Title

A MU-SR STUDY OF THE 1-K PHASE-TRANSITION IN THE HEAVY ELECTRON COMPOUND UCU5

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Recent applications of μ^+SR in magnetism: Novel magnetic features in *heavy electron* compounds

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This paper is dedicated to Prof. G. zu Putlitz on the occasion of his 60th birthday

Abstract. As an example for the power of μ^+SR-spectroscopy in solid state physics three applications to heavy electron systems, i.e. $CeA\ell_3$, $U_{1-x}Th_xBe_{13}$ and UCu_5 will be discussed. Each of these systems reveals very specific magnetic features of unusual characteristics which involve very small to extremely small magnetic moments and random or very short range magnetic order. This kind of small moment magnetic order can be studied relatively easily by μSR but will be hardly accessible by other methods such as neutron scattering.

1 Introduction

The fact that the μ^+ is a spin-1/2 particle and possesses a relatively large magnetic moment renders it a particularly well adapted probe for the investigation of magnetic phenomena in solids, especially weak ones, in view of the absence of any electrostatic quadrupolar interaction. In metals, due to its positive charge, the μ^+ is expected to reside in general at the center of an interstitial site (exhibiting, of course, a relatively large zero point vibration amplitude), where it interacts predominantly with conduction electrons and other less localized electron states, and therefore probes magnetism from a different perspective as compared with a substitutional probe as in nuclear magnetic resonance (NMR) and Mössbauer spectroscopy. Magnetic interactions of the implanted spin-polarized μ^+ are communicated to the outside world by means of the parity violating asymmetric emission of positrons from their decay (30% effect!) which allows to monitor the time evolution of the muon's spin auto correlation function, $< \mathbf{S}(t) \cdot \mathbf{S}(o) >$, quite easily, provided the involved time scale is compatible with the μ^+ lifetime of 2.2 μs [1]. Fortunately this latter requirement is in a majority of cases fullfilled.

Experiments can be performed with or without an external magnetic field $\mathbf{H}_{ext}$. Application of a transverse field ($\mathbf{H}_{ext} \perp \mathbf{S}(o)$) allows to observe the μ^+ Larmor precession and associated relaxation phenomena, a very important one being dephasing by a static inhomogeneous magnetic field distribution inside the specimen. Application of a longitudinal field ($\mathbf{H}_{ext} \parallel \mathbf{S}(o)$) allows to decouple the μ^+ spin from internal static field components and μ^+-spin relaxation is then only induced by fluctuating internal fields arising from the dynamical features of the host lattice magnetism (spin lattice relaxation). Of particular interest in certain cases is the possibility to study both static and dynamic aspects of internal fields in the absence of an external field which otherwise might interfere with and disturb the intrinsic magnetism. This proved to be of great importance in the study of spin glasses [2]. In ferro- or antiferromagnetically ordered systems one often finds a well defined internal field (e.g. the contact hyperfine field and or dipolar fields originating from the ordered moments) at a given type of interstitial site which then assumes the role of an external field and a well resolvable μ^+ Larmor-precession may become observable [3].

It is therefore no surprise that muon spin rotation spectroscopy (μSR) has found its widest application in the field of magnetism or in fields in which magnetic properties are an important feature, as e.g. in high temperature superconductivity [4]. In this contribution we would like to review applications to a particularly interesting family of compounds which are called **heavy electron** or **heavy fermion** systems and which, as it turned out, possess a wide variety of complex magnetic properties.

Heavy electron compounds are intermetallic compounds containing rare earth (Ce, Yb) or actinide (U, Np) elements and simple or transition metal elements [5]. Their most conspicuous feature is the occurance of a huge electronic specific heat at low temperatures (the Sommerfeld constant γ may assume values up to the order of severel J/mol K^2) and certain similarities to liquid ^{3}He suggesting that these systems may be described in terms of Fermi liquid theory, involving very large effec-

