

Effect of pressure on the magnetic properties of $U(\text{In}_{1-x}\text{Sn}_x)_3$: Moment suppression in $U(\text{In}_{0.6}\text{Sn}_{0.4})_3$

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The volume dependence of the magnetic properties of $U(\text{In}_{1-x}\text{Sn}_x)_3$, with $x=0.2$ and 0.4 , has been studied using ^{119}Sn nuclear forward scattering of synchrotron radiation and Mössbauer spectroscopy at high pressures up to 25 GPa. The results show that in $U(\text{In}_{0.8}\text{Sn}_{0.2})_3$ the $5f$ magnetic moment is almost localized. Pressure induces an increase of the Néel temperature, while the transferred magnetic hyperfine field shows no change. Conversely, in $U(\text{In}_{0.6}\text{Sn}_{0.4})_3$ the transferred field decreases monotonically with increasing pressure and the Néel temperature goes through a maximum, showing a clear delocalization of the $5f$ electrons. This is discussed in terms of $5f$ -ligand hybridization and appears to lead to the formation of a high-pressure state characterized by strong dynamical spin correlations.

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I. INTRODUCTION

Strongly correlated electron systems with $4f$ electrons (e.g., Ce and Yb) and $5f$ electrons (e.g., U) are the subject of continuous experimental and theoretical efforts. The main point of interest in these systems is that their ground state critically depends on the competition between two interactions: the indirect Ruderman-Kittel-Kasuya-Yosida (RKKY) exchange interaction between the local magnetic moments via the conduction electrons and the direct Kondo exchange interaction resulting in a compensation of the magnetic moment.¹ This competition leads to an extraordinary broad spectrum of ground states ranging from magnetic ordering of local moments, band magnetism of heavy electrons and non-magnetic states, to spin fluctuations and even unconventional superconductivity.²

In the case of light actinide systems, due to the larger spatial extent of the $5f$ orbitals, hybridization also plays a very important role. According to a model proposed by Hill,³ the ground-state magnetic properties of actinide compounds are determined by the direct interaction between the $5f$ electrons of neighboring actinide atoms, so that the distance between the actinides in the lattice is the controlling parameter: when the actinide-actinide distance is below a critical value of 3.4–3.6 Å, known as Hill's limit, the overlap of the $5f$ wave functions of neighboring actinides is so large that magnetic ordering is prevented by the delocalization of the $5f$ electrons (itinerant compounds). In the opposite case, where the distance between neighboring actinides is larger than Hill's limit, magnetic ordering occurs (local moment compounds). However, this simple classification does not apply to a number of U systems, among which are the UX_3 compounds, where X is an element of groups IIIA (Al, Ga, In, Tl) or IVA (Si, Ge, Sn, Pb) of the periodic table. They all crys-

tallize in the cubic AuCu_3 crystal structure. Although the lattice constants are, for all types of X atoms, considerably larger than Hill's limit, these compounds show a broad spectrum of magnetic properties, ranging from Pauli paramagnetism [USi_3 (Ref. 4) and UGe_3 (Ref. 5)] to spin fluctuations [USn_3 (Ref. 6) and UAl_3 (Ref. 7)] and antiferromagnetism [UPb_3 (Ref. 8), UGa_3 (Ref. 9), UIn_3 and UTl_3 (Ref. 10)]. Koelling and co-workers¹¹ have pointed out the importance of the hybridization of the U $5f$ electrons with the s , p , and d electrons of the ligands X . In their model, the f -ligand hybridization decreases as the size of the ligand increases (when moving down within a column of the periodic system), explaining the presence of long-range magnetic order only in the compounds of the heaviest X atoms. Moreover, this type of hybridization increases when going from group IIIA to group IVA and the transition from a more localized to a more itinerant state can proceed through a heavy-fermion state.

Within the family of UX_3 compounds, the series $U(\text{In}_{1-x}\text{Sn}_x)_3$ is particularly interesting, because the substitution of Sn for In changes the lattice parameter by only $\sim 0.2\%$ between UIn_3 and USn_3 .¹² This allows one to study the effects of the change of the electronic structure of the X atom on the strength of the f -ligand hybridization, without changes in the volume of the unit cell. The magnetic phase diagram of $U(\text{In}_{1-x}\text{Sn}_x)_3$, determined experimentally by Zhou *et al.*,¹² shows the presence of antiferromagnetic order in the In-rich side, with a Néel temperature decreasing from $T_N \approx 108$ K for UIn_3 to $T_N \approx 35$ K for $U(\text{In}_{0.6}\text{Sn}_{0.4})_3$. For larger Sn concentrations, no long-range magnetic order is observed: USn_3 is a spin fluctuator and for $0.45 \leq x \leq 0.8$ large values of Sommerfeld's coefficient are observed [$\gamma_{\text{max}} = 530$ mJ/(mol K²) for $x = 0.6$].

