

**Electrical resistivity of 5f-electron systems affected by static and dynamic spin disorder**L. Havela,<sup>1,\*</sup> M. Paukov,<sup>1</sup> V. Buturlim,<sup>1</sup> I. Tkach,<sup>1</sup> D. Drozdenko,<sup>1</sup> M. Cieslar,<sup>1</sup> S. Mašková,<sup>1</sup> M. Dopita,<sup>1</sup> and Z. Matěj<sup>2</sup><sup>1</sup>Charles University, Faculty of Mathematics and Physics, Ke Karlovu 3, Prague 2, Czech Republic<sup>2</sup>MAX IV Laboratory, Lund University, SE-221 00 Lund, Sweden

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Metallic 5f materials have very strong coupling of magnetic moments and electrons mediating electrical conduction. It is caused by strong spin-orbit interaction, coming with high atomic number  $Z$ , together with involvement of the 5f states in metallic bonding. We have used the recently discovered class of uranium (ultra)nanocrystalline hydrides, which are ferromagnets with high ordering temperature, to disentangle the origin of negative temperature coefficient of electrical resistivity. In general, the phenomenon of electrical resistivity decreasing with increasing temperature in metals can have several reasons. The magnetoresistivity study of these hydrides reveals that quantum effects related to spin-disorder scattering can explain the resistivity behavior of a broad class of actinide compounds.

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Metallic systems based on light actinides (Th-Pu) belong typically to 5f narrow-band materials [1]. Magnetic ordering of 5f moments can appear if the 5f atoms are not very close together and the hybridization with non- $f$  states is weak or moderate. The magnetic order is then of the band (itinerant) type, although it can have, similar to 3d metals, a local-moment character. In short, it means that transversal fluctuations (fluctuating direction) of moments are more important than longitudinal ones (fluctuating size), at least in the ordered state and in a limited temperature range above it. A special ingredient is the very strong spin-orbit interaction, which leads to sizable orbital moments even in a band case [2]. The involvement of  $f$  states in bonding and the hybridization with non- $f$  states has, in such a situation, a dramatic mutual influence of the magnetic subsystem and electrons responsible for metallic conduction [3]. It also leads to giant magnetic anisotropy related to the  $f$ - $f$  bonding directions [4].

Out of all the light actinides, most of the experimental data exist for uranium-based systems. Pure U-metal as well as U-rich compounds have the temperature dependences of electrical resistivity,  $\rho(T)$ , rather regular, which means increasing with increasing  $T$ , some with a weak tendency to saturation at high temperatures [5]. The same holds for compounds with lower U concentration but compact crystal structures, implying short U-U distances and broad 5f band, as, e.g., Laves phases [6]. More anomalous electrical resistivities have been observed in narrow-band compounds. Resistivities of such systems often substantially exceed  $\approx 200 \mu\Omega \text{ cm}$ , assumed as an upper limit for conventional metallic systems (known also as the Mott-Ioffe-Regel limit) [7], although the  $T$  dependence remains metallic. In such cases the temperature coefficient of resistivity,  $\text{TCR} = d\rho/dT$ , is frequently found to be negative at high temperatures. There are numerous such binary and ternary compounds, such as  $\text{USb}_2$  [8],  $\text{U}_3\text{Au}_3\text{Sn}_4$  [9],  $\text{UCu}_2\text{Sn}$ ,  $\text{UAu}_2\text{Al}$ ,  $\text{UPt}_2\text{Sn}$  [10], or  $\text{URuGa}$  [11]. In some cases the reason for the negative TCR could be seen in the context of a superzone boundary effect in antiferromagnets, at which an additional magnetic periodicity leads to a gapping of the

Fermi surface, or due to the Kondo effect (conduction electrons condensing around local moments due to spin-spin interaction, suppressing magnetic order, and increasing resistivity). There are, however, cases where the two mechanisms are hardly applicable. For example,  $\text{UGa}_2$  has a ferromagnetic ground state, so there are no superzones, and a large magnetic moment of  $3.0\mu_B/\text{U}$  leaves no space for spin compensation due to the Kondo effect [12]. Nevertheless, as the Kondo effect remains almost a synonym for negative TCR, there has been persisting discussion about its applicability even in such a case. One of the reasons is that the Kondo prediction of  $\rho$  proportional to  $-\ln T$ , describing the negative slope, is rather nonspecific, and such a term can be used successfully to fit over a certain  $T$  range any decreasing function with a saturating tendency.

