

## Delocalization of the U 5f magnetic moments in $U(\text{In}_{0.6}\text{Sn}_{0.4})_3$ and UNiSn under high pressure

This article has been downloaded from IOPscience. Please scroll down to see the full text article.

2005 J. Phys.: Condens. Matter 17 S859

(<http://iopscience.iop.org/0953-8984/17/11/015>)

View [the table of contents for this issue](#), or go to the [journal homepage](#) for more

Download details:

IP Address: 129.252.86.83

The article was downloaded on 27/05/2010 at 20:31

Please note that [terms and conditions apply](#).

## Delocalization of the U 5f magnetic moments in $U(\text{In}_{0.6}\text{Sn}_{0.4})_3$ and UNiSn under high pressure

A Barla<sup>1,2</sup>, J-P Sanchez<sup>1</sup>, A Aksungur<sup>3</sup>, R Lengsdorf<sup>3</sup>, J Plessel<sup>3</sup>,  
B P Doyle<sup>2,4</sup>, R Ruffer<sup>2</sup> and M M Abd-Elmeguid<sup>3</sup>

<sup>1</sup> Département de Recherche Fondamentale sur la Matière Condensée, CEA Grenoble, 17 rue des Martyrs, F-38054 Grenoble Cedex 9, France

<sup>2</sup> European Synchrotron Radiation Facility, 6 rue Jules Horowitz, F-38043 Grenoble Cedex 9, France

<sup>3</sup> II Physikalisches Institut, Universität zu Köln, Zùlpicher Strasse 77, D-50937 Köln, Germany

<sup>4</sup> Laboratorio TASC-INFN, Area Science Park, Basovizza SS 14 Km 163.5, I-34012 Trieste, Italy

E-mail: jsanchez@cea.fr

Received 5 January 2005

Published 4 March 2005

Online at [stacks.iop.org/JPhysCM/17/S859](http://stacks.iop.org/JPhysCM/17/S859)

### Abstract

The pressure–temperature dependence of the electronic and magnetic properties of the compounds  $U(\text{In}_{1-x}\text{Sn}_x)_3$  and UNiSn has been investigated by means of high pressure x-ray diffraction, <sup>119</sup>Sn nuclear forward scattering (NFS) of synchrotron radiation and Mössbauer spectroscopy (MS) measurements. We show that pressure has different effects on these systems: while  $U(\text{In}_{0.8}\text{Sn}_{0.2})_3$  shows some of the typical properties of nearly localized 5f systems, pressure induces the delocalization of the U 5f electrons in  $U(\text{In}_{0.6}\text{Sn}_{0.4})_3$  and UNiSn with a pressure dependent interplay between RKKY exchange interaction and the consequent collapse of the magnetic order. These results are discussed in terms of hybridization between U 5f electrons and conduction band electrons such as U 6d, Sn/In 5sp and Ni 3d electrons.

### 1. Introduction

The fascinating variety of ground state properties of strongly correlated electron systems with 5f electrons has been the subject of intensive experimental and theoretical efforts during the last decades [1]. A central issue is the question of the nature of the 5f electrons which behave, depending on the compound investigated, in more localized-like or itinerant-like ways. There is now growing evidence that in actinides localized and delocalized 5f electrons may coexist [2, 3]. This view, suggested by a large set of experiments, recently received strong theoretical support [4]. In this context, special interest was focused on systems close to a magnetic instability because of the observation of unconventional superconductivity at the quantum critical point where magnetic order vanishes [5]. In general, the electronic structure and magnetism of actinide compounds are intimately linked to the hybridization of the 5f

orbitals either by direct 5f–5f interaction (interactinide distance below the Hill limit) or by overlap with the conduction band electrons such as U 6d and ligand sp or d electrons. An elegant way to tune the properties of a material is to reduce its interatomic distances and thus to increase the hybridization by applying an external pressure. Pressure experiments have recently led to a number of outstanding results—for instance the pressure-induced enhancement of the tiny uranium moment in URu<sub>2</sub>Si<sub>2</sub> [6] and the appearance of superconductivity within the ferromagnetic phase of UGe<sub>2</sub> [5] and UIr [7]. In the present paper we discuss our recent studies on the magnetic and electronic properties of selected 5f compounds, U(In<sub>1-x</sub>Sn<sub>x</sub>)<sub>3</sub> and UNiSn, which exhibit different responses to external pressure due to the different degrees of hybridization between the U 5f electrons and ligand spd electrons.

### 1.1. The systems investigated

U(In<sub>1-x</sub>Sn<sub>x</sub>)<sub>3</sub> belongs to the series of UX<sub>3</sub> compounds (where X = Al, Ga, In, Tl, Si, Ge, Sn or Pb). They crystallize in the AuCu<sub>3</sub> structure and show a broad variety of magnetic properties (Pauli paramagnetism, spin fluctuations, antiferromagnetism). This spectrum of properties can be well described using the model of Koelling *et al* [8], where the f-ligand hybridization is considered to increase as the size of the ligand decreases or when going from group IIIA to group IVA of the periodic table, with a progressive transition from a more localized to a more itinerant state. This effect of hybridization can be particularly well studied in the family of compounds U(In<sub>1-x</sub>Sn<sub>x</sub>)<sub>3</sub>, because the lattice parameter shows almost no change across the whole series. The phase diagram as determined by Zhou *et al* [9] evidences the presence of antiferromagnetism for  $x \leq 0.4$ , with a Néel temperature that decreases from 108 K for UIn<sub>3</sub> to 35 K for U(In<sub>0.6</sub>Sn<sub>0.4</sub>)<sub>3</sub>. For larger Sn concentrations a spin fluctuating state is reached (USn<sub>3</sub>) after crossing a region of the phase diagram where large values of Sommerfeld's coefficient of the specific heat are observed ( $\gamma_{\max} = 530 \text{ mJ mol}^{-1} \text{ K}^{-2}$  for  $x = 0.6$ ).