In contrast to chemical substitution, the application of an

external pressure to compounds of the series $U(\text{In}_{1-x}\text{Sn}_x)_3$ offers the possibility to study the dependence of the magnetic properties of a given compound on changes of the volume of the unit cell, without modifying the electronic structure of the ligand. This allows one to verify how the magnetic ground state, corresponding to a given value of f -ligand hybridization (determined by the relative concentration of Sn in the sample), reacts to a volume change. Using ^{119}Sn nuclear forward scattering (NFS) of synchrotron radiation and ^{119}Sn Mössbauer spectroscopy (MS) in a diamond-anvil cell (DAC), we have determined the volume dependence of the magnetic properties of two selected samples of the series $U(\text{In}_{1-x}\text{Sn}_x)_3$, with $x=0.2$ and $x=0.4$. In this paper we discuss the effect of pressure on the Néel temperature and the transferred magnetic hyperfine field of the two compounds and interpret the results in terms of the competition between the indirect exchange RKKY interaction and the hybridization between the U $5f$ electrons and the electrons of the outer shells of the ligands (In and Sn).

II. EXPERIMENT

Samples of $U(\text{In}_{1-x}\text{Sn}_x)_3$, with $x=0.2$ and 0.4 , were prepared by arc melting the elemental constituents under titanium-gettered argon atmosphere and subsequent annealing of the melted buttons in vacuum at 600°C for two weeks. In order to increase the count rates in the ^{119}Sn NFS and MS experiments at high pressure the samples were enriched to 90% in ^{119}Sn . The quality of the samples was checked by x-ray powder diffraction. The alloys were found to be single phase with the expected cubic AuCu_3 -type crystal structure and the refined lattice parameters were in agreement with the literature data.¹²

The dc-magnetic susceptibility was measured over a wide temperature range (1.7–400 K) in a field of 0.5 T, using a Quantum Design superconducting quantum interference device magnetometer. The magnetization curves were recorded at 1.7 K in magnetic fields up to 5 T.

High pressure was applied to the samples by using a modified Merrill-Basset DAC.^{13,14} The cell is made of non-magnetic CuBe alloy, allowing for measurements in external magnetic fields. The diamonds had a culet diameter of 600 μm . The initial diameter of the sample cavity in the $\text{Ta}_{90}\text{W}_{10}$ gaskets was 300 μm and the thickness was $\sim 60 \mu\text{m}$. The samples were mixed with epoxy, in order to reduce the risk of dispersion of uranium dust in case of breakage of the high-pressure cell. Epoxy also acted as a pressure transmitting medium. The pressure was determined by the ruby fluorescence method^{15,16} at room temperature, on different rubies placed in the sample chamber. The pressure gradient in the cell was about 5%.

The ^{119}Sn NFS measurements were performed at the undulator beamline ID18 (Ref. 17) of the European Synchrotron Radiation Facility (ESRF) in Grenoble, France. The storage ring was operated in 16-bunch mode with a maximum current of $I_{\text{SR}} \approx 90$ mA. A high heat-load He gas-cooled monochromator reduced the energy bandwidth of the undulator radiation to $\Delta E \approx 5$ eV. This was further reduced to ≈ 0.7 meV by a high-resolution monochromator, as de-

scribed in Ref. 18, combined with a collimating Be compound refractive lens.^{19–21} The beam was focused horizontally by a sagittally bent crystal. The spot size at the sample was $\sim 200 \times 400 \mu\text{m}^2$ (horizontal \times vertical), so as to illuminate almost completely the sample, and the flux at the sample was $\sim 2 \times 10^7$ photons/s. The high-pressure cell was mounted in a liquid-helium cryomagnet system, allowing for measurements in the temperature range between 3 and 300 K and in external magnetic fields up to 6 T. The scattered radiation was measured by using four stacked avalanche photodiodes, with a detection efficiency of $\sim 40\%$.²² The typical measuring time for each spectrum was about 0.5–1 h.

The low-temperature ^{119}Sn MS measurements were performed in a top-loading liquid-helium bath cryostat, using a $\text{Ca}^{119\text{m}}\text{SnO}_3$ radioactive source kept at a temperature of 4.2 K. The source had an activity of 10 mCi. The velocity of the drive moving the source was modulated sinusoidally. The typical measuring time for each high-pressure spectrum was about 24–48 h.

High-pressure x-ray-diffraction measurements were carried out in order to determine the pressure-volume relationship of the compounds under study. These measurements were performed by energy dispersive x-ray diffraction at beamline F3 of the Hamburger Synchrotronstrahlungslabor (HASYLAB) in Hamburg, Germany, for pressures up to 30 GPa at room temperature.

