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Uranium moment collapse in magnetically diluted US

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Abstract

Muon spin rotation/relaxation (μSR) measurements were carried out on single crystalline specimens of $U_xLa_{1-x}S$ with $x = 1, 0.8, 0.55, 0.4, 0.15$ and 0 in the temperature range between 300 and 0.1 K. The limiting compounds US and LaS are ferromagnetic and diamagnetic, respectively. In the pseudo-binary system a collapse of the U magnetic moment was observed in bulk magnetic and neutron diffraction studies at $x < 0.6$. There neutrons no longer were able to detect magnetic order. Our μSR data verify the moment collapse below $x = 0.6$ but show that it leads to weak (between 0.3 and 0.05 μ_B), but not vanishing U moments. Furthermore, all componds except the diamagnetic LaS exhibit a magnetic transition with the transition temperature decreasing linearly with diminishing U content.

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Keywords: Muon spin rotation; Muon spin relaxation; Actinides; Localized moments

1. Introduction

Studies of the actinide compounds AnX (An = actinide) with the FCC NaCl structure have especially been instrumental in understanding the influences of ligand–5f electron hybridization on magnetic behavior [1]. Their simple crystal structure favors theoretical treatments [2]. The uranium monochalcogenides (US, USe, UTe) are collinear ferromagnets, while the corresponding monopnictides (UN, UP, UAs, USb, UBi) exhibit often fairly complex antiferromagnetic spin structures. In general, all the compounds show somewhat reduced magnetic moments (compared to the free-ion value of either U^{4+} or U^{3+}) which is thought to arise from the hybridization of the 5f electronic states with ligand electrons. The simple 'Hill picture' [3] based on 5f–5f overlap alone can be excluded on account of the fairly large An–An separation. Additional information can be obtained by extending the studies to diluted systems, where U is substituted in part by the diamagnetic La. The pseudo-binaries $U_xLa_{1-x}S$, in which the NaCl structure is maintained throughout, show quite unusual magnetic behavior. The results of previous

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bulk magnetic [4] and neutron diffraction [5] studies can be summarized as follows:

(1) The overall change of lattice constant is smooth and linear from 5.49 Å (US) to 5.87 Å (LaS). Neutron diffraction experiments suggest a possible irregularity just below $x = 0.6$, but the deviation is at the limit of resolution.
(2) The effective Curie–Weiss moment is practically independent of x at $\sim 2.25\ \mu_B$, while the paramagnetic Curie temperature decreases linearly from 184 K ($x = 1$) to -52 K ($x = 0.08$). The change in sign occurs at $x = 0.3$.
(3) The Curie temperature first decreases linearly from 177 K ($x = 1$) to ~ 100 K for $x = 0.6$. In this temperature range the ordered moment seen by neutrons changes little ($\mu_{ord} \approx 1.5\ \mu_B$). Below $x = 0.6$, no magnetic Bragg reflections could be detected any more by neutron diffraction, and it is concluded that the ordered moment has collapsed at least to a value $\mu_{ord} \leqslant 0.4\ \mu_B$, which is the sensitivity limit of the neutron study. The extrapolation of high field (9 T) magnetization data to zero field yields moments around 0.3–0.5 μ_B below the concentration region of collapse, but this procedure is considered unreliable due to domain effects.
(4) The specific heat shows a distinct anomaly around 100 K for $x = 0.6$ which is absent for $x = 0.55$ and below.

Some of these findings are summarized in Fig. 1.

The U moment collapse has theoretically been modelled by Cooper and Lin [6] on the basis of ab initio calculations within the frame of local density approximation. The essential feature is that the f electron spectral density at the U atom can be divided into two parts. One describes a localized f^3 configuration which is stable over a time span long enough to initiate magnetic order. The other refers to a rapidly fluctuating state between the f^3 and the non-magnetic delocalized f^2 configuration. This dynamic state is altogether inefficient in creating magnetism. Alloying with La (as well as changing the unit cell volume by applying pressure) increases the fluctuating part of the electron spectral density. At a certain critical concentration the

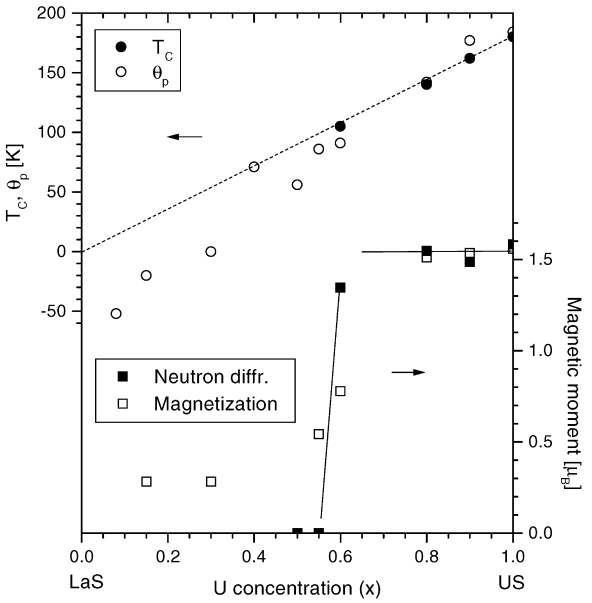

Fig. 1. Summary plot of the results from previous bulk magnetic and neutron diffraction work on $U_xLa_{1-x}S$. For details see text. (Figure adopted from Ref. [5].)

f^3 component falls below the critical value necessary to sustain a local magnetic moment and magnetic order. In the calculations the starting ratio of the relative weights of the static and dynamic portions in US is adjusted to the reduction of the local U moment with respect to the free ion value.

