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Itinerant Magnetism in Actinide Systems

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ABSTRACT

A survey is presented on the occurrence of itinerant-electron magnetism in actinide metals with emphasis on the uranium Laves phase- and the uranium-platinum compounds. The Laves phase compounds can be understood with concepts of magnetism that have been developed for the transition metals, including that of spin fluctuations. The absence of magnetic order in most of the uranium-platinum compounds in combination with large values for the coefficient of the electronic term in the specific heat points to a strong hybridisation of f and d electron states. The magnetic order of UPt is complex and can not simply be represented by an itinerant-electron model.

I. INTRODUCTION

The unusual electronic and magnetic properties of the actinide metals have attracted growing interest in the past recent years. In particular, uranium and its compounds are widely studied on polycrystalline as well as on single-crystalline samples. The electronic and magnetic properties of the actinide metals and their compounds are largely determined by the unfilled 5f shell. The 5f electrons are less localized than the 4f electrons in the corresponding rare earth series. This conclusion emerges from both experimental and theoretical work.[1-4] It is illustrated by the variation in the atomic volumes of the different d and f transition metal series, see Fig.1. The atomic volumes of the d-transition metals, plotted as a function of the d-electron concentration, show a parabolic behaviour that is ascribed to the d-electron

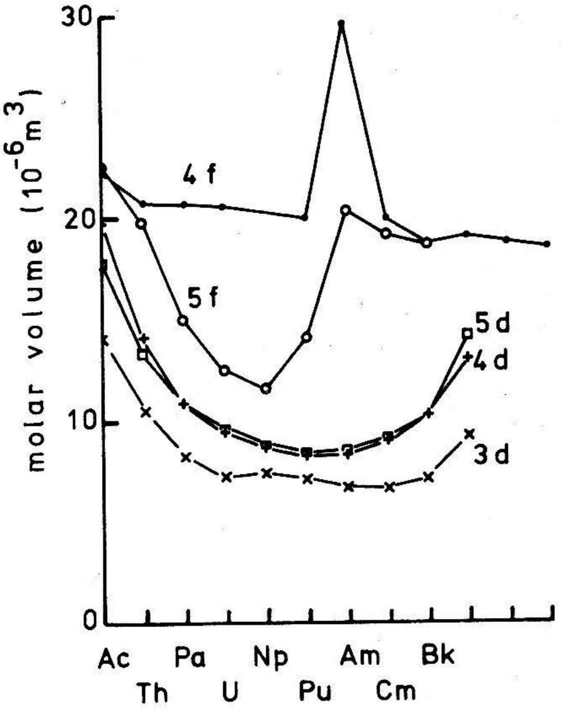

Fig.1.Molar volumes of d- and f-transition metal series.

contribution to the metallic bonding. It is considered as a proof for the itinerant character of these electrons. This parabolic behaviour is absent for the 4f series but appears again in the first half of the actinide series. In the light actinide elements Th to Pu, no magnetic order has been observed. On the contrary, superconductivity occurs in Th, Pa and U, whereas spin-fluctuation effects are reported for Np and Pu. Magnetic order has been found in Cm and Bk; Am is supposed to be in a non-magnetic $5f^6$, $J = 0$ ground state. The presence and the itinerant or localized character of the 5f electrons has large consequences for the crystallographic and magnetic structures. The heavy actinide metals behave like the rare earth metals, whereas for the light actinide metals several, sometimes unconventional, structural modifications have been found.[5] Magnetic structures in some uranium compounds are as complex as in some cerium compounds.[6] The overlap between the 5f wave functions of neighbouring atoms decreases going from the light to the heavy actinide metals. This overlap can also be varied in a series of suitably choosen compounds based on one of the actinide elements. In such a series the gradual change from itinerant-electron states (superconductivity) to localized states (complex magnetic structures with high magnetic anisotropies) via the intermediate spin-fluctuation state can be studied. Because most of the actinide elements, with the exception of Th and U, are available in small quantities only and since radio-activity and toxicity require other restrictions in handling these materials, the thorium and uranium compounds have been investigated most extensively. The strong reactivity of uranium asks for additional precautions in preparing compounds, especially in the single-crystalline state. Single crystals of some intermetallic uranium compounds have succesfully been grown by the Czochralski method using a cold-crucible technique in combination with an arc-melt installation.[7]

