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Mössbauer investigation of ^{119}Sn in U$(\mathrm{Ga}_{0.98}\mathrm{Sn}_{0.02})_3$ and f-p hybridization in the itinerant antiferromagnet UGa$_3$

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The magnetic properties of U$(\mathrm{Ga}_{0.98}\mathrm{Sn}_{0.02})_3$ were investigated by using the ^{119}Sn Mössbauer probe occupying the Ga position with the purpose of gaining information on the itinerant antiferromagnet UGa$_3$. In the ordered state, at $T<T_N\sim73\,\mathrm{K}$ the ^{119}Sn nuclei feel a transferred hyperfine field that orients perpendicular to the principal axis of the electric field gradient. This is in agreement with the proposed antiferromagnetic type-II structure for which only the anisotropic component of the transferred field does not cancel. From the magnitude of the transferred field we estimated the spin polarization of the $5p$ states of Sn that arises because of hybridization with the ordered $5f$ electrons. The magnetic moment carried by the Sn atoms is about $0.02\mu_B$. Below 35 K the uranium moments undergo a spin reorientation with the moment close to the [110] direction at low temperature.

I. INTRODUCTION

The actinide AnX_3 compounds, where X is a nontransition element from group IIIA or IVA, crystallize in the cubic AuCu$_3$-type structure where the interactinide separation $d(\text{An-An})$ is equal to the lattice parameter a. It turns out that $d(\text{An-An})$ of these compounds is far above the Hill limit, therefore the $5f$-ligand hybridization is the main mechanism controlling the delocalization of the $5f$ electrons.[1] The systematics of this hybridization has been well demonstrated for the UX_3 compounds for which large amounts of experimental data and band structure calculations are available.[2–4] These compounds either do not order magnetically ($X=\mathrm{Al, Si, Ge, Sn}$) or they exhibit an antiferromagnetic structure ($X=\mathrm{Ga, In, Tl, Pb}$), USn$_3$ being close to a magnetic instability while local moment character is observed in UTl$_3$ and UPb$_3$. From this behavior it was concluded that the $5f$-ligand hybridization is enhanced as the size of the X atom decreases, i.e., when moving upwards within each group. Furthermore, owing to the higher occupation of the p states as one moves from a group-IIIA element to a group-IVA element, the f-p hybridization is favored.[1]

Among the UX_3 compounds, special attention was paid to UGa$_3$ because experiments and band structure calculations strongly suggest an itinerant nature for the magnetism of UGa$_3$.[4–8] Besides, the availability of high-quality single crystal allowed new investigations. The single crystal neutron diffraction studies[9] of the magnetic structure of UGa$_3$ confirm earlier neutron investigations made on polycrystalline samples[10,11] but disagree with Mössbauer data.[12] According to the neutron diffraction results UGa$_3$ orders at $T_N\sim67\,\mathrm{K}$ with a type-II antiferromagnetic (AF) structure in which alternate (111) ferromagnetic planes of U moments are coupled antiferromagnetically. The ordered magnetic moment (m) is $0.74(8)\mu_B$ per U atom at 5 K and in-field experiments suggest that the transition at 40 K, where the single crystal magnetic susceptibility exhibits a singularity,[9,13] may be associated with a spin reorientation of the U moments from $m\parallel[100]$ at high temperature to $m\parallel[111]$ or [110] at low temperature.[9,13] Single crystals of UGa$_3$ were also examined by synchrotron radiation using the magnetic x-ray diffraction technique at the uranium M edges and at the Ga K edge.[14] The shape of the uranium M resonances was as found for most such compounds but, surprisingly, a great enhancement of the signal at the Ga K edge was observed. This is suggested to be due to the spin polarization of the Ga $4p$ states caused by f-p hybridization. These new findings prompted us to reinvestigate the magnetic properties of UGa$_3$ by means of Mössbauer spectroscopy using a ^{119}Sn enriched U$(\mathrm{Ga}_{0.98}\mathrm{Sn}_{0.02})_3$ sample. This microscopic technique was already shown to provide useful complementary information on the magnetic structure, e.g., type of spin arrangement, orientation of the moments with respect to the principal axis of the electric field gradient (EFG).[15,16] In addition, the magnitude of the transferred hyperfine field is expected to be related to the strength of the f-p hybridization and thus to give new insights into the $5f$ delocalization in UGa$_3$.

