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Electrical transport in URu_2Si_2

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Abstract. The temperature dependence of the resistivity, magnetic susceptibility and Hall coefficient have been measured in oriented single crystals of URu_2Si_2. The susceptibility is Curie–Weiss like at high temperatures and the anisotropy shows that the f moments on the U atoms are highly confined to lie along the *c* direction. The resistivity is analysed in terms of a Kondo-like behaviour at high temperatures and shows the onset of coherence, modified by electron-spin wave scattering, at low temperatures. There is a sharp superconducting transition at 1.33 K. The Hall coefficient is analysed by using a modification of the recent theory by Fert and Levy and suggests that the coherence temperature T_0 and the Kondo temperature T_K need not be sharply distinguished in URu_2Si_2. There is a large increase in the Hall coefficient as the temperature is lowered below 18 K which is attributed to a Fermi surface restructuring, possibly due to the existence of spin density waves in the antiferromagnetic state.

1. Introduction

URu_2Si_2 is a body-centred tetragonal system which has been shown to have properties that characterise it as a heavy-fermion metal. Formerly, there were three known ground states in heavy-fermion metals: superconductors ($CeCu_2Si_2$, UBe_{13}), antiferromagnets (UCd_{11}, U_2Zn_{17}) and normal metals down to 20 mK ($CeAl_3$, $CeCu_6$) (Lee *et al* 1986). Highly anisotropic URu_2Si_2 emerged as the first heavy-fermion metal that is superconducting, with a critical temperature of approximately 1 K, and orders magnetically with an antiferromagnetic Néel temperature of 17.5 K. Superconducting UPt_3 has been found very recently to be also antiferromagnetic (Aeppli *et al* 1988).

Since the initial work by Cordier *et al* (1985), a considerable amount of experimental data have been collected for URu_2Si_2. Specific heat and magnetic susceptibility measurements have been carried out by Palstra *et al* (1985), Schlabitz *et al* (1986) and Maple *et al* (1986). Measurements of the second critical field have been made by Palstra *et al* (1986) and Rauchschwalbe (1987) and Meissner effect measurements were done by Schlabitz *et al* (1986). Resistivity (Schlabitz *et al* 1986, Maple *et al* 1986, Palstra *et al* 1986), magnetoresistance (Palstra *et al* 1986) and Hall effect (Schoenes *et al* 1987) in URu_2Si_2 have also been investigated. Further measurements on URu_2Si_2 include elastic (Broholm *et al* 1987) and inelastic (Walter *et al* 1986, Holland-Moritz *et al* 1987) neutron scattering as well as light scattering (Cooper *et al* 1987).

The present work investigates single crystals of URu_2Si_2 with measurements of the DC resistivity, magnetic susceptibility and Hall effect. Section 2 provides a discussion of these three properties and a development of some of the theoretical ideas needed to

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understand them in URu_2Si_2. In § 3, the experimental method is described followed by a presentation of the measurements. Section 4 presents the analysis and the discussion of the results while § 5 summarises the important results of the work.

2. Background

The high-temperature, DC magnetic susceptibility of a heavy-fermion metal with N ions is Curie–Weiss like:

$$\chi_{\text{molar}} = N\mu_{\text{eff}}^2/3k_B(T - \theta)$$

with the Curie temperature θ less than zero and the effective magnetic moment, μ_{eff}, greater than $2\,\mu_B$. In no heavy-fermion metal does μ_{eff} correspond to one calculated from Hund's rules for a single electron configuration. In antiferromagnetic heavy-fermion metals, χ diverges from the Curie–Weiss fit at low temperatures, indicating that the antiferromagnetism may be itinerant (Stewart 1984).

In a normal metal, the resistivity can be written $\rho = m/ne^2\tau$ where m is a band mass and n is the carrier density. $1/\tau$ is the scattering probability of Bloch states and is the factor that leads to the temperature dependence of the resistivity in a normal metal. The resistivity of a Bloch metal above and below the Debye temperature, θ_D, is:

$$\rho = \rho_0 + bT^5 \qquad T \ll \theta_D$$

$$\rho = \rho_0 + cT \qquad T \gg \theta_D.$$

The two terms in both expressions are due to impurity and electron–phonon scattering. In normal metals, ρ is of order 1 $\mu\Omega$ cm at room temperature while ρ is enhanced by two or three orders of magnitude in heavy-fermion metals.

At low temperatures in metals with dilute magnetic impurities, the resistivity rises as the temperature is lowered. Known as the Kondo effect, this phenomenon is understood through a rather subtle argument accounting for second-order scattering processes of conduction electrons by localised magnetic electrons through an antiferromagnetic exchange interaction (Dugdale 1977). This scattering causes the magnetic resistivity to go as

$$\rho(T) = \rho_0 - \rho_s \ln T.$$

Above a characteristic Kondo temperature T_K, this behaviour is obeyed by a heavy-fermion metal and $d\rho/dT$ for $T \gg T_K$ is negative. The Kondo effect is enhanced with temperature and the material acts as a lattice of isolated Kondo impurities.

