

Hall effect of URu₂Si₂ in very high magnetic fields up to 40.5 T

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The Hall coefficient (R_H) of the heavy-fermion compound URu₂Si₂ has been measured in very high magnetic fields directed along the tetragonal axis at temperatures of 0.6, 1.5 and 4.2 K. At low fields, $R_H = 15 \times 10^{-3} \text{ cm}^3/\text{C}$, but is reduced to almost zero in the high-field limit (40.5 T). The three high-field transitions, previously observed in the magnetization and the magnetoresistance, are also identified in the Hall resistance at fields of 35.6, 36.2 and 39.2 T at $T = 1.5 \text{ K}$.

1. Introduction

The heavy-fermion compound URu₂Si₂ has attracted a great deal of attention because of the coexistence of antiferromagnetic order ($T_N = 17.5 \text{ K}$) and superconductivity ($T_c = 1.3 \text{ K}$) [1]. Specific heat and resistivity measurements indicate that the antiferromagnetic order is of the spin-density wave type, with a gap ($\Delta = 130 \text{ K}$) opening over a part of the Fermi surface. The magnetic properties can be explained to a certain extent with a crystalline electric field model with singlet–singlet-induced magnetic ordering [2]. However, theory has failed so far to account correctly for a particularly puzzling property of URu₂Si₂, namely the extremely small ordered moment ($0.03\mu_B/\text{U}$ atom, oriented along the tetragonal axis) detected by neutron scattering [3]. Reduced-moment antiferromagnetism has also been observed in several other heavy-fermion compounds, and is generally ascribed to the strong c–f hybridization.

However, by applying a high magnetic field along the tetragonal axis (*c*-axis), i.e., the easy axis for magnetization, a three-step magnetization process occurs, with steps located at $B_1 = 35.8 \text{ T}$, $B_2 = 37.3 \text{ T}$ and $B_3 = 39.4 \text{ T}$ at $T = 1.5 \text{ K}$ [4,5]. At the highest fields a substantial magnetic moment is induced ($1.4\mu_B/\text{U}$ atom at $B = 40 \text{ T}$) and the antiferromagnetic order is suppressed. Three transitions are also observed in the high-field magnetoresistance, where they occur as large jumps [5]. In order to explain the

complex magnetization process several mechanisms have been evoked, among which a reconstruction of the Fermi surface, a destruction of the Kondo screening, followed by changes in magnetic structure, and a crossing of crystalline electric field levels. However, a satisfactory model is still lacking.

In order to further elucidate the suppression of the antiferromagnetic state we have performed high-field Hall-effect measurements. The temperature variation of the low-field Hall resistance (R_H) has been investigated by several groups [6,7]. A comparison with the magnetic susceptibility $\chi(T)$ is made using the relation $R_H(T) = R_0 + 4\pi\chi(T)R_s$, where R_0 and R_s are the ordinary and extraordinary Hall coefficients, respectively. R_0 is due to the ordinary Hall effect and skew scattering by residual defects, while R_s is due to intrinsic skew scattering. Near the antiferromagnetic transition an enormous variation of R_H is observed ($B||c$), indicating an increase in R_0 by a factor of 6, as $\chi(T)$ shows only a kink at T_N . The increase in R_0 evidences a reconstruction of the Fermi surface at T_N . Encouraged by the large variation of R_0 near T_N , we have investigated the Hall resistance near the high-field antiferromagnetic phase boundary.

2. Experimental

A single-crystalline URu₂Si₂ sample (tetragonal ThCr₂Si₂ structure) was prepared in a modified tri-arc furnace under a continuously gettered argon atmosphere using the Czochralski technique. A thin plate-like sample (dimensions $0.1 \times 2 \times 4 \text{ mm}^3$) was cut from the single-crystalline batch by means of an accur-

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ate miniature spark erosion technique. The sample has six extrusions in the geometry of a Hall bridge, allowing for both magnetoresistance and Hall-effect measurements. Measurements of the Hall resistance with the field up and down showed that the resistive contribution to the Hall signal was less than 1%, indicating an almost perfect sample geometry. Electrical contacts on the sample were realized by silver paint. The Hall resistance and the magnetoresistance were measured with a DC method with the current along the *a*-axis ($I = 100$ mA at $T = 1.4$ and 4.2 K, $I = 25$ mA at 0.6 K). The magnetic field was applied along the *c*-axis (normal to the plate). The sign of the Hall coefficient was taken from ref. [6]. Long-pulse magnetic fields up to 40.5 T (total pulse time ~ 1 s) were produced in the High Magnetic Field Installation of the University of Amsterdam. The measurements at $T = 1.4$ and 4.2 K were performed using a standard bath cryostat, with the sample immersed directly in liquid ⁴He. Data at 0.6 K were taken using a simple ³He insert, with the sample immersed in pumped ³He.