II. ORIGIN OF THE TRANSFERRED HYPERFINE FIELD AT THE Sn NUCLEI

The magnetic hyperfine field acting on the nuclei of the nominally diamagnetic tin atoms, in the ordered state of the

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$U(Ga_{0.98}Sn_{0.02})_3$ solid solution, arises from the finite spin density at the ligand site produced by the uranium $5f$ spin via the magnetic exchange interactions. Two mechanisms contribute to the transferred hyperfine field (H^{tr}):[15–17] (i) the conduction electron polarization through the Ruderman-Kittel-Kasuya-Yosida interaction, and (ii) the mixing of the $5f$ electrons with the outer electrons of the Sn atoms (e.g., f-$5p$ hybridization). For a type-II antiferromagnetic structure, as found for UGa_3 and supposed to hold for $U(Ga_{0.98}Sn_{0.02})_3$, the isotropic part of the transferred field is vanishing as the contributions from the neighboring U atoms cancel pairwise. Only the anisotropic component has a residual contribution which, as shown below, orients in the (100) plane, i.e., perpendicular to the local EFG axis (it is worth mentioning that the anisotropic contribution to H^{tr} was disregarded in previous works). This could explain why the magnetic structures of UIn_3 and UGa_3 proposed from the Mössbauer data did not agree with the neutron results).[12,18] The anisotropic field can be viewed as a spin-dipolar term arising mainly from the unpaired spin density (f_{5p}) transferred into the Sn $5p$ orbitals through f-p hybridization (the regular dipolar field induced by the U moments is an order of magnitude smaller).[19] Writing explicitly the contributions to the hyperfine field in terms of spin density transfer along the bond direction (α), the components of the hyperfine field for a single U-Sn bond can be expressed as:[20,21]

$$H^{\mathrm{tr}}_{\alpha}=H_{\mathrm{an}}\frac{m_{\alpha}}{m},\quad H^{\mathrm{tr}}_{\beta,\gamma}=-\frac{1}{2}H_{\mathrm{an}}\frac{m_{\beta,\gamma}}{m},\tag{1}$$

where $H_{\mathrm{an}}=-\frac{4}{5}\mu_B\langle r^{-3}\rangle f_{5p}$ and $\langle r^{-3}\rangle=75.6\times10^{24}\,\mathrm{cm}^{-3}$ (Ref. 22) corresponds to the average radial distribution of $5p$ electrons of Sn. $m_{\alpha,\beta,\gamma}$ are the projections of the uranium moment m along the orthogonal system of axes (α,β,γ).

Considering now only the four nearest neighbor U atoms of an Sn atom, the total hyperfine field is obtained by summation over the four equivalent and independent U-Sn bonds. The components of the hyperfine field along the local x,y,z axes as defined in Fig. 1 are given by the expressions:

$$H^{\mathrm{tr}}_{x}=3H_{\mathrm{an}}\frac{m_x}{m},\quad H^{\mathrm{tr}}_{y}=3H_{\mathrm{an}}\frac{m_y}{m},\quad H^{\mathrm{tr}}_{z}=0.\tag{2}$$

They show that the transferred hyperfine field orients parallel to the U-Sn plane whatever the orientation of the U moments in the AF type-II magnetic structure. In the following we will show that the number of magnetically inequivalent sites as well as the magnitude of the hyperfine field depend on the actual direction of the U moments (Table I). For the most general case, i.e., when the U moments are along an arbitrary $[uvw]$ direction, the Sn nuclei in positions 1,2,3 (see Fig. 1) feel different hyperfine fields, which are given by