As the temperature is lowered, the resistivity goes through a maximum $\rho_{\max}$ at a temperature $T_{\max}$. $\rho_{\max}$ can be used to estimate the Fermi wavevector k_F from Friedel's rule:

$$\rho_{\max} = 4\pi\hbar x(2l + 1)/e^2k_F\nu.$$

The relation is valid if only one l-value dominates the scattering. x is the fraction of f-electron atoms and ν is the number of electrons per atom (Friedel 1958).

Below the coherence temperature T_0 which is approximately equal to $T_{\max}$, the scattering becomes Bloch like and the resistivity drops dramatically. Presumably due to spin fluctuations, the heavy-fermion metal exhibits Fermi liquid behaviour which leads to a resistivity ρ_{FL} proportional to T^2. It should be noted, however, that mechanisms other than spin fluctuations can lead to a T^2- dependence in the resistivity.

If an energy gap from spin waves is present in an antiferromagnet, putting the dispersion $\omega_{sw}(\boldsymbol{k}) = \Delta + bk^2$ into the linearised Boltzmann equation yields a resistivity due to electron-spin wave scattering (Anderson 1980):

$$\rho_{sw} = T/\Delta(1 + 2T/\Delta)\,e^{-\Delta/T} \qquad \text{at } T \ll \Delta.$$

Resistivities due to different scattering processes may be added by Matthiessen's rule to obtain the total resistivity.

In a normal metal, the classical Drude expression for the Hall coefficient $R_H = E_y/j_xH_z = 1/ne$ can be retained if n is the density of carriers of charge e and if there is no skew scattering; that is, if the electron scattering time $\tau(\boldsymbol{k}, s)$ is independent of the wavevector, $\boldsymbol{k}$, and the spin s. In a normal metal, the Hall coefficient should therefore be temperature independent since τ does not enter into it. A small temperature dependence of R_H is observed in some simple metals due to the freezing out of Umklapp electron–phonon scattering processes, which are clearly anisotropic in k-space (Hurd 1972).

In magnetic materials, the effect of skew scattering has been described by the empirical *ansatz*

$$R_H(T) = R_0 + 4\pi\chi_{volume}(T)R_s.$$

This relation will here be referred to as the Hurd relation. R_0 and R_s are the temperature independent ordinary and extraordinary Hall coefficients (Hurd 1972).

Fert and Levy (1987) have derived a general expression for the Hall coefficient resulting from intrinsic skew scattering in heavy Fermion metals:

$$R_H^{MAG} = \gamma\tilde{\chi}(T)\rho_m(T)$$

where $\tilde{\chi}(T)$ is the reduced susceptibility $\tilde{\chi}(T) = \chi(T)/C_{cw}$ with $C_{cw} = N\mu_{eff}/3k_B$, and ρ_m is the magnetic resistivity. γ takes on two different values in two different limits.

$$T \gg T_K \qquad \gamma_1 = (-5\,g\mu_B/7\,k_B)\sin\delta_2\cos\delta_2$$

$$T_0 \lesssim T \lesssim T_K \qquad \gamma_2 = (-5\pi g\mu_B/21\,k_B)\sin(2\delta_3 - \delta_2)\sin\delta_2/\sin^2\delta_3.$$

The δ_i are the phase shifts in the $l = i$ channel and T_0, T_K are the coherence and Kondo temperatures, respectively. The Fert–Levi (FL) model has been successfully applied to UPt_3, UAl_3, $CeRu_2Si_2$ and $CeCu_6$. No theory exists for $R_H^{MAG}(T)$ below T_0. In the low-temperature limit, $R_H(T)$ in different heavy-fermion metals has been shown to have a variety of power-law dependences (Hadzic-Leroux *et al* 1986).

Two quantities have emerged that are roughly constant for all heavy-fermion metals. They are the Wilson ratio, a weighted ratio of the susceptibility at $T = 0$ to the Sommerfeld $\gamma(0)$ (Lee *et al* 1986) and the Kadowaki–Woods (Kodowaki and Woods 1986) ratio, the ratio of the T^2 coefficient of the low-temperature resistivity to the square of $\gamma(0)$.

In summary, the following are the various temperatures and energies in URu_2Si_2: T_c is the superconducting critical temperature, T_N is the antiferromagnetic ordering temperature, T_0 is the coherence temperature and T_K is the Kondo temperature above which the resistivity is Kondo like. Δ_{SW} is the energy gap in the spin wave dispersion and Δ_{FS} is the Fermi surface energy gap measured in specific heat. θ is the temperature in the Curie–Weiss expression. There is also T_{max}, the temperature of the maximum of the resistivity.

