Dynamic spin correlations in $U(In_{1-x}Sn_x)_3$ heavy-fermion compounds: A perturbed angular correlation study

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By applying perturbed angular-correlation spectroscopy we have investigated spin correlations in the ''heavy-fermion'' compounds $U(In_{1-x}Sn_x)_3$, with $x=1.0$, 0.7, and 0.5, by measuring the induced magnetic hyperfine field B_{ind} at diamagnetic ^{111}Cd probe nuclei as a function of temperature and applied magnetic field. In zero applied field, the absence of any detectable magnetic hyperfine field at Cd reveals the absence of static magnetic correlations down to 4.2 K. However, from the field dependence of B_{ind} we find evidence for the presence of field-induced, short-ranged, and dynamic spin correlation between U f electrons at all compositions. The strength and dynamics of these correlated spins strongly depend on x, temperature, and applied magnetic field. As an important feature, for compositions near $x=0.5$, classified as a heavy-fermion material with electronic specific heat coefficient $\gamma=500$ mJ/mol K^2, these U spin correlations seem to set in from a relatively high temperature ($\geqslant 37$ K), and become very large on lowering temperature and/or increasing magnetic field, reflected in the measured Knight shift value K of about -32% at 4.2 K and $B_{app}=7$ T. We believe that these short-range spin correlations and their relaxation dynamics are responsible for the low temperature increase in magnetic susceptibility and electronic specific heat, previously considered to be an indication of heavy-fermion behavior in this system.

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I. INTRODUCTION

Strongly correlated electronic materials, especially the heavy-fermion systems, have been the focus of intense experimental and theoretical investigation in recent years.[1] As a characteristic feature these systems show a high value of the linear coefficient of specific heat ($\gamma > 200$–400 mJ/mol K^2, enhanced Pauli susceptibility (χ_0), and several other anomalous low temperature physical properties by which the effective electronic mass (m^*), as compared to normal metals, is enhanced by many orders of magnitude. Over the past several years a large number of compounds involving $4f$ (Ce, Yb) and light actinide elements (U, Np) have been discovered to show such behavior.[2] Key parameters influencing electron correlations in these materials are the degree of localization and the strength of hybridization between f and ligand electrons. Theoretically, it is generally accepted that in Ce-based systems heavy-fermion behavior arises mainly due to the Kondo effect, where the strong electron correlations arising from coherent many-body interactions between periodically arranged f ions (Kondo lattice) give rise to an enhanced effective electronic mass.[3] The origin of heavy fermions in U-based intermetallic compounds is more controversial, because the larger spatial extent of the $5f$ orbital has led to a debate over itinerant versus localized treatments.[4,5] Moreover, in the localized limit it is unclear whether U ions have a $5f^2$ (tetravalent) or $5f^3$ (trivalent) ground state configuration and if crystal-field excitations are relevant.[6] Notwithstanding these difficulties the heavy-fermion behavior in U systems is also discussed in terms of Kondo interaction, more recently as being due to the quadrupolar Kondo effect.[7]

Another question, intensively debated in recent times is the effect of spin fluctuations and/or spin correlations on low temperature physical properties of ''heavy-fermion'' materials. Over the years, extensive studies have been carried out in a large number of pseudobinary and pseudoternary intermetallic systems. It is generally observed that heavy-fermion behavior—with enhancement of γ and χ_0 as its telltale signals—usually occurs in alloys close to a magnetic to nonmagnetic phase boundary.[8] But precisely in these systems short-range magnetic interactions, either static or dynamic, are likely to be present. They can significantly influence the γ and χ_0 values, which in turn would make the identification of a true ''heavy-fermion'' material difficult. It is therefore important to investigate spin correlation in typical heavy-fermion alloys and examine its evolution as a function of temperature. In this connection, a number of microscopic investigations have been carried out using nuclear techniques such as neutron diffraction, NMR and muon spin resonance (μSR).[8–13] The results obtained in several well-established heavy-fermion compounds have revealed the presence of strong spin correlations, in some cases leading to magnetic ordering at very low temperatures.[8–13] In this respect, it is desirable to explore the possibility of using other microscopic techniques to study magnetic correlations and their relaxation dynamics in strongly correlated electron systems. For this purpose we have applied the perturbed angular correlation (PAC) technique to study static and dynamic spin correlations in some heavy-fermion compounds.