$$H^{\mathrm{tr}}(1)=3H_{\mathrm{an}}\sqrt{(u^2+v^2)/(u^2+v^2+w^2)},$$

$$H^{\mathrm{tr}}(2)=3H_{\mathrm{an}}\sqrt{(v^2+w^2)/(u^2+v^2+w^2)},$$

$$H^{\mathrm{tr}}(3)=3H_{\mathrm{an}}\sqrt{(u^2+w^2)/(u^2+v^2+w^2)}.\tag{3}$$

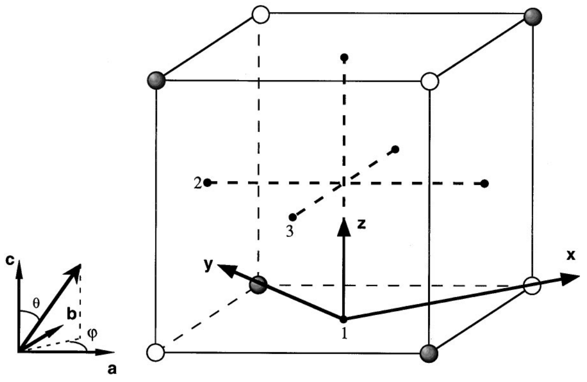

FIG. 1. 1/8 of the AF type-II unit cell for UGa_3. White and black spheres are U with antiparallel moments. The dots represent the Ga(Sn) atoms whose three inequivalent positions are numbered. The hyperfine field components are computed (see text) along the local (x,y,z) system of axes shown, e.g., for Ga(Sn) position 1. The principal axis of the electric field gradient is oriented along the normal to the face of the cube containing the considered Ga(Sn) position. The polar angles (θ,φ) shown in the inset give the orientation of the uranium magnetic moment in the system of axes attached to the crystal structure.

These expressions simplify considerably when the U moments points along particular directions like $[uv0]$ and $[uuw]$ or along the principal directions of a cube (Table I). Let us examine the latter cases. The U moments are along a [100] direction: tin atoms in positions 1 and 3 feel the same hyperfine field $H^{\mathrm{tr}}(1,3)=3H_{\mathrm{an}}$, while the Sn atoms in position 2 do not experience a magnetic field. This yields two inequivalent tin sites, the first A (1 and 3) experiences a combined quadrupole and magnetic interaction and the second $B(2)$ merely the quadrupole interaction. The intensity ratio $A{:}B=2{:}1$. The U moments are along a [110] direction: Here again there are two inequivalent tin sites but both experience a magnetic hyperfine field. For tin atoms in position $1(A)$, $H^{\mathrm{tr}}(1)=3H_{\mathrm{an}}$ and for tin atoms in positions $2,3(B)$, $H^{\mathrm{tr}}(2,3)=(3\sqrt{2}/2)H_{\mathrm{an}}$. The intensity ratio of the two sites is $A{:}B=1{:}2$. Finally if the U moments are along a [111] direction: all three tin sites are magnetically equivalent and $H^{\mathrm{tr}}(1,2,3)=\sqrt{6}H_{\mathrm{an}}$.

III. EXPERIMENTAL DETAILS

The polycrystalline sample of $U(Ga_{0.98}Sn_{0.02})_3$ enriched with ^{119}Sn was prepared by arc melting the constituent ele-

TABLE I. Transferred hyperfine field at the three inequivalent Sn positions for different U moment directions. θ and φ are the polar angles defining the orientation of the uranium magnetic moment in the AF type-II structure (see Fig. 1).

Moment direction	Hyperfine field at position (i)
$[uvw]$	$H^{\mathrm{tr}}(1)=3H_{\mathrm{an}}\sin\theta$, $H^{\mathrm{tr}}(2)=3H_{\mathrm{an}}\sqrt{1-\sin^2\theta\cos^2\varphi}$ $H^{\mathrm{tr}}(3)=3H_{\mathrm{an}}\sqrt{1-\sin^2\theta\sin^2\varphi}$
$[uv0]$	$H^{\mathrm{tr}}(1)=3H_{\mathrm{an}}$, $H^{\mathrm{tr}}(2)=3H_{\mathrm{an}}\sin\varphi$, $H^{\mathrm{tr}}(3)=3H_{\mathrm{an}}\cos\varphi$
$[uuw]$	$H^{\mathrm{tr}}(1)=3H_{\mathrm{an}}\sin\theta$, $H^{\mathrm{tr}}(2)=H^{\mathrm{tr}}(3)$ $=3H_{\mathrm{an}}\sqrt{\frac{1}{2}\sin^2\theta+\cos^2\theta}$
[100]	$H^{\mathrm{tr}}(1)=H^{\mathrm{tr}}(3)=3H_{\mathrm{an}}$, $H^{\mathrm{tr}}(2)=0$
[110]	$H^{\mathrm{tr}}(1)=3H_{\mathrm{an}}$, $H^{\mathrm{tr}}(2)=H^{\mathrm{tr}}(3)=\frac{3\sqrt{2}}{2}H_{\mathrm{an}}$
[111]	$H^{\mathrm{tr}}(1)=H^{\mathrm{tr}}(2)=H^{\mathrm{tr}}(3)=\sqrt{6}H_{\mathrm{an}}$