In discussing possibilities for localization of the 5f electrons, important parameters are the bandwith (W) that represents the hopping energy of the 5f electrons between the actinide ions, and the intra-atomic Coulomb repulsion (U) between the 5f electrons. For U much smaller than W we are in the limit of itinerant behaviour, whereas for $U \cong W$ a transition may occur from itinerant to localized electron states. By increasing the interatomic distance between the actinide atoms in different compounds, this transition is likely to occur. An interesting question is whether magnetic order is restricted to those compounds in which this localization is realized. Moreover, does the hybridization of the 5f states with d states prevent this localization in intermetallic compounds of uranium with d elements?

This paper is devoted to the phenomenon of itinerant-electron magnetism in actinide systems. It is mainly concerned with intermetallic compounds of uranium with d elements. Important parameters in a description of this type of magnetism are the density of electron states at the Fermi level, $N(E_F)$, and the effective interaction parameter I_{eff} between the itinerant electrons. In the Stoner model, the product of these two parameters should exceed a critical value in order to split the spin-up and spin-down subbands in energy. For the light actinide elements this criterion is apparently not fullfilled, although the product $I_{eff}N(E_F)$ approaches the critical value of 1. According to Skriver et al.[8], values for I_{eff} of the order of 0.5 eV can be expected for these elements. Values for the coefficient γ of the electronic term in the specific heat are of the order of 10 mJ/K^2mole, corresponding to a value for $N(E_F)$ of approximately 2 states/eV atom spin, see Table I. The γ-values of the pure actinide metals do not exceed the numbers that are usually found in the d-transition metals. In several non-magnetic intermetallic uranium compounds, however, giant γ-values are observed. It is sometimes difficult to understand why magnetic order is absent in those compounds. Criteria that are used in general for characterising a magnetically ordered compound as an itinerant-electron system are:

- the ratio between the effective and the ordered magnetic moments is much larger than 1;
- the magnetic moment remains susceptible even at 0 K;
- ferromagnetism is suppressed under hydrostatic pressures;
- the coefficient γ in the specific heat reaches large values;
- the magnetic entropy is much smaller than R ln 2.

None of these single criteria, however, delivers a sufficient proof for the itinerant nature of a ferromagnetic or antiferromagnetic compound. The collection of criteria, completed with additional information from resistivity, neutron diffraction, spectroscopic and other microscopic measurements, contributes to the classification of magnetic materials.

The systematics in the occurrence of superconductivity and magnetic order in actinide compounds is best represented by a critical value for the distance between nearest neighbour actinide ions, which value is known as the Hill limit. For uranium compounds this critical value amounts to 3.4 - 3.6 Å.[1]

Below this value no spontaneous magnetic order of the uranium sublattice exists, whereas beyong this value no superconductivity is found (see fig.2).