The basic principle of the PAC technique lies in the directional correlation of γ radiation emitted by nuclei with preferred spin orientations and its modification caused by hyperfine interactions between the nucleus and the electromagnetic fields produced by the surrounding atomic environ-

ment. Experimentally, a trace quantity of radioactive impurities is introduced into the samples under investigation and one measures the hyperfine fields acting on the daughter nuclei as a function of physical variables like temperature, pressure, concentration, etc. For the present investigation we have used ^{111}In which decays with a half-life of 2.83 days to ^{111}Cd via electron capture. The 247 keV, $I=5/2$ level in the daughter ^{111}Cd, having half-life $T_{1/2}=84$ ns, quadrupole moment $Q=0.84b$, and nuclear magnetic moment $\mu_I=0.765\mu_N$ (eV/T) serves as a suitable probe for the detection of electric and magnetic hyperfine fields, which in turn provide microscopic information on the magnetic interactions as well as electronic charge distributions in the sample under investigation. The experimental information is expressed by the quadrupole interaction frequency ω_0 (related to the principal component V_{zz} of the electric-field gradient tensor) and the Larmor precession frequency ω_L (related to the magnetic hyperfine field). In the discussion below, ω_L will sometimes be translated into hyperfine fields (T). Like other nuclear methods, e.g., Mössbauer spectroscopy, NMR, μSR, etc., the hyperfine parameters extracted from PAC measurements are very sensitive to details of the nearest-neighbor environment and are thus suitable for the study of short-range interactions. Furthermore, because of the short half-life of the probe state, the measured hyperfine fields are ideally suited to derive information on fast spin and charge processes in the solid. Further details on the PAC technique can be found elsewhere.[14,15]

As a case study we have applied this technique to investigate static and dynamic spin correlations in $U(In_{1-x}Sn_x)_3$. These intermetallic alloys crystallize in the cubic $AuCu_3$ structure, with U on the Au positions and In and Sn (almost) randomly distributed over the Cu sublattice. USn_3 is a well-established spin fluctuator[16] showing a large electronic specific heat coefficient $\gamma\sim 170$ mJ/mol K^2.[17] Upon substituting Sn for In, the system remains paramagnetic (at least down to 4.2 K) until the concentration $x=0.5$ has been reached. There a transition to long-range antiferromagnetic order occurs, with T_N increasing for decreasing x, up to $T_N\sim 108$ K for UIn_3.[17] This has been confirmed from microscopic studies using PAC (Refs. 18–20) and ^{119}Sn Mössbauer spectroscopies.[21,22] Being close to a magnetic transition, it was not a big surprise that in the intermediate composition range $0.5<x<0.8$ this system was classified as a heavy-fermion material, after measuring an enhanced electronic specific heat with the γ value exceeding 500 mJ/mol K^2.[17] In this paper we present results from our PAC study in the ''heavy-fermion'' alloys ($0.5<x<1.0$). In these macroscopically paramagnetic compounds, the PAC measurements, carried out as a function of temperature and external magnetic field, reveal a strong field-induced magnetic hyperfine field (B_{ind}) at the diamagnetic ^{111}Cd probe atoms, consistent with the presence of short-range dynamic correlations between the U moments. These spin correlations are most prominent for compositions near $x=0.5$, where the electronic specific heat coefficient γ reaches its maximum value. They can account for the increase of γ and χ, with no need to invoke a heavy-fermion state.

II. EXPERIMENTAL DETAILS

Polycrystalline samples of $U(In_{1-x}Sn_x)_3$ were prepared at the Institute for Transuranium Elements, Karlsruhe (Germany) by melting together stoichiometric quantities of high purity elements in an arc furnace under argon atmosphere. Care was taken to account for possible loss of In/Sn, which have low melting points. The samples were characterized by powder x-ray diffraction measurements and were confirmed to be single phase with the cubic $AuCu_3$ crystal structure. For PAC measurements, a trace amount of the parent ^{111}In activity was diffused into the samples by heating at 850 °C for 24 h in sealed quartz tubes. In order to reduce the loss of volatile In/Sn from the sample during the diffusion of ^{111}In radioactivity, the tube was first evacuated to high vacuum and then filled with Ar gas with a partial pressure of 0.25 atm. The hyperfine interaction parameters were extracted from the lifetime spectra of the 247 keV nuclear level in ^{111}Cd, recorded in a 90°/180° geometry using a setup consisting of four BaF_2 detectors and a standard slow-fast coincidence circuit having time resolution better than 800 ps. The perturbation factor $A_{22}G_{22}(t)$ in the angular correlation function $W(\theta,t)=\Sigma A_{kk}G_{kk}(t)P_k(\cos\theta)$ was obtained by constructing the appropriate ratio function, called the PAC time spectrum,

$$R(t)=\frac{2[W(180°,t)-W(90°,t)]}{[W(180°,t)+2W(90°,t)]}.$$

Measurements were performed in the temperature range of 4.2 K to 300 K and in external fields up to 7 T using a He cryostat fitted with a superconducting magnet.