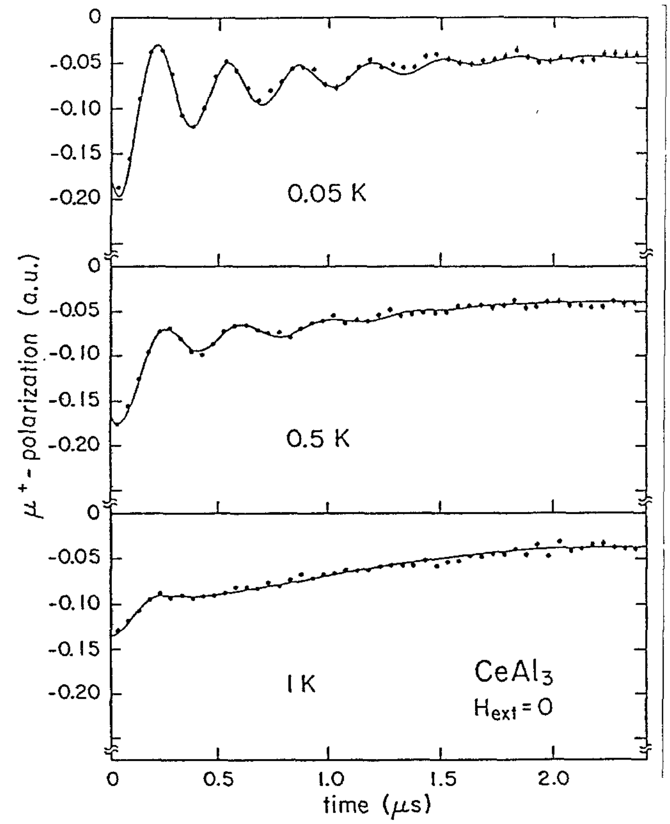

Fig. 1. ZF-μSR signal in $CeA\ell_3$ at three different temperatures. The solid lines represent three component fits. The low temperature frequency corresponds to an internal field of ~220 G (from ref. 13).

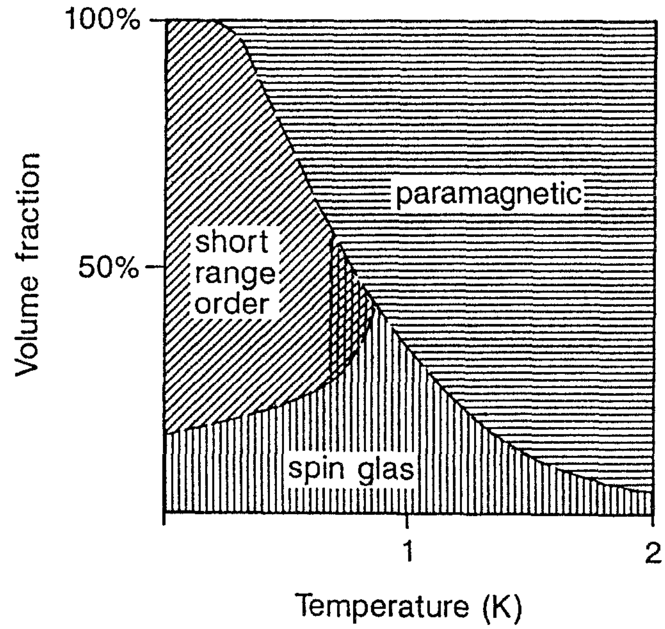

Fig. 2. Magnetic phase diagram of $CeA\ell_3$ as deduced from ZF- and TF-μSR data. Note the coexistence of different magnetic phases at a given temperature. No genuine phase transition can be identified.

tive quasi particle masses of the order of $200 \cdot m_e$ (i.e. of the order of the muon mass!) [6]. In other words these systems are characterized by strongly interacting and highly correlated electrons (f-electrons and conduction electrons) which — at low temperature — develop a ground state with unusual coherent features.

The groundstate may display other properties such as (perhaps unconventional) superconductivity ($CeCu_2Si_2$, $U_{1-x}Th_xBe_{13}$, UPt_3, URu_2Si_2) and antiferromagnetic (AF) order (U_2Zn_{17}, UCu_5, etc.) [5]. The magnetic properties seem to be determined by a competition between a Kondo type mechanism, screening the local f-moments, and the RKKY-interaction, which couples the f-moments via the conduction electrons by an indirect exchange mechanism. The Kondo type mechanism manifests itself by a change of the magnetic susceptibility from a Curie behavior at high temperatures to an essentially temperature independent strongly enhanced Pauli like behavior at low temperatures. Also the ordered moments are generally much smaller than the high temperature moments extracted from the Curie law [5].

The research on these, so far little understood systems, entered into a new phase when it was discovered first by μSR and subsequently also by neutron scattering that all those compounds exhibiting superconductivity also displayed — in coexistence with superconductivity! — some sort of magnetic ordering involving moments as small as possibly $10^{-3} \cdot \mu_B$ [7-12]. While the magnetic order in UPt_3 and URu_2Si_2 appears to be antiferromagnetic and long range in nature [10,11] the small moment magnetism in $U_{1-x}Th_xBe_{13}$ ($0.019 \leq x \leq 0.043$) [8,9] and in $CeCu_{2.1}Si_2$ [7] seems to be of a different type which may have more in common with spin glasses or very short range ordering. The $U_{1-x}Th_xBe_{13}$-results will be discussed in more detail in sect. 3.