μSR spectroscopy has been shown to be especially effective for the study of small moment magnetism since it can resolve magnetic moments down to 0.001 μ_B in favorable cases. This feature has been exploited in particular for heavy fermion compounds where magnetic features connected to small Ce or U moments are encountered in several instances and may play a decisive role in the superconducting properties (see Ref. [7] for a comprehensive overview). It is natural then to attempt a study of the moment collapse in $U_xLa_{1-x}S$ using the μSR technique.

2. The μSR method

We briefly present some of the essentials of μSR spectroscopy in order to aid the reader in the

understanding of the experimental results to be discussed. A general introduction into μSR can be found, for example, in the monograph of Schenck [8] or the article by Chappert [9].

Spin-polarized positive muons are stopped in the sample under study. The appropriate muon beams are available at the so-called 'meson factories'. Due to its positive charge the muon comes to rest at an interstitial site, which in many cases is unique. There it is exposed to the magnetic field generated by surrounding dipoles, either of atomic or nuclear nature. Of interest to us are electronic moments. The field B_μ at the muon site arises primarily from the direct dipolar fields of the magnetic moments in the vicinity to which in conducting compounds a Fermi contact field adds. Furthermore, an external field may be present. The muon spin will precess in B_μ unless the field is oriented parallel to the spin. The positive muon decays with a mean lifetime of 2.2 μs by emitting a positron and two neutrinos. This decay process is governed by the weak interaction which fully violates parity. As a consequence, the emitted positrons show a distinct backward–forward asymmetry with respect to the momentary direction of the muon spin. Using appropriate detector arrangements and electronic circuitry the time dependence of this asymmetry is traced. It reveals the motion of the muon spin in B_μ. The data are usually expressed as

$$A(t) = A_0 G(t) \cos(2\pi \nu_\mu t).$$

A_0 is the initial asymmetry at $t = 0$, the moment when the muon has been stopped in the sample, often also referred to as the signal amplitude. Its value is somewhat dependent on details of the spectrometer. Values around 0.2 are typical. The factor $G(t)$ describes the loss of muon spin polarization in time (muon spin relaxation). The oscillating term is visible only if the muon spin precession is coherent, i.e., if the muons see a sufficiently well-defined value of B_μ. In zero applied field (ZF) this condition is usually fulfilled in an ordered magnet where the moments are spatially aligned. One uses the term 'spontaneous spin precession'. In a paramagnet the spins point in random directions. To induce spin precession one then needs an external field applied perpendicular to the initial direction of the muon spin. This is described by the term transverse field (TF) μSR. The precession frequency is always directly proportional to the interstitial field:

$$\nu_\mu = (\gamma_\mu/2\pi) B_\mu,$$

with the gyromagnetic ratio $\gamma_\mu/2\pi \approx 135$ MHz/T. In TF measurements the observed frequency ν_μ is shifted slightly from the value $\nu_0 = (\gamma_\mu/2\pi) B_{\mathrm{app}}$ because the internal field adds to the applied field. One defines the Knight shift $\Delta\nu/\nu_0 = (\nu_\mu - \nu_0)/\nu_0$. It is a measure of the local interstitial susceptibility and scales in simple cases with the bulk susceptibility.

In ZF the field at the muon site in a paramagnet is widely distributed around its mean $\langle B_\mu \rangle = 0$ due to the random orientation of the magnetic moments. Coherent spin precession cannot take place, and $A(t)$ now only contains the relaxation function $G(t)$ which in this case is known as the Kubo–Toyabe function. For the usual Gaussian distribution of internal field components, a Gaussian decay $G(t) = \exp[-\Delta^2 t^2/2]$ of the muon spin polarization is seen at early times. Here Δ is the second moment of the field distribution. At later times, $G(t)$ will first go through a minimum followed by a recovery to $A_0/3$ *if the field is static*. This is often called the '1/3 tail' and is a distinct feature of a static situation in the spin ensemble. If, however, the local field fluctuates, one observes a decay of the '1/3 tail' at low fluctuation rates, while at high rates the muon spin relaxation attains exponential shape, $G(t) = \exp[-\lambda t]$. The muon spin 'relaxation rate' is given by $\lambda = \Delta^2 \tau_c$ where $1/\tau_c$ is the field fluctuation frequency and thus τ_c the correlation time of moment fluctuations. In the static limit the Kubo–Toyabe type muon spin relaxation can be suppressed by applying a longitudinal field (LF). This effect, mostly referred to as 'decoupling', is very sensitive to spin fluctuations. At low values of $1/\tau_c$, decoupling is still possible but much higher fields than in the static limit are needed. At high fluctuation rates decoupling becomes impossible with the fields commonly available at μSR spectrometers (~ 0.5 T).

In summary, the parameters of importance that can be extracted from a μSR spectrum are (a) the precession frequency ν_μ either in ZF (if long-range order exists) or in TF (in the paramagnetic regime), (b) the muon spin relaxation rate λ or, in the static

case, the field distribution width Δ and (c) the initial asymmetry A_0, which gives information if all or only a portion of the muons stopped in the sample contribute to the detected signal. The decoupling (LF) field is another quantity of interest. We shall make use of all these parameters.