III. RESULTS AND DISCUSSION

A. Magnetic susceptibility, ^{119}Sn NFS, and MS at ambient pressure

The temperature dependence of the magnetic susceptibility at ambient pressure of $U(\text{In}_{0.8}\text{Sn}_{0.2})_3$ and $U(\text{In}_{0.6}\text{Sn}_{0.4})_3$ are shown in Fig. 1. In both cases, the inverse magnetic susceptibility shows a clear minimum at the temperature at which magnetic ordering sets in. The obtained values of the Néel temperature ($T_{\text{N}} \approx 90$ K for $x=0.2$ and $T_{\text{N}} \approx 35$ K for $x=0.4$) are in good agreement with those reported in Ref. 12. In the paramagnetic region, the susceptibility of both alloys follows a modified Curie-Weiss law, with effective magnetic moments μ_{eff} of 3.02(1) and 2.57(1) μ_{B} , paramagnetic Curie temperatures θ_{p} of $-120(2)$ and $-27(3)$ K, and temperature-independent terms χ_0 of $2.89(1) \times 10^{-4}$ and $5.24(1) \times 10^{-4}$ emu/mole for $U(\text{In}_{0.8}\text{Sn}_{0.2})_3$ and $U(\text{In}_{0.6}\text{Sn}_{0.4})_3$, respectively. The low-temperature magnetization (σ) of both compounds (see the inset of Fig. 1) shows a linear dependence on the applied magnetic field with no hysteresis effect, thus corroborating an antiferromagnetic character of the ordered state with full compensation of the sublattice magnetic moments. Moreover, the $\sigma(B)$ results prove the high quality of the samples measured, which were free of any ferromagnetic-like impurities.

^{119}Sn MS and NFS measurements have been performed on both compounds at ambient pressure and different temperatures, mainly in order to further inspect the quality of the samples and to compare the results with previous Mössbauer spectroscopy work.²³ The spectra measured in the paramagnetic and in the magnetically ordered state with MS and NFS are shown in Figs. 2 and 3 together with their fits, performed

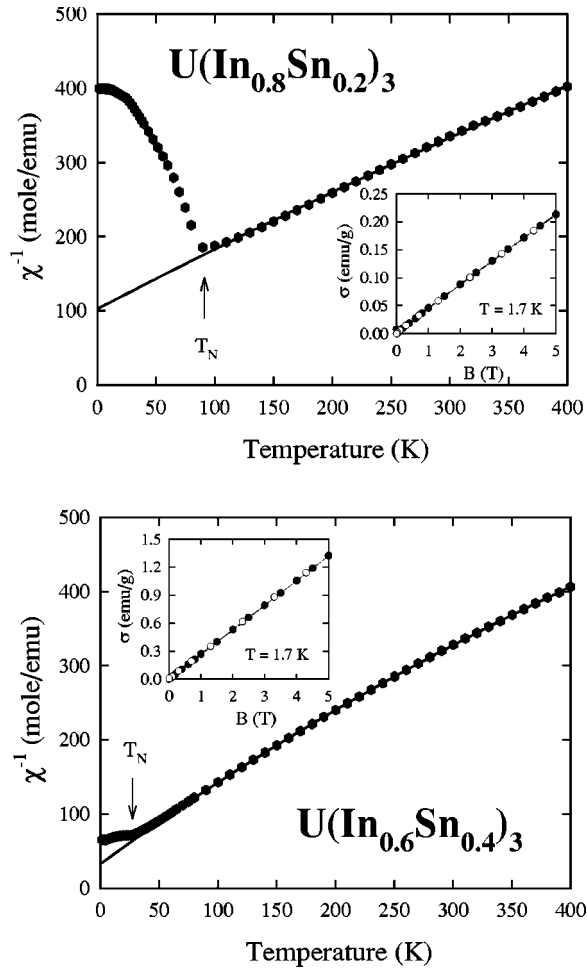


FIG. 1. Inverse magnetic susceptibility plotted as a function of temperature for the two $U(\text{In}_{1-x}\text{Sn}_x)_3$ compounds with $x=0.2$ and 0.4 . The full circles are experimental data points measured in a field of 0.5 T, while the solid lines are the modified Curie-Weiss fits with the parameters given in the text. The arrows mark the Néel temperature. In the inset of each graph the corresponding magnetization curve taken at 1.7 K is shown. The full and open circles denote the magnetization data measured with increasing and decreasing magnetic-field strength, respectively.

with the package CONUSS.²⁴ This fits both MS and NFS spectra by using the full dynamical theory of nuclear resonance scattering, including the diagonalization of the complete hyperfine Hamiltonian.

In the paramagnetic state ($T > 90$ K and $T > 35$ K for $x = 0.2$ and 0.4 , respectively), the spectra are characterized by a quadrupole splitting. This 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 Sn atoms. The quadrupole interaction parameter ($\Delta E_Q = \frac{1}{2} e Q V_{zz}$) has a value of $1.53(1)$ mm/s for $x=0.2$ and $1.50(1)$ mm/s for $x=0.4$ at $T = 125$ K, therefore showing a slight decrease as the Sn concentration increases. These results agree with the general trend observed in the series $U(\text{In}_{1-x}\text{Sn}_x)_3$, where ΔE_Q decreases from 1.64 mm/s in $U(\text{In}_{0.98}\text{Sn}_{0.02})_3$ to 1.46 mm/s in $U(\text{In}_{0.4}\text{Sn}_{0.6})_3$.²³ The quadrupole interaction parameter is sig-