Another interesting μSR result was obtained in $CeA\ell_3$ [13], which compound for a long time was considered to be an archetypical representative of a heavy electron system which remains paramagnetic down to the lowest temperatures. That this is not the case will be discussed in sect. 2 in which we will be confronted with a very strange magnetic phase diagram. Among the only remaining heavy electron systems not showing any sign of magnetic order are $CeCu_6$, $CeRu_2Si_2$, UPt_4Au and $YbCu_{4,.5}$. Future μSR studies on these compounds will show whether these compounds are truly paramagnetic at the lowest temperatures or not. We would not be surprised, if also these compounds would show some sort of small moment magnetism at low temperatures.

Finally also those heavy electron systems showing "conventional" AF order are perhaps more complicated than what is believed up to now. A good example in case is U_2Zn_{17} which has displayed in μSR studies an extremely complex behavior [14]. An even more interesting case is UCu_5 which seems to develop some small moment magnetism in coexistence with conventional AF-order [15]. This will be reviewed in sect. 4.

Finally, some future perspectives will be discussed in sect. 5.

2 Magnetic phase diagram of $CeA\ell_3$

The temperature dependence of the specific heat $c_p(T)$ of $CeA\ell_3$ does not show any anomaly which could be associated with a magnetic phase transition [16,17]. Usually, if there is a phase transition, one cannot fail to recognize it in form of a sharp cusp-like feature in $c_p(T)$. However, $c_p(T)/T$ displays a shallow maximum at around 0.5 K which was interpreted in terms of the onset of the heavy mass Fermi liquid state [17]. Also the susceptibility displays a shallow maximum at around 0.7 K [17] which in principle could be associated with an AF-phase transition. However, neutron diffraction results down to 60 mK provided no hint for the formation of static long range magnetic correlations [18]. The more it was a surprise when zero field (ZF) μSR measurements in polycrystalline $CeA\ell_3$ revealed the presence of static nonzero magnetic fields at the μ^+ below 2 K [13]. This is clearly visible in Fig. 1 which for temperatures below ~0.7 K, displays a nice Larmor precession signal associated with a local field of ~220 G. In fact the signal shown in Fig. 1, is composed of three components, two of which originate from the $CeA\ell_3$ sample and the third one from the Cu-target holder (the cold finger of a He^3-He^4 dilution refrigerator). This third component is of the well known Kubo-Toyabe type appropriate for Cu [1]. The second component arising from the $CeA\ell_3$ sample is also well described by a Kubo-Toyabe function, but describing a much faster relaxation than in the Cu-case. Hence μ^+ implanted in $CeA\ell_3$ experience two different magnetic environments: one associated with a nonzero magnetic field (220 G) and the other one associated with zero average field but a nonzero field spread around this value leading to the Kubo-Toyabe signal.

The signal showing the precession pattern must originate from domains inside the sample in which some coherent but short range magnetic order is established. The order cannot be long range otherwise it should have been seen in the neutron scattering study. The Kubo-Toyabe signal, on the other hand, must originate from domains exhibiting some static, random or extremely short range order resembling perhaps a spin glas phase.

The amplitudes of the two $CeA\ell_3$ related signals display a strong temperature dependence, their sum decreasing monotonously with rising temperature until the ZF-μSR signal becomes invisibly small on approaching 2 K. This behavior implies that there must be a third type of magnetic domain in which the μ^+ polarization is not affected suggesting that these domains are in a paramagnetic state. This was confirmed by transverse field (TF) μSR measurements which allowed to identify this component directly [19]. The amplitudes of the three components originating from the $CeA\ell_3$ sample provide a measure of the volume fractions occupied by the various types of domains. Their temperature dependence allows, therefore, to construct a phase diagram, which is shown somewhat schematically in Fig. 2.

The most surprising feature of this phase diagram is the obvious absence of any clear cut cooperative magnetic phase transition which involves the whole sample volume. Rather some random static order develops below ~2 K first in a vanishingly small fraction of the volume which fraction then increases as the temperature is decreased. In parallel the paramagnetic volume shrinks by the same proportion. Below (0.7–0.9) K, i.e. approximately where the susceptibility displays a shallow maximum, some new type of magnetic domain develops which is associated with coherent short range order. The corresponding volume grows with further decreasing temperature at the expense of both the paramagnetic and the "spinglas" like volume. The paramagnetic volume has essentially disappeared below 0.25 K. This phase diagram is very unusual and we do not know of any other example [20]. It is certainly consistent with the absence of any singularity in $c_p(T)/T$. A possible explanation may be given within the framework of magnetic frustration. Magnetic frustration is indeed not entirely impossible in view of the planar triangular arrangement of the Ce-moments in the hexagonal Ni_3Sn-type structure of $CeA\ell_3$ and intraplane AF-interactions between nearest neighbor Ce-moments [19].

