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EVIDENCE FOR COMPLEX MAGNETIC ORDER IN U_2Zn_{17}: A μ^+SR STUDY

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μ^+SR-measurements in transversally applied magnetic fields of 2000 G and 4000 G on heavy-electron single crystal U_2Zn_{17} are presented. They reveal that at least two types of interstitial sites are occupied by the positive muons. One of these sites (1/3, 2/3, 5/6) could be identified via induced local dipolar fields which above $T_N = 9.7$ K can exactly be derived from the magnetic susceptibility. The corresponding component of the μ^+-signal exhibits a steplike decrease by about 40% at T_N which is caused by the onset of a very broad distribution of static internal magnetic fields ($\Delta B \approx 1000$ G) with zero average. Such a field distribution is in distinct contrast to dipolar-field calculations performed for the simple antiferromagnetic structure deduced from neutron diffraction. The remaining 60% of the muons contributing to this component below T_N are subject to a narrow static field distribution ($\Delta B \approx 1$ G). The induced dipolar fields at the site (1/3, 2/3, 5/6) are temperature-independent below T_N. A weak dipolar coupling to the U-moments renders similar observations for muons occupying the second type of interstitial impossible.

1. Introduction

The ground state properties of magnetically-ordering heavy-electron materials are generally explained in terms of a phenomenological model based on the occurrence of two competing interactions between the f-electron moments [1]. One of these is a renormalized Kondo-interaction [2] and describes the screening of the local magnetic moments by antiferromagnetic coupling to the conduction electrons, the renormalization arising from interactions between the Kondo-scattering centers. Obviously, this Kondo-interaction favours a non-magnetic singlet ground state. The second interaction is the Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction [3-5] which establishes spatially oscillatory magnetic correlations between the local magnetic moments. As a possible consequence of

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these competing interactions complicated magnetic structures involving small effective moments or even suppression of long-range magnetic order may occur in these materials as evidenced by some recent experiments [6–8].

An important question concerns the degree of localization of the f-electrons. While at high temperatures a Curie-Weiss law of the magnetic susceptibility χ points to localized moments [1], a break of χ at low temperatures in a narrow temperature interval often leads to an almost temperature-independent behaviour favouring an itinerant description [1]. This is also reinforced by small effective moments in the ordered state [1]. The distances between the f-ions, however, generally lie well above the Hill-limit [9] by which traditionally localized and non-localized magnetic behaviour is separated. Another very interesting fact concerning the magnetic phase transition in heavy-electron systems is the remanence of a still considerable electronic contribution to the specific heat C_p as $T \rightarrow 0$ K [1]. This indicates that only part of the Fermi-surface is affected by magnetic order and may be compared to the occurrence of anisotropic superconducting gaps in heavy-electron superconductors [1]. Furthermore the extreme sensitivity of magnetic order to small amounts of impurities is a very puzzling property of these systems [1]. A famous paradigm exhibiting all these features is U_2Zn_{17} [10–13], which orders antiferromagnetically at $T_N = 9.7$ K. Neutron diffraction revealed a simple antiferromagnetic structure [11] which shows no doubling of the unit cell and involves small effective moments of 0.8 μ_B/U at 5 K which lie in the hexagonal basal plane. In the present μ^+SR-study we attempt to provide complementary microscopic information on the static and dynamical magnetic properties of U_2Zn_{17} by means of an interstitial spin probe, the positive muon.

2. Experimental results

Earlier μ^+SR-measurements [14,15] in U_2Zn_{17} confirmed the discontinuous onset of magnetic order at $T_N = 9.7$ K. Interestingly, the ordering does not affect all muons implanted into the sample in the same way. Below T_N, 80% of the muons are subject to a quasi-static magnetic field distribution of about 1 G width, whereas the other 20% feel a quasi-static magnetic field distribution of about 1000 G width. Both distributions have a vanishing average value. The occurrence of the broad field distribution of 1000 G width is rather unexpected because the dipolar fields calculated considering the simple antiferromagnetic structure [11] cancel at the most probable muon sites, i.e., (2/3, 1/3, 2/3), (1/2, 1/2, 0) and (1/3, 2/3, 5/6) shown in fig. 1.