3. Experimental procedure

Single crystals of URu_2Si_2 prepared from stoichiometric amounts of U, Ru and Si, were grown in a Reed-type triarc furnace. Extensive modifications have been made to the

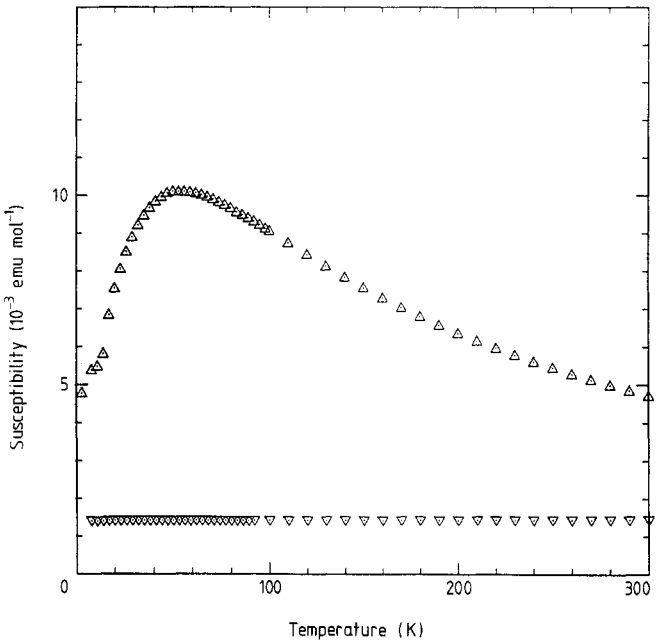

Figure 1. DC magnetic susceptibility of URu_2Si_2 with the magnetic field $\boldsymbol{B} \| \boldsymbol{c}$ (Δ) and $\boldsymbol{B} | \boldsymbol{c}$ (∇).

apparatus to provide direct water cooling to the Czochalski seed rod to the hearth. A commutator was installed on the hearth drive shaft to provide a consistent current path to ground. Gettered argon was used for the atmosphere in the furnace. For the initial growth, the melt was seeded with a tungsten rod. Subsequent growths of oriented crystals were nucleated with oriented seeds silver soldered onto the end of the seed rod in the place of the tungsten. For all growths, the seed rod rotation rate was 9 min^{-1}; the hearth rotation rate was 60 min^{-1} and the pull rate was 20 mm h^{-1}. Four samples were spark cut in a slice-like fashion from one single crystal. X-ray measurements established that the samples were single crystals with the c axis pointing perpendicular to the direction of the spark cut. The dimensions of the sample for resistivity and Hall effect measurements was $6.6 \times 5.1 \times 0.28$ mm^3. A cylindrical sample for resistivity measurements was a pulled crystal.

The DC magnetic susceptibility was measured with a SQUID magnetometer designed by Quantum Systems with the field perpendicular and parallel to the c axis of the sample as it was heated from 4.2 to 300 K. The molar susceptibility is shown in figure 1. With the magnetic field parallel to the c axis it shows a broad maximum at 50 K and is an order of magnitude larger than with the field perpendicular to the c axis.

Six silver paste contacts were attached to the sample, which was mounted on the brass sample holder with GE varnish so that the c axis was normal to the (vertical) probe direction. Current and resistance contacts were attached at each end of the sample, all four in a straight line parallel to the probe direction. The magnetic field applied perpendicular to the current direction was provided by an electromagnet with a maximum field of 1.9 T. The two remaining contacts were attached as accurately as possible on opposite sides of the sample so that they were in a line perpendicular to the direction of the current flow for Hall voltage measurements.

The resistance voltage was measured with a current of 10 mA as the sample was heated from 1.2 to 300 K. The absolute error in the resistivity is 30% because of the

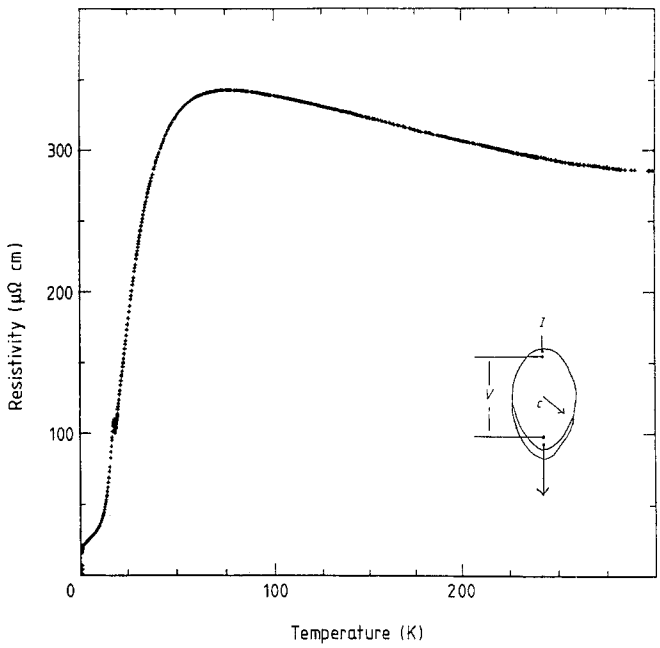

Figure 2. Temperature dependence of the DC resistivity of URu_2Si_2 with the current perpendicular to the c direction.

error introduced by the three dimension measurements required in its calculation and to a lesser extent by the unideal sample and contact geometry. However, the relative error is less than 0.2%. Absolute measurement of the susceptiobility and Hall coefficient are much better since they only depend on accurate measurement of the sample's mass and thickness, respectively.

The resistivity rises slowly with decreasing temperature to a maximum at 70 K and then drops quickly below about 50 K as shown in figure 2. The superconducting transition is at 1.33 K (defined as the temperature at which the resistivity drops by half) and the 10–90% transition width is 0.1 K. There is a small sharp peak at 17.5 K.