More recently the μSR-findings where partially confirmed by NMR measurements which showed a drastic increase of the linewidth below ~1.2 K and a peak of T_1^{-1} at 1.2 K [21]. These results seem to point to the presence of a cooperative phase transition at ~1.2 K, which was suggested to lead to spin density wave-type order. Such a conclusion is at variance both with the c_p/T- and the μSR-results. More studies are clearly needed.

Finally, we wish to mention that a consistent analysis of all the TF-μSR data also allowed to determine the magnitude of the ordered moment (~0.5 μ_B in both types of ordered domains) and the μ^+ site [19].

3 Superconductivity and magnetism in $U_{1-x}Th_xBe_{13}$

The system UBe_{13} has attracted particular attention because it was the second heavy electron compound which displayed superconductivity [22] (after $CeCu_2Si_2$ [23]) associated with the heavy electron state and the first compound in which superconductivity seems to be of an unconventional nature [24]. Additional excitement arouse when it was discovered that Th doped UBe_{13} ($U_{1-x}Th_xB_{13}$) for a certain range of Th-concentrations (1.9% $\leq$ x $\leq$ 4.2%) possesses another second order phase transition at T_{c2} below the onset of superconductivity at T_{c1} [25]. The additional phase transition manifests itself by a typical singularity in the specific heat but the nature of this phase transition remained obscure. Moreover the x-dependence of T_{c1} shows a highly non-monotonic behavior. The x-dependence of T_{c1} and T_{c2} is displayed in Fig. 3. In order to learn more about the nature of the phase transition at T_{c2} ZF-μSR measure-

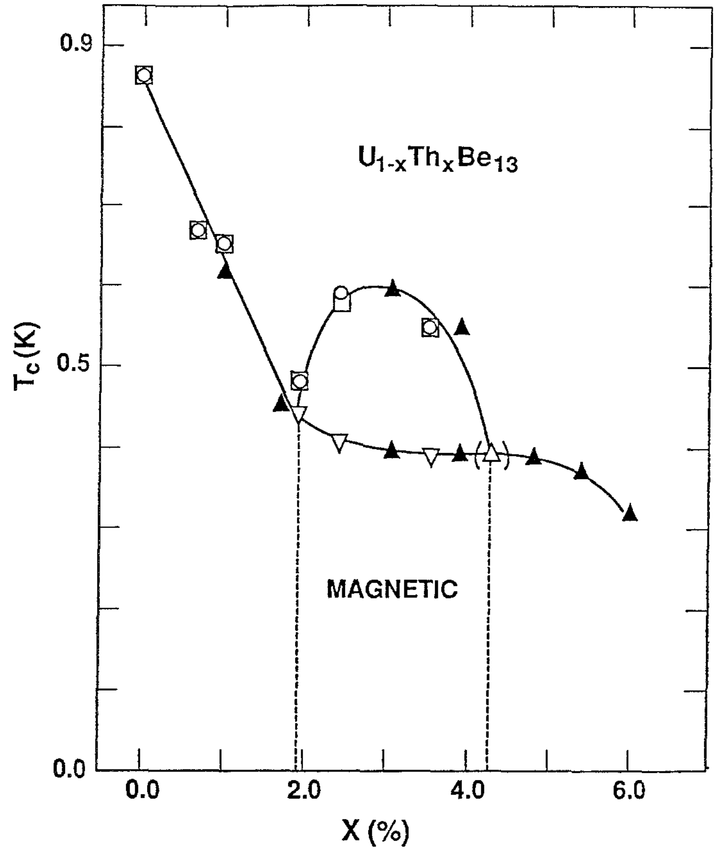

Fig. 3. Phase diagram for $U_{1-x}Th_xBe_{13}$. The data points stem from various techniques. Note the appearance of a second phase transition at T_{c2} between $x = 1.9\%$ and $x = 4.2\%$ (from ref. 28).

