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A COMPARATIVE STUDY OF THE MAGNETIC HEAVY - ELECTRON MATERIALS U_2Zn_{17} AND UCu_5 BY μ^+SR

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A systematic μ^+SR study of the magnetic heavy-electron systems U_2Zn_{17} and UCu_5 in the paramagnetic and in the magnetically ordered state is presented. In both systems the antiferromagnetic nature of the low-temperature phase could be at least partially confirmed, but the muon reveals significant differences with regard to the phase transition itself. UCu_5 behaves like a model-antiferromagnet showing a drastic increase of the relaxation rate both below and above T_N, two spontaneous frequencies in the ordered phase, and a Knight shift above T_N which scales with the bulk susceptibility. In contrast U_2Zn_{17} shows a loss of μ^+ asymmetry by 20% below T_N, which is independent of the external field but can be quenched in sufficiently strong longitudinal fields. No scaling of the Knight shift and the susceptibility was observed and no critical increase of the relaxation rate λ. Most astonishing is the strong and nonlinear field dependence of λ above and below T_N in both compounds. The absence of longitudinal relaxation demonstrates the static origin of λ.

1. Introduction

At temperatures well above 50 K rare-earth and actinide-based heavy-electron materials usually behave as if their unpaired f-electrons were localized on regular lattice sites leading to a Curie-Weiss-behaviour of the magnetic susceptibility with moments close to those of the free f-electron carrying ions. However, below a certain degeneracy temperature which is of the order of 10-100 K strong electron-electron interactions lead to the formation of narrow f-bands /1/. Macroscopically this is reflected in a drastic increase of the effective electron mass which enters critically into the specific heat, the electrical resistivity and other transport properties /2/. The ground state of these systems is found to be magnetically ordered as in the systems studied here, superconducting or neither of them /2/. The magnetic order resembles that of itinerant antiferromagnets, e.g. chromium, as it is deduced from a nearly field- and temperature-independent susceptibility below T_N /3/ and small effective magnetic moments. This is quite unexpected because the separation between the ions with the unfilled f-electron-shell in these compounds is quite large. Since it was suspected that the formation of the heavy-electron state is a consequence of suppressed magnetic order it was rather a surprise when it was recently discovered in UCu_5 that it can also emerge out of an already ordered state /4/. So far little experimental data exist on the microscopic properties of the magnetic heavy-electron systems, but it is of great importance to gain such information about the competing magnetic interactions among the f- and the conduction electrons. The dominating interactions hereby are the RKKY interaction which favours magnetic order and the Kondo interaction which tries to compensate the local magnetic moments. Earlier investigations with positive muons in Ce and Ce-Th-systems /5/ have demonstrated that the positive muon is well suited to monitor properties related to f-electrons and may complement the information that is obtained from neutron-scattering experiments on magnetic heavy-electron systems /6/.

2. Experimental Results

We performed transverse, longitudinal and zero field μ^+SR measurements in the paramagnetic and antiferromagnetic range of polycrystalline U_2Zn_{17} and UCu_5 samples. As it is shown in Fig. 1, in U_2Zn_{17} the transition into the magnetically ordered state at 9.7 K is accompanied by a loss of μ^+ asymmetry by about 20% which is nearly independent of the applied transverse field and was also observed

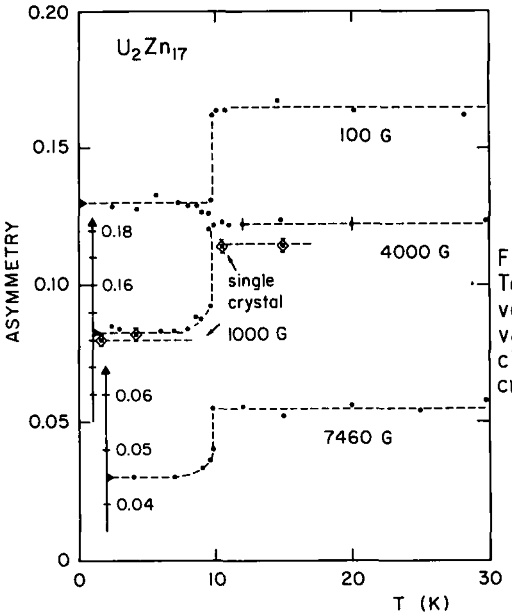

Fig. 1.
Temperature dependence of the transverse field asymmetry in U_2Zn_{17} for various external applied fields (including preliminary data of a single crystal).

