MAGNETIC AND ELECTRONIC PROPERTIES OF NEPTUNIUM AND PLUTONIUM COMPOUNDS

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Abstract. – The magnetic and electrical transport properties of the U, Np and Pu monopnictides and monochalcogenides are reviewed. The results of single crystal measurements on Np and Pu compounds are emphasized. Measurements on a single crystal of $NpRu_2Si_2$ are summarized.

The actinides form a transition serie characterized by the progressive filling of the 5f shell. They differentiate from the lanthanides, except the anomalous ones as Ce, by a more extended nature of the 5f wave function. Depending on the actinide element, the interactinide spacing and the actinide environnement in compounds, properties ranging from itinerant (3d like) to localized systems can occur. The properties of the actinides have been recently reviewed in the "Handbook on the Physics and Chemistry of the Actinides" which gives an exhaustive description of the studies performed up to 1984 concerning bulk properties [1], Mössbauer spectroscopy [2], elastic [3] and inelastic neutron scattering [4] to which the reader can refer for bibliography. It is clear that a large number of uranium compounds have been extensively studied for a long time but much less has been done on neptunium and plutonium compounds and the studies on transplutoniums are rather seldom. This is obviously a consequence of the high activity of Np and Pu and transplutonium elements the latter furthermore being avalaible only in very small quantities. A big effort has been devoted during the last ten years, to the sample preparation and crystal growth of Np and Pu compounds [5]. One of the most important progress, due to the joint efforts of Vogt at ETH Zurich and of Spirlet and Rebizant at EITU Karlsruhe, consists in the synthesis of large, good quality, single crystals of rocksalt Np and Pu compounds which allowed to perform many physical studies on these systems.

In this paper we will describe the properties of the structurally simple NaCl type monopnictides and monochalcogenides AnX with special attention to the transuranium Np and Pu compounds. The results obtained on $NpRu_2Si_2$ will be presented to illustrate the interest of the ternary actinide compounds AnM_2X_2 (isomorphous with the Ce counterparts). All these results are derived from magnetic measurements (susceptibility and magnetization), Mössbauer spectroscopy, electrical transport measurements and neutron scattering experiments. The scoop of this paper is limited and we apologize that many interesting studies on actinides are not covered.

Uranium monopnictides and monochalcogenides

The magnetic properties of the AnX monopnictides and monochalcogenides are summarized in table I.

All the uranium monopnictides are antiferromagnets with Neel temperature and ordered magnetic moments increasing from the nitride to the bismuthide, i.e. with interuranium spacing. The magnetic ordering of all these compounds is characterized by wave vectors along the cubic axes $\mathbf{k} = \langle k\ 0\ 0 \rangle$ and a longitudinal polarization corresponding to Fourier components $\mathbf{m(k)}$ parallel to $\mathbf{k}$. A similar situation occurs also in the cerium monopnictides CeBi and CeSb [6]. In these last compounds as in the U counterparts it is now well established that this is a consequence of highly anisotropic exchange interactions due to the proximity of the 4f (or 5f) electrons to the Fermi energy and to the hybridization of the f electrons with band electrons [7]. In some cases this anisotropy of $\mathbf{k}$ and $\mathbf{m(k)}$ disagrees with magnetization measurements which indicate another easy axis ([110], or [111]). This discrepancy is explained by the occurence of planar double-k or multiaxial triple-k structures instead of the collinear single-k ordering. All the uranium monochalcogenides are ferromagnets with magnetization along the [111] direction. From US to UTe the Curie temperature decreases while the ordered magnetic moments increase. Solid solutions of monopnictides and monochalcogenides can be easily formed as, for instance, $USb_{1-x}Te_x$ [3], or $UAs_{1-x}Se_x$ [8] in which a progressive evolution from the antiferromagnetic behaviour of the pnictide to the ferromagnetism of the chalcogenide can be studied. As regards to their electrical transport properties all the UX compounds are metallics. The evolution of the electrical resistivity, from US to UTe, indicates a progressive lo-