In order to determine the muon locations in U_2Zn_{17} we performed single crystal μ^+SR-measurements in transversely applied magnetic fields of 2000 G and 4000 G. Our sample was of cylindrical shape having 7 mm diameter and 12 mm length. It was mounted into the cryostat that by turning the axis of the

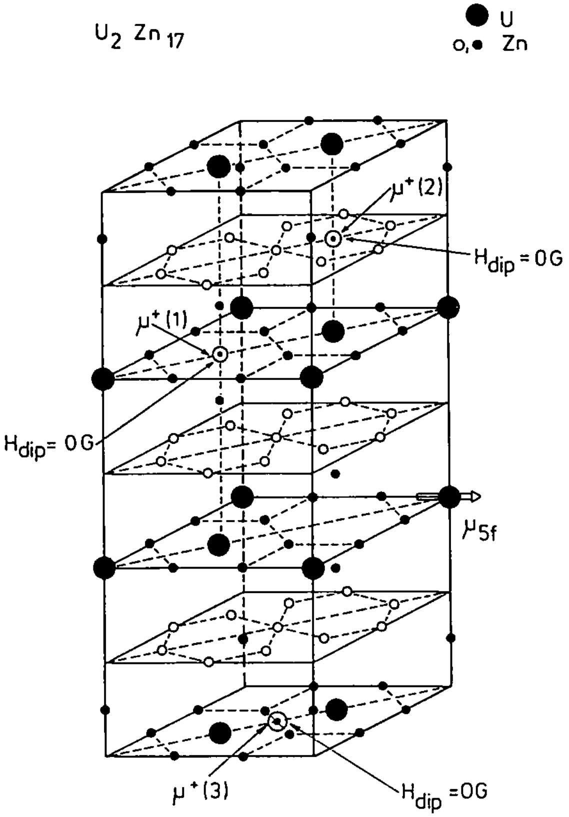

Fig. 1. Crystallographic unit cell of U_2Zn_{17} [16]. The most probable muon sites are $\mu^+(1)$ at (2/3, 1/3, 2/3), $\mu^+(2)$ at (1/3, 2/3, 5/6), and $\mu^+(3)$ and (1/2, 1/2, 0). The dipolar fields for the simple antiferromagnetic structure [11] cancel at each site.

Al-targetholder the angle θ between the external field and the c-axis could be varied while the angle ϕ between the external field and the a-axis remained constant. The spectra obtained consistent of two μ^+-polarization components with different precession frequencies $\nu_\mu(1)$ and $\nu_\mu(2)$. In fig. 2 we show the dependence of $\nu_\mu(1)$ and $\nu_\mu(2)$ on the angle θ for different temperatures in an external field of 4000 G, the angle ϕ being zero. Note that there is an offset of θ in the laboratory frame which amounts to 22.5°. $\nu_\mu(1)$ is temperature- and angle-independent within the possible experimental errors and therefore drawn as a dashed line. In contrary, $\nu_\mu(2)$ exhibits a pronounced angle- and temperature-dependence. A recently reported more complicated anisotropy of the μ^+-frequency in U_2Zn_{17} turned out the be an artifact of the fitting program due to the superposition of these two components [15]. The ratio of the μ^+-asymmetries or relative fractions of muons A(1) and A(2) is roughly equal. While A(1) is not influenced by the phase transition at all, a steplike decrease of A(2) occurs at T_N which amounts to 40% of the value of A(2) above T_N, i.e. 20% of all muons

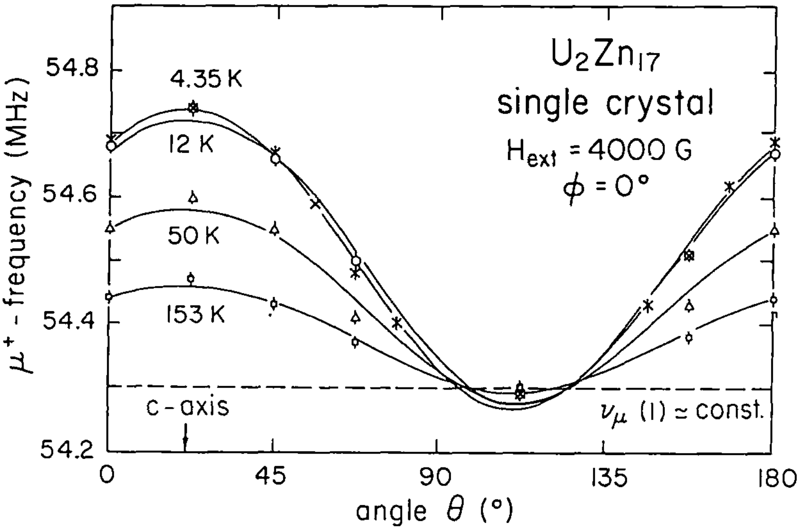

Fig. 2. Angular dependence of the frequency $\nu_\mu(2)$ on the angle θ with contact angle $\phi = 0°$ (definition see text) in U_2Zn_{17} at temperatures of 153 K, 50 K, 12 K and 4.35 K. $\nu_\mu(1)$ is temperature- and angle-independent and therefore drawn as a dashed line. A transverse external field of 4000 G was applied. The solid lines represent the anisotropy due to induced dipolar fields at the site (1/3, 2/3, 5/6).