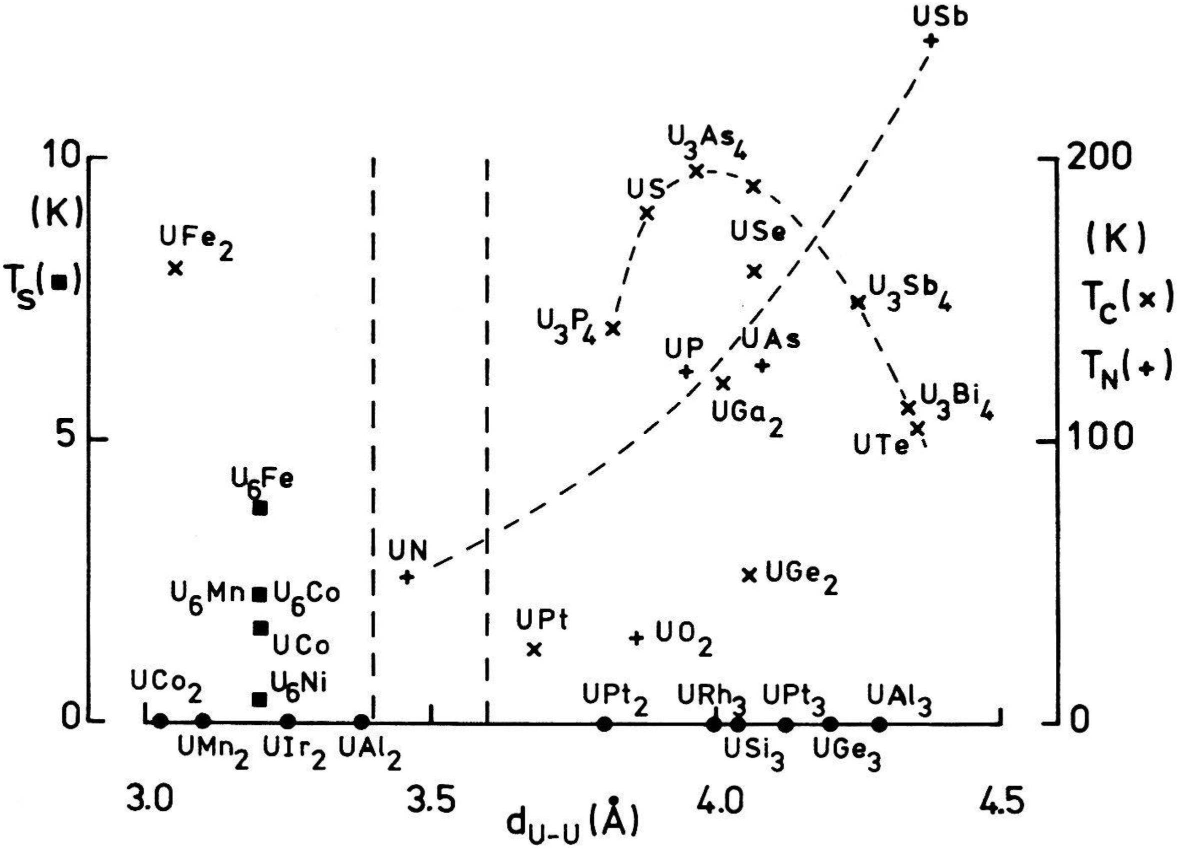

Fig.2. Hill plot for a selected number of superconducting, paramagnetic (●), ferromagnetic and antiferromagnetic uranium compounds; d_{U-U} is the nearest neighbour distance between uranium ions.

The Hill limit gives the restrictions under which magnetic order or superconductivity occurs in the actinide systems. It does not predict these phenomena as, for instance, the Stoner criterion does for the occurrence of itinerant-electron ferromagnetism. Nevertheless, it is a remarkable point that the same critical value is applicable for intermetallic compounds as well as for pnictides and chalcogenides. Apparent exceptions as UFe_2 and UNi_2 do not really disturb the systematics as will be discussed later. Of special importance for the subject of itinerant-electron magnetism are compounds in which the nearest neighbour distance between uranium atoms, d_{U-U}, approximates the critical value of 3.4 - 3.6 Å. To this category belong the uranium Laves phase compounds UX_2 with d_{U-U} values between 3.0 and 3.4 Å, orthorhombic UPt and cubic UN.

II. MAGNETIC PROPERTIES OF INTERMETALLIC URANIUM COMPOUNDS

Recent reviews of magnetism in actinides and their compounds have been presented by Brodsky[4] in 1978 and by Trzebiatowski[5] in 1980. Since that time progress has been made, not at least in the study of the intermetallic uranium compounds.

Up till now the uranium pnictides and chalcogenides have most extensively been investigated. Magnetic order frequently occurs in these compounds. Monopnictides with the NaCl-type of structure order antiferromagnetically, whereas in some monochalcogenides with the same structure ferromagnetic order is reported. Ferromagnetism is also found in U_3X_4 compounds ($X = P$, As, Sb, Bi) and antiferromagnetism for UX_2 compounds ($X = O$, P, As, Sb, Bi). Itinerant-electron magnetism is suggested for the antiferromagnetic uranium mononitride. Pressure effects on the magnetic parameters of UN are negative. The close agreement between values for the relative pressure dependence of the ordered magnetic moment and the Néel temperature is considered as a strong argument for the itinerant character of antiferromagnetism in this compound.[9] At the other side, however, Lemmer and Lowther[10] concluded on the basis of the magnetic data to a $5f^2$ configuration of uranium in UN and obtained a satisfactory description of the ordered magnetic moment and the paramagnetic susceptibility in a crystal-field calculation.