III. RESULTS AND DISCUSSION

Figure 1 displays some typical PAC spectra for different Sn concentrations, recorded at 293 K and 4.2 K and in zero applied magnetic field. The room temperature spectra for all samples show nearly the full anisotropy and could be simulated with a unique randomly oriented electric-field gradient (V_{zz}). Considering the point symmetries of different atomic sites in the $AuCu_3$-type crystal structure, the electric-field gradient at the U site is expected to be zero in USn_3 (point group $m3m$). On the Sn site, the $4/mmm$ point group yields an axially symmetric field gradient, as observed [see Fig. 1(a)]. The same is also seen for UIn_3,[18] and therefore we can conclude that ^{111}Cd probes substitutionally occupy the position of In/Sn atoms for all x. This substitutional site occupation of the probe atom is also supported from considerations of the atomic size and electronegativity of Cd and In/Sn. In the pseudobinary alloys $U(In_{1-x}Sn_x)_3$, the PAC spectra were found to be weakly damped [Fig. 1(b)]. This reflects a small distribution in the quadrupole interaction frequency ω_0 ($\Delta\omega_0<3\%$), due to the disorder on the In/Sn sublattice. In addition, a small asymmetry parameter $\eta\sim 0.2$ was clearly visible. This has been shown previously[23] to be due to short-range chemical order of In and Sn atoms: more Sn than average is in the first neighbor shell of the Cd probe atom, less in the second, and so on. Upon cooling, the spectra continued

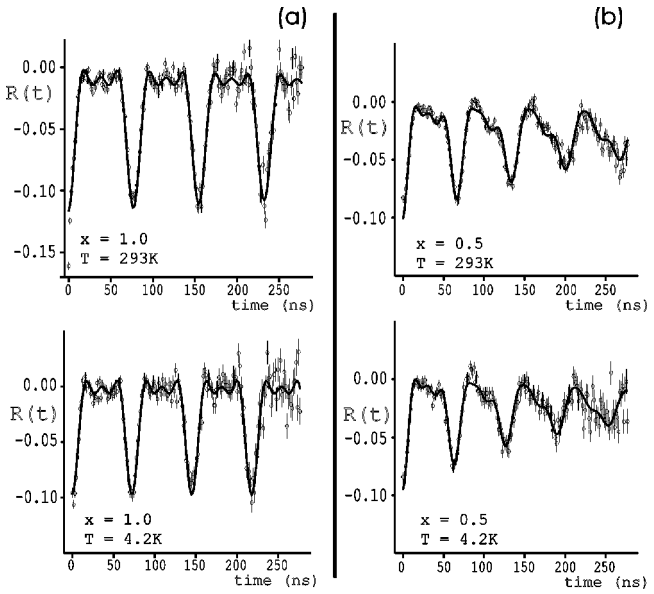

FIG. 1. Typical PAC time spectra taken at 4.2 K and 300 K under zero applied field for (a) USn_3 and (b) $U(In_{0.5}Sn_{0.5})_3$. Here, as in the other pictures, the vertical scale always spans 0.20 dimensionless units.

to exhibit pure quadrupole interaction with no indication of any measurable static magnetic hyperfine field down to 4.2 K, confirming the paramagnetic nature of the samples, previously inferred from bulk magnetization measurements.[17] The quadrupole interaction frequency ω_0 increased linearly with decreasing temperature with a weak anomaly near about 30 K for $x=0.5$ and $x=0.7$.[18] Further, for all x values studied here, the PAC spectra did not show any noticeable change in the damping down to the lowest measured temperature. All these features clearly indicate the absence of *static* magnetic correlations between U atoms. However, the presence of *dynamic* spin correlations cannot be ruled out. The minimal observable frequency for such correlated spins is about 12 MHz: faster processes will be averaged out during the half-life ($T_{1/2}=84$ ns) of the probe used in the present PAC experiment.