in a U_2Zn_{17} single crystal. This fraction of 20% depolarizes within less than 20 ns which is the spectrometer deadtime and must derive from an internal field spread of at least several hundred Gauss. The remaining fraction of 80% shows a precession according to the external field. While no spontaneous μ^+ signal occurred in U_2Zn_{17} below T_N, we detected two spontaneous frequencies in UCu_5 below T_N = 15 K which have rather different temperature dependences. In Fig. 2 the temperature dependence of the μ^+ signals in both substances are compared. There is a step-like discontinuity of the Knight shift in U_2Zn_{17} at T_N, the magnitude of which turned out to be a function of the applied field. Above T_N the Knight shift in U_2Zn_{17} does not reflect the bulk susceptibility χ while in UCu_5 we found a linear relaxation between both quantities. The temperature dependence of the corresponding relaxation rates is displayed in Fig. 3. "Critical" behaviour in UCu_5 is clearly monitored by the drastic increase of λ both below and above T_N. In contrast to this the relaxation rate in U_2Zn_{17} remains constant when approaching T_N from higher temperatures and increases in a manner reminiscent of a

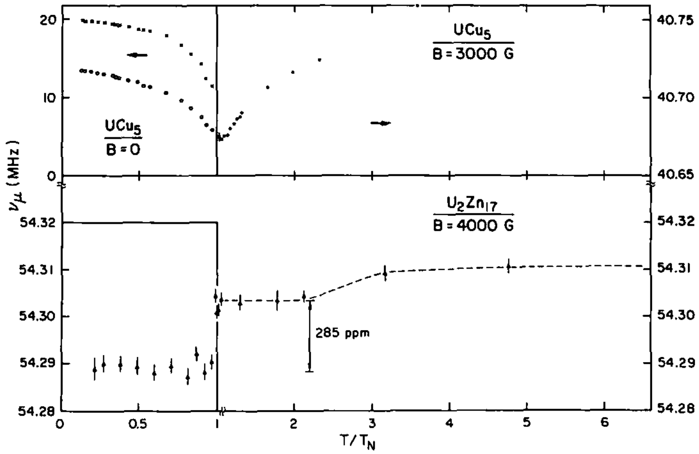

Fig.2
Local magnetic field or μ^+ frequency ν_μ in U_2Zn_{17} (T_N=9.7 K) and UCu_5 (T_N=15 K) versus normalized temperature. Note that only 80% of the muons in U_2Zn_{17} contribute to the signal below T_N.

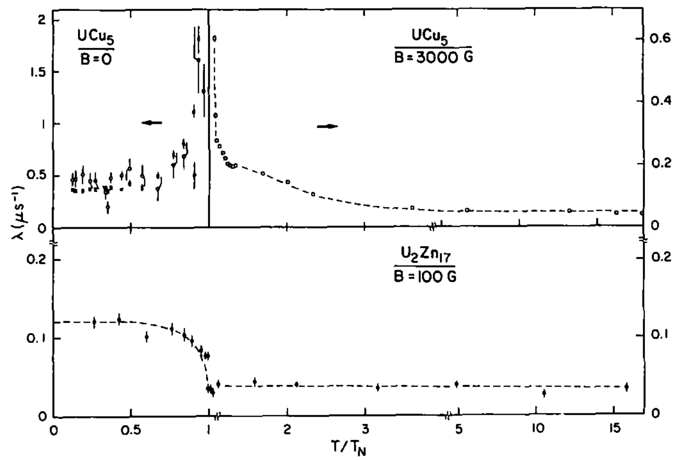

Fig. 3
Transverse field relaxation rate λ in U_2Zn_{17} (T_N=9.7 K) and UCu_5 (T_N= 15 K) versus normalized temperature.