UNiSn belongs to the class of so-called Kondo insulators or narrow gap semiconductors and has been extensively studied during the last two decades owing to its exceptional electronic and magnetic properties [10]. It crystallizes in a cubic structure (MgAgAs-type) and undergoes a first-order transition from a paramagnetic (P) semiconductor (S) to an antiferromagnetic (AF) metal (M) at  $T_N \sim 43 \text{ K}$  [11]. Its magnetic structure determined by means of neutron diffraction (see figure 1) was found to be of type I with ferromagnetic (001) planes stacked along the [001] axis in the sequence + - + - [12]. The ordered U moment oriented along the [001] axis amounts to  $\sim 1.55 \mu_B$ . Furthermore, it was shown that the S–M transition is accompanied by a concomitant tetragonal distortion and a ferroquadrupolar (Q) order at  $T_N \approx T_{SM} \approx T_Q$  [13]. In order to shed more light on these peculiar multiple phase transitions in UNiSn, magnetic field–temperature ( $H, T$ ) as well as pressure–temperature ( $p, T$ ) phase diagrams have been investigated.  $T_N$  is nearly field independent at least up to 15 T, while  $T_Q$ , identical to  $T_{SM}$ , raises slightly with  $H$  above 4 T ( $dT_Q/dH \sim 0.15 \text{ K T}^{-1}$ ) [13]. On the other hand, high pressure resistivity  $\rho(p, T)$  measurements up to 8 GPa [14–16] and very recently up to 20 GPa [17] show that the temperature-induced S–M transition is suppressed at  $p \geq 9 \text{ GPa}$ , resulting in a pressure-induced metallic state. Further details of the  $\rho(p, T)$  data on UNiSn will be published elsewhere [17].

Here we present high pressure results on U(In<sub>1-x</sub>Sn<sub>x</sub>)<sub>3</sub> and UNiSn obtained using combined macroscopic (x-ray diffraction) and microscopic (<sup>119</sup>Sn nuclear forward scattering (NFS) of synchrotron radiation and Mössbauer spectroscopy (MS)) techniques in a diamond anvil cell (DAC). These methods allowed us to determine the volume dependence of the electronic and magnetic properties of these compounds from the pressure variation of the Néel temperature and the transferred magnetic hyperfine field at the <sup>119</sup>Sn nuclei. We interpret the

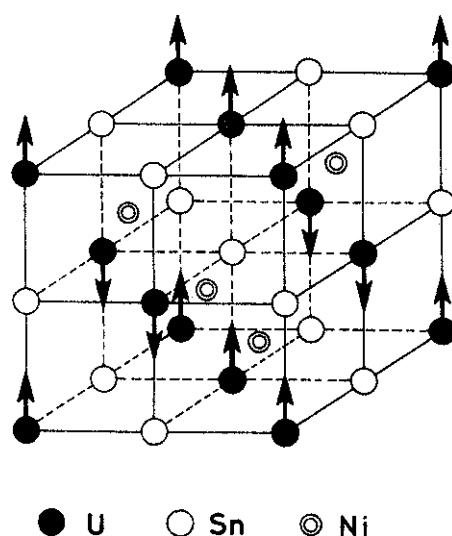


Figure 1. Crystallographic and magnetic structures of UNiSn taken from [12].

results in terms of the competition between the indirect Ruderman–Kittel–Kasuya–Yosida (RKKY) exchange interaction and the hybridization between the U 5f electrons and the electrons of the outer shells of the ligands (Sn, In and Ni). A more detailed description of some of these results can be found in [18, 17].

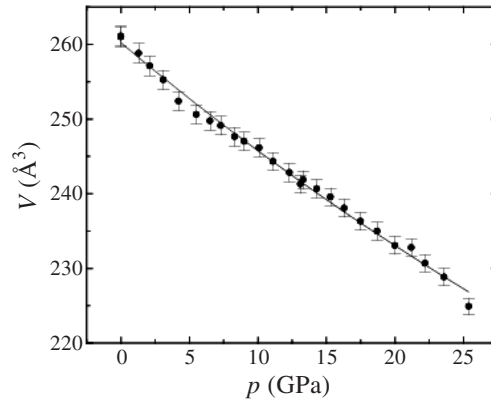
## 2. Experimental details

Polycrystalline samples of  $U(\text{In}_{1-x}\text{Sn}_x)_3$  with  $x = 0.2$  and  $0.4$  were prepared and characterized as described in [18]. Polycrystalline samples of UNiSn were prepared by arc melting of the elemental constituents and subsequent annealing following methods described in [14]. For the  $^{119}\text{Sn}$  NFS experiments the sample was isotopically enriched to 90% in  $^{119}\text{Sn}$ .

High pressure was applied to the samples using either modified Merrill–Basset [19] or piston–cylinder [20] DACs. The pressure was determined by the ruby fluorescence method at room temperature. In order to reduce the risk of dispersion of uranium dust in case of breakage of the pressure cell, the samples were mixed with epoxy which also acted as the pressure transmitting medium. The  $^{119}\text{Sn}$  MS measurements were performed at the University of Cologne and at the CEA Grenoble, while the  $^{119}\text{Sn}$  NFS experiments were carried out at the undulator beamline ID18 [21] of the European Synchrotron Radiation Facility (ESRF) in Grenoble. A more detailed description of the ESRF experimental set-up can be found in [18]. Note that the NFS technique is related to the Mössbauer effect; thus similar microscopic information to that inferred from conventional Mössbauer spectroscopy can be obtained. The x-ray diffraction measurements, carried out in order to determine the pressure–volume relationship, were performed at the energy dispersive x-ray diffraction beamline F3 at HASYLAB in Hamburg.