There is, however, another mechanism yielding  $\text{TCR} < 0$ , which is related to a strong electron scattering due to disorder. Mooij [13] pointed out that if the residual resistivity value of disordered alloys exceeds a certain limit ( $\approx 100 \mu\Omega \text{ cm}$ ), the overall temperature dependence flattens due to progressing nonadditivity of impurity and electron-phonon scattering, and the ultimate flat dependence starts to exhibit negative TCR for  $\approx 150 \mu\Omega \text{ cm}$ . Nowadays it can be understood as being due to weak localization, which increases resistivity as a consequence of quantum interference effects in conditions when the electron wavelength is comparable with the distance of two scattering sites. In particular, the resistivity is enhanced by a higher probability of scattering of electrons backwards compared to forward scattering. Elevated temperature destroys the quantum coherence, which leads to a resistivity reduction [14]. Such variations of resistivity in the conditions of increasing disorder can be parametrized considering that low-energy phonons are ineffective scatterers if their wavelength is longer than the electron mean free path. The effect of high-energy lattice vibrations, populated at high temperatures, can be approximated by the Debye-Waller factor, which is normally used to quantify the reducing effect of lattice vibrations on coherent scattering in diffraction experiments. This model developed by Cote and Meisel [15] gives the low-temperature behavior of resistivity approximated as  $\rho \sim 1 - aT^2$ .

A more general view, discussing different regimes depending on ratio of the inelastic mean free path  $l_{\text{ph}}$  (depending on phonons) and a characteristic localization length  $\xi$ , was

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given in Ref. [16]. The interplay of electron-impurity and electron-phonon scattering was already a basis of *ab initio* calculations trying to describe reasons for  $\text{TCR} < 0$  in elemental actinides [17], but the subject remained highly disputable.

We suggest that a quantum interference mechanism such as weak localization can be operational in narrow-band U intermetallics. Their high resistivity values are not due to strong impurity scattering, but they are introduced by strong scattering on  $5f$  spins, which are disordered in the paramagnetic state, and the spin-dependent part of the atomic potential is random. For example, for  $\text{UGa}_2$  it was indicated how the application of pressure or external field, which tends to suppress the paramagnetic transversal fluctuations, contributes to remove  $\text{TCR} < 0$  [12]. One has to adopt certain assumptions for the validity of this model. The paramagnetic state could be essentially approximated by a static model, and the only inelastic processes removing the coherence are phonons. But even if we consider the magnetic inelastic scattering, its energy should scale as  $k_B T$ , and it will work at high temperatures in parallel with phonons in suppression of electron coherence.

In the present work we use properties of nanocrystalline  $\text{UH}_3$ -based ferromagnets, which provide randomness on atomic scale as in glassy materials, but magnetic moments and ordering temperature remain untouched by the disorder. The materials studied belong to the alloyed uranium hydrides, which can be synthesized by room-temperature exposure of *bcc* U alloys to high  $\text{H}_2$  pressures. Such alloys are supersaturated solid solutions of a *d*-metal such as Mo, Zr, or Ti. They retain the *bcc* structure, which is for pure uranium metal stable only above 1049 K. The H uptake leads to different resulting structures. For example, the alloys with Zr yield the  $\alpha$ - $\text{UH}_3$  type, which was known only as a transient species and its properties could not be reliably established in the past [18]. For us, now, another structure modification is more interesting. The *bcc* U-Mo alloys yield hydrides, which were assumed as amorphous [19], but detailed structure study, which includes a pair distribution function analysis, indicates predominant crystalline  $\beta$ - $\text{UH}_3$  structure type. It is cubic with the lattice parameter  $a = 0.664$  nm, but the average grain size is merely 2-3 nm [20]. Hence the coherence length is only  $\approx 4a$ . We can denote such structure as ultrananocrystalline. The reason for such structure should be in a fast nucleation accompanied by a slow grain growth, likely impeded by the Mo atoms, entering one of the two U positions. Mo with low affinity to hydrogen appears to be a very efficient inhibitor of the grain growth, and such structure (revealed by TEM in Fig. 1) can accept many other alloying elements. Such situation has one important consequence. The slow grain growth which does not progress into larger grain size helps to avoid formation of a macroscopic interface between the hydrogenated and nonhydrogenated material. As the volume expansion across such interface normally leads to a sample decrepitation, the nanocrystalline hydrides tend to remain monolithic, although much more brittle than the parent alloy. Irrespective of structure and composition, all the hydrides are ferromagnets with ordering temperatures in the range 160–205 K, which is remarkable, especially for the nanocrystalline materials. It emphasizes that the U-H interaction may be more important than the U-U coordination. Previous experience with  $5f$  systems was