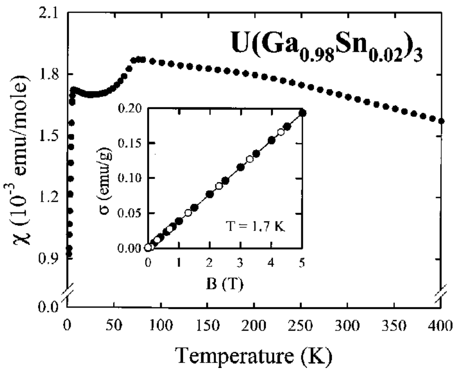

FIG. 2. Magnetic susceptibility vs temperature for $U(Ga_{0.98}Sn_{0.02})_3$ measured in a field of 0.5 T. The inset shows the field dependence of the magnetization at 1.7 K taken with increasing (filled circles) and decreasing (open circles) magnetic field

ments in an argon atmosphere and subsequent annealing in vacuum at 600 °C for 1 week. The sample checked by x-ray diffraction was found to be single phase with the expected cubic $AuCu_3$-type crystal structure ($a = 4.258$ Å). It is worth noticing that UGa_3 and USn_3 from a solid solution, therefore the ^{119}Sn atoms are expected to occupy the Ga position.[5] The dc-magnetic susceptibility was measured over a wide temperature range (1.7–300 K) in a field of 0.5 T using a Quantum Design MPMS-5 superconducting quantum interference device magnetometer. A magnetization curve was recorded at 1.7 K in fields up to 5 T.

The ^{119}Sn Mössbauer measurements were performed on a powder absorber made by mixing the sample with BN to ensure homogeneity and uniform thickness ($\sim$50 mg/cm^2). ^{119}Sn enriched tin to a level of $\sim$98% was used. The Mössbauer source of $Ca^{119m}SnO_3$ was kept at 4.2 K and the temperature of the absorber was varied from 4.2 to 290 K. Since the quadrupole and magnetic interactions were of comparable magnitude, the data were analyzed by diagonalization of the full Hamiltonian.

IV. RESULTS AND DISCUSSION

A. Magnetic susceptibility and magnetization

The temperature variation of the magnetic susceptibility $\chi(T)$ of $U(Ga_{0.98}Sn_{0.02})_3$ in a field of 0.5 T is shown in Fig. 2. As seen in this figure, the susceptibility is only weakly temperature dependent above 5 K and the Néel temperature of 73 K manifests itself only as an inflection point in the $\chi(T)$ curve. As expected from the study of the $U(Ga_{1-x}Sn_x)_3$ solid solutions,[5] substitution of Sn($x = 0.02$) for Ga shifts T_N to higher temperature ($\Delta T_N/T_N \sim 9\%$). In the paramagnetic region, there is no evidence for Curie-Weiss behavior, which has been explained by the itinerant nature of the $5f$ electrons.[5] As found previously for UGa_3 we observe an upturn of the susceptibility at low temperature, which is followed, in the $U(Ga_{0.98}Sn_{0.02})_3$ sample, by a rapid decrease of $\chi(T)$ below about 5 K. Also, some anomaly in $\chi(T)$ has been reported around 8 K for UGa_3 single crystalline samples[7,9] and it was furthermore shown that the lower the applied field the more rapid the drop in the susceptibility.[23] These low-temperature features observed in $U(Ga_{0.98}Sn_{0.02})_3$ and in UGa_3 single crystals may be ascribed to a peculiar itinerant nature of their magnetically ordered state.[7] This presumption was supported by Cornelius *et al.*[4] who suggested that due to the itinerant character of the $5f$ electrons the Fermi surface of UGa_3 can be drastically altered by the application of a magnetic field. Clearly more investigations of the origin of the low temperature $\chi(T)$ anomaly (also observed in the thermal conductivity[7]) and of the Fermi surface reconstruction are needed to fully understand the observed behavior.[4] The singularity seen at 40-K in UGa_3 single crystals[7,9] was not observed in the $U(Ga_{0.98}Sn_{0.02})_3$ solid solution and in polycrystalline UGa_3 samples.[5] It is interesting to note that this transition is best visible when the field is high and applied along the [100] direction.[7,9] This was recently confirmed by Aoki *et al.*[24] who showed that the 40-K anomaly in the $\chi(T)$ curve of UGa_3 is washed out when the field is applied along the [111] direction. Therefore it is not so surprising that, due to anisotropy, the 40-K transition escapes detection in polycrystalline samples.