## 3. Results and discussion

Before the presentation and discussion of the pressure dependences of the magnetic ordering temperature  $T_N$  and of the transferred magnetic hyperfine field  $B_{\text{thf}}$ , we stress that energy dispersive x-ray diffraction patterns recorded at 300 K up to 30 GPa ( $U(\text{In}_{0.8}\text{Sn}_{0.2})_3$ ) and

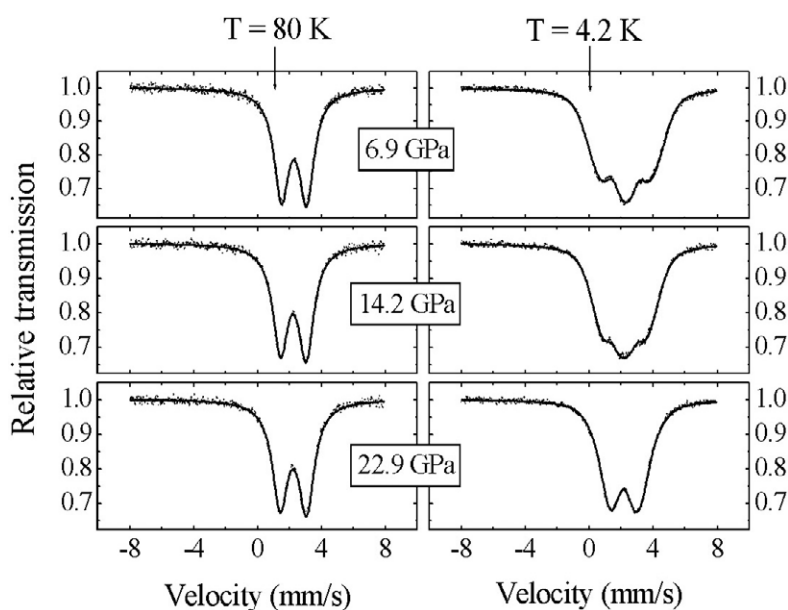


**Figure 2.** The volume–pressure relationship for UNiSn at room temperature. The full circles are the measured points while the curve is a fit with the Murnaghan equation of states.

25 GPa (UNiSn) indicate that both compounds retain their ambient pressure structures over the whole pressure range investigated. Figure 2 shows the pressure variation of the unit cell volume of UNiSn, where it is evident that there are no anomalies in the compression curve. The dependence can therefore be well accounted for with a Murnaghan equation of states, yielding a value for the ambient pressure bulk modulus of  $B_0 = 168(10)$  GPa for UNiSn and  $B_0 = 93(2)$  GPa for  $U(\text{In}_{0.8}\text{Sn}_{0.2})_3$ . As also both  $\text{UIn}_3$  and  $\text{USn}_3$  are stable in this pressure range and have comparable bulk moduli [22], in the following we will assume that  $U(\text{In}_{0.6}\text{Sn}_{0.4})_3$  is stable as well and we will use the same bulk modulus as for  $U(\text{In}_{0.8}\text{Sn}_{0.2})_3$ .

### 3.1. $^{119}\text{Sn}$ MS and NFS measurements

**3.1.1.  $U(\text{In}_{1-x}\text{Sn}_x)_3$ .** NFS measurements have been performed up to pressures of 15 and 25 GPa on the samples with  $x = 0.2$  and  $0.4$ , respectively. For  $U(\text{In}_{0.6}\text{Sn}_{0.4})_3$  the NFS study has been complemented with MS studies in the pressure range between 0 and 23 GPa. These studies have been carried out on the same sample as the NFS ones and some selected measured spectra are shown in figure 3 together with their least squares fits obtained with the package CONUSS [23]. Both MS and NFS spectra can be fitted with this program, which uses the full dynamical theory of nuclear resonance scattering including the diagonalization of the hyperfine Hamiltonian. In the paramagnetic state, the spectra are characteristic of a quadrupole split doublet, which originates from the presence of an axially symmetric electric field gradient (EFG) at the Sn nuclei, due to the tetragonal point symmetry  $4/mmm$  of the lattice positions occupied by the Sn atoms. The quadrupole interaction parameter ( $\Delta E_Q = \frac{1}{2}eQV_{zz}$ , where  $Q$  is the nuclear quadrupole moment of the  $I = 3/2$  excited state and  $V_{zz}$  is the principal component of the electric field gradient) has a value of  $1.53(1)$  mm s $^{-1}$  for  $x = 0.2$  and  $1.50(1)$  mm s $^{-1}$  for  $x = 0.4$  at  $T = 125$  K and ambient pressure and increases with increasing pressure. For each pressure  $p$  in the range reached by this study, below the ordering temperature  $T_N(p)$ , the spectra show the presence of a distribution of transferred magnetic hyperfine fields, with average  $\overline{B_{\text{thf}}}(p)$ , at the Sn nuclei, combined with the quadrupole interaction. The origin of  $B_{\text{thf}}$  at the Sn nucleus is twofold: an indirect polarization of the conduction electrons by localized 5f electrons mediated by the RKKY interaction is combined with the direct polarization of the outer 5sp electrons of the Sn atoms caused by their overlap with the U 5f electrons [24]. At ambient pressure one has  $\overline{B_{\text{thf}}}(0) = 3.0(1)$  T for  $x = 0.2$  and  $2.3(1)$  T for  $x = 0.4$  at 4 K.