To examine whether or not dynamic spin correlations are present in the ''heavy-fermion'' $U(In_{1-x}Sn_x)_3$ system, we performed a number of PAC measurements under external magnetic fields varying up to 7 T and at temperatures ranging between 4.2 K and 137 K. Figure 2 shows typical $R(t)$ spectra for ^{111}Cd in $U(In_{1-x}Sn_x)_3$ ($x=0.5$, 0.7, and 1.0) at 4.2 K at different fields together with spectra recorded for the isostructural nonmagnetic compound YIn_3. In the presence of an externally applied magnetic field the diamagnetic ^{111}Cd probe nuclei in their noncubic surroundings experience combined electric and magnetic hyperfine interactions. The orientation of the electric-field gradients is random with respect to the detectors (polycrystalline sample). The effective magnetic field $\vec{B}_{\rm eff}$ at the nuclei is a vector sum of the applied field $\vec{B}_{app}$ with a unique orientation, and an induced hyperfine field $\vec{B}_{ind}$ of unknown orientation. The magnetic hyperfine field at Cd—mainly arising from the polarization of valence 5s electrons—has the following contributions: (i) spin polarization due to the applied magnetic field, and (ii) transferred hyperfine field caused by the magnetic moments of neighboring U atoms. For paramagnetic systems—as in the present case—with a U 5f spin correlation time much shorter than the measurement time scale (i.e., the half-life of the probe level), the transferred hyperfine field due to U magnetic moments can be expected to be negligible. The hyperfine field experienced by the Cd probe will then be $\vec{B}_{ind}=K\vec{B}_{app}$. Here, $\vec{B}_{ind}$ is essentially parallel to the applied field and K represents the Knight shift of the probe. It should be noted that for a Cd impurity the magnitude of K has been found to be quite small, being less than a few percent.[24] Moreover, since the Knight shift is proportional to the macroscopic susceptibility of the host matrix ($\chi=M/H$), the former is expected to be independent of the applied-field strength.

However, if short-range spin correlations are present, the response of the Cd Knight shift to the applied magnetic field will depend both on the degree of spatial correlations between U f electrons and on their relaxation dynamics. In such a case, the magnitude of $\vec{B}_{ind}$ and hence K can be expected to increase with decreasing spin-fluctuation rate. Also, depending on the size of this correlated spin system, $\vec{B}_{ind}$ and therefore K can become large and vary strongly with applied magnetic field, as seen, for example, in superparamagnetic clusters.[25] If the fluctuation rate is not too slow, we can as a first approximation still assume that $\vec{B}_{ind}$ and $\vec{B}_{app}$ are parallel. The variation of B_{ind} with temperature and B_{app} can thus provide crucial information on the extent and dynamics of spin correlations. The PAC time spectra were fitted for combined interactions using the computer code NNFIT,[15] assuming the presence of an effective ''applied'' field only ($B_{eff}=K'B_{app}+B_{ind}=KB_{app}$). This approximation holds for fast fluctuations, and becomes progressively worse when the spin fluctuations slow down. Acceptable fits were obtained even for the cases with slowest fluctuations, indicating that in the studied concentration, field, and temperature ranges this model is valid. The fitted quadrupole and Larmor frequencies ω_0 and ω_L are reported in Table I.

Figure 3 displays the variation of induced magnetic hyperfine field of Cd in USn_3 and $U(In_{0.5}Sn_{0.5})_3$ as a function of temperature and applied magnetic field along with the results obtained in YIn_3. In the nonmagnetic YIn_3, the $B_{\rm eff}$ experienced by Cd came out to be very close to the applied magnetic field, indicating a negligible induced magnetic hyperfine field B_{ind}. In pure USn_3 too, the spectra measured at high temperatures $T>37$ K and low applied fields yielded small values of B_{ind}. However, measurements carried out at 4.2 K (see Fig. 2) indicated the presence of non-negligible magnetic hyperfine fields at Cd whose magnitude became larger at higher applied fields, yielding $B_{ind}=-1.33(14)$ T at $B_{app}=7$ T, which corresponds to a Knight shift of -19%. To illustrate the size of this effect, in Fig. 2 the dotted line represents the best fit with B_{ind} constrained to zero. Considering that USn_3 remains paramagnetic down to 4.2 K as seen from our zero field PAC measurements and bulk magnetization data,[9,10] this large B_{ind} experienced by ^{111}Cd at high applied magnetic fields implies the appearance of strong field-induced spin correlations between U atoms, at

