Giant magnetoresistance in uranium intermetallics: *Ab initio* calculations for U_2Pd_2In and U_2Pd_2Sn

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Among the magnetic uranium intermetallic compounds, a considerable number is exhibiting field induced magnetic phase transitions. Such transitions are frequently accompanied by giant magnetoresistance (GMR) effects. On the basis of density functional calculations we predict GMR to be present in the recently discovered ternary compounds U_2Pd_2In and U_2Pd_2Sn. In particular, it is shown by this exemplary calculation that GMR in $5f$ intermetallics can be produced by topological changes of the Fermi surface even if the size of the Brillouin zone is invariant under the metamagnetic transition. The possibility of an inverse GMR effect is demonstrated without assuming any spin dependence in the scattering. No qualitative changes of the results are found if a moderately spin dependent scattering is introduced. [S0163-1829(96)09342-3]

Much experimental and theoretical work was devoted to the magnetoresistance of transition metal multilayer systems in recent years. This booming interest was initiated by the discovery of a drastic change of the resistivity with increasing magnetic field in Fe/Cr multilayers,[1] the so-called giant magnetoresistance (GMR) effect. The effect occurs if the exchange coupling through the spacer (Cr) between subsequent, internally ferromagnetic layers (Fe) is antiparallel in zero external field. Application of an external magnetic field brings all moments into alignment, thus reducing the resistivity by up to a factor of 3.[2] Different theoretical explanations for the GMR in terms of spin-dependent scattering and magnetic state dependence of the electronic structure have been given. Recently, it was shown[3,4] that a semiquantitative understanding of the effect in thin multilayers is possible from the coherent electronic structure of the idealized system: Changes of the electronic structure as a function of the magnetic order, in particular doubling the Brillouin zone size, cause pronounced changes of Fermi surface and Fermi velocity. The effect can be enhanced, reduced, or even inverted by spin-dependent impurity scattering.[4] It seems natural that this coherent mechanism of the GMR is not restricted to artificially structured materials showing a metamagnetic transition. For instance, similar behavior was observed[5] and described theoretically[6,7] for the transition metal compound FeRh.

Another class of materials exhibiting large resistivity changes at field induced magnetic transitions is that of uranium intermetallics.[8] A resistivity drop by a factor of 7 was observed, e.g., at the antiferromagnetic (afm) $\rightarrow$ ferromagnetic (fm) transition in hexagonal UNiGa.[9] Possible mechanisms for the GMR in such compounds have been discussed in a recent publication.[10] There, arguments are given against the significance of the following: stacking faults in the magnetic structure (excluded by neutron diffraction); different size of the Brillouin zone for the fm and the afm state leading to a major reconstruction of the Fermi surface at the transition (less probable, since no change of the Sommerfeld parameter was observed at that time); the Kondo effect (no instability of the $5f$ moment is present). Spin-dependent scattering is favored as the remaining possibility to produce the effect.[10] A serious objection against this hypothesis is discussed in Ref. 10 as well: In a situation with strong spin-orbit interaction present spin-flip processes should be important and lead to a considerable reduction of the spin dependence in the scattering. Moreover, new measurements[11] show that there is indeed a 10% difference in the low temperature specific heat between the afm and the fm state of UNiGa, pointing to possible Fermi surface reconstructions.

The question to be solved is which mechanism could be responsible for the experimentally observed GMR in uranium intermetallics if none of the known ideas is convincing. From the discussion above it is evident that stacking faults in the magnetic structure and the Kondo effect can be excluded on the basis of the experimental information. The two other mechanisms discussed, reconstruction of the Fermi surface and spin-dependent scattering, will be separately investigated in the following. To try a clarification, we have chosen two representatives of the recently discovered system U_2T_2X.[12] The intermetallics U_2Pd_2In and U_2Pd_2Sn both

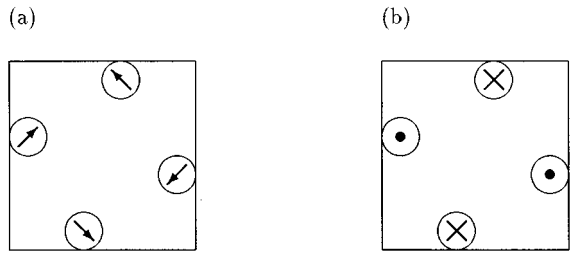

FIG. 1. (a) Noncollinear antiferromagnetic ground state of U_2Pd_2In and U_2Pd_2Sn (U atoms in the tetragonal basal plane); (b) assumed collinear arrangement.