3. Previous μSR studies on related compounds

UX compounds with the NaCl structure have been studied by μSR previously in some detail in the case of the pnictides (X = N, P, As, Sb) and one chalcogenide (X = Te). (See, for example, the summary by Asch [10] or the general review on μSR in magnetism by Schenck and Gygax [11].) The results of relevance from the earlier studies for the present work are the verification that influences of muon diffusion cannot be seen at least up to 250 K and that the muon is stopped at a site of high local symmetry: the interstitial position in the center of the cube formed by four U and four X atoms (Fig. 2). This stopping site for the muon has an important consequence. In all cases, even if a small tetragonal distortion is present, the dipolar field contributions to B_μ cancel. In a ferromagnet (like US), the contact field will still be present on account of spontaneous magnetization. Hence it follows roughly the temperature dependence of the domain magnetization. The staggered magnetization in an antiferromagnet (like UN or UP, etc.) leads to the full cancellation of the contact field as well and the muon feels essentially no field at all. In practice, a weak field distribution around $\langle B_\mu \rangle = 0$ is present due to dilute faults in the spin lattice (see Ref. [10]). UTe is the only mono-chalcogenide which has been studied previously [12]. A thick sample consisting of randomly oriented single crystal platelets was used. A strong rise in the paramagnetic frequency shift was seen on approach to $T_C = 104$ K from above. The temperature dependence of the shift followed well the Curie–Weiss behavior of the bulk susceptibility,

$$\frac{\Delta\nu}{\nu_0}(T) = [(T - T_C)/T_C]^w$$

with $w = -1$. Concomitantly, the muon spin relaxation rate λ increases as well due to the formation of paramagnetic spin correlations which slow down spin fluctuations. The temperature dependence $\lambda(T)$ can also be described by a critical power law, but the increase is steeper, resulting in $w = -1.3$. This somewhat unusual value remains unexplained. No spontaneous spin precession signal could be detected below T_C. Muon spin relaxation must be so fast that the signal is damped out within the initial dead time of the μSR spectrometer. In this earlier experiment a high-energy muon beam (a so-called decay beam) was used which leads to a somewhat bigger dead time than the low-energy muon beam (surface beam) used in the present study.

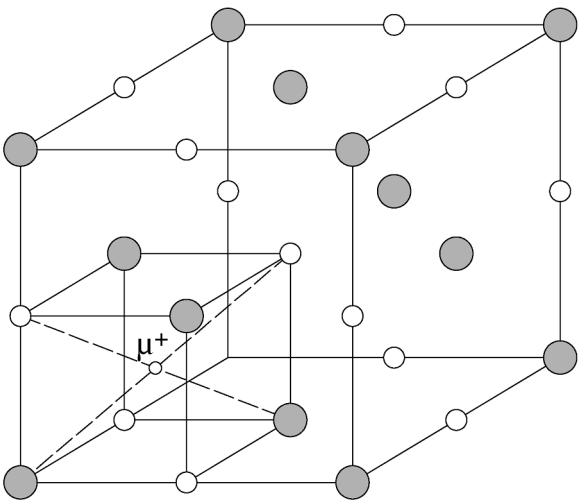

Fig. 2. Muon stopping site in UX compounds with the NaCl structure.

4. Experimental

The compounds were prepared using the methods described in Ref. [13] Small platelets of single crystalline material were glued to a silver backing, forming a thin mosaic sample. The platelets were oriented such that two of the three cubic [1 0 0] axes were in the plane of the sample and the third parallel to the beam direction. The πM3 beam at PSI with the general purpose spectrometer (GPS) was used. The πM3 beam provides 100%

spin-polarized positive muons with fairly low energy. These muons were safely stopped inside the thin single crystalline platelets. As usual, some muons are stopped in the sample holder (silver) rather than in the sample itself. They produced a background signal which was taken care of in the data analysis. This is fairly straightforward in the paramagnetic regime as pure silver does not cause muon spin depolarization at any temperature and hence is easy to handle. In the ferromagnetic regime, however, the remanent magnetization of the sample produces a weak field at the sample backing or distorts the applied field. In this regime background corrections are somewhat less reliable. This distortion of the background signal, however, can be taken as another indicator for the onset of ferromagnetism. The GPS uses an He flow cryostat allowing sample temperatures between 300 and ~ 2 K. For the highly dilute sample ($x = 0.15$) the measurements were extended using the low-temperature facility (LTF) at the same beam line, which comprises a ^{3}He/^{4}He dilution refrigerator. All data were analyzed using the least-squares fit program MSRFIT developed by J. Brewer at TRIUMF.

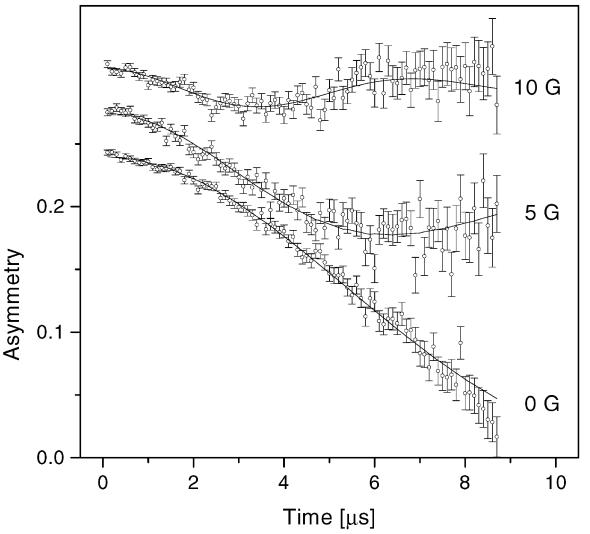

Fig. 3. Zero and LF spectra of LaS at 4 K. The different spectra are shifted vertically for clarity. In reality, all spectra show the same initial asymmetry $A_0 = 0.24$. The solid lines are fits to a static Gaussian Kubo–Toyabe function with $\Delta = 0.138\ \mu s^{-1}$. The characteristic minimum and the recovery to $A_0/3$ occur at late times outside the active time range of the μSR spectrometer.