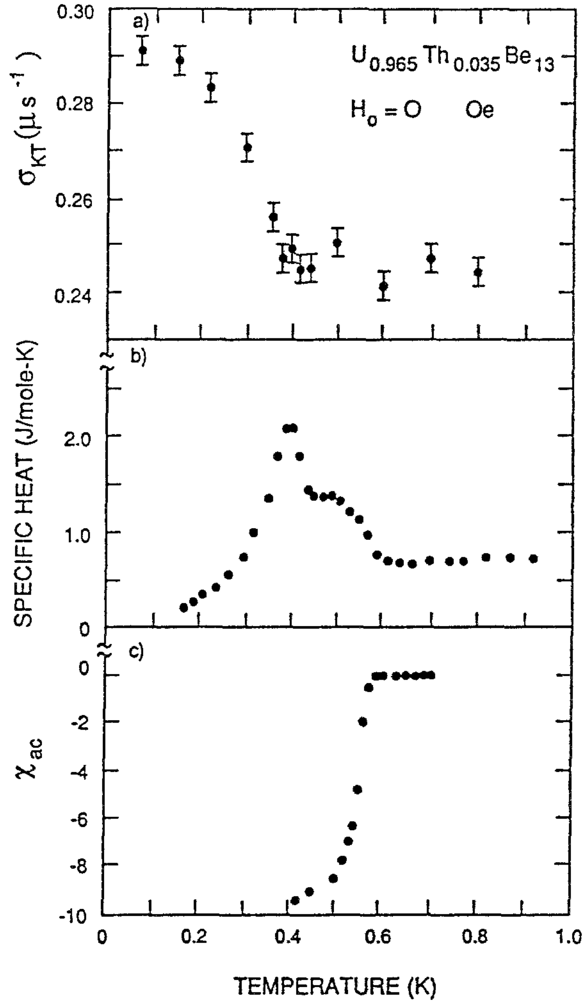

Fig. 4. Comparison of specific heat and ac-susceptibility data with ZF-μSR-relaxation rates in $U_{0.965}Th_{0.035}Be_{13}$. Note the onset of superconductivity at T_{c1} by the diamagnetic response in χ_{ac}, the appearance of the second phase transition at T_{c2} from the cusp in the specific heat and the increase of σ_{ZF} starting right at T_{c2} (from ref. 27).

ments were undertaken first at LAMPF [9,26] and — much improved — at PSI [27,28]. A typical result is displayed in Fig. 4. It shows the onset of superconductivity in ac-susceptibility data at T_{c1} and the position of the second phase transition in specific heat data at T_{c2}. At the latter temperature the ZF-μ^+ relaxation rate σ_{ZF} starts to increase significantly as the temperature is lowered below T_{c2}. The relaxation of the μ^+ polarization below and above T_{c2} is well described by a Kubo-Toyabe function. Above T_{c2} the observed temperature independent relaxation rate can be traced back to the spread in dipolar fields arising from the Be nuclear magnetic moments at a certain interstitial site. This site is shown in Fig. 5. For this site one calculates under certain additional assumptions $\sigma_{ZF} = 0.246\ \mu s^{-1}$ [24] which compares very well with the experimental value of $\sigma_{ZF}^{ex} = 0.245(2)\ \mu s^{-1}$.

Below T_{c2} an additional source for internal fields must become available which, since it cannot be of nuclear origin has to be of electronic origin. The preserved gaussian form of the internal field distribution below T_{c2}, as evidenced from the persisting Kubo-Toyabe type relaxation function, implies further that the involved electronic moments must be static, randomly oriented and fairly regularly positioned, e.g. at each U-site. The magnitude of these moments must be of the order of $(10^{-3}$–$10^{-2})\ \mu_B$, i.e. extremely reduced compared to the high temperature U-moment of $\mu_{5f} = 3.15\ \mu_B$ which results from the Curie behavior of the susceptibility. μSR measurements on samples with other x revealed that the enhanced relaxation rate shows only up in samples with $1.9\% \leq x \leq 4.2\%$ and the onset of the increase of σ_{ZF} is always correlated with T_{c2}. No anomalous features could be observed for $x < 1.9\%$ and $x > 4.2\%$.

The occurance of some sort of static magnetic order in coexistence with superconductivity has found so far two interpretations. The first one is more conventional and assumes the formation of a perhaps rather complex antiferromagnetic phase below T_{c2}. This view is not inconsistent with ultra sound attenuation data [27]. In view of the termination of the phase boundary associated with $T_{c2}(x)$ on the phase boundary $T_{c1}(x)$, separating the normal state from the superconducting state (see Fig. 3), the order parameters of the two phases must be strongly coupled.