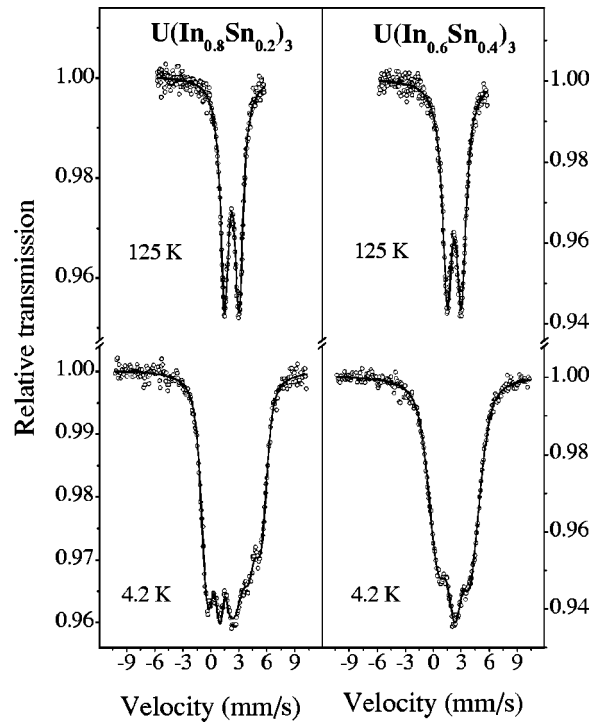


FIG. 2. Mössbauer spectra of $U(\text{In}_{0.8}\text{Sn}_{0.2})_3$ and of $U(\text{In}_{0.6}\text{Sn}_{0.4})_3$ at ambient pressure and different temperatures. The circles represent experimental data points, while the lines are fits.

nificantly larger than in isostructural $R\text{Sn}_3$ ($R = \text{rare-earth}$) compounds [0.9 – 1.2 mm/s (Ref. 25)] and NpSn_3 [1.23 mm/s (Ref. 26)], but lower than in $U(\text{Ga}_{0.98}\text{Sn}_{0.02})_3$ [1.68 mm/s (Ref. 27)]. This indicates a different distribution of the Sn $5p$ electrons in the various compounds.

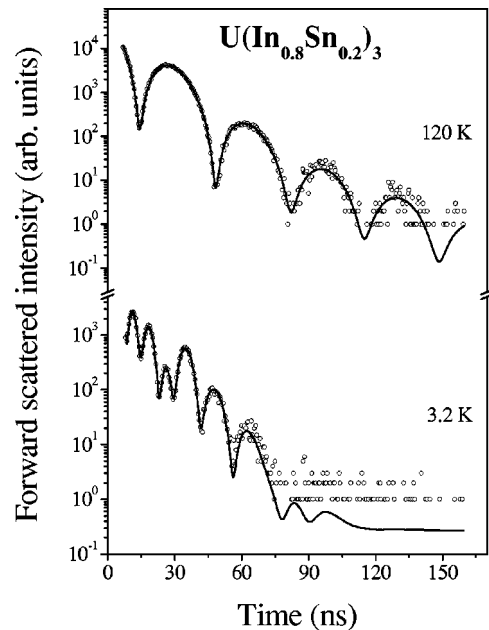


FIG. 3. NFS spectra of $U(\text{In}_{0.8}\text{Sn}_{0.2})_3$ at ambient pressure and different temperatures. The circles represent experimental data points, while the lines are fits.

The value of the isomer shift ΔS at 125 K [2.25(1) mm/s for $x=0.2$ and 2.27(1) mm/s for $x=0.4$] is found to be in the region which is typical for metallic Sn compounds (for β -Sn, $\Delta S=2.57$ mm/s at room temperature). It slightly increases as the Sn content increases, as already found in Ref. 23, indicating an increase of s -electron density at the Sn nuclei.

Although both absorbers were prepared so that they contain the same amount of Sn nuclei, the linewidth of the MS spectra is larger for $x=0.4$ than for $x=0.2$. This can be interpreted as caused by the increase of atomic disorder introduced when substituting Sn for In. This has also been demonstrated by perturbed angular correlation (PAC) studies on $U(\text{In}_{0.5}\text{Sn}_{0.5})_3$ at room temperature.²⁸ These authors have found that the effective charges on Sn and In atoms are different and that an attractive interaction between Sn and In exists, so that every In atom has $\approx 65\%$ of Sn atoms as nearest neighbors, instead of the 50%, which one would expect for a completely random distribution of Sn and In atoms. This causes a distortion of the EFG at every ligand site, with changes in its magnitude V_{zz} and asymmetry parameter η from site to site. Such a distortion is reflected in the Mössbauer spectra by a distribution of quadrupole splittings, which results in an increase of the linewidth. This would be the largest for equal concentrations of Sn and In and decrease as one of the two components prevails.