Table I. — *Magnetic properties of* U, Np, *and* Pu *monopnictides and monochalcogenides.*

	a (Å)	T_N (K)	T_c (K)	k	ordering	Easy axis	$m(T=0)$ μ_B	Ref.
UN	4.890	53		⟨001⟩	$1-k$ type I	⟨001⟩	0.75	1, 3
UP	5.589	122		⟨001⟩	$1-k$ type I	⟨001⟩	1.7	1, 3
		22		⟨001⟩	$2-k$ type I	⟨110⟩	1.9	1, 3
UAs	5.779	124		⟨001⟩	$1-k$ type I	⟨001⟩	1.9	1, 3
		62		⟨0 0 1/2⟩	$2-k$ type IA	⟨110⟩	2.2	1, 3
USb	6.191	213		⟨001⟩	$3-k$ type I	⟨111⟩	2.85	1, 3
NpN	4.897		87	0	ferro	⟨111⟩	1.4	13
NpP	5.615	130		⟨0 0 0.36⟩	Inc	⟨001⟩		13
		74		⟨0 0 1/3⟩	$+ + + - - -$	⟨001⟩	2.2	
NpAs	5.838	173		⟨0 0 1/4 $-\varepsilon$⟩	$1-k$, Inc	⟨001⟩		13, 15
		154		⟨0 0 1/4⟩	$4+, 4-, 1-k$	⟨001⟩		16
		138		⟨001⟩	$3-k$, type I	⟨111⟩	2.5	
NpSb	6.254	202		⟨001⟩	$3-k$, type I	⟨111⟩	2.5	13, 14
PuN	4.905	13?						20, 21
PuP	5.550		126	0	ferro	⟨100⟩	0.75	22
PuAs	5.780		125	0	ferro	⟨100⟩	0.67	23
PuSb	6.240	85		⟨0 0 0.13⟩	$1-k$, Inc	⟨100⟩		24
		70		0	ferro	⟨100⟩	0.74	
PuBi	6.350	58		⟨0 0 0.23⟩	$1-k$, Comm	⟨100⟩	0.50	27
US	5.489		170	0	ferro	⟨111⟩	1.70	1, 3
USe	5.750		160	0	ferro	⟨111⟩	2.0	1, 3
UTe	6.155		104	0	ferro	⟨111⟩	2.25	1, 3
NpS	5.527	23		⟨1/2 1/2 1/2⟩	Type II		0.9	17
NpSe	5.804	41		?	?		1.35	18, 19
NpTe	6.198	30		?	?		$\simeq 2.$	18, 19
PuS	5.536	Modified CW		behaviour,	$T_N = 4.5?$			28
PuSe	5.775	TIP						29
PuTe	6.151	TIP						30, 31

calization of the 5f electrons with a pronounced Kondo anomaly in UTe [9]. In the case of the monopnictides the resistivity above T_N is nearly constant. This is mainly due to spin fluctuation scattering and, in contrast with the monochalcogenides no evidence for a Kondo-type behaviour is observed. In the ordered state the behaviour of the electrical resistivity is rather complicated, reflecting the complexity of the magnetic phase diagram of these compounds [9]. In particular it has been found through measurements under high magnetic fields [10] or uniaxial stress [11] that the magnetic contribution to the electrical resistivity of UP and UAs is highly anisotropic and this can be related to the anisotropy of their magnetic properties. In the case of USb the triple-k magnetic ordering leads to a gap opening in the Fermi surface from which results a very large maximum in $\rho(T)$ below T_N [9, 12].

Np and Pu monopnictides and monochalcogenides

The neptunium nitride is a ferromagnet, NpP and NpAs order at $T_N = 130$ and 173 K respectively in incommensurate phases, still characterized by longitudinal waves with propagation vectors along the cubic axes and exhibit low temperature lock-in transition towards commensurate structures [13]. NpSb is a simple antiferromagnet with a triple-k type I structure at any temperature below $T_N = 202$ K [14]. The case of NpAs is particularly illustrative. A tetragonal distortion is associated with the ordering transition at T_N but this distortion vanishes at $T_0 = 138$ K, where a transition takes place from long period (4+, 4−) ordering toward antiferromagnetic type I ordering, with an apparent cubic symmetry [13], figure 1. Moreover the mag-
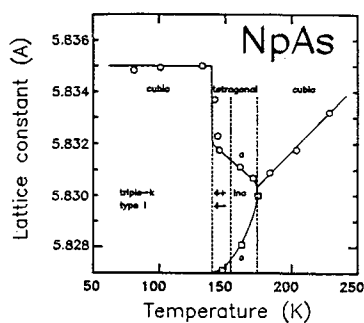
Fig. 1. – Cell parameters *versus* temperature in NpAs from reference [13].

