Journal of Alloys and Compounds, 207/208 (1994) 221–228
JALCOM 3097

Anisotropy of magnetism in actinide and lanthanide intermetallics

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Abstract

The magnitude of magnetic moments of 4f-electrons in lanthanide intermetallics is well understood in the single-ion picture considering a crystal field interaction which also determines the magnetocrystalline anisotropy in these materials. In intermetallic compounds of actinides, this concept fails due to a significant delocalization of 5f-electron states. Systematic occurrence of particular types of anisotropy in different type structures suggests that the type of magnetic anisotropy is related to a bonding anisotropy. A mechanism responsible for giant anisotropy energies in actinides can be found in the anisotropic bonding of 5f electrons yielding anisotropic exchange interaction between 5f magnetic moments.

1. Introduction

Magnetocrystalline anisotropy, which is manifest by coupling of magnetic moments to certain crystallographic directions, is one of the important characteristics of materials being considered for permanent magnet applications. In particular, the anisotropy energy E_A (or the anisotropy field H_A) is one of the key parameters determining the coercive field in ferromagnets. H_A can be practically defined as the magnitude of the external magnetic field in which the saturation magnetization value is achieved in the hard magnetization direction. As the spin-spin exchange interaction is essentially isotropic, the magnetocrystalline anisotropy appears as the effect of the orientation of orbital moments with respect to the crystallographic axes. Mechanisms responsible for this phenomenon can be the single-ion crystal field interaction and the anisotropic exchange interactions originating, *e.g.* in the anisotropy of bonding. The spin-orbit interaction is a necessary pre-requisite in both cases.

The electronic structure of lanthanide and actinide based materials is strongly influenced by partially occupied f-electron states (4f and 5f states, respectively). However, the magnetic and other electronic properties, especially those of light-lanthanide intermetallic compounds and their light-actinide counterparts, have only little in common. The main reason is the different degree of localization of the 4f and 5f electrons.

The aim of this paper is to discuss mechanisms of the magnetocrystalline anisotropy in these two classes of f-electron materials in the context of the fundamental features of their electronic structure. A special emphasis is put on a discussion of the mechanism of the magnetic anisotropy in uranium intermetallic compounds considering the anisotropy of bonding of 5f-electrons and the existence of large orbital moments, which may originate from itinerant electron states in the case of a strong spin-orbit interaction.

2. Intermetallic compounds of regular lanthanides

Since the maximum 4f electron density is deeply embedded in the core of lanthanide atoms, the interaction of the 4f states with the environment is weak, as a rule, also in intermetallic compounds and can usually be adequately represented by local exchange interactions between the 4f and conductions electrons. This fact is manifest by results of neutron spectroscopy experiments with various rare earth intermetallics [1] revealing a negligible mixing of the 4f states of regular lanthanides (Pr, Nd, Gd–Tm) with conduction electron states.

Consistently, the ground state magnetic moments of regular lanthanide ions even when located in a metallic surrounding usually agree well with the R^{3+} free-ion values ($g_J J$) calculated within the LS coupling scheme. Some discrepancies between the experimental and the calculated moments can be understood as due to crystal field effects lifting the degeneracy of the 4f states. These effects which originate from the electrostatic interaction

0925-8388/94/$07.00
SSDI 0925-8388(94)03097-G

between the charges surrounding the 4f electrons on the one hand and the aspherical 4f charge cloud on the other lead to the single-ion magnetocrystalline anisotropy of lanthanide moments. In this context, the magnetocrystalline anisotropy is a consequence of the directional dependence of the energy of the 4f charge cloud in the crystal field (CF) promoting the preferential orientation of corresponding 4f magnetic moments with respect to the crystallographic axes.

The difference in shape of the 4f cloud (which may be shaped like a cigar or like a pancake) is reflected in the difference in sign of the second-order Stevens factor (α_J) and leads to different easy magnetization directions in analogous materials where the rare earth ions experience the same crystal field. The 4f charge cloud in Gd has a spherical symmetry in the first approximation and therefore the crystal-field induced anisotropy is negligible in Gd based compounds.

As the crystal field depends on spatial distribution of the charge density surrounding the 4f electrons, it is closely connected with the point symmetry of rare earth sites in a crystal. Its nature can be conveniently expressed in terms of the microscopic crystal field Hamiltonian. The parameters V_n^m of this Hamiltonian, which describe the strength of the CF interaction, can be determined experimentally from inelastic neutron scattering measurements [1] combined with the results of magnetic measurements or theoretically from *ab initio* electronic structure calculations [2].

The situation in some lanthanide compounds (especially in those with magnetic transition metal elements) can be more complex and cannot be described only within the single-ion crystal field model. Additional terms are then usually added to a model Hamiltonian. Thus, we can resume that there exists a relatively straightforward theoretical concept for understanding the vast amount of experimental data on the magnetocrystalline anisotropy in many lanthanide compounds with well localized 4f electrons.

3. Actinide intermetallic compounds

The fact that the 5f intermetallic compounds display much stronger magnetic anisotropy than their lanthanide counterparts, has been known for a long time. The anisotropy field estimated as the magnetic field of the intercept of the easy and hard magnetization curves extrapolated to high fields is frequently of the order of 10^2–10^3 T [3,4]. These values are comparable with the anisotropy energy value of about 0.2 eV in US obtained by Brooks *et al.* [5] from the LMTO calculations.