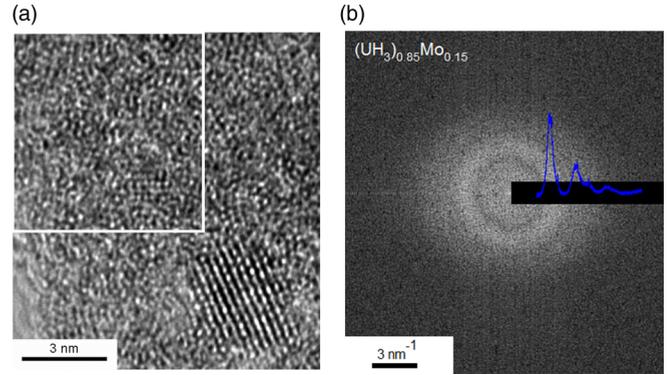


FIG. 1. High-resolution TEM micrograph (a) reveals the highly disordered structure of  $(\text{UH}_3)_{0.85}\text{Mo}_{0.15}$  contrasting with the crystalline precipitate of spurious uranium carbide UC in the right lower part. The Fourier transformed image within the white square shown in (b) correlates with the fuzzy x-ray diffraction lines of  $(\text{UH}_3)_{0.85}\text{Mo}_{0.15}$ , which are the diffraction lines of  $\beta$ - $\text{UH}_3$  structure type broadened due to extremely small size of coherently diffracting domains—crystallites. The small amount of spurious UC, seen by the weak sharper peaks, does not interfere with magnetization or magnetoresistance data as this material does not undergo magnetic ordering. The structure of  $(\text{UH}_3)_{0.78}\text{Mo}_{0.12}\text{Ti}_{0.10}$  is practically equivalent.

typically indicating a suppression of magnetism with a loss of long-range periodicity [21].

Considering the generally strong magnetic anisotropy in light actinides, randomly oriented nanograins mean a random distribution of easy magnetization directions and slow approach to saturation of magnetization in external magnetic field. The anisotropy is comparable with the exchange interaction. An estimate of the anisotropy energy per one U atom can be taken from the magnon gap width  $\Delta = 36$  K, obtained from the low- $T$  specific heat [19]. It means that the directions of individual magnetizations depend on exchange interaction across the grain boundary and on local easy magnetization directions, the latter being more important at very low temperatures. The total U magnetic moments of  $\approx 1.0 \mu_B$  represent, as usual for less-than-half filling of the  $5f$  shell, a difference between larger spin and orbital moments, being mutually antiparallel.

The electrical resistivity of U hydrides generally reaches very high values. The study of  $\text{UH}_3$  in a compact form, prepared by high-temperature synthesis at very high pressures, revealed very high resistivity ( $\approx 600 \mu\Omega \text{ cm}$ ) in the paramagnetic state, and strongly decreasing tendency below  $\approx 165$  K, which is the Curie temperature [22]. This means that the spin-disorder resistivity has a relatively high contribution in the paramagnetic state. However, such high values cannot be a consequence of strong scattering only. As expressed by the Mott-Ioffe-Regel limit, the electron mean free path cannot be shorter than the interatomic spacing. Therefore we can speculate about a low concentration of electrons with low effective mass, which are mainly responsible for the charge transport. Indeed a substantial depopulation of U  $6d$  and  $7s$  states due to the U-H interaction, indicated by *ab initio* calculations [18], suggests that mostly  $5f$  states, which have a higher effective mass (the Sommerfeld coefficient  $\gamma$  of electronic specific heat is enhanced to  $\approx 30 \text{ mJ/mol K}^2$ ) remain