Below the magnetic ordering temperature T_N , the spectra for both compositions show the presence of a transferred magnetic hyperfine field (B_{thf}) at the Sn nuclei, combined with the splitting due to the quadrupole interaction. B_{thf} is 3.0(1) T for $x=0.2$ and 2.3(1) T for $x=0.4$. In order to fit these spectra, an assumption has to be made about the magnetic structure of the compounds, as this will affect both the value and orientation of B_{thf} . The magnetic structure of $U\text{In}_3$ has been determined by neutron diffraction on powder samples as antiferromagnetic of type II.¹⁰ In the absence of measurements of the magnetic structure of compounds of the series $U(\text{In}_{1-x}\text{Sn}_x)_3$ with $0 < x < 0.45$, the fit of the MS and NFS spectra has been performed assuming that both measured compounds keep the same magnetic structure as $U\text{In}_3$. This is consistent with μSR studies on this series,²⁹ which show no magnetic field at muons located in the center of the cubic unit cell. The origin of the transferred magnetic hyperfine field at the Sn nuclei B_{thf} is twofold: an indirect polarization through the conduction electrons mediated by the RKKY interaction is combined with the direct polarization caused by the overlap of the U $5f$ electrons with the outer electrons of the Sn atoms (mainly belonging to the $5p$ shell).^{25,26,30} Due to the particular location of the Sn atoms in the unit cell of the AuCu_3 crystal structure, the isotropic part of B_{thf} must vanish for an antiferromagnet of type II because the contributions from the four neighboring U atoms cancel pairwise. Only the anisotropic component, due to the unpaired spin density transferred into the Sn $5p$ orbitals through f - p hybridization,²⁷ has a residual contribution. This orients always in the U-(Sn,In) planes and should therefore always be perpendicular to the principal axis of the local EFG. Although this is fulfilled in $U\text{In}_3$,³¹ the distortion of the EFG caused by the atomic disorder introduced by the substitution of In with Sn (as discussed above) can lead to changes

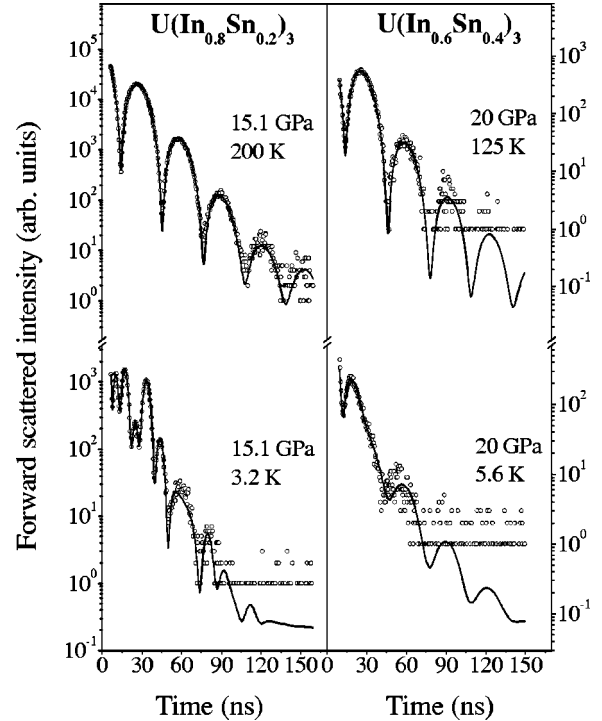


FIG. 4. NFS spectra of $U(\text{In}_{0.8}\text{Sn}_{0.2})_3$ at $p=15.1$ GPa and of $U(\text{In}_{0.6}\text{Sn}_{0.4})_3$ at $p=20$ GPa at different temperatures. The circles represent experimental data points, while the lines are fits.

in the direction of the EFG. The angle θ between the principal axis of the EFG and B_{thf} can therefore be different from 90° . Moreover, the value of the transferred field depends on the magnitude of the U magnetic moment and this is in turn determined by the local environment of each U atom. The atomic disorder between In and Sn causes different U atoms to have different local environments and therefore introduces a distribution in the values of B_{thf} around a mean value $\overline{B_{\text{thf}}}$. The MS and NFS spectra measured at temperatures below T_N have therefore been analyzed according to these observations, using θ , $\overline{B_{\text{thf}}}$ and the width of the distribution as free parameters during the fitting procedure. The quadrupole interaction parameter ΔE_Q has been constrained to its high temperature value, as it does not show any appreciable temperature dependence for $T > T_N$.

B. Volume dependence of the magnetic state

1. Stability of the magnetic state

NFS measurements have been performed up to pressures of 15 and 25 GPa on the samples with $x=0.2$ and 0.4, respectively. On the latter, the NFS measurements have been complemented by MS studies. Some selected NFS spectra are shown in Fig. 4, together with the least-squares fits performed using CONUSS.

