponding band energy, E_F the Fermi energy, $\langle n\mathbf{k}|\Pi_i|n\mathbf{k}\rangle$ is the matrix element of the relativistic momentum operator ($i=x,y,z$), V_{uc} the unit cell volume, and τ_n the phenomenological conduction electron lifetime, which may be band dependent. To evaluate Eq. (1) we employ the framework of density-functional theory in the local spin-density approximation (LSDA) [18]. For the calculation of the LSDA energy bands and wave functions we have used a fully relativistic, spin-polarized LMTO method [19,20]. The conduction electron lifetime τ_n is the only unknown quantity in Eq. (1), since it relates inherently to the sample purity. For simplicity, however, we adopt here the constant relaxation time approximation. In this approximation, the unknown, but identical τ_n's drop out when we consider the quotient of two ρ's ($\rho=1/\sigma$). This is the case for the resistivity anisotropy and also for the MR, for which we use the definition: $(\Delta\rho/\rho)=[\rho_F-\rho_{AF}]/\rho_{AF}$, with ρ_F, ρ_{AF} the resistivities of the F phase, and of the simplest AF phase, respectively. In experiment, however, one measures $\rho(B)$ and $\rho(0)$, the resistivity in an applied magnetic field and in zero field. To compare to the experimental MR, we make the identification $\rho_{AF}=\rho(0)$ and $\rho_F=\rho(B)$. This is evidently appropriate for UNi_2Ge_2, which has a simple AF groundstate [11], but for UNiGa it is an approximation, because its AF groundstate is more complicated [6,7].

To start with, we consider the resistivities of UNi_2Ge_2. From Eq. (1) it is clear that the conductivity stems at zero temperature from the FS and from the contribution of the squared Fermi moment $|\langle n\mathbf{k}|\Pi_i|n\mathbf{k}\rangle|^2$ on the FS. For UNi_2Ge_2 we calculate for the AF phase 3 doubly degenerate FS sheets. For the F ordered phase, we find that each of the doubly degenerate FS sheets splits into two distinct FS sheets. In Fig. 1 we show these three FS sheets calculated for the AF and F phase. The first FS sheet has a rather entangled shape, whereas the second and third FS sheets have an almost pillar-like shape. It is this pillar-like shape which gives rise to a large crystalline resistivity anisotropy. In Table 1 we give the computed values for the resistivity anisotropy, for each of the three FS sheets separately and for the whole FS. Since for each AF sheet the two corresponding non-degenerate ferromagnetic sheets are quite similar, we shall consider the average of the two non-degenerate sheets. In the AF phase the resistivity anisotropy varies substantially with the FS sheet: FS sheets No. 2 and 3 exhibit very large anisotropies, of 43 and 10, respectively. The first sheet, however, does not show such a large a isotropy. From Eq. (1) and the fact that the Fermi momentum ($\sim\partial E_n/\partial\mathbf{k}$) is perpendicular to the FS, it is immediately seen that for the FS sheets No. 2 and 3 there will be a big contribution to the conductivity in the plane, because at every point on the FS the Fermi momenturn has a large component in the basal plane. The component in the *z*-direction, however, will be very small. It would even vanish if instead of the obtained nearly pillar-like shape the FS would have a pure cylindrical shape. The third FS sheet is just a bit more rounded than the second sheet, therefore sheet No. 3 yields an already four times smaller resistivity anisotropy of 10.

The total calculated resistivity anisotropy $\rho_\parallel/\rho_\perp=2.1$ is quite small compared to the experimental value of 26 [10]. An explanation of this deviation could be that the itinerant SDA approach is not sufficiently warranted for UNi_2Ge_2.