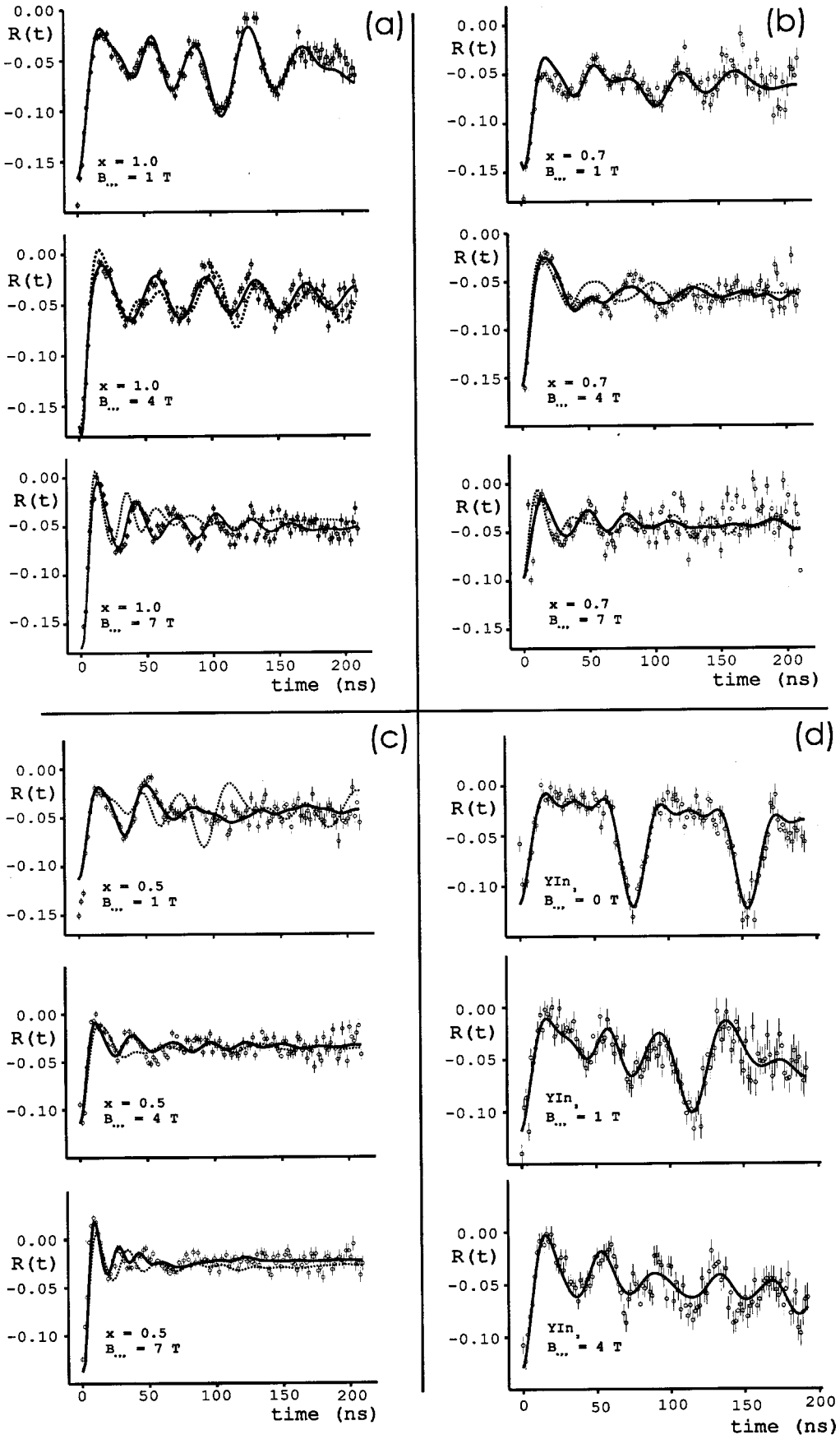

FIG. 2. PAC time spectra recorded as a function of applied magnetic field at 4.2 K. (a) USn_3, (b) $U(In_{0.3}Sn_{0.7})_3$, (c) $U(In_{0.5}Sn_{0.5})_3$, and (d) YIn_3. The dashed lines are the curves simulated with zero induced magnetic field, i.e., $B_{\mathrm{eff}}=B_{app}$ (not shown if coinciding with the actual fit).