In the case of light actinide intermetallics, there are arguments suggesting another principal anisotropy mechanism than the crystal field interaction. The fundamental difference in the character of 4f and 5f states in metals originates in a much larger spatial extent of the 5f wave functions than of the 4f ones. Therefore, much stronger interaction with the environment should be considered. The 5f electrons are strongly delocalized because of their participation in bonding and hence the strong hybridization of the 5f states with the valence electron states of the neighbouring atoms in the lattice. The delocalization of the 5f electrons has serious consequences, which make the use of the crystal field concept in the light-actinide intermetallics questionable. Here we quote mainly the results obtained on uranium intermetallics, for which the most systematic information is available:

(1) The 5f electron states constitute a more or less narrow 5f energy band (the bandwidth W is of the order of eV) rather than discrete energy levels. Consequently, the magnetic moments of the itinerant 5f electrons (similar to the 3d electrons in the 3d transition metals) are much smaller than expected for a free U ion (U^{3+} or U^{4+}).

(2) No crystal field excitations could be observed by neutron spectroscopy in uranium intermetallics (except for UPd_3) studied so far.

Depending on the environment of the actinide atoms in different intermetallics and on the extent of the bonding character of the 5f states (and consequently the extent of the 5f-ligand hybridization [6]), the 5f electrons delocalized to various degrees give rise to a large variety of magnetic properties. The extraordinary strong magnetocrystalline anisotropy is, however, observed in all compounds in which it was studied, as *e.g.* in an itinerant ferromagnet UNi_2 [7] with a tiny spontaneous magnetic moment of 0.08 μ_B/U atom, in UGa_2 [8], which is a ferromagnet with a large local U magnetic moment, or in the heavy-fermion antiferromagnet UPd_2Al_3 [9].

The magnetic anisotropy studies are usually limited only to rarely available single crystals (usually uranium compounds), and therefore systematic information on the actinide materials in this field is lacking. Materials of the uniaxial lattice symmetry (hexagonal and tetragonal crystal structures) are of particular interest not only from the point of view of potential applications but also as they offer the possibility of unambiguous interpretation of results of magnetization measurements. The easy magnetization direction in these structures may be either parallel (‖) or perpendicular (⊥) to the c-axis. A similar transparent situation, where the easy and hard magnetization directions are, as a rule, orthogonal, is offered also in orthorhombic structures. In the uniaxial structures, however, the anisotropy within the basal plane is difficult to determine due to the higher multiplicity of equivalent directions in this plane. Reliable magnetic anisotropy data in high symmetry systems can be obtained only by means of microscopic experiments, as has been demonstrated by Lander *et al.* [10] who were able to determine the anisotropy constant K_1 of cubic US ($K_1 \approx 10^{10}$ erg cm^{-1} at 0 K) from neutron scattering experiments.

In the case of low symmetry materials, especially those with the uniaxial magnetocrystalline anisotropy, single crystal studies can be successfully simulated by means of high-field magnetization experiments on oriented powders and careful analysis of the data obtained together with those measured on polycrystals brings valuable information [11].

In order to receive more systematic information about the magnetocrystalline anisotropy in uranium compounds, we have focused on magnetism in equiatomic ternary intermetallic compounds UTX of uranium with transition metals (T) and p-electron metals (X). There are few characteristic types of crystal structures in which members of the large UTX family crystallize. The most frequently adopted are the hexagonal ZrNiAl (Fe_2P)-type, the orthorhombic TiNiSi ($CeCu_2$)-type and the hexagonal GaGeLi ($CaIn_2$)-type. The large isostructural groups of UTX compounds are particularly suitable for systematic studies of magnetocrystalline anisotropy over a wide range of magnetic properties in corresponding materials, in which the uranium atoms appear in the same crystallographic environment. Several structure types, on the other hand, make it possible to reveal general aspects in the relationship between the anisotropy type and the symmetry of the environment of U atoms in the crystal lattice and hence, to collect evidence for determining the anisotropy mechanism in the 5f electron intermetallics.

The most extended information has been obtained on compounds with the hexagonal ZrNiAl-type structure. The development of ground state properties in these compounds correlates with the strength of 5f-ligand hybridization [3,12,13]. Thus, within this group of more than 20 compounds, one can find a Pauli paramagnet (UFeAl), weak spin-fluctuators with metamagnetic transitions in low (UCoAl) or high magnetic fields (URuAl), antiferromagnets (UNiX, UTIn) or ferromagnets (UCoGa, UCoSn, URhX, UIrX, UPtAl, UPtGa) with various size of uranium magnetic moments.