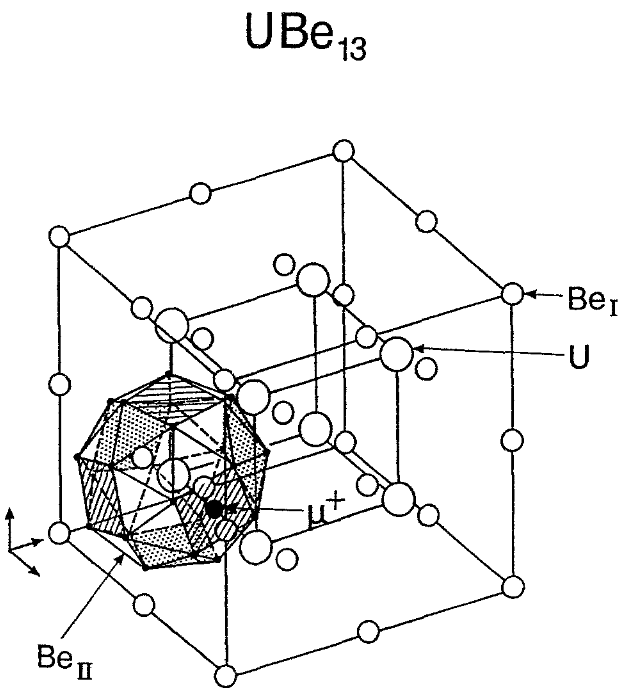

Fig. 5. Crystal structure of UBe_{13} and possible μ^+-position with two nearest U- and 4 nearest Be neighbors. Each U-atom is surrounded by a 'snub cube' of 32 Be-sites (Be_{II}). Not shown are additional Be_{II} positions which form icosahedrons around the Be_I positions at (0,0,0) and (0,0,1/2).

The second more exotic explanation postulates the existence of an intrinsically magnetic superconducting state below T_{c2} which would have to be described by a complex multicomponent order parameter and would violate time reversal invariance [30]. The phase transition at T_{c2} would be a phase transition between two different superconducting states which differ by their symmetry properties. Several other observations [28] add credibility to such an explanation, although the first interpretation cannot be ruled out on the basis of the present status of knowledge.

It should be emphasized that only the μSR technique up to now has the sensitivity to detect the kind of weak magnetic features associated with the phase-transition at T_{c2} in $U_{1-x}Th_xBe_{13}$.

4 'Strong' and 'weak' magnetism in UCu_5

UCu_5 shows a transition to an antiferromagnetic state close to 15 K [31]. The magnetic structure could be determined from neutron scattering measurements [32]. Interestingly the heavy electron state develops below 4 K, i.e. well inside the magnetically ordered state [31]. Specific heat measurements revealed a second continuous phase transition close to 1 K which displayed hysteretic features (see Fig. 6) but no latent heat [31]. In addition the electrical resistivity increased by an order of magnitude below ~1 K. This behavior is exceptional among all heavy electron systems which generally show a trend towards smaller resistivity values when the co-

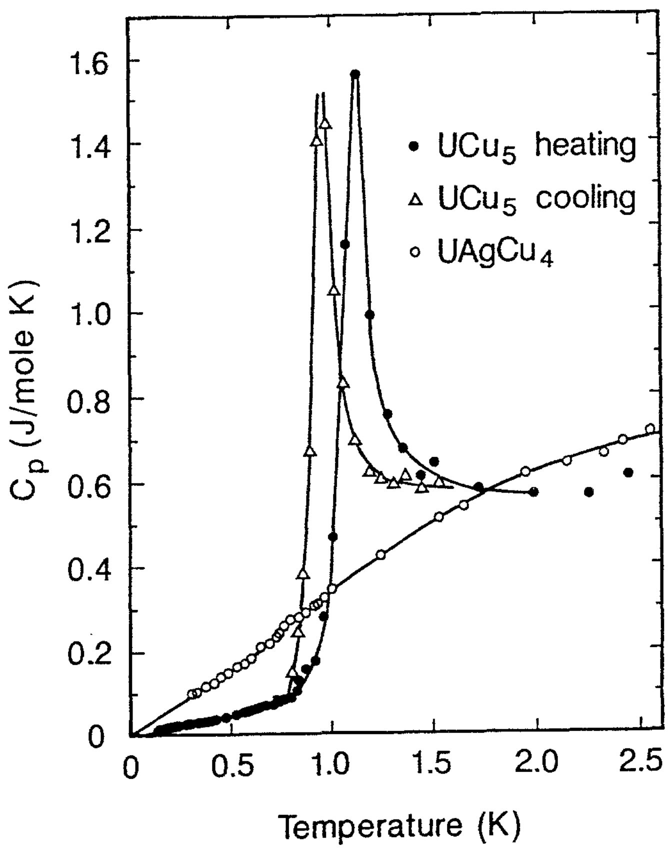

Fig. 6. Specific heat of UCu_5 and $UAgCu_5$ showing evidence for a phase transition in UCu_5 at ~1 K (from ref. 31).

herent heavy electron state develops. The nature of this additional phase transition could not be determined. Interestingly the 1 K anomaly in the specific heat and the rise in resistivity below this temperature are only observed in high quality UCu_5 samples.