Intermetallic compounds of uranium with the IIIA and IVA group of elements show interesting properties in between itinerant and localized magnetism. Ferromagnetism with in general large magnetic anisotropy occurs in U_2Ga_3, UGa_2, U_3Ge_4 and UGe_2 and antiferromagnetism in UX_3 compounds with $X = Ga$, In, Tl, Pb.[5] Materials with a strong magnetic anisotropy ask for magnetization studies on single crystals. These type of experiments have been performed on UGa_2[11] and UGe_2.[12] The large values for the high-field susceptibility along the easy axis in these compounds could indicate a substantial contribution of polarized electron states to the magnetic moment. Spin-fluctuation effects have been reported for several UX_3 compounds[13] ($X = Al$, Ga, In, Si, Ge, Sn) and for UAl_2 which compound is considered as the prototype of a spin-fluctuating material.

The number of magnetically ordered intermetallic compounds of uranium with transition elements is very limited. Of 46 binary compounds of uranium with d transition elements of group VIII, for instance, superconductivity has been reported so far for five compounds (U_6M with $M = Mn$, Fe, Co, Ni and UCo) and magnetic order for only four compounds (UPt, UFe_2, UPd_4 and

U_2Co_{11}). For several compounds belonging to this group contradictory results or interpretations of the magnetic phenomena have been reported, among them UNi_2, UPt_2 and UPt_3. A theoretical understanding of this category of compounds has to take into account the large hybridization of the 5f uranium- and the transition metal d states. The destructing effect of f-d hybridization on magnetic order is once more illustrated by the compounds UNi_5 (paramagnetic) and UCu_5 (antiferromagnetic, $T_N = 15$ K).[14] For the subject of itinerant-electron magnetism this hybridization is most interesting of course.

In the search for itinerant-electron magnetism in intermetallic uranium compounds we shall concentrate in this paper on two series of compounds:

- the Laves phase compounds with d_{U-U} values between 3.0 and 3.4 Å;
- uranium-platinum compounds: UPt, UPt_2, UPt_3 and UPt_5.

In this latter series the d_{U-U} values increase from 3.68 to 5.25 Å.

III. MAGNETIC PROPERTIES OF URANIUM LAVES PHASE COMPOUNDS

As can be expected from their position in the Hill plot, magnetic ordering is in general not observed in the Laves phase uranium compounds, although there are two exceptions: UFe_2 and UNi_2. In considering possibilities for itinerant-electron magnetism, the density of electron states at the Fermi level is the important parameter. Specific heat studies have been performed for several of the Laves phase compounds, see Table I. The coefficient γ of the

TABLE I

Values for the coefficient γ (in mJ/K^{2}mole f.u.) of the electronic specific heat for actinide metals, some intermetallic uranium Laves phase compounds and uranium-platinum compounds. Data for the pure elements from ref.4.

				ref.			ref.
Th	4.3	UMn_2	41	15	UPt	110	20,21
Pa	-	UFe_2	55	16	UPt_2	77	20,21
U	9.1	UCo_2	35*	16	UPt_3	420*	20,21
Np	12.4	UNi_2	65	17	UPt_5	85	20,21
Pu	17.0	URe_2	34	15			
Am	6.0	UIr_2	53	18			
		UAl_2	143*	19			

*enhanced by spin-fluctuation effects.

electronic term reaches values that are of the same order of magnitude as or larger than the corresponding value for ferromagnetic (YFe_2 [22]: 12.5- and $ZrFe_2$ [23]: 13 mJ/K^2mole) or nearly ferromagnetic (YCo_2) d-metal compounds of the same structure. For the strongly exchange-enhanced, Pd-like, paramagnetic compound YCo_2, a γ-value of 36 mJ/K^2mole has been reported[22], a value lower than most of the studied uranium compounds exhibit. Unless the effective electron-electron interaction is systematically smaller in these compounds than in the d metals, one would expect that the Stoner criterion for ferromagnetism could easily be passed in several of these compounds. Apart from large values for the density of state at E_F, the specific heat measurements on UAl_2 and UCo_2 reveal anomalies at low temperatures that can be represented by an additional term in the specific heat proportional to $T^3 \ln T/T_{SF}$, with T_{SF} the spin-fluctuation temperature. We return to these phenomena in section V. Photoemission studies on UAl_2 and UCo_2 [24] result in density of states schemes in which a narrow 5f band is placed near the Fermi energy. Due to a large hybridization of the uranium 5f states with the cobalt 3d states, the 5f intensity at E_F is expected to be weaker in UCo_2 than in UAl_2. These expectations have been confirmed by photoemission as well as by specific heat studies.