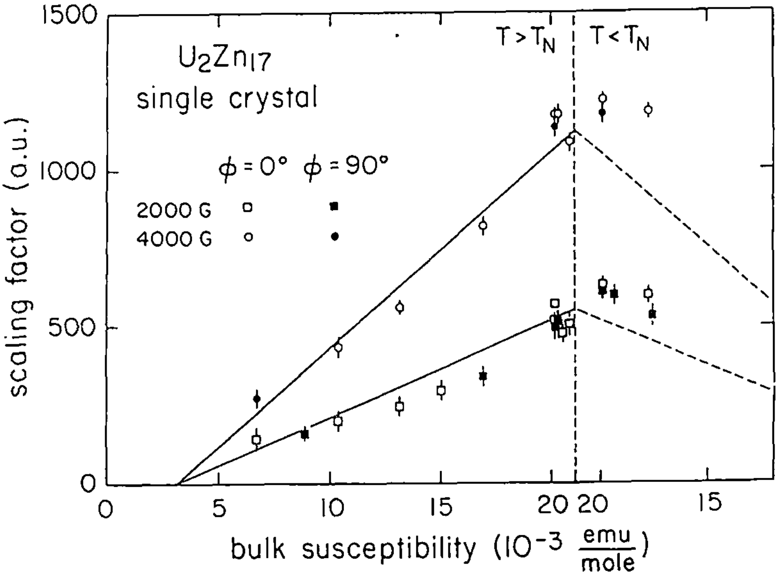

Fig. 3. Scaling behaviour of the anisotropy of the frequency $\nu_\mu(2)$ and the bulk susceptibility in external fields of 2000 G and 4000 G for $\phi = 0°$ and $\phi = 90°$. Note that the scale has been inverted at $\chi(T = T_N)$.

implanted. In order to explain the anisotropy of $\nu_\mu(2)$, the angular dependence of induced dipolar fields at the three possible muon sites shown in fig. 1 was calculated. As it turned out, above T_N, the anisotropy of $\nu_\mu(2)$ can be derived exactly from the induced dipolar fields at, and only at the site (1/3, 2/3, 5/6).

The scaling of the anisotropy of $\nu_\mu(2)$ with the bulk susceptibility is illustrated in fig. 3 for external fields of 2000 G and 4000 G, the temperature being implicit parameter. In contrast to the behaviour in the paramagnetic state, this scaling is no longer valid below T_N and the induced dipolar fields keep their values attained at T_N. This means that the local susceptibility of the U-moments producing the dipolar fields becomes temperature-independent. The behaviour expected from the scaling above T_N is drawn as a dashed line and clearly deviates from the experimental observations. Note that the scale has been inverted at χ $(T = T_N)$.

As a possible explanation for the occurrence of the frequency $\nu_\mu(1)$ it should be mentioned that the dipolar coupling is 30 times smaller at the site (2/3, 1/3, 2/3) and would therefore fall into the possible experimental errors. An occupation of the site (1/2, 1/2, 0) can be ruled out by symmetry considerations.

3. Conclusions

From the exact correspondence of the frequency $\nu_\mu(2)$ and the macroscopic susceptibility above T_N it follows unambiguously that 50% of the muons in U_2Zn_{17} are located at the site (1/3, 2/3, 5/6). The occurrence of a field distribution of 1000 G width below T_N at 40% of these sites is unexpected because, as mentioned above, the dipolar fields for the simple antiferromagnetic structure [11] cancel.

It seems unlikely that this behaviour involving 40% (!) of the (1/3, 2/3, 5/6) sites is caused by e.g. domain walls or possibly lattice defects. The latter in particular should show up also in the angular dependence above T_N. On the other hand, the angular dependence of $\nu_\mu(2)$ seen below T_N at the remaining 60% of these sites implies 5f-moments which align along the applied magnetic field direction and therefore do not take part in the antiferromagnetic ordering. In contrast to the total susceptibility, their behaviour below T_N seems to be described by a temperature-independent local susceptibility. It is unclear so far, whether this behaviour may be taken as evidence for a transition of a part of the 5f-moments into a dispersion band state without a pronounced change of the magnetic form factor or a spatially inhomogeneous temperature-dependent Kondo-screening.

In addition, it is to be noted here that the muons located at site (1/3, 2/3, 5/6) do not display any sizable isotropic Knight-shift while the muons associated with $\nu_\mu(1)$ possess a temperature independent isotropic Knight-shift of -0.2%.

Taken altogether, it seems that the actual magnetic structure of U_2Zn_{17} is much more complicated than the simple antiferromagnetic structure of strongly reduced moments suggested by the neutron data [11].

In summary, a site determination for positive muons in U_2Zn_{17} was performed via induced dipolar fields. No consistency between the magnetic structure deduced from neutron scattering and the local magnetic fields in the ordered state was found. The induced dipolar fields do not show the expected scaling with the susceptibility below T_N. One possible explanation is, of course, that the positive muon locally changes the magnetic order but the existence of a complicated super-structure on a microscopic level appears to be more likely. In either case, our measurements underline the complexity of magnetism in heavy-electron compounds.

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