Brillouin function below T_N at small applied fields. Most astonishing and in this form not observed before, is the strong field dependence of the transverse relaxation rate λ in all heavy-electron compounds. Fig. 4 illustrates this field dependence in U_2Zn_{17} and UCu_5. In U_2Zn_{17} λ shows a nonlinear field dependence saturating at 4 kG both below and above T_N. In UCu_5 a field dependence of λ is found up to 50 K. Here λ remains constant up to a certain field of at least 1500 G and then increases linearly with the external field B_{ext}. A trivial explanation for the field dependence, the influence of inhomogeneous demagnetization fields seems to be unlikely, because in this case λ should scale with χ and B_{ext}. Nevertheless the influence of inhomogeneous demagnetization fields was estimated to lead to corrections of not more than 10%. The zero field muon-spin-relaxation in UCu_5 is of the typical static Kubo-Toyabe form reflecting a quasi-static muon surrounding. The values obtained for Δ_{KT} between 15 K and 50 K are constant and of the order of 0.5 μs^{-1} which is compatible with lattice calculations of the second moment of the copper nuclear dipols. In U_2Zn_{17} also a non-vanishing zero field relaxation was found. U and Zn nuclei possessing no dipole moment it must be of electronic origin. The λ values are consistent with the

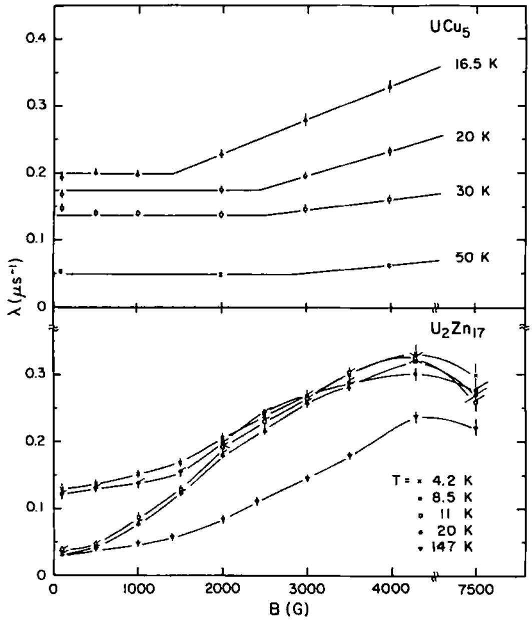

Fig. 4
Field dependence of the transverse field relaxation rate λ in U_2Zn_{17} and UCu_5.

extrapolated values of the transverse field data shown in Fig. 4. The absence of any longitudinal relaxation in both materials demonstrates the static origin of λ. In U_2Zn_{17} the lost asymmetry below T_N can be restored in sufficiently strong longitudinal fields as it is evident from Fig. 5. This permits the determination of the field spread B_Δ at the μ^+ sites assuming a Gaussian distribution of dipolar origin and using the Kubo-Toyabe formula in the limit $t \to \infty$:

$$g_z(t \to \infty) = 1 - 2x^2 + 2x^3 \exp\left(-\frac{1}{2x^2}\right) \cdot \int_0^{1/x} \exp\left(\frac{u^2}{2}\right) du \tag{1}$$

$$\text{with } x = \frac{B_\Delta}{B_{ext}} \text{ and } B_\Delta = \langle H^2_{dip,\rho} \rangle, \ \rho = x,y,z$$

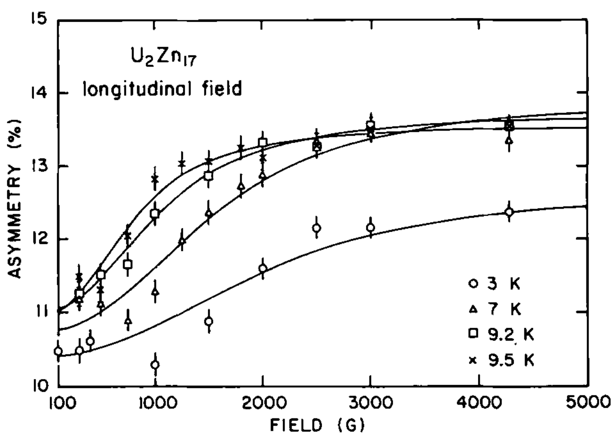

Fig. 5: Field dependence of the longitudinal field asymmetry in U_2Zn_{17} for various temperatures below T_N = 9.7 K.