In the paramagnetic state, the spectra are characteristic of a quadrupole split doublet, with the splitting increasing with 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

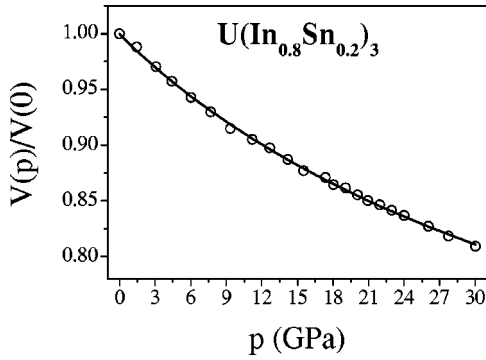


FIG. 5. Volume-pressure relationship for $U(\text{In}_{0.8}\text{Sn}_{0.2})_3$ at room temperature. The open circles are the measured points while the line is a fit with the Murnaghan equation.

hyperfine fields, with average $\overline{B_{\text{thf}}(p)}$, at the Sn nuclei. All spectra have been analyzed following the same procedure used for the spectra measured at ambient pressure, with the additional assumptions that neither the crystalline nor the magnetic structure are changed by pressure. Our high-pressure x-ray-diffraction measurements on $U(\text{In}_{0.8}\text{Sn}_{0.2})_3$ show indeed that the AuCu_3 -type structure is stable up to the highest pressure reached of 30 GPa. The volume of the unit cell (normalized to its ambient pressure value) is plotted as a function of the applied pressure in Fig. 5. The experimental curve has been fitted by a Murnaghan equation, yielding a bulk modulus $B_0=93(2)$ GPa. A previous x-ray diffraction study on UX_3 compounds³² has demonstrated that both UIn_3 and USn_3 have a stable AuCu_3 structure up to at least 40 GPa and that the bulk moduli of both compounds are rather similar (99 GPa for UIn_3 and 83 GPa for USn_3 , respectively). We therefore assume that also $U(\text{In}_{0.6}\text{Sn}_{0.4})_3$ is stable and use the same bulk modulus as determined for $U(\text{In}_{0.8}\text{Sn}_{0.2})_3$ to calculate the pressure-volume relationship for this compound.

The volume dependence of $\overline{B_{\text{thf}}}$, measured at low temperature $T \leq 5.6$ K, and T_N for the two compounds is shown in Fig. 6, where both quantities are plotted as a function of the relative decrease of the volume of the unit cell $1 - V(p)/V(0)$. For $x=0.2$, the average magnetic hyperfine field shows almost no dependence on pressure (up to $p = 15$ GPa, corresponding to a volume contraction of the unit cell of $\sim 12\%$). Assuming that not only the magnetic structure but also the orientation of the U moments does not change with pressure, this implies that the magnitude of the U moment does not decrease as the volume contracts. The Néel temperature shows a monotonic increase with pressure: T_N increases from 90(2) K at ambient pressure to 172(5) K at the highest pressure of 15 GPa. The case of $U(\text{In}_{0.6}\text{Sn}_{0.4})_3$ is considerably different. The average magnetic hyperfine field decreases monotonically with increasing pressure and reaches its lowest value, corresponding to $\sim 10\%$ of the initial one, at the highest pressure reached in this study, 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

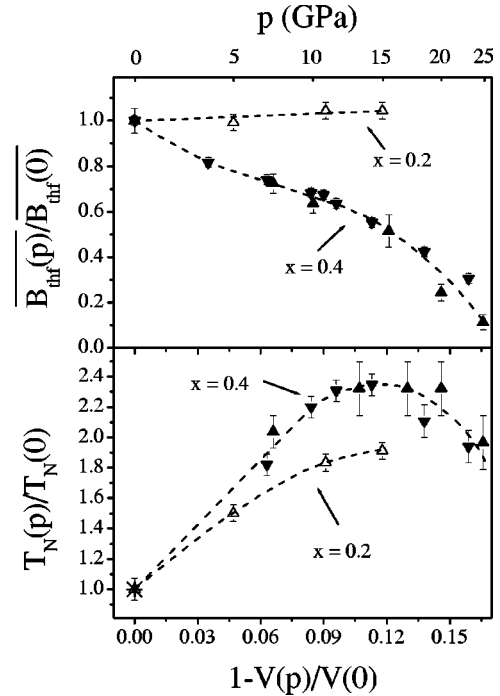


FIG. 6. Volume dependence of the average transferred hyperfine field $\overline{B_{\text{thf}}}$ (measured at low temperature $T \leq 5.6$ K) and of the Néel temperature T_N for $U(\text{In}_{0.8}\text{Sn}_{0.2})_3$ and $U(\text{In}_{0.6}\text{Sn}_{0.4})_3$. T_N and $\overline{B_{\text{thf}}}$ are plotted against the relative decrease of the volume of the unit cell $1 - V(p)/V(0)$. The open symbols refer to $x=0.2$, while the full symbols refer to $x=0.4$. The down triangles refer to the results of the MS measurements while the up triangles indicate the results of the NFS measurements. The value of T_N at ambient pressure indicated by an asterisk is determined by magnetic susceptibility for $x=0.4$. The dashed lines are a guide to the eye.

comparison with that of UIn_3 . On the other hand, the Néel temperature first increases with pressure, reaches its maximum value $T_N=65(2)$ K at $p \approx 14$ GPa and then starts decreasing for $p > 14$ GPa. However, its value at 25 GPa, 55(5) K, is still high although the value of the ordered magnetic moment ($\sim 0.1\mu_B$) is strongly reduced.