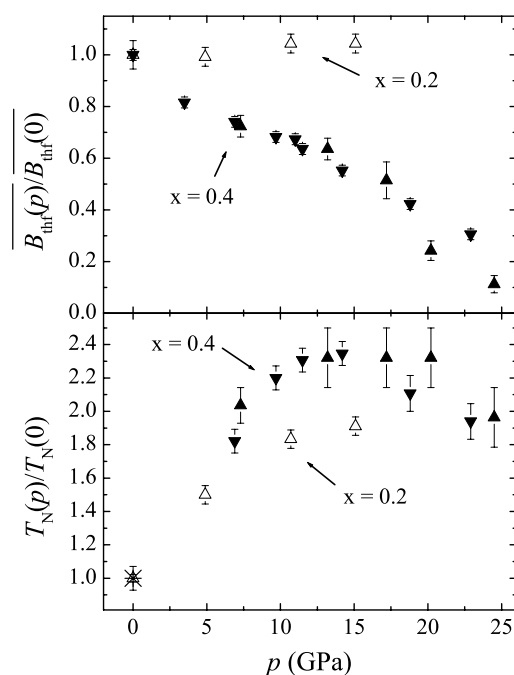


**Figure 3.**  $^{119}\text{Sn}$  Mössbauer spectra of  $\text{U}(\text{In}_{0.6}\text{Sn}_{0.4})_3$  at different temperatures and pressures up to 23 GPa. The dots represent experimental data points, while the curves are least squares fits.

A more detailed discussion of the model used to analyse the spectra below  $T_N$  may be found in [18]. All spectra at  $p > 0$  have been analysed following the same procedure as was used for those measured at ambient pressure, with the additional assumption that the magnetic structure is not changed by pressure.

The pressure dependence of  $\overline{B_{\text{thf}}}$ , measured at low temperature  $T \leq 5.6$  K, and  $T_N$  (both normalized to their ambient pressure values) for the two compounds is shown in figure 4. For  $x = 0.2$ , the average magnetic hyperfine field shows only a very weak dependence on pressure (up to  $p = 15$  GPa, corresponding to a volume contraction of the unit cell of  $\sim 12\%$ ). Under the assumption that the orientation of the U moments does not change with pressure, this implies that the magnitude of the U moment does not change appreciably as the volume contracts. The Néel temperature shows a monotonic increase with pressure, from 90(2) K at ambient pressure to 172(5) K at the highest pressure of 15 GPa. On the other hand, for  $\text{U}(\text{In}_{0.6}\text{Sn}_{0.4})_3$  the average magnetic hyperfine field decreases monotonically with increasing pressure and reaches a value corresponding to  $\sim 10\%$  of the ambient pressure one at 25 GPa (corresponding to a volume contraction of the unit cell of  $\sim 16.5\%$ ). This indicates a large reduction of the U ordered moment to a value below  $\sim 0.1 \mu_B$ , as compared to an ambient pressure value, which can be estimated as  $\sim 0.8 \mu_B$  by comparison with that of  $\text{UIn}_3$ . The Néel temperature first increases with pressure, reaching its maximum value of 65(2) K at  $p \approx 14$  GPa and then starts to decrease for  $p > 14$  GPa. However, its value at 25 GPa, 55(5) K, is still high even if the value of the ordered magnetic moment ( $\sim 0.1 \mu_B$ ) is strongly reduced.

**3.1.2.  $\text{UNiSn}$ .** The  $^{119}\text{Sn}$  Mössbauer spectra recorded at ambient pressure in the 4.2–300 K temperature range agree well with those published in the literature [11, 25]. At 4.2 K, i.e., in the magnetically ordered state, the spectrum consists of a superposition of a magnetically split sextet (with  $B_{\text{thf}} = 7.15$  T, vanishing quadrupolar interaction and isomer shift  $\Delta S \approx 1.8 \text{ mm s}^{-1}$  with respect to the  $\text{CaSnO}_3$  source) and a single line (about 15% of

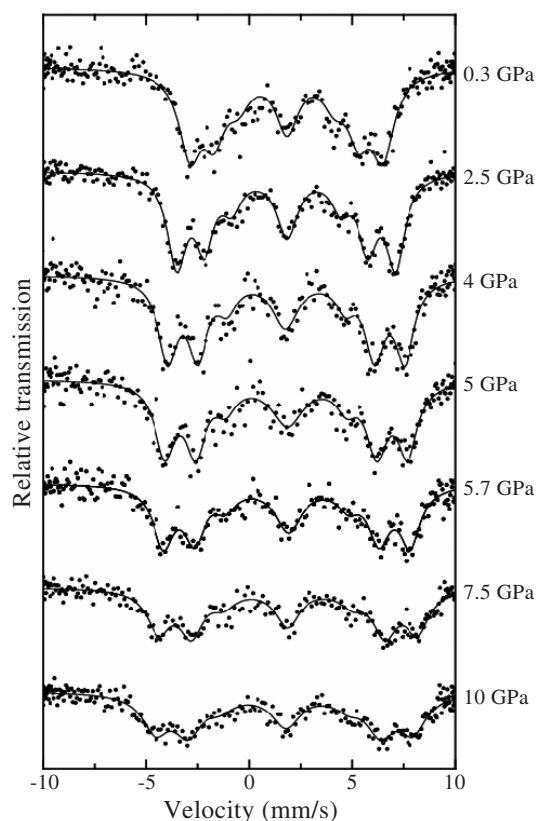


**Figure 4.** The pressure dependence of the normalized average transferred hyperfine field  $B_{\text{thf}}(p)/B_{\text{thf}}(0)$  (measured at low temperature  $T \leq 5.6$  K) and Néel temperature  $T_N(p)/T_N(0)$  for  $\text{U}(\text{In}_{0.8}\text{Sn}_{0.2})_3$  and  $\text{U}(\text{In}_{0.6}\text{Sn}_{0.4})_3$ . The open symbols refer to  $x = 0.2$ , while the full symbols refer to  $x = 0.4$ . The downward pointing triangles refer to the results of the MS measurements while the upward pointing triangles indicate the results of the NFS measurements. The value of  $T_N$  at ambient pressure indicated by an asterisk is determined by the magnetic susceptibility for  $x = 0.4$ . The figure is reproduced from [18].

the spectral area). The observation of a sextet is expected owing to the fact that each Sn atom, according to the known magnetic structure, is surrounded by six U nearest neighbours with four spins up and two spins down [12]. The nonmagnetic contribution (single line), whose intensity was previously shown to be dependent on the annealing conditions [11], is tentatively ascribed to Sn atoms occupying the Ni site. At temperatures above  $T_N \approx 43$  K a single absorption line is observed.