5. Results

We shall first report the results for the two limiting compounds LaS and US to establish a frame for the data of the pseudo-binary system $U_xLa_{1-x}S$ which then will be presented in descending order of U concentration x.

LaS: ZF and low LF spectra of diamagnetic LaS recorded at 4 K are shown in Fig. 3. The relaxation is governed by a static Gaussian Kubo–Toyabe function, which arises from the nuclear dipoles on ^{139}La ($\sim 100\%$ abundant). The static width $\Delta = 0.138\ \mu s^{-1}$ decreases somewhat (by $\sim 5\%$) on warming to room temperature. This is easily explained by thermal expansion. The Kubo–Toyabe spectra can fully be decoupled by an LF of 10 G, which confirms their expected static nature. The data further confirm that muon diffusion need not be considered over the full temperature range.

US: The known Curie temperature of $T_C = 177$ K is easily visible in the TF spectral parameters shown in Fig. 4a. Both, the precession frequency ν and the muon spin relaxation rate λ in applied field rise sharply on approach to T_C. This is the expected behavior in a strong paramagnet when moving towards a second-order magnetic transition. It is due to the formation of spin–spin correlations and the concomitant slowing down of spin fluctuations (see Ref. [14] for a discussion on these features). The signal amplitude A_0 (initial asymmetry) in TF breaks down at T_C as a consequence of the now present local field caused by the appearance of spontaneous magnetization. The combination of external and internal fields rises the width of field distribution and thus results in a sharp increase of muon spin relaxation rate, rendering the signal unobservable by shifting it into the region of initial spectrometer dead time.

In ZF, the expected exponential decay of muon spin polarization is observed above T_C, while well below T_C we found a spontaneous spin precession pattern (Fig. 5). The precession frequency at 100 K is $\nu_\mu = 63.4$ MHz which corresponds to an internal field of 4.7 kG. The signal is rather strongly damped by muon spin relaxation with a rate of

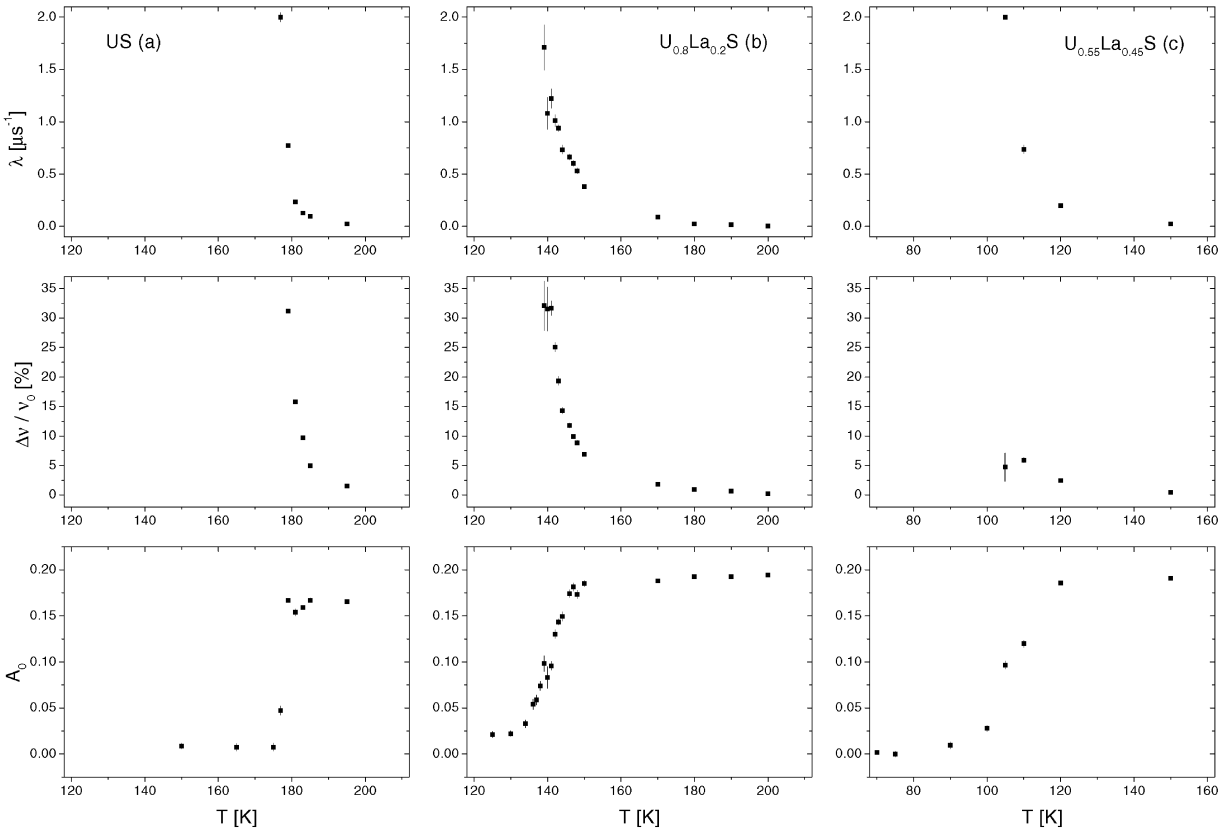

Fig. 4. Temperature dependencies of the muon spin relaxation rate λ, frequency shift $\Delta\nu/\nu_0$ and initial asymmetry A_0 (from top to bottom) obtained in low transverse field (TF = 50 G) measurements of (a) US, (b) $U_{0.8}La_{0.2}S$ and (c) $U_{0.55}La_{0.45}S$.

