Mechanism of the Giant Magnetoresistance in UNiGa from First-Principles Calculations

V. N. Antonov,* A. Ya. Perlov,* P. M. Oppeneer, A. N. Yaresko,* S. V. Halilov
Max-Planck Research Group "Theory of Complex and Correlated Electron Systems," University of Technology, D-01062 Dresden, Germany
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The giant magnetoresistance (MR) in UNiGa is investigated from first principles, using density-functional band-structure theory in the local approximation together with a linear-response ansatz for the conductivity. The MR is calculated to be very anisotropic, with a large value of $-(45 \pm 5)\%$ for current in plane and a giant MR of $-(64 \pm 5)\%$ for current perpendicular to plane. These values are in semiquantitative agreement with the measured MR. The basic mechanism of the giant MR is identified to be a superzone reconstruction of the Fermi surface at the field induced metamagnetic transition. [S0031-9007(96)01990-4]

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The discovery of the giant magnetoresistance (MR) in Fe/Cr multilayers [1] has lead to a world-wide interest in the MR phenomenon as well as to a nearly "avalanchelike" research activity. After the first report on the giant MR in Fe/Cr multilayers, giant MRs were discovered in Co/Cu multilayers [2], in heterogeneous Co/Cu and Co/Ag alloys [3], and also in a number of "ordinary" intermetallic compounds [4–7]. One of the essential conditions for the occurrence of the giant MR in transition-metal (TM) multilayers is an antiparallel interlayer exchange coupling across the spacer layer [8]. The giant MR effect appears when the magnetic moments, which are initially oriented antiparallel, are aligned parallel in an applied magnetic field. A similar condition is required for a giant MR in intermetallic compounds, which without exception is found to be intimately related to the occurrence of a ground state antiferromagnetic (AFM) phase. As compared to the MR in TM multilayers, the MR in intermetallics is attractive for theoretical investigations of the MR mechanism for two reasons: first, in a number of intermetallic compounds a MR was observed which exceeds by far the giant MR measured in TM multilayers. Examples are $La_{1-x}Ca_xMnO_3$ [4], and in particular also the uranium based intermetallics, as, e.g., UNiGa, UNiGe, UPdIn, and UNiSn, where magnetic field induced changes of the resistivity up to a factor of 7 were measured [5,6]. Second, the giant MR in intermetallics was in most cases measured on pure single crystals [5–7]. The latter situation is in contrast to that of TM multilayers, where, depending on the achieved superstructure quality, incoherent conduction electron scattering off impurities and interfacial roughness potentials may contribute in a distinct, though quantitatively unknown manner. It is therefore of a fundamental interest for the general understanding of the MR phenomenon to explain on a first-principles basis the nature of the giant MR in intermetallic compounds. In this Letter, we report on such an investigation of the giant MR in UNiGa.

It was discovered recently that in an external field the resistivity in UNiGa changes enormously [5]. UNiGa crystallizes in the hexagonal Fe_2P structure [9,10], which exhibits a natural layer structure, consisting of planes of uranium atoms admixed with 25% Ni atoms perpendicular to the c axis, each of which are separated from one another by a layer of Ni and Ga atoms. The magnetic moments of all uranium atoms in one layer are rigidly coupled parallel due to a strong intralayer exchange coupling. Uniaxial magnetic anisotropy leads to an orientation of the moments parallel to the c axis [10]. The interuranium layer exchange coupling is much weaker: With a relatively small magnetic field of less than 1 T it is possible to flip the magnetic orientation of one complete layer. The magnetic phase diagram of UNiGa, furthermore, is quite intricate: there are several AFM phases below the Néel temperature $T_N = 39.5$ K [10]. Some of these are rather complex, for instance, with a sequential stacking of the uranium moments of $++-+--+-$ in the consecutive layers perpendicular to the c direction. In this notation, "+" means $+c$ oriented, and "−" means $-c$ oriented moments, respectively. The ground state AFM structure is characterized by a sequential stacking $++-+--$ of the uranium moments [10]. Recent magnetotransport measurements on pure single crystals yielded a giant negative MR of -87% for current perpendicular to plane (CPP) ($i \parallel c \parallel B$), and a somewhat smaller MR of -58% for current in plane (CIP) ($i \perp c \parallel B$) [5]. With regard to the negative sign of the MR, we note that the definition customary for the MR in uranium intermetallics has been used:

$$\Delta\rho/\rho \equiv [\rho(B) - \rho(0)]/\rho(0). \quad (1)$$

This definition differs from the one commonly used for TM multilayers, in which $\rho(B)$ and $\rho(0)$ are interchanged. Using the multilayer convention, the MR in UNiGa would be about 650% for CPP and about 140% for CIP. Thus, the MR in UNiGa closely resembles that of the TM multilayers, which are also loosely exchange coupled magnetic layers, but the MR in UNiGa is considerably larger.