exhibit the same kind of noncollinear afm structure [Fig. 1(a)] below $T_N=36$ K and $T_N=41$ K, respectively, with the magnetic unit cell equivalent to the crystallographic cell.[13] That means no change of the size or shape of the Brillouin zone will occur at an assumed transition to the fm state. In experiment, metamagnetic transitions to still unknown (probably not fm) states have been observed in high fields, $B_{\mathrm{meta}}=25$ T for U_2Pd_2In and $B_{\mathrm{meta}}=39$ T, 45 T for U_2Pd_2Sn.[14] Here, in accordance with the exemplary character of this calculation, we approximate the more complicated noncollinear afm ground state of U_2Pd_2X by an in-plane afm arrangement with collinear moment orientation along the tetragonal c axis [Fig. 1(b)]. The field induced state is taken, in the same spirit, as ferromagnetic along c.

We now solve the quasiclassical Boltzmann equation in relaxation time approximation in a periodic system with low concentration of scattering centers, yielding the conductivity tensor

$$\sigma=\frac{e^2}{V}\sum_k \tau_k\delta(\varepsilon_k-\varepsilon_F)\mathbf{v}_k\mathbf{v}_k. \tag{1}$$

Here, k is the Bloch state index $(\mathbf{k}n)$, τ_k are quasiparticle lifetimes, and ε_k and $\mathbf{v}_k$ are related quasiparticle energies and Fermi velocities, respectively. The latter two quantities are taken from the Kohn-Sham band structure of relativistic local spin density calculations[15] where the respective afm and fm state is obtained through a suitable choice of the initial conditions (both states are local minima of the Hohnberg-Kohn energy functional). Quasiparticle lifetimes for the case of single impurity scattering in elemental ferromagnetic metals have been calculated recently.[16] It is not possible at the present state of the art, however, to obtain this information for the complicated systems considered here. Hence we will treat the τ_k as model parameters and introduce averaged lifetimes along the same lines as in the case of multilayer systems.[4] In this way, we do not restrict our consideration to any specific scattering mechanism, but still assume that k is a good quantum number. Moreover, this treatment has the advantage to allow an approximative separation of band structure and scattering properties. Averaging the lifetime over the Fermi surface gives

$$\langle\tau\rangle=\frac{\Sigma_k\tau_k\delta(\varepsilon_k-\varepsilon_F)}{\Sigma_k\delta(\varepsilon_k-\varepsilon_F)}, \tag{2}$$

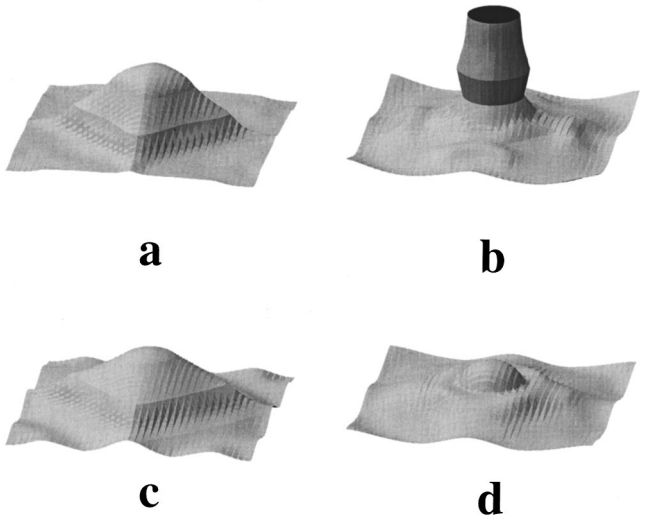

FIG. 2. Characteristic Fermi surface sheet of afm (a) and fm (b) U_2Pd_2Sn. The same band is presented for afm (c) and fm (d) U_2Pd_2In. The flat regions of the Fermi surfaces are oriented in the k_x-k_y plane. To avoid redundancy, only the upper half of the Brillouin zone is drawn.