The Hall voltage $V_H(B)$ was determined from the potential $V(B)$ between the transverse contacts with the magnetic field parallel to the c axis according to

$$V_H(B) = \tfrac{1}{2}(V(B) - V(-B)).$$

The magnetic field from the electromagnet was rotated to change the field direction. The Hall voltage was linear with field up to the maximum field of the magnet showing that the Hall coefficient is a relevant experimental quantity. The Hall coefficient was calculated from

$$R_H(T) = V_H(B)t/BI$$

where t is the sample thickness, B is the magnetic field and I is the current.

The uncertainty in R_H below 100 K has been estimated from the signal noise and above 100 K, from the standard deviation of the ten or so measurements taken. The sign of the Hall coefficient was determined by Schoenes *et al* (1987) and is positive as in all other heavy fermion metals. Shown in figure 3, the Hall coefficient has a relative maximum at 50 K. At 18 K, R_H jumps by a factor of more than four and then decreases slowly down to 2 K. The Hall coefficient at 4.2 K is 13×10^{-3} cm^3 C^{-1} which is slightly larger than that measured by Schoenes *et al* (1987).

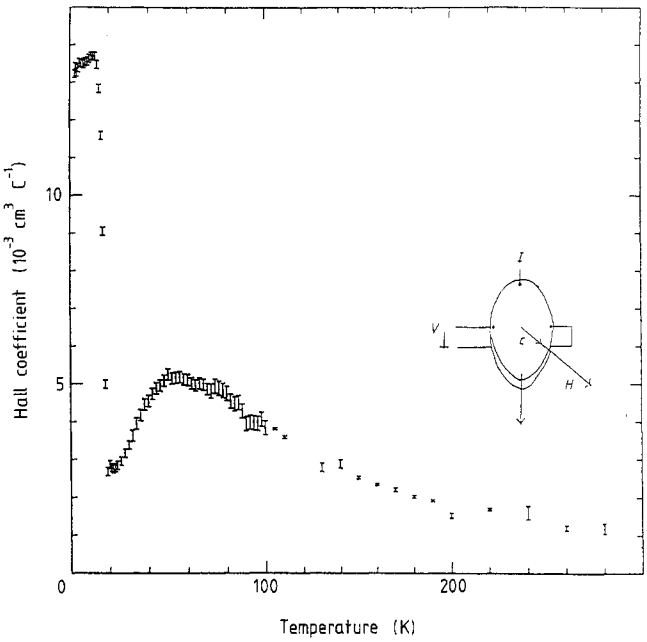

Figure 3. Temperature dependence of the DC Hall coefficient of URu_2Si_2 sample with the magnetic field parallel to and the current perpendicular to the c axis.

4. Discussion

At high temperature, a linear regression of χ^{-1} against T was calculated. Above 100 K, the susceptibility obeyed the Curie–Weiss law (figure 4) with $\mu_{\text{eff}} = (3.99 \pm 0.02)\mu_B$ and $\theta = (-110 \pm 3)$ K. μ_{eff} does not correspond to that of trivalent ($\mu_{\text{eff}} = 3.62\mu_B$) or tetravalent ($\mu_{\text{eff}} = 2.68\mu_B$) uranium. The anisotropy of χ is the reverse of what one usually expects for an antiferromagnet. Below T_N, it is easier to magnetise perpendicular to the direction of the staggered magnetisation, so $\chi(\boldsymbol{H} \perp c)$ should be greater. The opposite result indicates that an anisotropy of the crystal field strongly constrains the f moments to lie in the c direction. This is supported by the fact that the spin wave excitations are polarised along the c direction (Broholm *et al* 1987). The divergence of χ from Curie–Weiss behaviour is not surprising, since we know from the anisotropy that the levels are split by the crystal field and make the Curie–Weiss theory inapplicable.

There is a small anomaly in $\chi(T)$ at approximately 18 K. It comes from the transition to the antiferromagnetic state at low temperatures.

In analysing the high-temperature resistivity, we have scaled the expression determined by Schoenes *et al* (1987) from measurements up to 1200 K by the ratio of the maximum resistivity ρ_{max} measured in this work to the ρ_{max} measured by Schoenes *et al*

$$\rho(\mu\Omega\ \text{cm}) = 886.7 + 0.076T - 110 \ln T.$$

The second term is the first-order correction at high temperature due to electron–phonon scattering and the third is the Kondo term. As is shown in figure 5, the data diverge from the curve below 250 K, so the resistivity is Kondo like only above 250 K.

At the onset of coherence, between T_N and T_{max}, the resistivity drops rapidly and is presumably the region where the behaviour is crossing over from Kondo-like behaviour to Fermi liquid behaviour, modified by the onset of critical behaviour at T_N.