netization measurements, on single crystals indicate several metamagnetic transitions toward ferrimagnetic and ferromagnetic phases [15], figure 2. These transitions are strongly dependent on temperature and magnetic field direction yielding to different phase diagrams for field applied along the [001], [110] or [111] directions. Neutron diffraction experiments on NpAs single crystal [16] have established that, in zero field, an incommensurate, sinusoidally modulated phase of wave vector $(0, 0, 1/4 - \varepsilon)$ develops from $T_N = 173$ K down to a lock-in transition at $T_{IC} = 154$ K where a commensurate $(4+, 4-)$ phase $(\mathbf{k} = (0, 0, 1/4))$ takes place. The development below T_{IC} only of third order harmonics indicates a squaring of the moment modulation. At $T_0 = 138$ K another transition leads to the type I antiferromagnetic phase $(\mathbf{k} = (0, 0, 1))$. Whatever the temperature the three symmetry equivalent k-vectors are observed in zero field but in cooling under a small field $H = 6$ kOe $/\!/$ [110] only the wave vector perpendicular to $\mathbf{H}$ is observed in the two high temperature phases confirming the collinear nature of these phases evidenced by the tetragonal distortion of the unit cell. At T_0 the three equivalent Fourier components develop with equal amplitude establishing the triple-k nature of the low temperature type I phase and
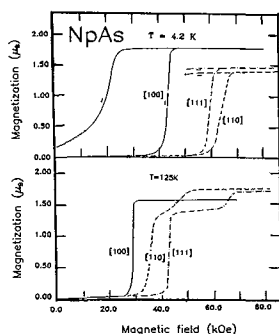
thus explaining the reentrance to the cubic symmetry. In larger field parallel to [1$\bar{1}$0], figure 3, the incommensurate and $(4+, 4-)$ phases are relatively unstable with respect to a ferromagnetic state. Just above the critical field the magnetization aligns along a cubic axis and rotates progressively towards the field direction $(\mathbf{m} /\!/ \mathbf{H})$. The antiferromagnetic triple-k type I phase also transforms into this ferromagnetic phase only in a small temperature range, just below T_0, but becomes much more stable at low temperature. Below 100 K no change occurs until a critical field of about 60 kOe where a ferrimagnetic state is induced. This state is characterized by an antiferromagnetic component perpendicular to $\mathbf{H}$ and a ferromagnetic ones, along a cubic axis at 45° from $\mathbf{H}$. This anisotropy of the ferromagnetic component is large, indeed the magnetization measured at 4.2 K in a field of 80 kOe along the [110] direction is only $1/\sqrt{2}$ the value of the ferromagnetic component deduced from neutron diffraction and moreover the critical fields in the [100] (40 kOe) and [110] (60 kOe) directions are in about the same ratio. To explain this magnetic properties of NpAs one must consider highly anisotropic exchange interactions from which results the quenching of the wave vector and of the Fourier components along the cubic axis. The value of $|k|$ depends essentially on the relative strength of the interactions between first (J_1) and second neighbours (J_2) and in NpAs the minimum of $\mathbf{J}(\mathbf{k})$ arises for $k = 1/4 - \varepsilon$ and an incommensurate ordering develops at T_N. On cooling the saturation of the magnetic moment is responsible for the lock-in transition to the commensurate k value 1/4. To account for the significant tetragonal distortion associated with these collinear phases large magnetoelastic terms must be considered. To drive the transition from a single-k to a triple-k structure higher order terms than bilinear ones must be important, the transition occuring when these fourth or higher order terms overcome the magnetoelastic energy. The subsequent turn back to cubic symmetry modifies the interplane distances, and then the corresponding exchange interactions, allowing for the change of the k value. Finally the very large anisotropy of the ferromagnetic component in the low temperature ferrimagnetic phase induced by the field is more difficult to interpret. It can have its origin
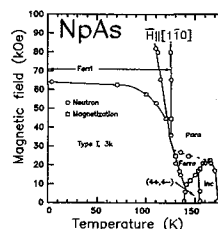

Fig. 2. – Magnetization at $T = 4.2$ and 125 K *versus* magnetic field along the [100], [110], and [111] directions of a single crystal of NpAs.