where spin-independent scattering was imposed but $\langle\tau\rangle$ still depends on the magnetic state. To retain only pure band structure quantities, additionally independence of $\langle\tau\rangle$ on the magnetic state is assumed,

$$\langle\tau\rangle^{\mathrm{fm}}=\langle\tau\rangle^{\mathrm{afm}}, \tag{3}$$

arriving at the following simple relation for the GMR:[4,17]

$$\mathrm{GMR}_i=\frac{\sigma_{ii}^{\mathrm{fm}}-\sigma_{ii}^{\mathrm{afm}}}{\sigma_{ii}^{\mathrm{afm}}}=\frac{\Sigma_k\delta(\varepsilon_k^{\mathrm{fm}}-\varepsilon_F)v_{ki}^{\mathrm{fm}}v_{ki}^{\mathrm{fm}}}{\Sigma_k\delta(\varepsilon_k^{\mathrm{afm}}-\varepsilon_F)v_{ki}^{\mathrm{afm}}v_{ki}^{\mathrm{afm}}}-1. \tag{4}$$

Here, $\sigma_{xx}=\sigma_{yy}$ and σ_{zz} mean the conductivity in the tetragonal plane and perpendicular to the plane, respectively. The corresponding notation in multilayer systems would be current-in-plane (CIP) and current-perpendicular-to-plane (CPP).

Evaluating Eq. (4), we find that a pronounced GMR effect can be produced by topological changes of particular Fermi surface sheets (Fig. 2) which are possible at a metamagnetic transition even if the size and shape of the Brillouin zone are preserved. In U_2Pd_2Sn, a GMR_x of 95% is found in the plane, whereas a much smaller effect of 3% is present in perpendicular direction. This difference can easily be understood from the Fermi surface topology. σ_{xx} is determined by the in-plane components of the Fermi velocity which mainly stem from cylindrical (in k_z-direction) parts of the Fermi surface. In U_2Pd_2Sn, such cylindrical parts appear if the afm configuration is changed to fm [Figs. 2(a) and 2(b)]. Consequently, a large GMR_x is obtained. On the other hand, σ_{zz} mainly originates from flat [in the (k_x-k_y)-plane] Fermi surface parts with large v_{kz}. The flat part area is not much changed under the metamagnetic transition, and a small value of GMR_z is found. In U_2Pd_2In, the shape of the Fermi surface is only marginally different for the two magnetic configurations investigated [Figs. 2(c) and 2(d)]. Hence the GMR effect is small in both directions. The related values for U_2Pd_2In are $\mathrm{GMR}_x=12\%$ and, interestingly, a small inverse effect of $\mathrm{GMR}_z=-5\%$. All these numbers should

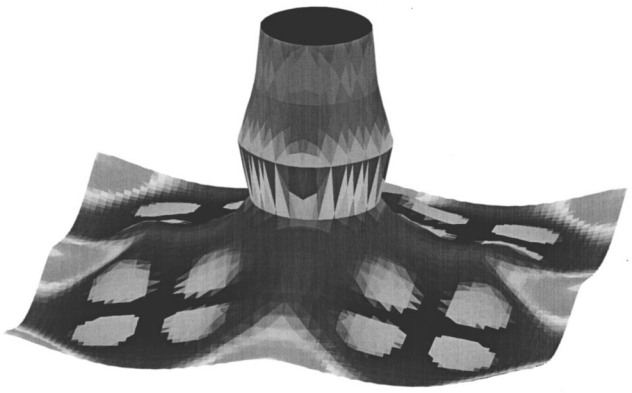

FIG. 3. Same as Fig. 2(b) with spin character of the states resolved. The darkest areas have 10% majority spin character, the lightest areas 90%.

be taken as examples for the possible size of the effect rather than accurate predictions. Unfortunately, experimental magnetoresistance data are available only for the related U_2Ni_2Sn compound[18] in fields much below the critical value. For the case of UNiGa, where the size of the magnetic cell changes under the transition, agreement of theoretical data[19] according to Eq. (4) with experiment[9] was found.