$\sim 23\,\mu s^{-1}$. The initial asymmetry is slightly reduced (by $\sim 10\%$) compared to the paramagnetic signal. This is a common observation and means that small parts of the sample (i.e., domain walls, surfaces, etc.) deviate from the proper spin structure of the bulk.

$U_{0.8}La_{0.2}S$: The TF results for this compound are shown in Fig. 4b. They are quite similar to those of pure US except for the trivial shift in transition temperature which agrees with the neutron data. Unfortunately, no spontaneous spin precession could be detected below T_C. The dilution of U causes a disorder in the configuration of magnetic moments around the muon leading to an increase of the muon spin relaxation rate which had already been high in ferromagnetic US. In consequence, the precession signal is damped out very quickly ($\lambda > 100\,\mu s^{-1}$) and vanishes in the initial dead time of the spectrometer. The situation worsens for the higher dilutions and spontaneous spin precession was not found in any of the quasi-binary compounds.

$U_{0.55}La_{0.45}S$: According to the neutron data this compound already exhibits moment collapse. The μSR spectral parameters derived from TF measurements (Fig. 4c) clearly show a magnetic transition around 110 K. Two differences to US are apparent. Firstly, the peak value of the frequency shift near T_C is greatly reduced, while for $x = 0.8$ practically no change had been observed in this respect. Secondly, the temperature region over which the TF signal loses its initial asymmetry is

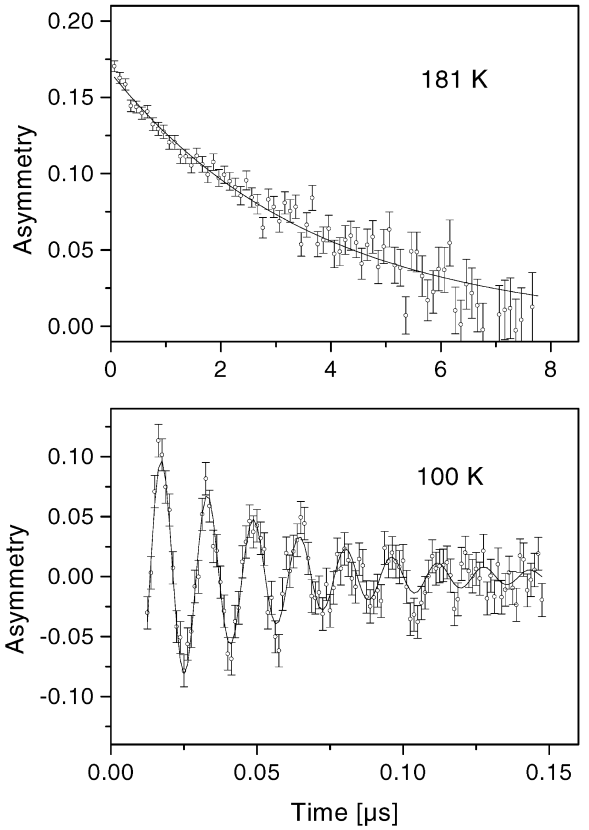

Fig. 5. ZF spectra of US. Relaxation spectrum at 181 K, slightly above T_C (top) and spontaneous spin precession signal at 100 K, well below T_C (bottom).

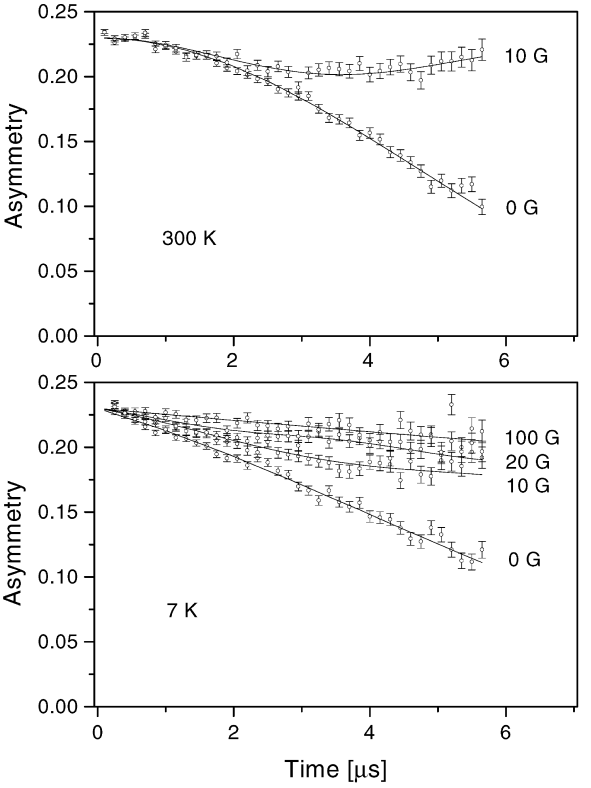

Fig. 6. Zero and longitudinal field spectra of $U_{0.15}La_{0.85}S$ at 300 K (top) and 7 K (bottom). While the 300 K data can be fitted with a Gaussian Kubo–Toyabe function arising from ^{139}La nuclear moments, the 7 K spectra require a fit using electronic–nuclear double relaxation. In LF the static width of the (nuclear) Kubo–Toyabe function was kept constant while the electronic relaxation rate was allowed to vary slightly with field.