In order to determine the nature of the 1 K phase transition we applied again the μSR-technique complemented this time by a neutron diffraction study at the reactor saphir of PSI [15]. The results were the following [15]. The neutron study showed no change in intensity and position of the nuclear and the magnetic Bragg peaks when changing the temperature from 1.3 K to 10 mK (see Fig. 7). Within the limits given by the accuracy of these measurements one has to conclude that at ~1 K no change in the magnetic structure nor in the crystalline structure takes place. The zero field μSR-signal below T_N proved to be quite complicated in that four components were contained in it. Three components showed a relaxing precession pattern, corresponding to average internal fields between ~1 kG and 1.46 kG, while the fourth component did not display any oscillation (corresponding to zero average field) but a significant

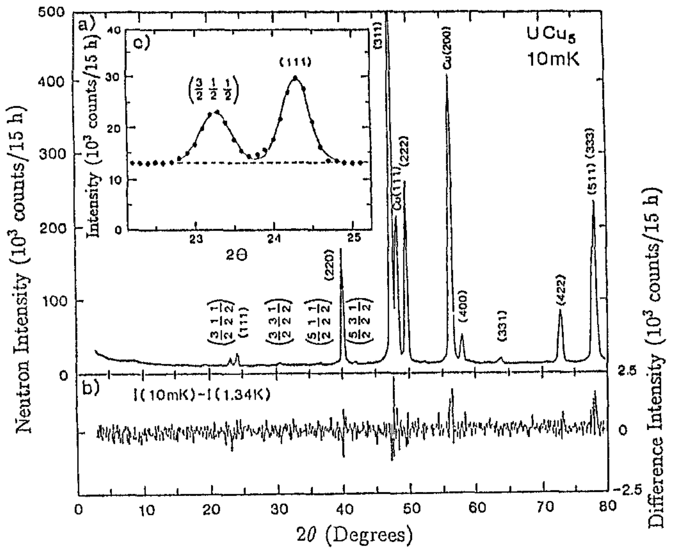

Fig. 7. (a) Neutron-diffraction pattern at 10 mK in UCu_5. (b) Difference neutron diagram I (10 mK) - I (1.39 K) showing the absence of any influence of the 1 K transition on the crystallographic and magnetic structure (from ref. 15). (c) A blow up of the magnetic (3/2,1/2,1/2) r eflection next to a nuclear peak.

relaxation. The temperature dependence of the precession frequencies did not show any conspicuous features at around 1 K, confirming therefore the conclusions drawn from the neutron results. Very drastic effects, however, were seen in the relaxation rates of three of the four components including the nonprecessing one (the other precessing component possesses an order of magnitude larger relaxation rate both above and below 1 K. It's origin is not clear to us). As can be seen from Fig. 8 the relaxation rates start to rise steeply as the temperature is lowered through ~1.1 K. This behavior implies that although the average fields remain unchanged the field spread around each average field rises dramatically below 1.1 K. The relaxation rates above 1.1 K can be explained in terms of the dipolar fields arising from the Cu nuclear moments. The question now is what causes this increase in field width sampled by the μ^+. (Another question concerns the origin of the four components and their different average fields. This question has not been answered yet.) Since the neutron diffraction results exclude more conventional explanations, including the possibility of a charge density wave [31], we interpret the increase in relaxation rate below 1.1 K along the same line as in $U_{1-x}Th_xBe_{13}$, URu_2Si_2 and UPt_3, namely as a result of the formation of small moments which order below 1.1 K in some random fashion or in a pattern which is incommensurable with the lattice structure. These small moments (~0.01 μ_B if we place them at the Cu sites) exist in parallel to the ordinary U-5f moments which are responsible for the antiferromagnetic order below 15 K.

If this interpretation is right, it has some far reaching consequences. First, the fact that the 1 K phase transition is seen both in the specific heat, which arises from the heavy electron ground state, and in the μSR linewidth data could imply that the small moment magnetism is carried by the heavy quasi particles. Secondly, the fact that the normal antiferromagnetic order is unaffected by the 1 K anomaly could imply that the U-5f moments are unrelated to the heavy electron state. In effect we are proposing that the ground state of UCu_5 is made up of two rather independent electron substates the first one involving those 5f-electrons which settle into an antiferromagnetic order below 15 K and a second one which forms the heavy electron state. The latter could be associated with itinerant electrons while the former one arises from localized electrons. Within such a picture the formation of a heavy electron state within an already established antiferromagnetic phase appears to pose no special problem. The concept of rather unrelated electron substates, if present in other heavy electron systems, may perhaps provide a basis for explaining other peculiarities in such systems as well.