Fig. 6 displays the temperature dependence of the second moment B_Δ which is well approximated by a Brillouin function.

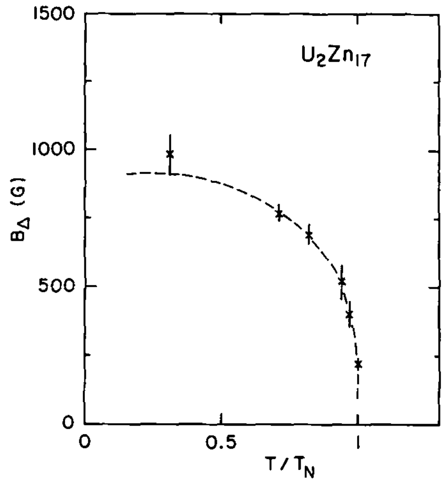

Fig.6
Field distribution width B_Δ in U_2Zn_{17} as a as a function of the normalized temperature.

3. Discussion

In both systems the magnetic nature of the the phase transition could be confirmed, but the muon reveals significant differences in the properties of the transition itself. UCu_5 behaves like a modell-antiferromagnet exhibiting a "critical" behaviour of λ below and above T_N which is attributed to almost static spin correlations persisting up to 50 K. There are two spontaneous μ^+ signals in the ordered phase. Dipolar field calculations for two tetrahedral sites in the host lattice give a vanishing dipolar field at one of them and a dipolar field of 0.7 kG at the other one. The difference equals the difference of the measured internal fields extrapolated to T=0. So the lower spontaneous frequency might reflect the hyperfine field B_{hf}. In that case the temperature dependence of B_{dip} and B_{hf} would turn out to be quite different. The two spontaneous signals should also help two clarify the nature of a second phase transition in UCu_5 at 1 K whose origin and character is unknown so far. This would be of great importance for the theoretical description of the ground state of heavy-electron systems. In contrast to these results only 20 % of the muons in U_2Zn_{17} feel large dipolar fields arising from magnetic order while the remaining fraction of 80% is hardly affected by the magnetic order. It is interesting to note that the dipolar fields cancel at the two most probable muon sites. Possible explanations for this behaviour are the occurrence of strong dipolar fields in domain walls or an incommensurate, modulated magnetic structure which has already been found in a number of uranium compounds /7/. The latter is, however, in contradiction to neutron-scattering data /6/, which indicate a simple antiferromagnetic order in the basal planes. The most surprising experimental result is the field dependence of λ, especially above T_N. It may be suspectedthat it arises from a frozen-in residual magnetization like that in a spin glass, but entropy considerations make this unlikely. Preliminary Knight shift data show that the muon Knight shift has also a complicated field dependence. Maybe λ reflects a distribution of Knight shifts, whose width is a function of the external field. It may be concluded that there is an additional field-induced magnetic moment. In conclusion this and other new phenomena observed in a first systematic μ^+SR study of magnetic heavy-electron compounds give incentive for both experimental and theoretical work in future.

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DISCUSSION

Boekema: Below T_N, two muon frequency signals were observed in UCu_5, and two possible muon sites were indicated. Why should the hyperfine field contribution at these different sites be the same, as you suggested? May I suggest to perform dipolar-field calculations, not only at highly symmetrical sites, which you mentioned, in order to search for the muon stopping sites in UCu_5.

Barth: Of couse the hyperfine field contribution need not be the same at the different site but at least the temperature dependence should be equal. However, we are planning to perform also calculations at less symmetrical sites.

Heffner: Do you know whether the μ^+ is diffusing in U_2Zn_{17}? Could diffusion be a source of the non-linear Knight shift versus susceptibiltiy relation.

Barth: We cannot totally exclude diffusion, but is seems plausible that the muon is not diffusing in U_2Zn_{17} because our zero field data in UCu_5 and some other compounds indicate that the muon is static at these low temperatures.