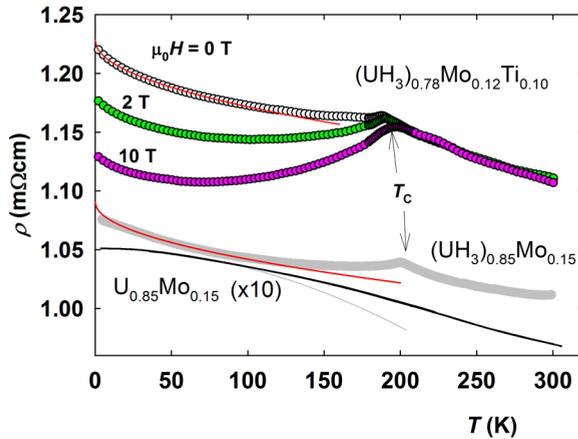


FIG. 2. Temperature dependence of electrical resistivity for  $(\text{UH}_3)_{0.78}\text{Mo}_{0.12}\text{Ti}_{0.10}$  measured in various magnetic fields. In the ferromagnetic state the data are strongly dependent on magnetic history. Here we compare data obtained when heating up after cooling in the given field, which was applied at  $T = 300$  K. The sequence was  $\mu_0 H = 10$  T, 0 T, and 2 T. The data are compared with similar data on  $(\text{UH}_3)_{0.85}\text{Mo}_{0.15}$  in zero field and with its precursor  $\text{U}_{0.85}\text{Mo}_{0.15}$  [19], which has  $\rho \approx 100 \mu\Omega \text{ cm}$  and was multiplied by a factor of 10 for easier comparison of the  $T$  dependence. The thin gray line indicates the  $\rho \sim -aT^2$  dependence, the red lines the  $\rho \sim -aT^{1/2}$  dependence, both mentioned in the text.

at the Fermi level, while the states of “light” electrons ( $6d$  and  $7s$ ), which are more efficient in the charge transport, are largely depopulated. The precursor  $bcc$  U alloys remain well metallic [23]—their resistivity reaches typically  $100 \mu\Omega \text{ cm}$ —but a strong disorder due to the randomness in the distribution of the alloying elements leads to a flat  $T$  dependence, which exhibits a weak negative TCR, as indicated in Fig. 2 for  $\text{U}_{0.85}\text{Mo}_{0.15}$ . It saturates towards the low- $T$  limit and can be reasonably well described by  $-aT^2$  up to about 100 K.

Their hydrides apparently combine both features, the resistivity enhanced by an order of magnitude to the vicinity of  $1 \text{ m}\Omega \text{ cm}$ , and flat  $T$  dependence with a negative TCR. Despite their ultranano-crystalline structure, they are more ordered from the point of view of atomic mixing, as one type of site is occupied purely by U atoms. This is likely the feature which leaves some space, besides the atomic disorder, for manifestation of magnetic disorder. The clear impact of magnetism can be seen in Fig. 2, showing a cusp, which can be clearly associated with  $T_C$ , detected by magnetization measurements.  $(\text{UH}_3)_{0.85}\text{Mo}_{0.15}$  has  $T_C = 200 \text{ K}$  [19]. In the present study we use  $(\text{UH}_3)_{0.78}\text{Mo}_{0.12}\text{Ti}_{0.10}$ , which has better mechanical properties, but structure and magnetic properties are almost the same. Its  $T_C = 188 \text{ K}$  is marginally lower. The temperature dependence of resistivity was measured by the  $ac$  four-probe technique also in magnetic field, applied perpendicular to the current direction (transversal magnetoresistance). The formation of the cusp can be understood as due to the interplay of conventional negative TCR and the ferromagnetic ordering at  $T_C$ , which starts to reduce the random spin fluctuations in the paramagnetic state. Close below  $T_C$ , the correlation length between individual magnetic moments is very short but increasing with decreasing  $T$ , as the exchange interactions are not restricted by the size of