TABLE I. Fitted quadrupole interaction frequencies ω_0 and Larmor frequencies ω_L. The fitting model assumes an effective applied magnetic field only, implying that induced internal fields are assumed to be parallel with the applied field. The effective field $B_{\rm eff}$ experienced by the nuclei and the induced field B_{ind} ($B_{\rm eff} = B_{app} + B_{ind}$, scalar sum), extracted from B_{app} and ω_L, are given too.

T (K)	B_{app} (T)	ω_0 (Mrad/s)	ω_L (Mrad/s)	$B_{\rm eff}$ (T)	B_{ind} (T)
		$x=1.0$			
293	0.0	80.1			
137	0.0	82.1			
	1.0	82.1	14.6	0.997	−0.003
	4.0	82.0	59.1	4.03	+0.03
	7.0	81.9	101.4	6.92	−0.08
37	0.0	86.1			
	1.0	86.1	14.7	1.005	+0.005
	4.0	86.0	60.4	4.12	+0.12
	7.0	84.2	100.1	6.83	−0.17
4.2	0.0	86.9			
	1.0	86.9	14.8	1.01	+0.01
	4.0	85.7	61.3	4.18	+0.18
	7.0	74.1	83.1	5.67	−1.33
		$x=0.7$			
4.2	0.0	93.8			
	1.0	93.3	18.6	1.27	+0.27
	4.0	80.2	66.0	4.50	+0.50
	7.0	59.1	72.7	4.96	−2.04
		$x=0.5$			
293	0.0	93.2			
137	0.0	95.4			
	1.0	96.4	15.0	1.02	+0.02
	4.0	93.2	59.1	4.03	+0.03
	7.0	91.6	101.5	6.92	−0.02
37	0.0	98.1			
	1.0	97.8	17.9	1.22	+0.22
	4.0	102.6	68.0	4.64	+0.64
	7.0	135.2	72.3	4.93	−2.07
4.2	0.0	96.7			
	1.0	98.1	27.4	1.87	+0.87
	4.0	112.9	84.0	5.72	+1.28
	7.0	167.3	69.7	4.75	−2.25
		YIn_3			
4.2	0.0	81.1			
	1.0	80.8	14.5	0.99	−0.01
	4.0	60.0	63.5	4.33	+0.03

least over microscopic length scales, whose dynamic relaxation rates are slower than the nuclear lifetime of the probe but fast compared to time scales of dc measurements. From band structure calculations it has been shown that in USn_3, a typical spin fluctuator with large Stoner enhancement factor[26] $I=3.03$, the U $5f$ electrons are on the threshold of being localized. The observations from the present PAC measurements indicate that at high magnetic fields U $5f$ spin fluctuations are significantly suppressed leading to strong,

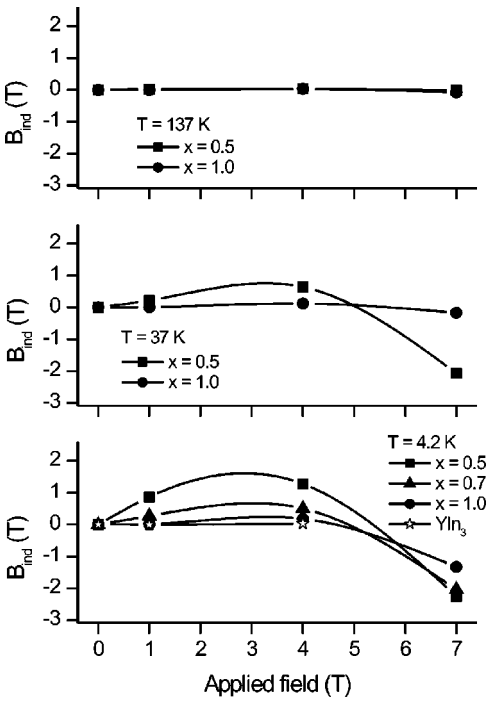

FIG. 3. Variation of the induced magnetic hyperfine field for ^{111}Cd in $U(In_{0.5}Sn_{0.5})_3$ (squares), $U(In_{0.3}Sn_{0.7})_3$ (triangles), and USn_3 (circles) at 137 K, 37 K, and 4.2 K. Data obtained in the nonmagnetic YIn_3 are shown as stars. The solid and dashed lines are guides to the eye.

