

HIGH-FIELD TRANSITIONS IN URu₂Si₂ OBSERVED BY MAGNETORESISTIVITY AND MAGNETIZATION EXPERIMENTS

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(Received 16 March 1987 by P. Wachter)

High-field magnetoresistivity and magnetization experiments have been performed on a monocrystalline sample of the antiferromagnetic superconductor URu₂Si₂, for a field direction along the tetragonal axis. At a temperature of 1.5 K three sharp transitions are observed at fields of 35.8, 37.3 and 39.4 T, suggesting a complex magnetization process. Large jumps in the magnetoresistance indicate that the magnetization process involves gap-like changes of the Fermi surface.

1. INTRODUCTION

SCHLABITZ *et al.* [1], PALSTRA *et al.* [2], and Maple *et al.* [3] have reported the existence of both an antiferromagnetic and a superconducting transition in the intermetallic compound URu₂Si₂. The transition to the magnetically ordered state, at $T_N = 17.5$ K, is discerned by a λ -like anomaly in the specific heat, a kink in the susceptibility curve and a pronounced anomaly in the electrical resistivity. Superconductivity is observed below $T_c \cong 1$ K. Specific heat measurements [1, 2, 3] and a study of the Meissner effect [1] demonstrate that superconductivity is a bulk property. Analyses of the uppercritical field, employing the relatively large value of the electronic specific heat just above T_c ($\gamma \cong 50$ mJ mol K⁻²), yield reported effective electron masses of 25 [3], 50 [1] and 147 [4] in units of m_e . Hence, URu₂Si₂ can be classified as a moderately-heavy-fermion system and is unique in the sense that it is the only heavy-fermion system that exhibits both an antiferromagnetic and a superconducting transition.

The exponential temperature dependence of the specific heat below T_N suggests that magnetic order is brought about by the opening of an energy gap over a part of the Fermi surface [2, 3]. This brings URu₂Si₂ close to the antiferromagnetic heavy-fermion systems U₂Zn₁₇ [5], UCd₁₁ [6] and UPt₃ alloyed with Pd [7]. For this class of compounds Fermi surface instabilities have been invoked to account for the transition to the antiferromagnetic ground state. On the other hand a comparison of URu₂Si₂ with order UT₂Si₂ com-

pounds (where T is a transition metal), brings to light that magnetic order is a general feature in this series of compounds [8, 9]. Based on such a comparison, Nieuwenhuys [10] suggested that magnetic order in URu₂Si₂ is induced by crystal-field effects. This is supported by a crystal-field calculation which qualitatively reproduces the observed temperature and field dependence of the magnetization [10], and the Schottky-like anomalies in the specific heat [1] and thermal expansion [11] above T_N .

Recently, neutron-diffraction experiments [12] have revealed that the ordered moment ($0.03 \pm 0.01 \mu_B$ /U-atom, directed along the *c*-axis) is nearly 2 orders of magnitude smaller than those (1.6–2.9 μ_B /U-atom) observed for other ordered compounds of the UT₂Si₂ series. However, in a previous publication [13], we reported magnetization measurements revealing that a magnetic moment of this order of magnitude is realized for URu₂Si₂, at 1.4 K, in a two-step process, between 35 and 40 T. These transitions, only observed for a field direction along the tetragonal axis, were taken as evidence for a complex magnetic structure. In order to elucidate these high-magnetic field transitions, we performed magnetoresistivity measurements up to 41 T, as well as additional magnetization measurements. Low-field ($B < 7$ T) magnetoresistance data obtained by Palstra *et al.* [4] revealed the presence of a quadratic term, $\Delta\rho/\rho = aB^2$, in the antiferromagnetic phase.

2. EXPERIMENTAL

Magnetoresistivity measurements were performed in the Amsterdam High Field Installation [14]. Magnetic fields up to 41 T were attained by means of

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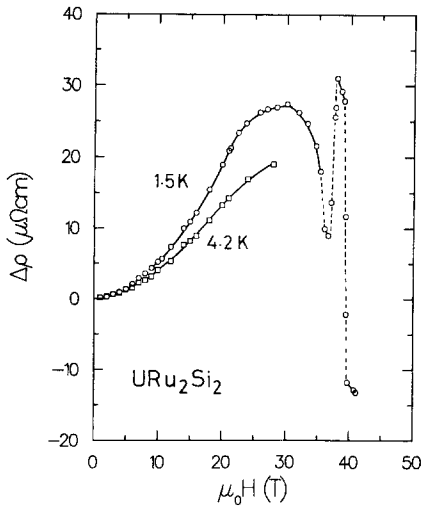


Fig. 1. Magnetoconductivity ($B \parallel I \parallel c$ -axis) of annealed URu₂Si₂ at temperatures indicated. The zero-field resistance equals 18.8 and 21.8 $\mu\Omega$ cm at 1.5 and 4.2 K, respectively. Dashed lines indicate transitions. Solid lines serve as a guide to the eye.