Except for the Pauli paramagnet, where no 5f magnetic moments can be expected, magnetization measurements reveal uniaxial magnetic anisotropy with the easy magnetization direction along the *c*-axis in all compounds from this group. The huge anisotropy fields far exceed 35 T, which is the maximum field in our experiments at the Amsterdam High-Field Installation. The values, estimated from the high-field extrapolation of the magnetization curves measured at 4.2 K on single crystals with magnetic field ∥ and ⊥ to the *c*-axis, fall in the range 300–400 T with the exception of UPdIn ($\mu_0 H_A \approx 110$ T) [4]. A common feature of all these compounds (see *e.g.* Fig. 1(a)) is the weak linear (paramagnetic) response of the basal plane magnetization M^{ab} to the magnetic field up to 35 T. The values M^{ab} (35 T) for all compounds (except for UPdIn, where it reaches 0.5 μ_B/f.u. [14]), fall into a narrow interval from 0.10 to 0.14 μ_B/f.u. [4] (note that the width of this interval is comparable with the experimental error). The Pauli paramagnet UFeAl at 4.2 K displays the isotropic paramagnetic magnetization yielding 0.11 μ_B/f.u in 35 T. From these findings we can conclude that the basal plane magnetization in all compounds of this group is reflecting paramagnetic behaviour essentially independent of the size and ordering of uranium magnetic moments which are strongly coupled to the *c*-

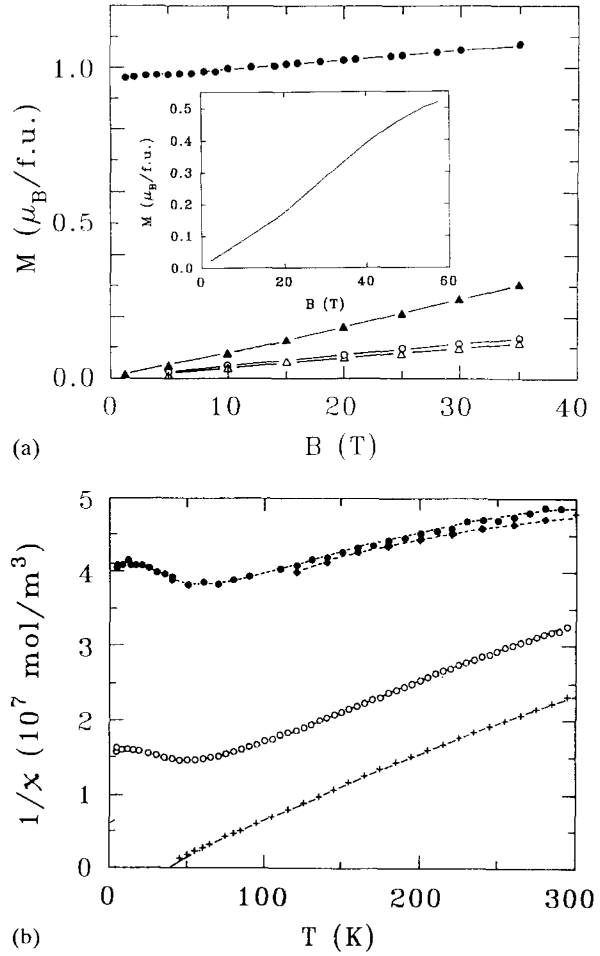

Fig. 1. (a) Magnetization curves measured at 4.2 K on the single crystals of URuAl in the magnetic field parallel (▲) and perpendicular (△) to the *c*-axis and URhAl in the magnetic field parallel (●) and perpendicular (○) to the *c*-axis. Inset: The *c*-axis magnetization curve of URuAl at 4.2 K in fields up to 60 T. (b) Temperature dependence of the inverse magnetic susceptibility in the single crystals of URuAl in the magnetic field parallel (○) and perpendicular (●) to the *c*-axis and URhAl in the magnetic field parallel (+) and perpendicular (◆) to the *c*-axes.

axis. Consistently, the neutron diffraction studies of UCoAl [15], UNiAl [16], UNiGa [17], URhAl [18] and UPdIn [19] have revealed collinear magnetic structures in all cases with the c-axis as the preferential direction. The universality of the uniaxial anisotropy in the UTX compounds with the ZrNiAl-type structure has also been documented in the studies of the onset of ferromagnetism in the pseudoternary system $U(Ru_{1-x}Rh_x)Ga$ [11]. Whereas the ordered uranium magnetic moments of more than 1 μ_B in URhGa are oriented along the c-axis, the weak magnetization response in the basal plane is almost the same as in paramagnetic URuGa, as well as in other compounds throughout the $U(Ru_{1-x}Rh_x)Ga$ series. The extremely strong coupling of uranium magnetic moments to the c-axis is also manifest by the immeasurable effect of the magnetic field oriented within the basal plane on the specific heat anomalies connected with the magnetic phase transitions.