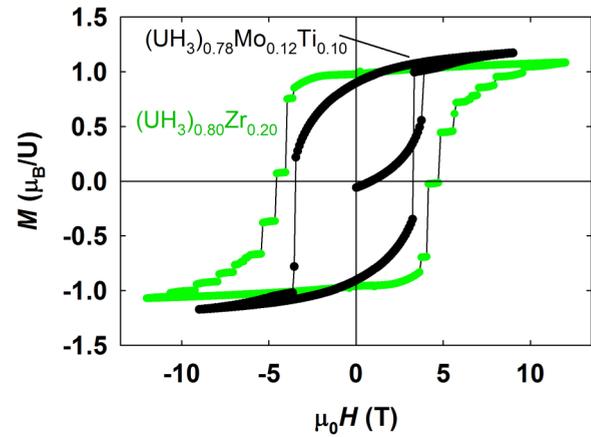


FIG. 3. Comparison of magnetic hysteresis loops at  $T = 2$  K for the ultranano-crystalline  $(\text{UH}_3)_{0.78}\text{Mo}_{0.12}\text{Ti}_{0.10}$  with that of  $(\text{UH}_3)_{0.80}\text{Zr}_{0.20}$  with the average grain size  $\approx 10 \text{ nm}$ .

the nanocrystals. Proceeding to lower temperatures, when the magnetic excitations progressively vanish, brings increasing magnetic anisotropy, and magnetizations in individual neighbor nanocrystals become gradually misoriented. This tendency continues down to the lowest temperatures, which can explain the lack of saturation observed for the precursor alloys. The magnetization of a ferromagnet itself has to saturate in the low- $T$  limit. Alternatively such behavior can be understood by the model of Imry [16], yielding the  $\rho \sim -aT^{1/2}$  scaling at low temperatures and  $-bT^{-1/2}$  or  $-bT^{-1/3}$  at high temperatures. The first type of dependence can describe, at least formally, the experimental data on the hydrides reasonably well, as shown by the fits in Fig. 2 for zero-field  $\rho(T)$ .

The picture of resistivity affected by magnetization in the ordered state is corroborated by  $\rho(T)$  measurements in applied magnetic fields. While in the paramagnetic state the fields of a few teslas have negligible impact on the spin dynamics, for  $T \leq T_C$  the effect is quite significant. The applied field tends to suppress both magnetic excitations in the ferromagnetic state and the misorientation of magnetization in individual nanograins, each grain representing several hundred atoms of  $\approx 1 \mu_B$ . However, the anisotropy energy at low temperatures is higher than the Zeeman energy of nanocrystals in external field (both scale with the number of magnetic atoms in the grain), and the misorientation, which increases resistivity, wins at very low temperatures. Such interplay gives rise to resistivity minima, which are shifted towards lower temperatures with increasing field.

The relation of magnetic state and electrical resistivity is even better illustrated when comparing magnetoresistance at particular  $T$  with the respective hysteresis loop. The loops of the U-alloy-based hydrides are very wide, especially in the low-temperature limit. The reason dwells in small monodomain grains and strong anisotropy increasing the field required for remagnetization. Figure 3 shows a comparison of the hysteresis loop of  $(\text{UH}_3)_{0.78}\text{Mo}_{0.12}\text{Ti}_{0.10}$ , with that of  $(\text{UH}_3)_{0.80}\text{Zr}_{0.20}$ . The latter material is also nano-crystalline, but on average has its grain size almost one order of magnitude larger ( $\approx 10 \text{ nm}$ ), and the crystal structure is of the  $\alpha$ - $\text{UH}_3$  type. Its hysteresis loop is rectangular, with very small field effect

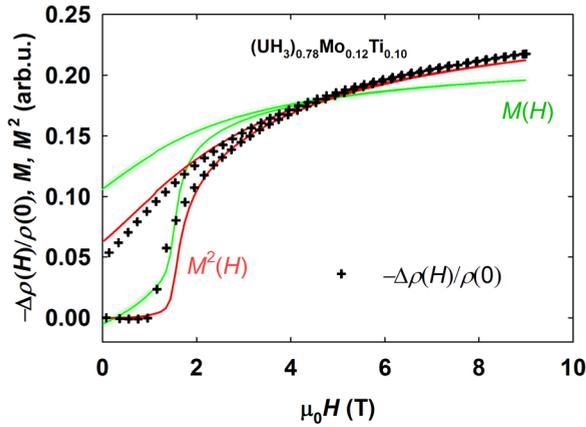


FIG. 4. Comparison of magnetoresistance and magnetization. The relative magnetoresistance as a function of magnetic field (crosses) at  $T = 50$  K started after cooling in zero magnetic field compared with the respective  $M(H)$  (green line) and  $M^2(H)$  (red line) data, which were rescaled arbitrarily to match the magnetoresistance. It illustrates that the magnetoresistance scales approximately as  $M^2$ .