3. Results

The field variation of the Hall coefficient for a current along the *a*-axis and a field along the *c*-axis at temperatures of 0.6 , 1.5 and 4.2 K is shown in fig. 1. At low fields ($B < 15$ T) R_H is quasi-constant, with a value amounting to 15×10^{-3} cm³/C, which is slightly larger than the value 13×10^{-3} cm³/C reported in literature [6,7]. At $T = 0.6$ K, the sharp increase at a field $B_{c2} = 2$ T, is caused by the superconducting to normal phase transition. In the high-field range $R_H(B)$ displays a significant structure. A first anomaly appears as a local maximum at $B_{\max} = 29$ T at $T = 0.6$ K. At increasing the temperature the anomaly weakens

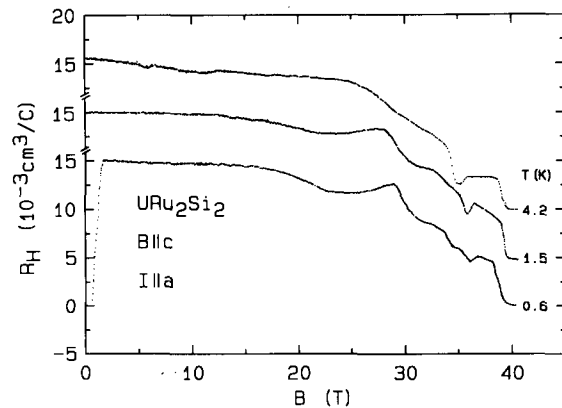


Fig. 1. The field variation of the Hall coefficient of URu₂Si₂ for $I||a$ and $B||c$ at temperatures as indicated.

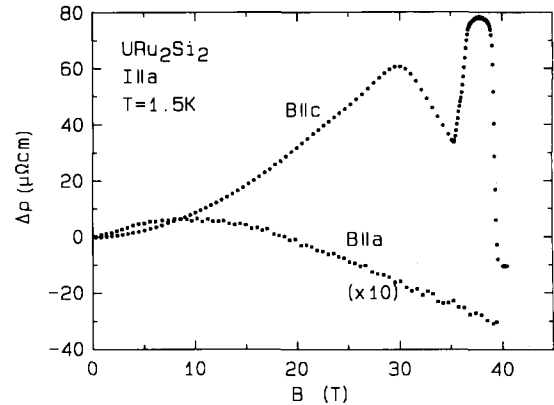


Fig. 2. The transverse ($B||c$ and $I||a$) and the longitudinal ($B||I||a$) magnetoresistance of URu₂Si₂ at $T = 1.5$ K. Note that the curve for $B||I||a$ is multiplied by a factor of 10. In zero field $\rho(0) = 16.3$ $\mu\Omega$ cm.

and shifts to lower fields: $B_{\max} = 28$ T at 1.5 K and $B_{\max} = 26$ T at 4.2 K. The three high-field transitions, previously observed in the magnetization and magnetoresistance [5], appear as consecutive steps (down, up and down), in the field range 34 – 40 T. The two lower transitions were found to exhibit a weak temperature dependence: $dB_1/dT \approx dB_2/dT \approx -0.3$ T/K, while the upper transition at B_3 is almost temperature independent. At $B_3 = 39.2$ T the Hall coefficient drops precipitously to a value close to zero.