The giant uniaxial anisotropy in the UTX compounds is clearly seen in the susceptibility behaviour in the paramagnetic state (see *e.g.* Fig. 1(b)). The $M^c(B)$ curves of UCoAl [20], URuAl [11,21] and URuGa [11] at 4.2 K are distinctly above the basal plane signals $M^{ab}(B)$ and moreover, the field induced transitions in UCoAl and URuAl (see the inset in Fig. 1(a)) are seen only in the c-axis magnetization. The basal plane susceptibility χ^{ab} is much weaker than χ^c even at room temperature and this difference increases rapidly with decreasing temperature. Both temperature dependencies $\chi^{ab}(T)$ and $\chi^c(T)$ can be reasonably well approximated by the modified Curie–Weiss law

$$\chi = \frac{C}{T-\Theta_p} + \chi_0$$

with the isotropic Curie constant C and temperature independent susceptibility χ_0, whereas the anisotropy is expressed by the difference $\Delta\Theta_p = |\Theta_p^c - \Theta_p^{ab}|$. The values of $\Delta\Theta_p$ obtained on available single crystals are all approximately 350–450 K; only the value for UPdIn is much lower (108 K). Closer inspection of $\Delta\Theta_p$ and $\mu_0 H_A$ values reveals that they correlate quite well [4]. There is, however, a large uncertainty in determining the anisotropy field by means of extrapolation to much higher high magnetic fields than the maximum field in the experiment (35 T). The distinct difference in both values $\mu_0 H_A$ and $\Delta\Theta_p$ for UPdIn in comparison with other compounds of this type, should be considered as meaningful.

Before a closer inspection of magnetic structures, some basic characteristics of the ZrNiAl-type structure should be mentioned. It is a layered structure obtained by stacking the $U–T_I$ and $T_{II}–X$ basal plane layers separated by $c/2$ (typically $c = 380–400$ pm). Uranium atoms are closer together within the U–T layer (4 nearest U neighbours at $d_{U-U} = 350–390$ pm). Moreover, much stronger hybridization of uranium 5f states with the d electron states of the transition metal within the $U–T_I$ layers has been concluded from the polarized neutron experiments on URhAl [18] and URuAl [22] revealing an order of magnitude larger magnetization density induced on the transition metal atoms in T_I position than in the T_{II} site. The ferromagnetic coupling within the basal plane may be attributed to the direct 5f–5f exchange interaction (due to the overlap of 5f wave functions from neighbouring U atoms) and/or the indirect 5f–d–5f exchange interaction induced by the 5f–d hybridization due to the strong bonding of uranium 5f wave functions within the U–T layers. Which of these two exchange mechanisms are more effective depends on the details in the interatomic distances within the U–T layer in a particular compound. The exchange interaction along the c-axis is typically much weaker than the strong ferromagnetic coupling within the basal plane, nevertheless its character finally determines the type of magnetic structure.

The second important ingredient of the magnetic anisotropy in UTX compounds is an orbital magnetic moment on U sites. Although the giant magnetic anisotropy in some light actinide intermetallics has been known for a long time, the existence of orbital moments in the system of itinerant 5f electrons was not commonly believed and the effect of the quenching similar to the 3d transition metals was supposed. Brooks and Kelly, when calculating the electronic structure in UN [23], were the first to consider the role of the strong spin-orbit coupling in the formation of orbital polarization in a narrow band system. An important difference between the 3d transition metals and the light actinides is the relation between the energy of the spin-orbit coupling Δ_{S-O} and the energy width of the 3d (5f) band W_{3d} (W_{5f}). Whereas $\Delta_{S-O} \ll W_{3d}$, the respective values in light actinides become comparable as Δ_{S-O} is of the order of eV. Since that time a large experimental effort by neutron scattering teams in magnetic form factor studies brought evidence of strong orbital components of the 5f magnetic moments in a number of light actinide intermetallics [24] and in numerous cases it has been corroborated by relevant electron structure calculations [25]. The spin-polarized calculations, including an orbital polarization term, predict in light actinides the reduction of the orbital moment due to the strong hybridization of the 5f states with other electron states. On the other hand, this orbital moment is very large compared to the d-transition metal systems. There are several interesting consequences of this orbital 5f-band magnetism as, *e.g.* "magnetic" materials with zero-ordered moment [25]. This can arise in cases where the orbital and spin contributions would exactly cancel ($\mu_{5f} = \mu_L - \mu_S$ in light actinides), which almost happen in UNi_2 [26] and UFe_2 ($\mu_{5f}=0.01\,\mu_B$, whereas $\mu_L=0.23\,\mu_B$ and $\mu_S=0.22\,\mu_B$) [24], so that the net moment is zero. Large orbital moments have been predicted and detected also in some UTX compounds such as UCoAl [15,27] and URhAl [22,28] and even in URuAl [18,28] in which the strong 5f electron delocalization prevents the magnetic ordering.

The giant uniaxial anisotropy in UTX compounds ZrNiAl-type structure can be then understood as follows. The strong bonding of 5f electrons within the U–T basal plane compresses the 5f charge density towards this plane, which leads to the orbital moment confinement in the *c*-axis (perpendicular to the strong bonding plane). As a consequence, the Ising type of magnetism is observed in these and other materials with strong bonding in uranium layers. A question remains whether the anisotropy energy of the order of hundreds T can be related to the energy of the ferromagnetic exchange interaction between uranium magnetic moments within the basal plane.

An interesting question arises about the application of large enough magnetic fields to break such anisotropy where the direction of magnetic moments is intimately connected with the bonding symmetry. Then the effort to reach states with another orientation of 5f magnetic moments may lead to the violation of this type of bonding. The estimated anisotropy energies in UTX compounds are indeed comparable with the difference in cohesion energies of different structures, which can be generally of the order of 0.1 eV.