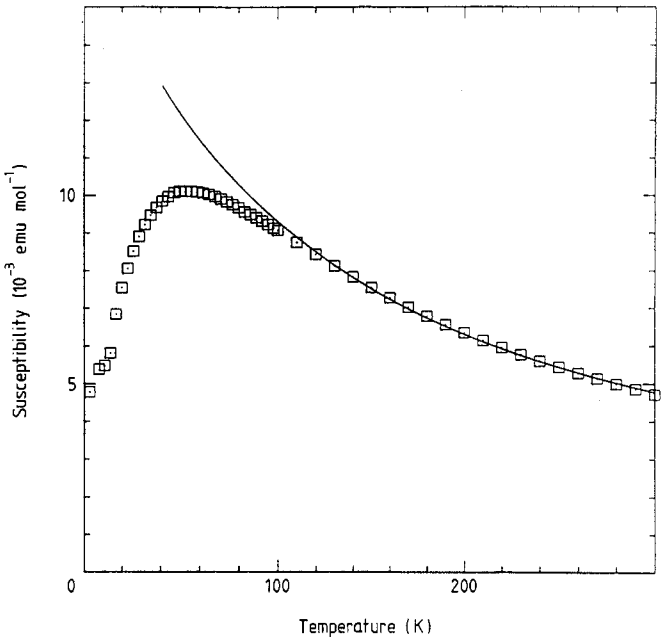

Figure 4. Magnetic susceptibility of URu_2Si_2 with $\boldsymbol{B} \| \boldsymbol{c}$. The solid line is the fitted Curie–Weiss curve.

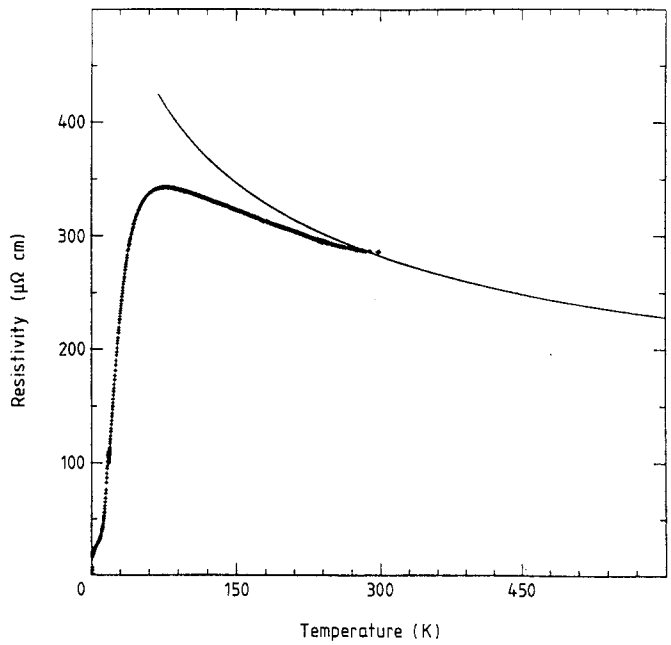

Figure 5. Temperature dependence of the resistivity of URu_2Si_2. The full curve is the fit after Schoenes *et al.*

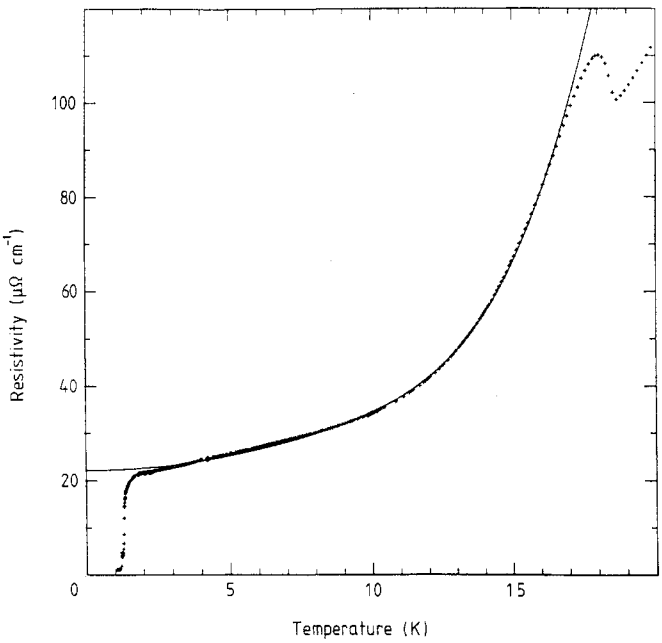

Figure 6. Low-temperature resistivity of URu_2Si_2 with the current perpendicular to the c axis. The full curve is the low-temperature fit described in the text.

Because URu_2Si_2 is an antiferromagnet with energy gap spin waves, the low-temperature resistivity was fitted to a T^2 Fermi liquid term plus an electron spin-wave scattering term. Since the Fermi surface gap measured in the specific heat is large (Palstra *et al* 1985) and since there are still 'free' carriers in the antiferromagnetic state as shown below, we need not consider the effect of the Fermi surface restructuring in the resistivity. The resistivity between 2 and 16 K was thus fitted to the expression

$$\rho(T) = \rho_0 + AT^2 + bT(1 + 2T/\Delta)\,e^{-\Delta/T}.$$

The results are: $\rho_0 = 22.2 \pm 0.2\ \mu\Omega$ cm, $A = 0.119 \pm 0.003\ \mu\Omega$ cm K^{-2}, $b = 648 \pm 100\ \mu\Omega$ cm, $\Delta = 99 \pm 3$ K.