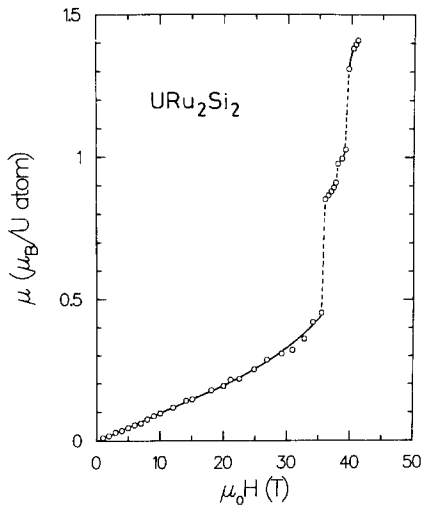


Fig. 2. Magnetization of annealed URu₂Si₂ along the c -axis at 1.5 K. Dashed lines indicate transitions. Solid lines serve as a guide to the eye.

a special pulse with a sweep-time of 0.6 s. (field up: 0.4 s., field down: 0.2 s.; see also [13]). The change in resistivity as function of field was measured with a standard d.c.-method. Both current ($I = 0.65$ A) and field were applied along the tetragonal axis. Data were taken at 1.5 and 4.2 K. Stable temperatures were guaranteed by immersing the samples directly in the cryogenic liquid. The relative accuracy is estimated to be 3%, whereas the absolute accuracy amounts to 7% due to the determination of the geometrical factor. A large single-crystalline sample of URu₂Si₂ was

prepared by means of the Czochralski technique. After annealing (at 900°C for a period of 9 days) a cylindrical specimen (diameter = 2 mm, length = 6 mm) was cut from the bulk crystal, by means of the spark erosion technique. At room temperature, the resistivity value of the annealed sample amounts to 264 $\mu\Omega$ cm, in good agreement with previous results on similar samples [4, 13]. Magnetization experiments have been performed by the method described in [13].

3. RESULTS

In Fig. 1, we present the magnetoconductance of URu₂Si₂, plotted as $\Delta\rho = \rho(T, B) - \rho(T, 0)$ vs B . The data points were sampled from a recorder trace, taken with increasing field. The zero-field resistivity values amount to 21.8 and 18.8 $\mu\Omega$ cm at T equals 4.2 and 1.5 K, respectively. At low fields ($B < 7$ T) $\Delta\rho$ is positive and follows a quadratic field dependence. The proportionality constant yields values of 0.046 and 0.060 $\mu\Omega$ cm/T² at 4.2 and 1.5 K, respectively, in agreement with the data of [4]. Focusing on the data at 1.5 K, the steady increase of $\Delta\rho$ is followed by a broad maximum at $B = 29$ T. The decrease of $\Delta\rho$ above 29 T seems to be a precursor of the rapid drop at higher fields. Above 35 T the most striking results of the present data are observed: 3 steps of $\Delta\rho$ (at 1.5 K) at fields of 35.8 ($\equiv B_1$), 37.3 ($\equiv B_2$) and 39.4 T ($\equiv B_3$), amounting to -8.1 , 22.0 and -39.7 $\mu\Omega$ cm, respectively. When compared with the $\rho(1.5\text{ K}, 0)$ -value these steps are surprisingly large: -43 , 117 and -211% , respectively. At 41 T the resistance of the specimen yields the low value of 5.6 $\mu\Omega$ cm. The transitions are rather sharp, as is indicated by a width, ΔB , of 0.7, 1.3 and 0.6 T, respectively. Hysteresis effects were not observed within experimental accuracy.

Magnetization measurements on this sample at 1.5 K, for a field applied along the tetragonal axis, are shown in Fig. 2. The magnetization is linear up to ≈ 15 T, with a slope $\chi = 66 \pm 2 \times 10^{-9}$ m³/molU-atoms. Just as in Fig. 1, 3 discontinuities are observed in the high-field region: at 35.7, 37.8 and 39.4 T. Taking into account the width, ΔB , of the transitions, 0.53, 0.33 and 0.44 T, the observed transition fields correspond nicely with the values for B_1 , B_2 and B_3 defined above. The increase of the magnetic moment at B_1 , B_2 and B_3 , amounts to 0.40, 0.06 and 0.28 μ_B /U-atom, respectively. At 41 T the magnetization yields a magnetic moment of 1.41 μ_B /U-atom. As in the case of the magnetoconductivity experiments, hysteresis effects were not observed.