In order to test one of basic features of the model presented leading to the easy magnetization axis perpendicular to strong bonding directions, we extended our experiments on UTX compounds crystallizing in other structure types, where we can expect other symmetries of bonding. Fourteen UTX compounds (X–Si or Ge) from the second largest isostructural group have in common the orthorhombic structure of the TiNiSi-type. The nearest interuranium distance (typically close to 350 pm) in these materials can be found in zig-zag chains of uranium atoms stretched along the *a*-axis with a small amplitude *c*/10. The second uranium nearest neighbours are at slightly larger distance on two of neighbouring chains, again generating zig-zag chains along the *b*-axis but with much larger amplitude along *c* (0.4*c*). Therefore *a* can be considered as the effectively strongest bonding axis for the 5f states.

The high-field magnetization measurements performed on UNiGe (see Fig. 2(a)) [29,30], UPdGe [30] and UPtGe [30] single crystals have revealed in all three cases the *a*-axis to be the hard magnetization axis. The weak linear response of the *a*-axis magnetization in UNiGe at 4.2 K to magnetic field yields 0.23 μ_B/f.u. in 35 T, whereas in the other two principal axes the magnetization reaches 1.43 μ_B/f.u. Similar high field magnetization values were also measured in the other two compounds. Neutron diffraction results [31–34] revealed, in agreement with the magnetization data, that the ground state magnetic structures in these three compounds have in common the uranium magnetic moments confined in the *b*–*c* plane and ferromagnetically coupled along the *a*-axis. In addition, the powder polycrystal magnetization measurements (see the above-mentioned method of oriented powder studies) of other UTSi and UTGe compounds [35], which were not available in a single crystal form, are consistent with the easy plane anisotropy, which is then the common feature of all UTX compounds crystallizing in the TiNiSi-type structure irrespective of the type of ground state (*e.g.* UNiGe and UPdGe are antiferromagnetic, whereas UPdGe rests in the ferromagnetic state).

Moreover, the temperature dependencies of the paramagnetic susceptibility measured on the single crystals of UNiGe (see Fig. 2(b)) [29,36], UPdGe [36] and UPtGe [36] show the same symmetry of the anisotropy in the paramagnetic state. The Curie–Weiss law (eqn. (1)) analysis of the temperature dependence susceptibility in UNiGe [29] in all three principal axes yields a moderate anisotropy in the *b*–*c* plane $\Delta_p^{ab}=|\Theta_p^c-\Theta_p^b|\approx 40$ K but the *a*-axis data reveal a much stronger anisotropy $\Delta\Theta_p^{ab}\approx 140$ K. Note that the high-field extrapolation of magnetization data yields an anisotropy field of about 210 T.

Another test structure of UTX compounds on which reliable experimental data exist (UPdSn, UAuSn, UCuSn and UPdSb) is the hexagonal structure of the $CaIn_2$ type (GaGeLi type for the ordered ternary version) [3,37]. Here, the nearest uranium atoms (d_{U-U} is typically 360 pm) form true linear chains along the *c*-axis with a large inter-chain separation. The first three compounds are antiferromagnetic at low temperatures [37–40] whereas UPdSb is a ferromagnet [41]. Results of magnetization measurements performed on the single crystals of UPdSn [37] and powders of the latter three compounds [37,42] are consistent with the easy plane anisotropy. The magnetic structure of UAuSn is collinear antiferromagnetic with magnetic moments parallel to the orthorhombic *b*-axis [38]. The ferromagnetic coupling along *c* conforms to the strongest bonding along the *c*-axis. The antiferromagnetic structure in UPdSn is non-collinear [39], nevertheless the effective hard magnetization direction is unambiguously the *c*-axis [37]. The estimated anisotropy energy in this compound ($\mu_0H_A\approx 110$ T, $\Delta\Theta_p)\approx 60$ K) is comparable with the values which we meet in UPdIn, the compound with the most localized 5f states in comparison to other ZrNiAl-type structure UTX materials. In comparison with UAuSn, it is not clear whether the non-collinear structure in UPdSn is only due to a higher degree of the 5f-electron localization [43], or whether UAuSn,