The fit is shown in figure 6. The value of A gives a value for the Kadowaki–Woods ratio of $R = A/\gamma^2 = 0.4 \times 10^{-5}\ \mu\Omega$ cm (mol K mJ^{-1})$^{-2}$, which is comparable with their value of 1.0×10^{-5}. The value of the spin wave gap Δ does not agree well with the gap of 21 K measured directly with neutrons (Broholm *et al* 1987) but is surprisingly close to the Fermi surface gap of 115 K determined from the specific heat (Palstra *et al* 1985).

From susceptibility measurements in $CeAl_3$ and $CeRu_2Si_2$ (Buschow and Fast 1985, Gupta *et al* 1983, Lapierre *et al* 1987) for the Kondo temperature T_K, the temperature was divided into two regions, $T_0 \leqslant T < T_K$ and $T \geqslant T_K$, which were used in fitting the Hall coefficient in these heavy-fermion metals. It seems that they have taken T_K as either θ in the Curie–Weiss expression or as the temperature at which the susceptibility diverges from Curie–Weiss-like behaviour. In our material both these temperatures are around 100 K, but in this work, we have assumed that we can use T_K as an additional parameter in the Fert–Levy fit. Our modified fitting procedure will therefore yield an experimental Kondo temperature.

The total Hall coefficient can be written as

$$R_H(T) = R_0 + R_H^{EX}(T) + [R_H(T = 0) - R_0]\chi(T)/\chi(T = 0).$$

R_0 is the ordinary temperature-independent Hall coefficient derived by extrapolating

the high-temperature data on the graph of $\chi(T)\rho(T)$ versus $R_H(T)$ to $\chi\rho = 0$. The last term accounts for the skew scattering caused by defects and impurities, and is assumed to follow a temperature dependence given by the Hurd (1972) expression.

In our analysis the last term has been dropped. Fert and Levy have shown that it is small in another heavy-fermion metal. Analysis using this term would be difficult considering the jump in R_H and the subsequent difficulty in determining $R_H(T = 0)$.

The resistivity due to resonant scattering is

$$\rho_m(T) = \rho_{TOT}(T) - \rho_{el\text{-}ph}(T) - \rho_0.$$

The residual resistivity is $\sim 22\ \mu\Omega$ cm and we can estimate $\rho_{el\text{-}ph}(T)$ from the linear contribution to the resistivity to be $(0.08\ \mu\Omega\ \text{cm K}^{-1})T$ which is $24\ \mu\Omega$ cm at 300 K. Since, in the region of interest, ρ_{TOT} is everywhere greater than $300\ \mu\Omega$ cm, we drop ρ_0 and $\rho_{el\text{-}ph}(T)$ and take $\rho_m(T) = \rho_{TOT}(T)$.

The problem has been reduced to fitting curves of $R_H(T)$ against $\chi(T)\rho(T)$. We therefore must know all three of R_{Hi}, ρ_i, and χ_i, at the same temperature T_i. The standard natural cubic spline interpolation technique, which fits cubic polynomials between adjacent data points, was implemented. While the natural spline fit creates a very smooth looking curve, it is constrained to go through all the data points, so that extraneous points are not 'filtered out' as in, say, a polynomial fit. a spline was done on each of the temperature dependence data sets and yielded interpolated points for $R_H(T_i)$, $\rho(T_i)$ and $\chi(T_i)$ for T_i between 4 and 300 K equally spaced by 0.5 K.

The coherence temperature T_0 is taken as 50 K, the temperature at which the Hall coefficient goes through a relative maximum. We then fitted two straight lines to the interpolated points $R_H(T_i)$ against $\chi(T_i)\rho(T_i)$ in the region $50\ \text{K} \leqslant T_i \leqslant 300\ \text{K}$. A computer fit was done with four variable parameters a_1, a_2, R_0 and T_K defined by

$$R_H(T_i) = R_0 + a_1\chi(T_i)\rho(T_i) \qquad \text{for } T_K < T_i$$

$$R_H(T_i) = R_0 + a_2\chi(T_i)\rho(T_i) \qquad \text{for } 50 \leqslant T_K \leqslant T_i.$$

The fit is optimised with a Kondo temperature of 90 ± 30 K and parameters

$$a_1 = 0.0156 \pm 0.0001\ (\mu\Omega\ \text{cm C})^{-1}$$

$$a_2 = 0.0182 \pm 0.0002\ (\mu\Omega\ \text{cm C})^{-1}$$

$$R_0 = (-1.18 \pm 0.37) \times 10^{-3}\ \text{cm}^3\ \text{C}^{-1}.$$

The uncertainties have been determined by calculating the extreme values of each quantity for which χ^2 is less than twice its optimum value.