Figure 5 shows  $^{119}\text{Sn}$  Mössbauer spectra recorded at 4.2 K at various pressures up to 10 GPa and figure 6 presents some selected  $^{119}\text{Sn}$  NFS patterns obtained at various temperatures (200 and 3 K) and pressures up to 21.4 GPa. At high temperatures (left panel in figure 6,  $T = 200$  K) and for all pressures the NFS spectra are characteristic of unsplit nuclear levels, as expected for Sn atoms in the absence of magnetic order and in an environment of cubic symmetry. Magnetically split sextets are observed in all Mössbauer spectra recorded at pressures up to 10 GPa (see figure 5), while clear quantum beat patterns, due to the action of a magnetic interaction on the nuclear levels of  $^{119}\text{Sn}$ , appear at low temperatures (right panel in figure 6,  $T = 3$  K) in the NFS spectra for pressures less than 18.5 GPa. The vanishing  $B_{\text{thf}}$  seen above 18.5 GPa is nicely illustrated by the similarity of the NFS spectral shapes at 200 and 3 K for  $p = 21.4$  GPa.

The pressure dependence of the transferred magnetic hyperfine field  $B_{\text{thf}}$  is illustrated in figure 7. In a first approximation  $B_{\text{thf}}$  is proportional to:



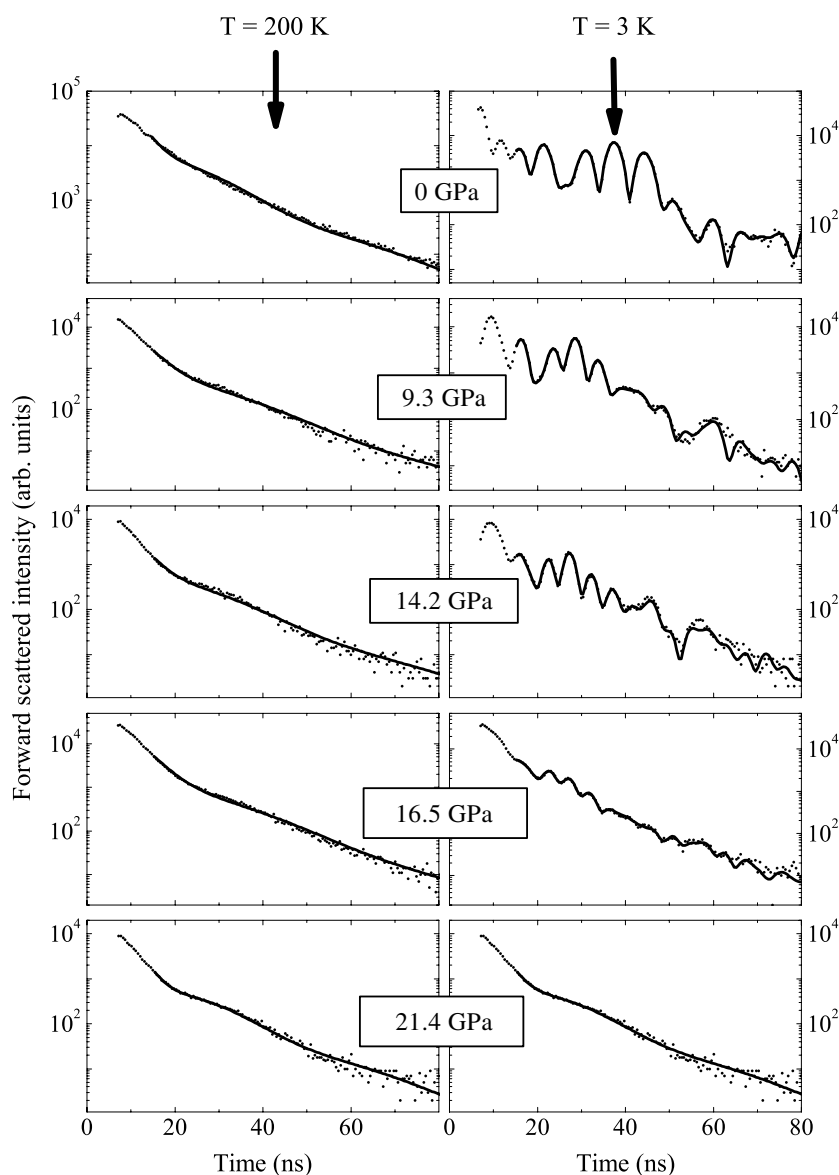
**Figure 5.**  $^{119}\text{Sn}$  Mössbauer spectra of UNiSn at  $T = 4.2$  K for different pressures up to 10 GPa. The dots represent experimental data points, while the curves are least squares fits.

- (i) a hyperfine coupling constant  $A$  which depends on the electronic structure of the material,
- (ii) the magnitude of the U magnetic moment,
- (iii) the weighted vector sum,  $\sum \vec{\mu}/\mu$  of the U magnetic moments in the immediate vicinity of the Sn atom ( $\sum \vec{\mu}/\mu = 2$  for the case of a type I antiferromagnetic structure as found for UNiSn at  $p = 0$  (see figure 1)).