on magnetization in high fields. On the other hand, the slow approach of the ultrananocrystalline  $(\text{UH}_3)_{0.78}\text{Mo}_{0.12}\text{Ti}_{0.10}$  to saturation can be interpreted as field variations of noncollinear ferromagnetism.

The electrical resistivity of  $(\text{UH}_3)_{0.80}\text{Zr}_{0.20}$  indeed behaves as in a regular ferromagnet, with decreasing  $\rho$  below  $T_C$  due to a suppression of the spin disorder, and only a small influence of magnetic field [18]. The magnetization processes responsible for the shape of the hysteresis loop are mainly the remagnetization of larger magnetic domains, and the distance between the domain boundaries is larger than the electron mean free path. On the other hand, the static spin incoherence on the atomic scale in  $(\text{UH}_3)_{0.78}\text{Mo}_{0.12}\text{Ti}_{0.10}$  makes the impact of field-induced moment alignment on the resistivity much more dramatic. We could assume from Fig. 3 that the resistivity will be decreasing as a function of magnetic field, and the decrease should be most pronounced in an intermediate temperature range, below  $T_C$  but not close to the 0 K limit, as the manipulation of moments at very low temperatures is impeded by increasing anisotropy, leading to a rigidity of the spin system.

Figure 4 displays the field dependence of resistivity. The width of the resistivity hysteresis after cooling down to 50 K corresponds to the magnetization hysteresis. The resistivity change is negative indeed (Fig. 4 shows the  $\rho$  value inverted for easier comparison with magnetization), reducing the initial

value by 22% in  $\mu_0 H = 9$  T. A quantitative analysis reveals that the  $\rho$  values do not scale with  $M$ , but rather with  $M^2$ . The transversal magnetoresistance consists generally of two terms. The normal magnetoresistance reflects the effect of magnetic field on conduction-electron trajectories. This term should increase the resistivity. The other term, anomalous magnetoresistance, reflects the field effect on magnetic moments. The resistivity decrease implies dominance of the latter term. The quadratic dependence can be related to the even character of magnetoresistance ( $M$  and  $-M$  should lead to the same  $\Delta\rho$ ). On a microscopic scale, the square comes from the dependence of  $\rho$  on  $\langle J_z^2 \rangle - \langle J_z \rangle^2$ , whereas  $M$  is proportional to  $\langle J_z \rangle$  (e.g., in rare earths with total quantum number  $J$ ).  $\langle \dots \rangle$  denotes the thermodynamic expectation value. The proportionality of  $\Delta\rho$  and  $M^2$  proves that the scattering on magnetic moments dominates the resistivity. The magnetoresistance effect is negligible above  $T_C$ .

From the very similar tendency of resistivity decrease with increasing  $T$  we can conclude that it is dominated by the spin disorder at the nanocrystalline U-based hydrides both in the paramagnetic (dynamical disorder) and ordered (static disorder) state. This may seem surprising, as the two types disorder are of rather different nature. However, one has to realize that the primary reason for resistivity decrease is the effect of inelastic scattering of electrons by phonons, leading to a quantum decoherence. The role of magnetic disorder is in reducing the electron mean free path and its details are not significant. This effect becomes important in a situation, in which the  $5f$  states, which carry spin and orbital magnetic moments, are involved in metallic bonding. Any change of orientation of magnetic moments profoundly impacts the electrical transport. The paramagnetic resistivity of “clean” single crystals can then be assumed equivalent to the ultrananocrystalline hydrides, representing a “dirty” limit. It means that the weak localization mechanism may be applicable in a wide class of  $5f$  intermetallics, which exhibit  $\text{TCR} < 0$  in the paramagnetic state, and which have been so far labeled as Kondo systems.

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