2. Pressure induced collapse of the magnetic state of $U(\text{In}_{0.6}\text{Sn}_{0.4})_3$

The pressure-induced changes of the magnetic state (magnetic moment and ordering temperature) of $4f$ - and $5f$ -electron systems give valuable information about the mechanisms underlying the delocalization of these electrons in such systems. For a number of systems, such as compounds of the $4f$ elements Ce and Yb, the volume dependence of the ordering temperature can be explained by the Kondo lattice model proposed by Doniach¹ and its extensions.³³⁻³⁵ According to these models, the properties of the ground state of a dense Kondo system depend on the competition between the RKKY interaction, which tends to stabilize long-range magnetic order, and the quenching of the local moments by the Kondo effect. The characteristic energies of these two interactions have a different dependence on the product $|JN(E_F)|$, where J is the exchange-coupling

strength and $N(E_F)$ the density of states at the Fermi level. In fact the energy scale of the RKKY interaction can be written as $k_B T_{\text{RKKY}} \propto J^2 N(E_F)$, whereas that of the Kondo singlet shows an exponential dependence of the type $k_B T_K \propto N(E_F)^{-1} \exp[-1/|JN(E_F)|]$. For small values of $|JN(E_F)|$, the RKKY interaction prevails and a magnetically ordered ground state is formed. As $|JN(E_F)|$ increases, the Kondo interaction becomes dominant, and long-range magnetic order is suppressed. The resulting phase diagram¹ shows an increase in the magnetic ordering temperature T_m with $|JN(E_F)|$, followed by a much faster decrease when $|JN(E_F)|$ exceeds a critical value. The application of pressure to Ce compounds has been experimentally shown to increase $|JN(E_F)|$ (see, for example, Refs. 36 and 37), while the opposite holds true for systems containing Yb (see, for example, Ref. 38). In the case of systems containing $5f$ electrons, whose wave functions have a considerably greater spatial extent than those of $4f$ electrons, one expects that hybridization plays a very important role in determining the ground-state properties of a compound. In this case, the demagnetization process is driven by the transition from a local to an itinerant (bandlike) 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,³⁹ the decrease of the atomic distance caused by pressure induces the $5f$ wave functions to diffuse more outside the core region. This in turn increases the hybridization between the $5f$ electrons and the band states of the compound, with a consequent loss of the $5f$ spectral weight and the reduction of the local moment. On the other hand, hybridization enhances the exchange-coupling J , thus strengthening the magnetic order. 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 volume dependence of the ordering temperature of U monochalcogenides.^{40,39,38}

The results of our measurements as shown in Fig. 6 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 $\text{U}(\text{In}_{0.8}\text{Sn}_{0.2})_3$. However, local moment compounds of the rare earths [for example EuAl_2 (Ref. 41) and DyAl_2 (Ref. 42)] and of the actinides [for example NpCo_2Si_2 (Ref. 43)] show a quadratic increase of T_N with pressure. In the case of $\text{U}(\text{In}_{0.8}\text{Sn}_{0.2})_3$ the increase of the Néel temperature with pressure is only linear 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, similarly to NpGa_3 .⁴⁴ On the contrary, $\text{U}(\text{In}_{0.6}\text{Sn}_{0.4})_3$ 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. For pressures lower than $p \approx 14$ GPa one can therefore conclude that the RKKY exchange interaction pre-

vails over the mechanisms, which tend to weaken or destroy magnetic order, whereas at higher pressures the latter dominate. In the current series of measurements it has not been possible to reach the critical pressure for the complete suppression of magnetic order. However, in the case of $\text{U}(\text{In}_{0.6}\text{Sn}_{0.4})_3$ a clear maximum is observed in the volume dependence of the Néel temperature. This curve appears to be symmetric with respect to the maximum: the rate at which T_N increases with pressure below its maximum is approximately the same as the rate at which T_N decreases above the maximum. This points rather towards hybridization as the cause for the delocalization of the U $5f$ moments, because the Kondo interaction would be expected to produce a much faster decrease of the ordering temperature.^{1,37} One can therefore conclude that it is the $5f$ -ligand hybridization, as a consequence of the increasing $5f$ bandwidth with increasing pressure, that drives the transition from the magnetic to a nonmagnetic state for $\text{U}(\text{In}_{1-x}\text{Sn}_x)_3$. In this respect it is important to compare the effect of increasing the Sn concentration on the stability of the U $5f$ moments with that of external pressure. As it is mentioned above (see Sec. I), increasing Sn concentration from $x=0$ to $x=0.4$ results in a reduction of the Néel temperature by about a factor of 3.¹² In such a case, the increase of the hybridization is predominantly due to an increase of the $5p$ density of states (band filling effect), which also results in a delocalization of the $5f$ moments. In fact, an increase of the $5p$ density of states should also strengthen the RKKY interaction. However, such an effect is weaker than that of the predominating hybridization. In contrast to this, when external pressure is applied, the two mechanisms compete and for $x=0.4$ the RKKY exchange interaction initially prevails, although the ordered magnetic moment decreases monotonically. Consequently, a pressure higher than 25 GPa is necessary to fully delocalize the $5f$ moments for $x=0.4$ and even higher pressures are expected to be necessary in the case of $x=0.2$, which is more localized at ambient pressure.