$B_{\text{thf}}$  increases almost linearly from 7.15 T to 9.4 T at  $p = 0$  and 7.5 GPa, respectively. A less steep increase of  $B_{\text{thf}}$  is then observed up to 12.7 GPa where  $B_{\text{thf}} = 9.7$  T. When pressure is increased further,  $B_{\text{thf}}$  remains almost constant up to 16.5 GPa. Above that pressure it starts to drop dramatically and at 18.9 GPa ( $1 - V(p)/V(0) \sim 10\%$ )  $B_{\text{thf}}$  was found to vanish to zero.

Figure 7 illustrates the pressure dependence of the Néel temperature as determined from the temperature variation of the NFS spectra (and the analysis of the resistance  $R(T, p)$  data as published in [17]).  $T_N(p)$  first increases with pressure ( $dT_N/dp \sim 4.8$  K GPa $^{-1}$ ), in agreement with previous high pressure resistivity measurements [14–16], reaches a maximum value of about 95 K at  $p \approx 13$  GPa and then rapidly decreases for  $p \geq 14$  GPa. At 18.7 GPa no magnetic transition was found down to the lowest measuring temperatures (3 and 1.8 K for NFS and  $R(T, p)$  measurements, respectively).

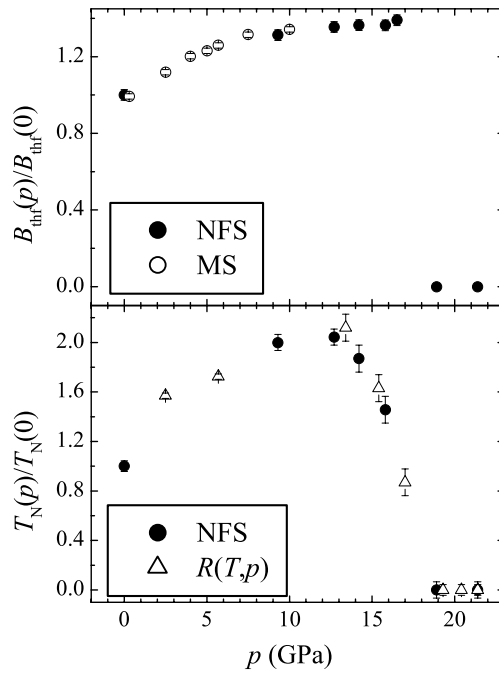




**Figure 6.**  $^{119}\text{Sn}$  NFS spectra of UNiSn at  $T = 200$  and 3 K for some selected pressures. The dots represent experimental data points, while the curves are least squares fits.

### 3.2. Pressure-induced collapse of the magnetic states in $U(\text{In}_{0.6}\text{Sn}_{0.4})_3$ and UNiSn

The pressure-induced changes of the magnetic state (magnetic moment/hyperfine field and ordering temperature) of f electron systems give valuable information about the mechanisms underlying the delocalization of these electrons. Doniach proposed several years ago [26] that the properties of the ground state of a Kondo lattice system depend on the competition between the magnetic ordering of local moments through the RKKY interaction and the weakening or destruction of the local moment by the Kondo spin compensation. The two characteristic



**Figure 7.** The dependence on pressure of the normalized transferred hyperfine field  $B_{\text{thf}}(p)/B_{\text{thf}}(0)$  (measured at low temperature  $T \leq 4.2$  K) and Néel temperature  $T_N(p)/T_N(0)$  for UNiSn. The full circles refer to the results of the NFS measurements, the open circles indicate the results of the MS measurements and the open triangles are the results of the resistance measurements described in [17].

energy scales given by the Kondo temperature ( $T_K$ ) and the RKKY interaction ( $T_{\text{RKKY}}$ ) are both related to the product  $|JN(E_F)|$  where  $J$  is the strength of the exchange coupling between the local moments and the conduction electrons and  $N(E_F)$  the density of states at the Fermi level:  $T_K \propto \exp[-1/|JN(E_F)|]$  and  $T_{\text{RKKY}} \propto J^2N(E_F)$ . For small values of  $|JN(E_F)|$ , the RKKY interaction prevails and the compound can sustain magnetic order. As  $|JN(E_F)|$  increases, the Kondo compensation of the local moments becomes dominant and suppresses the development of long range magnetic order. The resulting phase diagram [26, 27] shows an increase in the magnetic ordering temperature with  $|JN(E_F)|$ , followed by a much faster decrease when  $|JN(E_F)|$  exceeds a critical value. This simple model has been applied with great success to Ce (Yb) Kondo lattices where pressure was shown to increase (decrease)  $|JN(E_F)|$  [28–32]. Although less documented,  $|JN(E_F)|$  was also found to increase with pressure in uranium compounds [33]. However, due to the greater spatial extent of the 5f wavefunctions, one expects hybridization to play a very important role in determining the ground state properties of light actinide compounds. In this case, the demagnetization process is driven by the transition from a local to an itinerant (band-like) state rather than by the Kondo effect and this is reflected by a generally different dependence of the magnetic properties of a given compound on pressure. According to Sheng and Cooper [34], the decrease of the interatomic distances caused by pressure induces the 5f wavefunctions to diffuse more outside the core region. This in turn enhances the 5f-ligand hybridization and causes a gradual wash-out of the U moment and the consequent suppression of ordered magnetism. On the other hand, the increased overlap between the 5f and ligand orbitals enhances the exchange integrals

and this can cause a strengthening of the magnetic order and thus an increase of the ordering temperature. Although initially this latter mechanism may prevail, the moment reduction is always predominant at higher pressures. This model has been successfully applied to describe the pressure dependence of the ordering temperature of U monochalcogenides [30, 34, 35] and UPtAl and UNiAl [36]. The same model could explain the pressure dependence of  $T_N$  in UGa<sub>3</sub> [37] and in UPb<sub>3</sub> [38].