Previously several theories of the giant MR in TM multilayers were proposed [11–19]. Initially, these con-

centrated on an explanation of the MR in terms of spin-dependent electron scattering off impurities or interfaces [11–15], but simplified the electronic structure. Some recent publications took the effect of the electronic structure on the MR into account, which may be appreciable [16–19]. A general consensus exists on the appropriate formalism for the evaluation of the conductivity σ ($\sigma = 1/\rho$): mainly the Kubo-Greenwood linear-response formulation [20] and also the Boltzmann equation in constant relaxation time approximation [21] have recently been used [11–19]. An exception is the work of Schep *et al.* [22], where the MR of point contacts was considered.

We adopted the linear-response approach [20] to calculate the conductivity of UNiGa. The static, or so called intraband conductivity at zero temperature is given by

$$\sigma_{ii} \propto \tau \sum_{n\mathbf{k}} |\langle n\mathbf{k}|p_i|n\mathbf{k}\rangle|^2 \delta(E_{n\mathbf{k}} - E_F) . \qquad (2)$$

Here $|n\mathbf{k}\rangle$ is the Bloch wave function, $E_{n\mathbf{k}}$ the corresponding band energy, E_F the Fermi energy, $\langle n\mathbf{k}|p_i|n\mathbf{k}\rangle$ is the matrix element of the momentum operator ($i = x, y, z$), and τ the phenomenological conduction electron lifetime. The *inter*band contribution to σ_{ii}, which is important at elevated temperatures and for the frequency dependent conductivity, has been neglected here because the MR was measured at 4.2 K [5]. To evaluate Eq. (2) we calculated the electronic structure within the framework of the local spin-density approximation (LSDA) [23] to density-functional theory. UNiGa requires a relativistic treatment, wherefore we used a fully relativistic, spin-polarized linear muffin-tin orbital (LMTO) method with combined corrections [24,25]. For $\langle n\mathbf{k}|p_i|n\mathbf{k}\rangle$ the following relation holds: $\langle n\mathbf{k}|p_i|n\mathbf{k}\rangle\hbar = m(\partial E_{n\mathbf{k}}/\partial \mathbf{k})_i$ [26]. Substitution of this equality in Eq. (2) leads to an expression for σ_{ii} which is also found from the solution of the Boltzmann equation in constant relaxation time approximation [21]. Thus, in the appropriate limits, these theories yield the same expression for the conductivity, illustrating thereby that Eq. (2) is theoretically firmly established. The electron lifetime τ, which relates to the intrinsic sample purity, is unknown. It may in principle be band and spin (or magnetic ordering) dependent, but we adopted here an isotropic, constant lifetime. The spin dependence can in a good approximation be assumed to be isotropic on account of the spin-orbit interaction of uranium, which causes a strong mixing of minority and majority spin, wherefore no well defined spin states exist. This is an important difference with the MR discussed for TM multilayers. In this approximation the unknown τ drops out when the quotient of two conductivities is considered. The MR is therefore calculated without adjustable parameters.

The unit cell of UNiGa in the ferromagnetic (FM) phase of the Fe_2P structure contains nine atoms. The simplest AFM unit cell, i.e., the one which has $+-$ order of the moments in the c direction, contains 18 atoms. We considered only these two magnetic superstructures for reasons of computational feasibility. The ground state AFM structure corresponds to 54 atoms, which would make the relativistic self-consistent energy-band calculation extremely tedious. Consequently, the theoretical MR is calculated here using Eq. (1), with the identification $\rho_{\mathrm{FM}} = \rho(B)$ and $\rho_{\mathrm{AFM}} = \rho(0)$, with ρ_{FM}, ρ_{AFM} the calculated resistivities of the FM phase and of the simplest AFM phase, respectively. We note that the MR calculated in this way may be different from the one in which the true AFM reference state is used. In the calculations, care was taken to achieve numerical convergence with respect to the basis set size and k-point number. A basis set of spd functions on Ni and Ga, and $spdf$ on U was used. A large number of 24 576 tetrahedra, finally, was used to compute the Brillouin zone (BZ) integral in the irreducible wedge ($\frac{1}{12}$th) of the BZ.

Our band-structure calculations showed that in both the FM and AFM phases there are six bands crossing E_F, which contributed to the MR: for CPP we calculated $\Delta\rho/\rho = -64\%$, and for CIP $\Delta\rho/\rho = -45\%$. Numerical accuracy tests showed that these values may have an uncertainty of $\pm 5\%$. The *ab initio* calculated MRs are in semiquantitative agreement with the experimental values, which are -87% and -58%, respectively. The anisotropy in the CPP and CIP-MR, moreover, is quantitatively described. The trustworthiness of the computed MR depends of course on the applicability of the LSDA approach, which, in the case of UNiGa, has to be clarified.

