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FERMI SURFACE MEASUREMENTS ON UPd_3

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The Fermi surface of the DO24 compound UPd_3 has been investigated by the de Haas-van Alphen effect in magnetic fields up to 46 Tesla. Two sheets are detected, one with a high frequency, about 4 10^3 Tesla, and one having low frequencies, about 4 10^2 Tesla. Values of the effectives mass obtained are in accordance with a non-f character of the electron wave function on the orbits investigated.

1. Introduction

The compound UPd_3 crystallyzes in the hexagonal DO24 structure with four formula units per unit cell[1]. Each U-atom is surrounded by a cage of 12 Pd atoms; the closest U-U distance is 4.81 A, too large for the formation of f bands[2]. The material has been the subject of several studies, by different techniques. Most of them agree on U^{4+} ion's with a 2f configuration, a canonical localized 5f-system, in a non magnetic J=4 ground state. A neutron diffraction study resulted in the detection of two strong excitations at 3.4 and 0.4 THz (resp. 163 and 19 K)[3,4,5], understood in terms of crystal field splitting of the groundstate. The low temperature properties are intrigueing too. Two phase transitions are observed at temparatures T about 7 and 5 K, in resistivity, thermal expansion, specific heat and susceptibility[6,7]. Ott[8] suggests that the transition is driven by the non magnetic quadrapolar interaction between the U-atoms. A large change in the c/a ratio accompagnizes the transitions.

Apart from the peculiar phase transitions for UPd_3 there exists a general interest in 5f systems, both from the experimental and the theoretical side. All questions concentrate on the (non) localization of the f-electrons.

In UPd_3 one deals with a U^{4+} system. Assuming that the 4d Pd bands are filled one has 4 electrons per formula unit and 16 per unit cell. Consequently UPd_3 is a compensated metal, with electrons in the hybridised s-d bands. This assumption is validated by the low value of γ, where the linear term γT is the contribution to the specific heat from the conduction electrons. Estimates for γ run from 4 to 10 mJ/mole.K^2; however it is difficult in the experimental data to separate γ from the large specific heat arising from the phase transitions. A comparison of XPS and BIS data for UPd_3 and $ThPd_3$ indicates that 5f-levels are close to the Fermi level, to within 1.5 eV[9,10].

2. Experiment and results

The material was synthesized by arc melting the constituents. After zone refining the raw material a single crystal was grown by remelting the UPd_3 by high frequency induction and slowly reducing the temperature. Samples of typical size 0.5x0.5x3 mm^3 were spark cut from the crystal and annealed in vacuum at 900°C for 24 hours. The final resistivity ratio $\rho(300K)/\rho(4.2K)$ was about 25, depending on the orientation.

Anomalies accompagning the low temperature phase transition are observed in various properties; their expression is dependent on crystal quality[6]. We measured the resistivity as function of temperature for one of the samples used for the Fermi surface (FS) study. For the current parallel to the [120] direction the results are given in fig. 1. The two transitions are clearly observable at temperatures 6.7 ± 0.2 K and in 5.0 ± 0.3 K. The values are in accordance with the earlier resisitivity data, measured on material with resistivity ratio 100.

We determined the shape of the FS by measuring the de Haas - van Alphen (dHvA) effect. Two sets of equipment were used. A 15 Tesla dc Bitter magnet in the Nijmegen high magnetic field laboratory, in which two low frequency orbits were observed. Here we used large field modulation detection at 185 Hz. A 40 T pulsed field magnet, described in detail elsewhere[11], was mainly used for the high frequency orbit.

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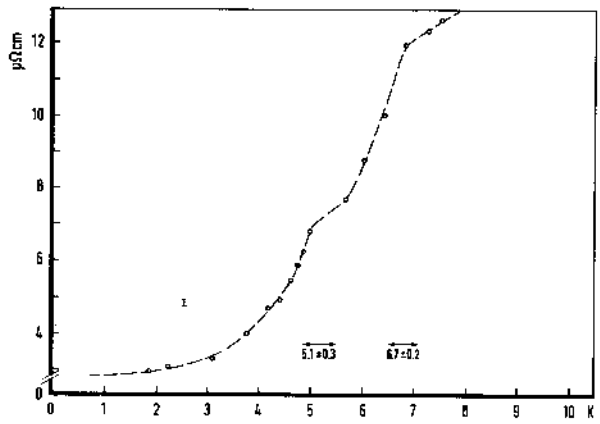

Fig. 1. Resistivity at low temperatures of one of the samples used for the dHvA investigation. The ordinate is units of absolute resistivity with an accuracy of 20%; The relative error is given by the vertical bar. The two anomalies due to the phase transition are indicated.

For the observation of the latter frequency the large field range of the pulsed system was essential in order to overcome the decrease in amplitude of the dHvA signal due to scattering. Data were taken at temperatures between 1.2 and 4.2 K.

The frequencies of the orbits observed are shown in fig. 2. Essentially two types of orbits are found, a high frequency branch labeled β, and low frequency branches labeled α_i. The amplitude of the β signal is weak in general; corresponding values for m^*T_D are about 16. A strong signal is observed only in the (010) plane at orientations indicated by the broken horizontal line. At 56° from the [001] axis (vertical broken line) a clear beat is seen in the β signal with a zero in amplitude at a field of 18.5 Tesla. The avarage frequency assumes a minimum value at this orientation. Consequently it is unlikely that the splitting is caused by mozaic spread in the sample.