At 1.7 K, the magnetization of $U(Ga_{0.98}Sn_{0.02})_3$ is a linear function of the applied field and does not exhibit any hysteresis effect (see inset to Fig. 2). In a field of 5 T the magnetization reaches only 0.193 emu/g, which corresponds to about $1.6 \times 10^{-2} \mu_B$/mole, i.e., the value found in UGa_3.[7]

All the above features show that a small ($\sim$2%) substitution of Sn for Ga does not change the basic aspects of the magnetic properties of UGa_3. We can thus safely assume that the AF type-II ordering observed for UGa_3 is retained in the $U(Ga_{0.98}Sn_{0.02})_3$ solid solution. This assumption was further supported by the Mössbauer data (see Sec. IV C): the small transferred hyperfine field felt by the ^{119}Sn nuclei and its perpendicular orientation with respect to the principal axis of the EFG are clear signatures of an AF type-II ordering (see Sec. II).

B. Quadrupole interaction, isomer shift, and Lamb-Mössbauer factor

The temperature dependence of ^{119}Sn Mössbauer spectra of $U(Ga_{0.98}Sn_{0.02})_3$ is shown in Fig. 3. In the paramagnetic state ($T > 73$ K), the spectra are characterized by a quadrupole split doublet whose linewidth $W \sim 1$ mm/s. This originates from the $4/mmm$ symmetry of the tin site which leads to an axially symmetric EFG. Due to the crystal symmetry, the principal component of this EFG is oriented along the normal of the face of the cube containing the considered Sn position (see Fig. 1). Within experimental errors the quadrupole splitting (QS) $= (1/2)e^2qQ = -1.68(3)$ mm/s is found to be constant from 290 K down to T_N (its negative sign was inferred from the data taken in the ordered state). QS in $U(Ga_{0.98}Sn_{0.02})_3$ is significantly larger than the values found in isostructural rare-earth Sn_3 (0.9–1.2 mm/s),[15] USn_3 (1.38 mm/s),[25] or $NpSn_3$ (1.23 mm/s) (Ref. 16) compounds. This indicates a different $5p$ electron distribution on Sn along these series of compounds. The isomer shift (δ_{IS}) of 2.15(3) mm/s (vs the $CaSnO_3$ source at 4.2 K) measured in $U(Ga_{0.98}Sn_{0.02})_3$ is smaller than the one observed in USn_3 ($\sim$2.45 mm/s), which is close to the value of β-tin (2.55 mm/s).[25] This behavior may be ascribed to a variation of the number of conduction electrons that increases with the tin content. The onset of magnetic order which shows up as a

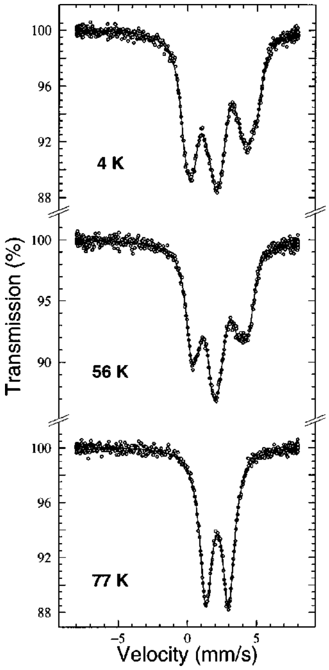

FIG. 3. ^{119}Sn Mössbauer spectra of $U(Ga_{0.98}Sn_{0.02})_3$ at variable temperatures. The full curves represent the least-squares fit to the data using the appropriate hyperfine Hamiltonian (see text).