The results of our measurements as shown in figures 4 and 7 can be interpreted in terms of the models mentioned above. The very weak dependence of the average transferred hyperfine field on pressure suggests that the moment is localized in U(In<sub>0.8</sub>Sn<sub>0.2</sub>)<sub>3</sub>. However, the Néel temperature increases with pressure linearly and the slope appears to decrease at the highest pressure reached of 15 GPa. This compound should therefore be regarded as a weakly delocalized system. In contrast, U(In<sub>0.6</sub>Sn<sub>0.4</sub>)<sub>3</sub> shows a monotonic decrease of the average transferred hyperfine field as pressure is increased. This suggests that the ordered magnetic moment is strongly delocalized as the U–U and U–Sn/In distances decrease. However, the initial increase with pressure of  $T_N$  indicates a strong increase of the exchange coupling constant  $J$ , which tends to stabilize the magnetically ordered state. A similar initial increase with pressure of the ordering temperature is observed for UNiSn as well, at least up to  $p \sim 12$  GPa. Here, however, such an increase is associated with a corresponding increase of  $B_{\text{thf}}$ , which can be ascribed to the strengthening of the hyperfine coupling constant rather than to an increase of the U moment, which is expected to remain constant as long as it may be considered (nearly) localized. Only above 12 GPa does the delocalization start to be visible in the pressure dependence of the ordering temperature, which starts to decrease with increasing pressure, and above 18.5 GPa we observe the complete collapse of the transferred hyperfine field. On the other hand, we do not observe for any of the compounds studied any anomaly in the  $T_N$  and  $B_{\text{thf}}$  pressure dependences which could be attributed to a sudden change of the magnetic structure. Thus, one can conclude that for pressures below a critical value ( $\sim 14$  GPa for U(In<sub>0.6</sub>Sn<sub>0.4</sub>)<sub>3</sub> and  $\sim 12$  GPa for UNiSn) the RKKY exchange interaction prevails over the mechanisms which tend to weaken or destroy the magnetic order, whereas at higher pressures the latter dominate. We suggest that increasing hybridization with increasing pressure is the cause of the delocalization of the 5f moments because the Kondo interaction would be expected to produce a much faster decrease of the ordering temperature [26, 30]. One can therefore conclude that it is the 5f-ligand (spd) hybridization, as a consequence of the increasing 5f bandwidth with increasing pressure, that drives U(In<sub>0.6</sub>Sn<sub>0.4</sub>)<sub>3</sub> and UNiSn from magnetic to nonmagnetic states.

#### 4. Conclusions

In conclusion, using high pressure x-ray diffraction, <sup>119</sup>Sn nuclear forward scattering and Mössbauer spectroscopy we were able to determine the volume dependence of the transferred magnetic hyperfine field and of the Néel temperature of U(In<sub>0.8</sub>Sn<sub>0.2</sub>)<sub>3</sub>, U(In<sub>0.6</sub>Sn<sub>0.4</sub>)<sub>3</sub> and UNiSn. We showed that pressure has a different effect on the various compounds owing to the different degrees of localization of the U 5f electrons. While in U(In<sub>0.8</sub>Sn<sub>0.2</sub>)<sub>3</sub> the U 5f moments appear to be nearly localized, with nearly no pressure dependence of the transferred hyperfine field and an increase of the ordering temperature up to 15 GPa, the other two compounds show a clear pressure-induced collapse of the magnetic order. For both U(In<sub>0.6</sub>Sn<sub>0.4</sub>)<sub>3</sub> and UNiSn,  $T_N$  shows an initial increase with pressure followed by a gradual decrease. However, while in U(In<sub>0.6</sub>Sn<sub>0.4</sub>)<sub>3</sub> the transferred hyperfine field decreases monotonically over the whole pressure range studied, thus indicating a gradual decrease of the U 5f moment, in UNiSn the U ordered moments appear to be more stable to external pressure and the collapse of the transferred

hyperfine field occurs rapidly at a pressure of about 18.5 GPa. It is shown that these different types of behaviour can be well understood by assuming a volume dependent competition between RKKY interaction and hybridization between U 5f electrons and conduction band electrons.

### Acknowledgments

The authors are grateful to D Kaczorowski for providing the  $U(\text{In}_{1-x}\text{Sn}_x)_3$  samples and to T Takabatake for providing the UNiSn samples.