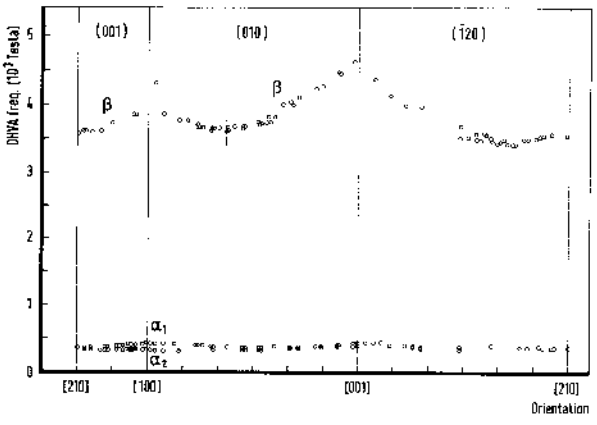

Fig. 2. Observed dHvA frequencies for UPd_3 in the main symmetry planes. Details are given in the text.

Two branches of low frequency are observed in the (010) and the (001) plane. In the (1̄20) plane only one orbit is detected clearly. For the field in the vincinity of the [100] axis the higher frequency α_1 orbit has the larger amplitude; along the [001] orientation the signal belonging to the lower frequency orbit is stronger. The two branches do not merge at the [210] orientation. From this and from the behaviour of the amplitude with orientation one concludes that two different sheets for the α_1 and α_2 branches exist.

Both the β and at least one of the α branches can be followed in all three high symmetry planes, indicating that the corresponding sheets are closed surfaces.

3. Discussion

We try to construct a model for the FS from the observed frequencies. The Brillouin zone (BZ) for UPd_3 is given in fig. 3; the area of the hexagonal basal plane correponds to a dHvA frequency F of $14.4\ 10^3$ T, the maximum cross section perpendicular to the line ΓM to a value F = $8.6\ 10^3$ T. The high frequency belongs to a deformed sphere; for B parallel to the [001] axis we find F = $4.8\ 10^3$ T, parallel to the [100] axis F = $4.2\ 10^3$ T. Its size fits in the BZ, its volume is about 0.2 electron per unit cell per spin. When looking for a centre of high symmetry one cannot place the orbit around the M, K, H, or L point; the orbits linear dimension does not fit in the side plane of the BZ. From this and from the observation of essentially only one β frequency (fig. 2.) one concludes that the spheroid has a single multiplicity, and has its centre at A or Γ.

For the α frequencies the situation is less clear. The angular dependence is small; a near degeneracy occurs for the field along [210]. At least one of the signals appear at all orientations, again suggesting closed sheets. The corresponding volume is about a factor 30 smaller than that of the β sheet. In UPd_3 the total

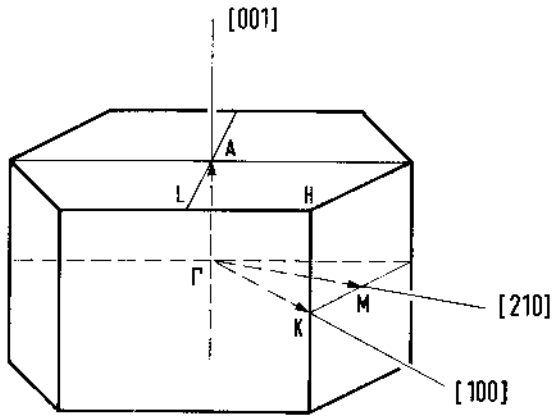

Fig. 3. Brillouin zone of UPd_3 with main symmetry points.

volume in the BZ enclosed by electron sheets must be equal to the one enclosed by hole sheets. From the dHvA data alone one cannot distinguish between the two types. In the simplest picture one assumes the small α pockets to belong to the other type of carriers as the β sheet. However, even when the pockets are at twelve symmetry connected points in the BZ, their volume cannot compensate for the one of the β sheet. Consequently, unobserved parts of the FS must exist. A large amount of scattering due to the limited crystal quality -for the β orbit m^*T_D values of about 16 are found- may account for their non observation.

For several directions of the magnetic field we derived the values of the effective mass from the temperature dependence of the amplitude of the dHvA magnetisation. For the field 62° from [001] towards [100] the value for m^* is 2.0 ± 0.2 for the β orbit. For the α_1 orbit we find m^* equal to 0.75 ± 0.20 with the field along [100] and 0.5 ± 0.1 along [001]. No temperature dependence is obtained for the α_2 frequency, having too small an amplitude. Both values for the electron mass are in accordance with a small or negligible contribution of the U f wavefunction to the electron states for the orbits investigated.

Taking the value of m^* as representative for the whole sheet, one can estimate its contribution to the density of states, aequivalent to γ of 4 $mJ/mole.K^2$. This value is of the same magnitude as experimental results for the total γ, derived from specific heat measurements. Consequently the unobserved parts of the FS cannot be of a too heavy mass.

In conclusion, dHvA measurements can be performed fort a material that undergoes two phase transistions at temperatures slightly higher than the ones employed in the experiment. The influence of the transitions on the FS and properties like m^* or T_D is not known; it presents itself as an interesting subject of an followup study.

The splitting observed for the β branch at 56° from the c to the a axis may tentatively be attributed to a spin split FS. The fact that two α braches are observed may have a similar cause. A bandstructure for UPd_3 has been calculated/12/; sheets of a size comparable to the one of the β orbit are found. The shape of the full calculated FS is sensitive to a number of parameters, eg. the Fermi level. A comparison with the dHvA data presented here proved not to be satisfactory yet.

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