broadening of the quadrupole doublet line shape appears at $T \sim 72\,K$. This temperature is in good agreement with T_N obtained from the susceptibility data (Fig. 2). As shown in Fig. 3 the spectral shape changes considerably below T_N. The analysis of the spectra taken in the ordered state will be considered in the next section. Here we will focus only on the temperature dependence of the spectral area $A(T)$ which is proportional to the Lamb-Mössbauer f factor. Figure 4 indicates that there is a large continuous increase in the f factor below T_N, while it exhibits a linear dependence above T_N. From the slope, $d\ln[A(T)/A(4.2\,K)]/dT = -6E_R/k_B\theta_D^2$, of the linear part one deduces a ‘‘Mössbauer’’ Debye temperature θ_D of 255(3) K using the atomic mass of ^{119}Sn for evaluating the recoil energy E_R, where k_B is Boltzmann’s constant. This θ_D value is very close to the one reported (250 K) for USn_3.[25] The unusual trend of the f factor below the ordering temperature, which physically means that the mean square displacement of the Sn atoms decreases, was already observed in other itinerant f-electron systems ($CeFe_2$,UFe_2).[26–28] Following the suggestion of Eremenko, Kaner, and Chercherskii[29] a strong magnon-phonon coupling could explain the large increase of the f factor. The study of the spin and lattice dynamical responses of UGa_3 by inelastic neutron scattering should give deeper insights into the mechanism responsible for the anomaly of the f factor.

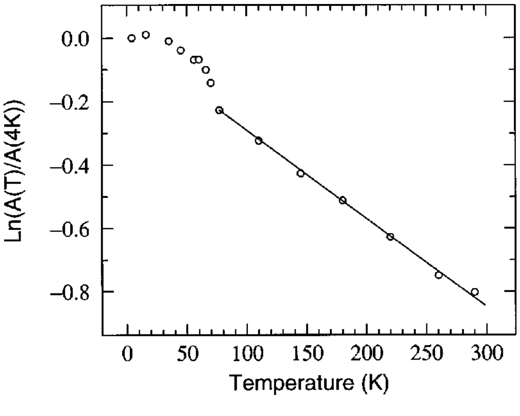

FIG. 4. Temperature dependence of the normalized spectral areas in $U(Ga_{0.98}Sn_{0.02})_3$. Notice the abrupt change of the Lamb-Mössbauer factor below T_N. The Debye temperature of 255 K was deduced from a fit of the linear part above T_N.

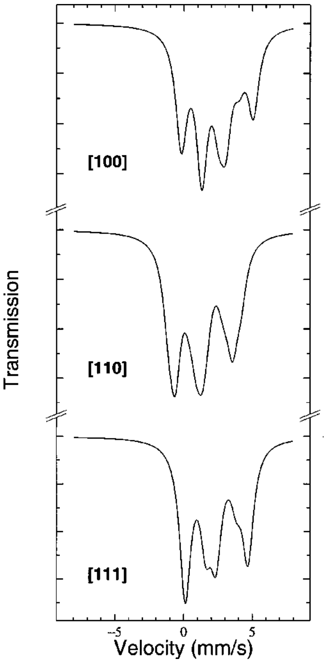

FIG. 5. ^{119}Sn Mössbauer spectral shape as a function of the orientation of the uranium moments along the principal directions of a cube. The hyperfine field was assumed to be perpendicular to the local EFG axis. The simulated spectra was computed with the following parameters: $H_{an} = 1.1\,T$; $\frac{1}{2}e^2qQ = -1.7\,mm/s$; $\delta_{IS} = 2.15\,mm/s$; $W = 1\,mm/s$ (see text and Table I).