### References

- [1] See for example  
Gschneidner K A *et al* (ed) 1993 *Handbook on the Physics and Chemistry of Rare Earths* vol 17 (Amsterdam: Elsevier Science)  
Gschneidner K A *et al* (ed) 1994 *Handbook on the Physics and Chemistry of Rare Earths* vol 19 (Amsterdam: North-Holland)
- [2] Bernhoeft N, Sato N, Roessli B, Aso N, Hiess A, Lander G H, Endoh Y and Komatsubara T 1998 *Phys. Rev. Lett.* **81** 4244
- [3] Kumigashira H, Ito T, Ashihara A, Hyeong-Do K, Aoki H, Suzuki T, Yamagami H, Takahashi T and Ochiai A 2000 *Phys. Rev. B* **61** 15707
- [4] Zwicknagl G and Fulde P 2003 *J. Phys.: Condens. Matter* **15** S1911
- [5] Saxena S S, Agarwal P, Ahilan K, Grosche F M, Haselwimmer R K W, Steiner M J, Pugh E, Walker I R, Julian S R, Monthoux P, Lonzarich G G, Huxley A, Sheikin I, Braithwaite D and Flouquet J 2000 *Nature* **406** 587
- [6] Amitsuka H, Sato M, Metoki N, Yokoyama M, Kuwahara K, Sakakibara T, Morimoto H, Kawarazaki S, Miyako Y and Mydosh J A 1999 *Phys. Rev. Lett.* **83** 5114
- [7] Akazawa T, Hidaka H, Fujiwara T, Kobayashi T C, Yamamoto E, Haga Y, Settai R and Onuki Y 2004 *J. Phys.: Condens. Matter* **16** L29
- [8] Koelling D D, Dunlap B D and Crabtree G W 1985 *Phys. Rev. B* **31** 4966
- [9] Zhou L W, Lin C L, Crow J E, Bloom S, Guertin R P and Foner S 1986 *Phys. Rev. B* **34** 483
- [10] See for example Sechovsky V and Havela L 1998 *Handbook of Magnetic Materials* vol 11 (Amsterdam: Elsevier Science) p 1
- [11] Akazawa T, Suzuki T, Nakamura F, Fujita T, Takabatake T and Fujii H 1996 *J. Phys. Soc. Japan* **65** 3661
- [12] Kawanaka H, Fujii H, Nishi M, Takabatake T, Motoya K, Uwatoko Y and Ito Y 1989 *J. Phys. Soc. Japan* **58** 3481
- [13] Akazawa T, Suzuki T, Goshima H, Tahara T, Fujita T, Takabatake T and Fujii H 1998 *J. Phys. Soc. Japan* **67** 3256
- [14] Fujii H, Kawanaka H, Takabatake T, Kurisu M, Yamaguchi Y, Sakurai J, Fujiwara H, Fujita T and Oguro I 1989 *J. Phys. Soc. Japan* **58** 2495
- [15] Kurisu M, Kawanaka H, Takabatake T and Fujii H 1991 *J. Phys. Soc. Japan* **60** 3792
- [16] Akazawa T, Suzuki T, Tahara T, Goto T, Hori J, Goshima H, Nakamura F, Fujita T, Takabatake T and Fujii H 1999 *Physica B* **259–261** 248
- [17] Barla A, Sanchez J P, Aksungur A, Lengsdorf R, Plessel J, Doyle B P, Ruffer R, Takabatake T and Abd-Elmeguid M M 2005 *Phys. Rev. B* **71** at press
- [18] Barla A, Sanchez J P, Ni B, Doyle B P, Vulliet P, Leupold O, Ruffer R, Kaczorowski D, Plessel J and Abd-Elmeguid M M 2002 *Phys. Rev. B* **66** 094425
- [19] Sterer E, Pasternak M and Taylor R D 1990 *Rev. Sci. Instrum.* **61** 1117
- [20] Machavariani G Yu, Pasternak M P, Hearne G R and Rozenberg G Kh 1998 *Rev. Sci. Instrum.* **69** 1423
- [21] Ruffer R and Chumakov A I 1996 *Hyperfine Interact.* **97/98** 589
- [22] Le Bihan T, Heathman S, Darracq S, Abraham C, Winand J-M and Benedict U 1996 *High Temp. High Pressures* **27/28** 157
- [23] Sturhahn W 2000 *Hyperfine Interact.* **125** 149
- [24] Sanchez J P, Spirlet J C, Rebizant J and Vogt O 1987 *J. Magn. Magn. Mater.* **63/64** 139
- [25] Bykovetz N, Herman W N, Yuen T, Chan-Soo J, Lin C L and Crow J E 1988 *J. Appl. Phys.* **63** 4127
- [26] Doniach S 1977 *Physica B* **91** 231

- 
- [27] Thompson J D and Lawrence J L 1994 *Handbook on the Physics and Chemistry of Rare Earths* vol 19, ed K A Gschneidner Jr *et al* (Amsterdam: North-Holland) p 383
- [28] Cornelius A L and Schilling J S 1994 *Phys. Rev. B* **49** 3955
- [29] Cornelius A L, Gangopadhyay A K, Schilling J S and Assmus W 1997 *Phys. Rev. B* **55** 14109
- [30] Cornelius A L, Schilling J S, Mandrus D and Thompson J D 1995 *Phys. Rev. B* **52** R15699
- [31] Winkelmann H, Abd-Elmeguid M M, Micklitz H, Sanchez J P, Geibel C and Steglich F 1998 *Phys. Rev. Lett.* **81** 4947
- [32] Winkelmann H, Abd-Elmeguid M M, Micklitz H, Sanchez J P, Vulliet P, Alami-Yadri K and Jaccard D 1999 *Phys. Rev. B* **60** 3324
- [33] de Novion C H, Konczykowski M and Haessler M 1982 *J. Phys. C: Solid State Phys.* **15** 1251
- [34] Sheng Q G and Cooper B R 1995 *J. Appl. Phys.* **75** 7035
- [35] Link P, Benedict U, Wittig J and Wühl H 1992 *J. Phys.: Condens. Matter* **4** 5585
- [36] Sechovsky V, Honda F, Prokes K, Syshchenko O, Andreev A V and Kamarad J 2003 *Acta Phys. Pol. B* **22** 159
- [37] Nakashima M, Haga Y, Honda F, Eto T, Oomi G, Kagayama T, Takeshita N, Nakanishi T, Mori N, Aoki D, Settai R and Onuki Y 2001 *J. Phys.: Condens. Matter* **13** L569
- [38] Haga Y, Yamamoto E, Onuki Y, Nakashima M, Aoki D, Onuki Y, Hedo M and Uwatoko Y 2003 *Acta Phys. Pol. B* **34** 1239