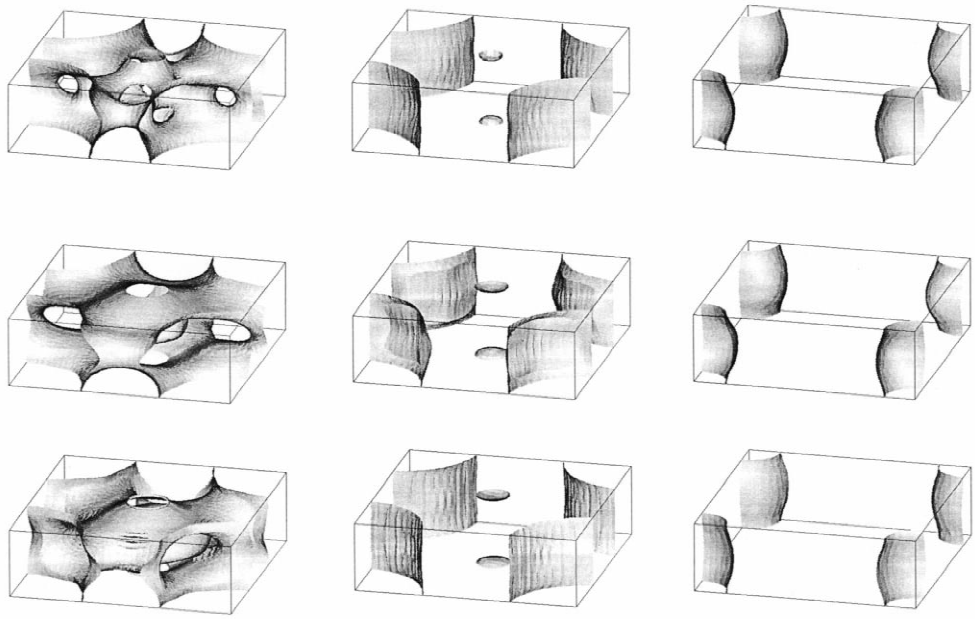

Fig. 1. Calculated FS sheets (Nos. 1–3, from left to right) of UNi_2Ge_2 in the AF and F phase, respectively. For comparison, all FS's are depicted in the AF tetragonal Brillouin zone. The doubly degenerated FS sheet of the AF phase is in all three panels shown in the top subfigure. The corresponding two FS sheets of the F phase are shown in the center and bottom subfigures.

The LSDA itinerant electron description is known to work very well for broad band metals, but it is also known to be limited for the description of narrow band or localized f-electron materials. If the U electrons are localized, the LSDA description can especially be insufficient. At present, however, not much is known about the degree of 5f localization in UNi_2Ge_2. Under these circumstances we can nevertheless conclude from the calculated results, that for both the FS sheets No. 2 and 3 we obtain an extremely large resistivity anisotropy of about the size of the experimental anisotropy. Although the anisotropy behavior of FS No. 1 does not follow the anisotropy trend observed for the other two FS sheets, we expect nevertheless that the calculated pillar-like FS shape is basically responsible for the huge resistivity anisotropy in UNi_2Ge_2. We further mention that the proper resistivity anisotropy, i.e. $\rho_\parallel > \rho_\perp$ is calculated, in agreement with experiment, although this behavior is opposite to the empirical rule proposed for this kind of compound [9].

The MR of UNi_2Ge_2 follows, in the constant relaxation time approximation, from the resistivities of the F and AF phase. The calculated MR values are given in Table 1. The

Table 1
Calculated resisitivity anisotropy $\rho_\parallel/\rho_\perp$ and MR ($\Delta\rho/\rho$) for each of the three FS sheets of UNi_2Ge_2

FS sheet	$\rho\parallel/\rho\perp$		$\left(\frac{\Delta\rho}{\rho}\right)_\parallel$	$\left(\frac{\Delta\rho}{\rho}\right)_\perp$	$\left(\frac{\Delta\rho}{\rho}\right)$
	AF	F			
1	0.7	0.4	−61	−27	−47
2	43.1	10.4	−77	−8	−11
3	10.4	11.7	+18	5	+6
All	2.1	0.9	−61	−14	−31

The MR is given in % for $i\parallel c$, $i\perp c$, and average value.

metamagnetic transition does not give rise to pronounced changes of the FS sheets No. 2 and 3 (see Fig. 1). The MR for $i\parallel c$ obtained TI for FS sheet No. 2 is large, −77%, but due to the nearly cylindrical shape of the main part of the FS, this large MR contribution stems practically only from the small cup-like part of the FS at the Z-point (see Fig. 1). Such a FS part is missing for FS sheet No. 3, wherefore the MR corresponding to this sheet is relatively small. The only FS sheet which gives rise to a large MR is No. 1. We have analyzed the MR of sheet No. 1, with the result that the large MR is mainly due to changes in the squared Fermi momentum $|\langle n\mathbf{k}|\Pi_i|n\mathbf{k}\rangle|^2$ at the metamagnetic transition. The calculated averaged total MR is moderately large, $\Delta\rho/\rho = -31\%$.