The fit is shown figure 7. Notice the 'crossover' region, centred about T_K, where the Hall coefficient changes from that of one limit to that of the other. The fit extended below T_0 seems to be satisfactory as Fert and Levy noticed for $CeAl_3$ and $CeCu_6$. We can calculate

$$\gamma_1 = a_1 C_{cw} = 0.306\ \text{K}/T$$

$$\gamma_2 = a_2 C_{cw} = 0.356\ \text{K}/T.$$

$\gamma_2/\gamma_1 > 1$ and γ_1 and γ_2 are three times those of $CeAl_3$ (where $\gamma_2/\gamma_1 > 1$) and $CeCu_6$ (where $\gamma_2/\gamma_1 < 1$).

From expressions for γ_i and using $g\mu_B = \mu_{eff}^{exp}$, we can deduce that

$$\sin(\delta_2)\cos(\delta_2) = -0.0823$$

$$\sin(2\delta_3 - \delta_2)\sin(\delta_2)/\sin^2(\delta_3) = -0.1791$$

so $\delta_2 = -0.0827$ and $\delta_3 = -0.0396$. The former is close to -0.078 and -0.09, the δ_2-values calculated by Fert and Levy for $CeAl_3$ and $CeCu_6$, respectively. These can only

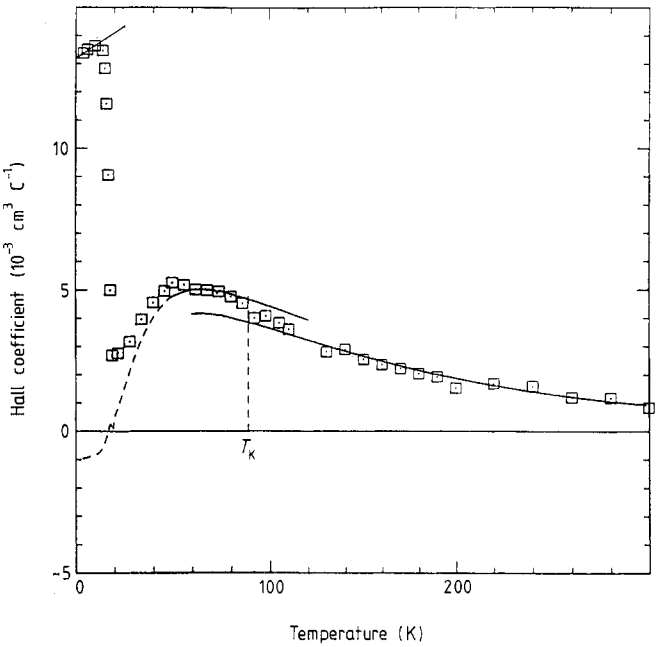

Figure 7. Temperature dependence of the DC Hall coefficient of URu_2Si_2 with Ic. The full curves are the Fert–Levy fit above 50 K and the low-temperature linear fit. The broken curve is the Fery–Levy fit extended below 50 K.

be used to estimate the phase shifts since the significant effect of crystal field splitting has not been taken into account in the expressions.

With their high-temperatures resistivity measurements, Schoenes *et al* (1987) calculated $T_K \sim 370$ K and stressed that, unlike in other heavy-fermion metals T_0 and T_K have to be sharply distinguished in URu_2Si_2. Our experimentally derived Kondo temperature, 90 ± 30 K, which agrees with 100 K, the temperature at which the susceptibility diverges from Curie–Weiss behaviour, shows, however, that the distinction between the Kondo temperature T_K and the coherence temperature T_0 is not a sharp one in URu_2Si_2.

The jump in the Hall coefficient occurs at the Néel temperature and is probably the result of a restructuring of the Fermi surface. Above T_N, we get from the Fert and Levy fit that $n_{eff} = p_V - n_c = 1/R_0 = 0.43 \pm 0.14$ electrons FU^{-1}, where p_V is the density of holes. Fitting a straight line to R_H below T_N, we find that $R_H(T = 0) = 13.2 \pm 0.7$ mm^3 C^{-1}, so $n_{eff} = 0.0392 \pm 0.002$ holes FU^{-1}. We can analyse this change in the density of carriers as if it occurred from a gap in the Fermi surface opened up by spin density waves.

In a one-dimensional model, if the effective number is equal to the total number of carriers, $n_{eff} = 0.4$ electrons FU^{-1}, then since there are two electron states per band, we get that the Fermi wavevector $k_F = 0.2k_{BZ}$ where k_{BZ} is the Brillouin zone edge. If there were some sort of coupling of the electrons to the antiferromagnetism, then below T_N a gap would be opened up at $k = k_{BZ}/2$ and not at k_F. The antiferromagnetism itself would therefore not be sufficient to cause the observed jump in the Hall coefficient. If, however, there were spin density waves present with a wavevector Q slightly greater than $0.2k_{BZ}$, then a gap would be opened sightly above k_F causing the number of carriers to change from electrons to a smaller number of holes. We therefore conclude that the jump can

be explained as resulting from the formation of spin density waves below the Néel temperature with a wavevector $Q = 0.2k_{BZ}$.