Previous magnetization data [13], taken on an unannealed specimen (field along the c -axis), revealed

only 2 transitions: at 36 T ($\Delta\mu = 0.45 \mu_B/\text{U-atom}$) and at 40 T ($\Delta\mu = 0.29 \mu_B/\text{U-atom}$). The fields at which these transitions occur, correspond well with the values for B_1 and B_3 . When repeating the magnetization experiments on the unannealed sample (with a better resolution of the recorder trace than in [13]) we observed a faint indication of the transition at B_2 . Apparently, in the unannealed sample, this transition is broadened into the field region $B_1 < B < B_3$. We emphasize that the transition at B_2 cannot be due to the presence of a slightly different oriented grain in the specimen (such a grain would yield a somewhat higher value for B_1 due to its misorientation). This is directly evidenced by the different signs of the steps in $\Delta\rho$: negative at B_1 and B_3 , positive at B_2 .

4. DISCUSSION

As mentioned in the introduction, the anomalies observed at 17.5 K in the susceptibility, specific heat and resistivity of URu₂Si₂ are interpreted to reflect the onset of antiferromagnetism. The quadratic field dependence of the magnetoresistance (for $B < 7$ T) is consistent with this interpretation [4]. Accepting such an analysis, it is plausible that at large fields the antiferromagnetic order is destroyed, and that, via a spin-flopped state and/or another intermediate spin structure, a ferromagnetic phase is induced. Assuming the magnetic moments to be the effective scattering centers, we might explain the high-field data as follows. The transitions at B_1 and B_3 are to a state with less effective scattering, suggesting the presence of a more coherent phase. The latter transition possibly being the one to the ferromagnetic phase, given the low ρ -value: $5.6 \mu\Omega \text{ cm}$. However, since $\Delta\rho$ and μ have not reached a constant value, the ferromagnetic alignment will not be perfect, yet. The sudden increase of $\Delta\rho$ at B_2 reflects a transition to an incoherent phase, possibly an incommensurate magnetic structure.

However, the interpretation of these anomalies as being caused merely by transitions to different magnetic structures with different effective scattering rates, is thwarted by the small magnitude of the magnetic moment ($\approx 0.03 \mu_B/\text{U-atom}$), as determined by neutron experiments [12]. Therefore, it is likely that the variation of the magnetic structure is accompanied by gap-like changes of the Fermi surface as is observed, for instance, in antiferromagnetic Ho. In this rare-earth metal a spiral \rightarrow fan \rightarrow ferromagnet transition occurs under influence of a magnetic field. The resulting magnetoresistivity [15] and magnetization [16] curves resemble the data shown in Figs. 1 and 2. In the case of URu₂Si₂ this would imply that the transition at B_1 restores a part of the Fermi surface, that previously

has been removed (at zero field) at T_N . At B_2 another gap is opened, followed by a reconstruction of the Fermi surface at B_3 . Hall-effect measurements performed by Schoenes *et al.* [17] support such an interpretation. The observed large jump in the Hall coefficient at T_N , indicates that the carrier concentration strongly alters when the gap opens. Analysing the data in this way the broadening of the transition at B_2 in the unannealed specimen can be explained by assuming a strong stress dependence of the Fermi surface.

At present, it is not clear whether crystal-field effects play a role in the high-field behaviour. According to crystal-field calculations performed by Nieuwenhuys [10], the ground state (assuming tetravalent U) consists of a singlet. Excited levels are found at 42 (singlet-magnetic), 169 (singlet-non-magnetic) and 550 K (doublet). In that picture, it cannot be excluded that the magnetic-singlet is populated at high fields. In that case, one of the transitions might be of the crystal-field type.

Recent measurements on URu₂Si₂ doped with Th [8] indicate that the magnetic correlations become weaker on alloying. Therefore, one may expect that in the doped compounds the transition fields are substantially lower. This would facilitate the further experimental investigation of these high-field phenomena.

In conclusion, we have presented high-magnetic field magnetoresistivity and magnetization data on the antiferromagnetic superconductor URu₂Si₂. At high fields 3 transitions are observed at 1.5 K, indicating a complex magnetization process, involving gap-like changes of the Fermi surface.

Acknowledgements — The authors are grateful to L.W. Roeland and R. Gersdorf for experimental support. This work was part of the research program of the Dutch "Stitching FOM".

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