most likely short-range spin correlations between U atoms, which give rise to a sizable induced magnetic hyperfine field at Cd. With partial substitution of Sn by In, the magnitude of B_{ind} increases quite substantially. As can be seen from Figs. 2 and 3, compared to pure USn_3, the spectra recorded for $x=0.5$ at all temperatures and applied fields yielded much higher values of B_{ind}. Figure 4 shows the variation of B_{ind} measured at 4.2 K and $B_{app}=7$ T as a function of x. It can be seen that the magnitude of B_{ind} increases from −1.33 T in USn_3 to a high −2.25 T in $U(In_{0.5}Sn_{0.5})_3$. For $x=0.5$, this large internal magnetic field is equivalent to a Knight shift as

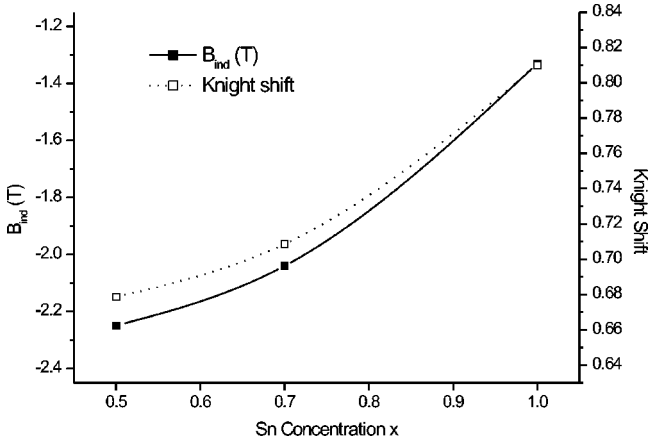

FIG. 4. Variation of the induced magnetic hyperfine field at ^{111}Cd (4.2 K, $B_{app}=7$ T) as a function of Sn concentration x in $U(In_{1-x}Sn_x)_3$ alloys. The right hand scale shows the changes in the Knight shift $K = B_{ind}/B_{app}$ (see text). The lines in the figure are guides to the eye.

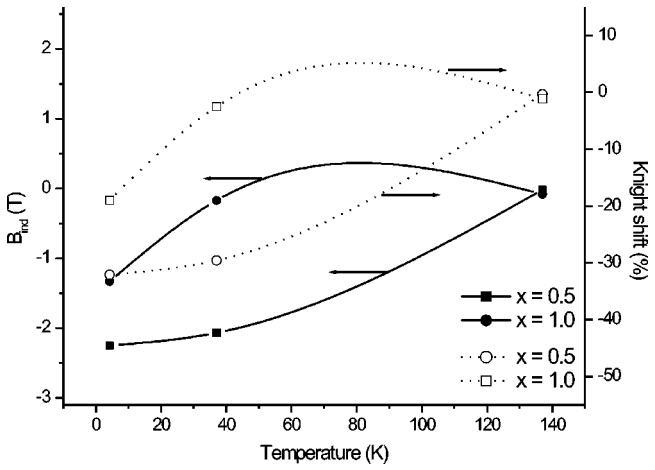

FIG. 5. Temperature dependence of B_{ind} and the corresponding Knight shift values for Cd in $U(In_{0.5}Sn_{0.5})_3$ and USn_3 at 7 T.

high as $K=-32\%$. This indicates that with increasing In concentration the spatial extent of spin correlations between U $5f$ electrons becomes large, and possibly that they also have slower relaxation times. In Fig. 5 we show the temperature variation of B_{ind} obtained from PAC measurements at $B_{app}=7$ T for $x=1.0$ and $x=0.5$. The result shows that, for a given composition, the magnitude of B_{ind} and hence the strength of U-U spin correlations systematically increases with decreasing temperature. It is also interesting to note that the variation of B_{ind} with applied field (see Fig. 3) shows oscillatory behavior, initially showing positive values and then becoming substantially negative at high magnetic fields. This variation of B_{ind} with applied magnetic field most probably arises due to field-induced modifications to the Ruderman-Kittel-Kasuya-Yosida (RKKY) type conduction electron polarization directly influencing the hyperfine field at the Cd probe site.

At this point it is worth mentioning that at low temperature and high magnetic fields the quadrupole interaction frequencies (ω_0) in all the samples came out quite different from the value obtained in zero field. For example, at 4.2 K the ω_0 for USn_3 was found to decrease from 86.9 Mrad/s at 0 T to 74.1 Mrad/s at $B_{app}=7$ T. A similar behavior was seen for $x=0.7$ and in YIn_3, while for $x=0.5$ at high fields there is an increase. The possibility that this could be an artifact of the fit model can be excluded: as soon as ω_0 was constrained to its zero field value, a satisfactory fit was impossible. As far as we know, such a large change in the electric-field gradient due to application of an external magnetic field has not been observed hitherto. It must be that in this compound the local density of states (DOS) at the Cd site is such that the shift between up-spin and down-spin DOS's due to the applied field favors a local electron density with an appreciably different deviation from cubic symmetry than without field. This point is currently being investigated in more detail.