3. Nature of the high-pressure state of $\text{U}(\text{In}_{0.6}\text{Sn}_{0.4})_3$

In order to gain a deeper insight into the nature of the high-pressure state of $\text{U}(\text{In}_{0.6}\text{Sn}_{0.4})_3$, we have performed a series of ^{119}Sn NMR measurements, at $p=25$ GPa, at various temperatures between 3.2 and 20 K in an external magnetic field of 6 T. The spectrum measured at 3.2 K is shown in Fig. 7. A fit to this spectrum shows that the effective hyperfine field at the Sn nuclei is a combination of the external field of 6 T and an average induced field of ~ 2.4 T, almost perpendicular to the external one. The large strength of the induced field points towards the presence of large dynamical spin correlations in the high-pressure state of this compound, as already observed at ambient pressure on the paramagnetic compounds ($x \geq 0.5$) of the series $\text{U}(\text{In}_{1-x}\text{Sn}_x)_3$ by several studies.^{45,46,29,6,47} In particular, Cottenier *et al.*⁴⁵ have demonstrated, using PAC on ^{111}Cd impurities occupying the Sn lattice positions in compounds with $x=0.5, 0.7$, and 1.0, that the induced field at 4.2 K is the highest for $x=0.5$, which is very close to the phase boundary between long-range magnetic order and paramagnetism. They measured an induced

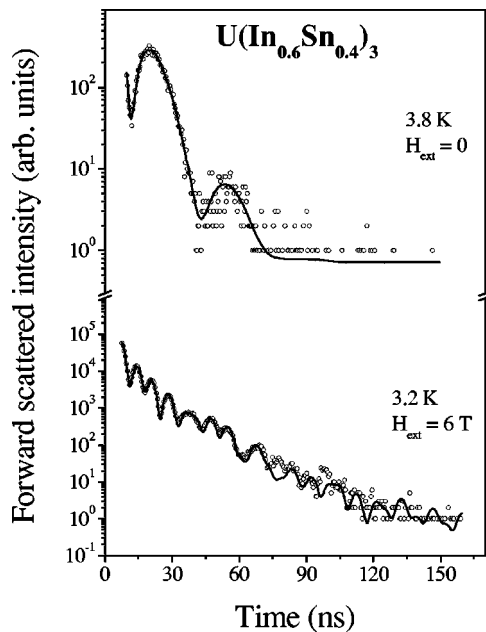


FIG. 7. NFS spectra of $U(\text{In}_{0.6}\text{Sn}_{0.4})_3$ at $p = 25$ GPa. The upper graph shows the spectrum measured at 3.8 K without any externally applied magnetic field, while the graph at the bottom displays the spectrum measured at 3.2 K in a field of 6 T. The circles represent experimental data points, while the lines are fits.

hyperfine field at the Cd nuclei whose value is comparable to that obtained in the present study. This demonstrates the similarity of the dynamic nature of the pressure induced state with that obtained by chemical substitution. However, from our results alone it is not possible to draw any conclusion about some of the properties of the high-pressure state of $U(\text{In}_{0.6}\text{Sn}_{0.4})_3$, such as possible large values of the Sommer-

field coefficient γ , and of the low-temperature magnetic susceptibility χ_0 , as is the case at ambient pressure for $x \geq 0.5$.

IV. CONCLUSIONS

In conclusion, using high-pressure ^{119}Sn nuclear forward scattering and Mössbauer spectroscopy we were able to determine the volume dependence of the Néel temperature and of the average transferred magnetic hyperfine field of $U(\text{In}_{1-x}\text{Sn}_x)_3$, for $x = 0.2$ and 0.4. While $U(\text{In}_{0.8}\text{Sn}_{0.2})_3$ behaves as a very weakly delocalized system up to 15 GPa, with its Néel temperature increasing with pressure but with only very weak volume dependence of the transferred field, $U(\text{In}_{0.6}\text{Sn}_{0.4})_3$ shows a typical behavior for a system being close to a magnetic instability. Although its Néel temperature increases initially with pressure, reaching a maximum at $p \approx 14$ GPa, the average transferred field decreases monotonically and its value at 25 GPa is reduced to only $\sim 10\%$ of the initial value. This behavior can be explained by the competition between the indirect exchange RKKY interaction and the $5f$ -ligand hybridization. The former prevails at lower pressure, where it is responsible for the increase of the Néel temperature, whereas the latter dominates at pressures above ~ 14 GPa, substantially delocalizing the $5f$ electrons. Measurements at 25 GPa in an external magnetic field of 6 T reveal the presence of a large induced magnetic hyperfine field. This points towards the presence of strong spin correlations of dynamical nature in $U(\text{In}_{0.6}\text{Sn}_{0.4})_3$, as already observed at ambient pressure in the paramagnetic region ($x \geq 0.5$) of the phase diagram of $U(\text{In}_{1-x}\text{Sn}_x)_3$.

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