Ferromagnetism in UFe_2 is due to the ordering of iron magnetic moments as neutron experiments have shown. For UNi_2 different interpretations of its magnetic behaviour have been proposed[16,26], one of them[16] ascribing the magnetic order to defects in the atomic structure of this compound. Ferromagnetism in UFe_2 is no exception in so far that it is found in several Laves phase compounds XFe_2 with X a transition metal element, whereas it is lacking in general in the corresponding XMn_2, XCo_2 and XNi_2 series. Within the series YFe_2, $ZrFe_2$, UFe_2, the extent of hybridization of the iron d electrons with the valence electrons of Y, Zr and U increases in that sequence and results in an enhanced itinerant character of the iron magnetism in UFe_2.[27] Between UFe_2 and the other actinide Laves phase compounds $NpFe_2$, $PuFe_2$ and $AmFe_2$, a marked difference is observed, the itineracy of UFe_2 being significantly higher.[28] So, for the subject of itinerant-electron magnetism the compound UFe_2 is of special importance.

Extensive magnetic studies have been performed on UFe_2 [29] and some ferromagnetic iron-rich $U(Fe,Co)_2$ compounds.[16] The zero temperature properties, especially the relative pressure dependence of the spontaneous magnetic moment, follow the Stoner-Wohlfarth relations, see Fig.3. Aldred observed, besides spin wave terms, a quadratic term in the temperature dependence of the spontaneous magnetization of UFe_2, indicating the presence of Stoner excitations. By studying the effect of pressure on the critical phenomena near T_c, Franse et al.[16,21] concluded that the magnetic moments in UFe_2 are partly reduced by Stoner excitations at $T = T_c$. Similar conclusions were drawn for pure iron from thermal expansion studies by Shiga.[30] These results can be understood in a local-band type description of the 3d ferromagnetism.

A survey of the Laves phase compounds with Np, Pu and Am[4] reveals that magnetic order is a common feature of the Np compounds, whereas it is absent for the Pu and Am compounds with the exception of $PuFe_2$, $AmFe_2$ and $PuPt_2$. The other AmX_2 compounds are non-magnetic probably due to a $5f^6$, $J = 0$ ground state of Am.

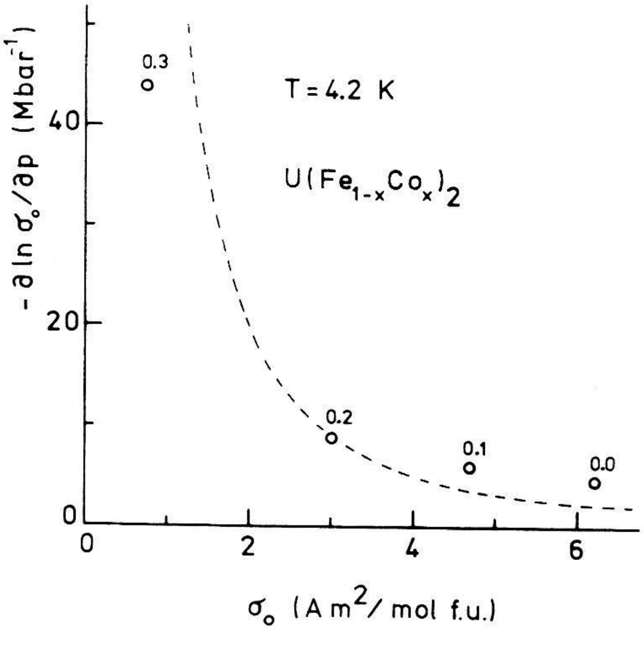

Fig.3. Relative pressure dependence of the spontaneous magnetization as a function of σ_o. The broken curve represents the Stoner-Wohlfarth model.