5 Future perspectives

The three presented examples prove without doubt the power of μSR-spectroscopy in the field of magnetism. In particular weak effects are now susceptible to thorough investigations. The field of magnetism itself, although quite venerable in age, seems to be an inexhaustible reservoir of new and unexpected phenomena. The last 15 years have witnessed the emergence of such important topics as spin glasses (to which μSR has contributed prominently (see e.g. [2])), magnetism in low dimensional materials, magnetism in mixed valence systems, magnetism in the family of the cuprate oxides, famous for their ability to develop high temperature superconductivity, and last but not least magnetism in heavy electron compounds. This development was accompanied by corresponding efforts in solid state theory. Magnetism, of course, is just but one side of a more basic aspect of solids which concerns their electronic structure. And since solids are made up of practically an infinite number of constituents one can imagine that the phenomenology of solid state systems (or more generally of condensed matter systems), including their magnetic properties may be as varied as there are degrees of freedom. This perspective promises lots of work in the future and μSR spectroscopy is bound to remain an indispensible tool as many other present day techniques such as NMR and neutron scattering.

One import aspect in this respect is the complementary character of neutron scattering (measures in k-space) and μSR (measures in r-space) which should be more fully and systematically exploited in the future. It is therefore of much advantage that at several laboratories μSR and neutron scattering can be practised next door to each other (e.g. at RAL, PSI, Dubna, KEK).

In this contribution we have only discussed applications of μSR spectroscopy in magnetism. To fully assess the future of μSR spectroscopy one would have to consider all the other applications in physical chemistry and solid state physics as well. New and exciting possibili-

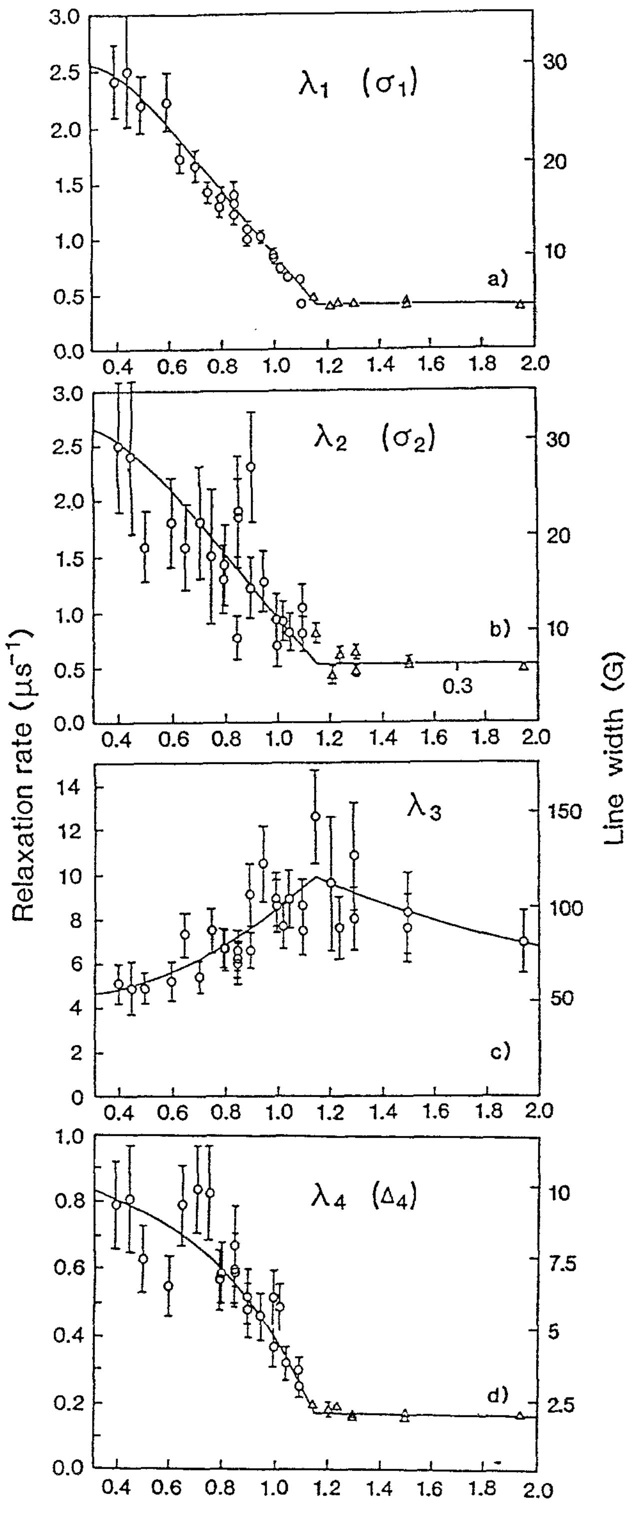

Fig. 8. Temperature dependence of the μ^+ relaxation rates associated with the four components. (a)-(c) are from the precessing components, (d) is from the non precessing one. (a), (b) and (d) clearly reflect the phase transition at ~1.1. K (from ref. 15).

ties would also arise with availability of ultra slow muon beams. It may then become possible to apply μSR to the study of magnetism in surfaces and thin layers and other artificial structures. In conclusion μSR spectroscopy will be a prime force in the demand for muon sources also in the future.

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