The concentration and field dependence of B_{ind} in $U(In_{1-x}Sn_x)_3$ as seen from the present PAC measurements can be perceived to arise from superparamagnetic clusters of U spins. Considering that UIn_3 shows long-range antiferromagnetism and USn_3 is paramagnetic down to very low temperatures,[17] in the pseudobinary system $U(In_{1-x}Sn_x)_3$ the formation of such strongly correlated spin clusters can be conceived to arise due to the disorder on the In/Sn sublattice. Such disorder means that many of different local environments are present, which could in turn cause a wide distribution in the magnetic interaction between U magnetic moments. Depending on the size and morphology of these spin clusters, the system then has a broad spectrum of magnetic relaxation times and can remain paramagnetic down to very low temperature. However, over short measurement time scales its response to an external magnetic field can yield substantial hyperfine fields, similar to the behavior seen in many disordered magnetic systems.[25] On increasing the In concentration beyond $x=0.5$, the spin correlations become very strong and have a much longer relaxation time, leading to the appearance of long-range static magnetic ordering, which has been observed in both bulk and microscopic measurements.[17,18,21,22]

Finally, we examine the consequences of the presence of spin fluctuations for alloy compositions near $x=0.5$, the concentration region where the electronic specific heat coefficient γ has been shown to have the highest value (~ 500 mJ/mol K^2). From the results presented above, the PAC measurements reveal that near this concentration the induced magnetic hyperfine field and hence the spin correlations become quite strong, while nevertheless the fluctuation rate remains rather high ($\geqslant 12$ MHz) even at 4.2 K. As an important feature, it should be noted that at these compositions the induced magnetic hyperfine field at Cd starts to increase at temperatures as high as 37 K (see the data for $x=0.5$ in Figs. 3 and 5 and Table I]. This implies that the evolution of dynamic short-range spin correlations discussed before sets in at somewhat higher temperatures. With decreasing temperature, the dynamics of such correlated spins gradually slow down but do not show up as long-range magnetic ordering. The features observed in our PAC data are consistent with the results obtained from other microscopic measurements such as μSR and ^{111}Sn Mössbauer spectroscopy. Indeed, recent μSR measurements in $U(In_{1-x}Sn_x)_3$ reveal that for $x\sim 0.5$ the muon relaxation rate increases quite rapidly below 30 K.[27] This is also reflected in the temperature dependence of the ^{119}Sn Mössbauer linewidth, which increases in the same temperature region.[21,28] It is reasonable to expect that at low temperatures such dynamically coupled spin clusters can also give rise to enhanced specific heat and magnetic susceptibility. We thus tend to believe that dynamic spin correlations, seen by μSR, Mössbauer, and now also by PAC measurements, are the factors responsible for the large γ value and the concomitant apparent heavy-fermion behavior of these alloys, reported from low temperature bulk measurements. In the terminology of Geschneidner *et al.*,[29] this compound is a '' false heavy-fermion material,'' with the high γ caused by nonmagnetic atom disorder.

IV. SUMMARY

To summarize, we have applied the technique of perturbed angular correlation to study static and dynamic spin

correlations in the ''heavy-fermion'' system $U(In_{1-x}Sn_x)_3$. The concentration, temperature, and field dependences of the induced magnetic hyperfine field experienced by the diamagnetic ^{111}Cd probe nuclei clearly manifest the presence of short-range spin correlations which become quite strong near the compositions where the electronic specific heat coefficient γ reaches its maximum. The results presented in this work, together with earlier μSR and Mössbauer measurements, illustrate that spin correlations and their relaxation dynamics strongly influence the low temperature physical properties of the material. We believe the large enhancement of the γ and χ values—leading to the classification of $U(In_{1-x}Sn_x)_3$ as a heavy-fermion system—is mainly due to these spin fluctuations. The present work also demonstrates PAC as a powerful tool for microscopic investigation of heavy-fermion materials and strongly correlated electron systems in general. Further investigations in other intermetallic systems with different electronic mass enhancement factors are in progress.

ACKNOWLEDGMENTS

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