C. Spin reorientation and spin density

The spectra recorded in the ordered state of $U(Ga_{0.98}Sn_{0.02})_3$ present complex line shapes (Fig. 3). In our computer data analysis, we have constrained several parameters: (i) all resonance widths were fixed to the value of 1 mm/s observed in the paramagnetic state, (ii) the isomer shift was set equal for all magnetically inequivalent tin sites, (iii) the quadrupole splitting was fixed to 1.68 mm/s, and (iv) the hyperfine field and the quadrupole interaction axes were assumed to be perpendicular. Attempts to fit the spectra following the model described in Sec. II with the U moments along one of the principal axes of a cube, i.e., $m \parallel [100]$, $[110]$, or $[111]$, proved to be unsuccessful. This is clearly demonstrated from a comparison between the experimental data (Fig. 3) and the simulated spectra shown in Fig. 5. Note, however, the strong similarity between the 4.2-K data and the spectrum calculated for $m \parallel [110]$. In a further step we considered the most general case, i.e., $m \parallel [uvw]$. This implies the occurrence of at most three magnetically inequivalent tin sites with relative intensities 1:1:1. Figure 6 shows that, from T_N down to 35 K, two of the three Sn sites feel the

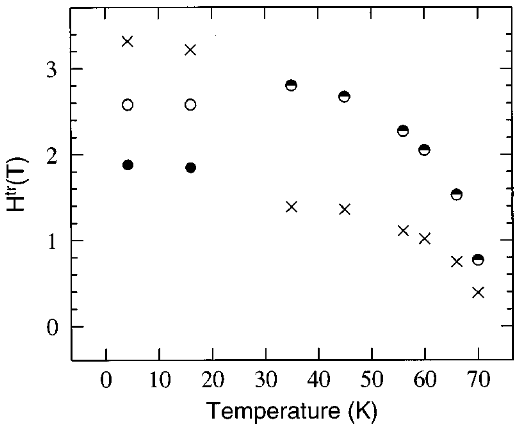

FIG. 6. Temperature dependence of the transferred hyperfine field at the three inequivalent Sn positions in U($Ga_{0.98}Sn_{0.02}$)$_3$. The error bars are smaller or equal to the size of the experimental points. From T_N down to ~35 K, two magnetic tin sites were observed. The intensity ratio of the high to low field component is 2:1 [position 1(×), positions 2 and 3 (◓)]. Below 35 K, the best fits were obtained assuming three equally populated magnetic sites [position 1(×), position 2(○), position 3(●)].

same hyperfine field. This means that the U moments should orient along a $[uuw]$ direction. Using the expressions given in Table I together with the data of Fig. 6 one deduces that $\theta \sim 28°$ and $\varphi = 45°$ ($m \| [338]$) when $35\,\mathrm{K} \leqslant T < T_N$. Below 35 K, the U moments undergo a spin reorientation: $\theta \sim 90°$ and $\varphi \sim 53°$ ($m \| [340]$); they are in the basal plane and slightly tilted (~8°) from the [110] direction. The saturated (4.2 K) hyperfine field values allow us to deduce $H_{\mathrm{an}} \sim 1.1\,\mathrm{T}$. It follows that the spin density f_{5p} transferred into the tin $5p$ orbitals amounts to 2%. Accordingly, the spin moment carried by the Sn atoms is about $0.02\mu_B$. At this stage, it is worth comparing our results to those obtained by ^{115}In nuclear quadrupole resonance measurement of the isostructural $CeIn_3$ compound.[30,31] In the ordered state of this type-II antiferromagnet, the In nuclei feel a hyperfine field of only 0.6 T and the observation of a single magnetic site means that the Ce moments ($\sim 0.5\mu_B$) orient along the [111] direction (see Table I). This leads to estimate that the spin density in the In $5p$ orbital amounts to only ~0.8% and to conclude to a strong decrease of the f-p hybridization from UGa_3 to $CeIn_3$.

In summary we have studied the magnetic properties of the U($Ga_{0.98}Sn_{0.02}$)$_3$ solid solution by ^{119}Sn Mössbauer spectroscopy. The transferred hyperfine field at the ^{119}Sn nuclei observed in the antiferromagnetic type-II phase has been shown to be *anisotropic* and directly connected to the strength of the f-p hybridization. Contrary to neutron diffraction measurements, the hyperfine data allow us to determine unambiguously the direction of the U moments. Our findings suggest that the transition observed in the vicinity of 40 K in UGa_3 is due to a spin reorientation with the U moment close to the [110] direction at low temperature. Finally, our results evidence a lattice ''hardening'' below T_N which may be due to a magnon-phonon interaction.

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