It is instructive to compare the resistivity and MR behavior of UNi_2Ge_2 to that of UNiGa. As mention before, UNiGa is one of the ternary U compounds for which a giant MR has been discovered [5], which exceeds by far the MR of TM multilayers [4]. We have very recently investigated the origin of the giant MR in UNiGa on the basis of first-principles electronic structure calculations, employing the above outlined formalism [7]. These calculations unambiguously provide a physical explanation of the mechanism taking place in UNiGa which is responsible for the giant MR: At the metamagnetic phase transition a substantial superzone reconstruction of the FS occurs [7]. This superzone FS reconstruction gives rise to a calculated anisotropic MR of $(\Delta\rho/\rho)_\parallel = -64\%$, and $(\Delta\rho/\rho)_\perp = -45\%$. These calculated MR values are smaller than the experimental data, but this may be related to the fact that we performed the calculations for the simple AF structure and not for the more complex experimental AF groundstate which has a three times larger magnetic unit

cell [21]. The superzone FS reconstruction arising at the metamagnetic transition is illustrated for one characteristic FS sheet of UNiGa in Fig. 2. As can be seen in Fig. 2, the magnetic phase transition brings a substantial reshaping of the FS shape about. Even the topology of the FS changes in the reconstruction of the FS. The FS sheet is initially connected along the $\mathbf{k}_z$-direction for the AF configuration, but it becomes disconnected upon the metamagnetic transition to the F configuration. The MR induced by this FS reconstruction is genuinely giant, and very anisotropic: $(\Delta\rho/\rho)_{\parallel} = -88\%$, $(\Delta\rho/\rho)_{\perp} = -40\%$, respectively. The total MR value, which stems from six calculated FS sheets is, however, smaller because not all sheets contribute as much as the one depicted in Fig. 2 [7].

We next consider the changes in the U moments upon the magnetic transition. Compared to the large resistivity modifications, the magnetic moments change very little. For UNiGa we find, for the spin (orbital) moment: -1.54 μ_B (1.98 μ_B), and -1.54 μ_B (1.99 μ_B), for the F, AF phase, respectively. The calculated total moment is thus smaller than the experimental moment of 1.4 μ_B [21]. For UNi_2Ge_2 the moments change somewhat more: -1.30 μ_B (2.25 μ_B), -1.47 μ_B (2.43 μ_B), for F, AF phase, respectively.

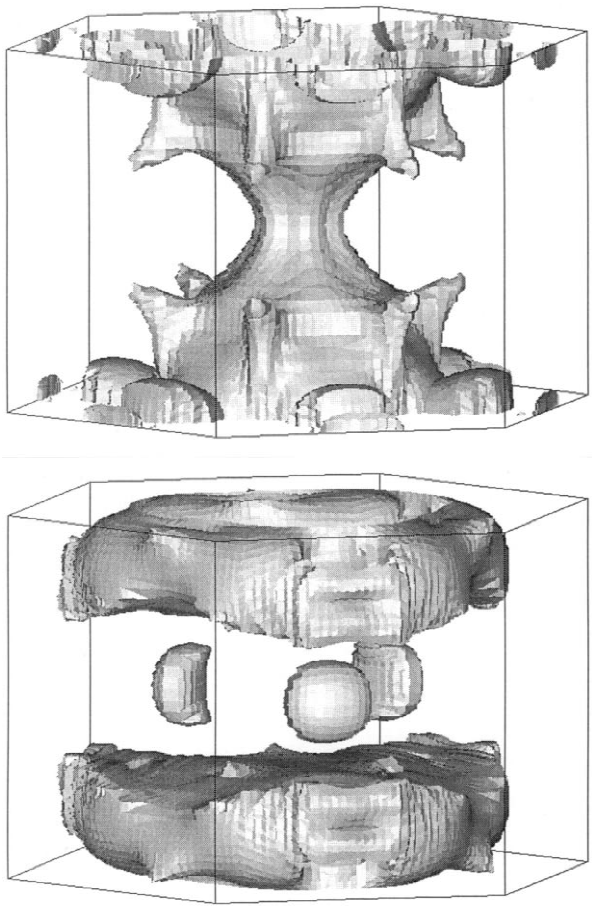

Fig. 2. Example of the characteristic FS reconstruction calculated to take place for one of the FS sheets of UNiGa. The upper figure gives the FS sheet for the AF configuration, the bottom one for the F configuration.

To summarize, we have investigated the resistivity anisotropy and MR in UNi_2Ge_2 and UNiGa. The mechanism which causes the giant MR in UNiGa is a pronounced superzone FS reconstruction at the metamagnetic transition. While the calculated MR values are in semiquantitative agreement with experiment for UNiGa, the enormous resistivity anisotropy measured in UNi_2Ge_2 is not reproduced within the present first-principles approach. The origin of this discrepancy is currently unclear, but it might well be related to the applicability of the LSDA to describe the electronic structure of UNi_2Ge_2. Explaining the quasi-two-dimensional conductance remains thus a challenge for the future. Our calculations predict a reasonably large MR for UNi_2Ge_2. The MR mechanism, however, which causes the giant MR in UNiGa is not found to occur in UNi_2Ge_2.

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