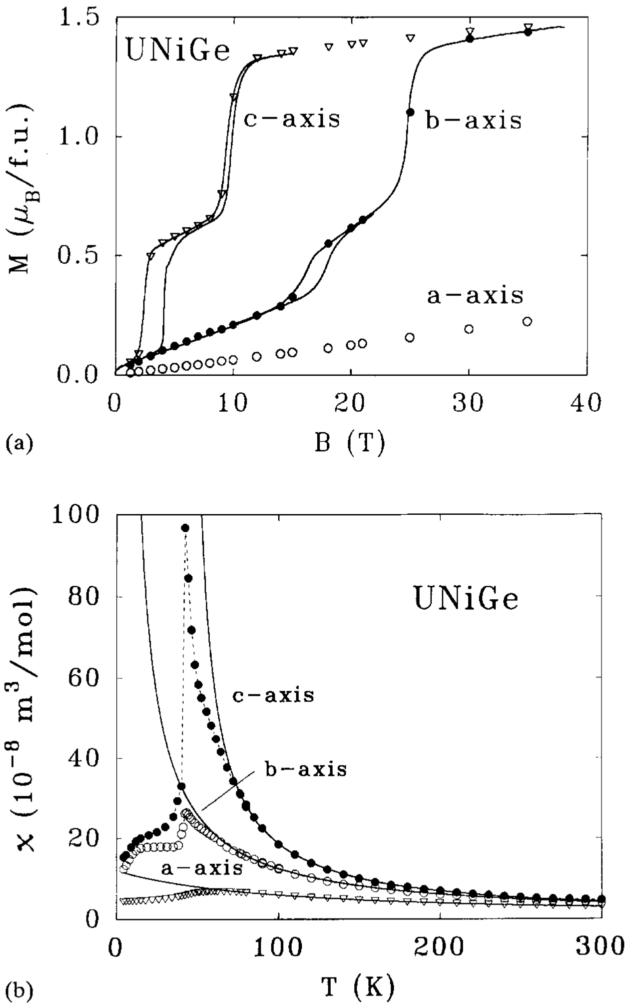

Fig. 2. (a) Magnetization curves measured at 4.2 K on the single crystal of UNiGe in the magnetic field applied along the *a*-, *b*- and *c*-axis. (b) Temperature dependence of the inverse magnetic susceptibility in the single crystal of UNiGe in the magnetic field applied along the *a*-, *b*- and *c*-axes.

with the presumably less localized 5f-states, would adopt a similar magnetic structure if it were allowed by the crystal symmetry (UPdSn has a lower symmetry of the structure as, unlike UAuSn, it is an ordered ternary compound).

From the analysis of the magnetic anisotropy in a large number of UTX compounds, which can be segregated into three characteristic crystal structures, we can conclude:

(1) There exists directions (planes) of strong bonding (hybridization) of the uranium 5f with nearest neighbour states in each particular structure, which leads to compression of the 5f charge density along these directions (within these planes).
(2) Orbital moments of 5f electrons orient perpendicular to these directions (planes) which leads to the easy-plane (easy-axis) anisotropy.
(3) The exchange interactions induced by this strong bonding (hybridization) are usually ferromagnetic which typically leads to collinear magnetic structures with strong ferromagnetic coupling of the 5f moments along these directions.
(4) The exchange interactions (ferromagnetic or antiferromagnetic) in perpendicular directions, which are weaker, determine the type of ground state magnetic structure (ferromagnetic antiferromagnetic).

It is interesting to inspect whether another uranium or other light-actinide compounds also display similar features. We can conclude that, at least in cases where we can easily determine the strong bonding directions, the easy magnetization directions are perpendicular to them irrespective to the degree of 5f electron delocalization (*e.g.* in the strong ferromagnet UGa_2 [8], heavy-fermion compounds such as UPt_3 [44] or UPd_2Al_3 [9]). Although, the model which relates the magnetic anisotropy to the bonding anisotropy works well for a number of uranium compounds and enables easy predictions of magnetic anisotropy type, it is not *a priori* clear which are the most decisive factors in determining the real bonding anisotropy. In particular, it is not possible to estimate how far the bonding strength in this case is assisted by the 5f–d hybridization, or, in other words, what is the importance of the layout of the non-f ligands. In order to shed more light on this issue, we were searching for compounds displaying different basic arrangement of uranium atoms, *i.e.* planar or linear within one particular type structure. Such a possibility is offered by the recently discovered system of U_2T_2X compounds [45,46] crystallizing in the tetragonal U_3Si_2-type structure, where a specific choice of ligands leads to compounds with the nearest uranium neighbours either within the basal plane or along the *c*-axis, *e.g.* U_2Ni_2Sn and U_2Pt_2Sn. This is one of the main reasons why we are going to concentrate on the system of U_2T_2X ternary intermetallics.

When looking for an appropriate microscopic picture to account for such a situation, we can use for example the concept of the hybridization-induced anisotropy developed by Cooper *et al.* [47], which has been applied up to now to cerium and actinide monopnictides and monochalcogenides. It is based on the Coqblin–Schrieffer theory describing the interaction of an f-ion with surroundings due to hybridization [48], but is extended towards two-ion effects. Although this approach is justified only for a moderate delocalization case, it explains for example that the ferromagnetic type of coupling along the bonding axis is preferred in systems with dominating hybridization-induced exchange interactions.

4. Conclusions

The regular lanthanide intermetallics and the intermetallic compounds based on light actinides can be considered as representing two limit cases of f-electron magnetism. Since the well localized 4f electron states forming discrete energy levels have a negligible mixing with other electron states, the magnitude of the 4f magnetic moments are well defined within the ionic picture. The 4f charge density distribution is modified only to a limited extent (with respect to the free atom case) due to the interaction with the crystal field. The 4f orbital moment is then choosing direction(s) satisfying the minimum energy of the crystal field interaction. Since the crystal field symmetry is closely connected with the point symmetry at the particular rare earth ion site, the magnetocrystalline anisotropy is reflecting this symmetry in conjunction with the symmetry of the 4f wave function. The crystal field parameters, which are mapping this situation, can be determined either experimentally or by means of relevant electronic structure calculations, and the character of the magnetocrystalline anisotropy can be understood in simple cases from analysing the microscopic crystal field Hamiltonian.