Two important questions that are to be addressed are: What is the mechanism that causes the giant MR in UNiGa, and what is the validity of the LSDA itinerant electron description for UNiGa? To start with the second point, it is known that the mean-field LSDA approach has limitations in describing strongly correlated electrons. The electrons in the uranium $5f$ shell can be more or less correlated, depending on their degree of localization. To investigate the applicability of the LSDA to UNiGa, we calculated the frequency dependent conductivity tensor using the linear-response formalism (including the interband contribution), from which the magnetooptical (MO) Kerr spectra can be derived [27]. Very recently, the MO Kerr spectra of UNiGa were measured [28], so that we could directly compare measured and calculated spectra. This

TABLE I. Calculated MR for each of the six FS sheets of UNiGa. MR values (in %) are given for CPP and CIP. The experimental MR data are those of Ref. [5]. For completeness' sake the calculated conductivities σ_{AFM} (for CPP and CIP, normalized to arbitrary units) are also given for each FS sheet.

	Fermi surface sheet number						
	1	2	3	4	5	6	Exp.
MR-CPP	−75	−55	−69	−88	+57	−70	−87
MR-CIP	−54	−48	−18	−40	−18	−87	−58
σ_{AFM}-CPP	0.22	0.43	2.35	4.39	9.70	1.42	
σ_{AFM}-CIP	0.28	0.94	2.34	3.30	9.09	1.17	

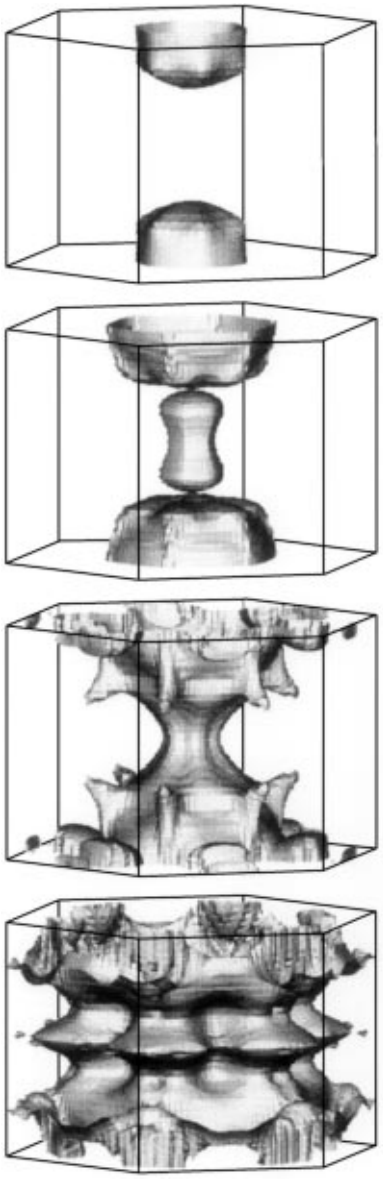

FIG. 1. Four characteristic FS sheets (Nos. 2–5, top to bottom) of AFM UNiGa. The FS sheets are shown in the hexagonal double unit cell.

comparison showed that the LSDA approach gives a fairly good description of the measured spectra. As a further test, we also calculated the electronic structure and MO spectra of UNiGa using the LSDA + U approach [29], which contains an additional Hubbard-like U to simulate strong on-site Coulomb correlations between the $5f$ electrons. The LSDA + U approach gave, however, much worse results for the MO spectra. Therefore it can be concluded that application of the LSDA approach to UNiGa is justified.

To investigate the mechanism of the giant MR, we analyzed the band-by-band contributions of each of the six Fermi surface (FS) sheets to the MR, which are listed in Table I, as well as are the normalized AFM conductivities for each sheet. All FS sheets, except No. 5, contribute substantially to the giant negative MR. In Figs. 1 and 2 we show four characteristic FS sheets (Nos. 2–5) for the AFM and FM phase, respectively. From a comparison of the FS sheets in both phases, it can be seen that *a large FS reconstruction occurs at the field induced metamagnetic transition.* In the case of FS sheets Nos. 3 and 4 even a reconstruction which changes the FS topology occurs: the FS sheets, which are initially connected along the k_z direction in the AFM configuration, become disconnected upon the metamagnetic transition. Such a reconstruction of the FS may naturally bring about pronounced changes in the crystalline anisotropy of the conductivity. Next, we consider the mechanism of the giant MR in detail from the example of FS sheet No. 3 (2nd FS in Figs. 1 and 2). For the CPP-MR, one basic mechanism that can be identified from this FS sheet is the following: the rather spherically curved AFM sheet is reshaped into

