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Magnetic Anisotropy of U_2Pd_2In

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Abstract

Magnetic susceptibility studies of the U_2Pd_2In single crystal show that in the paramagnetic state larger χ -values are found for field along the basal plane than along the c -axis. However, below $T_N = 37$ K, where U-moments form a non-collinear magnetic structure within the basal plane, a cross-over of the two χ -branches occurs. The single crystal results may explain the origin of the exotic magnetization results obtained on powder samples.

Keywords: U_2Pd_2In , magnetic anisotropy

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I. Introduction

$\text{U}_2\text{Pd}_2\text{In}$ is member of a large isostructural group of uranium compounds with the stoichiometry 2:2:1 crystallising in the tetragonal U_3Si_2 -type structure [1,2]. The results of electrical-resistivity, specific-heat and magnetic-susceptibility experiments on $\text{U}_2\text{Pd}_2\text{In}$ polycrystalline samples point to an antiferromagnetic transition at $T_N = 37$ K [3]. The antiferromagnetic order was confirmed by neutron powder diffraction, which reveal that U magnetic moments of about $1.6 \mu_B/\text{U}$ (at $T = 10$ K) confined to basal plane form a non-collinear magnetic structure [3].

Here additional data obtained on a single crystal are presented and the type and strength of magnetic anisotropy are discussed in the framework of the hybridization-induced anisotropy model.

II. Results and discussion

Polycrystalline $\text{U}_2\text{Pd}_2\text{In}$ has been prepared by arc-melting appropriate amounts of the constituting elements with 5% excess of In. The structure, the lattice parameters and the atomic positions were determined on a small single crystal using an Enraf-Nonius four-circle diffractometer [1]. Single crystals with the typical mass of about 100 mg have been synthesised by the mineralization method and checked and oriented using X-rays. DC magnetic susceptibility of a $\text{U}_2\text{Pd}_2\text{In}$ single crystal was studied using a SQUID magnetometer with the sample oriented with the principal directions along magnetic field. These results are given in the context of high-field magnetization measurements, which were performed in quasi-static fields at $T = 4.2$ K on two types of fine powder samples: one consisting of powder particles fixed in random orientations and the other with particles free to rotate in magnetic field [3]. In the latter case the magnetization

usually reaches larger values (representing the easy-axis magnetization) and the ratio to the fixed-powder magnetization (representing an ideal polycrystal) is influenced by the type and strength of magnetic anisotropy.

In the case of U_2Pd_2In , the magnetization curve for the fixed powder shows a pronounced metamagnetic transition, which sets on around 25 T, followed by a gradual saturation in higher fields (Fig. 1). For the free-powder sample, the initial susceptibility dM/dB is larger than in the fixed powder, but the metamagnetic transition is much less pronounced. Thus we encounter a rather curious case, because the magnetization measured on a random powder is in high fields larger than the free-powder magnetization.

The magnetic susceptibility χ on a single crystal was measured with field both parallel ($B||c$) and perpendicular ($B\perp c$) to the c -axis. Above 70 K, $\chi(T)$ curves for both field orientations (Fig.2) can be described by the modified Curie-Weiss law. The respective fits yield parameters $\mu_{\text{eff}} = 3.0 \mu_B/U$, $\Theta_p = -125$ K and $\chi_0 = 0.9 \cdot 10^{-8} \text{ m}^3/\text{mol f.u.}$ for $B||c$ and $\mu_{\text{eff}} = 2.5 \mu_B/U$, $\Theta_p = -23$ K and $\chi_0 = 2.0 \cdot 10^{-8} \text{ m}^3/\text{mol f.u.}$ for the perpendicular field. Thus the susceptibility above T_N reaches higher values for field along the basal plane. The anisotropy energy in the paramagnetic state, which can be estimated as the difference of both Θ_p -values, is about 100 K, which is a rather modest value comparing to other U-intermetallics. Below T_N , the $\chi^{\perp}(T)$ curve drops significantly as usual in antiferromagnets. On the other hand, χ^{\parallel} increases even faster than would be expected from the high temperature MCW behaviour. In the low temperature limit $\chi^{\parallel}(T) \gg 3\chi^{\perp}(T)$.

The temperature dependences of the magnetic susceptibilities may imply a scenario which could account for the unusual magnetization behaviour of powders mentioned above.

Due to a much higher susceptibility for field along the c -axis at 4.2 K, we can expect that the powder grains are oriented in moderate fields with $c \parallel B$. However, the spin reorientation transition can be reached at $B = 25$ T apparently only with $B \perp c$, and thus the grains oriented with $c \parallel B$ pertain their low-field alignment. For the fixed-powder sample, on the other hand, there is a large fraction of grains with the ab -plane not inclined much from the field direction, and thus magnetization can reach higher values in this case. There remains only to check this conclusion by high-field magnetization measurements on the single crystal.

The point which is less clear is the crossover of the two branches of $\chi(T)$ dependencies. In antiferromagnets with weak anisotropy, the configuration of magnetic moments perpendicular to applied field has naturally a lower energy (higher susceptibility) than that with moments along the field direction. Such situation is, however, not typical for actinide intermetallics with strong anisotropy. Thus what we observe here is in fact an exceptional situation. Since a more general evidence of anisotropy in U_2T_2X compounds for various transition metals T is up to now missing, we can deduce only speculative conclusions about the reasons for weaker anisotropy. The magnetic anisotropy in U-intermetallics can be often understood as due to the anisotropic $5f$ -hybridization, which forces U magnetic moments perpendicular to strong $5f$ -bonding directions [4]. Apparently, the $5f$ - $5f$ coordination is an important parameter in such approach.

In U_2Pd_2In , each U atom has 7 U neighbours (5 within the basal plane and two along the c -axis). The U nearest-neighbour links are found along the tetragonal c -axis, but the distance between the U neighbours within the basal plane is only slightly larger. Thus one expects the U magnetic moments oriented within the basal plane, but the anisotropy energy can be rather small.

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Figure Captions

Fig.1: High-field magnetization results obtained on the two forms of powder samples described in the text. Full lines represent measurements with field swept continuously up and down. Magnetization obtained on the single crystal in $B = 5$ T is shown as large open symbols, and the corresponding field dependence is extrapolated by dotted lines.

Fig.2: Temperature dependence of magnetic susceptibility of the single crystal U_2Pd_2In compared with the original polycrystalline data (full line). The experiment on single crystal was done in $B = 1, 3$ and 5 T. A slight field dependence was observed only by the low temperature end of the $B \parallel c$ curve, where the higher field tend to suppress the susceptibility values (see the inset - (Δ) - 1 T, (\square) - 3 T, (\circ) - 5 T). Dash-dotted lines represent the respective modified Curie-Weiss fits.

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