A large spatial extent of the 5f orbitals leads in typical light-actinide intermetallic compounds to participation of the 5f electrons in bonding and consequently a more or less wide 5f band pinned at E_F is formed. The itinerant character of the 5f electrons is reflected in reduced 5f magnetic moments. Such 5f moments have, however, a significant orbital component due to a strong spin-orbit interaction. The participation of the 5f states in bonding leads to a compression of the 5f charge densities towards the bonding directions, which, for a particular structure type, are given primarily by shortest inter-actinide directions. Then, for example, if the 5f-bonding within a plane is dominating (as in the case of ZrNiAl-type compounds), the compression of the 5f-charge towards the plane leads to the orientation of orbital moments perpendicular to the plane. Moreover, a strong ferromagnetic coupling between actinide moments appears as a rule along the strong bonding direction. As the preferential moment direction is also maintained for paramagnetic materials, we can specify for each structure type easy and hard-magnetization directions irrespective of the type of the ground state. Hence, the character of the anisotropy in itinerant uranium intermetallics reflects rather the layout of the nearest U atoms in the lattice than in the point symmetry at the actinide sites.

Acknowledgments

This work is part of the research program of the "Stichting voor Fundamenteel Onderzoek der Materie (FOM)", which is financially supported by the "Nederlandse organisatie voor Wetenschappelijk Onderzoek (NWO)". The research at Charles University in Prague was supported by the University grant no. 312. Part of the work of V.S. and L.H. was also supported by the Alexander von Humboldt Foundation.

References

1 P. Fulde and M. Loewenhaupt, *Adv. Phys., 34* (1986) 589 and refs. therein.

2 M. Divis and J. Kuriplach, *Physica B, 183* (1993) 25.

3 V. Sechovsky and L. Havela, in E.P. Wohlfarth and K.H.J. Buschow (eds.), *Ferromagnetic Materials*, Vol. 4, North-Holland, Amsterdam, 1988, pp. 309–491 and refs. cited therein.

4 V. Sechovsky, L. Havela, F.R. de Boer and E. Brück, *J. Alloys Comp., 181* (1992) 179.

5 M.S.S. Brooks, B. Johansson, O. Eriksson and H.L. Skriver, *Physica B, 144* (1986) 1.

6 D.D. Koelling, B.D. Dunlap and G.W. Crabtree, *Phys. Rev. B, 31* (1985) 4966.

7 P.H. Frings, J.J.M. Franse, A. Menovsky, S. Zemirli and B. Barbara, *J. Magn. Magn. Mater., 54–57* (1986) 541.

8 A.V. Andreev, K.P. Belov, A.V. Deriagin, R.Z. Levitin and A. Menovsky, *J. Phys., 40* (1979) C4–82.

9 A. de Visser, H. Nakotte, L.T. Tai, A.A. Menovsky, S.A.M. Mentink, G.J. Nieuwenhuys and J.A. Mydosh, *Physica B, 179* (1992) 84.

10 G.H. Lander, M.S.S. Brooks, B. Lebech, P.J. Brown, O. Vogt and K. Mattenberger, *Appl. Phys. Lett., 57* (1990) 989.

11 V. Sechovsky, L. Havela, F.R. de Boer, P.A. Veenhuizen, K. Sugiyama, T. Kuroda, E. Sugiura, M. Ono, M. Date and A. Yamagishi, *Physica B, 177* (1992) 164.

12 V. Sechovisky, L. Havela, E. Brück, F.R. de Boer and A.V. Andreev, *Physica B, 163* (1990) 103.

13 T. Gasche, S. Auluck, M.S.S. Brooks and B. Johansson, *J. Appl. Phys., 70* (1990) 6580.

14 H. Fujii, H. Kawanaka, M. Nagasawa, T. Takabatake, Y. Aoki, T. Suzuki, T. Fujita, E. Sugiura, K. Sugiyama and M. Date, *J. Magn. Magn. Mater., 90–91* (1990) 507.

15 M. Wulff, J.M. Fournier, A. Delapalme, B. Gillon, V. Sechovsky, L. Havela and A.V. Andreev, *Physica B, 163* (1990) 331.

16 J.M. Fournier and P. Burlet, *Proc. 21st Journées des Actinides*, Montechoro, Portugal, 1991, p. 126.

17 H. Maletta, R.A. Robinson, A.C. Lawson, V. Sechovsky, L. Havela, L. Jirman, M. Divis, E. Brück, F.R. de Boer and A.V. Andreev, *J. Magn. Magn. Mater., 104–107* (1992) 21.

18 J.A. Paixao, G.H. Lander, P.J. Brown, H. Nakotte, F.R. de Boer and E. Brück, *J. Phys.: Condensed Matter., 4* (1992) 829.

19 E. Sugiura, K. Sugiyama, H. Kawanaka, T. Takabatake, H. Fujii and M. Date, *J. Magn. Magn. Mater., 90–91* (1990) 65.

20 V. Sechovsky, L. Havela, F.R. de Boer, J.J.M. Franse, P.A. Veenhuizen, J. Sebek, J. Stehno and A.V. Andreev, *Physica B, 142* (1987) 283.