Transverse ($B||c, I||a$) and longitudinal ($B||I||a$) magnetoresistance data, taken on the same sample at $T = 1.5$ K, are shown in fig. 2 in a plot of $\Delta\rho(B) = \rho(B) - \rho(0)$ versus B . A broad maximum appears at 30 T, slightly above $B_{\max} = 28$ T observed in $R_H(B)$. In the high-field range B_1 and B_2 are observed as steps upwards, while B_3 is observed as a step downwards. The three high-field transitions are detailed in fig. 3, where we have plotted $\Delta\rho(B)$ and $R_H(B)$ on an

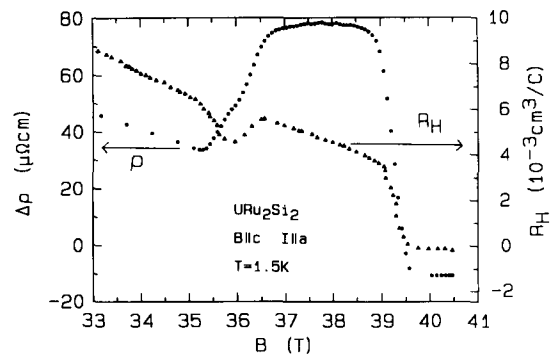


Fig. 3. Field variation of R_H (\blacktriangle) and $\Delta\rho$ (\bullet) of URu₂Si₂ near the three high-field transitions at 1.5 K ($B||c$ and $I||a$). The transitions (midpoints) are located at $B_1 = 35.6$ T, $B_2 = 36.2$ T and $B_3 = 39.2$ T.

extended scale. We deduce values for B_1 , B_2 and B_3 of 35.6, 36.2 and 39.2 T, respectively (at $T = 1.5$ K), in agreement with our previously reported values of 35.8, 37.3 and 39.4 T, respectively [5]. However, our previous magnetoresistance experiments ($B \parallel I \parallel c$) indicated a downward step in $\Delta\rho$ at B_1 . It is not clear whether this difference is caused by the different configuration (longitudinal versus transverse magnetoresistance) or the poorer resolution in the previous experiment. The high-field magnetization and the transverse high-field magnetoresistance ($B \parallel c, I \parallel a$) were also investigated by Sugiyama et al. [8]. Their magnetoresistance data are in good agreement with the data in fig. 2, but show a rounding at the transitions because of the shorter pulse times. From magnetization measurements at $T = 1.3$ K these authors deduced values for B_1 , B_2 and B_3 of 35.8, 36.5 and 39.6 T, respectively, in agreement with our values quoted above. The longitudinal magnetoresistance ($B \parallel I \parallel a$) is more than one order of magnitude smaller than the transverse magnetoresistance (fig. 2). It is positive for small fields, displays a maximum at 8 T, and becomes negative for $B > 19$ T.

4. Discussion

The most striking result of the present experiments is the strong reduction of the Hall coefficient above 40 T. At low fields the ordinary Hall coefficient amounts to $R_0 = 15 \times 10^{-3} \text{ cm}^3/\text{C}$, which corresponds to a carrier concentration of 0.03 holes/f.u. in a simple one-band model [6] (this is of course a rather crude approximation for a complicated metal as URu₂Si₂). At 40 T, $|R_H| < 0.5 \times 10^{-3} \text{ cm}^3/\text{C}$ (the experimental uncertainty does not allow for the determination of the sign), which indicates an increase of the carrier concentration to a value larger than ~ 1 electron or hole per f.u. Such a large variation of the carrier concentration strongly suggests a reconstruction of the Fermi surface, i.e., the gap, opened in zero field at T_N , is closed again. Note that for temperatures above T_N also a substantial carrier concentration of 0.5 electrons/f.u. was deduced [6] (again in the simple one-band model). The pronounced structure in R_H at high fields suggests that the reconstruction of the Fermi surface takes place in a complicated three-step process. This is further supported by the large changes in the magnetoresistance observed at B_1 , B_2 and B_3 .

The maximum in $R_H(B)$ and $\Delta\rho(B)$ near $B_{\text{max}} = 30$ T is yet another puzzling aspect of URu₂Si₂. In the

high-field magnetization no clear anomaly is observed at this field, although $M(B)$ becomes somewhat steeper. In our prior publication [5] it was suggested that the decrease in $\Delta\rho$ above B_{max} was a precursor of the transition at B_1 . However, the present experiments suggest that it is an independent phenomenon, which indicates a pronounced change in the electronic properties. An appealing interpretation of the anomaly at B_{max} is that it signals the destruction of the Kondo screening, leading to an increase in the size of the ordered moment (it increases up to $0.5\mu_B/\text{U}$ atom for $B \rightarrow B_1$). Subsequently, the antiferromagnetic order is suppressed in a three-step process, indicating the presence of two intermediate magnetic structures. The large variation in the Hall coefficient at high fields supports the idea that the suppression of the antiferromagnetic state is accompanied by a reconstruction of the Fermi surface.

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