significantly wider. This trend was already visible for $x = 0.8$ but has now become more pronounced. The diluted sample no longer shows a sharp transition into the magnetically ordered state. Different parts of the material order at slightly different temperatures. This might also reflect a small variation in U concentration in the different platelets comprising the μSR sample.

$U_{0.4}La_{0.6}S$: Peaking of the relaxation rate λ and of the precession frequency ν in applied field together with a loss of TF signal strength A_0 show that a magnetic transition still takes place at around 60 K. The temperature range of the transition is further widened and the peak frequency shift further reduced. Otherwise, no fundamental difference to the case $x = 0.55$ is found.

$U_{0.15}La_{0.85}S$: As stated in Ref. [4], this pseudobinary compound is at the percolation limit and the question whether a magnetic transition occurs is of particular interest. The high temperature (300 K) spectra in ZF and LF = 10 G are not significantly different from those of LaS. The muon spin relaxation is dominated by the action of static ^{139}La nuclear dipoles and can fully be decoupled in LF = 10 G (Fig. 6, top). The situation changes quite remarkably at 7 K (Fig. 6, bottom). The ZF spectrum can no longer be described by a Gaussian Kubo–Toyabe function. For example, it no longer shows the initial Gaussian decay seen at 300 K. To

reproduce the spectrum properly one has to use the formalism of electronic–nuclear double relaxation, which combines electronic exponential relaxation with nuclear Kubo–Toyabe relaxation (see, for example, Ref. [15] for more details). The essential conclusion is that moments on U are still present and make themselves felt in the μSR magnetic response. While the static nuclear part is easily decoupled in LF, the dynamic electronic part resists decoupling to much higher fields as should be the case. At high temperatures (300 K) the U electronic moments fluctuate rapidly and in this fast dynamic limit their influence on muon spin relaxation is hardly detectable. This is the well-known phenomenon called 'motional narrowing' in NMR. At low-temperatures spin–spin correlations develop and the spin fluctuations slow down, thus becoming more effective for muon spin relaxation. For the double relaxation fits shown in Fig. 6, bottom, a decrease of the electronic relaxation rate λ with increasing applied longitudinal field B_L had to be allowed for, while the static width due to nuclear moments was kept the same at all fields. The decrease of the electronic relaxation rate was found to be 50% at a longitudinal field $B_L = 30$ G. The field dependence of dynamic relaxation can be described by

$$\lambda(B_L) = \lambda(0)/[1 + (\gamma_\mu B_L \tau_c)^2]$$

(see Ref. [16]). Inserting our results gives $1/\tau_c \approx 3$ MHz, a very low frequency, already close to the static situation. Since the overall relaxation of muon spins is nevertheless fairly weak, one can immediately conclude that the U moments involved must be small. The influence of the U moments rises on further cooling. At 2.2 K it fully dominates muon spin relaxation and leads to an exponential relaxation shape. An indication of a magnetic transition was not seen, however.

Additional TF measurements at 1000 G were carried out at the LTF facility down to 0.1 K. The results are presented in Fig. 7. The temperature variations of the muon spin relaxation rate and of the frequency shift indicate a magnetic transition at around 0.45 K. The signal amplitude does not break down, however. We shall discuss this observation and the μSR information on the magnetic transition in general in the next section.

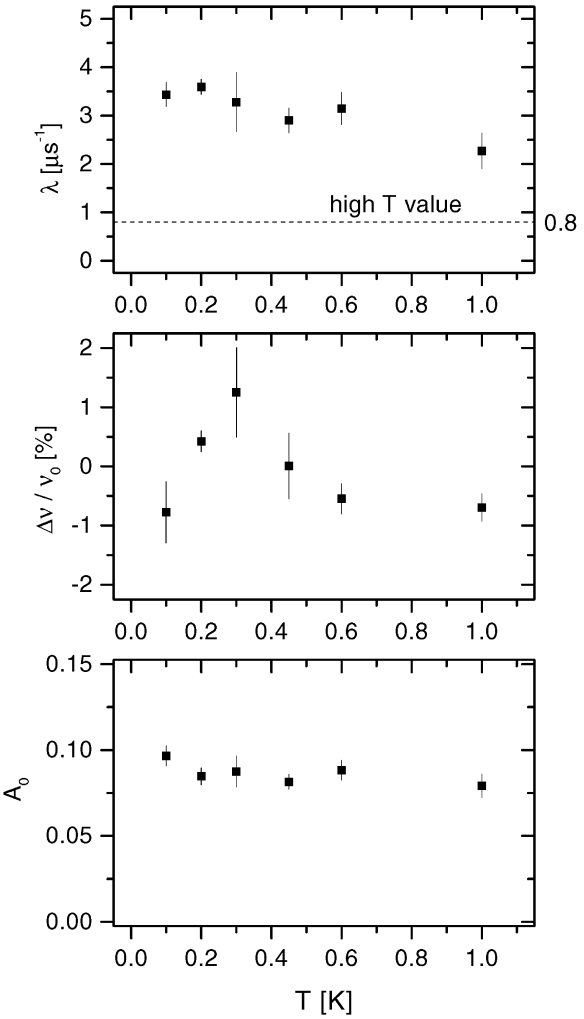

Fig. 7. Transverse field (1 kG) μSR results on $U_{0.15}La_{0.85}S$ at very low temperatures. From top to bottom: muon spin relaxation rate λ, frequency shift $\Delta\nu/\nu_0$ and initial asymmetry A_0. The sample was a single platelet and thus the signal strength is comparatively small.