To estimate an upper bound on the possible influence of spin-dependent scattering we use the following procedure. Since the spin is not a good quantum number in the considered systems due to strong spin-orbit interaction we subdivide all states k into a class k^+ with positive spin expectation value, $\langle k^+|\hat{\Sigma}_z|k^+\rangle>0$, and a class k^- with negative spin expectation value, $\langle k^-|\hat{\Sigma}_z|k^-\rangle<0$. $\hat{\Sigma}_z$ is the z component of the spin operator in standard Dirac notation, and in the absence of spin-orbit coupling pure spin states would exist with $\langle k^\uparrow|\hat{\Sigma}_z|k^\uparrow\rangle=1$, $\langle k^\downarrow|\hat{\Sigma}_z|k^\downarrow\rangle=-1$. According ''class dependent'' relaxation times are defined via

$$\langle\tau\rangle^{\pm}=\frac{\Sigma_{k^\pm}\tau_{k^\pm}\delta(\varepsilon_{k^\pm}-\varepsilon_F)}{\Sigma_{k^\pm}\delta(\varepsilon_{k^\pm}-\varepsilon_F)}, \tag{5}$$

resembling the spin-dependent relaxation times $\langle\tau\rangle^\sigma$ ($\sigma=\uparrow,\downarrow$) introduced in the nonrelativistic theory.[4] While $\langle\tau\rangle^\uparrow$ and $\langle\tau\rangle^\downarrow$ may differ by up to one order of magnitude,[16] much smaller differences can be expected for $\langle\tau\rangle^+$ and $\langle\tau\rangle^-$ in the case of actinide compounds, since k^+ and k^- are distributed over the same Fermi surface sheet (Fig. 3). The highest degree of spin polarization at the Fermi surface is $|\langle k|\hat{\Sigma}_z|k\rangle|=0.8$ (i.e., 90% majority and 10% minority spin character or vice versa). The amount of states of nearly pure spin character, $|\langle k|\hat{\Sigma}_z|k\rangle|>0.7$, is less than 5% of the total number of states at the Fermi surface. However, a polarization of $|\langle k|\hat{\Sigma}_z|k\rangle|<0.3$, that means an almost balanced mixture of majority and minority spin character, is found for half of these states. This strong spin mixing is another argument for a small difference between $\langle\tau\rangle^+$ and $\langle\tau\rangle^-$.

TABLE I. Calculated GMR of U_2Pd_2Sn and U_2Pd_2In for different values of the spin anisotropy β of the scattering.

	U_2Pd_2Sn		U_2Pd_2In	
β	GMR_x	GMR_z	GMR_x	GMR_z
0.5	100%	8%	11%	−3%
0.7			8%	−7%
0.8	91%	2%		
1	95%	3%	12%	−5%
1.5	110%	11%	24%	4%
2	140%	23%	41%	17%

A straightforward assumption for the relaxation time in the afm state is

$$\frac{1}{\langle\tau\rangle^{\text{afm}}}=\frac{1}{2}\left(\frac{1}{\langle\tau\rangle^+}+\frac{1}{\langle\tau\rangle^-}\right), \tag{6}$$

yielding

$$\text{GMR}_i=\frac{\Sigma_\pm\langle\tau\rangle^\pm\Sigma_{k^\pm}\delta(\varepsilon^{\text{fm}}_{k^\pm}-\varepsilon_F)v^{\text{fm}}_{k^\pm i}v^{\text{fm}}_{k^\pm i}}{\langle\tau\rangle^{\text{afm}}\Sigma_k\delta(\varepsilon^{\text{afm}}_k-\varepsilon_F)v^{\text{afm}}_{ki}v^{\text{afm}}_{ki}}-1. \tag{7}$$

Results of related calculations for given ratios $\beta=\langle\tau\rangle^+/\langle\tau\rangle^-$ are compiled in Table I. Obviously, no qualitative changes of the size of GMR are present if β is varied in reasonable bounds. Note, that the β values 0.7 and 0.8 given in Table I correspond to the minima of the function $\text{GMR}_i(\beta)$ for the respective compound.

In conclusion, we have demonstrated that a large magnetoresistance effect in $5f$ intermetallics can be produced by topological changes of the Fermi surface at a metamagnetic (afm → fm) transition with invariant size of the Brillouin zone. This mechanism may be particularly important if other sources of GMR, like spin-dependent scattering or changes of the magnetic unit cell, are not effective. On the basis of *ab initio* band structure calculations, we predict a GMR to be present in the ternary compounds U_2Pd_2In and U_2Pd_2Sn. Moreover, an inverse GMR effect has been shown to occur as a pure band structure effect without assuming any spin dependence in the scattering. If spin-dependent scattering is introduced, no qualitative changes of the results are obtained for reasonable values of the asymmetry parameter β. Calculations taking into account the correct magnetic ground state are in progress.

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