In three dimensions, the argument is less clear, but from Friedel's relation, using the density of electrons in the Fermi sphere, $\nu = \Omega k_F^3/3\pi^2$, where Ω is the volume per U atom, assuming $l = 3$ and taking $x = \frac{1}{5}$ we get

$$\rho_{max} = 12\pi^3\hbar\chi(2l + 1)/e^2\Omega k_F^4.$$

The result is that the Fermi wavevector in the c direction is 0.94 $Å^{-1}$. The Brillouin zone edge, which is at 0.65 $Å^{-1}$ in the c direction, may thus cut off a small pocket of electrons whose small Fermi surface is cut by spin density wave planes.

5. Conclusions

We have shown that the DC magnetic susceptibility of URu_2Si_2 is Curie–Weiss like at temperatures above 100 K with $\mu_{eff} = 3.99\mu_B$ and $\theta = 110$ K. μ_{eff} does not correspond to any value calculable from Hund's rule. The anisotropy of χ implies that the moments are strongly held in the c direction.

The high-temperature resistivity of URu_2Si_2 perpendicular to the c direction agrees with the behaviour observed by Schoenes *et al* (1987) and is described in terms of the U atoms behaving as a lattice of isolated Kondo impurities. The system becomes superconducting at 1.33 K and the low-temperature resistivity can be described as Fermi liquid behaviour combined with electron–boson scattering by antiferromagnetic spin waves with an energy gap of 99 ± 2 K. The discrepency of this value with that measured by neutron scattering remains unexplained.

The temperature dependence of the Hall coefficient of URu_2Si_2 with the field parallel to the c direction can be described with the model of Fert and Levy, which accounts for the intrinsic skew scattering of electrons in heavy Fermion metals. From our modified analysis, an experimental Kondo temperature of 90 K is derived which agrees well with 100 K, the temperature below which the susceptibility is no longer Curie–Weiss like. This is not surprising, since both are temperatures beyond which the f moments are expected to behave as local, isolated moments. Our work suggests that T_K and $T_0 \simeq$ 50 K, the coherence temperature, do not have to be sharply distinguished in URu_2Si_2.

From the Hall coefficient, it is evident that there is a change in the effective number of carriers at the Néel temperature. This change may result from the existence of spin density waves in the antiferromagnetic state which can open up an energy gap at the Fermi surface.

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References

Aeppli G, Bucher E, Broholm C, Kjelms J K, Baumann J and Hufnage J 1988 *Phys. Rev. Lett.* **60** 615

Anderson N H 1980 *Crystalline Field and Structural Effects in f-Electron Systems* ed. J E Crow, R P Guertin and T W Mihalism (New York: Plenum) p 373

Broholm C, Kjelms J K, Buyers W J L, Matthews P, Palstra T T M, Menovsky A A and Mydosh J A 1987 *Phys. Rev. Lett.* **58** 1467
Buschow K H J and Fast J F 1965 *Z. Phys. Chem., NF* **50** 1
Cooper S L, Klein M V, Maple M B and Torkachvii M S 1987 *Phys. Rev.* B **36** 5743
Cordier G, Czech E, Schafer H and Woll P 1985 *J. Less-Common Met.* **110** 327
Dugdale J S 1977 *Electrical Properties of Metals and Alloys* (London: Arnold)
Fert A and Levy P M 1987 *Phys. Rev.* B **36** 1907
Friedel J 1958 *Nuovo Cimento Suppl.* **7** 288
Gupta L C, MacLaughlin D E, Chen Tien, Godart C, Edwards M A and Parks R D 1983 *Phys. Rev.* B **28** 3673
Hadzic-Leroux M, Hamzic A, Fert A, Haen P, Lapierre F and Laborde O 1986 *Europhys. Lett.* **1** 579
Holland-Moritz E, Schlabitz W, Loewenhaupt M, Walter U and Loong C K 1987 *J. Magn. Magn. Mater.* **63** and **64** 187
Hurd C M 1972 *The Hall Effect in Metals and Alloys* (New York: Plenum)
Kadowaki F and Woods S B 1986 *Solid State Commun.* **58** 507
Lapierre F, Haen P, Briggs R, Hamzic A and Kappler J P 1987 *J. Magn. Magn. Mater.* **63**/**64** 338
Lee P A, Rice T M, Serene J W, Sham L J and Wilkins J W 1986 *Comment. Condens. Matter Phys.* **12** 99
Maple M B, Chen J W, Dalichaouch Y, Kohara T, Rossel C and Torikachvili M S 1976 *Phys. Rev. Lett.* **56** 185
Palstra T T M, Menovsky A A and Mydosh J A 1986 **33** 6527
Palstra T T M, Menovsky A A, Van Der Berg J, Dirkmaat A J, Kes P H, Nieuwenhuys G J and Mydosh J A 1985 *Phys. Rev. Lett.* **55** 2727
Rauchschwalbe U 1987 *Physica* B **147** 1
Schlabitz W, Bauman J, Pollit B, Rauchschwalbe U, Mayer H M, Ahlheim U and Brendl C D 1986 *Z. Phys.* B **62** 171
Schoenes J, Schonenberger C, Franse J J M and Menovsky A A 1987 *Phys. Rev.* B **35** 5375
Stewart C R 1984 *Rev. Mod. Phys.* **56** 755
Walter U, Loong C-K, Loewenhaupt M and Schlabitz W 1986 *Phys. Rev.* B **33** 7875