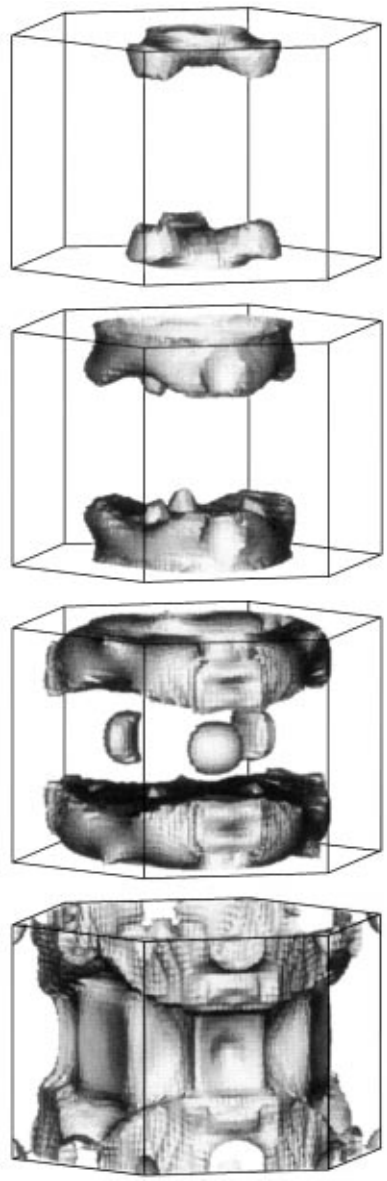

FIG. 2. As Fig. 1, but for FM UNiGa. Note the rearrangement of the FS sheets with respect to those shown in Fig. 1.

a flat, more planar FS sheet. Since the Fermi momentum $\partial E_{n\mathbf{k}}/\partial \mathbf{k}$ is normal to the surface, its z component contributes therefore particularly on the flat surface. The same mechanism operates for the not shown FS sheet No. 1, and it furthermore contributes to the MR stemming from sheets Nos. 2 and 4. A second mechanism which we identify here to contribute to the MR is a reduction of the total FS area in the transition from the FM to the AFM state: the computed area of the FS is in the AFM state 26% smaller than of the FM state. If we assumed $\partial E_{n\mathbf{k}}/\partial \mathbf{k}$ to be constant, this reduction of the FS area alone would lead to an *isotropic* MR of -26%. Such a reduction of the total FS is equivalent to the opening of a superzone gap in part of the BZ. In the situation where the metamagnetic transition would be accompanied with a metal-insulator transition, as happens, e.g., in UNiSn [30], a band gap in the whole BZ occurs, which causes a colossal MR. With regard to our result, we mention that recently the linear specific heat coefficient γ was measured for the ground state AFM phase and the FM phase of UNiGa [31]. A 10% smaller γ was obtained for the AFM phase, which was interpreted as the first direct evidence for a partial superzone gap [31]. Our calculations are consistent with this experimental observation.

The CPP-MR effect of FS sheet No. 5 is opposite to that of the other sheets (see Table I). We found that this band is extremely sensitive to the relative positions of the other bands and of E_F, because band 5 just touches at E_F in part of the BZ and is quasiparallel to E_F in other parts of the BZ. Therefore, a small shift of the Fermi level can drastically change the shape of FS No. 5 [32].

To summarize, we have presented the first *ab initio* calculations of the giant MR in UNiGa. The theoretical MR, which, as we emphasize, is calculated without adjustable parameters, is in semiquantitative agreement with experiment. Two basic mechanisms that may cause a giant MR have been recognized from our calculations. One of them is the occurrence of a superzone gap, which reduces the total FS area. This leads essentially to an isotropic MR. The second mechanism is the deformation of the FS sheets, which can lead to an extremely anisotropic MR. The decisive quantity for an anisotropic MR is the "flatness" of the FS in a principal direction, or in other words, the amount that a component $(\partial E_{n\mathbf{k}}/\partial k)_i$ contributes on the FS area. A theoretical recipe for "designing" a giant, anisotropic MR can be deduced from these observations: if an AFM FS consisting of a cylindrical surface along k_z is transformed into one or two parallel, flat surfaces perpendicular to the k_z axis, then σ_{zz} would vanish for the cylinder, but it would be very large for the planar surface. The resulting theoretical CPP-MR would be up to -100%. These findings lend to the general understanding of the MR phenomenon.

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*Permanent address: Institute of Metal Physics, Academy of Sciences of Ukraine, 252680 Kiev, Ukraine.

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