IV. MAGNETIC PROPERTIES OF URANIUM-PLATINUM COMPOUNDS

Magnetic order in this series of compounds has been reported for UPt, UPt_2 and UPt_3. A closer inspection of the magnetic data reveals that UPt_2 and UPt_3 are both paramagnetic. The divergence in the susceptibility of UPt_2 near 18 K, reported by Schneider and Laubschat[31], is most probably due to the presence of a small fraction of the UPt phase in their compound. The susceptibility of UPt exhibits a maximum around this temperature, indicating a change in magnetic structure. The maximum in the susceptibility of UPt_3 near 18 K that was considered as an indication of antiferromagnetic order, can equally well be explained by more sophisticated models. Schneider and Laubschat[32] performed XPS measurements on this series of compounds and deduced large contributions from 5f electron states to the density of states at the Fermi level. Specific heat measurements confirm these results. In UPt_3 the same type of anomaly in the specific heat is observed as for UAl_2, the enhanced γ-value being in this case as large as 420 mJ/K^2mole f.u. The absence of magnetic order in three of the four uranium-platinum compounds is rather puzzling. The d_{U-U}-values for UPt_2, UPt_3 and UPt_5 amount to 3.81, 4.12 and 5.25 Å, respectively, clearly at the upper side of the Hill limit where localized f-electron states can be developed. Hybridization with the 5d platinum states apparently prevents this localization. This hybridization leads to large values for the density of states at the Fermi level but does not imply the spontaneous splitting of the spin-up and spin-down subbands.

In discussing possibilities for itinerant-electron magnetism in this series of compounds we confine ourselves to the cases of UPt and UPt_3. Different interpretations of the electronic and magnetic phenomena in these compounds can be found in the literature. Assuming that the electronic specific heat coefficient of UPt is not appreciably different from that of ThPt, Luengo et al.[33] deduced the existence of spin waves below T_c and derived a value for the magnetic entropy of 0.62 R per mole by comparing the specific heat curves of UPt and ThPt below 30 K. This result is smaller by a factor of four than that expected for the possible 5f configurations $5f^3$ or $5f^2$, but is much larger than the values observed in general for very weak itinerant ferromagnets. Franse et al.[16], however, analyzed the specific heat of UPt below 10 K in electron, phonon and spin-wave terms and deduced a γ-value of 110 mJ/K^2mole f.u., much larger than the value of 3.5 mJ/K^2mole f.u., reported by Luengo et al. for ThPt. With this large γ-value the magnetic entropy, as determined by comparing the specific heats of UPt and ThPt, is at least one order of magnitude smaller than the value given above. Magnetization studies at 4.2 K yield values for the spontaneous magnetic moment per uranium atom between 0.40 and 0.45 μ_B. Preliminary results of neutron diffraction studies at the ILL in Grenoble[34] indicate a collinear magnetic structure for UPt at 4.2 K, with a magnetic moment per uranium site of $(0.78 \pm 0.10)\mu_B$, directed along the c-axis.

These results of magnetization and neutron diffraction studies probably mean that UPt is a strongly anisotropic uniaxial ferromagnetic material. Without preferred grain orientations one expects in that case a value for the bulk magnetic moment of $(0.4 \pm 0.05)\mu_B$ per uranium site, in agreement with the experimentally derived values. The neutron-diffraction value for the ordered magnetic moment is considerably smaller than the value of 3.5 μ_B for the effective moment derived from susceptibility measurements up to 1000 K. Ferromagnetism in UPt is suppressed under hydrostatic pressures. Magnetization curves up to 7 kbar also reveal a magnetic transition around 4 tesla. Thermal expansion, ac susceptibility and specific heat measurements at zero pressure clearly show an additional transition of magnetic origin near 19 K, well below the transition to the paramagnetic state at approximately 27 K.[35] The collinear magnetic structure apparently changes in a more complex magnetic structure either by pressurising the material at 4.2 K or by increasing the temperature above 19 K.

Summarizing the available information for UPt, we are in a dilemma at characterizing the type of magnetism in this compound. The strong magnetic anisotropy and the complex magnetic structure of UPt point to a more localized type of magnetism. The ratio of effective to spontaneous magnetic moments, the large electronic contribution to the specific heat, the small value for the magnetic entropy suggest an itinerant nature of the magnetic moment carriers. For the time being this dilemma remains unsolved.