Fig. 3. – Magnetic phase diagram (H, T) of NpAs in a field along the [110] direction.

in an anisotropy of the interactions ($J (k = 0)$ being minimum for $\mathbf{m}(\mathbf{k} = 0)$ $/\!/$ $\langle 100 \rangle$) because it is difficult to imagine another way to compensate the loss of magnetization energy when $\mathbf{H}$ is along [110]. The neptunium monochalcogenides are antiferromagnets. Up to now only NpS has been studied [17], it exhibits a type II ($\mathbf{k} = (1/2\ 1/2\ 1/2)$) antiferromagnetic ordering fundamentally different from that observed in the U monopnictides. Very recently Mössbauer experiments [18] and susceptibility measurements have shown that NpSe and NpTe order below T_N = 41 and 30 K respectively with a complex magnetic ordering still to be determined by neutron diffraction.

The nitride PuN exhibits a modified Curie-Weiss behaviour, with an anomaly in the sysceptibility at about 13 K [19], but no magnetic ordering has been detected at low temperature [20]. PuP [21] and PuAs [22, 23] are simple, strongly anisotropic ferromagnets. PuSb [23] orders in an incommensurate phase, again characterized by wave vector [0 0 k] and $\mathbf{m}(\mathbf{k})$ $/\!/$ $\mathbf{k}$. k varies continuously from $k = 0.130$ at T_N down to $k = 0.090$ at $T_{IC} = 70$ K where a ferromagnetic ordering takes place. The high temperature incommensurate phase is rather unstable with respect to an applied field. In moderate field ($H \leq 12$ kOe) a commensurate ordering with a wave vector $k = 1/8$ is stabilized in a finite temperature range and larger fields induce a ferromagnetic state (Fig. 4). PuSb has also been investigated by critical neutron scattering [24] and inelastic neutron scattering [25]. These studies evidence the strong anisotropy of the exchange interactions, very similar to that observed in many Ce and uranium isomorphous compounds. PuBi develops at any temperature below $T_N = 58$ K a long period incommensurate ordering [26]. The results relative to Pu monochalcogenides PuS [27] and PuSe [28] are limited. The telluride PuTe has been studied by magnetization measurement [29] and polarized neutron diffraction [30]. The susceptibility is very small, nearly constant with temperature in contrast with the expected behaviour for a Pu^{3+} ion suggested by the value of the lattice parameter $(a = 6.15\ \text{Å})$. The polarized neutron experiments confirm that only a very small magnetic moment ($5 \times 10^{-3}\ \mu_B/Pu$) can be field induced. Taking into account the behaviour of PuSb and PuTe, the investigation of the solid solutions $PuSb_{1-x}Te_x$ is particularly interesting. An anomaly occurs in the variation of
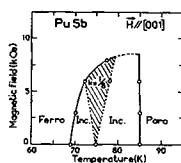
the lattice constant which exhibits a maximum around $x = 0.05$ [31]. Very small Te substitution (2 %) affects significantly the magnetic behaviour of PuSb. For larger tellurium concentrations the magnetic susceptibility can be interpreted by a modified Curie-Weiss law. On increasing x the effective moment and the paramagnetic Curie temperature decrease while the temperature independent susceptibility χ_0 increases almost linearly with a tendancy to saturate at the value observed in PuTe (Fig. 5). Mössbauer experiments [32] also indicate the loss of the localized magnetic moment on going from PuSb to PuTe.
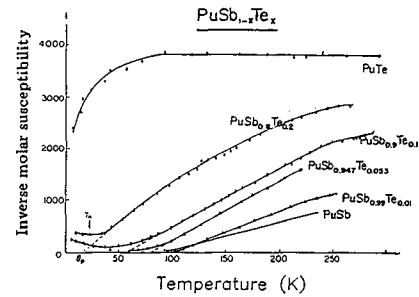
Fig. 5. – Inverse susceptibiity of solid solutions $PuSb_{1-x}Te_x$ from Reference [29].

The study of the electrical properties of transuranium AnX compounds, using single crystals, is only beginning. The first measurements on a NpX compounds, namely NpTe is presented at this conference [33]. The resistivity of NpTe exibits a logaritmic dependence at high temperature ($T \geq 50$ K), the ordering temperature is detected at $T = 30$ K and a large residual resistivity is observed at low temperature. Thus NpTe seems to behave like a heavy fermion system which develops a magnetic ordering at low temperature.