21 P.A. Veenhuizen, F.R. de Boer, A.A. Menovsky, V. Sechovsky and L. Havela, *J. Phys.(Paris)-Colloq., C8* (1988) 1485.

22 J.A. Paixao, G.H. Lander, P.J. Brown, H. Nakotte, F.R. de Boer and E. Brück, *Europhys. Lett., 24* (1993) 607.

23 M.S.S. Brooks and P.J. Kelly, *Phys. Rev. Lett., 51* (1983) 1708.

24 B. Lebech, M. Wulff and G.H. Lander, *J. Appl. Phys., 69* (1991) 5891 and refs. cited therein.

25 G.H. Lander, M.S.S. Brooks and B. Johansson, *Phys. Rev. B, 43* (1991) 13672 and refs. cited therein.

26 J.M. Fournier, A. Boeuf, P. Frings, M. Bonnet, J.X. Boucherle, A. Delapalme and A. Menovsky, *J. Less-Common Metals, 121* (1986) 249.

27 D. Eriksson, B. Johansson and M.S.S. Brooks, *J. Phys.: Condensed Matter, 1* (1989) 4005.

28 T. Gasche, B. Johansson and M.S.S. Brooks, Ground state properties of ternary uranium compounds, to be published.

29 L. Havela, V. Sechovsky, F.R. de Boer, E. Brück and H. Nakotte, *Physica B, 177* (1992) 159.

30 S. Kawamata, G. Kido, K. Ishimoto, Y. Yamaguchi, H. Iwasaki, N. Kobayashi and T. Komatsubara, *Physica B, 177* (1992) 169.

31 R.A. Robinson, A.C. Lawson, J.W. Lynn and K.H.J. Buschow, *Phys. Rev. B, 47* (1993) 6138.

32 S. Kawamata, K. Ishimoto, Y. Yamaguchi and T. Komatsubara, *J. Magn. Magn. Mater., 104–107* (1992) 51.

33 A. Murasik, P. Fisher, R. Troc and V.H. Tran, *J. Phys.: Condensed Matter, 3* (1991) 1841.

34 V. Sechovsky, L. Havela, K. Prokes, H. Nakotte, E. Brück, F.R. de Boer, P. Svoboda and H. Maletta, *Proc. 23rd Journées des Actinides*, Schwarzwald 1993, 05.3.

35 F.R. de Boer, E. Brück, V. Sechovsky, L. Havela and K.H.J. Buschow, *Physica B, 163* (1990) 175.

36 S. Kawamata, K. Ishimoto, H. Iwasaki, N. Kobayashi, Y. Yamaguchi, T. Komatsubara, G. Kido, T. Mitsugashira and Y. Muto, *J. Magn. Magn. Mater., 90–91* (1990) 513.

37 F.R. de Boer, E. Brück, H. Nakotte, A.V. Andreev, V. Sechovsky, L. Havela, P. Nozar, C. J. M. Denissen, K. H. J. Buschow, B. Vaziri, M. Meissner, H. Maletta, and P. Rogl, *Physica B, 176* (1992) 275.

38 R.A. Robinson, J.W. Lynn, V. Nunez, K.H.J. Buschow, H. Nakotte and A.C. Lawson, *Phys. Rev. B, 47* (1993) 5090.

39 R.A. Robinson, A.C. Lawson, K.H.J. Buschow, F.R. de Boer, V. Sechovský and R.B. von Dreele, *J. Magn. Magn. Mater., 98* (1991) 147.

40 H. Fujii, H. Kawanaka, T. Takabatake, E. Sugiura, K. Sugiyama and M. Date, *J. Magn. Magn. Mater., 87* (1990) 235.

41 T.T.M. Palstra, G.J. Nieuwenhuys, R.F.M. Vlastuin, J. van den Berg, J.A. Mydosh and K.H.J. Mydosh, *J. Magn. Magn. Mater., 67* (1987) 331.

42 V. Sechovský, unpublished results.

43 L. Havela, V. Sechovsky, J.R. Naegele, T. Almeida, E.H. Brück, H. Nakotte and F.R . de Boer, *J. Magn. Magn. Mater., 104–107* (1992) 23.

44 J.J.M. Franse, P.H. Frings, A. de Visser, A. Menovsky, T.T.M. Palstra, P.H. Kes and J.A. Mydosh, *Physica B, 126* (1984) 116.

45 F. Mirambet, P. Gravereau, B. Chevalier, L. Trut, and J. Etourneau, *J. Alloys Comp., 191* (1993) L1.

46 M.N. Peron, Y. Kergadallan, J. Rebizant, D. Meyer, S. Zwirner, L. Havela, H. Nakotte, J.C. Spirlet, G.M. Kalvius, E. Collineau, J.L. Oddou, C. Jeandey, J.P. Sanchez and J.M. Winand, *J. Alloys Comp., 201* (1993) 203.

47 B.R. Cooper, R. Siemann, D. Yang, P. Thayamballi and A. Banerjea, in A.J. Freeman and G.H. Lander (eds.), *Handbook on the Physics and Chemistry of the Actinides*, Vol. 2, North-Holland, Amsterdam, 1985, pp. 435–500.

48 B. Coqblin and J.R. Schrieffer, *Phys. Rev., 185* (1969) 847.