6. Discussion

The μSR results for the pure compounds LaS and US reflect the known properties of those materials and offer no surprises. The important result for this study is that they confirm that μSR correctly senses the magnetism in this series of compounds. The data on US may be compared to the earlier results on UTe [12]. Very good agreement is

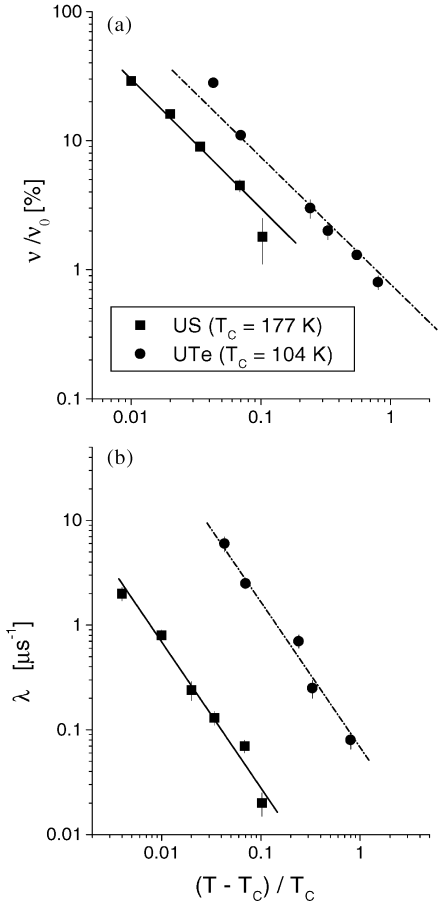

Fig. 8. Fit of the temperature dependencies of (a) the paramagnetic Knight shift and (b) the muon spin relaxation rate for US and UTe to a critical power law $[(T - T_C)/T_C]^w$. The exponent is $w = -1$ for the shift and $w \approx -1.3$ for the relaxation rate. The data for UTe were taken from Ref. [12].

obtained as presented in Fig. 8. It was mentioned that the temperature dependence of the paramagnetic frequency shift $\Delta\nu/\nu_0$ as well as that of the paramagnetic muon spin relaxation rate λ in UTe could be expressed satisfactorily in terms of a critical power law $[(T - T_C)/T_C]^w$ with $w = -1$ for $\Delta\nu/\nu_0$ and $w = -1.3$ for λ. The same results were obtained for US, as Fig. 8 demonstrates. The power $w = -1$ simply means that the paramagnetic frequency shift follows the Curie–Weiss law of the bulk susceptibility. The power $w = -1.3$ for the relaxation rate is somewhat unusual and we are not aware of any theoretical model which would predict this value. One expects values on the order of -0.5 [17] and such a value has been seen, for example, in Gd metal [18] WAK (except very close to T_C where it reduces to -0.15). In the ferromagnetic cubic Laves phase compounds RAl_2 (R = rare-earth) values between -0.35 and -0.7 were found [19]. In any case, the similarity of the results for UTe and US proves that with the high magnitude of w an intrinsic property is observed. One may thus conclude that the magnetic correlations which lead to this value of w must be the same in the two monochalcogenides.

Since no spontaneous spin precession was seen in UTe, we cannot compare the values of B_μ for the two materials. The field seen in US for $T = 100\,\mathrm{K} \ll T_C$ with good accuracy represents the saturation contact field. Its value of 4.7 kG is well in keeping with the results obtained for rare-earth elements or intermetallics. In Gd metal the saturation contact field is ~ 7 kG [20] in the RAl_2 intermetallics it is around ~ 1 kG for low-moment cases like $NdAl_2$ [19]. A reliable theoretical calculation procedure for contact fields does not exist, since the muon, due to its positive charge, enhances the conduction electron density and thus the spin density in its surroundings. The failure to observe a spontaneous spin precession in UTe is easy to understand when looking at the data shown in Fig. 8b. Even the paramagnetic muon spin relaxation rate for the UTe sample is about one order of magnitude higher than in the sample of US. This is probably due to a much wider distribution of fields on account of the large number of non-oriented platelets. The larger relaxation rate will damp the spontaneous spin precession signal too quickly to be observed, especially in connection with the poorer time resolution of the μSR spectrometer used in the UTe study.