Magnetization curves for UPt_3 along a and b directions differ from that in the c direction and show a type of magnetic transition that was originally believed to be caused by spin-flop phenomena[20], see Fig.4. Specific heat measurements on UPt_3, however, did not show any evidence for magnetic order below 30 K, but revealed a large, nearly field-independent, anomaly at the

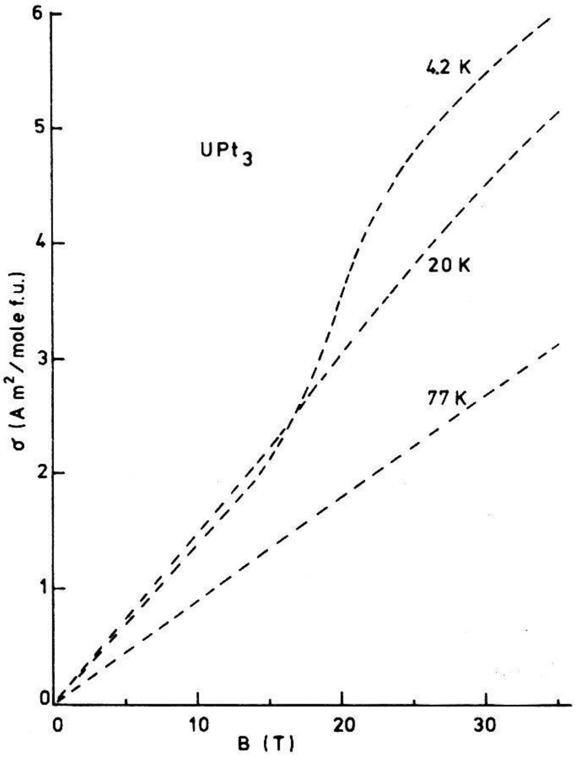

Fig.4. Magnetization curves along the a-direction for a single crystal of UPt_3.

lowest temperatures, leading to an extremely large γ-value. The observed behaviour could indicate the presence of spin fluctuations in the material. UPt_3 is under study now in neutron experiments.

V. SPIN FLUCTUATIONS IN INTERMETALLIC URANIUM COMPOUNDS

Exchange-enhanced paramagnetic transition metals often show anomalies at low-temperatures inproperties as the specific heat and the magnetic susceptibility. Examples are: Pd, YCo_2, $LuCo_2$, $TiBe_2$ etc. Magnetism in these materials is clearly of an itinerant nature. The anomalies either reflect fine details in the band structure, or point to many body effects, indicated as spin fluctuations. A survey of this latter subject has recently been presented by Béal-Monod.[36] Spin fluctuation phenomena are frequently observed in the actinide compounds as a consequence of the narrow 5f band of the actinide metals. Low-temperature anomalies in the resistivity, as they occur in Np and Pu and in some nearly magnetic compounds (UAl_4, UAl_3, UAl_2, UGa_3, UIn_3, UPt_3 etc.), in particular the T^2-dependence at the lowest temperature, are considered in general as a first indication. Specific heat, susceptibility and high-magnetic field experiments can provide further evidence. High-field magnetization measurements on some typical spin-fluctuation (UAl_2 and UCo_2) and valence-fluctuating (YbCuAl and YbCuAl-based quasiternary compounds) systems reveal common features.[37] For both systems the low-temperature magnetic isotherms exhibit abrupt changes in slope between 15 and 20 T, see Fig.5.Further, a correlation is found between the sign of the field dependence of the differential susceptibility and the sign of the temperature dependence of the initial susceptibility.[21] For the spin-fluctuation compounds the Zeeman energy connected with the field where the kink in the low-temperature isotherms occurs, is comparable with the thermal energy at T_{SF}, the characteristic temperature for spin fluctuations. Quantitatively, for

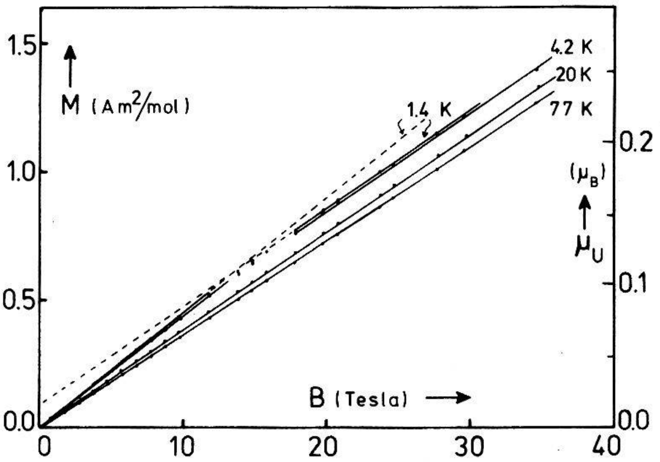

Fig.5. Magnetization curves of UAl_2.

