Discovery of novel magnetic features in the heavy-electron compound $\text{U}_2\text{Zn}_{17}$

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Transverse field $\mu^+\text{SR}$-measurements on a single crystal sample of $\text{U}_2\text{Zn}_{17}$ reveal a strikingly non uniform magnetic behavior. While part ($\sim 30\%$) of the U-5f moments remain paramagnetic below $T_N = 9.7$ K (at least for external fields $\geq 0.2$ T) another part ($\sim 20\%$) enters into a complex order below 9.7 K which is associated with huge internal field spreads at the $\mu^+$ sites. Above $T_N$ this part causes an anisotropic $\mu^+$ Knight shift involving direction cosines up to 8th order. Only the remaining fraction is compatible with the simple AF order suggested by neutron diffraction studies.

Measurements of the magnetic susceptibility, specific heat and resistivity have revealed that $\text{U}_2\text{Zn}_{17}$ undergoes a magnetic phase transition at 9.7 K [1], which was subsequently shown by neutron diffraction studies to lead to a simple Antiferromagnetic (AF) structure [2].

However, $\mu$SR measurements on both polycrystalline [3] and monocrystalline samples [4] have indicated that the magnetic properties are not as simple as implied by the neutron diffraction results. We have refined and extended the analysis of the single-crystal data and have found additional striking features which we wish to report in this contribution.

For details on the sample and the measurements see ref. [4]. The measurements to be discussed here were performed in external fields of $H_{\text{ext}} = 0.4$ and 0.03 T in the temperature range 4–295 K.

The observed transverse field $\mu^+\text{SR}$ signal consists of four components which are distinguished by their frequency or Knight-shifts which are different both respect to their temperature and orientational dependences. In the following we list some but not all of the striking features of the four components.

Component no. 1: Accounts for $\sim 67\%$ of the total signal (this number and the corresponding ones below seem to be temperature independent). It includes some background signal from $\mu^+$ stopping outside the single crystal specimen. Its frequency $\nu_1$ is, within the present accuracy, solely determined by $H_{\text{ext}}$ (no detectable Knight-shift). It does not reflect the phase transition at 9.7 K.

Component no. 2: Accounts for $\sim 20\%$ of the total signal. It shows a pronounced angular dependence of $\nu_2$, which is shown in fig. 1 for temperatures $\geq T_N$ and $H_{\text{ext}} = 0.4$ T. The angular dependence is well described by $\nu_2 = \nu_2^{(0)} + \nu_2^{(2)}P_2(\cos \theta) + \nu_2^{(4)}P_4(\cos \theta)$, where $\theta$ is the angle between $H_{\text{ext}}$ and the hexagonal c-axis. This angular dependence is also observed below $T_N$ for $H_{\text{ext}} \geq 0.2$ T. However, for $H_{\text{ext}} = 0.03$ T $\nu_2(\theta)$ assumes a different form as the temperature is lowered from 9.5 to 4 K (see fig. 2).

Fig. 1. Orientational dependence of $\nu_2$ at 0.4 T for various temperatures. $\theta_L$ is the angle between the c-axis and $H_{\text{ext}}$, shifted by an offset angle of $22^\circ$ ($\theta_L = \theta + \theta_{0\text{f}}$). The solid line represents a fit (see text).
Component no. 3 and 4: Both appear with the same amplitude and account together for the remaining 13% of the total signal. Again a very strong angular dependence is observed above $T_N$ which is very unusual in that 6th order and 8th order Legendre polynomials have to be included in order to describe $\nu_3(\theta)$ and $\nu_4(\theta)$ (the $T = 9.8$ K data are shown in fig. 3). In addition a strong dependence on temperature is manifest. Below $T_N$ these two components are not detectable leading to a missing $\mu_+^\ast$ polarization. This loss in polarization has been observed before also in a polycrystalline $U_2Zn_{17}$ sample [3] suggesting strongly that it reflects some intrinsic property of $U_2Zn_{17}$.

In the following we discuss some of the implications arising from the features of the four components. Component no. 2: Above $T_N$ the general features of this component can be understood consistently in terms of a particular $\mu_+^\ast$-site, e.g. $(\frac{1}{2}, \frac{1}{2}, \frac{1}{2})$, and dipole fields at this site originating from the neighboring effective U-5f-moments induced by $H_{\text{ext}}$ in accordance with the magnetic susceptibility [5]. The fact that the form of the angular dependence persists unchanged to $T < T_N$, at least for $H_{\text{ext}} > 0.2$ T, implies that the neighboring U-moments are still able to align freely along $H_{\text{ext}}$ (i.e. remain paramagnetic) and hence cannot take part in the AF order below 9.7 K. However, as fig. 2 reveals, the apparent paramagnetism seems to become suppressed for smaller $H_{\text{ext}}$ and temperatures well below $T_N$ signaling the presence of some residual interactions among the U-ions which is in competition with the Zeeman interaction.

Components no. 3 and 4: The disappearance of these components below $T_N$ has been shown to be the result of a huge static field spread which appears in part of the sample volume below $T_N$ [3]. The present analysis shows that this fraction of the volume is also associated with distinctly different properties above $T_N$ as manifested in the unusual angular dependence of $\nu_3(\theta)$ and $\nu_4(\theta)$. This angular dependence cannot originate from the angular dependence of dipole fields even when taking into account the tensorial character of the magnetic susceptibility, but must involve a complex dependence of moments at the U-sites on the direction and probably strength of $H_{\text{ext}}$. Such a complex behavior may again be a result of competing interactions, perhaps involving quadrupole-quadrupole interactions as seen in e.g. CeB$_6$ [6]. The presence of competing interactions may also lead to a complex AF order in this fraction of the sample volume producing the observed huge field spread below $T_N$.

Fig. 2. Orientational dependence of $\nu_2$ at 0.03 T. Note the change in the angular dependence at the lowest temperatures as compared to fig. 1.

Fig. 3. Orientational dependence of $\nu_3$ and $\nu_4$ at 0.4 T and 9.8 K. The solid lines represents fits (see text). The errors, except where explicitly shown, are smaller than the dot size.
Component no. 1: The independence of $\nu_1$ on angular orientation (and also its temperature independence) can be explained if some fraction of the $\mu_+^+$ are located at another interstitial site (e.g. (000)), where the net dipole field from the induced U-moments are reduced by a factor of 30 in comparison to the site ($\frac{1}{3}, \frac{1}{3}, \frac{1}{3}$). Below $T_N$ the dipole fields from the ordered moments will cancel to zero in case of the simple AF structure suggested in [2]. This would then leave component no. 1 unaffected.

$UZn_{17}$ shows a very peculiar magnetic behavior which suggests that several magnetically inequivalent regions or domains may coexist in a single crystal specimen. Moreover, the observations imply strongly that at least in part of the sample volume the behavior of the U-5f moments is governed by competing interactions. It is suggested that quadrupole–quadrupole interactions may be involved.

References

[1] H.R. Ott et al., Phys. Rev. Lett. 52 (1985) 1551.
[2] D.E. Cox et al., Phys. Rev. B 33 (1986) 3614.
[3] S. Barth et al., Hyperfine Interactions 31 (1986) 397.
[4] S. Barth et al., Hyperfine Interactions 50 (1989) 711.
[5] See e.g. J.M. Effantin et al., J. Magn. Magn. Mater. 47 & 48 (1985) 145.