The failure to observe spontaneous spin precession in the pseudo-binaries $U_xLa_{1-x}S$ forced us to use a different parameter as a measure of the local contact field at the muon site and in turn of the strength of the moments. At least a rough estimate of the moment can be drawn from the frequency shift $\Delta\nu/\nu_0$ very near T_C. We used the maximum observed value of $\Delta\nu/\nu_0$ which occurs about midway in the decrease of the initial asymmetry A_0 around T_C (see Fig. 4). This quantity

might be called 'frequency shift at T_C' although strictly speaking the shift should diverge at T_C. Since the shift follows the bulk susceptibility this is a coarse but valid approach. The concentration dependence of the U local moment derived in this fashion is plotted together with the concomitant variation of magnetic transition temperatures (defined by the midway point of the loss of TF signal amplitude) in Fig. 9. The collapse of moment between $x = 0.6$ and 0.5 is quite evident. It does not, however, lead to vanishing moments. There is still a resolvable frequency shift for $x \leqslant 0.55$ and in particular the low-temperature spectral shape for the most diluted sample, $U_{0.15}La_{0.85}S$, definitely proves the existence of a finite though small moment on U.

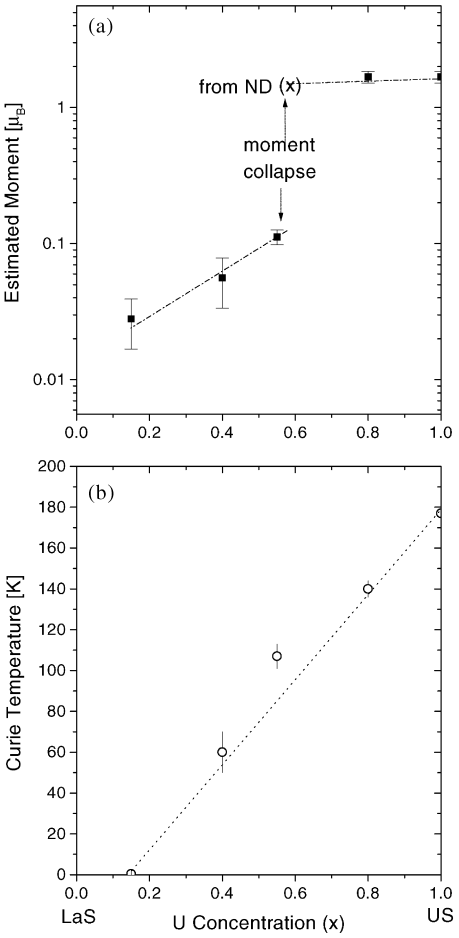

Fig. 9. Temperature dependencies of (a) the magnetic moment on U (estimated from the maximum frequency shift) and (b) the magnetic transition temperatures for $U_xLa_{1-x}S$. Note the logarithmic scale for (a). The transition temperature for $x = 0.15$ in (b) is 0.45 K.

Furthermore, we also observe magnetic transitions in the compounds with $x \leqslant 0.55$. An important question is the nature of this transition in the material with $x = 0.15$. According to bulk measurements the paramagnetic Curie temperature at this concentration is negative. This means that there should be a transition into an antiferromagnetic rather than a ferromagnetic state. In the TF data (Fig. 7) a loss of signal amplitude A_0 (initial asymmetry) is not observed in contrast to the data on compounds with higher U concentrations and positive paramagnetic Curie temperatures. If an antiferromagnetic structure exists, the mean field at the muon site is $\langle B_\mu \rangle = 0$. This means the muon spin precession signal in an applied field can be observed without difficulty, as has been demonstrated for the antiferromagnetic uranium monopnictides (see, for example, Ref. [21]). In a ferromagnet with the same simple cubic structure this is not the case due to the presence of a contact field. Hence, our data are in keeping with antiferromagnetic order. We cannot, however, definitely proclaim that the transition at 0.45 K is a true phase transition to a long-range ordered magnetic state. It could as well be a spin freezing temperature and the magnetic state a spin glass like state with dominating antiferromagnetic spin couplings.

With respect to the theoretical study described in Ref. [6] we conclude that the loss of moment is properly reproduced. But even for the region of dominantly delocalized f electron spectral density the coupling to initiate magnetic order is not broken. Extrapolating linearly the concentration dependence of the magnetic transition temperature (Fig. 9) towards $T_C \to 0$ yields a loss of magnetic order at $x \approx 0.13$. This is a concentration very close to the percolation limit.

7. Conclusions

The collapse of the U magnetic moment in $U_xLa_{1-x}S$ seen in bulk magnetic and neutron diffraction data for $x \geqslant 0.55$ has been basically verified by μSR. These latter data, however, clearly

demonstrate that the moments are not reduced to zero, but rather to small magnitudes on the order between 0.1 and 0.01 μ_B. These moments are smaller than those derived from high-field magnetization data extrapolated to zero field (Fig. 1). That procedure had been considered unreliable in Ref. [4] on account of the presence of domain structure. The μSR data show that this problem exists, but does not lead to fundamentally wrong results in the present case. In addition it is shown that all samples down to the lowest concentration measured ($x = 0.15$) exhibit a magnetic transition. The transition temperature varies linearly with concentration x. At $x = 0.15$ the μSR result is compatible with antiferromagnetic spin coupling as suggested by the negative paramagnetic Curie temperature.

The results on pure US were compared to earlier data on UTe. It was found that in both materials the temperature dependencies of the paramagnetic frequency shift and the muon spin relaxation rate follow a critical power law on approaching the Curie point from above with similar critical exponents ($w = -1$ for $\Delta\nu$ and $w = -1.3$ for λ). The former exponent just reflects Curie–Weiss behavior, but the size of the latter is unusual and remains unexplained. The similarity of exponents in US and UTe indicates that they reflect the same type of spin coupling in both compounds. A spontaneous spin precession was observed in US well below T_C, giving a saturation hyperfine field of 4.7 kG.

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