UAl_2 it could be established that the differential susceptibility of the high-field part of the magnetization is equal to the initial susceptibility above T_{SF}. For UCo_2, spurious contributions to the susceptibility make quantitative conclusions impossible. A few central questions regarding spin- and valence-fluctuations in actinide compounds can now be formulated in the following way:

- are some of the low-temperature anomalies in spin- or valence-fluctuating materials zero temperature properties or are they induced at elevated temperature?
- does the similarity in the low-temperature properties of spin- and valence fluctuating compounds have a profound significance?

The first question arises from the above quoted study of the suppression of spin fluctuations in UAl_2 in high magnetic fields. Magnetization curves at 4.2 K and 1.4 K do not differ significantly and both show the typical reduction of the differential susceptibility above 15 T. This reduction is absent at temperatures above the spin fluctuation temperature of about 25 K.
The second point is put forward by similarities in the temperature and field dependence of the susceptibility in spin- and valence-fluctuating systems. Yet, our theoretical understanding of these two classes of materials is based on two essentially different models: atomic or localized magnetism in the case of valence fluctuations, band magnetism in that of the spin fluctuation system. It must not be excluded that these distinct approaches come together in the actinides and that a unified model might exist which properly describes both classes of materials. This paper is especially concerned with problems of itinerant-electron magnetism in intermetallic compounds of uranium with transition elements. The occurrence of magnetic order was discussed in detail for the Laves phase uranium compounds and for the uranium-platinum compounds. Most compounds belonging to these two classes have been studied in specific heat experiments and large values for the coefficient of the electronic term is a rule. Since magnetic order in perfectly ordered compounds only occurs for UFe_2 and UPt, one might expect to find spin fluctuation effects in the other compounds. Among the Laves phase compounds this is the case indeed for UMn_2[38], UCo_2 and UAl_2. In UNi_2, these effects are depressed by the internal magnetic field produced by a small number of local magnetic moments. Anomalies in the specific heat of uranium-platinum compounds are very pronounced for UPt_3, although UPt_2 and UPt_5 are also abnormal. Whether these deviations are indicative for spin fluctuation phenomena is not yeat clear. Further experimental work is in progress.

VI. SUMMARY

Magnetic properties of the Laves phase uranium compounds can be understood with concepts of magnetism that have been developed for the transition metals. UFe_2 is a good example of the model for itinerant-electron ferromagnetism, although it must be realized that magnetism is mainly carried by the iron atoms in this compound. Spin-fluctuation phenomena that are characteristic for transition metals at the borderline of paramagnetism and magnetic order, have been reported for UAl_2, UCo_2 and UMn_2. The predominance of itinerant-electron magnetism in the Laves phase compounds is a consequence of the close distance between uranium atoms by which the overlap of the 5f wave functions is ensured. In uranium-platinum compounds with considerably larger distances between the uranium atoms, localization of the 5f electrons is impeded by hybridization of uranium 5f- and platinum 5d states. This hybridization, nevertheless, leads to large values for the density of states at the Fermi level as XPS- and specific heat experiments have shown. Why these large density of states values do not imply a fullfilment of the Stoner criterion for UPt_2, UPt_3 and UPt_5 remains an unsolved question. Spin fluctuation effects in these compounds can not be excluded.

Of special importance for the subject of itinerant-electron magnetism in 5f systems are actinide compounds with a non-magnetic element as the second constituent and with nearest neighbour distances between the actinide atoms close to the critical value that is known as the Hill limit. Two representatives of this class of compounds are UPt and UAl_2. In both compounds elements of itinerant-electron magnetism have been observed. The complex magnetic structures and the strong magnetic anisotropy of UPt, together with the similarities in the physical behaviour of spin- and valence-fluctuation systems prevent us, however, from characterizing these materials as pure examples of the model of itinerant-electron magnetism.

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