The plutonium compounds PuSe, PuTe and PuSb as well as $PuSb_{1-x}Te_x$ solid solutions have been investigated [34] (Fig. 6). The chalcogenides PuSe, and PuTe behave as semiconductors with very small gap (≤ 20 meV). PuSb is a semi-metal displaying Kondo effect. The $PuSb_{1-x}Te_x$ solid solutions evidence the development of a small hybridization gap as x increases
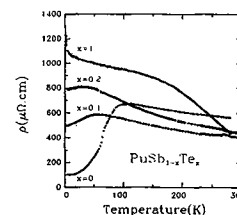
Fig. 4. – Magnetic phase diagram (H, T) of PuSb in a field along the [100] direction.

Fig. 6. – Resistivity *versus* temperature for solid solution $PuSb_{1-x}Te_x$.

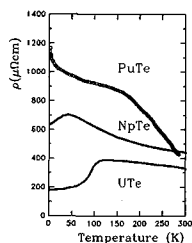(Fig. 6). The same trend is observed in the tellurides on going from UTe to PuTe through NpTe (Fig. 7).

Fig. 7. – Resistivity *versus* temperature of single crystal of UTe, NpTe, PuTe.

Actinides compounds with the $ThCr_2Si_2$ type structure

Since the discovery of heavy fermion behaviour and superconductivity in $CeCu_2Si_2$ [35] the tetragonal compounds with the $ThCr_2Si_2$ type structure especially those of cerium have been the object of many extensive studies. Ternary actinide compounds AnM_2X_2 where M = Ru, Rh and X = Ge or Si can be formed. Among them URu_2Si_2 is especially interesting [36-38], the low temperature specific heat coefficient $\gamma = 180$ mJ/mole. K^2 is rather large. At $T_N = 17.5$ K URu_2Si_2 orders with a antiferromagnetic structure (k = (001)) but with an extremely low moment value (0.05 μ_B/U). Furthermore a superconducting transition occurs at $T = 1.1$ K. Therefore the study of the Np isomorphous is quite interesting. The results of Mössbauer spectroscopy and neutron diffraction experiments reported at this conference [39] show that $NpRu_2Si_2$ orders at $T_N = 27.5$ K in a sinusoidally incommensurate modulated phase. The wave vector of the modulation, along the tetragonal axis, varies continuously with temperature and no lock-in transition has been detected down to 1.5 K. On cooling some squaring of the modulation develops but even at 1.5 K ($T_N/20$) the magnetic moments are not fully saturated. Preliminary magnetic and resistivity measurements have been performed, on a small single crystal ($\simeq 0.8$ mg) [40]. At high temperature the magnetic susceptibility follows a modified Curie-Weiss la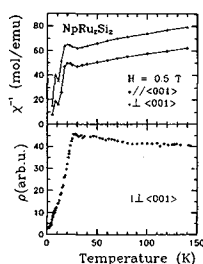w. The very small weight of the crystal prevents an accurate determination of X_0 ($\simeq 10^{-2}$ emu/mole) and of the effective moment ($\mu_{eff} \simeq 4$ μ_B). At low temperature the Neel temperature is detected at $T_N = 29 \pm 1$ K. Below T_N an anomaly is observed at $T = 6$ K both in magnetization and electrical resistivity curves (Fig. 8) which has not been detected in neutron and Mössbauer measurements.

Conclusion

Presently many informations about the magnetic behaviour of U, Np and Pu monopnictides have been obtained. Except for the nitrides in which the interactinide distance is small and just below the Hill limit for occurence of localized magnetism all the other compounds are characterized by highly anisotropic exchange interactions resulting from their electronic structure allowing large hybridization effects. Beside this common feature many different properties are observed ranging from ferromagnetic to antiferromagnetic behaviour through incommensurate and long period ordering, from collinear to planar (double-k) and multiaxial (triple-k) magnetic arrangements. This is a consequence of the details of the magnetic interactions along the direction of the propagation vector and of the competition between exchange anisotropy, single ion anistropy (CEF) and magneto-elastic term. A theoretical study of these systems has been developed by Cooper *et al.* [41] very consistent with the present experimental results.

The monochalcogenides exibit also rather complex properties ranging from ferromagnetism in uranium compounds to complex orderings and to weak magnetism in neptunium and plutonium compounds. Clearly more detailed studies are needed in order to understand their exciting properties.

The recent results on the ternary compound $NpRu_2Si_2$ indicate that the physical properties of intermallic transuranium systems are quite interesting to be investigated in a near future.

Acknowledgements

Fig. 8. – Inverse susceptibility and resistivity of a single crystal of $NpRu_2Si_2$.

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